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PhD Thesis Summary

Magnetization manipulation in the VCMA framework: a multiscale study, from fundamental material properties to magnetization dynamics

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1. Introduction

In an era becoming more and more information technology reliant, the computers are of crucial importance to our society and with the ascending trend of Internet of Things (IoT), they will be even more present in our daily life. Nonetheless, this started to draw the attention on the power consumption, as well. It is estimated that 1% of the world energy consumption is destinated to computers, data centers and network infrastructure [1] and from this quantity, 30% is consumed by computer terminals alone [2]. Even more interesting is the fact that 40% of a computer's power consumption is attributed to the memory solely, mainly because of its volatile character. Therefore, as our today's world is tormented by pollution effects and energy crises, reducing the energy consumption at least in areas where it can be saved is of crucial importance. From the memories' standpoint, the practical approach was to gradually replace the energy ravening volatile memories with non-volatile alternatives. However, this was a considerable challenge from two points of view. The first one would be that the replacement has a limitation: the non-volatile memories cannot fully replace the volatile ones in the current von-Neumann architectures, with the actual operating systems, but only a fraction, until the inferior levels of cache [3]. However, this issue along with other challenges, like speed and efficiency, led to blooming research in the optimization of non-von-Neumann architectures, such as the in- memory computing paradigm, which although it is not a completely new idea, it appears to more feasible than ever. The second one would be related to performances, but in this field, it seems that the magnetoresistive memories still struggle to keep up with the performances of the SRAM memories in terms of speed and with those of DRAM in terms of cell density [5]. A comparison in performances is revealed in the Figure 1.1.

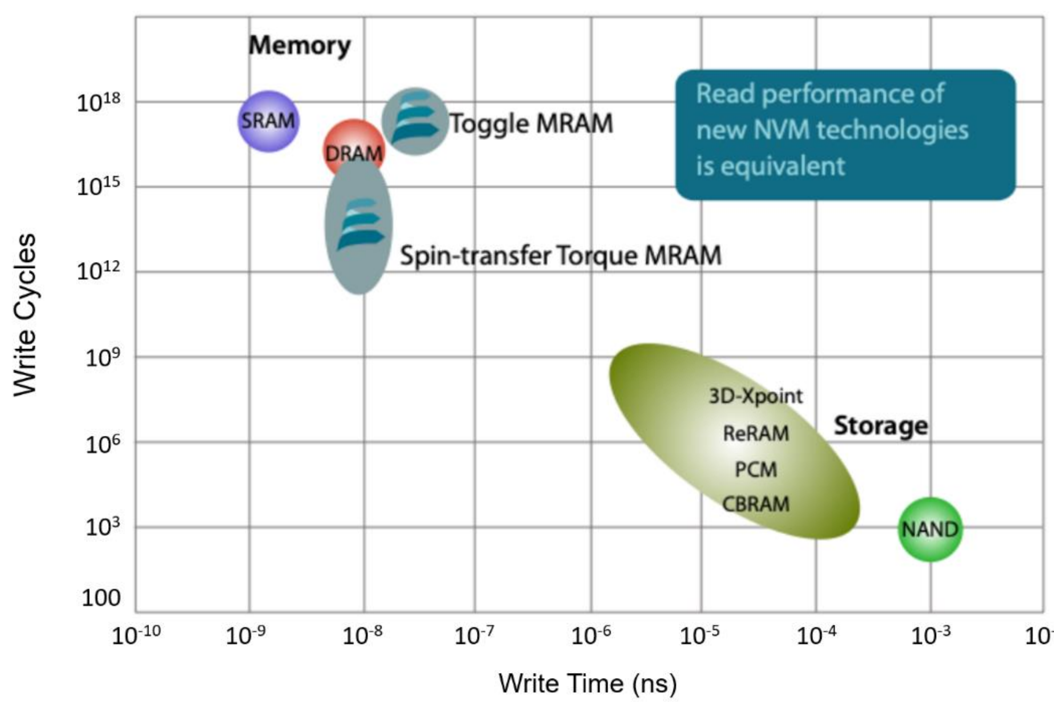

Figure 1.1: MRAM performances in relation to other memories (from Everspin recent data in [4])

1.1 Aim of the thesis

The electric field control of magnetization, which is synonym with voltage controlled magnetic anisotropy (VCMA) is the foundation of VC-MRAM (voltage-controlled MRAM) and the object of this thesis. Its main assumption is that in a magnetic system, with two energy minima, corresponding to two opposite magnetization orientations, the magnetization can be switched if the anisotropy energy barrier is reduced by an electric field acting at the ferromagnet/insulator interface. During this lowering of the barrier, any external stimulus (thermal, magnetic field, Rashba magnetic field) can trigger the switching by lowering one of the energy minima with respect to the other one.

Why does this method of magnetization switching deserve so much attention? It is because it comes with a set of advantages superior to any other type of switching method and that it could soon promote the MRAM technology in areas that are still dominated by SRAM. We will punctually enumerate these features:

• It is the most energy efficient magnetization switching technique known so far. For MRAM applications, it already demonstrated a write energy of two orders of magnitude under the state-of-the art STT-MRAM [6];

• It allows super-fast switching, either by itself (simulated) [7] or by assisting the SOT-MRAM (proved experimentally) [8];

• It is considered to be easily scalable, which is promising in terms of information storage density.

Its future applications would not be limited whatsoever to the basic information storage. It is a great candidate for spin-logic gates and also for the recently demonstrated in-memory computing [9]. However, as this is not a mature technology, it still requires in-depth knowledge to be added to it. Several open questions remained to be answered:

? What is the origin of this mechanism? (several hypotheses explain it, but they do not universally apply, regardless the system)

? What are the critical conditions to achieve a switching with a minimum energy consumption? (it is more energy efficient to cancel just a little from the anisotropy energy barrier)

? Which materials would be suitable in this type of applications?

