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Impact of the interface magnetic disorder on the exchange bias between ferromagnetic and antiferromagnetic layers

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Abstract

In a substrate/ferromagnetic/antiferromagnetic/ferromagnetic trilayer, the exchange coupling occurring at the bottom ferromagnetic/antiferromagnetic interface is always found to be weaker than the one at the top antiferromagnetic/ferromagnetic interface after thermal treatment. We show clearly in this paper that this effect is related to the degree of magnetic disorder at the ferromagnetic/antiferromagnetic interface during the multilayer growth. The larger the magnetic disorder the weaker is the exchange bias field in the as-deposited bilayer, as expected, but, counterintuitively, the stronger is the exchange bias field after a subsequent in-field annealing.

The exchange bias phenomena exerted by an antiferromagnetic (AF) layer on a ferromagnetic (FM) layer in close contact (in an FM/AF bilayer) has been shown, since the pioneering work done by Meiklejohn and Bean [1], to be strongly dependent on the AF layer grain size, crystallographic structure or roughness [2]. When the exchange bias is set only by applying a field during the growth it is always found to be much larger in substrate/FM/AF systems (top biased) than in substrate/AF/FM systems (bottom biased), as shown again recently [3]. However, when a thermo-magnetic annealing is used to set up the exchange after the growth, it is nearly always found that the exchange bias occurring in top biased substrate/FM/AF systems is smaller than in bottom biased substrate/AF/FM systems [2]. In a previous contribution on substrate/Py/IrMn/Py multilayers [4], we showed that this effect is associated to the different magnetization configurations in the Py layer at the Py/IrMn and IrMn/Py interface. Indeed, the IrMn/Py interface appears to be unbiased after the multilayer growth but it acquires a larger bias field than the Py/IrMn interface during the subsequent in-field annealing. However, those conclusions were built up by comparing the magnetic states of two different Py layers. In this paper, we consider the magnetic states of a single FM layer in contact with AF layers and show the influence of these states on the surface exchange

coupling energy J_{ex} . We clearly demonstrate that, after a thermo-magnetic annealing, the interface exchange bias energy increases continuously from the value $J_{\text{ex}}^{\text{inf}}$ it takes in a top biased FM/AF bilayer to the value $J_{\text{ex}}^{\text{sup}}$ observed in a bottom biased AF/FM bilayer when the remnant magnetization of the FM layer decreases.

Multilayers were deposited onto float-glass substrates by sputtering Ir₂₀Mn₈₀ (IrMn), Co, Pt and Ta targets in an Alcatel SCM 650 sputtering machine. Each multilayer was deposited at an operating Ar pressure fixed to 5×10^{-3} mbar and the substrates were maintained at room temperature. All stacks were grown onto a 5 nm thick Ta buffer layer and subsequently covered by a 2.5 nm thick Pt protection layer. Hereafter the thickness of the various layers is quoted between parentheses (in nm). Several sets of samples have been studied.

- Basic trilayer referenced as (S0): Co(10)/IrMn(x)/Co(5), with x varying from 5 to 20 nm.
- Trilayer with reduced top FM thickness referenced as (S1): Co(10)/IrMn(10)/Co(x) ($x = 1.5$ –5 nm).
- Reversed upper stack with the insertion of an exchange breaking Pt layer referenced as (S2): Co(10)/IrMn(10)/Pt(1)/Co(x)/IrMn(10) ($x = 1.5$ –5 nm).
- Double stack referenced as (S3): Co(10)/IrMn(10)/Co(x)/IrMn(10) ($x = 1.5$ –5 nm).

After growth, the magnetic properties of the multilayer stack were checked either in the virgin state or after a thermal annealing for 30 min at 200 °C with a 300 Oe in plane applied magnetic field. This thermal treatment was made after growth in a separate chamber with a base pressure of 5×10^{-7} mbar.

