MATERIAL DESIGNS AND FABRICATIONS FOR VARIOUS SPINTRONICS APPLICATIONS

by

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A dissertation submitted to the Faculty of the University of Delaware in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Physics

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Spintronics which deploys the spin in addition to or sometimes in place of the charge of the electron has exhibited rich physics and industrial potential in the past two decades. Intensive research has been carried out probing the interaction between spin and magnetic field, electric current, electromagnetic waves and more recently temperature gradient. Work presented in this dissertation focuses on utilizing spin dynamics in contemporary applications.

Generation of pure spin current in a magnetic material subjected to a temperature gradient is called the spin Seebeck effect (SSE). SSE measured for metallic ferromagnetic thin films in commonly used longitudinal configuration is contaminated by the anomalous Nernst effect (ANE), considered to arise from the bulk of the ferromagnet (FM) and the proximity-induced FM boundary layer. Thus, isolation of pure SSE from other parasitic effects in FM metals has been a matter of significant interest and a point of hot debate. We optimized an FM alloy with zero Nernst coefficient to mitigate the ANE contamination of SSE and inserted a thin layer of Cu to separate the heavy metal (HM) from the FM to avoid the influence of the proximity effect. These modifications to the experiment should permit the complete isolation of SSE from ANE. However, further thickness dependence studies and careful analysis of the results revealed, ANE of the isolated FM alloy is twofold, surface and bulk. The existence of a surface contribution to ANE in FM metals was first uncovered in our research. Magnitudes of surface and bulk contributions are
comparable to that of the SSE and can be modified by the neighboring layer. Hence, the surface contribution is an important effect that needs to be considered.

Nanomedicine, the amalgamation of nanotechnology with medical diagnostics has gained a lot of traction in recent years. Especially point of care (POC) assay systems using magnetic nanoparticles (MNPs), which provide improved sensitivity and specificity along with better reproducibility and reliability. Profound efforts have been made to fabricate a bio-detection system incorporating magnetic nanoparticles in a magnetoresistive (MR) sensor. Details of fabrication and optimization of a magnetic tunnel junction (MTJ) based sensor for bio-detection applications are discussed in this work. The goal of this project is to develop an assay for early risk assessment of genetically pre-disposed children with Type-I diabetes. We combine biotechnology with MNPs to enable enrichment of the diabetes autoantibodies in a test sample. Then, detect and measure MNPs with enhanced sensitivity on an MTJ sensor.

Local electrical manipulation of magnetization is a promising candidate for information storage and processing. Electrical control of magnetization can be achieved by leveraging the spin-orbit coupling (SOC), through which an electrical current exerts a spin-orbit torque (SOT) on the magnetization. While there are still debates over the dominant SOC mechanism, one thing is quite clear, the interfacial electronic structures and spin textures play a critical role in determining SOC phenomena. The interfacial electronic structures and spin textures are intimately related to materials and fabrication. They are very difficult to characterize directly. However, their characteristics are manifested in SOT behaviors. Most recent first principle and semi-classical calculations indicate SOTs will depend on the current distributions in HM and FM layers. To experimentally validate this theory, we
compare temperature dependent SOTs in structures with markedly different resistivities. Temperature-dependent Magneto-optical Kerr spectroscopy (MOKE) has been employed to probe the SOTs and the results of CoFeB(t)/Pt and Py(t)/Pt structures were compared. In addition, magnetic structures with perpendicular magnetic anisotropy (PMA) were developed and optimized. Temperature-dependent SOTs for these structures were extracted with DC-MOKE measurement. Moreover, PMA samples were used to perform time-resolved MOKE (TR-MOKE) study of magnetization switching.
Chapter 1
INTRODUCTION

1.1 Spintronics

An empirical observation turned prophecy Moore’s law inspired the information technology revolution. In the famous 1965 article [1] Gordon Moore, who went on to co-found Intel cooperation, estimated the number of electronic components per chip would double every year. In 1975 [2] he made a course correction and revised the estimation for doubling time to more realistic two years. Chip makers, fueled by the consumer demand, scrambled to materialize exponential growth conjectured by Moore and managed to stay on track largely due to a unified effort. Then the reality struck, unprecedented growth in the industry meant, more and more circuitry packed into the same area, and higher processing speeds required shuttling around electrons through cramped spaces at higher speeds. Inevitably chips got hot. Moreover, as feature sizes shrink, quantum mechanical effects would take precedence and transistors would become terribly unreliable. Thus, the need for an infallible alternate for charge-based electronics was strongly felt. Among other contenders spintronics or in other words spin-based electronics which deploys the spin, in addition to or sometimes in place of the charge of the electron seems to be the most promising alternative.

Unlike non-magnetic metals, the resistivity of ferromagnetic (FM) metals deviate significantly from Matthiessen’s rule, and in the process of solving this conundrum, Mott [3] proposed two-current model, where he described conductivity of
a metal as the sum of independent conductivities of up and down spin channels with no spin-flip scattering permitted. In addition, Mott postulated density of states and consequently, the mobility of two channels are different from each other. Thus, the influence of spin on the transport of electrons has been known since the mid-1930s. Significant advances in the field did not happen for a very long time, but all that changed in the year 1988 with the discovery of Giant Magnetoresistance (GMR) by two European physicists, Albert Fert [4] and Peter Grünberg [5], working independently. This remarkable discovery was recognized with the 2007 Nobel prize in Physics. Enlivened by the new turn of events spintronics gradually found its way into many applications, ranging from memory devices to medical. Advantages such as nonvolatility, increased data processing speeds, decreased electric power consumption, and higher integration densities can be achieved by amalgamating spin degree of freedom to conventional charge-based electronic devices or by utilizing the spin degree of freedom alone. However, the success of spintronics hinges on finding effective solutions to major issues abating the progress, including efficient spin polarization, spin current transport across relevant length scales, spin injection, controlling relaxation of injected spins, spin manipulation, and detection.

1.2 Galvanomagnetic Effects

Scottish scientist Professor W. Thompson (Lord Kelvin) discovered, that the applied external magnetic fields influence the electrical conductivity of ferromagnetic metals. His findings were reported to the Royal Society of London in 1857 [6]. Thompson could not measure any appreciable magnetic influence on the electrical conductivity of brass, a nonmagnetic metallic alloy. He attributed this to the inadequacy of sensitive instruments at the time. However, with the aid of more
sensitive equipment, found to be present in all metals, this new phenomenon was termed magnetoresistance (MR). Soon it was realized magnetoresistance belongs to a larger class of phenomena called galvanomagnetic effects, referring to the properties of metals and semiconductors under the simultaneous influence of electric and magnetic fields. Influence of magnetic fields on the resistivity of normal metals can be readily understood with the simple Drude model [7], where the microscopic behavior of electrons in a solid is treated classically. Classical equation of motion with the introduction of magnetic field reads,

\[ m \frac{d\vec{v}}{dt} + \frac{m}{\tau} \vec{v} = -e \left( \vec{E} + \vec{v} \times \vec{B} \right) \]

(1.1)

Where \( \vec{v} \) denotes the drift velocity of electrons decaying with the relaxation time \( \tau \). All other symbols have their usual meaning. For steady-state \( \frac{d\vec{v}}{dt} = 0 \). Thus,

\[ \vec{v} = -\frac{e\tau}{m} \left( \vec{E} + \vec{v} \times \vec{B} \right) \]

(1.2)

Hence current density,

\[ \vec{j} = -en\vec{v} = \frac{e^2n\tau}{m} \left( \vec{E} + \vec{v} \times \vec{B} \right) \]

(1.3)

With the mobility of electrons \( \mu = \frac{e\tau}{m} \), the current density can be rewritten as,

\[ \vec{j} = en\mu\vec{E} - \mu (\vec{j} \times \vec{B}) = \sigma_0\vec{E} - \mu (\vec{j} \times \vec{B}) \]

(1.4)

Here \( \sigma_0 \) is the conductivity of the material with no external magnetic field applied.

\[
\begin{pmatrix}
E_x \\
E_y \\
E_z
\end{pmatrix} = \frac{1}{\sigma_0} \begin{pmatrix}
1 & \mu B_z & 0 \\
-\mu B_z & 1 & 0 \\
0 & 0 & 1
\end{pmatrix} \begin{pmatrix}
j_x \\
j_y \\
j_z
\end{pmatrix}
\]

(1.5)
Assuming B-field to be aligned along the z-direction, components of the electric field shall be written in the above matrix form. Since $AA^{-1} = A^{-1}A = I$, components of the current density take the following matrix form,

$$
\begin{pmatrix}
  j_x \\
  j_y \\
  j_z
\end{pmatrix} = \sigma_0 \begin{pmatrix}
  1 & -\mu B_z & 0 \\
  \mu B_z & 1 & 0 \\
  0 & 0 & 1 + (\mu B_z)^2
\end{pmatrix} \begin{pmatrix}
  E_x \\
  E_y \\
  E_z
\end{pmatrix}
$$

(1.6)

Here diagonal elements correspond to ordinary magnetoresistance (OMR) while non-diagonal elements correspond to the Hall effect (HE). Galvanomagnetic effects of FM metals deviate greatly from the classical Drude model and require quantum mechanical treatment to fully comprehend. Mott’s two current model provides a framework with which galvanomagnetic response of FM metals can be understood.

### 1.2.1 Mott’s Two Current Model

There are two main ingredients to the Mott’s model,

1. As illustrated in Figure 1.1, d-electron states of FM materials are exchanged split. Thus, number of available states around Fermi level where spin up electrons may scatter into differs from that of spin down electrons. Consequently, the mobility of the two spin channels will be markedly different.

2. The probability of spin-flip scattering as compared to scattering events which preserve the spin orientation is negligibly small.
1.2.2 Anomalous Magnetoresistance

Dependence of electrical resistivity on the relative angle between the direction of sense-current and the local magnetization is called the anomalous magnetoresistance or AMR. Illustrated in Figure 1.2 (a), mathematical representation of AMR is as in Equation 1.7, where $\rho_\parallel, \rho_\perp$ are resistivities measured parallel and perpendicular to the magnetization direction and, $\theta$ is the angle between sense-current and the magnetization.

$$\rho(\theta) = \rho_\perp + (\rho_\parallel - \rho_\perp)\cos^2 \theta = \rho_\parallel \cos^2 \theta + \rho_\perp \sin^2 \theta \quad (1.7)$$

Spin-orbit interaction is the underlying mechanism which leads to AMR [8]. Some insight into the mechanism is provided in Figure 1.2 (b).
Figure 1.2 (a) Illustration of AMR setup. (b) Shows two instances where spin-orbit interaction is turned OFF and ON. Wave functions of the spin up and down d-states will intermix while the spin-orbit interaction is turned ON. Hence, allowing spin up s-electrons to scatter into the spin up part of the formerly spin down d-band.

Scattering of s-electrons into d-band primarily dictates the resistance. By invoking Mott’s model, one can easily understand, while spin-orbit coupling (soc) is turned OFF, spin up s-electrons cannot scatter into the d-band. However, with soc turned ON wave functions of the spin up and down, d-states will intermix, thereby allowing spin up s-electrons to scatter into the spin up part of the formerly spin down d-band. Scattering rate of spin up s-electrons into spin down d band depends on the direction of the s-electron momentum \( \vec{p} = \hbar \vec{k} \). Thus, explaining AMR, the dependence of resistance on the relative direction between current and magnetization.

1.2.3 Giant Magnetoresistance

This Nobel worthy discovery turned the field of spintronics on its head. Giant Magnetoresistance or GMR describes the electrical resistance change, in response to an applied external magnetic field of structures with alternating FM and nonmagnetic metallic(NM) layers. Figure 1.3 is a phenomenological depiction of the effect.
Figure 1.3  (a) Shows a non-magnetic layer sandwiched between two FM layers. The magnetization direction of FM$_1$ is pinned, whereas the magnetization direction of FM$_2$ is manipulated via an external magnetic field. (b) Illustrates electrical resistance in the alternating FM, NM metallic layered structure as a function of the applied external magnetic field. Resistance is low when magnetizations of adjacent magnetic layers are parallel and high when antiparallel.

Resistance is low when magnetizations of adjacent magnetic layers are parallel to each other and high when anti-parallel. Equation 1.8 describes the GMR, where $R_p$ and $R_{AP}$ are the respective resistance while magnetizations of adjacent magnetic layers are parallel and anti-parallel to each other.

$$GMR = \frac{R_{AP} - R_p}{R_p}$$  \hspace{1cm} (1.8)

Yet again this effect can be understood invoking the Mott’s two current model. Within the framework of Mott’s model, it is apparent that the electrons with spin parallel to the magnetization undergo weak scattering whereas electrons with spin antiparallel to the magnetization scatter strongly, reflecting the asymmetry of available d-states at the
Fermi energy for spin up and spin down mobile electrons. Figure 1.4 is a schematic representation of the above argument.

Figure 1.4  (a) Spin up electrons undergo weak scattering in both FM layers and spin down electrons undergo strong scattering in both FM layers. Since spin up and down electrons are carried through independent parallel channels resistance is low in this case. (b) Spin up and spin down electrons undergo a strong scattering in one of the FM layers leading to a large resistance.

As suggested by the name GMR is much larger, compared to OMR or AMR. It should be noted, though the magnetoresistive effects discussed thus far describe the response of the electrical resistance to an external magnetic field, underlying mechanisms leading to these effects are sharply contrasting. OMR is due to Lorenz force on electrons, AMR is due to spin-orbit interaction, whereas GMR is due to the magnetic configuration of adjacent magnetic layers. Typically, the biggest obstacle to commercializing a radical idea is the cost, GMR is no exception. A research team led
by IBM fellow Dr. Stuart Parkin were successful in fabricating GMR devices with sputtered multilayers [9] as opposed to molecular beam epitaxy (MBE), help cutting the manufacturing cost significantly. Soon followed the spin valve structures [10].

### 1.2.3.1 GMR Devices

There are four main types of GMR devices in use: multilayer GMR, spin valve GMR, pseudo-GMR and granular GMR. It is adequate to discuss only the multilayer and spin valve GMR devices within the scope of this thesis work. Operation of a GMR device as we clearly understand now depends on the relative orientation of the magnetization of the adjacent magnetic layers and the above device categories differ based on how these magnetic orientations are controlled. In multilayer GMR devices shown in Figure 1.5 (a) adjacent FM layers could be antiferromagnetically or ferromagnetically coupled depending on the thickness of the NM spacer layer which mediates the exchange coupling analogs to Ruderman-Kittel-Kasuya-Yosida (RKKY) [11] interaction. Spacer layer thickness can be chosen such that the neighboring FM layers are antiferromagnetically coupled, i.e. anti-parallel magnetic configuration and achieve parallel configuration via an applied external magnetic field. However, the lack of exchange coupling does not necessarily forbid achieving GMR. As illustrated in Figure 1.5 (b) in spin valve GMR devices, the magnetization of an FM layer is pinned to a neighboring antiferromagnetic layer, while the magnetization of the other FM layer is free to rotate with an applied external magnetic field. Even though GMR devices found applications in a wide variety of fields, the magnetic recording industry happens to be the most impacted, marking the dawn of a new era [12].
Figure 1.5  (a) Multilayer GMR device with alternating FM and NM layers. The thickness of the NM layer is chosen such that the FM layers are antiferromagnetically coupled. An applied external magnetic field helps achieve parallel magnetic configuration. (b) Spin valve GMR devices, where the FM\textsubscript{1} layer is pinned to an adjacent antiferromagnetic layer and the FM\textsubscript{2} layer is free to rotate with an applied external magnetic field.

1.2.4 Tunneling Magnetoresistance

A device with two FM electrodes separated by an ultrathin insulating spacer layer is called a magnetic tunnel junction (MTJ). The resistance of such devices depends on the relative orientation of the magnetization vectors of the FM layers, mimicking GMR devices. Ignoring defects in the insulator, through which electrons might traverse between the FM electrodes, the majority of electrons would tunnel through the insulator. Tunneling probability is low while magnetizations are aligned anti-parallelly resulting in a high resistance, whereas parallel alignment results in a high tunneling probability, thus a low resistance. This magnetoresistive phenomenon is termed tunneling magnetoresistance (TMR).
Prolonged history of MTJs, in a nutshell, is as follows. 1975 julliére, *et al.* [13] first reported the tunneling between ferromagnetic films with maximum TMR of 14% measured at 4.2K. A room temperature MTJ with reasonably large TMR would enhance the scope and capability of MTJs immensely. Those didn’t arrive till 1995, where Moodera, *et al.* [14] and Miyazaki, *et al.* [15] independently reported room temperature MTJs with TMR over 10%. Many researchers followed suit and came up with new FM, insulator combinations achieving astounding improvements in room temperature TMR. Work of Ikeda, *et al.* [16], reporting 604% TMR at 300K in Ta/Co$_{20}$Fe$_{60}$B$_{20}$/MgO/Ta pseudo spin valve MTJ structure puts the remarkable growth in the MTJ fabrication technology in perspective. MTJs have replaced spin valve structures in magnetic recording devices and with TMR values discussed above this should come as no surprise.

Now let us take an in-depth look at the process of tunneling magnetoresistance. In the discussion let us assume there is no direct coupling between the FM layers and the magnetization direction of FM layers can be altered independently. Further, ignore conducting channels between the FM layers through the insulator, thus electron transport purely due to tunneling. With a bias voltage $V$, only the electrons in a small band $eV$ around the Fermi level would participate in the tunneling process. Figure 1.6 illustrates the parallel and antiparallel alignment of the FM layers. Meanwhile, Equation 1.9 expresses the conductance for respective magnetic alignments [17]. Here $G_p, G_{AP}$ are the conductance for parallel and antiparallel magnetic alignments respectively. $N^{A(B)}_\uparrow$ and $N^{A(B)}_\downarrow$ represent the density of states for up and down spins at the Fermi energy in FM$_A$ and FM$_B$ layers.
Figure 1.6 Two FM layers are separated by an insulating layer, ignoring imperfections, electrons should tunnel through the barrier, a process which conserves the electron spin. (a) Magnetizations of FM layers A and B are parallel to each other, thus there are plenty of available state for the electrons to tunnel into, resulting in a low resistance (b) Magnetization of the FM layers are aligned antiparallelly, hence states available for electrons to tunnel into are scarce, resulting in a high resistance.

\[
\begin{align*}
G_p &= N_\uparrow^A N_\uparrow^B + N_\downarrow^A N_\downarrow^B \\
G_{AP} &= N_\uparrow^AN_\downarrow^B + N_\downarrow^AN_\uparrow^B
\end{align*}
\] (1.9)

Therefore, TMR can be written in terms of \(G_p, G_{AP}\) as shown in Equation 1.10,

\[
TMR = \frac{R_{AP} - R_p}{R_p} = \frac{1/G_{AP} - 1/G_p}{1/G_p} = \frac{G_p - G_{AP}}{G_{AP}}
\] (1.10)

By substituting Equation 1.9 in 1.10 we obtain,

\[
TMR = \frac{N_\uparrow^AN_\uparrow^B + N_\downarrow^AN_\downarrow^B - N_\uparrow^AN_\downarrow^B - N_\downarrow^AN_\uparrow^B}{N_\uparrow^AN_\downarrow^B + N_\downarrow^AN_\uparrow^B}
\] (1.11)

Defining degree of spin polarization \(P\) at Fermi level as: \(P = \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}\), TMR can be rewritten as:

\[
TMR = \frac{2PP_2}{1-P_1P_2}
\] (1.12)
1.3 Hall Effects

Under this section let us discuss Hall effect (HE) and anomalous Hall effect (AHE) which should have been discussed under the framework of galvanomagnetic effects but moved under the Hall effects theme just for the sake of clarity. Further, discuss the spin Hall effect (SHE) and inverse spin Hall effect (ISHE).

1.3.1 Hall Effect

Discovered by E.H. Hall in 1879 [18] Hall effect describes the electric field generated by a current carrying conductor under a magnetic field, orthogonal to both the current and the magnetic field. Relevant setup is shown in Figure 1.7, with the current flowing in +x-direction and external magnetic field in the +z-direction, electrons experience a Lorentz force in the +y-direction, leading to a charge accumulation. Thus, an electric field orthogonal to the current flow and the magnetic field. At equilibrium, we have \( e\vec{V} = e\vec{v} \times \vec{B} \), with drift velocity \( v = -I/(L \times d)en \), all the symbols here have their usual meaning and all dimensions are as in Figure 1.7.

Combining these expressions Hall voltage can be written as,

\[
\frac{V_H}{L} = -\frac{IB}{Lden} \Rightarrow V_H = -\frac{1}{(ne)} \cdot \frac{IB}{d} = R_0 \frac{IB}{d}
\]  

(1.13)

\(-\frac{1}{ne}\) is defined as the Hall coefficient \( R_0 \), which is negative for the free electrons and positive for the holes. Since \( \vec{j} = \sigma \vec{E} \) Hall resistivity \( \rho_{xy} \) can be defined as,

\[
\rho_{xy} = \frac{E_y}{j_x} = BR_0
\]

(1.14)

Hall resistivity is a linear function of applied external magnetic field.
Figure 1.7  Illustrates the Hall measurement setup with an electric current flowing in the +x-direction (electrons flow in -x-direction) and external magnetic field applied in the +z directions electrons deflect in the +y-direction due to Lorentz force and lead to a charge accumulation. This charge accumulation generates an electric field along the y-direction and the voltage measured here is called Hall voltage.

1.3.2 Anomalous Hall Effect

In 1881 Hall reported [19] magnitude of the transverse voltage measured with the same setup shown in Figure 1.7 for FM conductors is much larger compared to the non-magnetic conductors. Termed anomalous Hall effect (AHE), its origin was under scrutiny for over a century. Hall resistivity measured for magnetic metals varies non-linearly with the external magnetic field in contrast to non-magnetic metals, manifesting a sharp increase at the lower fields and, plateauing at the higher fields. With saturation values observed at higher fields being proportional to the magnetization of the material, an empirical formula shown in Equation 1.15 was established, relating the Hall resistivity \( \rho_{xy} \), external magnetic field \( B_z \) and the magnetization \( M_z \) [20]. Here \( R_0 \) is the Hall coefficient discussed earlier and \( R_s \), the
anomalous Hall coefficient. Anomalous Hall coefficient found to be substantially larger than Hall coefficient and strongly temperature dependent.

\[ \rho_{xy} = R_0 B_z + \mu_0 R_z M_z \]  

(1.15)

After many iterations and refinements, it is widely accepted AHE is a result of both intrinsic and extrinsic mechanisms, illustrated in Figure 1.8.

Karplus and Luttinger [21] proposed electrons in a solid acquire an additional component to their group velocity under an electric field, transverse to the field. In the case of an FM conductor, this anomalous velocity over all occupied states sums up to
a non-zero value, thus contributing to the Hall resistivity. Depending exclusively on
the band structure and hence independent of scattering. This mechanism was dubbed
intrinsic deflection. Skew scattering from impurities due to spin-orbit coupling [22]
and side jump mechanism [23] were suggested to account for the scattering from
impurities, ignored by the intrinsic mechanism. Further discussion involving the
confusion revolving around the true origin of AHE is beyond the scope of this thesis
work and a detailed account of the progression in the field can be found in the review
article by Nagaosa, et al. [24].

1.3.3 Spin Hall Effect

When a charge current is driven through a conductor, electrons undergo spin-
dependent deflection transverse to the flow, due to intrinsic and/or extrinsic
mechanisms discussed above. With spin imbalance in FM conductors, this leads to an
excess charge accumulation transverse to the current flow, thus a measurable voltage,
which we define as the anomalous Hall voltage, given in Equation. 1.16. Here $I$ is the
charge current and $R_{AHE}$ the anomalous Hall resistance.

$$V_{AHE} = IR_{AHE}$$  \hspace{1cm} (1.16)

With the same setup, lack of spin imbalance in NM conductors means, spin-dependent
deflection does not give rise to a charge accumulation rather, leads to a spin
accumulation. A phenomenon come to known as spin Hall effect (SHE). Illustrated in
Figure 1.9, with charge current in the -x-direction, i.e. net electron flow in the +x-
direction, spin up and down electrons deflect in the transverse direction which can be
viewed as a pure spin current flowing in the -y-direction with no net flow of charges.
Figure 1.9  External electric field drives a charge current in the -x-direction, thus a net flow of electrons in the +x-direction. Spin up and down electrons get deflected along a direction transverse to charge flow due to intrinsic and/or extrinsic mechanisms. Since the number of up spins equal to the number of down spins in a non-magnetic conductor, this does not lead to a charge accumulation but a spin current.

