Gated electric field control on multi-layered structures with perpendicular magnetic anisotropy

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**NANYANG
TECHNOLOGICAL
UNIVERSITY**

SINGAPORE

**Gated electric field control on multi-layered structures with
perpendicular magnetic anisotropy**

Tan Funan

SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

2019

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perpendicular magnetic anisotropy**

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SCHOOL OF PHYSICAL AND MATHEMATICAL
SCIENCES

A thesis submitted to the Nanyang Technological
University in partial fulfilment of the requirement for
the degree of Doctor of Philosophy

2019

Statement of Originality

I hereby certify that the work embodied in this thesis is the result of original research done by me except where otherwise stated in this thesis. The thesis work has not been submitted for a degree or professional qualification to any other university or institution. I declare that this thesis is written by myself and is free of plagiarism and of sufficient grammatical clarity to be examined. I confirm that the investigations were conducted in accord with the ethics policies and integrity standards of Nanyang Technological University and that the research data are presented honestly and without prejudice.

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Authorship Attribution Statement

This thesis contains material from one paper published in the following peer-reviewed journal(s) where I was the first and/or corresponding author.

Chapter 4 is published as Funan Tan, Gerard Joseph Lim, Wai Cheung Law, Fei Long Luo, Hongxi Liu, Francis Poh, Danny Shum, and Wen Siang Lew*, "Electric Field control on Gated Pt/Co/SiO₂ Heterostructure with Insulating Polymer", Journal of Physics D: Applied Physics, 51, 365001 (2018).

The contributions of the co-authors are as follows:

- I designed the study, fabricated the device, performed the measurements, analyzed the data and prepared the manuscript drafts.
- Gerard Joseph Lim and Wai Cheung Law carried out the thin film deposition process using magnetron sputtering and electron beam evaporator. They also assisted during the measurement setup.
- Fei Long Luo coded the Labview which the measurement process ran on.
- Dr Hongxi Liu, Francis Poh, Dr Danny Shum and Assoc. Prof Wen Siang Lew supervised the project and establish the collaboration between the university and the industry
- The manuscript was revised with all of the authors

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Tan Funan

Abstract

The ability to control the properties of ferro-metallic materials after device fabrication is a highly desirable feature for fundamental studies and technological advancements. In semiconductor-based devices, electric field control using a gate voltage to vary the conductivity is well established in commercialized devices. In comparison, the electric field control of spintronic devices is still in its infancy and there is a great deal of research interest surrounding this phenomenon.

This thesis presents a comprehensive study and experimental implementation of electric field control on ferro-metallic based spintronic device. Despite the electric field screening effect of metals, electric field control was realized on a multilayer ferro-metallic structure. This modulation of anisotropy energy with electric field broadens the application of electric field control to include thicker multi-layered magnetic films. This allows spintronic devices to have a larger magnetic volume, thereby enhancing the thermal stability of the device. Another investigation on electric field control focuses on the dynamics of the magneto-ionic effect using a polymer as the insulating layer. The results show that thinner polymer layers lead to a faster magneto-ionic effect. This allows the possibility of flexible spintronics devices with high fabrication throughput. In addition, electric field control was employed to assist in the injection of domain walls in a geometrically constrained Hall cross structure. The reduction in the required current density for domain wall injection illustrates the wide range of application for electric field control on spintronic devices. Such applications also include novel techniques proposed in this thesis which manipulates domain walls using electric field control. The proposed technique propagates domain walls deterministically at high velocities without relying on any intrinsic or permanent extrinsic pinning features.

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Table of content

Statement of Originality	i
Supervisor Declaration Statement.....	ii
Authorship Attribution Statement	iii
Abstract.....	1
Acknowledgements.....	2
Table of content	4
1. Introduction	9
1.1. Ferromagnetism	12
1.1.1. Exchange stiffness.....	13
1.1.2. Zeeman energy	13
1.1.3. Demagnetization energy.....	14
1.1.4. Domain wall energy	15
1.1.5. Dzyaloshinskii-Moriya interaction.....	17
1.1.6. Magneto-crystalline, strain induced and interfacial anisotropy energy	18
1.2. Stoner-Wohlfarth model	20
1.3. Domain wall propagation.....	23
1.4. Perpendicular magnetic anisotropy.....	23
1.5. Electric field control	25

1.5.1. Voltage controlled magnetic anisotropy	27
1.5.2. Magneto-ionic effect	29
1.6. Motivation and technical contributions	31
References	34
2. Methods.....	46
2.1. Micromagnetic simulations	46
2.1.1. Landau-Lifshitz-Gilbert equation.....	46
2.1.2. Finite difference/element method.....	47
2.2. Thin film deposition.....	49
2.2.1. Magnetron sputtering	49
2.2.2. Electron beam evaporation.....	51
2.3. Device fabrication.....	52
2.3.1. Electron beam lithography	53
2.3.2. Ion milling	57
2.4. Magnetic measurements	58
2.4.1. Vibrating sample magnetometer	58
2.4.2. Magneto-Optic Kerr effect	60
2.5. Electrical measurements	62
2.5.1. Anomalous Hall effect	62
Reference	64

3. Bulk effect of electric field control on multi-layered Pt/Co	67
3.1. Motivation	67
3.2. Magnetic Characterization	68
3.3. Electric field modulation of double Pt/Co stack	70
3.4. K_{eff}/E comparison with thermal stability of multi-layered Pt/Co	73
3.5. Micromagnetic validation	77
3.6. Summary	81
Reference.....	82
 4. Magneto-ionic effect on Pt/Co/SiO₂ Hall cross device using insulating polymer.....	85
4.1. Motivation	85
4.2. Effect of Co thickness on coercivity of magnetic material.....	86
4.3. Quantification of the Magneto-ionic effect	88
4.3.1. Coercivity measurement.....	89
4.3.2. Resistivity measurement	93
4.4. Polymer thickness contribution to the Magneto-ionic effect.....	94
4.5. Summary	99
Reference.....	100

5. Electric field control for energy efficient domain wall injection.....	102
5.1. Motivation.....	102
5.2. Domain wall injection using Hall cross structure.....	103
5.3. Electric field modulation of the Pt/Co/Pt stack	106
5.4. DW detection with electrical and magnetic optical Kerr imaging	109
5.5. Summary	114
Reference	115
 6. Domain wall propagation using voltage controlled magnetic anisotropy	 117
6.1. Motivation.....	117
6.2. Stepped anisotropy gradient	119
6.3. Optimization of gate electrode width	120
6.4. Domain wall dynamics due to gate electrode shape.....	123
6.4.1. Slanted gate electrodes	123
6.4.2. Chevron gate electrodes	129
6.5. Simulation parameters	137
6.6. Energy consumption	138
6.7. Summary	138

Reference.....	139
7. Conclusion and future work	142
7.1. Summary of thesis work	142
7.2. Outlook of future work	144
7.2.1. Ultra-fast magnetization switching of magnetic tunnel junction for domain injection.....	145
7.2.2. Electric field assisted domain wall propagation with spin orbit torque	146
7.2.3. Electric field control on antiferromagnet and synthetic antiferromagnetic structure.	147
Reference.....	149
Appendix A: Magnetic tunnel junction	153
Introduction	153
Results	154
Magnetization Characterization	154
Current-in-plane-tunnelling.....	156
Device tunnelling magneto-resistance measurement	157
Reference.....	159
List of publication	161
List of conference presentation.....	163

1. Introduction

The name “spintronics” is a blend of the word “spin” and “electronics”. Spintronics devices not only use the charge of an electron, but also the spin of the electron. Since the electron spin gives rise to magnetism, spintronics devices are devices that utilize the properties of magnetic materials. Magnetic materials in memory storage devices is a very old idea that has been revised and repurposed to keep up to date with technological advancements and market demands. In 1937, drum memory was invented¹ and was widely used in computers until the 1960s. The design and working mechanism of the drum memory is strikingly similar to the modern hard disk drive (HDD), where a rotating drum is used instead of a spinning disk. However, this form of memory fell out of favour in the 1950s partially due to the development of the core memory technology.^{2, 3} Core memory technology is a random access memory (RAM) using magnetic rings which can be magnetized in either the counter-clockwise or clockwise direction. The performance of the RAM was significantly superior to that of the drum memory technology and more cost efficient than the vacuum tubes or transistors of that time, making it a universal memory due to its non-volatility, high performance and almost unlimited endurance.

However, the expensive manual manufacturing process to produce core memory devices allowed the rise of static random access memory (SRAM) in the 1960s^{4, 5} and dynamic random access memory (DRAM) in 1970s.⁶⁻⁹ These semiconductor based memories use the highly automated Ultra-Violet (UV) lithography processes to take over the market. However, these semiconductor RAMs were volatile in nature, allowing magnetic memory devices to be retained in the market and used as non-volatile memory in the guise of magnetic tapes and floppy disk. The development of floppy disk technology eventually led to the HDD we use today. In the area of emerging memory technology, the

HDD serves as the drum technology of yesteryear, and the UV lithography process that drove core memory technology out of the market is being employed onto magnetic materials for non-volatile memory devices. Recent research interest in spintronics devices are mostly focused on the racetrack memory device and magnetic tunnel junctions as both have the required characteristics to fulfil the promises of being a universal memory.

Although spintronic devices have the potential to be a universal memory that can replace all memory devices in the future. The main obstacles facing spintronic devices has more to do with the market dynamics. CMOS technology does not stay stagnant, and further improvements in the current CMOS memory device makes them even harder to be replaced by spintronic devices. Furthermore, the industry wide adoption of CMOS based memory device incentives the foundries to veer towards CMOS based devices as CMOS technology is a matured technology and it is highly likely that most of the manufacturing processes for the new CMOS based device is transferable from the old processes.

Therefore, spintronic based devices need to occupy a niche market where the benefits of using a spintronic device far outweighs the use of a CMOS device. For example, MRAM are currently already in niche areas such as in satellites,^{10, 11} where the non-volatility and endurance is of paramount importance. A faulty CMOS device on Earth is cheap and easy to replace, but that is not true on satellites. This allows spintronic memory to enter the market, removing the initial barriers that prevent spintronic devices from landing a foothold in the market.

This thesis is arranged as follows:

In Chapter 1, a brief introduction of the physics governing ferromagnetic systems are discussed.

In Chapter 2, the simulation and experimental method employed in this thesis with their working principles are introduced.

In Chapter 3, the electric field control on single and double Pt/Co layer is characterized and the results on the electric field modulation and thermal stability is discussed.

Chapter 4 investigates the mechanism of magneto-ionic effect with an insulating polymer. The dependence of the magneto-ionic effect on the polymer thickness is discussed.

In Chapter 5, electric field control is employed to reduce the current density required for domain wall injection using a geometrically constrained Hall cross device.

Chapter 6 proposes a novel technique to propagate domain walls using multiple gate electrodes for local electric field control along the magnetic nanowire.

Chapter 7 summarizes the work presented on electric field control in this thesis. Future works that build on the work presented here in this thesis are also discussed.

Appendix A presents the experimental work on magnetic tunnel junction.

Below is a table of units with the conversion factor between cgs and SI units for easy reference.

Units for reference				
Quantity	Symbol	cgs	Conversion factor	SI
Magnetic moment	m	emu	10^{-3}	Am ²
Permeability	μ	dimensionless	$4\pi \times 10^{-7}$	H/m
Volume magnetization	M	emu/cm ³ (emu/cc)	10^3	A/m
Magnetic field strength	H	oersted (Oe)	$10^3/4\pi$	A/m

Table 1-1. Table of units for magnetic properties and conversion from cgs to SI units.

1.1. Ferromagnetism

Ferromagnetic materials can have spontaneous magnetization without any external magnetic field. This is due to the magnetic moments within the material that point in the same direction, resulting in the polarity of any magnets. The magnetic moments derive from the electronic structure of the atoms. Within the atoms, the spin of the electron gives rise to the spin angular momentum, which can be expressed as $\pm \mu_B$, where $\pm$ indicates the sign of the spin angular momentum quantum number¹²⁻¹⁵ and μ_B refers to the Bohr magneton. The electron's superposition around the nucleus gives rise to the orbital angular momentum. This orbital angular momentum of the electrons can be expressed as $\mu_B l$, where l is the orbital angular momentum quantum number.¹²⁻¹⁴

However, due to Pauli's exclusion principle,^{14, 16} an electron with up spin must be paired with an electron with down spin in the same energy band. Therefore, the spin and orbital momentum of the electrons are often cancelled out by the paired electrons, which is the main reason why most materials are not magnetic. However, the $3d$ orbital in Co, Fe and Ni is both close to the Fermi level and narrow enough such that the band splits up. This neatly circumvents the Pauli's exclusion principle, thereby allowing unpaired electrons to occupy the $3d$ orbital.^{12-14, 17} The combined spin and orbital angular momentum of these unpaired electrons contribute towards the magnetic moment of the atomic spin.

The rest of this section will explore the various types of energy contributions that determines how these atomic spins align and the direction of the magnetization in ferromagnetic materials.

1.1.1. Exchange stiffness

The electronic structure of the atom contributes to the magnetism property of a material. However, in most materials, these atoms do not usually exist alone. When the atoms are in proximity of each other, it leads to an exchange interaction between the neighbouring atomic spins which can be described as

$$E_{ex} = -J(\mathbf{S}_i \cdot \mathbf{S}_j), \quad (1.1.1)$$

where E_{ex} refers to the exchange energy between the pair of atomic spins, $\mathbf{S}$. In the many-body system, the parameter, J , is the exchange constant, and the sign determines if the exchange interaction results in a ferromagnetic or antiferromagnetic spin structure.¹⁸⁻²⁰ This is illustrated in Figure 1.1-1, where the arrows represent the atomic spins. Figure 1.1-1(a) shows a ferromagnetic ordering due to a positive J and 1.1-1(b) shows an anti-ferromagnetic ordering due to a negative J .

Figure 1.1-1. Schematic diagram representing the atomic spins interaction due to the exchange constant, J . (a) Positive J results in an energy minimization when the atomic spins are in parallel orientation, which allows the material to have a net magnetic moment. (b) Negative J results in an anti-parallel orientation of the atomic spins, resulting in zero net magnetization for anti-ferromagnetic materials.

1.1.2. Zeeman energy

Ferromagnetic materials do not require external magnetic field for spontaneous magnetization. However, an external magnetic field introduces an energy term to energetics

of the magnetization, $\mathbf{M}$. This energy term is known as Zeeman energy,²⁰⁻²² and can be described with the following equation

$$E_{ze} = -\mu_0 \mathbf{M} \cdot \mathbf{H}_{\text{ext}} \quad (1.1.2)$$

where μ_0 is the permeability of vacuum, $\mathbf{M}$ is the magnetization and $\mathbf{H}_{\text{ext}}$ is the external magnetic field. As with all systems, the preferred energy state is the lowest possible energy state. Therefore, when an external magnetic field is applied to a magnetization, the direction of the magnetization changes to lower the Zeeman energy.

1.1.3. Demagnetization energy

The demagnetizing energy, E_{demag} arises from the demagnetization field generated by $\mathbf{M}$ itself.^{14, 20, 21, 23, 24} This energy can be described in a similar manner as the Zeeman energy in Equation (1.1.3) as follows

$$E_{\text{demag}} = -\frac{\mu_0}{2} \mathbf{H}_{\text{demag}} \cdot \mathbf{M}, \quad (1.1.3)$$

where $\mathbf{H}_{\text{demag}}$ is the demagnetization field generated from the $\mathbf{M}$. Similar to Zeeman energy, the demagnetization energy can also be lowered. In practice, this is usually achieved by controlling the shape of the magnetic object. Therefore, this method of controlling the direction of magnetization is also known as shape anisotropy.^{12-14, 17, 25} Figure 1.1-2(a), illustrates using micromagnetic simulation that when a rectangular magnet with the direction of $\mathbf{M}$ along the longer axis, it results in fewer $\mathbf{H}_{\text{demag}}$ interaction with $\mathbf{M}$. The opposite is shown in Figure 1.1-2(b), where $\mathbf{M}$ is along the shorter axis which results in a higher E_{demag} . In this way, the E_{demag} determines the direction of $\mathbf{M}$ via shape anisotropy. The difference in the direction of $\mathbf{M}$ in the 24 nm by 2 nm rectangular magnet

results in a difference of E_{demag} by approximately 14 times. This anisotropy energy has been used in extremely high aspect ratio magnetic pillars to determine $\mathbf{M}$ direction for the spintronic device,²⁶ similar to the example shown in Figure 1.1-2.

Figure 1.1-2. Demagnetization energy calculated with micromagnetic simulations. The demagnetization fields are mapped as arrows in the figure and the magnetization direction is represented by the blue and red colour which illustrates the poles of the magnets. The magnetic blocks have a size of 56 nm $\times$ 32 nm $\times$ 1 nm with a magnetic saturation of 10^6 A/m. (a) The magnetization direction is along the long axis, resulting in a lower demagnetization energy density of $4.5 \times 10^3 \text{ J/m}^3$. (b) A higher demagnetization energy density of $5.7 \times 10^3 \text{ J/m}^3$ results from the magnetization pointing along the shorter axis.

1.1.4. Domain wall energy

The minimization of the demagnetization energy term can also give rise to the formation of a domain wall (DW) in a ferromagnetic material. The DW acts as a boundary between two neighbouring domains which have their local $\mathbf{M}$ in different directions.^{24, 27,}

²⁸ The formation of DWs is opposed by the J_{ex} , which is minimized when the neighbouring spins are aligned. Another energy term that encourages spins to be collinear is the

anisotropy term, which will be further discussed in section 1.1.6. Hence, the DW energy can be written as an integral as follows,

$$E_{dw} = \int_{-l/2}^{+l/2} [K_{anis}(x) + E_{ex}(x)]dx \quad (1.1.4)$$

where K_{anis} represents the anisotropy energy, E_{ex} represents the exchange energy, and the boundary conditions represent the start and end of the domain wall width, l . Using variational approach to find the minimum energy, the value of l can be given as,

$$l_{min} = \pi\sqrt{2}\sqrt{E_{ex} / K_{anis}} \quad (1.1.5)$$

Solving the integral in Equation (1.1.5) would result in $E_{dw} = K_{anis}l / 2 + E_{ex}\pi^2 / l$.