Along this work, we will attempt to provide each question with an answer.

1.2 Brief presentation of the thesis content

This work aims to answer the questions earlier exposed. To the first two, regarding the origins of the electric field effect on the magnetization and the minimum energy conditions required for a successful switching, we will answer punctually, dedicating them two chapters. However, for the third, although we also have an experimental approach, we must rely our answer on the existing literature, as well.

The structure of this thesis comprises eight chapters, out of which, the first one has the role of an overture, by briefly introducing the reader into the field's history and outcomes.

It presents the general applications of spintronic devices and the philosophy in which they have been conceived, until the latest breakthroughs. The practical applications are revealed as well, with relevance for day-to-day technology and not only.

If the first chapter was just a story level presentation of the VCMA and spintronics, the second chapter is bolder in terms of physical descriptions and explanations. The purpose of this chapter is to familiarize the reader with the magnetization manipulation techniques, not only to a declarative level, but with actual mathematical descriptions and to emphasize the necessity and the magnitude of the impact that VCMA has in this given context. The accent falls on the state of the art regarding the electric field manipulation of magnetization, from the first attempts with ferromagnetic semiconductors, to metallic ferromagnets. Within this framework, we present how our multiscale study is integrating in the overall view of this domain and, along with the research methodology, objectives, and results.

The third chapter is dedicated to the methodology. Although this work was intended to be purely experimental at the beginning, in the context of the pandemic and of other challenges independent of our will, the thesis trajectory suffered some readjustments. Therefore, along with the experimental methods, we will describe theoretical tools and methods, ranging from analytical quantum and ab initio calculations, to atomistic and micromagnetic simulations.

As we mentioned before, this thesis is in fact a multiscale approach of the VCMA phenomenon. In order to use such a backbone for our study, our approach started from an analytical model of one spin precessing under the influence of an electric field and only after this model provided explanations regarding the phenomenology, we incremented the general picture to superstructures, via ab initio, or to large real systems, via atomistic and micromagnetic simulations. Therefore, the fourth chapter is a description of the analytic model of electric field induced magnetization switching. It presents in detail a quantum mechanics approach for perpendicular magnetic anisotropy and VCMA. We start from a simple Stoner Hamiltonian to illustrate the static and dynamic behavior of a spin in an external magnetic field. Then, in order to describe the effect of an electric field, we introduce an additional corresponding Rashba term, determine the stationary eigenvalues and eigenstates and then, describe the time evolution characteristics, mainly the spin precession within the Stoner-Rashba mixed Hamiltonian situation. Last but not least, the analysis of the eigenvalues solution in this case illustrates the contributions to the total energy corresponding to the magnetic and electric field, allowing us to identify the electric field contributions to the anisotropy and the possibility to control it by varying the electric field. Our simple analytical quantum analysis developed in chapter four allows us to identify the Rashba mechanism responsible on the VCMA effects, predicts the processional capability to manipulate the spin by electric field pulses, aspects that will be developed in more detail in the chapters based on higher level approaches: ab initio and micromagnetic modeling.

The fifth chapter illustrates the results of some ab initio calculations performed within a fully relativistic scheme implemented in a framework based on the full-potential linearized augmented plane-wave + local orbitals method (LAPW+lo). Beyond the spin-orbit contribution to the Hamiltonian, allowing to describe the magnetic anisotropy, in our calculations we included the effect of an external electric field via a zig-zag shape potential. Based on thoroughly designed supercell models, we succeeded to calculate the magnetic anisotropy and its variation with an external biasing electric field in various magnetic heterostructures, e.g. M/Fe(100)MgO, M=V, Au, Ag, Pd, Pt, Cr; Pt(111)/Co/MgO. Then, we explained the physical origin of the PMA and the VCMA effect based on the Rashba spin-orbit splitting, and illustrate their dependence on the stacking sequence and layer composition. Furthermore, we theoretically illustrate the possibility to control the perpendicular magnetic anisotropy by interfacial insertion of impurities, demonstrate quantum well related oscillations of anisotropy with the thickness of the magnetic layer.

The sixth chapter is dedicated to magnetization dynamics. This chapter comprises a macrospin approach of the voltage-controlled magnetization switching, followed by a micromagnetic proof, demonstrating that the findings can be extended to real-life sizes and systems. This is a core chapter regarding our results, as it provides a clear insight to the minimum energy conditions for a switching.

The seventh chapter covers all our experimental struggles and results, ranging from sample preparation descriptions, lithography, magnetic and transport measurements. The experimental and the theoretical results are discussed in parallel at a qualitative level in this chapter, as well.

Last but not least, chapter eight is dedicated to general conclusions and perspectives. It is a summarization of all our efforts and results and an assessment of how our initial questions have been answered. The perspectives are also enumerated in this last chapter with the opening of new horizons.

2. State of the art and problematic in the field

In this chapter we attempt to emphasize the subject of this thesis in the magnetic memories' application landscape.