1.3.4 Inverse Spin Hall Effect

A net flow of charges can be easily measured by simply hooking up a multimeter. How about a net flow of spins, with no net flow of charges, in other words how to measure a pure spin current. Regardless of whether it is due to the intrinsic or extrinsic mechanism, the root cause for SHE is the spin-orbit coupling. With the aid of the same relativistic effect, we should be able to realize the inverse process, called inverse spin Hall effect (ISHE) [25]. Suppose, a pure spin current is injected into a material. Spin up and down electrons flow in the opposite directions and spin-orbit coupling would push both the spins in the same direction thus generating a charge current transverse to the spin current, enabling us to quantify the spin current. For converting spin current into charge current, materials with strong spin-orbit coupling
such as heavy metals (Pt, Ta, Nb) are preferred. Figure 1.10 is a schematic representation of the ISHE. The relation among charge current density \( j_c \), spin current density \( j_s \), and spin polarization \( \sigma \) is given by Equation 1.17, where \( D_{ISHE} \) is a coefficient representing ISHE in the material.

\[
\vec{j}_c = D_{ISHE} \vec{j}_s \times \vec{\sigma}
\]  

(1.17)

Figure 1.10 A pure spin current flows in the -x-direction. Electrons with spin polarization parallel and anti-parallel to +z-direction get deflected in the +y-direction due to spin-orbit coupling. Thus, generating a charge current transverse to the spin current.

1.4 Spin Caloritronics

The discussion thus far revolved around the interaction of charge and spin. We could conclude with absolute certainty that the amalgamation of spin has enriched the charge-based electronics. Would it be possible to enhance the field of thermoelectrics or in other words interaction among charge and heat by incorporating spintronics, all recent trends point to a grand success of this scheme, dubbed spin caloritronics. Before
we get into the discussion of spin caloritronics, let us review the fundamentals of thermoelectrics.

### 1.4.1 Wiedemann-Franz Law

Charge current \((\vec{J}_c)\) generated in response to a potential gradient \((\nabla V)\) in a conductor is given by Equation 1.18, where \(\sigma\) is the electrical conductivity.

\[
(\vec{J}_c)_{V_T=0} = -\sigma A \hat{\nabla} V
\]  

(1.18)

Meanwhile, heat current \((\vec{J}_q)\) generated by a temperature gradient \((\nabla T)\) in a conductor is given by Equation 1.19, where \(\kappa\) is the thermal conductivity.

\[
(\vec{J}_q)_{T_e=0} = -\kappa A \hat{\nabla} T
\]  

(1.19)

These are related through Wiedemann-Franz law, given in Equation 1.20, where \(L_0 = \frac{\pi^2 k_B^2}{3e^2}\) is the Lorenz number, a fundamental constant.

\[
\lim_{T \to 0} \frac{\kappa}{\sigma} = L_0 T
\]  

(1.20)

Wiedemann-Franz law says the ratio of the electrical component of the heat conductivity to the electrical conductivity is constant for different materials at a given temperature. This implies charge transport and heat transport are correlated, which is the basis of thermoelectrics.

### 1.4.2 Seebeck Effect

When a conductor is under a temperature gradient a voltage will build up, this phenomenon is called the Seebeck effect. The magnitude of the voltage is governed by the material property called the Seebeck constant \((S)\). The equation relating relevant parameters is as in Equation 1.21.
A thermocouple is a direct application of the Seebeck effect where materials with two different Seebeck coefficients \((S_A \neq S_B)\) are connected and kept at a temperature gradient as shown in Figure 1.11 (a), leading to a measurable voltage difference \(\Delta V = (S_A - S_B)\Delta T\).

1.4.3 Peltier Effect

The reverse of the Seebeck effect is defined as the Peltier effect. When a charge current is driven through a conductor, it leads to a heat current. Charge current, and the heat current dragged with it are related via the Peltier coefficient \((\Pi)\) as described by Equation 1.22.
If a current is driven through a loop with two different materials, thus with different Peltier coefficients \((\Pi_A \neq \Pi_B)\), as in Figure 1.11 (b), one terminal gets hot and the other gets cold, hence giving rise to a temperature gradient, where relevant parameters are related through, \(J_Q = (\Pi_A - \Pi_B)J_C\).

1.4.4 Thermodynamics of Thermoelectrics

If we have two reservoirs at a temperature gradient \((\nabla T)\) and potential gradient \((\nabla V)\), a heat current \((J_Q)\), as well as a charge current \((J_C)\) will be generated. \(J_C\) can be expressed combining ohm’s law with Seebeck effect and \(J_Q\) can be expressed combining Peltier effect with the Fourier’s law. Thermoelectric equations are as in Equation 1.23 which takes the matrix form shown in Equation 1.24 [26].

\[
\begin{align*}
\bar{J}_C &= -\sigma A \left( \bar{V}V + S \bar{V}T \right) \\
\bar{J}_Q &= \Pi \bar{J}_C - \kappa A \bar{V}T \\
\begin{pmatrix}
-\bar{V}V \\
\bar{J}_Q
\end{pmatrix} &= \begin{pmatrix}
1/\sigma A & S \\
\Pi & -\kappa A
\end{pmatrix} \begin{pmatrix}
\bar{J}_C \\
\bar{V}T
\end{pmatrix}
\end{align*}
\]

(1.23)

(1.24)

Currents and, forces driving the currents, can be separated by a transformation and presented in Equation 1.25.

\[
\begin{pmatrix}
\bar{J}_C \\
\bar{J}_Q
\end{pmatrix} = \sigma A \begin{pmatrix}
1 & ST \\
\Pi & (\kappa/\sigma + \Pi S)T
\end{pmatrix} \begin{pmatrix}
-\bar{V}V \\
-\bar{V}T/T
\end{pmatrix}
\]

(1.25)

With Onsager reciprocity \(L_{ij} = L_{ji}\), one can recover the Onsager-Kelvin relation \(\Pi = ST\), which implies that the physics describing the Seebeck effect and the Peltier effect is the same.
1.4.5 Role of Spin in Thermoelectrics

As we discussed previously incorporating spin degree of freedom with charge-based electronics has led to advancements in many fields. Now let us extend the discussion to, subsuming spin degree of freedom in thermoelectrics, a field come to known as spin caloritronics. With spin degree of freedom in play, an external electric field would drive distinct currents for spin up \( J_\uparrow \) and spin down \( J_\downarrow \) electrons.

Sum of spin up and spin down currents constitute the charge current, whereas the difference between spin up and spin down currents constitute the spin current, as shown in the Equation 1.26.

\[
J_{C(s)} = J_\uparrow \pm J_\downarrow
\]

Considering the interface between an FM and NM, an electric field or a temperature gradient would lead to a charge accumulation as dictated by the spin diffusion equation. We will take a deeper look into the spin diffusion formalism in Chapter 3.

Now we can rewrite the thermoelectric equations, similar to Equation 1.23, taking spin-dependent transport into consideration.

\[
J_\uparrow = -G_i \Delta V_i - G_i S_i \Delta T
\]

\[
J_\downarrow = -G_i \Delta V_i - G_i S_i \Delta T
\]

\[
\therefore J_{C(s)} = J_\uparrow \pm J_\downarrow
\]

\[
J_{C(s)} = -(G_i + G_j) \left( \frac{G_i \Delta V_i + G_i \Delta V_j + G_i S_i \Delta T + G_i S_i \Delta T}{G_i + G_j} \right)
\]

\[
J_{C(s)} = -(G_i + G_j) \left( \frac{G_i \Delta V_i + G_i \Delta V_j + G_i S_i \Delta T}{G_i + G_j} \right) - (G_j + G_i) \left( \frac{G_j S_j + G_j S_j}{G_j + G_i} \right) \Delta T
\]

\[
J_{C(s)} = \frac{G_i + G_j}{2} \left( \frac{2G_i \Delta V_i + 2G_i \Delta V_j + G_i S_i \Delta T + G_i S_i \Delta T - G_i \Delta V_i + G_i \Delta V_j - G_i \Delta V_j + G_i \Delta V_i - G_i \Delta V_j}{G_i + G_i} \right) - (G_j + G_i) \left( \frac{G_j S_j + G_j S_j}{G_j + G_i} \right) \Delta T
\]

\[
J_{C(s)} = \frac{G_i + G_j}{2} \left( -\frac{\Delta V_j}{2} - \frac{G_i - G_j}{G_i + G_j} \left( \frac{\Delta V_i - \Delta V_j}{2} \right) \right) - (G_j + G_i) \left( \frac{G_j S_j + G_j S_j}{G_j + G_i} \right) T \frac{\Delta T}{T}
\]

\[
\Rightarrow J_e = G(-\Delta V) + G P(\Delta V/2) + G S \left( -\frac{\Delta T}{T} \right)
\]
Similarly,

\[
J_j = J_\pi - J_z
\]

\[
J_\pi = -(G_i + G_j + G_k) \left( \frac{G_i \Delta V_i - G_j \Delta V_j + G_j S \Delta T - G_S \Delta T}{G_i + G_j + G_k} \right)
\]

\[
J_z = -(G_i + G_j + G_k) \left( \frac{G_i \Delta V_i - G_j \Delta V_j + G_j S \Delta T - G_S \Delta T}{G_i + G_j + G_k} \right)
\]

\[
J_S = -(G_i + G_j + G_k) \left( \frac{G_i \Delta V_i - G_j \Delta V_j + G_j S \Delta T - G_S \Delta T}{G_i + G_j + G_k} \right)
\]

\[
J_T = -(G_i + G_j + G_k) \left( \frac{2G_i \Delta V_i - 2G_j \Delta V_j + G_i \Delta V_i - G_j \Delta V_j + G_j \Delta V_j - G_i \Delta V_i + G_i \Delta V_i - G_j \Delta V_j + G_j \Delta V_j - G_i \Delta V_i}{2(G_i + G_j + G_k)} \right) + \left( \frac{G_i \Delta V_i - G_j \Delta V_j}{G_i + G_j + G_k} \right) \Delta T
\]

\[
J_T = -(G_i + G_j + G_k) \left( \frac{\Delta V_i + \Delta V_j}{2} \right) + \left( \frac{\Delta V_i - \Delta V_j}{2} \right) + \left( \frac{G_i \Delta V_i - G_j \Delta V_j}{G_i + G_j + G_k} \right) \Delta T
\]

\[
J_z = G \left[ p(-\Delta V) + \left( \frac{-\Delta V}{2} \right) + p' \Delta T \left( -\frac{\Delta T}{T} \right) \right]
\]

Here \( G = G_i - G_j, S = \frac{G_i S + G_j S}{G_i + G_j}, P = \frac{G_i - G_j}{G_i + G_j}, \Delta V = \frac{\Delta V_i + \Delta V_j}{2}, \Delta V = \Delta V_i - \Delta V_j, P' = \frac{G_i - G_j}{G_i + G_j} \).

\( G, S, P, \) and \( P' \) are total electrical conductance, total interface Seebeck coefficient, spin polarization and polarization of \( GS \) respectively. Further, heat current \( J_v \) can be written down as, \( J_v = -K \Delta T + TS J_z + TS J_z \), recall \( \Pi = ST \).
\[
J_o = -K\Delta T + T \left[ S, \left( -G, \Delta V_r - G, S, \Delta T \right) + S, \left( -G, \Delta V_s - G, S, \Delta T \right) \right]
\]

\[
J_o = -K\Delta T - (G, + G) \frac{T \left[ S, \left( G, \Delta V_r + G, S, \Delta T \right) + S, \left( G, \Delta V_s + G, S, \Delta T \right) \right]}{(G, + G)}
\]

\[
J_o = -K\Delta T - (G, + G) \frac{T \left[ S, \left( G, \Delta V_r + G, S, \Delta T \right) + S, \left( G, \Delta V_s + G, S, \Delta T \right) \right]}{(G, + G)}
\]

\[
J_o = -K\Delta T - (G, + G) \frac{T \left[ S, \left( G, \Delta V_r + G, S, \Delta T \right) + S, \left( G, \Delta V_s + G, S, \Delta T \right) \right]}{(G, + G)}
\]

\[
J_o = -K\Delta T - (G, + G) \frac{T \left[ S, \left( G, \Delta V_r + G, S, \Delta T \right) + S, \left( G, \Delta V_s + G, S, \Delta T \right) \right]}{(G, + G)}
\]

Thus, thermoelectric equations can be expressed in the following matrix form separating the currents and the forces such that the Onsager reciprocity is satisfied.

\[
\begin{bmatrix}
J_C \\
J_S \\
J_Q
\end{bmatrix} = G \begin{bmatrix}
1 & P & ST \\
P & 1 & P' ST \\
ST & P' ST & KT/G
\end{bmatrix} \begin{bmatrix}
-\Delta V \\
-\Delta V_s / 2 \\
-\Delta T / T
\end{bmatrix}
\]

(1.27)

A spin current can propagate as independent spins in metallic magnetic materials or as a collective excitation of spins (magnon) in magnetic insulators [27]. Thus, spin caloritronic effects should be classified based on how the spin current propagates in the medium. Spin-dependent Seebeck effect [28] where a spin current is injected across an FM/NM interface via a thermal gradient and Spin-dependent Peltier effect [29] where a heat current is induced by injecting a spin current into an FM are
prime examples of the spin caloritronic effects under independent spin propagation. Meanwhile spin Seebeck effect observed in a magnetic insulator is a classic example for spin caloritronic effects due to collective excitation of spins. Let us differ the detailed discussion of spin Seebeck effect to a later chapter. A comprehensive review of spin caloritronics can be found in the article by Bauer, et al. [30].

1.5 Dissertation Layout

Chapter 2 of this dissertation will provide an insight into the sample fabrication, characterization and measurement methods, and tools utilized in the work presented here. Chapter 3 is dedicated to discussing the efforts to isolate pure spin Seebeck effect from thermal measurements performed on FM metals in the longitudinal configuration. Chapter 4 details the deposition, microfabrication, testing and measurement processes involved in developing a magnetic tunnel junctions-based sensor for bio-detection applications. Though the primary goal of this sensor is to detect early childhood Type-II diabetes, its versatility would allow other types of detections with minor alterations. Chapter 5 is allotted to examine the influence of the current distributions in the FM/ NM bilayers on current induced SOTs. Thus, experimentally affirm or debunk recent theoretical studies. Chapter 6 outlines the continuation of work comparable to Chapter 5, but FM materials with perpendicular magnetic anisotropy are scrutinized to conclude the dissertation.
Chapter 2

THIN FILM DEPOSITION, ENGINEERING, AND CHARACTERIZATION AT THE MICRO AND NANO SCALES

2.1 Sputter Deposition

Thin film deposition techniques can be classified into two main categories Physical Vapor Deposition (PVD) and Chemical Vapor Deposition (CVD). Each technique has its pros and cons and the choice of method really depends on the material you are interested in depositing [31,32]. Sputtering, a PVD technique where surface atoms of a target material liberated by bombarding accelerated ions are collected at a substrate to form a film of the target material [33] is the method exclusively employed to fabricate samples used in this thesis work. Depicted in Figure 2.1 (a), sputtering allows you to deposit a wide variety of materials including metals, alloys, and insulators while closely replicating the target composition in the deposited films. Moreover, provides a better film quality and step coverage compared to evaporation. Which means it is a versatile, reliable and sophisticated technique. Thus, highly desired.

2.1.1 DC Diode Sputtering

As shown in Figure 2.1 (b) in DC sputtering, substrate and the target are placed on the anode and the cathode of two parallel electrodes respectively and the chamber is filled with an inert gas like argon since we want the process to be purely physical not chemical. Recall sputtering is a PVD technique. A high DC voltage is applied to
the diode, which will accelerate the free electrons in the chamber. Ar atoms will ionize giving up secondary electrons due to collisions with these energetic electrons \( (Ar + e^- \rightarrow Ar^+ + 2e^-) \). Secondary electrons will continue the ionization process resulting in a gas breakdown, thus a glow discharge (plasma).

![Diagram](image)

**Figure 2.1** (a) Illustration of the sputtering process where accelerated ions collide with the target surface. Depending on the energy carried by the ion, it either dislodges a surface atom from the target material or gets embedded in the substrate, called ion implantation. (b) DC sputtering setup. Here G-Gas, A-Anode, S-Substrate, T-Target, P-Pump, C-Chamber, HV-High Voltage. (c) RF sputtering setup, here RF refers to Radio Frequency power.

The voltage required \( (V_{bd}) \) for gas breakdown is given by Paschen’s law as in Equation 2.1 where \( P \) is the chamber pressure, \( L \) the electrode spacing and \( b \) a constant. From this relation, one can immediately see, self-sustaining the plasma depends on two key parameters: chamber pressure and the electrode separation.

\[
V_{bd} = \frac{PL}{\log(PL) + b}
\]  

(2.1)

If the electrode separation is smaller than the dark space, plasma cannot be sustained. Similarly, if the chamber pressure is too low, the mean free path \( \lambda \propto \frac{1}{\sqrt{P}} \) will be
large, hence secondary electrons will reach the anode before colliding with Ar atoms making it difficult to sustain the glow discharge. However, that does not mean very large pressure is favorable for sputter deposition. If the pressure is too high, mean free path will be very small, resulting in many collisions, leaving electrons with not enough time to gather sufficient energy to ionize Ar atoms, resulting in plasma being extinguished.

2.1.2 Magnetron Sputtering

![Magnetron Sputtering Setup](image)

Figure 2.2 Illustrates the magnetron sputtering setup, where electrons are trapped by applying a magnetic field at the cathode thus increasing the probability of ionizing collisions with Ar atoms.

\[
S = \frac{\text{Number of sputtered atoms}}{\text{Number of incident ions}}
\]

Sputter yield \(S\) is another crucial parameter in sputtering and largely a function of ion energy. If the ion energy is too low, they are unable to knock the target atoms off the surface resulting in zero yield. If the ion energy is too high, they get embedded in the target, called ion implantation, still resulting in zero sputter yield. Deposition rate from a target depends on the product of the sputter yield and the flux of incident ions. So how to increase the flux of incident
ions? One obvious way is to ramp up the pressure. As we discussed earlier, this is not ideal. An alternate way to achieve this is called magnetron sputtering [34], where a magnetic field is applied at the cathode (target) such that $\vec{E} \times \vec{B}$ drift currents close in on themselves, thus trapping primary and secondary electrons closer to the target increasing the probability of ionizing collisions resulting in much-improved deposition rates. Magnetron sputter system is illustrated in Figure 2.2.

### 2.1.3 RF Sputtering

As we discussed earlier sputtering technique is not limited to metallic thin film fabrication. It may be used to sputter deposit from insulating targets as well. But not with the aid of a DC source. Problem with using a DC source can be understood easily. When an insulating target is bombarded with Ar ions they stick to the target surface. Negative charges from the back of the target (cathode) cannot migrate through the insulating material to neutralize this positive charge buildup, which will effectively block further Ar ions from approaching the target, resulting in extinguishing the plasma. The solution is to use an AC source at a high frequency, typically 13.56MHz. Thus, this method is known as Radio Frequency or RF sputtering [35]. When the target is at negative polarity, Ar ions will be attracted and the target material will be sputtered off. In the meantime, electrons will bombard the positive substrate. During the subsequent half of the cycle, the target will be at positive polarity and substrate will be negative. Does that mean Ar ions will now bombard the substrate? If we were to use a lower frequency ($<100kHz$) answer is YES, which is not desirable. However, if we were to use a high-frequency ($>1MHz$) substrate will remain negative for only a very short period of time, hence providing heavy ions with not sufficient time to reach the substrate.
In order to deposit dielectric materials, we do not necessarily require RF sputtering. There is a cheaper alternative called reactive sputtering [36], where metallic targets are sputtered in the presence of a gas mixture consisting of the carrier gas argon and a reactive gas like nitrogen or oxygen. For example, if one wishes to deposit a layer of aluminum oxide, this can be achieved by DC sputtering an aluminum target in the presence of oxygen. Reactive gas will react with the target as well as the substrate to form a compound, though this is the basis of the technique, it turns out to be a major drawback as well. As we can see from Figure 2.3 (a) [37] when we start sputtering the metal in the presence of a reactive gas, we are at a very high sputtering rate. Sputtering at metallic rates is called the metal mode. However, as more reactive gas is introduced into the chamber target metal starts grabbing the reactive gas molecules (gettering) thus forming a compound, which in turn will cover the surface.
This is called the target poisoning. Now we are sputtering in the compound mode and the sputtering rates drop drastically. As seen in Figure 2.3 (a) if we cut down on the gas flow significantly we can clean the target surface and get back to the metal mode. Desirable stoichiometric compounds are obtained often while operating at the knee of the hysteresis. Thus, reactive sputtering requires very careful control of the gas flow rates.

### 2.1.5 Plasma Oxidation

With the emergence of spin-based memory devices fabricating high-quality magnetic tunnel junctions (MTJs) thus, high-quality oxide layers have become a key component of thin film technology. There are several ways to accomplish this. Sputtering an oxide target with an RF power source is the most obvious method. As discussed earlier sputtering the relevant metallic target in the presence of oxygen, the reactive gas is another. But it appears oxidizing a metallic film by exposing it to an oxygen plasma in-situ yields the best results. For example, to fabricate an aluminum oxide layer one can DC sputter an aluminum layer and wait for a little while to allow pump the argon out. Then introduce oxygen into the chamber and ionize it with the aid of an oxidation gun, one similar to shown in Figure 2.3 (b) and place the metal film on top of the oxygen plasma for a predetermined duration.

### 2.1.6 Sputtering System Used in This Thesis Work

Figure 2.4 is a schematic illustration of the magnetron sputtering system used in this thesis work, which consists of seven sputtering guns, one ion beam etching gun, and one oxidation gun. There are ten sample holders in the system, allowing fabrication of as many samples in each run. Sample holders are moved to sputtering
guns and rocked about the gun (to keep the film uniform) with the aid of a stepper motor. High vacuum is maintained with a CTI Cryo-torr 10 vacuum pump, capable of pumping air and water vapor at pumping speeds of $3000 \ell/s$, $9000 \ell/s$ respectively.

![Diagram](image.png)

Figure 2.4  Schematic illustration of the sputtering system used in this thesis work.

### 2.1.7 Wedge Technique

Structural, optical, magnetic and transport properties of thin films are markedly different from their bulk counterparts and often show a prominent thickness dependence. Thus, performing thickness dependence studies is a key component in understanding the behavior of thin films. However, fabricating different films for each thickness is a tedious and time-consuming endeavor. A wedge sample is a pragmatic
alternative, and Figure 2.5 shows the deposition of such a film. Here the sample holder is placed approximately 6° off the axis of the plasma. Therefore, the edge of the sample closer to the plasma grows the full intended thickness. Meanwhile, far edge grows thinner. Given substrate is long enough one can obtain the full thickness range.

Figure 2.5  Off angle sputtering for wedge sample deposition.

2.2 Microfabrication

Microfabrication techniques are at the heart of the chip-making process, thus anyone who uses as much as a cellphone is dependent on the success and the progress of the field. Typical microfabrication techniques can be classified as in Figure 2.6.
We have already discussed thin film deposition in some detail (additive processes). Deposited films need to be engineered to define desired structures. Photolithography is the most commonly used technique for structure fabrication and extensively used in this thesis work.

2.2.1 Photolithography

The basis of photolithography [38] lies in polymer chemistry, where UV light is used to define a polymer network on a wafer corresponding to a pattern on a photomask. A photomask blocks UV light where there is a pattern (chrome) and allows UV light to come in contact with the polymer (photoresist) elsewhere. Depending on the photoresist (PR) used, parts exposed to the UV light either degrade or hardens and called positive and negative photolithography respectively. Weaker
parts of PR dissolves upon developing. Steps involved in these processes are illustrated in Figure 2.7 and 2.8. Recipes used in each process are discussed below.

![Positive Photolithography](image)

**Figure 2.7  Positive photolithography steps**

Following steps were followed in positive lithography:

1. Spin coat PR (AZ1512) at 4000 rpm for 60 s. By changing the frequency and/or the time one can increase or decrease the thickness of the PR layer.