By substituting Equation (1.1.6) to the solution of Equation (1.1.5), the DW energy is given to be,

$$E_{dw} = \pi\sqrt{2}\sqrt{E_{ex}K_{anis}} \quad (1.1.6)$$

This simple model from Equation (1.1.5) show that E_{ex} would increase the DW width, whereas, K_{anis} increases the energy required for the domain wall, which decrease the DW width. The balance of the energy terms introduced thus far in a ferromagnetic system can be expressed simply in an illustration as shown in Figure 1.1-3, where E_{demag} and E_{dw} represent the energy terms for demagnetization and domain wall respectively. Although DWs reduce the E_{demag} , the DWs themselves require energy, which is related to the intrinsic E_{ex} and K_{anis} of the material as shown in Equation (1.1.7).

Figure 1.1-3. Schematic diagram illustrating the demagnetization and domain energy in a ferromagnetic system. (a) A single domain results in large demagnetization energy and negligible domain wall energy. (b) A two-domain structure lowers the demagnetization field. (c) Demagnetization energy becomes almost negligible as most of the flux are closed, the reduced demagnetization energy comes at the cost of a heightened domain wall energy.

1.1.5. Dzyaloshinskii-Moriya interaction

Dzyaloshinskii-Moriya interaction (DMI) is an antisymmetric exchange interaction which causes spin canting. Similar to exchange interaction, DMI derives from the interaction between atomic spins, however it can only arise from a mixture of atoms or at the interface of magnetic thin films.²⁹⁻³¹ It is an exchange interaction between two neighbouring atomic spins through a third atom illustrated in Figure 1.1-4, where the atomic spins are represented by the arrows, the third atom is represented by the vector pointing out of the plane, and the dotted lines represent the interactions. In Figure 1.1-4(a), the $J > 0$, which gives rise to a ferromagnetic order, and in Figure 1.1-4(b), the $J < 0$, giving rise to an anti-ferromagnetic order. In the case where $J < 0$, the spin canting caused by the DMI causes an overall $\mathbf{M}$ pointing towards the right as shown in Figure 1.1-4(b). The weak ferromagnetism due to the DMI in an anti-ferromagnet resulted in the first experimental demonstration of DMI.³⁰

Figure 1.1-4. Schematic diagram illustrating the DMI between two atomic spins. (a) Spin canting occurring in a ferromagnetic structure due to DMI. (b) Spin canting occurring in an anti-ferromagnetic structure due to DMI, resulting in a small net magnetization.

The energy associated with DMI is given as follows,

$$E_{DMI} = \mathbf{D} \cdot (\mathbf{S}_i \times \mathbf{S}_j) \quad (1.1.7)$$

where, $\mathbf{D}$ is the DMI vector and its direction determines the chirality of the spin-canting effect. Similar to the A_{ex} , the direction and magnitude of $\mathbf{D}$ depends on the atomic spins involved in the interaction, and can be describe as follows

$$\mathbf{D} = d\mathbf{u} \times \mathbf{z} \quad (1.1.8)$$

where, $\mathbf{u}$ is the unit distance vector between the two neighbouring spins, $\mathbf{z}$ is the unit vector perpendicular to the thin film plane, and d is a coefficient proportion to the exchange interaction of the third atomic spin to the two neighbouring atomic spins.

Due to the spin canting effect, the DMI in a ferromagnetic system affects the DW in the system.^{32, 33} This will be further discussed in Section 1.3 of Chapter 1 and in Chapter 6.

1.1.6. Magneto-crystalline, strain induced and interfacial anisotropy energy

Magneto-crystalline anisotropy energy derives from the crystal structure of the ferromagnetic material. Since the atomic spins are stack in a crystal lattice, this can deform

the orbitals, giving the spin angular momentum an anisotropy due to the deformation.^{34, 35} An illustration of this is shown in Figure 1.1-5, where the atomic spins in 1.1-5(a) favour a magnetic direction in the up-down direction, whereas in 1.1-5(b), a left-right direction would be favoured. Stress induced anisotropy can also stem from the same vein as magneto-crystalline anisotropy, whereby the lattice stress induced due to a lattice mismatch from an interface or dopants allows the lattice structure of the atomic spins to create an anisotropy.³⁶⁻³⁸ Spin orbit coupling at the interface also contributes towards interfacial anisotropy where the hybridization of the orbitals at the interface creates an anisotropy.³⁹⁻⁴¹ This predominately occurs with heavy metal with high spin orbit coupling such as Pt and Pd.⁴²⁻⁴⁵

Figure 1.1-5. Schematic diagram illustrating the change of magnetization direction due to the deformation of the electron orbitals. (a) An anisotropy direction is created in out-of-plane direction. (b) The magnetization points along the axis of anisotropy in the in-plane direction due to the deformation of the orbitals.

These anisotropy energies can allow ferro-metallic thin film to have perpendicular magnetic anisotropic (PMA) energy, which energetically favours $\mathbf{M}$ to have an out-of-plane easy axis. Without these anisotropy energies due to the lattice structure, shape anisotropy dominates, and magnetic thin films would only have an in-plane magnetic anisotropy (IMA) energy. The total effective anisotropy energy of a magnetic thin film can be described as follows,

$$K_{eff} = \frac{K_{interfacial}}{t} + \frac{K_{strain}}{t^2} + K_{bulk} - \frac{\mu_0}{2} M_s^2 \quad (1.1.9)$$

where K_{eff} represents the total effective anisotropy energy, $K_{interfacial}$ represents the interfacial anisotropy, K_{strain} represents the strain induce anisotropy, K_{bulk} represents the magneto-crystalline anisotropy, t represents the thickness of the thin film and $\frac{\mu_0}{2} M_s^2$ represents the shape anisotropy, also known as the demagnetization term. The equation can be simplified to represent the energy contributing towards PMA and the energy term contributing towards IMA, which is usually represented as follows,

$$K_{eff} = K_{anis} - \frac{\mu_0}{2} M_s^2 \quad (1.1.10)$$

where K_{anis} represents the combined energy terms of the magnet-crystalline, interfacial and strain induce anisotropy. Therefore, the anisotropy of a thin film material which determines the direction of $\mathbf{M}$ is a balance between the K_{anis} and the demagnetization term. If $K_{eff} > 0$, the thin film material will have PMA and when the $K_{eff} < 0$, the thin film material will have IMA.

1.2. Stoner-Wohlfarth model

Some of the energies introduced can be used to formulate the Stoner-Wohlfarth model,^{46, 47} which can be described as follows,

$$E_{total} = K_{anis} \sin^2 \theta - H \cdot M_s \cos(\theta - \phi) \quad (1.2.1)$$

where K_{anis} is the anisotropy energy, M_s is the magnetic saturation, and H refers to the applied magnetic field. Figure 1.2-1 illustrates the variables used in Equation (1.2.1), where $\mathbf{M}$ represents the magnetization vector.

Figure 1.2-1. Illustration of the Stoner-Wohlfarth model, where the easy axis of the ferromagnetic material is perpendicular to the applied magnetic field, H . The axes and angles are colour coded for visual aid.

In this simple model, several energy terms are not considered, such as DMI, exchange and DW energy. Therefore, this simple model is only effective for assuming that the magnetic structure behaves as a single domain. In Equation (1.2.1), the first term, $K_{anis} \sin^2 \theta$ is known as the anisotropy energy term, and it is at minimum when $\theta = 0$. The axis determined by θ is also known as the easy axis of the magnetic system. The second term of the equation, $H \cdot M_s \cos(\theta - \phi)$ describes the energy term of the applied magnetic field.

In order to present the interactions, the magnetization is defined into two components. The magnetization can be projected along the magnetic field in the

longitudinal configuration, or perpendicular to the magnetic field in the transverse configuration as shown in Figure 1.2-2(a) and 1.2-2(b) respectively.

Figure 1.2-2. Hysteresis curve from the Stoner-Wohlfarth model, in (a) the longitudinal configuration and (b) the transverse configuration. The figure legends show the angle of ϕ .

In Figure 1.2-2 shows the magnetization dependence on the applied magnetic field at various ϕ . At high magnetic fields, where $H > K_{anis}$, the magnetization is parallel to the magnetic field. When the magnetic field is lowered, the energy between anisotropy and Zeeman competes, resulting in magnetization switching or magnetization rotation based on the ϕ . When ϕ is at 90° , the magnetization saturates at K_{anis} , however, this rarely occurs in experimental works due to physical defects which greatly reduces the K_{anis} at random localized regions.⁴⁸ When ϕ is at 0° , this cause the localized magnetization to switch before the $H = K_{anis}$, allowing the magnetization of the material to saturate as the nucleated domain propagates with $H < K_{anis}$. Therefore, it is common to find the K_{anis} experimentally

with ϕ at 90° ,⁴⁹⁻⁵¹ with the magnetization gradually rotating as predicted by the Stoner-Wohlfarth model.

1.3. Domain wall propagation

A magnetic DW can be propagated by either a magnetic field and/or an electric current. However, it is less desirable for DWs to be propagated by magnetic field as it results in a bidirectional DW motion, causing the domain to become larger. The larger domains reduces memory density as illustrated in Figure 1.3-1(a). This issue is trivial for current induced DW propagation as the DWs move unidirectionally. All the domain walls move in the same direction and the magnetic configuration is eventually shifted as shown in Figure 1.3-1(b). Therefore, from memory application point of view, the current induced DW motion is considered to be superior than magnetic field induced DW motion.⁵²

Figure 1.3-1. Domain wall propagation by (a) magnetic field and (b) spin current. The domain walls propagated by magnetic field moves in opposite direction away from each other, whereas the domain walls propagated by a spin current moves in tandem.

1.4. Perpendicular magnetic anisotropy

It has been established since 1977 that PMA materials are superior for use as a practical magnetic memory storage device than magnetic materials with IMA.⁵³⁻⁵⁵ As

shown in Figure 1.4.1, this is due to the demagnetization energy from the magnetization bits, where magnetic bits of equal density and aspect ratio for PMA and IMA material is compared. The PMA material can further reduce the demagnetization energy with a thicker material. This increases the aspects ratio, which further reduces the demagnetization energy due to shape anisotropy as observed in Figure 1.1-2(b).

Figure 1.4-1. Demagnetization energy of magnetization bits in the (a) PMA and (b) IMA configuration calculated using micromagnetic simulation. The arrows inside the coloured squares represents the direction of magnetization for each individual magnetization bit, the arrows outside represent the demagnetization field. The magnetic blocks have a size of $56 \text{ nm} \times 32 \text{ nm} \times 1 \text{ nm}$ with a magnetic saturation of 10^6 A/m .

Studies also have shown that PMA is beneficial to many spintronic devices, such as in racetrack memory, where the DWs in PMA material require a lower current density to propagate.^{56, 57} Magnetic tunnel junctions also have increased thermal stability and a low required switching energy when PMA material with high anisotropy energy is used.⁵⁸⁻⁶¹ These reasons motivate researchers to focus on PMA materials for future spintronic device.

As introduced in Section 1.1, PMA is usually induced by either magneto-crystalline anisotropy, interfacial anisotropy and/or stressed induce anisotropy. In ferromagnetic thin films, the contribution from the interface plays a dominate role, as it can exceed the in-plane shape anisotropy created by the demagnetizing field. The interface contribution to the anisotropy of PMA materials has been well established.^{44, 62} Researchers focus on PMA

materials structure with heavy metal/ferromagnetic layer interface as the large spin orbit interaction of the heavy metal can contribute towards a stronger PMA.^{42, 44, 63-66}

1.5. Electric field control

Conventionally, spintronic devices can be controlled by magnetic field, such as in a HDD or toggle Magnetic Random Access Memory (MRAM).⁶⁷⁻⁶⁹ However, a strong localized magnetic field requires a large current, and has the detrimental effect of stray magnetic fields that limit the device size and density due to cross talk caused by the Oersted field.⁷⁰⁻⁷²

The introduction of spin-torque methods trivialize any potential issues arising from cross talk.⁷³⁻⁷⁷ Nonetheless, it is inherently a current-based method, typically requiring currents of $>1 \times 10^{11}$ A/m². Furthermore, since the current is running directly through the magnetic material, Joule heating caused by the current is highly undesirable as it reduces the thermal stability. These innate challenges that plagues current based techniques provided the impetus for the experimental realization of electric field control on magnetic materials. Electric field control on spintronic devices can be broadly divided into two categories.

Firstly, the electric field can induce a change in the insulating layer which in turn modulates the magnetic layer adjacent to it. This is usually achieved by using a piezoelectric or ferroelectric insulating layer, whereby the electric field induces a strain.⁷⁸⁻⁸⁰ However, the endurance of such systems raises concerns on the commercial viability due to the piezoelectric fatigue caused by the strain induced on the material.^{78, 81-85} In the case of multiferroics systems, the working mechanism is similar except that the insulating layer

also acts as the magnetic layer.⁸⁶⁻⁸⁸ However, the high resistance of the multiferroic materials requires high energy for any current to be in the system.

The other major group for electric field control is centred about the anisotropy modulation of ferro-metallic materials and will also be the focus of this thesis. Electric field control for spintronics devices allows current based techniques to either assist with the magnetic manipulation or to act as a direct comparison.⁸⁹⁻⁹² The first experimental demonstration of electric field control on ferro-metallic materials was performed on FePt and FePd thin films using a liquid electrolyte as the insulating layer. The electric control modulated the coercivity of the material.⁹³ Before this discovery, it was assumed that experimental observable of electric field control on ferro-metallic materials is virtually impossible due to the screening effect of conductive materials.⁹⁴ This breakthrough sparked the research interest in electric field control on ferro-metallic materials.^{50, 95-100} A solid oxide layer as an insulating layer is also found to produce the electric field control effect,¹⁰¹ and also in various ferro-metallic materials such as CoFeB/MgO,^{92, 102-104} Pt/Co,^{100, 102, 105, 106} and CoFeAl/MgO.¹⁰⁷ Various characterization techniques were employed to observe the electric field control, such as by using the tunnelling magnetoresistance in magnetic tunnel junctions,^{90, 108} Kerr microscopy,¹⁰⁹ ferromagnetic resonance,¹¹⁰ anomalous Hall effect,¹¹¹ and Brillouin light scattering.¹¹²

The energy efficient nature of electric field control requires only the minute energy for the charge or discharge of a capacitor. A gate voltage creates an electric field across an insulator that interfaces with the magnetic material. This enables the manipulation of the magnetic properties, similar to how the resistivity of semi-conductor devices are being controlled by a gate voltage.¹¹³⁻¹¹⁵ In the remaining part of this section, a few of the proposed mechanism for electric field control on ferro-metallic materials will be briefly discussed.

1.5.1. Voltage controlled magnetic anisotropy

The voltage controlled magnetic anisotropy (VCMA) effect describes the phenomena where the K_{anis} of the material is modulated by the electric field, this effect is reversible and dependent on the applied electric field strength and its polarity.¹¹⁶⁻¹¹⁸ Apart from modulating the magnetic properties statically with an electric field, the timescale of which VCMA operates in allows the effect to be utilized with very short nanoseconds electrical pulses. This generates a temporal electric field which causes a magnetization precession induced by the sudden change in the direction of magnetic anisotropy.^{90, 92, 119} However, in order for this technique to be viable, it requires a tight control on the duration of the electric pulse.

There are a few models that describe the working mechanism of the VCMA effect and the rest of this section will explore two of the leading models used to explain the phenomena.

1.5.1.1. Electron accumulation/decumulation

The VCMA effect can be attributed to the accumulation and decumulation of the conduction electrons at the oxide/ferro-metallic interface causing a change in the magnetic anisotropy.¹²⁰⁻¹²² The accumulation/decumulation of electrons changes the position of the Fermi level which changes the electron density in the $3d$ orbitals. As a result, the number of electrons with up spins and down spins changes which modulates the magnetization of the material at the vicinity of the insulator/ferro-metallic interface. This is schematically illustrated in Figure 1.5-1, where the Fermi level and density of states for the up and down spins are shown at the insulator/ferro metallic interface.

Figure 1.5-1. Schematic diagram illustrating the density of states at the insulator/ferro-metallic interface during (a) electron accumulation and (d) electron decumulation.

1.5.1.2. Orbital deformation

Another possible mechanism for VCMA is the orbital deformation of the atomic spins under an electric field. The deformation of the orbital enhances the spin orbital interaction due to the relative shift of the nucleus position in relation to the orbitals.^{120, 123-125} The dependence of this contribution can be either symmetrical or asymmetrical with respect to the direction of the applied electric field. This phenomenon is illustrated as a toy model in Figure 1.5-2 where 1.5-2 (a), (b) and (c) shows the symmetrical contributions and 1.5-2(c), (d) and (e) shows the asymmetrical contributions.

Figure 1.5-2. Schematic diagram of electron orbital shown in (a) and (d) and its deformation due to an applied electric field in (b), (c), (e) and (f).

In Figure 1.5-2, $\mathbf{E}$ represents the applied electric field and $\mathbf{H}_{so}$ represents the magnetic field due to spin orbit interaction. In Figure 1.5-2(a), (b) and (c) the nucleus is slightly shifted relative to the orbitals due the electric field. The direction of $\mathbf{H}_{so}$ does not depend on the direction of the electrical field. However, for Figure 1.5-2 (d), (e) and (f) where the orbitals are elliptical, the orbitals can be either elongated or contracted due to the electric field. The $\mathbf{H}_{so}$ is non-zero even without any electrical field as emphasised in Figure 1.5-2 (d). Therefore, the direction of an electrical field either reduces or increases the $\mathbf{H}_{so}$ without changing its direction. Although the $\mathbf{H}_{so}$ does not switch its direction, the changes of the $\mathbf{H}_{so}$ is still opposite for the opposite electric field direction. As shown in Figure 1.5-2 (e) and (f) the $\mathbf{H}_{so}$ is larger with an electric field pointing down and the $\mathbf{H}_{so}$ is conversely smaller when the electric field is pointing up.

