Scientific report

Project title:
*Advanced spintronic devices for communication and data storage technologies based on Heusler compounds - SPINCOD*

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According to the work plan, in the corresponding period, we performed activities corresponding to the two work packages (WP), as detailed below:

**WP1. Heusler alloy thin films with perpendicular magnetic anisotropy**

- **A1.1. Elaboration of full-Heusler (FH) thin films with tuned structural, electronic and magnetic properties.**
- **A1.2. Composition, structural, morphological and magnetic characterization of FH layers.**
- **A1.3. Patterning of FH layers for resistivity and Hall measurements.**
- **A1.4. Magneto-electric characterization of single FH layers.**
- **A1.5. Ab-initio band structure calculations. Correlations with experiments**

**WP2. Elaboration and characterization of MTJ, CIP, AHE spintronic devices.**

- **A2.1. Elaboration of MTJ, PMA-MTJ, CIP-AHE heterostructures.**
- **A2.2. Structural, morphologic and magnetic characterization of the heterostructures.**
- **A2.3 Micro and nano-patterning of of MTJ, PMA-MTJ, CIP-AHE heterostructures.**
- **A2.5 Magneto-electric characterization of of MTJ, PMA-MTJ, CIP-AHE heterostructures.**
- **Study of the efficiency of the patterned devices.**
I. Realization of the targeted objectives.

The objectives targeted have been achieved, a brief description of the targeted achieved objectives is presented below:

- deposition by magnetron sputtering of polycrystalline and epitaxial Heusler of the type $Co_2Mn_xFe_{1-x}Si$. Furthermore, we have deposited films of CFB and CoFe. Those two alloy (CFB - amorphous CoFe - crystalline) are some of the most important ferromagnetic materials in spintronic devices and were deposited in order to have a benchmark to compare their properties with the ones of Heusler alloys.
- an exhaustive study was made of the structural, magnetic and electronic transport properties of the deposited films and perpendicular magnetic anisotropy was demonstrated.
- band structure ab-initio calculations were performed for alloys of the type $Co_2Mn_xFe_{1-x}Si$ in order to identify the optimum composition for desired half-metallic properties.
- CIP AHE heterostructures were deposited and characterized in terms of their structural, magnetic and electronic properties and highly efficient current induced magnetization switching was demonstrated.

II. Dissemination of the scientific results

So far, the results have been disseminated in 5 ISI papers (3 papers published in Physical Review B and 2 papers in Journal of Physics D:Applied Physics) and one invited oral presentation (11th International Conference On Physics Of Advanced Materials, Cluj-Napoca, 2016)

In the next section, we briefly report on the main published scientific results. A very brief description of the results that are in the process of publication will be also given. The report will not strictly follow the schedule of work-packages and related activities. All the studies implicate elaboration of single films and/or planar devices spintronic (CIP-AHE) which corresponds to activities A1.1 and A2.1, followed by structural, morphological and magnetic characterization of stacks (A1.2, A1.3), patterning of micrometric planar devices by UV lithography (A1.3, A2.3), than the patterned devices were characterize in terms of magneto-electric properties (A1.4, A2.5).

All the above mention activities were fulfilled using the experimental facilities available in the implementation laboratory (TUCN) while additional ferromagnetic resonance and Brillouin Light Scattering experiments were conducted in collaboration with the group of dr. M. Belmeguenai (LSPM (CNRS-UPR 3407), University Paris 13, Sorbonne Paris, France).
Magnetization switching by current induced spin–orbit torques (SOTs) in heavy metal/ferromagnetic metal/oxide structures is of great research interest due to its potential applications in the field of low power consumption spintronic devices. Here, we study the Slonczewski-like and the field-like SOT effective fields in β-W/Co2FeAl/MgO structures showing perpendicular magnetic anisotropy (PMA). We characterize the SOT effective fields using harmonic Hall voltage measurements and we point out the essential role of the planar Hall effect corrections. The ferromagnetic layer we have chosen is a Co2FeAl full-Heusler alloy, which is an attractive material for spintronic applications, since it was shown to provide large spin polarization\(^1\) and to possess a low Gilbert damping\(^2\). The Si/SiO\(_2\)/W(t nm)/CFA(0.8 nm)/MgO(1.0 nm)/Ta(2 nm) structures, with t = 3.5, 4.5, 5.5, 6.5 and 7.5 nm, were elaborated using a magnetron sputtering system. After deposition, the samples were patterned for transport measurements, using UV lithography and Ar ion milling in the form of standard Hall bars with longitudinal and lateral dimensions of 100 μm and 30 μm, respectively (figure 1(a)). A second lithographic step was employed in order to allow the deposition of Ta (5 nm)/Cu(100 nm)/Ta(5 nm) contact pads. The magnetotransport measurements were performed using standard DC and AC lock-in techniques.

The crystal structure of the films was investigated using a four circle diffractometer, while the saturation magnetization was determined using a vibrating sample magnetometer.

