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Time-resolved domain wall motion in novel spin transfer torque devices

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A thesis submitted for the degree of Doctor of Philosophy

University of Bath
Department of Physics

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ABSTRACT

In the past decade, the interaction of spin polarised electrons with ferromagnetic domains and domain walls has stimulated an immense volume of research work in the field of spintronics worldwide. Perpendicular magnetic anisotropy (PMA) in thin film magnetic materials has been widely exploited as a building block for realising spintronic devices. Co/Pt multilayer thin films with PMA have been extensively investigated for their potential applications in magnetic recording media and MRAM-like devices. Much work has been done to develop ways to control the magnetic properties of these films. Ga\(^+\) focussed ion beam (FIB) irradiation is a well-established technique for controlling the magnetic properties of systems with perpendicular magnetic anisotropy (PMA).

Here we have systematically investigated the coercivity, magnetic anisotropy and surface roughness of Ta(4 nm)/Pt(3 nm)/Co(x nm)/Pt(1.8 nm) multilayer films as a function of Ga\(^+\) FIB irradiation dose. The influence of the thickness of Co and Pt layers on the coercivity and switching behaviour was systematically investigated and the conditions established for realising structures with medium coercivity (∼100 Oe) and sharp switching that are well-suited for current-driven domain wall motion studies. An unexpected increase in the coercivity at very low Ga\(^+\) ion doses followed by a reduction at higher irradiation levels is observed. This correlates with an increase in surface roughness which we tentatively attribute to the ion-induced formation of highly strained nanoscale regions of ordered CoPt alloy at the Co/Pt interface. The possibility to both increase and decrease coercivity with very low dose Ga\(^+\) ion irradiation could have important applications in the design of novel spintronic devices. In addition, annealing at relatively low temperatures (< 200 C) is shown to drastically increase the magnetic anisotropy and switching field for all Co thicknesses whereby the ratio of increase in coercivity and magnetic anisotropy is higher in thicker Co films.

Optimised Co/Pt multilayer films were lithographically patterned into nanowire devices for time-resolved Extraordinary Hall Effect (EHE) measurements. The devices were based on 50 Ω coplanar waveguides incorporating single Hall cross structures. The coercivity of the region surrounding the Co/Pt Hall crosses was reduced by local Ga\(^+\) FIB irradiation, allowing the controlled nucleation of domain walls (DWs) at the edges of these regions by application of an appropriate field sequence. Domain imaging was carried out to study the evolution of artificial domains and DW dynamics under the application of magnetic fields and current pulses. DW creep measurements were carried out to study the effect of bias field on DW motion. Using pulsed currents, spin transfer torque-driven domain wall motion was
demonstrated in these artificial domain structures and the DW position and velocity tracked using time-resolved EHE measurements.
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LIST OF ABBREVIATIONS

1) DW - Domain wall
2) DWM – Domain wall motion
3) STT – Spin transfer torque
4) ML – Multilayers
5) PMA – Perpendicular magnetic anisotropy
6) FIB – Focussed ion beam
7) EHE – Extraordinary Hall effect
8) XRD – X-ray diffraction
9) MOKE – Magneto optical Kerr effect
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Chapter 1 Overview

1.1 Introduction and Background

The 20\textsuperscript{th} century has witnessed some of the greatest revolutions in human history in the form of political, social and economic changes. All these changes have shaped the century, but none quite like the technological changes that have dominated every aspect of human life. The field of electronics lies at the very heart of these technological changes. In this new millennium, the emerging field of nanotechnology is pushing forward the boundaries of technological progress. Electronics, supported by physics, is at the heart of these advances. The presence of charged particles and the manipulation of their densities is the basis of nearly all present day electronics. Quantum physics tell us that electrons not only possess a charge but also another property called spin. Nobel Prize winner Sir Neville Francis Mott gave us the “Two Current Model” \cite{1} which assumes that the electron spin state is conserved, spin up or spin down. Even though this has been known since the 1920s, practical devices that manipulated the spin or at least took advantage of it were out of reach until recently. Research and industrial applications based on spin were a distant reality until the arrival of nanotechnology. Thin films, nanowires and quantum dots have made the study and manipulation of spin practical. This has led to the birth of a new field called spin-electronics or simply spintronics.

Spintronics has fascinated the research community all over the world ever since its development revealing its unique and fascinating properties. Spintronics represents a recognised breakthrough in the field of nanotechnology. It is a multi-disciplinary research area where the joint efforts of several related fields are required to achieve results in challenging themes. The technology behind spintronics relates to the ability to modify or influence the quantum spin state of electrons. Magnetic materials have their own advantageous properties and spintronic devices combine the properties of magnetic materials, thereby making them non-volatile, fast and less energy consuming. In modern technology, spintronics is widely used and finds wide applications in various fields, most notably in memory devices like MRAM and in read heads. These magnetic devices are based on spin polarised currents and their interaction with magnetic domains and domain walls.

By the 1960s and 70s there were substantial efforts aimed at studying the underlying physics of DW motion in magnetic thin films and multilayers \cite{2}. There were also theoretical efforts targeted at understanding magnetic domains and domain dynamics based on the Landau-Lifshitz (LL) or Landau-Lifshitz-Gilbert (LLG) models \cite{3, 4}. Experimental results
and theoretical predictions were in agreement in most cases and became the basis for our understanding of domain wall dynamics.

Recent efforts to generate and utilize spin polarised currents to control domain wall dynamics in ferromagnetic structures have led to several breakthroughs [5]. Slonczewski first demonstrated the reversal of magnetic moments in a ferromagnetic layer by the injection of spin polarised currents [6]. With the emergence of spin transfer torque (STT), which is the angular momentum transfer of spins resulting in the generation of a torque on domains and domain walls, several drawbacks that were present in the first generation of MRAMs and other magnetic devices were overcome. However, the current densities required to achieve spin transfer torque (STT) domain wall motion [7-12] are currently at \( \sim 6 \times 10^6 \) A/cm\(^2\) [13, 14] high for practical applications. Hence there is need to find an alternate approach which would require lower current densities in order to exploit current-induced domain wall motion.

In the past decade there have been several investigations of “artificial domains” realised by the local focused ion beam (FIB) irradiation of magnetic multilayer structures with perpendicular magnetic anisotropy (PMA) [15-17]. This approach allows one to define nanoscale ferromagnetic domains of arbitrary shape with controllable anisotropy and coercive fields. Not only do these eliminate the need for an additional field pulse to nucleate a domain wall, but the ability to tune the local magnetic anisotropy represents an additional control parameter that can be used to reduce critical current densities. The use of FIB irradiation is a well-established technique [18] for controlling the magnetic properties of systems with perpendicular magnetic anisotropy (PMA). PMA in Pt/Co/Pt trilayers with Co thickness less than 1nm [15] is influenced by the interface roughness, strain and the intermixing at the Co-Pt interfaces [15-17]. Ga\(^+\) FIB irradiation tends to relax the strain at interfaces, resulting in a reduction of the film coercivity and anisotropy [19].

The focus of this PhD is an exploration of the implementation of artificial magnetic domains in Pt/Co/Pt multilayer structures and the optimisation of these domains for STT domain wall motion. In this way artificial domains can be realised formed from adjacent regions with very similar magnetisation but different coercive fields, presenting a novel opportunity to reduce the current density for STT DW motion.
1.2 Thesis Structure

This thesis describes experimental research into artificial domains in Pt/Co/Pt multilayers realised by Ga⁺ ion irradiation and the demonstration that time-resolved extraordinary Hall Effect measurements represent a novel approach to characterise the velocity of driven domain walls. The research work was predominantly carried out at the University of Bath, while Co/Pt multilayer samples were grown using DC magnetron sputtering at University of Leeds. Ga⁺ ion irradiation using a focussed ion beam (FIB) was carried out at the Interface Analysis Centre, University of Bristol and magneto-optical imaging using Kerr microscopy was performed at the National Institute of Material Science, Japan.

Chapter 2 introduces the fundamentals of ferromagnetism; magnetic anisotropy, magnetic domains, domain walls and magnetisation reversal processes, material that is mostly found in undergraduate and postgraduate textbooks. This section provides a background in magnetism and establishes the context for the experimental results that follow.

Chapter 3 introduces the basic principles of spintronics; the two-current model, spin transfer torque (STT), extraordinary Hall effect (EHE) and the fundamental theories of domain wall motion by application of magnetic fields and currents, which motivate the experiments on ferromagnetic multilayer structures.

Chapter 4 describes the Co/Pt multilayer thin film growth, nanofabrication techniques and device designs employed in the fabrication of Hall probe devices as well as the techniques used to characterise the Hall probe devices.

Chapter 5 describes the experimental approach for low frequency EHE measurements, time-resolved EHE measurements and magneto-optical domain imaging, as well as the acquisition and analysis of data.

Chapter 6 details the optimisation of each layer in the Ta/Pt/Co/Pt multilayers using M-H loops and x-ray diffraction (XRD) characterisation.

Chapter 7 describes the realisation of artificial domains in Hall bar structures by Ga⁺ ion irradiation and how the anisotropy and coercive field of the Co/Pt multilayer can be controlled and optimised for the needs of the domain wall motion experiments.

Chapter 8 describes Kerr magneto-optical imaging of artificial domain switching, domain wall creep under the influence of a small bias field and time-resolved EHE measurements to monitor the DW motion in the current arm of the Hall bar.

A summary of the research results and proposals for future extensions of this work are contained in chapter 9 which concludes this thesis.
Chapter 2 Introduction to Ferromagnetism

Magnetic materials are classified based on the dependence of their magnetization on an external magnetic field. The three common types are diamagnetic, paramagnetic and ferromagnetic materials [20]. The phenomenon of having spontaneously aligned magnetic moments without any external field is known as ferromagnetism and materials that exhibit this are known as ferromagnets. In ferromagnetic materials the net magnetic moment is very large because all the atomic moments are aligned in one direction. This is mainly due to the coupling of the magnetic moments of neighbouring atoms leading to parallel alignment. The interaction responsible is called exchange coupling (exchange coupling is responsible for antiferromagnetic alignment as well) [21]. A region inside a ferromagnetic material in which the atomic magnetic moments point along the same direction is called a domain. The magnetic moments in a domain will orientate themselves under the influence of various energy terms like the exchange energy, magnetocrystalline anisotropy and Zeeman energy [22-24]. In addition the energy of a spin also depends on the dipole interaction with every other magnetic moment present inside a domain. Ferromagnetic materials exhibit spontaneous magnetisation below the ferromagnetic Curie temperature $T_C$ [20, 23, 24]. The material becomes paramagnetic without a spontaneous moment, when the temperature is greater than $T_C$. Ferromagnetic materials typically consist of a number of smaller domains that are spontaneously magnetized in different directions, with domain boundary or domain wall (DW) [20, 23, 24] separating the domains. Upon application of applied field, most of the magnetic changes occur at the DWs. Based on the physical and magnetic properties of the sample different types of domain walls are broadly classified as Head-to-Head DWs (Vortex and Transverse walls) [25] and Up-Down DWs (Bloch and Néel walls) [26]. The two common types of domain walls in out-of-plane magnetised systems with different spin structures are called the Bloch and Néel structure types [22-24, 27]. A Bloch wall has magnetisation parallel to the plane of the wall while the Néel wall has magnetisation perpendicular to the plane of the wall.

2.1 Diamagnetism, Paramagnetism & Ferromagnetism

Materials whose atoms lack permanent magnetic moments are classified as diamagnetic [20, 21, 24]. In diamagnetic materials the net magnetic moment is equal to zero when in zero applied magnetic field because the atoms or ions have complete electronic shells. The
magnetic susceptibility of diamagnetic materials often arises due to distortions of orbital motion by a magnetic field, and is small and negative in organic materials and light elements while it is large and negative in superconducting materials. Hence, when a diamagnetic material is placed in a magnetic field, it repels the magnetic lines of force as shown in figure 2.1

![Figure 2.1: (a) Behaviour of the flux lines ‘B’ in a diamagnetic material in the presence of an applied magnetic field. The magnetic lines of force are repelled by a diamagnetic material](image)

Atoms that carry a permanent magnetic moment but do not interact strongly with neighbouring atoms are classified as paramagnetic [20, 21, 24]. Electrons having both spin and orbital magnetic moments produce a net moment when the shells of the electrons are not completely filled. In the case of paramagnetic materials, the magnetic moments are randomly oriented in the absence of an external field resulting in net zero magnetisation as shown in figure 2.2(a). When a magnetic field is applied to a paramagnetic material, the magnetic dipoles of the atoms align themselves with the direction of the field producing a net magnetisation which is proportional to the field. The magnetic susceptibility (\( \chi = M/H \)) is positive and rather temperature dependent in these materials.
When permanent atomic magnetic moments in a material align themselves spontaneously in the absence of an external applied field, the material is called ferromagnetic with the moments parallel to each other. If the moments are antiparallel then it is called anti-ferromagnetic and the net magnetic moment is zero. If the opposing magnetic moments are unequal, then a spontaneous magnetisation exists and the material is said to be ferrimagnetic [20, 23, 24]. In ferromagnetic transition metals the spin moment is the main contribution to the net magnetic moment and the spontaneous magnetization is due to the exchange interaction between the neighbouring atoms. In this way the magnetic moments are coupled together resulting in an overall parallel alignment within a region called a domain. In the absence of applied field, there are many domains with different magnetisation present in a material. The magnetic susceptibility ($\chi$) of soft multi-domain ferromagnetic materials is positive and very large. Ferromagnetic materials often exhibit hysteresis due to domain wall pinning. Nickel (Ni), Iron (Fe) and Cobalt (Co) are the most common ferromagnetic metals.

Figure 2.2: (a) Magnetic moment orientation in a paramagnetic material in zero applied field. (b) Magnetic flux lines ‘B’ passing through a paramagnetic material in the presence of an applied magnetic field are attracted towards the material core. (c) The Susceptibility ($\chi = M/H$) decreases with increasing temperature.

\[ \chi_{\text{para}} = \frac{c}{T} \]
The Exchange interaction

Weber first suggested that every atom has its own magnetic moment which is present even in the absence of external field. It was later explained by Ampere that atomic magnetic moments arise because of the continuously circulating electric currents within the atoms. Magnetic moments in atoms are associated with orbital motion (L) and spin (S) of electrons as shown in figure 2.4. The sum of total spin and total orbital angular momentum of the electrons gives the total angular momentum (J) [21].

A magnetic moment experiences a torque in the presence of an external magnetic field. The resulting force is perpendicular to the applied field and the moment, and in an undamped system the moment precesses around the field axis. In a damped system the precessing moment eventually relaxes until it is parallel to the applied field.

Figure 2.3: (a) Magnetic moments orient parallel to each other in a single domain of a ferromagnetic material in zero applied field. (b) Magnetic lines of force passing through a ferromagnetic material in the presence of applied magnetic field originate in the material core and hence $B_{in} >> B_{out}$. (c) When the temperature is greater than Curie temperature ($T_c$) of the ferromagnet, the susceptibility ($\chi$) decreases and the material behaves as a paramagnet.

2.2 The Exchange interaction

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Several energies compete over different length scales to control the ordering of magnetic moments [22-24]. At the range of the inter-atomic spacing, the exchange interaction dominates. In ferromagnets this leads to parallel alignment or antiparallel alignment in antiferromagnets and the characteristic energy is the exchange energy. The exchange interaction will attempt to make the neighbouring magnetic moments lie parallel to one another if the material is ferromagnetic (or antiparallel to one another if antiferromagnetic), due to the overlapping of wave functions and Pauli’s exclusion principle.

Let us consider two electrons on adjacent atoms at a position $\hat{r}_1$ and $\hat{r}_2$ as shown in figure 2.5. A joint two-electron wave function is constructed from the product of one-electron wave functions $\psi_1(r_1)$ and $\psi_2(r_2)$. The overall electron wave function must be an antisymmetric so that when the spatial state is symmetric, the spin part of wave function will be in the antisymmetric singlet state ($\chi_S$) or when the spatial state is antisymmetric, the spin part of the wave function will be in the symmetric triplet state ($\chi_T$).
The singlet case (\(\Psi_s\)) and triplet (\(\Psi_T\)) case wave functions can be written [21] as,

\[
\Psi_S(r_1r_2) = \frac{1}{\sqrt{2}}[\psi_1(r_1)\psi_2(r_2) + \psi_2(r_1)\psi_1(r_2)]\chi_S
\]

(2.1)

Figure 2.6: Electron wave function for spins in antisymmetric singlet state

\[
\Psi_T(r_1r_2) = \frac{1}{\sqrt{2}}[\psi_1(r_1)\psi_2(r_2) - \psi_2(r_1)\psi_1(r_2)]\chi_T
\]

(2.2)

Figure 2.7: Electron wave function for spins in symmetric triplet state

Then, assuming that \(\chi_S\) and \(\chi_T\) are normalised, the energy of the two states are given by

\[
E_S = \int \Psi_S^* \hat{H}\Psi_S dr_1dr_2,
\]

(2.3)

\[
E_T = \int \Psi_T^* \hat{H}\Psi_T dr_1dr_2,
\]

(2.4)

where \(\hat{H}\) is the full Hamiltonian for the system.

Singlet and triplet states can be parametrised by the dot product \(S_1 \cdot S_2\) where \(S_1 \cdot S_2\) for the singlet state is equal to (-3/4) and for a triplet state it is (1/4). The effective Hamiltonian is then given by,

\[
\hat{H} = \frac{1}{4}(E_S + 3E_T) - (E_S - E_T)S_1 \cdot S_2.
\]

(2.5)

While the first term is a constant and can be absorbed into another constant energy term, the second term is dependent on the spin orientation. The exchange constant, \(\mathcal{J}\), is then defined as [21],

15
\[ J = \frac{E_S - E_T}{2} = \int \psi_1^*(r_1) \psi_2^*(r_2) \hat{H} \psi_1(r_2) \psi_2(r_1) \, dr_1 \, dr_2 . \quad (2.6) \]

Therefore the effective exchange Hamiltonian can be written as,

\[ \hat{H}_{\text{exch}} = -2 J S_1 \cdot S_2 \quad (2.7) \]

Using Heisenberg’s model, the exchange energy is described in an effective Hamiltonian [21],

\[ E = -2 J S_1 S_2 \cos \varphi_{12} , \quad (2.8) \]

where \( S_i \) is the spin operator of the \( i^{\text{th}} \) atom and \( \varphi_{12} \) is the angle between two adjacent magnetic moments. When \( J \) is positive and greater than 0, \( E_S > E_T \), and the favoured state is a triplet state \( S = 1 \) with parallel alignment of spins. When \( J \) is negative and less than 0, \( E_T > E_S \), and the favoured state is a singlet state \( S = 0 \) with antiparallel alignment of neighbouring spins. Due to the exchange interaction the material gains a spontaneous magnetization below the Curie temperature and this results in the development of a net magnetization per unit volume.

### 2.3 Ferromagnetism in metals

In transition metals the overlapping of wave functions of neighbouring atoms gives rise to a wide 4s energy band and a narrow 3d electron band due to the stronger localization of 3d orbitals [28]. Transfer of electrons from spin down to spin up states arises due to the exchange coupling in order to minimise the energy of the system (with more spins aligned parallel). According to Pauli’s exclusion principle these electrons have to fill up unoccupied majority spin states, leading to an increase in the kinetic energy (K.E). At equilibrium this rise in K.E is balanced by the decrease in exchange energy. This leads to an overall reduction of the total energy of the system and stabilises the ferromagnetic state. Stoner formulated this concept in his criterion for ferromagnetism, given by [28],

\[ J \cdot g_{\uparrow\downarrow}(E_f) > 1 , \quad (2.9) \]

where \( J \) is the exchange coupling and \( g_{\uparrow\downarrow}(E_f) \) is the spin up (down) density of states.
The exchange interaction is often introduced through a fictitious “exchange” magnetic field \( H_{\text{exch}} \).

\[
H_{\text{exch}} = E_{\text{exch}}/2\mu_0\mu_B.
\]  

(2.10)

The spin dependent orientation of magnetic moments in a ferromagnet is influenced by various energy terms like the exchange energy, magnetocrystalline anisotropy, Zeeman energy, etc.

### 2.4 Magnetic anisotropy

The dependence of magnetic properties on direction is called magnetic anisotropy. Magnetic anisotropy often arises from the crystallographic structure (magnetocrystalline anisotropy) or the physical dimensions of samples (shape anisotropy) [24]. Magnetic anisotropy can also be induced by mechanical strain leading to elongation/contraction of interatomic distances, called magneto-elastic anisotropy (magnetostriction) [24].

#### 2.4.1 Magnetocrystalline Anisotropy

The specific atomic lattice structure influences the total energy of the magnetized body. A ferromagnet has ‘easy’ crystallographic axes along which it can easily be magnetised. Magnetocrystalline anisotropy is the energy required to change the magnetization from the easy axis to the given magnetization direction. This anisotropy arises due to spin-orbit coupling, which couples the spin magnetic moments with the crystal lattice.
For hexagonally closed packed structures like cobalt, uniaxial anisotropy is observed with the easy magnetization direction along the ‘hexagonal’ c-axis.

\[
E_a = K_{u1} \sin^2 \theta ,
\]

where \(K_{u1}\) is the uniaxial anisotropy constant and \(\theta\) is the angle between the magnetization direction and the ‘hexagonal’ c-axis.

For cubic crystals, the anisotropy is much more complex and is often expressed in terms of the internal magnetization directions with respect to the crystallographic axes through the respective direction cosines. This is usually represented by a polynomial series.
\[ E_a = K_1 \left( \cos^2(\theta_x) \cos^2(\theta_y) + \cos^2(\theta_x) \cos^2(\theta_z) + \cos^2(\theta_y) \cos^2(\theta_z) \right) + \]
\[ K_2 \left( \cos^2(\theta_x) \cos^2(\theta_y) \cos^2(\theta_z) \right) \]  
(2.12)

where \( K_1 \) and \( K_2 \) are anisotropy constants. Depending on the sign of \( K_1 \), the energy \( E_a \) will be minimum in the <111> crystallographic direction (\( K_1 < 0 \)) or in the <100> direction (\( K_1 > 0 \)). For Fe (BCC), \( K_1 = +4.8 \times 10^4 \) J/m\(^3\) leading to <100> easy axes while for Ni (FCC), \( K_1 = -4.5 \times 10^3 \) J/m\(^3\) leading to <111> easy axes.

### 2.4.2 Shape Anisotropy

Any magnetic material has magnetic anisotropy due to its shape. Shape anisotropy has its origins in the demagnetizing field within the material. The demagnetization field opposes the magnetization of the material that produces it and will be smaller along the long axis of a non-uniform sample creating an easy axis in this direction.

By definition: \( H_d = -N_a M \), where \( N_a \) is the demagnetizing factor

![Figure 2.11: A thin film sample having in-plane magnetization due to shape anisotropy](image)

Hence we can write the total magnetostatic energy \( (E) \) per unit volume is

\[ E = \frac{1}{2} \mu_0 M_s^2 V \left( N_x m_x^2 + N_y m_y^2 + N_z m_z^2 \right), \]  
(2.13)

where \( V \) is the volume, \( M_s \) is the saturation magnetisation and \( N_x, N_y, N_z \) are the demagnetizing factors along different directions. In the case of a thin film lying perpendicular to the \( z \)-axis, \( N_x \approx N_y \approx 0 \) and \( N_z \approx 1 \).

In a uniformly magnetized structure, shape anisotropy will result in a field dependence of the magnetization direction. The magnetic dipolar interaction, whose contributions depend
on the shape of the sample, mediates the magnetic dipolar or shape anisotropy of the material. In the case of thin films, e.g. Co/Pt multilayers, shape anisotropy normally dominates and aligns the magnetic moments along the in-plane direction, regardless of crystallographic orientation.

In thin films, the shape anisotropy energy per unit volume is approximately given by [29]

\[ E = \frac{1}{2} \mu_0 M_s^2 \cos^2 \theta, \]  

(2.14)

where \( \theta \) is the angle between the magnetization direction and the normal to the plane of the sample.

### 2.4.3 Zeeman Energy

The Zeeman energy is another energy term that contributes in addition to the anisotropy energy when an external field is applied. It arises from the interaction between the magnetisation of the sample and the applied external field. The Zeeman energy is minimum when the magnetisation lies parallel to the external magnetic field.

\[ E_{\text{Zeeman}} = -\mu_0 H M_s \cos(\phi), \]  

(2.15)

where \( \phi \) is the angle between the magnetisation and the applied field (H).

