Spin pumping and spin transport in magnetic heterostructures

by

Eric Montoya

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Name: Eric Montoya
Degree: Doctor of Philosophy
Title: Spin pumping and spin transport in magnetic heterostructures

Examining Committee:

Dr. David Sivik (chair)
Assistant Professor

Dr. Bret Heinrich
Senior Supervisor
Emeritus Professor

Dr. Erol Girt
Co-Supervisor/
Associate Professor

Dr. Karen Kavanagh
Supervisor
Professor

Dr. David Broun
Supervisor
Associate Professor

Dr. Eundeok Mun
Internal Examiner
Assistant Professor
Department of Physics, SFU

Dr. Theodore Monchesky
External Examiner
Professor
Department of Physics & Atmospheric Science
Dalhousie University

Date Defended: 19 January 2016
Abstract

High quality, ultrathin magnetic films were prepared by means of molecular beam epitaxy (MBE). Magnetization dynamics and anisotropies were studied by means of ferromagnetic resonance (FMR) in GaAs|Fe|Au(001) structures as a function of the Fe layer thickness, allowing the determination of bulk and interface properties. Spin transport was studied in GaAs|Fe|Au|Pd structures, where two interesting results were found: (1) The spin pumping induced damping showed an oscillatory dependence on the Au spacer layer thickness when this layer’s thickness was less than the electron mean free path. This effect is attributed to the formation of quantum well states in the Au layer. (2) The spin pumping induced damping was quickly suppressed with the addition of the Au spacer layer as compared to GaAs|Fe|Pd samples. It is experimentally shown that this reduction is not related to the removal of magnetic proximity effect induced damping at the Fe|Pd interface. It is shown that the Pd layer can neither be treated as an ideal spin sink nor as a simple normal metal (diffusive spin scatterer) with respect to spin currents and that the reduction in damping is due to a reflection of spin currents at the Au|Pd interface.

Magnetization dynamics were investigated in ferrimagnetic insulator Yttrium Iron Garnet (YIG, Y$_3$Fe$_5$O$_{12}$). Ferromagnetic resonance was used to determine the spin pumping induced damping in YIG and YIG|Au|Fe|Au structures. In the YIG|Au|Fe|Au structures, the YIG acts as a spin pump and the Fe as a spin sink when the YIG layer undergoes ferromagnetic resonance. Comparing the damping in the YIG and YIG|Au|Fe|Au structures allows one to determine the efficiency of spin pumping at the YIG|Au interface given by the spin mixing conductance. It is found that the spin mixing conductance of as grown YIG films is 10% of that typically found at metallic FM|NM interfaces. Surface treatment of the YIG films by Ar$^+$ etching is able to improve the spin pumping efficiency, approaching closely to that obtained by first principle electron band calculations.

Keywords: ferromagnetic resonance; molecular beam epitaxy; spin pumping; spin transport; magnetic interfaces
Dedication

To Rachelle
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Chapter 1

Introduction

Today in the area of spintronics there exist two distinct conventions of using electron spin to create exciting new devices as well as to explore new fundamental physics. Most of the devices that have proven lucrative have been based on the phenomena such as giant magnetoresistance (GMR), tunneling magnetoresistance (TMR), and spin transfer torques (STT) that rely on a spin-polarized current. These devices are based on thin film heterostructures that utilize the properties of ultrathin magnetic films. First the GMR, and then the TMR effects, have lead to smaller and smaller computer hard drive read heads that have been a key component to the high density magnetic recording industry in past few decades. The STT effect has of recent become very popular within the magnetism community for its potential to make non-volatile magneto random access memory (STT-MRAM) a viable commercial product. Today, there are a handful of new companies as well as some very large ones working on making STT-MRAM a consumer product.

While these devices have ratified their utility and robustness, their transport of charge and spin is indissoluble and as such they are still plagued by issues concerning conventional electronics, including Joule heating, electromigration, and capacitive coupling. A second emerging approach to spintronics is one that separates the net flow of charge and spin, i.e. one that uses a pure spin current. Spin dynamics studies and spintronic devices are increasingly based on nonlocal spin transport using magnetic films where spin-dependent electron scattering plays a significant role; see, e.g., lateral spin valves [2–4], inverse spin Hall effects [5, 6], spin Seebeck effect [7, 8], and spin pumping [9–11].

This thesis is mainly dedicated to examining spin dynamics and spin transport. However, the understanding of magnetostatics is crucial to the design of the systems in which to search for new ideas and further understanding concerning spin dynamics and spin transport. Spin dynamics will be explored in both ferromagnetic metals (FM) and ferrimagnetic insulators (FI), while spin transport will be examined exclusively in metallic structures.
the all metallic systems, high quality single crystalline multilayer samples were prepared by molecular beam epitaxy (MBE) in ultra high vacuum (UHV) at the Surface Science Lab at Simon Fraser University. Ferrimagnetic insulator films were grown by our collaborators from the Microwave Magnetics and Nanomagnetism Group of Prof. Mingzhong Wu at Colorado State University.

The all metallic systems in this thesis were mainly based on GaAs/Fe(001) samples. The GaAs substrate provides a good template on which to grow high quality epitaxial single crystalline Fe ultrathin films. This is mainly due to the good lattice match $\approx 1.4\%$ between GaAs and Fe. Fe is an attractive material owing to its robust ferromagnetism (Currie temperature much greater than room temperature even for ultrathin films), large magnetization, and its elemental abundance on Earth. The other metals used in these studies were Au and Pd. The spin transport in Au is well described by spin diffusion theory where absorption of spin currents is governed by the spin-orbit interaction. Pd represents an extreme limit in which to apply spin diffusion theory. Heterostructures of Fe|Pd have previously been studied [1], and it was found that spin currents are absorbed on a much shorter length scale in Pd than in typical normal metals (NMs, no long range magnetic order, e.g. Cu, Ag, Au). Applying spin diffusion theory to Pd results in a spin diffusion length that is less than the electron mean free path in Pd. The rapid attenuation of spin currents in Pd was attributed to interaction with fluctuation paramagnons in the Pd due to the large Stoner enhancement of the paramagnetic susceptibility. One of the questions addressed in this thesis is how spin currents behave in heterostructures involving both Au and Pd, where spin current absorption is governed by different processes, and what happens at the Au|Pd interface. The systems studied are heterostructures of Fe|Au|Pd, which differ from the typical FM|NM and FM|NM|FM2 heterostructures previously studied, allowing one to investigate the role of the Au|Pd interface in spin current transport in the Au layer confined between Fe and Pd.

The ferrimagnetic insulator (FI) Yttrium Iron Garnet (YIG,$Y_3Fe_5O_{12}$) is used in the ferrimagnetic insulator/metallic systems. YIG is a very attractive FI that is commonly used in today’s rf electronics. The recent development of the growth of quality ultrathin YIG films on Gadolinium Gallium Garnet (GGG, Gd$_3$Ga$_5$O$_{12}$) substrates has garnered much attention as this allows the study and application of YIG in spintronic devices, including using thermal gradients to create spin currents (spin caloritronics). One of the questions addressed in this thesis is whether or not one can spin pump from the FI|NM interface using traditional FMR induced spin pumping. The YIG systems studied in this thesis involve heterostructures with Au and Fe.
This thesis is organized as follows: In Chapter 2, the experimental tools and methods used in the ultra high vacuum system are described. Details of preparing the GaAs(001) surface, specifically for the \( p(4 \times 6) \) surface reconstruction, and the subsequent growth of metallic layers of Fe, Au, and Pd are also given. In Chapter 3, the theoretical considerations needed to understand the ferromagnetic resonance studies in later chapters are provided. Chapter 4 details the different ferromagnetic resonance spectrometers used to study the magnetic properties of the samples in this thesis. The new spectrometers that allow wide frequency dependences in a single sample mounting that were built during this thesis work, one using a multimode cavity and the other a coplanar waveguide, are presented and measurements of test sample are compared. Chapter 5 explores magnetic anisotropies and spin dynamics of crystalline Fe deposited on \( p(4 \times 6) – \text{GaAs(001)} \) in GaAs|Fe|Au heterostructures. The thickness of the Fe layer is varied, allowing one to separate bulk and surface contributions to both the anisotropies and the damping. In chapter 6, theories of spin pumping and spin transport are overviewed. The spin transport models presented are spin diffusion, ballistic spin decoherence, and modified spin decoherence. In chapter 7, spin pumping and spin transport in GaAs|Fe|Au|Pd heterostructures is explored. It is shown that the spin pumping contribution to the magnetic damping \( \alpha_{sp} \) displays an oscillatory-like behavior as a function of the thickness of the Au spacer layer. It is also shown that the thick Pd layer should not be treated as an ideal sink or as a diffusive spin scatterer and that an effective reflection of spin current happens at the Au|Pd interface. In chapter 8, spin pumping at the ferrimagnetic insulator YIG|Au interface is presented. It is shown that surface treatment by grazing angle Ar\(^+\) etching can lead to enhancement of the spin pumping efficiency, approaching the theoretically predicted value \cite{12}, showing that YIG can spin pump nearly as efficiently as metallic ferromagnets.
Chapter 2

Experimental Methods

In this chapter, most of the experimental systems and techniques used in the work presented in this thesis are described. The ferromagnetic resonance theory and ferromagnetic spectrometer are described in Chapters 3 and 4, respectively. For the samples presented in this thesis, thin metallic films were prepared in ultra-high vacuum (UHV) by means of molecular beam epitaxy (MBE) unless specifically stated otherwise. The MBE system was equipped with in situ tools for both surface treatment and characterization: Ar$^+$ etching, atomic H cleaning, Auger electron spectroscopy (AES), X-ray photoemission spectroscopy (XPS), and reflection high energy electron diffraction (RHEED), low energy electron diffraction (LEED), electric transport measurement, and scanning tunneling microscope (STM).

2.1 Ultra high vacuum system

The primary system used in these studies to prepare samples was the ultra high vacuum (UHV) system in the Surface Science Lab at Simon Fraser University. The system consists of 5 separable sub-chambers. The sub-chambers are: (i) Load-Lock (base pressure: $\sim 10^{-8}$ Torr): this chamber is used for insertion of sample and to achieve intermediate vacuum between ambient and primary UHV system. (ii) Intro Chamber (base pressure: $< 2 \times 10^{-10}$ Torr): this chamber is used to outgas and atomic H clean substrates. (iii) Analysis Chamber (base pressure: $\sim 10^{-11}$ Torr): this chamber is used for in situ characterization of samples and houses the tools for AES, XPS, LEED, and electric transport measurements. It also contains the Ar$^+$ sputter gun for surface cleaning and preparation. (iv) STM Chamber (base pressure: $\sim 10^{-11}$ Torr). (v) MBE chamber (base pressure: low $10^{-11}$ Torr): this is the most important chamber in the system as it is used for sample growth by means of molecular beam epitaxy. In fact, the whole UHV system is commonly referred to as the MBE system with this chamber then being referred to as the growth (or deposition)
chamber. The MBE chamber also houses the reflection high energy electron diffraction (RHEED) system.

2.1.1 $\text{Ar}^+$ sputtering

Ionized Ar ($\text{Ar}^+$) sputtering is a widely used tool for in situ substrate and surface cleaning in UHV systems. In the SFU MBE system, $\text{Ar}^+$ sputtering is performed in the Analysis Chamber by means of a standard differential ion gun. Ar gas is allowed to flow into the ion gun through a bleeding valve. The Ar back pressure inside the chamber is generally maintained at $\sim 2 \times 10^{-7}$ Torr. The differential ion gun is used to create a focused beam of $\text{Ar}^+$ ions at room temperature that is directed towards the sample holder.

The $\text{Ar}^+$ beam is set to an angle of incidence of $\sim 65^\circ$ from substrate surface normal. During the sputtering process, the wafer is rotated about its normal axis by $90^\circ$ back and forth and the $\text{Ar}^+$ beam is rastered over an area greater than the wafer size. This is done in order to obtain as uniform of a sputtered surface as possible.

2.1.2 $\text{H}$. cleaning

GaAs substrates, while marketed as epi-ready (or epitaxy ready), are exposed to air and substrate cutting tools before being placed into the UHV system, which leads to the formation of a layer of oxides and adsorbed water and carbon contaminants. While the adsorbed water can easily be desorbed at low temperatures ($\sim 100^\circ$C), the native oxides require much higher temperatures to desorbed; As oxides $450^\circ$C and Ga oxides $580^\circ$C. This thermal cleaning or "out gassing" cannot completely remove C contaminates and can lead to surface roughness and increased substrate impurities [13].

While $\text{Ar}^+$ sputtering can be useful for removing surface contaminates, it is a fairly slow process. Atomic Hydrogen ($\text{H}$.) treatment can lead to a much faster route to in situ substrate cleaning [13–16]. In $\text{H}$. treatment, $\text{H}$. is produced from $\text{H}_2$ gas by a process known as thermal cracking. The system used in these studies was an Oxford Scientific thermal cracker in which $\text{H}_2$ gas is introduced into one end of a narrow tungsten tube that is heated by electron bombardment. As the $\text{H}_2$ gas collides with the inner wall of the hot tube, it is dissociated into neutral $\text{H}$. , which then leaves the other end of the tube in a beam that is directed toward the sample.

The temperature for efficient removal of GaAs native oxides is reduced when $\text{H}$.-treatment is used in conjunction with out gassing. The chemical cleaning process can be broken down into two main stages. 1) The removal of the As oxides $\text{As}_2\text{O}_x$, where $x = 1, 3$, or 5 for the assortment of oxides of arsenic, follows the chemical process [15, 16],
As$_2$O$_x$ + 2xH· $\rightarrow$ xH$_2$O ↑ + As$_2$ $\left(\frac{1}{2}A$s$_4$\right)$ ↑, \hspace{1cm} (2.1)

where the ↑ indicates the species removed from the surface. 2) The removal of Ga$_2$O$_3$ follows the chemical process,

Ga$_2$O$_3$ + 4H· $\rightarrow$ Ga$_2$O ↑ + 2H$_2$O ↑. \hspace{1cm} (2.2)

One point of note is that the reactant Ga$_2$O in Equation (2.2) only becomes volatile at temperatures greater (200°C), therefore H-treatment does not lead to the removal of the Ga$_2$O layer at lower temperatures. However, the chemical process,

Ga$_2$O + 2H· $\rightarrow$ 2Ga + H$_2$O ↑, \hspace{1cm} (2.3)

can lead to a Ga rich surface, which is typically undesirable. The efficient liberation of As$_2$($\left(\frac{1}{2}\right)$As$_4$) in Equation (2.1) and the Ga$_2$O species in Equation (2.2) has been shown to begin at 400°C \cite{15}.

The removal of surface C is through the creation of CH$_4$ and the removal of O is through the creation of H$_2$O, with both reactants being volatile and easily desorbed at the necessary temperatures to remove the native oxides \cite{14, 17}.

\textbf{2.1.3 Electron spectroscopy}

Auger electron spectroscopy (AES) and X-ray photoemission spectroscopy (XPS) are very useful tools to have in situ in a UHV system, allowing the elemental composition and chemical state analysis of samples during the growth process. The techniques are based on the energy analysis of electrons emitted from the surface of samples either by an incident electron beam (AES) or an incident X-ray beam (XPS).

The kinetic energy spectrum of the emitted electrons gives rise to peaks that are element specific; the peaks correspond to energy level differences between specific atomic levels in AES and the binding energy of excited electrons in XPS. The position of the peaks gives information on the elemental composition and chemical state, while the intensity of the peaks provides information on their concentration.

Both AES and XPS are surface sensitive techniques. In the range of energies of the emitted electrons (100 eV - 2 keV), the mean free path is less than 2 nm. In our system, the sample holder can be rotated in such a way as to change the angle of incidence of the excitation species, allowing a certain degree of depth resolution.
The AES electron source, XPS X-ray source, and the electron analyzer are in the Analysis Chamber. The positioning is set up in such a way that the Ar$^+$ etching can be performed at the same time as AES. A diagram of the AES and XPS processes is shown in Figure 2.1.

$$E_p$$ is the energy of the incident electron beam for AES and $$h\nu$$ is the energy of the incident X-ray beam in XPS. KE refers to the kinetic energy and BE refers to the binding energy of electrons. $$E_{F,S}$$ and $$E_{F,A}$$ are the Fermi levels of the sample and analyzer respectively. $$E_{F,S}$$ is defined to have zero binding energy and the BEs are measured from this level. $$V_A$$ is the retarding potential of the analyzer which allows the energy dependence of both AES and XPS to be scanned. The energy levels of the sample are indicated in the left, while the potential of the electron analyzer is shown relative to the sample potential on the right. The details of each process are in the text.

**Auger electron spectroscopy (AES)**

Auger electron spectroscopy (AES) is used for elemental and chemical characterization of sample surfaces in vacuum. An Auger electron spectrum is the plot of the number of emitted electrons as a function of the incident excitation energy. In our system, the excitation is by means of an incident beam of primary 3 keV electrons. The primary function of AES in the studies in this thesis is to monitor the concentration of contaminating species during surface cleaning procedures Ar$^+$ and H$^-$. 
An example Auger process is as follows, see Figure 2.1: a) A primary electron beam of energy \( E_p \) is directed towards the sample surface. b) When the energy of the incident electron is greater than the binding energy of a core level \( K \) electron, a \( K \) level electron can be ionized out of the sample. c) An outer \( L \) level electron (shown in Figure 2.1 as \( L_{II} \)) then relaxes to this inner vacancy with the release of energy in the form of a photon. d) This photon’s energy, \( (E_K - E_{L_{II}}) \), is absorbed by the release of another outer shell \( L \) electron (shown in Figure 2.1 as \( L_{III} \)), known as the Auger electron. The two electron coulombic rearrangement leaves a final state with two vacancies; in Figure 2.1 the core level \( K \) state is reoccupied in the AES process.

The kinetic energy of the Auger electron shown in Figure 2.1 is

\[
KE_{KLL} = BE_K - BE_{L_{II}} - BE_{L_{III}},
\]

(2.4)

where \( BE_K \), \( BE_{L_{II}} \), and \( BE_{L_{III}} \) are the binding energies of the \( K \), \( L_{II} \), and \( L_{III} \) level electrons, respectively.

In practice, the electron energy spectrum is analyzed by sweeping the retarding potential \( V_A \) of the analyzer, see Figure 2.1; this process shifts the analyzer Fermi level \( (E_{F,A}) \) relative to the sample Fermi level \( (E_{F,S}) \). The kinetic energy of the counted Auger electron is

\[
KE_{AES} = KE_{KLL} - \Phi_A - V_A,
\]

(2.5)

where \( \Phi_A \) is the work function of the analyzer.

The emission of the Auger electron is one of two possible processes occurring with the ionization of the core level \( K \) electron. The other possible process is X-ray fluorescence, where a real photon is emitted. For lighter elements, the probability of the Auger process is nearly unity. For heavier elements, atomic number greater than 30, the X-ray fluorescence process dominates and the AES signal is attenuated. In this case, XPS becomes a more sensitive characterization technique.

**X-ray photoemission spectroscopy (XPS)**

Sample surface analysis by means of X-ray photoemission spectroscopy involves the irradiation of the sample with monoenergetic soft X-rays in vacuum and analyzing the kinetic energy of the emitted electrons. In our UHV system, the X-ray source is an electron gun in combination with Mg or Al targets. In the electron gun, electrons are accelerated to 15 keV towards one of the targets. Upon impact with the target an X-ray beam is created with a strong component at an energy specific to the target; in our system, the available X-ray beams are the Mg-\( K_\alpha \) (\( h\nu = 1253.6 \text{ eV} \)) or Al-\( K_\alpha \) (\( h\nu = 1486.6 \text{ eV} \)).
The X-rays have a limited penetration into the sample, on the order of 1-10 microns\[18\]. The X-rays interact with the atoms in the surface region via the photoelectric effect; an example process is shown in Figure 2.1. In this example an X-ray with energy $h\nu$ interacts with a core $K$ level electron which is ionized and is called a photoelectron. The kinetic energy of the photoelectron is

$$KE_{\text{photo}} = h\nu - BE_K.$$

(2.6)

Just as in AES, the photoelectron energy spectrum is analyzed by sweeping the retarding potential $V_A$ of the analyzer, see Figure 2.1. The kinetic energy of the counted photoelectron is

$$KE_{\text{XPS}} = KE_{\text{photo}} - \Phi_A - V_A.$$

(2.7)

One can consider the binding energy to be the ionization energy of the atom for the specific electron shell involved in the process. Each atom has a variety\[131\] of possible ions which gives rise to a heterogeneity of kinetic energies of the photoelectrons. The shifts in $KE_{\text{XPS}}$ associated with the different ionization energies gives information on the chemical state of atom. Furthermore, each X-ray photoemission process has a different probability (sensitivity) which leads to different intensities for each process. The sensitivity factors for a given XPS process for many elements, for example O 1s and Fe 2p, are well documented and can be found in XPS handbooks, for example ref. [18]. Therefore, the XPS positions and lineshapes give quantitative information about the chemical state and composition at the sample surface.