1. **Spin–orbit torques and magnetization switching in W/Co2FeAl/MgO structures**


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Fig1. (a) Optical microscopy image of the Hall patterned device and the dc experiment configuration. (b) Electrical conductance of the patterned devices versus the thickness of the W layer. The red line is a linear fit of the data. (c) Anomalous Hall measurements for the W(5.5 nm)/CFA(0.8 nm)/MgO(1.0 nm) structure, showing the presence of the Perpendicular Magnetic Anisotropy.

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The SOT fields were quantified using harmonic Hall voltage measurements. A 593 Hz sinusoidal current was passed through the micro-strip and the first ($V_\omega$) and second harmonic ($V_{2\omega}$) transverse voltages were measured using the lock-in amplifier technique. The magnetotransport experiments were performed in two different configurations: the longitudinal one, with the external magnetic field applied along the x-axis (HL); and the transverse one, with the external magnetic field applied along the y-axis (HT). The effective spin Hall angle was calculated using the extracted value for the Slonczewski-like SOT induced effective field and it is depicted in figure 2(a).

\[ \theta_{\text{SH}}^\omega = 0.30 \pm 0.03 \]
\[ \lambda_{ef} = 2.2 \pm 0.3 \text{ nm} \]

**Fig2** (a) Effective spin Hall angle versus the W layer thickness, the red line stands for theoretical fitting. The values of the bulk spin Hall angle and spin diffusion length are given. (b) Switching current density versus the longitudinal bias field for the W (5.5 nm)/CFA (0.8 nm)/MgO (1.0 nm) structure. (c) and (d) Typical current induced magnetization switching loops measured for negative and positive longitudinal bias fields.
In order to demonstrate the potential of this system for spin orbitronic devices with spin-torque driven magnetization manipulation, we have performed current induced magnetization switching experiments. They have been carried out for the sample having a 5.5 nm thick W layer, using a pulsed dc current with a pulse width of 100 μs and a 2 ms interval between pulses. The switching was detected by measuring the Hall voltage for each pulse. In order to ensure a deterministic switching of the magnetization a longitudinal bias field was applied. Figures 2(c) and (d) show typical switching cycles recorded for negative and positive longitudinal bias fields. The switching current density was determined as an average between the positive and negative switching current densities. Each measurement cycle was repeated 10 times and the error bars correspond to the standard deviation from the mean value. The switching current density shows a rapid decrease with the bias field up to a field of about 50 Oe, above which the decrease shows a much lower rate. We experimentally observe that for bias fields above 50 Oe a sharp switching can be obtained for charge current densities of about $1.5 \times 10^6$ A cm$^{-2}$. The low switching current density observed in our samples is a consequence of several factors: the relatively large spin Hall angle of β-W, the relatively low damping and anisotropy field of the Heusler ferromagnet and the relatively large field like torque.

The relative low value of the current density needed for spin orbit induced switching of the perpendicular magnetized Co2FeAl layer makes the β-W/Co2FeAl/MgO system an interesting candidate for spin–orbit torque spintronic devices.

2. Annealing temperature and thickness dependencies of structural and magnetic properties of Co2FeAl thin films

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Materials with high spin polarization, such Heusler alloys, are eligible for spintronic applications. These materials are considered as a key technology to solve some spintronic challenges, especially large magnetoresistance ratios, low critical current densities for spin transfer torque magnetization switching, and the injection and the detection of spin-polarized currents from metallic ferromagnets into semiconductors. Among the full Heusler alloys, Co2FeAl (CFA) is a very attractive material due to its high Curie temperature (TC ≈ 1000 K) and its relatively high spin polarization leading to high tunnel magnetoresistance (TMR) ratios up to 360%, as achieved in CoFe/MgO/Co2FeAl structures$^3$. In addition to its lowest magnetic damping parameter among

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Heusler compounds\textsuperscript{5}, making it suitable for magnonic\textsuperscript{6} devices, the relatively small lattice mismatch between MgO(001) and CFA(001) enables the fabrication of high quality CFA/MgO(001) epitaxial heterostructures with low resistance area product magnetic tunnel junctions (MTJs), which are essential for spin transfer switching. However, in such alloys, there is always some degree of chemical disorder, which strongly influences many of their physical properties. In reality, the totally ordered phase (L2\textsubscript{1}) is difficult to achieve and there are a variety of possible disorder types. Furthermore, we recently showed that CFA thin films (with a thickness down to 10 nm) grown on MgO substrates exhibit a strong negative perpendicular anisotropy (reinforcing the in-plane easy plane). This turns out to be a purely interfacial contribution. Therefore, it is of great interest for both fundamental and technological reasons to investigate the magnetic behavior of the CFA ultrathin films (with a thickness down to 2 nm) grown on MgO and to point out the annealing temperature dependencies of the interfacial anisotropy over a large range of CFA thicknesses, in order to allow for the manufacturing of CFA films with the desired properties. The experimental strategy employed in this paper is a complex correlated structural, static, and dynamic magnetic analysis. Therefore, the x-ray diffraction (XRD), ferromagnetic resonance in microstrip line (MS-FMR) under in-plane applied magnetic field, combined with vibrating sample magnetometry (VSM) allowed us to correlate the structural and magnetic properties of CFA thin films grown on MgO substrates and annealed at different temperatures. Our results demonstrate the presence of in-plane and perpendicular to the plane interface anisotropies, which are \( T_a \) dependent and their signs depend on the CFA thickness, offering the possibility of versatile sample design with skillfully tuned magnetic properties.