### 2.4.4 Magneto-elastic anisotropy

Applying an external field to a magnetic material will result in elongation/change in length of the material in the applied field direction. This is called magnetostriction (\( \lambda \)).

\[ \lambda = \frac{\Delta l}{l}, \]  

(2.16)

where \( \Delta l \) is the elongation length of the material and \( l \) is the initial length of the material before strain.
In a saturated state, the magnetisation is aligned parallel to the applied field and so as the strain is as shown in figure 2.12. This is given by the expression,

\[ \lambda = \frac{2}{3} e, \]  

(2.17)

where \( e \) is the spontaneous strain within the material due to magnetisation along a particular direction.

In practical applications all solids exhibit a certain degree of anisotropy and therefore it is necessary to define \( \lambda \) in relation to the crystal axis along which the magnetisation lies. For cubic structure materials, magnetostriction is given by

\[ \lambda = \frac{3}{2} \lambda_{100} \left( \alpha_1^2 \beta_1^2 + \alpha_2^2 \beta_2^2 + \alpha_3^2 \beta_3^2 - \frac{1}{3} \right) + 3 \lambda_{111} (\alpha_1 \alpha_2 \beta_1 \beta_2 + \alpha_2 \alpha_3 \beta_2 \beta_3 + \alpha_3 \alpha_1 \beta_3 \beta_1), \]  

(2.18)

where \( \lambda_{100} \) and \( \lambda_{111} \) are magnetostriction constants measured along the \( <100> \) and \( <111> \) directions. \( \alpha_1, \alpha_2, \alpha_3 \) are direction cosines for the axis along which magnetisation is saturated and \( \beta_1, \beta_2, \beta_3 \) are direction cosines along the magnetostriction direction (c.f. figure 2.13).
In the case of magnetostriction along the same direction as the applied field, then \( \alpha_i = \beta_i \). Then Eq. 2.18 becomes

\[
\lambda = \lambda_{100} + 3(\lambda_{111} - \lambda_{100})(\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2) \quad (2.19)
\]

In the reverse process to magnetostriction, strain in a magnetic material will induce a change in magnetisation direction. The spin magnetic moments are coupled to the lattice structure through orbital electrons. Stain in a magnetic material will elongate/reduce the atomic spacing of the lattice structure changing the spin-orbit interaction energies and this creating magneto-elastic anisotropy. The magneto-elastic energy under the application of a field is given by

\[
E_{ME} = -\frac{3}{2} \lambda \sigma \left( \cos^2 \theta - \frac{1}{3} \right), \quad (2.20)
\]

where \( \sigma \) is the stress induced and \( \theta \) is the angle between \( \sigma \) and \( M_s \).

### 2.4.5 Magnetic anisotropy in Co/Pt multilayers

Magnetic thin films grown on crystalline substrates will be subject to interface interactions that can change the magnetization direction of the film with respect to a similar free standing layer. Cobalt at room temperature has a hexagonally closed packed (hcp) structure and can be grown epitaxially on a fcc <111> platinum/palladium buffer by choosing a suitable underlying substrate and optimising the growth conditions. Since the discovery of perpendicular magnetic anisotropy (PMA) in Co/Pt multilayers (MLs) \[30\], extensive research reports have been published on its origins with a focus on the film morphology and crystal structure \[30-34\]. Using x-ray magnetic circular dichroism (XRMCD) Nakahima et al., \[34\] observed a strongly enhanced perpendicular Co orbital moment in Co/Pt MLs. This out-of-plane anisotropy arises due to spin-orbit coupling. This is because of a strong increase in orbital moment in thin Co films of less than 6 to 8 monolayer thickness due to the strong interfacial 3d-5d hybridisation between Co and Pt atoms and also the hcp structure has greater orbital moment. The results of these are presented in \[35\]. Furthermore, spin polarisation also induces a substantial Pt orbital moment that favours PMA. Due to the manner of interfacial hybridisation and spin polarisation, the intermixing, roughness and possible alloy formation at the Co/Pt interface plays a crucial role for PMA in Co/Pt multilayer system. In addition to the
interfacial term, magnetocrystalline anisotropy due to the <0001> Co texture also contributes to the effective anisotropy in the multilayer sample.

The anisotropy energy \( E_{an} \) of a thin ferromagnetic layer with PMA is approximated by,

\[
E_{an} = K_{eff} \sin^2 \theta ,
\]

where \( K_{eff} \) is the effective anisotropy constant and \( \theta \) is the angle between the surface normal and the magnetisation direction. The effective anisotropy is expressed by Draaisma \textit{et al.},\cite{36} as,

\[
K_{eff} = \frac{2K_s}{t} + K_V - \mu_o M_s^2 ,
\]

where \( K_s \) is the surface anisotropy constant, \( K_V \) is the bulk volume anisotropy constant and \( M_s \) is the saturation magnetisation. At the surface of thin films the symmetry of atoms is not the same as those in the bulk phase of the films. This reduced symmetry gives rise to the surface/interface anisotropy \( K_s \) that has an effect on the magnetisation easy axis. The second term \( K_V \) is the volume anisotropy which is the combination of magnetoelastic anisotropy, \( K_{ME} \) and magnetocrystalline anisotropy, \( K_{MC} \). \( K_{ME} \) is largely due to the tensile strain in the Co layer. Strain is another important parameter that contributes to the PMA. The strain in the Co layer arises from the misfit of Co and Pt atoms at the interface and is often seen in magnetoelastic anisotropy. The last term in the Eq. 2.22 is the shape anisotropy term which is due to the dipolar contributions and can be greatly reduced by the presence of interfacial roughness.

The effect of crystallographic orientation on the easy axis direction was seen by Lin \textit{et al.}\cite{33} whereby the film texture can lead the easy axis to lie along the plane of the film even if the interfacial anisotropy is in the out-of-plane direction. In this case, epitaxially grown Co(3.7Å)/Pt(16.8Å) films with three different Pt seed layer orientations <111>, <110> and <100>, were used. With the exception of MLs having Pt <111> orientation, they all exhibited an easy axis parallel to the plane of the sample. This was confirmed with polar MOKE measurements in a perpendicular magnetic field, where MLs with Pt <111> seed layers exhibit a square hysteresis loop while the rest display clear in-plane magnetization. Magnetocrystalline anisotropy has to be optimised during Co/Pt ML growth and a Pt seed layer with <111> texture is ideal to realise strong PMA.
2.5 Domain formation

In ferromagnets, uniform magnetic ordering exists within small region called domains. Each domain has its own net magnetic moment which will in general differ from nearby domains. In a fully demagnetised sample all these net moments sum approximately to zero. Weiss built on the previous work of Weber, Ampere and Ewing to suggest the existence of domains in ferromagnets.

The magnetisation orientation within a domain and the size of the domain is dependent on the different energies of the material, e.g., the exchange energy, magnetostatic energy and magnetocrystalline anisotropy. A magnet, say rectangle in shape as shown in fig 2.14, usually consists of a single domain having high magnetostatic energy. By splitting this into two magnetic domains with opposite magnetisation, a domain boundary or domain wall is formed. The magnetostatic energy is greatly decreased by the reduction in the stray fields represented by flux lines that run between the poles of the magnet. The number of flux lines flowing outside and through the magnet has now decreased because these lines are confined to the end poles of adjacent domains. Breaking the larger domains into more domains leads to an increase in the number of domain walls being formed. The exchange energy and anisotropy energy are the two contributors to a domain wall. A larger number of domain walls means an increase in exchange and anisotropy energy while the magnetostatic energy of the material decreases further. Indeed the formation of 90° domain walls can lead to a fully flux closed structure with no magnetostatic energy. This is likely to be found in materials with cubic anisotropy. However forming domain walls does cost additional energy.

![Diagram showing domain formation](image)

Figure 2.14: Schematic diagram showing how the creation of domains can reduce the stray fields, reducing the magnetostatic energy. This is the main driving force for the formation of domains
2.6 Domain walls (Bloch, Néel)

The region separating the domains where the direction of magnetisation changes are called domain wall (c.f. fig 2.15). In magnetic nanostructures domain walls commonly exist at sharp bends, asymmetric & symmetric notches [37] and nearby dots and at any irregular shapes in the nano structure. There commonly exist two types of 180° domain walls with different spin structures called Bloch and Néel walls [23, 24].

A Bloch wall is shown in figure 2.16 (a) and is a narrow transition region between the adjacent magnetic domains, where the magnetization direction rotates in a plane parallel to the plane of the wall. In a Néel wall the magnetic moments rotate perpendicular to the plane of the wall. The Néel wall (c.f. fig 2.16 (b)) consists of a narrow core with long tails on both sides. Neel walls are found in in-plane magnetised thin films due to the exchange length being greater than the material thickness while Bloch walls are generally found in thicker films.

The exchange energy and the anisotropy energy combine to govern the total energy per unit area of the domain wall (σ).
\[ \sigma_{BW} = JS^2 \frac{\pi^2}{Na^2} + \frac{NKa}{2} \]  \hspace{1cm} (2.23)

Eq. 2.23 is valid for a Bloch wall, where \( J \) is the exchange integral, \( S \) is the net spin, \( N \) is the number of sites over which the spin undergoes a rotation of \( \pi \), \( K \) is the anisotropy constant and \( 'a' \) is the lattice spacing. The first term is the contribution of the exchange energy and the second is the anisotropy energy. The first term, proportional to \( J/N \), tends to unwind the DW making it wider and the second term, proportional to \( N \), tends to tighten the DW and make it narrower. The value of \( N \) can be derived for the equilibrium configuration solving \( d\sigma_{bw}/dN = 0 \),

\[ \therefore N = \pi S\sqrt{2J/Ka^2}. \]  \hspace{1cm} (2.24)

The contributions of the exchange energy and the magnetocrystalline energy balance each other and establish the width of the domain wall (\( \delta_{BW} \)).

\[ \delta_{BW} = Na = \pi S \frac{2J}{\sqrt{K}a}. \]  \hspace{1cm} (2.25)

For large values of \( J \) the DW gets wider and for large values of \( K \) it becomes smaller. The DW energy per unit area is given by,

\[ \sigma_{BW} = \pi S \frac{2J}{\sqrt{K}a}. \]  \hspace{1cm} (2.26)

An analytic continuum approximation for the DW energy yields the DW width (\( \delta_{BW} \)) and DW energy per unit area (\( \sigma_{BW} \)) in an alternate form

\[ \delta_{BW} = \pi \frac{A}{\sqrt{K}}, \]  \hspace{1cm} (2.27)

\[ \sigma_{BW} = \pi \sqrt{AK}, \]  \hspace{1cm} (2.28)

where \( A \) is the exchange stiffness constant.

Wide walls are favoured by the exchange energy which tries to align adjacent magnetic moments as close to parallel as possible, whereas magnetocrystalline anisotropy promotes rapid changes in the transition between easy axis directions thereby making moments point along the hard axis directions as little as possible.
In our Co/Pt films we expect to see a $180^\circ$ Bloch like domain wall structures including where we have created DWs by Ga$^+$ irradiation followed by a magnetic field cycle. In such cases, the irradiated region has a lower anisotropy compared to the non-irradiated region [19, 38]. Hence the DW that is created upon minor loop magnetisation switching will have two different anisotropy energies contributing to the DW energy. In an ideal case, the DW is split into two, with one half in the high anisotropy region and the other half in the low anisotropy region.

![Diagram](image)

Figure 2.17: Half irradiated ferromagnetic material with an interface that hosts a DW upon undergoing a minor hysteresis loop. The pink region is the Ga$^+$ irradiated region and has a reduced anisotropy (anisotropy constant $K_1$). The blue area is non-irradiated part and has the original anisotropy (anisotropy constant $K_2$).

For the example shown in figure 2.17, the DW energy ($\sigma_{BW}$) will contain two anisotropy constant $K_1$ and $K_2$, corresponding to the irradiated and non-irradiated regions. In this case the anisotropy constant $K_1 < K_2$. The DW energy ($\sigma$) will draw half its energy from the non-irradiated part and the other half from the irradiated region.

$$\sigma_{BW} \approx \frac{\pi S}{2} \left( \sqrt{\frac{2JK_1}{a_1}} + \sqrt{\frac{2JK_2}{a_2}} \right). \quad (2.29)$$

2.7 Magnetic reversal and hysteresis

The magnetisation reversal process arises from a combination of magnetisation rotation within domains and domain wall motion. A plot of magnetisation as a function of applied field normally produces a hysteresis loop [c.f. fig 2.18] reflecting the hysteresis of DW motion or magnetisation rotation. A sample can reach magnetisation saturation $M_s$ by applying an
increasing field. When the field is reversed and brought to zero, the magnetisation is reduced to remanence or the remanent magnetisation, $M_r$. The magnetisation doesn’t fall to zero at this point and instead retains a value often only slightly reduced from saturation. To switch the magnetisation, a negative field is applied and when the magnetic field is equal to the coercive field, $H_c$, the magnetisation falls to zero. $M_r$ and $H_c$ are generally used to characterise a ferromagnetic material.

![Diagram of hysteresis loop](http://mxp.physics.umn.edu/s03/projects/s03moke/theory.htm)

**Figure 2.18:** Generalised hysteresis loop showing different magnetic states of a magnetic material. Reproduced from [http://mxp.physics.umn.edu/s03/projects/s03moke/theory.htm](http://mxp.physics.umn.edu/s03/projects/s03moke/theory.htm)

The lowest energy state is generally a demagnetised state at zero field containing many domains and DWs. Application of a (positive) field magnetises the films to saturation. First there is domain wall motion (DWM), i.e. domains whose magnetisation is aligned parallel to the applied field will grow at the expense of the domains with anti-parallel alignment. As the magnetic field is increased to higher values, there is a coherent rotation of domains towards the direction of the magnetic field and this takes place irrespective of the hard and easy axes of the films. Applying a reverse magnetic field, the magnetisation switching/reversal starts with the nucleation of domains and later by domain wall motion to complete the magnetisation reversal process. In the case of perpendicularly magnetised ferromagnetic thin films (e.g.
Co/Pt multilayers), magnetisation at remanence is generally fully saturated ($M_s$) and approximately remains so until $H_c$. Structural defects, impurities in the material and rough edges are common sites for domain nucleation centres. Domain nucleation can happen at one or multiple centres in the sample under reverse fields. It leads to the formation of reverse domains surrounded by DWs. The growth of the nucleated domain occurs by DW motion. In some cases, the domain nucleates from the sides or edges of the material and propagates in random directions. When the nucleated volume of reversed domains is equal to that of unreversed domains, the material is in a demagnetised state. The magnetisation will fall to zero at $H_c$ and will then become more negative until magnetisation reversal is complete.

The magnetisation curve for a single domain particle can be calculated using the Stoner-Wohlfarth model. This assumes an ellipsoidal particle shape so that the magnetisation is uniform throughout the sample. The easy axis is assumed to lie along the elongated axis of the particle.

For the geometry of Fig 2.19, the free energy of a Stoner-Wohlfarth particle is expressed as,

$$E = (K_1 \sin^2 \theta - \mu_0 MH \cos \theta)V,$$  \hspace{1cm} (2.30)

where $K_1$ is the uniaxial anisotropy constant and $\theta$ is the angle between the magnetisation ($M$) and the easy axis. The free energy has a minimum at $\theta = 0$. The magnetisation will only reverse when the applied reverse field is greater than or equal to the anisotropy field, $H_k$. When this condition is satisfied, the energy minimum at $\theta = 0$ becomes a maximum. The field at which the magnetisation falls to zero is the switching field and in this case gives rise to rotational hysteresis.

Figure 2.19: Schematic representation of easy axis magnetization and the associated M-H loop. The saturation states (double headed arrows) in the first and third quadrants indicate the reversible process while the single headed arrows indicate irreversible processes.
A positive second derivative of the free energy and $\partial E/\partial \theta=0$ for equation 2.30 gives the stability condition and the anisotropy field for locally stable magnetisation.

\[
\frac{\partial E}{\partial \theta} = 2K_1 \sin \theta \cos \theta + \mu_0 MH \sin \theta = 0, \quad (2.31)
\]

or \( \sin \theta (K_1 \sin 2\theta + \mu_0 MH) = 0 \).

\[
\frac{\partial^2 E_{\text{tot}}}{\partial \theta^2} = 2K_1 \cos^2 \theta + \mu_0 MH \cos \theta > 0. \quad (2.32)
\]

\[
\therefore H_k = \pm \frac{2K_1}{\mu_0 M}; \text{For } \theta = 0 \text{ and } \pi. \quad (2.33)
\]

A square hysteresis loop with high remanence generally results from the application of a field along the easy axis of the sample.

In the case of a field applied perpendicular to the easy axis of magnetisation, $\partial E/\partial \theta = 0$ and positive second derivative defines the energy minimum condition.

\[
E = K_1 \cos^2 \theta - \mu_0 M_s H \cos \theta, \quad K_1 > 0. \quad (2.34)
\]

\[
\frac{\partial E}{\partial \theta} = (-2K_1 \cos \theta + \mu_0 M_s H) \sin \theta = 0. \quad (2.35)
\]

Figure 2.20: Schematic representation of hard axis magnetization and the associated M-H loop

Two solutions arise from this condition. The one of interest is

\[
2K_1 \cos \theta = \mu_0 M_s H. \quad (2.36)
\]
For fields below saturation, eq. 2.36 describes the equation of motion for magnetisation. At saturation \( \cos(\theta) = 1 \), and this saturation field is defined as being equal to the anisotropy field (\( H_{\text{sat}} = H_k \)).

\[
\therefore H_k = \frac{2K_1}{\mu_0 M_s}.
\] (2.37)

When the field is increased from zero, a smooth rotation of the magnetisation direction takes place and the magnetisation along the field direction increases linearly up to the anisotropy field \( H_k \) where the material is saturated.

**Summary**

The fundamentals of ferromagnetism and magnetic properties such as magnetic anisotropy, exchange interaction and magnetisation process have been described. Domain formation and types of domain walls are explained in detail. PMA in Co/Pt multilayers and the domain wall type in Co/Pt MLs have been discussed. This chapter provides the background knowledge in ferromagnetism required to understand the experimental research work performed.
Chapter 3 Introduction to Spintronics

The driving forces behind greatly increased efforts in the research and development of nanostructured magnetic materials and nanoparticles include ultra-high density magnetic storage devices [39], magnetic random access memory (MRAM) [40], spintronics [41], magnetic semiconductors [42] and nano-granular magnetic materials [43]. Domain walls in ferromagnetic nanostructures also have potential applications in spintronic devices. In solid state physics spintronics refers to the study of the electron spin devices that exploit its spin properties rather than just its charge. There are two main physical principles underlying the interest in spintronics. They are the inherent quantum mechanical nature of spin as a dynamical variable (leading to the possibility of novel quantum spintronic devices which are not feasible with present day charge-based electronics) and the inherently long relaxation or coherence times associated with spin states (compared with their momentum states). The operation [44, 45] of spintronic devices relies on the magnetic configuration (parallel or antiparallel) of ferromagnetic layers which control the flow of charge carriers having opposite spin orientation.

The primary requirement for making a spintronic device is to have a system that can generate a current of spin polarized electrons and a system which is sensitive to the spin polarization of the electrons. The spin polarization is defined as net fractional moment parallel to the quantization direction [46]. Electrons passing through a magnetic material will have their spins aligned parallel to the local magnetisation and when it leaves the magnetic material it generates a spin polarised current. In order to fully comprehend the behaviour of spintronic devices, it is necessary to understand the properties and propagation of domain walls [47-50]. The motion of domain walls in spintronic devices can be achieved by applying pulsed electric currents instead of a magnetic field, resulting in improved device performance. Whenever an electron passes through a domain wall, the direction of the electron spin will change, leading to transfer of angular momentum from electrons in the domain wall and a resulting torque leading to domain wall motion [51-53]. This torque is called the ‘Spin Transfer Torque (STT)’

Key outstanding issues for fully exploiting the spintronic devices include understanding and controlling the behaviour of domain wall motion and large critical currents to induce domain wall motion. For example, a domain wall may propagate and get pinned [54, 55], change its structure [56] or be annihilated [57], depending on a number of factors. Therefore it is necessary to find a way to control the domain wall motion (velocity), its structural stability and its time dependent behaviour. In the rest of this chapter, the principles
of control and measurement of domain wall motion in artificially created domains will be discussed.

3.1 Basic principles

The electrical resistance of a magnetic material depends on the relative orientation of the local magnetic moments or magnetization of the domain with respect to the current direction. Using Drude transport theory we can directly relate the current density to the electric field to obtain an expression for electrical conductivity \( \sigma \) \([24]\).

\[
J = \left( \frac{ne^2\tau}{m^*} \right) E, \tag{3.1}
\]

\[
\sigma = \frac{ne^2\tau}{m^*}, \tag{3.2}
\]

where \( \sigma \) is the electrical conductivity, \( n \) is the carrier density, \( e \) is the electronic charge, \( \tau \) is the scattering rate and \( m \) is the carrier mass. The electrical resistivity \( \rho \) is given by

\[
\rho = \frac{1}{\sigma}. \tag{3.3}
\]

We can apply this to spin polarised ferromagnetic systems in which the resistance increases when the ferromagnetic material is heated above Curie temperature. This is explained using Mott’s Two Current Model which approximately describes the resistance of ferromagnetic materials \([1, 58]\).

Mott first recognised that at temperatures well below the Curie temperature, the spin direction of the electrons is conserved in most scattering events, that is spin-up to spin-down transitions or vice versa are most unlikely. This is because spin-waves, which mix spin-up and spin-down states, are not excited at low temperatures. Thus the spin-up and spin-down charge carriers can be represented as two parallel paths along which conduction can take place – as shown in the two current model in fig 3.1. The 4s electrons have a low effective mass compared to 3d electrons and hence the 4s electrons dominate conductivity \([58]\).
The spin-up and spin-down electrons have different scattering rates irrespective of the nature of scattering centres. This is due to the fact that the density of states of the spin-up and spin-down electrons at the Fermi energy is different as the band structure of the ferromagnet is exchange split (c.f. figure 3.2). Since scattering rates are proportional to the density of states, it is clear that the scattering rates and resistivities are different for electrons with different spins. Figure 3.2, it is seen that the spin-up electron has a completely filled electrons in 3d state, while the spin-down electron band is partially occupied leading to spin dependent s-d scattering. Commonly the majority charge carriers will be the spin-up electrons and will experience negligible amounts of s-d scattering. Hence they have a relatively low resistivity as compared to minority electrons. The density of states for the spin-up and spin-down electrons are equal in non-magnetic metals.

Figure 3.1: Mott - two current model, where $\rho_{ss}$ and $\rho_{sd}$ are the resistivity due to s-s and s-d scattering respectively.

Figure 3.2: Energy band structure is an exchange split ferromagnetic metal
The resistivities of spin-up ($\rho^\uparrow$) and spin-down ($\rho^\downarrow$) charge carriers are given by,

$$\rho^\uparrow = \rho_{ss}^\uparrow + \rho_{sd}^\uparrow,$$  \hspace{1cm} (3.4a)

$$\rho^\downarrow = \rho_{ss}^\downarrow + \rho_{sd}^\downarrow.$$  \hspace{1cm} (3.4b)

The total resistivity ($\rho$) of a metal in a two-current model is then given by,

$$\rho = \frac{\rho^\uparrow \rho^\downarrow}{\rho^\uparrow + \rho^\downarrow}.$$ \hspace{1cm} (3.5)

### 3.2 Magnetoresistance

Charge carriers in a conductor collide with defects and impurities to cause resistance, and in a magnetic field the number of collisions increases due to the magnetic force acting on them. This dependence of resistance on the magnetic field is called magnetoresistance.

#### 3.2.1 Giant magnetoresistance (GMR)

Magnetoresistance in ferromagnetic materials was found to be quite small and the need for much larger magnetoresistance effects led to the discovery of ‘Giant Magnetoresistance (GMR)’ in the late 1980s [59, 60]. The discovery of GMR had a major technological impact on storage devices. When a magnetic field is applied, the magnetic moments of two separated ferromagnetic layers are aligned parallel along the field direction and the resistance of the multilayer structure drops to a minimum. This is due to lower scattering of majority (spin-up) charge carriers as they are the majority carriers in both ferromagnetic layers. When the magnetisation of the multilayers are aligned anti-parallel then the majority charge carriers in one layer becomes minority carriers in the other and the overall resistance is high. This phenomenon gives rise to giant magnetoresistance (GMR). This phenomenon is shown schematically in figure 3.3.
The overall spin-dependent scattering is higher when a majority carrier in one layer becomes a minority spin in the other layer. In each layer the mean free path of the majority spin channel will be longer, and resistivity lower, than for the other spin channel. Figure 3.4.a shows a trilayer with two ferromagnetic layers whose magnetisation is parallel. In such a system, the majority spin electrons whose spin is aligned parallel to the ferromagnet magnetisation experiences little scattering and has low resistance. Minority spin electrons will have high scattering rates and hence high resistance.
Figure 3.4: (a) Schematic representation of electron scattering in a ferromagnetically coupled multilayer system (b) A schematic diagram of the density of states showing the transfer of charge carriers from one ferromagnetic layer to another. Red arrows represent the high scattering/resistance path.