2.1 NV-MRAM: limitations of current magnetization manipulation techniques

The aim of the microelectronic industry is to continuously downscale the memories physical size, while increasing their density and improving efficiency. The transition from the hard disk storage technology to the MTJ based memories meant a significant technologic leap in data storage, since the access to the stored data does not imply a mechanical component anymore and thus the access can be faster. The broadening road to miniaturization and embedded applications almost made forgotten the era of hard disk drives. The next leap is already in progress, since the advances in computer science are both memory and power hungry. We now have on the market magnetic memories based on MTJs, with promising performances in terms of endurance and energy consumption for read/write cycles, such as toggle and STT-MRAM, but for the MRAM to reach the status of universal memory, the range of needed improvements spans from the MTJ intrinsic characteristics, to magnetization manipulation techniques and memory architecture. Such an improved memory requires a balanced solution that simultaneously satisfies two requirements: the PMA should be large enough, so that the thermal stability would be retained, but small enough so that the magnetization switching would not require a significant energy consumption [10]. The most recent family of non-volatile magnetic random-access memories (NV-MRAM) is represented by the Spin Transfer-Torque (STT) MRAMs. Despite the optimism that met the STT-MRAM, the non-volatile magnetoresistive memories are a technology that needs to mature even more. The challenges faced by the STT-MRAM include: writing efficiency, read margin, and even reliability issues, along with some process control deficiencies (due to narrow pitches, with variable distribution uniformity) [11]. Aside from this set of issues, it remains the energy efficiency problem. The STT-MRAM typically requires several hundreds of fJ per bit write event, although recently the energy consumption descended to 45 fJ/bit [12]. Other magnetization manipulation techniques could tremendously reduce this energy waste, to only a few fJ/bit like the VCMA does [6][13].

2.2 VCMA framework

The philosophy of electric field switching of magnetization relies on a series of intricated phenomena that occur at the interfaces. We could say that it might be attributed to a set of collective mechanisms and not just to a single one, since the evidences do not exclude each other in the majority of cases. Beyond the origins of VCMA, an important objective is to maximize the yield of this phenomenon in applications. It is of great interest to find ways of manipulating the magnetic anisotropy energy, so that we could decrease the energy barrier between two magnetization orientations and trigger a switching event. To do so, we should maximize the parameter known as the VCMA coefficient, known as β in the first theoretical studies, or ξ, as it is currently widely known. This parameter reflects nothing else, but the modulation rate of the magnetic anisotropy by an electric field, and it is measured in fJ/Vm. The typical values for this parameter range between tens of fJ/Vm and hundreds of fJ/Vm, with the notable observation that the experimental values are typically larger than the theoretical ones, which theory has failed until now to explain. It is believed that since the VCMA effect is the result of collective mechanisms, a good agreement between theory and experiment cannot be obtained. From a strictly dynamic viewpoint, switching the magnetization with a voltage pulse is conditioned by two elements: an electric field must produce notable effects on the surface anisotropy, i.e., a significant decrease, and a secondary stimulus generating a torque, must topple the magnetization in the opposite direction. To fulfill these conditions, several experimental setups have been tested, such as associations between the VCMA effect and STT or SOT (spin-orbit torque).

We described in general the state of the art and attempted a chronological description of the progress in the field of the voltage controlled magnetic anisotropy. We provided the details related to this field, a justification for the strive of achieving a precise control of electric field switching, but we did not provide information related to our pursuit. To provide more context about our quest, we will describe which question we tried to answer. Since the origin of the voltage controlled magnetic anisotropy is not attributed to a single phenomenon and it is often under discussion, our first objective was to understand the underlying phenomena. Starting from simple systems, several key elements cannot be explained simply by interfacial charge doping effects. Let us take for instance a HM/Fe/MgO heterostructure, which is subjected to an electric field. Depending on the
heavy metal layer (HM), the sign of the VCMA coefficient and the PMA vary dramatically. Such findings do not have an explanation based on the surface charge doping approach. We coupled analytical and ab initio calculations in order to understand ourselves what is the origin of these experimental discrepancies.

Moving upwards, to a "bigger universe" of spins, the collective reaction of the spin magnetic moments as a response to an electric field, materializes itself into magnetization switching. Achieving control over this dynamic process, means to harness the parameters to which the magnetization is susceptible: length of stimuli action and effective fields. We did not find in the whole literature a study on magnetization dynamics, that portrays the switching probability in the least energy consumption paradigm. Therefore, our contribution to the field materialized itself into a description of the critical conditions (with the minimum energy consumption) in which the voltage-controlled magnetization switching takes place. Not only

we focused on the critical voltage, but we were interested about the minimum required in-plane field with which the switching can be achieved, and along the way we discovered the importance of a large damping constant, which is a change of
paradigm, compared to the STT switching.

3. Thesis results in relation to the advances in the field

3.1 Analytic approach

Since the most fundamental phenomena occur at spin magnetic moment level, we decided that this should be our starting point. Just like Barnes et al. [14], in Chapter 4 we started from the simplest building blocks. We analyzed Hamiltonians and deduced the eigenvalues and the eigenvectors for different situations: when a spin is subjected to a molecular field (Stoner Hamiltonian), then we briefly talked about the spin precession due to an electric field (Rashba Hamiltonian). However, the emphasis was put on the combined conditions, as we described the situation in which a spin subjected to a molecular field is simultaneously affected by an electric field (mixed Stoner-Rashba Hamiltonian).

$$\hat{H} = \frac{\hbar^2 k^2}{2m} - J_0 \vec{m}\hat{\sigma} + \alpha_R(\sigma_x k_y - \sigma_y k_x)$$

All these steps aimed to describe analytically the phenomenology at the most basic level and the consequences reflected in the dynamics. We show in detail how the Rashba mechanism contributes both the PMA and to the electric field modulation of anisotropy through a term dependent on E^2.

3.2 Ab initio investigation

In the fifth chapter, we explored the effects of the electric field on the magnetic anisotropy through ab initio methods, within a scale of tens of atoms. We studied how the PMA, generated in some systems, responds in completely opposite manners to the electric field, based on the stack componence.
These observations are reflected in the sign of the VCMA coefficient β, describing the anisotropy reaction to the electric field. This coefficient is defined as the ratio between the anisotropy energy variation and the electric field:

$$\beta = \frac{\Delta MAE}{\Delta E}$$

and its typical values range from a few fJ/Vm to tens, or even hundreds of fJ/Vm. To accomplish these investigations, we used the Wien2k software package, in which an electric field is applied across a supercell as a zig-zag potential gradient. We studied HM/FM/MgO stacks, where HM is a heavy metal or a 3*d* metal, FM is substituted either by Fe or Co, covered by an insulating layer of MgO. Chapter 5 from the thesis discusses in a broad manner how the heavy metal underlayer influences the sign of the β coefficient. We saw that although all the investigated systems share similar FM/MgO top interfaces, the HM/FM interface dictates the VCMA behavior and the PMA values. The table below (Table 3.1) provides a brief description of our results.