As previously mentioned, the excitation in the AES process need not be electrons. From the example shown in Figure 2.1, it should be clear that the X-ray beam can also lead to the Auger process. The advantage of having two X-ray beam energies (Mg-K\(\alpha\), Al-K\(\alpha\)) can be seen when comparing Equation (2.5) to Equation (2.7). The kinetic energy of the Auger electron $KE_{\text{AES}}$ is independent of the excitation energy, while the kinetic energy of the photoelectron $E_{\text{XPS}}$ is dependent on the excitation energy. By changing the X-ray source between the Mg-K\(\alpha\) and Al-K\(\alpha\), the XPS peaks will shift in energy while the AES peaks will remain unchanged in position. Comparing the two spectrums allows one to quickly distinguish between the XPS and AES peaks.

### 2.1.4 Molecular beam epitaxy (MBE)

Molecular beam epitaxy is a thermal deposition technique that is performed in UHV ($< 3 \times 10^{-10}$ Torr in these studies). An example of the furnace (thermal source) design used is shown in Figure 2.2. The material desired to be evaporated is placed inside a crucible
and heated. The resistive heater is made up of two thin sheets of tantalum formed in a
two half cylinder configuration with space between the sheets. The top portions of the half
cylinders are connected to the inside of a ring made of tantalum wire. A heat shield made of
wrapped tantalum sheet is connected to the outside of the ring at the top. Multiple tantalum
wires are connected to the bottom of each half cylinder in an even distribution and serve as
current leads. All of the above connections are made by means of spot welding. The current
leads are connected to water cooled copper leads that are housed in a vacuum feedthrough
flange. The copper leads are connected to DC power supplies which are generally operated
in voltage control mode. Water cooled, impedance matched resistors are placed in series
with each furnace to help stabilize the power delivered to the furnace. The crucible sits
on top of the current lead connections inside the resistive heater. For the Fe, Au, and Pd
sources, alumina (Al₂O₃) crucibles are used. All the furnaces are surrounded by a liquid
nitrogen cooled cryoshroud to absorb unwanted gasses that may be produced when heating
the furnaces. Additionally, the furnaces have retractable shutters that block the flux of
atoms from reaching the sample.

Figure 2.2: Schematic of the MBE furnace used for thermal deposition. The resistive heater
is made up of two thin sheets of tantalum formed in a two half cylinder configuration. The
two halves are connected to the positive or negative current leads at the bottom and are
connected together with a ring made of tantalum wire at the top. Also connected to the
ring at the top is a heat shield made of wrapped tantalum sheet. The crucible sits atop the
current lead connections inside the resistive heater and is filled with the desired material to
be evaporated.
Figure 2.3: Schematic of the Ewald sphere interpretation of RHEED. The radius of the Ewald sphere (shown in green) is given by the magnitude of the incident electron beam $|k_i|$. The reciprocal lattice consists of reciprocal rods (shown in red) that extend perpendicular to the sample surface plane in the $Z$ direction and have a spacing given by the inplane reciprocal spacing. The intersection of the reciprocal rods and the Ewald sphere defines the diffracted beams and satisfies the conservation of energy and conservation of momentum up to a reciprocal lattice vector $G$, see Equations (2.9) to (2.11). The diffracted beams fluoresce on a detector screen. (a) A perspective view to show the origin of the Laue circle. (b) A top view.
2.1.5 Reflection high energy electron diffraction (RHEED)

RHEED is a tool that can provide real time information about surface symmetry and morphology of a sample. It can also be used to provide thickness information during sample growth with sub atomic layer accuracy. Reflection high energy electron diffraction (RHEED) is a common and very useful tool incorporated into many MBE and UHV systems.

A RHEED system requires two main components, a high energy electron beam source and a photoluminescent detector screen. The electron beam is directed towards the sample at a grazing angle of a few degrees with respect to the sample surface. The diffracted beam pattern is illuminated on the detector screen, where it is typically monitored by means of a charge coupled device (CCD) camera and computer for recording its time evolution during growth. The SFU RHEED system makes use of KSA400 computer software.

Since the angle of incidence is very small, there is very little momentum perpendicular to the sample surface; thus the incident beam interacts primarily with atoms at the sample surface. The high energy electron beam corresponds to a short wavelength for the electrons. The 10 keV electron beam in our system corresponds to \( \lambda_{\text{RHEED}} = 0.123 \, \text{Å} \) which is an order of magnitude smaller than the typical atomic spacings of the samples; therefore, the electron beam is sensitive to atomic arrangements at the surface of the sample.

A schematic of the RHEED system is shown in Figure 2.3. In order to understand the RHEED patterns it is useful to use the concept of the Ewald sphere. The Ewald sphere representation is a simple kinematic analysis of the diffraction pattern; for more detailed analysis that considers intensity of diffraction patterns, one must consider dynamic scattering as well. In the kinematic model, the diffracted beams maintain the energy of the incident beam, but the electron beam momentum changes. To construct the Ewald sphere, one defines the tip of the incident beam’s wavevector \( \mathbf{k}_i \) to be the coordinate system origin and centers a sphere of radius equal to the magnitude of the wavevector

\[
|\mathbf{k}_i| = \frac{2\pi}{\lambda_{\text{RHEED}}}. \tag{2.8}
\]

Thus the Ewald sphere gives all the possible diffracted wavevectors \( \mathbf{k}_d \) that satisfy the conservation of energy for the incident beam,

\[
|\mathbf{k}_d| = |\mathbf{k}_i|. \tag{2.9}
\]

Due to the grazing angle of the electron beam, only the first few layers at the film surface contribute to diffraction and there are no well defined diffraction conditions in the direction perpendicular to the sample surface. The typical three dimensional lattice of the bulk crystal is replaced with a two dimensional lattice of reciprocal rods in the surface plane,
with rods originating at the conventional inplane lattice points of the sample surface and extending in the perpendicular to surface direction, as shown in Figure 2.3. The diffraction pattern is given by the intersection of the reciprocal rods and the Ewald sphere. The intersections are given by the diffraction conditions

\[ k_d - k_i = G. \]  

(2.10)

where the reciprocal lattice vector

\[ G = m \frac{2\pi}{a} X + n \frac{2\pi}{a} Y + Z Z, \]  

(2.11)

where \( a \) is the inplane lattice constant (assuming a cubic lattice), \( m \) and \( n \) are integers, and \( Z \) is the perpendicular lattice vector chosen such that Equation (2.9) is satisfied.

The wavevectors \( k_d \) are labeled such that the \( k \)-vector that forms the lowest angle to the sample surface is called 0th order beam. The 0th order beam is commonly referred to as the specular beam and the spot created on the detector screen is commonly called the specular spot. The specular spot generally has the greatest intensity on a RHEED pattern. Each successive intersection of a reciprocal rod and the Ewald sphere at higher angles to the sample surface is labeled as a sequentially higher order reflection. If one considers a row of reciprocal rods that is perpendicular to both the incident beam direction and the sample surface as a plane, then this intersection of this plane and the Ewald sphere would be a circle referred to as a Laue circle, see small circle on Ewald sphere in Figure 2.3 (a). The orders of the Laue circles follow the same convention as the \( k \)-vectors. The first order diffraction spots, specular spot, and the direct beam lie on one circle on the detector screen, as illustrated in Figure 2.3 (a).

From Equations (2.9) and (2.11) as well as Figure 2.3 (b), one can see that there are many reciprocal lattice rods that intersect the Ewald sphere, thus satisfying the diffraction laws. However, as shown in Figure 2.3 (a) only low order diffraction beams hit the detector screen in a typical RHEED system. The RHEED pattern viewed at the detector screen can only contain the \( k \)-vectors that project into the angular range subtended by the detector screen. Aligning the incident electron beam along one of the main symmetry azimuths allows one to determine the surface lattice symmetry from the RHEED pattern. Using either the geometry of the RHEED configuration and the beam energy or a reference sample of known lattice spacing, the separation between the diffracted beams can be used to determine the inplane lattice spacings. Secondary diffraction patterns between the main diffraction pattern give information about the surface reconstruction when present.
RHEED oscillations

RHEED is a powerful technique for monitoring epitaxial growth and monitoring the film thickness. Oscillations in the intensity of RHEED patterns during the epitaxial growth of GaAs by means of MBE were discovered in the early 1980s [19]. The oscillatory behavior proved to be particularly useful for the control of sample growth as they allow one to observe the completion of individual atomic layers [20, 21]. Purcell et al. [22] reported for the first time that RHEED oscillations of the specular beam could be observed for metallic films using Ni deposited on Fe and Ni substrates.

During ideal layer-by-layer growth, the intensity of the specular spot will oscillate with film thickness. Each period of oscillation corresponds to a growth of 1 atomic layer (AL). In this growth mode, the RHEED beam is sensitive to the top two layers of the film. The 2nd layer (bottom) is completely filled and the 1st layer (top) coverage ranges from 0 – 1. When the 1st layer coverage becomes 1, it becomes the new 2nd layer and a new 1st layer starts to grow. The RHEED oscillations have two contributions: oscillating diffuse scattering with changing coverage of layer 1 and interference from scattering from layers 1 and 2. The interference contribution is most prominent if the RHEED beam is set at an anti-Bragg angle where the reflected beams from layers 1 and 2 will be out of phase. With changing surface coverage the destructive interference will oscillate. Fortunately the contributions from both diffusive scattering and anti-Bragg scattering are in phase; the maximum RHEED intensity for the specular spot is with a 1st layer coverage of 0 (or 1) and the minimum intensity corresponds to a 1st layer coverage of 0.5. In practice, the growth mode is quasi layer-by-layer in which another layer can start to form before the 1st layer mentioned above reaches a coverage of 1. In order to observe well pronounced RHEED oscillations, good layer-by-layer growth is required. Example RHEED oscillations during the growth of a 17 (atomic layer) AL Fe film on GaAs (001) are show in Figure 2.4.

2.1.6 Quartz crystal thickness monitor

In addition to RHEED oscillations, film thicknesses are monitored by means of a quartz crystal thickness monitor, also known as a quartz crystal microbalance. A schematic of the thickness monitor is shown in Figure 2.5. The thickness monitor allows an indirect measurement of the film thickness by measuring the film thickness grown on its own surface rather than the sample surface.

The quartz crystal monitor operates on the piezoelectric effect. Applying an ac voltage to the crystal induces oscillations and the crystal behaves as a resonator. The ac voltage is provided by an external oscillator circuit. At constant temperature, the resonant frequency remains constant unless the crystal’s mass is changed. The quality factor of today’s quartz
Figure 2.4: Example oscillations in the specular spot intensity during the growth of Fe on GaAs (001). The MBE furnace shutter is opened at time \( \simeq 46 \) s starting the Fe film deposition. The shutter is closed at time \( \simeq 998 \) s ending the film deposition. Each oscillation corresponds to the deposition of 1 AL. The observed oscillations are indicative of smooth quasi layer-by-layer growth. The intensity is initially reduced during the first four oscillations before the Fe forms a continuous film [33].
crystals is $Q \gtrsim 10^5$, allowing very precise measurements of the resonant frequency. Here $Q = f_r/\Delta f$ describes the sharpness of the resonance, where $f_r$ is the resonant frequency of the quartz crystal and $\Delta f$ is the bandwidth of the resonance given at the full width at half maximum. The crystal used had a starting resonant frequency $f_r = 6$ MHz. During film deposition the crystal’s mass is increased, lowering the resonant frequency of the crystal. The shift in frequency is measured and converted to a corresponding thickness by an external monitor.

Since the quartz crystal monitor is an indirect measurement of the film thickness, the placement of the monitor is often very important for accurate results. In our UHV system, the quartz crystal holder feed through is mounted on an adjustable bellows flange. This allows one to place the crystal in the same location as the sample will be during growth in order to measure direct growth rates as a function of furnace power. The crystal can then be retracted to a desired position and the rate/thickness can be scaled with a tooling factor. However, we typically calibrate the crystal monitor in its desired growth position using RHEED oscillations on a test sample. The calibrations are steady over time when using the same furnace power.

Since changes in temperature lead to changes in resonance frequency of the crystal, the crystal holder is water cooled with a temperature controlled water pump and bath. With the opening and closing of the furnace shutters during film growth, the molecular beam represents a moderate thermal load that leads to jumps/drops in thickness readings, see Figure 2.6. The time scale to reach the steady state is $\sim 50$ s. The thickness monitor is calibrated to the RHEED oscillations in the steady state. The jumps/drops in thickness reading are dependent on the growth rate (thermal load), so it is important to calibrate the thickness monitor for the intended growth rate for precise control of the deposited film thicknesses. If only an accurate thickness determination is needed, the thickness of the
Figure 2.6: Example thickness monitor reading during the growth of Fe on GaAs (001). The MBE furnace shutter is opened at time $\approx 46$ s starting the Fe film deposition. The shutter is closed at time $\approx 998$ s ending the film deposition. The dotted black line is a linear fit to the steady state growth. The drop in thickness monitor reading at the beginning of growth to the extrapolated line is to within hundredths of an Å of the difference between the jump at the end of the growth.
sample can be determined after the growth without prior calibration of the jump/drop value.

In Figure 2.6, the thickness \( d \) is listed as \( \text{Å}^* \) as it is not the true thickness scale at the sample. This is due to the facts that the thickness monitor and sample are not in the same area and that we set the material density to a low value to obtain more digits of accuracy. The RHEED oscillations are used to find the quartz thickness calibration parameter \( T \) (in units of \( \text{Å}^* \)) such that the total change in thickness reading divided by \( T \) is equal to the thickness in AL grown, see Section 2.2.2.

2.2 MBE sample preparation

All samples using GaAs as the substrate, see Chapters 5 and 7, were grown on undoped commercially available (Wafer Technology Ltd.) GaAs(0 0 1) substrates. The substrates are circular with a diameter of 50.5 ± 0.5 cm. The substrates have two flat portions oriented 90° to each other, one of which is the longer major flat (16 ± 2 mm) and one of which is the shorter minor flat (8 ± 1 mm). The substrates used were a European/Japanese (EJ) cut, where the major flat is on the (TT0) and the minor flat is on the (T00). The substrates are polished on one side, the same side on which the films are deposited. In the lab, small lines are made with a diamond scribe parallel to the major flat on the unpolished side before the wafer is cleaved into seven 10 × 10 mm squares; this allows one to know the orientation of the smaller substrates when mounting on the sample holder in the UHV transfer arm. GaAs cleaves very cleanly along the \{1 1 0\} planes as they contain equal numbers of As and Ga atoms per unit area and are therefore neutral [23].

Surface reconstruction of GaAs (0 0 1)

GaAs is a III-V compound semiconductor consisting of an fcc lattice with a two atom basis Ga and As, known as a zincblende structure. The Ga and As are shifted from one another by 1/4 of the body diagonal or \( \sqrt{3} a_{\text{GaAs}}/4 \) along the [1 1 1] direction, where \( a_{\text{GaAs}} (= 5.6535 \text{ Å}) \) [24] is the lattice constant of GaAs. The As–Ga bonds are mostly covalent in nature, but also have an ionic component due to the difference in electronegativities for the As and Ga; the Ga atoms represent cations while the As represent anions. sp\(^3\) hybridization of the atomic orbitals leads to a tetragonal coordination of the next nearest neighbor environment for the As and Ga; the Ga atoms represent cations while the As represent anions. The \{1 0 0\} planes of bulk GaAs consist of alternating planes entirely of either all As or Ga; therefore the ideal polar GaAs(0 0 1) surface could be either As or Ga terminated. In either case, each surface atom would have two dangling bonds that are unstable. The atoms move from their bulk positions in order
character, but also an ionic part since the electronegativities are smaller for the metal (Ga) than for the non-metal (As). By convention the cation Ga and the anion As are also referred to as type A and type B atoms respectively [20, 21].

For the following discussion it is important to recognize that all the surfaces are crystallographically equivalent. To improve consistency within this review all the GaAs$_{100}$ surfaces used by different research groups will be quoted as GaAs(001) and the in-plane directions will be transformed accordingly (see also Appendix B).

GaAs cleaves along $f_{110}$ planes, which contain the same number of Ga and As atoms per unit area and are thus intrinsically neutral. Each surface atom has only three nearest neighbours leaving one unpaired electron (dangling bond) per atom. The surface termination is almost bulk-like with planar zigzag chains of alternating cations and anions. No reconstructions are observed, but the surface atoms relax slightly from their positions in the bulk leading to a tilting of the cation–anion zigzag chains [21].

$f_{110}$ planes in bulk GaAs are occupied by either cations or anions in an alternating fashion. An ideally terminated GaAs(001) surface is therefore polar with a Ga–As-stoichiometry of either 1 : 0 (purely Ga terminated) or 0 : 1 (purely As terminated) and in either case two dangling bonds per surface atom. The composition of real GaAs(001) surfaces deviates usually from these ideal stoichiometric ratios.

Figure 1. Zincblende crystal structure of GaAs. The inset details the directions of the tetrahedral bonds on a Ga atom.

Figure 2.7: The crystal structure of GaAs. The insert shows the directions of the tetrahedral bonds on a Ga atom. Figure reproduced from ref. [25] with kind permission from Taylor and Francis (www.tandfonline.com).

to make new bonds. The result is that the surface stoichiometry deviates from purely As or Ga and a rather large range of surface reconstructions can occur depending on whether the surface is As or Ga rich amongst other factors. The surface reconstructions serve to reduce the large surface energy associated with the above mentioned dangling bonds.

While the quantitative understanding of surface reconstructions of semiconductors is quite complicated, Duke [26] presented a series of general principles derived from both calculations and experiments describing the surface structures of semiconductors in a qualitative manner. The principles are in the context of the main features of the surface chemistry. The principles relevant to GaAs(001) are the ones pertaining to tetrahedrally coordinated compound semiconductors, as further detailed by Wastlbauer and Bland [25], and are summarized in the following:

1. The surface structure observed will be the lowest free energy structure kinetically accessible under the preparation conditions.

2. For a given surface stoichiometry, the surface atomic geometry is determined primarily by a rehybridization induced lowering of the surface state bands associated with the (filled) anion dangling bond states.
3. Surfaces tend to be autocompensated.

Principle 1 states that each possible surface reconstruction corresponds to a local minimum in surface free energy and that the final minimum reached depends on the energetics. The energy difference between the different surface structures and stoichiometries is often small [25]. The final surface reconstruction will correspond to the lowest free energy minimum that can be reached by the process conditions used to prepare the system, i.e. the surface reconstruction will depend strongly on the condition of preparation. While modifications in the surface preparation can lead to very different surface reconstructions, carefully controlled preparation will lead to reproducible surface reconstructions.

Principle 2 leads to the paired bonding of neighboring surface atoms called dimerization. Dimerization serves to cut the number of dangling bonds in half, as compared to the ideal bulk terminated surface. The paired atoms are called surface dimers and form along the direction of the dangling bonds. The dimers typically form in rows with high long range order. The dangling bonds lie in the (1\(\bar{1}\)0) plane for the Ga terminated surface and the (1\(\bar{1}\)0) plane for the As terminated surface. The dimer directions are [1\(\bar{1}\)0] on As terminated surfaces and [1\(\bar{1}\)0] on Ga terminated surfaces. Dimerization leads to a surface periodicity 2 times the bulk value parallel to the dimer directions.

Principle 3 states an important constraint which limits the possible stoichiometries of compound semiconductor surfaces, the requirement that no charge accumulate at the surface. Polar compound semiconductor surfaces will only form structures whose dangling bond valence band states are filled and dangling bond conduction band states are empty. At the GaAs(001) surface, the As dangling bond states are below the valence band and the Ga dangling bond states are in the conduction band, both with respect to bulk GaAs [25]. Principle 3 requires filling of all the As dangling bond states while emptying the Ga dangling bond states. This is not possible on entirely As or Ga terminated GaAs(001) surface. Instead, missing dimer states are formed that serve to compensate the deficiency (excess) of electrons in the outer surface layer by transfer of charge from (to) the second partially exposed atomic layer; this process is autocompensation. Autocompensation at the GaAs(001) surface produces an N-fold periodicity normal to the 2-fold periodicity caused by principle 2.

The convention for expressing the surface reconstruction for GaAs(001) is to list the periodicity along the [1\(\bar{1}\)0] direction (\(A\)) first and along the [1\(\bar{1}\)0] direction (\(B\)) second, i.e. \(A \times B\). \(A\) and \(B\) are integer multiples of the atomic spacing in their respective directions. Xue et al. [27] have presented a rather comprehensive scanning tunneling microscopy (STM) study of the varying GaAs(001) surfaces.
2.2.1 GaAs substrate preparation

Since the substrates are exposed to air prior to loading into the UHV system, they require surface cleaning to remove oxides and other contaminants. In order to have a high reproducibility of the GaAs(001) surface reconstruction, the following substrate preparation recipe is performed:

1. Outgassing to 400°C.
2. 20 minutes of H· cleaning as described in Section 2.1.2.
3. 3+ hours of Ar+ etching as described in Section 2.1.1.
4. 40-50 minutes of annealing to a temperature of up to 600°C.