1.5.2. Magneto-ionic effect

Unlike the other previous electric field control mechanism, magneto-ionic effect involves the physical diffusion of oxygen ions. The negatively charged oxygen ions are able to migrate in and out of the magnetic material adjacent to it especially in oxides with high oxygen mobility such as GdO_x .^{89, 106, 126-131} The migration of the oxygen ions leads to two effects that modulates the magnetic properties. In magnetic materials that depends on interfacial anisotropy. Oxygen ion significantly deforms the $3d$ orbitals of the magnetic material, which induces PMA. When oxygen ions migrate into the magnetic material, the spin-orbit interaction of the oxygen ion and d orbitals is enhanced. This consequently

increases the PMA energy. The reverse effect can also be observed, resulting in lower PMA energy when oxygen near the interface migrates away,

Another effect due to the migration of the oxygen ions is due to the increase or decrease of the magnetic dead layer. The magnetic dead layer is a non-magnetically active layer that is caused by the interdiffusion between magnetic and non-magnetic material occurring at the interface.¹³² The magnetic dead layer naturally occurs from the process of the growth deposition, where the deposited material has energy to diffusion into another material. Hence the migration of oxygen in and out of the magnetic material alters the effective magnetic layer thickness, which greatly determines the total magnetization, shape anisotropy and anisotropic energies that has a thickness dependence.⁸⁹

Figure 1.5-3 shows a schematic diagram to illustrate the magneto-ionic effect from 1.5-3(a) to (e), the graphs from 1.5-3 (f) to (j) show the corresponding hysteresis of the magnetic layer in the out-plane-direction. In Figure 1.5-3(a) and (f), the magnetic layer has PMA due to surface and stress induced anisotropy. An application of electric field allows oxygen ions to migrate out of the magnetic dead layer at the interface as shown in Figure 1.5-3(b) and (g), where the total magnetic saturation increases due to increased magnetic volume. Continuous application of the electric field in Figure 1.5-3(c) and (h) allows the shape anisotropy of the thin film to dominate over the surface and stress induced anisotropy, resulting in an in-plane easy axis. This process is reversed in Figure 1.5-3(d) and (h), where the opposite polarity of the electric field is applied, causing the oxygen ions to migrate back into the magnetic layer at the interface, achieving the same state as shown in Figure 1.5-3 (a) and (f). The prolonged application of this electric field in Figure 1.5-3(e) and (j) allows the oxygen ions to convert the entire magnetic layer into a dead layer.

Recently, the magneto-ionic effect has been reproduced using hydrogen as the migration ions instead of oxygen.¹³³ This result potentially unleashes the wide array of chemical bonding available to ferro-metallic materials for fundamental research and technological advancements.

Figure 1.5-3. Schematic illustration of the magneto-ionic effect and the resulting hysteresis curve. (a)/(f) A naturally occurring dead layer reduces the magnetic layer thickness, allowing it to have PMA. (b)/(g) Oxygen moves out of the magnetic layer due to the applied electric field, resulting in a larger magnetization signal. (c)/(h) The lack of a dead layer results in a much thicker magnetic layer, causing it to display IMA. (d)/(i) The reapplication of electric field in an opposite polarity allows oxygen to move back into the magnetic layer, creating a dead layer and obtaining the results shown in (a)/(f). (e)/(j) Further application of the electric field results in a non-magnetic material as the entire layer becomes a dead layer. Figure redrawn from ⁸⁹.

1.6. Motivation and technical contributions

Current based techniques are the conventional way to manipulate the magnetization of the material. The most direct and simple way to change the magnetization state of a magnetic material is to generate a magnetic field by passing a current. However, the stray magnetic field caused by such methods does not scale well. However, the electrical charges

passing through a spintronic device is only a means to an end. It is to influence the state of the magnetic material which is the basis for all spintronic devices. This is unlike in a CMOS device, where the charges involved are a fundamental aspect of the device. For spintronic devices, there is no fundamental requirement for the amount of charges to be used in order to influence the state of the magnetic material.

Instead of using magnetic field, an alternative approach is to apply an electric field. This method promises high energy efficiency as negligible amount of current is required. A gate voltage can be applied onto an insulating material adjacent to a magnetic material. This simple setup would be the basic premise for the work demonstrated in this thesis.

The main work in this thesis is presented in Chapter 3, 4, 5 and 6. In Chapter 3, the issue of electrical field decay into a ferro-metallic material was highlighted. The decay limits any electric field effect to the surface of the magnetic material. The work here demonstrates that despite the electric field decay, a bulk component of the electric field effect exists in a Pt/Co multilayer. By demonstrating that the properties of bulk or multilayered ferro-metallic material can be modulated with electric field, a larger engineering flexibility is allowed when designing a spintronic device. This is further shown in this work where the thermal stability of the material is greatly increased with a double Pt/Co layer as compared to the single Pt/Co layer.

In Chapter 4, an alternative insulating layer was employed. In principle, a thin layer of insulating layer is sufficient to generate the electric field required. However, defects in the insulating layer limits the thickness of the insulating layer. Another idea is use a liquid based material which avoids defect formation, the liquid can simply be dripped onto the device. However, practical usage of such a device involves keeping the liquid at its freezing temperature or some form of encapsulation. Here, a polymer was proposed as the insulating

layer due to its liquid nature, thereby avoid defect formation. After baking, the polymer provides the rigidity for use as practical device. The demonstration in this work shows results from a magneto-ionic effect and provides a fast and reliable method that employs an insulating polymer layer instead of depositing a thick layer of oxide.

In Chapter 5, a novel application of the electric field modulation effect was demonstrated. The anisotropy of a magnetic material was lowered with the application of a gate voltage before a current pulse was applied to inject a DW. This lowers the required current density for DW formation. The results from this work pave the way toward an energy efficient spintronic device based on electric field effect.

In Chapter 6, a technique to propagate DW using electric field effect was proposed and demonstrated using micromagnetic simulation. Gate electrodes of slanted and chevron shapes were compared for their efficiency at propagating DWs. An analysis of results demonstrate a clear advantage in the utilization of chevron-shaped gate electrodes for higher DW velocity. The obtained DW velocity from the proposed technique is comparable to the DW velocity from conventional current based techniques. The proposed technique allows an energy efficient DW propagation that is free from pinning issues that plagues the current based propagation techniques.

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2. Methods

This chapter will introduce the micromagnetic simulation framework that builds on the ideas introduced in Chapter 1. In this thesis, the micromagnetic simulations serves in a variety of ways to either supplement the experimental studies or to propose novel techniques that would be either technologically difficult or tedious to implement in a research lab. The experimental techniques used in this thesis are also introduced in this Chapter.

2.1. Micromagnetic simulations

2.1.1. Landau-Lifshitz-Gilbert equation

The Landau-Lifshitz-Gilbert (LLG) equation allows the dynamics of the magnetization to be formalized along with all the energy terms introduced in Chapter 1 as follows,

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma_0 (\mathbf{M} \times \mathbf{H}_{\text{eff}}) + \frac{\alpha}{M_s} (\mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t}) \quad (2.1.1)$$

where $\mathbf{M}$ represents the magnetization, t represents time, γ_0 represents the gyromagnetic ratio, $\mathbf{H}_{\text{eff}}$ represents the effective field, α represents the damping constant and M_s represents magnetic saturation.

Since Equation (2.1.1) shows the relation of two vectors, $\mathbf{M}$ and $\mathbf{H}_{\text{eff}}$, it is useful to visualize the magnetization dynamics with the two vectors as shown in Figure 2.1-1. The torques acting on the magnetization is illustrated by the blue and green arrows, which is derived from the LLG equation. The blue arrow, $\mathbf{M} \times \mathbf{H}_{\text{eff}}$ represents the first torque term in the LLG equation, it causes the magnetization to precess around the $\mathbf{H}_{\text{eff}}$. The second

torque term, $\mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t}$ is presented by the green arrow and is also known as the damping torque, which allows the magnetization to eventually align with the $\mathbf{H}_{\text{eff}}$. The damping constant, α , is associated with a transfer of energy away from the magnetic system to other energy system such as the lattice structure through the spin-orbit interaction or disperse as heat energy. The data points from Figure 2.1-1 were obtained using micromagnetic simulation, which calculates using the LLG equation. The rest of Section 2.1 will briefly explore how micromagnetic simulation software packages solve the LLG equation.

Figure 2.1-1. Magnetization dynamics calculated by micromagnetic simulation package, Mumax3. ¹ The blue arrow represents the precession term and the green arrow represents the damping term. The colour of the data point represents the evolution of magnetization with time. The total simulation time spans 0.5 ns as indicated by the colour map on the left of the figure.

2.1.2. Finite difference/element method

It is often difficult to solve the LLG equation for a given magnetic system analytically. Therefore, micromagnetic simulations solve the LGG equation using finite

difference method by employing techniques such as the Euler method or the Runge-Kutta method. These methods solve the LLG equation numerically by finite difference approximation of the derivatives. In this thesis, the micromagnetic simulations are calculated using the Runge-Kutta method. This is a fourth order method, where the weighted average of four interval increments defines the estimated rate of change. This allows a very accurate numerical solution to be obtained as compared to the 1st order Euler method.²

The solving of the LLG equation is further complicated by the geometry of the magnetic system, which has a significant effect on the $\mathbf{H}_{\text{eff}}$ due to shape anisotropy. Hence, the geometry of the magnetic system is considered into micromagnetic simulation by the finite element method. The finite element method discretizes the geometry of the system into small cells. Each cell represents a magnetization which allows the LLG equation to be solved whereby the $\mathbf{H}_{\text{eff}}$ for each cell is recalculated after each time step using the finite difference method.

Although micromagnetic simulation packages offer great flexibility in the investigation of magnetization dynamics, both finite difference and finite element methods are approximation methods. Hence, the parameters used for the micromagnetic simulations have to be carefully chosen for the results to have any significance for experimental implementation. For example, the cell size needs to be smaller than the exchange length of the magnetic material, otherwise any study involving the exchange energy parameter is inaccurate. Furthermore, the discrete cells do not accurately represent a smooth curve, resulting in a staircase-like artefact at the edges as shown in Figure 2.1-2. Hence, geometrical shapes with straight lines are generally preferred for micromagnetic

simulations, otherwise smaller cell size for curved geometry structures are required which results in longer computation time.

Figure 2.1-2. Schematic illustration of the jagged edge due to the discrete cells used to simulate object with curvatures.

Graphic cards with large number of processing cores (~100-1000) allows the micromagnetic simulations to run quickly due to the parallel processing capabilities of the multiple cores. Micromagnetic simulators that make use of graphic cards are Mumax3,^{1, 3} GoParallel⁴ and MicroMagnum.⁵ In this thesis, all of the micromagnetic simulations are carried out using the open source software, Mumax3. The simulator is designed for low memory use and can solve the LLG equation for 16 million cells with 2GB of GPU memory, hence it is ideal for inexpensive GPUs.

2.2. Thin film deposition

2.2.1. Magnetron sputtering

Sputtering involves the bombardment of ionized Ar gas onto the target material. The target material ejected by the bombardment moves towards the substrate and forms a thin layer of the desired material on the substrate. Strong permanent magnets under the target form a magnetic trap for the electrons, which enables the sputtering process to be

much faster. A schematic diagram for the magnetron sputtering process is shown in Figure 2.2-1.

Figure 2.2-1. Schematic diagram for magnetron sputtering. The target material is negatively charged which accelerates the Ar ion towards itself. The Ar bombardment releases the target material which can be deposited on the substrate.

All magnetic thin films were deposited using a high vacuum magnetron sputtering system developed by AJA. Figure 2.2-2(a) and 2.2-2(b) show the setup of the sputtering system and interior of the sputtering chamber respectively. The base pressure is maintained by a cryogenic pump at $\sim 10^{-8}$ Torr, and the sputtering pressure is kept at 2-4 mTorr by maintaining an Ar flow of 20 sccm and varying valve of the cryogenic pump. As the desired thin film in this thesis is a smooth homogenous layer of material, the lowest sputtering pressure is chosen to sustain the Ar plasma to prevent grain formation on the thin film.^{6, 7} This sputter pressure differs for target/material sputtered but is kept within the range of 2-4 mTorr.

Figure 2.2-2. Sputtering chamber with a six-stage loadlock with control tower.⁸ Interior of the sputtering chamber. Eight target materials can be installed, two DC and two RF supply are used for sputtering, requiring a change of cables when the desired material to be sputter is needed.

2.2.2. Electron beam evaporation

An Edwards 306 system was used for the oxide deposition using electron beam evaporation. Figure 2.2-3 shows the electron beam evaporation system used and a schematic diagram of an electron beam evaporation system is shown in Figure 2.2-4.

Figure 2.2-3. Edwards 306 evaporation system with the control panel on the right and evaporation chamber on the left. The evaporation chamber is maintained by a turbo molecular pump below it.

Figure 2.2-4. Schematic diagram of electron beam evaporation. The magnetic field alters the trajectory of the electron beam, guiding it towards the evaporation material.

Figure 2.2-4 illustrates the working principle of an electron beam evaporation system. The electrons are generated by thermionic emission from a filament and accelerated by high voltage. A magnetic field bends the electron beam upwards and into the evaporation material. The oxides used in this thesis such as SiO_2 and MgO sublime when heated with the electron beam, allowing epitaxial growth with a controlled growth rate of $\sim 1 \times 10^{-3}$ nm/s.^{9, 10} The pressure in the evaporation chamber is maintained by turbo molecular pump and the base pressure for the evaporation is $< 5 \times 10^{-7}$ Torr

2.3. Device fabrication

Thermally oxidized silicon wafers with SiO_2 of 200 – 300 nm were used. The wafers were cleaved into squares of $\sim 1 \times 1 \text{ cm}^2$ before cleaning. The samples are soaked in an

ultrasonic acetone bath for 15 mins before rinsing with Iso-Propyl Alcohol (IPA) and a 15 mins ultrasonic IPA bath. The samples are blown dry with N₂ gas to remove any IPA left on the sample. The following section will briefly explore electron beam lithography (EBL), which was used in this thesis for device patterning.

2.3.1. Electron beam lithography

The EBL system used in this thesis is the Raith e-Line system which is shown in Figure 2.3-1. The electrons are accelerated by high voltages ranging from 10 to 20 keV. These electrons are collimated into a beam using electromagnetic lenses. A schematic diagram of an EBL system is shown in Figure 2.3-2.

Figure 2.3-1. EBL system with control tower by Raith. EBL system is in yellow light clean room, image processing was carried out to remove the yellow light.

Figure 2.3-2. Schematic diagram illustrating the main EBL system components during electron beam exposure. The desired feature to be patterned is sent from the computer to deflection coil and sample stage driver to trace the feature using a raster scan.

There are two main types of resist used for EBL lithography, the positive and negative resist. These resists are spin coated at 2000-8000 RPM for 60 s to obtain a thin and flat resist layer on the sample. After spin coating, the samples are baked on a hotplate at to remove excess water from the resist. The positive and negative resists were used for device patterning and their steps are detailed in Figure 2.3-3 and Figure 2.3-4 for positive and negative resist fabrication respectively.

Figure 2.3-3. Schematic diagram illustrating the process flow of EBL using a negative resist.

Figure 2.3-3 shows the EBL process using a negative resist by etching the unexposed regions with ion milling. The recipe for the lithography process is briefly explained as follows.

The thin film to be patterned is deposited onto the substrate using magnetron sputtering. The film is then spin coated with negative resist, MaN2401 at 2000-6000 RPM.¹¹ The substrate with the coated photoresist is then baked on a hot plate at 100°C for 1 minute. Next, the sample is exposed to electron beams using the Raith EBL system. The unexposed regions are dissolved away by soaking and gently stirring in a developer solution, MaD525 for 90 s.¹² This is followed by rinsing the sample in de-ionized water before blow drying it with N_2 gas. To transfer the pattern of the resist to the thin film, the sample undergoes ion beam milling which is detailed in Section 2.3.2. The resist acts as a mask, protecting the thin film beneath it from being etched. Finally, to ensure that all the resist is then stripped off, the sample is placed into an ultrasonic chemical bath of mr-Rem 400¹³ for 6 hours before rinsing with IPA and blow drying with N_2 gas.

Figure 2.3-4. Schematic diagram illustrating the process flow of EBL using a positive resist.

A positive photoresist and a lift-off technique is also employed for device fabrication. Figure 2.3-4 shows the schematic diagrams for the lithography steps involved. The recipe for the lithography process using a positive resist is briefly explained as follows.

First, the substrate to be patterned is spin coated with PMMA 950K ¹⁴ positive photoresist at 4000 RPM to obtain a resist thickness of ~ 200 nm. This is followed by a 120°C bake for 3 minutes on a hotplate. After the resist hardens, the sample is exposed to electron beams using the Raith EBL. The regions exposed to the electron beam become significantly more soluble to the developer solution of methyl isobutyl ketone (MIBK):IPA with a 1:3 ratio. The sample is rinsed in the MIBK:IPA solution for 30 s before a rinse with only IPA for another 30 s. Before of the deposition of the required thin film on the patterned sample, reverse sputtering using RF bias is performed for 60 s at 30 W RF power to remove any oxides or remnant material. Lastly, the resist along with the deposited material on top of the resist is removed by lift-off using an acetone bath under ultrasonic followed by IPA rinse and blow drying with N_2 gas.

2.3.2. Ion milling

Ion milling is a dry etching technique where ionized gas bombards the surface of the sample to remove material from it, thereby etching the material ^{15, 16}. In this thesis, the ion-milling system used is AJA ATC-2020-IM,¹⁷ as shown in Figure 2.3-5, where the exterior with the control tower is shown in 2.3-5(a) and the interior which shows the sample stage is shown in 2.3-5(b). A turbomolecular pump brings the milling chamber to a base pressure of 10^{-8} Torr with a working pressure of 10^{-3} Torr. The Ar flow rate into the milling chamber is maintained at 12 sccm and power of etching is kept at 64 W.