Table 3.1 Different HM/FM/MgO systems, investigated by ab initio methods

System	E_{ani} (meV)	E_{ani} (erg/cm^2=mJ/m^2)	β	$a = b$	crystal
V3Fe5MgO	0.54	0.94	68.44	3.02996	cubic(100)
Au3Fe5MgO	0.71	1.37	-32.26	2.883162	cubic(100)
Au3Fe5Pt1MgO	5.86	11.28	-56.65	2.883162	cubic(100)
Pd3Fe5MgO	0.43	0.83	17.3	2.883162	cubic(100)
Pd3Fe5Pt1MgO	5.13	9.87	87.46	2.883162	cubic(100)
Ag3Fe5MgO	1.35	2.59	-	2.888263	cubic(100)
MgO-Fe3-MgO	1.91	3.71	-	2.8689	cubic(100)
Pt3Co5MgO-ot-aPt	0.23	0.58	-43.43	2.7709	hexa(111)

More than that, we observed that the insertion of a fine HM layer at the FM/MgO interface enhances the PMA by almost one order of magnitude.

3.3 Magnetization dynamics

The sixth chapter of the thesis is dedicated to thorough study of the magnetization dynamics in the VCMA framework. We used the macrospin framework to show what the VCMA dynamics looks like, for synchronized voltage and magnetic field pulses, at a nanosecond time scale. We learned that the dynamics has periodic features, but we identified a condition where the pulse length is no longer important, as the switching appears to be pulse length independent. When this condition is fulfilled, it leads to the generation of horizontal "critical" bands in the switching diagram, and the Gilbert damping constant improves the switching time is these bands. On this occasion, we emphasize the importance of the damping in the VCMA dynamics. If in STT switching large damping values are avoided in order to limit the critical current densities, here the damping ensures brings a beneficial effect. We show that it is not necessary to completely cancel the anisotropy barrier to achieve a successful switching event, but only a fraction of it. In this context, the conditions for the critical anisotropy modulation were determined. Within the same chapter, we also determined the conditions for the minimum energy consumption that should be targeted and optimized at device specific level in applications.

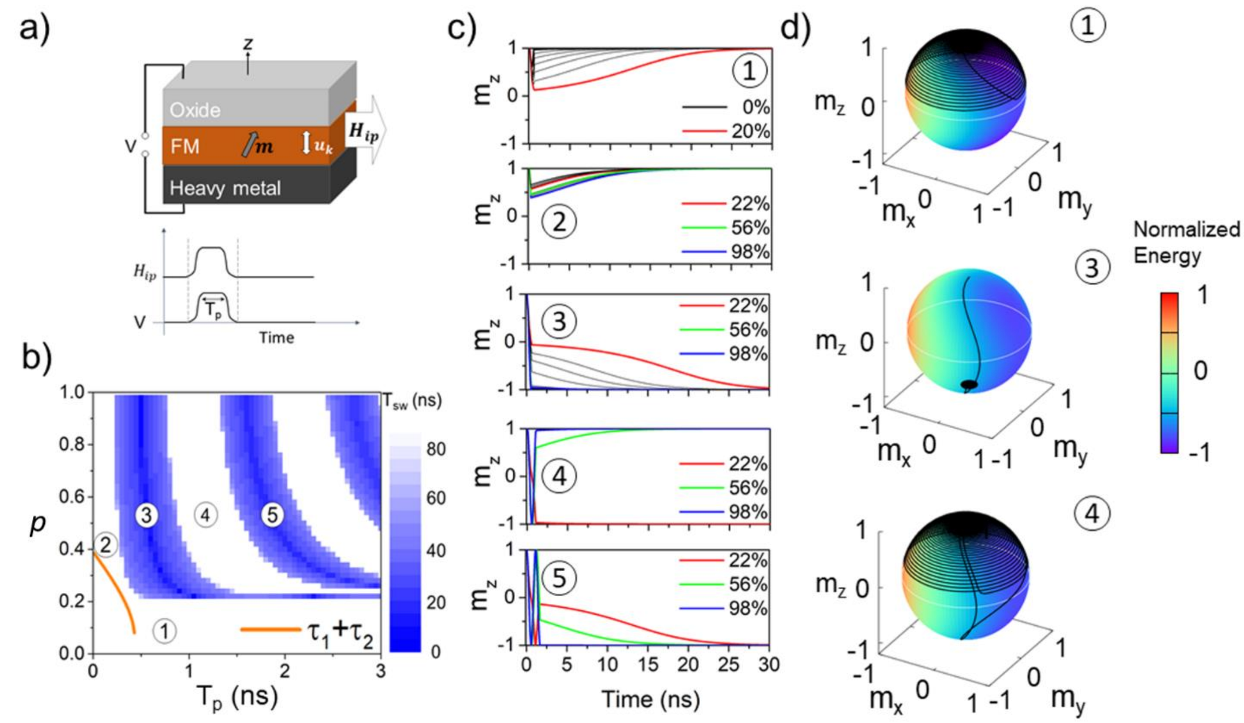

Figure 3.1 (a) Sketch illustrating the simulated system, with the anisotropy field oriented along the uk direction. The electric field is applied simultaneously with the in-plane magnetic field, for a Tp pulse duration. (b) Magnetization reversal diagram, for Hip = 0.0324 T and α = 0.01. The switching probability is correlated with the switching time, Tsw. Additionally, in orange is represented the xOy plane crossing for a minimum pulse length (τ1 + τ2) from the analytical estimation. (c) The Oz projection variation with time for several zones of the switching diagram. (c) The Oz magnetization projection variation with time for several zones of the switching diagram. (d) Magnetization dynamics on the normalized energy sphere, for various switching attempts, representative for some areas in the switching diagram. Reproduced from [7].