Outgassing is performed in the Intro chamber by slowly increasing the substrate temperature to \(\sim 400^\circ\) C until the pressure in the Intro chamber recovers to \(\sim 2 \times 10^{-10}\) Torr. The largest pressure increase is \(\sim 100\) C, indicating that outgassing mainly desorbs water from the substrate.

Longer H· cleaning times were found to reduce the Ar+ sputtering times required to remove O and C contaminants fully; however, it was found that much longer times lead to a less desirable surface, which result in long range inhomogeneities in the deposited Fe films as evident in an increase of the zero frequency offset in the ferromagnetic resonance linewidth as function of frequency. Prior to Ar+ sputtering, AES is used to check the contamination of the surface. After H· cleaning, residual amounts of C and O surface contaminants are generally still present. After 3 hours of Ar+ sputtering, AES is again used to check for contaminants O and C. This can be performed simultaneously to sputtering. The Ar+ sputtering is continued until the O and C contaminants are removed. The total Ar+ sputtering time generally needed is between 3 – 4 hours.

GaAs surface annealing

Once the substrate has determined to be free of surface contaminants, it is then moved into the MBE chamber for annealing. The pseudo \((4 \times 6)\) surface reconstructed GaAs has been shown to be a suitable template for the growth of high quality single crystal Fe layers \([28, 29]\). The pseudo \((4 \times 6)\) reconstruction is a hybrid of Ga rich \((4 \times 2)\) reconstructed regions with Ga dimers forming in the \([110]\) direction and As rich \((2 \times 6)\) reconstructed regions with As dimers forming along the \([1\bar{1}0]\) direction. The notation of referring to the pseudo \((4 \times 6)\) reconstructions as \(p(4 \times 6)\) will be adopted in the remained of this thesis.

At the annealing temperatures, the surface atoms have enough energy to become mobile and are free to self-assemble into a structure having a minimum surface energy. During the
annealing process, the GaAs surface is monitored by RHEED. The RHEED pattern data allows one to observe surface transformations in real time and provides continuous data for monitoring the surface symmetry.

Annealing is performed until a well ordered $p(4 \times 6)$ reconstruction is observed. In practice, the surface periodicity along the $[1 \overline{1} 0]$ direction is monitored with the RHEED beam oriented along the $[\overline{1} \overline{1} 0]$ crystallographic axis of GaAs. The $\times 6$ periodicity becomes faintly observable at $\sim 500^\circ$ C. At temperatures $\sim 600^\circ$ C the $\times 6$ reconstruction becomes well defined and the annealing is stopped. The $4\times$ reconstruction along the $[1 \overline{1} 0]$ can be observed with RHEED beam oriented along the $[1 \overline{1} 0]$ direction when the annealing temperature reaches $\sim 400^\circ$ C and remains throughout the annealing process. When the substrate is cooled, the surface reconstruction remains the same since it is in its lowest local minimum energy accessible during this annealing process, see Figure 2.9. Previous scanning tunneling microscopy (STM) studies at SFU have shown that the $p(4 \times 6)$ reconstruction obtained by using this surface preparation technique is dominated by the As rich $(2 \times 6)$ reconstruction with 95% coverage [30]. Example STM images provided by the authors of ref. [30] and ball-and-stick models by Xue et al. [27] of the $p(4 \times 6)$ reconstruction are shown in Figure 2.8.

2.2.2 MBE growth technique

All metallic structures grown by means of MBE in this study were deposited at pressures $< 3 \times 10^{-10}$ Torr. Deposition was performed at room temperature. However, the thermal load from the furnace can cause the sample temperature to rise upwards of 65$^\circ$ C. The deposition rate was selected to be $\sim 1$ AL/min. The film thickness were monitored by means of RHEED oscillations and a quartz crystal thickness monitor, see Figures 2.4 and 2.6. The RHEED intensity and thickness are both tracked by computer software (KSA400) as a function of time. By matching up the time axes, one can plot the RHEED intensity as a function of the thickness monitor reading.

The RHEED intensity as a function of thickness for quasi layer-by-layer growth can generally be fit with a damped sinusoid on an experimental background

$$I_{\text{RHEED}}(\Delta d) = A_1 e^{-\frac{\Delta d}{\lambda_1}} \sin \left( \frac{2\pi}{T} (\Delta d - \phi) \right) + A_2 e^{-\frac{\Delta d}{\lambda_2}} + A_3 (\Delta d - d_0)^2,$$

(2.12)

where $\Delta d$ is the increase in film thickness during growth, $A_1 e^{-\frac{\Delta d}{\lambda_1}}$ describes the damping of oscillation amplitude on the length scale $\lambda_1$, $T$ is the period of oscillation (desired thickness calibration parameter), $\phi$ describes the phase of the oscillation at the beginning of the considered fit, $A_2 e^{-\frac{\Delta d}{\lambda_2}}$ describes a decaying background on the length scale $\lambda_2$, and
Figure 2.8: STM images of the $p\,(4\times6)$ reconstruction for (a) a region displaying only the $(2\times6)$ reconstruction and (b) a region displaying areas of both the $(2\times6)$ and $(4\times2)$ reconstructions (images courtesy of the authors of ref. [30]). The directions labeled in (a) apply for (b) as well. Ball-and-stick model for the (c) $(2\times6)$ and (d) $(4\times2)$ reconstructions (reproduced from ref. [27] with permission from Elsevier).
Figure 2.9: Example RHEED images of the preparation of the GaAs(001) substrate with the RHEED beam along the [1\bar{1}0] direction. The top image shows the room temperature RHEED pattern of the GaAs substrate after 20 min. of H· cleaning and 3 hrs. of Ar⁺ sputtering. The higher order diffraction spots show that the surface is 3 dimensional. The middle image shows the surface reconstruction at 585°C. The ×6 reconstruction has become sharp and annealing is stopped. The bottom image shows the surface reconstruction after the sample has cooled to room temperature, where the sharp ×6 reconstruction remains. The diffracted images are below the direct beam as the sample holder faces down in the MBE chamber.
$A_3(\Delta d - d_0)^2$ allows for curvature beyond a decaying exponential background. The oscillations are fit in the growth region where there is steady layer-by-layer growth. Therefore $\Delta d = d(t) - d(t_0)$, where $t_0$ is the time corresponding to the start of the fit region.

Figure 2.10: RHEED specular spot intensity as a function of Fe layer thickness. The solid blueish line is the experimental data. The solid black line is the fit using Equation (2.12). The dashed grey line is the contribution from the background given by the last two terms on the RHS of Equation (2.12). The period of oscillation is used to define the thickness calibration parameter $T = 1.87 \pm 0.01 \text{Å}/\text{AL}$.

**Growth of Fe on GaAs(0 0 1)-p(4 × 6)**

The first layer deposited on all GaAs substrates was bcc Fe. The lattice constant for bcc Fe is $a_{\text{Fe}} = 2.865 \text{ Å}$ and for GaAs $a_{\text{GaAs}} = 5.6535 \text{ Å}$; this means that $a_{\text{Fe}}$ is 1.4% larger than the half unit cell of GaAs. For GaAs(0 0 1)|Fe, the inplane epitaxial orientations is $[1 0 0]_{\text{Fe}} \parallel [1 0 0]_{\text{GaAs}}$.

Example RHEED oscillations during the growth of 17 AL of Fe are shown in Figure 2.4. The growth from 48-300 s illustrates the transition from the GaAs|Fe interface to a continuous Fe layer by the deposition of the equivalent of $\sim 4 \text{ AL}$. After 300 s, the increase in the specular spot intensity and the regular RHEED oscillations indicate good quasi layer-by-layer growth. In this region, the RHEED diffraction pattern for Fe emerges and the presence and spacing of the diffracted beams reveal that the Fe grown is a bcc single crystal structure, see Figures 2.9 and 2.11, which are at the same scale.
An example thickness monitor calibration fitting using Equation (2.12) for the Fe growth shown in Figures 2.4 and 2.6 is shown in Figure 2.10. The fit thickness calibration parameter is $T = 1.87 \pm 0.01$ Å*/AL. The total growth read by the thickness monitor is $31.65 \pm 0.05$ Å*, resulting in a total growth of $16.9 \pm 0.1$ AL.

**Growth of Au and Pd films**

Both fcc Au and fcc Pd are known to grow well on GaAs|Fe(001). The lattice constant for fcc Au is $a_{Au} = 4.0786$ Å[31], which represents a less than 1% mismatch with the Fe(001) diagonal ($= 4.0537$ Å). The inplane epitaxial orientations is $[100]_{Au}||[110]_{Fe}$, representing a 45° rotation of the Au(001) layer with respect to both the GaAs(001) and Fe(001) layers. The lattice constant for fcc Pd is $a_{Pd} = 3.8898$ Å[32], which represents a 4% mismatch with the Fe(001) diagonal. The inplane epitaxial orientations is $[100]_{Pd}||[110]_{Fe}$, again representing a 45° rotation of the Pd(001) layer with respect to both the GaAs(001) and Fe(001) layers. The lattice mismatch between the Au and Pd layers is 4.6% and the orientation of each deposited on one another is $[100]_{Au}||[100]_{Pd}$. Example RHEED patterns for the surfaces of Au and Pd are shown in Figure 2.11.

For all films where Au is not already the final layer in the desired structure, 20 AL of Au is deposited as the final capping layer to prevent oxidation and other contamination of the films during ex situ characterization.
Figure 2.11: RHEED images during the growth of the sample GaAs|17Fe|100Au|50Pd|20Au with the beam oriented along the [1 1 0] direction of GaAs and Fe. The images are presented at the same scale as Figure 2.9 for spacing comparisons. (top left) The diffraction pattern for 17Fe. (bottom left) The diffraction pattern for 100Au. The ×2 reconstruction is consistent with As acting as a surfactant [33]. (top right) The diffraction pattern for the 50Pd layer. (bottom right) The diffraction pattern for the 20Au capping layer. The spacing well defined streaks are a consequence of the reciprocal lattice rods of the smooth surface intersecting an Ewald sphere of finite thickness. The Ewald sphere has a finite thickness as the RHEED beam consists of electrons within a narrow band of energies. The spacing is given by the plane spacing perpendicular to the incident beam, except for the 100Au layer where the spacing is halved by the ×2 reconstruction.
Chapter 3

Ferromagnetic Resonance -
Theoretical Considerations

This chapter serves as an introduction to the well established concepts required to understand the experimental ferromagnetic resonance results presented throughout this thesis. While most of the described concepts are true for magnetism in general, the focus of this chapter will be on how these concepts relate to the ferromagnetic resonance measurements and the magnetic properties of thin films. The theoretical background for the concepts of spin pumping and spin transport will be described in Chapter 6.

3.1 Magnetic free energy

The crystallographic direction of the magnetization is determined by the minimum of the total free-energy density $E_{\text{tot}}$, which in an external magnetic field can be expressed as the sum

$$E_{\text{tot}} = E_{\text{ex}} + E_{\text{Zeeman}} + E_{\text{ani}} + E_{\text{demag}},$$

(3.1)

where $E_{\text{ex}}$ is the energy associated with the exchange interaction, $E_{\text{ani}}$ is the energy associated with the crystallographic anisotropy, $E_{\text{demag}}$ is the demagnetization energy associated with the dipole-dipole interaction, and $E_{\text{Zeeman}}$ is the energy associated with the magnetization and the external magnetic field. $E_{\text{ani}}$ and $E_{\text{demag}}$ are phenomenologically determined by symmetry considerations of the sample. I will describe in detail only the $E_{\text{ani}}$ and $E_{\text{demag}}$ symmetries relevant to this thesis. A detailed overview of common crystalline symmetries and related anisotropies is given by Lindner and Farle [34] and Heinrich [35]; the magnetoelastic anisotropies by Sander [36]; and the dipolar interaction by Hurben and Patton [37].
3.1.1 Exchange energy

In ferromagnetic materials, the exchange energy favors the parallel alignment of spins (individual magnetic moments). The length scale over which the exchange energy dominates the dipole energy is called the exchange length given as

\[ \delta_{\text{ex}} = \sqrt{\frac{A_{\text{ex}}}{2\pi M_s^2}}, \]  

(3.2)

where \( A_{\text{ex}} \) is the exchange constant and \( M_s \) is the saturation magnetization [38]. For Fe, \( A_{\text{ex}} = 2 \times 10^{-6} \) erg/cm and \( M_s = 1710 \) emu/cm\(^3\), resulting in \( \delta_{\text{ex}} = 3.3 \) nm [38]. Magnetic films whose thickness is comparable to or smaller then \( \delta_{\text{ex}} \) have their spins locked together by the exchange interaction across the film thickness; such magnetic films are referred to as ultrathin and can be treated as a single macrospin. All of the films in this thesis will be treated in the ultrathin limit.

3.1.2 Zeeman energy

The presence of an external magnetic field introduces the Zeeman energy density term

\[ \mathcal{E}_{\text{Zeeman}} = -M \cdot H_{\text{dc}}, \]  

(3.3)

where \( M \) is the saturation magnetization vector and \( H_{\text{dc}} \) is the external dc magnetic field vector with magnitude \( H \).

3.1.3 Magnetic anisotropy energy (MAE)

In ferromagnets, the magnetization has energetically preferential directions governed by the symmetry of the lattice and shape of the sample. The magnetic energy is therefore dependent on the orientation of \( M \), this dependence is referred often to as magnetic energy anisotropy (MAE). The MAE is given by the work required to rotate the magnetization from an energy minimum (easy axis or plane) to any arbitrary direction. Assuming a constant temperature, the work required given by the change in free energy, \( dF = dW - SDT \) reduces to \( dF = dW \), where \( F \) is the free energy, \( W \) is the work, \( S \) is entropy, and \( T \) is temperature. Therefore the MAE is

\[ \mathcal{E}_{\text{MAE}} = F_2 - F_1 = \int_1^2 dW_{\text{MAE}}, \]  

(3.4)

where 1 represents the initial and 2 represents the final direction of the magnetization. Symmetry consideration are usually used to find the phenomenological expressions for \( F \).
There are two fundamental sources of magnetic anisotropy: (i) the spin-orbit interaction and (ii) the magnetic dipole-dipole interaction. In general, both the spin-orbit interaction and the dipole-dipole interaction are a few orders of magnitude weaker than the exchange interaction; however, they tie the magnetic moments to positions in space. It is worth noting that in the literature, the term MAE often refers to the magnetic anisotropy energy due to the spin-orbit interaction only, i.e. the magneto-crystalline energy and magneto-elastic energy. The magnetic anisotropy energies due to the dipole-dipole interaction are often called magneto-static anisotropies or simply shape anisotropies.

In the ferromagnetic resonance experiment, the experimentally accessible parameters are fields and angles. An expression for the effective anisotropy field can be found by considering the torque exerted on $\mathbf{M}$ by the effective magnetic field inside the sample. In equilibrium, the torque exerted $\mathbf{M} \times \mathbf{H}_{dc}$ by the external field on the magnetization must be compensated by the torque exerted by the anisotropy field.

**Crystalline anisotropy energy**

In crystalline ferromagnets, the magnetization has energetically preferential directions governed by the symmetry of the lattice. The magnetic energy is therefore dependent on the orientation of the magnetization with respect to the crystallographic axes; this dependence is often called magneto-crystalline anisotropy.

The treatment of anisotropies is made easier by the concept of the directional cosines of the magnetization $\mathbf{M}$. The directional cosines are given by

$$\alpha_i = \frac{\mathbf{M}}{M_s} \cdot e_i,$$

(3.5)

where $M_s$ is the magnitude of the magnetization and $e_i$ are unit vectors in the $i = X, Y, Z$ laboratory system of coordinates. In this thesis, the $X, Y, Z$ directions will be defined by principle crystallographic axes $\langle 100 \rangle$. Therefore,

$$\alpha_X = \sin \phi_M \cos \theta_M$$

(3.6)

$$\alpha_Y = \sin \phi_M \sin \theta_M$$

(3.7)

$$\alpha_Z = \cos \phi_M,$$

(3.8)

see Figure 3.1.
Figure 3.1: Schematic of the laboratory and magnetization coordinate systems. The laboratory system of coordinates is described by $X, Y, Z$. $\mathbf{M}$ is the magnetization and $\mathbf{H}_{dc}$ is the applied magnetic field. The $x, y, z$ coordinates are used to describe the magnetization dynamics, with $x \parallel \mathbf{M}$ and $y \parallel XY$ plane.

Cubic and Tetragonal anisotropy

The Fe films in this thesis in general have an approximate tetragonal symmetry. The lattice mismatch at the GaAs|Fe interface leads to an elastic deformation of the crystal structure. For this reason, it is convenient to replace the cubic symmetry of the bulk Fe with an inplane fourfold symmetry and a perpendicular to plane uniaxial symmetry. This leads to the tetragonal energy density [34, 35]

$$E_{\text{tetra}} = -\frac{1}{2}K_1^\parallel (\alpha_x^4 + \alpha_y^4) - \frac{1}{2}K_1^\perp \alpha_Z^4, \quad (3.9)$$

where $K_1^\parallel$ is the inplane fourfold anisotropy energy constant and $K_1^\perp$ is the perpendicular to plane fourfold anisotropy energy constant. If $K_1^\perp = K_1^\parallel \equiv K_1$, then one recovers the
cubic energy density,
\[ E_{\text{cubic}} = K_1 \left( \alpha_\chi^2 \alpha_Y^2 + \alpha_\chi^2 \alpha_Z^2 + \alpha_Y^2 \alpha_Z^2 \right). \]  
(3.10)

This can easily be seen from the simple expansion,
\[ \alpha_\chi^4 + \alpha_Y^4 + \alpha_Z^4 = 1 - 2 \left( \alpha_\chi^2 \alpha_Y^2 + \alpha_\chi^2 \alpha_Z^2 + \alpha_Y^2 \alpha_Z^2 \right). \]  
(3.11)

**Bulk and surface anisotropies**

The reduced symmetry at the surfaces of the ferromagnet can lead to enhancements of the spin-orbit interaction, which contributes to the magneto-crystalline anisotropy energy. For ultrathin films, this surface anisotropy energy is shared by means of the exchange interaction with the entire film.

The bulk and surface contributions to the magneto-crystalline anisotropy energy densities \( K \) can be distinguished by their dependence on the ferromagnetic layer thickness \( d_{\text{FM}} \).

For an extended film with two surfaces, top and bottom,
\[ K_j = K_j^\text{bulk} + \frac{K_j^\text{top}}{d_{\text{FM}}} + \frac{K_j^\text{bot}}{d_{\text{FM}}}, \]  
(3.12)

where \( j \) stands for any anisotropy energy density constant.

The ultrathin films also have a perpendicular uniaxial anisotropy energy density of the form
\[ E_\perp^\perp = -K_\perp^u \alpha_Z^2, \]  
(3.13)

where \( K_\perp^u \) is the perpendicular uniaxial energy constant. This term is due to the sharp change in spin-orbit coupling at the interfaces of the ferromagnet as compared to the bulk [35]. It is not a surface enhancement of the perpendicular fourfold anisotropy as it has a different angular dependence than that associated with \( K_\perp^1 \). It is in general present regardless of the internal symmetry of the ferromagnet. In fact for ultrathin films, this term often is the dominant surface anisotropy.

The Fe samples grown on the 4×6 surface reconstructed GaAs(0 0 1) also have a inplane uniaxial anisotropy,
\[ E_\perp^\perp = -K_\perp^u \frac{(u \cdot M)^2}{M^2} = -K_\perp^u \sin \phi_M \cos^2 (\theta_u - \theta_M), \]  
(3.14)

where \( K_\perp^u \) is the inplane uniaxial energy density constant, \( u = (\cos \theta_u, \sin \theta_u, 0)_{X,Y,Z} \) is the unit vector in the direction of the uniaxial anisotropy with the angle \( \theta_u \) measured from the \( X \) axis. In these films, \( u \) points in the \( [1 \bar{1} 0] \) (\( \theta_u = -45^\circ \)).
Demagnetization energy

The finiteness of a given ferromagnetic sample means that there will be pseudo magnetic surface charges resulting in a dipole-dipole interaction energy. In the limit of ultrathin films, the inplane dimensions of the sample, \(l_X\) and \(l_Y\), are much larger than the thickness of the sample \(d\) \((l_Z = d)\). In such a case, the thin film can be approximated as an infinite sheet; when the magnetization lies in the film plane, the pseudo magnetic surface charges are avoided, corresponding to a minimum in the demagnetization energy. If the magnetization is tilted with respect to the sample plane, the pseudo magnetic surface charges lead to a demagnetizing (restoring) energy density given by

\[
\mathcal{E}_{\text{demag}} = 2\pi DM_s^2\alpha Z^2, \tag{3.15}
\]

where \(D\) is the demagnetization factor and \(\phi_M\) is the azimuthal angle of the magnetization. For ultrathin films thicker than a few atomic layers (AL), \(D \approx 1\). The demagnetization energy has a form corresponding to a uniaxial anisotropy with a hard axis in the \(Z\) direction and an easy \(XY\) plane. For ultrathin films, the demagnetization energy can often be the dominating internal energy term.

