SPIN DEPENDENT TRANSPORT AND SPIN TRANSFER IN NANOCONSTRICTIONS AND CURRENT CONFINED NANOMAGNETS

A Dissertation

Presented to the Faculty of the Graduate School of Cornell University in Partial Fulfillment of the Requirements for the Degree of Doctor of Philosophy

by

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In this thesis, I have employed point contact spectroscopy to determine the nature of electron transport across constrained domain walls in a ferromagnetic nanocontact and to uncover the relationship between ballisticity of electron transport and domain wall magnetoresistance. In the range of hole sizes studied (from 10 to 3 nm) the resulting magnetoresistance was found to be less than 0.5% and one that increases with decreasing contact size.

I have used point contacts as local probes, to study the spin dependent transport across Ferromagnet/Normal Metal/Ferromagnet (FM/NM/FM) trilayers as well as the consequences of localized spin polarized current injection into a nano-magnet on spin angular momentum transfer and high frequency magnetization dynamics. I have demonstrated that absolute values for spin transfer switching critical currents are reduced in this new geometry as compared to uniform current injection. I have also performed micromagnetic simulations to determine the evolution of magnetization under the application of magnetic fields and currents to gain more insights into experimental results.

I have used Scanning Transmission Electron Microscopy (STEM), X-Ray Photoemission Spectroscopy (XPS) and Electron Energy Loss Spectroscopy (EELS) techniques to characterize the interfacial mixing and oxygen diffusion in the metal-
lic multilayers of interest. I have shown that the $Ta/CuO_x$ bilayer structure provides a smooth substrate by improving interfacial roughness due to grain boundary diffusion of oxygen and reaction with Ta that fills in the grain boundary gaps in Cu. Analysis of the $Py/AlO_x$ interface proved a strong oxidation passivation on the Py surface by Al coating accompanied by Fe segregation into the alumina. I have utilized the characterization results to design a new nanomagnet whose sidewalls are protected from adventitious sidewall oxide layers and yields improved device performance.

The oxide layers that naturally develop at the sidewalls of Py nanomagnets cause an enhancement in magnetic damping especially for temperatures below the blocking temperature of the AFM layer ($\leq 40K$). Studies with pillars protected by Al coating and ones with more NiO coating ($\sim 2.5nm$) shed light onto the role of surface oxides in determining temperature dependent behaviour of both spin torque and field driven switching characteristics.
Ozhan Ozatay was born in Istanbul, Turkey on April 26th 1978. He enjoyed most of his childhood spending a lot of time with his grandmother, playing cards and chatting about life in general. At around the same time as the unexpected loss of this beloved person, he started struggling with the real world. He spent seven years in one of the most prestigious schools in Istanbul, Kadikoy Anatolian High School to master English as a foreign language, math and science. He graduated in 1996 with a valedictorian.

In 1989, he got interested in acting and joined the theater club to act in a musical play Midas’ Ears by Gungor Dilmen. This performance received the first prize in an acting competition between high schools in the Marmara Region of Turkey. Then his extra-curricular interests shifted to classical music and he started taking piano lessons in 1990. He performed in two recitals playing Chopin, Bach, Beethoven and Mozart. He had the greatest honor in getting a chance to work with Sevin Erel, a well-respected pianist in 1993. He not only developed a lot more musical skills with the aid of his teacher but also received a lot of moral support and advise about life from this very special person which he will appreciate for the rest of his life.

After graduation from high school, he was lucky enough to be admitted to Bogazici University Electrical and Electronics Engineering department, ranking 152nd out of a million candidates in the university entrance exam. He was most interested in learning the working principles of solid state electronic devices but soon he realized that a good understanding of any electronic device is only possible with a strong physics background. For that reason he decided to do a double major with physics. He got his bachelors degree from both departments in year 2000.
Then he decided to take a big leap by going overseas to suffer from the cold weather in Ithaca, NY. He started his graduate studies in Cornell in 2001 with the idea that he could work on anything but magnetism. After joining Buhrman group, it didn’t take him very long to realize that magnetism is actually a fascinating field, especially in the nano-domain. After having a very enjoyable time in Ithaca, he is planning to take a post-doc position somewhere warmer.
To him,

who without any doubt,

knows it all
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CHAPTER 1

INTRODUCTION

*Nanomagnetism* is the study of novel magnetic phenomena specific to magnetic structures geometrically confined to dimensions in the submicron range. It offers a broad range of challenges and potential applications which can only be realized with a deep understanding of the spin-dependent transport and spin dynamic properties. Figure 1.1 summarizes some of the most important challenges appearing in this field that are currently under investigation which includes creating energy efficient ultrastrong magnets, achieving ultra-high density information storage while maintaining thermal stability, a spin transistor with gain, integrating magnetoelectronic devices with the existing semiconductor technology through the usage of room temperature magnetic semiconductors, magnetic random access memory (MRAM) for instant boot-up computers, magnetic logic, spin-based qubits for quantum computing, biomagnetic sensors, on-chip microwave oscillators etc. [1]

A new branch of nanomagnetism that utilizes the spin-degree of freedom alone or in addition to the conventional electron charge is called *spintronics* [2]. The idea of employing electron spin for information transportation has unique advantages over the conventional charge-based electronics because it potentially enables nonvolatile data storage, improved data processing speed, more efficient power consumption as well as high integration density [3]. For successful implementation of these goals a good understanding of three basic steps in the information transportation is necessary:

1-) How can we generate spin polarized current carriers?

2-) How can we conserve spin coherence during transport?
3-) How can we detect the spin orientation?

The first goal can be achieved by electrical transport through a ferromagnet. This results in a net spin polarization of conduction electrons due to an asymmetry in the scattering probabilities of electrons with different spin orientations [4]. In a simplistic picture, spin-dependent scattering probabilities are dominantly determined by the spin split energy band-mismatch at the Fermi level in a normal metal (NM)/ferromagnet (FM) interface in a spin valve structure [5] or the spin-polarized density of states of the two ferromagnetic electrodes in a magnetic tunnel junction [6]. By using a thin paramagnetic metal spacer with a long spin diffusion length or a thin tunnel barrier the spin coherence is conserved. This asymmetry in the conduction probabilities of different electron spins forms the basis for a number of interesting physical phenomena such as giant magnetoresistance effect (GMR) [7], tunneling magnetoresistance effect (TMR) [8] and their inverse effect, spin-momentum transfer (SMT) [9]. The magnetoresistance effects can be used to sense the spin orientation whereas SMT can be used to write information onto magnetic bits.

Chapter 2 describes the basic phenomenology of spin-dependent transport in magnetic multilayers. First I will discuss the generation of spin-polarization via electrical transport through a ferromagnetic layer. I will review two ways of measuring the spin polarization by Andreev reflections at a superconductor/ferromagnet interface and by Tedrow-Meservey tunneling spectroscopy. Then I will move on to a basic description of spin-dependent scattering in ferromagnet/normal metal interfaces via the simple 2-channel model which treats up-spin and down-spin currents to be independent of each other. Large magnetoresistance effects easily ex-
Figure 1.1: Grand Challenges in Nanomagnetism (modified from Ref. [1]). Nanomagnetism is a very exciting and challenging field with a variety of ambitious goals all of which rely upon creating and combining new magnetic materials and understanding their spin-dependent transport and spin dynamic properties.
ceeding 100% can be achieved in magnetic tunnel junctions. The section on tun-
neling magnetoresistance will introduce the basic physical mechanisms by which
such large effects can be realized. This chapter will conclude with a discussion
on spin-polarized current induced magnetic excitations within the framework of
Landau-Lifshitz-Gilbert (LLG) equation with a spin-torque term. A review of
macrospin analysis for nanomagnets will be followed by a more realistic approach
using micromagnetic simulations. This section will focus on the main shortcomings
of the macrospin approximation in comparison to the micromagnetic approach.

Chapter 3 presents the basic nano-fabrication procedures followed, to make the
devices under consideration in this thesis. This includes making point contacts to
extended thin films by in-situ deposition onto both sides of a nitride membrane
and defining a nano-aperture inside a nanopillar by using advanced electron beam
alignment techniques. The nonlinear etch technique used to make nanoconstric-
tions to thin films results in contact diameters less than 10nm whereas the ion-mill
etch for current confinement in nanopillars results in hole sizes between 15 to
30nm. This procedure may introduce non-idealities in the magnetic behaviour of
the nanomagnets such as domain wall pinning due to ion-mill induced defects. This
chapter will conclude with a discussion on ion-mill damage and possible solutions
to reduce and/or eliminate it.

In bulk magnetic materials such as Ni, the domain wall width determined by
an interplay between anisotropy and exchange energies is of the order of 100nm.
However in a magnetic point contact this width is determined by the constriction
size [10]. Chapter 4 presents a systematic study of spin-transport across a domain
wall geometrically constrained to fit at a 3D point contact between two otherwise
uncoupled ferromagnetic thin films covering from diffusive to ballistic transport
regimes. Here I introduce point contact spectroscopy as a tool to determine the electrical transport regime in magnetic nanocontacts. The contact size can be manipulated by applying a large current density at the contact region (≈ 10^10 A/cm^2) leading to electromigration. The resulting magnetoresistance is less than 0.5% even in the smallest contact sizes attainable (≈ 3nm).

The second half of chapter 4 describes possible artifacts that can occur in domain wall magnetoresistance measurements in experiments with sharp Ni tips. These include contact deformation through magnetostriction as well as magneto-static forces. I will include evidence for magnetostriction induced deformation even in nanoconstrictions due to surface anisotropy effects. For contact diameters less than 3nm, 1/f noise due to defect fluctuations obscures the measurement of such small magnetoresistance effects.

In the experimental geometry described in Chapter 4 a domain wall is nucleated by exchange biasing one side of the point contact with an antiferromagnet. The idea that a point contact makes an ideal site for domain wall nucleation is utilized in chapter 5 to achieve current induced magnetization reversal in a nanomagnet. A point contact inside a nanomagnet acts to concentrate the current away from device edges and therefore can maintain a strong local spin torque effect. This local spin torque leads to domain wall nucleation under the nanocontact which expands by current pressure to eventually cause full reversal of the nanomagnet. In chapter 5, I present low temperature data for spin torque experiments with a 150x250nm^2 nanomagnet with a 20-30nm nanoaperture that focuses the current to a local spot on a thin permalloy free layer. I will also show that nonuniform current injection does not only lead to full reversal but also microwave dynamics with threshold current levels being an order of magnitude smaller compared to
uniform injection to a same size pillar. I will show that the effect can be understood using micromagnetic simulations of the LLG equation with spin torque term which suggest a reversal mechanism via localized domain wall nucleation and propagation.

Chapter 6 describes an atomic scale characterization of the magnetic multilayers used in magnetic nanopillar devices using a combination of scanning transmission electron microscopy (STEM), electron energy loss spectroscopy (EELS) and x-ray photoemission spectroscopy (XPS) techniques. The first half of this chapter begins with the basic experimental procedures followed in sample preparation followed by a basic introduction to the theory of EELS and XPS techniques and their limitations. The second half is about the results obtained for different constituents in the magnetic multilayer stack, starting from the smooth $Ta/CuO_x$ seeding layers, moving onto Py/Cu interface subject to moderate temperature treatments and ending with chemical interactions in the $Py/AlO_x$ interface that appears in current-confined nanopillars. XPS investigations of the surface chemistry of Py exposed to air show a combination of Fe and Ni oxides. In studying $Py/AlO_x$ interface we evaluate the effectiveness of thin Al coating to passivate such unwanted oxidation. A discussion on possible artifacts in the characterization measurements and additional experiments that either rule out or reduce these artifacts is included in the appendix.

Temperature dependent pulse measurements and magnetoresistance measurements on Py/Cu nanopillar spin valves have recently shown anomalous low temperature damping enhancement and non-ideal switching characteristics below a critical temperature ($\sim 40K$) [11]. Chapter 7 focuses on the adverse effects of sidewall antiferromagnetic oxides in Py nanomagnets. First damping enhancement mechanisms due to surface oxides are introduced. Then I will show that the
anomalous low temperature effects due to a very thin surface oxide layer can not be explained within the framework of a strong exchange coupling model but is rather due to a weak, rotatable anisotropy.

Experimental results on three types of magnetic pillars subject to different sidewall treatments are presented in the second half of this chapter. These are pillars with additional NiO coating on the sidewalls, pillars whose sidewall oxidation is at least partially passivated by Al coating and pillars subject to no unusual treatment. Differences between both field driven and current driven switching characteristics as a function of temperature clarifies the role of unprotected sidewalls in the device performance. The effect of these oxides on the anomalous increase in the magnetic damping at low temperatures is revealed by the comparison of the spin-torque phase diagrams at 4.2K.

Finally, Chapter 8 will conclude with a discussion of the most important results obtained from experiments described in chapters 4 through 7 and also will provide a perspective for future investigations.
References for Chapter 1


[2] The term Spintronics was introduced by S. A. Wolf in 1996 as a name for a DARPA initiative for novel magnetic materials and devices.


Early studies of the temperature dependence of the resistivity of Stoner ferromagnets, where the moment is itinerant and delocalized, displayed a kink at the Curie temperature $T_C$ [1]. This simple observation led to the idea of spin-polarized current transport. The unusual change in the resistivity of a Stoner ferromagnet at its $T_C$ can be related to the exchange-splitting in the band structure of the ferromagnet which modifies the transport properties of the two spin populations, spin-$\uparrow$ and spin-$\downarrow$ at the Fermi level. This creates an imbalance in the ability of the ferromagnet to conduct electrons with different spins so that it favors one spin sub-band (called majority spins) over the other (called minority spins). These two components of the electrical current can be treated as a parallel combination of two conduction channels for spin-$\uparrow$ and spin-$\downarrow$ components as long as the spin-flip scattering events are rare [2].

When the spin-polarized current is transmitted by conduction through a medium with a sufficiently large spin-diffusion length such as $Cu$ or by tunneling through a thin barrier such as $AlO_x$, the spin polarization generated via spin filtering in the ferromagnet is mostly conserved. However, magnetoresistance effects occur when the electrons have to scatter into different momentum states during interaction with another ferromagnet whose magnetization orientation is not parallel to the direction of spin-polarization. The resulting giant magnetoresistance (GMR) and tunneling magnetoresistance (TMR) effects with a normal metal spacer and a thin tunnel barrier spacer respectively are manifestations of the requirement on the
conduction electrons to make the necessary changes in their spin angular momentum during these interactions. For high enough current densities carrying a spin polarization non-collinear with the magnetization direction of the free layer, the reaction torque exerted by electrons on the scattering center can also lead to an instability of the magnetic moment via depositing the transverse component of their spin on the lattice of the ferromagnet. Such spin momentum transfer (SMT) induced instabilities can give rise to full magnetization reversal as well as persistent spin dynamics in the ferromagnet on which the spin-polarized current impinges. Even within a ferromagnet a domain wall can experience a strong reaction force from such spin torque interactions that lead to mobilization and propagation.

2.1 Spin Polarization and Spin Filtering

The imbalance in the ability of a ferromagnet to conduct one type of spins more than the other due to its exchange-split band structure implies that important transport properties such as Fermi velocity $v_f$ and the density of states at the Fermi energy $g(E_F)$ are also spin-dependent. The resulting spin polarization is defined as:

$$P = \frac{n_\uparrow - n_\downarrow}{n_\uparrow + n_\downarrow}$$  \hspace{1cm} (2.1)

where $n$ is a spin-dependent quantity. For a spin polarized current $P$ is the ratio of the spin current $I_s$ to the charge current $I$, so that

$$P = \frac{I_s e}{I \mu_B}$$  \hspace{1cm} (2.2)

For a normal metal with equal density of states for both spin sub-bands $P = 0$, whereas for half-metals, which by definition have a band gap at Fermi level for one spin sub-band, $P = 1$. However, a fully polarized current at a finite temperature is
Figure 2.1: Total density of states as a function of electron energy for Ni, Fe and Co obtained from first principles band structure calculations [3]. The dotted line shows the Fermi level. The total density of states for electrons to occupy are spin-dependent due to an exchange coupling mechanism. This asymmetry for the number of states available for different spins to scatter into gives rise to spin-polarized currents.

yet to be measured. CrO$_2$ with 98% spin polarization is the highest measured so far. This discrepancy between the theoretical calculations and experimental results stems from the fact that most measurements of spin polarization are only sensitive to the polarization at a surface or at an interface where the spin-dependent band structure is not necessarily the same as the bulk. The simplest definition of $P$ is a description of an imbalance in the density of states for spin−↑ and spin−↓ electrons:

$$P = \frac{g_\uparrow(E_F) - g_\downarrow(E_F)}{g_\uparrow(E_F) + g_\downarrow(E_F)}$$

(2.3)

where $g_\uparrow(E_F)$ and $g_\downarrow(E_F)$ are the density of states at the Fermi level for spin−↑ and spin−↓ electrons respectively. The calculated total density of states for Ni, Fe and Co are shown in Fig. 2.1. Although this definition gives a quite intuitive
picture for spin polarization, it can only be measured through spin-polarized pho-
toemission experiments. The complex electronic band structure of ferromagnets
prohibits such a simple definition of polarization because of the differences in the
mobilities of electrons in different bands crossing the Fermi level when transport
measurements are made.

When a spin polarized current is injected into a paramagnetic normal metal,
the non-equilibrium spin population at the normal metal/ferromagnet interface
is subject to an exponential attenuation with a characteristic length scale known
as the spin diffusion length $L_s$ [4]. The first experiment to detect the coupling
between electron charge and spin at a ferromagnet/normal metal interface is the
pioneering work of Johnson and Silsbee [5]. The experimental geometry shown in
Fig. 2.2 consists of two permalloy (Py)/Al interfaces where one electrode injects
a non-equilibrium spin accumulation into Al and the other electrode detects the
spin polarization via a change in its relative chemical potential. A field sweep
that changes the relative orientation of the two Py electrodes from parallel to
antiparallel appears as a change of sign in the voltage across the detector. This
experiment also clearly showed that the spin information can be transmitted from
one ferromagnet to the other with a thin paramagnetic spacer quite easily.

Another way of injecting spins is using a ferromagnet/tunnel barrier interface.
In the famous Tedrow-Meservey experiment [6] a planar ferromagnet/thin insu-
lator was used to inject spins which can be detected by a superconductor. The
junction was cooled below the transition temperature of the superconductor and
a field was applied to Zeeman split the spin density of states of the superconduc-
tor so that perfectly spin-polarized states become available at the gap edges. A
measurement of the differential conductance as a function of bias can be fit to
Figure 2.2: The pioneering work of Johnson and Silsbee [5](a) The experimental geometry. Two permalloy (Py) Al interfaces are separated by a distance smaller then the spin diffusion length in Al. The first interface acts as a spin polarizer by injecting spins into the Al which are detected as a voltage appearing on the detector Py-Al interface which acts as a spin analyzer. (b) The detector voltage acts as a sensor of the relative orientation of the magnetization of the two Py electrodes(inset). When the field is swept to switch the two electrodes parallel and antiparallel, the detector voltage changes sign accordingly.
calculate the spin polarization of the ferromagnet. However a correct interpretation of the results must take into account the fact that tunnelling is dominated mostly by itinerant electrons. This leads to a new definition of spin polarization which involves weighting the density of states for each spin orientation with the appropriate tunneling matrix elements $\Gamma$ so that:

$$P = \frac{g_\uparrow(E_F)|\Gamma \uparrow|^2 - g_\downarrow(E_F)|\Gamma \downarrow|^2}{g_\uparrow(E_F)|\Gamma \uparrow|^2 + g_\downarrow(E_F)|\Gamma \downarrow|^2}$$  \hspace{1cm} (2.4)$$

However, the tunneling matrix elements also depend on the tunnel barrier of choice which makes it even harder to interpret the results. To avoid the complications in tunneling measurements of polarization, an alternative measurement technique called \textit{Point Contact Andreev Reflection} (PCAR) can be used to measure the spin polarization of a ballistic current. This technique was first used by Soulen et al. [7] and Upadhyay et al. [8].

For a conventional BCS superconductor for applied biases within the gap of the superconductor only Cooper pairs with opposite spins are allowed to be injected into or extracted out of the superconductor. At a normal metal superconductor interface this requirement leads to the Andreev reflection of holes of the opposite spin of an injected electron. Since only pairs of electrons can be injected at the subgap energies whereas single electrons can be injected at higher biases, the differential conductance is twice that of the high bias conductance.

In the case of a ferromagnet/superconductor interface the subgap differential conductance is suppressed due to the fact that it is not possible for every electron to find another one with opposite spin to be injected into the superconductor. Therefore the ratio of subgap conductance to the conductance at high bias is a measure of spin polarization of the current. This simple picture was modelled by Blonder, Tinkam and Klapwick [10] and is known to be the \textit{BTK model}. This model was
extended to the ferromagnet/superconductor case by De Jong and Beenakker [11].

Fitting the differential conductance to the extended-BTK model is possible with three parameters: $Z$, the transparency of the interface and $P$, the spin polarization and $\Delta$ the energy gap of the superconductor. However for the ballistic current case the definition of spin polarization is modified to be:

$$P = \frac{g_\uparrow(E_F)|v_F\uparrow|^2 - g_\downarrow(E_F)|v_F\downarrow|^2}{g_\uparrow(E_F)|v_F\uparrow|^2 + g_\downarrow(E_F)|v_F\downarrow|^2} \quad (2.5)$$

where the density of states are weighted by the Fermi velocities $v_F$ for each spin.

An example of BTK fits for Cu-Pb and Co-Pb interfaces are shown in Fig.2.4. Although this technique has been regarded to be a reliable measure of spin polarization, it is subject to errors from finite temperature effects, scattering from disorder at the interface, proximity effects, inelastic scattering processes etc...

This technique was applied by Upadhyay et al. [13] to show that Co films acquire
Figure 2.4: Differential conductance as a function of applied bias for a Cu-Pb and Co-Pb point contact sample. The data is fit to the extended-BTK model to extract the spin polarization, interfacial transparency and the superconductor band gap (from ref. [12]).
75% of their bulk polarization only with two monolayers. This important result shows the dominant role of interfacial scattering over bulk scattering in generating a spin-polarized current. In other words spin filtering is predominantly due to spin dependent scattering at a ferromagnet surface as a result of band mismatch.

### 2.2 Giant Magnetoresistance

Once a spin polarized current is generated via the spin filtering effect there are several possible scattering mechanisms that spins can experience. A clear distinction has to be made between spin-dependent scattering and spin-flip scattering because the first one gives rise to magnetoresistance effects whereas the later one is detrimental to it. Spin-dependent scattering occurs when electrons conserve their spin in the scattering event but the probability of the scattering event depends upon the spin orientation of the electron. On the other hand in the case of spin-flip scattering both the electron spin orientation and the spin of the scattering center change to satisfy conservation of angular momentum. Spin-orbit interactions, scattering of magnetic impurities in a normal metal spacer and scattering from spin waves within a ferromagnet are a few examples of cases that can cause spin-flip scattering.

When two magnetic electrodes are separated by a normal metal spacer, for certain spacer thickness values, an antiferromagnetic coupling exists between the two electrodes. This means that it is possible to achieve antiparallel configuration at zero applied field by virtue of the exchange coupling between the magnetic electrodes. This is known to be Oscillatory Exchange Coupling and was first reported by Parkin et al. [15](see fig.2.5). Under the application of a bias, the resulting electrical current is spin polarized and is subject to spin-dependent scattering at
Figure 2.5: (a) Magnetoresistance of three Fe/Cr superlattices at 4.2K (from ref. [14]). (b) Oscillatory Exchange Coupling in Fe/Cr superlattices at 4.5K (from ref. [15])
Figure 2.6: Band structure of Fe for minority spins gives a perfect match with that of Cr but the match is very poor for majority spins (from ref. [16]).

There are two contributions to the spin-dependent scattering in giant magnetoresistance (GMR) stacks. One is the bulk spin-dependent scattering which stems from the aforementioned exchange split density of states within a ferromagnet. Another important contribution is the interfacial spin-dependent scattering due to the mismatch between up spin and down spin bands and the band structure of the normal metal spacer. A good example is the Fe/Cr superlattice where the bands match almost perfectly for down spins but very poorly for the up spins (see fig 2.6). The resulting GMR was measured by Baibich et al. [14] and shown in fig.2.5. The GMR signal is defined as the percentage change in resistance from
parallel to antiparallel states so that GMR ratio (with the pessimistic definition) becomes:

\[ \frac{\Delta R}{R} = \frac{R_{AP} - R_P}{R_{AP}} \]  \hspace{1cm} (2.6)

where \( R_{AP} \) is the resistance in the antiparallel configuration and \( R_P \) is the resistance of the parallel configuration.

When the spin-flip scattering events are rare as is usually the case, the spin up and spin down conduction can be modelled to be occurring in two parallel independent channels [17]. When the local magnetization is parallel to the spin direction scattering is minimal with a small resistance \( r \) but an opposite orientation results in large scattering and large resistance \( R \). As seen in fig.2.7 the effective resistance of two parallel electrodes are determined by the parallel combination of these resistances for each case. Assuming \( R >> r \) one would get an equivalent resistance of \( 2r \) and \( R/2 \) for parallel and antiparallel cases respectively which implies:

\[ \frac{\Delta R}{R} \sim \frac{R - 4r}{R} \]  \hspace{1cm} (2.7)

There are two experimental geometries where GMR effects can be measured. The first one is the current-in-plane (CIP) geometry. This is experimentally easy to probe but theoretically more complicated. In CIP geometry, the current flows through low resistance normal metal layers and gets scattered at the ferromagnet/normal metal interfaces and the rougher the interfaces the higher the GMR. The second one is the current-perpendicular-to-plane (CPP) geometry which is the one used in this thesis. The resistance values are usually too small to measure unless one enters the nanoscale world. The CPP GMR is dominated by the interfacial scattering.
Figure 2.7: An illustration of the 2-channel model for spin dependent transport. When both ferromagnetic (FM) layers are parallel only one spin channel gets scattered at the normal metal (NM)/FM interface (low resistance state) whereas the antiparallel alignment causes both spin subbands to get scattered (high resistance state) [17].
2.3 Tunneling Magneto-resistance

When the normal metal spacer is replaced with a thin insulator in a GMR structure, the resulting device also shows magnetoresistance effects but this time due to tunneling of a spin-polarized current. Tunneling between two ferromagnetic electrodes with a thin insulating barrier was first studied by Julliere [18] for a Fe/GeO$_x$/Co structure. His treatment is along the same lines with the 2-channel model used for GMR structures. In the tunneling version of the 2-channel model spins that are emitted from one ferromagnetic electrode do not experience any spin-flip scattering events while tunneling through the thin barrier. Instead depending on the magnetic orientation of the destination electrode they are accommodated in the corresponding spin channel (see fig.2.8). The resulting conductance is determined by the density of states at the Fermi level $g(E_F)$ available for each spin orientation in both the emitter and the detector electrodes.

When the two ferromagnetic electrodes have parallel magnetizations $M_1//M_2$ then the majority/minority spins tunnel into the majority/minority subband of the detector electrode. Because majority spin population sees a high number of states available in the detector electrode this results in a low resistance state whereas in the antiparallel case majority spins experience a lot of spin-dependent scattering being a minority in the destination electrode giving a high resistance state. Using the simplest definition of spin polarization (2.3) combined with the simple Julliere model for 2-spin channels tunneling independently one arrives at the Julliere formula for the "optimistic" TMR ratio:

$$TMR = \frac{R_{AP} - R_P}{R_P} = \frac{2P_1P_2}{1 - P_1P_2}$$  \hspace{1cm} (2.8)

where $P_1$ and $P_2$ are the polarizations of the emitter and detector ferromagnetic...
Figure 2.8: Illustration of spin polarized electron tunneling in between two ferromagnetic electrodes. When the magnetization of the first electrode M1 is parallel to that of the second one M2, the majority of the current carriers that are polarized parallel (P) to M1 have a large number of states available to scatter into. This results in a low resistance state. However an antiparallel (AP) configuration necessitates that majority of the spin current that is polarized to be parallel to M1 can not be accommodated by the second electrode in which they are bound to become minorities. This leads to the high resistance state. Modified from ref. [19].
Figure 2.9: Giant tunneling magnetoresistance in $CoFeB/MgO/CoFeB$ magnetic tunnel junctions. (a) The magnetoresistance ratio of a $CoFeB/Mg(4\text{ Å})/MgO(8.5\text{ Å})/CoFeB$ 100x200nm tunnel junction. The arrows show the relative orientations of the magnetizations of the two CoFeB electrodes. (b) Bias dependence of the TMR ratio. (c) A cross-sectional high resolution TEM image of a CoFeB /MgO /CoFeB tunnel junction. (from Ref [20])

electrodes respectively.

Julliere’s model proves to be useful to give a basic insight into the problem of spin-polarized tunneling but fails to account for many important experimental observations such as the bias dependence of TMR as shown in fig. 2.9(b) for a $CoFeB/MgO/CoFeB$ tunnel junction or the dependence of the effect on the tunnel barrier of choice. More insights into the physical mechanisms that lead to experimental observations necessitate more complex theories that take into account a realistic band structure of the ferromagnetic electrodes [21], [22] as well as the details of the electronic structure of the tunneling barrier(see for example ref [23]).

Effects as large as 230% have been observed at room temperature in $CoFeB/MgO/CoFeB$ magnetic tunnel junctions [24] (data for one such large TMR device is shown in fig.2.9). However such high TMR ratios are usually accompanied
by large resistance-area products (RA) which limits the operating frequencies and increases both Johnson and shot noise. Since the device resistance scales exponentially with the barrier thickness, this can be accounted for by going to ultra-thin barriers ($6 - 7\AA$) to get RA products less than $4\Omega \mu^2$ as required for read head applications. This becomes a real challenge in terms of device fabrication since the interfacial roughness between the magnetic electrodes and the tunnel barrier can become comparable to the barrier thickness creating possible metallic conduction paths (pinholes) as well as causing magnetic coupling between the electrodes through magnetic edge charges developing at the interfaces. Later in Chapter 6 I will describe a recipe to get smooth metallic seed layers to avoid such problems. One can test to identify such potential problems with a device in question by measuring to get nonlinear I-V characteristics as well as testing to see if the temperature dependence of the device resistance is that of a weak insulator.

To understand the physical origin of the giant TMR effect one needs to look at realistic band structures of the electrodes as well as the the transmissivity of the tunnel barrier for electrons coming from each subband. Such a treatment has been used to argue that very large TMR effects should be expected in the Fe(100)/MgO(100)/Fe(100) junction [25]. The tunneling density of states for both parallel and antiparallel electrodes in this system are shown in Fig. 2.10. As can be seen in the majority density of states of Fe/MgO/Fe the $\Delta_1$ subband tail extend with minimal attenuation into the destination electrode. The tunneling current is dominated by the s electrons in this band giving a large current. However in the antiparallel case as can be seen in density of states of Fe(majority)/MgO/Fe(minority) case this band is attenuated giving a significant reduction in the tunneling current.
Figure 2.10: Theory of Giant Tunneling Magnetoresistance in Fe(001)/MgO(001)/Fe(001) tunnel junctions. Majority of the tunneling electrons are s-type which can be accommodated in the $\Delta_1$ band in Fe(001). Although these states are readily available for majority spins in Fe(001) (top left) they are non-existent in the minority band (top right). This implies a high conductance for the parallel case (top) when the majority spin states for s-electrons are available on both sides of the tunnel barrier. In the antiparallel case (bottom) the attenuation of $\Delta_1$ band in MgO in majority-minority tunneling coupled with its unavailability in minority-majority tunneling decreases the conductance. (from ref. [25])
Figure 2.11: Current induced switching in a CoFeB/AlOx/CoFeB nanopillar at 77K. Inset shows the minor loop of the same device. (from ref. [27])

The large effects also seen in MgO based junctions with amorphous CoFeB electrodes can also be attributed to a similar mechanism (coherent spin-polarized tunneling of $\Delta_1$ electrons because it has been shown that MgO(001) acts as a template to crystallize CoFeB via annealing at temperatures above 300°C [26]. This crystallization is believed to take place because of a very good lattice match between bcc CoFeB(001) and MgO(001).

Nanoscale magnetic tunnel junctions with their high resistances ($> 1k\Omega$) and large TMR ratios at room temperature ($> 100\%$) are ideal candidates for magnetic random access memory (MRAM) applications that offer a combination of nonvolatility, high density and high speed. In fact at the time this thesis is being written, Freescale Semiconductor has already announced a 4Mbit commercial MRAM chip using tunnel junctions [28]. In the current generation of devices these tunnel junctions are manipulated via the Oersted field generated by electrical cur-
rent. An alternative switching scheme by transferring angular momentum from a spin polarized current is under investigation [27]. Main advantages over conventional switching include superior scalability and elimination of half select issues. The first demonstration of spin transfer switching for a nanoscale magnetic tunnel junction is shown in Fig. 2.11. For more information on spin transfer in nano-scale tunnel junctions see ref. [29].