In order to determine the crystal structure and chemical order degree of the CFA films, we have performed x-ray diffraction experiments. Figure 1(a) shows typical 2\( \theta \)-\( \omega \) diffraction patterns for the 50 nm film as functions of the annealing temperature. One can observe that, besides the peak corresponding to the MgO substrate, the patterns only show the (002) and (004) CFA peaks. The (002) superlattice peak is characteristic of the B2 phase of CFA and, therefore, the absence of this peak is a signature of the A2 phase in which Fe, Al, and Co randomly occupy the atomic sites. Since the (004) reflection is a fundamental one corresponding to the cubic CFA structure, the ratio \( A(002)/A(004) \) of the integrated intensities of the (002) and of the (004) peaks, which increases with the film thickness and the annealing temperature, represents the measure of the order degree on the Co sites. This ratio, shown in Fig. 1(b) as a function of \( T_a \) for thicker films, increases with \( T_a \) above 300 \( ^\circ \)C, suggesting a monotonous enhancement of the chemical order from the A2 towards the B2 phase as the thickness and \( T_a \) increase. For CFA thicknesses below 10 nm, the films present an A2 phase for all annealing temperatures.

\textsuperscript{6} H. Ulrichs, B. Lenk, and M. M\’unzenberg, Appl. Phys. Lett. 97, 092506 (2010).
Fig. 1a) Typical example of 2θ/ω (out-of-plane) x-ray diffraction patterns for 50 nm thick CFA films annealed at different temperatures. The patterns have been shifted vertically for better visibility. (b) Evolution of the integral intensities of the (002) and (004) CFA peaks [A(002)/A(004)] with respect to the annealing temperature for different film thicknesses $t$.

In order to quantify the perpendicular magnetic anisotropy in our films we have performed ferromagnetic resonance experiments in perpendicular configuration and extracted the effective magnetization $4\pi M_{\text{eff}}$. The extracted effective magnetizations from FMR measurements are shown in Fig. 2 as a function of $1/t$ for the different $T_a$.

Depending on $t$, two different regimes, separated by a critical thickness ($4 \text{ nm} < t_c < 7 \text{ nm}$, depending on $T_a$), can be distinguished. Indeed, for $t > t_c$, $M_{\text{eff}}$ linearly increases with $1/t$ while it linearly decreases with $1/t$ for $t < t_c$. Therefore, a separate interpretation of the magnetic

Fig2. 3D plot of the thickness dependence of the effective magnetization ($4\pi M_{\text{eff}}$)
anisotropy must be made in the regions above and below tc. According to the above structural investigation, $K_{v\perp}$ and $K_{s\perp}$ may include contributions of magnetocrystalline and magnetoelastic origin. In the case of epitaxial growth with a lattice misfit between the constituents, the particular form of strain encountered can contribute not only to $K_{v\perp}$ (coherent growth regime (regime I): $t<t_c$), but also to $K_{s\perp}$ (incoherent growth regime (regime II): $t_c<t$) [17]. Therefore, in order to analysis the results of Fig2 $K_{v\perp}$ and $K_{s\perp}$ are given by:

\[
\begin{align*}
K_{v\perp} &= K_{me\perp} + K'_{me,v\perp} \\
K_{s\perp} &= K_{N\perp} \\
K_{v\perp} &= K_{me\perp} + K''_{me,s\perp} \\
K_{s\perp} &= K_{N\perp} + K_{me,s\perp}
\end{align*}
\]

Where $K_{me\perp}$ is the perpendicular magnetocrystalline anisotropy constant, $K_{me,v\perp}$, $K_{me,s\perp}$ are the perpendicular volume and interface strain anisotropy constants and $K_{N\perp}$ is Néel-type perpendicular interface anisotropy constant induced by the broken symmetry at the interfaces. According to this model, in region I, the influence of misfit strain appears as a volume contribution to the anisotropy, while it leads to an apparent interface contribution in regime II.