3.2 Magnetic equations of motion

This section describes the equations of motion of the magnetization vector \(M\). Throughout this thesis, the Landau-Lifshitz-Gilbert equation of motion is used and is described in Section 3.2.1. Section 3.2.2 describes the Landau-Lifshitz equation of motion and compares it to the Landau-Lifshitz-Gilbert equation of motion; Section 3.2.2 can be passed over if one is not interested in the subtleties.

3.2.1 Landau-Lifshitz-Gilbert equation of magnetization dynamics

The dynamics of the magnetization \(M\) in an effective field \(H_{\text{eff}}\) is given by the Landau-Lifshitz-Gilbert (LLG) equation of motion

\[
\frac{\partial M}{\partial t} = -\gamma [M \times H_{\text{eff}}] + \alpha \left[M \times \frac{\partial n}{\partial t}\right], \tag{3.16}
\]

where \(\gamma\) is the absolute value of the gyromagnetic ratio, \(\alpha\) is the dimensionless Gilbert damping parameter, \(n\) is the direction of \(M\), and \(t\) is time.

The gyromagnetic ratio gives the magnetization per spin,

\[
\gamma = \frac{g\mu_B}{\hbar}, \tag{3.17}
\]
where \( g \) is the Landé factor for an electron with both spin and orbital momenta, \( \mu_B (= 9.27401 \times 10^{-21} \text{ erg/G}) \) is the Bohr magneton, and \( h (= 1.054572 \times 10^{-27} \text{ erg} \cdot \text{s}) \) is the reduced Planck constant. The free energies described in Section 3.1 enter the magnetization equations of motion in the form of internal effective fields by the relation [39]

\[
H_{\text{int}} = -\frac{\partial \mathcal{E}_{\text{tot}}}{\partial M}.
\]

(3.18)

These effective fields due to MAE and/or shape anisotropy are often referred to as internal fields; this is in contrast to externally applied magnetic fields, which are typically referred to as external fields.

The first term on the right hand side of Equation (3.16) describes a precessional torque on the magnetization by the effective \( H \) field. Even in the static case, \( M \) and \( H_{\text{dc}} \) do not have to be parallel. This fact is especially conspicuous when the crystalline anisotropy energy is comparable to the Zeeman energy and \( H_{\text{dc}} \) is not applied along an easy axis or plane. In this case, the magnetization will point somewhere between the easy direction and the applied \( H_{\text{dc}} \) field, that is \( m \) will be parallel to \( H_{\text{eff}} \). This effect is often referred to as magnetization dragging.

The second term on the right hand side of Equation (3.16) describes the magnetization relaxation as proposed by Gilbert. The dimensionless Gilbert damping parameter relates to the relaxation time by the relation

\[
\alpha = \frac{G}{\gamma M_s},
\]

(3.19)

where \( G \) is the original Gilbert damping parameter in units of \( s^{-1} \). For the experimental portions of this thesis (and often in the literature), it is the dimensionless Gilbert damping parameter \( \alpha \) being referred to as 'the Gilbert damping parameter' unless specifically stated otherwise.

### 3.2.2 Landau-Lifshitz equation of motion

The Gilbert damping in the LLG is a modification of Landau-Lifshitz (LL) equation of motion,

\[
\frac{\partial M}{\partial t} = -\gamma_{\text{LL}} [M \times H_{\text{eff}}] + \frac{G}{\gamma_{\text{LL}} M_s^2} [M \times [M \times H_{\text{eff}}]],
\]

(3.20)

where \( G \) is the Landau-Lifshitz damping parameter and \( \gamma_{\text{LL}} \) is again the absolute value of the gyromagnetic ratio.
The LL and LLG equations of motion are the same when $\alpha \ll 1$. Equation (3.20) can be transformed into Equation (3.16) by the substitutions

$$\frac{G}{\gamma_{\text{LL}} M_s} = \alpha$$

(3.21)

$$\gamma_{\text{LL}} = \gamma \frac{1}{1 + \alpha^2}.$$  

(3.22)

Both the LL and LLG equations of motion preserve the absolute magnetization, i.e $M \cdot M = M^2$.

The LL and LLG equations diverge for large values of $\alpha$ ($\approx 1$). In the case of LL, the damping term becomes dominant over the precessional torque term with increasing damping, representing an increasingly fast relaxation of $M$. For very large damping, the system would be in a constant quasi-equilibrium state. It is not surprising that Equation (3.20) leads to such physically unrealistic behavior for large damping, since the LL equation of motion was derived in the limit of small damping from general thermodynamic principles.

Gilbert’s modification to the equation of motion assumes a viscous like damping. Again with large damping, the damping term dominates over the precessional torque term; however in the LLG case, $\partial M / \partial t \to 0$. In the limit of infinite damping, the magnetic system approaches equilibrium at an infinitely slow rate.

### 3.3 Ferromagnetic resonance

In the ferromagnetic resonance (FMR) experiment, a microwave field $h_{\text{rf}} = h e^{i\omega t}$ excites the magnetization dynamics described by the LLG equation of motion, see Equation (3.16), where $\omega = 2\pi f$ is the microwave angular frequency. In this thesis, all the FMR measurements are done at a fixed microwave frequency $f$ while the external field $H_{\text{dc}}$ is stepped through resonance. Changing the external field $H_{\text{dc}}$ changes the precessional frequency of the magnetization $M$; when the precessional frequency is equal to the microwave frequency $f$, the sample undergoes FMR which is detected as an additional loss in the microwave signal.

Additionally, all of the FMR experiments in this thesis were done in the low (precession) angle limit. In this case, the magnitude of the microwave (rf) component of the magnetization $|m| \ll M_s$, and one can linearize Equation (3.16) with the approximation

$$M = M_s + m.$$  

(3.23)
Here $M_s$ is the longitudinal and $m$ is the transverse component of the total magnetization $M$ (described with respect to the static $M$).

The total effective field in Equation (3.16) is given by the sum of the internal and external fields,

$$H_{\text{eff}} = H_{\text{int}} + H_{\text{dc}} + h_{\text{hf}}.$$  (3.24)

It is often convenient to carry out the linearization of $M$ in the magnetization coordinate system $(x, y, z)$ rather than the laboratory coordinate system $(X, Y, Z)$. In the magnetization coordinate system, $x \parallel M$ and $y \parallel h_{\text{hf}}$ which lies in the $XY$ plane. For the small precessional angle $(|m| \ll M_s)$, the directional cosines in the laboratory system can be written in terms of the magnetization components in the magnetization coordinate system [35],

\[
\alpha_X = \frac{M_x}{M_s} \cos \theta_M \sin \phi_M - \frac{M_y}{M_s} \sin \theta_M - \frac{M_z}{M_s} \cos \theta_M \cos \phi_M
\]  (3.25)

\[
\alpha_Y = \frac{M_x}{M_s} \sin \theta_M \sin \phi_M + \frac{M_y}{M_s} \cos \theta_M - \frac{M_z}{M_s} \sin \theta_M \cos \phi_M
\]  (3.26)

\[
\alpha_Z = \frac{M_x}{M_s} \cos \phi_M + \frac{M_z}{M_s} \sin \phi_M
\]  (3.27)

see Figure 3.1 for diagram defining each angle.

In the following two subsections, I will go through examples of the two most common measurement configurations of the FMR experiment, the inplane configuration Section 3.3.1 and the perpendicular to plane configuration Section 3.3.2.

### 3.3.1 Inplane configuration

In the inplane configuration for FMR measurements, both the magnetization $M$ and the applied magnetic dc field $H_{\text{dc}}$ lie in the plane of the film ($XY$ plane), therefore $\phi_M = \phi_H = 90^\circ$.

Using Equations (3.1), (3.18) and (3.25) to (3.27), one can calculate the effective internal field $H_{\text{int}}$ components due to magnetic anisotropies in the magnetization coordinate system $(x, y, z)$. The components are...
The external dc field is expressed in the \((x,y,z)\) system as

\[ H_{dc} = H \left[ \cos (\theta_M - \theta_H) x - \sin (\theta_M - \theta_H) y \right] \quad (3.31) \]

and the external microwave driving field is \( h_{rf} = h_{rf} e^{i\omega t} \). The total effective field interacting with the magnetization in the LLG equation, Equation (3.16), is given by Equation (3.24).

In the limit of small precession, it is convenient to write the magnetization vector as

\[ M = M_s x + m_{rf}^y y + m_{rf}^z z, \quad (3.32) \]

where \( x, y, z \) are unit vectors. Here I have made the substitutions \( M_x \rightarrow M_s \), \( M_y \rightarrow m_{rf}^y \), and \( M_z \rightarrow m_{rf}^z \), with the lowercase font used to illustrate that these components are small deviations from their equilibrium and are the consequence of microwave excitation.

One can then linearize the LLG in terms of the small \( rf \) components \( m_{rf}^y, m_{rf}^z, \) and \( h_{rf} \). Assuming the time dependence of the \( rf \) components is of the nature \( e^{i\omega t} \), the result is a set of two coupled equations in terms of \( m_{rf}^y \) and \( m_{rf}^z \):

\[ 0 = i\frac{\omega}{\gamma} m_{rf}^z + \left( B_{eff} + i\alpha \frac{\omega}{\gamma} \right) m_{rf}^y \quad (3.33) \]

\[ M_s h_{rf} = \left( H_{eff} + i\alpha \frac{\omega}{\gamma} \right) m_{rf}^y - i\frac{\omega}{\gamma} m_{rf}^z \quad (3.34) \]
where

$$B_{\text{eff}} = H \cos (\theta_M - \theta_H) + 4\pi M_{\text{eff}} + \frac{K_1^\parallel}{2M_s} (3 + \cos 4\theta_M) + \frac{K_\perp}{M_s} \left(1 + \cos (2(\theta_M - \theta_U))\right)$$

(3.35)

$$H_{\text{eff}} = H \cos (\theta_M - \theta_H) + \frac{2K_1^\parallel}{M_s} \cos 4\theta_M + \frac{2K_\perp}{M_s} \cos (2(\theta_M - \theta_U)).$$

(3.36)

Since the demagnetization field and the perpendicular uniaxial anisotropy have the same angular dependence $\alpha_2^Z$, see Equations (3.13) and (3.15), they add linearly and one can express an effective demagnetization field as

$$4\pi M_{\text{eff}} = 4\pi M_s - \frac{2K_\perp}{M_s}.$$

(3.37)

The solution of the $x$ component of the LLG yields the result

$$H \sin (\theta_M - \theta_H) + \frac{K_1^\parallel}{2M_s} \sin 4\theta_M + \frac{K_\perp}{M_s} \sin (2(\theta_M - \theta_U)) = 0.$$

(3.38)

This equation is the static solution for the magnetization vector. It follows from the fact that in the static equilibrium case there can be no torque on the magnetization vector.

The transverse magnetic susceptibility can be determined from Equations (3.33) and (3.34) [35],

$$\chi_y \equiv \chi_y' + i\chi_y'' \equiv \frac{m_{y}}{\hbar} = \frac{M_s \left(B_{\text{eff}} + i\alpha_{\frac{\omega}{\gamma}}\right)}{\left(B_{\text{eff}} + i\alpha_{\frac{\omega}{\gamma}}\right) \left(H_{\text{eff}} + i\alpha_{\frac{\omega}{\gamma}}\right) - \left(\frac{\omega}{\gamma}\right)^2},$$

(3.39)

where $\chi'$ is the dispersive portion and $\chi''$ is the absorptive portion of the transversal rf susceptibility. Ferromagnetic resonance occurs when the denominator of the susceptibility function Equation (3.39) is minimized. Neglecting the small damping contributions $\alpha_{\frac{\omega}{\gamma}}$, the resonance condition in the parallel configuration is given as

$$\left(\frac{\omega}{\gamma}\right)^2 = B_{\text{eff}} H_{\text{eff}}|_{H=H_{\text{FMR}}}$$

(3.40)

In the linear precession regime, the $\chi_y'$ has a so called anti-Lorentzian lineshape and $\chi_y''$ has a Lorentzian lineshape (of the form $A/(1 - x^2)$). This can be seen by replacing $H$ in Equation (3.39) with $H_{\text{FMR}} + \delta H$, using Equation (3.40) to simplify the denominator, and linearizing in terms of small fields $\delta H$ and $\alpha_{\frac{\omega}{\gamma}}$, the result is:

$$\chi_y \simeq \frac{M_s}{B_{\text{FMR}} + H_{\text{FMR}}} \frac{B_{\text{FMR}} + \delta H + i\alpha_{\frac{\omega}{\gamma}}}{\delta H - i\alpha_{\frac{\omega}{\gamma}}},$$

(3.41)
\[ H_{\text{FMR}} = H_{\text{eff}} |_{H=H_{\text{FMR}}} \quad \text{and} \quad B_{\text{FMR}} = B_{\text{eff}} |_{H=H_{\text{FMR}}}. \]

Using the relation \( \delta H = H - H_{\text{FMR}} \) one can write

\[
\chi' \equiv \Re (\chi_y) \simeq M_s \frac{B_{\text{FMR}} - H_{\text{FMR}}}{B_{\text{FMR}} + H_{\text{FMR}}} \left( \frac{1}{B_{\text{FMR}}} + \frac{H - H_{\text{FMR}}}{(H - H_{\text{FMR}})^2 + \Delta H^2} \right) \tag{3.42}
\]

\[
\chi'' \equiv \Im (\chi_y) \simeq -M_s \frac{B_{\text{FMR}}}{(B_{\text{FMR}} + H_{\text{FMR}})} \left( \frac{\Delta H}{(H - H_{\text{FMR}})^2 + \Delta H^2} \right) \tag{3.43}
\]

where \( \Delta H = \alpha \omega \gamma \) is the half width at half maximum (HWHM) linewidth and \( H_{\text{FMR}} \) is the field value of \( H \) corresponding to the center of the absorption line, i.e. the resonance field. The term \( B_{\text{FMR}} / (B_{\text{FMR}} + H_{\text{FMR}}) \) is often called the ellipticity factor.

In general \( \theta_M \neq \theta_H \) due to the anisotropy fields. For large external fields the approximation \( \theta_M \approx \theta_H \) can be used to simplify the fitting of anisotropy fields or constants. In general however, it is preferable to use Equation (3.38) for the self consistent determination of \( \theta_M \) and the effective anisotropy fields.

### 3.3.2 Perpendicular configuration

In the perpendicular configuration for FMR measurements, both the magnetization \( M \) and the applied magnetic dc field \( H_{\text{dc}} \) are oriented perpendicular to the plane of the film (XY plane), therefore \( \phi_M = \phi_H = 0^\circ \).

Again in the small angle precession regime, \( M \) is given as Equation (3.32). Using Equations (3.1), (3.18) and (3.25) to (3.27), one can calculate the effective internal field \( H_{\text{int}} \) components due to magnetic anisotropies in the magnetization coordinate system \((x, y, z)\). The components linearized in terms of \( m_y^{\text{rf}} \) and \( m_z^{\text{rf}} \) are:

\[
H_{x,\text{int}} = \frac{2K_{u}^{\perp}}{M_s} + \frac{2K_{u}^{\perp}}{M_s} - 4\pi M_s \tag{3.44}
\]

\[
H_{y,\text{int}} = \frac{2K_{u}^{\parallel}}{M_s} M_y \tag{3.45}
\]

\[
H_{z,\text{int}} = 0 \tag{3.46}
\]

The external magnetic dc field is expressed in the \((x, y, z)\) system as \( H_{\text{dc}} = H x \) and the external microwave driving field is \( h_{\text{rf}} = h_{\text{rf}}^* e^{i\omega t} y \). The total effective field interacting with the magnetization in the LLG equation, Equation (3.16), is given by Equation (3.24).

The linearization of the LLG in terms of the small rf components \( m_y^{\text{rf}} \), \( m_z^{\text{rf}} \), and \( h_{\text{rf}} \), with the time dependence of the rf components of the nature \( e^{i\omega t} \), again results in a set of two coupled equations in terms of \( m_y^{\text{rf}} \) and \( m_z^{\text{rf}} \).
\[
0 = i\omega m_y^{rf} + \left(\mathcal{A}_1 + i\alpha \frac{\omega}{\gamma}\right) m_z^{rf}
\]  \hspace{.5cm} (3.47)

\[
M_s h^{rf} = \left(\mathcal{A}_1 + i\alpha \frac{\omega}{\gamma}\right) m_y^{rf} - i\omega m_z^{rf},
\]  \hspace{.5cm} (3.48)

where

\[
\mathcal{A}_1 = H - 4\pi M_{\text{eff}} + \frac{2K_\perp}{M_s}
\]  \hspace{.5cm} (3.49)

\[
\mathcal{A}_2 = \mathcal{A}_1 - \frac{2K_\parallel}{M_s}.
\]  \hspace{.5cm} (3.50)

The transverse magnetic susceptibility for the perpendicular configuration can be determined from Equations (3.47) and (3.48),

\[
\chi_y \equiv \chi_y' + i\chi_y'' \equiv \frac{m_y^{rf}}{h^{rf}} = \frac{M_s \left(\mathcal{A}_1 + i\alpha \frac{\omega}{\gamma}\right)}{\left(\mathcal{A}_1 + i\alpha \frac{\omega}{\gamma}\right) \left(\mathcal{A}_2 + i\alpha \frac{\omega}{\gamma}\right) - \left(\frac{\omega}{\gamma}\right)^2}.
\]  \hspace{.5cm} (3.51)

Neglecting the small damping contributions \(\alpha \frac{\omega}{\gamma}\), the resulting resonance condition for the perpendicular configuration is

\[
\left(\frac{\omega}{\gamma}\right)^2 = \mathcal{A}_1\mathcal{A}_2|_{H=H_{\text{FMR}}}.
\]  \hspace{.5cm} (3.52)

Noting that Equations (3.47) and (3.48) have the same form as Equations (3.33) and (3.34) with \(B_{\text{eff}} \rightarrow \mathcal{A}_1\) and \(H_{\text{eff}} \rightarrow \mathcal{A}_2\) and following a similar analysis as in Section 3.3.1, one can express

\[
\chi_y' \equiv \text{Re} (\chi_y) \simeq M_s \frac{\mathcal{A}_{1,\text{FMR}}}{\mathcal{A}_{1,\text{FMR}} + \mathcal{A}_{2,\text{FMR}}} \left(\frac{1}{\mathcal{A}_{1,\text{FMR}}} + \frac{H - H_{\text{FMR}}}{(H - H_{\text{FMR}})^2 + \Delta H^2}\right)
\]  \hspace{.5cm} (3.53)

\[
\chi_y'' \equiv \text{Im} (\chi_y) \simeq -M_s \frac{\mathcal{A}_{1,\text{FMR}}}{\mathcal{A}_{1,\text{FMR}} + \mathcal{A}_{2,\text{FMR}}} \left(\frac{\Delta H}{(H - H_{\text{FMR}})^2 + \Delta H^2}\right),
\]  \hspace{.5cm} (3.54)

where \(\mathcal{A}_{1,\text{FMR}} = \mathcal{A}_1|_{H=H_{\text{FMR}}}\) and \(\mathcal{A}_{2,\text{FMR}} = \mathcal{A}_2|_{H=H_{\text{FMR}}}\). One finds that the lineshape for the perpendicular configuration is the same as for the inplane configuration, with the HWHM given as \(\Delta H = \alpha \frac{\omega}{\gamma}\).

To be in the perpendicular configuration (\(M \parallel H_{\text{dc}}\)) requires an external field \(H_{\text{dc}}\) such that \(\mathcal{A}_2 > 0\).
3.4 Ferromagnetic resonance parameter extraction

In the FMR experiment, the microwave response signal \( \epsilon \chi \) is proportional to the rf magnetic susceptibility \( \chi = \chi' + i\chi'' \) in the direction of an applied rf field. In our analysis of data we are concerned mainly with line position and shape and not the total amplitude. The line position, or resonance field \( H_{\text{FMR}} \), is used to determine the anisotropy fields \( H_{\text{eff}} \) and \( \gamma \). Absolute measurements are possible and can be used to determine \( M_s \), however this process requires more measurements and more analysis time, see for example Celinski et al. [40].

In the linear response regime of Equation (3.16), the rf signal for the in-phase component (dispersion), for both inplane (Equation (3.42)) and perpendicular (Equation (3.53)) configurations, can be expressed as

\[
\epsilon \chi' = \mathcal{A} \left( \frac{1}{B_{\text{FMR}}} + \frac{H - H_{\text{FMR}}}{\Delta H^2 + (H - H_{\text{FMR}})^2} \right),
\]

(3.55)

and for the out-of-phase component (absorption), for both inplane (Equation (3.43)) and perpendicular (Equation (3.54)) configurations, can be expressed as

\[
\epsilon \chi'' = -\mathcal{A} \frac{\Delta H}{\Delta H^2 + (H - H_{\text{FMR}})^2},
\]

(3.56)

where

\[
\mathcal{A} = a \Delta S M_s \frac{d_{\text{FM}} B_{\text{FMR}}}{B_{\text{FMR}} + H_{\text{FMR}}},
\]

(3.57)

and \( \Delta S \) is the film surface area, \( d_{\text{FM}} \) is the FM film thickness, and \( a \) is a proportionality factor. Typically, FMR lineshapes are fit with \( \mathcal{A} \) as an arbitrary amplitude parameter.