2.4 Spin Momentum Transfer

Both magnetoresistance effects I have discussed so far are the results of an interaction between a spin polarized current and the localized moment in the magnetic electrodes. If the current carrier electrons can undergo spin-dependent scattering, a natural question follows, that is: Are there any special circumstances in which the localized moments themselves can be scattered? First theoretical investigation to answer this question was undertaken by Berger [30] for the case of a high density spin polarized current incident on a domain wall. His findings indicated that domain walls can be dragged via the application of a spin torque from incoming electrons. This situation is illustrated in Fig. 2.12. An electron whose spin is close to being parallel to the local magnetization experiences a mutual torque with the local moments on a domain wall. Since the electron spin has to accommodate the local change in magnetization while traversing the domain wall it has to deposit its transverse component on the domain wall dragging it in the direction of its drift velocity.

A more recent theoretical treatment of current-driven domain wall motion was undertaken by Tatara et al. [31]. In this work it is shown that there are two contributions of the current to the domain wall motion. One is the momentum
Figure 2.12: A hypothetical domain wall in a 1-d ferromagnetic wire is shown. Under the application of a high density of spin polarized current the domain wall is dragged in the direction of the electrons as a result of the spin torque which deposits the transverse component of the electron spin on the wall.

Transfer from the charge current and the second one being spin transfer from spin current. The first momentum transfer occurs during reflection of the conduction electrons from the wall whereas the second one is due to a spin torque that develops while the conduction electrons are traversing the wall with their spin following the local magnetization adiabatically. In the case of thick metallic wires the spin transfer driven domain wall motion is the dominant term with a threshold spin current

$$j^{sr}_{cr} \sim \frac{e \lambda}{a^3} \max\left\{K_\perp, \alpha V_0\right\} \quad (2.9)$$

where $e$ is the electron charge, $\lambda$ is the domain wall width, $a$ is the lattice constant, $K_\perp$ is the hard-axis magnetic anisotropy, $\alpha$ is the Gilbert damping and $V_0$ is the pinning potential. This means that the wall motion can easily be controlled by the sample shape through $K_\perp$. The average wall velocity after depinning is given by

$$v_{dw} \sim \sqrt{\left(\frac{j_s}{j^{sr}_{cr}}\right)^2 - 1} \quad (2.10)$$

In the case of narrow domain walls constrained to nanocontacts for instance, the depinning current is dominantly determined by $j^{sr} \sim \frac{V_0}{\rho_w}$ where $\rho_w$ is the domain wall resistivity. The domain wall velocities driven by spin polarized currents were recently measured in permalloy nanowires by the IBM group [32] by us-
Figure 2.13: A proposed nonvolatile memory device using spin polarized current driven domain walls. It consists of a micron tall 100 nm wide track where each track holds about 100 bits of data. Current pulses are used to move domain walls in between bits from one notch to the other. A magnetic tunnel junction is used to sense the fringing fields from the domain walls via TMR effect. Current pulses through a separate write line create an Oersted field which writes information onto individual bits. Courtesy of IBM

The current driven domain wall motion was used in a proposal for a novel magnetic memory device called "magnetic race track memory" by S. S. P. Parkin. The design is shown in Fig. 2.13. It is based upon the current-driven motion of domain walls in a permalloy nanowire with nanoscale diameter but about a micron or so tall to increase data storage density by taking advantage of the third dimension. The magnetic bits are distinguished by notches fabricated in between them that act to pin domain walls after a current pulse. The information is stored in the fringing fields of the domain walls so that for example existence of a domain wall is a one but nonexistence is a zero. The information is read by an MgO tunnel junction sensor using the TMR effect and domain walls are nucleated or annihilated.
by using Oersted fields of a current pulse in the write line. This design offers several advantages over the existing hard-drive technology such as the possibility of making extremely high density of magnetic bits, being a reliable nonvolatile memory with no moving parts which also makes it relatively cheap compared to both MRAM and hard disks (However, one has to come up with a cheap and easy way to pattern the race tracks). The main problem for this approach is currently the threshold currents required to move the domain walls being too high.

In Chapter 4 we will see that the inverse of this effect, namely the domain wall magnetoresistance due to reflection of spins from a domain wall is also possible. The same exchange interaction that leads to domain wall motion can also be used manipulate the magnetic orientations of thin magnetic electrodes in current perpendicular to plane (CPP) geometry [33], [34]. In this case the spin transfer torques occur at the normal metal (NM)-ferromagnet (FM) interface such that the transverse component of the incoming spins are absorbed by the magnetic lattice in the ferromagnet. Such an interaction most dominantly takes place at the interface due to the mismatch between band structures of the FM and the NM for each spin subband, exactly the same thing that leads to the GMR effect. The band structure of the ferromagnet itself also causes electron spins to rapidly precess around the local magnetization which averages out their transverse components to zero. The nature of spin torque driven magnetic excitations in nanoscale CPP magnetic trilayers can be understood quite well within the framework of a single domain model. However to get better insights into real experiments a more realistic micromagnetic model is almost certainly necessary.
2.4.1 Macrospin Model

Just like any other magnetization dynamics, the magnetic excitations due to a spin-transfer torque are governed by the Landau-Lifshitz-Gilbert equation with a spin torque term (LLGS).

\[
\frac{d\hat{m}}{dt} = \gamma \hat{m} \times \overrightarrow{H}_{\text{eff}} - \alpha \hat{m} \times (\hat{m} \times \overrightarrow{H}_{\text{eff}}) - \left( \frac{\gamma \hbar}{e |\overrightarrow{m}|} \right) \eta I \hat{m} \times \hat{p} \times \hat{m} \tag{2.11}
\]

The time evolution of a magnetic moment \( \overrightarrow{m} \) is determined by the total effective torque acting on it. These torque terms are represented at the right hand side of the LLGS equation. The first one is the torque due to field driven precession. Here \( \gamma \) is the gyromagnetic ratio and \( \overrightarrow{H}_{\text{eff}} \) is the effective field seen by the moment including applied field, demagnetization field and any other field that is being applied by another magnet such as dipole field due to surface edge charges. The second term is the damping torque. As the moment precesses around the effective field it loses some of its energy to the lattice via spin orbit coupling. The resulting damping will reduce the angle of precession causing the magnetic moment to spiral down to be parallel to the effective field. The strength of damping is measured in terms of a phenomenological damping parameter \( \alpha \) also known as the Gilbert damping. Spin orbit coupling is not the only mechanism that determines the damping. Others include spin pumping, slowly relaxing impurities, eddy current damping, exchange bias etc.. These will be discussed in detail in Chapter 7.

The effect of the spin torque due to a spin polarized current is accounted for in the last term with I being the applied DC current, \( \eta \) being the polarization and \( \hat{p} \) the polarization direction. This torque term acts against the damping by pumping energy into the system. This may have two important consequences. If
the spin torque is much larger than the damping torque then the precession will occur in such a way to increase the angle between the magnetic moment and the effective field which will lead to magnetization reversal. The other possibility is to cancel out the damping torque in the presence of an applied field larger than the in-plane anisotropy field so that the precession never dies out. The diagram in Fig. 2.14 shows a summary of all the torque terms acting on the moment. Within this simple picture one can qualitatively understand how current driven excitations occur in a nanoscale CPP spin valve structure. This is illustrated in Fig. 2.15. For electrons flowing from the fixed layer to the free layer the current is polarized by the fixed layer so that the polarization direction is more or less parallel to the magnetization of the fixed layer. Note that for spin torque to occur there should be a small misalignment that can easily be achieved just from thermal fluctuations. The spin polarized current when incident upon the free layer applies a torque in such a way to orient the magnetization to be parallel to the polarization direction. If the spin torque is large enough (i.e. a threshold current density is reached), the free layer switches to satisfy this requirement.

However when the two layers are parallel, with this current direction this magnetic orientation is stable. When the current flows in the opposite direction (electron flow from the free to the fixed layer), the current is polarized by spin filtering in the free layer. Since the magnetic volume is too large in the fixed layer for spin torque to excite any dynamics, majority electrons are transmitted whereas the minority electrons are reflected. It is these reflected electrons when impinging back upon the free layer/spacer interface that apply a spin torque to switch the free layer to be antiparallel to the fixed layer.

Another possible outcome is that under the application of a large field that
Figure 2.14: Illustration of all the torque terms that act on a magnetic moment $\vec{m}$. The moment precesses around the effective field due to the precessional torque. Damping torque acts to reduce the precession angle whereas the spin torque acts to increase it when the spin polarization is antiparallel to the effective field.
Figure 2.15: A CPP spin valve with a thick fixed layer and a thin free layer is shown. When the magnetizations of the two electrodes are antiparallel as depicted at the top the antiparallel configuration becomes unstable whereas the parallel configuration in unstable for the opposite electron flow direction.

keeps the two magnetic electrodes parallel the spin torque can balance out the damping so that the magnetization precesses around the effective field persistently. This precession takes place at ferromagnetic resonance frequencies (1-10GHz) so that the device acts as an on-chip microwave oscillator.

A numerical study of the spin-current induced magnetic precession dynamics using LLGS equation for a single-domain body was performed by Jonathan Sun [35] and a summary of the results are presented in Fig. 2.16. Here $h_s$ is the
dimensionless spin-current amplitude and $\alpha$ is the damping parameter. In the presence of uniaxial anisotropy alone (top) with no spin current, the magnetization comes to a rest parallel to the easy axis due to damping. However when the spin current exceeds a certain threshold value the magnetization reversal occurs. A low damping decreases the threshold current needed to complete the reversal. However the ring-down time for the $x$-component of the magnetization is considerably longer in this case. The shape of the trajectory is determined by the uniaxial anisotropy alone to be a spiral.

When there is an additional easy-plane anisotropy (bottom) as would be the case in a thin film, the shape of the trajectory changes to be elliptically distorted with the cone angle spread out more in the easy plane but confined more perpendicular to the easy plane. This effect appears as an additional ringing during switching in the $z$-component. Another possibility is to have the spin current cancel out the effect of damping to get persistent oscillations under the application of a field larger than the in-plane anisotropy field. The resulting elliptical precession has been extensively studied and is discussed in ref. [36–39]. These numerical simulations have been reproduced and discussed in detail in ref. [40].

If the polarization of the incident electron spins lie in the plane of the film, the critical current levels for the onset of dynamic precession is given by:

$$I_c = \frac{\alpha 2e}{\eta \hbar} V (H + H_k + 2\pi M_s)$$

(2.12)

where $\alpha$ is the damping parameter, $\eta$ is the polarization, $V$ is the volume, $H$ is applied field, $H_k$ is uniaxial anisotropy field and $2\pi M_s$ is the demagnetization field where $M_s$ is the saturation magnetization. From this equation it becomes quite clear that to get small switching currents one needs to use a low $M_s$ material with low damping and a small volume. However to maintain thermal stability of the
magnet a high aspect ratio may be desirable. For details on a work along these lines see ref. [41]. In Chapters 5 and 7 I will present an alternative approach to get low threshold currents by local current injection as well as by decreasing extrinsic damping through sidewall oxide passivation.

2.4.2 Micromagnetic Effects

The macrospin model is an ideal situation where the nanomagnet acts like a Stoner-Wohlfarth single domain nanoparticle. However in real experiments discrepancies between the predictions of this model and experimental data for both field driven and current driven switching are inevitable. There are a number of reasons why a nanomagnet would not act ideally. The first category is geometry related issues such as lithographic edge roughness, redeposition of a magnetic material that is being etched to the sidewalls, the pattern of choice (elliptical, hexagonal, circular etc.). The second category includes structural problems including crystalline structure (magnetocrystalline anisotropy effects, defects, grain boundaries etc.) and thin film interfacial interactions (thermal diffusion and interfacial reactions, impurity effects.). The third category includes self-field effects from applied DC currents in spin torque experiments and nonuniformity of the effective dipole field that magnetic layers experience from magnetostatic edge charges on other layers and local pinning effects from antiferromagnetic sidewall oxides. Another potential problem is the Joule heating that becomes more pronounced at high applied currents, which would affect any temperature dependent intrinsic property of the nanomagnet including magnetization, spin polarization etc. and can also lead to device failure by electromigration.

All these non-ideal effects can be systematically included in a more realistic
**Figure 2.16:** The time evolution of magnetization under the application of a spin polarized current for uniaxial anisotropy only top and uniaxial and easy plane anisotropy bottom. The easy axis points in the z-direction. (a) and (d) the evolution of $z$-component. (b) and (e) the evolution of $x$-component. (c) and (f) Trajectory of the magnetization. From ref. [35]
micromagnetic model where the nanomagnet is considered as a collection of interacting spins whose magnetization is governed by the LLG equation and evolves together with the other magnetic layers rather than a single-macrospin. To be able to include at least some of the micromagnetic effects in modelling magnetic devices, a public software called object-oriented micromagnetic framework (OOMMF) is available from National Institute of Standards and Technology [42]. This code is written in C++ and Tcl/Tk. It does not take into account finite temperature effects and requires a script to be written to define a simulation problem. An example script that simulates the field driven switching of a 150x250nm² elliptical nanopillar with 5nm Py free layer, 20 nm Py fixed layer and 8 nm Cu spacer is shown below:

```plaintext
# MIF 2.1
# 150 by 250 nm ellipse with 5nm Py/8nm Cu/20nm Py
set pi [expr 4*atan(1.0)]
set mu0 [expr 4*$pi*1e-7]
set TIMEDRIVER 0 #Set the variable TIMEDRIVER to 0
Specify Oxs_MultiAtlas:atlas {
atlas { Oxs_BoxAtlas: {
name top
xrange {0 250e-9}
yrange {0 150e-9}
zrange {28e-9 33e-9}
} }
atlas { Oxs_BoxAtlas: {
name spacer
```
xrange \{0 250e-9\} 
ynge \{0 150e-9\} 
zrange \{20e-9 28e-9\} 

\} \} 

atlas \{ Oxs_BoxAtlas: \{ 
name \{bottom\} 
xrange \{0 250e-9\} 
yrange \{0 150e-9\} 
zrange \{0 20e-9\} 
\} \} \} 

Specify Oxs_RectangularMesh:mesh \{ 
cellsie \{5e-9 5e-9 1e-9\} 
atlas :atlas 
\} #The simulation unit cell is a 5nm x 5nm x 1nm cube within which the magnetization will be assumed to be uniform. 

# K1 is the crystalline anisotropy constant in J/m\(^3\) #axis is the anisotropy direction. For permalloy we assume that K1 is 0. No anisotropy. 

# Exchange 
Specify Oxs_UniformExchange \{ 
A 13e-12 
\} 

#The exchange constant is uniform across all space.A=13e-12 in J/m for permalloy. 

Specify Oxs_UZeeman " 
multiplier [expr 0.001/$\mu_0]
Hrange {
{0 1 1 400 1 1 20}
{400 1 1 -400 1 1 40}
{-400 1 1 0 1 1 20}
}"

# Demag
Specify Oxs_Demag {}

#The demag energy term is computed in each cell assuming constant magnetization in each cell.
if {$TIMEDRIVER}
{
  # Evolver
  Specify Oxs_EulerEvolve {
    alpha 1
    gamma_LL 1.76e5
    start_dm 0.01
  }

  #The damping parameter is 0.008 (JAP 91,10(2002)8659-8661) but we are taking 1 to reduce simulation time. The maximum change in the direction of magnetization m is 0.01 degrees per iteration.