The angle of the stage which the sample is attached on can be adjusted manually and is kept 10° to the incident angle from the direction of Ar bombardment. This helps ensure that any re-deposition of the material to the side wall of the etched pattern is kept to a minimum. The stage is also water cooled, which reduces the heating generated during the Ar bombardment of the sample and stage. Despite the cooling stage, the heat generated can still harden the resist, resulting in difficulties during the resist stripping process. An intermittent milling process was employed to allow the stage and sample to cool sufficiently, thereby reducing resist stripping issues.

Figure 2.3-5. (a) AJA ion milling system with control tower. ¹⁷ On the left of the milling chamber, a secondary-ion mass detector is attached which can be used to detect composition of the etched sample surface. (b) Interior of the milling chamber, the rotating

sample stage is highlighted, the samples can be mounted in this position before tilting it to face the back of the milling chamber for the etching process.

2.4. Magnetic measurements

2.4.1. Vibrating sample magnetometer

Vibrating Sample Magnetometer (VSM) is a measurement equipment which determines the magnetic characteristics of a sample with very high precision. It is based on Faraday's law of induction which accounts for the generation of an electromagnetic force due to a change in magnetic flux. This is commonly adapted in VSM systems using a set of pickup coils, whereby the change in flux is amplified by the number of windings in the pickup coils. The pickup coils detect the magnetic flux from the magnetic sample, which can be simply described as follows,

$$\phi_B = \mu_0 (H + M)A \quad (2.4.1)$$

where ϕ_B represents the flux in the coil, μ_0 represents the magnetic permeability, H is the applied magnetic field, M is the magnetization of the sample, and A is the area of the pickup coil. The change in flux can be found by measuring the voltage induced, which can be described simply with,

$$v = -N \frac{dB}{dt} \quad (2.4.2)$$

where v is the induced voltage, and N is the number of turns in the pickup coils. Hence, by measuring the induced voltage from the pickup coil, and with accurate calibration of the applied magnetic field, H . The flux from the magnetization of the sample, M , can be easily found.

The change in flux is achieved by vibrating the magnetic sample near the two pickup coils as schematically shown in Figure 2.4-1. The oscillating driver provides a sinusoidal vibration, which allows the signal to be picked up by a lock-in amplifier. This enables magnetic signals in the range of 10^{-9} emu to be detected by the Lakeshore 8600 VSM system used in this thesis work. Figure 2.4-2 displays the VSM system used.¹⁸ As with most VSM systems, the pickup coils are placed at the pole caps of the electromagnets. Therefore, the hysteresis curve obtained using this VSM system shows the magnetization in the longitudinal configuration as predicted by the Stoner-Wohlfarth model.

Figure 2.4-1. Schematic diagram illustrating the working principles of the VSM system.

Figure 2.4-2. Lakeshore VSM system for magnetization characterization.¹⁸

2.4.2. Magneto-Optic Kerr effect

Magneto-optical Kerr effect (MOKE) provides another technique to characterize the magnetic properties of the PMA thin films. This technique is based on the Kerr effect, where polarised light is reflected light off the magnetic material to be elliptically polarized with the major axis of the ellipse slightly rotated from the polarized plane of the incident light. This rotation is known as the Kerr rotation and it can be detected using the setup as shown in Figure 2.4-3.

Figure 2.4-3. Schematic diagram of a polar MOKE setup for magnetization characterization of samples with PMA. The electromagnet pole cap has a hole in the middle to allow the light to pass through it.

Figure 2.4-3 shows a polar MOKE setup, where the incident angle of the light is kept to a small degree. The light is polarized before it is reflected by the thin film sample. The Kerr rotation from the reflected light is detected from the intensity detected by the photodetector. The beam splitter and lens help to direct the light path as shown in Figure 2.4-3.

Figure 2.4-3. Polar MOKE setup with optical chopper and lock-in amplifier to help reduce noise. Inset shows a Kerr microscopy setup which also uses the principles of MOKE.

Although the MOKE does not measure the absolute values of the magnetic moments from the device like a VSM, it has several advantages over VSM, such as a depth sensitive characterization by tuning the penetration depth of the laser beam.^{19,20} The signal to noise ratio can also be easily increased by increasing the light intensity. Additionally, a camera in place of the photodetector enables device level characterization by detecting the change in local magnetization. This allows the direct observation of the domains.²¹⁻²⁴

The observation of the magnetization is achieved by differential imaging, where the image of the magnetic device is first taken after saturation. After magnetization switching, the image of the device is taken again, and the differential image shows the Kerr contrast. Figure 2.4-4 shows the Kerr imaging of a Hall cross device with opposite magnetization states, where the Hall cross is fully magnetized in opposite out-of-plane directions.

Figure 2.4-4. Differential imaging from a Kerr microscopy system illustrating the Kerr contrast of a Hall cross device with PMA.

2.5. Electrical measurements

2.5.1. Anomalous Hall effect

The anomalous Hall effect (AHE) is the ferro-metallic counterpart to the non-magnetic Hall effect. Both effects are discovered by Edwin Hall, and the AHE is discovered to produce a much larger effect.²⁵⁻²⁷ Despite its wide use in magnetic characterization, its fundamental principles are seldom delved into. Ostensibly, the AHE seems like the ordinary Hall effect due to the magnetization of magnetic material forming a magnetic field. However, experimental results show that this assumption results in a magnetization difference of up to 10^2 from the ordinary Hall effect. This difference became known as the anomalous factor.²⁸

As a result, an intrinsic and extrinsic effect has been proposed, whereby the intrinsic contribution derives from the band states, independent of scattering effects.^{28,29} Conversely the extrinsic effect is based on the scattering effects of the conduction electrons with the orbitals to produce the AHE phenomena.^{30,31}

The effects dominate with varying degrees for different regimes. In this thesis, the ferro-metallic materials at room temperature belong to the regime of high conductivity materials ($\sigma > 10^6 \Omega^{-1} \cdot \text{cm}^{-1}$), where skew scattering dominates.^{25, 32, 33} A schematic

diagram is shown in Figure 2.5-1(b) to illustrate the skew scattering phenomena, where an asymmetric scattering of the electrons occurs due to the spin-orbit interaction of the impurity. However, the origins of AHE is still very much in debate due to the nature of the many-body problem especially with the multitude of ferro-metallic materials with varying composition and stack structures where the AHE phenomena can be observed from.²⁵

Figure 2.5-1. Schematic diagram of AHE mechanism. (a) Intrinsic contribution and (b) skew scattering effect that is the dominant effect in magnetic materials with high conductivity.^{25, 32, 33}

The veil that shrouds the exact mechanism of the AHE does not hinder the use of AHE as a method to characterize the magnetization. The hysteresis curves obtained from AHE measurements on Hall cross devices can be simply be modelled using the Stoner-Wohlfarth model introduced in Section 1.2. These measurements can extract the coercivity, anisotropy energy and electric field control modulation of the Hall cross devices. The AHE measurements form the main bulk of the electric measurements in this thesis.

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3. Bulk effect of electric field control on multi-layered Pt/Co

3.1. Motivation

The Thomas-Fermi model describes the electric field screening effect. In metals, this yields a screening length of $\sim <10^{-1}$ nm, thereby limiting the VCMA effect to be predominately an interfacial effect.¹⁻³ As such, research on electric field control of ferro-metallic materials mainly focus on thin ferro-metallic layers with special emphasis on materials that have interfacial PMA such as a CoFeB/MgO heterostructures.^{4,5} However, recently researchers have begun to abandon this preconception and investigate on the possibility of electric field control on thicker magnetic layers. Studies with increasing thickness of the ferro-metallic layer have revealed that there might be a higher order bulk component to the VCMA effect.^{6,7} At the same time, electric field control has also been successfully demonstrated across heavy metals such as Pd and Pt on a Co layer due to the proximity induced magnetism (PIM) effect, causing the heavy metal to be magnetically polarized.⁸⁻¹⁰ These recent discoveries opens a myriad of possibilities for magnetic materials previously thought to be incompatible for electric field control. These include bulk and multi-layer magnetic materials, which provides enhanced performance for spintronic devices such as having a higher thermal stability.

The work presented in this chapter demonstrates that electric field control is not only limited to single layer thin film structures. This enables a much greater flexibility for material engineering to improve the performance of any spintronic device using the electric field effect. In this chapter, the thermal stability is shown to improve significantly while maintaining the utility of electric field control.

3.2. Magnetic Characterization

Pt/Co based PMA thin films were grown using magnetron sputtering, the stack information of the thin films is as shown in Figure 3.2-1 where the numbers in parenthesis represent the nominal thickness in nanometre. The convention where the material on the left starts from substrate is used in this thesis. The thin films are characterized using a VSM and results are also presented in Figure 3.2-1. The square hystereses indicate that the easy axis of the ferro-metallic thin films are out-of-plane. The coercivities (H_c) of the double Pt/Co stacks is larger than a single Pt/Co stack, and a thicker Pt spacer thickness also increases the H_c of the thin film. The increase in H_c is a good indication that the Pt spacer increases the PMA strength of the material. The magnetic saturation (M_s) of the thin films can also be extracted from the hysteresis loop and is shown in the Table 3.2-1 where the numbers in parentheses represent the thickness in nm. The magnetic volume is estimated with consideration for the magnetic dead layer as shown in Figure 3.2-2. The magnetic dead layers for the various interfacial layers are also obtained using VSM. The Co thickness dependence on the M_s indicates the magnetic dead layer caused by the capping layer. The Co/MgO interface has a ~ 0.36 nm dead layer while the Co/Pt (t) interface has a dead layer of negligible thickness. Table 3.2-1 shows the results of the magnetic characterization of the thin film stacks obtained from VSM measurement.

Figure 3.2-1. VSM measurement of the various PMA Pt/Co stack structure. The single Pt/Co stack shows the lowest coercivity and magnetic saturation.

Figure 3.2-2. Magnetic dead layer approximation using VSM measurement.

3.3. Electric field modulation of double Pt/Co stack

The Pt/Co based PMA thin films were fabricated into 3 μm wide Hall crosses for Hall resistance measurement as schematically shown in Figure 3.3-1. The normalized Hall resistance (R_H^n) dependence on the applied out-of-plane magnetic field ($H_\perp$) and in-plane magnetic field longitudinal to the applied current ($H_\parallel$) is shown in Figure 3.2-2(a), (b) and (c), (d) respectively. Figure 3.2-2(a) and (c) shows the R_H^n signal from the single Pt/Co stack while Figure 3.2-2(c) and (d) shows the R_H^n signal from the double Pt/Co stack.

Figure 3.3-1. Schematic diagram detailing the electrical setup during the Hall resistance measurement with a sweeping magnetic field.

Figure 3.3-2. Hall resistance measurement with applied electric field, where (a) and (b) shows the out-of-plane magnetic field sweep for a single and double Pt/Co stack respectively. (c) and (d) shows the in-plane Hall resistance measurement from the in-plane magnetic field sweep a single and double Pt/Co stack respectively.

Figure 3.3-2 (a) (b) shows a square shaped hysteresis curve indicating that the PMA characteristic of the thin film was retained after the fabrication process. The overall increase in coercivity (H_c) for the fabricated device compared to the thin film counterparts can be attributed to the reduction of domain nucleation from surface irregularities or physical defects.¹¹ In both the single and double Pt/Co stacks, the H_c was modulated by the applied electric field, where a positive/negative (± 250 MV/m) electric field reduces/increases the H_c . This allows the magnetization of the Hall crosses to switch along its easy axis with different magnetic field strength by varying the applied electric field.

In Figure 3.2-2(c), the R_H^n dependence on $H_{//}$ shows that at a high $H_{//}$ (~ 14 kOe) as indicated as part 1 in the figure, there is insignificant R_H as the magnetization of the Hall cross was saturated along the direction of the $H_{//}$. As the $H_{//}$ reduces, the magnetization goes towards its out-of-plane easy axis, resulting in an increase in R_H^n . When the $H_{//}$ increases in the opposite direction, the R_H^n drops as the magnetization begins to tilt towards the direction of $H_{//}$ as shown in part 2. The sharp drop observed at ~ 5 kOe as indicated as part 3, occurs due to the small misalignment between the plane of the device and the $H_{//}$. This results in a small amount of $H_{\perp}$ during the application of a large $H_{//}$, causing the magnetization to switch along its easy out-of-plane axis before full saturation along the $H_{//}$ as finalized in part 4. The process repeats itself from the opposite direction, leading towards the magnetization reversal as shown in part 5 to 8. The magnetization dynamics are similar for the double Pt/Co stack as shown in Figure 3.2-2 (d), albeit with a larger magnetic field required for saturation. Both single and double Pt/Co stack exhibit lower/higher saturation field when a positive/negative electric field is applied. This agrees well with the change in H_c coercivity observed in (a) and (b) as well as similar experiments conducted with Pt/Co based PMA structures.^{9, 12}

3.4. K_{eff}/E comparison with thermal stability of multi-layered Pt/Co

From the R_H^n measurements obtained in Figure 3.3-2 (c) and (d), the normalized in-plane component of magnetization ($M_{||}^n$) can be calculated by using the equation,

$$M_{||} = \sin(\cos^{-1}(R_H^n)) \quad (3.4.1)$$

Hence, the $M_{||}^n$ dependence on $H_{||}$ can be extracted from Figure 3.3-2 (c) and (d), to be presented as shown in Figure 3.4-1 and 3.4-2. From the obtained $M_{||}^n$ dependence to the $H_{||}$, the effective anisotropy strength (K_{eff}) of the Pt/Co stacks are calculated by using the following integral,

$$K_{eff} = M_s \int H_{||} dM_{||}^n \quad (3.4.2)$$

where the M_s is obtained from thin film VSM measurement. Figure 3.4-3 shows the K_{eff} dependence on the electric field for all the Pt/Co Hall crosses. The K_{eff} of all the Pt/Co stacks shows a linear relation to the electric field used in this work, where a negative/positive electric field results in an increase/decrease in K_{eff} . The sign of the gradient observed agrees to those demonstrated for similar Pt/Co^{9, 12} and CoFeB/Mgo¹³ PMA structures, however, FePd based structures show an opposite sign.¹⁴ The gradient of the fitted lines represents the electric field modulation of the K_{eff} and is taken to be the magnetic anisotropy modulation efficiency, K_{eff}/E , the K_{eff} dependence on the applied electric field for all the Pt/Co stacks are presented in Figure 3.4-3.

Figure 3.4-1. Normalized in-plane component of the Hall resistant for a single Pt/Co layer with ± 250 MV/m applied.

Figure 3.4-2. Normalized in-plane component of the Hall resistant for a double Pt/Co layer with ± 250 MV/m applied.

Figure 3.4-3. The K_{eff} dependence on the applied electric field for the single Pt/Co and double Pt/Co stacks with varying Pt spacer layers.

Figure 3.4-4 shows the K_{eff}/E for the double Co layers as a percentage with respect to the K_{eff}/E obtained from the single Pt/Co stack. By considering the effective magnetic thickness between the top and bottom Co layers, the K_{eff}/E obtained from the R_H^n measurements would be ~32% of the single Pt/Co stack if the magnetic anisotropy modulation only occurs for the top Co layer without any coupling effects.^{15, 16} However, as shown in Figure 3.4-4, the K_{eff}/E of the double Pt/Co stack is ~80% of the single Pt/Co stack for all Pt spacer thicknesses, suggesting that the magnetic anisotropy modulation occurs for both the top and bottom Co layers. This can be attributed to the PIM in Pt due to exchange interactions with the Co, where the growth of Pt on Co has a larger PIM than Co on Pt.¹⁷ There is also a marginal decrease in K_{eff}/E with thicker Pt spacer layer resulting in a change of ~8% between Pt spacer

thickness of 0.4 nm and 1.6 nm. This decrease may not constitute a trend as the change is smaller than the error margin.

Figure 3.4-4 also compares the thermal stability of the Pt/Co stacks. The thermal stability factor, Δ , where $\Delta = K_{eff}V / k_B T$ and K_{eff} , V , k_B and T represents the effective anisotropy energy, volume of magnetic material, Boltzmann constant and temperature in Kelvin respectively. The Δ increases with an additional layer of Pt/Co due to the larger volume. The combined effect of V and K_{eff} results in an increase in thermal stability, allowing double Pt/Co stack to retain its magnetic properties better than single layer Pt/Co stack after exposure to high temperatures.¹⁸ Since the thickness of the Co layer is proportional to V , a comparison of the thermal stability between the single and double Pt/Co stacks can be made for the equally sized Hall cross devices used in the conducted experiments. The K_{eff} of the Pt/Co stacks were obtained from the R_H measurement done at 0 MV/m, as shown in Figure 3.4-3. The large difference in K_{eff} between the single and double Pt/Co stacks can be attributed to the additional interfacial anisotropy from the Pt(111) spacer layer which encourages a larger magneto crystalline anisotropy in the Co layers.¹⁹⁻²² Comparison between the double Pt/Co stacks in Figure 3.4-4 is shown as a percentage with respect to the single Pt/Co stack. The boost in K_{eff} of the material along with the increase in magnetic thickness due to the additional Co layer, allowed the double Pt/Co stacks to have a thermal stability of ~ 3.9 to ~ 5.2 times larger than the thermal stability of a single Pt/Co stack. As the Co thickness is equal across the double Pt/Co stacks, the increase in thermal stability with a thicker Pt spacer layer is solely due to the increase in K_{eff} , where the interfacial anisotropy is greatly enhanced from a Pt spacer thickness of 0.4 nm to 0.8 nm, but starts saturating from 0.8 to 1.6 nm.¹⁶ The change in Pt spacer thickness also show no oscillating behaviour of the K_{eff} , implying that there is no RKKY coupling-like

effect which can arise due to a non-magnetic layer between two ferromagnetic layer.^{15, 23} This supports and extends the finding for thicker Pt layers sandwiched by ferro-metallic materials.