In certain circumstances, one does not necessarily measure the orthogonal \( \epsilon \chi' \) and \( \epsilon \chi'' \) separately, but one can measure a mixture of these components, which can be viewed as their projection onto a rotated coordinate system of the rf susceptibility complex plane given by,

\[
\begin{pmatrix}
\epsilon_{\chi'}^{\text{mix}} \\
\epsilon_{\chi''}^{\text{mix}}
\end{pmatrix} =
\begin{pmatrix}
\cos \varphi & \sin \varphi \\
-\sin \varphi & \cos \varphi
\end{pmatrix}
\begin{pmatrix}
\epsilon_{\chi'} \\
\epsilon_{\chi''}
\end{pmatrix},
\]

(3.58)

where \( \varphi \) is the angle of rotation in the complex plane.

In this thesis, the FMR spectra has been fit as a mixed absorption signal,

\[
\epsilon_{\chi''}^{\text{mix}} = \epsilon_{\chi'} \sin \varphi + \epsilon_{\chi''} \cos \varphi.
\]

(3.59)
When using lock-in detection, one measures the derivative of the mixed absorption signal with respect to $H$, 

$$\chi'_{\text{mix,der}} = -\frac{2(H - H_{\text{FMR}}) \Delta H \cos \varphi + \left((H - H_{\text{FMR}})^2 - \Delta H^2\right) \sin \varphi}{\left((H - H_{\text{FMR}})^2 + \Delta H^2\right)^2}. \quad (3.60)$$

Measuring only one signal is equivalent to having an admixture of the in-phase and out-of-phase components of the rf susceptibility signal.

### 3.5 Magnetic damping in ultrathin ferromagnets

Magnetic damping is in general sample dependent. The contributions to the damping can however be partitioned into two generally distinct parts, intrinsic and extrinsic damping. Some damping mechanisms are unavoidable and these are referred to as intrinsic. In ultrathin films, the Gilbert damping is caused by spin-orbit interaction and contains both bulk and interface contributions. Since the interfaces cannot be avoided, their contribution to the Gilbert damping is considered intrinsic. Damping mechanisms that are related to structural defects or sample impurities are referred to as extrinsic since in principle they are possible to avoid. Experimentally, the smallest measured damping under well defined thermodynamic conditions is considered to be intrinsic to the system[41]. The intrinsic Gilbert damping due to spin-orbit interaction is described in Section 3.5.1. The intrinsic contributions to the damping due to Eddy currents and phonon drag have been shown to be negligible in ultrathin films and will not be discussed, see ref. [41]. The extrinsic contribution due to two magnon scattering is described in Section 3.5.3. The extrinsic contribution to the damping due to the spin pumping effect is described later in Chapter 6. The following subsections are based in part on the book chapter on spin relaxation by Heinrich [41].

#### 3.5.1 Gilbert damping in ferromagnets

Magnetic relaxation, or damping, is phenomenologically described by the second term on the right hand side of Equation (3.16). The Gilbert damping parameter is usually determined from the frequency dependence of the FMR linewidth:

$$\Delta H(f) = \frac{G M_s}{\gamma^2} \omega \left(=\frac{G M_s}{\gamma}\right). \quad (3.61)$$

Equation (3.61) shows the two hallmarks of Gilbert damping: (1) $\Delta H$ has a strictly linear dependence on the microwave frequency and is inversely proportional to the saturation
magnetization $M_s$. (2) There is no explicit dependence on the magnetic anisotropies or the applied field $H_{dc}$.

Kamberský [42, 43] presented models of the magnetic damping mechanisms in metallic ferromagnets based on the observation that the Fermi surface changes with respect to changes in the direction of the magnetization. As the precessing magnetization evolves in time and space, the Fermi surface also evolves periodically in time and space. This effect is known as a breathing Fermi surface. The electrons try to repopulate the changing Fermi surface through the creation of electron hole pairs, but are time delayed by the finite relaxation time of the electrons $\tau_m$. The relaxation of the electron hole pairs happens through scattering at defects or with phonons, transferring energy and angular momentum to the lattice. The time delay of these processes results in a phase lag between the Fermi surface evolution and the precessing magnetization. The phase lag for the breathing Fermi surface is directly proportional to the precession frequency $\omega$, the typical behavior for the viscous-like damping described by the LLG.

The repopulation of energy levels due the breathing Fermi surface is a dissipative process. The energy of the system contributing to the dissipative process depends on the magnetization and thus enters the LLG as an effective field. In a very simple model [42], the energy associated with the repopulation of the breathing Fermi surface can be expressed as

$$\mathcal{E}_{\text{dam}} = \sum_k n_k \mathcal{E}_k,$$

(3.62)

where $n_k$ is the population number per unit volume of an electron with wavevector $k$ with associated energy $\mathcal{E}_k$ near the Fermi level. When the magnetization precession rate is slower than the relaxation time of the electrons, the time dependence of the population number is given by

$$n_k(t) = n_{k,\text{eq}} - \tau_m \frac{\partial n_k(t)}{\partial t},$$

(3.63)

where $n_{k,\text{eq}}$ is the equilibrium population of the instantaneous magnetization. The time derivative of $n_k$ can be expressed as

$$\frac{\partial n_k}{\partial t} = \frac{\partial n_k}{\partial \mathcal{E}_k} \frac{\partial \mathcal{E}_k}{\partial M_i} \frac{\partial M_i}{\partial t},$$

(3.64)

43
with $M_i$ being the $i (=x, y, z)$ components of the instantaneous magnetization vector. The effective damping field can then be written using Equation (3.18) as

$$
H_{\text{dam},i} = -\sum_k n_k(t) \frac{\partial E_k}{\partial M_i}
$$

$$
= \frac{\tau_m}{M_s} \sum_k \delta (E_k - E_F) \left( \frac{\partial E_k}{\partial \alpha_i} \right)^2 \frac{\partial M_i}{\partial t},
$$

where $\alpha_i$ are the directional cosines, see Equation (3.5), and $\partial n_k/\partial E_k$ has been replaced with the delta function in the sum. One can see that the torque on the magnetization due to the effective damping field has the form of the Gilbert damping term in Equation (3.16),

$$
\frac{1}{\gamma} \frac{\partial M}{\partial t} = -[M \times H_{\text{dam}}]
$$

$$
\propto \left[ M \times \frac{\partial n}{\partial t} \right].
$$

Kamberský [43] showed that the intrinsic damping in metallic ferromagnets can be treated generally by using the spin-orbit interaction Hamiltonian. The spin-orbit Hamiltonian for the transverse spin and angular momentum can be expressed as a three particle interaction Hamiltonian [41],

$$
H_{\text{so}} = \frac{1}{2} \left( \frac{2S}{N} \right)^{1/2} \sum_k \sum_{\eta, \eta', \sigma} \langle \beta | L^+ | \eta \rangle c^+_{\beta, k+q, \sigma} c_{\eta, k, \sigma} b_q + \text{h.c.},
$$

where $N$ is the number of atomic sites, $S = M_s (T)/M_s (0)$ is the reduced spin, and $\xi$ is the coefficient of the spin-orbit interaction. $L^+ = L_x + iL_y$ is the right handed component of the transverse angular momentum $L_t$ at the atomic site. $c_{\eta, k, \sigma}$ and $c^+_{\beta, k+q, \sigma}$ annihilate and create electrons in the appropriate Bloch states with spin $\sigma$. The operator $b_q$ annihilates the spinwave with wavevector $q$. The indices $\eta$ and $\beta$ represent the projected local orbitals of the Bloch states and identify the individual electron bands. The rf susceptibility can be calculated using the Kubo Green function formalism in the Random Phase Approximation. The resulting damping field is

$$
\frac{G \omega}{\gamma M_s \gamma} = \frac{\langle S \rangle^2}{2M_s \xi^2} \left( \frac{1}{2\pi} \right)^3 \int dk^3 \sum_{\eta, \beta, \sigma} \langle \beta | L^+ | \eta \rangle \langle \eta | L^- | \beta \rangle
$$

$$
\times \delta (\epsilon_{\eta, k, \sigma} - \epsilon_F) \hbar \omega \frac{h/\tau_m}{(h\omega + \epsilon_{\eta, k, \sigma} - \epsilon_{\beta, k+q, \sigma})^2 + (h/\tau_m)^2},
$$

where $\tau_m$ is the electron momentum relaxation time, $\epsilon_{\eta, k, \sigma}$ is the energy of an electron in the $\eta$ band with wave vector $k$ and spin $\sigma$, and $\epsilon_F$ is the Fermi energy.
Intraband transitions ($\eta = \beta$): The electron energy balance term in Equation (3.70) can be expressed as $\hbar \omega + \epsilon_{\eta,k,\sigma} - \epsilon_{\beta,k+q,\sigma} = \hbar \omega - (\hbar^2/2m) (2k \cdot q + q^2)$. For low frequency spinwaves satisfying $q \ll k_F$, the electron energy balance term can be significantly less than $\hbar/\tau_m$. This limit is satisfied above cryogenic temperatures. The resulting Gilbert damping due to intraband transition can be approximated as

$$G \simeq \langle S \rangle^2 \left( \frac{\xi}{\hbar} \right)^2 \left( \sum_{\eta} \chi_{p}\eta \langle \eta \mid L^+ \mid \eta \rangle \langle \eta \mid L^- \mid \eta \rangle \right) \tau_m, \tag{3.71}$$

where $\chi_{p}\eta$ is the Pauli susceptibility of the states that participate in the intraband transitions and satisfy the low frequency limit. This approximation shows that the Gilbert damping in this limit is directly proportional to $\tau_m$ and therefore scales with the conductivity of the ferromagnet.

Interband transitions ($\eta \neq \beta$): Interband transitions are associated with energy gaps $\Delta\epsilon_{\beta,\eta}$ and are accompanied by changes to the electron wave functions. The electron hole pair energy can be dominated by the energy gaps. When the energy gaps are larger than the relaxation frequency $\hbar/\tau_m$, the Gilbert damping can be approximated as

$$G \simeq \langle S \rangle^2 \sum_{\eta} \chi_{p}\eta \left( \Delta g_{\eta} \right)^2 \frac{1}{\tau_m}, \tag{3.72}$$

where

$$\Delta g_{\eta} = 4\xi \sum_{\beta} \frac{\langle \eta \mid L_x \mid \beta \rangle \langle \beta \mid L_x \mid \beta \rangle}{\Delta\epsilon_{\beta,\eta}} \tag{3.73}$$

is the deviation of the g-factor from its purely electron spin value $g_s \simeq 2$ due to the spin-orbit contribution to the Landé g-factor. $\chi_{p}\eta$ only includes the electron states that have a change in energy during the interband transition that is much greater than the relaxation frequency, $\Delta\epsilon \gg \hbar/\tau_m$. Equation (3.72) shows that interband transitions lead to a Gilbert damping that is proportional to $1/\tau_m$ and thus scales with resistivity. However, this dependence is only valid at low temperatures. With increasing temperatures, the relaxation rate becomes comparable to the energy gaps, resulting in a saturation of the interband Gilbert damping [43].

3.5.2 Two magnon scattering

In addition to the intrinsic damping due to spin-orbit interaction, structural defects and inhomogeneities can have an important effect on the magnetic relaxation. One particularly important scattering process that affects the damping is the so-called two magnon scattering. Inhomogeneous magnetic properties can lead to the scattering of magnons. In FMR, the
uniform precession mode $k \sim 0$ can be scattered into nonuniform modes $k \neq 0$. Conservation of energy requires the scattered magnon to have the same energy of the uniform precession mode, but the loss of translational invariance means that the momentum need not be conserved.

In FMR, the $k = 0$ mode is scattered into magnons with non-zero $k$ with a scattering matrix that is proportional to the Fourier transform of the sample inhomogeneities \[44\]

$$A(k) = \int \Delta U(k) e^{-i \mathbf{k} \cdot \mathbf{r}} d\mathbf{r}$$

(3.74)

where $\Delta U(k)$ represents variations of the local magnetic anisotropy energy density.

Arias and Mills \[45\] introduced a theory of two magnon scattering for ultrathin ferromagnetic films. In ultrathin films the magnon $k$-vectors are confined to the plane of the film. Neglecting magnetocrystalline anisotropies and considering a magnetization oriented parallel to the external in-plane field, the spin wave manifold has the form:

$$\left( \frac{\omega_k}{\gamma} \right)^2 = \left( H_{FMR} + 4\pi M_{\text{eff}} - 2\pi M_s kd + Dk^2 \right) \left( H_{FMR} + 2\pi M_s kd \sin^2(\phi_k) + Dk^2 \right) ,$$

(3.75)

where $\phi_k$ is the angle between the magnetization and the scattered magnon $k (= k(\phi_k))$ and $D = 2A_{ex}/M_s$ is the exchange stiffness ($A_{ex}$ being the exchange energy). The term $-2\pi M_s kd$ in the first set of parenthesis on the RHS of Equation (3.75) is negative and proportional to $k$ and thus lowers the resonance frequency for magnons with $k > 0$. The next term $Dk^2$ is positive and proportional to $k^2$ and thus raises the resonance frequency for magnons with $k > 0$. For small $k$, the magnon resonance frequency initially decreases with increasing $k$. For further increasing $k$, the quadratic term begins to dominate, increasing the magnon resonance frequency. Eventually for a magnon $k_{\text{deg}}$, the magnon resonance frequency will be equal to the FMR frequency given by the $k = 0$ mode; at this point $k_{\text{deg}}$ and $k = 0$ are degenerate and two magnon scattering can occur. The terms $-2\pi M_s kd$ and $Dk^2$ represent the competition between the dipolar energy and the exchange energy. The magnitude of $k(\phi_k)$ reduces with increasing $\phi_k$. One can see from a simple multiplication of the terms in parenthesis on the RHS of Equation (3.75), that above the critical angle $\phi_{k,max}$, which is defined by

$$\sin^2(\phi_{k,max}) = \frac{H_{FMR}}{H_{FMR} + 4\pi M_{\text{eff}}} ,$$

(3.76)

the dipolar terms no longer lead to degenerate modes available for two magnon scattering.

A treatment of two magnon scattering for external fields applied at angles out of the plane can be found in ref. [41]. One important result is that no degenerate modes exist when the
external field is applied perpendicular to the film plane [45], and therefore the two magnon scattering process is absent in the perpendicular FMR configuration.

The two magnon scattering process leads to an increase of the FMR linewidth $\Delta H$. However, it has been shown that the two magnon scattering contribution to the linewidth saturates at material dependent frequencies. Below the saturation frequency, two magnon scattering leads to a nonlinear dependence of $\Delta H (f)$. The slope of $\Delta H (f)$ at higher frequencies than the saturation frequency can be used to recover the intrinsic damping parameter $\alpha$, see [44, 46]. This is because the two magnon scattering process is a mode conversion: the magnetic excitations do not disappear and still needs to be relaxed to the lattice system by the intrinsic damping mechanism governed by the spin-orbit interaction. The $k \neq 0$ modes are still damped to equilibrium by intrinsic damping.

3.5.3 Long range inhomogeneity line broadening

The intrinsic magnetic damping was shown to lead to an FMR linewidth that is linearly proportional to the microwave frequency, see Equation (3.61). In practice, however, one usually finds that even when the linewidth has a linear dependence, it is usually accompanied by an extrapolated offset $\Delta H (0)$. $\Delta H (0)$ is commonly referred to as the zero frequency offset. In this case, the experimentally measured linewidth as a function of frequency is interpreted as

$$\Delta H (f) = \alpha \frac{\omega}{\gamma} + \Delta H (0).$$

Equation (3.77)

The linear term in Equation (3.77) is the same as in Equation (3.61) and is taken to be a measure of the intrinsic damping.

McMichael et al. [47] have shown that the zero frequency offset is caused by parameter variations that cause different ferromagnetic resonance conditions in different areas of the film. One such variation is long range magnetic inhomogeneities, where the magnetic anisotropy effective fields can vary within the film. Assuming local inhomogeneous fields do not interact, they can simply add to the applied field giving a superposition of local resonances. The long range inhomogeneity line broadening results in a true zero frequency offset.

The long range inhomogeneity line broadening is very different than the two magnon scattering line broadening. In fact, $\Delta H (0)$ increases in the perpendicular FMR configuration where the inhomogeneous magnetic anisotropy fields contribute more effectively to the FMR field due to the lack of strong elliptical polarization in the parallel FMR configuration [44]. The two magnon scattering contribution has the opposite relation as it is absent in the perpendicular FMR configuration.
Chapter 4

Ferromagnetic Resonance Spectrometer

Ferromagnetic resonance (FMR) has been extensively made use of to study the magnetic properties in this thesis. FMR is a well understood experimental technique [48] that has proven to be an invaluable tool to probe the static (saturation magnetization and anisotropies) and dynamic (Landé g-factor and damping) magnetic properties of ultrathin films and multilayer nanostructures.

FMR occurs in the range of microwave frequencies. In field swept FMR, the FMR signal is determined by measuring the microwave losses in the studied sample at a constant frequency as a function of the applied external dc magnetic field $H_{dc}$. One can also measure FMR signals at constant $H_{dc}$ while sweeping the frequency. This is commonly performed using a vector network analyzer (VNA) [49–53].

In order to determine the spectrum of magnetic parameters, one needs to carry out FMR measurements at a wide range of frequencies. For this reason, it is of great advantage to have a broadband system where measurements can be made at multiple frequencies without the need to remount samples in different waveguides or cavities.

4.1 Traditional narrowband FMR spectrometer

A typical traditional ferromagnetic resonance spectrometer is based upon rectangular waveguides and generally narrow bandwidth microwave sources. The ferromagnetic resonance measurements presented in Chapters 5 and 8 were performed using FMR spectrometers of this nature.

The microwave sources for the ~ 9, 14, and 72 GHz measurements were reflex klystrons. A reflex klystron is a vacuum tube device that has more or less become obsolete as it
has been effectively replaced by semiconductor based devices. In the reflex klystron, an
electron beam is shot into one end of the vacuum tube by an electron gun and is made to
pass through grids in a single resonant cavity. The electron beam is velocity modulated as it
passes through the cavity grids which results in a bunching of electrons. The electron beam
is then reflected by a negatively charged repeller cathode and passes through the resonant
cavity a second time. The reflector voltage is adjusted to maximize the electron bunching so
that the maximum of energy is transferred to the rf oscillations in the cavity. The output
frequency is primarily determined by the geometry of the cavity; however, the reflector
voltage can be perturbed slightly to achieve a degree of frequency tuning (∼100 MHz).
The microwave sources for the ∼24,36 GHz spectrometers were Gunn diodes.

![Schematic of the FMR spectrometer using microwave cavities](image)

Figure 4.1: Schematic of the FMR spectrometer using microwave cavities. The microwave
generator used was either reflex klystron power supplies, Gunn diodes with varactor tuning,
or a broadband microwave generator. The sample is mounted in a microwave cavity and
FMR is detected by monitoring the reflected microwave power at the detector while stepping
the field.

A schematic representation of the FMR spectrometers is shown in Figure 4.1. The sam-
amples are mounted in tunable microwave cavities that are tuned to the microwave frequency
of the source (reflex klystron or Gunn diode), except for 72 Ghz measurements, which are
performed using a terminated waveguide. The tunable cavities are necessary due to the
limited frequency tuning of both the reflex klystron and Gunn diode microwave sources.

The microwave absorption is measured by monitoring the reflected power from the
microwave cavity using a diode detector. When the sample undergoes FMR, the microwave
losses are increased and the amplitude of the reflected signal changes. The microwave cavity
is placed between the pole pieces of a Varian 3800 electromagnet such that the sample is in
the center of the line connecting the centers of the magnetic pole pieces. Measurements are
made at a fixed frequency while the external field $H_{dc}$ is stepped through resonance. The Varian 3800 electromagnetic can be rotated to make an arbitrary angle with respect to the sample axes with a precision of 0.1°.

The cavities for the $\sim$24 and 36 GHz measurements were cylindrical and operated in the $\text{TE}_{011}$ mode, sometimes referred to as the doughnut mode. The cavity is coupled to the waveguide track through a small hole in the top end plate located approximately at the half radius. The sample is mounted on the top end plate inside the cavity, opposite the coupling hole. The cavity for the 9 GHz measurements was rectangular and operated in the $\text{TE}_{101}$ mode. The cavity is coupled to the waveguide track through a small hole in the center of the top end plate. The sample is mounted on the bottom end plate in the center.