This can be explained in terms of the density of states at the Fermi level as shown in figure 3.4(b). Due to the exchange interaction (field) in ferromagnetic materials, the spin-up and spin-down electron bands are shifted in energy leading to spin-dependent scattering of carriers. The spin-up electrons are the majority charge carriers and pass through the ferromagnetic layers (FM$_1$ and FM$_2$) and non-magnetic layer (NM) unaffected. The spin-down electrons suffer much more scattering and exhibit much higher resistance than the spin-up ones. Hence the spin-up charge carriers generally dominate the electronic transport. The resistance of the spin-up and spin-down electron paths depends on the relative magnetisation of the two ferromagnetic layers. In ferromagnetically aligned multilayers the resistance due to electron scattering is represented in figure 3.5 when one spin direction has low resistance ($r$) and the other high resistance ($R$).
The total resistance ($R_{\text{parallel}}$) experienced by the charge carriers in the ferromagnetically coupled circuit is

$$R_{\text{parallel}} = \frac{(2r_1 + R_0)(2R_1 + R_0)}{2(R_1 + r_1 + R_0)}, \quad (3.6)$$

where $r_1$ is the low resistance ($R_1$ is the high resistance) of majority (minority) spins.

For anti-ferromagnetically aligned multilayers, conduction electrons of either spin always have one high resistance leg as they pass through the structure. Figure 3.6.a shows a trilayer with two ferromagnetic layers whose magnetisation is anti-ferromagnetically aligned. In such a system, the electrons encounter minority carrier scattering in at least one layer irrespective of its spin orientation. This can be seen in the density of states diagram in figure 3.6.b where minority (spin-down) moments have unfilled states in $\text{FM}_1$ promoting the scattering of spin-down electrons to take place, while the spin-down band is fully occupied in $\text{FM}_2$. The opposite is the case for majority spin carriers (spin-up). Spin-up electrons pass through $\text{FM}_1$ without scattering due to the completely filled 3d sub-band while strong scattering occurs in $\text{FM}_2$. 

Figure 3.5: Circuit representation of the conduction paths for spin-up and spin-down charge carriers in a ferromagnetically aligned multilayer. The resistances ‘r’ and ‘R’, represent the low and high values of majority and minority spins and $R_0$ is the resistance of the non-magnetic layer.
Figure 3.6: (a) Schematic representation of electron scattering in an anti-ferromagnetically aligned multilayer system (b) The parallel paths of spin-up and spin-down electrons. Green arrows represent low scattering/resistance paths and red arrow represent high scattering/resistance paths.

In the anti-parallel magnetisation configuration each spin channel will act as a majority carrier in half the structure and as a minority spin in the other half. A circuit representation of the resistances due to scattering of spin-up and spin-down electrons is shown in figure 3.7. Majority electrons in FM1 (c.f. fig 3.4.a) experience low scattering rates but strongly scattered in FM2. This leads to an overall increase in scattering rates and higher resistance for spin-up charge carriers. The same processes take place for spin-down charge carriers but in reverse.

Figure 3.7: Circuit representation of the conduction paths of spin-up and spin-down charge carriers in an anti-ferromagnetically aligned multilayer. The resistances ‘r’ and ‘R’, represents the low and high values of majority and minority spins and $R_0$ is the resistance of the non-magnetic layer.
In this case, the total resistance \( R_{\text{antiparallel}} \) experienced by the charge carriers in the anti-ferromagnetically coupled circuit is

\[
R_{\text{antiparallel}} = \frac{1}{2} (R_{\downarrow} + r_{\uparrow} + R_{0}),
\]

where \( r_{\uparrow} \) is the low resistance (\( R_{\downarrow} \) is the high resistance) of majority (minority) spins.

The GMR ratio is then defined by,

\[
GMR = \frac{R_{\text{antiparallel}} - R_{\text{parallel}}}{R_{\text{antiparallel}}} = \frac{(R_{\downarrow} - r_{\uparrow})^2}{(R_{\downarrow} + r_{\uparrow} + R_{0})^2}
\]

Normally current is applied either in the plane (current in-plane – CIP) or perpendicular to the plane (current perpendicular to plane – CPP) of the structure. In the case of CIP-GMR, the electronic mean free path is much longer than the thickness of the layers and carriers hence cross the multilayer interfaces many times as they drift along the in-plane direction.

### 3.2.2 Anisotropic magnetoresistance (AMR)

In ferromagnetic materials, the magnetoresistance is a function of the applied field strength and the magnetisation, and the resistivity \( \rho \) depends on the angle \( \phi \) between the direction of current \( \vec{j} \) and the magnetisation \( \vec{M} \) directions. The resulting anisotropic magnetoresistance (AMR) has its origins in the spin-orbit interaction since there is a higher probability of s-d scattering of electrons in the magnetisation direction. AMR is large in 3-d transition metals like Ni-Fe and Ni-Co and can be greater than 5% [61]. When the direction of current and magnetisation are parallel, the electrical resistance has a maximum value while the resistance is low when they are perpendicular to each other (c.f. fig 3.8). The angle, \( \phi \), between the magnetisation and current determines the strength of AMR in the material.
Figure 3.8: Anisotropic magnetoresistance (AMR) is (a) low and (b) high depending on the angle, $\varphi$, between the current and the magnetisation (c) Plot sketching resistance as a function of angle, $\varphi$.

The AMR resistivity $\rho(\varphi)$ is generally well approximated by,

$$\rho(\varphi) = \rho_\perp + (\rho_\parallel - \rho_\perp)\cos^2 \varphi$$  \hspace{1cm} (3.9)

where $\rho_\perp$ and $\rho_\parallel$ are the resistivities when the current and magnetisation are oriented perpendicular and parallel to one another respectively.

### 3.3 Extraordinary Hall effect (EHE)

An electrical conductor carrying a current in a magnetic field normal to its plane, will produce a voltage difference perpendicular to the current direction as charges move to one side of the conductor under the Lorentz force. This is known as the ordinary Hall effect and the voltage measured is called the Hall voltage. A schematic picture of the Hall effect in an out-of-plane field is shown in figure 3.9.
The ordinary Hall electric field $E_H$, when a magnetic induction is applied perpendicular to the current is described by

$$\vec{E}_H = R_H (\vec{J} \times \vec{B}),$$  
(3.10)

where $R_H$ is the Hall coefficient and $\vec{J}$ is the current density.

This has its origin in the Lorentz force,

$$\vec{F} = q(\vec{v} \times \vec{B}),$$  
(3.11)

and the relation,

$$\vec{J} = ne \vec{v},$$  
(3.12)

where $n$ is the electron density and $\vec{v}$ is the carrier velocity. Comparing this with Eq. 3.9, we identify the Hall coefficient as

$$R_H = (ne)^{-1}. \quad (3.13)$$

The Hall resistivity $\rho_H$ which describes the Hall Effect is

$$\rho_H = \frac{E_H}{J_x} = R_H B.$$

(3.14)

In the case where the electrical conductor is a ferromagnetic material there is an extra transverse field called the Extraordinary Hall Effect (EHE). EHE signals often have a very good signal-to-noise ratio in perpendicularly magnetised materials and can be directly correlated with the magnetization of the underlying ferromagnetic material. This makes it a valuable indirect way to measure the magnetization of a material. EHE is usually measured in
a planar structure as shown in figure 3.10, with a magnetic field applied perpendicular to the sample plane.

![Diagram of a planar structure with a magnetic field](image)

Figure 3.10: schematic representations of the Extraordinary Hall Effect in a Hall cross structure with a change in magnetisation reflecting the applied magnetic field direction.

With a constant current $I$ flowing in the current arms of the Hall cross, the measured Hall voltage in the voltage arms has an ordinary Hall component and an Extraordinary Hall component. The Hall electric field is given by

$$
\bar{E}_{\text{Hall}} = \rho_{\text{OHE}} \bar{B} \times \bar{J} + \rho_{\text{EHE}} \mu_0 \bar{M} \times \bar{J}.
$$

Here $\rho_{\text{OHE}}$ is the ordinary Hall resistivity and $\rho_{\text{EHE}}$ is the extraordinary Hall resistivity. The former is due to the Lorentz force experienced by the charge carriers and is proportional to the applied magnetic induction ($B$). $\rho_{\text{EHE}}$ is directly proportional to the magnetisation [62] and is generally much larger than $R_{\text{H}}$ at low fields. EHE arises from the spin-orbit interaction between the electrons and local magnetic moments. Charge carriers carry an orbital angular momentum $L$ about a scattering centre whose sign depends on whether the charge carrier passes either to the left or to the right of the centre. Due to the spin orbit coupling proportional to $L.S$, which is an interaction between the electron spin and the orbital part of electron’s wave function in an atom, charge carriers find it energetically more favourable to pass through one specific side of the scattering centre. This leads to asymmetrical scattering of charge carriers and the generation of a transverse electric field that is responsible for the EHE.

Two types of spin-orbit scattering are responsible for EHE, namely the skew scattering and side-jump scattering (c.f. fig 3.11). Skew scattering is the deflection of the electron from its original path when it strikes an impurity atom [63], while side jump scattering is the lateral displacement of the electron path upon striking an impurity atom [64].
Combining the two types of scattering, the extraordinary Hall coefficient can generally be expressed as

\[ \rho_{EHE} = a \rho + b \rho^2, \]  

(3.16)

where \( \rho \) is the resistivity and the first term represents skew scattering and the second term is attributed to side-jump scattering. This can in turn be expressed in terms of a spontaneous Hall angle \( \phi_{EHE} = \rho_{EHE}/\rho = \phi_{sk} + b \rho \), where \( \phi_{sk} \) refers to the skew scattering angle and \( b \rho \) relates to the side jump scattering.

Because of the small value of \( \rho \) in pure metals the skew scattering is generally the major contribution to the extraordinary Hall component, while for large values of \( \rho \) the side-jump scattering starts to dominate. In these limiting cases Eq. 3.16 becomes,

\[ R_{EHE} = c \rho^n, \]  

(3.17)

where \( n=1 \) for skew scattering and \( n=2 \) for side-jump scattering.

EHE is often large in perpendicularly magnetised thin film multilayers containing metals like Co, Pt, Au and rare earth metals that have a strong spin-orbit interaction. EHE measurements
of PMA thin films in a perpendicular magnetic field will generally produce square hysteresis loops, like the one in figure 3.12, reflecting the easy axis switching of the sample.

Figure 3.12: Hysteresis loop of a ferromagnetic multilayer (Ta/Pt/Co/Pt) obtained from an EHE measurement

3.4 Spin transfer torque
Spin transfer torque (STT) is best explained in the case of two ferromagnetic layers (F1 and F2) separated by a non-magnetic layer, whose magnetisation directions ($\vec{M}_1$ and $\vec{M}_2$) are misaligned at an angle $\theta$. This type of a model is schematically represented in figure 3.13. The nonmagnetic layer can be a metal, e.g. Copper, or a dielectric material, e.g. MgO. When an electric current flows perpendicular to the plane of the magnetic layers, the electrons passing through the interfaces and across the layers are spin polarised along the direction of magnetisation of the source ferromagnetic layer. Let us assume that the magnetisation in F1 is pinned to one specific direction and the spin polarised electrons have their magnetic moments, $\mu_1$, aligned parallel to $\vec{M}_1$. The electrons leaving F2 have magnetic moments, $\mu_2$, aligned parallel to $\vec{M}_2$. The change in magnetic moment from $\vec{M}_1$ to $\vec{M}_2$ causes the electron to lose some transverse angular momentum and which is transferred to $\vec{M}_2$ in the form a torque acting to align it in the direction of $\vec{M}_1$. This is called spin transfer torque.
Figure 3.13: (a) Schematic representation of Spin Transfer Torque in a magnetic multilayer film (b) Vector representation of the change in magnetic moment of an incident carrier and the resulting torque

The torque ($\vec{T}$) induced by the spins on the free layer is given by

$$\vec{T} = \gamma_o \frac{a_j}{M_s} \vec{M} \times (\vec{M} \times \vec{M}_1), \quad (3.17)$$

where $\gamma_o$ is the gyromagnetic ratio, $a_j$ is a prefactor characteristic of the magnetic multilayers, $M_s$ is the saturation magnetisation and $\vec{M}_1$ is the magnetisation of the fixed layer.

When the current first passes through the free layer, the spin polarised carriers are mostly reflected back into the free layer at the interface to the pinned layer [65]. This also results in a torque acting on the free layer, effectively switching the magnetisation away from that of the fixed layer.

3.5 Domain Wall Motion (DWM)

Various energies and magnetic anisotropies compete with one another to control the alignment of magnetic moments within a domain. The moments tend to be fixed in a particular direction because of the strong exchange interaction. When a weak field is applied in the direction of moments pointing upward (downward), it will not affect domains with moments pointing down (up) because it acts on a deep energy well. However, a weak field can tip the balance of moments in a DW in favour of the ‘up’ (down) direction. This can then result in the rotation of moments inside the wall in the direction of the applied field which leads to the DW moving laterally. This is called domain wall motion.
3.5.1 DWM induced by a magnetic field

On application of a field, the moments will rotate in the direction of the field and the final resting direction will be a balance of anisotropy and exchange energies. The energy change due to movement of a domain wall position to a new location is given by [20],

\[ E = -2\mu_0 M_s \cdot Hx, \]  

where \( A \) is the area of the wall, \( M_s \) is the spontaneous magnetization, \( H \) is the applied field and \( x \) is the distance moved by the domain wall. The force per unit area \( (F) \) exerted on the wall (the DW pressure) is given by,

\[ F = -\left( \frac{1}{A} \right) \left( \frac{dE}{dx} \right), \]

and hence \( F = 2\mu_0 M_s \cdot H. \)

The existence of defects plays a key role in domain wall motion. Defects can be of any form (e.g. sharp corners in certain shaped materials, inhomogeneous surfaces, second-phase particles, impurities, dislocations in the crystal structure and the associated inhomogeneous strain) and all impede DWM. The domain wall once pinned by these defects cannot move or propagate unless a field (or current) is applied in order to de-pin the domain wall.

As in the magnetisation rotation process, domain wall motion can be either reversible or irreversible and a transition from reversible to irreversible DWM is observed as a function of the magnitude of the applied field. Domain wall motion consists of two typical effects; domain wall bowing and the translation of domain walls. Upon application of a weak field, the DW expands like an elastic membrane and once the field is removed it contracts back to its original length/position. This is called DW bowing and can be irreversible when the DW is significantly deformed and continues to expand without any increase in the applied field. DW translation is normally irreversible due to pinning associated with dislocations, etc. in the material or due to its inhomogeneous microstructure [66].

On application of a strong magnetic field, the Zeeman energy overcomes the pinning potential energy barrier. When this happens, the moments within the domain rotate from their original crystallographic easy axis to an easy axis closest to the field direction, causing the domain boundaries to expand (or contract) or a domain wall to move. The pinning and depinning of domain walls is a key process in reversible and irreversible wall propagation. The ability to depin a domain wall from pinning centres depends on is the strength of the
pinning potential, the domain wall energy and the overall geometry of the structure (e.g., thickness, etc.) [67]. Defects pin the domain wall as the potential well represented by the pinning site is deeper than the domain wall energy. The pinning strength depends on geometrical parameters like the depth, width or area of the defect. Domain wall pinning leads to an overall reduction in wall energy (the wall area times the domain wall energy per unit area). Hence an applied field must be higher than the pinning field if the domain wall is to depin and move.

Consider the case of a Hall cross structure and a domain wall in one of the current arms as shown in figure 3.14. The four corners in the active region of the Hall cross have a high pinning potential. Depending on the applied field strength to move an $180^\circ$ Bloch wall (BW), wall bending can occur once it becomes pinned at two corners.

Figure 3.14: Schematic representation of DW bowing under the application of a field. The DW propagates on application of a field, gets pinned at the two corners and starts to expand under the continuous application of the field. Once the DW is depinned, DW bowing occurs towards the right hand lead of the Hall bar.

$180^\circ$ DWs in cobalt have the highest energy ($\sigma_w = 7.6 \times 10^{-3} \text{ J.m}^{-2}$) compared to Fe ($\sigma_w = 2.9 \times 10^{-3} \text{ J.m}^{-2}$) and Ni ($\sigma_w = 0.7 \times 10^{-3} \text{ J.m}^{-2}$) [20]. Therefore one would expect less bending to occur in Co. If the applied field strength is equal to or slightly greater than the depinning field, the DW propagates after first bending.

The energy difference ($\Delta E$) in the domain wall energy due to bending is given by [20],

$$\Delta E = \gamma_{BW} [A(H) - A(0)],$$  \hspace{1cm} (3.22)
where $\gamma_{BW}$ is the Bloch wall energy, $A(H)$ is the area of DW under an applied field and $A(0)$ is the area at zero applied field. For simplicity, let us assume that the wall deformation is cylindrical. Under this assumption the force per unit area ($F$) is

$$F = \frac{\gamma_{BW}}{r},$$  \hspace{1cm} (3.23)

where $r$ is the radius of curvature of the bowed wall.

This wall bending is reversible until a critical value of $r$ is reached, after which the domain along the field direction grows and the wall propagates, first depinning from one of the two corners and becoming irreversible. When the domain wall bends under the influence of the field ($H$), there is a pressure ($\gamma_{bw}/r$) on the wall which tends to balance the force due to the applied field. For the case of a $180^\circ$ domain wall [20],

$$\frac{\gamma}{r} = 2\mu_0 M_s H \cos \theta,$$  \hspace{1cm} (3.24)

where $\theta$ is the angle between applied field direction and the magnetisation within the two growing domains. The critical value of $r$ is reached when $r = l/2$ ($l$ is the distance between the two pinning sites).

Eq 3.24 can be used to determine the coercivity ($H_c$) of the material [20],

$$H_c = \frac{\gamma}{\mu_0 M_s l \cos \theta}.$$  \hspace{1cm} (3.25)

This allows us to understand that the critical depinning field, and hence the coercivity, is dependent on the number density of pinning centres which is inversely proportional to the pinning site spacing $l$.

**DW Creep**

At a finite temperature (e.g., $T \sim 290$ K), domain wall motion occurs at any drive field below the coercive field, $H_c$, when the pressure experienced by the DW is the critical force, $F_c$. The DW structure at $F_c$ and the velocity-force ($v$-$F$) characteristic are important to determine the DW velocity-field characteristic. At low temperatures and very small drive fields, the force on the DW is less than $F_c$ and one expects the DW motion to be slow. This was assumed to be taking place due to DW ‘hopping’ between pinning sites [68]. It was then realised that,
because of the glassy nature of the pinned DWs, the effective barriers present between pinned states diverge as the applied DW pressure reduces. As a result of this, a phenomenological creep theory was proposed [69, 70] having non-linear velocity-force characteristics of the form

\[ v \propto \exp(-\beta U_c (F_c/F)^{\mu}), \tag{3.26} \]

where \( v \) is the DW velocity, \( \beta = 1/kT \), \( U_c \) is the pinning energy and \( \mu \) is the dynamic constant.

To estimate the DW velocity as a function of field, consider a DW segment of length \( L \) and displacement \( u \). The total free energy can then be expressed as

\[ E(u, L) = \varepsilon_{el} \frac{u^2}{L} - (\Delta \xi^2 L)^{1/2} - M_s H t L u, \tag{3.27} \]

where \( t \) is the thickness of the magnetic layer, \( \varepsilon_{el} \) is the DW energy density per unit length. \( \Delta = F_{pin} n_i \xi \) is a measure of the pinning strength, where \( n_i \) is the surface density of pinning centres, \( \xi \) is their characteristic length and \( F_{pin} \) is the local pinning force. The second term in equation 3.27 represents the pinning energy and describes the contributions of the amplitude of the pinning force and/or the fluctuations of the pinning centre density to the pinning the DW [69, 71]. The third term in the equation is the Zeeman energy.

The characteristic pinning length \((L_c)\) of the DW is given by [72]

\[ L_c = \left( \frac{\varepsilon_{el} \xi^2}{\Delta} \right)^{1/3}. \tag{3.28} \]

Making a quasistatic approximation, Lemerle et al. rewrite the free energy of equation 3.27 as

\[ E(u, L) = U_c \left( \frac{L}{L_c} \right)^{2\xi - 1} - 2M_s H t L_c u_c \left( \frac{L}{L_c} \right)^{\xi + 1}, \tag{3.29} \]

where \( u_c \) is the transverse scaling parameter. The first term in this equation tends to lead to a decrease in the DW length while the second term tries to increase it.

Minimising equation 3.29 with respect to \( L \) and assuming an Arrhenius law as in reference [69], the DW velocity becomes
\[ v(H) = v_0 \exp\left[-\beta U_c \left(\frac{H}{H_c}\right)^\mu\right] \text{ for } H < H_c, \]  

where \( v_0 \) is a prefactor and \( \mu \) is the dynamic constant which is expected to be \( \frac{1}{4} \) for a 1D domain wall moving in a 2D disordered medium [72-74].

### 3.5.2 Current induced DWM

Domain walls present in a ferromagnetic wire will yield to the ‘pressure’ exerted by the spin current of flowing electrons thereby driving the domain wall in the direction of electron flow [75]. Current driven domain wall motion finds its main application in memory devices, example MRAMs, shift registers and racetrack memory [40, 76, 77], where the magnetic domains store information. On application of current the domain walls present between the domains will move past the output device to be ‘read’. This can in principle result in high storage density that is fast to read out and non-volatile.

![Diagram of current driven DW motion](image)

**Figure 3.15**: Schematic representation of current driven DW motion

The theory of current driven DW motion has been developed by a number of spintronics research groups. Works by Tatara *et al* [78] and Shibata *et al* [52] describe different models for current-induced DW motion (CIDWM). We restrict ourselves to those needed to describe and analyse experimental observations. Berger introduced the idea of transfer of spin angular momentum from the conduction electrons to the localised moments of a DW in the form of a torque which results in the displacement of the DW to a new position [75]. The electrons in the current are spin polarised after passing through a ferromagnetic metal, and as they traverse the DW the electron spins align parallel to the local magnetisation due to the exchange interaction. In order to conserve the total spin angular momentum a component is transferred
to the localised moments. This is like a torque acting on the local magnetisation of the DW resulting in DW displacement along the electron flow direction. This effect is independent of the film thickness and readily dominates the magnetic drag that arises due to the Lorentz force associated with the Hall Effect [79]. Interaction of the domain wall with the spin polarised current depends on the DW width and the lengthscales defining spin angular momentum transfer (i.e., the Larmor precession length or the Fermi wavelength depending on the model used), usually a few nanometres in 3d transition metals [80-84]. The domain wall width defines wide and narrow domain walls [85]. For wide DWs the conduction electron spins will adiabatically follow the local magnetisation (adiabatic STT) while for narrow DWs there is a non-adiabatic effect.

Considering a 1D system with a spin current flowing along the x-axis, the adiabatic spin torque $\vec{\tau}_{ST}$ is given by [81, 86],

$$\vec{\tau}_{ST} = -\frac{\partial J_s}{\partial x}, \quad (3.31)$$

where $J_s$ is the spin current density and can be written as a function of charge current density $J$ and current spin polarisation $P$, as $J_s = J\Phi/2e$.

$$\therefore \vec{\tau}_{ST} = \frac{J\Phi}{2e} \frac{\partial m}{\partial x}, \quad (3.32)$$

The adiabatic spin torque $\vec{\tau}_{ST}$ can be converted into a time derivative of the unit magnetisation by multiplying Eq. 3.32 by $-\gamma/M_s = -g\mu_b/\hbar M_s$

$$\frac{\partial m}{\partial \tau}_{ST} = -u \frac{\partial m}{\partial x}, \quad (3.33)$$

where the spin drift velocity ($u$) is given by,

$$u = \frac{Jg_\Phi \mu_b}{2eM_s}, \quad (3.34)$$

and where $J$ is the current density, $g$ is the Landé factor, $\mu_b$ is the Bohr magnetron, $P$ is the spin polarisation, $e$ is the electronic charge, $M_s$ is the saturation magnetisation and $u$ is the maximum DW velocity when all of the rotating spin moments are converted into DW displacement.

Every electron that crosses the DW will undergo a change in its angular momentum of $\hbar$ and this results in the addition of a magnetic moment of $2\mu_B$ to the domain wall [87].
Therefore, the change in magnetic moment in a domain due to an electric current flowing for a time \( \Delta t \) is,

\[
\delta m = \frac{2P\mu_B J A \Delta t}{e},
\]

where \( A \) is the cross-sectional area of the domain.