The structural and magnetic properties of all series were carefully studied. As shown in our previous study on Py/IrMn(t_{IrMn})/Py and Py/IrMn(t_{IrMn})/Pt/Py/IrMn multilayers [4], the layer thicknesses are in agreement with deposited thicknesses, and the crystallographic quality of the multilayer stack and the interface roughness do not depend on t_{IrMn} nor on the insertion of a thin 1 nm thick Pt layer into the layer stack. Then, any change in the surface exchange coupling energy can only be related to the magnetic history of the sample. However, when both Py layers are replaced by Co layers, the IrMn layer is less textured because of the quasi amorphous structure of the Co underlayer. That results in a clear reduction of the (111) IrMn peak intensity in the $\theta/2\theta$ geometry.

The magnetic properties of series (S0) were checked after thermal annealing as a function of IrMn layer thickness using Kerr magnetometry or vibrating sample magnetometry at room temperature. As observed in Py/IrMn(x)/Py multilayers [4], the exchange bias strength at the top IrMn/Co interface is very weak in the as-deposited stacks but, after thermo-magnetic annealing, it is always found to be larger than at the bottom Co/IrMn interface (figure 1). This extends the universality of the effect to other FM materials. Here again, the symmetry breaking can only be explained by differences in the way exchange anisotropy is written at the interface. The disordered magnetic structure of an unbiased antiferromagnetic layer, grown before the ferromagnetic one, allows a better polarization after in-field annealing than the partly oriented structure of the same layer grown onto a saturated ferromagnetic layer.

As in our previous study of Py/IrMn(x)/Py multilayers, the conclusions are drawn by comparing the magnetic states of two different (bottom and top) Co layers. In order to have a more direct insight on the effect of the AF layer polarization during the growth we have undertaken another series of measurements in which the degree of saturation of the FM layers could be varied.

The magnetic properties of our polycrystalline Co layers made of small magnetic grains coupled by exchange interactions can be understood on the basis of models for magnetization reversal relying on the well-known ripple domain configuration in thin magnetic films [5–10]. On a macroscopic scale, the layers are magnetically isotropic due to a random orientation of the

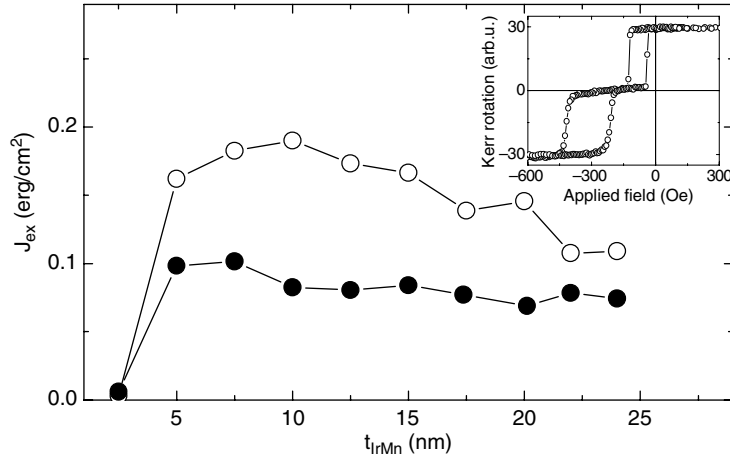


Figure 1. Surface coupling energy J_{ex} evaluated from equation (3) for glass/Ta(5 nm)/Co(10 nm)/IrMn(t_{IrMn} nm)/Co(5 nm)/Ta(5 nm) trilayers ((O) for Co(5 nm) and (●) for Co(10 nm)), with t_{IrMn} varying from 2.5 to 24 nm. Inset: magnetization curve measured on a glass/Ta(5 nm)/Co(10 nm)/IrMn(10 nm)/Co(5 nm)/Ta(5 nm) trilayer.