The linear fit of the measurements allows determining the perpendicular surface and volume anisotropy constants for both regimes using equations (1) and (2). Their variations as function of $T_a$ are shown in figures 3a and 3b. The volume constant (Fig. 3a), which has a magnetocrystalline origin in regime II according to equation (3), is positive over the whole range of $T_a$. It decreases slightly as $T_a$ increases due to the enhancement of the chemical order with $T_a$ where magnetocrystalline anisotropy would be higher in phase A2 than in phase B2. The minimal value of $K_{v\perp}$ in this regime is obtained around an annealing temperature of 500°C. In regime I, $K_{v\perp}$ is negative, much higher (in absolute value) than that of region II and increases significantly for $T_a>200°C$, due to the magnetoelastic contribution as predicted by equation (1). For the surface anisotropy (Fig. 3b), its absolute value increases with $T_a$ in both regimes. However, compared to $K_{v\perp}$, it behavior versus $T_a$ is different: it becomes positive for $T_a>200°C$ in regime I, where it is pure Néel-type interface anisotropy (see equation (1)), while it is negative over the studied $T_a$ range for regime II, due to the contribution of strain to the surface anisotropy as predicted by equation (2). Therefore, we conclude that the linear thickness dependence of the effective magnetization is mainly governed by pure Néel-type surface interface anisotropy in region I, reinforcing perpendicular easy axis, while it given by the interface strain anisotropy favoring in-plane easy axes in regime II. The origin of this pure surface interface can be attributed to the CFA/MgO interface due to the hybridization of the O 2$p$ and metal-alloy 3$d$ orbitals.
In this paper we have performed an exhaustive study of magnetic anisotropy components of epitaxial CFA films grown on MgO, as a function of the thickness and annealing temperature. We have pointed out two different regimes governed by surface and magnetoelastic strain induced magnetic anisotropies. We have shown that for thicker films, where strains are relaxed, the overall surface anisotropy favors in-plane anisotropy, while for thinner films, where the magnetoelastic strain induced magnetic anisotropies contributes only to the volume component of the anisotropy, the pure surface anisotropy attributed to the CFA/MgO interface favors perpendicular magnetic anisotropy. Another important result of this paper is that it provides a mean of attaining perpendicular magnetic anisotropy in thicker CFA films by realizing a compressive biaxial strain on the CFA film, for example by using a suitable buffer layer of another type of cubic monocristaline substrate with a lower lattice constant that MgO.
Heavy metal/ferromagnet (HM/FM) heterostructures are drawing rapidly increasing interest in scientific community due to their outstanding potential for technological innovations capable of revolutionizing the whole philosophy of magnetic memory and logic devices. However, no technological breakthrough is possible without major developments in fundamental science. Thus the discovery of spin Hall effect (SHE), spin orbit torques [8] and most recently the skyrmion (spin configurations at the nanoscale) and domain wall (DW) chirality inversion in HM/FM bi-layers resulted in a profound re-assessment of the roles played by major physical mechanisms, typically involved in nano-magnetism and spintronics. In particular, much higher velocities of current-driven DW were reported in ultrathin ferromagnetic layers displaying large perpendicular magnetic anisotropy, which is only possible in the presence of spin-orbit torques in metals of large spin-orbit coupling, i.e. heavy metals.

The Dzyaloshinskii-Moriya effect is antisymmetric exchange interaction that appears in ferromagnetic materials in the absence of spatial inversion symmetry. Nowadays, practically all the applications of thin magnetic films, however different, are unthinkable without nano-structuring unavoidably leading to breaking of symmetry. The latter applies directly to HM/FM bi-layers in which case DMI is localized in the immediate proximity of the interface and consequently is referred to as interfacial. It changes the static and dynamic properties of domain walls. It is also involved in creating and stabilizing chiral spin textures such as magnetic skyrmions, generally considered as promising candidates for the role of magnetic bits to carry and store information in the future. In this work, we use Brilouin light Scattering combined with vibrating sample magnetometry to measure the CoFe thickness dependence of DMI constants in Pt/CoFe ultrathin heterostructures. We show that both in-plane and perpendicularly magnetized CoFe films are subjected to DMI. Moreover, the effective constant is thickness and interface dependent, its sign is influenced dependent by on the stacks order.

Two types of structures are investigated one grown on Si and having a (111) texturing for the Pt layer and the other grown on MgO(001) showing epitaxial growth, as indicated by X-ray diffraction experiments (Fig. 1).
Fig. 1. X-ray 2θ-ω (out-of-plane) diffraction patterns for samples grown on (a) Si and (b) MgO; Φ-scan measurements performed at a tilt angle $\Psi = 54.7^\circ$ probing (c) MgO and Pt (111) type reflections and (d) Pt (002) and Si (111) type reflections for samples grown on MgO and Si, respectively. The measurement indicated the epitaxial growth of Pt on MgO with the Pt(001)[100]//MgO(001)[100] epitaxial relation and the (111) out-of-plane texturing growth on Si.