  # Driver
  Specify Oxs_TimeDriver {
    evolver Oxs_EulerEvolve
    stopping_dm_dt 0.01
    mesh :mesh
    stage_count_check 0
```
stage_iteration_limit 0

total_iteration_limit 0

Ms { Oxs_ScriptScalarField {
script {Ellipsoid 6.5e5}
atlas :atlas
}
}
m0 { Oxs_AtlasVectorField {
atlas :atlas
norm 1
default_value { 1 0 0 }
values {
top { -1 0 0 }
}} } } } }

else

{ #The precision of the equilibrium is within 0.01 degrees. There are no limits to
number of iterations. #The saturation magnetization is 8.6e5 A/m. The initial
magnetic orientation is antiparallel.
Specify Oxs_CGEvolve:evolve {
gradiant_reset_count 1

minimum_bracket_step 0.00002 comment { atan(0.002) is approx. 0.1 degrees }
maximum_bracket_step 0.2 comment { atan(0.2) is approx. 11 degrees }
line_minimum_relwidth 1e-6
}
}

#If the the starting point of a line is not the result of a previous minimization the
first bracket attempt step is sized so that the fastest moving spin rotates by 0.1
degrees. The maximum angle the fastest spin is allowed to rotate is 11 degrees. The equilibrium is reached when the line bracketing interval is less than 1e-6.

Specify Oxs_MinDriver {
evolver :evolve
stopping_mxHxm 0.05
mesh :mesh
stage_count_check 0
stage_iteration_limit 0
total_iteration_limit 0
Ms { Oxs_ScriptScalarField {
  script {Ellipsoid 6.5e5}
atlas :atlas
}
} }
m0 { Oxs_AtlasVectorField {
atlas :atlas
  norm 1
  default_value { 1 0 0 }
  values {
    top { -1 0 0 }
  }
} }

#The equilibrium condition is mxHxm being less than 0.05
proc Ellipsoid { Ms x y z } {
set xrad [expr 2.*$x - 1. ]
set yrad [expr 2.*$y - 1. ]
set test [expr $xrad*$xrad+$yrad*$yrad]
if \{test > 1.0 | \| 0.606 < z \& \& 0.848 > z\} \{return 0\}
return $Ms$

The simulation universe is defined as a 3D box in what is called a box atlas. Regions with different magnetic properties are labeled separately as top, bottom and spacer. In this simulation top refers to the free Py layer and bottom is the fixed layer whereas, the spacer refers to the Cu in between. This box is divided into unit cells with dimensions $5x5x1nm^3$ within which the magnetization is assumed to be uniform (a single spin resides or with zero magnetization). The next task is to define intrinsic anistropy constants of the magnetic material of choice. For Py, magnetocrystalline anisotropy is negligible. Then the exchange stiffness in $J/m$ is specified. The "UZeeman" class specifies the field range and number of steps that will be simulated. For this example simulation the applied field ranges from -400 to 400 mT. The evolvers are used to update the magnetization configuration from one step to the other. There are two choices with one being the Euler evolver that keeps track of LLG dynamics and conjugate gradient minimizer that uses direct energy minimization. From the experience we have so far with Oommf the second one seems to run faster and pretty much gives the same results as the Euler evolver.

The actual cross-sectional geometry is defined in a procedure called Ellipsoid in this case. This procedure sets the magnetization outside a 150 by 250 nm ellipse as well as within the spacer to be zero and elsewhere $6.5x10^5emu/cm^3$ as measured for sputtered Py films at 4.2K with a superconducting quantum interference device (SQUID) \[43\]. The simulation is initialized to start with an antiparallel configuration between the two magnetic layers, which was experimentally found to be the case.

An example of deviations from single-domain behaviour can be seen in Fig.
2.17 for the simulation of the 150 by 250 nm elliptical $Py/Cu/Py$ nanopillar spin valve. First thing to notice is a geometrical effect that causes curling of the spins at the edges of the pillar in the free layer. This configuration is called a leaf state and has been observed in previous micromagnetic studies as well [44]. The next deviation from single domain behaviour becomes apparent from the sideview of two magnetic layers where there is an additional curling at the tips of the pillars due to dipole coupling between the two layers.

The micromagnetic simulations for ideal elliptical structures almost always overestimate the coercive field and dipole fields of the free layer by 100 to 200 Oe when compared to data taken at 4.2K. A closer look at the fabricated nanopillar structures by a scanning electron microscope (SEM) as shown in Fig. 2.18 reveals that in reality the nanopillar shapes are often far from being ideal. The lithographic edge roughness in combination with a tapering at the bottom due to reposition effects are observable. Careful simulations of these nonideal features have shown that increasing lithographic roughness seems to decrease the coercive field of the free layer whereas the dipole field is more dominantly affected by the tapered structure. These effects have also been extensively studied by Cowburn and coworkers [45] and they found that increasing edge roughness for permalloy rectangles larger than 200nm in width results in an increase in coercivity due to multi-domain structure whereas the opposite is seen in smaller structures.

Another possible deviation from the macrospin model is the possible nucleation of domain walls during the switching process which can be pinned by the structural defects in the nanomagnet during the reversal process. A micromagnetic study of nanosecond magnetization reversal for a $Co(20nm)/Cu(5nm)/Co(2.5nm)$ 70x130nm$^2$ was performed by Finocchio et al. [46]. Their results show that the
Figure 2.17: The simulated equilibrium state of a 150 by 250 nm elliptical $Py/Cu/Py$ spin valve. At the top is a top cross section through the 5nm Py free layer displaying a leaf state and at the bottom is a sideview cross section of the fixed and free layers.
Figure 2.18: SEM images of nanopillars demonstrate the lithographic edge roughness effects as well as tapering due to redeposition.
switching is by domain wall nucleation under the application of $10^8 \text{A/cm}^2$ current. The switching proceeds as shown in Fig. 2.19. At the initial stage the oscillations of spins at the boundaries of the ellipse give rise to domain formation. The domains expand quickly to decrease the central domain size. The confined domain collapses to complete the switching process.

Investigation of the microwave modes excited by a spin polarized current by Montigny et al. [47] have shown that micromagnetic simulations and macrospin calculations show a good agreement except for high currents ($\sim 10^8 \text{A/cm}^2$) where spin transfer induced disorder becomes a dominant effect. So in conclusion, although macrospin calculations give good insights into the basic physics underlying spin transfer in nanostructures a more complete micromagnetic model can be necessary to fully understand the spin dynamics induced by spin polarized currents in these nanomagnets.
Figure 2.19: Evolution of the magnetization reversal with nanosecond pulse switching in Co/Cu/Co spin valve nanopillars. (a) the initial stage of switching (b) domain wall propagation (c) final step with domain confinement. From ref. [46].
References for Chapter 2


CHAPTER 3

NANOFABRICATION OF POINT CONTACTS TO MAGNETIC THIN FILMS AND NANOMAGNETS

Current perpendicular to plane (CPP) magnetoresistance measurements as well as current-induced magnetization dynamics measurements require fabrication of magnetic nanostructures that allow for concentrated current flow. There are two conventional geometries that have been used to achieve the high current densities required. The first one is the nanoconstriction geometry where two extended metallic electrodes are in contact through a 5-20nm diameter aperture and otherwise are isolated from each other. This process was developed by Kristin Ralls [1] and was used for a number of experiments including giant magnetoresistance in magnetic multilayers and spin transfer [2], [3]. The second possibility is the nanopillar geometry where all the magnetic layers are either fully or partially patterned to usually a pancake shape with elliptical cross section with a minor diameter less than 100nm. This process was developed by Jordan Katine and Frank Albert and later modified by Nathan Emley and is well described in ref. [4, 5].

The experiments that are discussed in Chapters 4 through 7 include spin-dependent measurements on both types of structures in addition to one which combines the two ideas to make a nanocontact inside a nanomagnet for further current confinement. Since nanofabrication is as much of an art as being a science, every person has a different perspective on how to execute it. In the next few sections I summarize the nanoconstriction process from my perspective and describe a new process that I have developed to achieve concentrated nonuniform current injection into a nanomagnet.
3.1 Nanoconstrictions on Nitride Membranes

The fabrication procedure that I followed to make nanoholes on suspended nitride membranes starts with a 3”, double-side polished, 1-10$\mu\Omega$cm resistivity, $<100>$ oriented Si wafer. Here it is important to calculate what thickness of Si one needs because the thickness determines the size of the final nitride window. For details on how to choose the wafer thickness and the proper mask see the discussion on $KOH$ etch below. Next step is to deposit 100nm of low stress low-pressure-chemical-vapor-deposition (LPCVD) nitride. However to avoid contaminating the furnace and also to get high-quality films a standard metal-oxide-semiconductor (MOS) clean needs to be performed. Any metal and/or organic contaminants at the surface of the wafer will diffuse into Si as well as react with the reagents in the furnace during a high temperature deposition. The guidelines for MOS clean is as follows:

1. Since handling strong acids and bases are involved green nitrile gloves should be used.

2. These gloves usually have a Na residue on them as they come from the manufacturer so they need to be rinsed under running distilled (DI) water for about a min to clean the residue off.

3. Blow dry the gloves with nitrogen.

4. Rinse all the chemical baths with DI water and turn on the nitrogen bubbler until the resistivity sensor reads 16$\mu\Omega$.cm or more to make sure the baths are neutral.

5. Then the baths should be refilled with about 3.6L DI water and heated up to 75$^\circ$C

6. Rinse a 250ml beaker with DI water and blow dry
7. Prepare the base bath with 600ml of $NH_3OH$ and 600ml of $H_2O_2$.
8. Prepare the first acid bath with 600ml $HCl$ and 600ml of $H_2O_2$.
9. The last acid bath is a concentrated $HF$ solution which can only be prepared by a staff person.
10. Place wafers on a MOS clean boat and use a teflon holder to grab onto the boat.
11. When all baths reach back to $75^\circ C$ the wafers can be placed into the first acid bath for 10 min.
12. It is important to quickly transfer the boat to a DI rinse bath with a nitrogen bubbler to avoid any chemical residue.
13. After a 10min rinse in DI water where the resistivity finally reads about $16\mu \Omega cm$ the wafers can be transferred into the first acid bath to stay there for 10 min.
14. After this one they can quickly be transferred to the $HF$ bath to clean off the native oxide.
15. After a final rinse that brings the pH back to 7 the wafers are placed in a nitrogen drier.

Following this cleaning procedure wafers are considered to be MOS clean for about 6 hours. It is best to follow the MOS clean directly with the 100nm LPCVD nitride deposition. The exact thickness is not very important but it is important to have it thick enough to protect from scratches and holes that can develop during $KOH$ processing. The low stress recipe includes 84sccm $SiCl_2H_2$ and 20 sccm $NH_3$ mixture with a deposition rate of about 3nm/min. The final nitride quality depends mostly on the condition of the furnace. It is highly recommended to run a deposition within 2 days of a furnace clean. The uniformity of the nitride thickness
over a batch of wafers as well as the exact chemistry also depend on the location of the wafers with respect to the center as well as the separation between the wafers. For best results single spacing with central location are recommended. The final quality of the nitride can be checked by measuring the index of refraction with an ellipsometer ($\sim 2.15 - 2.17$) as well as measuring the stress (should be less than 200MPa tensile). A high stress nitride membrane can break to form cracks during subsequent processing.

The nitride deposition is followed by a photolithography step where a mask with a window size that is appropriate, given the thickness of the wafer for the final membrane window size is chosen. Since it is important to keep both sides of the wafer free of scratches this photolithography is done in two steps. First the front side is spin coated with Shipley 1813 resist spinning at 2000 rpm for 30 seconds. This gives a resist thickness of about 2 $\mu$m. It is not a good idea to bake the wafer on a hot plate under vacuum because that can potentially cause scratches on the back side. For that reason this bake is performed on a boat made from Al foil in a 90$^\circ$C oven for 10 minutes. After cooling down in air (not by using a heat sink) the opposite side (back side) is coated with resist following the same procedure. After cooling in air the back side is exposed the window pattern under deep UV for 6s by contact lithography. The resulting pattern after developing in MIF300 for 1 minute, rinsing in DI water and final nitrogen blow drying is shown in Fig. 3.1. In addition to the etch windows the pattern includes break lines which get partially etched in the subsequent $KOH$ etch step. These make it a lot easier to cleave the wafer. Any scratches that develop at the edges of the wafer due to handling with tweezers and/or from the pyrex where the development was done could cause problems in the $KOH$ step. For that reason all unprotected areas
should be wiped with a cotton swab soaked in resist. Then the wafer is hard baked another 20 min in the oven.

After photolithography the resulting pattern is transferred onto the wafer by dry etching the nitride that is exposed through the windows. To avoid any contamination from the etch chamber the first thing to do is a 15 min $O_2$ cleaning of the chamber. The standard recipe is to use 30 sccm $O_2$ at 30 mTorr pressure and at 100W RF power. To get reproducible etch rates for the nitride, the chamber is conditioned with the nitride etch gases. These are 30 sccm $CHF_3$ and 0.7sccm $O_2$. Again at 30 mTorr pressure and 100W power a 45sec conditioning is usually enough. I have found that using clean Si wafers to suspend the wafer instead of placing the wafer directly on the plate is effective in avoiding scratches. The pattern at the back side of the wafer is transferred assuming about 1 nm/min etch

**Figure 3.1:** (a) A schematic of the wafer cross-section after LPCVD nitride deposition and (b) top view after contact lithography.
Figure 3.2: (a) A schematic of the wafer cross-section after KOH etch and (b) after overhang removal and front side thinning.

It is important that all of the nitride in the window is removed so over-etching would be ok. After this the wafers are soaked in acetone and isopropanol to strip off the remaining resist to get ready for the KOH etch.

The most important step in the process is the subsequent KOH etching. KOH etches \{100\} planes leaving behind \{111\} planes in silicon [6]. Since the \{111\} planes are at 54.74 degrees with respect to the \{100\} planes, the nitride window size can be estimated by:

\[ x = w - \frac{2t}{\tan(54.74)} + u \]  \hspace{1cm} (3.1)

where \( x \) is the final window size that the nitride membrane will be suspended on, \( w \) is the window size in the exposed pattern, \( t \) is the wafer thickness and \( u \) is the undercut from KOH etch which is about 40\( \mu \)m. A window size of about 50\( \mu \)m is big enough for rough e-beam alignments. The overhang size depends on the
temperature at which \( \text{KOH} \) etch was done and also the etch time. So if after an inspection under the microscope the windows are found to be too small it is possible to widen them more by more \( \text{KOH} \) etch. The \( \text{KOH} \) etch procedure is as follows:

1. If the resist refuses to come off, residual amounts can be dissolved into \( \text{KOH} \). However too much resist on the other hand would contaminate the solution and decrease the etch rates.

2. DI rinse a pair of nitrile gloves, 3 2000ml Pyrex beakers, a wafer boat, teflon tweezers, magnetic stirrer and the teflon thermocouple of the hotplate and blow dry.

3. Weigh with a balance about 375g of white \( \text{KOH} \) pellets and place them into one of the beakers and fill it up with DI water to 1500ml level.

4. Set the stirring to 500rpm and probe temperature to 95\(^\circ\)C, lower the thermocouple into the solution. Too hot of a solution results in a more violent reaction of \( \text{KOH} \) with Si that can cause cracks.

5. Insert the boat with the wafers using a holder. The pattern should be facing out so that one can see bubbles coming out.

6. If the bubbles do not start coming out within 10-15 secs that means there is left over nitride which can be removed with \( \text{HF} \) etch but this never happened to me.

7. Keep another beaker full of DI water boiling on a second hot plate but do not let it boil too violently because that may break the membranes.

8. The etch rate is about 2\( \mu \text{m} \) per minute so that a 3-4 hour etch is usually enough but it is good to check every hour by holding the wafers to the light to see if the windows are transparent (it means Si has been etched away). Be careful \( \text{KOH} \) makes everything very slippery.
9. Once all windows are transparent the wafers can be transferred into the hot DI water to avoid any thermal shocks that can cause cracks in the membrane.

10. Prepare the third beaker full of DI water and replace the etch solution with this on the hot plate. Transfer the wafers to the new DI rinse bath once it is close to boiling (10 minutes).

11. Let them cool for half an hour and then place them under slowly running cold DI water.

12. Lift each individual wafer and gently spray DI water and at the end drip dry them all (No nitrogen...that will destroy the membranes!).

The resulting cross section after the KOH etch is shown in Fig. 3.2(a). The overhang from the KOH etch is removed by placing and aligning a Cu grid on the back side of the wafer which acts as an etch mask for the nitride membranes. Using the same nitride etch recipe described above the overhang nitride is removed by plasma etching. Then the front side nitride is thinned down to 50 nm by first assuming 1 nm/sec etch rate and targeting 75 nm thickness and then measuring the actual thickness to recalibrate the etch rate and finally going down to 50 nm. However, I have found that a high quality nitride is robust down to 30 nm. The cross section of the final structure is shown in Fig. 3.2(b).

At this point to define the nanoholes a single layer 5.5% PMMA e-beam resist is spun onto the wafers at 3500 rpm giving 250 nm thick resist. There is a special chuck which holds a wafer by clips instead of vacuum. For membranes it is necessary to use clips because vacuum will destroy the membranes. Again hot plate baking should be avoided to protect the membranes but instead the resist can be baked at a 170°C oven for 1 hour in an Al foil boat. In case of a problem with PMMA spinning spraying acetone and IPA to remove the resist is a bad idea.
because it is destructive to the membrane. Alternatively the resist can be etched in \(O_2\) plasma. The actual exposure is done after a coarse alignment so that the beam ends up exposing a single pixel anywhere on the nitride membrane in each window. Since a much more advanced alignment is used in the process that I developed later on I will focus on the alignment issue in the next section. The required electron beam dose to get a single pixel exposure is about 15000 – 20000\(\mu C/cm^2\).

After the exposure the wafers are developed in MIBK : IPA 1:1 solution for 90 sec without agitation. After development the wafers are gently rinsed with IPA and drip dried Fig. 3.3(a).

The resulting holes in the \(PMMA\) are 20-50nm wide and can be transferred into the nitride membrane by a special nitride etch recipe known as the nonlinear etch. This time the etcher is cleaned with \(O_2\) plasma in both linear and nonlinear etch modes. By feeding the etch gases from the bottom electrode instead of the shower and placing the wafer at 2 o’clock position on the plate the nonlinear etch mode works isotropically to give a bowl shaped hole so that the final hole size is

![Figure 3.3](image-url)
Figure 3.4: The cross sectional view of the nanoconstriction after isotropic nitride etch.

smaller than the one defined on the resist (about 5-20nm in diameter) Fig. 3.3(b), 3.4. The etch is divided into two steps in between which the wafer is rotated by 180 degrees to increase uniformity. The final device is prepared by in situ deposition of metallic layers onto both sides of the membrane but starting with the bowl side first for mechanical stability.

3.2 Drilling a Nanoaperture Inside a Nanomagnet

The nanoconstriction process enables making a three dimensional nanocontact to an extended film. However the bowl shaped contact imposes some limitations on the type of magnetic devices one can study. Micromagnetic simulations indicate that the bowl shape in addition to having a thick nitride membrane, promotes a vortex state or a C type bending (as will be seen in Chapter 4) when filled with a magnetic material at zero applied field, which can lead to non-ideal magnetic switching characteristics. Therefore it is desirable to have a point contact defined on an insulating layer as thin as possible to have better control on the magnetic structure around the contact region. Such a device has much better chances of working when patterned to nanoscale dimensions because it is easier to have a pin-hole free, thin insulating barrier. On the other hand the barrier needs to be thick
enough so that tunneling effects are negligible. Nanopillars with their nanoscale dimensions and well defined magnetic configuration due to shape anisotropy become an ideal choice. The outline of the process that I have developed to achieve these goals is as follows:

1. Sputter the multilayer

Start with a $<100>$ Si wafer with 2000Å thermal oxide. It is important to minimize the roughness of the layers to minimize orange neel coupling and avoid pinhole defects in the oxide barrier. One way to do this is to use a multilayer structure involving a $Cu/CuO_x/Ta$ stack. Another important point is that it is preferable to have oxidizable materials and if needed any the ones that are not oxidizable should be at the top of the pillar. A good example would be: $Py_{40}$(for good adhesion)/$Cu_{20}/CuO_x_{200}/Ta_{50}/CuO_{x_{200}}/Ta_{250}$/magnetic layers/Al/10Å..air oxidized. The sputtering can be done in AJA at a base pressure lower than or equal to $5\times10^{-8}$ Torr. The sputtering procedure for this type of a structure would be as follows:

First check all the power supply connections and condition all targets at appropriate powers (150-250W range) for 1.5 minute at 1mTorr Ar pressure, 8sccm flow rate (magnetron source) with manual control. Then sputter $Py_{40}/Cu_{20}$ stack at 1mTorr Ar pressure, 8 sccm. Then stabilize the pressure at 1 mTorr with 8 sccm $Ar+15\%O_2$ mixture (substrate source). Sputter $CuO_x$ at a high power setting (350W). At this point wait until the $O_2$ is pumped out, the base pressure goes below $5\times10^{-8}$ Torr. Recondition all the targets for 1.5 minute. Then sputter Ta 50 using $Ar$ only. Stabilize the pressure at 1 mTorr with $Ar+15\%O_2$ (substrate source) and sputter $CuO_x$ again at a high power setting. Wait until the pressure goes below $5\times10^{-8}$ Torr. Recondition all the targets for 1.5 minute. Sputter the
magnetic layer(s). Coat it with 10Å of Al. This has to be sputtered at 160W with 8 mTorr Ar pressure using 20sccm flow rate. This ensures the deposition rate is quite low (0.3 Å/s) which gives good surface coverage.

2) Ion Beam Deposition of Al₂O₃

To minimize impurity contamination it is best to transfer the wafer from AJA to IBD as soon as possible. For that reason keep the wafer in the AJA load lock until you are done preparing IBD (clean off the stage and vacuum out the chamber). Air transfer the wafer to IBD. XPS studies show that 10Å of Al will completely oxidize to give about 14Å of AlOₓ by exposing it to atmosphere. The acceptable base pressure range of the IBD is below 7 − 8×10⁻⁷ Torr. IBD deposition of Al₂O₃ is done at φ = 180° with the rotation on at room temperature using Ar (2.24 sccm) and Ar − 15%O₂ (1.92 sccm) mixture at 0.6 mTorr pressure. This needs to be calibrated frequently due to long-term fluctuations in the Ar + O₂ flow. Deposit about 56Å of Al₂O₃.

3) Photolith 1 (Coarse Alignment Marks)

Al₂O₃ etches in MIF300 developer because of the TMAH (tetra-methyl ammonium hydroxide) content. It needs to be protected by PMMA. Adjust one of the SU8 hot plates in the photolithography room to 1600rpm. Do an acetone/IPA spin cleaning on the surface. First spin 2% 495k PMMA at 3000 rpm which gives about 57 nm thickness. Bake on a 170°C hot plate for 15 minutes. Spin at 4000 rpm while blowing nitrogen to get rid of any contaminant particles on the surface. LOR (lift-off resist)10Å is too sensitive to the particulates on the surface.

Spin LOR10Å on PMMA in the photolithography hood where SU8 is normally processed. Spin speed of 2000 rpm gives about 1.4 microns thickness. Bake it on 160° hot plate for 5 minutes. Cool it down on the chill block for 10 sec. Spin
Shipley 1813 at 2000 rpm which also gives about 1.5 micron resist thickness. Bake the wafer at 115° for 1 minute. Chill block 10 sec.

Expose the latest Coarse Alignment Marks mask using HTG contact aligner. The exposure time is 60 sec. Use the contact-alignment plexiglass piece back side to align the mask to the wafer and to get good contact. The first development in MIF300 is only for 2 minutes. We will develop it more later on. The second photolithography step should follow right after this one.

3.2.1 Fine Alignment Marks

The sharpness of the edges of the fine alignment marks is what determines the ultimate alignment accuracy in electron beam alignment. There are two acceptable methods for making fine alignment marks. The cheaper, dirtier and also more time consuming method involves a lift-off photolithography procedure. This results in 20-30nm alignment accuracy whereas the more costly method in which the marks themselves are defined by e-beam lithography gives better than 10nm alignment accuracy. I have found that on a lucky day one can even achieve 5nm alignment. The lift off method is as follows:

4-) Photolithography 4: Without exposing the wafer to UV light bring the wafer to the stepper room. Expose the 'fine alignment marks' mask using the 5x stepper. This mask is to be aligned to the HTG mask. Use the Ozhan program pass1 on 5x stepper. Make sure the main switch is in Opaque position and you are in the 3" wafer mode with the appropriate loading corrections. (Chucks) Execute the 5x jobfile 'Ozhan'. The exposure time needs to be 3 seconds.

Develop the wafer in MIF300 manually for 1 minute. Never use the Hamatech
developer! It sometimes leaves MIF 300 residue on the wafer which slowly etches the metallic multilayer. This development gives about 0.5 microns undercut which is enough for a clean lift off process. $O_2$ clean RIEx for 10 min. $O_2$ clean at 100W with 30 sccm $O_2$ at 30 mTorr for 1 minute (2minutes in PT72) to descum as well as to remove the PMMA protection layer. PMMA etches at 210 nm/minute. So 1 minute should be more than enough to clean off the surface.

5) E-Beam Evaporation of Ti 20nm/120 nm Gold

Use the odd hours or even hours SC4500 evaporator to evaporate 20nm of Ti seed layer for adhesion/120 nm of gold onto the wafer. This can be done at 2-3 /sec deposition rate with e-beam evaporation. The tooling factor for this, I have found to be about 71.8% instead of the posted number 80% for odd hours evaporator.

6) Lift-Off Process of LOR10A

Clean 2 beakers with detergent and scotch brite. Rinse 3 times with DI water in TOL. Blow dry. Pour about 3-4 inch full of Nanoremover PG. One of them is for coarse lift-off and the other one is for fine lift-off. Put the wafer first into the coarse lift-off beaker and put the beaker on 900 hot plate and let it sit there until the large flakes come off (overnight). PG vaporizes very slowly. You will still have enough solution remaining even after 24 hours on the 90 degree hot plate.

Leave the door open and do not cover the beaker but put a label on the hood. The flash point of Nanoremover is 95°. Although I’ve measured the solution temperature to be about 60° when the plate is at 90° due to heat loss, we have to make sure there is no excessive pressure build up in the beaker or in the hood that could cause the nanoremover vapor to ignite.

Transfer the wafer to the fine lift-off solution and leave it on the 90° hot plate for 1 hour and ultrasound for 5 minutes. Repeat this 3 times. You can use the
automatic turn off feature of the hot plate and make sure there is enough solution that it won't totally evaporate during this time.

Immediately after taking out of the solution rinse with running DI water for 2-3 minutes. Blow dry and inspect the wafer to see if there is any residual Nanoremove left. Inspect the wafer under the optical microscope to make sure the lift-off is clean and there are no sidewalls on the alignment marks. The alignment marks you get should be 2 sets of 4 micron squares.

The second method of using e-beam lithography to define the marks is very similar to the way nanopillars are defined and can be summarized as follows:

4’) E-Beam Lithography to define fine alignment marks using bilayer PMMA

Place the wafer on a 3” chuck and spin at 3200 rpm. Blow nitrogen while spinning to clean off the surface dust particles. Alternatively spraying acetone, acetone + isopropanol and then finishing with isopropanol while spinning also works. Take a pipette and blow nitrogen both at the tip and also to the inside to clean off any dust particles. Rinse the inside of the pipette by filling it with PMMA and dumping the rinse solution into the spinner sink. Then take 495k PMMA (4%) in Anisole from the top. Let the first few droplets fall out into the sink to get rid of trapped air. Quickly squeeze the pipette at the center of the wafer and make sure the resist covers all the surface. Spin at 3200 rpm for 1min. Bake on a 170°C hot plate for 15 minutes and cool down on a chill block for 20 seconds. Next clean up any dust particle that fell on the wafer during the bake by blowing the surface with nitrogen. This time repeat the same procedure except use 950k PMMA (2%) in MIBK. Spin at 2000 rpm for 1 min and follow the same bake procedure as before. These two e-beam resist layers have different sensitivity to electron beam exposure with 495K being more sensitive. So at a given dose
an exposure results in an undercut structure similar to the lift-off resist used in photolithography.

5') E-Beam Exposure of Alignment Marks and Ti/Gold evaporation

This step is a straightforward e-beam exposure with no alignment. If using this method the coarse marks as well as the fine alignment marks can be exposed in a single e-beam step which substantially reduced processing time. It is important that the fine alignment marks are designed so that they can be exposed within one field which is 300 microns in Leica VB6. This means that each set of fine marks are exposed with no stage motion and only by beam deflection. If stage motion is involved that will introduce more alignment error. For details on how to do a basic e-beam exposure see Ref. [5].

After the exposure the wafer is developed in a 7 : 3 IPA:DI water solution by ultrasounding for 1 minute. Then spray the wafer with IPA and blow dry with nitrogen.

At this point the same Ti/Au evaporation procedure that is described above is used to deposit the marks. After the deposition do an acetone lift-off to strip the resist.

The resulting marks either made by lift-off photolithography or e-beam lithography are shown in figures 3.5, 3.6 and 3.7. The coarse alignment marks are squares with 40 microns on a side and located at (20,20),(20,-20),(-20,20) and (-20,-20) with all coordinates in millimeters and are with respect to the center of the wafer (Fig. 3.5). The fine alignment marks are at each die. One set of fine alignment marks consists of four 4 micron squares with the center of the pattern being offset 75 microns from the center of the device in the -y direction (Fig. 3.7). Each die has 16 devices with a Cu block in the middle for reference to the easy axis of the
3.2.2 Electron-Beam Alignment

7-) E-beam exposure: The instructions for electron beam alignment are as follows:
Spin 2% 495K PMMA at 3000 rpm and bake on 170° hot plate for 15 minutes, chill block 10 sec.
Align the flat of the wafer to be parallel to the stage motion. Locate the global alignment marks with respect to circle1 (top rightmost circle on the stage) and note these numbers. Always use the same magnification and focus that you used to locate the circle1.
Figure 3.6: Each die is designed to have 16 devices. The easy axis of the nanopillars are parallel to the Cu block. A dose test array is also exposed at the center of each die to check dimensions.
Figure 3.7: Two sets of fine alignment marks (4 micron squares) are shown in reference to the bonding pads for one device. Each set of marks forms a square with 250 microns and 300 microns on a side for the inner and outer set respectively. The center of the squares are 75 microns away from the device location.
Figure 3.8: A dose test array which consists of two arrays of nano-apertures and nanopillars that is exposed at the center of each die is shown. Four data types are used for 4 different doses to achieve variation in hole sizes.

For this step the focus and stigmation are very important. Use 0.2nA database LOADTINY followed by LOADTEST. Measure the current with MEA-C to make sure. Initialize the stage first with h c1 for the 3” holder. Check to see if the focus mark is located properly by MVSP FM/SSEM. Center the mark using LOCM FM O1 /POSM. These marks are spaced at 1.125 mm distance in x. Center a mark and do DDIA (measure the beam diameter) 5 times and average to check the beam diameter. The smaller the better.. When you find the best mark type SP FM . This will store the current stage position as the location for new focus mark.

Run focus and job calibration using FOC and JC. If you get a ’Fine Focus Not Good Enough For Conjugate Beam Blanking’ warning, do foc and jc once again. MVSP FM/SSEM/SFAB to check if the final focus is good. Also adjust the video levels by decreasing BACK OFF and increasing GAIN.
Locate circle 1 and go to the first expected location in the 'GLOBAL\_ALIGN\_COM' (one of the coarse alignment mark locations) and after centering the mark, locate the 3 remaining global alignment marks. If you can't immediately find them use circle1 and relative motion MVSP CIRCLE1 and MVRL to find the mark locations. The coarse marks are located at (20 -20), (20 20), (-20 20) and (-20 -20). On each mark first manually run LOCM PIT On /POSM for the nth mark. Then SP On. When the alignment is done load the absolute coordinate system with DWMO ABS/LOAD. Now you need to align to the 5X mask. MVSP O1 then MVRL -2.05 -1.875 to go to the first fine alignment mark location. Run FINEMARK\_COM. Then LOCM FINEMARK O5 /POSM. Then run GLOBAL\_FINE\_ALIGN\_NANOHOLE\_COM. At each mark location store the position as SP O5-8. At the end check your alignment by going to one of the device alignment marks. For example MVPO -6.9 -6.725..then MVRL +0.150 +0.150. Run the job file FINE\_ALIGN\_NANOHOLE\_COM. The electron beam will go to the expected position of each mark and scan a 40 micron window to look for a change in the secondary electron signal. Once it locates the gold marks it will do a fine scan to locate the center of each mark. At the end the computer displays the mark locations and the pattern read will be exposed in the new chip coordinate system. In this case a nano-hole is exposed 75 microns away from the center of the chip. A schematic of the alignment is shown in Fig. 3.9. The alignment procedure for nanopillars is identical except this time the marks at +0.125 mm locations are used. These are the inner set of marks. Develop in 7:3 IPA/DI Water by ultrasounding for 1 minute and rinse the wafer with IPA. Blow dry. AFM to check the array of holes to make sure they came out fine and to check your alignment. An example AFM image is shown in Fig. 3.10. The biggest holes are DT6 and
Figure 3.9: A schematic of the electron beam alignment procedure for each device is shown. The electron beam scans each mark and locates the mark center by analyzing the strong secondary electron signal from gold marks. Then each device is exposed 75 microns away from the center which coincides with the center of the bonding pads.
Table 3.1: An example table of device locations. To move from one die to the other all the coordinates should be augmented/diminished by 5mm. Devices 1 through 16 are shown with their x and y coordinates for one die.

<table>
<thead>
<tr>
<th>Device</th>
<th>X Coordinate</th>
<th>Y Coordinate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1/16</td>
<td>-1.900</td>
<td>-1.725</td>
</tr>
<tr>
<td>2/16</td>
<td>-1.900</td>
<td>-0.525</td>
</tr>
<tr>
<td>3/16</td>
<td>-1.900</td>
<td>0.675</td>
</tr>
<tr>
<td>4/16</td>
<td>-1.900</td>
<td>1.875</td>
</tr>
<tr>
<td>5/16</td>
<td>-0.700</td>
<td>-1.725</td>
</tr>
<tr>
<td>6/16</td>
<td>-0.700</td>
<td>-0.525</td>
</tr>
<tr>
<td>7/16</td>
<td>-0.700</td>
<td>0.675</td>
</tr>
<tr>
<td>8/16</td>
<td>-0.700</td>
<td>1.875</td>
</tr>
<tr>
<td>9/16</td>
<td>0.700</td>
<td>-1.725</td>
</tr>
<tr>
<td>10/16</td>
<td>0.700</td>
<td>-0.525</td>
</tr>
<tr>
<td>11/16</td>
<td>0.700</td>
<td>0.675</td>
</tr>
<tr>
<td>12/16</td>
<td>0.700</td>
<td>1.875</td>
</tr>
<tr>
<td>13/16</td>
<td>1.900</td>
<td>-1.725</td>
</tr>
<tr>
<td>14/16</td>
<td>1.900</td>
<td>-0.525</td>
</tr>
<tr>
<td>15/16</td>
<td>1.900</td>
<td>0.675</td>
</tr>
<tr>
<td>16/16</td>
<td>1.900</td>
<td>1.875</td>
</tr>
</tbody>
</table>

Smallest holes are DT3 each having 4 columns. The intermediate hole sizes each have 3 columns. For AFM imaging always use a new tip to start with. The tip diameter must be less than 10nm. (Olympus AC160TS). A better but more expensive choice would be to use C diamond tips with 1nm diameter. However these tips require extra care because they are very fragile. One should start imaging at the boundary of largest holes. Avoid imaging the frame... The tip can not recover once it goes into the sides of the frame and gives very poor contrast for holes. Once you find the holes go to the hole that is further into the frame and use offset to center the hole... Do a 1 micron scan with 0.1 Hz scan speed. I have found that I could get the tip to go into the hole as much as 10nm deep. With the 5 micron scan it only goes about 3nm into the hole. WHEN MEASURING HOLE DIAMETERS ONLY SECTION PERPENDICULAR TO SCAN. Scan lines increase the true diameter about 1 tip size. For example a 60nm hole measured along the scan direction is actually 50 nm in diameter. Most of the tip artifacts can be minimized by scanning at a 45 degree to the hole. Sometimes if the tip hits the surface hard enough you will get an image of the tip rather than the hole. This is shown in Fig. 3.10(a). An example AFM image of an array of largest holes is shown in Fig. 3.11. The SEM imaging of the holes is very tricky. You need to be in the range 1 to 2 keV with a working distance of about 3 mm to minimize spherical aberration and
Figure 3.10: (a) shows an artifact that can occur in imaging of the holes that happens when the tip is not sharp enough to go deep into the hole resulting in an image of the tip. (b) An AFM image of a 20 nm nano-aperture on PMMA.

stigmation corrections. Imaging holes etched into PMMA that end in $\text{AlO}_x$ is very difficult because of charging problems of both layers. That’s why this is not the preferred method. An example SEM image of a smallest hole is shown in Fig. 3.12.

### 3.2.3 Finishing the hole (Ion-Mill Damage)

8) Ion-mill to define holes (TRASHCAN)

Ion mill the wafer at 0 degrees to the normal with 11.5 mA cathode current and
Figure 3.11: An AFM image of an array of largest holes obtained which are about 100nm in diameter.
Figure 3.12: An SEM image of one of the smallest holes obtained in $PMMA$ is shown. The surface appears darker due to C signal in $PMMA$. 
10 mA emission current at 500V beam voltage, 200V accelerator voltage and 40V discharge voltage. The mill rate of $\text{Al}_2\text{O}_3$ is 0.37\(\pm\)0.02 Å/sec and that of PMMA is 3.13 Å/sec. Aim for half way through the $\text{Al}_2\text{O}_3$ (the mill rate can drift in time so it should be recalibrated before milling the actual devices). This mill is at room temperature with no heat sink compound and no rotation. The shutter is open for 20 sec and closed for 40 sec for each mill step.

9) Photolithography #3 (Protect Alignment Marks) RIE $\text{O}_2$ clean the residual PMMA and acetone/IPA clean, respin 2% 495k PMMA at 3000 rpm and bake at 170°C for 15 minutes, chill block 15 seconds. Spin and bake LOR10A, spin and bake Shipley 1813. Expose the ‘Protect Alignment Marks’ Mask on 5x with 3 sec exposure using ‘Ozhan’ jobfile and develop for 5 minutes in MIF300.(2’+2’+1’). RIE $\text{O}_2$ clean for 1 minute to remove the PMMA underlayer.

10) Ion Mill #2 (Finish the holes and the multilayer)(TRASHCAN) Ionmill through the rest of $\text{Al}_2\text{O}_3$ to open up the nanoholes. Sputter the top multilayer. Cap it with Cu and transfer to AJA to finish with Pt cap.