24

Figure 3.4-4. The thermal stability and anisotropy modulation efficiency for the double Pt/Co stack is shown as a percentage of the single Pt/Co stack. The table inset shows the values presented on the main figure.

3.5. Micromagnetic validation

To validate that the anisotropy change occurs in both the top and bottom Co layer, micromagnetic simulation is used to replicate the experimental procedures. By considering the magnetic dead layer, the top and bottom Co layers of the double Pt/Co stack have an average thickness of ~ 0.36 nm and ~ 0.95 nm and the thickness of the single Co layer is ~ 0.64 nm. Hence, a mesh of 0.2 nm^3 was chosen, where the Co thickness are rounded by to the nearest 0.2 nm as shown in the schematic diagrams of Figure 3.5-1. The parameters used in the

simulation were obtained from an average of the experimental results, with $M_s = 658$ emu/cc and $K_{anis} = 675$ kJ/m³. K_{anis} was obtained from an averaged K_{eff} value of 403 kJ/m³ by using the equation as follows,

$$K_{eff} = K_{anis} - \frac{\mu_0}{2} M_s^2 \quad (3.5.1)$$

where μ_0 represents the permeability of free space and $\frac{\mu_0}{2} M_s^2$ represents the demagnetization term.

The $M_{||}^n$ dependence on $H_{||}$ with varying electric field was obtained by altering the uniaxial anisotropy constant, K_{anis} . The results are shown in Figure 3.5-1 (a) and (b), and they closely resemble the experimental results in Figure 3(c) and (d) from the previous section. In Figure 3.5-1(a), a single Co layer was used, where the K_{anis} of the 0.6 nm Co is varied from 80% to 120% of its original value. In Figure 3.3-5(b), a double Co layer was used, where only the K_{anis} of the top 0.4 nm Co layer is varied between 80% to 120%. The distinct 2nd order arising from the double Co layers is absent in Figure 3.5-1(a) and the experimental results, suggesting that the top and bottom Co layers in the experimental double Pt/Co stacks are strongly coupled, causing it to behave more like a single magnetic film rather than two magnetic films separated by a non-magnetic spacer. This can be attributed to the PIM of the Pt spacer layer which enables the strong interaction between the top and bottom Co layers.

Figure 3.5-1. Normalized in-plane magnetization with varying anisotropy energy for (a) single Pt/Co stack and (b) double Pt/Co stack. The anisotropy energy is only varied for the top Co layer in (b).

In Figure 3.5-1, the simulations are repeated with varying Pt spacer layer and the percentage of K_{eff}/E is calculated in the same manner as shown for the experimental data in

Figure 3.5-2. The K_{eff} / E of the double Pt/Co stacks is only $\sim 29\%$ of the single Pt/Co stack as compared to the $\sim 80\%$ obtained in the experimental results. This strongly suggests that the experimental results indicate that both the top and bottom Co layers are modulated by the electric field. Since the demagnetization field coupling between the top and bottom Co layers is included in the simulations, this eliminates demagnetization field as the dominant interaction between the top and bottom Co layers. The demagnetization field from the bottom 1 nm Co layer is also shown to be $\sim 300\text{-}250$ Oe, which is insignificant compared to the anisotropy field of ~ 20 kOe for the top Co layer.

Figure 3.5-2. The K_{eff} / E dependence on the Pt spacer thickness is shown. The low stray field from the bottom Co layer results in the similar K_{eff} / E for Pt spacer thickness 0 – 6 nm.

3.6. Summary

Electric field control was successfully demonstrated for double Pt/Co stacks. The results obtained in this work show that the electric field control is not just an interfacial effect at the ferro-metallic/insulator interface. The electric field modulation of the bottom Co layer can be attributed to the PIM of the Pt spacer layer, allowing the double Pt/Co structures to have <20% decrease in K_{eff}/E as compared to a single Co layer. The experiment was replicated using micromagnetic simulation and the K_{eff}/E from the simulation results support PIM of the Pt space layer by eliminating demagnetization field as a coupling mechanism between the top and bottom Co layer. The minimal decrease of K_{eff}/E coincides with a huge ~390-520% increase in thermal stability for double Pt/Co structures. This increase can be credited to the additional magnetic volume along with the increase in K_{eff} . The results in this work establish the possibility of electric field control on multilayer ferro-metallic materials, leading towards energy efficient control of spintronic devices with high thermal stability.

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4. Magneto-ionic effect on Pt/Co/SiO₂ Hall cross device using insulating polymer

4.1. Motivation

An electric field can be applied on to a ferro-metallic material by applying a gate voltage across an insulating layer with the ferro-metallic material as ground. In the current state of art, the insulating layer used in electric field control devices consist of high- κ dielectrics such as hafnium oxide,^{1, 2} zirconium oxide,^{3, 4} gadolinium oxide,⁵⁻⁷ and magnesium oxide.^{8, 9} The depositions of these materials are usually carried out using atomic layer deposition, molecular beam epitaxial or sputtering deposition techniques. All these methods require a relatively long deposition duration to ensure high quality insulating layer of a suitable thickness (~30-50 nm) to avoid dielectric breakdown due to physical defects. Another insulating technique is based on using a liquid electrolyte.¹⁰⁻¹² However, to make a practical device using liquid electrolyte, specialized techniques are required to encapsulate the liquid electrolyte. Alternatively, the freezing temperature of the electrolyte must be reached to make a practical device such that the liquid does not flow.

In this work, the use of an MaN2401, an insulating polymer (IP) for electric field control was investigated. The IP was chosen to be the insulating layer as its viscosity ensures little to no major defects within the layer. Furthermore, after baking, the IP does not require encapsulation to remain in place. The IP can be spin coated onto a device, which is a much faster process than any conventional physical or chemical vapour deposition methods, requiring only minutes instead of hours. A polymer layer also allows the possibility of future spintronic devices on flexible substrates.

4.2. Effect of Co thickness on coercivity of magnetic material

A trilayer stack consisting of Ta (3 nm)/Pt (3 nm)/Co (t nm) was grown on thermally oxidised silicon dioxide substrates by magnetron sputtering with a base pressure of $< 3 \times 10^{-8}$ Torr. Nominal thicknesses of Co were deposited ranging from 0.8 to 2.3 nm. As a break in vacuum was required, the Co layer was allowed to naturally oxidise at room temperature for ~ 10 min before it was capped with 5 nm of SiO₂ deposited by electron-beam evaporation.

Measurements carried out using VSM with a sweeping magnetic field out-of-plane to the thin film show, sharp coherent switching of the thin films. This indicates that the Ta/Pt/Co/SiO₂ thin films exhibit PMA. The hysteresis results from the VSM measurements are shown in Figure 4.2-1, where Figure 4.2-1(a) is for Co thickness that is ≤ 2 nm and Figure 4.2-1(b) is for Co thickness that is ≥ 2 nm. It is evident that H_c increases with Co thickness until it reaches 2 nm. When the Co thickness is thicker than 2 nm, the H_c reduces significantly due to the in-plane magnetic anisotropy from the thick Co layer after the *fcc/hcp* growth.¹³ The in-plane magnetic anisotropy also results in the gradual magnetization switching observed in thin films with Co thickness of 2.2 and 2.3 nm. From VSM measurements, the H_c dependence on Co thickness is plotted in Figure 4.2-2, where H_c of the thin film peaks at a Co thickness of 2 nm before decreasing sharply. The inset shows the stack structure of the thin film.

Figure 4.2-1. VSM measurement for Ta/Pt/Co/SiO₂ thin films with varying Co thicknesses, where (a) Co thickness < 2 nm and (b) Co thickness $\geq$ 2 nm.

Figure 4.2-2. Coercivity dependence on the Co thickness for the PMA thin film stack extracted from Figure 4.2-1.

4.3. Quantification of the Magneto-ionic effect

The thin film structure containing 0.9 nm of Co was patterned into Hall cross devices using EBL. Figure 4.3-1 shows a schematic diagram of the measurement setup with the fabricated device. The inset shows the Hall cross under scanning electron microscopy before the gate electrode was deposited. The insulating polymer (IP) used in this work is MicroChem MaN2401, a negative polymer resist for electron beam lithography. All the IP used in this work were baked at 100 °C for 1 min after spin coating. An insulating layer of MgO was deposited to smoothen the step height from the Hall cross. It should be noted that the MgO is not on top of the Hall cross and therefore plays no active role in the magneto-ionic effect. The electrodes for the gate voltage and at the ends of the Hall cross consist of Ta (5 nm)/ Cu (100 nm)/ Au (5

nm) and were grown by using magnetron sputtering. All transport measurements done in this work were carried out at room temperature.

Figure 4.3-1. Schematic diagram illustrating the electrical setup during electrical measurement. Inset shows the Hall cross device using scanning electron microscopy.

4.3.1. Coercivity measurement

Hall resistance (R_H) measurements were carried out with a sweeping out-of-plane magnetic field. The device has a 152 nm thick layer of IP and was measured in its pristine state without any gate voltage and was measured again after applying ± 12.5 V (± 0.82 MV/cm) for 2000 s. A low current density ($< 1 \times 10^{10}$ A/m²) was used in all the R_H measurements in this work to avoid Joule heating which might affect the H_c of the device. The results of the R_H measurements are shown in Figure 4.3-2, where an applied electric field of ± 82 MV/m results

in the H_c of the device becoming larger (smaller) when +82 MV/m (−82 MV/m) gate voltage was applied. This results in a percentage change of +68% and −47% of the H_c for the negative and positive gate voltage respectively. The R_H measurements were carried out at zero gate voltage after the application of the electric field, demonstrating the nonvolatility of the device which lasted for at least ~48 hours in room temperature.

Figure 4.3-2. Hall resistance measurements illustrating the change in coercivity with an applied electric field of ± 82 MV/m.

To validate that the magneto-ionic effect modulates the thickness of the magnetic layer, a comparison between the H_c change due to electric field control and Co thickness was made. To find the effective magnetic Co thickness, the magnetic dead layer of the Pt/Co/SiO₂ was obtained using VSM measurements as shown in Figure 4.3-3, which indicates that the thin films have a magnetic dead layer of $0.47 \text{ nm} \pm 0.03 \text{ nm}$. This was subtracted from the nominal

Co thickness to provide the effective Co thickness. Selected thin films of various Co thicknesses from Figure 4.2-2 were fabricated into 500 nm width Hall crosses and R_H measurements were carried out with the results shown in Figure 4.3-4. The overall increase of the H_c compared to their thin films counterparts can be attributed to the lack of domain nucleation from surface irregularities or physical defects.¹⁴

Figure 4.3-3. Magnetic dead layer approximation from VSM measurements. The red line represents the linear fit used to extract the magnetic dead layer of ~0.47 nm.

Figure 4.3-4. Normalized Hall resistance measurement illustrating the coercivity dependence on the Co thickness.

The H_c from Figure 4.3-4 is compared with the H_c modulated *via* electric field in Figure 4.3-5. The range of H_c shared by the devices is demarcated by dotted lines and the corresponding range of effective Co thickness is demarcated by dashed lines. This suggests that the change in H_c of the device due to electric field control is due to a Co thickness range of ~ 0.32 to ~ 0.75 nm. By comparing the H_c , it supports and directly demonstrates the mechanism of the magneto-ionic effect of modulating the Co thickness. It is noted that due to experimental limits, the thinnest effective Co, with PMA is ~ 0.32 nm, which resulted in a device H_c of 76 Oe. However, since the smallest H_c obtained by applying a negative electric field is 61 Oe, it is likely that the effective Co thickness is thinner than 0.32 nm after the application of the negative electric field.

Figure 4.3-5. Direct comparison of coercivity from a device ($\text{Co} = 0.9\text{nm}$) under gate voltage and from devices with various Co thicknesses. The error bars arise from the instrument error of the Hall probe during measurement.

4.3.2. Resistivity measurement

To further validate the change in Co thickness caused by the magneto-ionic effect, Figure 4.3-6 shows the resistance of the nanowire under various electric field. When negative electric field is applied, the resistance is higher, indicating that the nanowire thickness decreases. Similarly, when positive electric field is applied, the resistance decreases. Furthermore, the increase in resistance saturates after an electric field of -49 MV/m , which is mirrored by the saturation in H_c as observed in Figure 4.3-5. This can also be seen when a positive electric field is applied. In Figure 4.3-6, the resistance dependence on the Co thickness obtained from Figure 4.3-5 shows a linear relation, providing further evidence for the mechanism of the magneto-ionic effect.

Figure 4.3-6. Resistance of nanowire changing with the applied electric field. The Co thickness is obtained from the comparison in Figure 4.3-4.

4.4. Polymer thickness contribution to the Magneto-ionic effect

The thickness of the insulating polymer (IP) on the devices are determined with direct step height measurement also using atomic force microscopy. Due to the nonvolatility of its H_c , dynamic studies were carried out from a reference state. For the R_H measurement shown in Figure 4.4-1(a), the reference state is reached when a gate voltage of -15 V (-0.85 MV/cm) was applied for 2000 seconds. During the R_H measurement, a gate voltage of opposite polarity, $+15$ V ($+0.85$ MV/cm) is applied. This measurement process is illustrated as a flowchart in Figure 4.4-1(b) as a reference for the measurements done to obtain the results in Figure 4.4-1(a). Each R_H measurements was carried out after a gate voltage of $+15$ V ($+0.85$ MV/cm) was applied for 500 seconds across a 177 nm thick IP layer. This procedure was repeated for 4 times, up to a total culmulative period of 2000 seconds during which the gate voltage was

applied. The total cumulative period is not inclusive of the time taken during the R_H measurements with the sweeping magnetic field.

Figure 4.4-1. (a) Anomalous Hall effect measurement with applied gate voltage of +15 V ($+0.85 \text{ MV cm}^{-1}$) from 0 to 2000 s on a 177nm thick IP. (b) Flowchart of measurement process in figure (a).

The thickness of the IP was varied by controlling the rotation speed during the spin coating process (2000 to 7000 RPM). In Figure 4.4-2 (a) and 4.4-2 (b) the electric field was applied across a IP thickness of 177nm, the H_c dependence at the applied electric fields between $\pm 0.85 \text{ MV/cm}$ follows a linear trend. This linear behaviour of the H_c change occurs for IP thickness between 157 to 233 nm for all the gate voltages used in this work. The solid black line in Figures 4.4-2 and 4.4-3 provides a visual aid of the linear fitting. However, with a thinner IP layer of 152 nm and at an electric field of ± 0.49 , ± 0.66 and $\pm 0.82 \text{ MV/cm}$, the H_c appears

to reach a saturation state where the $\frac{dH_c}{dt}$ is significantly different. In Figure 4.4-3 (a) and 4.4-3 (b), this saturating behaviour is highlighted in blue, indicating that a shorter time is required to reach saturation with a thinner IP layer. The black dotted line in Figure 4.4-3 provides a visual aid which extrapolates the linear fitting from the non-saturated regime, emphasising the difference in $\frac{dH_c}{dt}$ between the saturated and non-saturated regime. The saturation of $\frac{dH_c}{dt}$ demonstrates that a thinner IP layer is more efficient due to the shorter distance involved. The $\frac{dH_c}{dt}$ at various electric field up to ± 0.96 MV/cm was obtained for the various IP thickness and the results are shown in Figure 4.4-4.

Figure 4.4-2. Coercivity of Hall cross with varying applied electric field, values are offset arbitrarily in the y axis for visual representation. (a) Varying positive electric field with 177 nm of insulating polymer. (b) Varying negative electric field with 177 nm of insulating polymer.

Figure 4.4-3. Coercivity of Hall cross with varying applied electric field, values are offset in the y axis arbitrarily for visual representation. (a) Varying positive electric field with 152 nm of insulating polymer. (b) Varying negative electric field with 152 nm of insulating polymer.

In Figure 4.4-4(a), the $\frac{dH_c}{dt}$ of the devices with IP of thickness 157 to 233 nm was obtained with a linear fit. For the device with IP thickness of 152 nm, the $\frac{dH_c}{dt}$ was extracted with the linear fit from only the non-saturated regime. For all the of IP thicknesses, the results of the $\frac{dH_c}{dt}$ dependence on the electric field also shows a linear relation, and the gradient of linear fit is shown in Figure 4.4-4(b). As the thickness of the IP gets reduced, the effect that the electric field has on the $\frac{dH_c}{dt}$ becomes greater. This trend of increasing $\frac{\partial^2 H_c}{\partial t \partial E}$ with decreasing IP thickness agrees with the observations from Figure 4.4-3. The increased $\frac{dH_c}{dt}$ due to the thinner IP layer results in a faster saturation of the magneto-ionic effect. Thinner IP layers allows the possibility of optimizing spintronics devices using IP as an insulating layer.

Figure 4.4-4. (a) The obtained $\frac{\partial H_c}{\partial t}$ against electric field for the various polymer thicknesses from the linear regime as shown in Figure 4.4-3 and 4.4-4. (b) $\frac{\partial H_c}{\partial t \partial E}$ is obtained from the gradient of the inset and plotted against the polymer thickness of the Hall cross device. The error bar reflects the standard error from the fitting of the inset. The value of $\frac{\partial H_c}{\partial t \partial E}$ indicates how the electric field affects the H_c of the Hall cross device.

4.5. Summary

In conclusion, the viability of using an IP for electric field control has been demonstrated on 500 nm width Hall crosses consisting of a Ta/Pt/Co/SiO₂. The magneto-ionic effect is reversible and has non-volatile retention of H_c . The applied electric field results in a percentage change in H_c of -47 and $+68\%$ which correlates to a change in Co thickness from 0.3 to 0.75 nm. The transport measurements from the R_H and resistance of the nanoscale device presented in this section supports the mechanisms for the magneto-ionic effect. The dynamics of the H_c is characterized and it is indicative that a thinner layer of polymer results in faster magneto-ionic effect for tuning the H_c of the magnetic layer. Using an IP for electric field control of magnetic material provides an unique approach which ensures low physically defect and also has a higher throughput compared to conventional insulation deposition approaches. Furthermore, the usage of an IP allows the possibility of future spintronic devices on flexible substrates.