Since the DMI is an interface effect, its strength is expected to be influenced by the different surfaces (111) and (001) with different packings of Pt in contact with the ferromagnetic material. Therefore, for systematic comparison between the two sets of samples, $D_s$ should be determined. The evolution of the obtained values of $D_{eff}$ as function of the effective inverse of the CoFe films nominal thickness are shown in Fig. 2. The effective thickness dependence of the DMI effective constant shows two regimes due to the degradation of the interfaces as the CoFe thickness decreases. By the linear fit of the data of Fig. 2 for $t_{CoFe} \geq 1.2$ nm (red lines) for Pt/CoFe and for $t_{CoFe} \geq 1$ nm (black lines) for the reversed structure, $D_s$ has been found to be -1.27 pJ/m and 1.32 pJ/m for Pt/CoFe and its reversed system, respectively. The above obtained result for Pt/CoFe films allows classifying the investigated bi-layers in terms of their applicability for skyrmion stabilization. According to recent theoretical predictions\textsuperscript{7}, there exist two threshold values distinguishing conventional bubbles from skyrmions, namely $D_{eff} = 1.6$ mJ/m$^2$ for isolated skyrmion and $D_{eff} = 2.1$ mJ/m$^2$ for skyrmion lattice ground state. This means that our samples fall into three major categories. First, the thickest sample with nominal thickness 1.6 nm is not suitable

Fig2. Thickness dependence of the effective DMI constants of Pt/CoFe(t<sub>CoFe</sub>)<sub> ICommand</sub>/MgO grown on Si substrate and MgO/CoFe(t<sub>CoFe</sub>)<sub> ICommand</sub>/Pt grown on MgO substrate. red (blue) and black (olive) fits correspond to t<sub>CoFe</sub>≥1.2 nm (t<sub>CoFe</sub>≤1.2 nm) and t<sub>CoFe</sub>≥1 nm (t<sub>CoFe</sub>≤1 nm) for Pt/CoFe and its reversed structure, respectively.

for skyrmions. On the contrary, the thinnest with nominal thickness 1.0 nm is capable of stabilizing arrays of skyrmions. All the rest are good for individual skyrmions but not for their arrays.

4. Interface Dzyaloshinskii-Moriya interaction in the interlayer antiferromagnetic-exchange coupled Pt/CFB/Ru/CFB systems


The exchange interaction can be direct (involving an overlap of electron wave functions from the neighboring atoms and Coulomb electrostatic interaction) or indirect (little or no direct overlap between neighboring electrons and mediated through intermediary atoms). The direct exchange interaction between electrons may contain symmetric and asymmetric terms. The symmetric term, commonly known as the Heisenberg interaction, usually leads to collinear magnetic structures. The asymmetric exchange, referred to as the Dzyaloshinskii–Moriya interaction<sup>8</sup> (DMI), favors canted neighboring spins leading to various magnetization structures at the nanoscale such as helices and skyrmions. It changes the static and dynamic properties of domain walls and leads to different energy (non-reciprocity) of two spin waves (SW) having the same wavelength and propagating along two opposite directions. DMI can be induced by a lack of inversion symmetry of the compound and a strong spin-orbit coupling. This can be achieved by using heavy metal/ferromagnet (HM/FM) heterostructures, giving rise to interfacial DMI. Indirect exchange interactions such as coupling between two magnetic layers separated by a non-magnetic spacer layer is mediated by conduction electrons of the spacer layer which are scattered successively by the magnetic layers. The coupling, which oscillates in sign as a function of the thickness of the spacer layer was first observed by Grünberg<sup>9</sup> for transition metal systems. It is crucial for many applications in modern magnetic storage devices and spin electronics.

Recently, Chen et al. demonstrates an experimental approach to stabilize a room temperature skyrmion ground state in chiral magnetic films via interlayer exchange coupling (IEC). Moreover, it is utmost important to investigate the spin waves spectrum in the presence of both DMI and IEC. Therefore, both experimental and theoretical investigations of this aspect will be reported in this work. We thus use BLS combined with vibrating sample magnetometry (VSM) to measure the combined effects of the IEC strength and of the DMI constant on SW non-reciprocity in Pt/Co$_{20}$Fe$_{60}$B$_{20}$/Ru/Co$_{20}$Fe$_{60}$B$_{20}$. We show that although the two ferromagnetic (FM) layers are similar with the same thickness, caution should be paid to the interpretation of the SWs non-reciprocity. Indeed, the frequency difference between the two counter propagating SWs, usually attributed to DMI is also IEC strength dependent when the two FM layers present different perpendicular surface anisotropies.