11) Evaporate C 800 Å-This step should follow right after step 10. The C source is ours which we prepare by breaking C rods in Clark. Put them as packed as possible into the crucible. Make a line of C that the beam will sweep on. Acceptable base pressure is below 2 x 10^{-6} Torr. The frequency of the sweep should be very low. Increase the power at 1%/min rate. The correct power setting is around 11.1 to 13%. The EVAPORATION RATE=20 Å /min. Count a min to check if the rate is about right and adjust the power accordingly. Having it go down too fast causes adhesion problems. This rate is very important.

12) Lift-Off Process same as 6. At this point we are ready to start the standard nanopillar process with an aligned exposure. To be able to protect C from coming
Figure 3.13: (modified from ref. [7]) A schematic representation of the possible outcomes from the interaction of energetic ions and a metal surface.

off during ultrasound do a 10 sec on 10 sec off routine for 10 min. 5 min total ultrasound time seems to be enough. We now have a wafer with a complete multilayer and an embedded nano-aperture with gold marks that tell us where the aperture is located. Follow the same alignment procedure and start the regular nanopillar process. This process is well-described in ref. [5].

The only problematic part of this process that needs further discussion is the possible outcomes of the ion milling of the nano-apertures. The ion mill process is a mechanical one where Ar atoms are ionized by electrons that are boiled off and accelerated from a filament. The kinetic energy of the Ar ions makes them hit the cathode and remove material. When the energetic ion hits the surface as shown in Fig. 3.13 there are four possible outcomes. For ion energies smaller than 10 eV there will be a minor heating on the surface by the transfer or energy to phonons. For energies higher than 10 keV ion implantation occurs deep into the structure. This can change the physical structure and introduce defects. Another possibility
is the redeposition of atoms extracted from the surface. There will always be reflected ions and secondary electrons as well.

For the energy range used in this process with a beam voltage of 500V and accelerating voltage of 100V although not a substantial damage to the magnetic films is expected, introduction of some defects into the nano-contact region is inevitable. One way to get around this problem is to insert the nano-contact in the midst of the spacer normal metal layer so that the ion mill damage if any does not extend into the magnetic layer and the magnetic switching characteristics are not affected by this. We will see some consequences of the ion-mill damage in Chapter 5 in current-confined nanopillars.
References for Chapter 3


CHAPTER 4
GEOMETRICALLY CONSTRAINED DOMAIN WALL MAGNETORESISTANCE AND MAGNETOSTRICTION EFFECTS IN 3D MAGNETIC NANOCONTACTS

When a contact of nano-scale dimensions is established between two ferromagnetic electrodes (also known as a magnetic nanocontact) it is energetically favorable for any domain wall that is developed between the two layers to be constrained at the contact region [1]. Such geometrically constrained domain walls behave differently from Bloch and Neel walls in the bulk material in the sense that the domain wall thickness is expected to scale linearly with the minimum diameter of the nanocontact.

In an unconstrained bulk ferromagnet the wall structure is determined by a trade off between the exchange and anisotropy energies (Bloch wall) [2] whereas in a thin ferromagnetic film the dipolar interaction also plays an important role leading to Neel walls where the spins rotate in the film plane [3], Fig. 4.1. The corresponding situation in a magnetic nanocontact can be understood by considering the interplay between the wall area and the exchange energy. When the cross-sectional area of the constriction is a lot smaller than the cross section of the wide area, the wall structure loses its dependence on the material properties such as magnetization, exchange stiffness and anisotropy constant. If we assume an infinitely narrow wall to be nucleated at the nanocontact region, the exchange energy for such a configuration becomes too high and can be reduced by letting the wall expand at the cost of an increase in anisotropy and wall area. If the cross-sectional area changes rapidly as the wall expands away from the nanocontact then the cost of increase in the wall area becomes the dominant term. This means that
Figure 4.1: Types of domain walls in bulk ferromagnets. (from ref. [4]) (a) Bloch wall shows a 3 dimensional spin rotation with the wall width determined by an interplay between exchange and anisotropy energies (b) Neel wall shows an in-plane spin rotation in thin ferromagnetic films with a wall width determined by exchange, anisotropy and dipolar energies.

the wall structure in this case is determined by the geometry. A more abrupt magnetic transition as compared to the bulk can occur from one magnetic layer to the other. Fig. (4.2) displays the results of an energy minimization calculation for a contact size d and unconstrained wall width $w_0$ with $d/w_0 = 0.1$ and a factor of 10 increase in cross-sectional area for distances larger than d. It can be seen that a constrained domain wall as compared to an unconstrained Bloch wall ends up being a lot narrower with its width being comparable to the contact size. When a domain wall width becomes much smaller than the spin-flip mean-free-path of
Figure 4.2: Comparison of domain wall widths in constrained and unconstrained geometries (from ref. [1]) The $z$-component of the magnetization $M_z$ normalized to the saturation magnetization $M_s$ representing the domain wall is shown for an unconstrained Bloch wall (dashed line) and a constrained domain wall (solid line) as a function of distance $x$ from the wall center. $w_0$ is the width of the unconstrained Bloch wall.
electrons in the magnetic layers, spin transport across the wall becomes ballistic. Due to the non-adiabatic nature of such transport the spin scattering rate at the interface is expected to increase which implies an enhanced magnetoresistance as compared to one due to an unconstrained domain wall. Especially in the case of atomically sharp point contacts one can expect to see a large magnetoresistance effect. The enhancement of the magnetoresistance with decreasing domain wall width has also been shown for a Py/Gd/Py trilayer where the domain wall is trapped in the Gd layer instead of a point contact. [5].

Recently there have been reports of very large magnetoresistance effects in electrodeposited Ni nanocontacts [6], [7] and such effects have also been extensively studied for nanocontacts prepared by electrical breakdown of an oxide layer between a Ni microcluster and a magnetic film [8] as well as mechanical contacts between sharp tips etched on Ni wires [9]. There have also been reports on high magnetoresistance effects in half-metallic oxide point contacts prepared by break-junction technique [10]. However the exact contact geometry in most of these techniques is not known, and in general the interface purity and quality can not be easily controlled. Mechanical stability is also limited in break junctions. Clean, stable, and reproducible contacts formed under at least near-UHV conditions are likely to be required for device applications.

In pursuit of this goal, in this experiment (see ref. [11]) we employ nanofabrication and thin film deposition techniques as described in Chapter 3 section 3.1 to form Ni and Co nanocontacts that have a three-dimensional point contact geometry with a minimum contact diameter in the range of 1 - 10 nm (see fig.4.10). The electron transport through these nanocontacts has been demonstrated to be ballistic or at least quasi-ballistic. Measurements of the magnetoresistance aris-
ing from the creation and removal of a domain wall have yielded a small effect, < 1%, but one which increases with decreasing contact diameter. The smallest contacts are subject to a large $1/f$ noise which obscures the measurement of the small magnetoresistance effect. Depending upon the substrate preparation, the nanocontacts can exhibit a strong magnetostriction behavior in the presence of an in-plane magnetic field. The experimental geometries under consideration in most previous experiments are subject to measurement artifacts due to the existence of magnetostatic forces and magnetostriction induced contact deformation as shown in a systematic study by Egelhoff et al. [12]. We have shown that a lithographic 3 dimensional point contact geometry can successfully avoid these complications and makes it possible to uniquely identify the domain wall magnetoresistance signal.

4.1 Point Contact Spectroscopy

In the development of microelectronic devices the problem of making reliable metallic contacts to small scale devices is a very important challenge. For this reason, understanding electrical transport through metallic contacts both in the diffusive and the ballistic regime will be very helpful in designing reliable contacts. In the so-called diffusive regime the contact size $a$ between two metal electrodes is much larger than the electron mean free path $\lambda$. In this regime under the application of a voltage the resulting electron flow consists of highly nonlinear trajectories. The diffusive regime is also called Ohmic regime because Ohm’s law applies to the contact resistance. A very insignificant parabolic background appears in the R-V characteristic due to ohmic heating. If one plots $dR/dV$ as a function of $V$, the resulting plot is linear with a very small slope reflecting the heating effect. However when the contact size is reduced so that it becomes comparable to the electron
mean free path the conduction enters the ballistic regime where the electron trajectories are linear. Interesting nonlinear features appear both in the $dV/dI$ vs $V$ and $d^2V/dI^2$ vs $V$ plots especially at low temperatures. This interesting feature was first observed by Yanson in 1974 [13] while measuring I-V characteristics of a pinhole on an oxide barrier. To his surprise he realized that the peaks that he observed correspond to the phonon frequencies of the bulk metal.

This nonlinear effect can qualitatively be understood as follows: within a mean free path, electrons are accelerated due to the electric field caused by the applied voltage and are injected from one side through the point contact to the other. These accelerated electrons are brought back into equilibrium by inelastic collisions usually with phonons. After an inelastic scattering process electrons can backflow through the contact which gives rise to a voltage dependent correction to the current.

To be able to describe the voltage dependence of the resistance of a metallic point contact quantitatively, one needs a general expression for the resistance of a point contact. This problem was classically considered by Maxwell [14] where he solved the Laplace equation for a small classical metallic contact (i.e one which has a diameter much larger than the electron mean free path) as a function of the position $r$ in oblate spherical coordinates (see Fig. 4.3). For a current flowing perpendicular to an orifice of radius $a$ the contours of constant potential vary roughly by an inverse tangent dependence on position. The potential $V$ is calculated using oblate spherical coordinates with the Laplace equation:

$$\Delta^2 V(r) = 0$$ (4.1)

with solutions:

$$V(r) = \pm \frac{1}{2} eV[1 - (\frac{2}{\pi})tan^{-1}(\frac{1}{\xi})]$$ (4.2)
Figure 4.3: A point contact which consists of a circular orifice with radius $a$. An arbitrary position is described with coordinates $(r, \phi, z)$.

Figure 4.4: (Equipotential surfaces and electric field lines in a Maxwell point contact..from ref. [15]) The equipotential surfaces around a point contact are ellipsoids with electric fields lines concentrated within a diameter of the contact.

where $\xi$ is given by the coordinate transformation:

$$\frac{r^2}{a^2} = (1 + \xi^2)(1 - \frac{z^2}{\xi^2 a^2})$$  \hspace{1cm} (4.3)

An expression for the resistance of the point contact in the ohmic (diffusive) regime can be obtained from the Maxwell expression of the potential distribution around the contact region (equation 4.2) by using Ohm’s law. The total current is given by:
The resulting expression for the Maxwell resistance $R_M$ of a point contact with radius $a$ in the diffusive limit i.e for $a \gg \lambda$ in between two metal electrodes with resistivity $\rho$ is:

$$R_M = \frac{\rho}{2a} \quad (4.6)$$

When the contact width is comparable to the electron mean free path Ohm’s law does not apply any more. Following Drude model the change in the velocity $\Delta v$ of an electron as it traverses through such a point contact junction is directly proportional to the applied potential $V$ and inversely proportional to the Fermi momentum $p_F$. For a contact radius $a$, electron density $n_0$ and mean free path $\lambda$ the change in velocity, the total current and the resistivity are given by:

$$\Delta v = \frac{eV}{p_F} \quad (4.7)$$

$$I = \pi a^2 n_0 e \Delta v \quad (4.8)$$

$$\rho = \frac{p_F}{n_0 e^2 \lambda} \quad (4.9)$$

Substituting these equations into $V/I$ and integrating over all angles in the limit where the electron mean free path $\lambda$ is much larger than the contact radius $a$ one gets the Sharvin resistance in the ballistic limit $R_S$(ref. [16]):

$$R_S = \frac{4\rho \lambda}{3\pi a^2} \quad (4.10)$$

One important point to note here is that the Sharvin resistance is independent of the electron mean free path. In fact the resistivity mean free path product ($\rho \lambda$)
is a material dependent constant. For a 1 Ω Cu sample Maxwell formula gives 
\( a = 4\AA \) whereas Sharvin gives 120Å. So for reasonably pure metals, point contacts 
usually operate in the ballistic regime.

For the intermediate regime in between ballistic and diffusive contacts a simple 
interpolation formula was proposed by Wexler [17]. He solved the Boltzmann 
equation using the variational principle for resistance of a contact. His result is that 
the contribution from the diffusive part has to be weighed by a gamma function 
\( \Gamma(K) \) which depends on the ratio of electron mean free path \( \lambda \) and contact radius 
a. This ratio is known as the Knudsen number \( K \).

\[
R_W = \frac{4\rho\lambda}{3\pi a^2} + \Gamma(K) \frac{(\rho\lambda)}{2a\lambda} 
\] (4.11)

In the limit of a purely ballistic contact i.e. as lambda goes to infinity the 
resistance is Sharvin resistance whereas in the limit of purely diffusive contacts i.e. 
as lambda goes to 0 one sees the resistance goes to infinity as expected. Fig. 4.5 
shows resistance vs Knudsen number for a 10 nm Cu point contact. We can see 
that the Wexler resistance asymptotically approaches the Sharvin resistance as the 
Knudsen number increases.

Now that we have a general expression for the resistance of a metallic point 
contact, to be able to find the source of the spectroscopic signal we need to answer 
the question where the voltage dependence comes from. The resistivity mean free 
path product is a material dependent constant and \( a \) is the contact radius. So the 
Sharvin like term does not give any voltage dependence but it serves the purpose of 
allowing an electric field within a metal. The spectroscopic signal comes from the 
energy dependence of the electron mean free path. So the question boils down to 
understanding where the energy dependence of the electron mean free path comes
Figure 4.5: (Resistance vs. Knudsen number for a 10Ω Cu point contact.) The Wexler resistance asymptotically approaches the Sharvin resistance in the large Knudsen number limit.

from.

\[ \frac{dR}{dV} = \Gamma(K) \rho \lambda d \frac{1}{\lambda(V)} \]  \hspace{1cm} (4.12)

The total scattering length of the electrons is determined by the elastic scattering length with the impurities and the inelastic scattering length with the phonons. The two scattering lengths for electron-impurity \( \lambda_{\text{imp}} \) and electron-phonon scattering \( \lambda_{\text{ep}} \) add up in accordance with Matthiessen’s rule. The energy \( \epsilon \) dependent part namely the electron-phonon scattering is related to the scattering time \( \tau_{\text{ep}} \) with this relationship.:  

\[ \frac{1}{\lambda} = \frac{1}{\lambda_{\text{imp}}} + \frac{1}{\lambda_{\text{ep}}} \quad , \quad \tau_{\text{ep}}(\epsilon) = \frac{\lambda(\epsilon)}{v_F} \]  \hspace{1cm} (4.13)

Using Fermi’s golden rule at 0 temperature to calculate the scattering rate, i.e \( \frac{1}{\tau} \) over the scattering time. One sees that the electron-phonon scattering rate changes with energy and the rate of change is proportional to the phonon density of states. If we plug in this result we come up with the final expression for the point contact
Figure 4.6: (First three solutions to Boltzmann transport equation for a metallic point contact junction) Leftmost is the zeroth order Sharvin solution with no back flow. The subsequent solutions are first order i.e. one collision and second order i.e. double collision back flow mechanisms that give rise to the energy dependent correction to the current.

\[
\frac{dR}{dV}(V) = \frac{\rho \lambda}{2a} \Gamma(K) \frac{2\pi e}{\hbar v_F} \alpha^2 F(eV)
\] (4.14)

In this equation \( \alpha \) is a geometrical factor and \( F(eV) \) is the phonon density of states. So what one measures in a point contact in the second derivative of voltage with respect to current is nothing but the phonon density of states of that metal times a geometrical factor.

This result can alternatively be derived from Boltzmann transport equation [18]. A schematic representation of the first three solutions are shown in Fig.4.6. The zeroth order solution is the Sharvin current which does not give any energy dependence. It is the first-order single collision back- flow and second-order double collision solutions so on and so forth that give rise to a nonlinear \( dR/dV \) signal.

Fig. 4.7 compares the phonon density of states of Au as measured from inelastic neutron scattering compared to point contact spectroscopy. As expected, there is a clear agreement in the voltage dependence of the signal with the frequencies of the
Figure 4.7: (Inelastic neutron scattering vs. point contact spectroscopy, from ref. [15], [19]) Phonon density of states of gold for transverse and longitudinal phonons as measured by inelastic neutron scattering (long-dashed line) and point contact spectroscopy (solid line). Background signal due to spontaneous phonon emission (short-dashed line).

The point contact spectrum shows a weaker coupling of the electrons with longitudinal phonons, and this is known to be the case for all noble-metal point contacts and is due to the d-band character of the noble metals being important for the electron-phonon interaction. By taking into account the hybridization of the s and d bands, the electrons couple strongly to the transverse phonons via umklapp processes. The non-equilibrium spontaneous phonon emission gives rise to a background signal which saturates above the Debye energy in the point contact spectrum.

In fact the point contact spectroscopy is not limited to probing electron-phonon interactions. The solution to Boltzmann equation says as long as the inelastic
scattering mean free path is large compared to the linear dimension $a$ of the contact i.e in the clean contact limit every interaction process of the conduction electrons can be studied such as electron-magnon scattering. In this work point contact spectroscopy has been used to check the electrical transport regime and investigate any correlations between the transport regime and the magnetoresistance due to a constricted domain wall. First we need a mechanism to manipulate the contact size in magnetic nanocontacts in order to be able to access all the transport regimes to systematically study the spin-dependent transport across the constrained domain wall. The mechanism that I have used is electromigration.

### 4.2 Electromigration

Electromigration is the net motion of atomic defects which results from a current density high enough to mobilize the atomic defects. A high current density in addition to the bulk electric field induced motion, also provides a momentum transfer from the electrons to the defect. The driving force of electromigration has two origins. One is the direct effect of the external bulk electric field on the charge of the migrating ion and the second one is the so called "electron wind force" term which describes the scattering of conduction electrons by the defect under consideration. These two potentially competing effects can be visualized as in Fig. 4.8.

Since the force due to electron wind is directly proportional to current one can write the total force acting on the defect as:

$$F = Z_0 e E + F_{\text{wind}} = Z^* e E \quad (4.15)$$

where $Z_0 e$ is the nominal charge of the defect and $Z^* e$ is the effective charge which
Figure 4.8: (Direct force and electron wind force) Two effective forces during electromigration are the direct force due to electrostatic interaction of the ion with the external field and the electron wind force due to momentum transfer during scattering of current carrying electrons.

is the sum of the defect charge and a contribution from the electron wind term. So for electromigration to occur a lot of electrons and a lot of electron scattering are needed. That’s why electromigration is not observed in semiconductors unless they are heavily doped.

Electromigration is one of the most important failure mechanisms in narrow interconnects. The microstructure parameters of the conductor such as grain size distribution, the distribution of grain boundary misorientation angles etc. cause a non-uniform distribution of atomic flow. A non-zero atomic flux divergence implies either mass accumulation (hillocks) or mass depletion (voids) in the conductor.

When a void starts forming, a thermally accelerated electromigration process takes place. As the cross-section of the contact gets smaller and smaller, local current density increases (current-crowding). Since joule heating is proportional to the square of the current density, a local heating effect further accelerates the
void formation. This thermal runaway process is summarized in Fig. 4.9.

There is also a back-flow of atoms during and/or after electromigration, that is in opposite direction to the electron wind force. This so-called ”healing effect” is a diffusion process due to inhomogeneities in the temperature and concentration gradients occurring via the electromigration damage.

In addition to the defects already in the sample the defects created during electromigration can sometimes be pinned and become unable to diffuse out of the sample region affected by electromigration. Such a generation of defects will make the sample more resistive to electromigration. On the other hand if the defects are readily mobile, they instead can enhance the electromigration. The mobility of defects at a given temperature correlates with the melting point of the sample. So, as a rule of thumb, irreversible electromigration starts when the effective sample temperature is above half the melting point. This means that metals with low melting points are more prone to electromigration even at relatively low bias voltages. For comparison Al has a melting point of 933.5K whereas Cu melts at 1356.6K. So Al is more susceptible to electromigration than Cu.

The devices used in this work were Ni-Ni and Co-Co nanocontacts. The contact
geometry was a bowl-shaped nanometer-sized hole defined on an insulating Si$_3$N$_4$ membrane by e-beam lithography and reactive ion etching techniques as described in Chapter 3. The metallization of the contact was achieved by DC magnetron sputtering with a base pressure of $5 \times 10^{-8}$ Torr, in a parallel in-plane field of 1200e, which provided a magnetic orientation to the growing film. A Cu or Pt seed layer was deposited first on the side of the membrane having the smaller opening to produce $<111>$ texture in the pinned layer. X-ray diffraction experiments confirmed $<111>$ preferred orientation for such films. This seed layer deposition was then followed by the deposition of the free magnetic layer on the opposite side of the membrane, which also filled the bowl. The fixed-ferromagnetic electrode was then deposited on top of the seed layer and was magnetically pinned by deposition of a succeeding antiferromagnetic pinning layer (FeMn). In order to be able to obtain significant exchange fields the thickness of the fixed layer (10nm) was chosen to be much smaller than that of the free layer (100nm)(see Fig. 4.10). The effectiveness of this pinning layer was further enhanced by thermally annealing the final device in field. In this manner nanocontact devices with various contact sizes (1-10nm) were fabricated through the use of different e-beam doses, which determined the size of the hole in the membrane which could also be manipulated through electromigration. The contact sizes were determined using the Sharvin resistance for ballistic contacts.

Once fabricated, the size of the nanocontacts could also be varied through the application of high current biases to the nanocontact, which results in further necking down of the contact through controlled-electromigration [20], [21]. Electromigration in this case is the net atomic flux due to local high-current densities which preferably proceeds through grain boundaries and surfaces. The schematic
Figure 4.10: A schematic representation of a nanocontact device formed by filling a bowl-shaped hole etched in a Si$_3$N$_4$ membrane. The top or bowl side of the nitride layer is covered by the free magnetic layer whereas the pinned magnetic layer is deposited on the bottom or flat side, which is first seeded by a 1-2 nm Cu or Pt layer. The free layer is 100nm thick whereas the pinned layer is 10nm. The pinning is achieved by a 50 nm FeMn antiferromagnetic layer.

drawing for electromigration setup and typical electromigration data are shown in Fig. 4.11 and Fig. 4.12.

The R-V data shows 2 distinct regions. As the current across the device is ramped up the initial increase in the resistance is at a relatively steady rate as opposed to the region where the current bias is above the threshold point where there is an accelerated rate of increase for the sample resistance. The first portion corresponds to electron heating due to electron-phonon scattering events whereas the second portion shows the characteristic electromigration behaviour. When the ramping process is stopped in the first region, the original resistance at low measurement biases is quickly recovered because the excess heat built up at the nanocontact gets dissipated by the surrounding thermal reservoir provided by the metal electrodes. Once the threshold for electromigration is exceeded, the atoms in the contact region gain enough mobility to move and hence the device resistance
Figure 4.11: Schematic Drawing of the Electromigration Setup. Voltage across the sample and a sense resistor in series with it is ramped up which acts to current bias the sample. The process is controlled by a computer which stops the ramping either when a preset resistance is reached or the rate of increase in resistance goes above a threshold value. This was found to be necessary to avoid blowing up the samples. The zero bias resistance of the sample is measured after each cycle.
Figure 4.12: Typical electromigration data for a Ni-Ni point contact is shown. The ions at the contact region are excited by momentum transfer from incoming electrons. Once the local temperature exceeds approximately half the melting point atoms start migrating along grain boundaries and surfaces under the influence of electron-wind leading to a permanent change in the sample resistance.
and the contact size changes permanently. Depending on the current direction it is possible to move atoms out of the contact point to generate voids which results in an increase in sample resistance, as well as pushing atoms to the contact region to fill in the voids which results in a decrease in sample resistance. This threshold current is of the order of $10^{10} - 10^{11}$ A/cm$^2$ for Ni and Co.

In a recent similar nice experiment by Bolotin et al. break junctions of Py were used and the contact size was similarly manipulated by electromigration [22]. This allowed a study of magnetoresistance both in the 1-10nm regime that is targeted in this work but also in the tunneling regime.

### 4.3 Magnetoresistance in Ferromagnetic Constrictions

As a result of irreversible electromigration to manipulate contact sizes or comparing devices with different contact sizes achieved by variation of e-beam exposure dose, it becomes possible to access different electron transport regimes. The character of the electron transport through these magnetic nanocontacts was established by measurement of their ”point contact spectra” $d^2V/dI^2$ (PCS). If the nanocontact diameter $c \gg \lambda$ the electron mean free path, the resistance is that of a diffusive contact $R_M = \rho/c$ where $\rho$ is the film resistivity. In this regime $d^2V/dI^2$ is featureless. For $c \leq \lambda$, transport through the contact becomes at least quasi-ballistic and $d^2V/dI^2$ begins to exhibit the phonon spectrum of the conductor as some of the ballistic electrons emit phonons during their transit through the contact regime and are inelastically backscattered. For $c < \lambda$ the contact resistance is the Sharvin resistance for ballistic contacts $R_S = \frac{16\rho\lambda}{\pi c^2}$. For near ballistic contacts ($c \simeq \lambda$) a good approximation is $R \sim R_M + R_S$ [17] as derived in section 4.1.

In Fig. 4.13 the point contact spectra for a 66Ω and for a 214Ω Ni nanocontact
Figure 4.13: The point contact spectrum of a Ni-Ni nanocontact at 4.2K is shown. The point contact spectrum of two Ni-Ni nanocontacts at 4.2K. The "zero bias anomaly,” which is particularly prominent in the 214Ω device is attributable to atomic scale structural defect instabilities in the nanocontact [24] and is seen to decrease in amplitude over time as the defects anneal away.

are shown. In both cases the rapid turn-on of the phonon emission by the electrons once the bias exceeds $\sim 10$ mV is clearly seen. The clearly resolved peak in $d^2V/dI^2$ for the 214Ω contact at $\sim 20$ mV coincides with the peak in the phonon density of states of Ni [23]. This indicates that for this contact $c < \lambda$ and from $R_S$ we obtain $c = 1.8nm$

From temperature dependent 4-point resistance measurements for 200nm thick sputtered Ni and Co films we can estimate the electron mean free path from:

$$\lambda = \frac{(r_s/a_0)}{\rho_\mu} \times 92\AA$$  \hspace{1cm} (4.16)
Table 4.1: T dependence of $\rho$ and $\lambda$ for Ni and Co

<table>
<thead>
<tr>
<th>$\rho_{Ni}(\mu\Omega.cm)$</th>
<th>$\lambda_{Ni}(nm)$</th>
<th>$\rho_{Co}(\mu\Omega.cm)$</th>
<th>$\lambda_{Co}(nm)$</th>
<th>T(K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>19.94</td>
<td>2</td>
<td>25.3</td>
<td>1.6</td>
<td>300</td>
</tr>
<tr>
<td>7.25</td>
<td>5.4</td>
<td>13.5</td>
<td>2.9</td>
<td>77</td>
</tr>
<tr>
<td>5.44</td>
<td>7.2</td>
<td>12</td>
<td>3.3</td>
<td>4.2</td>
</tr>
</tbody>
</table>

where $r_s$ is the radius of a sphere whose volume is equal to the volume per conduction electron and $a_0$ is the Bohr radius. (see ref. [25]). Table 4.1 summarizes the results of this calculation for sputtered Ni and Co films.

The bulk resistivities of Ni and Co at room temperature are around $6-7\mu\Omega.cm$ [26]. The high residual resistivity of sputtered films reflects the existence of a high impurity content. The zero-bias anomalies present at the point contact spectra (more so in Co than in Ni) are consistent with the presence of impurities and atomic defects around the contact region.

In-plane magnetic-field superconducting-quantum-interference-device magnetometry (SQUID) measurements on the samples reveal two hysteresis loops in the magnetic behavior, corresponding to the separate magnetic reversal of the pinned and free layers (see Fig. 4.15(a)). Since the in-plane dimensions of both electrodes are the same, the total magnetic moments measured are proportional to the electrode thicknesses. Thus, starting at high, saturating negative fields, at $+50$Oe the free layer switches its magnetization direction and the plateau between $+50$ and $+120$ Oe fields corresponds to the region when there is an antiparallel alignment between the two layers. At $+120$Oe the pinned layer switches its magnetization direction which leads to saturation at high positive fields. All these transitions occur in the reverse order as the field is increased in the negative direction. The
Figure 4.14: The point contact spectrum and bias dependent differential resistance of a ballistic Co nanocontact. The bottom graph shows the differential resistance as a function of bias voltage whereas the top graph demonstrates the second derivative of voltage with respect to current implying a fully ballistic Co nanocontact. Again zero bias anomalies are present. [23]
transitions corresponding to separate switching events of the magnetic layers are confirmed by the magnetoresistance measurements as illustrated in Fig. 4.15(b). Here, the typical exchange field as measured from the shift in the symmetry axis of the field scans was about 50 Oe. The switching events at the contact region that are detected by the magnetoresistance happen at slightly different fields, as compared to the switching of the bulk magnetic layers, due to the existence of an intermediate vortex state in the bowl as indicated by micromagnetic simulations (see Fig. 4.16). Given the small diameter c of the three dimensional contact, the device resistance is only sensitive to the magnetic state of the film within a distance c from the minimum contact cross section whereas the SQUID measurements are not sensitive to the magnetic configuration in the bowl. Thus the sharp transitions in the field scans reflect the domain wall formation and elimination at the nanocontact. In Fig. 4.17, the in-plane magnetoresistance signal obtained from a number of Ni and Co nanocontacts is plotted versus the nanocontact resistance. There is a general trend of the magnetoresistance increasing, more or less linearly, with contact resistance (decreasing contact area) but the maximum magnetoresistance change observed was only 0.45% and 0.25% in a 100Ω device for Ni and Co respectively. Assuming that, as indicated by the point contact spectra, all of the devices are in either the quasi-ballistic or ballistic regime when the device resistance is above 20Ω, the 20 – 100Ω resistance range corresponds to a contact diameter varying from 6 nm to 2.6 nm. It is interesting to compare these results with the domain wall magnetoresistance in bulk Ni calculated using band-structure arguments [27]. Such a calculation predicts a domain wall magnetoresistance of 0.11% from an unconstrained domain wall in Ni. Thus we conclude that for clean Ni and Co nanocontacts with minimum diameters as small
Figure 4.15: (a) Room-temperature SQUID magnetometry measurement of M vs H for a Ni-Ni point contact device. The thick and thin horizontal arrows represent the magnetization directions of the free and pinned layers respectively. The vertical arrows represent the field scan directions. There are two independent hysteresis loops for the free and fixed layers. (b) Room temperature magnetoresistance scan for a Ni-Ni nanocontact. The difference in the switching fields arises from an intermediate magnetic state in the bowl (see text).
Figure 4.16: Micromagnetic simulations of the magnetization configuration in the experimental geometry used displays a local energy minimum for vortex nucleation. This configuration is shown with the gray color scale together with the arrows indicate the domain structure.
Figure 4.17: Magnetoresistance as a function of device resistance is shown. Solid circles and solid squares are the data for Ni-Ni and Co-Co point contacts respectively. The dashed line is a linear fit to the data. In the ballistic transport regime, a 100 Ω device is expected to have a minimum contact diameter of about $\simeq 3\text{nm}$.
as 3nm, and with ballistic transport across the domain wall, although there is an enhanced magnetoresistance over the bulk values, there is no exceptionally large magnetoresistance effect.

### 4.4 Magnetoresistance in Atomically Sharp Ni Tips

Previous studies on geometrically constrained domain walls in Ni pioneered by Garcia et al. [9] resulted in several reports of extremely large magnetoresistance values (more than 100,000%. see ref. [6]). The experimental findings have been interpreted as being due to a ballistic magnetoresistance effect (BMR) where an atomically sharp domain wall pinned at a constriction site between two Ni electrodes provides a high reflection probability for spins depending on the relative orientation of the magnetizations of the two magnetic electrodes.

There are two very commonly used experimental geometries for the BMR type measurements of Ni nanocontacts. In the first geometry [9] (shown in Fig. 4.18) two Ni wires with 2mm diameter with round tips are put into contact. The wires are glued by epoxy resin onto a teflon tube to give them rigidity. A force is applied until a contact of a few atom size (\(<\sim 1\text{nm}\)) is formed. The wires have two coils at the ends with a square wave being applied to one coil whereas a constant direct current is applied to the other to be able to manipulate the magnetization orientation in one of the Ni wires but not the other. The resistance of the nanocontact is monitored with an oscilloscope as a function of changes in the applied field. With this technique magnetoresistance values over 200% have been obtained.

The most important concern in terms of possible artifacts in these measurements is contact deformation either induced by magnetostatic forces or magnetostriction. In the first conventional geometry because of the large shape anisotropy
Figure 4.18: First BMR experiment setup by Garcia et al. (from ref. [9]). Two Ni wires glued onto a teflon tube are put into contact and their magnetizations are manipulated by applying current to coils present at the end of the wires. The resistance change of the nanocontact at the tip correlated with an applied square wave magnetic field is monitored with an oscilloscope.
**Figure 4.19:** Artifacts due to magnetostatic forces (from ref. [12]). A resistance change that mimics the BMR signal can be obtained by the consecutive stretch-retract cycle due to large magnetostatic forces at the contact region.

In the wire geometry, in addition to the applied field direction, the magnetic configuration of the electrodes are most likely to be either parallel or antiparallel. This implies that a 180° domain wall is expected to nucleate at the contact region. This eliminates the possibility of magnetostriction playing a role since there is no expected elongation or contraction of the contact region for parallel and antiparallel states. However a detailed analysis of the magnetostatic forces carried out by Egelhoff et al. [12] has shown that a stretching and retraction cycle of 1-3nm at the contact region should be expected due to large magnetostatic forces (about 0.01N) that develop at the contact region (see Fig. 4.19). Unfortunately the modulus of elasticity for epoxy is about 100 times smaller than Ni which implies that epoxy can largely deform to accommodate the Ni-Ni contact deformation.

In the second common geometry the nanocontact is formed between two Ni wires in a T configuration [6]. Ni is electrodeposited at the gap (a few microns in size) between the sharp tip (40-400nm in diameter) and the other Ni electrode. The resistance of the junction is monitored and is controlled by a feedback system.
The envisioned domain configuration at the T junction nanocontact consists of a parallel state at zero field which by application of an axial field leads to a 90° domain wall which is supposedly very narrow (see Fig. 4.21). Unfortunately, on top of magnetostatic forces that develop at the contact region, this geometry suffers from artifacts due to magnetostriction. The magnetostriction coefficient for bulk polycrystalline Ni is $-34 \times 10^{-6}$ which implies that as a result of magnetization rotation needed to make it parallel to the applied field in a 4mm long wire one would expect 136nm contraction (see Fig. 4.22). This type of a deformation at high fields in the contact region would indeed cause a large increase in the resistance as can be seen from the typical data shown in Fig. 4.21 that mimics the BMR effect.

To be able to study magnetoresistance due to a constrained domain wall systematically avoiding magnetostatic and magnetostriction artifacts as well as to assure absence of contamination at the contact region, one needs to work with a nanocontact prepared in a vacuum environment. Also the domain configuration at
Figure 4.21: A domain configuration model for T junctions (from ref. [28]) A possible domain configuration that could potentially explain the BMR data for nanocontacts in the T configuration.

Figure 4.22: Magnetostriction in a T type magnetic nanocontact (from ref. [12]) Magnetostrictive forces that develop at high fields in the T junction is shown. This can lead to a deformation as large as 136nm.