easy magnetic axis of each grain. However, on a microscopic scale an effective local anisotropy can be defined as well as an effective correlation, the length scale of which is characterized by the exchange correlation length, l_{ex} . This correlation length is often larger than the grain size and is very sensitive to spatial variations of the anisotropy, to the magnetic moment, and to the coupling strength between grains. Therefore the local effective anisotropy varies over a distance l_{ex} , and one can define a maximum deviation angle, Λ_{max} , of the local magnetization from the mean magnetization in the magnetic layer. The larger Λ_{max} , the lower is the resulting remnant magnetization. Since, in polycrystalline thin films, the inter-grain exchange coupling strength depends on the thickness of the film [6, 10], Λ_{max} and the remnant magnetization can be tuned by varying the layer thickness according to the equation [6]

$$\Lambda_{\text{max}} \propto \frac{K_{\text{FM}}}{M_{\text{S}}^{1/2} t_{\text{FM}}^{1/4}} \quad (1)$$

where M_{S} and t_{FM} are the magnetization and thickness of the FM layer respectively and K_{FM} is the local anisotropy. Therefore the reduction of the ferromagnetic layer thickness provides a means to increase its magnetic disorder despite the presence of the magnetron stray fields during the growth.

The simplest experiment would have been to vary the degree of saturation of the FM underlayer in an FM/AF structure. As far as a single Co(x) layer is concerned we have shown [11] that the remnant magnetization M_{r} decreases from 1 to 0.85 when x is brought from 5 to 1.5 nm. Thus, and unfortunately, M_{r} keeps a significant value, a sign of a small value of the angular dispersion Λ_{max} . Furthermore, the coercive field of the bottom Co layer keeps a small value, less than the stray field of the magnetron, and even the thinnest layer is still saturated during the deposition of the capping IrMn layers.

Therefore, we tailored the properties of the top Co layer by changing its thickness. As the top surface of the antiferromagnetic layer is not polarized there is no exchange bias of the top ferromagnetic layer, on average, but each grain is still subjected to an exchange field that is random both in strength and orientation. Hence the exchange coupling between the ferromagnetic and the antiferromagnetic grains gives rise to a supplementary contribution to

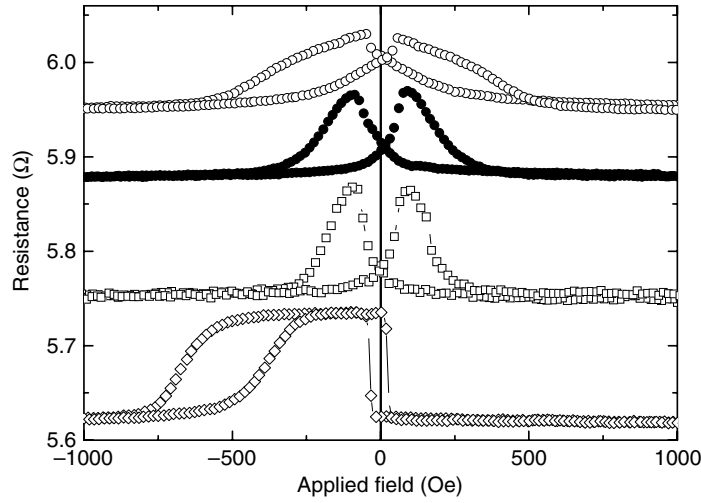


Figure 2. Resistance versus magnetic field of Co(10)/IrMn(10)/Co(x)/Cu(3.5)/Co(7) SV with $x = 2$ nm (○), $x = 4$ nm (●) and $x = 5$ nm (□) measured before thermal treatment. (◇): resistance versus magnetic field of Co(10)/IrMn(10)/Co(2)/Cu(3.5)/Co(7) SV measured after thermal treatment.

the effective anisotropy of the ferromagnetic grains. Since this is a surface contribution K_{ex} to the effective grain anisotropy the formula (1) has to be adapted to take it into account. Then the maximum deviation angle, Λ_{max}^* , of the local magnetization from the mean magnetization in the top magnetic layer becomes

$$\Lambda_{\text{max}}^* \propto \frac{\frac{K_{\text{ex}}}{t_{\text{FM}}} + K_{\text{FM}}}{M_{\text{S}}^{1/2} t_{\text{FM}}^{1/4}}. \quad (2)$$

Now Λ_{max}^* shows a much larger dependence on t_{FM} than the mere $t_{\text{FM}}^{1/4}$ of the FM layer alone, which has been checked on series (S1).