3.4 Experimental investigations

Since the chapter dedicated to ab-initio investigations revealed that some systems exhibit PMA and how the PMA can be enhanced by a fine heavy metal insertion at the FM/MgO interface, we decided to explore experimentally a system that could probe all these observations. We chose the Pt/Co/MgO system, with different variations, as presented in Table 3.2.

Table 3.2 Sample composition

Sample type	Composition and remarks
Reference sample (R)	**Si/SiO2// Ta(3nm)/Pt(4nm)/Co(1nm)/Pt(4nm)** Symmetric top and bottom Co/Pt interfaces Sample with PMA, but no VCMA effect expected.
Standard sample (S)	**Si/SiO2// Ta(3nm)/Pt(4nm)/Co(1nm)/MgO(1nm)/Ta(2nm)** Sample with PMA, but the Co/MgO interface is expected to be strongly influenced by an applied electric field, due to its inherent dipole.
Sample with tailored top interface (T)	**Si/SiO2// Ta(3nm)/Pt(4nm)/Co(1nm)/Pt(1nm)/MgO(1nm)/Ta(2nm)** Sample expected to manifest a PMA enhancement and an improved response to the electric-field application.

These stacks were then subjected to several optical lithography steps, so that they would be prepared for Anomalous Hall Effect magnetometry. The Hall devices are represented Figure 3.2.

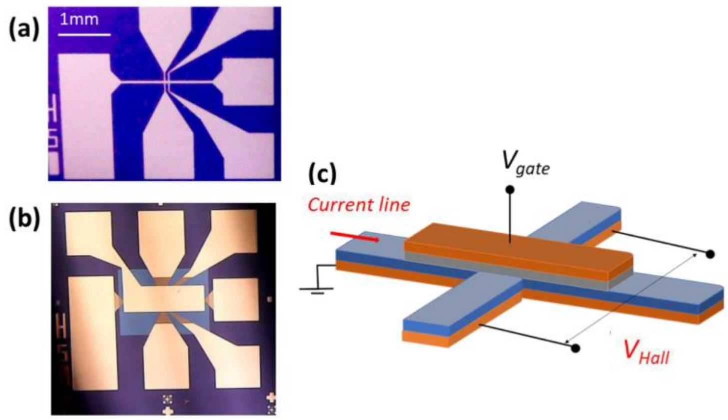

Figure 3.2 AHE setup for measurements. Reproduced from [15] ©2021 IEEE.

For the reference sample (R) no notable effect was observed for an electric field application, since no electric field penetrates across the structure.
However, for the test sample (T), two major observations have been made: a coercive field variation occurs as a response to the applied electric field, and the
Hall voltage decreases with the increase of the applied electric field. We discuss the potential reasons of these observations in depth in Chapter 7 of the thesis, and provide the proof of these observation in Figure 3.3.
To briefly summarize our results, first, we confirmed experimentally the existence of PMA in the samples, as predicted by the ab initio calculations. Second, we observed an asymmetric variation of the anisotropy with the electric field, which also validates the theoretical prediction. However, the main discrepancy is related to the VCMA parameter, β, which in the theoretical estimations is much larger than the obtained experimental values. Then, the variation of Ks in relation to the electric field does not respect the theoretically predicted curve.

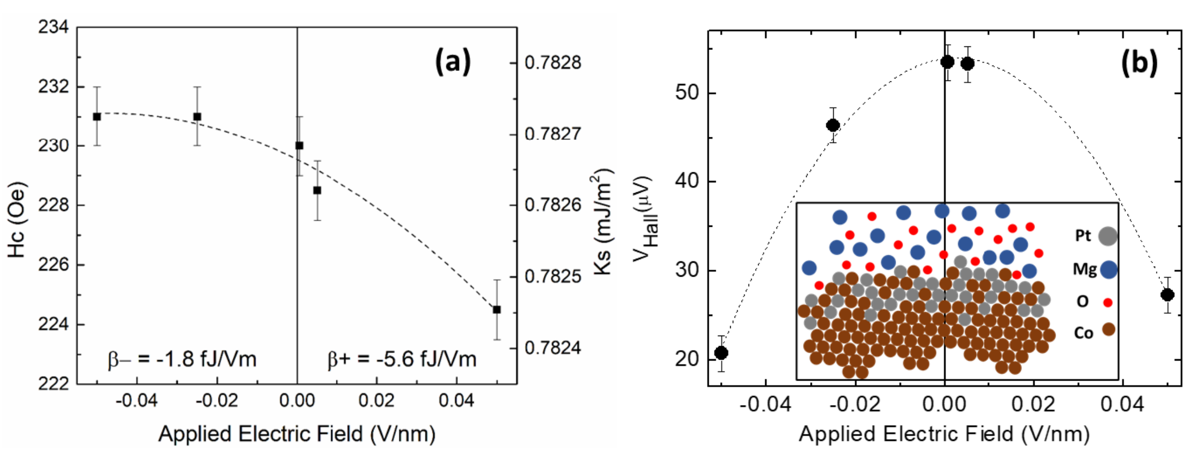

Figure 3.3 (a) The coercive field variation with the electric field; (b) The top interface of sample (T). Reproduced from [15] ©2021 IEEE.