2. Bake at 110°C for 60 s.

3. Align the spin-coated sample with the mask (Mask Aligner: OAI Hybralign 200) and expose to UV light for 6 s. Parts of the photoresist exposed to the UV light or in other words not blocked by the mask degrades under photo-exposure.

4. Rinse in the developer MF - 319 until degraded parts of the PR are completely removed, roughly 20 - 30 s.

5. A network of PR corresponding to the mask will remain on top of the sample.
Following steps were followed in positive lithography:

1. Spin coat PR AZ5214 at 4000rpm for 60s.
2. Bake at 90°C for 60s (soft bake).
3. Align the spin-coated sample with the mask and expose to UV light for 0.7s.
4. Bake at 110°C for 60s (hard bake).
5. Expose to UV light for 45s (NO mask).
6. Rinse in the developer AZ300 until a clear contrasting pattern with sharp edges emerges (check under the microscope after developing for short periods of time).
7. Parts of the photoresist exposed to the UV light or in other words not blocked by the mask hardens under photo-exposure rest of the photoresist will be removed by the developer.
2.2.2 Ion Beam Etching

Etching a subtractive process refers to the removal of material from a wafer surface. There are two main forms of etching wet and dry. In wet etching, the wafer is immersed in an etchant and a chemical reaction between the etchant and the wafer help remove the unwanted material. However, there are several drawbacks to wet etching. Hazardous chemicals used pose a safety concern, etch conditions may change during the process thus making it difficult to control etching time, it is an isotropic process which may lead to an undesired undercut. We have employed dry etching, specifically ion milling in the majority of sample fabrication in this thesis work. Figure 2.9 is a graphical representation of an ion beam etching system. Here, an inert gas like argon is pumped into the chamber and ionized by electrons emitted at the hot
cathode. These ions are accelerated toward the wafer with the aid of an accelerator grid. Ions bombarding the wafer surface will etch away regions not covered with the photoresist. Thus, it is apparent that the ion milling is highly directional but not selective technique. If one wishes to etch only up to a certain layer, this can be achieved with the aid of a reference, deposited on a glass substrate. The reference with a compatible paper pasted on the back should be placed next to the sample and etching process should be continued until paper on the back of the reference becomes clearly visible.

2.2.3 Lift-Off Process

Figure 2.10 Lift-off process.
Some materials may have very low etching rates making it nearly impossible to etch thicker layers. Further, etching may alter the properties of the underlayer or simply one may not have access to an etching tool. Lift-off, an additive technique, is a cheaper alternative to etching. General steps involved in the lift-off process are illustrated in Figure 2.10 and are listed below.

1. Spin coat appropriate photoresist.
2. Form inverse of the desired pattern.
3. Metallization: target material will be deposited everywhere.
4. Use a solvent like Acetone or 1-Methyl-2-pyrrolidone remove photoresist.

During metallization step of the lift-off process sidewalls of the PR get coated with the target material. In some cases, this could lead to a continues film, as shown in Figure 2.11 (a). Therefore, the solvent could strip off the metal film along with the PR. There are several methods to address this concern. We employed the bilayer lift-off process to avoid this issue. Steps involved in the bilayer lift-off process are illustrated in Figure 2.11 (b) and are listed below.

1. Spin coat \textit{LOR5A} (lift-off resist) at 2000\textit{rpm} for 60s.
2. Bake at 170\textdegree C for 5min.
3. Spin coat \textit{nlof 2020} (imaging resist) at 6000\textit{rpm} for 60s.
4. Bake at 110\textdegree C for 60s.
5. Align the spin-coated sample with the mask and expose to UV light for 20s.
6. Bake at 110\textdegree C for 60s.
7. Rinse in the developer AZ300 for 90s. During the rinse, nlof 2020 gets developed first. Thereafter, LOR5A gets developed both vertically and horizontally, leading to an undercut. One can control the degree of undercut by developing for the appropriate amount of time.

![Figure 2.11](image)

(a) Illustrates deposited film grown continues across the step, thus the possibility of being removed by the solvent along with the photoresist. (b) Bilayer photoresist process, we can control the degree of undercut by developing for the appropriate amount of time.

### 2.3 Sample Characterization

#### 2.3.1 X-Ray Diffraction

X-ray diffraction (XRD) is a tool extensively employed to extract structural information such as composition, defects, and thickness of thin films. Regularly spaced atoms in a crystal lattice cause the incident beam of collimated X-rays to
diffract, resulting in the familiar XRD pattern. Bragg’s law relates the wavelength ($\lambda$) of the incident X-rays to the spacing of the atomic planes ($d$) through Equation 2.2, where $\theta$ is the incident angle and $n$ an integer known as the order of reflection [39–41]. Diffraction of X-rays by a crystal is illustrated in Figure 2.12 (a).

$$n\lambda = 2d \sin(\theta)$$  \hspace{1cm} (2.2)

Figure 2.12  (a) Illustration of X-ray diffraction from atomic planes. (b) Typical XRD setup.

XRD data usually measures scattered X-ray intensity ($I$) as a function of incident angle ($\omega$) and/or diffracted angle ($2\theta$). Figure 2.12 (b) is a graphical representation of the XRD setup. We can perform different scans listed below to extract various information regarding the sample structure.

1. Rocking Curve: Plots $I$ Vs $\omega$, used to study defects.
2. Detector Scan: Plots $I$ Vs $2\theta$. 
3. Coupled Scan: Plots $I$ Vs $2\theta$ but $\omega$ also changes such that 
$\omega = \theta + \text{offset}$, used to study lattice mismatch, relaxation, thickness and more.

The structure factor ($F$) describes the effect of the crystal structure on the intensity of the diffracted beam. The integrated intensity of a diffracted beam is given by Equation 2.3.

$$I = F^2 p \left( 1 + \frac{\cos^2 2\theta}{\sin^2 \theta \cos \theta} \right) e^{-2M}$$

(2.3)

Here $p$ is the multiplicity factor, term in the parenthesis is the Lorentz-polarization factor and $e^{-2M}$ is the temperature factor [42]. Structure factor is described by Equation 2.4. Where $f$ is the atomic scattering factor, $u, v$ and $w$ are the atomic positions in the unit cell, $h, k$ and $l$ are the Miller indices.

$$F = \sum_i f_i e^{2\pi i (hu_i + kv_i + lw_i)}$$

(2.4)

Refractive index of a material is less than 1 for X-rays. Thus, X-rays undergo total reflection when incident on a flat material surface at a grazing angle, where incident angle $\theta < \theta_c$ (critical angle), as shown in Figure 2.13 (a). X-rays reflected from the material surface and the interface between the material and substrate, interfere, leading to oscillations (Kiessing fringes) in the reflectivity profile. Shown in Figure 2.13 (b), these oscillations depend on the thickness of the film (thicker film higher frequency of oscillations) and can be used as a tool to determine film thicknesses. Apart from X-ray diffraction, other tools such as SEM, AFM, XPS, ellipsometry, profilometry were used for structural characterization in this thesis work.
2.3.2 Vibrating Sample Magnetometer

Operating on the basis of Faraday’s law, vibrating sample magnetometer or the VSM [43,44] is a widely used tool for characterizing magnetic properties of samples. Measurement setup is illustrated in Figure 2.14 where, a sample fixed to a holder vibrates with an amplitude and frequency set by the oscillator, at the center of two pole pieces of an electromagnet producing a homogeneous magnetic field. A voltage will be induced on the stationary pick-up coils mounted on the electromagnet due to the change in flux caused by the vertical vibration of the sample. Lock-in technique is employed to extract the signal. We can understand the principle of operation as follows.

1. Sample vibrating at frequency $\omega$ give rise to a signal with the same frequency and amplitude $A$ in the pick-up coil through magnetic induction.

2. If this response is fed to a lock-in along with the signal driving the sample (reference), we obtain a signal described in Equation 2.5. Note,
here we assumed amplitude of signal driving the sample is $B$ with the same frequency but with a phase difference $\varphi$.

$$S = A \cos(\omega t) B \cos(\omega t + \varphi)$$

$$= \frac{1}{2} AB \cos(\varphi) + \frac{1}{2} AB \cos(2\omega t + \varphi) \tag{2.5}$$

3. The fast-varying component can be removed with the aid of a low-pass filter. Thus, only the constant voltage term $\frac{1}{2} AB \cos(\varphi)$ proportional to the signal from the sample remains.

4. Let us assume the signal from the pick-up coil interferes with external electromagnetic radiation $\left[ \sum A_i \cos(\omega_i t + \varphi_i) \right]$. Multiplying this with the reference signal $\left[ B \cos(\omega, t + \varphi) \right]$ we get the expression shown in Equation 2.6. Thus, it is easy to see, any electromagnetic interference with a frequency different from the reference signal, i.e. the signal driving the oscillation of the sample will be ignored with the measurement mechanism of the VSM. Emphasizing its importance in magnetometry.

$$S' = \sum_i \left( \frac{1}{2} A_i B \cos((\omega_i - \omega) t - \varphi_i) + \frac{1}{2} A_i B \cos((\omega_i + \omega) t + \varphi_i) \right) \tag{2.6}$$
2.3.3 Transport Measurements

Resistivity measurements for thin films are greatly influenced by the lead and the contact resistance. A four-probe measurement could help avoid such errors associated with the traditional two-probe measurements. Figure 2.15 compares these two techniques. In four-probe, negligible current flows through the voltmeter, the only voltage drop measured is across the sample. However, two-probe measure the sample, contact and the lead resistance. This would lead to significant errors while measuring thin films since sample resistance is comparable to the lead and contact resistance. Mathematical justification of this fact is as follows.
\[
\begin{aligned}
V_{2\text{-probe}} &= I \left( R_{\text{contact}} + R_{\text{sample}} + R_{\text{lead}} \right) \\
V_{4\text{-probe}} &= (I + i) R_{\text{sample}} + i \left( R_{\text{contact}} + R_{\text{lead}} \right) \\
V_{4\text{-probe}} &= IR_{\text{sample}} + i \left( R_{\text{contact}} + R_{\text{sample}} + R_{\text{lead}} \right)
\end{aligned}
\]

(2.7)

Figure 2.15 (a) Two-probe measurement setup where voltage drop is measured across the sample, leads and the contact. (b) Four-probe measurement setup, where voltage drop measured only across the sample.

Majority of the DC transport measurements in Xiao lab are performed at a four-probe station, where Keithley 220 current source provides the DC current, Keithley 2001 multimeter, and 2182A nanovoltmeter are used to measure appropriate voltages. Microprobes with tip diameter ranging from 1.2\(\mu\)m to 12\(\mu\)m being used to make contact with the samples. The external magnetic field is applied through a set of Helmholtz coils. Current through the magnet is controlled by a 20-20 KEPCO power supply.
supply. National Instruments LabView software is used to control instruments remotely and acquire data.
3.1 Introduction

Spintronics which deploys the spin, in addition to or sometimes in place of the charge of the electron has exhibited rich physics and industrial potential in the past two decades. Intensive research has been carried out investigating the interaction among, spin and magnetic field [45], electric current [46,47], electromagnetic waves [48], and recently temperature gradient [49].

Figure 3.1 Illustrates the interaction among electron charge and spin called spintronics. The interaction between charge and heat, thermoelectrics and amalgamation of spintronics and heat, spin caloritronics.
As we discussed earlier, incorporating the spin degree of freedom
revolutionized the charge-based electronics and lead to many industrial applications in
a wide variety of fields. Similarly, numerous applications have emerged in the field of
thermoelectrics, where interaction among electron charge and heat is exploited. For
example, modern warfare heavily depends on thermoelectric applications, ranging
from military avionics to body cooling systems. Further, internal combustion engines
lose more than half of the energy as heat, thermoelectric generators (TEGs) scavenge
this waste heat and convert into electricity. In other words, thermoelectrics has

Figure 3.2  (a) Spintronics applications: spin-torque MRAM and a magnetic
biosensor we are working on at Xiao lab for early childhood diabetes
detection. (b) Thermoelectric applications: Thermoelectric generator to
scavenge waste heat from automobile engines and a soldier equipped
with gear based on thermoelectrics. (c) Spin caloritronic application:
magnetic nano-scale heat engine, where domain walls in a nanowire are
moved with a temperature gradient resulting in a rotation of the wire.
Figure reprint from ref. 30, © 2012 NPG.
become an integral part of the day-to-day life. Impact of thermoelectrics can grow tremendously with the amalgamation of spin degree of freedom, a field comes to known as spin caloritronics. Figure 3.1 summarizes the underlying interactions of spintronics, thermoelectrics, and spin caloritronics. Whereas, Figure 3.2 represents few of the countless applications immerged from these fields. This chapter primarily deals with spin caloritronics, specifically spin Seebeck effect.

### 3.2 Spin Seebeck Effect

Figure 3.3 (a) Illustration of a thermocouple, most widely known application of Seebeck effect. Here two different metals are connected at the ends and kept at a temperature gradient generating a net flow of charges. (b) Ferromagnetic metal kept at a temperature gradient giving rise to a spin accumulation.

...It is always useful to compare novel concepts such as spin Seebeck effect to more familiar phenomenon like the Seebeck effect. Generation of a charge...
accumulation by placing a conductor in a thermal gradient is called the Seebeck effect (SE). Whereas, spin Seebeck effect (SSE) refers to the thermal generation of a spin accumulation. A temperature gradient in a ferromagnetic (FM) metal generates a spin chemical potential splitting between spin-up and spin-down electrons, owing to the spin-dependent density of states. This leads to a spin current with no net charge flow in an open circuit and is called the Spin Seebeck effect. First reported in ferromagnetic metals [50], SSE was later found to be present in ferromagnetic semiconductors [51] and insulators [52, 53]. Figure 3.3 (a) illustrates the most widely known application of SE, the thermocouple. Where two different metals connected at the ends are placed in a temperature gradient leading to a net flow of charges. Figure 3.3 (b) depicts SSE, where a magnetic metal placed in a temperature gradient leading to a spin accumulation. Here $J_c, J_s$ are the charge and spin current densities, $\nabla T$ the temperature gradient and $\mu_\uparrow - \mu_\downarrow$ the spin voltage.

A charge accumulation can be easily measured by simply hooking up a multimeter, but how to measure a spin accumulation with no net flow of charges? We employ a technique introduced in Chapter 1, the inverse spin Hall effect (ISHE) [25], where we exploit the strong spin-orbit coupling in a heavy metal. A temperature gradient will give rise to a spin accumulation in an FM due to SSE and injects a spin current into an adjacent heavy metal (HM) like Pt. Which converts the spin current into a measurable charge current due to spin-orbit coupling. Recall the formula for ISHE $J_c \propto J_s \times \sigma$, where $\sigma$ is the spin polarization vector. Thermal measurements are typically carried out in the longitudinal and transverse geometries [54]. As illustrated in Figure 3.4, in the longitudinal configuration, the temperature gradient is
applied normal to the plane of the ferromagnet whereas, in the transverse configuration, the temperature gradient is applied in the plane of the ferromagnet.

Figure 3.4  (a) Longitudinal configuration for measuring SSE, where the thermal gradient is applied perpendicular to the plane of the sample. A spin current will be injected from the FM to the HM, along the z-direction. Spin polarization vector aligns with the external magnetic field (y-direction). Thus, a measurable voltage builds up along the x-direction $(J_c \propto J_s \times \sigma)$. (b) Illustration of the transverse configuration for measuring SSE. Where a thermal gradient is applied along the plane of the sample.

**3.3 Drift-Diffusion Model (Bilayer)**

In a simplified scenario, assuming a transparent interface, the spin diffusion in the FM/HM bilayer, in the longitudinal configuration can be described by the one-dimensional drift-diffusion model [55] in Equation 3.1.

\[
\nabla^2 (\mu_\uparrow - \mu_\downarrow) = \frac{1}{\lambda^2} (\mu_\uparrow - \mu_\downarrow)
\]

\[
J_c = -\nabla \left( \sigma_\uparrow \mu_\uparrow + \sigma_\downarrow \mu_\downarrow \right) + \nabla \left( S_\uparrow \mu_\uparrow + S_\downarrow \mu_\downarrow \right) (-\nabla T) \tag{3.1}
\]

\[
J_s = -\nabla \left( \sigma_\uparrow \mu_\uparrow - \sigma_\downarrow \mu_\downarrow \right) + \nabla \left( S_\uparrow \mu_\uparrow - S_\downarrow \mu_\downarrow \right) (-\nabla T)
\]
Where $J_C$ the charge current density, $J_S$ the spin current density, $\lambda$ the spin diffusion length, and $\nabla T$ the temperature gradient. $\mu, \sigma$ and $S$ are spin electrochemical potential, electrical conductivity, and Seebeck coefficient respectively. Here the subscripts $\uparrow$ and $\downarrow$ denote spin-up and spin-down electrons, respectively. We refer to Figure 3.5 while solving drift-diffusion (DD).

![Figure 3.5](image)

Figure 3.5 Defines the coordinates for solving the drift-diffusion model for a bilayer system.

Upon solving drift-diffusion, we obtain,

\[
\begin{align*}
\left( \mu_{\uparrow} - \mu_{\downarrow} \right)_{FM} &= A_{FM} e^{\frac{z}{\lambda_{FW}}} + B_{FM} e^{-\frac{z}{\lambda_{FW}}} \\
\left( \mu_{\uparrow} - \mu_{\downarrow} \right)_{HM} &= A_{HM} e^{\frac{z}{\lambda_{HW}}} + B_{HM} e^{-\frac{z}{\lambda_{HW}}}
\end{align*}
\] (3.2)
Here \( A_{\text{FM}}, B_{\text{FM}}, A_{\text{HM}}, B_{\text{HM}} \) are arbitrary constants. Spin current distribution is obtained by applying boundary conditions, that the charge current vanishes everywhere, and the spin current vanishes at the two surfaces while being continuous at the FM/HM interface.

\[
(J_C)_{\text{FM}} = (J_C)_{\text{HM}} = 0 \tag{3.3}
\]

\[
(J_S)_{\text{FM}} \bigg|_{z=-d_{\text{FM}}} = 0
\]

\[
(J_S)_{\text{HM}} \bigg|_{z=+d_{\text{HM}}} = 0
\]

\[
(J_S)_{\text{FM}} \bigg|_{z=0} = (J_S)_{\text{HM}} \bigg|_{z=0}
\]

\[
J_{1,\text{rr}} (z) = \sigma_{\text{rr}} (S_r, -S_r) \forall T \left[ 1 - \frac{\lambda_{\text{rr}}}{\lambda_{\text{ru}}} C \left( \frac{d_{\text{ru}}}{\lambda_{\text{ru}}}, \frac{d_{\text{ru}}}{\lambda_{\text{ru}}}, \frac{d_{\text{ru}}}{\lambda_{\text{ru}}} \right) \right] + \frac{\sigma_{\text{ru}}}{\lambda_{\text{ru}}} C \left( \frac{d_{\text{ru}}}{\lambda_{\text{ru}}}, \frac{d_{\text{ru}}}{\lambda_{\text{ru}}}, \frac{d_{\text{ru}}}{\lambda_{\text{ru}}} \right)
\]

\[
J_{1,\text{ru}} (z) = \sigma_{\text{ru}} (S_r, -S_r) \forall T \left[ \frac{\lambda_{\text{ru}}}{\lambda_{\text{ru}}} C \left( \frac{d_{\text{ru}}}{\lambda_{\text{ru}}}, \frac{d_{\text{ru}}}{\lambda_{\text{ru}}}, \frac{d_{\text{ru}}}{\lambda_{\text{ru}}} \right) \right] + \frac{\sigma_{\text{ru}}}{\lambda_{\text{ru}}} C \left( \frac{d_{\text{ru}}}{\lambda_{\text{ru}}}, \frac{d_{\text{ru}}}{\lambda_{\text{ru}}}, \frac{d_{\text{ru}}}{\lambda_{\text{ru}}} \right)
\]

(3.4)

Here \( \sigma = \frac{2\sigma_+\sigma_-}{\sigma_+ + \sigma_-} \). The spin current in the HM measured through ISHE can be used to determine the strength of the SSE in the FM. It can be understood from Equation. 3.4 that the more spin current goes into the HM when its spin diffusion length is shorter and conductivity is higher than those of the FM [56–59]. Therefore, Pt is the optimum candidate as the HM for the longitudinal SSE measurement.

### 3.4 Motivation

Though SSE can be measured in the transverse or the longitudinal configuration as in Figure 3.4, it has been demonstrated that the SSE signal measured in the transverse configuration may be contaminated with an out-of-plane temperature gradient due to the finite thermal conductivity of the substrate [60]. The longitudinal
configuration has a well-defined direction for the temperature gradient and is suitable for the propagation of spin current due to the short spin-diffusion length. Thus, we have performed experiments exclusively in the longitudinal configuration. However, the transverse configuration may be suitable for substrate free samples [61]. By opting to perform experiments in the longitudinal configuration, we have managed to avoid a major source of ambiguity. However, there are several obstacles in implementing the longitudinal configuration to the SSE. Before going into the detailed discussion, let us briefly examine anomalous Nernst effect, in FM metals.

### 3.4.1 Anomalous Nernst Effect

![Anomalous Nernst Effect Diagram](image)

Figure 3.6 (a) Anomalous Nernst setup for FM metals. (b) Illustrates the magnetic proximity effect, where an HM placed on top of an FM material gets partially magnetized.
An FM metal with an emphasis on metal, under a temperature gradient, produces an anomalous Nernst signal given by Equation 3.5 and illustrated in Figure 3.6 (a).

\[ \tilde{V} V_{ANE} = \alpha \tilde{V} T \times \bar{\sigma} \]  (3.5)

Here \( V_{ANE} \) is the anomalous Nernst voltage, \( \alpha \) the anomalous Nernst coefficient. Side jump and skew scattering mechanisms due to spin-orbit coupling give rise to anomalous Nernst effect (ANE) similar to anomalous Hall effect (AHE) discussed in Chapter 1. Nevertheless, AHE is driven by an electric current whereas ANE is driven by a temperature gradient.

If you compare the longitudinal SSE (LSSE) setup in Figure 3.4 (a) with the anomalous Nernst setup in Figure 3.6 (a), they possess the exact same symmetry. In other words, the SSE signal measured in the longitudinal configuration is contaminated with the anomalous Nernst signal generated by the FM metal. Further, the HM placed on top of the FM may get partially magnetized due to magnetic proximity effect [62], as shown in Figure 3.6 (b). Magnetized HM layer generates its own ANE signal under a temperature gradient, further contaminating the LSSE signal. Therefore, isolation of pure SSE from other parasitic effects in an FM metal has been a matter of significant interest and a point of hot debate ever since SSE was first reported in an FM metal back in 2008. While many attempted to resolve the puzzle, others deviated from FM metals and lean towards magnetic insulators. Though there was noteworthy work done to settle the issue, none have been fully successful. Here we propose an elegant technique to disentangle SSE from proximity induced and ferromagnet induced anomalous Nernst signals in an FM metal.
3.5 Methods and Results

3.5.1 Anomalous Nernst Contribution to Thermal Measurements from FM Metal

The problem we are trying to address here is two-fold, let us try to deal with one at a time. Upon examining Equation. 3.5 describing ANE, one can immediately see the use of an FM metal with zero anomalous Nernst coefficient will be an ideal experimental solution to eliminate the ANE signal originating from the FM metal. To fabricate an FM with zero anomalous Nernst coefficient, two typical FM metals, Fe and Ni, known to have opposite anomalous Nernst coefficients [63,64] can be alloyed. All necessary thin films were fabricated on a wet thermally oxidized Silicon wafer (Si/SiO$_2$) with magnetron sputtering at a base pressure lower than $5 \times 10^{-7}$torr, 4.5mtorr working pressure and 10sccm argon flow. All samples were capped with a 5nm SiO$_2$ layer. Which serves two purposes, it prevents samples from oxidation and electrically insulates the heater from the sample.

Few batches of samples were fabricated with the co-sputtering technique. Where, Fe and Ni targets were mounted on adjacent guns in the sputtering chamber, ignited simultaneously and the substrate swept between the guns. If the substrates were to be swept at a sufficiently higher rpm, we can form a NiFe alloy rather than alternating films of Ni and Fe. By controlling the power of the dc sources igniting and sustaining the Fe and Ni plasmas we can control the composition of the deposited film. However, we quickly realized films were not uniform. Thus, switched to forging composite sputtering targets.