If this angular momentum is all used to create a DW displacement \( \Delta l \), this would also result in a change in magnetic moment,

\[
\delta m = 2M_s A \Delta l .
\] (3.36)

The velocity \((v)\) of the DW can be obtained by balancing Eq 3.35 and Eq 3.36 leading to,

\[
v = \frac{\Delta l}{\Delta t} = \frac{P\mu_B}{eM_s} J = u .
\] (3.37)

When deviations from adiabaticity cannot be neglected, we have a non-local torque, called the non-adiabatic torque that is perpendicular to the adiabatic torque. The non-adiabatic torque was introduced by Zhang and Li [88] and Thiaville et al [89] as the spin torque that originates from the mis-tracking of electron spins and the local moments. This happens when the conduction electron spin does not follow the orientation of the local magnetic moments and gets reflected back from them. The reflected spin changes its direction of motion and can transfer linear momentum from the conduction electron spins to the local magnetic moments which might also lead to DW motion. This is called non-adiabatic spin transfer torque \((T_\beta)\) and it is given [88] by,

\[
\frac{\partial \vec{m}}{\partial t} = \beta \vec{m} \times [(\vec{u} \cdot \nabla)\vec{m}] ,
\] (3.38)

where \( \beta \) is the parameter characterised by the relative contribution of the non-adiabatic spin transfer torque to the adiabatic spin transfer torque term. The non-adiabatic torque is physically distinct from the adiabatic torque and is something called the ‘beta’ term in the non-adiabatic STT equation.

The non-adiabatic STT alters the DW dynamics significantly and can determine the DW velocity and critical current density. The torque has two contributions; the first due to spin relaxation in DWs. Spin relaxation can occur by spin-flip scattering at impurities and due
to spin-orbit scattering events where the spin is not conserved [78, 80, 83, 88-92]. The second contribution is purely a non-adiabatic effect when there is a large spatial gradient of the magnetisation and it becomes difficult for the current spin polarisation to follow the direction of the local magnetisation [81, 83, 84, 93, 94].

The Landau, Lifshitz and Gilbert equation [95] is commonly used in micromagnetic description of DW dynamics. Spin transfer torque terms, both the adiabatic and non-adiabatic terms were added to the Landau-Lifschitz-Gilbert (LLG) equation iby Zhang et al. [87] and Thiaville et al. [89] to study the response of the magnetisation dynamics of domain walls to current excitation.

\[
\frac{\partial \vec{m}}{\partial t} = (-\gamma_0 \vec{m} \times \vec{H}_{\text{eff}}) + \left( \alpha \vec{m} \times \frac{\partial \vec{m}}{\partial t} \right) - \left[ \left( u(\vec{j}.\nabla) \vec{m} \right) + (\beta m \times (\vec{j} \cdot \nabla) \vec{m} \right],
\]

(3.39)

where \(\alpha\) is the damping constant and \(\vec{H}_{\text{eff}}\) is the effective field given by,

\[
\vec{H}_{\text{eff}} = \frac{1}{M_s} \frac{\partial E}{\partial \vec{m}},
\]

(3.40)

where \(\vec{m}\) is the unit vector representing direction of local magnetic moment and \(E\) is the total energy density of the system.

![Diagram](image)

**Figure 3.16:** Various terms present in the LLG equation describing current-induced DW dynamics

The first two terms in the LLG equation 3.39 are the precession term and the damping torque, graphically represented in figure 3.16. The precession term describes the rotation of moments around an applied field while the damping torque will act on the magnetic moment to align it along the field direction. The third term is the adiabatic and non-adiabatic STT which acts parallel to the damping torque or against it depending on the current direction. Depending on the magnitude and direction of current, three situations can be achieved. When the damping
torque equals the STT they balance each other and a stationary precession of moments around the applied field takes place [53]. When the STT is greater than the damping torque, it will push the magnetic moments away from the direction of the applied field until a critical angle is reached. At this point the moments will switch and reorient itself into another direction leading to complete switching of moments. If the STT is smaller than the damping torque, then the magnetic moment will experience the usual damping.

Figure 3.17: Sketch of a magnetic material or a wire with out-of-plane easy axis having a Bloch wall, $\psi$ is the tilt angle of the DW’s and $q$ is the position of the centre of mass of the DW. Reproduced from [51]

Eq 3.39 can be simplified into two sets of coupled differential equations within a 1D model under the assumption that the DW has a constant profile and defining the magnetisation angle $\phi$ and DW position $q$ as shown in figure 3.17 [11, 80, 89, 96, 97]

$$\dot{\psi} + \frac{\alpha \dot{q}}{\Delta} = \gamma \mu_0 H + \frac{\beta u}{\Delta} - \frac{\gamma}{2M_s} \frac{\partial V_{pin}}{\partial q} .$$  \hspace{1cm} (3.41)

$$\frac{\dot{q}}{\Delta} - \alpha \dot{\psi} = \frac{\gamma \mu_0 H_k}{2} \sin 2\psi + \frac{u}{\Delta} ,$$  \hspace{1cm} (3.42)

where $H$ is the applied magnetic field along the easy axis, $H_k$ is the restoring field for transverse orientation of the DW, $V_{pin}$ is the pinning potential which may depend on $q$. The domain wall width $\Delta(\psi) = (A/(K_0 + K \sin^2 \psi))^{1/2}$ for uniaxial anisotropy $K_0$ and transverse anisotropy due to magnetostatic effects $K = \mu_0 M_s H_k / 2$ [98]. The non-adiabatic torque is seen in Eq 3.41 in an analytical form that is analogue to an externally applied field $H_I; \mu_0 H_I = \beta u / \Delta \gamma$. The 1D model is used to estimate the DW velocity and the critical current density for current driven DW motion without pinning [52].
For $\beta = 0$, there is a critical current density above which the DW starts to propagate while there is no steady DW motion below the threshold current value [88]. The onset of periodic DW transformations corresponds to this threshold value [99-101]. The non-adiabatic torque acts as a magnetic field to sustain a steady DW propagation which results in the disappearance of the intrinsic threshold current even for $\beta < 1$. The DW velocity increases linearly up to a critical velocity [88, 90, 101].

$$v_{\text{final}} = \frac{\beta u}{\alpha}. \quad (3.43)$$

Another important factor to consider in current driven domain wall motion is the Joule heating effect in the measured samples due to the rise in temperature caused by the applied current. This effect has been observed in many investigations where a 5K temperature rise was measured for a current density less than $10^{11}$ A/m$^2$ [102-104]. A.Yamaguchi et al. were able to measure this effect by correlating the temperature with the sample resistance while observing domain wall motion after application of an electric current [105]. Their results suggest an increase in temperature close to 830K for current densities of 7.5x$10^{11}$A/m$^2$, which is close to the critical current density for permalloy (Ni$_{80}$Fe$_{20}$). It also suggests the possible formation of multidomain structures due to demagnetisation when the temperature rises above the Curie temperature.

**Summary**

The basic principles involved in spintronics along with giant magnetoresistance and extraordinary Hall effect have been explained in detail. Most of the experimental techniques for magneto-transport studies and time-resolved studies are based on the Extraordinary Hall effect. The background theory for domain wall motion by applied fields was described. The concept of spin transfer torque was introduced to explain the underlying physics of current-induced domain wall motion, which forms the basis for experiments on domain wall dynamics.
Chapter 4 Device Fabrication

4.1 Magnetic materials under study

We chose a Ta/Pt/Co/Pt material system for our experiments with an ultrathin ferromagnetic Co layer, non-magnetic Pt and Ta buffer layers and a thin protective Pt capping layer to prevent the oxidation of cobalt. The Co film thickness was optimised to exhibit perpendicular magnetic anisotropy. Si/SiO$_2$ was chosen as a substrate material with a surface roughness of less than 0.3 nm. A schematic diagram of the layer structure is shown in figure 4.1.

![Figure 4.1: Sketch of Ta/Pt/Co/Pt layer structure](image)

4.2 Sample growth by sputtering

Deposition of thin metal films by sputtering is one of the most widely used deposition methods for thin film device applications. Over the years it has been demonstrated to be a powerful tool for growing high quality ultra-thin metallic films [106] and a variety of material properties can be achieved by varying the deposition parameters [107]. To grow our Co/Pt films on the Si/SiO$_2$ substrate, we used a magnetron sputtering system in Leeds collaborating with Dr. Thomas Moore in the School of Astronomy and Physics.
A schematic diagram of the sputter system is shown in figure 4.2. The sputtering process [108] takes place in a vacuum chamber when an ionised gas molecule makes an impact on a target material; this is likely to create a series of atomic collisions resulting in the ejection of some atoms from the surface. The ejected atoms hit a substrate material onto which they get deposited at an atomic scale, bonding strongly to the substrate. In a sputter chamber, an atom of a material from the ‘target’ is hence ejected onto a substrate on which the thin films are grown. In magnetron sputtering a permanent magnet is placed behind the target helping to achieve plasma confinement near the target surface. For efficient momentum transfer, argon is employed as sputtering gas as it can closely match the atomic weight of cobalt and platinum. Since argon is an inert gas, it also does not react with the target material. A high negative voltage applied to the target attracts the positive argon ions thereby causing a high energy impact for sputter atoms. The direction of the sputtered atoms is controlled by the magnetron (magnetic fields which influence the deposition) onto the substrate sample. By controlling the path of the sputtered atoms we can control the rate and thickness of the material to be deposited onto the substrate.

Ta/Pt/Co/Pt multilayers were grown on Si/SiO₂ (90 nm) substrates using DC magnetron sputtering with a base pressure of 1.4±0.2 x 10⁻⁷ Torr and Ar pressure of 2.5±0.3 mTorr. A schematic diagram of the layer structure is shown in figure 4.1. The cobalt thickness range for good PMA was investigated around 0.3-0.85 nm Cobalt thickness when sandwiched
between Pt layers (1.8 nm and 3 nm). The thick Pt layer acts as a buffer layer to improve Co growth morphology and the thin Pt layer is a capping layer to avoid oxidation.

### 4.3 Polar MOKE characterisation of multilayer films

Characterization of ferromagnetic thin films can be effectively achieved using the magneto-optical Kerr effect (MOKE) [20, 23]. The Kerr effect describes the change in polarization and reflectivity of light (usually a laser beam) reflected from a magnetic surface. There are three types of Kerr effect as shown in figure 4.3, depending on the direction of the magnetic field and plane of incidence of light on the sample surface [109]. These are polar, longitudinal and transverse. In the MOKE setup for the transverse Kerr mode, the external magnetic field and the magnetization of the sample are perpendicular to the plane of the incident laser beam. In the longitudinal MOKE setup, the magnetization of the sample is in-plane and parallel to the plane of the incident laser beam. A polar MOKE mode setup has the sample magnetization parallel to the plane of incident laser beam and normal to the material surface. The polar MOKE mode setup shown in fig. 4.4 is suitable for characterizing the magnetic anisotropy and magnetization switching of out-of plane anisotropy thin film materials [17].

![Figure 4.3: Three possible modes of magneto-optical Kerr effect. Reproduced from: http://www.study-on-line.co.uk/whoami/thesis/images/img2_4.gif](http://www.study-on-line.co.uk/whoami/thesis/images/img2_4.gif)
Characterization of the thin films grown was performed in a polar geometry using a 633 nm He-Ne laser. The reflection of light takes place at near normal incidence as both incident and reflected beams have to pass through a hole in one of the poles of the electromagnet. The field range was set from -400 Oe to +400 Oe and we obtained a hysteresis loop of the magnetization of the sample with field $H_z$ in Oe versus photodiode voltage (MOKE signal). Fig 4.5 shows a polar MOKE plot obtained for one of the Co/Pt samples grown using DC magnetron sputtering that exhibits good PMA.
4.4 Design of device patterns

After MOKE characterization of the thin films grown we proceeded to create device designs for the Hall probe structure and contact lead patterns.

Figure 4.6: (a) and (b) Design patterns used for fabrication of a single Hall cross device for time-resolved domain wall motion studies. The red colour represents the coplanar wave guide structure with connecting leads and the surrounding ground plane. The blue region in (b) represents the single Hall cross.

Using the commercially available nanolithography software tool, “Elphy plus”, a single Hall cross and 5x5 mm² 50 Ω coplanar waveguide (CPW) structures, impedance-matched to the voltage pulse generator, were designed for a single Hall probe device (shown in Fig 4.6: (a) & (b)). This was based on an established model for a coplanar waveguide structure with a finite thickness of dielectric material [Appendix A]. The presence of the tantalum buffer layer lowers the device resistances and improves impedance matching in our 50 Ω coplanar waveguide structures.

The CPW impedance value of $Z_0$ is calculated as

$$Z_0 = \frac{1}{c C_{air} \sqrt{\varepsilon_{eff}}} \quad \varepsilon_{eff} = \frac{C_{CPW}}{C_{air}},$$  \hspace{1cm} (4.1)

where $C_{CPW}$ is the total capacitance of the coplanar waveguide, $c$ is the velocity of light in free space, $\varepsilon_{eff}$ is the effective dielectric constant and $C_{air}$ is the partial capacitance of the CPW in the absence of all the dielectric layers.
To calibrate the optimum Ga$^+$ dose level for irradiation of a single Hall probe sample, a 3.75 mm$^2$ device was also designed having an array of 5 Hall probes connected in series separated by 150 µm from each other (c.f., Figs 4.7: (a) & (b)).

Figure 4.7 (a) & (b): Design patterns employed for fabrication of Hall array devices. The gold colour in (a) represents the contact pads and the grey region is the Co/Pt leads connecting the Hall probes

4.5 Electron beam lithography and photolithography

Once the Hall device structures were defined, we proceeded to the, patterning of these designs in Co/Pt samples. We employed photolithography for fabrication of Hall array devices. Photolithography is one of the most common and oldest processes for pattern definition, first used in the 19th century. The process involves a pattern in a photo mask being transferred into a resist coated surface by exposing it to UV light. The photoresist is used to make patterns on thin films for device fabrication. The resist used here was Shipley S1813, a light sensitive polymer whose molecular chains break up upon exposure to UV light. The resist is spun onto the sample at 3000 rpm using a spin coater and subsequently baked at 90° for 30 minutes. The thickness of the resist depends on the spin speed and it was optimized for this process. Upon illumination with UV light for 8-10 seconds in contact with the mask, the required pattern was exposed. The sample pattern was then developed in Microposit 351 developer (mixture of water & 351 in the ratio of 3.5:1) for about 30-45 seconds and the sample then washed with deionized water. The exposed resist dissolved in the developer transferring the design pattern onto the sample. The sample with the pattern was baked for nearly 2 hours before dry etching using an argon ion miller. A pictorial representation of the entire procedure for fabricating samples by photo-lithography is shown in figure 4.10 ahead of fig. 4.8 and fig. 4.9.
Electron beam lithography (EBL) is a modern technique for writing patterns on resist-coated thin films. We used EBL for fabrication of single Hall cross devices. The resist used here was an electron sensitive bi-layer resist containing polymethyl methacrylate (PMMA) and copolymer (MMA 8.5). Each resist layer was spin coated on the Ta/Pt/Co/Pt samples and baked separately at 125 °C (for MMA8.5) and 150 °C (for PMMA) in an oven prior electron beam exposure. A beam of 25 keV electrons with a dose range of 300-500 μC/cm², was used to expose the resist on the coated surface pixel by pixel, allowing for patterning of complex nanoscale-sized structures with great accuracy. The nanoscale diameter (~1.5 nm) focused electron beam exposed the sample using the ‘Raith Elphy Plus’ pattern generator which controls the electron beam write trajectory and dose. The exposed sample is placed in a developer containing a mixture of methyl isobutyl ketone (MIBK) and isopropanol (IPA) in the ratio of 1:3 for 10 seconds. The weakened polymer bonds of the exposed resist dissolved in the developer mixture. EBL was used to define the single Hall cross pattern for deposition and lift-off of Al (10 nm)/Ti (10 nm) films onto the Ta/Pt/Co/Pt sample wafers. The Al/Ti Hall cross film structure provides a well-defined layer pattern with sharp edges and the precise design dimensions and acts as a mask for subsequent Ar-ion milling. Post Ar-milling, Al/Ti layers are chemically etched away using MF319 developer leaving the defined Co/Pt Hall cross structures on the Si/SiO₂ substrate.

4.6 Argon ion milling and thermal evaporation

Argon ion milling is used to transfer nanoscale patterns in conjunction with lithographic techniques [110-112]. This technique is preferred over wet chemical etching techniques when it is important to obtain a high quality edge definition and avoid undesirable side wall degradation. In the ion miller similar to the one in figure 4.8, a broad beam DC ion source is used to generate the argon ion beam, with the beam energy in the range of 500 eV. The plasma formed by electrical discharge contains argon ions which are accelerated towards the target by a pair of optically aligned grids. The target is mounted on a rotating sample holder, bombarding the target surface. A neutralizer filament grid present inside the chamber prevents the building up of positive charges on the target surface. The duration of this process was about 20 seconds and was based on the milling rate for Co/Pt by Ar-ions. The ion beam etches away the entire film surface which has not been protected by a mask. In the case of the Al(10 nm)/Ti(10 nm) mask the ion beam only etched away ~1 nm of Ti layer in the same time period. The entire process takes place in a vacuum chamber. Once the milling is finished, the
sample is retrieved and soaked in MF319 in an ultrasonic bath to remove the Al/Ti mask (or acetone to remove an S1813 mask) and reveal the milled sample pattern.

![Schematic diagram of an Ar-ion Miller](http://www.ionbeammilling.com/about_the_ion_milling_process)

Figure 4.8: Schematic diagram of an Ar-ion Miller.
Reproduced from: http://www.ionbeammilling.com/about_the_ion_milling_process

In the next step, Cr/Au contacts were deposited, using thermal evaporation through an optical lithography pattern. Thermal vapour deposition is another conventional technique used for thin film growth. It involves melting of a material until it is in vapour form and depositing it as a thin film on a sample surface. The entire process takes place inside a vacuum chamber at very low pressures of the order of $2.5\pm0.2 \times 10^{-6}$ Torr at the deposition rate of 0.5-0.7 nm/sec. This avoids reactions between residual gases present inside the chamber (c.f. fig. 4.9). The coating material is placed in a tungsten boat connected to electrical terminals. The amount of current necessary to melt the metals and the rate of deposition will vary for different materials. Under low pressures, the atoms of the material in the vapour state have a mean free path that is comparable to the vacuum chamber dimensions. This results in atoms travelling in straight lines towards the target. An oscillating quartz crystal monitor is placed inside the chamber near the target to monitor the rate and thickness of the material being deposited on the target.
Figure 4.9: Schematic diagram of a thermal evaporator.
Reproduced from: http://www.old.ece.utep.edu/research/webedl/cdte/Fabrication/index.htm

Figure 4.10: Sketch of the ion milling and contact deposition process

Figure 4.11: (a): Hall array device fabricated by photo lithography; (b): SEM image of one of the Ga⁺ FIB irradiated Hall cross structures
The completed Hall array consisting of five $2 \, \mu m \times 2 \, \mu m$ Hall crosses on a $3.75 \times 3.75 \, mm^2$ chip is shown in figure 4.11. The chip was glued in a leadless chip carrier (see figure 4.12) for wire bonding and electrical measurements.

Figure 4.12: The Hall array wire bonded in a ceramic leadless chip carrier

Figure 4.13: Sketch of the fabrication process for a single Hall cross device for time-resolved domain wall motion investigations.
The fabrication procedure for single Hall probes by e-beam lithography was designed to make a robust Hall probe device free of any electrical leakage. \(5 \times 5 \text{ mm}^2\) Co/Pt wafers were diced from larger pieces of wafer and patterned into 2 \(\mu\text{m}\) wide single Hall cross structures using e-beam lithography as shown in figure 4.14 (b). Cr/Au contacts, designed to form a 50 \(\Omega\) coplanar waveguide structure, were defined by evaporation and lift-off (c.f. fig. 4.14 (a)). After FIB irradiation the Hall probe was then fixed to a piece of PCB board as shown in figure 4.15.

![Completed single Hall cross structure bonded to a 50 \(\Omega\) coplanar waveguide structure on a PCB.](image)

Figure 4.14: (a) Optical image of a single Hall probe device fabricated by e-beam lithography; (b) SEM image of a single 2 \(\mu\text{m}\) Hall cross structure

Figure 4.15: Completed single Hall cross structure bonded to a 50 \(\Omega\) coplanar waveguide structure on a PCB.
4.7 Focused ion beam irradiation

Focused Ion Beam (FIB) is commonly used in materials and biological science and finds many applications in the semiconductor industry [113-115]. Using a FIB is similar to the operation of a Scanning Electron Microscope (SEM). SEM uses a beam of electrons to image the sample surface, whereas FIB uses a beam of ions to image while using low beam currents and mill the surface/samples using high beam currents. In our experiments we have used a 30 keV Ga\(^{+}\) ion source. When Ga\(^{+}\) ions hit the sample surface, secondary ions and secondary electrons are produced. At very low doses the Ga\(^{+}\) ions hardly damage the surface layer, which was estimated to be less than 0.013 nm thick [116], however there is a possibility that Ga\(^{+}\) implantation could occur before sputtering takes place at a higher dose [117, 118]. At high doses the surface gets bombarded heavily and can be milled with precision to pattern films [119]. Surface roughening, ion implantation and residual deposition of the sputtered material are drawbacks of FIB [120, 121]. Irradiation with Ga\(^{+}\) ions at lower doses has been shown to be an effective technique for reducing the perpendicular anisotropy of Co/Pt thin films while retaining overall out-of plane anisotropy [19]. An ‘FEI Helios 600’ combined SEM-FIB was used to irradiate the active region of individual Hall crosses with a range of Ga\(^{+}\) dose levels spanning 0 - 0.144 pC/\(\mu \text{m}^2\) with a 30 keV ion energy at 1 pA beam current. The beam diameter was set to 10 nm which was much larger than the pixel spacing in order to achieve a spatially homogeneous dose.
4.8 Atomic force microscopy

The atomic force microscope (AFM) belongs to the family of scanning probe microscopes. It is used as a characterization tool for measuring structural properties such as the surface roughness and height of the Co/Pt thin film microstructures. Unlike electron microscopes AFM uses a probe made of a thin cantilever with a sharp conical tip. The probe tip is usually a few micrometres in height and the tip radius varies from 1-15 nm. The AFM tip scans over an area of the sample while a laser beam is reflected off the back of the cantilever and falls on a photodetector, registering deflections. In this way the force between probe and sample surface is measured and an image of the sample surface is created. A sketch of an AFM is shown in figure 4.17(a). There are two basic types of scan mode; contact mode and tapping mode. In contact mode, the tip is continuously dragged in contact with the surface and an image is created based on the deflections of the cantilever. In tapping mode, the tip is brought near to the surface and a piezoelectric element is used to oscillate the tip up and down gently tapping the surface at each single point. This mode avoids any damage to the surface. We have used an ‘Asylum Research Instruments’ AFM and tip. The tip radius was 9±2 nm. Careful AFM
imaging of the active regions of Hall crosses (2 μm² area active regions as shown in figure 4.17(b)) was performed in tapping mode before and after irradiation in order to study the relationship between the magnetic anisotropy and the film microstructure and surface roughness.

Figure 4.17: (a) : Sketch of an AFM during operation. (b) One element of a Hall array device showing the AFM scan area, i.e. the active region of the Hall bar.

(a) is reproduced from: http://www.nanoscience.gatech.edu/zlwang/image/research/afm2.jpg

4.9 X-ray powder diffractometer

X-rays have wavelengths that approximately correspond to the interatomic spacing of ~1-3 Å in typical crystal structures. When fast moving electrons strike a copper target, a small amount of their kinetic energy is converted into radiation producing characteristic x-rays which are highly intense and have a particular wavelength. The x-rays are named after their emission lines, namely Kα, Kβ, etc. Kα has high intensity and is predominantly used in x-ray diffraction studies. X-ray diffraction works on the principle of Bragg diffraction which is described by Bragg’s law

\[ 2d \sin \theta = n\lambda \; ; \text{for } n= 1, 2 ,..., \]  

where d is the crystal plane spacing, \( \lambda \) is the wavelength of the x-ray beam, \( \theta \) is the angle of incidence of the x-ray beam on the sample surface and n is the diffraction order.