As the field is stepped through resonance, the resonant frequency of the cavity can be slightly perturbed. The frequency of the microwave source is locked to the resonance frequency of the cavity. Locking is achieved by externally modulating the microwave frequency at 60 kHz using the reference output of a PAR lock-in amplifier. For the reflex klystron the reflector voltage is modulated and for the Gunn diodes the varactor voltage is modulated. The reflected power from the cavity is converted to a signal voltage by the detector diode which is monitored on the input of the PAR lock-in and is at a minimum when the source and cavity resonance frequencies are equal; in this case, the signal at the detector diode contains only the second harmonic (120 kHz) of the 60 kHz modulation and the signal on the PAR lock-in is zero. If the source and cavity resonance frequencies are not equal, the voltage at the detector diode will have a 60 kHz component whose phase depends on whether the source frequency is above or below the cavity resonance frequency. In this case the reading on the PAR lock-in is non-zero and the sign depends on the phase of the detector diode signal and therefore can be used as an error feedback signal to shift the source frequency to the cavity resonance frequency. The analog output voltage of the PAR lock-in is proportional to the input reading and is mixed with the reference modulation signal to provide feedback.

The FMR signals from ultrathin films are usually only a small perturbation to the reflected (or transmitted) microwave signal; for this reason, it is common practice to use lock-in detection to enhance the signal to noise ratio (SNR). It is most common to use field modulation (HM) to this end. HM was achieved by using a Kepco bipolar operational power supply/amplifier (model: BOP72-5M) to drive a set of Helmholtz modulation coils. A modulation frequency of 104-105 Hz was used. Greater modulation frequencies in principle could be used to allow a shorter time constant on the Lock-in amplifier, but the amplitude of modulation in our system is limited by the inductive reactance of the coils. Lock-in detection is performed with a Stanford Research Systems SR830 DSP Lock-In Amplifier.
It is important to note that the use of rectangular waveguide tracks and narrowband microwave sources essentially requires one to use a different FMR spectrometer for each frequency. This requires significant time to remount and align samples as well as hook up all necessary equipment. In the next section, I will describe two FMR spectrometer setups that are broadband in nature.

4.2 Broadband FMR spectrometer

In this section, it is demonstrated that one is able to create a broadband FMR system which has the sensitivity capable of detecting FMR signals in ultrathin films. This system is based on broadband, low noise components: an Anritsu MG3696B microwave generator (0.1 to 70 GHz) and an Agilent 8474E planar-doped barrier diode microwave detector (0.01 to 50 GHz).

Two microwave spectrometers were built during the work shown in this thesis:

4.2.1 A typical microwave spectrometer based on rectangular microwave waveguides with samples inserted inside a microwave cavity. In this case, however, a multimode cavity was machined and was used to allow the frequency dependence of the FMR spectrum to be collected with a single sample mounting. Using the multimode cavity one has the ability to measure four frequencies (∼27 – 40 GHz) and benefit from the sensitivity enhancement of the cavity measurement.

4.2.2 A coaxial microwave track with samples mounted on a coplanar waveguide.

Either the multimode cavity and coplanar waveguide can be mounted on the same mounting apparatus. The mounting apparatus has two tracks of WR-28 waveguide as well as coaxial cables.

Despite the different coupling between the film and microwave cavity compared to that of a coplanar waveguide, the FMR results can be analyzed in a simple manner using Equations (3.55), (3.56) and (3.59) and an instrumental background. The resulting magnetic properties are in very good agreement, as shown in Section 4.3.

4.2.1 Multimode cavity

The experimental setup for the multimode cavity with field modulation configuration (Cavity-HM), is the same as shown in figure Figure 4.1. The microwave signal is brought by means of coax cable from the microwave generator to a coax to waveguide coupler (Quinstar MN:QWA-28S24F). The coax to waveguide coupler is mounted on a waveguide directional coupler and the forward signal is directed to the center of the magnet by means of a WR-28 waveguide. The microwave signal is reflected from a termination plate mounted at the end.
of the waveguide, where the sample is mounted, and is detected by means of a zero-bias microwave detector (Quinstar MN:QEA-FBFBAP) which is mounted on the return port of the directional coupler.

FMR measurements can be performed in this configuration using the whole bandwidth of the waveguide. This is commonly referred to as terminated waveguide FMR. However, high sensitivity in this case can be achieved using a microwave cavity. The cavity used is a 25.7 mm resonant section of WR-28 waveguide and is coupled to the waveguide track through a small aperture. The sample is mounted in the center of a terminating plate on the other side of the rectangular cavity. Within the WR-28 waveguide band the cavity has four single mode resonances at \( f \simeq 27.23 \) (\( \text{TE}_{103} \)), 31.22 (\( \text{TE}_{104} \)), 35.68 (\( \text{TE}_{105} \)), and 40.45 (\( \text{TE}_{106} \)) GHz.

![Figure 4.2: Example cavity modes for the multimode cavity used for ferromagnetic resonance measurements.](image)

The WR-28 waveguide is specified to support the Ka band (26.5 - 40.0 GHz) of microwave frequencies. The waveguide will propagate higher frequencies, however other propagation modes become possible, including degenerate modes. The 28 in WR-28 refers to the length of the \( H \)-axis (longside of cross section) of the waveguide in hundredths of inches. The typical rectangular waveguide has an aspect ratio of 2:1. The cavity was designed to fit 4 \( \text{TE}_{10p} \) cavity modes in the Ka band where only single mode propagation is possible in the waveguide. In this case with the sample located on a terminating plate opposite the
coupling aperture, \( h_{rf} \) for the sample is known to be along the H-axis (long direction in cross section) of the cavity.

### 4.2.2 Coplanar waveguide

![Schematic diagram of the FMR spectrometer using a coplanar waveguide (CPW) with field modulation (HM) or microwave power amplitude pulse modulation (PM).](image)

Figure 4.3: Schematic of the FMR spectrometer using a coplanar waveguide (CPW) with field modulation (HM) or microwave power amplitude pulse modulation (PM). The coplanar waveguide is mounted in the center of a Varian electromagnet that provides the dc field and two Helmholtz coils (Modulation Coils) that can provide HM. The rf microwave source is an Anritsu MG3696B microwave generator. The rf FMR signal is converted to a low frequency output signal, with the frequency determined by HM or PM, by means of an Agilent 8474E planar-doped barrier diode detector (0.01 to 50 GHz) (Detector 1). The SR830 Lock-in Amplifier is used to provide both the modulation reference signal and to control the Varian electromagnet to step the dc \( H \) field. **CPW-HM:** The 105 Hz modulation signal is provided by the reference out (field mod. ref. out.) of the lock-in amplifier and is amplified by a Kepco power amp that supplies current to the modulation coils. An Aeroflex 8179 dc Block is used to filter out signals that can be induced by modulation. **CPW-PM:** The 10 kHz pulse modulation is supplied by the lock-in amplifier TTL out port to the microwave generator pulse reference port. The result is a on/off chopping of the microwave signal. To enhance sensitivity, a directional coupler is used to take a -13 dB portion of microwave signal that bypasses the coplanar waveguide. This microwave signal is turned into a low frequency signal by a Krytar 704s planar-doped barrier diode detector (0.01 to 50 GHz) (Detector 2). The the signals from Detector 1 and Detector 2 are detected by the lock-in amplifier in A-B mode.

The experimental setup for the coplanar waveguide configuration (CPW) is shown in Figure 4.3. In our case, the system is designed to be compatible with a liquid He flow cryostat to allow FMR measurements between room and liquid He temperatures. The rf signal is brought down to the center of the electromagnet by using semi-rigid coax cable. Heat losses at cryogenic temperatures are decreased by using two segments (input and output) of stainless steel cables inserted between the room temperature (outside dewar)
and cryogenic (inside dewar) areas. However, inside the dewar, Cu low loss non-magnetic semi-rigid coax cables (UT-85C-LL) are connected to the stainless steel cables and fed into a copper box containing a coplanar waveguide located in the middle of the electromagnet. On two opposite sides of the box, the outer coax conductors are soldered to the box feedthroughs while the center conductors are soldered directly to the opposite sides of the center line of the coplanar waveguide. Two wires are soldered between the outer coax conductor and the outer conductors of the coplanar waveguide. This is done on both sides and is the main reason we switch from stainless steel to Cu cables. The rf transmission output is converted to a dc voltage by means of a planar-doped barrier diode detector. The samples is usually mounted in a "flip-chip" configuration where the sample film is placed face down on the coplanar waveguide as shown in Figure 4.4. A thin mica sheet is placed between the sample and the coplanar waveguide to prevent electric current from entering the sample.

The sample filling factor for CPW is much better than that in the microwave cavity, therefore, no cavity was needed for CPW to achieve a similar sensitivity as that obtained for the multimode cavity system. FMR measurements can be carried out using (i) HM and (ii) microwave power amplitude pulse modulation (PM).

(i) The coplanar waveguide with field modulation mode (CPW-HM) is essentially the same as the Cavity-HM mode with the WR-28 waveguides and cavity being replaced with broadband coax cables and the segment of coplanar waveguide, respectively. The other main difference is that microwave transmission is monitored when using the coplanar waveguide.

(ii) In the coplanar waveguide with microwave power amplitude pulse modulation configuration (CPW-PM), the low frequency square wave signal generator TTL output port of the lock-in is connected to the Pulse Trig In port on the Anritsu microwave generator. The microwave output is generally pulsed at 10 kHz. This has the advantage over HM as it moves away from $1/f$ noise, allowing a shorter time constant for the lock-in amplifier than that for HM. In ultrathin films, the FMR signal is usually very small compared to the direct transmission signal. In order to enhance the sensitivity in PM mode, a Marki
C13-0150 microwave directional coupler is used to take a −13 dB portion of the direct signal before the coplanar waveguide and is detected by a second planar-doped barrier detector diode (Krytar 704S). An attenuator is then used to nearly match the amplitude of the coplanar waveguide signal and direct signal off resonance. The additional signal is fed into the B input of the SR830 lock-in amplifier and the data collection is performed in A-B mode. A-B mode allows one to use the lock-in at higher sensitivity in order to avoid being digital noise limited. Another advantage of measuring the transmission of the modulated microwave power, as opposed to the derivative of the transmission, is that one measures the total transmission signal and considering that the FMR signal is only a very small fraction even of the A-B signal allows one to add both phases of the lock-in amplifier in quadrature. Simply in this case one does not have to optimize the phase of the reference signal. This fact alone can greatly speed up the set up of the measurement, especially for very thin samples with wide FMR lines.

4.3 Example FMR data

To compare these different FMR techniques, a GaAs(00 1)|16(2.3nm)Fe|20(4nm)Au sample was prepared by means of MBE, where the integers refer to the layer thickness in atomic layers (AL). For details on sample preparation see Section 2.2.

4.3.1 Comparison of anisotropy field measurements

The free energy (magnetic anisotropies) needs to follow the symmetry of the ultrathin Fe grown on the GaAs(00 1) reconstructed surface. The magnetic anisotropies are as follows:

(i) The four fold in-plane anisotropy energy due to the cubic symmetry of the Fe lattice,

\[ \mathcal{E}_{\text{cubic}} = K_1 \left( \alpha_X^2 \alpha_Y^2 + \alpha_Z^2 \alpha_Y^2 + \alpha_Z^2 \alpha_Y^2 \right) \],

where \( \alpha_{X,Y} \) are the directional cosines of \( \mathbf{M} \) with respect the Fe cube axes. A positive \( K_1 \) results with the easy axes parallel to the cube axes \( \{100\} \).

(ii) The in-plane uniaxial anisotropy energy \( \mathcal{E}_u^\parallel = -K_u^\parallel \sin \phi_M \cos^2 (\theta_u - \theta_M) \) due to the reconstructed GaAs(00 1)-(4 \times 6) surface, where \( \theta_u = -45^\circ \) is parallel to the As dimers at the Fe|GaAs interface. By this definition, a negative \( K_u^\parallel \) corresponds to a hard uniaxial axis.

(iii) The magneto-elastic energy due to the lattice mismatch between the Fe(001) and GaAs(001) and the lack of cubic symmetry at the interfaces results in the perpendicular uniaxial anisotropy energy \( -K_u^\perp \alpha_Z^2 \) [35].

(iv) The demagnetization energy \( 2\pi M_S^2 \alpha_Z^2 \) due to the semi-infinite sheet shape of ultrathin films.
The following FMR data was collected in the in-plane FMR configuration ($\phi H = \phi M = 90^\circ$, see Section 3.3.1) where the resonance condition is,

$$\left(\frac{\omega}{\gamma}\right)^2 = B_{\text{eff}} H_{\text{eff}} |H_{\text{FMR}}|.$$  (4.1)

The above mentioned anisotropies lead to the effective fields

$$B_{\text{eff}} = H_{\text{dc}} \cos (\theta_M - \theta_H) + 4\pi M_{\text{eff}} + \frac{K_1^\parallel}{2M_s} (3 + \cos 4\theta_M) + \frac{K_u^\parallel}{M_s} (1 + \cos (2(\theta_M - \theta_U))).$$  (4.2)

$$H_{\text{eff}} = H_{\text{dc}} \cos (\theta_M - \theta_H) + \frac{2K_1^\parallel}{M_s} \cos 4\theta_M + \frac{2K_u^\parallel}{M_s} \cos (2(\theta_M - \theta_U)).$$  (4.3)

Here $4\pi M_{\text{eff}} = 4\pi M_{s,\text{eff}} - 2K_u^\perp/M_s$, $\theta_M$ is the angle of the dc magnetic moment, and $\theta_H$ is the angle of the applied external field $H_{\text{dc}}$, with the angles being measured from the Fe $[100]$ crystallographic direction. The origins of Equations (4.1) to (4.3) are described in Chapter 3.

Note that in FMR experiments in thin films, one measures the effective anisotropy fields $H_K$ and not the anisotropy parameters $K$ directly. Thus in this sample one measures $H_{K_1} = 2K_1/M_s$ and $H_{K_u^\parallel} = 2K_u^\parallel/M_s$. Note that (iii) and (iv) are both perpendicular uniaxial anisotropies (depend on $\alpha_2^2$), therefore they enter into the rf susceptibility on an equal footing; thus, in the FMR experiment one measures an effective demagnetization field $4\pi M_{\text{eff}}$. Other magnetometry techniques such as superconducting quantum interference devices (SQUID) and vibrating sample magnetometers (VSM) can be used to determine $M_s$. Then from the FMR measured effective fields, one can determine the energy of crystalline anisotropies. For the Fe layer $M_s = 1710$ G and $g = 2.09$ [54, 55], leading to $\gamma = 1.84 \times 10^7$ Hz/Oe.

In order to determine the in-plane anisotropy of the sample, the resonance field $H_{\text{FMR}}$ was measured as a function of angle $\theta_H$ at 35.6802 GHz. Example FMR data for each technique together with fits using (3.59) and an instrumental background (2nd order polynomial) are shown in Figure 4.5 (a-c), the angular dependences of $H_{\text{FMR}}$ are shown in Figure 4.6, and the resulting fitted anisotropy fields are listed in Table 4.1. All three techniques agree very well and are described well by Equations (4.2) and (4.3). Notice that FMR measurements using CPW can result in signals that are far away from measuring the microwave absorption $\chi''$ (common to measurements in microwave cavity). In Figure 4.5 (b-c), the measured FMR signal is very close to $\chi'$ and yet fitting parameters used for FMR
Figure 4.5: FMR measurements of GaAs|16Fe|20Au at $f = 35.6802$ GHz along the hard uniaxial axis [1\bar{1}0] direction are shown for (a) the multimode cavity with field modulation configuration with fit parameters $H_{\text{FMR}} = 7179.5(\pm 0.1)$ Oe, $\Delta H = 51.6(\pm 0.1)$ Oe, $\varphi = 2.8^\circ$, with collection time $t = 240$ s; (b) the coplanar waveguide with field modulation configuration with $H_{\text{FMR}} = 7170.2(\pm 0.1)$ Oe, $\Delta H = 52.1(\pm 0.1)$ Oe, $\varphi = 83.7^\circ$, $t = 704$ s; and (c) the coplanar waveguide with microwave power amplitude pulse modulation (CPW-PM) configuration with $H_{\text{FMR}} = 7165.7(\pm 0.1)$ Oe, $\Delta H = 52.9(\pm 0.1)$ Oe, $\varphi = 88.7^\circ$, $t = 335$ s. The colored dots are the collected data and the corresponding solid lines are the theoretical fit as given by Equation (3.60) for (a) and (b) and Equation (3.59) for (c) and a (first order (a),(b) and second order (c)) polynomial background (dotted black lines).

signals obtained in the microwave cavity (Figure 4.5 (a) and using CPW Figure 4.5 (b-c) are in great agreement, testifying to the fact that using (3.59) is a very reliable procedure, see Table 4.1.

4.3.2 Comparison of damping measurements

In order to compare the different techniques for determining the Gilbert damping parameter $\alpha$, the frequency dependence of the FMR signal was measured along the [1\bar{1}0] direction. The damping was Gilbert like as the FMR linewidth as a function of frequency was well described by

$$\Delta H (f) = \alpha \frac{\omega}{\gamma} + \Delta H (0),$$

(4.4)

where $\omega(= 2\pi f)$ is the microwave angular frequency and $\gamma$ is the absolute value of the gyromagnetic ratio. When $\Delta H (f)$ is linear, $\Delta H (0)$ is caused by long range magnetic inhomogeneities in the internal effective fields in the films [41, 47]. The zero frequency offset $\Delta H (0)$ was modest and $\Delta H (f)$ displayed linear dependence, see Figure 4.7.

The results of using Cavity-HM and CPW-HM were in excellent agreement. CPW-PM proved to be less well suited to measure individual FMR linewidths. This was caused by small drifts in the measured transmission amplitude as a result of the detector diode response to room temperature fluctuations. The HM techniques do not have this issue as the
Figure 4.6: A plot of the ferromagnetic resonance field $H_{\text{FMR}}$ as a function of the in-plane angle $\theta_{H_{dc}}$ for the sample GaAs|16Fe(0 0 1) measured at 35.6802 GHz for ⊗ multimode cavity with field modulation, ⋄ coplanar waveguide with field modulation, and ⊕ coplanar waveguide with pulse frequency modulation.

Table 4.1: GaAs|16Fe|20Au Magnetic Properties

<table>
<thead>
<tr>
<th>Measurement configuration</th>
<th>$4\pi M_{\text{eff}}$</th>
<th>$H_{K}\parallel$</th>
<th>$H_{K}\perp$</th>
<th>$\alpha$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cavity-HM</td>
<td>16.07</td>
<td>313</td>
<td>-399</td>
<td>3.52</td>
</tr>
<tr>
<td>CPW-HM</td>
<td>16.08</td>
<td>313</td>
<td>-393</td>
<td>3.52</td>
</tr>
<tr>
<td>CPW-PM</td>
<td>16.07</td>
<td>315</td>
<td>-384</td>
<td>3.6</td>
</tr>
</tbody>
</table>

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Figure 4.7: A plot of the ferromagnetic resonance linewidth $\Delta H$ as a function of microwave frequency for the sample GaAs$_{16}$Fe(001) measured along the Fe $[1\overline{1}0]$ direction for $\otimes$ multimode cavity with field modulation, $\odot$ coplanar waveguide with field modulation, and $\oplus$ coplanar waveguide with pulse frequency modulation.

Lineshape changes negligibly with these small drifts and thus the derivative is little effected. This obstacle was offset by measuring CPW-PM FMR by sweeping the dc field back and forth and averaging power fluctuations. Furthermore, the broadband nature of the CPW allowed measurements at a number of microwave frequencies and the fitted Gilbert damping parameter $\alpha$ agrees with the field modulation techniques within $\sim 1\%$, see Table 4.1 and Figure 4.7.

4.3.3 Sputter deposited samples on Si wafers

The FMR spectrometer based on coplanar waveguide is well suited for determining the magnetic properties of samples grown by sputter deposition on Si substrates. This system is easier to use than the cavity based systems as it does not involve the tuning of the microwave frequency to the cavity (or cavity to the microwave frequency) and can be fully controlled using a software interface.
The data presented here is meant to illustrate that the dynamic magnetic properties can now be measured accurately and more easily in ultrathin films deposited on Si substrates. The goal of this work is to study spin pumping at the Py|Ta interface and spin diffusion in Ta, where Py = Ni\textsubscript{80}Fe\textsubscript{20} (permalloy). Spin pumping and spin diffusion in Ta is currently a controversial topic. Some work has shown that pumping at the Py|Ta interface doesn’t occur [56], while other work has shown large pumping [57, 58]. Ta is currently very attractive for device applications as it is one of the best converters of spin currents to voltage and vice versa using the inverse spin Hall effect [59] and spin Hall effect [60, 61], respectively. The work at this point is still in progress, but the FMR data shows that the spectrometer is well suited to characterize the magnetization dynamics of these types of samples. Figure 4.8 shows FMR data measured for these sputtered samples. Figure 4.9 (a) shows examples FMR linewidth as a function of frequency. $\Delta H(f)$ displays the linear dependence a Gilbert damping and a very small zero frequency offset.