In order to measure separately the magnetic response of the top Co(x) layer in the Co(10)/IrMn(10)/Co(x) trilayers, these were covered by a /Cu(3.5)/Co(7) bilayer so as to build a spinvalve (SV) structure Co(10)/IrMn(10)/Co(x)/Cu(3.5)/Co(7). Owing to the low saturation field of the topmost Co(7) layer (figure 2, narrow loop around $H = 0$, (◇)), the variation of the spin valve resistance is characteristic of the Co(x) magnetic response only. Indeed, the 2% of magnetoresistance signal, GMR, cover the anisotropic magnetoresistance of each Co layer estimated to 0.1% and the spin valve resistance variation is only linked to the layers in contact with the Cu spacer.

In a first step, the resistance versus magnetic field characteristics, $R(H)$, of the SV were measured before thermal treatment (figure 2, ○, ● and □). It appears clearly that no exchange bias exists at the IrMn/Co(x) interface in the virgin state: the IrMn/Co(x) interface is not polarized, and this fact is independent of the Co thickness. This result was understood [4] by considering that the in-plane crystallographic grain axes are randomly distributed and that the IrMn magnetic moments at the top of the IrMn layer orient themselves in any easy direction since they are not constrained yet by exchange coupling with a polarized ferromagnetic layer. Therefore, through the local exchange between IrMn and Co(x), a very thin Co(x) layer in contact with IrMn is demagnetized. Then, with increasing thickness, it is more and more easily saturated. This is clearly observed in the figure as a decrease of the loop width in accordance

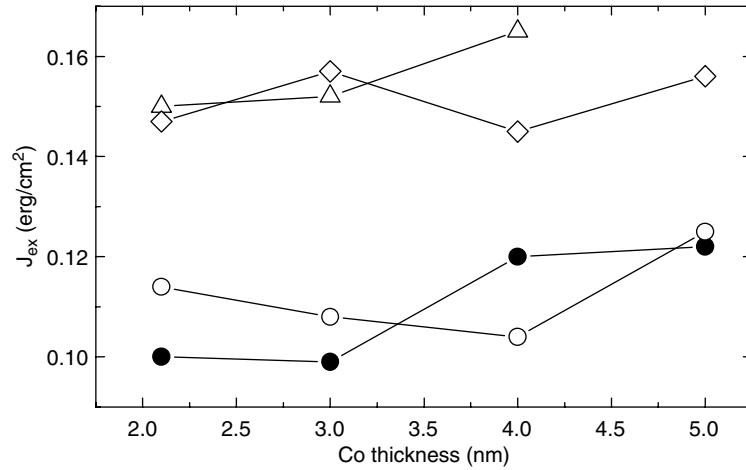


Figure 3. Variation of $J_{\text{ex}}^{\text{sup}}$ as a function of x in two series of Ta(5 nm)/Co(10 nm)/IrMn(10 nm)/Co(x)/Pt multilayers (Δ and \diamond). Variation of $J_{\text{ex}}^{\text{inf}}$ as a function of x in two series of Ta(5 nm)/Co(10 nm)/IrMn(10 nm)/Pt(1 nm)/Co(x)/IrMn(10 nm)/Pt multilayers (\circ and \bullet).

with the $\frac{1}{t_{\text{FM}}}$ dependence of (2). Furthermore, with increasing x , the GMR at zero applied field decreases, whereas its maximum amplitude increases, which shows that the angular dispersion of the layer magnetization decreases as expected from (2) while its remanence increases.