VSM hysteresis loops for a CFB/Ru/CFB trilayer with different Ru layer thicknesses are shown in figure 1. These loops clearly show that for all the samples, the magnetizations of the two CFB layers are antiferromagnetically coupled. Indeed, in zero applied magnetic fields, the magnetizations of successive magnetic layers are antiparallel to each other, resulting in zero remnant magnetization. When an external magnetic field is applied, the Zeeman energy tends to align the magnetizations of both layers in the field direction, so that the magnetizations progressively increase until a saturation field is reached. This saturation field is Ru thickness dependent as shown in figure 1. The typical BLS spectra are displayed in figure 2 for two Ru thicknesses at $k_{sw} = 20.45$ and $8.08 \mu$m$^{-1}$ and for two in-plane applied fields sufficient to saturate the magnetizations. Two main features are noticeable: one line (mode 1) is observable in the S and AS parts of each spectrum; the positions of these lines are not symmetrical. As the structure is made of two coupled FM layers, one expects two magnetic modes (optic and acoustic modes) in Stokes and anti-Stokes parts, as mentioned above. Nevertheless, the intensity of the second line (optic mode) is very weak and thus experimentally unobservable. This is because the CFB layers have the same thickness and not very different perpendicular anisotropy fields.

Fig1: VSM hysteresis loops showing the normalized in-plane magnetization component as function of the in-plane applied magnetic field for Pt/CFB(1.12 nm)/Ru (t$_{Ru}$)/CFB(1.12 nm)/MgO systems of various Ru thicknesses (t$_{Ru}$).

The experimental $k_{sw}$ dependences of $\Delta F$ for various Ru thicknesses as well as those of the Ta(3 nm)/Pt(3 nm)/CFB(1.12 nm)/Ru(0.8 nm)/Ta(3 nm) and Ta(3 nm)/Pt(3 nm)/Ru(0.8 nm)/CFB(1.12 nm)/MgO individual layers are shown in figure 3. Note the negative sign of $\Delta F$, the variation of its slope with the Ru thickness and its small value compared to that of the single CFB layer [Ta(3 nm)/Pt(3 nm)/CFB(1.8 nm)/Ru(0.8 nm)/Ta(3 nm)]. The effective iDMI constants ($D_{eff}$) of the individual layers, deduced from the slope of $k_{sw}$ dependences of $\Delta F$ using the above mentioned magnetization at saturation and the gyromagnetic ratio values, are found to be -0.84 mJ/m$^2$ and -0.3 mJ/m$^2$ for Pt/CFB(1.12 nm)/Ru (0.8 nm) and Pt/Ru(0.8 nm)/CFB(1.12 nm)/MgO, respectively. Furthermore, the experimental observed IEC dependence of $\Delta F$ is thus an indication that the bottom and the upper CFB layers have a different perpendicular anisotropy.

In summary, the spin wave observations by means of Brillouin light scattering reveal a Stokes and anti-Stokes frequency difference that could be related to the interfacial Dzyaloshinskii Moryia interaction. Nevertheless, simulations show that the frequency difference is also influenced by the...
coupling between the ferromagnetic layers when they possess different anisotropies. Magnetization profile calculations allowed for explaining the frequency mismatch by including anisotropy field difference between the top and bottom ferromagnetic layers of the stack. Therefore, the interfacial Dzyaloshinskii Moryia interaction parameter is correctly derived once the different anisotropies are evaluated.

5. Interlayer exchange coupling in perpendicularly magnetized Pt/Co/Ir/Co/Pt structures

Interlayer exchange coupling (IEC) between ferromagnetic films separated by a non-magnetic layer is a quantum interference phenomena with significant implications in the field of spintronic devices and information recording magnetic media. This effect is oscillatory with respect to the thickness of the non-magnetic layer and, thus, can induce ferromagnetic or antiferromagnetic coupling between the magnetic moments of the two ferromagnetic layers. Synthetic antiferromagnets (SAF), based on interlayer antiferromagnetically exchanged coupled ferromagnetic films, have been commonly used in magnetic tunnel junctions, spin valves and magnetic recording media to decrease the stray fields and the net magnetic moment. Recently, perpendicular magnetized SAF structures have attracted significant research interest. They are used to provide higher densities, prevent the read/write disturbances caused by thermal fluctuation or stray fields in spin transfer torque magnetic tunnel junctions and to improve the speed of spin torque driven domain wall motion in racetrack nanowires\textsuperscript{11}. The typical non-magnetic layer used in perpendicularly magnetized SAF structures is Ru. Recently\textsuperscript{12}, it was shown that Ir can provide even larger IEC in structures employing artificial Co/Pt superlattices consisting of Co and Pt monoatomic layer stacking. Therefore, it is of interest to study the IEC in structures employing perpendicularly magnetized Co films separated by Ir interlayers. In this paper, we thus deposited perpendicularly magnetized Pt/Co/Ir/Co/Pt thin films structures. We first analyze the magnetic anisotropy properties of the individual Co layers and we point out the important effect of lattice strains on the perpendicular magnetic anisotropy (PMA). Following, we investigate the interlayer exchange coupling as a function of the Ir layer thickness and we study its annealing temperature stability.