the contact region needs to be well-understood. The exchange bias scheme used to establish a $180^\circ$ domain wall in a 3D bowl shaped nanocontact meets all of these requirements. However as we shall see in the next two sections it is very hard to achieve a one atom size point contact and there are limitations due to $1/f$ noise. In a similar experiment by Bolotin et al. [22] two elliptical permalloy ($Ni_{81}Fe_{19}$) nanodots were used in a break junction with further contact size control through electromigration. The shape anisotropy provided by the elliptical shape serves to establish a well-defined $180^\circ$ domain wall. On the other hand permalloy is a well-known soft magnet with practically zero magnetostriction. The results of this experiment summarized in Fig. 4.23 also confirm that there is no large BMR effect in the metallic conduction regime. However a reasonable magnetoresistance value can be obtained in the tunneling regime due to tunneling magnetoresistance (TMR) effect. Similar results for Ni have also been obtained by Keane et al. [29].

4.5 Giant Surface Magnetostriction

Through the course of the experiment on Ni-Ni contacts in the bowl-shaped geometry we have discovered two factors that can be important when measuring ferromagnetic nanocontact samples. The first one is the giant surface magnetostriction effect of thin Ni films, which depends on the growth conditions, and the second one appears to be a $1/f$ noise issue (see section 4.6).

In most cases when the contact sizes were less than 3 nm the resistance signal was dominated by $1/f$ noise arising from atomic defect fluctuations in these very small, mechanically stressed nanocontacts [20]. The exception was in the case where a thin Cu layer was deposited instead of Pt to seed the Ni film growth. Due to the greatly enhanced surface magnetostriction effect that results from the use
Figure 4.23: Magnetoresistance vs resistance for permalloy nanocontacts (from ref. [22]). Magnetoresistance versus resistance in permalloy nanocontacts both in the low resistance regime (top) and in the high resistance regime (bottom) is shown. The inset shows typical magnetoresistance scans.
of this type of seed, the Cu seeded Ni nanocontacts were subjected to very high stress, and underwent plastic deformation as they were cycled through several field reversals. If we compare the surface free energies of Ni, Pt and Cu calculated from their internal free enthalpies of atomization (see ref. [30]), at room temperature they are 2691, 2364 and 1934 $\text{J/m}^2$ for Pt, Ni and Cu respectively. Recalling that when the surface energy of the substrate is large compared to the deposited film good film adhesion is expected (see ref. [31]), good wetting properties for Ni films deposited on Pt substrates are expected whereas Ni films deposited on Cu substrates are under more tensile stress.

This effect was the strongest in samples that had been electromigrated, which resulted in void formation at the contact and induced a perpendicular anisotropy at the contact region. In those cases when a parallel in-plane field was swept back and forth the magnetostriction forces building up across the voids, accompanied by the

\textbf{Figure 4.24:} A schematic drawing of a possible magnetic orientation that develops in a Ni-Ni nanocontact that allows magnetostriction forces to deform the contact during field scans.
existence of a tensile stress were high enough to gradually pull the nanocontact apart (see Fig. 4.24). This resulted in a continuous increase in resistance (see Fig. 4.25) during the application of the field due to the substrate dependent giant surface magnetostriction of Ni [32]. Such magnetostrictive effects were not observed in Pt seeded Ni samples [33].

The fact that magnetostriction behaviour of magnetic thin films may differ from that of the bulk has previously been shown to be highly dependent on the choice of the substrate [32], [33]. For instance for polycrystalline Ni films deposited on SiO$_2$, below 10 nm thickness, the magnetostriction can be enhanced as much as by a factor of 3 as compared to the bulk value. This enhancement may as well be due to microstructural changes near the surface and/or due to intrinsic effects associated with the electronic structure in the highly strained, low-symmetry surface environment. However for Ni films on 3Å of Pt, it has been shown that the magnetostriction is driven more positive as the film thickness is reduced and becomes zero at about 7Å of film thickness [33]. As the contact size is reduced through electromigration, the surface effects become more and more pronounced. Since Ni has negative magnetostriction regardless of the crystal direction, a parallel in plane field acts against the local shape anisotropy to induce magnetostriction forces which tend to shrink the contact parallel to its radial axis and pull it apart in the perpendicular direction.

### 4.6 1/f noise

The systematic study of magnetoresistance versus contact size that I set to carry out in this experiment was limited by noise in the smallest contact sizes. The signal to noise ratio gradually decreased as the contacts were electromigrated and
Figure 4.25: An example of contact deformation in electromigrated Cu seeded Ni nanocontacts. Resistance versus field and time are shown. The continuous increase in the resistance is irreversible and implies a change in the contact geometry.
Figure 4.26: A magnetoresistance scan of a 100Ω Ni-Ni nanocontact that demonstrates the obstruction of the measurement of the small magnetoresistance signal by the presence of a strong 1/f noise.
made it impossible to unambiguously measure the magnetoresistance signal. An example magnetoresistance scan for a 100Ω Ni-Ni nanocontact is shown in Fig.4.26. Further investigations of the source of this noise with a spectrum analyzer proved that the noise was intrinsic to the samples and had a $1/f$ character as shown in Fig. 4.27 for a 1nm diameter Co-Co nanocontact. Nevertheless from the trend in Fig. 4.17 we see that the magnetoresistance signal more or less increases linearly with sample resistance. If we were to extrapolate this data to the single electron conduction channel regime the resulting magnetoresistance behaviour when it is normalized to its maximum value is not too far off when compared to results from a selection of experiments (see ref. [34] and Fig. 4.28)

Metallic nanocontacts are sensitive to individual defect fluctuations that manifest themselves as fluctuations in the number of electron channels available for conduction through the aperture. If we define $S_R$ as the noise power spectral density then the noise amplitude $S_R/R^2$ has previously been shown to vary as $R^{1.5}$ whereas the expected behavior for a constant defect density is $R^{1/2}$ (see ref. [21] and Fig. 4.29). This behavior suggests an increase in the defect density at the contact region in the smallest nanocontacts, possibly due to stress. Since the magnetoresistance signal measured in Ni-Ni and Co-Co nanocontacts increase linearly with the contact resistance it becomes clear that the signal to noise ratio should indeed decrease for the smallest nanocontacts. The defect fluctuations between metastable configurations give rise to a $1/f$ spectrum that dominates the magnetoresistance signal for contact diameters less than 3nm. It seems that the break junction geometry used in similar experiments are preferable for studies in the atomic size limit in terms of stress induced changes in the number of defects that contribute to a $1/f$ noise source.
Figure 4.27: Noise level as a function of frequency in a 1nm diameter Co-Co nanocontact
Figure 4.28: A comparison of the results of magnetoconductance measurements (from ref. [34]) from a selected set of sources with an extrapolation of the data on Ni-Ni and Co-Co nanocontacts to the single electron channel regime.
Figure 4.29: $1/f$ noise magnitude vs sample resistance for a Cu-Cu nanocontact (from ref. [21].) As the contact size decreases i.e. the sample resistance increases the noise level increases with an $R^{1.5}$ rule.
References for Chapter 4


Nanocontacts to extended metallic thin films have long been used for fundamental studies of spin dependent transport in magnetic multilayers in the Buhrman and Ralph groups at Cornell. Of special interest has been nanocontact studies of current perpendicular to plane giant magnetoresistance (CPP-GMR) effect due to spin-dependent scattering (see ref. [1]) and of spin momentum transfer due to a spin torque that develops at the interface between a spin polarized current and a thin magnetic layer (see ref. [2]). However the bowl-shaped nanocontact geometry defined on a nitride membrane has its own limitations for such studies.

The CPP-GMR signal obtained from point contact measurements of spin valves turn out to be an order of magnitude larger than the domain wall magnetoresistance signal in magnetic nanocontacts. However the acquired signal does not scale with the contact area if one considers the $\Delta RA$ product. A good example is a Co/Cu/Co trilayer structure whose $\Delta RA$ product has been measured in nanopillars to be $\sim 0.5 m\Omega \cdot \mu m^2$ for a $\sim 3nm$ thick Co free layer (see ref. [3]). For a 10nm diameter Cu nanocontact to a Co/Cu/Co trilayer one would expect a $\Delta R$ signal of 5$\Omega$. The results of a magnetoresistance scan for such a device is shown in Fig. 5.1 (from ref. [4]). The measured $\Delta R$ value is 0.4$\Omega$ from the nanocontact with a 10 nm diameter with a magnetoresistance of 3%. The magnitude of the magnetoresistance signal is about an order of magnitude smaller than one would expect from a cylindrical 10nm diameter device.

There are two main reasons for this reduction in the magnetoresistance signal, namely the interfacial backscattering of electrons and current spreading. The first one is due to the fact that not all electrons that undergo spin dependent scattering
Figure 5.1: Magnetoresistance of a Co/Cu/Co spin valve point contact with a 4nm thick Co free layer (from ref. [4]). The device schematic is shown at the bottom. The resulting $\Delta R$ signal (top) is $0.4\Omega$. 
Figure 5.2: A schematic drawing of current spreading under a point contact in a Co/Cu/Co spin valve for transmitted electrons with an average incident angle $\gamma$. The one dimensional flow of all electrons shown by the white cylindrical area is unrealistic.

at the ferromagnet/normal metal interface make it back through the nanocontact but instead can go through multiple scattering events at the Cu-Co interfaces. The inability of the point contact to detect every spin-dependent scattering at the relevant interfaces causes a loss of $\Delta R$ signal. A second important factor is the fact the incident electrons at the nanocontact come from several different angles and those coming with a large incident angle will cause a spreading of the current underneath the nanocontact. The electron trajectories under the conditions of ballistic transport will be more or less linear but not necessarily perpendicular to the nanocontact. This spreading of the current is schematically shown for transmitted electrons in Fig. 5.2. Accompanied with the loss of the $\Delta R$ signal, there is a more important problem with the non-ideal shape of the magnetoresistance curves. It becomes clear from Fig. 5.1 that the device is far from acting like a
single-domain nanoparticle. The gradual relaxation of the magnetization at high fields as well as signs of multiple-step magnetic switching point to existence of stress-induced defects (also causing 1/f noise as seen in section 4.6) at the contact region that act as pinning centers and cause non-abrupt switching characteristics. For experiments that involve a magnetic layer deposited in the bowl, from micromagnetic simulations (see Fig. 4.16) it becomes clear that vortex nucleation and annihilation may be involved in the switching process which would also cause non-abrupt transitions in the switching characteristics. To get a more ideal magnetic configuration at the contact region the thickness of the nitride membrane needs to be comparable to or less than the contact size. However experiments on thin low-stress nitride membranes have shown that the minimum reliable pinhole free nitride thickness attainable is 30nm.

Optimization of current-induced switching characteristics of nanocontacts to extended magnetic multilayers also are subject to limitations due to the fact that the spin-momentum transfer effect results in a domain switching process where the spin torque has to overcome the exchange torque due to neighboring domains in the extended film. Although this is possible (see ref. [5]), the critical current densities required to observe switching are an order of magnitude larger than what is required to switch a single domain in a nanomagnet. A good example is a Co/Cu/Co spin valve where the experimental threshold current for switching is $\sim 10^9 A/cm^2$ [5] whereas it is $\sim 10^8 A/cm^2$ for the same structure patterned to a nanopillar [3]. This coupled with the fact that the GMR values are intrinsically low makes the point-contact geometry non-ideal for magnetic memory applications. A better approach that would improve the GMR of point contacts by reducing current spreading and backscattering effects would be to bring the true spin-dependent
scattering interfaces as close as possible to the nanocontact as well as to provide a current concentration mechanism in a nanomagnet to reduce the absolute current values needed to observe spin transfer switching. Such a geometry is shown in Fig. 5.3. The devices used in this experiment (see Chapter 3 section 3.2 for fabrication details) are designed to pursue this goal.

5.1 How to Reduce Spin Transfer Switching Currents

Manipulation of the thin film magnetization by transferring spin angular momentum from a spin-polarized current in nanomagnets both to drive precessional dynamics and to trigger magnetization reversal presents promising opportunities for better scalability characteristics in nanoscale microwave oscillators and magnetic memory devices such as magnetic random access memory (MRAM). However one of the major practical concerns is the current level needed to write magnetic bits in an error-free fashion at high operating speeds. This indeed poses a limit on the areal density one could achieve in a memory circuit using spin transfer because the required current level determines the size of the write transistor needed. This potential technological applicability has triggered an intense effort to optimize the device geometry and choice of materials to obtain reduction in switching currents.
To face the challenge of reducing critical currents needed for spin transfer switching we need to review what determines the switching speed and the switching currents of a nanomagnet in this scheme. Following a single-domain model by J. Z. Sun (ref. [6]) the switching speed \( \frac{1}{\tau} \) where \( \tau \) is the switching time should scale linearly with \( |I - I_{c0}| \ell \ln(\theta_0) \) where \( I \) is the applied pulse current level, \( I_{c0} \) is the zero temperature threshold current for instability and \( \theta_0 \) is the initial misalignment angle between the current polarization and the macrospin. The linear scaling of the switching speed with the applied current level has indeed been shown experimentally using time domain pulse measurements of a permalloy nanomagnet (see [7]). It becomes clear that fast switching can be observed by overdriving the precession with higher applied current levels. However for a given current level to implement fast switching, an increase in the initial misalignment angle \( \theta_0 \) and/or a decrease in the current levels required to achieve onset of dynamic instability \( I_{c0} \) would suffice. The onset of zero temperature dynamic instability on the other hand is given by:

\[
I_{c0} = \frac{\alpha 2e}{\eta h} V M_s (H + H_k + 2\pi M_s)
\] (5.1)

where \( \alpha \) is the damping parameter, \( \eta \) is the polarization, \( V \) is the volume, \( H \) is applied field, \( H_k \) is uniaxial anisotropy field and \( 2\pi M_s \) is the demagnetization field where \( M_s \) is the saturation magnetization. This equation implies that a reduction in \( I_{c0} \) is possible with proper choice of material parameters and device geometry. One can group the experimental strategies to reduce critical currents to two categories, one that involves initialization of reversal by either applied or built-in fields at an angle and the other one that involves changes in the device structure and geometry.

One way to achieve a non-zero misalignment angle between the free layer mag-
Figure 5.4: Using exchange-bias to reduce critical currents (from ref. [8]).
(a) The spin valve GMR effect is used to calculate the equilibrium misalignment angle between the exchange biased and free Py layers. (b) Time-domain measurements of turn on times for precessional dynamics.
netization and the current polarization is using exchange bias with an antiferromagnet in proximity to the fixed layer to induce an easy direction misaligned with the easy axis defined by shape anisotropy. This strategy has been used in an experiment by Krivorotov et al. [8] to demonstrate the effect of the misalignment angle on the turn-on times of steady state precessional modes by time-resolved studies. In this experiment the equilibrium orientation of the free layer was controlled by shape anistropy provided by a 60 x 130 nm elliptical pattern whereas the fixed layer equilibrium magnetization orientation was determined by the combined effects of a 45° exchange bias with IrMn, the shape anisotropy and dipolar coupling between the two layers. The device structure is shown in the inset to Fig. 5.4 (a). The equilibrium misalignment angle was measured from the spin valve GMR effect and a subsequent fit to a Stoner-Wohlfarth model to be 30°. Fig. 5.4 (b) shows the results of a time-domain measurement of the voltage oscillations in response to a voltage step. It has been shown that sub-ns turn-on times with applied currents of 6-7mA and 10ns pulse duration (with a rise time of less than 0.1ns) can be achieved and increasing the offset angle does indeed reduce the response time but the read-out signal to noise ratio is also reduced due to a decrease in the GMR signal.

Another way to induce offset angle between the free layer magnetization and the current polarization is combining nano-second pulses with fields applied along the hard axis of the device. An experiment along these lines was implemented by Devolder et al. [9]. The pre-establishment of an offset angle with a hard-axis field increases the spin transfer torque susceptibility and ensures a uniform spatial response of the magnetization. A plot of the pulse current level needed for error-free switching as a function of pulse duration and applied hard axis field is shown
Figure 5.5: Using hard-axis fields to reduce critical currents (from ref. [9]). The pulse current levels and durations needed for error-free switching of a 1nm CoFe/1.8nm Py free layer in the presence of different hard-axis fields is shown.

in Fig. 5.5. When more than a fifth of the anisotropy field is applied along the hard axis a significant reduction in the critical currents can be obtained but potentially at the cost of reducing the magnetic stability. An alternative scheme utilizes a DC precharging current to excite the magnetization to a precession trajectory and thereby accelerate the reversal induced by a subsequent current pulse (see ref. [10]). However the polarity of the precharging current has to be the same with that of the current pulse to accelerate reversal. Using an AC magnetic field is also possible (see ref. [11]) to replace the precharging current to overcome this limitation but probably is not very practical in a real memory device.

There have been several experiments in the second category that involve material exploration and changes in the device geometry in an effort to reduce critical currents. In the work by Braganca et al. (see ref. [12]) permalloy and permalloy-copper alloys, being low $M_s$ materials, were tested for reduction in critical currents in a $40 \times 120 \text{nm}^2$, small but high aspect ratio ellipse (for details about the fabrica-
Combining a magnetic tunnel junction with a spin valve to boost the total spin torque incident on a free layer has been demonstrated by Fuchs et al. (see ref. [14]). This approach not only proves to be beneficial in terms of reducing critical currents for anti-aligned fixed layers but also results in an increase in the read-out signal.
Figure 5.7: (a) Top-SEM image of a 150x250 nm$^2$ nanopillar. Bottom- AFM image of a 20nm hole in e-beam resist. (b) A cross sectional view of the nanoscale point contact device where the nano-aperture is aligned to be at the center of the pillar.

with a large TMR effect. In this scheme when the two fixed layers are antiparallel and electrons are tunnel injected from the fixed layer to the free layer (see Fig. 5.6), the transmitted spin polarization from the fixed layer adjacent to the tunnel junction and the reflected spin polarization from the fixed layer that is part of the spin valve add up to facilitate spin-torque switching of the free layer resulting in a $20 - 60\%$ reduction in switching currents (for details see [15]). However, using spin torque switching in tunnel junctions has its own challenges such as a substantial heating effect and the limitation on current levels that can be applied before reaching the high voltage breakdown limit.

In this work we have developed a fabrication technique to insert a nano-aperture inside a nanopillar (the fabrication details for this device were presented in Chapter
3). The device schematic and an SEM image of the pillar and an AFM image of a nano-aperture are shown in Fig. 5.7. This geometry allows the concentrated, local injection of a spin-polarized current into a nanomagnet, as well as the local probing of the result of the spin transfer effect via the spin-valve magnetoresistance effect. The strong concentrated spin torque effect potentially enables a new nonuniform spin torque switching mechanism. Indeed I present in section 5.3 that this localized injection results in a novel, efficient switching mechanism with as much as two orders of magnitude reduction in critical currents, albeit possibly at the cost of a decreased switching speed.

5.2 Point Contact Oscillators

Aside from spin transfer induced magnetization reversal, there is also another important effect that occurs under the application of a spin polarized current in the presence of an applied field that is large enough to effectively oppose hysteretic switching. In this case the spin transfer torque can drive steady state magnetic precession. In the early investigations of the spin transfer effect in the point contact geometry, the peaks and valleys that can be seen in the differential resistance were attributed to the onset of such precessional dynamics and/or generation of spin waves under the contact region (see ref. [5]). Similar excitations have been observed in nanopillar spin valves and studied extensively both in the time domain (ref. [8]) and frequency domain (ref. [16]). For a detailed study of microwave oscillations in the nanopillar geometry see ref. [17]. Such nanoscale microwave oscillators whose frequency can be tuned with applied currents and fields and that can be integrated with semiconductor electronics potentially could have important applications in the telecommunication industry.
Figure 5.8: Spin Transfer Induced Microwave Dynamics in a Point Contact (from ref. [18]) (a) Differential resistance versus current scan at 0.1T applied in-plane field (b) The corresponding frequency spectrum (inset) Excitation frequency as a function of current.
The first direct observation of coherent high-frequency magnetization dynamics induced by a spin-polarized dc current in the point-contact geometry was achieved by Rippard et al. (ref. [18]). A 40nm diameter nanoaperture was used to inject current into a $Ni_{80}Fe_{20}(\text{permalloy})(5nm)/Cu(6nm)/Co_{90}Fe_{10}(20nm)$ where the thin permalloy layer acts as the free layer and the thick CoFe layer acts as the fixed reference layer due to its larger volume, exchange stiffness and saturation magnetization. Fig. 5.8 (a) shows a typical spin transfer scan at 0.1T applied in plane field that makes magnetic orientation of the two layers be parallel at zero applied current. The peak observed at around 4mA corresponds to what had previously been interpreted as the transition from a static parallel state to a dynamic precession state. This peak in the differential resistance is nothing but the result of a measurement of the derivative of a step function for the static to dynamic transition. The observed dynamic response is only for the direction of electron flow from the free to the fixed layer such that the reflected spins at the fixed layer/spacer interface apply a torque at the free layer to drive it out of the static equilibrium by cancelling out the damping torque. The direct evidence that these non-hysteretic peaks correspond to onset of current induced microwave dynamics is seen in Fig. 5.8 (b) which shows the frequency spectrum of the voltage signal that is directly measured with a spectrum analyzer.

As seen in the inset of Fig. 5.8 (b) the frequency of the dynamics shows a linear dependence on the applied dc-current. In general the peak frequency shows a red shift with applied current for in plane applied fields whereas an out of plane field results in a blue shift of peak frequencies with current. From the field dependence of frequencies fit to Kittel equation (ref. [19]) for in-plane magnon generation a magnon wavelength of about 400nm is obtained. This implies that the spin transfer
excites long wavelength spin waves under the contact region.

In a similar experiment (ref. [20]) where a point contact was used to excite dynamics in a Co/Cu multilayer it was shown that the requirement of an applied field larger than the anisotropy field to impose parallel alignment is not a necessary condition but using interlayer exchange coupling that strongly favors a parallel alignment is just as effective to observe microwave dynamics in these devices where the spin torque is applied in a direction that favors antiparallel alignment. Fig. 5.9 shows the field dependence of the onset of dynamics on the applied in-plane field for these devices at the inset along with the differential resistance scans that give indirect evidence for the onset of dynamics. This important observation is encouraging for future experiments to eliminate the need to apply external fields by using built-in fields that originate from dipolar coupling and/or exchange bias.
with an antiferromagnet.

Experiments in which the out-of-plane angle of the applied field is varied ref. [21]) showed that for mostly out-of-plane fields (more than 55° out of plane) the frequency dependence on current is more complicated and starts showing a blue shift for large applied currents as well as frequency jumps. Single-domain simulations indicate that the fixed layer also starts precessing for large out-of-plane fields that causes the free layer precession frequency to increase. An important observation for out-of-plane applied fields is that despite the more complicated dependence of precession frequency on current, this scheme could be more beneficial for applications since the power output also increases by two orders of magnitude. All the above experiments proved that highly coherent magnetization dynamics in the magnetic point contact geometry can be obtained in a 1-40 GHz frequency range with narrow linewidths (1-50 MHz) which gives quality factors \( f/\Delta f \) exceeding 10000 that can be tuned with applied field strength, direction and applied DC current.

A finite-temperature single-domain model calculation based on Sloncweski theory (ref. [23]) has been performed by Russek et al. (ref. [22]) to gain more insights into the expected excitation spectra and linewidths. The temperature is added in as a random thermal field whose squared rms value is directly proportional to the temperature and Gilbert damping whereas inversely proportional to the saturation magnetization and the sample volume (ref. [24]). The results show that temperature plays an important role in determining the linewidths by two mechanisms; by inducing disorder in the magnetization orbits and/or by hopping between degenerate orbits via thermal fluctuations. Sankey et al. ( [25]) has also found that thermal fluctuations of the precession angle at low temperatures and
Figure 5.10: Macrospin Simulations of Magnetization Dynamics in Nanoscale Spin-Transfer Oscillator (from ref. [22]). Excitation spectra for a 2.5nm x 50nm x 100nm permalloy nanomagnet with a 50mT applied field along the easy axis at room temperature (a) and (b) x and y components of the magnetization respectively. (c) (d) (e) and (f) are the calculated time evolution of the magnetization as a function of increasing applied current level.
the thermally activated mode switching at high temperatures play the dominant role in determining the linewidths. Since a smaller magnet will be more prone to thermal fluctuations the difference in the excitation volume between nanopillars and nanocontacts can possibly account at least qualitatively for smaller linewidths observed in the latter.

The calculated magnetization trajectories of the free layer as a function of applied DC current level for a 2.5nm x 50nm x100nm Py/Cu/Py CPP spin valve for 50mT applied in-plane field is shown in Fig. 5.10. There are three distinguishable regions. Small applied currents (less than 0.6mA) results in thermally activated elliptical motion. Intermediate current levels (1-4 mA) yield bent elliptical motion with a bistable trajectory which leads to degeneracy for even higher currents. The thermal hopping between these degenerate orbits gives rise to a low frequency noise. At high enough currents a tilted out of plane orbit is obtained. The oscillation frequency is found to red-shift with increasing current until degeneracy is fully established and the linewidths are broadened due to hopping between the degenerate orbits. None of the calculated orbits are perfectly harmonic and they have been found to be very sensitive to the misalignment between the fixed and free layer magnetizations.

The above calculations give valuable qualitative insights into the physics of current induced microwave oscillations. However a lot of the features in the experimental data such as frequency jumps and more complicated dependencies of frequency on current and field are not accounted for. This points to the fact that the micromagnetic structure involving the complex coupling between fixed layer and free layer dynamics also play an important role. Other secondary effects such as the Oersted field, the angular dependence of damping and spin-transfer
efficiency, structural defects and non-uniform response of the magnetization due to antiferromagnetic coupling with sidewall oxides at the edges of nanomagnets in the case of nanopillars also need to be considered to gain a better handle on optimization of the microwave dynamics for applications.

For communication applications where the oscillator provides a reference signal to carry information some type of modulation scheme has to be used. The frequency modulation of the spin transfer point contact oscillators was investigated by Pufall et al. (ref. [26]). When a small 40MHz ac signal is injected through the capacitor branch of a bias tee along with the DC current coming through the inductor branch the resulting excitation spectrum displays sidebands whose power and amplitude can be tuned by adjusting the drive amplitude. By biasing the oscillator in a regime where the frequency changes in a non-linear fashion with applied DC current, an asymmetric sideband suppression can be obtained. Changing the drive amplitude was found to shift the center frequency as well. This can qualitatively be understood from the macrospin simulations which suggest that the change in the frequency of precession is due to a change in the effective field through changes in the demag field. The demag field is modified since the drive signal acts to modify the cone angle of precession of the magnetization.

If instead of applying a small AC signal with frequencies in the MHz range, one applies an RF signal whose frequency is close to the free running frequency of the point contact oscillator, the oscillator frequency locks into the drive frequency (ref. [27]). Fig. 5.11 (a),(b) and (c) shows the excitation frequency, differential resistance and power output versus DC current for a free running point contact oscillator. When a 10.86GHz rf drive is added injection locking is observed in (d),(e) and (f). The locked range shows up as a broad peak in the rectified voltage
**Figure 5.11**: Injection Locking of Nanocontact oscillators (from ref. [27]) (a), (b) and (c) Frequency, differential resistance and output power as a function of applied dc only current (d), (e) and (f) the same as (a) (b) and (c) but in the presence of a 10.86 GHz ac drive. (inset) Device and measurement schematic
output. The phase (relative angle of the oscillator signal with respect to the injected ac signal can be tuned by changing the applied DC current between \(+90^\circ\) to \(-90^\circ\).

Another important result that has been published by two independent groups (ref. [28], [29]) is that large coherent power can be obtained from an array of spin transfer point contact oscillators via phase locking of the radiated spin waves. Fig. 5.12 shows the individual nanocontact oscillator responses and the phase locked signal when DC current is injected in parallel to both oscillators in proximity. A large output power can be obtained using a phase controller to tune the power to its optimum value. Studies using focused ion milling to separate the two nanocontacts ( [28]) have shown that the coupling between the oscillators is spin-wave-mediated rather than by any ac fields generated by the time varying magnetizations. In fact the spin waves emitted under one point contact can be detected by measuring the GMR of another point contact in proximity. The spin wave coupling length scale is of the order of a few 100nms and phase locking can still be obtained for contact sizes as large as 300nm in diameter ( [30]).

The ability to tune the oscillation frequency and phase accompanied by synchronization of multiple oscillators to generate large output powers makes these nanoscale oscillators ideal candidates for various applications such as telecommunication. In this experiment although the dynamic response of point contacts embedded in nanopillars was not the main focus, it certainly is a potentially interesting system to investigate whether or not more coherent modes can be excited by nonuniform injection into patterned nanomagnets. A related question whether or not one can achieve phase locking in nanopillars through dipolar coupling is currently under investigation (see ref. [13]).
Figure 5.12: Phase Locking of Double Point Contact Oscillators (from ref. [28]) (a) Frequency versus current characteristics of individual point contacts with 0.8T 80 degrees out-of-plane field (inset) power spectral density as a function of frequency with a Lorentzian fit. (b) Combined spectra of two point contacts with one at constant bias (c) Comparison of output powers from individual contacts and different summing schemes. The measurement scheme of phase locked oscillators
5.3 Low Temperature Measurements

The devices of interest in the current work are 150$x$250nm$^2$ elliptical nanopillar multilayers that consist of 20nm Py/8nm Cu/5nm Py spin valve with a 20-30nm diameter nanohole located between the spacer layer and the thin, "free" Py layer. The thick Py layer acts as the reference for all magnetoresistance measurements because of its larger coercivity due to the larger volume. The elliptical shape helps improve the device stability with the help of shape anisotropy (uniaxial). The results of a 2D micromagnetic calculation of anisotropy field versus aspect ratio for a 40nm minor axis ellipse is shown in Fig. 5.13. This trend is qualitatively observed in experiments with elliptical nanomagnets. However for quantitative agreement more detailed simulations taking into account the exact device geometry as well as nonuniform dipole coupling with the fixed layer should be performed. The device schematic is shown in Fig. 5.7. In this case the small aspect ratio causes thermal instability at room temperature.

All measurements were taken at 4.2K to ensure thermal stability of the relatively low aspect ratio free layer nanomagnet. We examined 15 samples, whose resistances varied between 10 to 15 Ω, as compared to the 3-5 Ω resistance exhibited by 100$x$200nm$^2$ elliptical spin valve nanopillars with the same magnetic multilayer structure but without the oxide aperture. This indicates that the device resistance in the former case is indeed determined by the aperture size rather than by the pillar area.

The minor hysteresis loop of the free layer for a sample with a nano-aperture is shown in the inset to Fig. 5.14(a). This was generated by fixing the thick Py layer magnetization by applying a large external field $H_{ex}$ and then sweeping $H_{ex}$ back and forth about the value that cancels the average dipole field on the free layer
Figure 5.13: The results of a 2D micromagnetic calculation of anisotropy field versus aspect ration for a 5nm thick elliptical permalloy nanomagnet with 40nm minor axis.

Figure 5.14(a) shows a resistance vs current scan taken at 4.2K of the same device taken with $H_{ex}$ that on average cancels $H_d$. The measured spin-transfer switching
Figure 5.14: (a) A typical $dV/dI$ vs $I$ scan taken at 4.2K with an applied field cancelling the average dipolar field from the fixed layer, displaying AP-P switching at $-50\mu A$ and P-to-AP switching at $180\mu A$. Inset: The magnetoresistance minor loop behavior of the free layer of the same nanopillar device shows a multiple step switching behavior attributable to domain wall pinning by defects induced at the contact region by the nano-aperture fabrication process. (b) A current scan of the same device taken at an applied field of $\sim 500Oe$ that displays peaks in the positive current branch that are generally indicative of the onset of persistent microwave dynamics within the free layer.
currents, 180µA for parallel (P) to antiparallel (AP) and -50µA for AP-to-P are much smaller than those of a 100x200nm$^2$ pillar without the oxide aperture, 7.8 mA for P-to-AP and -4 mA for AP-to-P (not shown). The critical current densities for spin-torque switching of the free layer at the aperture and for free layer switching in a pillar without the aperture are both around $\sim 1\times10^7 A/cm^2$. If, for comparison, the switching current for the nano-aperture device were distributed uniformly over the entire nanopillar cross-sectional area, this would be equivalent to a spin torque switching current density of $\sim 4\times10^5 A/cm^2$, far lower than seen in any non-apertured nanopillar device (see table 5.1). We do need to note that the total change in resistance does not correspond to the $\Delta R$ for full reversal as seen in the magnetoresistance minor loop. The $\Delta R$ value for full magnetization reversal obtained from the GMR measurement is 250 mΩ whereas the $\Delta R$ from spin torque switching is 150 mΩ. We tentatively explain this as due to the domain wall nucleated by a spin polarized current being pinned by a defect in the vicinity of contact region. Evidence for distinct multi-step switching has been occasionally seen in other samples. If such pinning occurs in the vicinity of the aperture, so as to be observable in the magnetoresistance but somewhat away from the location of the minimum contact area where the current density is lower, an increased critical current for reversal can be expected.

Fig. 5.14 (b) shows a resistance vs. current scan of the same nano-aperture device taken with an applied field $H_{ex} > H_d$ which puts the free layer parallel to the fixed layer in the absence of a sufficiently large positive bias current. The peaks in $dV/dI$ for positive currents indicate the onset of a dynamic precession regime within the free layer [16] at applied currents of less than 1 mA. This low onset current is consistent with the data in Fig. 2(a) that collectively point to a
Table 5.1: Comparison of Uniform and Nonuniform Current Injection Devices

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<th>150 x 250 pillar</th>
<th>100 x 200 pillar</th>
<th>uniform injection</th>
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<tr>
<td></td>
<td>30nm aperture</td>
<td>uniform injection</td>
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<tr>
<td>P-AP</td>
<td>180 $\mu$A</td>
<td>7.8 mA</td>
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<tr>
<td>AP-P</td>
<td>50 $\mu$A</td>
<td>4 mA</td>
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<tr>
<td>$J_c$</td>
<td>$4 \times 10^5 A/cm^2$</td>
<td>$1.2 \times 10^7 A/cm^2$</td>
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</tr>
<tr>
<td>$R$</td>
<td>12$\Omega$</td>
<td>3$\Omega$</td>
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strong, highly-localized spin torque at the aperture region of the free layer. Fig. 2(b) also shows a substantial parabolic background in the $dV/dI$ vs. $I$ curve due to joule heating. The asymmetry of this background with current direction is due to the Oersted field $H_I$ generated by the current flowing through the aperture which results in the free layer taking the form of a C state [32] bending at the contact region (see the discussion on simulations in the next section). For negative bias currents this leads to an increasing misalignment with bias between the otherwise parallel fixed and free layer magnetizations resulting in an increased resistance that adds to the parabolic background caused by joule heating. For positive bias currents the contribution to the resistance is negative in the antiparallel case which exists above the switching current level as $H_I$ bending acts to close the angle in between the magnetizations of the two layers.