The samples are single magnetic layer Si|(3 nm)Ta|(3.45 nm)Py|(\(d_{Ta}\))Ta|(3.6 nm)Au and double magnetic layer Si|(3.0 nm)Ta|(3.45 nm)Py|(\(d_{Ta}\))Ta|(2.0 nm) Py|(4.0 nm) Fe|(3.6 nm)Au. The films were deposited at room temperature on Si (1 0 0) substrates by means of RF magnetron sputtering. The (2.0 nm)Py|(4.0 nm)Fe layers act as a single ferromagnet and the Fe layer is used to shift the resonance field away from the (3.45 nm)Py layer, see Figure 4.8 (d); this is due to the fact that \(4\pi M_{eff}\) is much larger in Fe than Py, see Equation (3.40). The magnetic damping of the (3.45 nm)Py layer as a function of the thickness of the Ta overlayer \(d_{Ta}\) can be used to determine the spin mixing conductance at the Py|Ta interface and spin diffusion length in Ta, as described later in Chapter 6. However, the determination of these parameters requires other measurements, which were still in progress at the time of writing. The data in Figure 4.9 (d), however, already shows qualitatively that there is an increase in damping with increasing \(d_{Ta}\) that quickly approaches saturation on the length scale of \(\sim 1\) nm. The increase in damping in the magnetic double layer compared to the magnetic single layer for \(d_{Ta} < 2\) nm shows that in fact there is a spin pumping contribution to the damping.

### 4.4 Magnetic field calibrations

#### External dc magnetic field

The magnetic field is measured by means of a Hall probe mounted to one of the poles of the magnet. The Hall probe reading is calibrated using nuclear magnetic resonance (NMR) of protons in H\textsubscript{2}O.
Figure 4.8: Example FMR data for sputtered samples: (a) 12 GHz measurement of Si|(3 nm)Ta|(3.45 nm)Py|(3.6 nm)Au with fit parameters $H_{\text{FMR}} = 1518.5$ Oe, $\Delta H = 43.4$ Oe, $\varphi = -28.1^\circ$, with collection time $t = 254$ s; (b) 20 GHz measurement of Si|(3 nm)Ta|(3.45 nm)Py|(5.0 nm)Ta|(3.6 nm)Au with $H_{\text{FMR}} = 3602.0$ Oe, $\Delta H = 90.0$ Oe, $\varphi = -2.7^\circ$, $t = 506$ s; and (c) 30 GHz measurement of Si|(3 nm)Ta|(3.45 nm)Py|(1.0 nm)Ta|(2.0 nm)Py|(4.0 nm)Fe|(3.6 nm)Au with $H_{\text{FMR}} = 6558.5$ Oe, $\Delta H = 139.2$ Oe, $\varphi = 111.7^\circ$, $t = 768$ s. The colored dots are the collected data and the corresponding solid lines are the theoretical fit as given by Equation (3.60) and a first order polynomial background (dotted black lines). (d) 6.5 GHz measurement of Si|(3 nm)Ta|(3.45 nm)Py|(2.0 nm)Ta|(2.0 nm)Py|(4.0 nm)Fe|(3.6 nm)Au to illustrate the separation in resonance field for the (3.45 nm)Py layer ($H_{\text{FMR}} = 464.5$ Oe) and the (2.0 nm)Py|(4.0 nm)Fe ($H_{\text{FMR}} = 228.6$ Oe).
Figure 4.9: (a) FMR linewidth as a function of frequency $\Delta H (f)$ for samples SL: Si|(3 nm)Ta|(3.45 nm)Py|(d$_{Ta}$)Ta|(3.6 nm)Au with $d_{Ta} = 0$ nm (●), 0.6 nm (⊗), 5.0 nm (⊙), and DL: Si|(3.0 nm)Ta|(3.45 nm)Py|(d$_{Ta}$)Ta|(2.0 nm Py)|(4.0 nm Fe)|(3.6 nm)Au for $d_{Ta} = 0.6$ nm (⊕). (b) $\alpha$ as a function of $d_{Ta}$ for SL (●) and DL (■).

The control of the magnetic field by the computer also needs to be calibrated for accurate control of measurements. The field can be externally controlled by an external voltage supply, in this case the analog output of the lock-in amplifier. The magnetic power supply has controls for field offset and scan range. The scan range sets the overall field step resolution, as the SR-850 lock-in used in these experiments allows voltage output of $\pm 10$ V with a resolution of 1 mV; therefore, one can use up to 20,000 field steps in the field bandwidth. The calibrated Hall probe measured field is always stored by the FMR software.
Chapter 5

Magnetic anisotropy and spin dynamics at the Fe|GaAs(001) – p (4 × 6) interface

In this chapter, the static and dynamic properties of GaAs(001) Fe Au as a function of Fe thickness will be presented. Specifically the p (4 × 6) reconstructed surface of GaAs will be used as a growth template. It will be shown that ultrathin ferromagnetic films display properties that deviate from the bulk, allowing the tuning of specific magnetic properties simply by varying the ferromagnetic layer thickness. The understanding and tunability of the magnetic properties of ultrathin Fe deposited on GaAs can be of great importance in the design of the experiments such as those presented in Chapter 7.

In the field of spintronics, GaAs(001) Fe structures have played a very important role [44, 62, 63]. One of the reasons is that the two lattices are well matched allowing the growth of high quality ultrathin Fe films using molecular beam epitaxy, see Section 2.2. Additionally the two materials on their own have desirable qualities. Fe has the desirable ferromagnetic characteristics of large Curie temperature, $M_s$, and spin polarization, as well as being a very common and affordable material. GaAs is one of the most studied III-V semiconductor compounds and is found in many physical devices such as microwave integrated circuits, light emitting diodes, and solar cells; therefore, high quality substrates with well know characteristics are readily available.

Extensive studies of the magnetic properties of Fe films grown on GaAs(001) templates have been carried out by a number of groups, see the review article by Wastlbauer and Bland [25]. Conflicting results have been found particularly concerning the inplane uniaxial anisotropy. Magnetic disorder at the Fe|GaAs(001) interface can also be expected to affect spin dynamics. So far, little is known about the interface spin dynamics in ferromagnetic-
metal/semiconductor structures. The purpose of this work was to extend the study of magnetic anisotropies and spin dynamics in Fe|GaAs(0 0 1) structures using Fe films in the thickness range from 5 to 90 atomic layers where the interface and bulk contributions to the magnetic anisotropies and spin dynamics can be demonstrated.

5.1 Sample preparation

Single crystal GaAs(0 0 1)|dFe|20Au samples were prepared by means of molecular beam epitaxy (MBE), where the integers refer to the layer thickness in atomic layers (AL). The film thickness, dFe, used were 5, 6, 7, 10, 12, 14, 16, 20, 25, 30, 45, and 90 AL (1 AL Fe = 0.1433 nm). The 20Au layer was used to protect the Fe film from ambient conditions.

Commercially available, epi-ready, and semi-insulating GaAs(0 0 1) substrates were cleaned for 20 minutes in an H· beam at 400° C. Generally, after 20 min of H· treatment very little C and O contamination is left on the GaAs surface, as determined by means of Auger electron spectroscopy (AES). Longer H· treatments times were found to fully remove the C and O, but were prone to damaging the GaAs surface. The H· treatment was followed by 3+ hr of 650 V Ar+ sputter etching at room temperature. After the Ar+ sputtering process, the film surface was checked for the complete removal of C and O contamination by means of AES. The substrates were then annealed to 585° C resulting in a well defined 4x6 surface reconstruction; annealing was monitored by means of reflection high energy electron diffraction (RHEED) along the 110 direction.

The Fe and Au films were deposited at temperatures between 40 – 60° C from thermal sources. The base pressure in the UHV growth chamber was in the low to mid 10⁻¹¹ Torr. The growth pressure was < 3 × 10⁻¹⁰ Torr. Both layers were grown at deposition rates of ≈ 1 AL/min. The film thicknesses and rates were monitored by means of oscillations in the intensity of the specular spot at an anti-Bragg RHEED reflection in conjunction with a quartz crystal thickness monitor, see Section 2.2 for more details.

5.2 Ferromagnetic resonance

Ferromagnetic resonance measurements were performed in single mode cavities at 9.1, 23.99, and 36 GHz and in the terminated waveguide configuration at 72.1 GHz. The microwave power sources were Gunn diodes for the 23.99 and 36 GHz measurements and reflex Klystron power supplies for the 9.1 and 72.1 GHz measurements. The measurements were performed as described in section 4.2.1 and the FMR spectra were fit using the methods described in Section 3.4.
The FMR spectra were well represented by a single FMR line showing a high level of sample homogeneity, see Figure 5.1. Varying both the angle of the external magnetic field $H_{dc}$ and the microwave frequency allowed the characterization of both the static and dynamic magnetic properties of the samples.

5.3 GaAs(001)/Fe magnetic anisotropy

Although the main focus of this thesis is to explore dynamic properties of spin pumping and spin transport, it is very important to explore the parameter space of the static magnetic properties; this allows one to better design the experiments in which to search for new physics and to create usable technological devices. One can design heterostructures with two ferromagnets (even of the same material) that will undergo ferromagnetic resonance at different external fields (frequencies) for a given frequency (external field). This is done by tuning the internal fields in the material using the interface components of the anisotropies. This is a very useful technique for both fundamental research as well as device technologies.

The effective inplane fourfold (cubic) anisotropy field $H_{1||}^{\text{eff}} = 2K_{1,\text{eff}}^{||}/M_s$, inplane uniaxial anisotropy field $H_{u||}^{\text{eff}} = 2K_{u,\text{eff}}^{||}/M_s$, and the effective demagnetizing field $4\pi M_{\text{eff}}$ were determined from the inplane FMR measurements at 72.1 GHz at room temperature, see Figure 5.2. The anisotropy fields are determined from the inplane angular dependence of $H_{\text{FMR}}$, where the resonance condition is

$$\left(\frac{\omega}{\gamma}\right)^2 = B_{\text{eff}} H_{\text{eff}} |_{H_{dc}=H_{\text{FMR}}},$$

with

$$B_{\text{eff}} = H_{dc} \cos (\theta_M - \theta_H) + 4\pi M_{\text{eff}} + \frac{K_{1,\text{eff}}^{||}}{2M_s} (3 + \cos 4\theta_M) + \frac{K_{u,\text{eff}}^{||}}{M_s} (1 + \cos (2(\theta_M - \theta_U)))$$

$$H_{\text{eff}} = H_{dc} \cos (\theta_M - \theta_H) + \frac{2K_{1,\text{eff}}^{||}}{M_s} \cos 4\theta_M + \frac{2K_{u,\text{eff}}^{||}}{M_s} \cos (2(\theta_M - \theta_U)).$$

Figure 5.3 shows $H_{1||}^{\text{eff}}$, $H_{u||}^{\text{eff}}$, and $4\pi M_{\text{eff}}$ as a function of the inverse Fe film thickness $1/d_{\text{Fe}}$. As the surface energy is shared evenly across the film thickness due to exchange for ultrathin ferromagnetic films, one expects the surface anistropies to have a $1/d_{\text{Fe}}$ dependence. All of the Fe films had their Curie temperature $T_C$ well above room temperature. In similar studies, Bensch et al. [64] have shown that $T_C > 500$ K even for 5 AL Fe on the $p(4 \times 6)$ reconstructed GaAs surface.
Figure 5.1: Example FMR data for the samples GaAs|$d_{Fe}$Fe|20Au. 72.1 GHz inplane FMR measurements are shown for $d_{Fe} = (a) 6$, (b) 16, (c) 30 AL. The measurements shown were taken along the Fe (GaAS) [110] direction, corresponding to the uniaxial hard axis. 36 GHz perpendicular FMR measurements are shown for (d) $d_{Fe} = 12$. 
Figure 5.2: $H_{\text{FMR}}$ as a function of inplane angle $\theta_H$ measured at 72.1 GHz used for determining the anisotropy effective fields. The data sets are for the samples GaAs|$d_{Fe}$|20Au with $d_{Fe}$ = ■ 5, ◊ 6, ⊙ 7, ○ 16, ⊕ 30, and ● 45 AL.
Figure 5.3: The inplane fourfold (cubic) \( H_\parallel = 2K_{1,\text{eff}} / M_s \) (\( \triangledown \)) and uniaxial \( H_u^\parallel = 2K_u^\parallel / M_s \) (\( \bigcirc \)) effective anisotropy fields and the effective demagnetization field \( 4\pi M_{\text{eff}} \) (\( \square \)) as a function of the inverse Fe film thickness \( 1/d_{\text{Fe}} \). The solid lines are linear fits to the data, where the y-axis intercept represents the bulk contribution and the slope represents the interface contribution. The inplane linear uniaxial behavior is fit between \( d_{\text{Fe}} = 6 - 20 \) AL. The grey dashed line represents fitting the inplane uniaxial behavior as a purely interface effect. The error bars are smaller than the points shown.
Both the inplane fourfold anisotropy field and $4\pi M_{\text{eff}}$ can be explained as a combination of bulk and interface contributions,

$$\frac{2K_{\parallel,\text{eff}}}{M_s} = \frac{2K_1^\parallel}{M_s} + \frac{2K_{\parallel,s}}{M_sd_{\text{Fe}}}$$  \hfill (5.4)

$$4\pi M_{\text{eff}} = 4\pi M_{s,\text{eff}} - \frac{2K_{\perp,u,s}}{M_sd_{\text{Fe}}}.$$  \hfill (5.5)

Here $2K_{1,s}/M_sd_{\text{Fe}}$ is the surface inplane fourfold anisotropy field associated with the inplane fourfold anisotropy energy $K_{1,s}^\parallel$, $M_{s,\text{eff}}$ is the effective saturation magnetization, and $K_{u,s}/M_sd_{\text{Fe}}$ is the surface perpendicular anisotropy field associated with the surface perpendicular anisotropy energy $K_{u,s}$. The first terms on the RHS of both Equations (5.4) and (5.5) represent the bulk contributions and the second terms represent the surface (interface) contribution.

The bulk contribution to the four fold anisotropy energy, $K_1^\parallel = 4.3 \times (0.1) \times 10^5$ erg/cm$^3$, is in good agreement with the values of the bulk cubic anisotropy listed by Bozorth [65]. The surface contribution to the four fold anisotropy energy is $K_{1,s}^\parallel = -0.033 \times (0.001)$ erg/cm$^2$, which is consistent with magneto-optic Kerr effect (MOKE) studies of 4-14 AL of Fe on GaAs by Moosbühler et al. [66] ($K_{1,s}^\parallel = -0.032 \times (0.002)$ erg/cm$^2$). The bulk contribution to the demagnetization field, $4\pi M_{s,\text{eff}} = 20.8 \times (0.1)$ kOe, is about 0.7 kOe less than the saturation induction $4\pi M_s = 21.5$ kOe [40]. For the extended ultrathin film, one would expect the demagnetization factor to be $D = 1$, resulting in a demagnetization field equal to the saturation induction. The surface contribution to the perpendicular uniaxial anisotropy is $K_{u,s}^\perp = 1.01 \times (0.02)$ erg/cm$^2$ is in reasonable agreement with previous studies by Urban et al. [67] ($K_{u,s}^\perp = 0.9$ erg/cm$^2$).

The difference in $4\pi M_{\text{eff}}$ can be explained by the axial magneto-elastic contribution to the anisotropy caused by strain due to the lattice mismatch in the Fe on the GaAs(001) template. The lattice strain leads to a tetragonal distortion of Fe grown on GaAs that modifies the effective demagnetization field [35, 68, 69],

$$4\pi M_{\text{eff}} = 4\pi M_s - \frac{2K_{u,s}^\perp}{M_sd_{\text{Fe}}} + \frac{2B_1(e_\perp - e_\parallel)}{M_s},$$  \hfill (5.6)

where $B_1$ is the magneto-elastic coupling coefficient, $e_\perp$ and $e_\parallel$ are the strains perpendicular and parallel to the substrate. The inplane lattice strains are given by the inplane lattice mismatch, giving $e_\parallel = -0.0139$. Allowing for the stress to be zero along the film normal
results in the vertical strain relation

\[ e_\perp = -\frac{2C_{12}}{C_{11}} e_\parallel, \]  

(5.7)

where \( C_{11} \) and \( C_{12} \) are the elastic moduli. Sander [70] lists the values for bcc Fe of \( C_{11} = 2.29 \times 10^{12} \) erg/cm\(^3\) and \( C_{12} = 1.34 \times 10^{12} \) erg/cm\(^3\). The upper limit on the magnitude of the magneto-elastic anisotropy field can be estimated by using \( B_1 = -3.43 \times 10^7 \) erg/cm\(^3\) from bulk Fe [36], assuming the strain is through the whole lattice. Using these values for \( C_{11}, \) \( C_{12}, \) and \( B_1 \) and Equations (5.6) and (5.7), the effective magneto-elastic field perpendicular to the film surface can be upwards of 1.2 kOe. Partial relaxation of strain would reduce the magnitude of the magnetoelastic field. The magnetoelastic field in this case is opposite the demagnetization field and thus serves to reduce \( 4\pi M_{s,\text{eff}} \). Furthermore, the \( B_1 \) term has been shown to be fairly complex as it can change with strain [36]. However, the important quantity for devices and FMR experimental design is \( 4\pi M_{\text{eff}} \), which can be tuned by varying the Fe thickness as the \( K_{\perp,\text{Fe}} \) is the dominant term. In fact, the change in \( 4\pi M_{\text{eff}} \) from \( d_{\text{Fe}} = 5 \) to \( d_{\text{Fe}} = 90 \) represents a change of \( 4\pi M_{\text{eff}} \) of \( \approx 15 \) kOe.

The \( d_{\text{Fe}} \) dependence of the inplane uniaxial anisotropy field \( H_{\alpha}^{\parallel} = 2K_{\text{u,eff}}^{\parallel}/M_s \) is more complicated than \( H_1^{\parallel} \) and \( 4\pi M_{\text{eff}} \). For \( d_{\text{Fe}} = 6-20 \) AL, the thickness dependence takes on the form of

\[ \frac{2K_{\text{u,eff}}^{\parallel}}{M_s} = \frac{2K_{\alpha}^{\parallel}}{M_s} + \frac{2K_{\text{u,s}}^{\parallel}}{M_s d_{\text{Fe}}}. \]  

(5.8)

The bulk contribution to the uniaxial anisotropy energy \( K_{\alpha}^{\parallel} = 0.5 (\pm 0.5) \times 10^5 \) erg/cm\(^3\) is negligible compared to the other internal energies. The interface contribution to the uniaxial anisotropy energy was found to be \( K_{\text{u,s}}^{\parallel} = -0.133 (\pm 0.006) \) erg/cm\(^2\). However, one would expect the inplane uniaxial anisotropy to be a purely interface effect, supported by the negligible \( K_{\alpha}^{\parallel} \) term in the previous fit. Fitting the data using only the interface term yields \( K_{\text{u,s}}^{\parallel} = -0.127 (\pm 0.003) \) erg/cm\(^2\), see gray dashed line in Figure 5.3. The values of \( K_{\text{u,s}}^{\parallel} \) are in good agreement with the MOKE studies by Moosbühler et al. [66], \( K_{\text{u,s}}^{\parallel} = -0.130 (\pm 0.001) \) erg/cm\(^2\), and the FMR studies in ref. [69], \( K_{\text{u,s}}^{\parallel} = -0.10 (\pm 0.01) \) erg/cm\(^2\).