X-ray powder diffractometry (c.f. figure 4.18) is widely used to determine crystal structures and can also be used for polycrystalline thin films. A beam of x-rays passes through a slit which collimates the diverging beam, and then hits the thin film sample placed midway between the emitter and detector at an angle \( \theta \) (between x-ray tube and counter in fig. 4.18). The reflected beam is collected at the detector which is at an angle of 2θ from the transmitted
x-ray beam path. It lies on the surface of a cone (radiation scatters in a conical pattern) whose apex is at the incident point of the x-ray beam on the sample. Crystal planes that make an angle $\theta$ to the incident beam have a number of different orientations. All the crystal planes of spacing, $d$, reflect at the same Bragg angle, if they have the right orientation. A ‘Bruker D8 advanced powder diffractometer’ was used to characterise the crystallinity of the Co/Pt multilayer samples. The diffractometer source is monochromatic copper K$_\alpha$ radiation emitted from a 0.6 mm wide slit at a wavelength of 1.54 Å. All measurements were taken with a step time of 1.5 seconds per degree of angle.

Figure 4.18: Schematic representation of an X-ray diffractometer.
Summary

The methodology of sample fabrication is explained and the working principles of all the equipment and techniques used in the fabrication process are described in detail. Characterisation techniques such as MOKE were used to characterise the Ta/Pt/Co/Pt films after growth to check for out-of-plane anisotropy, and x-ray diffractometry was used to analyse the crystal structure and crystallinity of the grown films. Using optical lithography Hall array devices were patterned and single Hall cross devices fabricated by EBL. Focussed ion beam irradiation was used for very low dose irradiation of Hall cross structures after fabrication. Surface roughness analysis of the irradiated Hall cross structures was carried out using atomic force microscopy to identify changes in roughness and thickness after FIB irradiation.
Chapter 5 Experimental methods

5.1 Low frequency extraordinary Hall effect (EHE) measurements

The Extraordinary Hall Effect (EHE) is sensitive to the magnetisation in a ferromagnetic material [24, 122]. Changes in the magnetization are reflected in the Hall voltage generated at the voltage leads. The EHE signal can be large in ultrathin films and hence it is often used as an effective tool for measuring the magnetisation state and magnetic anisotropy of ferromagnetic thin films [123, 124].

A low frequency EHE setup was constructed, as shown in Figure 5.1, with current and voltage leads connected to a single Hall probe. The setup was designed to characterise the magnetotransport properties of the microfabricated multilayer structures. The Hall probe device was placed inside a hollow cylindrical coil connected to a regulated bipolar power supply to generate controlled magnetic field cycles with the field normal to the plane of the sample. The Hall probe was driven by a constant (32 Hz) 4 μA AC current from a function generator and the detected Hall voltage signal was detected using a ‘Stanford Research 830’ digital lock-in amplifier. The entire experiment was controlled with LabView programme.

The Hall voltage obtained was a tiny signal on the order of μV. When the magnetic state in the ferromagnetic material switched from one (e.g. up) direction to the other (e.g. down direction), the Hall voltage changed its sign signalling magnetisation switching. Sweeping the external magnetic field between two extreme field values of opposite sign, the Hall voltage for each field step was recorded and the output saved. A plot of the data for applied field versus Hall voltage produces a hysteresis loop. For a perpendicularly magnetised material placed in an out-of-plane field, the hysteresis loop generated is a square shaped loop, with sharp switching at well-defined values of coercive field.

The magnetization direction and the coercivity of the material are determined from the hysteresis loops. The squareness of the loops indicates the presence of perpendicular magnetic anisotropy (PMA) in the material and the coercivity, which is controlled by the anisotropy, indicates the strength of the PMA. We also measured arrays of several Hall probe structures on a single chip. This was used as the first characterization tool after fabrication of Hall probe structures. After Ga⁺ irradiation of the active crosses of the Hall devices, the anisotropy was reduced in this region as reflected in a narrowing of the hysteresis loop as shown in figure 5.2(a). Since the central cross-section of the Hall bar is the active area (c.f fig. 5.2(b)), Hall voltage switching upon magnetisation reversal at lower external applied fields confirms the
changes in anisotropy of the irradiated region. Using this setup we studied the effect of Ga\(^+\) irradiation on the magnetic properties of our magnetic multilayers.
Figure 5.1: Circuit diagram of the Low frequency Extraordinary Hall Effect setup.

Note: Analogue Lock-in-An amplifier is depicted in the circuit diagram to simplify the explanation of signal processing. In our measurements we use a digital Lock-in-amplifier.
Figure 5.2: (a) Low frequency EHE measurement of a Ta(4 nm)/Pt(3 nm)/Co(0.35 nm)/Pt(1.8 nm) 2 μm Hall cross before and after FIB irradiation at a dose of 0.143 pC/μm². The magnetisation axis has been normalized by the saturation magnetisation before irradiation. (b) The active region whose magnetization state is detected in EHE measurements.

5.2 Scanning MOKE domain imaging

Kerr microscopy is arguably the most versatile and flexible magnetic imaging technique. This works on the principle of the magneto-optical Kerr effect (MOKE) where a plane-polarised light beam undergoes a polarisation rotation upon reflection (refer to Chapter 4, section 4.3). With the use of a polarisation analyser in a microscope, the rotation of the reflected light beam can be converted into a map of magnetic domain contrast. Using image processing this domain contrast from the surface of the ferromagnetic layers can be enhanced and extracted. The basic components of Kerr Microscopy are shown in figure 5.3. Generally there is no need for a specific surface treatment for magnetic domain imaging and one can even have thin nonmagnetic multilayers above the ferromagnetic thin film. External magnetic fields, both in-plane and out-of-plane, can be applied to image the magnetization process of the domains and simultaneously measure magnetisation loops.
Figure 5.3: Magnetic imaging setup of a Kerr Microscope.

‘Neoark BH-786IP-SP2’ Kerr microscopy was used to image the magnetization switching of low and high anisotropy regions in the Co/Pt thin film samples, i.e., magnetization switching of the Ga⁺ irradiated region and the non-irradiated region upon sweeping the applied out-of-plane field. Kerr imaging was carried out at the National Institute of Material Science (NIMS), Japan under the guidance of Dr. Masamitsu Hayashi. Kerr imaging shows distinct contrast between the two switching magnetic states, shown in figure 5.4 when the low anisotropy region switches first before the high anisotropy region. It also directly shows how domain walls propagate along the Hall bar sections under the influence of fields and currents (shown in chapter 8), as well as domain wall pinning/de-pinning and the annihilation of domain walls. This technique is very useful in understanding the influence of spin-transfer torque when multiple domain walls are present under the influence of a drive current.
5.3 EHE measurements with pulsed currents

A pulsed voltage (current) was added to the existing EHE measurement setup as a preliminary experiment to determine the minimum/threshold pulse amplitude necessary to move the domain wall in the Ga\(^+\) irradiated region of the Hall cross structure. The pulsed circuit part consisted of a National Instruments Digital to Analogue Controller (DAC) ‘NI USB-6210’ operated from a computer and was connected to an ‘Avtech AVMP-2-C-P’ pulse generator. Upon receiving the command from the PC, the DAC triggered the pulse generator to send out a voltage (current) pulse. Sweeping the magnetic field from positive maximum towards the reversal field, the magnetization in the low anisotropy irradiated region was switched with respect to the unirradiated parts; domain walls were hence created at the boundaries of the irradiated region in the Hall probe. Without completely reversing the rest of the sample into saturation (i.e., not switching the unirradiated regions), the field is swept back towards a positive value where it is held below where the magnetization in the irradiated region switches back. In this way we are able to capture a minor hysteresis loop (c.f. fig 5.5). The fixed external field at the end of the cycle is called the ‘bias field’.

By holding the bias field fixed and switching to a pulsed setup, a single voltage pulse with a fixed amplitude (~1-5 V) and pulse width (~50 ns) was fired into the current line of the Hall probe. The setup was connected back in AC current mode and the Hall voltage at the lock-in amplifier recorded. Any increase (decrease) in the Hall voltage signal indicates the displacement of the domain wall from its initial position in the irradiated region. The experiment was repeated for increasing pulse amplitudes and pulse durations. A data set as a function of pulse amplitude with a bias field increasing from zero was generated. A threshold value for the pulse amplitude required to move the domain wall from its original position was...
thus determined. Pulse amplitudes above the threshold value were then used for the fast time resolved EHE measurements.

Figure 5.5: Minor Loop of a Co 0.35 nm thin film sample. The magnetic switching takes place in the irradiated region at -86 Oe and the sweep direction reversed before the unirradiated region switches. This can be seen in the bottom Kerr image where the Co/Pt leads have not switched but the magnetization of the irradiated region has switched. The bias field at the end is held at 60 Oe.
Figure 5.6: Circuit diagram of lo frequency EHE setup with pulsed dc voltages (currents)
Note: An analogue Lock-in-Amplifier is depicted in the circuit diagram to simplify the explanation of signal processing. In our measurements we use a digital Lock-in-amplifier.
5.4 T-resolved EHE measurements

A time-resolved magneto-transport measurement circuit was constructed based on impedance-matched transmission lines, an ‘Avtech AVMP-2-C-P’ voltage pulse generator and an ‘Agilent DSA90604A’ high frequency fast digital oscilloscope with a differential probe, allowing Hall voltages to be sampled (c.f. figure 5.7). This setup was primarily used for measuring the critical current density ($J_c$) for domain wall motion and the velocity of driven domain walls in Hall probe devices. A synchronised pulse generator sends out a single square voltage pulse once the artificial domain state has been created after a minor hysteresis loop. The pulsed current displaces the domain wall and the position of the moving wall was recorded by monitoring the EHE at a single pair of Hall voltage contacts as it crossed them [10, 125]. There was no need to simultaneously nucleate domain walls with current pulses as these were prepared in the minor loop generation step using a quasi-static field cycle. This process was repeated 2048 times for varying bias fields for averaging purpose to reduce the noise in the signal. The averaged signal subtracted from a reference signal where it was known the no DW motion took place. The reference signal was an average of 8000 iterations of the above procedure with a negative bias field region where there was no domain wall motion irrespective of the pulse amplitude. Signal subtraction yielded a time-resolved pulse waveform having a peak at the start of the pulse and a gradual shift towards negative voltage amplitudes over the pulse duration. Figure 5.8 shows an idealised sketch of how a subtracted pulse signal might appear. The critical current density for DW motion was measured as a function of applied perpendicular bias field.
Figure 5.7: Circuit diagram of the time-resolved EHE measurement setup.
Figure 5.8: Idealised sketch of a time resolved pulse waveform. The inset is the input square voltage
(current) pulse applied to the current line with a pulse rise time $\leq 100$ ps and fall time $\leq 135$ ps.

5.5 Anisotropy field ($H_k$) measurement by EHE

A magnetometric technique based on the extraordinary Hall Effect was used to measure the
perpendicular magnetic anisotropy field ($H_k$) in thin ferromagnetic multilayer films [124]. The
EHE voltage was measured while rotating an external applied field in small angle steps around
the easy axis. The measured data were analysed using a Stoner-Wohlfarth based model in the
limit of small magnetisation deviation angle, in order to extract the anisotropy field. The
experimental setup showing the connections to a Hall bar is shown in figure 5.9. This
experiment was carried out in Leeds in collaboration with Dr. Thomas Moors in the School of
Astronomy and Physics. A constant current of 25 $\mu$A DC current was fed into the current arm
and a transverse Hall voltage detected in the voltage arm of the Hall bar. The Hall voltage
carries the extraordinary Hall effect signal which is directly proportional to the magnetisation
of the sample [122].
The magnetic energy of a single domain ferromagnetic particle is given by

\[ E = -K_u \cos^2 \theta - M_s H \cos(\theta - \phi), \]  

(5.1)

where \( K_u \) is the uniaxial anisotropy, \( H \) is the externally applied field, \( M_s \) is the saturation magnetisation and \( \theta \) and \( \phi \) are defined as the magnetisations angle and external field angle with respect to the easy axis.

Normalising Eq.5.1 by \( K_u \) gives

\[ \varepsilon = \frac{E}{K_u} = -\cos^2 \theta - \alpha \cos(\theta - \phi), \]  

(5.2)

where \( \alpha = M_s H / 2K_u \).

For small values of \( \theta \) and \( \phi \), the solution of this equation can be expressed in a Fourier series,

\[ \cos \theta = 1 + A_2 \phi^2 + A_4 \phi^4 + A_6 \phi^6 \ldots \]  

(5.3)

In [124], the Fourier coefficients \( A_4 \) and \( A_6 \) were found to be very small. \( A_2 \) can be expressed as

\[ A_2 = \frac{\alpha^2}{2!(1 + \alpha^2)}. \]  

(5.4)

We can hence write the Fourier series solution in the approximate form,
\[
\cos \theta \approx 1 + \frac{(M_s H)^2}{2(2K_u + M_s H)^2} \phi^2
\]  
(5.5)

In the anisotropy field measurements we restricted the external field angle, \( \phi \leq \pi/6 \) and the magnetisation angle, \( \theta \) was measured experimentally by means of the Extraordinary Hall Effect. Hence, we could deduce the out-of-plane components of the external field and magnetisation which are proportional to \( \cos \phi \) and \( \cos \theta \).

To measure the dependence of \( \phi \) on the magnetisation angle, \( \theta \), a constant magnetic field is first applied normal to the sample plane and then the sample is rotated in steps of 10° from \(-180^\circ\) to \(180^\circ\) about a rotation axis parallel to the current direction. The Hall voltage that is generated at a rotation angle of \( \phi \) is given by

\[
V_H = R_{OHE} H_x + R_{EHE} M_z,
\]
(5.6)

where \( H_x = H \cos \phi \) and \( M_z = M_s \cos \theta \).

Since the ordinary Hall effect is very small compared to the EHE, \( H < M_s \) and \( \cos \theta \) can be obtained from the relation

\[
\cos \theta = \left( V_H - \frac{V_{H_{\text{max}}} + V_{H_{\text{min}}}}{2} \right) \left( \frac{V_{H_{\text{max}}} - V_{H_{\text{min}}}}{2} \right),
\]
(5.7)

where \( V_{H_{\text{max}}} \) and \( V_{H_{\text{min}}} \) are the maximum and minimum Hall voltage values in one complete rotation measurement from \(-180^\circ\) to \(180^\circ\).

Plotting the extraordinary Hall voltage (\( V_{EHE} \)) data for every angular step with respect to the rotation angle, \( V_{EHE} \) flips to a maximum or a minimum at \(-90^\circ\) and \(90^\circ\) due to magnetisation reversal (c.f figure 5.10 inset). The magnetisation rotation at angles near \(0^\circ\) and \(180^\circ\) does not show any significant difference confirming that the magnetisation processes at these angles are reversible. The value of \( \cos \theta \) is fitted to the parabolic function of Eq. 5.5 with respect to the rotation angle and the line of best fit calculated as shown in Fig. 5.10. From this fit the Fourier coefficient \( A_2 \) can be obtained and used to calculate the anisotropy field (\( H_k \)) as
\[ H_k = \frac{2K_u}{M_s} = H \frac{1 - \sqrt{2A_2}}{\sqrt{2A_2}} \]  \hspace{1cm} (5.8)

Measurement of the anisotropy field using the EHE is most appropriate for ultrathin films with strong intrinsic signals compared with more conventional techniques in which signals rely on the volume and geometry of the sample.

Figure 5.10: \([\cos \theta - 1]\) as a function of rotation angle. For field \(H = 650 \text{ mT}\) and \(A_2 = 0.123 \pm 0.001\), \(H_k = 660 \pm 6 \text{ mT}\). The curve through the data points is the \(\chi^2\)-squared best fit to Eq 5.5. Inset is a sketch of the Hall voltage as a function of rotation angle.
Summary
A low frequency AC measurement setup was constructed to measure EHE in perpendicularly magnetised Ta/Pt/Co/Pt Hall cross devices. A pulse generator was added to the low frequency EHE setup. A quasi-static field sweep created a minor hysteresis loop and using measurements at low frequency AC currents, the extraordinary Hall voltage ($V_{EHE}$) was recorded. The current source was switched to a pulse generator firing a single pulse and reconnected back to the AC source to measure the final $V_{EHE}$. In this way the minimum/threshold pulse current needed for domain wall motion was investigated. Kerr imaging was used to visualise the magnetic switching of Ga⁺ irradiated domains and domain wall dynamics in the Hall cross devices. A time-resolved EHE setup was constructed to carry out fast time-resolved domain wall motion studies and measure the domain wall velocity. Anisotropy field measurements were carried out using an angle-dependent EHE set-up in Leeds.
Chapter 6  Optimisation of Pt/Co/Pt multilayers for STT applications

6.1  Introduction

Magnetic domain wall (DW) motion in perpendicularly magnetised thin films has been widely studied for several years because of its potential applications in spintronic devices [126, 127] and in high density magnetic optical recording media due to the large Kerr rotation angles at short wavelengths [128, 129]. The advantages of having PMA in magnetic thin films are a stable magnetisation state and narrow domain walls, especially valuable for effective current-induced domain wall motion (CI-DWM) [130] by spin transfer torque (STT). On the other hand disadvantages can include the large magnetic switching fields due to strong pinning potentials and an associated increase in the threshold current density for STT DW motion [86].

Narrower Bloch like DWs have been found in materials with perpendicular magnetic anisotropy (PMA)[29], the archetype for which is a thin Co film ≤ 1 nm thick in a Co/Pt multilayer. In this thickness range, the DW pinning potential and pinning density are low [131] and reflected in easy DW motion which dominates the magnetisation reversal [132]. Co/Pt MLs have long been viewed as the best multilayer stack system for studying domain wall (DW) dynamics, particularly DW motion induced by current flow [133-136].

Non-magnetic Pt buffer layers are typically used in Pt/Co/Pt stacks for inducing (111) texture and to promote PMA in the Co layer. A Pt capping layer also is used to protect the Co layer against oxidation. While there has be no conclusive explanation for the origins of PMA, it has been strongly linked to the Co-Pt lattice mismatch at the interface inducing strain in the Co layer [137, 138].

The extraordinary Hall effect (EHE) can be used to characterise the magnetization of Co/Pt ML thin films. The only problem with EHE measurements of Co/Pt MLs is that the majority of the applied current is shunted through the thick Pt buffer layer and only a small fraction flows through the magnetic layer. This gives a relatively small Hall voltage signal and the same applies to current-induced STT DW motion where the current flowing into the nonmagnetic layers contributes very little to STT but generates significant Joule heating in the Co/Pt ML stack [135]. Hence it is important to minimize the thickness of the Pt buffer and capping layers.

In this chapter Co/Pt MLs with different Co and Pt thicknesses were characterised and optimised to have a sharp switching perpendicular anisotropy at a medium coercive field,
allowing our experiments to be performed at relatively low applied fields when coil heating is not an issue. Sharp switching of the magnetisation at the coercive field \( (H_c) \) is indicative of fast magnetisation reversal due to weak DW pinning. In this case the magnetic anisotropy controls the coercive field and hence the dependence of magnetic switching and coercivity on Co and Pt layer thicknesses was investigated. The effect of an additional ‘Ta’ buffer layer below the Pt/Co/Pt stack on the magnetic anisotropy was also explored. Finally, the effect of varying the Pt capping layer thickness on the magnetic anisotropy was studied with a view to minimising current shunting through non-magnetic layers.

6.2 Ta/Pt buffer layer thickness

The effect of variation in Pt buffer and capping layer thickness was investigated earlier by Wang et al., a former member of the group working on Pt/Co/Pt multilayer grown on Si/SiO\(_2\) substrate [139]. To limit the current shunting in non-magnetic layers while still inducing PMA in the Co layer, optimisation of Pt (111) buffer layer was carried out for a fixed Co and Pt capping layer thicknesses of 0.5 nm and 2.2 nm with different Pt buffer layer thicknesses ranging from 4.7 nm to 8.8 nm, based on similar thickness of Pt used in reference [139]. The results of Wang et al., are shown in figure 6.1 where the rms surface roughness decreased for increasing buffer layer thickness and the coercivity increased. The increase in coercivity can be attributed to an overall enhancement of (111) FCC texture of the Co and Pt layers [32, 138, 140, 141]. RMS surface roughness for thin 0.4 nm Pt thickness was high and this could be due to a larger SiO\(_2\) substrate roughness of ~1-1.3 nm on which Pt/Co/Pt films were grown. As mentioned above, owing to the maximum field limitations of our solenoid, our optimal coercive field range for the samples was 100-200 Oe. While this condition is satisfied for the entire Pt buffer layer thickness range in figure 6.1, the problem of strong current shunting limits the optimal Pt buffer layer thickness to thinner layers. Moreover, PMA in the thinnest Pt buffer layer was not reproduced consistently in Pt/Co/Pt MLs. To overcome problems experienced by Wang et al. in obtaining strong PMA in Pt/Co/Pt structures, we included Ta as an additional buffer layer as this has been shown to enhance the (111) texture of Co/Pt multilayers [142]. The 4 nm of Ta under layer thickness was optimised by our collaborators in Leeds and was recommended to use in our Ta/Pt/Co/Pt ML film growth.
Polar-MOKE M-H loops for three Co samples with different Pt thicknesses (2 nm and 3 nm) and 4 nm Ta combinations are shown in figure 6.2. The coercivity of the samples with the additional Ta buffer layer is 145±5 Oe, which is well within our target field range. Also, as mentioned in the device design section 4.4 of chapter 4, Ta 4 nm and Pt 3 nm thickness improves the impedance matching between the Hall cross and the 50 Ω coplanar waveguide structures. Although this doesn’t solve the Joule heating problem, it reduces reflections at the Hall bar structure. From figure 6.2 one sees that the coercivity of the Co/Pt sample without Ta is increased to 194 Oe and is greater than the value for samples with a Ta under layer, which would have been fine for our experiments had it not been for the lack of reproducibility of $H_c$ in other MLs having the same stack and thickness values. A comparison of two different Pt thicknesses, with Ta under layer, as a function of varying Co thicknesses in shown in figure 6.7. This will be discussed in the following section.
Figure 6.2: MOKE hysteresis loops for three different multilayer samples showing the influence of Ta and Pt buffer layer thicknesses. The PMA is strong and coercivity is reduced in Co/Pt MLs with the additional Ta buffer layer.

An X-Ray diffraction (XRD) measurement of the three samples is shown in figure 6.3 and gives an insight into the structural role of the Ta underlayer. It has been shown by Fukami et al.,[143] that the Ta buffer layer facilitates layer by layer growth of Pt and Co layers, which develop a thermodynamically stable (111) orientation. The diffraction patterns show two strong peaks in intensity for the samples with the Ta (4 nm) underlayer. There is no significant development of (111) texture for the Pt layer grown directly on a Si/SiO$_2$ substrate. There is only a broad and weak diffraction peak near the Pt (111) position at $2\theta = 39.765^\circ$, despite the Pt thickness being the same as in the case of the Ta underlayer sample. In contrast multilayer samples with the Ta underlayer exhibit significant (111) texture as inferred from the Pt (111) peak.

The Pt/Co/Pt multilayer shows a broad diffraction peak and has a low integrated peak area which may be due to the poor ordering of the Pt (3 nm) buffer layer. Ta/Pt/Co/Pt samples have narrower peak widths at full width half maximum (FWHM) suggesting improved crystalline order and homogeneity. Since the Co layer is very thin, of the order of 2-5 monolayers, any diffraction peak for bulk Co would be expected to be very weak [144]. The strongest diffraction peak for the Ta(4 nm)/Pt(3 nm)/Co(0.55 nm)/Pt(1.8 nm) sample lies
between the predicted position of Pt(111) and ordered fcc CoPt₃ (111) peaks. CoPt₃ alloy is an ordered face centred cubic phase whose (111) reflection is predicted to be at 2θ = 40.56° [145]. From the X-ray diffraction measurements, samples with a Ta (4 nm) and Pt (3 nm) underlayers exhibit significantly better crystalline order and were deemed to be superior for our magnetotransport experiments [144, 146].

Figure 6.3: X-ray diffraction data for Pt/Co(x)/Pt multilayers with and without a Ta underlayer.