Here, Fe and Ni powders, -325 mesh and 99.9% purity were mixed and pounded together and then cold pressed into a die with a diameter 1.75". Targets formed as described were placed in a crucible and annealed at 1100°C in a gas
atmosphere of 5% H₂, balance Ar (forming gas) for 1h. The furnace was sealed and purged with forming gas for about 30min prior to the annealing process to drive away any air trapped in the tube. Annealing profile is illustrated in Figure 3.7. Subsequent to target preparation all necessary samples were deposited with magnetron sputtering. Fabricated samples were defined into 15mm×2mm pieces and pasted on to a thick Cu block using thermal grease. The heater is placed on top and maintained at a fixed power. All electrical contacts were made with indium press.

Figure 3.7 Illustrates target annealing profile. The furnace temperature is elevated to the target value of 1100°C at a rate of 5°C/min. After a soak time of 1h, the system was allowed to cool to room temperature. Entire annealing process takes place in a forming gas environment.

Figure 3.8 (a) shows the anomalous Nernst measurements for select NiₓFe₁₀₀₋ₓ(5) nominal compositions. The number in the parentheses is the thicknesses in nanometers. By tuning the composition of the alloy NiₓFe₁₀₀₋ₓ, where we invoke the
competition between opposite anomalous Nernst signals of constituent elements, achieved zero Nernst coefficient in \( \text{Ni}_{7}\text{Fe}_{93} \) at a thickness of 5nm. Figure 3.8 (b) compares the anomalous Nernst signal of optimized composition \( \text{Ni}_{7}\text{Fe}_{93}(5) \) with that of pure \( \text{Ni}(5) \) and \( \text{Fe}(5) \). The optimized composition has a profile similar to pure Fe and an anomalous Nernst coefficient at least 20 times smaller than that of Ni or Fe, and hence considered here as the “zero Nernst material”.

Figure 3.8  (a) Anomalous Nernst effect measurement for select \( \text{Ni}_{x}\text{Fe}_{100-x}(5) \) compositions. (b) Comparison of anomalous Nernst signal of \( \text{Ni}(5) \), \( \text{Fe}(5) \) and, \( \text{Ni}_{7}\text{Fe}_{93}(5) \).

Since we have established an FM metal with “zero anomalous Nernst coefficient”, we may use this composition to perform SSE measurements by placing an HM like Pt adjacent to the FM material. In Figure 3.9 (a), a representative curve for \( \text{Ni}_{7}\text{Fe}_{93}(5)/\text{Pt}(1) \) is shown. When an HM, Pt is placed adjacent to \( \text{Ni}_{7}\text{Fe}_{93}(5) \) the “zero Nernst material”, a Nernst-like voltage can be observed due to the ISHE in the Pt. Measured Nernst-like voltage is in the same polarity as that of pure Ni, discussed
above. Recall signal for Ni$_7$Fe$_{93}$(5) was in the same polarity as pure Fe. As a comparison, the curve for a control sample Ni$_7$Fe$_{93}$(5)/Cu(1) is shown in the same figure, with a signal 20 times smaller. An electric field arises in Pt due to the ISHE, $E_{\text{ISHE}} = \rho_{\text{Pt}} \theta_{\text{SH}} J_{s_{-H_M}}$ where $\rho_{\text{Pt}}$ is the resistivity and $\theta_{\text{SH}}$ is the spin Hall angle of the Pt layer. Considering the shorting of the adjacent FM layer, the measured voltage can be derived as Equation 3.6, where the length of the sample under a temperature gradient is $L$ and, $R$ is the sheet resistance of the sample.

![Graph](image.png)

Figure 3.9 (a) The thermal voltage measured for Ni$_7$Fe$_{93}$(5)/Pt(1) and control sample Ni$_7$Fe$_{93}$(5)/Cu(1). Films are heated under the same heater power. The signal for the sample with Pt is more than 20 times larger than that of Cu. (b) The thickness dependence of the thermal voltage. Curve for Ni$_7$Fe$_{93}$(5)/Pt(x) peaked when Pt thickness is about 2nm. The red curve represents the drift-diffusion fit, parameter for the fitting are: $\lambda_{\text{Pt}} = 1.3 \text{nm}$, $\lambda_{\text{FM}} = 5 \text{nm}$, $d_{\text{FM}} = 5 \text{nm}$, $\sigma_{\text{Pt}} = 5 \text{a.u.}$, $\sigma_{\text{FM}} = 1 \text{a.u.}$ and $(S_{\uparrow} - S_{\downarrow}) \nabla T = 1 \text{a.u.}$. Curve for Ni$_7$Fe$_{93}$(5)/Cu(x) does not show any significant variation with the thickness of Cu, except a little bump around 2nm.
thickness dependence study of Pt and control sample with Cu is shown in Figure 3.9 (b). Since the spin diffusion length is very short in Pt, the spin current distribution in Ni\textsubscript{7}Fe\textsubscript{93}(5)/Pt(x) bilayer will remain unchanged as Pt gets thicker. Thus, measured voltage scales with the total resistance. Indeed, as shown in Figure 3.9 (b), the SSE voltage increases quickly with the Pt thickness up to about 2nm, followed by monotonic decay as the Pt thickness increase further. The red curve is a fitting based on Equation 3.6, which predicts a spin diffusion length in Pt to be about 1.3nm. This result is consistent with the values extracted from the spin pumping measurements [65]. It is important to note that this technique can be used to determine spin diffusion lengths of HMs. The signal for the control sample Ni\textsubscript{7}Fe\textsubscript{93}(5)/Cu(x) does not show any significant variation except a little bump around 2nm.

3.5.2 Anomalous Nernst Contribution to Thermal Measurement from Proximity Induced FM Layer

An interface effect such as proximity effect [62] can be surfaced as a possible explanation for the signal measured for Ni\textsubscript{7}Fe\textsubscript{93}(5)/Pt(x). When Pt is in contact with an FM, it may be partially magnetized giving rise to anomalous Nernst signal that has the same profile as the SSE. Due to the short spin diffusion length, it is difficult to distinguish between the SSE and the proximity effect from the length scale fitting in Figure 3.9 (b). Insertion of a thin piece of Cu in between Ni\textsubscript{7}Fe\textsubscript{93} and Pt help eliminate the proximity effect while preserving the SSE. Following questions may arise naturally regarding insertion of the non-magnetic metal (NM) Cu between the FM and HM,

1. Could Cu play the role of an FM metal and give rise to an SSE signal?
2. Could Cu play the role of an HM and give rise to an inverse spin Hall signal?

3. Could insertion of Cu hinder the propagation of spin current into the HM?

Cu is a typical diamagnetic metal, far from Stoner instability [66,67], which is neither inherently magnetic nor becomes magnetic by being in proximity to the FM metal. Hence Cu will not give rise to any spin Seebeck signal and should be ruled out as a source of spin Seebeck effect. Apart from not being a source of SSE, the decision to use Cu as the light metal in the experiment was primarily driven by two factors. It does not produce any appreciable inverse spin Hall voltage [68,69], or in other words, spin Hall angle of Cu is negligibly small. Moreover, with a very large spin diffusion length [70], it does not hinder the propagation of spin current. Hence, we can rule out any inverse spin Hall contribution from Cu to the measured thermal voltage as well. Based on these facts, Cu is an ideal choice for the insertion layer between the FM and the HM. As illustrated in Figure 3.10, Ni$_7$Fe$_{93}$(5)/Cu(2)/Pt(x) sample still shows a sizable Nernst-like voltage. The magnitude is reduced due to the shunting effect, that the Cu shorts the voltage. Another possible reason for the reduction of the magnitude is the change of transparency to spin current at the interface so that the spin current entering Pt is reduced. Despite the reduction of the magnitude, comparing Figure 3.9 (b) and 3.10 one can still observe the same trend in the thickness dependence with and without the insertion of Cu.
Figure 3.10  Thermal voltage measured for Ni$_7$Fe$_{93}$Cu(2)/Pt(x). Thickness dependence is similar to the case without Cu insertion, except for a reduction in magnitude, owing to shorting due to Cu and change in the transparency for spin current with the insertion of the Cu layer.

One may wonder how we can ensure that the temperature gradient is the same when we change the top layer thickness, as shown in Figure 3.8 (b). Let us try to handle this subtlety with the following proof. Thin films stack along with the substrate shall be treated as series thermal resistors for heat conduction purposes, as shown in Figure 3.11 (a). Fourier equation, for steady conduction through a constant area plane interface, can be written as Equation 3.7. Where $\dot{Q}/A$ (Wm$^{-2}$) is the local heat flux density, $k$ (Wm$^{-1}$K$^{-1}$) is the thermal conductivity, $\nabla T$ (Km$^{-1}$) is the temperature gradient, $T_h$ (K) the high temperature, $T_l$ (K) the low temperature, and $t$ (m) the film thickness.
\[ \dot{Q} = -kA \frac{T_H - T_L}{t} \]  

(3.7)

Equation 3.7 can be rearranged as \( \dot{Q} = \frac{T_H - T_L}{R} \), where \( R = \frac{t}{kA} \) is the thermal resistance.

Figure 3.11 (a) Stack of thin films along with the substrate treated as series thermal resistors for heat conduction purposes. (b) Power dependence of measured anomalous Nernst signal for different thicknesses of Ni_{7}Fe_{93} sample.

Thin films stack shall be modeled as a thermal resistor network. Where

\[ R_{total} = R_{Pt} + R_{Cu} + R_{NiFe} + R_{Sub}. \]

\[ \dot{Q} = \frac{T_1 - T_2}{R_{Pt}} = \frac{T_2 - T_3}{R_{Cu}} = \frac{T_3 - T_4}{R_{NiFe}} = \frac{T_4 - T_{Sink}}{R_{Sub}} \]

\[ \Rightarrow \dot{Q} = \frac{T_1 - T_{Sink}}{R_{total}} \]  

(3.8)

Substrate thickness \( t_{Sub} = 500 \mu m \) as compared to \( nm \) scale thicknesses of other films and the thermal conductivity of the substrate \( k_{Sub} \ll k_{Pt}, k_{Cu}, k_{NiFe} \).
\[ R_{\text{Sub}} \left( \frac{T_{\text{Sub}}}{k_{\text{Sub}}A} \right) \gg R_p, R_{\text{Cu}}, R_{\text{NiFe}}. \] Thus, the substrate completely overwhelms the thermal conduction process. Therefore, within the thickness range of the top layer we probed, variation in the temperature gradient can be considered negligibly small. Moreover, as shown in Figure 3.11 (b), the measured thermal signal is linear with the heater power.

By adopting Ni\textsubscript{7}Fe\textsubscript{93}(5), a “zero Nernst material” as the FM and by inserting a thin layer of Cu between Ni\textsubscript{7}Fe\textsubscript{93}(5) and Pt, we have mitigated, two major contributions, ANE and proximity effect, eclipsing the LSSE measurements. To further solidify the results, a thickness dependence study has been performed on the Ni\textsubscript{7}Fe\textsubscript{93}, the FM metal used in the experiment. The results are shown in Figure 3.12 (a). Inverse thickness dependence is as in Figure 3.12 (b). This result is very interesting and merits detailed discussion.

### 3.6 Discussion

As illustrated in Figure 3.12 (a), the anomalous Nernst voltage measured for Ni\textsubscript{7}Fe\textsubscript{93}, varies with the thickness, suggesting the bulk anomalous Nernst contribution is not absolute, whereas a linear inverse thickness dependence shown in Figure 3.12 (b), suggests the existence of a surface anomalous Nernst contribution, a result never been reported before. The inverse thickness dependence shown in the Figure 3.12 (b) is fitted with a linear function given in Equation 3.9. It should be noted, though the resistivity of the Ni\textsubscript{7}Fe\textsubscript{93} depends on the film thickness, behavior of the ANE signal does not directly correspond to the resistivity, a sign change in the ANE signal can no way be explained by the resistivity.
Here \( V_b, V_s \) and \( V \) are the bulk, surface, and total anomalous Nernst contributions, respectively and \( t \) denotes the thickness of the FM. Linear fit yields \( V_s = 5.47 \mu V \) and \( V_b = -29.51 \mu V \text{nm} \) Hence bulk ANE with a 5nm thick layer of Ni\(_7\)Fe\(_{93}\) is about \(-5.90 \mu V \) and with a surface ANE contribution of \( 5.47 \mu V \), results in a total anomalous Nernst signal of \(-0.43 \mu V \), consistent with the data presented in Figure 3.8 (b). Therefore in the pursuit of a zero Nernst material, what we have truly achieved is the pivotal point where bulk ANE cancels off the surface ANE of the composition Ni\(_7\)Fe\(_{93}\) at a thickness of 5nm.

Many noteworthy works have been done to isolate SSE from the thermal measurement performed in the longitudinal configuration for an FM metal. Ramos, et al. [71] performed the thermal measurements on conducting films of magnetite, where they measured the ANE of magnetite directly and in the LSSE configuration. Further, estimated the ANE contribution due to proximity effect by dividing the Pt layer into
magnetic and non-magnetic regions. Since magnetite in the metallic phase has a resistivity of two orders of magnitude larger than that of Pt, ANE contribution of the magnetite is greatly suppressed by the Pt. In fact, the authors report ANE contribution from the magnetite to the measured thermal signal is only 3%. This result cannot be extended to isolate the SSE from ANE in the present work. Since FM and HM used here have resistivities of the same order. Jiang, et al. [72] studied the Topological Insulator (TI)/ Yttrium Iron Garnet (YIG) structure. Where they eliminated the contribution from ANE to the measured thermal signal by choosing YIG, a ferrimagnetic insulator as the source of SSE. Methodology to remove proximity induced ANE signal from the TI here is very innovative. By means of anomalous Hall measurements, authors establish a critical temperature beyond which proximity induced anomalous Nernst contribution from the TI layer is negligibly small and performed SSE measurements at a temperature well beyond the critical temperature. However, TI/ YIG system is starkly different from ours. Firstly, we use an FM metal as oppose to YIG, a ferrimagnetic insulator, where anomalous Nernst signal from the magnetic material cannot be avoided. Moreover, the critical temperature to avoid proximity induced anomalous Nernst contribution from the TI is well below 300K. Without extensive studies, we cannot guarantee such a methodology is suitable for an all metallic system. Even if such a technique is suitable, we should pay attention to the appropriate critical temperature, as this might be too high for an FM metal where it would lose its magnetic properties resulting in no signal.

Xu, et al [69] performed similar experiments with NiFe/Pd structure. In addition, they replaced the HM with β-Ta which is known to have a spin Hall angle opposite to that of Pd [73] and yet observed a thermal signal with the same sign as Pd.
Having eliminated proximity ANE by a Cu insertion layer Xu, et al. concluded, the majority of the signal arose from the ISHE in the magnetic layer, which was caused by the spin current redistribution in the magnetic layer induced by the presence of the probe layer. From Equation 3.4 it is clear, that the neighboring heavy metal will significantly modify the spin current distribution in the FM.

Figure 3.13 (a) Calculated spin current distribution for an FM/HM (Pt) bilayer in the longitudinal configuration where \( \lambda_{HM} = 1\text{nm} \), \( \lambda_{FM} = 5\text{nm} \), \( d_{HM} = 5\text{nm} \), \( d_{FM} = 5\text{nm} \), \( \sigma_{HM} = 1\text{a.u.} \), \( \sigma_{FM} = 1\text{a.u.} \), \( (S_{\uparrow} - S_{\downarrow})\nabla T = 1\text{a.u.} \).

(b) Calculated spin current distribution for a FM/HM (Ta) bilayer in the longitudinal configuration where \( \lambda_{HM} = 1\text{nm} \), \( \lambda_{FM} = 5\text{nm} \), \( d_{HM} = 5\text{nm} \), \( d_{FM} = 5\text{nm} \), \( \sigma_{HM} = 0.1\text{a.u.} \), \( \sigma_{FM} = 1\text{a.u.} \), \( (S_{\uparrow} - S_{\downarrow})\nabla T = 1\text{a.u.} \).

(c) Calculated spin current distribution for a FM/LM (Cu) bilayer in the longitudinal configuration where \( \lambda_{HM} = 50\text{nm} \), \( \lambda_{FM} = 5\text{nm} \), \( d_{HM} = 5\text{nm} \), \( d_{FM} = 5\text{nm} \), \( \sigma_{HM} = 1\text{a.u.} \), \( \sigma_{FM} = 1\text{a.u.} \), \( (S_{\uparrow} - S_{\downarrow})\nabla T = 1\text{a.u.} \).

Figure 3.13 shows representative SSE driven spin current distributions calculated with Equation 3.4. Here we strived to uncover the effect on the spin current distribution when different types of metals were placed on top of the FM. We compare the spin current distributions with Pt and Ta placed on top of the FM. Both HMs have similar spin diffusion lengths. However, Ta is highly resistive compared to Pt.
Furthermore, spin current distribution with a light metal Cu placed on top of the FM has been derived as well. Spin diffusion length of Cu is (200nm-300nm) much larger compared to the spin diffusion length of Pt/Ta (~1.5nm). Spin current distributions derived for different cases indeed confirm the claims of Xu, et al. Though this result is appreciable, SSE has been separated from ANE only exploring the large thickness ($t > 2\lambda$) limit, detailed studies of the thin end is necessary to understand the full picture. With experiments performed probing the entire thickness range, it is clear, that the ANE of the isolated FM is not a pure bulk effect, rather contains a surface component, which will be modified by the presence of any neighboring layer. More importantly, what is considered by Xu, et al. as intrinsic ANE, coming from the bulk of the FM, is not truly intrinsic rather changes with the introduction of an adjacent metal, particularly with an introduction of an HM.

In Figure 3.8 (a) “zero Nernst material”, Ni$_{7}$Fe$_{93}$(5) has a much smaller ANE magnitude, however, undergoes a polarity change as Ni concentration in the composition is decreased. What drives such a polarity change? As shown in Figure 3.8 (a), when Ni concentration in the composition is gradually reduced, the polarity of the measured signal switches from Ni-like to Fe like and, if Ni concentration is reduced further, the signal becomes prominently Fe like. This result comes with no surprise, as our initial goal was to obtain a zero Nernst material, purely based on the competition between opposite anomalous Nernst signals of Ni and Fe. Thus, Ni and Fe having opposite polarity play a decisive role in obtaining a zero Nernst material. However, based on the thickness dependence study of Ni$_{7}$Fe$_{93}$ shown in Figure 3.12 (a), the measured thermal voltage for the composition can be tuned with the thickness, invoking the competition between the surface and the bulk effects. Therefore,
competition between opposite polarities of the constituent elements in the composition as well as competition between surface and bulk contributions both are important in fabricating a zero Nernst material, where total anomalous Nernst coefficient is minimized.

Keeping this in mind let us try to interpret some of the experimental data which are hard to reconcile otherwise. Comparing the data presented in Figure 3.8 (b) and 3.9 (a), we can observe some peculiar behavior. The polarity of the ANE signal measured for $\text{Ni}_7\text{Fe}_{93}(5)/\text{Cu}(1)$ is opposite to that of $\text{Ni}_7\text{Fe}_{93}(5)$. As we discussed earlier, Cu will not give rise to any SSE signal or any appreciable inverse spin Hall signal. Hence polarity change in the ANE signal, as a Cu layer is introduced on top the FM should originate elsewhere. It is apparent from the analysis of the inverse thickness dependence of $\text{Ni}_7\text{Fe}_{93}$ shown in Figure 3.12 (b), ANE signal bears the sign of the dominant contribution of surface or bulk. Since at a thickness of 5nm magnitude of the bulk contribution is larger ($-5.9 \mu V$) than that of the surface ($5.47 \mu V$), the ANE signal carries the sign of the bulk contribution (negative). However, as a Cu layer is introduced on top of the $\text{Ni}_7\text{Fe}_{93}(5)$, the surface contribution is enhanced and if the magnitude of the enhanced surface contribution is larger than that of the bulk, ANE signal will now carry the sign of the surface contribution (positive). This is a strong evidence, reiterating the importance of the surface contribution to the ANE.

Cu, layer thickness required to guarantee total surface coverage of the FM is about 2nm. Cu placed on top of the FM will influence the surface until full surface coverage is reached and any Cu placed beyond this thickness does not influence the surface, rather only contribute to shunting. Therefore, surface contribution to the measured thermal voltage will vary until full surface coverage is reached and plateau
thereafter. Thus, explaining the bump around 2nm in the thermal voltage for Ni$_7$Fe$_{93}$Cu(x) shown in Figure 3.9 (b). Incidentally, the spin diffusion length of Pt($\lambda_{sp}$) is around 1.3nm and, as shown in Figure 3.9 (b), measured thermal voltage for FM/Pt structure peaks around that thickness, hence appearing to have a peak position similar to that of FM/Cu.

3.7 Conclusion

In summary, we employed the zero Nernst material approach to mitigate the ANE contribution in the thermal measurement of FM metal, and a thin Cu insertion layer to mitigate the magnetic proximity effect. These modifications were substantial in the process of developing a technique to isolate SSE from other parasitic effects in an FM metal. Our studies have confirmed that FM alloys can indeed have zero Nernst coefficient, which arises not only from the cancellation from, constituent elements having opposite Nernst coefficients but also from the competition among bulk and surface contributions. Effectively, zero Nernst coefficient is achieved when the bulk and surface contributions are equal but opposite in sign. The bulk contribution, which should be related to spin current distribution, will be modified with a good spin sink neighboring layer such as an HM layer. The surface contribution will also be affected by the neighboring layer arising from multiple factors such as spin accumulation, proximity effect, or density of states modification. Considering the magnitude of both bulk and the surface contributions in the isolated FM are approximately as large as 6$\mu$V, which is comparable to the observed “SSE” signal around 8$\mu$V, surface contribution to the ANE is a substantial effect in the thermal measurement of an FM metal. Our efforts uncovered the presence of both surface and bulk ANE and any
future work aimed at isolating SSE from the thermal measurement of FM metals should take this phenomenon into consideration.
Chapter 4

MAGNETIC TUNNEL JUNCTION BASED SENSORS FOR EARLY CHILDHOOD DIABETES DETECTION

4.1 Introduction

According to the National Health Expenditure Accounts (NHEA), the official estimate of total healthcare spending in the United States reached $3.3 trillion in the year 2016. Which means per capita national health expenditure is over $10,000, a truly staggering number. Are these numbers real or artificially inflated? Let me shed some light on this controversy by means of a couple of examples. Depending on the complexity of the analysis, it costs between $100-$3,000 to get blood work done at a laboratory. Similarly, it costs anywhere between $30 to $250 for a urine test. These are the two most commonly ordered tests by doctors. A simple urine test involves a plastic cup, a dipstick and a few minutes of a technician’s time. Even when we factor in the overheads this should not cost more than $5. It appears there is a big discrepancy in the math. Insurance companies are aware of these costs and will only pay the lowest acceptable amount to the service provider. Thus, hospitals and labs will bill the patient an amount far exceeding the cost, to get every penny possible from the insurance companies. The tragic part of this story is someone without insurance will end up paying the full amount billed which is unfair, to say the least. Therefore, making tools and techniques which would help alleviate some of this expenditure a clinical reality should be of utmost importance. Diagnostic tests account for a substantial portion of the healthcare expenditure. Point of care testing (POCT) referring to tests performed in
the presence of the patient, typically with a portable hand-held device or a small benchtop analyzer can be a viable alternative in this regard. Simpler sample collection, simpler pre-analytical process, faster test results, increased patient satisfaction are all advantages of this technology. Moreover, instant test results allow physicians to make better and faster decisions leading to an improved health outcome. In countries with poor medical infrastructure or limited access to healthcare, POCT can be potential lifesavers [74,75]. POC testing technologies are no longer limited to simpler diagnostics such as glucose monitoring, pregnancy testing, and drug-abuse screening, with ever-increasing demand much research and funding, go into improving the existing technologies and adding new capabilities. Nanomedicine, the amalgamation of nanotechnology with medical diagnostics has gained a lot of traction in recent years. Specially POC assay systems using magnetic nanoparticles (MNPs), providing improved sensitivity and specificity along with better reproducibility and reliability.