In a second step, after an in-field thermal annealing, the GMR signal shows a large exchange bias, independent of x (figure 2, \diamond) and equal to the GMR measured before annealing for the thickest layer ($x = 5$ nm). This shows that, in opposition to the virgin state, the angular dispersion of the magnetization has become narrow, whatever the value of x , while the initial random magnetization at the top of the AFM layer has become unidirectional.

This first set of experiments confirms the theoretical expectation that we are able to vary significantly the magnetization dispersion of the Co(x) layer when grown onto an unpolarized IrMn surface. Then we measured the surface exchange coupling energy J_{ex} at the interfaces of the top Co(x) layer: lower interface only (S1), upper interface only (S2) and both interfaces simultaneously (S3) as a function of the layer thickness.

The surface exchange coupling energy J_{ex} after in-field annealing was evaluated from the shift of the magnetization curve according to

$$J_{\text{ex}} = H_{\text{ex}} M_s t_F \quad (3)$$

where H_{ex} is the shift of the FM layer hysteresis loop, and M_s and t_{FM} are the magnetization and thickness of the FM layer, respectively.

The exchange coupling energy at the IrMn(10)/Co(x) interface (S1) is found to be independent of x and the largest of all (figure 3, Δ and \diamond). We label it $J_{\text{ex}}^{\text{sup}}$. Its value is around $0.155 \text{ erg cm}^{-2}$.

The exchange coupling energy in samples of series (S2) Co(10)/IrMn(10)/Pt(1)/Co(x)/IrMn(10) corresponds to the Co(x)/IrMn(10) interface only, thanks to the exchange breaking Pt layer. It is also found to be independent of x and its strength is the smallest of all, around $0.115 \text{ erg cm}^{-2}$. We label it $J_{\text{ex}}^{\text{inf}}$. One can note that $J_{\text{ex}}^{\text{inf}}$ is also the strength of the exchange coupling strength at the saturated bottom Co(10) layer, which is understood because Co(x) is decoupled from its IrMn underlayer and, thus, is saturated during the growth for any thickness.

Finally, the surface exchange coupling energy on the Co(x) layer in samples of series (S3) Co(10)/IrMn₁(10)/Co(x)/IrMn₂(10) was measured. The label IrMn₁ (respectively IrMn₂)

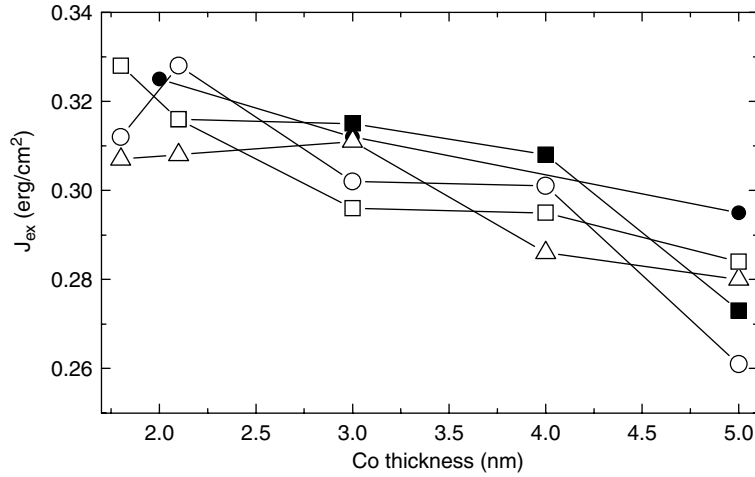


Figure 4. Variation of $J_{\text{ex}}^{\text{eff}}$ for five series of samples as a function of x in Ta(5 nm)/Co(10 nm)/IrMn(10 nm)/Co(x)/IrMn(10 nm)/Pt multilayers.