To address the discrepancies, one must be aware that they stem both from theoretical and experimental reasons, namely from methods, models, and technical limitations. For instance, in the ab initio calculations there are different computational methods by which the electric field can be applied across a supercell. While Wien2k employs a zig-zag potential, VASP uses localized charges place in the vacuum region and therefore the ab initio calculations performed on the same structure will generate different results. Another issue is represented by the fidelity of the model, since the superstructures might not accurately replicate the real experimental systems. This is not due to a lack of understanding of the experimental systems, but more because of the theoretical tools limitations, which cannot take into account the micromagnetic aspects, defects, etc. Often, we need to combine theoretical tools and frameworks and study a system at different scales in order to be able to characterize it properly. In consequence, it is only at a qualitative level where we can make comparisons between theoretical and experimental results.

4. Conclusions and perspectives

4.1 General conclusions

At the beginning of this thesis, we started with a set of challenging issues, formulated a list of initial questions and established and developed a quite complex multiscale approach to tackle them. Arriving to the end of the dedicated time for this thesis project, we can make a critical assessment of the results. Certainly, some major answers have been brought to our initial questions. We successfully answered to important questions related to the complex physical mechanisms governing the origin of the perpendicular magnetic anisotropy in ultrathin magnetic heterostructures. Using simple analytical quantum models, more sophisticated ab initio techniques and micromagnetic models, we successfully modelled the effect of the electric field on the PMA in systems with modulated architecture of the stacking sequence and interfaces. We successfully identified the critical issues governing the precessional switch of the magnetization under a pulse of electric field. Moreover, even within the complex context of the pandemic period, we succeeded
to get important experimental results confirming our theoretical predictions and expectations related to the electric field control of the magnetic anisotropy. And the experimental development is not an easy task in Spintronics. The ultimate goal concerning the magneto-electric characterization of a spintronic device implicates many fabrication steps: elaboration of the multilayer thin film heterostructure stacks by Physical Vapor Deposition tools (e.g. sputtering and/or Molecular Beam Epitaxy in Ultra-High Vacuum conditions), multistep lithography stages preceded the mask design and mask fabrication, sample characterization from morphological, structural, magnetic and magneto-electric point of view. These activities are often complex, time consuming and susceptible to implementation risks, and, therefore they have been in our case significantly affected by the pandemic context. Within these circumstances, the center of gravity of our activities has been gradually moved towards theoretical modelling at various scales. Therefore, critically, we cannot say that we closed all the initially opened points.

The remaining questions, and the other arisen during the research developed within the framework of the thesis, represent interesting perspectives of the current work. Let's summarize few of them. Concerning the analytical and the ab initio modelling, we successfully investigated fundamental phenomenological aspects concerning the PMA and the capability of its control by electric field. To study the complex aspects related to the multilayer stacks architecture, we designed special supercell models and obtain very interesting and promising theoretical predictions. However, these theoretical expectations require further experimental confirmation. Dedicated samples have to be elaborated, as perspective, for instance by using the Molecular Beam Epitaxy tools available via existing research collaboration with the formal laboratory of the PhD adviser (the Jean-Lamour Institute, Nancy, France). Then, these multilayered thin film heterostructures must be further lithographically patterned in dedicated devices, suitable for static and dynamic VCMA experiments. Beyond the static experiments (e.g. the electric field dependence of the anisotropy) it would be of great interest for us to characterize the dynamics of such systems (e.g. magnetization reversal in patterned nanopillars) in order to validate and refine our theoretical findings. Concerning the magnetization dynamics, based on what we have learned about the damping broadening of the switching probability window, it

would be useful to investigate in perspective the magnetization dynamics in systems with anisotropic damping. A first guess would be that switching in one direction would be more probable that in the opposite one, but this depends entirely on the energy landscape. We see it interesting from a strictly fundamental point of view.

Concerning the potential applications, for the VCMA efficient exploitation, we saw

that it is crucial to have variation rate of the anisotropy with the electric field coefficients (β or ξ) as large as possible. Based on our findings and understanding, one way to achieve this would be to thoroughly analyze the contributing factors of the VCMA effect and to enhance the ones that lead to the increase of β. Certainly, a major ingredient would be the control of the spin-orbit interaction, via the choice of the heavy-metal underlayer. Moreover, the additive contributions of the bottom and top magnetic thin film interfaces to the PMA and VCMA, often with opposite sign, opens an interesting perspective to control the magnitude and the sign of the PMA and β. The design and the elaboration of innovative materials and multilayered systems with enhanced spin-orbit properties is expected to boost the VCMA coefficient. Indeed Bauer et al. [16], reports values of 960 fJ/Vm and 910 fJ/Vm for a simple dusting of a Co/MgO interface with Tb^{3+} and Dy^{3+} ions.On the other hand, the interest for rare-earths based compounds is not limited to the VCMA applications. Recently, the electric field modulation of anisotropy has been

proposed as a strategy for solitonic chiral structures (e.g. skyrmion) manipulation [17]. Since their nucleation and stabilization depend tremendously on the DMI, PMA and dipole field, the electric field switching seems to be an elegant tool to control the skyrmion core orientation. Concerning this topic, during the implementation period of this thesis, we performed dedicated activities concerning the skyrmionic phase diagrams simulations and skyrmion manipulation strategies [18]. see Appendix B for more details. Beyond the possibility to tune the PMA, we consider that the ability to tune the Dzyaloshinskii-Moriya interaction with a voltage will remain a major topic in the future having in view

the ascending interest of skyrmionic devices for classic, neuromorphic and quantum applications. Moreover, due to their special spin-orbit related properties, the rare-earth-3*d* alloys proved their value in application with ultrafast chiral spin textures [17]. Skyrmion nucleation has already been found in ferrimagnetic materials such as $DyCo_3$ thin films [19] and in perpendicularly magnetized ferromagnetic $SmCo_5$ multilayer stacks [20] With respect to this topic, for comparison, we also fabricated and studied Rare-Earth materials, e.g. $PrCo_3$

[21], $DyFe_2$ and $ErFe_2$ (for more details, see the Appendix C of this thesis). These types of studies, currently ongoing based on our laboratory's experience with the rare-earth transition metal alloys for magnetocaloric effect, are also considering other candidates, mainly ferrimagnetic materials based on Rare-Earths and transition metals (RE-TM).