Figure 1 shows representative hysteresis loops measured for the Pt 3 nm/Co1 0.9 – 1.8 nm/Ir 1.6 nm/Pt 3 nm and the Pt 3 nm/Ir 1.6 nm/ Co2 0.9 – 1.8 nm/Pt 3 nm structures, with the


magnetic field applied perpendicular or parallel with the films surface. Irrespective of the nature of the underlayer, the 0.9 nm thick Co films show square shaped out-of-plane hysteresis loops with

**FIG. 1.** In-plane (a-d) and out-of-plane (e-h) hysteresis loops measured for the Pt 3 nm/Co1 0.9 – 1.8 nm/Ir 1.6 nm/Pt 3 nm and the Pt 3 nm/Ir 1.6 nm/Co2 0.9 – 1.8 nm/Pt 3 nm structures.
full remanence, indicating the presence of PMA. The in-plane hysteresis loops have a behavior typical for a hard axis of magnetization, showing a continuous rotation of the magnetization up to saturation. In the case of the Co1 layer the saturation field is around 6500 Oe, while for the Co2 layer shows a slight decrease down to around 6100 Oe. In the case of the structures with 1.8 nm thick Co layers, the easy magnetization axis turns in plane and the out-of-plane hysteresis loop shows typical behaviors for hard axis of magnetization [Fig. 1(g-h)]. The saturation field for the Co1 layer is around 3800 Oe, while for the Co2 layer is increasing up to 5100 Oe. All these features indicate that, although both Co1 and Co2 show PMA, for the same effective Co thickness the PMA is stronger for the structures with Pt underlayers, as compared with the ones having Ir underlayers.

In order to study the IEC we have grown a series of samples with fixed Co layers and variable Ir layer thicknesses. The thicknesses of the Co layers were chosen so they would provide similar perpendicular magnetic anisotropy fields (around 7 kOe). Depending on the thickness of the Ir layer the IEC was found to be oscillatory promoting ferromagnetic (FM) or antiferromagnetic (AF) alignment of the magnetizations of the two ferromagnetic layers. Furthermore, the amplitude of the IEC increases with decreasing Ir layer thickness. Depending on the strength of the AF-IEC two types of behaviors can be observed. Figure 2(a) shows a perpendicular to the plane hysteresis loop measured for the Pt 3nm/Co 0.9 nm/Ir 1.35nm/Co 0.7nm/Pt 3nm sample, having an exchange field lower than the perpendicular anisotropy field. At relatively large applied magnetic fields the Zeeman energy dominates and the magnetizations of both layers are saturated and aligned parallel with the field. By decreasing the field, due to AF-IEC, the magnetization of one layer switches abruptly, through nucleation and propagation, and the magnetizations become antiparallel aligned. If the field is further increased in negative direction the Zeeman energy dominates again and

**FIG. 2.** Perpendicular applied field VSM loops measured for the (a) Pt 3nm/Co 0.9 nm/Ir 1.35nm/Co 0.7nm/Pt 3nm and (b) Pt 3nm/Co 0.9 nm/Ir 0.54nm/Co 0.7nm/Pt 3nm samples, respectively. The red and blue arrows schematically depict the relative orientations of the magnetizations of the Co layers in different field regions. The exchange ($H_{ex}$) is also indicated.
another switching event takes place and both magnetizations become parallel with the applied field. Figure 2(b) shows a perpendicular to the plane hysteresis loops recorded for the Pt 3nm/Co 0.9 nm/Ir 0.54 nm/Co 0.7 nm/Pt 3nm sample, having an exchange field larger than the perpendicular anisotropy field. At relative large applied magnetic fields the magnetization of both Co layers are saturated and aligned parallel with the field, thus minimizing the Zeeman energy. By decreasing the field, due to the strong AF-IEC, the magnetization of both layers start to rotate in opposite directions away from the perpendicular magnetic anisotropy easy axis. The magnetizations rotation was confirmed by numerical simulations within a Stoner-Wohlfarth model (not shown here). By further decreasing the field, both layers magnetizations undergo a nucleation propagation event and become antiparallel and aligned with the perpendicular magnetic anisotropy easy axis, thus minimizing both AF-IEC and anisotropy energy at the expense of Zeeman contribution. This behavior is also replicated in the negative magnetic field part of the loop.