The objective of this project is to develop an assay for early risk assessment in genetically pre-disposed children with Type-I diabetes. Current methods for diagnosing Type-I diabetes in children are very limited due to detection techniques, poor sensitivity, bleaching, and quenching. We combine biotechnology with MNPs to enable enrichment of the diabetes autoantibodies in a test sample. Then, detect and measure MNPs with enhanced sensitivity on a magnetic tunnel junction (MTJ) sensor.

4.2 General Principle of Operation

Though specific steps may vary depending on the target analyte being detected, the general principle of operation of MTJ based bio-sensors remains the same. Figure 4.1 is a schematic illustration of the operational procedure and the steps involved are listed below [76].
Figure 4.1  Schematic illustration of the operational procedure of MTJ based magnetic bio-sensor.

1. **Step 1:** Sensing area is functionalized with probe molecules capable of selectively binding to target analyte molecules. For example, in DNA sequence detection, the sensor surface is functionalized with complementary DNA to capture the target DNA.

2. **Step 2:** Target analytes are tagged with MNPs.

3. **Step 3:** Sample solution is introduced to the sensor surface.

4. **Step 4:** Sensor surface is “washed”, we will get to the detailed discussion of this later in the chapter. If the target analyte is present in the sample solution it will selectively bind to the probe molecule and will not be removed upon washing.

5. **Step 5:** Magnetic labels will produce a stray field when an external magnetic field is turned on, which can be detected with the MTJ, hence the presence of the target analyte.
4.3 Type-I Diabetes

Type-1 diabetes is characterized by severe insulin deficiency resulting from an autoimmune destruction of insulin-producing beta cells from the islets of Langerhans in genetically susceptible individuals. Type-1 diabetes is also characterized by the presence of autoantibodies against islet cell antigens (ICAs) that recognize insulin, glutamate decarboxylase (GAD65) and protein tyrosine phosphatase-related insulinoma-associated protein 2 (IA-2). These autoantibodies appear to develop sequentially. Antibodies against insulin are often expressed first, especially in young children. Anti-GAD65 antibodies may represent a propensity for general autoimmunity, whereas antibodies against IA-2 are a more specific marker of beta cell destruction. Autoantibodies against ICAs are important preclinical markers as they may be present for years before the diagnosis of diabetes. In approximately 90% of new-onset Type-1 diabetes patients, one or more beta cell autoantibodies are present. Detection of two or more islet autoantibodies is associated with a higher risk of Type-1 diabetes. In addition, the risk of diabetes for first-degree relatives depends on the number and type of antibodies that are present. Therefore, testing for multiple antibodies is important for diabetes risk assessment. The autoantibodies seen in children are often different than those seen in adults. Autoantibody against insulin (IAA) is usually the first marker to appear in young children, and about 50% of children with new-onset Type-1 diabetes will be IAA-positive.

4.4 Fabrication and Optimizing Magnetic Tunnel Junctions

As we discussed in Chapter 1 magnetic tunnel junctions or MTJs consist of two ferromagnetic (FM) layers separated by an ultrathin insulating layer. The resistance of such devices depends on the relative orientation of the magnetization
vectors of the FM layers. Electrons would tunnel through the insulator. Tunneling probability is low while magnetizations are aligned anti-parallelly, resulting in a high resistance, whereas parallel alignment results in a high tunneling probability, thus a low resistance.

4.4.1 Anti-Ferromagnetic/ Synthetic Anti-Ferromagnetic Pinning

Since sensor response is based on the relative orientation of the magnetization vectors of the FM layers a hard-soft configuration is crucial to achieving optimal sensitivity. In other words, it is best to have one FM layer with a magnetization vector fixed (pinned) whereas the other FM layer rotates with the sensing field. Of many
schemes attempted exchange biased systems depicted in Figure 4.3 (a) yields the most promising results in this regard [77,78].

Figure 4.3 Depicts the scheme to achieve interface coupling at Anti-ferromagnet/Ferromagnet interface due to exchange anisotropy.

Here an anti-ferromagnetic (AFM) layer is attached to the FM layer and an external magnetic field is turned on at elevated temperatures (T) larger than the Néel temperature ($T_N$) of the AFM, which is necessarily smaller than the Curie temperature, $T_C$ ($> T_N$) of the FM. Under these conditions, AFM spins remain random and FM spins align with the external field (Figure 3 (a)-I). Now the system is cooled below the Néel temperature, maintaining the field. AFM spins at the interface align ferromagnetically with the FM spins due to interaction. However, other spins in the AFM layer will be in anti-ferromagnetic order (Figure 3 (a)-II). When the external field is reversed, AFM spin remains in the same configuration for sufficiently large AFM anisotropy, whereas
FM spins try to follow the external field. Due to interfacial interaction at the AFM-FM interface, AFM spins at the interface exert a torque on the FM spins making it difficult to rotate the magnetization of the FM layer. Thus, this scheme can help pin the magnetization of one FM layer in the MTJ.

However, systems with AF pinning limits the operational temperature of the sensors to well below the Néel temperature of the AF layer. Thus, for applications demanding higher operational temperatures, systems with synthetic anti-ferromagnetic pinning (SAF) are better suited [79,80]. Here two ferromagnetic layers are separated by a non-magnetic metallic spacer as shown in Figure 4.3 (b). The thickness of the spacer layer can be chosen such that the two FM layers are strongly anti-ferromagnetically coupled [9]. Moreover, one FM layer in the SAF layers is exchange coupled to an AF. Without such exchange coupling, the antiparallel axis would irreversibly flop to a direction orthogonal to the external field [81–83].

4.4.2 Sample Fabrication

All necessary thin films were fabricated on wet thermally oxidized Silicon wafers (Si/SiO$_2$) with magnetron sputtering. Si wafers were cut into 1.75"×1" pieces and cleaned with Acetone and Ethanol while sonicating to remove any contaminants from the substrate surface. Subsequently flushed with deionized water and dried with compressed nitrogen.

Two different types of MTJs one with aluminum oxide (Al$_2$O$_3$) barrier and the other with magnesium oxide (MgO) barrier were deposited. Figure 4.4. illustrates the thin film stacks.
Figure 4.4  (a) Illustrates the thin film stack for alumina junction with anti-ferromagnetic pinning. Numbers in the parenthesis represent the thickness in nm. (b) represents the thin film stack for the MgO junction with synthetic anti-ferromagnetic pinning.

Alumina (AlO$_x$) Junction:

Ta(10)/Cu(120)/CoFe(4)/IrMn(15)/CoFe(3)/AlO$_x$(2)/Py(20)/Ta(20)/Cu(50)/Au(50)

Numbers in the parenthesis correspond to the film thickness in nanometers.

Role of each layer in the alumina junction with AF pinning is as defined above.
Ta(10)/Cu(120) serves as the seed layer which provides a surface with minimal roughness for the further growth [84]. As we discussed earlier in the chapter CoFe/IrMn/CoFe is the bottom ferromagnetic layer (FM$_1$) with anti-ferromagnetic pinning. With high exchange bias energy, high blocking temperature and low critical thickness [85], choosing IrMn as the AF material is justified. Further, in CoFe$^{\text{Bottom}}$/IrMn / CoFe$^{\text{Top}}$, bottom CoFe facilitate the proper growth of IrMn [86] while top CoFe is pinned. AlO$_x$ the insulating barrier. Permalloy (Py-Ni$_{80}$Fe$_{20}$) the free/sensing
layer (FM\textsubscript{2}) and Ta/Cu/Au constitute the top electrode. All metallic layers have been deposited with DC sputtering. However, the tunneling barrier can be formed in several ways. One can sputter an Al\textsubscript{2}O\textsubscript{3} target with RF sputtering or sputter metallic aluminum (Al) in argon and oxygen environment (reactive sputtering), but we opt to deposit an Al layer and expose it to oxygen plasma in-situ. This technique, in general, facilitates the growth of nearly pin-hole free tunneling barriers.

### 4.4.2.1 Forming Aluminum Oxide Barrier

To optimize the aluminum oxide barrier formation, a 2nm thick aluminum layer is deposited on multiple substrates with DC magnetron sputtering at 26W power, 4.5mtorr pressure and 10sccm Ar flow. Then we cut off the Ar flow and leave the sputtering chamber idle for about 30min to flush out all the Ar from the system. Now oxygen is introduced to the chamber and the plasma is activated at the 80W power and 60mtorr pressure. Al layers are exposed to the plasma in two steps. Several short exposures typical lasting 1s each followed by a long exposure lasting anywhere between 1.5-4min. If Al layers were exposed to the oxygen plasma for prolonged periods (few seconds) initially, a thin Al\textsubscript{2}O\textsubscript{3} layer forms at the surface thus preventing further oxidation. However, several short exposures result in the formation of less stoichiometric aluminum oxide at the top. Thus, allowing the oxidation of the whole metallic layer. All alumina junctions with oxidation time less than 2min happen to be short, in other words, FM layers were not electrically isolated by the alumina barrier. This is likely due to the under oxidation of Al layer, thus not forming a barrier, resulting in electrical conduction, not tunneling. Optimal oxidation time for 2nm Al layer is 3min under the prescribed conditions.
Magnesium Oxide Junction:

\[
\text{Buffer SAF Pinned-FM Barrier FM Top Electrode} \\
\begin{array}{c|c|c|c}
\text{Ta(7)/Ru(30)/Ta(7)} & \text{CoFe(4)/IrMn(15)/CoFe(3)/Ru(1)/CoFeB(3)} & \text{MgO(2)/CoFeB(3)/Ta(10)/Ru(10)/Au(50)} \\
\end{array}
\]

Role of each layer in the MgO junction with SAF pinning is as defined above. Ta/Ru/Ta layers serve as the buffer with minimal roughness, enabling good pinning and thermal stability upon annealing [87]. Bottom CoFeB (FM\(_1\)) layer is synthetic anti-ferromagnetically pinned to the CoFe through Ru. Whereas, the IrMn layer is there to prevent the anti-parallel axis from flipping irreversibly. MgO is the tunneling barrier. Fabrication of the MgO tunnel barrier requires utmost care, attention to chamber conditions and special fabrication techniques. Since we will be dealing with this extensively in Chapter 6, let us defer detailed discussion till then. Finally, the CoFeB (FM\(_2\)) act as the free layer, meanwhile Ta/Ru/Au constitute the top electrode.

\section*{4.4.3 Testing Magnetic Tunnel Junctions}

It is essential to test the fabricated MTJs to make sure tunneling is established. However, at this stage, it does not make sense to go through rigorous microfabrication steps to fully pattern the sample since, if we fail to achieve tunneling all efforts would be in vain. Thus, a small portion of the sample is patterned into simple pillar structures and measurements were performed using a probe station.

To microfabricate pillar structures, the photoresist was spin coated on to the sample and pillars of varying diameters (10µm-200µm) were defined with photolithography. Subsequently, samples were ion milled up to the bottom electrode and excess photoresist washed off in Acetone. Figure 4.5 is a graphical representation of the microfabrication steps.
Figure 4.5 Graphical representation of microfabrication steps to define MTJs into pillar structures for testing purposes. Includes an image of a real set of pillars captured with a CCD camera mounted on an optical microscope.

A non-linear I-V characteristic is a signature of tunneling in these devices. Thus, we perform the I-V curve and magnetotransport measurements on pillar structures with the two-probe method at room temperature, where the external field is swept in the plane of the sample.

While performing such experiments one needs to take great care to not introduce any voltage spikes, which would result in breaking the junction. This includes grounding yourself. Furthermore, not to apply too large of a biasing current. It is a good practice to start off with a smaller current and make incremental changes. Figure 3.6 shows I-V characteristic curve and magnetotransport measurements performed on MgO based MTJs with the structure, Ta(7)/Ru(20)/Ta(7)/CoFeB(3)/MgO(3)/CoFeB(3)/Ta(7)/Cu(50)/Ru(10). Though MgO MTJs were fabricated with TMR up to 250%, considering rigorous conditions need to be met to form high-quality MgO barriers and given the sensitivity of alumina-based junctions were sufficient for
In this application, we decided to fabricate sensors exclusively with the alumina junctions. However, should the need for better sensitivity arises, we always have the option to switch to the MgO junctions.

Figure 4.6  (a) Non-linear I-V characteristic curve. (b) Illustrates the magnetotransport measurements on the MgO junction performed with the two-probe method, sweeping an external magnetic field in the plane of the sample at room temperature.

4.5 Shape Anisotropy and Linear Response

A linear, hysteresis-free response is desired from sensors. MTJ sensor output is given by Equation 4.1 [88].

$$\Delta V = \left( \frac{1}{2} \right) TMR \cdot I \cdot \left( \frac{RA}{wh} \right) \langle \cos(\theta_f - \theta_p) \rangle$$

(4.1)

As shown in Figure 4.7, $I$ is the Sensor-current, $R$ the Resistance, $A$ the sensor area, $w$ the sensor width, $h$ the sensor height, and $\theta_f(\theta_p)$ the angle between free (pinned) layer magnetization and longitudinal direction.
Figure 4.7 Illustrates MTJ structure with bottom pinned layer magnetization fixed at right angles to the longitudinal direction, meanwhile uniaxial anisotropy easy-axis of the free layer aligned in the same direction, when no sensing field is applied.

If the pinned layer magnetization is fixed in the transverse direction \((\theta_p = \pi/2)\) along with the uniaxial anisotropy hard axis of the free layer, MTJ sensor output can be rewritten as follows.

\[
\Delta V = \left( \frac{1}{2} \right) TMR \cdot I \cdot \left( RA \right) \left( \cos \left( \theta_f - \frac{\pi}{2} \right) \right)
\]

\[
\Rightarrow \Delta V = \left( \frac{1}{2} \right) TMR \cdot I \cdot \left( RA \right) \left( \sin \left( \theta_f \right) \right)
\]

For the case \( w \gg h \), i.e. samples with very large aspect ratios,

\[
\langle \sin \left( \theta_f \right) \rangle = \frac{H_{\text{Ext}} + H_{\text{Coupling}}}{H_{\text{Keff}}}, \text{ where } H_{\text{Ext}} \text{ is the applied external field, and }
\]

\[
H_{\text{Keff}} = H_K + H_{\text{demag}} \cdot H_{\text{Keff}} \text{ is the effective anisotropy field, which includes the free layer shape anisotropy field } \left( H_{\text{demag}} \right) \text{ and uniaxial anisotropy field } \left( H_K \right). H_{\text{Coupling}} \text{ corresponds to the sum of Néel coupling and magnetostatic field coupling between the free and pinned layers. Configuration discussed above results in a linear magnetic}
\]
signal response \( \left( \sin \left( \theta_f \right) \right) \propto H_{Ext} \), thus a linear sensor response. Equation 4.2 expresses sensor response in terms of fields.

\[
\Delta V = \left( \frac{1}{2} \right) TMR \cdot I \cdot \left( \frac{RA}{Wh} \right) \cdot \left( \frac{H_{Ext} + H_{Coupling}}{H_k + H_{demag}} \right)
\]  

(4.2)

### 4.5.1 Microfabrication

As described above, to attain liner hysteresis-free response, pinned layer magnetization and the uniaxial anisotropy hard axis of the free layer need to be engineered in the transverse direction. We achieve this exclusively through shape anisotropy. Steps involved in defining MTJ film stacks into structures with large aspect ratios are discussed hereafter.

1. **Step 1:** We start with MTJ film stacks deposited with magnetron sputtering and define bottom electrodes with positive photolithography. This will protect the bottom electrode region in the shape of dumbbells with photoresist.

2. **Step 2:** Etch the samples down to the substrate with ion milling. This will remove all material from areas unprotected with photoresist while leaving dumbbell shaped regions with a full stack of materials with photoresist on top. Now we can wash away the excess photoresist with N-Methyl-2-pyrrolidone (NMP) or Acetone. NMP is recommended since Acetone tend to leave residue behind due to its high vapor pressure.

3. **Step 3:** Again, with positive lithography, we define sensors with various aspect ratios in the dumbbell-shaped regions realized previously and etch down to the bottom electrode. To control this etching process, we deposit the film stack for the top electrode on a glass substrate (reference). Load both the sample and the reference simultaneously and etch the sample until the glass becomes clear and continue to over-etch for a minute or so to get to the bottom electrode. Figure 4.9 is a graphical representation of the elongated sensor structures along with the various aspect ratios tested in the experiments.
Figure 4.8 Depicts microfabrication steps involved in defining elongated sensor structures.

4. **Step 4:** However, if the top and bottom electrodes were to be shorted by any means, we would lose tunneling and structures would behave
like a mere ohmic device. Thus, top and bottom electrodes were isolated with a SiO2 layer deposited in-situ by sputter deposition at the end of the etching step described above. The excess photoresist was washed away with NMP.

5. **Step 5:** Next we define the top electrode with double layer negative lithography such that everywhere other than the electrode region is protected with photoresist and deposit Ta(10)/ Cu (50)/ Au (100). A subsequent lift-off process is done in NMP.

6. **Step 6:** MNPs would bind specifically to areas with Au, which means to the sensor regions as well as to the contact pads. We could use this area specific binding to our advantage by leaving exposed Au layers only in the sensor regions by depositing an Al layer on top of the contact pads with a process identical to Step 5.

![Figure 4.9](image)

**Figure 4.9** Optical microscope images taken at each step of the microfabrication process along with various aspect ratios tested in the experiments.

### 4.6 Optimizing Annealing Conditions

Optimizing annealing conditions is decisive to properly pin the magnetization of the bottom FM layer to an adjacent AFM layer (for alumina junctions). We employ rapid thermal annealing (RTA) and field cooling to accomplish this. For structures with large aspect ratios, free layer magnetization is stabilized along the long axis. Thus, the magnetization of the bottom electrode should be pinned along the short axis. Here, the sample is placed on a hot plate inside a glove box at the desired temperature.
for a predetermined amount of time. Subsequently, removed from the heat and placed between the poles of a permanent magnet of strength 4kG as quick as possible. The sample should be allowed to cool between the magnetic poles at least the amount of time it was warmed. The external magnetic field should be parallel to the short axis to help freeze the bottom FM layer magnetization in that direction.

![Graphs and Diagram](image)

Figure 4.10 (a) Depicts magneto-transport measurements on alumina-based MTJs defined in pillar structures. Samples have been annealed at 180°C for 2-4min and field cooled in a 4kG magnetic field in the plane of the sample for the same duration sample was warmed. (b) illustrates annealing setup.

Samples have been annealed at various temperatures for different amounts of time. Figure 4.10 exhibit the magneto-transport (R-H) curves obtained for alumina-based junctions, annealed at 180°C for various durations, along with the annealing setup. This is the most favorable temperature out of tested temperatures. As seen in
Figure 4.10 (a), pinning deteriorates gradually with the annealing time. Least rotation in the bottom FM layer in response to the external magnetic field is realized with 2min of annealing time. Please note R-H curves shown in Figure 4.10 are obtained with alumina junctions defined in simple pillar structures. Thus, the response is neither linear nor non-hysteretic. However, clearly indicates the effect of annealing conditions on the quality of pinning achieved.

4.7 Testing Sensors

Testing sensors, in general, comprises of three steps:

1. Detecting sensor response without MNPs.
2. Surface functionalization and attaching MNPs bound to target analytes.
3. Detecting sensor response with MNPs to compare with one without MNPs.

4.7.1 Sensor Response Without Magnetic Nanoparticles

Start with alumina-based junctions defined in various aspect ratios and anneal under the optimum condition and field cool as described above. Ideally, free layer magnetization would stabilize along the long-axis (transverse direction) and the magnetization of the bottom FM is pinned along the short axis (longitudinal direction). To measure, the sensor is placed between the coils such that the sensing field aligns with the short axis and sweep the external magnetic field while sourcing a current, typically about 1μA, through the sensor. Figure 4.11 illustrates the measurement setup.
Figure 4.11 Illustration of measurement setup with sensing field swept parallel to the short axis and a fixed bias field applied along the long axis. Current is sourced with the aid of a KEITHLY 220 programmable current source and the voltage is measured with a KEITHLY 2182A nanovoltmeter.

Here current is sourced with a KEITHLY 220 programmable current source, voltage is measured with KEITHLY 2182A nanovoltmeter, sensing field is controlled by a Kepco BoP 20-20m bipolar power supply. MTJs with proper aspect ratio should manifest a linear non-hysteretic (zero coercivity) signal under these measurement conditions.
Figure 4.12 Compares TMR and coercive fields observed while not applying a DC biasing field with applying a DC biasing field of 400e, 550e and 600e. Stronger biasing fields help reduce the coercivity. In the meantime, slashes TMR by few percentage points.

However, the observed signal deviates from the ideal case due to fluctuations in the free layer [89,90]. Implying, shape anisotropy alone is not enough to fully linearize the sensor. The help of a fixed bias field applied transverse to the sensing field is required [88,91]. Unfortunately, this improvement comes at the expense of decreased sensitivity. With the mask design used in the project, we get access to 30 sensors with the same aspect ratio. Labeled A-H, junctions possess the same width of 6μm and increasing length, up to 176μm. Figure 4.12 compares measurements
performed with and without a biasing field. Without a DC biasing field TMR of 17.1% and coercive field of 13.5Oe is observed. By applying a DC biasing field transverse to the sensing field, coercivity can be reduced dramatically. As illustrated in Figure 4.12, a DC biasing field of 40Oe, 55Oe and 60Oe reduces coercivity to 4Ω, 3Ω, and 2Ω respectively. In the meantime, TMR gets slashed by few percentage points. A happy medium between the TMR and the coercivity can be achieved with a biasing filed of about 50Oe. Absolute linearity and zero coercivity can be achieved, but this comes at a steep decline in TMR. Please note, TMR of devices with various aspect ratios does not vary significantly. However, coercivity decreases gradually with increasing aspect ratios, even without a DC biasing field [92].

4.7.2 Research Design

Before going into the details of the research design it is worthwhile to give a brief introduction into a couple of biological terms used frequently here, antigen and antibody. An antigen is a molecule capable of inducing an immune response in the host organism [93]. An antibody is a protein produced mainly by the plasma cells and used to neutralize pathogens. The immune response can be easily understood by the cartoon in Figure 4.13. Let us consider people in black suits as body cells and the suit itself represents an antigen. Thus, the antigen is like a uniform you wear letting others know you are part of the organization. Everyone in a black suit is recognized as harmless. However, if a pathogen (person in the blue suit) is present, that will be marked with a different antigen, here the blue suit. This blue suit is recognized as foreign, which triggers an immune response. Here balls are thrown to neutralize the person in a blue suit (pathogen), representing the immune response and the balls are analogs to antibodies.
Figure 4.13 Immune response illustrated in a cartoon.

The sensor is designed to detect the early phase emergence of ICAs, autoimmune antibodies against three markers GAD65 [94], IA-2 and insulin. The detection of these antibodies years before the onset of diabetes clinical symptoms will identify individuals at high risk, who may benefit from early therapeutic or preventive intervention. It is estimated that by the time clinical symptoms emerge, 60-80% of beta cells would have already been destroyed.

4.7.2.1 Operational Procedure

Sensor undergoes following modifications prior to being tested for the presence of target analytes:
1. **Step 1**: Covalent attachment of antigens produced from plants (by Fraunhofer team) to MNPs.

2. **Step 2**: Anti-human IgG capture antibody attachment to the sensor surface. These antibodies are complementary to target antibodies.

3. **Step 3**: Add MNPs to the serum and incubate to bind the autoantibodies from the patient and collect MNPs. Note, autoantibodies bind to antigens, not to MNPs. Thus, explaining the need for step 1. Where we attach MNPs with antigens recognized by target autoantibodies.

4. **Step 4**: Add MNPs collected in Step 3 to the sensor surface modified in Step 2. Incubate, wash and measure. MNPs not bound to target analytes.

---

**Figure 4.14** Graphical representation of sensor modification procedure, prior to being tested for the presence of target analytes.
antibodies will not be captured by complementary antibodies on the sensor surface and will be removed upon washing.

Figure 4.14 is a graphical representation of the sensor modification steps prior to being tested for the target analytes. Sensor surface functionalization and MNP synthesis were done by Dr. Aruna Sigdel. Protein synthesis and MNP coating have been performed by our collaborators at Fraunhofer USA. Thus, will not be discussed in detail here.