4.2 Main original results

Beyond the main perspectives, listed in the previous section of this last chapter, we would like to briefly underline the main original results that we consider that we obtained during this thesis work.

First, from simple, intuitive and analytically solvable quantum models we pointed out the main physical mechanisms governing the spin interaction with a magnetic and electric field. We underlined the physical origin of the PMA and the possibility to manipulate the PMA and the spin by precession within pulses of electric fields. Thes results, motivated us to perform further dynamic theoretical simulation within more complex atomistic and micromagnetic tools. On the other hand, from the ab initio perspective, we made important observations related to the anisotropy origins, its intrinsic mechanisms (Rashba, charge depletion/ charge doping) and its behavior with respect to an applied electric field. Before this work, we would have thought that having a heavy metal beneath a 3*d*-ferromagnet with a MgO layer on top (standard spintronic architecture for spin-orbitronic applications), would lead to PMA for the thickness of the ferromagnet below a maximum threshold value. This PMA would the result of the cumulative contribution of the two interfaces. However, in agreement with quite limited preliminary indications from the existing literature, we discovered that there is a wide variety of phenomena taking place in very similar structures, which lead to completely opposite behaviors when the systems are subjected to an electric field. Therefore, our predictive theoretical results indicate what type of system and architecture would be the most interesting to be designed and fabricated for applications. An important finding of this work, is related to the boost of anisotropy and the improvement of the electric field modulation capability in systems where a fine platinum insertion was added at the ferromagnet/insulator interface. Moreover, we also illustrate the fact that a nonmetallic adjunction at the interface has an opposite negative effect on PMA and VCMA properties. A very interesting and original theoretical result, with potential significant impact concerning the properties of realistic experimental systems and devices, is the demonstration of the oscillations of the PMA with the thickness of the ferromagnetic ultra-thin film, attributed to a Bloch symmetry dependent quantum well effect.

Beyond these results, perhaps the most important achievement of this thesis is contained in the sixth chapter. In this chapter we describe in detail the physical basis of the magnetization dynamics within the VCMA framework. We illustrate the precessional reversal mechanisms of the magnetization within an LLG-macrospin model. Then, we demonstrate the fact that it is not mandatory to completely cancel the anisotropy barrier that separates two opposite magnetization orientation to be able to have a precessional switch. This result, pushes the VCMA framework even closer to applications, as it further demonstrates its enhanced energetic efficiency. Moreover, our results lead to the design of an innovative three terminal spintronic VCMA device, in which the precessional switch of the magnetization can be triggered by an electric current density flowing in a planar stripe. This innovative architecture could replace a commonly used one, in which the precessional effective field is induced by an in-plane magnetic field. Beyond this phenomenological design

and analysis, our micromagnetic simulations led to a quantitative estimation of the current density pulse characteristic (intensity, duration) necessary for the switch.
Another important finding of the micromagnetic simulations is related to the demonstration of a critical band regime/window in which the magnetization switching takes place disregarding the pulse length. This regime is controlled by a positive contribution from the damping constant, that broadens the switching window where the switching probability is 100%. Our results demonstrates that for the VCMA dynamics, the damping constant has a positive effect. This, represents a change of paradigm compared to the STT landscape, where the damping might come with damaging effects and increase the energy consumption: the driving force of the precessional switch by STT has to compete with the intrinsic damping of the ferromagnetic material. Beyond the LLG-macrospin approach, we further performed micromagnetic simulations in realistic nanopillars with variable lateral diameter, including both PMA, DMI and demagnetizing field contributions. Within this more complex framework, we showed that the DMI seems to contribute to the switching, by multiplying the switch domains (bands). Compared to the macrospin LLG model, the DMI is lowering the critical modulation necessary to achieve the switching and optimistically, the energetic efficiency will be enhanced, correspondingly.

Although we did not achieve as much as we initially scheduled, even the experimental side of this thesis brought some satisfying results. We successfully elaborated thin film heterostructures demonstrating PMA, as continuous films. Then, we successfully designed and fabricated UV lithography masks, for multistep patterning of spintronic devices suitable for Anomalous Hall Magnetometry experiments under applied gating field.
Despite the clearly identified problems, specific to the lithographic pattering, we successfully obtained a couple of spintronic devices on which we were able to perform the magneto-transport VCMA experiments. On these samples, we unambiguously witnessed the electric field modulation of the anisotropy and we are convinced about the unfolding phenomenology. These results are in good qualitative agreement with the theoretical expectations issued from our theoretical modelling. Moreover, during the micro-lithographic patterning steps, we identified a complete list of problems and issues that could hinder the device fabrication and quality of their properties, and functional VCMA performance. To mitigate these blocking points, we designed and fabricated a second-generation UV lithography mask that can be used in the future perspective projects. To add a personal note to this chapter, I would say that beside the scientific observations value, to me it was a deep learning experience and tough adaptation exercise, because the technical limitations and the pandemic context changed the trajectory of this work several times. However, I could not be more grateful for this entropic path, because otherwise we would have missed the multiscale analysis of this phenomenon, and perhaps we would have only acquired knowledge limited to the experimental analysis. I often think that it would have been very frustrating to perform measurements with electric field pulses whose lengths would have been even multiples of the half-precession period. Also, we would have missed all the knowledge related to the Rashba origin of PMA and its response to electric field and perhaps we would have accepted the surface charge doping explanation as a given in the absence of conflicting results, like those from X/Fe/MgO system described in Chapter 5.