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5. Electric field control for energy efficient domain wall injection

5.1. Motivation

Domain wall (DW) based memory has the potential to be a universal memory, it is a solid state and non-volatile device which can potentially also be fast and cheap. It can potentially offer the density that HDD offers without any drawback due to its mechanical nature such as the slow read and write speed.

DW based memory devices require three basic operations: read, propagate and write. The write operation in domain wall memory devices is conventionally achieved by applying an electrical current into a conductive stripline fabricated on top of the magnetic wire. The applied current generates an Oersted field which causes local magnetization switching at the region around the stripline.¹⁻⁴ The amount of energy required to inject a DW in a PMA magnetic wire can be estimated with $K_{anis} \cdot V$ where K_{anis} refers to magnetic anisotropy energy and V represents the magnetic volume.⁵ However, the thermal stability of a material can be written as, $\Delta = K_{anis} V / k_B T$, where k_B and T represents the Boltzmann constant and temperature respectively. Therefore, while a high K_{anis} is desirable for thermal robustness⁶⁻⁸, such a memory device would require a large amount of energy to inject DWs.

In this chapter, electric field control is employed to modulate the K_{anis} of the magnetic wire, allowing DWs to be injected with a lower current density. The experimental results obtained using Kerr microscopy showed that the electric field modulation was able to reduce the required current density by ~20%.

5.2. Domain wall injection using Hall cross structure

Electric field control requires an insulating interface with the magnetic wire, this is usually achieved by oxide deposition on top the magnetic wire as demonstrated in Chapter 3 and 4. However, the stripline which generates the Oersted field is also conventionally deposited on top of the magnetic wire.¹⁻⁴ The simple solution would be to shift either the stripline or the insulating layer below the magnetic nanowire. However, this would involve complicated fabrication processes that includes planarization to ensure flatness before the deposition of the magnetic film. An uneven surface can give rise to pinning or nucleation sites around the domain injection region which might lead to false positive readings. To avoid unwanted domain wall nucleation, a Hall cross structure is proposed as a DW injection device, where the current is applied across the Hall bar. In order for sufficient current to pass through the Hall bar for significant Oersted field generation, a Ta(3) / [Cu (25) / Ta(1)] $\times$ 4 stack is deposited below the magnetic wire to increase conductivity. The resulting PMA film stack is Ta(3) / [Cu (25) / Ta(1)] $\times$ 4 / Ta(1) / Pt(3) / Co(1) / Pt(0.4), where the number in parenthesis represents the thickness in nanometer. Figure 5.2-1(a) shows a schematic diagram and 5.2-1(b) shows the microscope image of the Hall cross structure designed for DW injection. The HfO_x is the insulating layer and the ITO is the conductive and transparent gate electrode for Kerr microscopy.

Figure 5.2-1. (a) Schematic diagram illustrating the DW injection observed using Kerr imaging and electric measurement. (b) Microscope image of Hall cross device. The white regions are caused by the thin-film interference of the HfO_x .

The Hall cross has a geometrical constriction designed to increase the current density and Oersted field at the injection region. Simulations performed with COMSOL Multiphysics show the current density and Oersted field with the proposed Hall cross structure used in this Chapter. The current density is the largest at the corners of the Hall

cross structure, which in turn gives rise to larger Oersted field at the corners (H_0). A comparison of the proposed structure with two other conventional Hall crosses of different width is shown in Figure 5.2-2. From comparison, the proposed structure is able to generate a larger H_0 than the conventional Hall cross structures. The larger H_0 enables DWs to be injected using a lower J_{input} .

Figure 5.2-2. Current density and Oersted field mapped on to the various Hall cross structure. The J_{input} in the middle Hall crosses of 1.5 μ m is twice of the J_{input} from the other Hall crosses to ensure that the total current (A) in the Hall cross structure is kept constant.

5.3. Electric field modulation of the Pt/Co/Pt stack

The Hall cross structure is also used for Hall resistance (R_H) measurements to demonstrate the electric field modulation on the effective anisotropy energy (K_{eff}) of the material. Figure 5.3-1(a) shows the normalized R_H dependence on the out-of-plane magnetic field with an applied electric field of ± 250 MV/m. This demonstrates that the coercivity (H_c) of the material can be modulated, which reduces the amount of Oersted field required to switch local magnetization. The K_{eff} of the material can be extracted from the normalized R_H measurement with an in-plane magnetic field. This R_H measurement is shown in Figure 5.3-1(b), where the applied magnetic field, $H_x \ll H_k$. The inset shows the VSM measurements of the thin film as deposited to obtain the magnetic saturation of the material.

Figure. 5.3-1. Normalized R_H measurement of the Hall cross device with (a) out-of-plane magnetic field and (b) in-plane magnetic field. The line shows the fitting using Equation (5.3.3). The inset shows the results of the thin film measurement using vibrating sample to obtain the magnetic saturation of the material. (c) K_i dependence on applied electric field. The line shows the linear fit, where the obtained gradient of 59 ± 4 fJ/Vm is the VCMA coefficient.

The normalized R_H measurement can be modelled to be written as follows,

$$R_H^n = \frac{1}{2} \Delta R_H^n \cos(\theta) \quad (5.3.1)$$

where R_H^n is the normalized Hall resistance and ΔR_H^n is the maximum resistance change between the up magnetization and down magnetization state of the R_H^n .

Maclaurin expansion is applied, taking only the first two terms, and since $\Delta R_H^n = 2$, Equation (5.3.1) can be rewritten to be,

$$R_H^n = 1 - \frac{1}{2}\theta^2 = 1 - \frac{H_x^2}{2H_k^2} \quad (5.3.2)$$

This allows H_k to be obtained with a parabolic fitting,

$$R_H^n = aH_x^2 \pm 1 \quad (5.3.3)$$

where $\pm$ refers to the up and down magnetization state. Therefore, the H_k of the material can be found with the following equation,

$$H_k = \pm \sqrt{\frac{1}{2a}} \quad (5.3.4)$$

where a is the parameter from the parabolic fitting. The effective anisotropy of the material can be obtained with the equation,⁹

$$K_{eff} = \frac{1}{2} M_s H_k \quad (5.3.5)$$

where K_{eff} refers to the effective anisotropy energy and M_s refers to the magnetic saturation of the material.

If the change in interfacial anisotropy (K_i) is assumed to be the dominant cause of the anisotropy modulation, this can be modelled as follows,⁹

$$K_{eff} = K_{anis} - \frac{\mu_0}{2} M_s^2 = \frac{K_i}{t} - \frac{\mu_0}{2} M_s^2 \quad (5.3.6)$$

where K_i refers to the interfacial anisotropy, t is the thickness of the magnetic material, and μ_0 refers to the permeability of free space. Therefore, the K_i dependence on electric field can be obtained and is shown in Figure 5.3-1(c), giving the VCMA coefficient, ζ to be 59 ± 4 fJ/Vm.

5.4. DW detection with electrical and magnetic optical Kerr imaging

Electrical pulse of 50 ns is applied through the Hall cross to inject the DWs and the presence of the DW can be found from both electrical and Kerr microscopy. Figure 5.4-1 shows the R_H measurement of the device with and without an injection pulse to nucleate the DWs. When a domain is formed by the injection pulse, the magnetic field required for domain expansion is significantly lower than the H_c of the material. Figure 5.4-1 shows the Hall cross under an applied electric field of +250 MV/m, which reduces the H_c of the material to 1590 Oe. After a J_{input} current pulse of $\sim 6 \times 10^{11}$ A/m² was sent into the Hall cross structure, a domain nucleates in the device. Upon the application of magnetic field, the injected domain expands at 264 Oe, thereby switching the magnetization of the Hall cross

Figure 5.4-1. Electric measurement for domain detection on the Hall cross device. A lower magnetic field is required to switch the magnetization. This is characteristics of an injected domain.

Kerr imaging of the Hall cross reveals that partial domains can also be formed which do not go across the width of the magnetic wire such as the Kerr image shown in Figure 5.4-2(a). The partially formed domains can also be propagated by a magnetic field lower than the H_c . Hence, the electrical measurements shown in Figure 5.4-1 are only used as a reference. In this study, DWs are only considered be successfully injected when visual inspection of the Kerr images show that the DWs are stretched across the entire width of the wire as shown in Figure 5.4-2(b).

Figure 5.4-2. Kerr images obtained after a current pulse is applied across the Hall cross device (right to left) illustrating (a) a fully formed domain with a larger J_{input} of 5.94×10^{11} A/m and (b) a partially formed domain with a J_{input} of 5.29×10^{11} A/m.

Electric field is applied onto the Hall cross during the injection current pulse. When a positive electric field is applied, the anisotropy of the material decreases, enabling DWs to be injected with a lower current density. The opposite is also true when a negative electric field is applied, resulting in a higher required current density to inject DWs. The dependence between the required current density to inject DWs and the applied electric field is shown in Figure 5.4-3. The application of a +250 MV/m electric field results in a ~20% decrease in the required current density for injection of DWs injection. This change in the required current density demonstrates the role which electric field control has in lowering the energy requirement for a DW injection.

In Figure 5.4-3(b), the H_c modulation of the material by the electric field is compared with the simulated Oersted field generated at the corners of the Hall cross structure, H_0 . The difference in the H_0 and the H_c of the material suggests that there are other effects contributing to the injection of DWs. One such effect could be Joule heating, where the heat energy lowers the thermal activation threshold for magnetization switching.^{1, 10} Also, since the current is

passing through the magnetic material, this can potentially induce spin-torque effects. The Co layer is sandwiched between two Pt layer of different thicknesses. This stack structure is known to generate a spin-orbit torque.^{11, 12} The thicker Pt layer below the Co layer contributes a larger spin-orbit torque than the top Pt layer due to spin Hall effect (SHE). This allows the overall spin-orbit torque to assist in the local magnetization switching.^{13, 14} The inset of Figure 5.4-3(b) illustrates how the SHE can assist the local magnetization switching process. During the current injection, spin accumulation occurs at the boundaries of the Pt layer due to SHE. The accumulated spins at the Pt/Co interface applies a torque to the magnetization in the same direction as the Oersted field lines, possibility resulting in a spin orbit torque assisted magnetization switching during the injection of DWs.

Figure 5.4-3. (a) The dependence of the required J_{input} to inject a fully formed domain across the device at various applied electric field. (b) Comparison of the H_0 generated by the J_{input} and the H_c of the material. The inset shows the direction of the Oersted field and the spin of electrons in the Pt layer due to the spin Hall effect with the current running out of the current sectional plane.

5.5. Summary

A DW injection technique assisted by electric field control was demonstrated to lower the required current density for DW injection in PMA magnetic wires by up to ~20%. The geometrically constrained Hall cross structure allows a larger current density at the domain injection region. This gives rise to the Oersted field, heat and spin-torque effects which can possibly contribute towards the local magnetization switching. Although the amount of contribution from the various effects that results in the injection of DWs is not entirely clear. Electric field control was demonstrated to modulate the magnetic anisotropy and coercivity of the Hall cross device. This leads to the reduction in the required current density for the DW injection, paving the way towards developing an energy efficient DW injection device.

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6. Domain wall propagation using voltage controlled magnetic anisotropy

6.1. Motivation

Spin torque techniques to drive DWs require high current densities which inherently cause resistive loss, resulting in Joule heating. To circumvent the power loss and prevent Joule heating, electric field control methods can be used to manipulate the properties of magnetic devices due to its energy efficient nature.¹⁻³ The energy of a DW can be expressed as σ , where

$$\sigma \propto \sqrt{AK_u} \quad (6.1.1)$$

here, A represents exchange stiffness parameter and K_u represents uniaxial magnetic anisotropy energy. Therefore, DW energy can be reduced by lowering the K_{anis} of the ferro-metallic material through VCMA. This has been demonstrated to assist the propagation of DWs.² This is schematically shown in Figure 6.1-1 (a) as a cross-section of the electrode/insulator/ferro-metallic stack, where a gate voltage is used to apply an electric field onto the ferro-metallic material, resulting in a higher creep velocity of the DW propagated by magnetic field.

Figure 6.1-1. Schematic diagram illustrating how VCMA can be used to propagate a DW. (a) VCMA is used to assist magnetic field propagation of the DW. (b) A sloped electric field is employed which can propagate the DW without any magnetic field assistance.

A force exerting on the DW in the x direction can be described using the DW energy and K_u of the magnetic nanowire (NW) as follows,

$$F(x) = -(\partial\sigma / \partial K_u) \cdot (\partial K_u / \partial x) \quad (6.1.2)$$

Hence, a magnetic anisotropy gradient in the x direction, along the NW (dK_u / dx) will exert a force on the DW. By using a wedged insulator as shown schematically in 5.1-1(a), a sloped electric field can be generated, with an increasing electric field for the thinning insulator layer. This gives rise to a dK_u / dx which propagates the DW.^{4, 5} However, such a wedge structure does not scale well for a longer NW, as the lengthened NW would result in increasingly thicker or thinner insulators at the ends of the NW. At the end with the thinner insulating layer, the strong electric fields may lead to a dielectric breakdown of the insulating layer. Furthermore, it is recently reported in MTJs that the VCMA coefficient is only linear at

low electric field but saturates at higher electric field.^{6,7} This poses a limitation on the length of the NW for a sloped VCMA DW memory device.

In this work, we demonstrate an energy efficient DW propagation technique whereby a dK_u/dx is used to propagate DWs at high velocities with deterministic displacements on any NW length. The DW dynamics due to the manipulation of K_u using gate electrodes are also discussed, leading towards an optimized DW propagation technique. The results provide a promising path towards high velocity and accurate DW memory device using VCMA.

6.2. Stepped anisotropy gradient

A series of gate electrode can be used create a stepped dK_u/dx . This alleviates any length considerations, allowing the DW device to propagate much further and storing more data, as schematically shown in Figure 6.2-1(a). However, since the width of a DW in a material with perpendicular magnetic anisotropy is usually <10 nm, and the minimum feature size of deep UV lithography techniques is much larger than 10 nm. This results in instances whereby the entire DW is under the same gate electrode as shown in Figure 6.2-1(a), resulting in a flat magnetic anisotropy gradient. Hence, slanted gate electrodes are proposed to overcome this challenge and provide a uniform average dK_u/dx across the DW which is perpendicular across the NW as schematically shown in Figure 6.2-1(b). The differences in insulator thickness in both Figure 6.2-1 (a) and (b) is to ensure that the gate electrodes are not in electrical contact. This ensures that the anisotropy of the local regions can be modulated without a gap in between them, allowing a smooth dK_u/dx across the NW.

Figure 6.2-1. (a) DW trapped in a region with zero dK_u/dx . (b) At any point along the NW, the DW is under ≥ 2 gate electrode, allowing the VCMA effect to create a dK_u/dx which propagates the DW.

6.3. Optimization of gate electrode width

A repeating series of dK_u/dx is created when the adjacent regions of the NW have different K_u due to the modulation of the VCMA effect. A repeating series of three gate electrodes results in a high-medium-low K_u region as shown in the upper diagram (solid border) in Figure 6.3-1(a), where the high-medium-low region is labelled as 1, 2 and 3 respectively. In this instance, the DW energy is at a minimum as the DW is at the low K_u region, the DW is represented by a green line ending with solid dots. By switching the gate voltage to shift the K_u region of the NW towards the right, as shown in the lower diagram (dotted border), the position of lowest energy is shifted by 1 gate electrode length. This results in a dK_u/dx experienced by the DW which cause it to be propagated towards the lowest K_u region.

Figure 6.3-1. (a) DW propagation on a dK_u/dx by having modulated the anisotropy of adjacent regions on the left and right of the normal anisotropy region to have higher and lower anisotropy. (b) The average K_u in the y direction is shown along the NW with varying gate width.

The low K_u regions are sandwiched by high K_u regions, forming a potential well. This allows the DW to move in a step-wise manner deterministically. The probability of the DW crossing the high K_u regions at room temperature can be calculated using

$$P = \frac{e^{-\sigma_{high}/k_B T}}{e^{-\sigma_{high}/k_B T} + e^{-\sigma_{low}/k_B T}},$$
 where P is the probability, k_B is the Boltzmann constant, T is temperature and σ is the DW energy. The calculated probability of $\sim 1 \times 10^{-8}$ is similar to the write error rate for NAND flash and MTJ devices. However, the high K_u region which ensures deterministic motion also introduces undesired DW dynamics during propagation that will be discussed later in this section.

Figure 6.3-1(b) shows the K_u along the NW with the gate width (GW) as a percentage of the width of the NW. The DW position illustrated in Figure 6.2-2(a) and 6.2-2(b) schematically shows where the position of the DW would be before the gate voltages are changed. As shown in Figure 6.3-1(b), to create a uniform dK_u/dx , the GW is designed to be half of the NW width. A GW larger than half of the NW width results in a three-part segmented gradient, and a GW lower than half of the NW results in a stepped gradient as shown in the blue and black graph respectively. Furthermore, when GW is half of the NW width, it produces the highest dK_u/dx , ideal for high speed DW propagation. This is shown in Figure 6.3-2, where the dependence of the average dK_u/dx in the y direction with varying gate width is shown.

Figure 6.3-2. dK_u/dx dependence on the GW. The largest dK_u/dx occurs when the GW is half of the NW width.

6.4. Domain wall dynamics due to gate electrode shape

6.4.1. Slanted gate electrodes

Figure 6.4-1 shows an up-down (UD) and down-up (DU) DW propagated by discrete slanted gate electrodes or a moving wedge electrode in (a) and (b) respectively. The moving wedge creates a continuous dK_u/dx that continuously detect the position of the DW and moves along with the DW as it is propagated, acting as a control to illustrate the DW dynamics caused by the slanted gate electrodes.

Figure 6.4-1. Schematic diagram of the UD and DU DW propagated by the (a) slanted gate electrodes, and the (b) sliding gate electrodes that provides a uniform dK_u/dx along with the movement of the DW.