In order to quantify the coupling strength we have defined the exchange constant as $J = -H_{ex}M_s t$, where $M_s$ is the saturation magnetization, $t$ is the thickness of the Co layers and $H_{ex}$ is the exchange field described in Fig. 2. The calculated coupling constant together with the $H_{ex}$ are shown in Fig. 3 as a function of Ir layer thickness. The first maximum of AF coupling was obtained for a Ir layer thickness $t_{Ir}$ of 0.45 nm, with $J = -2.5\pm0.3$ erg/cm² (equivalent to a $H_{ex}$ above 12 kOe). This value for the coupling strength is similar to the values obtained for in-plane magnetized Co films separated by Ir interlayers or for CoPt/Ir/CoPt superlattice structures. It should be mentioned that a relative large exchange coupling is maintained in a relative large Ir layer thickness window around the first AF-IEC maximum, which is interesting from an application point of view. For $t_{Ir}$ between than 0.7 nm and 1.1 nm the coupling becomes FM. The second maximum of AF coupling was obtained for a Ir layer thickness of 1.25 nm, with $J = -0.12\pm0.03$ erg/cm². In order to quantify the oscillations period of the coupling strength we have

**FIG. 3.** The interlayer exchange coupling constant and the exchange field as a function of the Ir layer thickness. The red line is a fit of the experimental data using the relation given in the text.
fitted the data using the relation $J \propto \sin(\phi + 2\pi t_\parallel / \lambda) / t_\parallel^2$, which gives an oscillation period $\lambda = 3.7$ monolayers, in agreement with the value reported for in-plane magnetized structures.

In summary, we have studied the interlayer exchange coupling in Pt/Co/Ir/Co/Pt perpendicularly magnetized structures. We first analyzed the magnetic anisotropy properties of the individual Co layers and we pointed out the important effect of lattice strains on the perpendicular magnetic anisotropy. We showed that the surface anisotropy depends on the positioning of the Co layers within the stack, being around 1.76 erg/cm² for the lower one and around 1.5 erg/cm² for the upper one, respectively. Following, we investigated the interlayer exchange coupling as a function of the Ir layer thickness and we studied its annealing temperature stability. We demonstrate an exceptionally high interlayer exchange constant of around -2.5 erg/cm² for a Ir thickness of 0.45 nm which corresponds to a an exchange field larger than 12 kOe. Depending on the Ir layer thickness the stability of the AF-IEC is preserved for annealing temperatures up to 400 °C.

Results that are in the publication process

- Using as a benchmark the results obtained for the CoFe films interfaced with Pt(111) or Pt(001), we have extended our studies and performed similar studies on Co₂FeAl Heusler alloy thin films interfaced with Pt (111) and Ir (111). The Heusler alloy thin films are in-plane or perpendicular-to-plane magnetized.

- In order to understand the effect of the Heusler alloy composition on the perpendicular magnetic anisotropy we have grown on a series of samples Pt/Co₂(FeₓMnₓ)(Al₁₋₅Si₅)/MgO samples. The magnetic anisotropy of the films was precisely accessed using the ferromagnetic resonance technique. A typical example of ferromagnetic resonance experiments measured at different field angles is shown bellow
Using the values of the resonance field one can easily have access to the effective magnetization of the films, which contains information on the magnetic anisotropies of the films. Since the data are not yet published, we just show bellow an example of the effective magnetization versus the thickness for two different composition of Heusler alloys showing two different regions: one favoring perpendicular magnetic anisotropy (lower thickness) and the other favoring in-plane magnetic anisotropy.

*Effective perpendicular anisotropy versus effective thickness for Heusler alloy thin films with two different compositions: Co$_2$FeAl and Co$_2$FeAl$_{0.5}$Si$_{0.5}$.*
In order to understand the effect of the Heusler alloy composition on the half metallic properties we have employed \textit{ab-initio} band structure calculations using the Wien2k code\textsuperscript{13}. Below we show the minority bands structure calculated for different compositions.

It can be observed that by changing the composition of the alloy from Co\textsubscript{2}FeSi to Co\textsubscript{2}FeAl we have a movement of the Fermi level inside the minority gap from the bottom of the conduction band to the top of the valence band. Essentially the bands have the same structure except for the shift in energy. In the case of the alloy with the middle composition Co\textsubscript{2}FeAl\textsubscript{0.5}Si\textsubscript{0.5}, the Fermi level sits in the middle of the minority gap, giving the optimum half metallic properties. However, the band structure becomes more complicated due to the

\textsuperscript{13} P. Blaha, K. Schwarz, G. K. H. Madsen, D. Kvasnicka and J. Luitz, Wien2k. An augmented plane wave local orbitals program for calculating crystal properties (Technical University of Wien, Austria, 2001.)
decrease in the symmetry of the elementary cell of the compound. We were also interested in the effect of changing the 3d element will have on the band structure. Therefore, we have also calculated the band structure for the Co$_2$MnSi. In the previous case, when we change the $sp$ element, the effect was just a virtual movement of the Fermi level however, in this case we have an important modification of the band structure, especially along the $\Gamma\Delta X$ direction. The bands in the previous mentioned direction move up in energy opening a clear half-metallic gap. In this sense, Co$_2$MnSi given that it can provide perpendicular magnetic anisotropy seems to be a better choice than the more conventional Co$_2$FeSi, in terms of half metallic stability. Again, the middle composition Co$_2$Fe$_{0.5}$Mn$_{0.5}$Si compound retains band structure characteristics of the parent compositions compounds with the Fermi level situated close to the middle of the minority gap.

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