corresponds to the lower (respectively upper) IrMn layer, both coupled to the Co(x) layer. The coupling energy $J_{\text{ex}}^{\text{eff}}$ now results from the sum of the exchange energies J_{ex}^1 at the IrMn₁(10)/Co(x) interface and J_{ex}^2 at the Co(x)/IrMn₂(10) interface:

$$J_{\text{ex}}^{\text{eff}}(x) = J_{\text{ex}}^1(x) + J_{\text{ex}}^2(x). \quad (4)$$

Since J_{ex}^1 and J_{ex}^2 are unaffected by the degree of saturation of Co(x) during the growth, one would expect $J_{\text{ex}}^{\text{eff}}(x)$ to be equal to $J_{\text{ex}}^{\text{inf}} + J_{\text{ex}}^{\text{sup}} = 0.27 \text{ erg cm}^{-2}$ independent on x . Actually this is the value found for the thickest Co layer ($x = 5 \text{ nm}$) (figure 4). However, as soon as x is reduced, we can observe that $J_{\text{ex}}^{\text{eff}}(x)$ increases and reaches $J_{\text{ex}}^{\text{eff}}(1.5) = 0.31 \text{ erg cm}^{-2}$, that is twice the value of $J_{\text{ex}}^{\text{sup}}$. This is shown in figure 4 for five different series of samples. Since during the whole process the IrMn₁(10)/Co(x) interface has experienced the same conditions as in the (S1) series, the variation of $J_{\text{ex}}^{\text{eff}}(x)$ cannot be attributed to that of J_{ex}^1 , which must be constant and equal to $J_{\text{ex}}^{\text{sup}}$. Hence one must conclude that, in series (S3), J_{ex}^2 increases from $J_{\text{ex}}^{\text{inf}}$ to $J_{\text{ex}}^{\text{sup}}$ when x decreases from 5 nm down to 1.5 nm, whereas, in series (S2), it remains equal to $J_{\text{ex}}^{\text{inf}}$. The difference in the elaboration process of series (S2) and (S3) is that the top antiferromagnetic layer IrMn₂(10) is grown onto a saturated Co(x) layer in the former while, in the latter, it is grown onto a Co(x) layer with a large magnetization dispersion for small x values. As a consequence, in (S2) the IrMn₂ layer acquires a surface unidirectional polarization during its growth, whereas, in (S3), the angular distribution of the interfacial moments of the IrMn₂ layer gets larger and larger as the thickness of the Co underlayer decreases. Eventually the bottom interface of the IrMn₂ layer is not polarized for the thinnest Co layers: a random moment configuration is initiated during the growth of IrMn₂ on top of Co($x = 2 \text{ nm}$) similar to the one stabilized at the top of the IrMn₁ before the growth of Co(x). Thus, at both IrMn₁(10)/Co(x) and Co(x)/IrMn₂(10) interfaces, a random moment configuration exists in IrMn. It appears that such zero bias configuration after the growth leads to the largest J_{ex} after in-field annealing.

To our knowledge, this paper reports for the first time a clear experimental view of the effect of magnetic history on the exchange bias strength in AF/FM or FM/AF bilayers. We have given an unambiguous experimental proof that a larger exchange bias is finally obtained from a demagnetized AF interface, which can be induced either by depositing the AF layer

before the FM one or onto a demagnetized FM layer. In particular the in-field growth of FM/AF bilayers does not appear to be the best way to get the largest exchange bias strength. Indeed we have shown that the disordered magnetic structure of an unpolarized antiferromagnetic layer allows a better biasing after in-field annealing than the partly oriented structure of the same layer grown onto a saturated ferromagnetic layer. During the in-field annealing the induced domain configuration in the AF layer is thus strongly dependent on its initial magnetic state (polarized or not) and the early work of Malozemoff [12] considering different winding states of the bubble domains in the antiferromagnetic layer provides a good basis to understand the large difference between the exchange bias strength in the two cases.

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