6.3 Co layer optimisation
Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) multilayer films with varying Co layer thicknesses in the range 0.1-0.85 nm were grown and characterised using Polar MOKE measurements. Figure 6.4 shows MOKE M-H loops of different Co layer thicknesses with fixed Pt and Ta layer thicknesses. All samples show a sharp switching behaviour with the switching field increasing approximately linearly as the Co layer thickness increases. The sharpness of the rectangular M-H loops suggests strong perpendicular anisotropy in these samples. (111) FCC texture is known to lead to strained Co/Pt interfaces and strong out-of-plane anisotropy (Kₐeff) in Pt/Co(0.1-1 nm)/Pt multilayer stacks [33, 147]. In Pt/Co/Pt ML stacks with the Pt buffer layer
grown directly on the Si/SiO₂ substrate a strong (111) FCC texture does not develop. This is also observed by Emori et al.,[20] where it was shown that the addition of Ta underlayer to the Pt/Co/Pt stack enhances the (111) FCC ordering and increases the magneto-crystalline anisotropy (K_MC) along the crystalline hard-axis and increases the magnetoelastic ‘strain’ anisotropy (K_ME) due to the lattice mismatch between the Co and Pt layers [29, 148]. This can be described by the expression,

\[ K_{\text{eff} \ t_{Co}} = 2K_S + K_V t_{Co} , \]  

(6.1)

where \[ K_V = -2\pi M_S^2 + K_{MC} + K_{ME} , \]  

(6.2)

and K_S and K_V are the surface and volume contributions to the magnetic anisotropy. K_S is large in Co/Pt MLs due to the interfacial strain and asymmetry in the spin orbit coupling of the Co-Pt atoms near the interface. From [142], a plot of the product K_{eff} * t_{Co} for different Co layer thicknesses produces a shallow linear slope for Co/Pt MLs with a Ta underlayer as opposed to a sharp downwards slope for Pt/Co/Pt ML stacks. This is interpreted as the presence of an additional positive volume contribution to K_V counteracting the shape anisotropy (-2πM_S² in eq.6.2). This extra positive volume-like contribution, along with large contributions from K_MS and K_ME, result in it being easy to overcome the shape anisotropy. This could be the reason for an increase in the Co film thickness (t_{Co}) range over which perpendicular anisotropy is observed. In our Ta/Pt/Co/Pt ML films we observe PMA over the full range of Co film thicknesses 0.1-0.85 nm (c.f. Fig.6.4(a) & (b)), which is significantly wider than the range observed in Wang et al., for Co/Pt grown directly on Si/SiO₂ (t_{Co} : 0.4-0.7 nm) as shown in figures 6.5 (a) and (b) [139].
Figure 6.4: (a) MOKE hysteresis loops for Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) multilayer films with varying Co thicknesses. (b) Coercivity of Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) as a function of Co layer thickness.

Figure 6.5 taken from Wang et al.[139]: (a) MOKE hysteresis loops for Pt(4.7 nm)/Co(x)/Pt(2.2 nm) multilayer films with varying Co thicknesses. (b) Coercivity of Pt(4.7 nm)/Co(x)/Pt(2.2 nm) multilayer films as a function of Co layer thickness. Vertical lines divide regions of out-of-plane and in-plane anisotropy.

Figure 6.6 shows X-ray diffraction data for Ta (4)/Pt(3)/Co(x)/Pt(1.8) films as a function of Co thickness. We see that the Pt peak is largest and sharpest for the thinnest Co film, and weakens and very marginally shifts to higher angles as the Co thickness increases. This suggests that the degree of Co/Pt ordering decreases with increasing Co thicknesses. The shift to higher angles could reflect a change in average composition and/or a reduction of strain.
Finally figure 6.7 shows a comparison of the coercive field for Ta(4)/Pt(y)/Co(x)/Pt(1.8) multilayers with different Pt buffer layer thicknesses (y=2 and 3 nm) as a function of x(Co). The coercivity of films with y = 3 nm is uniformly higher than y = 2 nm and in both cases falls roughly linearly with approximately the same slope as a function of Co thickness.
Figure 6.7: Coercivity of Ta(4)/Pt(2/3)/Co(x)/Pt(1.8) multilayers as a function of Co layer thickness

6.4 Pt capping layer optimisation

Pt capping layer optimisation for a fixed Pt(4.7 nm) buffer and Co(0.5 nm) layer thicknesses in a Pt/Co/Pt multilayer stack (c.f. fig 6.8) was carried out by Wang et al., [139]. Change in Pt capping layer thickness did not yield any systematic trend in $H_c$ as shown in figure 6.8, taken from reference [139]. Even for the thinnest capping layer thickness of 2.2 nm, there is no sign of sample degradation (e.g. due to oxidation), indeed this sample has the highest $H_c$. MOKE data for a Pt capping thickness of 1.8nm, presently not available to show, had a very similar $H_c$ compared to other Pt capping thicknesses used in Wang et al. This Pt capping layer thickness 1.8 nm was considered as optimum and used in the Ta/Pt/Co/Pt ML stacks to minimise current shunting.
Figure 6.8: MOKE hysteresis loops for Pt(4.7)/Co(5)/Pt(x) multilayer films with different capping layer thicknesses. Reproduced from [92].

Summary

By optimisation of Ta/Pt/Co/Pt multilayer films, we could obtain a multilayer stack exhibiting strong PMA and sharp magnetisation switching indicative of weak DW pinning. Although the addition of a Ta buffer increases the current shunting through non-magnetic layers, it enhances the PMA and improves the impedance matching between the device and coplanar waveguide structures. With a Ta under layer, the impedance mismatch was greatly reduced by at least 33% compared with no Ta. This is crucial for our time-resolved EHE measurements. From the analysis so far, a Ta under layer thickness of 4 nm and a Pt capping layer thickness of 1.8 nm appear well suited for our EHE experiments. A Co layer thickness in the range of $x = 0.3–0.6$ nm and Pt buffer layer thickness of $y = 3$ nm in a Ta(4)/Pt(y)/Co(x)/Pt(1.8) exhibit strong PMA with a coercivity in the range of 130-165 Oe. The total thickness of the multilayer stack and $H_c$ with strong PMA appear to be well suited for time-resolved current-driven domain wall dynamics experiments.
Chapter 7 Control of coercive field

7.1 Introduction
Thin film magnetic materials with perpendicular magnetic anisotropy (PMA) have been widely exploited as building blocks for realising spintronic devices. Of these, Co/Pt thin films with PMA have been most intensively investigated due to their potential applications in magnetic recording media [30, 33, 149], and much work has been done to develop ways to control their magnetic properties. The relationship between the atomic structure and the magnetic properties in magnetic thin films/multilayers has been widely researched. In the case of Co/Au, a direct relation between the interface roughness and the magnetic anisotropy was shown [150]. In a perpendicularly magnetised multilayer system like Co/Pt, substantial changes to the Co/Pt atomic ordering at the interface set in at temperatures above 350 °C. At such high temperatures, the coercivity of the ferromagnetic material diminishes [151-155]. Interface sharpening in Ni/Cu multilayer was experimentally observed at 500 °C by atom probe tomography [156], but in this case the thicknesses of the layers were greater than 2 nm. In the case of ultra-thin Co films (< 1 nm), the Co/Pt interface is expected to be ordered with a high lattice strain. In such films, thermal effects such as annealing can also play a major role in controlling the magnetic properties. Any effect due to thermal effects is difficult to quantify microstructurally at the atomic level, but the magnetic properties, like coercivity and anisotropy, can be quantified using standard magnetic characterisation techniques.

Modifying the magnetic anisotropy by ion irradiation has become an established technique using Ga⁺ [19], He⁺ [157] and Ar⁺ [158] ions. In all cases increasing ion dose levels generally leads to a magnetization reorientation transition (MRT) from perpendicular to in-plane. This is accompanied by dramatic changes in the magnetic properties of the films, in particular the magnetic anisotropy [159] and coercivity [38]. These changes are believed to be related to light intermixing and strain relaxation at the Co/Pt interfaces as the ions pass through the multilayer interfaces at low dose levels, releasing the stress on Co atoms that is present due to the Co-Pt lattice mismatch [17]. At high dose levels it leads to milling [119] and implantation of ions inside the metals [116]. Ga⁺ ions, being heavier than He⁺ and Ar⁺ ions, are most effective at achieving this and lead to longer inter-diffusion lengths [17]. Hence even a rather low dose of Ga⁺ ions is sufficient to induce substantial changes in the magnetic properties of these systems.
7.2 Non-monotonic dependence of $H_c$ on FIB Ga$^+$ irradiation dose

Under some conditions, however, it has also been shown that interfacial mixing can lead to the formation of ordered Co-Pt alloys and an increase in the magnetic anisotropy [160]. Here we demonstrate such an increase in the coercive field of Ta/Pt/Co/Pt multilayers with PMA at very low Ga$^+$ ion doses.

Thin cobalt films exhibit PMA over a thickness range of about 0.3-1.0 nm when sandwiched between Pt buffer and capping layers. Here we used a 3 nm thick Pt buffer layer and a 1.8 nm Pt capping layer to prevent oxidation, in which case the strongest out-of-plane anisotropy is generally observed at Co thicknesses in the range 0.35-0.85 nm [139]. An additional 4 nm Ta buffer layer was included as it is known to promote (111) crystalline order, enhance the interfacial anisotropy and extend PMA over a wider range of Co film thicknesses [142]. Perpendicular magnetic anisotropy in Ta(4 nm)/Pt(3 nm)/Co(0.35-0.85 nm)/Pt(1.8 nm) MLs was confirmed using polar magneto-optical Kerr effect (MOKE) measurements. The perpendicular magnetization is inferred from the rotation of polarisation of the reflected laser beam that is incident on the sample in an applied out of plane magnetic field.

A linear Hall array consisting of five Hall crosses was used for these studies. Extraordinary Hall Effect (EHE) measurements were performed to characterize the M-H behavior of the multilayer films with a 32 Hz ac transport current of 4 µA and a magnetic field applied perpendicular to the plane of the multilayer. The coercivity ($H_c$) for unirradiated Hall crosses was typically ~100 Oe with a small variation from device to device due to slight sample inhomogeneities across the growth substrates. An ‘FEI Helio600’ combined SEM-FIB was used to lightly irradiate the active region of individual Hall crosses over an area of about 10×10 µm$^2$. A range of Ga$^+$ dose levels spanning 0 - 0.144 pC/µm$^2$ with a 30 keV ion energy, was explored to cover the evolution of anisotropy all the way from perpendicular to in-plane magnetization. Care was taken to strongly overlap adjacent Ga$^+$ ion pixels to ensure homogeneous irradiation of the film. After irradiation, EHE was again used to characterize the changes in the magnetic properties of the films.

A very large number of prior studies of the influence of low dose Ga$^+$ ion irradiation on Co/Pt multilayer samples with PMA have predominantly revealed a reduction of the magnetic anisotropy and coercivity followed by a spin reorientation transition to in-plane magnetization at very high Ga$^+$ doses [15, 19]. Recently, however, a spin reorientation from in-plane to out-of-plane magnetization upon irradiation with low doses of Ga$^+$ ions and vice versa at increasing doses has been observed in relatively thick in-plane magnetized Pt/Co(2.6
nm)/Pt samples [161]. Here we show a similar effect in much thinner Co-Pt samples which exhibited perpendicular magnetization as grown. EHE measurements as a function of Ga\(^+\) irradiation dose are shown for films with mean Co thicknesses of 0.35 nm, 0.55 nm and 0.85 nm in Figures 7.1(a), (b) and (c) respectively.

![EHE loops for different Co thicknesses](image)

Figure 7.1: EHE voltage as a function of applied magnetic field after various Ga\(^+\) FIB irradiation doses for Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) multilayer samples containing Co films of thickness (a) 0.35 nm, (b) 0.55 nm and (c) 0.85 nm.

The normalized coercive fields (normalized to \(H_c\) measured before irradiation) estimated from these EHE loops are plotted in Figure 7.2 as a function of Ga\(^+\) dose for the three different Co
thicknesses. Surprisingly, in all three samples we initially see the normalised coercivity increasing with increasing Ga\textsuperscript{+} ion dose. However, once the irradiation dose reaches a critical level the coercivity starts to drop again, and at higher doses the anisotropy goes in-plane in the two thicker Co films as revealed by a loss of loop squareness. This was confirmed by directly comparing the out-of-plane and in-plane EHE measurement of the highest Ga\textsuperscript{+} dosed (0.144 pC/\(\mu\)m\(^2\)) 0.85 nm Co sample. With the field applied in the sample plane only a very small maximum EHE voltage was observed which dropped to zero at high fields, as expected for a sample with in-plane magnetic anisotropy. A reduction of coercivity with Ga\textsuperscript{+} dose is well documented and believed to be due to weak intermixing at the Co-Pt interface combined with a reduction of strain, leading to a reduction in the interfacial anisotropy [15, 17, 19, 38].

![Graph showing coercivity normalized by that of the as-fabricated devices after Ga\textsuperscript{+} irradiation for three different Co thicknesses in Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) ML. Error bars were derived from the variation in the measured coercivity for several magnetization cycles. Solid lines are a guide to the eye. For the 0.55 nm thick Co sample, the value of \(H_c\) after FIB irradiation was a little higher than \(H_{Co}\) for 0 pC/\(\mu\)m\(^2\).]

7.3 Correlation between \(H_c\) and surface roughness

In order to try and establish a link between the coercivity and the sample microstructure, very careful Atomic Force Microscopy (AFM) was performed to characterize the surface roughness of active Hall crosses both before and after irradiation. Figs 7.3 (a) and (b) show topographic maps of a 0.85 nm Co sample after Ga\textsuperscript{+} doses of 0.0479 pC/\(\mu\)m\(^2\) and 0.0959 pC/\(\mu\)m\(^2\) respectively, corresponding to maximum and minimum coercivity in Figure 7.2. Typical line scans across both samples are shown in fig 7.3(c), and one sees immediately that the surface
roughness is much higher for the sample with highest coercivity. The characteristic rms surface roughness is plotted against Ga⁺ dose for all three samples in Figure 7.4 and, upon comparison with Fig. 7.2, a clear correlation between coercivity and surface roughness is observed at low irradiation doses.

![Figure 7.3](image)

Figure 7.3: Topographic AFM images of the central part of Hall crosses fabricated from a 0.85 nm Co film after Ga⁺ ion irradiation with doses of (a) 0.0479 pC/µm² and (b) 0.0959 pC/µm². (c) Line-scans along the directions indicated in the images (a), (b).
Figure 7.4: RMS surface roughness of films with three different Co thicknesses as a function of Ga\(^+\) irradiation dose. Error bars were derived from the variation in the roughness at several different regions of the sample.

However, at very high doses in the regime where the sample anisotropy becomes in-plane, the roughness starts to increase again. These trends are shown more explicitly for the 0.85 nm Co sample in Figure 7.5 where both coercive field and rms roughness have been plotted versus Ga\(^+\) ion dose in the same figure.

Figure 7.5: Detailed comparison of the normalized coercivity and the rms surface roughness as a function of Ga\(^+\) irradiation dose for the 0.85 nm thick Co sample.
7.4 Correlation between coercivity ($H_c$) and magnetic anisotropy ($H_k$) field

Though figures 7.2 and 7.4 strongly point to an increase in perpendicular magnetic anisotropy, coercivity can sometimes be influenced by the pinning potentials due to structural imperfections. A direct measurement of the magnetic anisotropy field ($H_k$) represents conclusive evidence for an increase/decrease in interfacial anisotropy after Ga$^+$ irradiation. The anisotropy field ($H_k$) was measured for the Co 0.85 nm sample using an EHE-based technique (c.f. fig 7.6). $H_k$ increases at low Ga$^+$ doses before falling again at high doses. The increase in anisotropy field is correlated with the rise in coercivity at low Ga$^+$ doses and both $H_k$ and $H_c$ follow the expected trend that at higher Ga$^+$ dose levels PMA reduces due to intermixing and/or strain relaxation at the interface and the magnetization becomes in-plane. Figure 7.6 represents strong evidence for an increase in PMA at low Ga$^+$ doses and shows that the increased surface roughness is linked to an enhancement in the interface anisotropy.

![Diagram showing the correlation between coercivity ($H_c$) and magnetic anisotropy ($H_k$) as a function of Ga$^+$ irradiation dose for a 0.85 nm thick Co sample. Dotted and solid lines are guides to the eye.]

Figure 7.6: Detailed comparison of the normalized coercivity and magnetic anisotropy field ($H_k$) as a function of Ga$^+$ irradiation dose for a 0.85 nm thick Co sample. Dotted and solid lines are guides to the eye.

7.5 Discussion

Our EHE measurements reveal that the coercivity of all our Cobalt thin films increases at very low Ga$^+$ doses. This enhancement grows up to a maximum Ga$^+$ ion dose, $D_{\text{max}}$, that increases from 0.036 pC/$\mu$m$^2$ to 0.048 pC/$\mu$m$^2$ as the Co thickness is increased from 0.35 nm to 0.85
nm. Above $D_{\text{max}}$ the coercivity starts to drop again and in-plane anisotropy is eventually recovered at very high doses (c.f., Figs. 1 and 2). Earlier studies [17, 19, 162] have established that Ga$^+$ irradiation leads to strain relaxation at the Co/Pt interface due to slight intermixing and a reduction of the abruptness of the chemical gradient. However, at low Ga$^+$ dose levels Co-Pt intermixing can be associated with the formation of ordered Co-Pt alloys at the interface, in particular CoPt$_3$ [160, 163]. The formation of ordered compounds would modify both the bulk and interfacial anisotropy terms in the effective anisotropy expression, $K_{\text{eff}}$, and have been previously implicated in a relatively thick Pt/Co(2.6 nm)/Pt sandwich structure [161]. In this case the as-grown film had in-plane anisotropy and the formation of CoPt$_3$ after low dose Ga$^+$ irradiation is believed to lead to PMA, with a magnetization reorientation into the plane at much higher doses. Maziewski et al. [164] have studied the influence of Ga$^+$ ion irradiation on a similar sandwich structure incorporating a Co wedge of thickness ranging from 0 to 5 nm. For Co thicknesses larger than about 1.3 nm, samples exhibited multiple magnetization reorientation transitions (MRTs). For example, in a 3.3 nm thick Co sample they initially observed a reorientation from in-plane to perpendicular magnetization at low Ga$^+$ doses related to the possible formation of Co$_1$Pt$_1$ alloy at the interface, followed by a return to in-plane magnetization at larger doses. At still higher Ga$^+$ doses a further magnetization reorientation transition to in-plane and then back to perpendicular magnetization was observed [164]. This second PMA regime at high doses was tentatively related to the formation of CoPt$_3$ alloy at the interfaces. It is noteworthy, however, that multiple magnetization reorientation was not observed in samples with Co thickness less than 1 nm. Indeed, a monotonic reduction in coercive field was observed in a 0.9 nm Co sample exhibiting perpendicular anisotropy before irradiation. Moreover the Ga$^+$ dose levels at which these authors see their first MRT is an order of magnitude larger than that at the coercive field peak in our samples.

Within the accepted expression for the effective anisotropy, $K_{\text{eff}}$, of our Ta/Pt/Co/Pt system shown in equation 6.1, we expect that both the volume anisotropy, $K_V$ and the interface anisotropy, $K_S$, will increase due to the formation of interfacial CoPt$_3$ [19, 165] at low Ga$^+$ dose irradiation. Hence both $K_V$ and $K_S$ could contribute to enhance PMA. The Ga$^+$ irradiation-induced formation of an ordered interfacial CoPt$_3$ alloy, leading to an enhancement in PMA, has been previously observed in a very similar system by Maziewski et al. [164] where it was investigated by HRTEM and XMCD. Results from these two techniques strongly indicated that increased perpendicular anisotropy could be associated with the presence of ordered Co-Pt alloys.
In Figures 7.4 and 7.5 we see that an initial increase in coercive field correlates well with an increase in surface roughness for all the three Co thicknesses studied. The roughening is most pronounced in the sample with the thickest 0.85 nm Co film, possibly because it has a larger source of Co for forming ordered Co-Pt alloys. Although ion-induced intermixing at the interface is generally expected to decrease strain, an increased strain would arise if Co combines with Pt to form CoPt\textsubscript{3} since the compound has a larger molar volume than the combined volumes of the elemental constituents. It is then reasonable that the increased strain will, in turn, lead to increased roughness at the surface of the multilayer stack as we observe. We can exclude the possibility that increased rms surface roughness arises from the swelling effect due to inhomogeneous irradiation at low doses [117, 118] since our FIB irradiation is performed with overlapping pixels and a pixel spacing such that each is dosed with >10 ions. Moreover, such swelling effects should be approximately independent of the specific thickness of the layers in our multilayer films, while we actually see a rather pronounced difference for samples with different Co thicknesses.

In practice the composition of the interfacial Co-Pt alloy will vary depending on the total Ga\textsuperscript{+} ion dose [160] and a continuous increase in both $H_c$ and roughness is expected up to the maximum dose, $D_{\text{max}}$. For dose levels higher than this the regions of ordered Co-Pt alloy probably become more granular at the nanoscale, with an associated strain relaxation coupled with a reduction in the surface roughness. This is particularly visible in Figs. 7.5 and 7.6 where one sees that in the 0.85 nm Co sample both the surface roughness and magnetic anisotropy ($H_k$) fall together along with coercivity between 0.05 pC/\textmu m\textsuperscript{2} and 0.09 pC/\textmu m\textsuperscript{2}. At still higher Ga\textsuperscript{+} dose levels intermixing broadens the interface still further leading to complete strain relaxation and in-plane shape anisotropy overcomes the bulk anisotropy. In this regime the surface roughness increases again consistent with earlier studies that revealed a high interfacial roughness for in-plane magnetized samples [166], possibly due to creation of a coarser-scaled granularity for the Co-Pt alloy formed. These observations open up unique possibilities to both increase and decrease the coercivity of ferromagnetic multilayer films with low Ga\textsuperscript{+} ion doses, and this could find important applications in novel spintronic devices.

7.6 Control of $H_c$ by low temperature annealing

Studies of the effect of annealing on ferromagnetic MLs have so far reported an increase in perpendicular magnetic anisotropy (PMA) for temperatures above 300 °C when annealed in vacuum [167]. In this case, Pt(3nm)/Co(0.6nm)/Al(2nm) samples grown on SiO\textsubscript{2} substrates
were subjected to varying oxidation times and annealing temperatures resulting in an enhancement in PMA due to the diffusion of and chemical changes induced by oxygen incorporation at both Co interfaces [167]. Oxidation of a Co film grown by evaporation on a Pt (111) substrate was shown to increase the Kerr rotation angle due to the formation of a Co/Co$_3$O$_4$ bilayer [168] for annealing temperatures in the range of 250-300 °C. Perpendicularly magnetised Co films 4-8 atomic layers thick grown by magnetron sputtering on single crystal Pt(111) and annealed in vacuum at 300 °C have shown similar increases in coercivity to values in excess of 1.2 kOe [169]. This was attributed to a partial structural rearrangement of the Co film from FCC to HCP, specifically a combination of 75% fcc(111) and 25% hcp Co layer lattice structure [169]. This observation is very unlikely to be related to changes seen in Pt/Co/Pt multilayer stacks grown on SiO$_2$ substrates, partly because of the presence of a polycrystalline Pt ‘substrate’ rather than single crystal Pt. The effect of annealing on Pt/Fe thin films grown using MBE also demonstrated a pronounced enhancement in perpendicular magnetic anisotropy at 523 °C leading to a large coercivity in the range of few kOe [170, 171].

The effect of annealing in air on the magnetic coercivity and anisotropy of Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) multilayers (ML) was studied using Extraordinary Hall Effect (EHE) measurements. Three Co thicknesses, t$_{Co}$, from the same set of samples used to study the coercivity enhancement by Ga$^+$ irradiation were investigated as a function of low temperature annealing in air.

Ta/Pt/Co/Pt samples were fabricated into Hall array devices and EHE measurements were performed to characterize the M-H behaviour of the multilayer films with a 32 Hz ac transport current of 4 μA and a magnetic field applied perpendicular to the plane of the multilayer. The coercivity (H$_c$) for unannealed Hall crosses was typically ~85±5 Oe for Co(0.35 nm), ~96±4 Oe for Co(0.55 nm) and ~105±3 Oe for Co(0.85 nm) with a small variation from device to device due to slight sample inhomogeneities across the growth substrates. After characterising the Hall crosses, the samples were annealed in air on a hot plate for 30 minutes at temperatures in the range 90 °C to 180 °C and then cooled to room temperature for measurement. The same set of samples were annealed for 30 minutes at every annealing temperature measured. Hall crosses of all three samples with different t$_{Co}$ were characterised using EHE M-H hysteresis loops to determine the increase/decrease in coercivity at each annealing temperature.
Figure 7.7 shows M-H hysteresis loops as a function of $t_{Co}$ revealing a gradual increase in coercivity up to 160 °C and suddenly rising above 1 kOe at 180 °C. The coercivity in all 3 MLs increased by a relatively smaller margin for every 10 °C increase in annealing temperature up to 160 °C.