Figure 6.4-2. Velocity dependence on the dK_u/dx for UD and DU DWs in propagated by the slanted gate electrodes and sliding gate electrode shown in Figure 6.4-3.

The steady state DW velocity dependence to the dK_u/dx of the UD and DU DW are shown in Figure 6.4-2. The abrupt decrease in velocity with increased dK_u/dx in all the configurations shown in Figure 6.4.2 is due to the well-studied Walker breakdown phenomenon.^{5, 8, 9} The Walker breakdown for a DW propagated by a sliding gate electrode and propagation by a magnetic field is phenomenologically similar and is shown in the Figure 6.4-3.

Figure 6.4-3. (a) and (b) shows the DW position along the NW for DW propagated by a sloped electric field and magnetic field respectively. The insets show the magnetization of the DW at the highlighted regions/points along the NW where the top half of the inset shows the out-of-plane magnetization and the bottom half shows the in-plane magnetization. (c) and (d) shows the corresponding DW velocity for (a) and (b) respectively. Both propagation methods show similar results and have a Walker breakdown velocity of 345 m/s. The DMI of the material for these results is 1 mJ/m^2 .

The UD and DU DWs in Figure 6.4-2 achieve distinctly different DW velocity when propagated using discrete slanted gate electrodes. However, this was not observed when the DW is propagated by the continuous dK_u/dx . The difference in velocities hints at DW dynamics occurring from more than just the smooth dK_u/dx as propagating force of the DW. Instead the discretisation of the gate electrodes has contributed to the propagation mechanism which causes the difference in DW velocities.

The DW dynamics was investigated by examining the 2-D magnetization of the DW, the averaged dK_u/dx in the y direction, and the applied K_u region of the NW. This is elaborated in Figure 6.4-4, where the DW centre position is represented by a dotted line for visual aid. Both UD and DU DWs are shown on Figure 6.4-4 respectively. In Figure 6.4-4(a), both UD and DU DWs experiences a dK_u/dx , causing the DWs to move rightwards as illustrated in Figure 6.4-4(b) where the bottom of the UD DW tilts towards the high K_u region. The red circle in the enlarged image, highlights the DW clearly encroaching into the high K_u region. However, this encroachment is not present in the DU DW as shown in the blue circle of the enlarged image. This difference is caused by the mismatch between the DW tilt and gate electrode slant angle. Due to the NW symmetry, an UD DW moving to the right is equivalent to a DU DW moving to the left. Hence, only the motion to the right is reported for conciseness.

Figure 6.4-4. 3. A comparison of a UD and DU DW being propagated using slanted gate electrodes in from (a) to (b). (a) The DWs move towards the right under the effect of the dK_u/dx . (b) The DW reaches the lowest averaged K_u in the x direction, highlighting the encroachment caused by the high K_u region in the UD DW and lack thereof in the DU DW.

The velocity of the UD and DU DWs are shown as it propagates along the nanowire. The higher average velocity for the DU DW stems from the results shown in Figure 6.4-5. The cyclical nature of the velocity derives from the switching of the gate electrodes. A spectral analysis of Figure 6.4-5(a), where fast Fourier transform is performed is presented in Figure 6.4-5(b). Here, it is clear that for the DU DW, the velocity has a periodicity of 32 nm, arising due to the gate electrodes switching after every 32 nm travelled by the DW, where the width of the gate electrode is 32 nm. However, for the UD DW, there are two peaks in the spectral analysis at ~ 16 nm and ~ 32 nm as shown in Figure 6.4-5(b). The periodicity of 16 nm occurs due to the encroachment of DW caused by the DW tilt angle mismatch after the DW moves ~ 16 nm from when the gate electrodes switches. This is further illustrated for visual representation in Figure 6.4-5(c), where the middle of the DW along the centre line of the NW is given as the DW position. In Figure 6.4-5(c)(i), the DW reaches the end of the dK_u/dx , triggering the gate electrodes to switch as shown in Figure 6.4-5(c)(ii), where the DW also propagates 16 nm before the lower portion of the DW starts encroaching into the high K_u region.

Figure 6.4-5. (a) DW velocity dependence on the position along the NW at the DW is propagated for the UD and DU DW. (b) Fast Fourier transform of the velocity plot, showing periodicity peak at 16 and 32 nm. (c) Superimposed image of the K_u region and the

magnetization, illustrating that the DW moves 16 nm before the lower end the DW reaches the high K_u region.

6.4.2. Chevron gate electrodes

While it is feasible to propagate a single DW using the slanted gate electrodes, the difference in velocity of the UD and DU DWs will result in a multi-DW propagation velocity that is limited by the slower DW. To prevent this asymmetry of DW propagation, the gate electrode shape should be symmetrical along the NW axis. As such, the use of chevron-shaped gate electrodes to propagate the DWs was investigated. The chevron gate electrodes were designed to have a steeper angle of 31° compared to 45° for the slanted gate electrodes so that the average dK_u/dx for both systems are kept constant, as shown in Figure 6.4-6.

Figure 6.4-6. Average K_u of the NW in the y direction, the background shows the K_u map of the NW modulated by slanted and chevron gate electrodes. The angle of the chevron gate

electrode has to be kept at 31° to have the same average K_u in the y direction of slanted gate electrode with an angle of 45° .

It is clear from Figure 6.4-6 that the tilt of the DW plays a role in the dynamics of the DW propagation. Hence, the averaged K_u is extracted with a tilted angle similar to a tilted DW at steady state traversing along the NW. This is shown in Figure 6.4-7 for both the slanted and chevron gate electrodes. With a tilt of 0° , the K_u is averaged across the y direction, showing the same results as previously shown in Figure 6.3-1, 6.4-4 and 6.4-6. However, with a $\pm 15^\circ$ tilt, the averaged K_u shows a distinct difference between slanted and chevron gate electrodes as shown in Figure 6.4-7(a) and 6.4-7(b) respectively, the graphs in Figure 6.4-7(b) is translated slightly in the y axis for visual clarity as the graphs are very similar. When the averaged K_u is obtained with a -15° tilt, the regions highlighted in yellow emphasize the beginning of the high K_u region, which causes the DW to slow down upon encroaching into the region. This angle mismatch only occurs for a negative tilt, however, for the chevron electrodes in Figure 6.4-7(b), the symmetry along the NW axis allows both a positive and negative tilt to have an averaged K_u similar to the one obtained without any tilt.

Figure 6.4-7. Average K_u of the NW at an angle w.r.t the y direction. (a) For slanted gate electrode, a positive tilt does not significantly alter the dK_u/dx of the propagation region while a negative tilt results in a step shaped averaged K_u . The highlighted regions in yellow shows the start of the high K_u , which caused the asymmetric results shown in Figure 6.4-2 and the periodicity in Figure 6.4-5. (b) For chevron gate electrodes, there is no significant change to the average K_u of the NW with either a tilt in either direction. The average K_u has a y offset for visual aid.

The tilt of the DW in steady state was varied by changing the DMI of the magnetic material.^{5, 10, 11} Figure 6.4-8(a) and 6.4-8(b) shows the steady state DW velocity and tilt respectively for DWs propagated using chevron and slanted gate electrodes. For slanted gate electrodes, the results in Figure 6.4-8(a) confirm that the increase in DW tilt angle mismatch results in a lower DW velocity. As the DMI increases, the difference in velocity and DW tilt between the two configurations increases, indicating that the slower velocity is due to the DW tilt angle mismatch. The difference in DW tilt angle between the configurations in Figure 6.4-

8(b) stems from the encroachment of the DW into the high K_u region, where the DW without angle mismatch with slanted gate electrodes has the largest tilt. The difference in tilt angle between configurations is mirrored by the DW velocities.

Due to the axial symmetry of the chevron gate electrodes, there is no difference in steady state DW velocity and DW tilt for a DU or UD DW during steady state propagation travelling in the same direction. However, as the chevron shape does not have lateral asymmetry, the steady state of the DW moving in opposite direction differs. The direction of DW motion is indicated by the arrow in the legend. As the DW moves towards the left, the DW tilt angle mismatch illustrated in Figure 6.4-4 happens for both UD and DU DW due to the high K_u region at both the upper and lower edge of the NW. The steeper angles of the chevron gate electrode reduce the extent of which the DW encroaches into the high K_u region as compared to using slanted gate electrodes. This results in different DW velocity and DW tilt for the slanted and chevron gate electrodes shown in Figure 6.4-8. When the chevron gate electrodes are used to propagate the DW rightwards, the higher K_u region is at the middle of the NW, hence, the tilt of DW does not increase the DW encroachment into the higher K_u region. This results in the higher DW velocity for DWs moving towards the right than moving towards the left.

Figure 6.4-8. (a) DW velocity dependence on DMI using UD and DU DW propagated with slanted and chevron gate electrodes. (b) DW tilt dependence on DMI using UD and DU DW propagated with slanted and chevron gate electrodes.

Since UD and DU DWs need to propagate at similar velocities when travelling in the same direction to avoid write errors, a NW containing multiple DWs needs to be propagated by chevron gate electrodes. This is illustrated in the inset of Figure 6.4-9, where snapshots of DWs are propagated using chevron and slanted gate electrodes. Instead of allowing the DW's

position to determine the gate electrode switching frequency, a driving frequency is applied to the NWs. The driving gate electrode switching frequency is shown in Figure 6.4-9, where the frequency increases from ~ 4 GHz to 8 GHz within 1 ns and subsequently remains constant at 8 GHz. The y axis on the right depicts the velocity of a DW driven by the gate electrode switching frequency, such as the DWs with the chevron gate electrodes in Figure 6.4-9. The domain propagated using chevron gate electrode does not suffer from any deformation throughout the propagation from (i) to (iv) of the figure inset, whereas the domain propagated using the slanted gate electrodes becomes larger or stops moving due to the difference in velocity between the leading and trailing DW.

Figure 6.4-9. Chevron and slanted gate electrodes propagating two DWs along the NWs with frequency accelerated towards 8 GHz. The NWs are 2048 nm in length and 64 nm in width. The domain propagated by chevron gate electrodes does not result in any deformation apart from the DW tilt. However, for the slanted electrodes, the DW tilt angle mismatch causes one of the DW to be slower. In the middle NW, the domain elongates as the trailing DW is unable to catch up with the frequency of the switching gate electrode. In the bottom NW, the leading DW is the one unable to catch up with the frequency of the switching gate electrode, which blocks the propagation of the trailing DW. As the trailing DW approaches the leading DW, DW-DW repulsion occurs as shown from (ii) to (iii).

As the DWs move faster towards the right than the towards left, a commercial realization for such a device would employ a mono-directional propagation for faster DW device operation. The magnetization bits can be stored as a delay line memory or looped into a circle much like a magnetic platter in HDD. Figure 6.4-10 shows the UD and DU DW velocities dependence on using the chevron gate electrodes. The DMI is chosen to be 1 mJ/m^2 , matching experimental results from CoFeB/MgO based PMA material.¹² The DWs have similar velocity and both can achieve a steady state DW velocity of up to 316 m/s at an average dK_u/dx of $\sim 900 \text{ GJ/m}^4$ before Walker breakdown occurs.

Figure 6.4-10. Velocity of DW in with chevron gate electrodes. The UD and DU DWs have similar velocities and can reach up to $\sim 316 \text{ m/s}$. The DMI is chosen to be 1 mJ/m^2 , matching experimental results from CoFeB based PMA material.¹²

Apart from Walker breakdown, another limiting factor of the DW velocity is the switching frequency of the gate voltage. Assuming a switching frequency 8 GHz, a steady state DW velocity of 256 m/s can be achieved, which is comparable to the DW velocities achieved using spin torque techniques.¹³⁻¹⁵ Also, VCMA coefficient of 100 fJ/Vm and higher has been reported,¹⁶⁻²⁰ requiring only 0.8 V on an insulating MgO thickness of 2 nm to drive the DWs at 833 GJ/m⁴ for a DW velocity of 256 m/s, which is much lower than the output voltage of most processors today. A larger gate and nanowire width would also result in a higher velocity as the DW would move across the longer gate width per cycle. However, increasing the gate and nanowire width comes at the cost of data density. A relationship between the DW velocity and data density using the proposed model is as shown in Figure 6.4-11, where the dotted line represents the DW velocity achievable using a 64 nm width NW used in this section.

Figure 6.4-11. Relation between the DW velocity and data density with varying nanowire width. The y axes are in log scale for better visual representation. The black dotted lines represent the 64 nm NW width used in this work, corresponding to a maximum DW velocity of 256 m/s with a clock speed of 8 GHz.

6.5. Energy consumption

Due to its dissipationless nature, electric field control has been shown to be very energy efficient in many techniques across different spintronic devices.^{1-3, 21-24} A simple comparison to give a general idea of the energy consumption can be easily calculated with the parameters provided in Table 6.6-1 below. For a DW to propagate 96 nm, a current based spintronic device would require ~ 6 times more energy compared to the proposed device in this Chapter. The values provided in the table does not consider the additional energy required for current based devices to depin from pinning sites.

(a) Table of parameters for proposed device

Parameter	Value
VCMA coefficient ²⁵	100 fJ/Vm
Insulator thickness	2 nm
Applied electric field	0.04 V/nm
Relative permittivity ^{26, 27}	9
Area	32 nm $\times$ 64 nm
Energy required	0.05 fJ

(b) Table of parameters for current based device

Parameter	Value
Current density ¹³	1×10^{12} A/m ²
DW speed ¹³	300 m/s
Cross sectional area	64 nm $\times$ 5 nm
Resistivity ²⁸	1×10^{-7} Ω m
Distance	96 nm
Energy required	0.3 fJ

Table 6.6-1. Comparison table for energy consumption for (a) proposed device and (b) current based device

6.6. Simulation parameters

The work presented in this Chapter was carried out using a mesh of $1\text{ nm} \times 1\text{ nm} \times 1\text{ nm}$, which is sufficiently shorter than the exchange length and the DW width. The material parameters are meant to simulate a CoFeB/ MgO-based structure and are as follows: saturation magnetization = $8 \times 10^5\text{ A/m}$, exchange stiffness = $22 \times 10^{-12}\text{ J/m}^2$, anisotropy energy = $0.8 \times 10^6\text{ J/m}^3$ and the Gilbert damping coefficient = 0.03. The discretization in time is based on the Runge-Kutta method as introduced in the 1st Chapter, allowing an adaptive time step with a maximum error per step of 10^{-5} to provide accurate results.²⁹

6.7. Summary

In conclusion, a comprehensive simulation study on the viability of slanted- and chevron-shaped gate electrodes to propagate DWs at high velocities was conducted. The discretized DW movement induced by the array of gate electrodes confines DW to precise locations, thus removing the need to create permanent pinning sites. With slanted gate electrodes, the DW tilt angle mismatch results in the velocity difference between UD and DU DW configurations, this results in domain propagation being limited by the slower DW. However, when axially symmetrical chevron gate electrodes were used, the UD and DU DWs were propagated at the same velocity on the NW, achieving high DW velocities of up to 316 m/s before Walker breakdown, which is ~ 6 times faster than a DW propagated by a slope electric field.⁵ The results presented in this work paves the way for energy efficient high velocity DW devices using the VCMA effect.

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7. Conclusion and future work

7.1. Summary of thesis work

In this thesis, electric field control of magnetic material is investigated using Hall bar devices. In the majority of research work on electric field control of magnetic materials, there is an assumption that the effect is based on the interface between the insulator and ferrometallic material as electric field decays rapidly in a conductive material.¹⁻⁵ However, in Chapter 3 the electric field control of multi-layered Pt/Co based PMA structure is investigated. Recently, research on the bulk component of VCMA has been attributed to magneto-elastic effects.^{6, 7} Also, PIM of heavy metal such as Pt and Pd has also been touted as the mechanism which allows electric field control of a magnetic layer under a metallic layer.⁸⁻¹⁰ These works signal the shift in focus into thermal robust magnetic materials and support the findings of Chapter 3, whereby the electric field modulation of Pt/Co multilayer structure is investigated. By comparison with a single Co layer and replicating the experiment using micromagnetic simulations. The electric field modulation of the bottom Co layer can be attributed to the PIM of the Pt spacer layer, allowing the double Pt/Co structures to have <20% decrease in K_{eff}/E as compared to a single Co layer. The decrease of K_{eff}/E is hugely compensated by the increase in thermal robustness of ~390-520% when compared to single Pt/CO structures.

Apart from the magnetic material investigation, the insulating layer used for electric field control is also investigated in Chapter 4. An insulating polymer was chosen due to its similar nature to a liquid electrolyte, ensuring that there is little to no major defects within the layer. After baking, the insulating polymer hardens, keeping it in place and ready for use as a device. Furthermore, spin coating of the insulating polymer is a much faster process than any conventional physical or chemical vapour deposition methods, requiring only minutes instead

of hours. The insulating polymer is spin coated on a PMA thin film consisting of Ta/Pt/Co/SiO₂. The magneto-ionic effect observed is reversible and has non-volatile retention of H_c . The applied electric field results in a change in H_c of -47% and $+68\%$ which correlates to a change in Co thickness from 0.3 to 0.75 nm. The transport measurements from the R_H and resistance of the nanoscale device also support the mechanisms for the magneto-ionic effect. The dynamics of the H_c is characterized and it is indicative that a thinner layer of polymer results in faster magneto-ionic effect for tuning the H_c of the magnetic layer. Furthermore, using an insulating polymer for electric field control of magnetic material the usage of an insulating polymer allows the possibility of future spintronic devices on flexible substrates.

In Chapter 5, electric field control is experimentally demonstrated to assist DW injection for memory devices. DW injection is conventionally achieved by applying a current pulse into a stripline on top of the magnetic wire. The current pulse creates an Oersted field which causes local magnetization switching, thereby injecting DWs.¹¹⁻¹⁴ A geometrically constrained Hall cross structure is proposed to be a DW injection device. The geometrical constrain causes larger current density at the domain injection region which gives rise to larger Oersted field, heat and spin-torque effects which contributes towards the local magnetization switching. Electric field control was demonstrated to modulate the magnetic anisotropy and coercivity of the Hall cross device. This leads to the reduction in the required current density for the DW injection by $\sim 20\%$. The result leads towards the realization an energy efficient DW based memory device.