Although the microwave dynamics induced by a non-uniform spin torque have not yet been thoroughly investigated in these devices, the results of a preliminary measurement at zero applied field in the low frequency range is shown in figures 7.11 and 5.16. The non-hysteretic peaks that appear in the differential resistance provide indirect evidence for switching to a dynamic state. A direct measurement of the frequency spectra with a spectrum analyzer at points 1 and 2 are given in
Figure 5.15: Current scan for a nano-aperture device with no applied field. Non-hysteretic peaks imply that points 1 and 2 should correspond to different dynamic states.

Figure 5.16: Direct measurement of the frequency spectra that correspond to points 1 and 2 in Fig. 7.11 with Lorentzian fits.
Fig. 5.16 which clarifies that the device has indeed switched to a low frequency dynamic state with a surprisingly high quality factor of 400. The presence of dynamic excitations at zero applied field can be expected if one considers the fact that the free layer sees a net dipole field from the fixed layer. This built-in field as we have seen in the previous section can indeed be an effective tool to relax the requirements on external fields to generate microwave dynamics.

Apart from spin transfer characteristics, in the GMR response we need to note that if one calculates the $\Delta RA$ product for a nanopillar device with a nanoaperture of diameter 30 nm that would be $250m \times \pi \times 15 \times 15 \times 10^{-6}$ which gives $0.18m\Omega \cdot \mu m^2$. As we will see in Chapter 7 a typical $\Delta R$ value is $180m\Omega$ for a 70x130 Py/Cu/Py spin valve nanopillar which gives a $\Delta RA$ product of $1.28m\Omega \cdot \mu m^2$. This implies that a $\Delta R$ value of about 43$m\Omega$ is expected for uniform injection into a device of 150x250 dimensions as in this experiment. This is a strong evidence for the existence of a concentrated current flow inside the nanopillar. However the ratio of the expected $\Delta RA$ for a 30 nm aperture to the measured value $1.28/0.18 = 7.1$ is still off. This is not very surprising considering the fact that in nanocontacts to extended films this discrepancy in the ratio of $\Delta RA$ products is measured to be 10. The discrepancy is due to the assumption that the electron flow through the point contact is one dimensional which is certainly not true in my nano-aperture device due to the current spreading and interfacial backscattering effects. The small improvement from a factor of 10 to a factor of 7 is a result of the fact that one of the spin dependent scattering interfaces is where the point contact is defined. However there will still be current spreading on the fixed layer side since the spacer is not truly confined as suggested in the idealized scheme of Fig. 5.3. A future experiment is proposed in Chapter 8 that involves a slight modification
Figure 5.17: The Oersted field due to a nanowire whose radius is equal to the radius of the aperture. The spatial dependence of the Oersted field is shown on a 3D plot (left) and a vector plot (right)

to the fabrication to achieve this.

5.4 Magnetization Reversal by Domain Wall Propagation

The non-uniform current injection scheme used in this experiment prohibits a macrospin approach to understand the switching mechanism since a localized torque will inevitably induce micromagnetic effects. For that reason we have performed zero temperature 3D micromagnetic simulations using OOMMF (ref. [31]) code extended to include the Slonczewski torque term [33] to investigate the switching mechanism of our devices. The Oersted field $H_I$ was modelled assuming a current flow at the center of the pillar through a wire whose radius is equal to the contact radius. The resulting field increases linearly with distance axially and reached a maximum at the boundaries of the point contact and die out as $1/r$ outside the contact region (see Fig. 5.17).

A static, field-switching simulation was performed first to find the micromagnetic state of the free layer for the case of an applied field $H_{ex}$ that cancels the average value of the dipolar field seen by the free layer. Under these conditions
the simulation indicated that the free layer would be in a leaf state [34]. Fig. 5.18 shows a cross sectional view of the fixed layer and free layer magnetizations cut along the easy axis. The nonuniform dipole field causes a curling of spins at the edges perpendicular to the film plane. A top view (right) of the free layer shows that there is additional in plane curling to accommodate the elliptical shape at the edges of the pillar. The augmented Landau-Lifshitz-Gilbert simulation was initialized to start with the free layer in this micromagnetic state. The simulated spin torque equation is in the form (see ref. [12])

\[ g(\theta) = \frac{A \sin(\theta)}{1 + B \cos(\theta)} \]  

(5.2)

where A is the torque amplitude and is proportional to the current polarization P and B is the spin torque asymmetry parameter.

The simulation parameters were chosen to be as follows: The saturation magnetization of Py films was measured to be 650emu/cm\(^3\) at 4.2K. The current polarization at 4.2K was calculated from the magnetoresistance behaviour with a 2 channel Valet-Fert [35] model to be 0.54. We assumed that the polarization direction is misaligned from the easy axis by 1 degree and the exchange stiffness
is $13 \times 10^{-12}$ J/m. The Gilbert damping parameter was chosen to be 0.014, which is larger than the typical value for uniform Py thin films, but actually somewhat smaller than that deduced from Py nanopillar switching experiments [12]. The torque asymmetry parameter $B$ was chosen to be 0.6. The simulation cell size in all directions was taken to be 2.5nm.

Fig. 5.19 shows the simulated evolution of magnetization reversal for AP-to-P switching at 0.5mA applied current and with $H_{ex} = -H_d$. The spin-polarized
current injection at this current amplitude causes a domain nucleation under the
contact region. The domain wall near the contact is forced to propagate into
the nanomagnet via current pressure. The domain wall then expands as in the
magnetic balloon effect [36] and interacts with the edge charges. The nanocontact
acts as a source of domain walls, which are continuously pushed to apply a torque
to the nanomagnet. Once the domain underneath the contact grows to be large
enough to touch the device edges, magnetic reversal is triggered.

This domain wall-mediated magnetization reversal has the disadvantage of being a slower reversal process than reversal by uniformly driven nanomagnet preces-
sion, especially in the presence of defects that act as domain wall pinning centers.
Simulation results for the free layer switching time vs. I are shown in Fig. 5.20.
The simulations indicate that the reduction in switching speed can be compensated
for by applying current pulses of larger amplitude. However the final micromag-
netic configuration to which the device switches is affected by the amplitude of the
Figure 5.21: Final Micromagnetic States as a Function of the Switching Current. From left to right: vortex state with highest applied currents, C state and S state with lowest applied currents

applied current as shown in Fig. 5.21. When the applied current is low enough that $H_I$ is negligible, the domain wall-mediated reversal results in an S state. For somewhat higher current levels $H_I$ results in the free layer magnetization ending in a C state, while for very large current levels this C state can even evolve into a vortex where the center of the vortex is displaced from the aperture due to the strong spin torque. When the current pulse is removed in the simulation, the different micromagnetic states persist, with the exception that the vortex that can be created at very high current pulse levels moves to a position centered over the aperture.

A recent micromagnetic calculation by Finocchio et al. [37] has shown that 1ns switching is possible with current densities of the order of $10^6 A/cm^2$ when considered with respect to the pillar area in these devices and the disturbance to the local moments under the contact region caused by the spin torque gives a substantial out-of-plane local contribution to the switching.

In conclusion, we have fabricated elliptical Py/Cu nanopillar spin valves with a 20-30nm diameter nano-aperture at the interface between the Cu spacer and the Py free layer. Low temperature spin transfer measurements yield very low absolute critical current levels for spin-torque switching, with the required current density at
the hole being comparable to experimental values required for spin torque switching of the same size nanopillar with uniform injection. 3D micromagnetic simulations indicate that the reversal process is via the nucleation of a localized domain wall which then is driven across the free layer by the magnetic balloon effect. Thus this mechanism, especially in the absence of defect pinning results in reduced switching currents. The data indicates that spin injection via a highly localized spin valve contact to a ferromagnetic line could be a much more efficient spin torque mechanism for initiating domain wall displacement than current injection across a domain wall [38]. However this reduced switching current is accompanied by a slower free layer reversal rate. The simulations indicate that faster switching can be obtained by applying progressively larger amplitude current pulses.
References for Chapter 5


[33] Donahue M.J., private communication.


CHAPTER 6

ATOMIC SCALE CHARACTERIZATION OF INTERFACIAL
ROUGHNESS AND CHEMICAL DIFFUSENESS IN PY BASED
MAGNETIC DEVICES

The asymmetry in the conduction probabilities of different electron spins in heterogeneous magnetic multilayer thin film systems forms the basis for a number of interesting physical phenomena such as giant magnetoresistance effect (GMR) (ref. [1]), tunneling magnetoresistance effect (TMR) (ref. [2]) and their inverse effect, spin-momentum transfer (SMT) (ref. [3]) as we have seen in Chapter 2. All of these quantum mechanical effects are not only important for fundamental science but also play a big role in determining the direction of future information technologies.

The hysteretic magnetization reversal process in a spin valve geometry leads to parallel and anti-parallel alignments of the pinned and sense layers that gives rise to GMR effects. In magnetic tunnel junctions the non-magnetic spacer is a thin insulating barrier whose uniformity of thickness, crystallinity and structure at the atomic scale has profound effects on the resulting tunneling current sensitivity on the misalignment of the two electrodes. Magnetic devices utilizing these effects have a tremendous number of applications in the data storage industry with some of the most spectacular examples including magnetoresistive read head sensors and magnetic random access memory (MRAM).

The increasing demand for ultra-high information storage density necessitates a better understanding of the nano-scale physics of magnetic thin film multilayer systems. This can only be achieved by a thorough investigation of the complex microstructure including interfacial interactions and roughness. In the present study I have used scanning transmission electron microscopy (STEM) and electron-energy
loss spectroscopy (EELS) to probe the atomic-scale interactions of one of the most
commonly used soft magnetic materials, permalloy (Py) along with a commonly
used seeding stack of $\text{Ta/CuO}_x$ to achieve a smooth conducting substrate. Py
with its ultra-low coercive fields and high spin polarization (45%) proves to be
an ideal choice for magnetic sensing applications via the GMR and TMR effects
(ref. [4]). Its relatively low saturation magnetization makes it easier to manipu-
late by spin-polarized currents via the spin-torque effect (ref. [5]). Any possible
surface oxidation for film thicknesses less than 5nm or sidewall oxidation in the
case of nanomagnets, changes the magnetic properties such as saturation magne-
tization, thermal stability, magnetic damping, switching fields and high frequency
characteristics (see Chapter 7). In collaboration with John Read we have also
employed x-ray photoemission spectroscopy (XPS) to study the nature of these
oxides that form on Py surface by exposure to air and evaluate the effectiveness of
thin Al over-coating to avoid such unwanted oxidation to be utilized in magnetic
nanoparticles.

6.1 STEM sample preparation

For the purpose of preparing a specimen of metallic multilayers for cross-sectional
imaging and composition analysis in the STEM, a thinning technique must be
employed to reach thicknesses below approximately 500Å to achieve transparency
to 100 keV electrons. In those applications, where a topographic image is desired,
such as imaging the shape and measuring the size of nanoparticles on a surface, the
sample preparation is substantially easier. One possible straight forward solution
for the plane view studies is to use thin nitride membranes ( for fabrication details
of nitride membranes see Chapter 3) as a substrate.
Although several thinning techniques exist, to prepare specimens for cross-sectional STEM studies such as mechanical polishing, electrochemical polishing, ion milling etc., the optimal choice depends on the materials to be studied. In this work I used a combination of mechanical wedge-polishing and low angle ion-mill polishing to prepare cross-sectional TEM specimens of metallic multilayers on silicon. This technique was found to work well for metallic multilayers on silicon in an earlier work (see ref. [6]).

The metallic films under study were deposited on a $<100>$ Si wafer with 50nm thermal oxide coating. To be able to have a good reference to make sure that the electron beam ends up being aligned perpendicular to the cross section, samples were cleaved to be two pieces of 5mm by 5mm squares with one side parallel to the flat of the wafer. This means that the electron beam will be perpendicular to the $<110>$ planes in the STEM.

Once the films were deposited, the TEM specimens were prepared as follows. Each of the surfaces was cleaned to be free of any oil contamination by using a droplet of ivory detergent, a gentle rubbing with a gloved hand and a subsequent rinsing with DI water. The surfaces were then dried by using a gentle touch with a Kim Wipe at the corners of the chips while holding them from another corner with sharp tipped tweezers to drip the residual water droplets. This was to make sure that there is no contamination close to the center of the two metal surfaces. EpoxyBond 110 from Allied High Tech Products was used to glue the two metal surfaces together. The epoxy was mixed with a hardener in a 10 to 1 ratio with a glass stirrer in a clean plastic container. A clean glass pipette was used to transfer as well as to evenly distribute the prepared epoxy on the clean metal surface. In this step it is important to not let the pipette directly touch the surface to
avoid scratches. It should only be rolled on a thin layer of epoxy to wipe out the surface. The resulting stack of two chips was placed on a teflon surface in a spring loaded vise to get a glue layer as thin as possible which assures a better mechanical stability for the subsequent polishing steps. The epoxy was cured at 130 – 150°C in an oven for about an hour (see Fig. 6.1). After the cool down the sample could be removed from the teflon using a razor blade. The residual epoxy at the back side of the two chips could be removed by manually polishing with a SiC polishing pad for about 5min (15 and 5 µ pads should be enough).

The next step was to cut 2mm by 1mm pieces with a diamond saw. The sample was mounted on a clean glass slide aligning one side to the edge of the slide with hot mounting wax on a 120º hot plate. The glass slide was mounted and aligned to have the edge with the sample be flat with the diamond saw wheel. The side of the glass slide with the sample on was placed down on the saw to cut pieces parallel to the slide edge (Fig. 6.2). The sample on the glass slide could be rotated for the second cut by heating and melting the mounting wax to reorient the sample. The polishing head was adjusted to be parallel to the polishing pad surface using an indicator. A pyrex wedge polishing rod was polished first to make sure the surface is as flat as possible.

One of the resulting 2mm × 1mm pieces was mounted on the pyrex wedge polishing rod again using hot mounting wax. At this step it is important to avoid bubbles forming in the hot wax by checking under the microscope before the specimen is mounted. The polishing pads were placed on the polish disc which has DI water distributed uniformly on the surface. The surface tension of the water holds the pads in place. The pad surface was swept by a rubber pad to get rid of any air trapped underneath and make the surface as flat and smooth as possible.
Figure 6.1: First three steps of the sample preparation for cross sectional TEM imaging. 5mm×5mm chips are glued together by epoxy and mounted onto a spring loaded vise and cured in a hot oven.
The polishing rod on a copper holder was mounted onto a tripod polishing jig. The first two polishing steps were perpendicular to the layers. SiC polish pads of 15$\mu$m and 5$\mu$m diameter grains were used for rough polishing, and then starting from 3$\mu$m going down to 0.5$\mu$m grain size diamond polishing pads were used to finish the polishing. In each polish step the amount of material to remove was at least 3 times the grain size of the previous pad used. The rotation speed was about 30 rpm and soap and water (Ivory detergent + DI water) was used as slurry.

The polish head could be set to slowly oscillate on the radial axis to polish uniformly over the pad making sure the swing range did not go beyond the limits of the polishing pad. It was important to keep track of the orientation of the sample since the polishing process gave one rough and one smooth edge. The polishing pad rotates from the rough to the smooth edge also pushing silicon dust to this edge. These particles were eventually cleaned off with a manual polish using a soft pad with a special citon slurry that contains silicon particles. As soon as
the polishing was done the specimen was rinsed in DI water to get rid of residual particles and also polished more manually with a second soft pad but using DI water as the slurry. The removal of the specimen from the pyrex to flip and polish the other side was done by leaving the pyrex rod on a filter paper in acetone until the mounting wax dissolves to release the specimen. A pipette was used to slowly remove acetone by gradually replacing it with isopropanol. Then the specimen was transferred onto another filter paper in an isopropanol bath. The same procedure was used to switch to the final methanol bath for the cleanest surface finish.

The other side to be polished was again mounted keeping track of the rough and smooth edges and polished following the same procedure perpendicular to the layers first. Then the head was tilted about 4° and the specimen was polished until about a length of about 1.5mm is achieved. A molybdenum washer was cleaned with acetone, isopropanol and finally by subjecting it to oxygen plasma for 20 min. The specimen that was removed from the pyrex rod following the same procedure as above was epoxied onto the Mo washer using an M bond adhesive. The specimen was mounted to have the tip aligned with the center of the washer hole (see Fig.
6.3. The washer was placed on a teflon piece and baked at 130° C for an hour to cure the epoxy. A microscope investigation of the tip now displayed interference fringes at the tip due to the presence of ultra-thin silicon.

Since metallic layers do not polish nearly as well as silicon, an ion-mill step was necessary to get an electron transparent tip. A low angle high energy two step ion-mill polish process was used to get a sharp tip in the metallic multilayer region that of course was of the primary interest. The first step process parameters were 3kV beam voltage, 5mA current with a 12° angle for 10 min and the final step was 1kV, 5mA with 10° angle. The specimen was oxygen plasma cleaned for 15 min to get rid of any C contamination before going into the microscope. To reduce contamination of the microscope there was a final 100°C chamber bake of the specimen in the load lock for an hour before it was transferred into the microscope for imaging.

6.2 Electron Energy Loss Spectroscopy

When an electron beam interacts with a specimen there are several possible outcomes that can be utilized to obtain information about topography as well as composition (see Fig. 6.4). In those cases when the specimen is thin enough to be electron transparent we can categorize the results of the interactions into two groups, those being due to reflected electrons and the others due to transmitted electrons.

When an incident electron collides with an atom of the specimen nearly normal to its trajectory it can backscatter close to normal. The probability of backscattering increases with increasing atomic number providing a bright region for higher atomic number species.
If an incident electron while interacting with the specimen loses some of its energy to excite a lower energy electron (usually from the K-shell), this excited electron leaves the surface with a low kinetic energy ($\sim 5$eV). When these secondary electrons are collected, the collection efficiency will depend on how far a feature at the surface is from the collector and thus will provide topographic information. After a secondary electron process the vacancy created in the inner shell can be filled in by the transition of a higher energy electron in the atom. The released energy can be absorbed by an outer shell electron to be released with a characteristic kinetic energy. These so called Auger electrons with their atom specific characteristic energies can be utilized to obtain chemical information about the specimen surface. Similarly if the released energy does not cause an Auger process it gets emitted in the form of X-rays. These X-rays contain information about element specific binding energies that is measured by energy-dispersive x-ray analysis (EDXA). When the resulting transitions between the energy levels yield photon emission, this causes cathodoluminescence as in the television screen.
The transmitted electrons are either unscattered, elastically scattered or inelastically scattered. The unscattered electrons transmit with a probability that decreases exponentially with the specimen thickness. So a thinner specimen will have more unscattered electrons. The elastically scattered electrons follow Bragg's law to yield a diffraction pattern that can be used to get information about atomic planes and orientations. The amount of energy lost by the inelastically scattered electrons is a characteristic feature of atomic species and can be used to get compositional as well as bonding information using electron energy loss spectroscopy (EELS).

The Cornell STEM used in this experiment is VG-HB501A with an accelerating voltage of 100 kV. The spatial drift rate is less than 0.3 nm/min and the energy drift rate is less than 0.03 eV/min. The energy resolution is better than 0.7 eV. (see ref. [8]). Fig. 6.5 shows a schematic of the STEM. A 2 Å diameter 100 keV electron beam is scanned over the sample to get a 2 dimensional image. For imaging purposes forward-scattered electrons are collected and form the bright field (BF) image whereas the electrons with high scattering angles are collected to give the annular dark field (ADF) image which is also atomic number sensitive. The ADF signal varies as $Z^{1.7}$ due to high-angle Rutherford scattering. Those electrons that form the BF image are directed to the EELS detector to be sorted according to their energies in an EELS measurement. An energy dispersive x-ray detector is also available, since it has a better sensitivity to core level excitations in high Z materials.

The electron-specimen interaction results in various excitations as a result of the energy transfer from the electron to the specimen. Figure 6.6 is an example of primary excitations detectable by EELS. The peak at zero energy is the zero
Figure 6.5: A schematic drawing of the Cornell STEM displaying the available EELS, ADF, BF and EDX detectors

loss peak due to elastically scattered or unscattered electrons. The full-width at half-max (FWHM) of the zero loss peak is a measure of the energy resolution of the instrument. The energy range from a few eV to about 40eV spans the losses due to plasmon excitations which can be composed of volume as well as surface plasmons in the case of very thin (< 30nm) specimens. This so called low loss region also contains energy losses due to interband transitions. For higher energies, element-specific core-level excitations are detected. These excitations correspond to transitions from the inner shell to empty states above the Fermi level. An example as shown in Fig. 6.6 would be the K-edge of oxygen that has an onset at 532 eV. This corresponds to electron excitations from 1s to empty states in the 2p orbital in oxygen. Similarly $L_{2,3}$ and $M_{2,3}$ edges correspond to transitions from $2p^{1/2}$, $2p^{3/2}$ and $3p^{1/2}$, $3p^{3/2}$ respectively to the fermi level. The area under these
characteristic peaks are proportional to the number of atoms present under the probe. With a 2Å diameter beam a composition profile across a sample in can easily be obtained with atomic scale resolution.

As the specimen gets thicker the number of electrons that give rise to the zero loss peak decreases but on the other hand the intensity of the plasmon peak increases due to the increasing contribution from volume plasmons. If we denote the intensity of the zero loss peak by $I_o$ and that of the plasmon peak $I_p$, we can use this information to estimate the specimen thickness by the equation:

$$t = \lambda \frac{I_p}{I_o}$$  \hspace{1cm} (6.1)
Figure 6.7: The usage of low loss region data to estimate the specimen thickness is shown.

where $\lambda$ is the electron mean free path, which is typically 100-120nm for 100keV electrons [8]. The experiments described in this chapter were performed with specimen thicknesses in the range of 10-75 nm with the measurements repeated three times in different thickness regions.

Reliable measurements of the composition profiles from EELS spectra requires a careful background subtraction procedure to be employed to individual spectra before any further analysis is done (see Fig. 6.8). The raw data is first corrected for spectrometer shifts. The calibration curve for this correction can be obtained by using reference materials with a well-known composition. The recently mea-
sured energy shift as a function of energy is a line with a slope $3.14 \times 10^{-3}$ and a zero intercept (measurement by K.A. Mkhoyan with NiO, C and BN reference materials).

The EELS detector used in this work is the parallel EELS detector (PEELS) which has a lower gain for channels near the detector edges compared to the center. This gain variation is due to a geometrical consequence of having a rectangular CCD array detector with a circular scintillator. The detector induced distortion can be corrected for by taking an EELS background spectrum that has no core loss edges in the energy range of interest by taking it from a region of the specimen where the atomic species in question is known to be not present. The ratio of a power-law fit to the central region of the PEELS spectrum to the measured signal provides a weighting function to account for the decreasing gain as a function of distance from the detector center. After the energy shift correction the spectra need to be weighted by this function.

The last step is to do an exponential fit to the pre-edge part of the spectrum to determine the background signal. The subtraction of the background signal yields the final core loss edge from the area under which one can calculate the composition profile. To avoid a complicated deconvolution, the effects of plural scattering in the low loss region were avoided by only working with high-loss energy spectra.
Figure 6.8: The background subtraction procedure steps for a Cu $L_{2,3}$-edge spectrum are shown as performed in a Labview code.
In addition to the points discussed here there are other important aspects of EELS that must be carefully considered when making such studies. Some of the most important of these are discussed in Appendix.

### 6.3 X-ray Photoemission Spectroscopy

If an x-ray beam instead of an energetic electron beam (as in EELS) is incident on a specimen, the resulting interactions can also be used to probe the chemical composition of the specimen. This technique is known as electron spectroscopy for chemical analysis (ESCA) or x-ray photoelectron spectroscopy (XPS) (for detailed discussion of this technique see ref. [9] and [10]). A simplified schematic of an XPS system is shown in Fig. 6.9. When the sample surface is bombarded with photons of energy $\hbar \nu$, photoelectrons originating from the core level of the atoms with a kinetic energy $E_k$ can be generated. When these photoelectrons are collected and
sorted out according to their kinetic energies it is possible to determine the binding energies (BE) of the atomic species from which they were ejected by using energy conservation:

\[ h\nu = E_k + BE + \phi \]  \hspace{1cm} (6.2)

where \( \phi \) is the work function of the material. Usually a known element present at the surface such as C is used to calibrate the absolute energy scale to avoid complications coming from the unavailability of an accurate value of the work function. Since the emitted photoelectrons and Auger electrons have mean free paths of the order of a few nm, only those electrons that are close to the sample surface are detectable, which implies that XPS can only probe the top 5-10nm of the specimen, making this a near surface sensitive technique. Another big difference between the EELS and XPS techniques is that the latter has a beam spot size of the order of 100s of microns (800\( \mu \text{m} \) was used in my experiment) so obviously the resulting signal does not give atomic scale information but rather an average over a large area on the surface.

In this experiment, the XPS spectra were acquired using a Surface Science Laboratories SSX-100 spectrometer equipped with a hemispherical energy analyzer with 0.6eV energy resolution in a UHV chamber. Al K\( \alpha \) radiation with a narrow linewidth of about 0.2eV and a clean, flat background was used as the x-ray source (for more details of this instrument see ref. [9] and [10]). The data acquired on each sample consists of a survey scan (a low resolution large energy window scan) to roughly determine the atomic species at the surface, followed by high resolution scans in different regions in the vicinity of the binding energies of the surface constituents including the inevitable C content. In this experiment the presence of sharp metallic peaks enabled the calibration of the energy scale without needing to
Figure 6.10: A survey scan of thin aluminum oxide coating on permalloy

refer to C, but C spectra could still be used to double-check the calibration required
due to spectrometer shifts. An example of a survey scan for a 50nmPy/4nmAlO_x
stack is shown in Fig. 6.10.

The individual regions of interest that are scanned with high resolution (0.6eV,)
after being corrected for spectrometer shifts, provide information about different
oxidation states present at the surface. This is because when an atom forms a
chemical bond such as in an oxide the core shell electrons experience an altered
potential. When a metal loses its electron to form an oxide species, because of
its positive valence, core shell electrons are now more tightly bound to the ion
yielding a positive shift in the binding energy. An example of this situation is seen
in the XPS spectrum in the Al 2p region of an ion-beam deposited stoichiometric
aluminum oxide (Al_2O_3) (see Fig. 6.11). The presence of one oxide peak at about
77 eV and the absence of the metallic Al peak at 73 eV proves that the Al content
Figure 6.11: A high resolution scan in the Al 2p region

is completely oxidized. In my XPS investigation, the XPS spectra were fitted using the standard approach of first applying a Shirley background (ref. [11]) and then fitting the peaks using a 90% gaussian line shape with 10% lorentzian. The precise identification of the atomic species present near the surface could be performed using a database for the peak locations supplied by National Institute of Standards and Technology (NIST) (ref. [12]).

6.4 Improving Roughness via Oxygen Diffusion in $\text{Ta/CuO}_x$ Bilayers

The ability to grow smooth metallic multilayer thin-films has long been a subject of intensive research (ref. [13], [14], [15]) and is very crucial for a number of emerging technological applications (ref. [16], [17]). In the area of metallic spintronics, which utilizes thin, $\sim$ 1-5 nm, multiple ferromagnetic layers separated by either thin
normal-metal spacer layers or very thin insulating tunnel-barrier layers for spin-valve magnetic sensor and magnetic-tunnel-junction applications respectively, it is desirable to have the metallic layers be as smooth as possible to enhance the magnetoresistance effects in the multilayer stack by minimizing undesirable Neel or "orange-peel" magnetic-dipole coupling between adjacent ferromagnetic layers, and by providing a smooth template for the growth of uniform tunnel-barrier layers on ferromagnetic electrodes (ref. [18]). In addition, it is usually required that these thin ferromagnetic layers be grown on a relatively thick, \( > 50 \text{nm} \), normal metal film layer, typically Cu, to accommodate good electrical connections to the spintronics device when it has been patterned to the nanoscale dimensions required for many applications. This presents a challenge since low-resistivity, low-reactivity metals such as Cu tend to form islands on amorphous substrates and transition metal films during their growth by virtue of their high surface energies and hence have large grain sizes and a rough surface.

Approaches to reduce the surface roughness of such films include deposition onto a cooled substrate (ref. [19]), and sputter deposition in a reactive ambient such as an \( \text{O}_2/\text{Ar} \) or \( \text{N}_2/\text{Ar} \) mixture (ref. [20]). This latter technique results in the nucleation of a higher density of islands during the initial stages of the film growth, and hence leads to an overall smaller grain size in the final film. A process that has been developed to improve upon this approach has been to deposit a compound electrode consisting of Cu layers deposited in a partial pressure of \( \text{O}_2 \) or \( \text{N}_2 \) separated by thin layers of a more reactive transition metal, such as Ta, deposited, for example, by sputtering in a pure Ar ambient (ref. [21], [22]). This can result in a substantial smoothing of the surface of the top Cu layer, in comparison to that of the first layer in such a \( \text{CuO}_x/\text{Ta}/\text{CuO}_x/\text{Ta} \) multilayer.
Here we report on an analytical scanning transmission electron microscopy (STEM) study of $CuO_x/Ta/CuO_x/Ta/permalloy(Py)$ thin film stacks that investigates the mechanism responsible for this effect (ref. [23]). Our measurements reveal that it is the relative differences in the reactivity between Ta and Cu, together with grain boundary diffusion of oxygen that is primarily responsible for the smoothing process.

The multilayer structure studied in this work consisted of the following configuration: $Py(4)/Cu(O_x)(20)/Ta(5)/Cu(O_x)(20)/Ta(25)/Py(20)/Cu(8)/Py(5)/AlO_x(7)$ with all the thicknesses in nm. A STEM bright-field cross-section image of this multilayer is shown in Fig. 6.12(a). All the layers were prepared by dc-magnetron sputter-deposition onto a Si (100) oriented substrate covered with 500nm of thermally grown $SiO_2$. A thin layer of Py was deposited first as an adhesion layer for the rest of the stack. Both $CuO_x$ layers were deposited from a Cu target in an $O_2/Ar$ (15/85) mixture at 1 mTorr. To avoid oxidation of the other layers, each $CuO_x$ deposition was followed by 30 min. pump down to $5 \times 10^{-8}$ Torr base pressure and a pre-sputter conditioning of all the targets in a 1mTorr Ar plasma for 75 seconds to clean off any surface oxides from the sputter sources. To passivate the surface of the metallic multilayer, the topmost Py deposition was followed by the final deposition of a 7 nm alumina layer. This deposition was done in two steps. The first 1 nm was sputter-deposited Al metal which then was thermally oxidized in air resulting in a $\sim 1.5$nm oxide layer and the rest of the alumina was ion-beam deposited from a stoichiometric $Al_2O_3$ target.

Cross-sectional STEM specimens from these thin film multilayers were prepared by the mechanical wedge-polish method followed by low angle ion-milling as described in Section 6.1. All the samples were cleaned in dry oxygen plasma for
15 minutes and baked at 120°C for 1 hour before going into the microscope. High resolution bright field (BF) and composition sensitive annular dark field (ADF) images were obtained with Cornell 100kV VG HB501 STEM. This microscope has a field emission gun and a high resolution pole piece with a spherical aberration coefficient $C_s = 1.3\, mm$, chromatic aberration coefficient $C_c \sim 1.5\, mm$, and a focused incident beam of $2\, Å$ diameter with convergent angle of $10\, mrad$ (objective angle). The specimen was aligned so that the electron beam becomes parallel to $[110]$ direction of the Si substrate and subsequent lattice imaging of the Si substrate was used to check the specimen alignment and spatial resolution. Electron energy loss spectroscopy (EELS) measurements were taken with an electron spectrometer that was operated for electrons scattered into $20\, mrad$ collection aperture (collection angle). The energy resolution of the EELS spectrometer was $0.5$ to $0.7\, eV$ with a $0.03\, eV/min$ energy drift (ref. [8]).

Figure 6.12: (a) STEM Bright Field image of $Py(4)/Cu(O_x)(20)/Ta(5)/Cu(O_x)(20)/Ta(25)/Py(20)/Cu(8)/Py(5)/AlO_x(7)$ with all the thicknesses in nm. The film growth direction is from bottom to top. (b) ADF image of the same stack. The gray interfacial lines represent the 50% threshold for changes in ADF intensity from one layer to the other across the interface.
Fig. 6.12 (a) and (b) show the BF and ADF images respectively of the multilayer of interest. The ADF intensity is due to high-angle Rutherford scattering and has been shown to vary approximately as $Z^{1.7}$ where $Z$ is the atomic number of the specimen (ref. [24]). For Ta with a relatively high atomic number ($Z=73$), the ADF signal is strong whereas for Ni, Fe and Cu with $Z=28$, 26 and 29 respectively, the relative intensity is very weak and results in poor contrast, making it difficult to distinguish Cu from Py in the Py/Cu/Py spin valve without a spectroscopic analysis. The BF image as shown in Fig. 6.12(a) displays a better contrast for the spin valve layers due to the presence of Fresnel fringes in the image.

By using the gradients in the ADF intensity from one layer to the other and defining a 50% threshold for an interfacial boundary, we have estimated the evolution of interfacial roughness from the bottom up. The resulting interfacial boundaries are highlighted on the ADF image in Fig 6.12(b). As a result of one repetition of a $CuO_x/Ta$ bilayer, we have found that the interfacial roughness improves by a factor of three, going from 1.2nm to 0.4nm rms roughness compared to a $CuO_x$-only bottom electrode. This result is quite consistent with standard surface roughness measurements made with an atomic force microscope on Cu films and $CuO_x/Ta$ multilayers grown in the same manner as the STEM sample. Due to the small grain size of the Ta films, the top Ta film can be grown relatively thick on such a $Cu(O_x)/Ta$ multilayer while still maintaining the low level of surface roughness, and thus even then it can serve as a very good template for the subsequent growth of a Py/Cu/Py spin valve, as demonstrated in Fig. 6.12, or of a magnetic tunnel junction.

Fig. 6.13 (a) shows a higher magnification BF image of the $CuO_x/Ta/CuO_x/Ta$ portion of the sample cross-section and provides insights into the mechanism that
Figure 6.13: (a) ADF-STEM image of the Ta/Cu(O_x)/Ta section of the thin film multilayer. The ellipses highlight the areas where a material, other than Ta metal, appears to fill in the surface crevasses in the Cu(O_x) layer caused by the presence of grain boundaries. (b) A high resolution ADF-STEM image of the Cu(O_x)Ta/Cu(O_x) section of the multilayer. The red dotted ovals highlight the grain boundary gaps filled with an interfacial layer of interfacial material between the Ta and Cu(O_x) films.
is responsible for the substantial smoothing that results from the deposition of the $CuO_x/Ta$ multilayers. Near the center of the image is a large Cu grain, 20 nm in lateral extent, which spans the distance between the bottom and top Ta layers, with clearly defined boundaries between it and its two neighboring Cu grains in this bright field image. The most notable aspect of this image is that there appears to be a thin layer of material of different bright field contrast, at both interfaces between the $Cu(O_x)$ and $Ta$ films. This material is somewhat thicker at the Cu-Cu grain boundaries than elsewhere and hence smoothes out the height variations, i.e. dips in vertical position, that these grain boundaries would otherwise cause in a uniform layer of Ta film deposited on top of the $Cu(O_x)$.