### 4.7.3 Drop-Dry Test for Detecting Magnetic Nanoparticles

![Figure 4.15 TEM image of as-synthesized (a) 20nm FeO NPs (b) 35nm NPs and (c) SEM image showing selective binding of MNP to sensor region.](image)

Fe$_3$O$_4$ MNPs with 25nm diameter were used in the development of the sensor and details of the synthesis can be found elsewhere [95]. While optimization of the sensor surface functionalization and MNP attachment are in progress, proper operation of the sensor can be verified by a drop dry test. It is essential to obtain magnetic signals only from the sensor regions. MNPs selectively bind to Au surfaces and during
the microfabrication process, we made sure, exposed Au layers are present only in the sensor regions. Thus, allowing MNPs to bind to the sensor area during the selective binding process. Figure 4.15 illustrates TEM images of synthesized MNPs of various sizes along with the SEM image of the MTJ structures after the selective binding process. In Figure 4.15 (c) white dots are the MNPs bound to sensor area and the contrasting dark region is the substrate, demonstrating the success of selective binding mechanism.

Figure 4.16  Illustrates stray field generated opposite to the applied sensing field by MNPs bound to the sensor surface.

To test for the desired output, the sensor response is compared with and without bound MNPs, using the setup shown in Figure 4.11. MNPs bound to the
sensor surface generate a stray magnetic field opposite to the sensing field as depicted in Figure 4.16.

Equations relating magnetic induction \( \vec{B} \) magnetic field strength \( \vec{H} \) and magnetization \( \vec{M} \) can be written as,

\[
\vec{B} = \mu_0 (\vec{H} + \vec{M})
\]
\[
\vec{M} = \chi_m \vec{H}_{app}
\]

(4.3)

Where \( \chi_m \) is the magnetic susceptibility. A spherical superparamagnetic or ferromagnetic nanoparticle in the presence of an external magnetic field will be uniformly magnetized and will give rise to a dipole distribution from an effective dipole moment \( \vec{p} \) [96],

\[
\vec{p} = \vec{M} \left( \frac{4\pi}{3} \right) a^3
\]

(4.4)

Vector potential \( \vec{A} \) is as Equation 4.5,

\[
\vec{A} = \frac{\mu_0}{4\pi} \frac{\vec{m} \times \hat{r}}{|\hat{r}|^2} \left( \vec{B} = \nabla \times \vec{A} \right)
\]

(4.5)

Without any loss of generality, we can orient the dipole such that \( \vec{m} = m\hat{z} \). Since,

\[
\hat{x} = \text{Sin } \theta \text{Cos } \phi \hat{r} + \text{Cos } \theta \text{Cos } \phi \hat{\theta} - \text{Sin } \phi \hat{\phi}
\]

\[
\hat{y} = \text{Sin } \theta \text{Sin } \phi \hat{r} + \text{Cos } \theta \text{Sin } \phi \hat{\theta} + \text{Cos } \phi \hat{\phi}
\]

\[
\hat{z} = \text{Cos } \theta \hat{r} - \text{Sin } \theta \hat{\theta}
\]

\[
\therefore \vec{m} = m\hat{z} \text{ & } \vec{\hat{z}} = \text{Cos } (\theta) \hat{r} - \text{Sin } (\theta) \hat{\theta}
\]

\[
\vec{m} \times \hat{r} = m \left[ \text{Cos } (\theta) \hat{r} - \text{Sin } (\theta) \hat{\theta} \right] \times \hat{r}
\]

\[
\Rightarrow \vec{m} \times \hat{r} = m \text{Sin } (\theta) \hat{\phi}
\]

\[
\vec{A} = \frac{\mu_0}{4\pi} \frac{m \text{Sin } (\theta)}{|\hat{r}|^2} \hat{\phi}
\]
Therefore,
\[
\nabla \times \mathbf{A} = \frac{\mu_0}{4\pi} m \left\{ \frac{\mathbf{r}}{r \sin \theta} \left[ \frac{1}{r} \frac{\partial}{\partial \theta} \left( \frac{\sin^2(\theta)}{r^2} \right) \right] - \frac{\frac{\hat{\theta}}{r} \frac{\partial}{\partial r} \left( \frac{\sin(\theta)}{r^2} \right) }{r} \right\}
\]
\[
\nabla \times \mathbf{A} = \frac{\mu_0}{4\pi} m \left\{ \frac{\mathbf{r}}{r^3} \left[ 2 \cos \theta \hat{\mathbf{r}} + \frac{\hat{\theta}}{r} \sin \theta \right] \right\}
\]
\[
\nabla \times \mathbf{A} = \frac{\mu_0}{4\pi} \left( \frac{m}{r^3} \right) \left[ 2 \cos \theta \hat{\mathbf{r}} + \sin \theta \hat{\theta} \right]
\]
\[
\tilde{H}_r = \left( \frac{1}{r^3} \right) \left( \frac{4}{3} \pi a^3 \right) \left[ 2 \cos(\theta) \hat{r} \right]
\]
\[
\therefore \tilde{\mathbf{M}} = \chi_m \tilde{H}_{\text{app}}
\]

Hence,
\[
\tilde{H}_r = \chi_m \left( \frac{8\pi}{3} \right) \left( \frac{a^3}{r^3} \right) \cos(\theta) \tilde{H}_{\text{app}} \quad \& \quad \tilde{H}_\theta = \chi_m \left( \frac{4\pi}{3} \right) \left( \frac{a^3}{r^3} \right) \sin(\theta) \tilde{H}_{\text{app}} \quad (4.6)
\]

Note $\theta$ refers to the angle between $\mathbf{r}$ and $\tilde{\mathbf{M}}$. Clearly, the effect is limited to the region immediately beneath the nanoparticle and the field distortion due to the nanoparticles fall off as $\left( \frac{1}{r^3} \right)$. Due to this stray field generated by the MNPs, the effective field experienced by the sensor is $H_{\text{app}} - H_{\text{Stray}}$. Therefore, the gradient of the linear portion of the response curve must have a smaller value with MNPs bound to the sensor surface. By comparing the sensor response with and without MNPs one can establish proper operation of the sensor.
Figure 4.17 Compares sensor response with and without MNPs. Response with the MNPs present in the sensor area has a gradient smaller than that without MNPs, verifying the proper operation of the sensor.

Figure 4.17 Compares sensor response before placing nanoparticle on top of the sensor versus after selective binding of MNPs. Signal realized with MNPs bound to the sensor area has a gradient smaller than that of the bare sensor. Thus, we have achieved the desired outcome from the sensor development procedure established here.

4.8 Discussion

The tool developed here is quite powerful, though the surface functionalization is carried out to capture three analytes specific for Type-I diabetes in this project,
range of modification can be done and the number of analytes that can be identified with this sensor is limitless. We are expecting to expand the capabilities of this sensor to detect environmental pollutants in the foreseeable future. In addition to the demonstration of the sensor system, studies will be performed on the selectivity and sensitivity of the system for ICAs. Using ICAs positive and ICAs negative, control experiments will be carried out with different concentration of MNPs present in the tagging solution for the optimum results. Furthermore, optimization will be done with antigen peptides engineered for the maximum binding to ICAs. This will further be benefited by reducing the size of MNPs for better stability and agility in solution. We will also perform studies on the sensor surface modified with anti-human IgG to optimize long-term stability and storage. Microfabrication processes can be modified to include wires adjacent to the sensor, which can be utilized to generate sensing and biasing fields required to measure the response of the sensor. Therefore, I cannot foresee any obvious obstacle that would prevent this system from being transformed into a hand-held device. Fulfilling all the expectations of a POCT system, the potential socioeconomic impact of commercializing the systems discussed here is immense.
Chapter 5

SPIN-ORBIT TORQUE MECHANISM IN HEAVY METAL/ FERROMAGNETIC BILAYERS

5.1 Introduction

Digital data volume in the world is increasing at a rapid rate. In fact, last two years alone produced 90% of the digital data that exists today. As much as 50% of the global population use the internet, consuming colossal amounts of digital content. Thus, the need for fast reliable memory devices is more imminent than ever before. Due to shortcomings of the conventional data storage devices, alternate memory technologies such as magnetic random-access memory (MRAM), resistive RAM (RRAM), phase change RAM (PRAM) are being explored. Among these, MRAMs have the edge due to their superior qualities and ability to seamlessly integrate to the CMOS process. First proposed by Slonczewski [97] and Berger [98], manipulation of the magnetic state of a ferromagnetic (FM) layer, with a spin-polarized current induced torque is called spin-transfer-torque (STT). Knowledge of STT led to spin-transfer-torque MRAMs (STT-MRAM). However, the practicality of STT driven memory devices suffers from having to find a balance between a small junction resistance to allow a large write current and a large magnetoresistance to enable reading. A promising alternate actuation technique, utilizing both spin-polarized currents and static electric fields has emerged. Termed spin-orbit torque (SOT), this scheme is fundamentally different from STT, despite the close resemblance. SOTs in ferromagnetic (FM)/ heavy metal (HM) bilayers have been scrutinized
extensively [99–102]. However, results seem to be strongly dependent on the system probed. In this chapter, we will investigate the validity of a proposed theoretical scenario [103] potentially explaining the reasoning behind discrepancies.

5.1.1 Spin Transfer Torque

![Figure 5.1 Illustrates spin transfer torque mechanism.](image)

As illustrated in Figure 5.1, an unpolarized charge current injected into a ferromagnetic layer with fixed magnetization (FM₁) will be spin-polarized along the magnetization vector (M\text{fixed}). When this spin-polarized current enters the second ferromagnetic layer (FM₂), spin components transverse to the magnetization vector (M\text{free}) are absorbed. Thus, to conserve the angular momentum, a torque is applied on the free layer magnetization, causing it to rotate. Termed spin transfer torque
(STT) [104], this mechanism shall be used to switch the free layer magnetization, provided a sufficiently large charge current is applied.

### 5.1.2 Spin-Orbit Torque

![Figure 5.2](image)

(a) SHE: Electric current through the HM layer generates a spin current via SHE. The spin current exerts an effective torque on the adjacent FM layer via the spin transfer effect. (b) Rashba effect: an electric current through the interface between HM and FM layer experience a Rashba field via the spin-orbit interaction under the structure inversion asymmetry. Through the exchange coupling and spin relaxation, the Rashba effect exerts an effective torque on the FM. Figure reprinted with permission from ref. 99, © 2013 NPG.

One major drawback of STT driven systems is that they require a non-collinear magnetic configuration where the spin-polarized current is generated via a reference FM layer. In contrast, strong SOC of HMs or interfaces can be exploited to create non-equilibrium spin-polarized electrons [105–107]. Which in turn interact with the
magnetization through exchange coupling and manipulates the magnetization for sufficiently large current densities. This scheme which does not require a non-collinear magnetic configuration is called spin-orbit torque (SOT). In commonly used FM/HM bilayers, the origin of SOT is credited to two major sources. Spin Hall effect in the bulk of the HM and the Rashba-Edelstein effect \([108,109]\) (inverse spin Galvanic effect) at the interface.

1. **Spin Hall Effect (SHE):** When a charge current is driven through an HM like Ta or Pt with large spin Hall angle, electrons undergo spin-dependent deflection transverse to the flow due to SOC. This results in the injection of pure spin current into the adjacent FM layer. Spin current absorbed by the FM transfers angular momentum to the magnetization through spin-transfer torque.

2. **Rashba Effect (RE):** Due to lack of inversion symmetry at the interface, electrons flowing in the plane of the sample experience an effective electric field perpendicular to the film plane. In the rest frame of the electron, this electric field transforms into the Rashba magnetic field. Thus, leading to a spin accumulation at the interface, in the plane of the samples, perpendicular to the flow of electrons. Such nonequilibrium spins accumulated at the interface interact with and manipulate the magnetization through exchange coupling \([110]\).

### 5.1.3 Damping Like Torque and Field Like Torque

An electrical current through an FM/HM bilayer exerts two types of torques on the magnetization of the FM. A field-like torque (FLT): \(a\vec{m}\times\vec{\sigma}\) and a damping-like torque (DLT): \(b\vec{m}\times(\vec{\sigma}\times\vec{m})\). Here, \(\vec{m}\) is the magnetization vector and \(\vec{\sigma}\) (collinear with the effective field) is the spin polarization vector, whereas \(a, b\) are parameters related to excitation \([111,112]\). As shown in Figure 5.3, DLT rotates \(\vec{m}\) towards the effective field \(H_{\text{eff}}\), meanwhile FLT induces precession of \(\vec{m}\) about the effective field. RE driven SOT is expected to be dominated by the FLT and SHE driven SOT is
expected to be dominated by the DLT. However, since they both follow the same geometrical form distinguishing the origin of SOT has been a matter of great interest.

Figure 5.3  (a) DLT rotates magnetization towards the effective field while FLT causes magnetization to precess around the effective field. (b) Red arrow corresponds to the DLT and the Blue arrow corresponds to the FLT. Figure reprinted with permission from ref. 112, © 2012 NPG.

5.2 Motivation

Interfacial electronic structures and spin textures, intimately related to materials and fabrication, play a critical role in determining various SOC driven phenomena. The interfacial electronic structures and spin textures are very difficult to characterize directly. Their characteristics are manifested in the SOT behavior and its counterpart spin pumping phenomena. Significant progress has been made in both experiments and theory to understand various SOC driven phenomena. For instance, progress in experimental techniques has allowed researchers to reliably measure DL
and FL torques [113,114]. First principle calculations that include interface SOC and spin memory loss (SML) due to spin-flip scattering have finally explained spin pumping experiments very well [115,116]. However, the final link between SOTs and interfacial electronic structures and spin textures is still missing. Measured DL and FL torques, even for the same material system with different interfaces vary significantly [117,118]. The first-principles calculation suggests the interfacial electronic structures sensitively depend on material systems and strong SOC proximity effect from HM into FM, leading to spin textures that cannot be explained with simple Rashba model [119,120]. Kim, et al. [103] have developed analytical expressions for interface contribution to current-induced SOTs, with the scattering matrix approach, while treating interfacial SOT as a perturbation. Independent of the details of the interface, these expressions provide a generalized view of the influence of the interface on SOTs. In contrast to previous studies, they have concluded, currents in both FM and non-magnetic (NM) layers contribute to the SOTs with additive FLT and canceling DLT. It is worth noting that this model includes a bulk contribution to SOTs excluding the intrinsic contribution from the Berry phase. Work we present here will validate these theoretical studies by investigating the effect of current distributions in HM and FM layers on DL and FL torques by comparing their temperature dependence in NiFe/ Pt and CoFeB/ Pt systems. FM/HM model systems are ideal for the study due to their experimental simplicity and repeatability. In the typical thickness regime, resistivities of the materials chosen for the study are related as: $\rho_{Pt} \sim \rho_{NiFe} << \rho_{CoFeB}$. Hence, we can achieve starkly contrasting current distributions in the bilayer combinations under investigation. Furthermore, the temperature coefficients of resistivity ($\alpha$) for the materials of interest are related as $\alpha_{CoFeB} << \alpha_{NiFe} \sim \alpha_{Pt}$ [121–
123], allowing us to perform comparisons within the structures by studying the temperature dependence of FLT and DLTs.

5.3 Methods

All necessary thin films were fabricated on a wet thermally oxidized Silicon wafer (Si/SiO$_2$) with magnetron sputtering at a base pressure lower than $5 \times 10^{-7}$torr, 4.5mtorr working pressure and 10sccm argon flow. Py(0~10)/Pt(4) and Co$_{40}$Fe$_{40}$B$_{20}$(0~10)/Pt(4) are the specific structures grown for the study. Where numbers in parenthesis indicate film thickness in nanometers. FM layers were grown in a wedge shape with the aid of off-angle sputtering. This would allow us to perform thickness dependence studies without having to grow individual films for each thickness.

Samples need to go through extensive microfabrication process before they are ready to be measured. However, with a carefully designed pattern, one may fabricate structures necessary to perform a variety of measurements. Thus, go through the microfabrication process only once. Figure 5.4 (a) illustrates the pattern designed for this purpose. Figure 5.4 (b) depicts structures used for DC-MOKE measurements. This includes two calibration wires adjacent to the central strip which carries the magnetic sample. An electric current applied through the calibration wires would generate a uniform magnetic field through the sample. Moreover, they allow measurements to be performed while applying a longitudinal or transverse electric current. Pads for central devices are shared to accommodate a maximum number of structures within the area permissible on the sample holder of the cryostat where we performed optical measurements. Hall-bar structures shown in Figure 5.4 (c) shall be used for transport
measurements. Whereas, the structures shown in Figure 5.4 (d) could be used for ST-FMR measurements.

Figure 5.4 (a) Illustrates the pattern designed for measurements including cleaving markers. (b) Devices used for DC-MOKE measurements which include calibration wires. Enables measurements while applying the current in longitudinal or transverse directions. (c) Hall-bar structure for transport measurements. (b) Devices designed for ST-FMR measurements.

5.3.1 Microfabrication

We have adopted an all lift-off process for the microfabrication as outlined below. A bilayer photolithography process has been implemented to achieve an undercut to facilitate the lift-off. Figure 5.5 illustrates samples realized with the microfabrication process.

1. **Step 1:** Use the bilayer lithography process to define the sample region with dimensions $250\,\mu m \times 50\,\mu m$ such that everywhere other than the sample region is protected with photoresist (PR).
2. **Step 2 (Metallization):** Deposit FM films with desired thickness as described above with magnetron sputtering.

3. **Step 3 (Lift-off):** Wash away excess PR in NMP.

4. **Step 4:** Use the bilayer lithography process to define electrodes. A portion of the electrode would coincide with the sample such that the dimension of the exposed magnetic region will now reduce to 50$\mu m \times 50 \mu m$.

5. **Step 5:** Deposit electrodes Ti(1)/ Cu(150)/ Au(100) with magnetron sputtering.

6. **Step 6:** Wash away excess PR in NMP.

![Figure 5.5](image)

Figure 5.5 Depicts optical microscope images of samples realized with the microfabrication process.

### 5.3.2 Magnetic Transport Measurements

Before going through taxing temperature dependent DCMOKE measurements we performed magnetic transport measurements with the aid of a four-probe station to ensure fabricated samples show typical $1/H_{Ext}$ behavior, where $H_{Ext}$ denote the external magnetic field. Samples were tested with d.c. planer Hall measurements (PHE) as outlined in ref. [99]. In the planer Hall geometry shown in Figure 5.6 (a), a charge current is applied parallel to the external magnetic field (xx-direction) and the voltage is measured along the transverse direction (xy-direction) while sweeping an
external magnetic field. Figure 5.6 (b) shows PHE measurements for Py(0~10)/Pt(4) samples, i.e. for varying Py thicknesses. While this measurement in itself does not provide any quantitative insight, it is a qualitative proof to establish the proper formation of sample structures.

Figure 5.6  (a) Shows PHE geometry with current and external magnetic field applied along the longitudinal direction and the voltage measured along the transverse direction while sweeping the field. (b) PHE measurements on Py(0~10)/Pt(4) samples with varying Py thickness.

Subsequent to the sample fabrication and testing, we performed temperature dependent DC-magneto optical Kerr measurements (DC-MOKE) to quantify the DL and FL torques in Py(0~10)/Pt(4) and CoFeB(0~10)/Pt(4) samples. I will discuss in detail the technique and the setup for this measurement in the following section.
5.3.3 Temperature Dependent Magneto-Optical Kerr Measurements

Schematic in Figure 5.7 illustrates the layout of the low-temperature MOKE microscopy set up used for the optical study of SOTs. A 780nm fiber pulsed (TOPTICA) laser with a pulse width ~100fs and repetition rate 80MHz is employed to conduct MOKE measurement and a white light source is used to perform the imaging of samples. All the MOKE measurement is performed inside a Montana Instruments closed cycle magneto-optical cryostation. The red line in Figure 5.7 denotes the laser optical beam path and the grey line denotes the white light beam path. The laser beam first passes through a linear polarizer with the polarization axis aligned along the x-axis to yield a well-defined polarization (p-polarized with the extinction ratio > 10^5:1). It then passes through an 80 (T):20 (R) beam splitter that is used to direct incoherent white light onto samples for the imaging purpose, and then a 10 (T):90 (R) beam splitter that is used to direct the reflected beam from the sample into the detection optics. A waveplate (either half waveplate or quarter waveplate) is mounted before a 50x objective lens to control the polarization of the laser when interacting with samples. The focused spot size is ~1.1μm. Proper selection of the polarization allows us to selectively probe in-plane and out-of-plane magnetization rotations (polar- and quadratic-MOKE, respectively) and their contributions to the overall MOKE signal can be distinguished through magnetic field scans. The reflected laser which is collinear to the incident beam is separated into s and p branches by a polarizing beam splitter with the respective polarizations routed to the two channels of a balanced bridge photodetector (BBP, Nirvana 2007). A half-wave plate rotates the polarization to balance the two channels of the BBP so that there is no signal in the absence of current-driven magnetization reorientation. When the current pulse induces a change of the magnetization along the optical axis (z-axis, i.e. polar-MOKE), the rotation of
the polarization of the reflected light, i.e. Kerr rotation, increases the signal on one channel of the BBP while reducing the other. To further increase the signal to noise ratio of this measurement, we perform the lock-in detection by sending an AC current directly from the Sine Out of the Stanford Research SR830 lock-in amplifier to drive a modulated MOKE signal and then sending the signal output from the BBP into the Stanford Research SR830 lock-in amplifier.

![Figure 5.7 Schematic illustration of the low-temperature MOKE microscopy setup. This setup is place inside a Montana Instruments closed cycle magneto-optical cryostation to enable low-temperature measurements.](image)

### 5.3.4 Temperature Dependent Resistivity Measurements

Temperature-dependent resistivity measurements ($\rho_{xx}$) necessary to derive the current distributions in the HM and FM layer for each sample has been performed in a Quantum Design (Model 6000) Physical Property Measurement System (PPMS) capable of executing measurements in a stable temperature ranging between 5K-300K.
As shown in Figure 5.8 (a), a fixed current of 5mA is applied along the longitudinal direction in the Hall-bar structure and longitudinal voltage is recorded while sweeping the temperature from 5K to 300K. Figure 5.8 (b) represents a resistance (R) Vs temperature (T) graph acquired from PPMS.

![Figure 5.8 (a)](image)

**Figure 5.8** (a) Hall bar structure used for temperature dependent resistivity measurements. A 5mA current is applied along the longitudinal direction and, longitudinal voltage was recorded while varying the temperature between 5K-300K. (b) Temperature-dependent resistance measured for the Py(2.5)/Pt(4) sample.

### 5.4 Results and Analysis

The goal of this study as described earlier is to experimentally substantiate the claim that the current distribution in the FM and HM layer influence the SOTs. Thus, link interfacial electronic structures and spin textures to SOC driven phenomena. Therefore, the first logical step in this study is to establish the current distributions in each layer for two sample structures under investigation.
5.4.1 Current Distributions

R vs T graphs for select FM layer thickness for Py(0~10)/Pt(4) and CoFeB(0~10)/Pt(4) samples have been collected with PPMS. Fitting functions for these graphs were obtained with the following procedure.

1. Considering FM/HM films to be a parallel set of resistors one can write:

\[
\frac{1}{R_{\text{tot}}} = \frac{1}{R_{\text{HM}}} + \left[ \frac{w}{\rho_{\text{FM}}l} \right] t,
\]

where \( R_{\text{tot}} \) is the total resistance, \( R_{\text{HM}} \) the resistance in HM layer, \( \rho_{\text{FM}} \) the resistivity of the FM layer, \( w \) the width of the sample, \( l \) the length of the sample and \( t \) the thickness of the FM layer.

2. Plotting \( 1/R_{\text{tot}} \) vs FM thickness at a given temperature yields a linear relationship. For example, if we were to acquire resistance vs temperature data for samples with different Py thickness but fixed Pt thickness and plot inverse total resistance at a fixed temperature (say 300K) against the Py thickness we can obtain a linear relationship. Thus, the gradient corresponds to the inverse resistance of the HM layer at that temperature.

3. Total temperature range is divided into three segments

\[ 100K \leq T_H \leq 300K, \quad 20K \leq T_M \leq 100K \quad \text{and} \quad 5K \leq T_L \leq 20K, \]

and extract \( R_{\text{HM}} \) for different temperatures within each segment. By plotting \( R_{\text{HM}} \) vs Temperature one can derive a function for the temperature dependence of HM resistance, thus temperature dependent HM resistivity \( \rho_{\text{HM}}(T) \) within a particular temperature range.