DWs are usually propagated by current based techniques which utilizes either the STT or the SOT effect. However, both current-based techniques required a large amount of energy and incur losses such as heat energy. A technique to propagate domain walls using VCMA is proposed and demonstrated by micromagnetic simulation in Chapter 6. The techniques involve discrete gate electrodes to propagate DW. The discretized DW movement induced by the array

of gate electrodes confines DW to precise locations, thus removing the need to create permanent pinning sites. With slanted gate electrodes, the DW tilt angle mismatch results in the velocity difference between up-down and down-up DW configurations, this results in domain propagation being limited by the slower DW. However, when axially symmetrical chevron gate electrodes were used, the up-down and down-up DWs were propagated at the same velocity on the NW, achieving high DW velocities of up to 316 m/s before Walker breakdown, which is ~ 6 times faster than a DW propagated by a slope electric field.¹⁵

7.2. Outlook of future work

The formalisation of electric field control is usually attributed to the change in effective anisotropy which can be written as,

$$K_{eff} = K_B + \frac{K_i}{t} - \frac{\mu_0}{2} M_s^2 \quad (7.2.1)$$

where K_{eff} represents the effective anisotropy, K_B represents the bulk anisotropy, K_i represents the interfacial anisotropy, t represents the thickness, μ_0 represents the permeability of free space and M_s represents the magnetic saturation. In micromagnetic simulation, K_B and K_i is combined to be represented as K_{anis} , shown in Equation (3.5.1).

In Chapter 3, the electric field modulation is taken to be the change to the K_{eff} due to the applied electric field, K_{eff} / E . Whereas in Chapter 6, the electric field modulation is assumed to be derived fully from the interfacial anisotropy. In order to separate the electric field modulation into the bulk and interfacial component, a thickness dependence study needs to be conducted, which can be also used to find the magneto-elastic term.⁶

7.2.1. Ultra-fast magnetization switching of magnetic tunnel junction for domain injection

Magnetic tunnel junction (MTJ) has been discovered since 1994¹⁶ and is widely used in HDD as magnetic sensors today. An MTJ can be used as a basis for non-volatile magnetic random access (MRAM). Appendix A will explore more about the mechanism of a MTJ.

The magnetization switching of a MTJ determines the write speed of the memory device. Therefore, it is imperative that the magnetization switching happens at the nanosecond scale or below. Nanosecond electrical pulse can be used to harness the VCMA effect to induce precession of the magnetization.^{5, 17} The precessional nature of the magnetization is shown in the switching probability which has an oscillatory behaviour. However, the nanosecond control of the electrical pulse has to be very precise due to the oscillatory behaviour otherwise it will lead to high write errors rates. Hence, a combination of VCMA with STT is used to increase the switching reliability.¹⁸ Two electrical pulses are used, a higher voltage pulse to precess the magnetization and a second longer pulse to finalized the switching process.

The MTJ can be grown on top of domain wall device, as shown in the schematic diagram in Figure 7.2-1. The track of the domain wall device acts as the free layer of the MTJ. Hence, the MTJ can sense the magnetization of the domain wall device. The combination of VCMA and STT technique can also inject a domain into the domain wall device at a nanosecond timescale.

Figure 7.2-1. Schematic diagram illustrating the use of a MTJ on a domain wall device. The pulse generator generates the electrical pulse for both the VCMA and STT.

7.2.2. Electric field assisted domain wall propagation with spin orbit torque

The mechanism of SOT has fascinated the spintronics community for several years, sparking interest from its contributing Rashba or spin Hall effect.¹⁹⁻²² However, the electric field control on SOT has not been thoroughly investigated. Following the work on Chapter 3, 4 and 5, electrical measurements using the Hall cross devices can be measured using harmonic measurements,²³ where the 1st and 2nd harmonic signal can be written as follows,

$$V_{\omega(t,l)} \approx \pm \frac{1}{2} \Delta R_H I_0 \left(1 - \frac{H_{(t,l)}}{2H_k}\right)^2 \quad (7.2.2)$$

$$V_{2\omega(t,l)} \approx -\left(\pm \frac{1}{4} \Delta R_H I_0\right) \Delta H_{(x,y)} \frac{H_{(t,l)}}{H_k^2} \quad (7.2.3)$$

where V_{ω} and $V_{2\omega}$ represents the 1st harmonic and 2nd harmonic signal respectively. R_H represents the Hall resistance, I_0 represents the current, H_l and H_t represents the transverse and longitudinal magnetic field respectively, H_k represents the effective anisotropy field and the $\pm$ represents the up and down magnetization states.

The damping and field like terms corresponds to the H_l and H_t respectively, and they can be found with the following equation,

$$\Delta H_{(t,l)} = -2 \frac{\partial V_{2\omega}}{\partial H_{(t,l)}} / \frac{\partial^2 V_{\omega}}{\partial H_{(t,l)}^2} \quad (7.2.4)$$

Since electric field control has been demonstrated to modulate the H_k of the material, this would in turn modulate the field-like and damping-like terms as shown in Equation (7.2.1) to (7.2.3). Experimental verification on the electric field control on the SOT terms will provide fundamental understanding towards both electric field control and SOT.

7.2.3. Electric field control on antiferromagnet and synthetic antiferromagnetic structure.

Antiferromagnets have been gaining a lot of research interest due to the possibility of terahertz computing through the use of the strong exchange coupling. Furthermore, antiferromagnetic materials have no net magnetic moment, hence it circumvents any issues related to stray fields.

Recently, the VCMA effect has been studied on anti-ferromagnetic materials using *ab initio* calculations, predicting a large VCMA coefficient for antiferromagnetic materials. Additionally, micromagnetic simulations for the VCMA effect modelled onto anti-ferromagnetic material, ²⁴ showed a switching time of < 10 ps. Experimentally, magnetic samples capped with a 15 nm IrMn has also demonstrated that VCMA is able to modulate the exchange constant of the antiferromagnetic material. ²⁵ These recent results point towards the possibility of enabling antiferromagnets as a memory storage material. Hence, the next progression would be to experimentally demonstrate electrical switching of the antiferromagnet using VCMA.

The logical progression from Chapter 3, which demonstrated the electric field control on Pt/Co multilayer would be to apply the same electric field control on a synthetic antiferromagnetic (SAF) stack structure. A SAF stack structure has an antiferromagnetic coupling occurs interlayer across a non-magnetic spacer, which is usually Ru.²⁶⁻²⁸ This has been widely used in MTJ stacks to reduce the stray field from the fixed layer.²⁹⁻³¹ Furthermore, domain wall motion on SAF stacks has been demonstrated to propagate at a higher velocity when driven by SOT.^{32, 33} Similar to the work in Chapter 3, the SAF structure consists of Pt/Co/PMA structures, the Pt/Co layers sandwich a Ru layer which encourages the antiferromagnetic coupling between the PMA layers. Recently, micromagnetic simulations have already shown that the antiferromagnetic coupling between the PMA layers can be modulated by electric field control.³⁴ Therefore, the work carried out in Chapter 3 is a step towards experimental demonstration of electric field control on SAF structures. This can be experimentally conducted using Hall crosses, similar to the work carried out in Chapter 3. Other methods such as applying it to the free layer switching of a MTJ³⁵ or towards improving DW velocity on a SAF structure can also be carried out.

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Appendix A: Magnetic tunnel junction

Introduction

A magnetic tunnel junction (MTJ) is a basic unit of a magnetic random-access memory (MRAM). MRAM devices based on spin transfer torque are already commercially available in the market for niche applications.^{1,2} However, these devices requires high writing current (~ 10 times more energy required than SRAM) which poses an issue due to Joule heating and degrades device durability.^{3,4} Hence, electric field control is touted to allow the magnetization switching to occur at high speed while consuming low energy.⁵⁻⁸

A MTJ is made up of two ferromagnetic layers sandwiching an insulating tunnel barrier. The two ferromagnetic layer differs in their coercivity, the layer with the lower coercivity is known as the free layer and the layer with higher coercivity known as the fixed layer. The difference in coercivity allows an external magnetic field to selectively switch the magnetization direction of free layer without switching the fixed layer. This can occur when the applied magnetic field is larger than the free layer coercivity yet smaller than the coercivity of the fixed layer. Therefore, both parallel and anti-parallel alignment between the two ferromagnetic layers can be achieved by changing the direction of the applied magnetic field.

A generalised model by Jullière⁹ assumes that the electron spin is conserved during tunnelling, causing spin-down and spin-up electrons to act as a two-spin channels. When the ferromagnetic layers are in the parallel alignment, the minority spin electrons can only tunnel to the empty minority states, and *vice versa* for the majority spin electrons. However, when the ferromagnetic layers are in the antiparallel alignment, the majority-spin electrons only tunnel to the minority-spin states and *vice versa* for the minority-spin electrons. This results in the difference of the tunnelling resistance between parallel and anti-parallel alignment, which forms the basis of the binary “1” and “0” bit in a MTJ device. The difference of resistance

between the parallel and anti-parallel state of a MTJ is general referred to as the tunneling magneto-resistance ratio (TMR). The equation to determine the TMR of a MTJ is $\frac{R_{ap} - R_p}{R_{ap}}$,

where R_{ap} and R_p are the resistance in the anti-parallel and parallel alignment.

A MTJ can also be used as a sensing unit in a DW memory device. It can either be placed close to the DW memory device to sense the stray field, much like the read operation in HDD, or it can be directly grown onto the domain wall device itself, with the domains acting as the free layer of the MTJ.

Results

Magnetization Characterization

Co₂₀Fe₆₀B₂₀ is the material used as the ferromagnetic layer in the MTJ where the subscript refers to the ratio. Stacks using CoFeB and MgO as the tunnel barrier are deposited and annealed at 280 °C. The MOKE result of Ta(5) CoFeB(1.2) / MgO(2) / CoFeB 1.6) / Ta(5) is shown in Figure A.1(a), where the parentheses represents the thickness in nanometers. The figure shows clear decoupling of the two CoFeB layer, with a coercivity of 9 Oe and 51 Oe for the free and fixed layer respectively.

The MTJ stack is grown with an additional SAF structure on the bottom. The SAF layer is made up of Co/Pt multilayers sandwiching a thin Ru layer. Hence, the eventual deposited stack is, Ta(5) / Pt(5) / [Co(0.6)/Pt(0.3)]×4 Co(0.6) / Ru(0.4) / [Co(0.6)/Pt(0.3)]×4 Co(0.6) / Ta(1) / CoFeB(1.2) / MgO(2) / CoFeB(1.6) / Ta(5). The MOKE result of the SAF MTJ is as shown in Figure A.1(b). The arrows represent the magnetization direction during the magnetic field sweep, the top arrow represents the CoFeB free layer, middle arrow represents the [Co/Pt]×4 / Ta / CoFeB ferromagnetically coupled layer, and the bottom arrow represents the

bottom [Co/Pt] $\times$ 4 layer. The free layer has a coercivity of ~ 12 Oe and the fixed layer's coercivity is ~ 3498 Oe.

Adding the SAF layer beneath the fixed layer of the MTJ helps to pin the fixed layer and reduces stray field from the fixed layer to the free layer. The SAF layer also helps to reduce the threshold critical current density for switching using STT effect.¹⁰

Figure A.1. (a) MOKE result of a Ta/CoFeB/MgO/CoFeB/Ta sample after annealing at 280 °C for 1 hour. (b) MOKE result of a SAF thin film consisting of Ta / [Co/Pt] $\times$ 4 / Ru / [Co/Pt] $\times$ 4 / Ta / CoFeB / MgO / CoFeB / Ta.

Current-in-plane-tunnelling

The TMR of the thin films were determined using a technique known as current-in-plane-tunnelling (CIPT). The working principle of CIPT involves the use of a four-point probe setup with varying pitch width in order to find the tunnelling resistance. At smaller probe spacing, the resistance is higher as most of the current is shunted only through the top conducting layer. However, as the probe spacing increases, the electrical configuration changes to a parallel circuit consisting of the top and bottom electrode bottom electrode. Figure A.2 (a) shows a schematic diagram illustrating the current path as the probe distance changes, and Figure A.2 (b) shows the change in sheet resistance of the measured SAF MTJ sample with varying mean pitch width.

Figure A.2. (a) Schematic diagram of current path using CIPT for thin film MTJ stack at different probe spacing. (b) CIPT result of SAF MTJ thin film stack. Blue and red circle represents the sheet resistance measured when magnetic field of opposite polarity is being applied. Blue and red crosses represent the fitting function of the sheet resistance. (c) CIPT result of SAF MTJ thin film stack, showing the percentage difference of sheet resistance measured in magnetic field of opposite polarity. The green line shows the fitted curve.

As the mean pitch width continue to increase, contribution of the tunnelling resistance to the total resistance decreases and its change in magneto-resistance may not be significant enough to detect. Hence, the mean pitch width of the probes to detect the tunnelling resistance must be somewhere in between shunting through the top electrode and the tunnelling resistance being negligible. From dimension analysis the length scale of the mean pitch width to detect the tunnelling resistance is expressed as, $\lambda = \sqrt{RA / (R_T + R_B)}$, where λ represents the mean pitch width, RA represent resistance area of the tunnel barrier, and R_T and R_B represents the sheet resistance of the top and bottom electrode respectively.

Figure A.2 (c) shows the change in sheet resistance at an applied magnetic field of opposite polarity at varying mean pitch width. By detecting the change in tunnelling resistance with various probe distance, the TMR of the MTJ stack can be known by finding the percentage change. The results show the thin film achieving a TMR of 74.2%. This is similar to the TMR widely reported for CoFeB/MgO/CoFeB MTJ stacks.¹¹⁻¹³ The CIPT measurement reported in this section is carried out using a Capres CIPTech M-200.

Device tunnelling magneto-resistance measurement

The thin film SAF MTJ stack is patterned into circular junctions 5 μ m in diameter using a combination of ultra violet lithography and ion milling. Figure A.3 (a) shows the fabricated device under a microscope. The magneto-resistance measurement carried out with a sweeping out-of-plane magnetic field. Figure A.3 (b) shows the averaged results over 5 measurements of the magneto-resistance measurement with clear TMR of 2.09 ± 0.04 %. From Figure A.3 (b), it can be clearly seen that the SAF fixed layer switches at 2376 ± 1 Oe and the free layer switches at 457 ± 1 Oe.

Figure A.3. (a) Fabricated MTJ device under an optical microscope. (b) Magnetoresistance measurement of SAF MTJ. The arrows show the magnetic direction of the layers during the field sweep. The top arrow represents the free layer and the bottom two layers represents the SAF magnetic layers.

The coercivity from magneto-resistance measurements of the fabricated MTJ differs from the MOKE measurement of thin film MTJ, which had coercivity of ~ 12 Oe and ~ 3498 Oe for the free and fixed layer respectively. This difference may be attributed to the sidewall damage during the fabrication process. The scaling of MTJ from a thin film stack to a micron sized junction also changes the magnetic anisotropy and switching field of the material.^{14, 15} These results show the basic functions of the MTJ device. The MTJ device can be incorporated into DW devices for magnetization sensing and domain injection purposes.

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1. **Funan Tan**, Gerard Joseph Lim, Wai Cheung Law, Fei Long Luo, Hong Xi Liu, Francis Poh, Danny Shum, and Wen Siang Lew*, "Electric Field control on Gated Pt/Co/SiO₂ Heterostructure with Insulating Polymer", Journal of Physics D: Applied Physics, 51, 365001 (2018)
2. **Funan Tan**, Qi Ying Wong, Weiliang Gan, Si Hua Li, Hongxi Liu, Francis Poh and Wen Siang. Lew*, " Electric field control for energy efficient domain wall injection ", submitted.
3. **Funan Tan**, Gerard J. Lim, Tianli Jin, Hongxi Liu, Francis Poh and Wen Siang. Lew*, "Electric field control on single and double Pt/Co heterostructure for enhanced thermal stability", submitted.
4. **Funan Tan**, Weiliang. Gan, Calvin C.I. Ang, Grayson.D.H. Wong, Hongxi Liu, Francis Poh, Danny Shum, and Wen Siang. Lew*, "High velocity domain wall propagation using voltage controlled magnetic anisotropy", Scientific Reports, revision.
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1. **Funan Tan**, Grayson D.H. Wong, Hongxi Liu, Francis Poh, and Wen Siang Lew, "Electric field control on single and double Pt Co heterostructure for enhanced thermal stability", Globalfoundries Technology Forum 2019, Singapore

2. **Funan Tan**, Grayson D.H. Wong, Qi Ying Wong, and Wen Siang Lew, "Electric field control on Pt/Co multilayer for enhanced thermal stability", IEEE Magnetic Symposium 2018, Singapore

3. **Funan Tan**, Weiliang Gan, Calvin C.I. Ang, and Lew Wen Siang, "Domain wall motion driven by voltage controlled magnetic anisotropy effect", Institute of Physics Singapore Meeting 2018, Singapore

4. Tian Li Jin, **Funan Tan**, Weiliang. Gan, Jiang Wei Cao, Wen Siang Lew, and S. Piramanayagam, "Pinning sites with tiled magnetisation for domain wall motion control in racetrack memory", Intermag 2018, Singapore

5. **Funan Tan**, Weiliang Gan, Wen Siang Lew. "Evaluation of voltage controlled magnetic anisotropy assisted domain wall injection in perpendicular magnetic anisotropy nanowire", International Conference on. Materials for Advanced Technologies 2017, Singapore

6. **Funan Tan**, Gerard J. Lim, Wai Cheung Law, Fei Long Luo, Hong Xi Liu, Francis Poh, Dannyn Shum and Wen Siang Lew, "Reversible Oxidation on Pt/Co Nanowire Using Gate Polymer", IEEE Magnetic Symposium 2017, Singapore

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