Fig. 6.13 (b) is a still higher magnification ADF image taken at optimal focus, established from a defocus series of images, that more clearly reveals the presence of this interfacial layer on both sides of the thin 5 nm Ta film sandwiched between the two $Cu(O_x)$ layers. As noted above, dark field imaging depends upon differences in Z rather than differences in crystal orientation to provide contrast when examining a uniform layer of material in the STEM. Thus the thin layers on both sides of the central bright Ta film must be composed of material that is intermediate in average atomic number between that of Ta and Cu. Since dark field imaging is not sensitive to crystal orientation at this magnification the Cu grain boundaries are not readily detectable, but the thicker intermediate layer on the bottom side of the Ta film in the region indicated by the dotted ellipse, and the thinner Ta layer above it suggests that, when considered in conjunction with Fig. 6.13 (a), there is a grain boundary in the $Cu(O_x)$ film below this region.

We have used EELS to determine the spatial variation in the chemical composition of the material between the bottom $Cu(O_x)$ and top $Cu(O_x)$ layers. Since
the Ta absorption edge is outside the energy range of our EELS spectrometer, this measurement was limited to determination of the Cu and O composition profiles as obtained from O K-edge and Cu $L_{2,3}$-edge measurements. In Fig. 6.14 we show an example of such a measurement made at 1 nm intervals across the layers. As can be seen, the O composition in the bulk of the $Cu(O_x)$ layers is quite low despite the Cu being sputter-deposited in a constant partial-pressure of oxygen. Instead the O content is concentrated in the intermediate layers on both sides of the Ta film, but with there being oxygen content even within the middle of this nominally 5 nm thick Ta layer. Since the Ta film was sputter-deposited in pure Ar, any Ta oxidation process must have occurred through a reaction with the oxygen that was previously incorporated into the bottom $Cu(O_x)$ film during its deposition, and with the oxygen that is introduced into the system during the reactive deposition of the top $Cu(O_x)$ film. We have used energy dispersive X-ray (EDX) analysis at the interfacial regions between the Ta and $Cu(O_x)$ layers and established that the Ta/O ratio in these regions is 1.2±0.1, suggesting that the interfacial layer seen in ADF is principally the Ta sub-oxide TaO, which is a semiconductor material that does not add substantially to the resistivity of a patterned thin film multilayer that incorporates such interfacial layers.

These EELS and EDX data indicate that the low electron-affinity of Ta relative to that of Cu, 31 kJ/mole vs. 118 kJ/mole (ref. [25]) results in the Ta deposition reducing Cu oxides in the underlying $Cu(O_x)$ film, via a process which pulls the oxygen ions up from the bulk of the Cu to form a $TaO_x$ transition layer at the interface between the Cu and Ta layers. At the top $Ta/Cu(O_x)$ interface the oxygen ions created during the sputter-deposition of the $Cu(O_x)$ can of course directly oxidize the exposed surface of the Ta layer during the initial stages of that
Figure 6.14: The relative concentration profiles of O (black) and Cu (gray) across the Ta/CuO$_x$ interface calculated using integrated intensities of EELS spectra of O K-edge and CuL$_{2,3}$ edge.

deposition step. However, the absence of significant oxygen content in the bulk of the top Cu(O$_x$) layer, and the lack of any signature of Cu oxide in the Cu EELS signal, indicates that a solid state diffusion and reaction are partially responsible for the formation of the top TaO$_x$ interfacial layer as well.

According to EELS measurements (see Fig. 6.14), the Cu concentration changes rather abruptly at the bottom Cu(O$_x$)/Ta interface and much more gradually at the top Ta/Cu(O$_x$) interface. This may be due to differences in the internal free enthalpies of atomization for Cu and Ta, with that of Cu (1934 mJ/m$^2$) being significantly lower than that of Ta (3018 mJ/m$^2$) (ref. [26]). However, the differences in the formation process for the two interfacial layers, Ar sputter-deposition of Ta and a solid state reaction to form TaO$_x$ at the bottom interface, as opposed to reactive sputtering of Cu(O$_x$) in combination with both gas and solid state oxi-
dation of Ta to form the top interface, may be playing an important role in the enhanced Ta/Cu intermixing of the top interface.

The apparent role of solid state O diffusion in forming the $TaO_x$ interfacial layers provides the explanation for the smoothing effect observed in the STEM imaging and by AFM measurements of these $Cu(O_x)/Ta$ multilayers. This diffusion is enhanced at the Cu grain boundaries and thus during the initial phase of the Ta deposition step the Ta atoms are likely to react preferentially at grain boundary sites where the supply of O is the greatest, a process which acts to fill in the dips or crevasses created on the surface of the Cu film by these boundaries. To maximize this smoothing effect it is important that the amount of O incorporated into the Cu films is optimally controlled, so that the grain boundary crevasses are not over-filled by this Ta oxidation process. We note (see Fig. 6.13 (b)) that such an overfilling may be responsible for the slight bulging of the thin Ta layer.

In summary, we have examined the process which can substantially reduce the surface roughness of thin film multilayer stacks that incorporate relatively thick layers of polycrystalline metal such as Cu that forms large grains during its deposition. This is accomplished by using a thin overlayer of a more reactive material, e.g. a transition metal such as Ta, that will reduce, by an out-diffusion and solid state reaction process, relatively unstable compounds, e.g. $CuO_x$, that are purposely incorporated into the base film during its deposition and thus form an interfacial transition layer, e.g. $TaO_x$, between the less reactive and more reactive metal films. This interfacial layer tends to be thicker at points adjacent to grain boundaries in the less reactive film due to enhanced out-diffusion of the reactant via the grain boundaries and thus it acts to smooth out the surface roughness created by large crystalline grains in the base film. While we have examined the
$Cu(O_x)/Ta$ multilayer system in this study, it seems reasonable to expect that the surface smoothing process that occurs in other multilayer systems such as $Cu(N_x)/Ta$ proceeds in a similar manner (ref. [27]).

6.5 Degradation Mechanisms of GMR in $Py/Cu/Py$ Spin Valves

Understanding the structural details of the interfaces in nanoscale magnetic multilayers and how they correlate to the spin-dependent transport characteristics is of great technological and scientific interest because of their utility in data storage and magnetic sensor applications. $Py/Cu$ multilayers with their high sensitivity, low hysteresis characteristics and also low spin-transfer switching currents when patterned to nanoscale dimensions make them an ideal choice for such device applications. There has been an extensive effort to understand the evolution of interfacial microstructure of $Py/Cu$ multilayers after various thermal treatments by 3-dimensional atomic probe analysis (ref. [28]), Auger electron spectroscopy, x-ray diffraction (ref. [29]) and resistivity measurements (ref. [30]). These measurements revealed that there are modifications in the interfacial microstructure of $Py/Cu$ multilayers even at moderate temperatures of 100 to 150 °C that leads to irreversible changes in the resistivity of these multilayers. The degradation of magnetoresistance with annealing at these moderate temperatures is thought to be dominated by short range interdiffusion induced by non-equilibrium point defects introduced during the deposition process whereas higher temperature treatments (above 350°C) lead to significant grain-boundary diffusion of Ni into Cu as well as Cu into Py that destroys the well defined layer structure. In the present study
we have employed EELS to probe the interfacial interactions between Py and Cu subject to moderate temperature annealing (\( \sim 120^\circ C \)) to determine the degree of interfacial intermixing that dominates the spin dependent scattering characteristics at each interface.

Figure 6.15 shows a series of spot mode EELS spectra offset for clarity with 0.92 nm spacing across the \( \text{Py/Cu/Py} \) trilayer taken with a large energy window to accommodate Ni, Fe and Cu \( L_{2,3} \) spectra at the same time with the onsets of 708, 855 and 931 eV respectively. The inset shows the ADF image of the trilayer region grown along the direction shown by the arrow where the spectra were collected as indicated by gray dots. Assuming conformal roughness at the \( \text{Py/Cu} \) interface and using the calibrated thickness of the Py film deposited on Cu (5 nm), the interfacial line between Py and Cu is shown with the gray line. A significant short range interdiffusion between Ni and Cu can be seen at both interfaces within 1 nm with traces of Ni present almost everywhere inside the Cu spacer layer. Although molecular dynamics simulations suggest, that Py grown on Cu is more chemically diffuse then Cu grown on Py (see ref. [28]), it is hard to come to this conclusion from the EELS measurement since the uncertainty of the position of the interface is determined by the \( \text{Py/Cu} \) interfacial roughness which is about 1-1.2 nm. Since Cu and Ni form a broad range of solid solutions, the tendency of intermixing between these two elements is understandable. We also find that \( \text{Fe/Cu} \) intermixing is much less than that of \( \text{Ni/Cu} \), which is consistent with the fact that Fe is not highly soluble in Cu (see ref. [31]). In any case my preliminary studies for this system show that the trace amounts of Ni if any, seen within the bulk of Cu spacer are less than 1-2 at.% confirming that the dominant chemical interaction between Py and Cu in this case is short-range interdiffusion of mostly Cu and Ni within 2
Figure 6.15: A series of spot mode EELS $NiL_{2,3}$, $FeL_{2,3}$ and $CuL_{2,3}$ spectra taken at the same time with 0.9 nm steps across $Py/Cu/Py$. The top inset shows the ADF image with the white arrow showing the growth direction and dots are the spots where the EELS spectra were collected.
nm of the interface due to the moderate temperature treatment and also possibly due to the nature of the sputter process as well.

The consequences of the Ni - Cu intermixing both at the interface and also extending into the bulk of the Cu spacer can be quite significant in terms of the spin dependent scattering properties of this system. The presence of a magnetic impurity such as Ni in a paramagnetic spacer such as Cu implies the introduction of spin-flip scattering centers which can modify the spin diffusion length of the spacer layer which would reduce the spin polarization of the incident current. This would imply that the critical currents required to achieve spin transfer switching of a \( Py/Cu \) nanomagnet would be larger than ideal and furthermore the resulting GMR signal would also be less than ideal. However, the transport measurements on our \( Py/Cu/Py \) nanopillar spin-valve devices show that the GMR signal is reasonably high, which implies that the short range \( Ni/Cu \) intermixing resulting from a moderate temperature treatment is not detrimental to GMR effect in this case.

6.6 Air Oxidized Py and Passivation of the Surface Oxidation by Thin Al Coating

As I discuss in more detail in the next chapter, the native oxides that form on the surfaces of ferromagnets as a result of exposure to air can alter their magnetic properties by introducing additional anisotropies such as unidirectional anisotropy via exchange bias effects below a critical temperature, \( T_B \) (the blocking temperature of the antiferromagnetic oxide layer)(ref. [32]). The establishment of this interfacial coupling leads to hysteresis loop shifts as a manifestation of a change in
the energy landscape that makes one magnetic orientation of a ferromagnetic element more favorable than the other. Additionally, these shifts are accompanied by an increase of the coercivity. This coupling can be quite strong in the case of thin film devices and magnetic nanostructures such as magnetic nanopillars, nanowires and nanodots. More complex magnetic behaviour due to higher-order anisotropies (ref. [33]), perpendicular coupling (ref. [32]) and rotatable anisotropies (ref. [34]) between a ferromagnet and its surface oxide can arise. The strength of this exchange coupling is a strong function of the microstructural properties of both the ferromagnetic material and its oxide, such as grain size distribution, structural defects, interfacial roughness and variations in chemical composition, because of their dominant role in determining the density of uncompensated interfacial spins. In this section I report on my spectroscopic measurements of the native oxide on Py thin films and evaluate the usage of thin Al coating to passivate such surface oxidation (or sidewall oxidation in magnetic nanostructures). The implications of these observations in the spin-dependent transport properties of patterned Py/Cu/Py spin valve devices are presented in Chapter 7.

Fig 6.16 shows a series of Ni 2p high resolution XPS spectra taken on a NiO reference (a), on an air oxidized Py surface (b), and on a Py surface that was coated with 1.5 nm Al before exposure to air (c). The NiO reference was sputter deposited from a Ni target following the procedure by Yu et al. (ref. [35]). The computer fitted peaks 1 and 2 at 854.4 and 856.14 eV are characteristic peaks of Ni$^{2+}$ and Ni$^{3+}$ in Ni 2p$_{3/2}$ region that are indicative of NiO, and possibly of Ni$_2$O$_3$ components but since the samples are air transferred into the XPS chamber it is quite possible to have a surface Ni(OH)$_2$ component whose peak structure is indistinguishable from that of Ni$_2$O$_3$. Peaks 3 and 4 at 861.6 eV and 865.7 eV,
Figure 6.16: High resolution Ni 2p XPS spectra taken from (a) A NiO reference sample (b) Py surface exposed to atmosphere (c) Py surface with thin 1.5 nm Al coating for oxidation passivation. The intensities are normalized for comparison. The dashed lines are shown to clarify the relationship between different peak structures.
are the shake-up peaks that accompany peaks 1 and 2 respectively. Secondary peaks in the XPS spectra can occur due to Auger transitions, metal plasmons and shake-up and shake-off energy loss mechanism. Shake-up peaks occur when the emitted core level electron causes some other excitation during its departure whereas the shake-off peaks are due to a secondary ionization process. This causes additional energy loss to the outgoing electron and leads to new peaks appearing at higher binding energy. Peaks 5 and 6 at locations 871.2 eV and 880.17 eV are the accompanying peaks for $Ni^{2+}$ and $Ni^{3+}$ components in the $Ni\ 2p_{1/2}$ region.

The computer fitted peak structure for a thin Py film exposed to air consists of 4 peaks in the $Ni\ 2p_{3/2}$ region as can be seen in Fig.6.16 (b). The first peak 1* that appears at 852.3 eV is characteristic of metallic $Ni\ 2p_{1/2}$. There is also a plasmon peak 3* at 858.2 eV that accompanies the metallic peak 1*. The second peak 2* with its shake-up peak 4* line up reasonably well with that of the NiO components with peak locations 854.7 eV and 860.8 eV for peaks 2* and 4* respectively. The broadening and the slightly lower binding energy of the $Ni^{2+}$ peak in the air oxidized Py can be attributed to a small $Ni(OH)_2$ component (possibly less than a monolayer) that develops during air transfer. Peaks 5* and 6* at locations 869.5 eV and 872.6 eV are Ni metal $2p_{1/2}$ and $Ni^{2+}\ 2p_{1/2}$ peaks with 7* and 8* being the accompanying plasmon and shake-up peaks respectively. $Ni_2O_3$ is not an antiferromagnet and its existence is known to weaken exchange coupling at the $NiO/Py$ interface. From the peak structure there is no direct evidence for a $Ni_2O_3$ component which means one can expect quite strong exchange bias effects develop due to coupling to a surface NiO component. Using the reference value for $Ni\ 2p_{3/2}$ photoelectron attenuation length in NiO (ref. [12]) we estimate the NiO thickness to be 13 Å. Fig 6.16(c) shows the corresponding Ni 2p XPS spectrum for a Py film.
**Figure 6.17:** High resolution Fe 2p XPS spectra taken from (a)Py surface exposed to atmosphere. The intensities are normalized for comparison. The dashed lines are shown to clarify the relationship between different peak structures (b)Py surface with thin 1.5 nm Al coating for oxidation passivation

that was coated with 15 Å Al passivation layer. It can be seen that this treatment successfully avoids the formation of NiO component. All the peak structure in this XPS spectrum can be explained with the existence of a pure metallic Ni.

Fig 6.17 shows a series of Fe 2p high resolution XPS spectra taken on the air oxidized Py surface (a) and Py with 1.5 nm Al coating before exposure to air (b). Fe 2p\textsubscript{3/2} region can be fitted to four peaks in the case of the air oxidized Py. Peak 1 located at 706.7 eV is the characteristic metallic Fe peak. Peak 2 at 708.6 eV and peak 3 at 711 eV are due to oxidized iron corresponding to Fe\textsuperscript{2+} 2p\textsubscript{3/2} and Fe\textsuperscript{3+} 2p\textsubscript{3/2} (ref. [36]). The oxide components are most likely to be in the
form of FeO and Fe$_2$O$_3$. However Fe$_3$O$_4$ and FeOOH characteristic peaks in the Fe 2$p_{3/2}$ region are also in the vicinity of the peak 3. Since the samples were air transferred, the existence of the FeOOH as well as Ni(OH)$_2$ is quite likely. Peak 4 located at 712.4 eV is a Ni Auger peak. For the film with the thin Al over-coating before exposure to air, Fig 6.17(a) shows that the peak structure is now reduced to 2 peaks with peak 1* at 706.9 eV being the metallic iron and peak 2* at 711.2 eV being due to a Fe$^{3+}$ component. Using Fe 2$p_{3/2}$ photoelectron attenuation length (ref. [12]) in FeO and Fe$_2$O$_3$ we estimate the total iron oxide thickness to be 7Å. This time we do not expect to have a hydroxide component due to the Py surface being protected by Al thin coating. Thus the Al passivation layer successfully impedes the formation of a FeO component but apparently does not stop the formation of the higher order oxidation state of Fe. We also note however that the amount of iron oxide that has formed is considerably reduced by the Al passivation layer. In particular the iron oxide to iron metal ratio decreases from 5.316 to 1.34 as a result of the Al passivation.

Further investigation of the Py/AlO$_x$ interface with STEM and EELS measurements revealed that the explanation for the disappearance of the Fe$^{2+}$, but not of the Fe$^{3+}$ signal when the Al passivation layer is employed is that there is diffusion of Fe$^{3+}$ ions into the AlO$_x$ layer. Fig. 6.18 shows high-resolution relative concentration profiles of Ni, Fe and O through the Py/AlO$_x$ interface as obtained from integrated intensity of Ni L$_{2,3}$, Fe L$_{2,3}$ and O K edge spectra. The corresponding ADF image and ADF intensity profiles before and after the EELS measurements taken along the path indicated by the dashed line with 0.6nm steps are shown in Fig. 6.18(b). There is no detectable drift along the scan direction as can be seen by the close match between before and after ADF intensity profiles. Our results
Figure 6.18: (a) The integrated relative concentration profiles of Ni, Fe and O across Py/AlO$_x$ interface obtained from spot mode EELS measurements of the Ni $L_{2,3}$, Fe $L_{2,3}$ and O K edge with 0.6nm steps. (b) ADF intensity profiles before and after the EELS measurements. The corresponding ADF-STEM image is shown on the left inset with the dots representing the approximate points where the spectra were collected.
suggest that there is more Fe-Al rather than Ni-Al intermixing at the $Py/AlO_x$ interface. A significant portion of the Fe segregation into the $AlO_x$ occurs within 1.5nm of the $Py/AlO_x$ interface. Furthermore there is quite a sharp drop (within a nm from 75% to 25% concentrations) in the oxygen signal going from $AlO_x$ layer into the Py layer. It is quite likely that Fe is being pulled out of Py by the oxygen content in the top $AlO_x$ layer. A comparison of the free energy of formation between NiO and $Fe_2O_3$ with -50.6 and -177.4 kcal/mol respectively shows that the iron oxidation is thermodynamically favorable (ref. [37]).

Fig. 6.19 (a) shows Ni $L_{2,3}$ edge measurements from two different spots, the top (1) one taken from Ni within the bulk of the Py layer whereas the bottom (2) is at the $Py/AlO_x$ interface. The spectra are very similar in form except, the reduced signal amplitude at the interface points to the fact that Ni concentration is less there. The peak structure lining up so well is a further confirmation that Ni stays at its metallic state even at the $Py/AlO_x$ interface. In comparison Fig. 6.19 (b) shows the corresponding $Fe L_{2,3}$ edge measurement from two different spots with the top one (1*) being from the bulk of Py and the bottom spectrum (2*) is from Fe that got segregated into the $AlO_x$ layer. This time it is clearly seen that the peak structure is quite different and is shifted towards higher energies by about 1 eV. This is a good indication that the Fe segregated into the $AlO_x$ is not at a metallic state but not so suprisingly oxidized. In conjunction with the findings from XPS measurements of this interface it becomes clear that a thin Al passivation layer does successfully eliminate Ni oxidation and also increases the metal to oxide ratio for iron component. The remaining iron in oxidized form is a result of Fe segregation during deposition and following the moderate temperature annealing into the $AlO_x$ layer.
Figure 6.19: (a) Individual Ni $L_{2,3}$ EELS spectra taken from Ni at the bulk of Py top and from the Py/AlO$_x$ interface bottom. The solid lines are drawn for comparison. (b) Individual Fe $L_{2,3}$ EELS spectra taken from Fe at the bulk of Py (top) and from Fe that diffused into about the first 2nm thickness of the AlO$_x$ layer (bottom). The solid lines are drawn for comparison.
In conclusion, this XPS investigation proved that an unprotected permalloy surface consists of antiferromagnetic oxide species namely, NiO, FeO and Fe$_2$O$_3$ with a total thickness of about 2nm. A 1.5 nm thick Al coating prior to exposure to air substantially passivates the surface oxidation. In the next chapter, I present the consequences of sidewall oxide passivation with the thin Al coating on both magnetoresistance and spin torque switching behaviour of Py/Cu/Py nanopillar spin-valves.
References for Chapter 6


CHAPTER 7
THE EFFECT OF SIDEWALL OXIDE PASSIVATION ON SPIN
DEPENDENT TRANSPORT IN PERMALLOY/CU/PERMALLOY
NANOPILLAR SPIN-VALVES

The presence of an adventitious antiferromagnetic oxide layer at the perimeter of a nanomagnet can have a major impact on both field-driven and spin-torque switching characteristics, such as an anomalous increase in magnetic damping at low temperatures (see ref. [1]) through the domain dragging process. Additional damping enhancement mechanisms such as spin pumping ( [2]) have also been shown to contribute to the extrinsic damping ( [3]) in permalloy nanomagnets but the effect from such mechanisms are relatively small ( [4]) and expected to have a much weaker temperature dependence.

In this work we have systematically studied the influence of the exchange coupling between a permalloy/Cu/permalloy nanopillar and its sidewall oxide in three distinct limits of strong, moderate, and reduced exchange anisotropy on the temperature-dependent field-driven and spin-torque switching characteristics of the Py free layer. For this purpose we have patterned Py(20nm)/Cu(12nm)/Py(4nm) spin valves into 70x130nm$^2$ ellipses and subjected them to three different surface treatments. The first group of devices was coated with a 2.5 nm antiferromagnetic NiO layer on top of the native sidewalls, the second group was exposed to air after patterning with no special surface treatment, and the third group was protected from sidewall oxidation by in-situ deposition of a 1.5nm Al passivation layer subsequent to ion-mill patterning. Analytical electron microscopy and x-ray photoemission spectroscopy measurements presented in chapter 6 reveal that the native oxide at the Py surface for our untreated samples consists of NiO, FeO and
$Fe_2O_3$ antiferromagnetic oxides with a total thickness of $\sim 2$nm.

Our results confirm the earlier observation of stochastic fluctuations in coercive fields and loop displacement also accompanied by the stabilization of intermediate states during the switching process ([1]). Low temperature measurements of the switching fields in the case of oxidized sidewalls display persistent fluctuations in the order of 20-50 Oe for the native oxide samples, and transient fluctuations that, due to the training effect, decrease as a function of minor-loop scan iteration number for the NiO coated samples. This unusual behavior is attributable to the weak rotatable anisotropy in the native oxides as opposed to the stronger pinning anisotropy effects from the thicker deposited NiO coating. In contrast to the native oxide samples the loop shifts in the case of $AlO_x$ passivated samples show a 15–20% gradual increase at low temperatures mainly due to the dipolar interlayer coupling that scales with the temperature dependent saturation magnetization. The corresponding loop widths also have a smooth temperature dependence with no unusual low temperature turn-on that are indicative of the antiferromagnetic surface oxide blocking.

For the samples with surface oxides, the temperature dependence of the spin-torque switching currents shows an anomalous increase at the blocking temperature of the oxides present, suggesting an increase in damping. However the strong low temperature local pinning effect at the perimeter of the nanomagnet may in fact modify the effective angle between the fixed and free layer magnetizations inducing a reduction in the spin torque amplitude as well. The sidewall oxide passivation by the thin Al coating successfully establishes stable switching fields accompanied by reduced spin-torque switching currents that also have a reduced dependence on temperature. Measurements of the energy barrier for spin torque switching suggest
that the surface oxide passivation also improves the thermal stability of the Py free layer.

7.1 Ferromagnetic Relaxation in Permalloy Films and Nanomagnets

The dynamic response of magnetization due to an applied field and/or a spin transfer torque is governed by the Landau-Lifshitz-Gilbert (LLG) equation with a spin torque term (see equation 2.11). Ferromagnetic relaxation or damping is the physical process of energy loss by which a previously excited GHz frequency magnetization precession is damped out to reach a static equilibrium state in which the magnetization is aligned with an effective field. The relaxation rate, which is governed by a phenomenological damping parameter $\alpha$ in the LLG equation, plays a key role in controlling the performance of magnetoelectronic devices such as magnetic microwave oscillators by determining resonant frequency linewidths and also by setting the threshold currents for spin torque switching applications (see equation 2.12). It is also important to note that this parameter represents an average behaviour of the magnetization since the non-ideal local variation in the microstructure of the materials such as defects, dislocations and impurity content will inevitably cause the damping strength to vary over the entire sample volume.

The microscopic origin of $\alpha$ is not well understood but several different relaxation mechanisms have been proposed that account for specific cases. The damping parameter can be written as:

$$\alpha = \alpha_{in} + \alpha_{ex}$$

(7.1)

where $\alpha_{in}$ stands for the intrinsic damping and $\alpha_{ex}$ represents any extrinsic contributions to the damping value. The intrinsic relaxation models were recently
Figure 7.1: (from ref. [1]) The temperature dependence of the damping parameter of a permalloy nanomagnet free layer in Py/Cu/Py nanopillar spin valves as obtained from the macrospin simulation fits to 1ns switching data for parallel to antiparallel (P-to-AP) as well as antiparallel to parallel (AP-to-P) switching.

reviewed by Vlad Pribiag (see ref. [5]). The intrinsic contributions are mainly due to the interaction of spin waves with phonons and/or conduction electrons through spin-orbit coupling ([6], [7]) and s-d exchange coupling ([8]). The energy transferred from magnons to the lattice is eventually dissipated as heat. A recently measured value of the intrinsic damping in bulk permalloy films capped with 5nm Cu using in-plane and out-of-plane ferromagnetic resonance (FMR) techniques is 0.0081 ± 0.0003 (see ref. [9]).

A direct, time-domain measurement of magnetization dynamics triggered by the spin transfer torque in a permalloy nanomagnet free layer in a nanopillar spin valve, performed at 40K revealed that the damping parameter is 0.025 (see ref. [10]) which is larger than the 0.008 value for bulk permalloy films. Furthermore, temperature dependent studies of the damping parameter which employ the macrospin
approximation to fit the temperature dependence of the high speed switching behaviour in these devices, display an anomalously high damping value of about 0.065 at low temperatures with it increasing fairly slowly until decreasing the temperature down to approximately 80 K and then increasing much more rapidly within a turn-on temperature region of 40-80K (see ref. [1]). An example of this damping behaviour as determined by the pulse switching measurements is shown in Fig. 7.1.

More recently, there have been direct, small angle precession measurements of the damping parameter in $P_{y65}Cu_{35}$ nanomagnet free layers in a $P_{y65}Cu_{35}/Cu/P_y$ spin valve nanopillar at low temperatures ($< 10 \text{K}$) using spin torque FMR by Sankey et al. (see ref. [11]). These measurements also show a damping parameter of $0.04 \pm 0.001$, which is substantially higher than the bulk thin film value of $0.021 \pm 0.003$ obtained from conventional FMR measurements (performed by J. M. L. Beajour, A.D. Kent and R. D. McMicheal. Also see ref. [9]). Both in this work ( [11]) and also in a follow-up work by Fuchs et al. ( [12]), it has been shown that application of high dc biases can excite nonuniform spatial modes that do not necessarily contribute to the magnetization reversal. Therefore the absolute value of the damping measured in high speed switching experiments obtained by fitting either the probability of switching as a function of dc bias for a series of pulse widths ( [13]) or the time domain switching behaviour for a given pulse width ( [1]), to the macrospin LLG simulations may include additional energy losses to the nonuniform modes that increases the effective damping measured. Nevertheless, both pulse measurements and spin torque FMR measurements suggest an anomalous increase in damping at low temperatures in Py and $P_{y65}Cu_{35}$ nanomagnets.

There are several extrinsic damping mechanism candidates that could poten-
Figure 7.2: A schematic drawing of the induced eddy currents in a conductor subject to a time-varying magnetic flux

Tially account for the observed low temperature damping enhancement. The first well-known mechanism is the eddy current damping. When a conducting material is subject to a time-varying magnetic flux as shown in Fig. 7.2, circulating eddy currents are induced to flow in such a direction to generate a magnetic field that opposes the change in the magnetic flux in accordance with Faraday’s law. However these currents are eventually dissipated as heat by the resistance of the conductor. The eddy current damping is inversely proportional to the film thickness but scales with the material conductivity. Fig. 7.3 shows the temperature dependence of the conductance of a Py/Cu/Py nanopillar spin-valve. There is about a 20% increase at low temperatures so indeed one expects to have a higher contribution to the extrinsic damping from eddy currents. However eddy current damping does not account for the observed damping enhancement for the following reasons. First of all the expected enhancement in the contribution of eddy
current damping to the extrinsic damping only increases by about 20% since it scales with the temperature dependence of the conductance as compared to the observed 100% increase in damping at low temperatures. There have been two important micromagnetic studies of the effect of eddy current damping to spin-torque driven switching by Hrkac and Wadhwa et al. (see ref. [14, 15]) for Co and Py nanomagnets. This mechanism is in effect only in the presence of time-varying magnetization. Therefore it does not appear to control the onset of magnetization dynamics. In the presence of dynamics, it increases the effective damping for a 20nm thick Py nanomagnet by 0.02 which leads to even faster switching compared to spin-torque only. The micromagnetic simulations by Hrkac et al. and Wadhwa et al both show that the eddy currents create non-uniformity in the magnetization providing a pathway to trigger as well as to facilitate the reversal process which implies that eddy currents appearing only at the onset of dynamics are not expected to increase the critical switching currents at low temperatures.
Figure 7.3: The Temperature Dependence of The Conductance of a $70 \times 130 \text{nm}^2$ Elliptical Permalloy/Cu/Permalloy Nanopillar Spin Valve
Figure 7.4: A schematic drawing of the spin pumping in a magnetic trilayer. The first ferromagnet-free layer F1 acts as a source of spin angular momentum which is transferred to and dissipated in a second ferromagnet F2 which acts as a spin sink due to its larger magnetization and/or volume.

Another important extrinsic damping mechanism called spin pumping (see ref. [2, 16]) is mainly due to the fact that the magnetization dynamics are non-local and actually governed by what happens in the total space where the spins are coherent. In the presence of a spin sink which can be another ferromagnet or a high spin orbit coupling lead such as Pt in close proximity to the excited magnetization dynamics in a ferromagnetic layer (spin source), the transverse component of the magnetization gets transported from the source via a spin current and dissipated in the spin sink. This situation is depicted in Fig. 7.4 where the spin transport medium is a normal metal such as Cu. This additional energy loss mechanism can be thought of being complementary to the spin-torque effect where a spin current generates magnetization dynamics. The effective additional damping from such a mechanism scales as:

\[ \Delta \alpha \sim \frac{G^{\uparrow\downarrow}}{M_s d} \]  

where \( G^{\uparrow\downarrow} \) stands for the mixing conductance which is a measure of interfacial
spin dependent scattering. For not too assymetric (two ferromagnetic electrodes made out of the same material with similar thicknesses) structures $\Delta R.G^{\uparrow\downarrow} \simeq 1$ (see ref. [4]). This condition is not strictly met in this experiment but to first order approximation I assume the temperature dependence of the mixing conductance will be inversely proportional to that of $\Delta R$. $M_s$ is the saturation magnetization and $d$ is the film thickness. $M_s$ and $\Delta R$ as a function of temperature for a 4nm thick permalloy free layer in a 70x130nm$^2$ elliptical $Py/Cu/Cu$ nanopillar spin valve is shown in Fig. 7.5. The saturation magnetization increases from 560emu/cm$^3$ at room temperature to 650emu/cm$^3$ at 4.2K. On the other hand $\Delta R$ doubles in going to low temperatures. Both of these effects imply a reduction in spin-pumping-induced extrinsic damping at low temperatures as opposed to the observed damping enhancement. Therefore the temperature dependence of spin pumping does not account for the extra low temperature damping.

A better understanding of the anomalous low temperature damping of permalloy nanomagnets can be achieved by recalling earlier studies of the hysteresis parameters in unprotected permalloy thin films with a surface oxide present. These studies revealed that the oxidized permalloy films develop a unidirectional anisotropy at low temperatures. The results of such a measurement for an 80 nm thick permalloy film is shown in Fig. 7.6.(see ref. [17]). Most important observations in this work can be summarized as follows. The presence of a surface oxide layer manifests itself in a turn-on of unidirectional anisotropy and a significant loop displacement as the sample is cooled below a threshold temperature of about 40K. These effects increase with the increasing surface oxide thickness (realized by a high temperature oxidation treatment) for a given film thickness implying that the surface oxide material acts as an antiferromagnet below a blocking temperature that depends
Figure 7.5: Saturation magnetization of a 4nm thick permalloy film (from Andrei Garcia) (a) and magnetoresistance (b) as a function of temperature for a 4nm thick permalloy free layer in a 70x130nm$^2$ elliptical spin valve nanopillar.
on the oxide thickness. Furthermore the temperature dependence of the film coercivity seems to be dominated by two different mechanisms, with one being the thermally activated domain wall motion above 40K and the coupling to the antiferromagnetic oxide layer below 40K being the other. The presence of thicker oxide layers also shift this threshold temperature to higher values with the typical blocking temperatures varying between 40 and 80K. In the work by Hagedorn et al. a model based on pseudo-unidirectional anisotropy meaning that a certain percentage of the antiferromagnetic spins are reorientable during the switching process was developed. Since the ultra-thin antiferromagnetic oxide layer has a distribution of grain sizes, not all of the grains get blocked together at the same temperature. The fraction of these "weak" antiferromagnetic spins decrease with the increasing oxide thickness. Therefore the strength of the exchange coupling of the permalloy spins with its surface oxides is dependent on the oxide thickness.
Figure 7.7: (from ref. [18]) The effect of oxidation-reduction cycles at 150° and 50 Oe in-plane field on the FMR linewidth in unprotected, hence oxidized 45nm thick Permalloy films at room temperature and 77K.

The work by Hagedorn et al. suggests that an attempt to measure any intrinsic parameters of unprotected permalloy films at low temperatures ($T < 40-80K$) will yield erroneous results. This conclusion has been confirmed for microwave measurements of damping by Patton et al. (see ref. [18]). From that study, a series of measurements of the linewidth following sequential oxidation and hydrogen reduction treatments at two different temperatures are shown in Fig. 7.7. A significant change (85%) in the linewidths is observed at 77 K, with the 300 K data showing a much smaller effect. This clearly indicates that the damping enhancement of oxidized films is much stronger at low temperatures. So the evidence is quite clear that surface oxides can cause enhanced magnetic damping at low temperatures. The questions then which oxides are actually responsible for this behaviour and what is the microscopic mechanism of damping enhancement.

The XPS studies of the air-oxidized permalloy surface presented in Chapter 6 reveals that there are three types of oxides present on a thermally oxidized
permalloy film, namely NiO, FeO and Fe$_2$O$_3$ with an approximate total thickness of 2nm (\(\sim 1.3\)nm NiO and the rest being iron oxides). All of these oxides, in the bulk show antiferromagnetic properties below their Neel temperatures of 525K, 200K and 950K respectively (with the blocking temperatures being essentially the same as Neel temperatures). A naive observation of the bulk properties would suggest that FeO with the lowest Neel temperature is the most likely candidate antiferromagnetic oxide for the observed low temperature behaviour. However, detailed measurements of the blocking temperatures of ultrathin films of these oxides show that both NiO and FeO can have effective blocking temperatures as low as 34K with the blocking temperature approaching the bulk values as the thickness is increased (see ref. [19]) suggesting that all the ultrathin surface oxides can be acting collectively to induce the observed low temperature surface exchange coupling.

Microscopically one likely explanation for the damping enhancement is the domain drag process where part of the energy loss during damping of the magnetization dynamics is deposited onto the surface oxides, which causes a large fraction of the spins to be reoriented. There is one more well-known extrinsic damping mechanism known to induce low temperature damping enhancement in spinel ferrites that is worth mentioning (see ref. [20]). In the work by Yager et al. it appears that a microwave study of the damping in \((NiO)_{0.95}(FeO)_{0.05}Fe$_2$O$_3\) which shows low temperature anomalies can be accounted for by considering a relaxation process (known as valence-exchange), where most of the energy loss is due to a rearrangement of valence electrons on the iron ions as the magnetization rotates. The strong evidence for this interpretation comes from the observation that the damping enhancement effect disappears if one of the iron oxide compo-
ments is not present in the ferrite. This rearrangement does not occur as fast as the magnetization rotation and consequently provides a net energy loss mechanism.

7.2 Sidewall Oxide Effects on Spin Torque Switching