4. Since \( \rho_{\text{FM}} = \frac{wT_{\text{FM}}}{l \left( \frac{1}{R_{\text{tot}}} - \frac{1}{R_{\text{FM}}(T)} \right)} \), plotting \( \rho_{\text{FM}} \) as a function of temperature at a given thickness, a function for the temperature dependence of FM resistivity \( \rho_{\text{FM}}(T) \) can be derived.

All necessary fittings have been performed with the aid of MATLAB. Scripts executed to accomplish the tasks are shown in Appendix A. Figure 5.9 (a) illustrates
the temperature dependent resistance data and fitting for Py(t)/Pt(4) sample, with various Py thicknesses. Whereas Figure 5.9 (b) shows data and fitting pertaining to CoFeB(t)/Pt(4) sample.

![Figure 5.9](image_url)

Figure 5.9  (a) Temperature dependent resistance data and fitting for Py(t)/Pt(4) sample with various Py thicknesses. (b) Temperature dependent data and fitting for CoFeB(t)/Pt(4) sample with various CoFeB thicknesses.

Using the dimensions of the Hall-bar structure used for the measurements, length \( l = 270\mu m \), width \( w = 50\mu m \), and data shown in Figure 5.9 we derived the temperature dependent resistivities for two sample structures as depicted in Figure 5.10. Fitting functions for temperature dependent resistivities are detailed in Appendix A.
Figure 5.10 (a) Temperature dependent resistivity for Py(t) / Pt(4) for various Py thicknesses. (b) Temperature dependent resistivity for CoFeB(t) / Pt(4) for various CoFeB thicknesses.

Figure 5.11 Temperature dependent normalized current distributions in Py and Pt layers in the Py(t) / Pt(4) sample.
Assuming a parallel resistor model for the bilayer structures, an input current $I$ get distributed among the HM and FM layers as given in Equation 5.1. Temperature-dependent, normalized current distributions derived thereby for the Py/Pt sample are shown in Figure 5.11. Temperature-dependent normalized current distributions in CoFeB/ Pt sample are shown in Figure 5.12.

\[
I_{HM} = \frac{\rho_{FM}t_{HM}}{\rho_{FM}t_{HM} + \rho_{HM}t_{FM}} I \\
I_{FM} = \frac{\rho_{HM}t_{FM}}{\rho_{FM}t_{HM} + \rho_{HM}t_{FM}} I
\]  

(5.1)
Choice of materials for the study is justified with the current distributions shown in Figure 5.11 and 5.12, i.e. current distributions are starkly different for each bilayer subjected to scrutiny.

5.4.2 Temperature Dependent MOKE Measurements

Our collaborator Prof. Matthew Dotty’s group from the Department of Material Science and Engineering at the University of Delaware performed the temperature dependent MOKE measurements for this project. We followed the scheme proposed by Fan, et al. [124] to perform the temperature dependent DC-MOKE measurements. For normally incident light with linear polarization, the rotation of the polarization angle $\psi(\vec{m})$ due to the magnetization is given by Equation 5.2. Where, $\alpha_{polar}$ and $\beta_{Quadratic}$ are the coefficients for the polar MOKE and quadratic MOKE. The z-direction is perpendicular to the magnetic film plane and x-direction is parallel to the plane of the incident polarization.

$$\psi(\vec{m}) = \alpha_{polar} m_z + \beta_{Quadratic} m_x$$  \hspace{1cm} (5.2)

Defining $\theta_M, \phi_M$ to be the polar and azimuthal angles of magnetization along with $\phi_{pol}$ the angle of the plane of polarization, Equation 5.2 can be rewritten as follows.

$$\psi(\vec{m}) = \alpha_{polar} \cos \theta_M + \frac{1}{2} \beta_{Quadratic} \sin^2 \theta_M \sin \left[ 2(\phi_M - \phi_{pol}) \right]$$  \hspace{1cm} (5.3)

Thus, small current induced rotations of the magnetization about an initial state with $\phi_M = 0, \theta_M = \pi/2$ would cause a Kerr rotation described by Equation 5.4.

$$-\psi(\vec{m}) = -\alpha_{polar} \Delta \theta_M + \beta_{Quadratic} \cos 2\phi_{pol} \Delta \phi_M$$  \hspace{1cm} (5.4)

In accordance with Equation 5.4, Polar MOKE response is independent of $\phi_{pol}$ and quadratic MOKE response vanishes for $\phi_{pol} = 45^\circ$. Hence, light linearly polarized at
$45^\circ$ was used to singularize polar MOKE response. Similarly, if we were to use circularly polarized light, polar MOKE component does not contribute to polarization change. However, the quadratic MOKE component causes the polarization to change from circular to elliptical. Thus, with this approach, one can quantify both polar and quadratic responses with normal incidence light and avoid cumbersome oblique light incidence experiments.

Figure 5.13 Illustrates magnetization reorientation under a current induced FL and DL torques. $\theta_M$ and $\varphi_M$ are the polar and azimuthal angles of magnetization. $h_{\text{SOT}} \vec{m} \times \vec{\sigma}$ and $h_{\text{SOF}} \vec{\sigma}$ represents out of the plane and in-plane effective fields forcing the magnetization reorientation under an applied current. The magnetization of the FM layer is saturated in the plane of the sample ($x'y'$) with an external magnetic field and the current is applied in the same direction.
In-plane \((\varphi_M)\) and out of the plane \((\theta_M)\) rotation of the magnetization in response to current induced SOTs can be parametrized as two orthogonal effective fields \(h_y\) and \(h_z\) as shown in Figure 5.13. These fields, to the first order, for an in-plane magnetized sample can be written in terms of in-plane anisotropy field \((H_{\text{anis}})\), out of plane anisotropy field \((H_{\text{anis},\perp}})\), saturation magnetization \((M_s)\) and applied external magnetic field \((H_{\text{ext}})\) as in Equation 5.5.

\[
\Delta \varphi_M = \frac{h_y}{H_{\text{ext}} + H_{\text{anis}}} \\
\Delta \theta_M = \frac{h_z}{H_{\text{ext}} + H_{\text{anis}} + M_s - H_{\text{anis},\perp}}
\]

(5.5)

It is clear from Equation 5.5 that the quadratic MOKE response is inversely proportional to the external magnetic field \((1/H_{\text{ext}})\). Meanwhile, polar MOKE response is independent of the external magnetic field for \((M_s \gg H_{\text{ext}})\).

Current-induced SOT measurements were performed with the aid of the setup shown in Figure 5.8. An AC current \(I_{\text{ac}} \cos \omega t\) \((I_{\text{ac}} = 15mA, f = 3.4kHz)\) was applied along the \(x'\) direction. An external magnetic field applied along the current direction saturates the magnetization initially. Effective magnetic fields corresponding to the current induced SOTs are eclipsed by a uniform in-plane Oersted field at the center and out of plane Oersted field at the edges of the sample. However, we can disentangle SOTs from the measurements exploiting distinct symmetries of two effects.

### 5.4.3 Damping Like Torque Measurements

For measuring temperature dependent DLT in the range \(10K - 300K\), we employed linearly polarized light at 45° as justified earlier. Out of plane, Oersted field does not change sign upon reversing the magnetization direction in contrast to out of plane.
plane effective SOT field. Hence, by plotting the Kerr signal $\psi (+m_z) + \psi (-m_z)$ one can isolate the out of plane oersted field. Similarly, plotting $\psi (+m_z) - \psi (-m_z)$ yields out of plane effective SOT field. With this method, we still need to go through a fitting process and linear regression to isolate the effective SOT field from Oersted field.

Hence, to simplify the isolation process we chose a different avenue. As shown in Figure 5.5, a calibration wire was added on either side of the central strip carrying the magnetic sample in the structure used for DC-MOKE measurements. If the current is driven through the central strip, signal observed with the MOKE setup contains both the DLT and Oersted field contributions. However, if the current is driven through the calibration wire, this generates an Oersted field at the sample, which we can compute using Ampere’s law as in Equation 5.6. Where $I$ is the current flowing through the wire, $w(50\mu m)$ the width of the sample and $y'(100\mu m)$ is the position of interest measured perpendicular to the current direction. This equation is valid for the sample with thickness $d (\ll w)$.

$$h_{z,oe} = \frac{I}{2\pi w} \ln \frac{y'}{w - y'}$$

(5.6)

While driving the current through the calibration wire say we measure a MOKE response of $V_{cal}$ and the Oersted field generated at the sample center is $h_{z,oe}$.

Similarly, running the current through the sample and probing the center of the sample say we measure a MOKE response of $V_{dl}$ corresponding to a field $h_z$. These parameters can be related as $\frac{V_{cal}}{h_{z,oe}} = \frac{V_{dl}}{h_z}$ yielding the desired effective field corresponding to damping like torque. Figure 5.14 exhibit effective DL field measured for Py(~6)/ Pt(4) and CoFeB(~7)/ Pt(4) samples.
Figure 5.14 (a) Temperature dependent effective DL field measured in the range $10K – 300K$ for Py(~6)/ Pt(4) sample. (b) Temperature dependent effective DL field measured for CoFeB(~7)/ Pt(4).

5.4.4 Field Like Torque Measurements

As justified earlier FLT term is probed with the circularly polarized light. Yet again measured MOKE signal is veiled by the in-plane oersted field contribution. AC current driven through a metal strip attached to the back of the sample can be used to generate an oscillatory in plane Oersted field at the sample. MOKE signal measured under these conditions would help isolate the effective field corresponding to FLT, with a similar approach used above. Nevertheless, Figure 5.15 presents normalized effective field corresponding to FLT for Py(~6)/ Pt(4) and CoFeB(~7)/ Pt(4) samples in the temperature range $10K – 300K$. Please note data has been normalized to the effective field measured at $10K$. This is sufficient for qualitative comparisons.
5.5 Discussion

As discussed in the motivation section Kim, et al. unraveled the influence of the current distributions in the bilayer structure on DLT and FLT. Primary results of the study are found in Figure 5.16, where SOTs are computed for the FM/ NM bilayer structure. Dashed and dotted-dashed lines represent FLTs, while solid lines represent DLTs. As prescribed by these results, the influence of the current distributions in the FM and NM layers on the SOTs can be summarized as follows. Let the current density in the NM layer be $j_N$ and current density in the FM layer be $j_F$. Electrical resistivity in the NM layer be $\rho_{NM}$ and in FM layer be $\rho_{FM}$. Then if,

$$j_N \gg j_F \Rightarrow FLT \sim DLT \left( \rho_{FM} \gg \rho_{HM} \right)$$

$$j_N \sim j_F \Rightarrow FLT \gg DLT \left( \rho_{FM} \sim \rho_{HM} \right)$$

(5.7)

Moreover, if the current in the NM layer generates a DLT given by $DLT \left( j_N \right)$ and the current in the FM layer generate a DLT given by $DLT \left( j_F \right)$. Similarly, the FLT counterparts be $FLT \left( j_N \right)$ and $FLT \left( j_F \right)$. Then, as shown in Equation 5.8 DLTs are
mostly subtractive and the FLTs are mostly additive. Which means the current in the FM layer tends to decrease DLT and not change FLT.

\[
\begin{align*}
DLT(j_N) - DLT(j_F) & \quad j_F \ \text{DECREASE DLT} \\
FLT(j_N) + FLT(j_F) & \quad j_F \ \text{NOT CHANGE FLT}
\end{align*}
\] (5.8)

Figure 5.16 Depicts the influence of current densities in the FM and NM layers on SOTs. Dashed and dotted-dashed lines represent FLTs, while solid lines represent DLTs. Figure reprinted with permission from ref. 103, © 2017 APS.

Figure 5.17 compares the effective DL field normalized to the current in the Pt layer for Py(~6)/Pt(4) sample with the corresponding current distribution. According to the theoretical description, current in the FM layer tends to decrease the DL field. Thus, we should expect the measured DL field to behave opposite to that of FM layer current. As shown in Figure 5.17 current in the Py layer decreases with the temperature. Measured DL field also manifests the same trend, contradicting theoretical predictions.
Figure 5.17  (a) Illustrates normalized current distribution in Py and Pt layers in Py(~6)/Pt(4) sample. (b) Shows effective DL field normalized to the current in the Pt layer measured for Py(~6)/Pt(4) sample.

Figure 5.18  (a) Illustrates normalized current distribution in CoFeB and Pt layers in CoFeB(~7)/Pt(4) sample. (b) Shows effective DL field normalized to the current in the Pt layer measured for CoFeB(~7)/Pt(4) sample.

Similarly, Figure 5.18 compares the effective DL field normalized to the current in the Pt layer for CoFeB(~7)/Pt(4) sample with the corresponding current.
distribution. Yet again measured DL field mimics the current in the CoFeB layer contrary to theoretical prognosis.

What could potentially lead to such a discrepancy between theoretical and experimental results? To answer this question let us take a closer look at the temperature dependent current distributions in the two FM layers probed in this study. If we revisit the current distributions shown in Figure 5.11 and 5.12 it is obvious that there is a stark contrast between the currents in the FM and HM layers in the bilayer structures. To be precise, for the Py(~6)/ Pt (4) sample roughly 69% of the current flows through the Py layer at the room temperature. Whereas, for the CoFeB(~7)/ Pt(4) sample roughly 35% of the current flows through the CoFeB layer at the room temperature. However, within the full temperature range probed (300K-5K), current through the Py layer increases only up to a maximum of 73%. A 4% change compared to the room temperature. Likewise, the current through the CoFeB layer decreases to an absolute minimum of 29%. A 6% change compared to the room temperature.

Considering temperature coefficient of resistance for Py and Pt are comparable to each other, the observed temperature dependence of the current distribution in the Py layer was expected. However, with a substantially small temperature coefficient of resistance as compared to Pt, observed temperature dependence of the current distribution in CoFeB layer defied expectations. To make sure the data and the analysis are reasonable, the spin Hall angle of Pt for both Py(~6)/ Pt (4) and CoFeB(~7)/ Pt(4) samples were computed at 150K with the aid of Equation 5.9. Following parameters were used in the calculations: saturation magnetization ($\mu_0M_S$) of Py(=1T) and CoFeB(=1.6T), width of the sample w(=50μm), DC current through the sample $I_C(=30mA)$, thickness of the Pt layer $t_Pt(=4nm)$ and, finally thickness of the
FM layers $t_{Py}(=5.92\text{nm})$ and $t_{CoFeB}(=6.8\text{nm})$. Spin Hall angle ($\theta_{SH}$) of nearly 10% was obtained for Pt with the Py/Pt sample, meanwhile spin Hall angle of nearly 7% was obtained with CoFeB/Pt sample. Latter value agrees perfectly with the comparable experiments [99,100], meanwhile former is a slight overestimation. However, spin Hall angle estimated for Pt with both the samples are within acceptable range. 

$$\frac{h_{DL}}{J_{Pt}} = \left( \frac{h}{2e} \right) \frac{\theta_{SH}}{\mu_B M_s t_{FM}}$$  \hspace{1cm} (5.9)$$

We were forced to perform cross-comparison between the two systems. As illustrated in Figure 5.19 current flowing through the CoFeB layer is substantially small as compared to the Py layer. Thus, DL field normalized to the current in the Pt layer measured for the CoFeB/Pt sample expected to dominate that of Py/Pt sample. However, we still observed the opposite trend.

Figure 5.19  (a) Normalized current distributions in Py and CoFeB layers. (b) DL fields normalized to the current in the Pt layer, measured for Py/Pt and CoFeB/Pt samples.
Though all available data points to a SOT behavior opposite to what is envisioned by the theoretical studies, with the available information we cannot with confidence affirm or debunk theoretical claims. Thus, experiments were designed to perform temperature dependent MOKE measurements on magnetic samples with perpendicular magnetic anisotropy. Steps involved in designing these experiments are detailed in the next chapter. It is worth noting, work of Kim, et al. predicts an additive FLT and canceling DLT regardless of the nature of the HM used. It will be interesting to see how DLT and FLT would behave when HM is replaced by Ta with a spin Hall angle opposite to that of Pt.
Chapter 6

FUTURE DIRECTION

6.1 Introduction

Chapter 5 detailed the efforts to experimentally validate or debunk recent theoretical studies describing the influence of current distributions on spin-orbital torques (SOTs) in a ferromagnet (FM)/ heavy metal (HM) bilayer structures with temperature dependent DC-MOKE measurements. In this regard, we scrutinized Py and CoFeB, FM layers in contact with Pt, the HM layer. However, experimental data exhibit a trend opposite to what is predicted by the theoretical studies. Thus, further experiments are necessary to realize our goal. FM layers utilized for the aforementioned study possess in plane magnetizations. Hence, extracting SOTs with temperature dependent DC-MOKE measurements for samples with perpendicular magnetic anisotropy (PMA) could shed some light on the observed disagreement between experiments and theory. Therefore, the first logical step for this project is to develop and optimize a magnetic structure with PMA. Optimized PMA structures need to go through microfabrication before measurements. We have already detailed this process in the previous chapter. However, we are also planning to use the PMA samples to perform time-resolved MOKE (TR-MOKE) study of magnetization switching, which requires samples to be defined into CPW structures. In what follows, we will describe in detail, methods to achieve optimized PMA samples. In addition, explain the rationale behind the CPW design and the fabrication steps. We conclude
the chapter with a brief discussion of associated measurement techniques to extract SOTs from PMA samples.

6.2 Perpendicular Magnetic Anisotropy Optimization

Materials with perpendicular magnetic anisotropy (PMA) are of significant interest, especially for storage media. High-density, non-volatile memory devices with high thermal stability can be achieved with these materials. Moreover, magnetization switching shall be realized with low current density for current assisted reading and writing [125,126]. Structures with PMA have been successfully constructed with various material combinations. Co/Pd multilayers (ML) [127], Co/Pt ML [128], Co/Ni ML [129] are some of the most frequently used material combinations. Recently PMA has been demonstrated in ultrathin CoFeB layers sandwiched between Au or Pt [130,131] as well. However, CoFeB/ MgO-based perpendicular MTJs reported by Ikeda, et al. [132] with sufficiently high thermal stability, low switching current seems to be the most promising candidate. In widely used Ta/CoFeB/MgO structure with the Ta buffer layer, PMA is attributed to interfacial anisotropy between MgO and CoFeB. Wang, et al. [133] claimed this interfacial anisotropy is solely responsible for PMA. Meanwhile, a few other studies argued Ta/CoFeB interface either significantly influence or assist PMA [134,135]. Regardless of this ambiguity, Ta/CoFeB/MgO layer structure is a reliable way to produce PMA samples.

6.2.1 Fabrication

Yet again all samples have been deposited directly from respective targets Ta, Co$_{40}$Fe$_{40}$B$_{20}$ and MgO with DC or RF magnetron sputtering. DC-MOKE samples were
fabricated on thermally oxidized Silicon substrates (Si/SiO$_2$), while TR-MOKE samples were fabricated on GaAs substrates.

Obtaining high-quality MgO layers with proper stoichiometry is contingent upon the reduction of residual gas molecules (O$_2$, H$_2$O) inside the chamber, especially H$_2$O. For this purpose, we bake the chamber for at least 36h. Thereafter, use Ta getter to further reduce H$_2$O content [136]. With this method, a base pressure better than $5 \times 10^{-8}$ torr, along with the water partial pressure under $5 \times 10^{-9}$ torr were attained.

Figure 6.1 (a) Microfabrication step 1, defines the thin bottom electrodes. (b) Step 2 defines a region of 205μm×10μm, to deposit the magnetic samples. (c) Step 3 defines the thick top electrodes. (d) Illustrates a single sample to be loaded into the cryostat. (e) Shows sample mount design which fits into the cryostat. Dimensions in the sample structures were determined so that this sample mount could accommodate the samples.
The deposition was followed by a multistep photolithography process to define the samples into desired structures to perform measurements. For DC-MOKE, magnetic and electric transport measurements we used the same pattern introduced in the previous chapter. However, for TR-MOKE measurements we had to pattern the samples into coplanar waveguides (CPW), for which the mask shown in Figure 6.1 was designed. Microfabrication steps are as follows:

1. **Step 1:** Define CPW structures (bottom electrode) as shown in Figure 6.1 (a).

2. **Step 2:** Deposit thin bottom electrodes Ti(1)/ Au(10) with magnetron sputtering and lift-off with NMP.

3. **Step 3:** Define the magnetic sample region of dimensions 205 \( \mu m \times 10 \mu m \) on the central strip (signal line), as shown in Figure 6.1 (b).

4. **Step 4:** Deposit the sample stack Ta/ CoFeB/MgO/SiO\(_2\) with magnetron sputtering and lift-off with NMP.

5. **Step 5:** Define CPW structures (top electrode) as shown in Figure 6.1 (c). some portion of the top electrode will overlap with the magnetic sample such that an exposed magnetic region will now be limited to 5 \( \mu m \times 10 \mu m \).

6. **Step 6:** Deposit top electrode Ti(1)/Cu(600)/Au(100) and lift-off with NMP.

Plenty of details were considered in designing the mask for TR-MOKE samples. In what follows I will outline some of these features. As shown in Figure 6.1 samples are accompanied by a calibration wire. This will help eliminate the Oersted field contribution to the current induced SOTs as described in Chapter 5. Since the largest sample width cryostat can accommodate is 5 mm, cleaving markers were placed within the dimensions 9100 \( \mu m \times 4800 \mu m \), as illustrated in Figure 6.1 (d). Furthermore, SMP connector to feed microwaves only extends out 800 \( \mu m \). So, the
contact between the pin and the signal line is guaranteed by placing the cleaving markers only 600\(\mu m\) away from the signal line. Horizontal and vertically aligned samples fall within a radius of 2.5\(mm\), this will guarantee the ability to probe both samples with the laser since the mobility of the laser spot is limited to a circle of the same radius. The rectangular strip between the samples was included to place and locate the initial laser spot within the permissible range to probe both samples.

### 6.2.2 Optimizing Film Thickness

The interfacial PMA in Ta/CoFeB/MgO trilayer arises from the hybridization between the ferromagnetic atoms (Co and Fe) and the oxygen atoms in MgO [137]. A more recent study suggested that the bottom metallic layer, like Ta also significantly influence the perpendicular anisotropy [138]. We investigated the influence of the thickness of individual layers on the development of PMA. Three sets of samples were fabricated. In each, the thickness of a single layer in the trilayer structure was varied holding the other two constant. Specific structures are, Ta(x)/CoFeB(1.2)/MgO(3), Ta(2)/CoFeB(y)/MgO(3), Ta(2)/CoFeB(1.2)/MgO(z). Moreover, annealing treatments are necessary for the development of PMA. We adopted the annealing conditions, 300\(^\circ\)C for 3min and field cool under a 4kG out-of-plane magnetic field. Even though some recent studies suggest the enhancement of thermal robustness by replacing the Ta with Mo [139], Hf [135] or W [140], we did not explore these avenues. Mainly because for our study thermal stability up to 300\(^\circ\)C is adequate. The PMA of each sample was investigated via the anomalous Hall effect (AHE). Where a dc current is applied along the longitudinal direction (x-direction) in the hall bar structure and transverse voltage (y-direction) was measured while sweeping an out of the plane magnetic field (z-direction). Results are shown in Figure 6.2.
A square shaped hysteresis loop is an indication of good PMA. The optimum thickness range for each layer are summarized as follows: \( t_{Ta} > 1\,nm \), \( 1.5\,nm > t_{CoFeB} > 0.8\,nm \) and \( t_{MgO} > 1.1\,nm \). We therefore choose the structure Ta(3)/CoFeB(1)/MgO(2) for the study. Further details and applications of this study are reported in our publication [141].

### 6.3 Spin-Orbit Torque Measurements

In samples with PMA, where magnetization vector is aligned perpendicular to the sample plane, polar MOKE response is much stronger than the quadratic MOKE.
response \( \alpha_{\text{Polar}} \gg \beta_{\text{Quadratic}} \). Thus, both damping like torque (DLT) and field like torque (FLT) are determined with the aid of polar MOKE signal in two different geometries illustrated in Figure 6.3.

Figure 6.3 (a) Illustrates configuration to measure effective field due to DLT, where the external magnetic field is in the same direction as current. Hence, this is called the parallel configuration. (b) Depicts the configuration to measure FLT, where the external magnetic field is applied perpendicular to the current direction. This is called the perpendicular configuration.