Last but not least, the development of this thesis clearly demonstrates the complexity of the PMA and VCMA topics and the necessity of a multiscale approach to tackle the critical issues. This point of view is in agreement with the more and more accepted idea, nowadays, that the Spintronics requires a holistic approach, considering its current and future development perspectives. This would be a compulsory approach for the research methodology in Spintronics to fulfil the integration requests in rapidly emerging neuromorphic and quantum technologies.

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List of Publications

Scientific Papers

• Roxana-Alina One, Hélène Béa, Sever Mican, Marius Joldos, Pedro Brandao Veiga, Bernard Dieny, Liliana D Buda-Prejbeanu, Coriolan Tiusan, "Route towards efficient magnetization reversal driven by voltage control of magnetic anisotropy"
Scientific Reports, vol. 11, 1, 8801, 2021. (IF=4.997, AIS=1.207)
• R.-A. One, S Mican, A Mesaros, M Gabor, T Petrisor, M Joldos, L.D. Buda-Prejbeanu, C Tiusan, "Perpendicular Magnetic Anisotropy Electric Field Modulation in Magnetron-Sputtered Pt/Co/X/MgO Ultrathin Structures With Chemically Tailored Top Interface"
IEEE Transactions on Magnetics, vol. 57, 6, 1-10, 2021 (IF=1.848, AIS=0.399)
• Sever Mican, Roxana-Alina One, Razvan-Claudiu Pop, Coriolan Viorel Tiusan, Romulus Tetean, "Influence of Cu addition on the structural, magnetic and magnetocaloric properties of the PrCo3 intermetallic compound" Journal of Alloys and Compounds, 905, 164248, 2022. (IF=6.2, AIS=0.736)
• Roxana-Alina One, Sever Mican, Angela-Georgiana Cimpoes, u, Marius Joldos, Romulus Tetean, Coriolan Viorel Tius,an, "Micromagnetic design of skyrmionic materials and chiral magnetic configurations in patterned nanostructures for neuromorphic and qubit applications" Nanomaterials, vol. 12, 24, 4411, 2022. (IF=5.3, AIS=0.707)
• Roxana-Alina One, Sever Mican, Coriolan Tiusan "Perpendicular magnetic anisotropy and its electric field manipulation in magnetic multilayered heterostructures" Studia Universitatis Physica, vol. 66, No. 1-2, 2021
• Roxana-Alina One, Coriolan Tiusan "Rashba field contribution and electric field control of the magnetic anisotropy" Studia Universitatis Physica, Volume 67, No. 1-2, 2022

Conferences: oral contributions

- MMM 2020, 2-6 November 2020, Virtual due to pandemic context "Optimization of the VCMA-Driven Magnetization Reversal" Roxana-Alina One, Hélène Béa, Charles-Élie Fillion, Sever Mican, Marius Joldos, Bernard Dieny, Liliana D. Buda-Prejbeanu, Coriolan Tiușan
- International Balkan Workshop on Applied Physics and Materials Science 2019, 16-19 July 2019, Contanta, Romania "Electric field switching of magnetization in HM/Co-MgO system: experiments, ab-initio, micromagnetic and atomistic studies" Roxana-Alina One, Sever Mican, Amalia Mesaros, Mihai Gabor, Traian Petrișor Jr, Liliana Buda-Prejbeanu, Marius Joldos, Coriolan Viorel Tiușan
- Processes in Isotopes and Molecules - 12th International Conference, 25.09.2019-27.09.2019 Cluj-Napoca, Romania "Magnetization dynamics in perpendicularly magnetized media under the influence of an electric field" Roxana-Alina One, Sever Mican, Amalia Mesaros, Mihai Gabor, Traian Petrișor, Liliana Buda-Prejbeanu, Marius Joldos, Coriolan Viorel Tiușan
- Processes in Isotopes and Molecules, Cluj-Napoca, 22-24 September, 2021 "Voltage control of perpendicular magnetic anisotropy in systems displaying quantum well effects" Roxana-Alina One, Sever Mican, Coriolan Viorel Tiușan
- International Balkan Workshop on Applied Physics and Materials Science, July 12-15, 2022 "Electric Field Assisted Magnetization Reversal In A Au/Fe/MgO Disk In A Pulse Length Independent Regime" Roxana-Alina One, Sever Mican, Coriolan Viorel Tiușan

Conferences: posters

- European School on Magnetism – "Magnetism by Light", 17th-28th September 2018, Krak´ow, Poland "Magnetization manipulation by electric field in perpendicularly magnetized thin films" R. One, A. Mesaro, s, M. Gabor, T. Petrișor Jr, V. Pop, C. Tiușan
- ESONN Grenoble (European School On Nanosciences & Nanotechnologies), 25th August - 14th September2019, Grenoble, France "Multiscale approach for electric field switching of magnetization in HM-3d-MgO systems" Roxana-Alina One, Sever Mican, Amalia Mesaros, Mihai Gabor, Traian Petrișor, Liliana Buda-Prejbeanu, Marius Joldos, Coriolan Viorel Tiușan
- INTERMAG, 2nd – 30th April 2021, Virtual due to pandemic context "Perpendicular magnetic anisotropy electric field modulation in magnetron sputtered Pt/Co/X/MgO ultra-thin structures with chemically tailored top interface" R. A. One, S. Mican, A. Mesaros, M. Gabor, T. Petrișor Jr, M. Joldos, L.D. Buda-Prejbeanu, and C. Tiușan

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