Figure 7.7: EHE voltage as a function of applied magnetic field for Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) multilayer samples containing Co films of thickness (a) 0.85 nm, (b) 0.55 nm and (c) 0.35 nm after annealing for 30 mins at the indicated temperatures.
Such large increases in coercivity reflect strong enhancements in perpendicular magnetic anisotropy, i.e. the interfacial anisotropy at the Co/Pt interfaces, upon annealing. A plot of normalised coercivity (the ratio of coercivity after annealing divided by coercivity after initial device fabrication) versus annealing temperatures is shown in figure 7.8. The enhancement in interfacial anisotropy at 180 °C is directly shown to be linked to an increase of perpendicular anisotropy field (Hk) using an EHE-based technique. The difference in Hk at 90 °C and 180 °C is represented in a bar graph in figure 7.9. All three Co thicknesses tCo show ≥ 80-100% increase in anisotropy field upon annealing at 180 °C further promoting out-of-plane magnetisation. We are interested in temperatures in the range of 140°C– 180°C, as these are used for baking the resist coated samples for e-beam lithography.

Figure 7.8: Coercivity normalized by that of the as-fabricated device as a function of annealing temperature for three different Co thicknesses in Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) ML.
Figure 7.9: Anisotropy field ($H_k$) measurement on Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) ML stack for three different Co thicknesses at 180 °C and immediately after fabrication of Hall cross structures (90 °C). During fabrication the samples are coated with S1813 resist and baked at 90 °C for 30 minutes to realise lithographic patterns.

7.7 Discussion

Here, in the case of Ta/Pt/Co/Pt MLs, annealing in air above 170 °C increases the coercivity more than 10 times. Even at EBL resist baking temperatures of 150 °C there is a 20% increase in coercivity for all Co thicknesses. In contrast, similar work on annealing of Si/Ta(4 nm)/[Pt(0.2 nm)/Co(0.2 nm)]3/Pt(2 nm) MLs at 350 °C in high vacuum yielded a small increase in coercivity and anisotropy [172]. This was attributed to chemical sharpening of the interface and an increase in asymmetric spin-orbit interaction at the Co/Pt interface. The XRD data in chapter 6 for Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) MLs with three different Co thicknesses, show a reduced intensity and peak area/FWHM of Co/Pt (111) for thicker Co samples indicating a lower degree of atomic ordering. We speculate that annealing our Co/Pt MLs sharpens the interfaces and increases the interfacial strain, thereby increasing the PMA. A possible increase in Neel type surface anisotropy due to asymmetric spin-orbit coupling at magnetic/non-magnetic interfaces may also be contributing.

However this was not thought to be the reason behind the enhancement of PMA in Co(0.85 nm)/Pt(2.3 nm) bilayers grown on SiO2 [155]. X-ray Photoemission Spectra (XPS) for a Co/Pt bilayer annealed at 300 °C as a function of time [155] reveal that PMA increases due to strain effects induced by the Pt diffusion into Co. The normalised anisotropy and normalised coercivity for different Co thicknesses drawn from the data in figures 7.8 and 7.9 are shown in figure 7.10. In all cases there is a strong correlation between $H_k$ and $H_c$. From
figure 77.10 we can see that, for thicker Co films both the anisotropy and coercivity ratios are larger than for \( t_{Co} = 0.35 \) nm. One infers that the increased amount of interfacial strain is dependent on the volume of the ferromagnetic layer. This can be concluded from figure 7.10, where for \( t_{Co} = 0.85 \) nm, the anisotropy increases by a factor of 2.5 for \( t_{Co} = 0.85 \) nm and only \( \sim 1.6 \) for \( t_{Co} = 0.35 \) nm.

![Graph showing comparison of normalised coercivity and normalised anisotropy as a function of Co film thickness in Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) ML. In each case the parameter after annealing at 180 °C has been normalised by the value after annealing at 90 °C.](image)

**Figure 7.10:** Comparison of the normalised coercivity and normalised anisotropy as a function of Co film thickness in Ta(4 nm)/Pt(3 nm)/Co(x)/Pt(1.8 nm) ML. In each case the parameter after annealing at 180 °C has been normalised by the value after annealing at 90 °C.

**Summary**

Controlling or modifying the magnetic properties of ferromagnetic thin films could open up new ways to optimize processes for building spintronic devices. \( \text{Ga}^+ \) irradiation offers a whole new range of possibilities for increasing or decreasing the magnetic anisotropy in Co/Pt multilayer thin films, specifically after growth and fabrication. This was demonstrated to increase and then decrease coercivity in Hall cross structures fabricated from Co/Pt ML stacks. The interfacial roughness, which was inferred from the rms surface roughness measurement, correlates well with the coercivity data for different Co thicknesses. An increase in the measured anisotropy field, \( H_k \), at low \( \text{Ga}^+ \) doses for 0.85 nm Co samples confirms the increase in PMA, probably due to ordered Co/Pt alloy formation at interfaces. Large enhancements in \( H_c \) and \( H_k \) are observed upon annealing the Co/Pt ML films in air at relatively low temperatures. Although all Co thicknesses show a drastic increase in coercivity and anisotropy, the rate of increase in \( H_c \) and \( H_k \) is highest in thicker Co films. We attribute
the increased anisotropy on low temperature annealing to a sharpening of the interfaces associated with increased interfacial strain. Higher strains appear to be achieved in samples with thicker Co films.
Chapter 8 Domain imaging and time-resolved EHE measurements

8.1 Introduction

Continued advances in information technology are driving an increasing need to develop new forms of non-volatile magnetic memory devices. Recent efforts to generate and utilize spin polarised currents to control domain wall dynamics in ferromagnetic structures have led to several breakthroughs [5]. However, the current densities required to achieve spin transfer torque domain wall (STT-DW) motion are currently $\sim 5 \times 10^6$ A/cm$^2$ [13, 14] and are too high for practical applications.

The use of FIB irradiation is a well-established technique [18] for realising artificial domains and controlling the magnetic properties of systems with perpendicular magnetic anisotropy (PMA). The strength of PMA in these structures has already been discussed in chapter 6 as well as the influence of interface roughness, strain and intermixing at the Co-Pt interfaces [15, 16, 173]. We have also seen how Ga$^+$ FIB irradiation can relax the strain at interfaces, resulting in an increase or reduction of the film coercivity as described in chapter 7. Several investigations of artificial domain structures based on the local focused ion beam (FIB) irradiation of magnetic multilayer structures with perpendicular magnetic anisotropy have been published [15, 16, 173]. However, STT-DW motion in artificial magnetic domains in ferromagnetic multilayers has not yet been extensively explored. Artificial domains eliminate the need for an additional field pulse to nucleate a domain wall in a nanowire, and the local magnetic anisotropy can be tuned to control the parameters with the goal of achieving enhanced STT-DW motion. Magneto-optical domain imaging gives one a direct visual representation of domain wall dynamics in these artificial domain structures. Time-resolved experiments are a powerful approach to study the effect of pulsed currents on domain wall displacements, as well as to quantify the local/edge pinning potentials in the irradiated multilayer films. By monitoring the EHE at a single pair of Hall voltage contacts as the DW crosses them [125], the domain wall velocity can be systematically investigated in pulsed-current ‘time-of-flight’ experiments on uniformly irradiated single Hall bar devices.

8.2 Domain imaging in a non-irradiated nanowire

A study of domain nucleation and the propagation of a DW driven by an applied out-of-plane field were carried out using magneto-optical Kerr microscopy. For this study a Ta(4nm)/Pt(3nm)/Co(0.35nm)/Pt(1.8nm) sample was fabricated into a 5 μm wide 30 μm long
microwire attached to a large square pad as shown in figure 8.1. Ta/Au was deposited to realise contact pads for pulsed current injection and a ground connection respectively. The resistance of the microwire was 404 Ω. The magneto-optical images have all been subtracted from a reference image for clarity. The reference image was captured in a uniformly magnetised state with an out-of-plane magnetisation along the z direction.

![Figure 8.1](image)

Figure 8.1: Schematic of the experimental setup showing an optical image of the 5 μm wide Ta(4 nm)/Pt(3 nm)/Co(0.35 nm)/Pt(1.8 nm) microwire with a large contact pad at one end. The yellow coloured regions are Ta/Au and used as contact pads. A voltage pulse is fed into the Ta/Au pad electrodes on the left and the other end is grounded as shown.

**8.2.1. Domain nucleation with a pulsed current**

The Ta/Pt/Co/Pt sample is initially magnetised uniformly along +z by applying an out-of-plane field (H_z) and then the field is brought back to zero. Togawa et al have shown that current pulses can transform the magnetic structure in a uniformly magnetised state below the Curie temperature [174]. This can lead to the nucleation of reversed magnetic domains and vortices, either due to the excitation of spin waves or as a consequence of thermal fluctuations. Current pulses ranging from 2 V to 18 V with 100 ns duration were applied to nucleate reverse domains in the wire region. This created one or two domain walls (DWs) within the wire region. The large area pad region does not experience any reverse domain nucleation at these pulsed voltage amplitudes. Figure 8.2 shows the aftermath of pulsed current excitation in a 0.35 nm thick Co sample.
Figure 8.2: Kerr images showing multidomain nucleation in a 0.35 nm Co sample after (a) positive and (b) negative voltage pulses.

It can be seen that the current pulse does not create a uniform reverse domain state. Instead a multi-domain or a demagnetised state is created without a well-defined domain wall structure. For pulse voltages up to +10 V, there are no current-induced effects while for negative pulsed currents, reverse domain nucleation begins at -10 V. The nucleation of multiple domains is plotted in figure 8.3 where the fraction of reverse domains is plotted as a function of pulse voltage. From this experiment we infer a pulse voltage of less than 10 V can be used to induce current-driven DW motion, while above this value multi-domain nucleation will take place.
8.2.2. Current nucleation followed by field-driven domain wall motion (F-DWM)

Conventionally DWs are created by applying a reverse field to a uniformly in-plane magnetised nanowire [175]. In such cases the domain nucleation takes place in a large pad region; the domain grows and fully reverses in the pad region before entering the much narrower wire. Controlling the desired DW position in this way is challenging in PMA systems (here in this case one would try to place the DW at the pad end of the wire). Alternatively, a single current pulse can be used to create a multi-domain state and a small reverse field (-Hz) of ~10 Oe applied to coalesce the tiny reverse domains into a single domain state, thereby creating a DW near the pad end of the wire as shown in figure 8.4.

Figure 8.4: Kerr image showing a nucleated reverse domain with a DW near the pad end of the microwire. A 12 V voltage pulse was applied to the wire to nucleate multiple reverse domains followed by application of a 10 Oe bias field to form a single domain.
Field-driven DW motion describes the behaviour of DWs under the application of an out-of-plane field. After the domain nucleation process, the field required to move a domain wall was estimated from the magneto-optical Kerr microscopy images. This is called the propagation field. The magnitude of the out-of-plane field was increased, either positive or negative, and the magnetic state was monitored in real time. The field at which the change in the Kerr signal is 50% of that at complete switching is defined as the propagation field, $H_p$.

A single domain was created using a pulse voltage of 12 V followed by a small reverse field of about 10 Oe, after which the field was brought to zero. An out-of-plane field was applied with positive or negative sign as shown in figures 8.5(a) & (b). The propagation field, $H_p$, in positive fields was ~70-74 Oe and in negative fields $H_p$ ~ -111 Oe as shown in figure 8.6. Some DW bowing was seen in figure 8.6a as expected due to the sharp corners at the end of the microwire, before the DW completely depins, elongates and propagates into the pad region. This is the usual behaviour of DW motion when driven by a field in out-of-plane magnetised systems. Applying a negative field to the DW leads to transverse creep rather than true propagation up the wire.

![Kerr images after application of (a) positive and (b) negative fields to drive the DW out of or into the wire.](image)

Figure 8.5: Kerr images after application of (a) positive and (b) negative fields to drive the DW out of or into the wire.
Figure 8.6: Field driven DW motion starting from a uniform reverse domain state. The field amplitude is increased in a positive (red) and negative (blue) sense. The Kerr signal is represented as the mean pixel value of the up and down domains.

8.3 Ga\(^+\) irradiated artificial domains

Attempting to drive DWs by pulsed currents in unirradiated Ta/Pt/Co/Pt wires did not result in DWM below the domain nucleation pulse threshold (< 10 V). Above this value a multidomain state was formed in the wire. This was also the case after creating a single reverse domain in the wire with a bias field. Using Ga\(^+\) irradiation we have also explored artificial magnetic domains with a view to optimising structures for STT domain wall motion. An area of 10 µm \(\times\) 5 µm near the middle of the wire was irradiated with a range of Ga\(^+\) dose levels spanning 0 - 0.04 pC/µm\(^2\) with a 30 keV ion energy (indicated in figure 8.7 (a) to (g) with red dotted line), in order to find the dose level that realizes a suitable reduction in perpendicular anisotropy. Care was taken to strongly overlap adjacent Ga\(^+\) ion pixels to ensure homogeneous irradiation of the film. A clear difference in the magnetization switching was observed in the irradiated region after a FIB dose of 0.04 pC/µm\(^2\). Figure 8.7(a) shows the reverse saturated state (-\(H_z\)).
Figures 8.7(b) & (e) show the magnetisation reversal of the irradiated region at $|H_{\text{Ir}}|= 142 \pm 2$ Oe while the non-irradiated region switches at $450 \pm 5$ Oe as shown in figures 8.7 (d) & (g). However, the switching of the pad region occurred earlier at $330 \pm 10$ Oe (figures 8.7 (c) & (f)) reflecting the influence of the much larger pad area.

Figure 8.7: (a-g) Kerr images of a field cycle showing clear switching of the irradiated artificial domain before the non-irradiated regions reverse. Red dotted line indicates the irradiated area of the wire (b) A schematic M-H loop showing corresponding regions of magnetisation reversal in (a) to (g).

8.3.1 Current-driven domain wall motion (C-DWM)

In order to study current-driven DW motion, a minor loop state was created by applying a small field to switch the irradiated part of the wire as shown in figure 8.7(b) and the field brought to zero without proceeding to full saturation as shown in 8.7(c) & (d). Applying a DC pulsed current should induce a spin transfer torque on the DW causing it to propagate. Based on the domain nucleation by pulsed current experiments in section 8.2.1 we know that a maximum threshold pulse voltage of $\pm 8$ V can be applied to move the domain wall without demagnetising the domain configuration in the irradiated region.

Figure 8.8 (a) shows no DW displacement after application of as single 8 V voltage pulse of 100 ns duration. The DW on the right hand side is expected to move first along the electron
flow direction as it can easily climb down from a high anisotropy to a low anisotropy region. After applying a chain of 275 pulses to the wire, part of the right hand side domain wall (RHS-DW) bends at the top as seen in figure 8.8(b). This situation is only maintained while the pulses are being applied and the DW reverts to its original position once the pulses stop. This kind of DW bowing is related to the Oersted field due to the current pulses [176, 177]. There is no influence on the left hand side DW (LHS-DW), in large part due to the fact that LHS-DW has to climb over the anisotropy energy barrier (from the low anisotropy region to the high anisotropy region) at the boundary of the irradiated region. Based on the fact that the electron flow direction is to the left one expects that the right hand DW will move first. We infer that DWs are quite strongly pinned at the boundaries of the irradiated region. Even the RHS-DW, which is moving down the anisotropy barrier, seems to exhibit quite strong pinning.

Figure 8.8: (a) Kerr image with an artificial domain in the wire after a single 8 V pulse with 100 ns duration. (b) Image during application of a continuous chain of 8 V pulses with 100 ns duration, and a repetition period of 10 ms. Bowing of the RHS-DW can be seen.

### 8.3.2 Field-biased current-driven domain wall motion in artificial domains

To overcome the DW pinning potential, a small bias field of less than the switching field of the irradiated region can be applied. An applied bias field allows the voltage pulse amplitude for DW motion to be minimised. Figure 8.9 shows field-biased DW displacements for different pulse amplitudes and a bias field $H_B = 52$ Oe, ($H_{\text{irrad}} = 142 \pm 2$ Oe). After creation of a minor loop artificial domain state and application of $H_B$, a voltage pulse with amplitude in the range $\pm 4-6$ V and 100 ns duration was applied. The number of pulses applied varies in each case. Figs. 8.9 (a) & (b) show that 170(275) positive(negative) 4 V voltage pulses with 100 ns duration at a repetition period of 10 ms, were required to completely reverse artificial
domains. Figs. 8.9 (c) & (d) show that 115(120) positive (negative) 6 V voltage pulses with 100 ns duration at a repetition period of 10 ms were required to fully reverse the artificial domains by DW motion. In figs. 8.9 (a) and (c) with a positive pulse amplitude, little influence of the spin transfer torque (STT) acting on DWs is observed. Instead more gradual DW creep appears to be taking place, in part due to the applied bias field. In figs. 8.9 (b) and (d) a negative pulse amplitude applied to the wire reveals some influence of the STT on the motion of the LHS-DW. The bias field probably also contributes to the DW motion in this case. The fact that the RHS-DW does not creep much into the low anisotropy region suggests that STT is driving it to the left where it is pinned due to the anisotropy barrier into the high anisotropy region. The reason why we observe a clear effect of STT on DW motion with negative pulse voltages and not positive ones is not clearly understood. However we note that the two DWs on the left and right are not exactly equivalent as shown in fig. 8.7. This is also reflected in the fact that during domain nucleation by voltage pulses as shown in figure 8.2, multidomain nucleation occurs for -10 V pulse amplitudes and not for +10 V. It appears that the proximity of the DW to the large pad influences the efficiency of STT-driven motion, possibly due to the modified magnetostatic energy in its vicinity.

DW velocity and critical current density estimates cannot be made in this case due to the strong pinning of DWs in this device and the very slow DW propagation. These magneto-optical imaging experiments were, however, particularly useful in helping us to understand the structure of artificial domains and the dynamics of DW motion in Pt/Co/Pt artificial domain structures. This was extremely valuable in informing the design of artificial domain structures for time-resolved DW motion experiments.
Figure 8.9: Kerr images showing the effect of multiple voltage pulses on DW motion in artificial domain structures. A magneto-optical image of a reversed artificial domain is used as a reference image for subtraction in order to resolve DW motion. Therefore the DW motion represents magnetisation reversal of the artificial domain towards the direction of non-irradiated regions.


8.4 Domain wall creep in Ga\(^+\) irradiated artificial domains

Having used Kerr microscopy to confirm that Ga\(^+\) irradiation produces an isolated pair of DWs at the artificial domain boundaries in Ta(4 nm)/Pt(3 nm)/Co(0.35 nm)/Pt(1.8 nm) multilayer samples, a study of magnetization reversal processes was carried out using low frequency ac-transport measurements to detect DW motion. A 2 μm wide single Hall bar was fabricated by optical lithography with the same sample layer structure as was used for domain imaging. For the ac transport measurements, an AC current of 4.5 μA at a frequency of 32 Hz was applied to the current arm of the Hall bar and the Hall voltage measured between an orthogonal pair of voltage contacts. The current density was small enough (~2.5×10\(^4\) A/cm\(^2\)) so that any spin transfer torque effects could be neglected [85] and the applied field was solely responsible for DW motion. Ga\(^+\) FIB irradiation was used to realise artificial domains (in this case the dose was 0.0716 pC/μm\(^2\) which will be called by the name ‘H-1’) around the active region of the Hall bar as shown in figure 8.10(a). The resulting structure was characterised by measuring M-H loops to confirm the perpendicular anisotropy (c.f. figure 8.10(b)) and coercive field (\(H_c^{H-1} = 43\) Oe) of the low anisotropy region in the sample.

Using Kerr microscopy, we were able to observe the magnetization switching of the irradiated and non-irradiated regions (c.f. figure 8.11 (a) to (d)) in a 0.35 nm thick Co sample. The same

![Figure 8.10](image_url)
reversal sequence is observed at negative magnetic fields. The stepwise switching process can be clearly be seen in the hysteresis loop generated from the Kerr microscopy imaging data as shown in figure 8.11 (d). The FIB irradiation has evidently reduced the magnetic anisotropy in the central Hall cross region leading to DWs at the boundaries. These results agree well with the EHE measurements obtained on the same sample though the coercive field on the Kerr microscopy is a little over estimated, because of the considerable height of the sample above the magnetic pole piece where the field is calibrated.

Figure 8.11: 0.35 nm thick Co sample locally irradiated with a 0.0716 pC/μm² Ga⁺ dose. The Kerr signal data were obtained from the red rectangular irradiated region of the Hall cross. (a) to (c) represent different stages of the magnetic switching of the irradiated and non-irradiated regions. (a) Represents the initial saturated state in a large negative field. (b) On application of a small reverse field the irradiated region switches. (c) At higher reverse fields the non-irradiated parts of the sample switch. (d) Hysteresis loop generated from the Kerr microscopy data clearly showing the different magnetisation states in the Hall bar.
Before starting creep measurements, the sample was saturated in a positive field to bring it into a single domain saturated state (c.f., fig. 8.10(b)) followed by a small reverse field (~ -80 Oe) to nucleate the reverse artificial domain in the irradiated area similar to the magnetic state seen in figure 8.11(b), without reversing the non-irradiated region. The applied field was brought to zero and a positive bias field, \( H_B \), less than the coercive field (fig. 8.10(b)) of the irradiated region is set to initiate DW propagation. Magnetic imaging in section 8.4 has shown both DWs creeping into the irradiated region under the influence of a small \( H_B \). In the case of a Hall bar with the central cross region irradiated, at least one DW has to cross the active region for a change in Hall voltage to be detected. In order to ‘set’ the DW that would first cross the voltage contacts, the irradiated area is asymmetric along the current arm of Hall bar, i.e., the edges of the 9.5 × 10 \( \mu \)m\(^2\) irradiated area are not equidistant from the centre of the Hall cross. Hence, when both DWs propagate in the bias field, the nearest DW in the current arm will reach the active region first and be detected.

DW creep was studied by monitoring the EHE voltage (c.f., fig. 8.12(a)) for various bias fields below \( H_c^{H-1} \). The time, \( \tau \), for the nearest DW to propagate a known distance \( d \) into the centre of the Hall cross is assumed equal to the time for \( V_{EHE} \) to reach half the full reversal voltage. For \( H_B \) above 38 Oe the wall propagation is fast as seen in the sharp rise in estimated velocity in figure 8.12(c). To study the effect of Ga\(^+\) irradiation on DW motion, another 2 \( \mu \)m wide Hall bar of the same multilayer was irradiated with a Ga\(^+\) dose of 0.076 pC/\( \mu \)m\(^2\) (called by the device name ‘H-2’) with \( H_c^{H-2} = 34 \) Oe. As in the previous case there was a sharp rise in creep velocity for \( H_B > 27 \) Oe (c.f. fig. 8.12(c)) and for bias fields above 31 Oe DW motion was too fast to measure with our low bandwidth setup. For these two radiation doses, there was only a marginal difference in DW creep velocity, though it is slightly faster in the more strongly irradiated device. The maximum measurable velocity of ~1 \( \mu \)m/s is a little faster than was previously reported for Pt/Co/Pt multilayers with PMA [72, 73].

In the creep regime the field-dependent DW velocity \( v(H) \) is given by [72, 73, 178],

\[
v(H) = v_0 \exp \left[ -\beta U_c \left( \frac{H_c}{H} \right) \right]^\mu, \tag{8.1}
\]

where \( \beta = 1/kT \), \( U_c \) is the pinning energy, \( v_0 \) is a prefactor and \( \mu \) is the dynamic constant which is expected to be \( \frac{1}{4} \) for a 1D domain wall moving into a 2D disordered medium [72-74].
Figure 8.12: Raw DW creep data for a Ta(4 nm)/Pt(3 nm)/Co(0.35 nm)/Pt(1.8 nm) ML Hall bar irradiated with a (a) 0.0716 pC/μm² Ga⁺ dose on device ‘H-1’ and (b) 0.076 pC/μm² Ga⁺ dose on device ‘H-2’. (c) Plot of estimated DW creep velocity as a function of bias field for the two different Ga⁺ irradiation doses.
The data from figure 8.12(c) are plotted as \( \ln(v) \) versus \( H^{-1/4} \) in figure 8.13 and show relatively good linear behaviour. This points towards a thermally-activated DW creep mechanism. The velocity for the irradiated Hall bar with the highest dose is clearly higher which points to a decreased pinning energy for DW motion. Linear fits to the data in Fig. 8.13 allow one to deduce the effective critical field \( H_c^{\text{eff}} = (\beta U_c)^4 H_c \) [73] from the fitted slope. For device H-1, the pinning energy, \( U_c = 1.7 \pm 0.16 \text{ eV} \) and \( H_c^{\text{eff}} = 9.4 \pm 3.6 \times 10^8 \text{ Oe} \) and for H-2, \( U_c = 1.3 \pm 0.14 \text{ eV} \) and \( H_c^{\text{eff}} = 2.4 \pm 1.5 \times 10^8 \text{ Oe} \). There is a significant reduction in magnitude of \( H_c^{\text{eff}} \) for H-2 compared to H-1 as expected for a sample with higher irradiation dose. The pinning potential \( U_c \) is also significantly lower for H-2 leading to faster DW creep.