Figure 6.3 (a) depicts the parallel configuration where the current is applied along the \( x \) directions and the external magnetic field is applied in the \( x-z \) plane such that, the azimuthal angle of the field \( \varphi_H = 0 \) and the polar angle \( \theta_H = 85^\circ \). In this case, the effective field due to DLT \( H_d \sim \vec{M} \times \vec{\sigma} \) is solely responsible for the change in polar angle of the magnetization. Where \( \vec{M} \) denotes the magnetization and \( \vec{\sigma} \) represents the spin polarization vector. Figure 6.3 (b) illustrates the perpendicular
configuration, where the current is applied along the $x$ directions and the external magnetic field is applied in the $y - z$ plane. Here the azimuthal angle of the external magnetic field $\varphi_H = 90^\circ$ and the polar angle of the external field $\theta_H = 85^\circ$. Effective magnetic field due to FLT, $H_f (\hat{\sigma})$ alone reorients the polar angle of magnetization in this case. Therefore, DLT and FLT can be quantified by only measuring the polar signal in two different configurations.

We are planning to perform a temperature-dependent study similar to the one reported in the previous chapter on the samples with PMA. Hopefully, these results would help resolve the conflict of signs observed between experimental and theoretical studies reporting the influence of current distributions on SOTs. Furthermore, PMA samples will be used to study the dynamics of magnetization switching with pump-probe spectroscopy.
REFERENCES


(n.d.).


Appendix A

MATLAB SCRIPTS TO EXECUTE FITTINGS AND FITTING FUNCTIONS

MATLAB code to fit resistance data for Py(t)/Pt(4) Samples:

```matlab
%%----Resistivity comparison Py(t)/Pt----------------------------------------
HiT=linspace(300,105,40);
MiT=linspace(100,25,16);
LoT=linspace(20,5,4);
RhoPtH=47.252+0.1126*HiT-4.1904e-004*HiT.^2+5.0570e-007*HiT.^3;
RhoPtI=50.022+0.0127*MiT+7.8370e-004*MiT.^2-4.3015e-006*MiT.^3;
RhoPtL=50.7037-0.0174*LoT+6.9630e-004*LoT.^2-2.4681e-017*LoT.^3;
figure
PtHwPy=plot(HiT,RhoPtH,'ro');
saveas(PtHwPy,'PtHwPy.fig');
figure
PtIwPy=plot(MiT,RhoPtI,'ro');
saveas(PtIwPy,'PtIwPy.fig');
figure
PtLwPy=plot(LoT,RhoPtL,'ro');
saveas(PtLwPy,'PtLwPy.fig');
HiTFM=linspace(300,105,40);
MiTFM=linspace(100,25,16);
LoTFM=linspace(20,5,4);

%%---------------------2.51nm-----------------------------------------------
RhoPyH_2=28.37-0.005267*HiTFM+0.000178*(HiTFM.^2)-1.733e-007*(HiTFM.^3);
RhoPyI_2=28.34-0.01166*MiTFM+0.0003299*MiTFM.^2-1.024e-006*(MiTFM.^3);
RhoPyL_2=28.16+0.004333*LoTFM-0.0006*LoTFM.^2+2.667e-005*LoTFM.^3;
figure
PyH_2=plot(HiTFM,RhoPyH_2,'r^');
saveas(PyH_2,'PyH_2.fig');
figure
PyI_2=plot(MiTFM,RhoPyI_2,'r^');
saveas(PyI_2,'PyI_2.fig');
figure
```

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PyL_2=plot(LoTFM,RhoPyL_2,'r^');
saveas(PyL_2,'PyL_2.fig');
%------------------------------------------------------------------------

%------------------------4.14nm-----------------------------------------
RhoPyH_3=28.64+0.001062*HiTFM+0.0001414*(HiTFM.^2)-1.333e-007*(HiTFM.^3);
RhoPyI_3=28.78-0.007643*MiTFM+0.0001414*(HiTFM.^2)-8.323e-007*(HiTFM.^3);
RhoPyL_3=28.69+0.0006667*LoTFM-0.0002*LoTFM.^2+1.333e-005*LoTFM.^3;
figure
PyH_3=plot(HiTFM,RhoPyH_3,'r^');
saveas(PyH_3,'PyH_3.fig');
figure
PyI_3=plot(MiTFM,RhoPyI_3,'r^');
saveas(PyI_3,'PyI_3.fig');
figure
PyL_3=plot(LoTFM,RhoPyL_3,'r^');
saveas(PyL_3,'PyL_3.fig');
%------------------------------------------------------------------------

%------------------------5.92nm-----------------------------------------
RhoPyH=28.53+0.001317*HiTFM+0.00014*(HiTFM.^2)-1.267e-007*(HiTFM.^3);
RhoPyI=28.68-0.007985*MiTFM+0.0002881*MiTFM.^2-8.352*10^(-7)*(MiTFM.^3);
RhoPyL=28.59+0.0006667*LoTFM-0.0002*LoTFM.^2+1.333e-005*LoTFM.^3;
figure
PyH=plot(HiTFM,RhoPyH,'r^');
saveas(PyH,'PyH.fig');
figure
PyI=plot(MiTFM,RhoPyI,'r^');
saveas(PyI,'PyI.fig');
figure
PyL=plot(LoTFM,RhoPyL,'r^');
saveas(PyL,'PyL.fig');
%------------------------------------------------------------------------

%------------------------7.63nm-----------------------------------------
RhoPyH_5=28.53-0.003271*HiTFM+0.0001654*(HiTFM.^2)-1.6e-007*(HiTFM.^3);
RhoPyI_5=28.52-0.00976*MiTFM+0.0003086*MiTFM.^2-9.327*10^(-7)*(MiTFM.^3);
RhoPyL_5=28.37+0.005667*LoTFM-0.0004*LoTFM.^2+1.333e-005*LoTFM.^3;
Fitting functions obtained in each temperature range is as follows:

Pt Fitting Functions:

High Temperature (300K-100K)
\[ \rho(T) = 47.252 + 0.1126 T_H - (4.19 \times 10^{-4}) T_H^2 + (5.06 \times 10^{-5}) T_H^3 \]

Intermediate Temperature (100K-20K)
\[ \rho(T) = 50.022 + 0.0127 T_I + (7.84 \times 10^{-4}) T_I^2 - (4.30 \times 10^{-5}) T_I^3 \]

Low Temperature (<20K)
\[ \rho(T) = 50.704 - 0.017 T_L + (6.963 \times 10^{-4}) T_L^2 - (2.468 \times 10^{-5}) T_L^3 \]

Py Fitting Functions:

High Temperature (300K-100K)
\[ \rho(T) = 28.37 - 0.00527 T_H + 0.00018 T_H^2 - (1.733 \times 10^{-3}) T_H^3 \rightarrow t_r (2.51 nm) \]
\[ \rho(T) = 28.64 + 0.00106 T_H + 0.00014 T_H^2 - (1.333 \times 10^{-3}) T_H^3 \rightarrow t_r (4.14 nm) \]
\[ \rho(T) = 28.53 + 0.00132 T_H + 0.00014 T_H^2 - (1.267 \times 10^{-3}) T_H^3 \rightarrow t_r (5.92 nm) \]
\[ \rho(T) = 28.53 - 0.00327 T_H + 0.00017 T_H^2 - (1.600 \times 10^{-3}) T_H^3 \rightarrow t_r (7.63 nm) \]

Intermediate Temperature (100K-20K)
\[ \rho(T) = 28.34 - 0.0117 T_I + 0.00033 T_I^2 - (1.024 \times 10^{-3}) T_I^3 \rightarrow t_r (2.51 nm) \]
\[ \rho(T) = 28.78 - 0.0076 T_I + 0.00028 T_I^2 - (8.323 \times 10^{-4}) T_I^3 \rightarrow t_r (4.14 nm) \]
\[ \rho(T) = 28.68 - 0.0080 T_I + 0.00029 T_I^2 - (8.352 \times 10^{-4}) T_I^3 \rightarrow t_r (5.92 nm) \]
\[ \rho(T) = 28.52 - 0.0098 T_I + 0.00031 T_I^2 - (9.327 \times 10^{-4}) T_I^3 \rightarrow t_r (7.63 nm) \]
Low Temperature (<20K)
\[ \rho(T) = 28.16 + 0.0043TL - 0.0006TL + (2.667 \times 10^{-1}) TL \rightarrow t_o \ (2.51nm) \]
\[ \rho(T) = 28.69 + 0.0007TL - 0.0002TL + (1.333 \times 10^{-1}) TL \rightarrow t_o \ (4.14nm) \]
\[ \rho(T) = 28.59 + 0.0007TL - 0.0002TL + (1.333 \times 10^{-1}) TL \rightarrow t_o \ (5.92nm) \]
\[ \rho(T) = 28.37 + 0.0057TL - 0.0004TL + (1.333 \times 10^{-1}) TL \rightarrow t_o \ (7.63nm) \]

MATLAB code to fit resistance data for CoFeB(t)/Pt(4) samples:

```matlab
clear all;
close all;
clc;

%----Resistivity comparison CoFeB(t)/Pt---------------------------------------
TH=linspace(300,105,41);
TI=linspace(100,25,16);
TL=linspace(20,5,4);
RhoPtHCFB=27.918+0.0350*TH+2.3422e-005*TH.^2-6.9459e-008*TH.^3;
RhoPtICFB=28.467-0.0024*TI+6.1889e-004*TI.^2-2.8252e-006*(TI.^3);
RhoPtLCFB=28.622-0.0067*TL+4.4444e-004*TL.^2-1.0259e-017*TL.^3;
figure
PtHwCFB=plot(TH,RhoPtHCFB,'bo');
saveas(PtHwCFB,'PtHwCFB.fig');
figure
PtIwCFB=plot(TI,RhoPtICFB,'bo');
saveas(PtIwCFB,'PtIwCFB.fig');
figure
PtLwCFB=plot(TL,RhoPtLCFB,'bo');
saveas(PtLwCFB,'PtLwCFB.fig');

%---------------------3.1nm---------------------------------------------------
RhoCFBH_2=119.5+0.05805*TH-0.0003815*(TH.^2)+7.337e-007*(TH.^3);
RhoCFBI_2=124.5+0.001529*TI-0.0004533*TI.^2+2.049*10^(-6)*(TI.^3);
RhoCFBL_2=123.8+0.1804*TL-0.01566*TL.^2+0.0004107*TL.^3;
figure
RhoCFBH_2=plot(TH,RhoCFBH_2,'r^');
saveas(RhoCFBH_2,'RhoCFBH_2.fig');
figure
RhoCFBI_2=plot(TI,RhoCFBI_2,'r^');
saveas(RhoCFBI_2,'RhoCFBI_2.fig');
figure
RhoCFBL_2=plot(TL,RhoCFBL_2,'r^');
```

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saveas(RhoCFBH_2,'RhoCFBH_2.fig');
%-------------------------------------------------------------------------------------------------

%---------------------5.88nm-----------------------------------------------
RhoCFBH_3=103.8+0.1178*TH-0.0003205*(TH.^2)+4.833e-007*(TH.^3);
RhoCFBI_3=107.3+0.02754*TI+0.0005315*TI.^2-2.553*10^(-6)*(TI.^3);
RhoCFBL_3=108+0.01907*TL-0.00286*TL.^2+0.0001213*TL.^3;
figure
RhoCFBH_3=plot(TH,RhoCFBH_3,'r^');
saveas(RhoCFBH_3,'RhoCFBH_3.fig');
figure
RhoCFBI_3=plot(TI,RhoCFBI_3,'r^');
saveas(RhoCFBI_3,'RhoCFBI_3.fig');
figure
RhoCFBL_3=plot(TL,RhoCFBL_3,'r^');
saveas(RhoCFBL_3,'RhoCFBL_3.fig');
%-------------------------------------------------------------------------------------------------

%---------------------6.8nm-----------------------------------------------
RhoCFBH_4=112.3+0.08354*TH-0.000336*(TH.^2)+5.797e-007*(TH.^3);
RhoCFBI_4=116.1+0.02207*TI-0.0001394*TI.^2+8.878e-007*(TI.^3);
RhoCFBL_4=116.6+0.0179*TL-0.00268*TL.^2+0.0001*TL.^3;
figure
RhoCFBH_4=plot(TH,RhoCFBH_4,'r^');
saveas(RhoCFBH_4,'RhoCFBH_4.fig');
figure
RhoCFBI_4=plot(TI,RhoCFBI_4,'r^');
saveas(RhoCFBI_4,'RhoCFBI_4.fig');
figure
RhoCFBL_4=plot(TL,RhoCFBL_4,'r^');
saveas(RhoCFBL_4,'RhoCFBL_4.fig');
%-------------------------------------------------------------------------------------------------

%---------------------7.8nm-----------------------------------------------
RhoCFBH_5=118.3+0.03693*TH-0.0001974*(TH.^2)+4.013e-007*(TH.^3);
RhoCFBI_5=121.2+0.003095*TI-0.0002183*TI.^2+1.172e-006*(TI.^3);
RhoCFBL_5=121.3-0.002933*TL-0.00106*TL.^2+4.933e-005*TL.^3;
figure
RhoCFBH_5=plot(TH,RhoCFBH_5,'r^');
saveas(RhoCFBH_5,'RhoCFBH_5.fig');
figure
RhoCFBI_5=plot(TI,RhoCFBI_5,'r^');
saveas(RhoCFBI_5,'RhoCFBI_5.fig');
Fitting functions obtained in each temperature range is as follows:

Pt Fitting Functions:

**High Temperature (300K-100K)**
\[ \rho(T) = 27.918 + 0.035TH + (2.34 \times 10^3)TH^2 - (6.95 \times 10^6)TH^3 \]

**Intermediate Temperature (100K-20K)**
\[ \rho(T) = 28.467 - 0.00024TI + (6.19 \times 10^3)TI^2 - (2.83 \times 10^6)TI^3 \]

**Low Temperature (<20K)**
\[ \rho(T) = 28.622 - 0.0067TL + (4.444 \times 10^3)TL - (1.026 \times 10^6)TL^3 \]

CoFeB Fitting Functions:

**High Temperature (300K-100K)**
\[ \rho(T) = 119.5 + 0.05805TH - 0.00038TH^2 + (7.337 \times 10^3)TH^3 \]
\[ \rightarrow t_n (3.1nm) \]
\[ \rho(T) = 103.8 + 0.11780TH - 0.00032TH^2 + (4.833 \times 10^3)TH^3 \]
\[ \rightarrow t_n (5.8nm) \]
\[ \rho(T) = 112.3 + 0.08354TH - 0.00034TH^2 + (5.797 \times 10^3)TH^3 \]
\[ \rightarrow t_n (6.8nm) \]
\[ \rho(T) = 118.3 + 0.03693TH - 0.00020TH^2 + (4.013 \times 10^3)TH^3 \]
\[ \rightarrow t_n (7.8nm) \]

**Intermediate Temperature (100K-20K)**
\[ \rho(T) = 124.5 + 0.1804TI - 0.01566TI^2 + 0.00041TI^3 \]
\[ \rightarrow t_n (3.1nm) \]
\[ \rho(T) = 107.3 + 0.0191TI - 0.00286TI^2 + 0.000121TI^3 \]
\[ \rightarrow t_n (5.8nm) \]
\[ \rho(T) = 116.1 + 0.0179TI - 0.00268TI^2 + 0.000100TI^3 \]
\[ \rightarrow t_n (6.8nm) \]
\[ \rho(T) = 121.2 - 0.0029TI - 0.00106TI^2 + (4.933 \times 10^3)TI^3 \]
\[ \rightarrow t_n (7.8nm) \]

**Low Temperature (<20K)**
\[ \rho(T) = 123.8 + 0.1804TI - 0.01566TI^2 + 0.00041TI^3 \]
\[ \rightarrow t_n (3.1nm) \]
\[ \rho(T) = 108.0 + 0.0191TI - 0.00286TI^2 + 0.000121TI^3 \]
\[ \rightarrow t_n (5.8nm) \]
\[ \rho(T) = 116.6 + 0.0179TI - 0.00268TI^2 + 0.000100TI^3 \]
\[ \rightarrow t_n (6.8nm) \]
\[ \rho(T) = 121.3 - 0.0029TI - 0.00106TI^2 + (4.933 \times 10^3)TI^3 \]
\[ \rightarrow t_n (7.8nm) \]
Appendix B

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Appendix C

PUBLICATIONS

Parts of this thesis work has been published in peer-reviewed journals. While some manuscripts are still under review, following provides a detailed account of already published or accepted papers.
Thickness dependence of anomalously large Nernst coefficient and longitudinal spin Seebeck effect in ferromagnetic Ni$_x$Fe$_{100-x}$ films

Harshad Kannan$^1$, Xin Fan$^2$, Halise Celik$^3$, Xiaowen Han$^4$ & John G. Xiao$^5$

Spin Seebeck effect (SSE) measured for metallic ferromagnetic thin films in commonly used longitudinal configuration contains the contribution from anomalous Nernst effect (ANE). The ANE is considered to arise from the bulk of the ferromagnet (FM) and the proximity-induced FM boundary layer. We fabricate a FM alloy with zero Nernst coefficient to mitigate the ANE contamination of SSE and insert a thin layer of Cu to separate the heavy metal (HIM) from the FM to avoid the proximity contribution. These modifications to the experiment should permit complete isolation of SSE from ANE in the longitudinal configuration. However, further thickness dependence studies and careful analysis of the results revealed, ANE contribution of the isolated FM alloy is twofold, surface and bulk. Both surface and bulk contributions, whose magnitudes are comparable to that of the SSE, can be modified by the neighboring layer. Hence, surface contribution to the ANE in FM metals is an important effect that needs to be considered.

Spintronics, which deploys the spin, in addition to or sometimes in place of the charge of the electron, has exhibited rich physics and industrial potential in the past two decades. Intensive research has been carried out investigating the interaction among spin and magnetic field$^6$, electric current$^{2,7}$, electromagnetic waves$^8$ and recently temperature gradient$^9$. A temperature gradient in a ferromagnetic metal generates a spin chemical potential splitting between spin up and spin down electrons, owing to the spin dependent density of states$^7$. This leads to a spin current with no net charge flow in an open circuit, and is called the Spin Seebeck effect. First reported in ferromagnetic metals$^7$, SSE was later found to be present in ferromagnetic semiconductors$^{9,10}$ and insulators$^{11,12}$. The inverse Spin Hall Effect (ISHE)$^{13}$ is a common tool employed to measure SSE, where a FM, for example Pt, placed
Quantifying angular dependence of spin-orbit torques in Ta/CoFeB/MgO trilayers with perpendicular magnetic anisotropy

Yunpeng Chen,1 Halise Celik,1 Tao Wang,1 Harsha Kannan,1 Lyza N. Krivorotov,2 and John Q. Xiao1
1Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, USA
2Department of Physics and Astronomy, University of California, Irvine, California 92697, USA
(Received 25 September 2016; revised manuscript received 18 January 2017; published 7 April 2017)

The spin-orbit interactions in heavy-metal/ferromagnet heterostructures have attracted considerable attention because they provide an efficient way to manipulate the magnetization with strong current-driven spin-orbit torques (SOTs) via the spin Hall effect in the heavy metal or Rashba effect due to the symmetry breaking at the interface. Theoretical calculations predict no dependence of the SOTs on the out-of-plane angle of magnetization due to spin Hall effect, but Rashba effect induces a nontrivial angular dependence of SOTs. Quantitative measurements with adiabatic harmonic Hall technique have observed the angular dependence in Ta/CoFeB/MgO or Pt/Co/Au with perpendicular magnetic anisotropy. However, this method is complicated because the signal consists of both anomalous and planar Hall contributions. In addition, the fitting of the measurement data is sensitive to the fitting parameters, particularly to the perpendicular anisotropy, in a certain angle region (40°–70°). To avoid this uncertainty, we have developed a scheme to quantify the angular dependence of SOTs based on the magneto-optic Kerr effect with field calibration. Without fitting procedures, we precisely determine the SOTs and their angle dependence on the magnetization orientation. We observe a strong angular dependence that is different from the previous experimental observations. Based on this strong dependence, we conclude that a Rashba effect at the same interface, that is responsible for the perpendicular magnetic anisotropy, is the dominant mechanism for the current-driven SOTs in this system.

DOI: 10.1103/PhysRevB.95.144405

I. INTRODUCTION

In-plane current in heavy metal (HM)/ferromagnet (FM)/metal-oxide (MOX) trilayers generates spin-orbit torques (SOTs) that enable an efficient method to control the magnetization of FM layers. The spin-orbit driven magnetic dynamics such as magnetic reversal [1–6], high-frequency oscillation [7–11], domain-wall motion [12–14] and skyrmion manipulation [15,16] have been demonstrated and may find critical applications in magnetic memory and logic devices. In a typical structure depicted in Fig. 1, the charge current along the x direction generates dampinglike torque of $\mathbf{M} \times (\mathbf{M} \times \hat{x})$ [17] and fieldlike torque $\mathbf{M} \times \hat{x}$ [18] on the FM layer. The dampinglike torque also refers to Słonczewski-like torque or spin-transfer torque that describes the transfer of respectively. Because $\mathbf{H}_J$ and $\mathbf{H}_K$ have different dependence on magnetization vector $\mathbf{M}$, one can separately detect them in designed experiment configurations.

Two mechanisms have been proposed to explain the generation of spin-orbit torques: the spin Hall effect [1,4,8,23–27] (SHE) in the bulk materials with strong spin-orbit interaction and the Rashba-Edelstein effects [14,18,27–30] due to the interfacial spin-orbit coupling. Much effort has been dedicated to identify the dominant mechanism of the SOTs, because, phenomenologically, both the SHE and the Rashba effect cause the torques with the same expressions. Although it has been pointed out that the dampinglike and fieldlike torques are dominated by the SHE and the Rashba effect, respectively [31–33], each torque contains the contributions from both
Vector-Resolved Magnetooptic Kerr Effect
Measurements of Spin–Orbit Torque

Halise Celik¹, Harsha Kannan², Tao Wang¹, Alex R. Mellnik²,³, Xin Fan⁴, Xinran Zhou⁵, Rasoul Barri¹, Daniel C. Ralph², Matthew F. Doty⁴,⁵, Virginia O. Lorenz⁴, and John Q. Xiao¹

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⁶Department of Physics, University of Illinois at Urbana–Champaign, Urbana, IL 61801 USA

We demonstrate simultaneous detection of current-driven dampinglike and fieldlike spin–orbit torques in heavy metal/ferrimagnetic metal bilayers by measuring all three magnetization components \(m_x\), \(m_y\), and \(m_z\) using a vector-resolved magnetooptic Kerr effect (MOKE) technique based on quadrant detection. We investigate the magnitude and direction of spin–orbit torques in a series of platinum/ferromagnetic samples, finding good agreement with results obtained via polar and quadrature MOKE measurements without quadrant detection.

Index Terms—XX

I. INTRODUCTION

SPIN–ORBIT coupling-driven phenomena such as the spin Hall effect (SHE) [1] and Rashba effect [2] enable manipulation of magnetization via electric current. By using electric current to control the magnetization of nanoscale elements, it is possible to efficiently integrate magnetic functionalities into electronic circuits [3] and accelerate the technological development of high-performance and high-density magnetic memory devices [4]–[11].

In heavy metal (HM)/ferromagnetic metal (FM) bilayers, an electric current will generate dampinglike spin–orbit torque (DT) and fieldlike spin–orbit torque (FT), which will change the magnetization direction. In order to quantify the magnitude and the direction of magnetization reorientation due to DT and FT generated by the SHE and Rashba effects, electrical measurement techniques such as the anomalous Hall balanced detector. Such MOKE techniques do not suffer from electrical artifacts, and for high-sensitivity measurements, potential optical artifacts such as reflectivity changes can be separated from the MOKE signal harmonically [16].

In ferromagnetic thin films, there have been several studies to determine the magnetization components vectorially [17], [18]. For example, Ding et al. [19] proposed a method to distinguish the pure longitudinal and polar Kerr contributions via two separate measurements, interchanging the positions of a light source and a detector. Yang and Scheinfein [20] showed the detection of three magnetization components by changing the different relative orientations of the optical devices: polarizer, modulator, and analyzer. As an alternative that does not require changing the position of optical elements or data analysis to separate overlapping signals from different vector components, Keatley et al. [21] used a scanning Kerr...