From considerations of the expected temperature dependence of the known extrinsic damping enhancement mechanisms, both in patterned and bulk permalloy films, I conclude that the damping enhancement at low temperature is most likely due to the existence of magnetic disorder caused by the presence of an adventitious sidewall oxide at the perimeter of the Py free layer. These native oxides act as additional sources of dissipation by causing local fluctuations on exchange coupling, an effect which is likely enhanced by lithographic edge roughness. Therefore for optimum device performance, it is desirable to carry out a detailed study of the adverse effects the surface oxides may have on both current-driven and field-driven switching characteristics. For this purpose in this work we have designed and fabricated Py(20nm)/Cu(12nm)/Py(4nm) spin valves with 70x130nm$^2$ cross sectional area and elliptical shape. The devices were subjected to three different surface treatments which are expected to give varying thicknesses of surface oxides for a systematic study of the strength of exchange coupling and device performance.

The device schematics are shown in Fig. 7.8. The first group of devices was exposed to air after patterning, with no special surface treatment. These devices formed the control group for comparison to other surface treatments. The second group of devices was coated with a 2.5 nm antiferromagnetic NiO layer on top of the native sidewalls. The NiO layer was sputter deposited from a Ni metal target at 4.2mTorr pressure in a 7 : 1 Ar/O$_2$ atmosphere with 150W power following a procedure by Yu et al. to get a stoichiometric NiO ( [21]). The third group was
protected at least in part, from sidewall oxidation by in-situ deposition of a 1.5nm Al passivation layer subsequent to ion-mill patterning. This pre-mill deposition was carried out in an ion-beam deposition system equipped with a base pressure $1 \times 10^{-7}$ Torr. The Al deposition was performed at three different angles of 20, 45 and 75 degrees with respect to the normal of the stage which was also rotating in plane at each deposition angle. An Argon pressure of 5.15 mTorr was used to help achieve a slow and as conformal as possible deposition to uniformly coat the permalloy nanomagnet sidewalls. The XPS studies presented in Chapter 6 suggest that such a thin Al coating is able to successfully passivate/substantially reduce the surface oxidation of permalloy.

A cross sectional bright field TEM image of a device (courtesy of Intel) with a thin Al surface oxide passivation layer is shown in Fig. 7.8 i and ii. The presence of a low Z material layer at the sidewalls and also at the top of the Pt capping layer is clear. However the TEM with which this measurement was done is not equipped with an EELS detector so atomic scale composition information for the sidewalls was not available. The low magnification image displays a tapering at the bottom of the device due to ion mill re-deposition effects. This tapering effect only causes a maximum of 10 nm excursion in the minor and major axis of the bottom of the fixed layer and has no effect on the nominal dimensions of the free layer nanomagnet. The bottom SEM plane-view image (iii) confirms the nominal dimensions of the 70 by 130 nm elliptical nanopillar structure.

Figure 7.9 shows a spin-torque response curve and the current convention on a device schematic and spin torque switching parameters as obtained from a typical spin transfer scan taken at room temperature with an Al-coated device. Positive current flow is defined to be from top down so that the electron flow is from
Figure 7.8: The schematics of three types of devices with different sidewall treatments are shown in A ($AlO_x$ passivated), B (air oxidized sidewalls) and C (air oxidized and NiO coated sidewalls). (i) and (ii) are high resolution and low magnification cross sectional TEM images of the $AlO_x$ coated devices respectively (courtesy of Intel corp.). (iii) is an SEM image of the top view of nanopillars which have similar dimensions of nominally 70 nm by 130 nm.
Figure 7.9: Left-nanopillar device schematic Right-Spin torque hysteretic switching curve at room temperature. The current convention is such that a positive current flows from the free to the fixed layer. The width of the loop is $\Delta I_c$ with $I_c^-$ being the anti-parallel to parallel (AP-P) switching current and $I_c^+$ being the parallel to anti-parallel (P-AP) switching current.

The fixed to the free layer which promotes parallel alignment, whereas the opposite direction promotes antiparallel alignment. $I_c^+$ and $I_c^-$ are the absolute values of the critical currents for P-AP and AP-P switching events respectively. We define the width of the loop as $\Delta I_c$ so that $\Delta I_c = I_c^+ + I_c^-$. There is a parabolic background in the data due to joule heating which due to Oersted field is asymmetric with respect to current. That is because in the parallel alignment the Oersted field tries to increase the angle in between the two layers having a positive $\Delta R$ contribution whereas in the AP state the oersted field acts to close the angle in between the two layers contributing a negative $\Delta R$ whereas the heating effect is symmetric with respect to current bias. The joule heating from the current is expected to increase the sample temperature above the bath temperature. Assuming the heat flow mostly comes from conduction to the contacts in a metallic wire the sample
temperature $T_{\text{sample}}$ can be approximated as:

$$T_{\text{sample}} = \sqrt{T_{\text{bath}}^2 + \left(\frac{3eI_R}{4\pi k_B}\right)^2} \quad (7.3)$$

where $T_{\text{bath}}$ is the bath temperature (see ref. [22]). Such a calculation gives an upper bound for the temperature of our samples to be 50K at 4.2K bath temperature for the typical resistance range of 4 – 5Ω but a simple calculation from the typical 4.2K resistivity values of the materials involved would give 1 – 2Ω sample resistance suggesting a maximum heating of 10-20K.

Figure 7.10 shows the temperature dependence of critical currents as determined from spin-torque switching measurements for a set of samples with three different surface treatments. Both in the case of P-AP switching ($I_{c^+}$) and AP-P switching ($I_{c^-}$) it can be seen that those samples whose sidewalls are NiO coated and air-oxidized have a stronger temperature dependence than the samples with Al oxide passivation layer. There is also a clear reduction in the amount of critical current increase at low temperatures for the Al-passivated samples. However there is still a noticeable increase in critical currents as the sample is cooled below 40K or so possibly reflecting the existence of a residual FeO within the sidewalls as was revealed by XPS measurements presented in Chapter 6 or less than complete coverage of the sidewalls by the Al oxide passivation layer. Both the loop widths ($\Delta I_c$) and AP-P switching currents ($I_{c^-}$) of the NiO coated samples indicate that for the case of the thicker antiferromagnetic there is a blocking effect both at 200K and in the usual 40 K region as revealed by the change in the temperature dependence of the spin-torque switching currents.

A comparison of the low temperature spin torque switching currents shows that the increasing amount of sidewall oxide introduces additional asymmetry to the spin torque switching. The 4.2K critical currents at an applied field that cancels
Figure 7.10: The temperature dependence of $I_c^+$, $I_c^-$ and $\Delta I_c$ is shown for all types of samples.
out the average dipole field for the samples presented in Fig. 7.10 are $I_{c^+} = 2.8mA$ and $I_{c^-} = 1.54mA$ for a NiO coated sample, $I_{c^+} = 2.6mA$ and $I_{c^-} = 2.04mA$ for an air oxidized sample and $I_{c^+} = 1.8mA$ and $I_{c^-} = 1.64mA$ for a sample with sidewall oxide passivation. We need to note that the switching for AP-P direction involves a transfer of spin torque from electrons transmitted through the fixed layer to the free layer (majority spins) and absorption of the transverse component of the spins at the free layer-spacer interface, whereas the P-AP switching events originate from those spins that get reflected at the spacer-fixed layer interface (minority spins) and are back incident upon the free layer-spacer interface. Since the transmission and reflection coefficients for majority and minority spins for transport from the ferromagnet to the spacer and vice versa are not necessarily the same, the spin-torque switching currents are inherently asymmetric. However, the fact that the spin-torque asymmetry is enhanced at the samples with a native sidewall-oxide present and even more so in the NiO coated samples, suggest that these oxides provide an extrinsic contribution to the asymmetry of the spin-torque.

Figure 7.11 shows current scans taken at different fields for a sample with the Al oxide passivation layer. The average dipole field seen by the free layer due to the edge charges of the fixed layer is determined from the minor loop to be about 450 Oe. The spin torque switching at 4.2K looks quite different from the one at room temperature shown in Fig. 7.9. The pre-switching dynamics at room temperature are suppressed due to thermally assisted switching. However at 4.2K the signature of the onset of pre-switching dynamic excitations are clearly seen in the differential resistance. For low applied fields since the dipole field from the fixed layer favors the antiparallel alignment the dynamics in the P-AP switching are suppressed, in that as soon as the spin-torque excitation is generated it is a sufficient perturbation
Figure 7.11: The field dependence of spin torque switching behaviour at 4.2K for an AlO$_x$ passivated sample is shown. The external field $H_{ex}$ is applied in the direction to oppose the dipole field $H_d$. Top- $H_{ex} < H_d$ the switching dynamics for parallel to antiparallel switching are suppressed, Middle- $H_{ex} \simeq H_d$ the switching dynamics are observed in both directions, Bottom- $H_{ex} > H_d$ the dynamics are suppressed for the AP-P switching. In this case a small ripple is seen in the AP state for the negative current branch, possibly pointing to fixed layer dynamics. $I_d^+$ is the threshold current for the onset of dynamics for P-AP switching and $I_d^-$ is the threshold current for the onset of dynamics for AP-P switching.
that the net field seen by the free layer drives a complete reversal. In the presence of an applied field larger than the average dipole field seen by the free layer, the dynamics in the AP-P switching are suppressed since this switching event is now field assisted. The onsets of the instability for magnetic excitations are denoted by \( I^+_d \) for P-AP switching and \( I^-_d \) for AP-P switching. Measurements of the microwave spectrum with a spectrum analyzer at these threshold currents give direct evidence of the onset of GHz frequency excitations (see [12]). Although due to the larger magnetic volume it is less likely to observe fixed layer excitations, in some samples we do find evidence of this as well. The broad peak in the differential resistance in the AP state for the case of large applied field but in the negative branch where the effect of the spin polarized current is to drive precession in the fixed layer seen in Fig. 7.11 is one good indication of the excitation of the fixed layer dynamics.

An exhaustive compilation of the field dependence of spin torque switching curves at a given temperature yields a "phase diagram" highlighting the boundaries between pre-switching dynamic modes (denoted by D), the bistable (hysteretic regime-AP/P) and the stable P and AP states. The phase diagrams of three samples with different sidewall treatments at 4.2K are shown in Fig. 7.12. In order to obtain a good comparison between different sample types and to bring out the hysteretic regime more clearly, a background subtraction procedure was applied to individual current scans so that a decreasing current direction was subtracted from an increasing current scan and the resulting differential resistance values were normalized to \( \Delta R \) of the full switching between P and AP states. Using this procedure it was possible to compare the phase diagrams with a common color scale. The horizontal dashed lines show the scans corresponding to an applied field that cancels out the average dipole field. All the other dashed lines highlight
Figure 7.12: Spin-torque switching phase diagrams at 4.2K for AlO$_x$ passivated, air oxidized and NiO coated samples. The horizontal dashed lines indicate the dipole field as measured from the individual minor loops. All the other dashed lines indicate the onset of dynamic region (D).
the phase boundaries for the onset of pre-switching dynamic excitations.

The first striking observation one can make from the phase diagram plots is the apparent shift of the hysteresis loop as a function of increasing sidewall-oxide content to more negative currents accompanied by an increasing asymmetry between AP-P and P-AP switching currents. A simple argument based on exchange bias effects would suggest that the antiferromagnetic oxides at the perimeter of the free layer were blocked during cooling when the free layer was at the AP state due to the net dipole field it sees from the fixed layer. Therefore the exchange coupling at the boundaries with the oxide layer should favor the AP state over the P state which would lead to a shift of the hysteretic regime to the positive current branch since more spin torque would be needed to switch to the P state to overcome the exchange bias. However the observed behaviour suggests that the exchange bias is not strong enough to cause this loop shift. On the other hand the antiferromagnetic oxide blocking at the AP state would imply that the AP state magnetization is more uniform compared to the P state where the local variations of the exchange coupling at the perimeter should create magnetic disorder. Therefore the presence of more magnetic disorder at the ferromagnet/antiferromagnet interface for the P state can increase the damping of magnetic excitations in the free layer causing a shift of the phase diagram to the negative branch.

It is possible to get an idea of how damping evolves with the increasing magnetic disorder present at the sidewalls using the slope of the measured field dependence as a function of current, of the phase boundaries for the onsets of pre-switching dynamics. Before discussing the results of the macrospin-based model, I should note that such calculations strictly speaking do not fully describe the magnetic behaviour of these spin valve nanopillars due to complications arising from the
local variations in the exchange coupling present on the free layer both at the
boundaries due to the magnetic oxides and also due to the possible presence of
fixed layer excitations, the complex coupling of the excited modes between the
free and the fixed layer (see ref. [12]), the possibility of non-uniform free layer
excitations due to non-uniform dipole field and edge effects etc. Therefore more
detailed micromagnetic simulations to determine the effective damping as well as
the nature of dynamic excitations in the presence of magnetic disorder at the pillar
boundaries are necessary but that is beyond the scope of this work.

If I ignore the complications and employ the macrospin model, the onset of
macrospin instability is given (see ref. [23] in SI units by:

\[ I_d = \mu_0 \frac{e}{h \eta} \frac{\alpha}{M_s \text{Vol}(H + H_k + \frac{M_s}{2})} \]  \hspace{1cm} (7.4)

where \( \alpha \) is the damping parameter, \( M_s \) is the saturation magnetization, \( \text{Vol} \) is the
free layer volume, \( H \) is the total field on the free layer, \( H_k \) is the in-plane anisotropy
field, \( M_s/2 \) is the out-of plane anisotropy field and \( \eta \) is the spin torque, which is
given by the Slonczweski (1996) model as

\[ \eta = \left[ -4 + (1 + P)^3(3 + \cos(\theta))/4P^{3/2} \right]^{-1} \]

(see ref. [24]) where \( P \) is the spin polarization and \( \theta \) is the initial angle between the
free and the fixed layer magnetizations. In the typical case when the applied field
cancels out the average dipole field on the free layer \( H = 0 \) and the out of plane
anisotropy is 2 orders of magnitude larger than the in-plane anisotropy, equation
7.4 becomes:

\[ I_d = \mu_0 \frac{e \alpha}{h \eta} \frac{M_s^2}{2 \text{Vol}} \]  \hspace{1cm} (7.5)

and the slope of the onset of dynamics is given by:

\[ \frac{dI_d}{dH} = \mu_0 \frac{e \alpha}{h \eta} M_s \text{Vol} \]  \hspace{1cm} (7.6)
Table 7.1: The results of a macrospin calculation for the damping to spin torque ratio both from the slopes of the field dependence of the onset of macrospin instability and also the measured critical currents for the onset of dynamics at an applied field that cancels out the average dipole field for all three types of samples are shown.

<table>
<thead>
<tr>
<th></th>
<th>AP-P slope</th>
<th>AP-P onset</th>
<th>P-AP slope</th>
<th>P-AP onset</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al-coated</td>
<td>0.176</td>
<td>0.052</td>
<td>0.165</td>
<td>0.108</td>
</tr>
<tr>
<td>Air-oxidized</td>
<td>0.412</td>
<td>0.056</td>
<td>0.517</td>
<td>0.16</td>
</tr>
<tr>
<td>NiO-coated</td>
<td>0.604</td>
<td>0.089</td>
<td>0.604</td>
<td>0.17</td>
</tr>
</tbody>
</table>

From such a calculation, the resulting $\alpha/\eta$ values for the three samples shown in Fig. 7.12 were obtained from the slopes and the onset of dynamic currents and are as shown in Table 7.1. The significant improvement of the slopes (a factor of 3-4) with the oxide passivation indicates that reducing the amount of magnetic oxides indeed does lead to a more ideal macrospin-like behaviour. On the other hand, measurements using the onset of dynamics currents also suggests that the $\alpha/\eta$ ratio is decreased by the Al oxide passivation but more so in the P orientation than for the AP orientation. This supports the idea that the P state for the air oxidized sample and the NiO sample has the larger amount of magnetic disorder at the perimeter of the free layer nanomagnet compared to what is obtained in the AP state. Furthermore, increasing oxide content introduced more complicated, nonuniform dynamic modes that lead to non-linear phase boundaries in the more extreme case of NiO coated samples as can be seen in the phase diagram plot. This is a clear indication that the nanomagnet spin-torque switching behaviour deviates more from an ideal macrospin-like behaviour.
7.3 Sidewall Oxide Effects on Field-Driven Switching

The earlier observation by Emley et al. ([1]) of random variations in the switching fields of the permalloy free layer at 4.2K was attributed to the stochastic nature of the incomplete reorientation of the antiferromagnetic domains. The three types of surface treatments used in this work allows us to access the limits of strong, weak and reduced exchange anisotropy which elucidates the role of the adventitious surface oxides in the magnetic field driven switching behaviour of the permalloy free layer.

The hysteresis parameters of interest defined on a minor loop scan of the permalloy free layer is shown in Fig. 7.13. The minor loop field scans are performed subsequent to the application of a large negative field which fixes the orientation of the pinned layer. The half width of the minor loop is defined as the coercive field \( H_c \) which ideally is a measure of the intrinsic in-plane anisotropy of a single domain Stoner-Wohlfarth particle in the absence of thermally assisted switching, i.e. when measured at low temperatures. The amount of loop shift from the origin is defined as \( H_d \). The loop shift is normally attributed to the average dipole field that the free layer sees due to the fixed layer nanomagnet in proximity. In reality, as identified by micromagnetic simulations the dipole field is non-uniform, especially at the boundaries of the pillar.

The temperature dependent studies of the minor loop hysteresis parameters presented in Fig. 7.14 clarify the role the adventitious antiferromagnetic oxides at the perimeter of the nanomagnets play in determining the low temperature magnetic behaviour. Although the exchange coupling between the oxide layer and permalloy is expected to be weak, in this study we find that given the nanoscale dimensions of the permalloy free layer the effects are significant. The three types
Figure 7.13: An example of the room temperature minor loop of the 4nm thick permalloy free layer. The coercive field $H_c$ is defined as half the minor loop width whereas the dipolar field $H_d$ is defined as the shift of the center of the hysteresis loop from origin.

of devices shown in Fig. 7.14 have very similar coercive fields at room temperature, yet very different temperature dependencies. The 40 K turn-on observed in the control (air-oxidized) sample points to an additional anisotropy beginning to become effective below 40 K indicating that the blocking temperature of the native antiferromagnetic oxides present are of the order of 40K. The temperature dependence of the coercive field for the Al-oxide coated sample shown displays a smooth behaviour with no detectable low temperature turn-ons. However, perhaps due to the difficulty of achieving uniform sidewall coating across the wafer we find that in some cases there is a measurable turn-up in $H_c$ in those Al-passivated samples that are somewhat away from the center of the wafer. Usually these low temperature turn-ups are less than half of what is observed in air-oxidized samples and they tend to occur at even smaller temperatures (20-30K). For the NiO coated samples the usual turn-on in the temperature dependence of $H_c$ at 40K is observed, but it
is almost 3 times as large as that of the control samples indicating a much stronger exchange anisotropy. We attribute the additional change in the behaviour seen at 200K in the NiO coated samples to the blocking of the stoichiometric 2.5nm NiO coating. This difference in the blocking temperatures of the extrinsic and intrinsic oxides can be attributed to the difference between the thicknesses as well as the average grain sizes since for blocking temperature is expected to scale with average grain size and magnetic volume for magnetic nanoparticles ([25]). Indeed in a study of coupling between ultrathin NiO (6.5nm) and permalloy layers, Khapikov et al. ([26]) has observed blocking at 250K.

Ideally the loop shift parameter $H_d$ is expected to have a very weak temperature dependence as it should solely stem from the dipolar interaction with the fixed layer. On average one would expect the loop shift to scale more or less with the temperature dependence of the saturation magnetization (see Fig. 7.5). Figure 7.14 (b) shows the observed temperature dependence for the three types of samples under study. The temperature dependence of Al-oxide passivated sample shown does follow the temperature dependence of the saturation magnetization quite well. In other samples with oxide-passivation layer we find that the temperature dependence of the loop shift is usually weak and results in not more than a 15-20% overall increase at 4.2K. However, we observe a maximum of 50-60Oe low temperature increase below the blocking temperature of the native oxides on air-oxidized samples and likewise turn-ups in NiO samples that follow the coercive field behaviour with the two-stage blocking.

Another important aspect of the exchange bias in antiferromagnet/ferromagnet bilayers is the training effect whereby a gradual degradation of the exchange bias field is observed upon cycling the ferromagnetic layer through consecutive hystere-
Figure 7.14: Temperature dependence of the coercive field top and the loop shift bottom are shown for all three types of devices.
Figure 7.15: The stability of the coercive field - top and the loop shift - bottom as a function of scan iteration at 4.2K for all three types of devices.
sis loops. The microscopic origin of this effect is still unclear and may be different for different bilayer systems. A recent work by Dho et al. ([27]) suggests that the relaxation is related to the measurement time as well as the scan number and is caused by a time relaxation behaviour of the antiferromagnetic domain configuration to minimize the total magnetic energy. In our experiment the sidewall structure in unprotected and NiO coated samples is expected to have a distribution of grain sizes of antiferromagnetic oxide particles that may cause a distribution of blocking temperatures for different regions of the oxide as well. In the presence of relatively strong exchange coupling in NiO coated samples both the loop shifts and the coercive fields display large decaying oscillations (as large as 300-350 Oe) (see Fig. 7.15). In the case of native oxides in air-oxidized sidewalls we observe stochastic oscillations in both parameters whereas in the samples with oxide passivated sidewalls we do not observe a measurable change in both the coercive field and the loop shift. The unusual persistent fluctuations in the air-oxidized samples may be attributed to the presence of a distribution of antiferromagnetic grains some of which may have a blocking temperature a lot smaller than the others which act as sources of stochastic changes in the antiferromagnetic domain configuration leading to a weak, oscillatory coupling. Such a component also manifests itself in the decaying fluctuations observed in NiO coated samples.

The decrease in the amount of sidewall oxide reduces any magnetic disorder caused by local changes in the exchange coupling which acts as a source of nonuniformity in the magnetization. The energy barriers for both field driven and current driven switching are expected to be affected by the presence of this nonuniformity since any deviations from the ordered structure will facilitate the switching and reorientation of the magnetic configuration. Furthermore the total effective mag-
Figure 7.16: Kurkijarvi current ramp rate measurements at room temperature for an AlO_x passivated-top and air-oxidized sample-bottom is shown for both P-AP and AP-P switching events at the dipole field.
netic volume of the nanomagnet will increase as a result of the oxide passivation. The effective magnetic volume at room temperature is generally half of what it should be. Figure 7.16 shows the results of a current ramp rate measurement to determine and compare the energy barriers of an air-oxidized sample and a sample with sidewall oxide passivation. These two samples have similar coercive fields and $\Delta R_s$ at room temperature. However the energy barriers for the current induced switching obtained from current ramp rates using a model by Kurkijarvi ([28]) are 1.91 eV and 1.42 eV for AP-P and P-AP switching respectively for an Al-oxide passivated sample as compared to 1.05 and 1.37 eV for AP-P and P-AP switching in an unprotected sample. The apparent increase in the energy barriers as a function of reduction in the amount of sidewall oxide suggests the overall uniformity of the magnetization plays the dominant role in determining the thermal stability of the nanomagnets.
References for Chapter 7


CHAPTER 8
CONCLUSION

In this thesis, I have discussed experiments designed to probe different aspects of spin-dependent transport in three different nanoscale magnetic multilayer device geometries. The first geometry is a three dimensional nanoconstriction defined on a silicon nitride membrane (originally developed by Kristin Ralls) ([1]) fabricated using state of the art nanofabrication tools. I have used this geometry to study spin dependent transport across constrained domain walls in Ni-Ni and Co-Co magnetic nanocontacts. When the magnetizations of the two magnetic electrodes are manipulated so that one electrode is pinned by exchange coupling with an antiferromagnet while the other one is free to rotate, a domain wall gets nucleated at the constriction site due to energy minimization arguments ([2]). If the domain wall width is larger than the spin-flip relaxation length the spin transport across the wall is adiabatic but for constrained domain walls, the wall width can be manipulated by varying the contact size and in this case a measurable domain wall magnetoresistance effect is obtained due to spin dependent scattering at the sharp domain wall. Detailed studies of this magnetoresistance effect in diffusive to ballistic conduction regimes presented in chapter 4 showed that the domain wall magnetoresistance scales linearly with the contact resistance (decreasing contact size). However as the contact size gets smaller, 1/f noise due to atomic defect fluctuations dominates over the small magnetoresistance signal ($\sim 0.1 - 0.5\%$) in these mechanically stressed point contacts which limits the smallest measurable contact sizes to about 3nm. Due to the presence of the film stresses and the giant magnetostrictive properties of Ni depending on the substrates used I have found that the smallest contacts can suffer from irreversible deformation as a function of
measurement iterations.

The magnetoresistance values obtained from the point contact studies of magnetic multilayer spin valves are about an order of magnitude larger than the domain wall magnetoresistance attainable in the smallest contact sizes. However still most of the spin dependent scattering signal is lost due to the fact that the current is not truly confined as it is in a spin valve nanopillar geometry where larger values of magnetoresistance are available. To be able to take advantage of better current confinement I have modified the nanopillar fabrication process to include an orifice (20-30nm in diameter) at the free layer, spacer interface defined on a thin (3.5nm thick) insulating alumina layer. Aside from an improvement in magnetoresistance, the concentrated spin polarized current at the contact region disturbs the local magnetization of the free layer nanomagnet leading to dynamic excitations and reversal via the process of spin angular momentum transfer ([3]). The absolute value of the threshold switching currents using this nonuniform current injection scheme was found to be two orders of magnitude smaller than those required in the case of uniform current injection into nanomagnets. Detailed micromagnetic simulations of the Landau-Lifshitz-Gilbert equation of motion including a spin torque term presented in chapter 5 showed that the reversal is via domain nucleation and propagation away from the contact region.

This geometry can also be used in the future to generate and study spin torque driven microwave dynamics in a nanomagnet to be compared to extended film point contact oscillators ([4]). A small modification to the fabrication process would be etching of a nano-channel on the alumina to make contact to the free layer which then gets filled with a spacer layer by in-situ deposition. An in-situ low angle ion mill step before depositing the rest of the film stack can be used to
planarize and remove the extended Cu film generating a truly confined channel for spin transport to eliminate all the sources of the loss of spin dependent scattering signal.

A deep understanding the microscopic origins of the magnetic behaviour of the magnetic multilayer devices necessitates an atomic scale characterization of the chemical structure and surface morphology at the thin film interfaces. In chapter 6 I present a series of experiments using analytical electron microscopy, electron energy loss spectroscopy (EELS) and surface sensitive x-ray photoemission spectroscopy (XPS) techniques that probe the chemical micro-structure of permalloy based magnetic devices.

XPS studies ([5]), allow the identification of native oxides present on a permalloy surface which consists of NiO, FeO and Fe$_2$O$_3$ components and also reveal that a thin Al coating before exposure to air substantially passivates the surface oxidation of permalloy. These adventitious antiferromagnetic oxides also present at the perimeter of a permalloy nanomagnet are found to act as sources of additional extrinsic damping below their blocking temperatures ($\sim 40K$) for spin torque switching. I have fabricated permalloy/Cu/permalloy nanopillar spin valves with 2.5nm NiO coating, 1.5nm Al-oxide passivation layer and native oxides present on the sidewalls. A systematic study of the effect of local exchange coupling at the boundaries of a permalloy nanomagnet both on field driven and spin torque switching (presented in chapter 7) using these devices reveal that in addition to extra low temperature damping, other adverse effects such as more asymmetric spin torque, fluctuations in switching fields and reduction in thermal stability are induced in the presence of the coupling to sidewall oxides and are reduced/eliminated with the oxide passivation. For future studies it should yet be possible to obtain bet-
ter results with a H reduction treatment before and after the deposition of the passivation layer to get completely oxide free sidewalls.

Apart from the oxygen chemistry at the permalloy/AlO$_x$ EELS studies of the interfacial interactions presented in chapter 6 reveal the microscopic origin of smooth Ta/CuO$_x$ seeding (used in magnetic tunnel junctions and also the nano-orifice device presented in chapter 5) as the preferential reaction of Ta with the oxygen content which moves along the grain boundaries of Cu and forms a TaO$_x$ layer that fills in the gaps introduced by the grain structure of Cu. These smooth layers are extremely important to get uniform, pinhole free, ultra thin tunnel barriers in magnetic tunnel junctions and also minimize roughness induced coupling between magnetic electrodes.
References for Chapter 8


APPENDIX A

POSSIBLE ARTIFACTS IN TEM MEASUREMENTS

The proper interpretation of electron microscopy measurements requires an in-depth understanding of the possible sources of error in the results that may arise due to both the measurement procedure used as well as the sample preparation technique. The most common sources of errors one can encounter in a study like the one presented in this thesis can be categorized as follows:

1. TILT: The cross-section specimen is not oriented properly so that the electron beam cuts perpendicular to the multilayers. In this experiment the substrate is a $<100>$ oriented Si wafer so that the electron beam must be parallel to the [110] direction to be perpendicular to the cross-section. The specimen is oriented in the microscope until a microdiffraction pattern corresponding to $<110>$ planes is obtained. However it is important to note that getting too close to the tip, it is possible to get a curling which may modify the relative orientation of the specimen with respect to the electron beam. A good example of this situation is shown in Fig. A.1. Microdiffraction patterns obtained from 5 different spots as shown in the bright field image with the same specimen orientation shows that although the alignment looks very good for 4 points into the specimen the point closest to the tip where the Si gets so thin that it starts curling up is misaligned. An alternative way to check the alignment as well as the spatial resolution of the measurement is to image the Si lattice. An example of the Si lattice image for this experiment is shown in Fig. A.2

The effect of misalignment on the oxygen composition profile across the $Py/AlO_x$ interface was studied systematically to reliably measure the oxygen diffusion profile. Fig.A.3 shows that even a small (100mrad) misalignment can be quite mis-
Figure A.1: The bright field image left shows roughly the regions where microdiffraction was performed to check tilt alignment. It is clear that the specimen is e-beam transparent up until approximately the rightmost spot. The tilt is a little bit off at the thinnest spot as expected. The corresponding dark field image is shown to the right with an inset of the original microdiffraction pattern obtained to prove that the specimen was aligned properly.

Figure A.2: A high resolution dark field image of the Si lattice.
Figure A.3: Oxygen Concentration Profile as a Function of Tilt Angle across the $Py/AlO_x$ Interface for three different tilt angles. The corresponding microdiffraction patterns are shown to the right.

leading.

2. RADIATION DAMAGE: When the specimen is hit with energetic electrons for a long time the structure of the specimen may be modified due to radiation damage itself. Metals being good conductors avoid the build up of large electric fields whereas in insulators migration of atoms are more likely to occur in the irradiated region due to both knock out and ionization damage.

In this study to be able make sure the measurement itself is not changing the resulting oxygen profile across $Py/AlO_x$ interface, we have performed a series of EELS measurements on the same spot in the middle of the $AlO_x$ layer with a 100 pA beam current. During each measurement the same spot was exposed for 15 secs. The summary of the results of 20 consecutive measurements are shown in Fig. A.4. The first 4 measurements are almost identical. After the fourth measurement a decrease in the intensity of the oxygen peak is observed accompanied by the development of a small peak at 532 eV indicative of molecular oxygen due to ionization damage (clearly seen in scan number 10)(also see Monica Plisch Ph.D.
Figure A.4: Summary of the results from 20 consecutive O-K edge measurements from the same spot in the AlO$_x$ layer. The development of a small peak at 532eV indicates the presence of molecular oxygen.

Thesis, Cornell University (2001)). This implies that the oxide layer is relatively resistant to radiation damage in the measurement time scale and such damage is not of concern in this experiment, as I also verified by repeating the measurements at different spots on the specimen. When the radiation damage does become a concern, one can spread the beam to a line of 2Å width to decrease the electron density.

3. C CONTAMINATION: Every specimen after the sample preparation will inevitably have C contaminants on its surface. A 15 min O$_2$ plasma cleaning is enough to get rid of the surface C contamination and it should be performed right before the specimen is transferred into the microscope. If for any reason there is contamination in the microscope itself or on the specimen charging effects during the EELS measurement are obscured. A before EELS and after EELS image can
be used to assess whether or not charge build up occurred. Also EELS spectrum of the C K-edge in the region of question gives the ultimate answer. An example of this problem is shown in Fig. A.5.

4. THERMAL DRIFT: During the EELS measurement a thermal drift can occur due to local heating of the specimen. The specified drift rate is $< 0.3 \text{ nm/min}$ (see K. A. Mkhoyan Ph.D. Thesis, Cornell University (2004)). However the drift can be minimized if the radiation damage is not an issue by repeated EELS measurements. The amount of drift during each measurement should be measured to be able to measure the chemical composition profile across an interface more accurately. Fig. A.6 shows the ADF intensity measured before and after spot mode EELS measurements across a $P_y/AlO_x$ interface which shows about a 0.4nm drift during the measurement which implies that the drift-corrected data will have $\pm 0.2 \text{ nm}$ uncertainty.

5. INTERFACIAL ROUGHNESS: The determination of interfacial chemical diffuseness requires a good knowledge of the interfacial roughness because in the transmission mode as the electrons traverse perpendicular to the interface they interact with both interfacial layers in a region whose thickness is determined by the interfacial roughness. To achieve an accurate measurement of interfacial
Figure A.6: Before and after spot mode EELS measurement ADF profiles of a Py/AlO$_x$ interface showing 0.4nm drift.

Figure A.7: Determination of best focus and interfacial roughness from a defocus series of Py/AlO$_x$ interface. Top-bright field, bottom-dark field images with 31.5nm full scale.
roughness, a defocus series of images by changing the current to the objective lens to determine the best focus condition can be performed. Fig. A.7 shows the results of such a measurement for the Py/AlO$_x$ interface. ADF signal from a series of defocussed images determines the exact interface position. Interfacial roughness has to be quantified from a low mag image and taken into account as an error source while discussing chemical diffusivity. The determined interfacial roughness in this case is about 1nm.

6. SURFACE OXYGEN: For measurements performed at very thin regions of the specimen (<15 nm) the surface oxide present both due to exposure to air and also due to oxygen plasma cleaning contributes as a background oxygen signal to the O-K edge EELS spectra. The repetition of the composition profile measurements in different thickness regions of the specimen where the surface effects become negligible can be performed to avoid surface effects as an error source.

7. ION MILL REDEPOSITION: Since the metals do not polish very well a low angle ion mill polish was found to be necessary to get a smooth tip. The ion mill process can cause redeposition problems on the surface of the specimen. However the usage of rocking from side to side during the mill process assures a more uniform milling so the error from the ion-mill redeposited material is minimized and more uniform throughout the surface. I have repeated the measurements with two different specimens prepared at different times to assure that the profiles measured are not dominated by errors due to ion-mill redeposition effects.

8. SPECIMEN BAKE: The last important point is to be aware of all the baking and curing steps the specimen goes through during the sample preparation. These bake steps include the curing of the epoxy and the sample bake to avoid
contaminating the chamber. Such annealing steps which are generally present in almost all nanofabrication schemes may also play a role in the chemical diffuseness across the interfaces.