![Figure 8.13: Log of DW velocity data versus \( H^{-1/4} \) in Ta(4 nm)/Pt(3 nm)/Co(0.35 nm)/Pt(1.8 nm) ML Hall bar. Straight lines are linear fits to equation 8.1 (see text).](image)

### 8.5 Time-resolved extraordinary Hall effect (EHE) in artificial domains

For the time-resolved EHE measurements, a Ta/Pt/Co/Pt sample with \( t_{\text{Co}} = 0.55 \text{ nm} \) was chosen, based on the optimisation of the ferromagnetic layer thickness to minimise current shunting with the aim of seeing a stronger STT effect in DW motion studies. The sample was patterned into a 2 \( \mu \text{m} \) wire width Hall bar using electron beam lithography (EBL). The resistance of the current arm of the Hall bar was \( \sim 475 \Omega \). The structure was designed with an asymmetry in the length of current leads either side of the voltage leads, and the shortest
current arm is grounded through a 50 Ω surface mount resistor as shown in figure 8.14(a). The EBL pattern was asymmetric to allow asymmetric irradiation with the lowest possible current lead resistances. The device was asymmetrically FIB irradiated to reduce the local anisotropy in the active region over an area of 4.2 × 4 μm² containing 0.55 μm of the short current arm and 1.65 μm of the long current arm on either side of the central active region. The 0.55 nm thick Co sample had to be irradiated twice in order to sufficiently reduce the coercivity of the irradiated region with total Ga⁺ dose of 0.0791+0.053 = 0.1321 pC/μm². The asymmetry in the irradiated area means that when DWs are formed at the edges of artificial domains, the DW in the shortest current arm will be closest to the active region while the other DW in the long current arm will be far from it. The coercivity of the irradiated Hall bar was characterised using the low-frequency EHE setup and the switching field of the artificial domain was found to be ~43±2 Oe and PMA in the irradiated region confirmed (c.f., figure 8.14(b)). A systematic investigation of different pulse voltages & durations was carried out to determine the critical pulse current required to achieve DW motion using the circuit shown in figure 5.6.

![Figure 8.14](image.png)

Figure 8.14: (a) SEM image of a 2 μm wide Hall bar grounded through a 50 Ω resistor at one end of the current lead. The red square shows the Ga⁺ irradiated area. (b) M-H loop of the Co 0.55 nm sample before (blue line) and after (red line) irradiation measured using the low-frequency EHE measurement set-up. The coercivity of the M-H loop before irradiation (blue line) was far greater than the field range shown.

The device was then connected to the time-resolved EHE experimental setup described in chapter 5, section 5.4. As a first step the time-resolved extraordinary Hall voltage, V_EHE, was measured in saturation. First the entire Hall bar was magnetised to a uniform up (down) state and a voltage pulse of 5 V amplitude with 110 ns duration was fired from the pulse generator into the current arm of the Hall bar, generating a current density ~5.6×10⁷ A/cm². The V_EHE
detected at the Hall voltage leads was collected by the differential probe and fed into the
digital signal analyser (DSA) where the output pulse voltage was measured. For averaging
purposes a train of 2048 voltage pulses was fired into the current arm of the Hall bar in an up
(down) state and the averaged $V_{EHE}^{Up(Down)}$ obtained. There is a substantial offset voltage in the
two $V_{EHE}^{Up(Down)}$ datasets which is probably due to inhomogeneities in the sample material
leading to inhomogeneous current flow in the Hall device. As a consequence the longitudinal
sample voltage is 'rotated' into the Hall voltage. This was eliminated by repeating the above
process for a uniform down(up) domain state and the EHE data for these two saturated domain
states (shown in figure 8.15a) were subtracted to remove the offset voltage and obtain twice
the true $V_{EHE}$. Half the voltage difference ($V_{EHE}/2$) reflects the extraordinary Hall voltage of a
single domain state. This is shown in fig. 8.15b where a value of around 625±5 μV is
estimated.
Figure 8.15: (a) EHE voltage signal obtained for two reverse saturated single domain states. (b) Difference of up and down EHE signals constructed to remove the offset voltage. The red line is the raw data after Gaussian smoothing.

Due to imperfect impedance matching in our circuit the pulse current 'rings' after the initial abrupt rise exhibiting a peak at the onset with the current settling down to a steady value in the second half of the pulse. The amplitude of the initial current peak in figure 8.15(b) is about twice the steady state value.

8.6 Time-of-flight measurements of domain wall motion driven by spin transfer torque in artificial domains

Before performing time-resolved EHE measurements, the Hall bar was driven to saturation in the positive field direction followed by field reversal until the artificial domain switches at -22 Oe, when the field was brought back to zero (i.e., a minor loop). A bias field, $H_b$, was then applied and a single voltage pulse of known pulse length was fired into the current leads of the Hall bar. This entire process was repeated 2048 times to obtain the average EHE voltage signal. The entire measurement process was then repeated for different bias fields and pulse voltages. A reference signal was obtained in the same way except that the reversal field applied to switch the artificial domains was held at -170 Oe and not brought to zero to try and ensure that there is no DW motion when the pulse fires. Raw $V_{\text{EHE}}(t)$ data are shown in figure 8.16. The reference signal has then been subtracted from the Hall voltage signals at different
values of $H_b$ to obtain the raw time-resolved extraordinary Hall voltage data. In figure 8.17 these subtracted Hall voltage data are plotted at different bias fields and compared with the $V_{EHE}$ expected after full reversal of the artificial domain which is shown as a red dotted line.

The spin polarised electrons travelling opposite to the classical current flow direction will induce a STT on the right hand DW causing it to propagate to the left. A DW moving across the active Hall bar region will be detected in the Hall voltage due to a change in magnetisation of the active region. Care had to be taken to ensure that bias field applied did not cause the DWs in the Hall bar to creep as seen in magneto-optical images of 5 μm wide microwires in figures 8.9 (a) & (c). This could usually be neglected due to fact that the entire measurement takes place much faster than the characteristic time for DWs to creep and induce magnetisation reversal in the active region. Note that the right hand DW in the shortest current arm is in very close proximity to the active region and will be assumed to reach the active region first in all experiments.
Figure 8.16: Raw EHE data obtained at different bias fields and a reference EHE signal at $H_B = -170$ Oe which is assumed to represent zero DW motion. The voltage pulse applied to the Hall bar is of 5 V amplitude and of 110 ns duration.
Figure 8.17 (a) & (b): Time-dependent $V_{\text{EHE}}$ obtained at different bias fields, $H_B$, for a voltage pulse amplitude of 5 V and 110 ns pulse length. The red dotted line is the extraordinary Hall voltage expected after full magnetisation reversal of the artificial domain as estimated from figure 8.2.

For $H_B = 10\text{--}16\ Oe$, the $V_{\text{EHE}}$ in the constant current region increases and this is seen as evidence that the nearest DW has entered the active region, although reversal is incomplete.
For $H_B = 18-20$ Oe, the value of $V_{EHE}(t)$ in the constant current region is still less than the estimated fully reversed magnetisation value of 1.25 mV. This end magnetisation state at large $H_B$ could reflect a DW pinned diagonally between two corners of the Hall cross as was observed in the Kerr microscopy image of figure 8.18 captured after a 4 V pulse voltage at $H_B = 35$ Oe, in a 2 μm wide optically-patterned Hall bar containing an artificial domain. This observation was reproduced 3 times consecutively after which the device failed to survive.

In the constant current region the $V_{EHE} = 625 \pm 5 \mu V$ measured in figure 8.15b represents the extraordinary Hall effect of one uniform saturation magnetisation direction and a switch from $V_{EHE}^{down} = -625 \pm 5 \mu V$ to $V_{EHE}^{up} = +625 \pm 5 \mu V$ represents full magnetisation reversal (~1.25±0.05 mV) of the artificial domain. In the subtracted saturation magnetisation signals in figure 8.17(a), $V_{EHE}$ of the constant current region is ~650 μV, which is close to half the value of full magnetisation reversal, consistent with a picture of DW pinning diagonally across the active Hall bar region. Under the assumption that the subtracted reference trace represents no DW motion, the extracted $V_{EHE}(t)$ must always be greater than zero in constant current part. However for $H_B = 0$ to -20 Oe, the value of $V_{EHE}$ falls below 0 V in the constant current region. This is currently not well understood and may be related to incorrect assumptions about the reference signal.
The initial peak in figure 8.17 for all the EHE measurements at $H_B \geq 10$ Oe results from a combination of current ringing and dynamic magnetisation reversal that possibly precedes a DW rebound before settling down to a stable value. We assume that the time to the first $V_{EHE}$ peak represents the initial domain wall motion and that the peak EHE voltage is proportional to the initial DW displacement. The much smaller initial peak for $H_B < 10$ Oe is consistent with this picture since the DW will not have entered far into the active region.

In order to determine the DW velocity, the peak $V_{EHE}$ at the initial rise in pulse voltage ($V_{EHE}^{\text{Peak}}$) is analysed. In Fig. 8.15(b) we assume that the Hall current ($I_H$) is doubled at the peak (c.f., Fig. 8.15(b)). Hence full magnetisation reversal leads to a change in $V_{EHE}$ of $2 \times 1.25$ mV. DW motion is only detected when it passes through the ~2 $\mu$m wide active region, and the distance the DW propagates in this region, $\Delta x$, is given by

$$\Delta x \approx \frac{2 \ \mu m}{2 \times 1.25 \ \text{mV}} \times V_{EHE}^{\text{Peak}}, \quad (8.2)$$

The time taken for the DW to reach $V_{EHE}^{\text{Peak}}$ is called the DW propagation time, $t_{\text{peak}}$. The estimated velocity, $v$, of the nearest DW (under bias field $H_B$) is hence given by

$$v_{DW} \approx \frac{0.55 \ \mu m + \Delta x}{t_{\text{peak}}}, \quad (8.3)$$

where 0.55 $\mu$m is the distance the DW has to travel in the irradiated length of the short current arm before entering the active region and $\Delta x$ is the distance travelled by the DW in the active region. The estimated DW velocity as a function of bias field is plotted in figure 8.19. The current density, $j$, in the 0.55 nm Co layer along the current arm is calculated as follows,

$$\text{Resistance of Co layer } (R_{Co}) = \rho \frac{l}{A} = \rho \frac{l}{(w \times t_{Co})},$$

where $l$ is the length of the current arm (6 $\mu$m), $\rho_{Co}$ is the resistivity of Co layer ~ 1 $\mu$Ω.m and $A$ is the cross-sectional area, which is width of the current arm (2 $\mu$m) times the thickness of the Co layer (0.55 nm). The resistance of the cobalt layer is calculated to be 5.45 kΩ.

$$\text{Current through the Co layer } (I_{Co}) = \frac{V}{R_{Co}},$$

where $V$ is the pulse amplitude voltage = 5 V.
Current density in the Co layer \( J_{Co} = \frac{I_{Co}}{A} \),

\[ J_{Co} = \frac{V}{(w \times t_{Co})R_{Co}} = 8.3 \times 10^7 \text{ A/cm}^2 \]

Figure 8.19: DW velocity estimated by relating the \( V_{EHE}(t) \) peak to the DW displacement. The current density of the Co layer, \( J_{Co} = 8.3 \times 10^7 \text{ A/cm}^2 \).

As expected the DW velocity is highest (~ 112±4 m/s) near the reverse switching field of the irradiated domain and is about 20% smaller (~90±4 m/s) at a bias field of 10 Oe, which is slightly less than half the switching field in this case. There is a linear increase in DW velocity from a small bias field of 12 Oe up to \( H_B = 20 \) Oe. There is no evidence of Walker breakdown in our data [100]; the DW velocity increases monotonically with bias field and would be expected to decrease once the Walker breakdown field [100] is exceeded. This is probably due to the relatively low pulse current density used. The average steady state current density of the current arm of the Hall bar ~5.6 \times 10^7 \text{ A/cm}^2 is less than the estimated density in just the Co layer alone. This suggests that a little more of the current flows in the magnetic Co layer than might initially be expected, but current shunting through Pt and Ta layers is still very significant.

In the works published so far for STT driven DW motion [179-181], current densities are in the range of \( 1 \times 10^7 – 10^8 \text{ A/cm}^2 \) for DW velocities of order 100 – 150 m/s. In such
experiments, a strong spin torque was cited as the main reason for DW motion at these velocities. Moreover, other explored materials like CoFeB [182, 183] and TbFeCo [184, 185] have resulted in similar DW velocities for current densities \(< 1 \times 10^7 \text{ A/cm}^2\). This is less than current densities for our velocity measurements and attributed to a strong spin Hall current. Recent efforts to inject current vertically has resulted in a large increase in DW velocity up to \(\sim 500 \text{ m/s}\) for current densities in the range of \(2-7 \times 10^6 \text{ A/cm}^2\) [14]. Probing DW dynamics using time resolved EHE with vertical current injection could be useful in achieving higher DW velocities at lower current densities.

Summary

Magnetic domain imaging experiments on a 0.35 nm thick Co sample reveal that pulsed current injection leads to the nucleation of a multidomain state in the wire with net magnetisation close to zero. The maximum pulse voltage amplitude of \(\pm 8 \text{ V}\) that could be applied to drive the DWs was also inferred. Application of a small bias field allowed a single domain to be formed with DWs at each end. Field-driven DW propagation was studied by applying positive and negative fields perpendicular to the film. Anisotropic DW creep was observed under negative fields which are expected to push the DW uniformly into the wire without changing its structure. Artificial domains were realised in the wire by FIB Ga\(^+\) irradiation and field-biased current-driven DW motion experiments performed to study the effect of spin transfer torque on DW motion. With pulse amplitudes in the range \(\pm 4-6 \text{ V}\) and pulse lengths of 100 ns, the current density in the wire region was \(2.16-3.24 \times 10^7 \text{ A/cm}^2\). STT-DW motion was observed that was dependent on the current direction. DW creep measurements were carried out as a function of bias field to determine the effect of locally reduced coercive fields in artificial domain structures on DW creep velocity. These showed a reduced pinning potential after higher irradiation levels leading to faster DW creep. Time-of-flight DW motion experiments were carried out to investigate STT-induced DW motion in the artificial domain region of the Hall bar by measuring the time-resolved EHE voltage. Under some simple assumptions the time-resolved EHE measurements were analysed to estimate the velocity of DW motion as a function of bias field. For an average pulse current density of \(5.6\pm0.2 \times10^7 \text{ A/cm}^2\), the velocity was found to be in the range of 80-120 m/s.
Chapter 9 Summary and future work

Co/Pt multilayer films have been grown using magnetron sputtering and strong PMA achieved for small Co thicknesses. Optimised films for Hall cross structures comprising Ta(4 nm)/Pt(3 nm)/Co(0.35-0.85 nm)/Pt(1.8 nm) multilayers were micropatterned on 5 mm² and 3.75 mm² Si(525 μm)/SiO₂(90 nm) substrates using standard lithographic techniques. Artificial magnetic domains were realised by tuning the magnetic anisotropy using Ga⁺ FIB irradiation. Low frequency EHE measurements were used to characterise the magnetic properties of unirradiated and irradiated samples, allowing the calibration of the FIB irradiation dose required to realise optimised structures for STT experiments.

After realising artificial domains, domain walls were created in the current leads of a Hall bar structure for current-driven domain wall motion experiments at static bias fields. To study the domain wall dynamics upon application of a current and a bias field in a magnetic wire, domain imaging experiments were carried out on Ga⁺ FIB-irradiated 5 μm wide wires fabricated by optical lithography. Application of large currents at H = 0 produces a multi-domain state with zero net magnetisation, and the maximum amplitude of pulsed currents that could be applied to drive domain wall motion (DWM) in the wire was inferred. The effect of spin transfer torque driven DWM was observed in the wire when pulsed currents were applied together with a small bias field. The domain wall creep was investigated in a 2 μm wide Hall bar composed of a similar ML stack. EHE measurements were used to determine the effect of static bias fields on the domain wall creep velocity without a drive current.

A time-resolved EHE measurement setup was designed and built to measure the transient Hall voltage generated under the application of short current pulses in static bias magnetic fields. A 2 μm wide Hall bar was lithographically fabricated and artificial domains and domain walls realised using Ga⁺ FIB irradiation. A 5 V pulse amplitude with 100 ns duration was applied to trigger DWM in the current leads of the Hall bar and the time-resolved EHE voltage measured. EHE transients were analysed under some simple assumptions to estimate the velocity of DW motion as a function of bias field. The range of the estimated DW velocity was around 80-120 m/s for current density in the Co layer to be ~8.3×10⁷ A/cm². These results are comparable with other domain wall velocity measurements (100-150 m/s) for a current density of ~0.5-1×10⁷ A/cm² in the literature. We conclude that the time-resolved EHE technique represents a powerful way of analysing domain wall dynamics in MRAM-like structures.
Pt/Co/Pt MLs with structural inversion asymmetry (SIA) (a very thick capping layer and a thin buffer layer), were shown to influence the direction of DWM [186]. In future, time-resolved measurement on SIA multilayer systems with different capping layer materials such as Ir, TaOx and Ta, would be interesting to explore. It is important to investigate the domain wall dynamics in pulsed current-driven time-of-flight experiments, by monitoring the change in Hall resistance due to DWM between two pairs of Hall probe contacts, as function of (a) Ga+ irradiation dose (b) the wire width of the Hall device and (c) the angle between the current direction and the DW normal. These data could be corroborated with parallel DW velocity measurements made using polar MOKE.

Recently CoFeB has been intensely investigated to improve the performance of DWM devices. Research into these materials has shown a change in DW orientation and velocity depending on the underlayer materials such as Hf, Ta, TaN and W and their thicknesses [187], thereby changing its direction of motion. (i.e., DWM against electron flow departing from the conventional DWM along electron flow due to STT) [126, 186, 188-191]. Ferrimagnetic rare earth metals like TbFeCo show a DW velocity in the range of 30-60 m/s for a much reduced current density in the range of 5-25 × 10⁶ A/cm² [192]. It would be interesting to investigate These materials with irradiation induced artificial domains using time-resolved EHE.

When it comes to developing small numbers of prototype devices, FIB irradiation is a very convenient tool. For mass production on an industrial scale, blanket irradiation of masked substrates is preferable [16]. Two possible applications that could evolve from extensions to this study are magnetoresistive sensors and MRAM-like storage elements. Magnetoresistive sensors based on Ga⁺ FIB irradiation of Pt/Co/Pt PMA MLs would be composed of magnetic ‘superlattices’ containing irradiated and non-irradiated regions exhibiting high magnetoresistance due to the number of domain walls created as the irradiated region switches first. The opposite sides of the artificial domain walls formed will contain magnetic regions that switch independently with out-of-plane or in-plane fields, adding valuable functionality. In the case of MRAM storage elements, they could be composed of three adjacent strip domains (I, II, III) with different anisotropies and coercive fields, that are tuned in such a way that $H_c^I > H_c^{III} > H_c^{II}$. Using quasi-static magnetic fields, the magnetisation in domain II and III can be reversed with respect to I and on application of current pulses a DW can be made to switch stably between the I/II and II/III interfaces. The logical state of the device can be read out using the EHE at the voltage leads in domain II. A schematic of this operation is shown in figure 9.1.
Figure 9.1: Schematic of a MRAM like structure containing Ga⁺ irradiated artificial domains (I, II, III) with intermediate DWs

9.1 Ga⁺ FIB irradiation and annealing

Thin films with PMA are promising candidates for high density magnetic storage, sensors and fast memory applications. Ga⁺ ion induced PMA has potential applications in these magnetic thin film systems. The use of FIB irradiation has allowed us to precisely control the local coercivity and anisotropy of domains after multilayer growth and device fabrication. Low dose Ga⁺ FIB irradiation of fabricated Hall cross structures led first to an increase in coercivity followed by a decrease at higher radiation levels. The same behaviour was observed at various Co thicknesses in the range 0.35 nm – 0.85 nm. The surface roughness, measured on the active regions of Hall bar structures, was shown to be correlated with the measured anisotropy in the irradiated Co/Pt multilayers. This correlation was most pronounced in the thickest (0.85 nm) Co films studied. This correspondence was also observed in direct measurements of the anisotropy field, $H_k$. An increase in $H_k$ at low Ga⁺ dose for Co 0.85 nm samples was confirmed as the mechanism for the increase in coercivity after irradiation, something we tentatively attribute to ordered CoPt alloy formation at Co/Pt interfaces.

Annealing of Co/Pt ML films in air at relatively low temperatures ($< 200 \, ^\circ C$) led to very large enhancements in $H_c$ and $H_k$. A gradual increase in coercivity and anisotropy was observed for temperatures less than 160 °C after which a drastic increase in $H_c$ and $H_k$ occurred above 180 °C. The increase in anisotropy at these rather low temperatures could be attributed to the sharpening of the chemical interfaces resulting in increased interfacial strain. Once again these effects were most pronounced in thicker (0.85 nm) Co films. In the immediate future it would be interesting to study the film morphology, composition and interfacial microstructure by high resolution transmission electron microscopy (HR-TEM) and electron energy loss
spectroscopy (EELS). Such investigations would also give us important information about the composition and phase of the CoPt alloys formed after Ga$^+$ FIB irradiation.

Ga$^+$ irradiation induced changes to magnetic properties represent a highly promising approach for the design of high resolution patterned planar structures with PMA. The success of Ga$^+$ FIB irradiation depends on the control of the energy and dose of Ga$^+$ ions. This method offers a novel way to tailor magnetic properties, especially the coercivity and magnetic anisotropy. It would also be interesting to modify the exchange bias between the ferromagnetic/antiferromagnetic multilayers in spintronic devices by inducing subtle changes to the interface by Ga$^+$ FIB irradiation. Using this technique the creation of chemical ordering in ferromagnetic alloys is also possible. The advantage of FIB irradiation is the ability to combine modifications to the magnetic properties with nanoscale patterning of device structures. Ga$^+$ FIB irradiation offers a whole new range of possibilities for increasing or decreasing the magnetic anisotropy of Co/Pt multilayer films after thin film growth and device fabrication. This technique could possibly evolve from the research lab to be used for large scale industrial applications.
Appendix A. Coplanar waveguide (CPW) on a dielectric of finite thickness

A single Hall cross and 5x5 mm² coplanar waveguide structures with impedance $Z_0 = 50 \, \Omega$ matched to the output of a voltage pulse generator were designed for a single Hall probe device. This was modelled by a conventional coplanar waveguide with a finite thickness of dielectric material, as shown in figure A.1.

![Figure A.1: Schematic representation of a coplanar waveguide structure on a dielectric of finite thickness](image)

The total capacitance of the coplanar waveguide (CPW) is the sum of dielectric capacitance $C_1$ and $C_{air}$.

$$C_{CPW} = C_1 + C_{air} \quad (A.1)$$

The capacitance of the dielectric, $C_1$, is given by

$$C_1 = 2 \varepsilon_0 (\varepsilon_r - 1) \frac{K(k_1)}{K(k_1')} \quad (A.2)$$

where $\varepsilon_0$ is permittivity of free space and $\varepsilon_r$ is relative permittivity. $K(k_1)$ and $K(k_1')$ are the complete elliptical integrals [193] given by

$$k_1 = \frac{\sinh(\pi S/4h_1)}{\sinh(\{\pi(S+2W)/4h_1\})} \quad (A.3)$$

$$k_1' = \sqrt{1 - k_1^2} \quad (A.4)$$

The capacitance of the air space, $C_{air}$, is given by

$$C_{air} = 4 \varepsilon_0 \frac{K(k_0)}{K(k_0')} \quad \text{where} \quad k_0 = \frac{S}{S+2W} \quad (A.5a)$$

Substituting the values of $C_1$ and $C_{air}$ into Eq. A.1 gives

$$C_{CPW} = 2 \varepsilon_0 (\varepsilon_r - 1) \frac{K(k_1)}{K(k_1')} + 4 \varepsilon_0 \frac{K(k_0)}{K(k_0')} \quad (A.6)$$
The ratio $C_{CPW}/C_{air}$ will yield the effective dielectric constant $\varepsilon_{eff}$ under the quasi-static approximation, which is given [3] by

$$\varepsilon_{eff} = \frac{C_{CPW}}{C_{air}} = 1 + \frac{(\varepsilon_r - 1) K(k_1) K(k_0')}{2 K(k_1') K(k_0)} .$$  \hspace{1cm} (A.7)

Finally the CPW impedance, $Z_0$ is calculated as

$$Z_0 = \frac{1}{c C_{air} \sqrt{\varepsilon_{eff}}} = \frac{30\pi}{\sqrt{\varepsilon_{eff}}} \frac{K(k_0')}{K(k_0)} ,$$  \hspace{1cm} (A.8)

where $c$ is the velocity of light in free space.
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