For \( d_{\text{Fe}} > 20 \) AL, the magnitude of \( K_{\text{u,eff}}^{\parallel} \) decreased faster than was expected when modeled by a simple interface term. \( K_{\text{u,eff}}^{\parallel} \approx 0 \) for the 45 AL Fe film; while for the 90 AL Fe film, \( K_{\text{u,eff}}^{\parallel} = 5.9 (\pm 0.4) \times 10^4 \) erg/cm\(^3\), indicating that \( K_{\text{u,eff}}^{\parallel} \) changed sign in its approach to zero, although it’s magnitude at this point is very small. Kardasz et al. [69, 71] have shown that the origin of \( K_{\text{u,s}}^{\parallel} \) is not only due to the surface chemistry with the dangling bonds of As at the GaAs\(\mid\)Fe interface. In fact, Gordon and Crozier [72] have shown by polarization dependent X-ray absorption fine-structure spectroscopy (XAFS) studies on \( p (4 \times 6) \) - GaAs\(\mid 001\)\|2Fe samples that the Fe nearest neighbor distance along the \( [\overline{1}0]\)
direction is longer than the distance along the [110] directions; this difference in spacing is attributed to an interface lattice shear consistent with an anisotropic inplane strain [73]. Furthermore, Gordon and Crozier [72] showed that the shear was rapidly decreasing away from the interface and was not observable in the GaAs|5Fe sample, suggesting that the lattice shear is confined to the GaAs|Fe interface and that it is not a bulk property. The symmetry at the GaAs interface is uniaxial due to the As dimers and the lattice shear is a consequence of this symmetry. The magnetic anisotropy at the interface is associated with the role of spin-orbit interaction at the GaAs|Fe interface. The magneto-elastic anisotropy energy for an inplane lattice shear is given by [74]

\[ E_{\text{shear}} = \frac{B_2 \epsilon_6}{2} \sin(2\theta) \]  

(5.9)

where \( B_2 \) is the magneto-elastic coupling coefficient relevant to the uniaxial shear [36], \( \epsilon_6 \) is the magnitude of the shear component, and \( \theta \) is the angle between the magnetization and the [1 0 0] direction. The approximately 1% shear measured by Gordon and Crozier [72] corresponds to \( \epsilon_6 = -0.013 \) [74]. Using \( B_2 = 7.83 \times 10^7 \text{ erg/cm}^3 \) for bulk bcc Fe [70] in Equation (5.9), and multiplying by the 2 AL Fe thickness \( (2.865 \times 10^{-8} \text{ cm}) \) to convert to an interface anisotropy, one can estimate \( E_{\text{shear, s}} = -0.014 \text{ erg/cm}^2 \), which gives the right sign to have a uniaxial hard axis along the [1 1 0] direction. However, the magnitude of the anisotropy using the bulk value of \( B_2 \) as an estimate is an order of magnitude smaller than the observed uniaxial anisotropy \( K_{\parallel, u} = -0.127 (\pm 0.003) \text{ erg/cm}^2 \). To explain the observed inplane uniaxial anisotropy would require an enhanced \( B_2 \) term at the interface or a larger shear than measured [74]. The onset of misfit dislocations relaxes the inplane strain at the GaAs|Fe interface and could be expected to alter the surface chemistry leading to the lattice shear in a complex manner, reducing the magnitude of \( K_{\parallel, \text{eff}} \) with increasing \( d_{\text{Fe}} \) faster than a simple interface term. Furthermore, conversion electron Mössbauer spectroscopy studies [69, 71] show that the Fe|GaAs(001) interface is complex and involves a degree of intermixing that changes the Fe nearest neighbor environment.

Both the large perpendicular uniaxial surface anisotropy and inplane uniaxial surface anisotropy allow one to create multilayer structures involving two ferromagnetic Fe layers separated by at least one nonmagnetic normal metal layer (NM), where the two layers can have largely different resonance fields (frequencies). In fact due the large inplane uniaxial anisotropy, in a structure of the form GaAs|Fe$_1$|NM|Fe$_2$, the thickness of the Fe$_1$ and Fe$_2$ can be tuned such that along certain crystallographic axes the two Fe layers will be on resonance at the same field (frequency) while along other crystallographic axes they are off resonance from each other. This feature was exploited to provide unambiguous experimental evidence for the spin pumping effect [75].
5.4 GaAs(001)/Fe spin dynamics

The spin dynamics of Fe on GaAs is a very important topic in this thesis. The thickness dependence of the Gilbert damping of ultrathin Fe films deposited on GaAs(001)–p (4 × 6) surface will be explored in this section. The Fe film thickness, $d_{Fe} = 5–45$ AL, indeed show a range where both the interface and the bulk contribution to the spin dynamics can be demonstrated. The spin dynamics at the GaAs(001)/Fe interface explored in this section will be of fundamental importance in some of the following chapters.

Inplane FMR

The damping for all films was measured in the inplane FMR configuration, see Section 3.3.1. For films $d_{Fe} \leq 20$ AL, the magnetization dynamics in the system followed the Gilbert like damping described in Section 3.2.1. The zero frequency offset $\Delta H (0)$ was small and $\Delta H (f)$ displayed linear dependence, see Figure 5.4. The FMR linewidth can simply be described by

$$\Delta H (f) = \alpha \frac{\omega}{\gamma} + \Delta H (0), \quad (5.10)$$

where $\omega (= 2\pi f)$ is the microwave angular frequency and $\gamma$ is the absolute value of the gyromagnetic ratio. When $\Delta H (f)$ is linear, $\Delta H (0)$ is caused by long range magnetic inhomogeneities in the internal effective fields in the films [41, 47].

For thicker films $d_{Fe} \geq 25$ AL, the films displayed a large $\Delta H (0)$ and a nonlinear dependence of $\Delta H (f)$ at lower frequencies, see Figure 5.4. The nonlinear dependence of $\Delta H (f)$ is consistent with the onset of two magnon scattering due to short range magnetic inhomogeneities. Furthermore, 9.1 GHz perpendicular FMR measurements showed a much smaller linewidth in these films. The two magnon scattering process is confined inplane and is not present in the perpendicular FMR configuration [44]. If the extrinsic contribution to the linewidth was cause by long range inhomogeneities, the linewidth in the perpendicular FMR configuration would be larger than in the inplane configuration, see Figure 5.6. This onset of two magnon scattering has been shown to be caused by the onset of misfit dislocations of thicker Fe films on GaAs(001) due to the lattice mismatch between Fe and GaAs [46].

The contribution of two magnon scattering to $\Delta H (f)$ saturates above a material dependent frequency; for Fe, the two magnon scattering contribution has been shown to be saturated before 36 GHz [44]. For this reason, the thicker films $d_{Fe} > 25$ AL, were determined using Equation (5.10) for the 36 and 72.1 GHz measurements.

The $d_{Fe}$ dependence of the Gilbert damping parameter $\alpha$ in the GaAs(001)/Fe|20Au system are shown in Figure 5.5. As one might expect, $\alpha$ increases rapidly with decreasing $d_{Fe}$, indicating a large interface contribution to the Gilbert damping. The thinnest Fe film
Figure 5.4: The inplane FMR linewidth $\Delta H (f)$ (half width at half maximum) as a function of microwave frequency for GaAs(001)$|d_{Fe}|20Au$ samples with $d_{Fe} = 7$ (○), 10 (▼), 16 (▲), and 45 (□) AL. The $d_{Fe} = 7, 10,$ and 16 AL samples have a linear dependence with modest zero frequency offset $\Delta H (0)$. The $d_{Fe} = 45$ AL sample has a deviation from linear dependence at lower frequencies, consistent with the onset of two magnon scattering due to misfit dislocations. The 9.1 GHz perpendicular measurement for the sample $d_{Fe} = 45$ (⊗) is also shown. Note that this has a much smaller linewidth than the inplane measurements, further supporting the existence of two magnon scattering.
Figure 5.5: The dependence of the Gilbert damping parameter $\alpha$ on the inverse Fe film thickness $1/d_{Fe}$ in the GaAs(001)$|d_{Fe}|20$Au samples with (○) inplane damping measurements made along the Fe [110] direction and (□) perpendicular to the film surface.
Fe = 5 AL) exhibits ~300% increase in damping over the thickest Fe film (d_{Fe} = 45 AL). For d_{Fe} ≥ 15 AL, α decreases slowly with increasing d_{Fe}.

The damping dependence on the Fe film thickness, shown in Figure 5.5, can be described by

$$\alpha = \alpha_{\text{bulk}} + \frac{\alpha_s}{d_{Fe}},$$  (5.11)

where \(\alpha_{\text{bulk}}\) is the bulk Gilbert damping in Fe, \(\alpha_s\) is the interface contribution to the damping. The resulting fit parameters are \(\alpha_{\text{bulk}} = 2.4 (\pm 0.3) \times 10^{-3}\), \(\alpha_s = 19 (\pm 1) \times 10^{-3}\) AL. The resulting \(\alpha_{\text{bulk}}\) is in agreement with \(\alpha_{\text{bulk}} = 2.2 \times 10^{-3}\) measured by Bhagat [76] in Fe whiskers and is larger than \(\alpha_{\text{bulk}} = 1.8 \times 10^{-3}\) measured in by Frait and Fraitová [77]. Recent Mössbauer studies [69, 71] using MBE and pulsed laser deposition of Fe on GaAs(001) templates have shown that the magnetic state of interface Fe atoms consists of a number of Fe, Ga, and As nearest neighbor configurations. The magnetic disorder at the Fe/GaAs(001) interface can be expected to introduce disorder in the spin-orbit interaction and change the interface electron band structure, which plays an essential role in spin dynamics, and most likely results in the interface contribution to the Gilbert damping.

### Perpendicular FMR

Damping was also measured in the perpendicular configuration, Section 3.3.2, for the samples \(d_{Fe} = 5, 6, 7, 10, 12\) AL. Perpendicular FMR measurements were performed at 9.1, 23.99, and 36 GHz. For thicknesses greater than 10 AL, the required resonance fields at the available microwave frequencies were higher the fields than reachable by the electromagnet.

The FMR linewidth as a function of microwave frequency is plotted in Figure 5.6. The dependence is also described by Equation (5.10). The zero frequency line broadening is larger in the perpendicular configuration, which is consistent with long range magnetic anisotropy inhomogeneities. The dependence of \(\alpha\) on \(1/d_{Fe}\) for the perpendicular configuration is plotted in Figure 5.5. The dependence shows a qualitative agreement with the inplane \(\alpha\), there is a clear interface contribution. Fitting the perpendicular \(\alpha\) data using Equation (5.11) results in \(\alpha_{\text{bulk}} = 1.7 (\pm 0.5) \times 10^{-3}\) and \(\alpha_s = 18 (\pm 3) \times 10^{-3}\) AL.

### 5.5 Summary

For Fe deposited by means of molecular beam epitaxy on GaAs(001), it has been shown that the inplane fourfold anisotropy \(K_{1eff}^{||}\) and perpendicular uniaxial anisotropy \(4\pi M_{eff}\) are well described by contributions from both bulk and interface terms for Fe film thickness ranging from \(d_{Fe} = 5 - 90\) AL. The bulk contributions were shown to be in good agreement with the parameters for bulk Fe. For \(d_{Fe} ≤ 25\) AL, the inplane uniaxial anisotropy \(K_{u,eff}^{||}\)
Figure 5.6: (a) The perpendicular FMR linewidth $\Delta H (f)$ (half width at half maximum) as a function of microwave frequency for GaAs(001)$|d_{Fe}|20Au$ samples with $d_{Fe} = 5 (\bigcirc)$, 6 ($\triangle$), 7 ($\square$), 10 ($\triangledown$), and 12 ($\otimes$) AL.
can be described well by only an interface term. For $d_{Fe} > 25$ AL, $K_{u,eff}^\parallel$ relaxes faster than $1/d_{Fe}$ as expected for an interfacial term alone. This is consistent with the interface chemistry changing by the onset of misfit dislocations at the Fe|GaAs interface. Thick Fe films grown on GaAs(001) have no significant inplane uniaxial anisotropy.

The damping measurements in both the inplane and perpendicular FMR configurations have shown that the GaAs|Fe interface leads to a significant contribution to the Gilbert damping. The fit bulk damping parameter $\alpha_{bulk} = 2.4 (\pm 0.2) \times 10^{-3}$ from inplane measurements and $\alpha_{bulk} = 1.7 (\pm 0.5) \times 10^{-3}$ from perpendicular measurements is in agreement with the damping parameter measured in bulk Fe $\alpha = 2.2 \times 10^{-3}$ [76]. For Fe films with $d_{Fe} \leq 25$ AL, one can avoid large extrinsic contributions to the damping.
Chapter 6

Spin pumping and spin transport theories

In this chapter, the concepts of spin pumping, spin diffusion, and spin decoherence will be introduced which are essential for understanding the experimental results later in this thesis. Spin pumping acts as the source of pure spin currents, where the net flow of spin is accompanied by zero net charge flow. The basic principles of spin pumping from both metallic and insulating ferromagnets will be described for the small FMR precession angle (linear regime). For more details, see the original theory papers by Tserkovnyak et al. [78] and Šimánek and Heinrich [79]. Spin diffusion theory is the standard theory used to describe the propagation of pure spin currents in metallic systems [2, 9, 80–82]. As will be shown, theory applies when the spin flip scattering rate is smaller than the electron momentum scattering rate, as is the case for most typical nonmagnetic metals. However, there are certain nonmagnetic metals where the spin flip scattering rate is about the same as or shorter than the electron momentum scattering rate, in which case a diffusive spin transport description is invalid. In this situation, the spin current relaxation is happening on length scales less than or equal to the mean free path of the electron transport, i.e. the spin transport is in a ballistic regime. Under this condition the spin transport is more appropriately described by spin decoherence theory.

6.1 Spin pumping theory

Ferromagnetic resonance experiments on ultrathin ferromagnetic films have shown that the Gilbert damping parameter $\alpha$ can depend heavily on the substrate and capping layer materials as well as their thickness [56, 67, 83–86]. In these studies, a decrease in ferromagnetic film thickness was accompanied by an increase in damping. In 2002, Tserkovnyak et al.
[78] introduced a theory to quantitatively explain the increase in damping. The underlying concept is that the damping occurs when a spin current leaks into a normal metal (NM) in contact with the ferromagnet (FM). It can be seen as the reciprocal process of the spin transfer torque, where a spin current can exert a finite torque on the magnetic order parameter leading to magnetization dynamics, see [87–89]. In spin pumping, the moving magnetization vector loses torque by emitting a spin current.

The ferromagnetic damping is the consequence of the adiabatic pumping of spins into adjacent normal metals by the precessing ferromagnet. The transfer of spin is governed by the reflection and transmission parameters of electrons impinging on the FM|NM interface. This is analogous to interlayer exchange coupling. In fact, Šimánek and Heinrich [79] showed that the spin pumping mechanism can be explained by the time retarded response to a dynamic interlayer exchange coupling.

![Figure 6.1: Ferromagnetic film (FM) sandwiched between two normal metal (NM) reservoirs. FM and both NMs are in thermal equilibrium. The reflection and transmission parameters $r$ and $t'$ shown govern the spin pumping into the right NM reservoir.](image)

A schematic of a NM|FM|NM system is shown in Figure 6.1. Coherent motion of the magnetization, as in the FMR mode, leads to the emitted spin current into each NM [78]

$$I_{sp} = \frac{\hbar}{4\pi} \left( \text{Re} [A_{\uparrow\downarrow}] \left[ n \times \frac{\partial n}{\partial t} \right] + \text{Im} [A_{\uparrow\downarrow}] \frac{\partial n}{\partial t} \right), \quad (6.1)$$

per unit area of the FM|NM interface, where for the right NM

$$\text{Re} [A_{\uparrow\downarrow}] = \frac{1}{2} \sum_{mn} \left( |r_{\uparrow,mn} - r_{\downarrow,mn}|^2 + |t'_{\uparrow,mn} - t'_{\downarrow,mn}|^2 \right) \quad (6.2)$$

and

$$\text{Im} [A_{\uparrow\downarrow}] = \text{Im} \left[ \sum_{mn} \left( r_{\uparrow,mn} (r_{\downarrow,mn})^* + t'_{\uparrow,mn} (t'_{\downarrow,mn})^* \right) \right]. \quad (6.3)$$

Here $r_{\uparrow(\downarrow),mn}$ is the complex spin majority (minority) reflection parameter of a NM electron impinging on the FM|NM interface and $t'_{\uparrow(\downarrow),mn}$ is the complex spin majority (minority)
transmission parameter for an electron crossing the ferromagnetic from an electron crossing
the FM (from right NM to left NM), where \(m\) and \(n\) label the incoming and outgoing
transverse modes at the Fermi energy in NM. The reflection parameters can be expressed
as \(r_\uparrow = |r_\uparrow|e^{i\theta_\uparrow}\) and \(r_\downarrow = |r_\downarrow|e^{i\theta_\downarrow}\), where \(\theta_\uparrow\) and \(\theta_\downarrow\) are the phases of the reflected majority
and minority spins. Similar expressions can be made for the transmission parameters.

Equations (6.2) and (6.3) can be expressed as [90]

\[
A_{\uparrow\downarrow} = \text{Re} [A_{\uparrow\downarrow}] + i \text{Im} [A_{\uparrow\downarrow}] = \langle g_{\uparrow\downarrow}^r - g_{\uparrow\downarrow}^t, \quad (6.4)
\]

where

\[
g_{\uparrow\downarrow}^r = \sum_{mn} (\delta_{mn} - r_{\uparrow, mn} (r_{\downarrow, mn})^*) \tag{6.5}
\]

and

\[
g_{\uparrow\downarrow}^t = \sum_{mn} (t'_{\uparrow, mn} (t'_{\downarrow, mn})^*). \tag{6.6}
\]

In the case of a metallic FM that is thicker than the transverse spin coherence length,
the terms \(t'_{\uparrow, mn} (t'_{\downarrow, mn})^*\) in Equation (6.5) are 0 and \(g_{\uparrow\downarrow}^t\) vanishes. The transverse spin
decoherence length for ferromagnets is on the order of a few atomic layers; ferromagnetic
layers thicker than a few nm will act as ideal spin sinks for transverse spin currents. The spin
pumping through the right FM\(\mid\)NM interface is then given by the interfacial spin mixing
conductance \(A_{\uparrow\downarrow} = g_{\uparrow\downarrow}^r\) and does not depend on the thickness of the FM [80]. The spin
pumping into the left normal metal is symmetric and is given by letting \(r \to r'\) and \(t' \to t\).

### 6.1.1 Spin pumping at the FM\(\mid\)NM interface

In the case where the FM is metallic and is thicker than the transverse spin decoherence
length, it has been shown that the real part of the spin mixing conductance dominates
the spin momentum transfer. Zwierzycki et al. [90] pointed out that the cross terms
\(r_{\uparrow, mn} (r_{\downarrow, mn})^*\) in Equation (6.5) for the metallic FM\(\mid\)NM interface typically have a negligible
magnitude owing to the fact that majority and minority reflection coefficients have indepen-
dently varying phases that lead to phase cancellation when summed up over all impinging
electrons at the FM\(\mid\)NM interface[90]. Equation (6.5) then reduces to counting the number
of single spin conduction channels at the FM\(\mid\)NM interface. For the spherical Fermi surface
this leads to

\[
g_{\uparrow\downarrow} = \text{Re} [A_{\uparrow\downarrow}] = \text{Re} \left[ g_{\uparrow\downarrow}^r \right] \approx \frac{k_F^2}{4\pi}, \tag{6.7}
\]

where \(k_F\) is the magnitude of the Fermi vectors.
The pumped spin current from the FM|NM interface by the precessing magnetization in Equation (6.1) can then be written as,

$$I'_{sp} = \frac{\hbar}{4\pi g_{\uparrow\downarrow}} \left[ n \times \frac{\partial n}{\partial t} \right].$$

(6.8)

Spin pumping into a diffusive medium (non-ballistic) must be modified by replacing the spin mixing conductance $g_{\uparrow\downarrow}$ with $\tilde{g}_{\uparrow\downarrow}$ [80]. The pumped spin current then reads

$$I_{sp} = \frac{\hbar}{4\pi \tilde{g}_{\uparrow\downarrow}} \left[ n \times \frac{\partial n}{\partial t} \right].$$

(6.9)

$\tilde{g}_{\uparrow\downarrow}$ is the renormalized spin mixing conductance, obtained by the subtraction of a spurious Sharvin conductance $g_{Sh}$ [91],

$$\frac{1}{\tilde{g}_{\uparrow\downarrow}} = \frac{1}{g_{\uparrow\downarrow}} - \frac{1}{2g_{Sh}}.$$  

(6.10)

Since the Sharvin conductance is calculated by counting the number of conduction channels, and for intermetallic interfaces $g_{\uparrow\downarrow}$ is also mainly determined by counting the number of single spin conduction channels [90], then

$$g_{\uparrow\downarrow} \approx g_{Sh}. $$

(6.11)

This approximation leads to the renormalized spin mixing conductance:

$$\tilde{g}_{\uparrow\downarrow} \approx 2g_{\uparrow\downarrow}. $$

(6.12)

The treatment of spin pumping becomes easier by the presence of at least partially diffuse electron momentum scattering at the FM|NM interface. This allows one to introduce the chemical potential for the majority ($\mu_{major}$) and minority ($\mu_{minor}$) spins in NM which results in an accumulated spin density $S$ in NM. The accumulated spin density $S$ propagates across the NM spacer by a spin diffusion current. The accumulated spin density is given by

$$S = \frac{\hbar}{2} N(E_F) \Delta \mu, $$

(6.13)

where $\Delta \mu = \mu_{major} - \mu_{minor}$. $N(E_F)$ is the single spin density of states at the Fermi level in NM, where for the free electron approximation

$$N(E_F) = \frac{k_F^2}{2\pi^2 \hbar v_F^2}. $$

(6.14)

where $v_F$ is the Fermi velocity in the NM.
The accumulation of spin density at the NM|FM interface together with the requirement of no net electron flow leads to a backflow spin current. The backflow of spin current is given by the magneto-electronics equations. The transverse (perpendicular to the instantaneous FM magnetization vector) spin current is fully absorbed at the FM|NM interface and is given by [80]

\[ I_{bf} = -\frac{1}{4\pi} \tilde{g} \n \times \Delta \mu \times n. \]  

(6.15)

From Equations (6.13) and (6.15) it follows that the spin current backflow \( I_{bf} \) is proportional to the accumulated spin density \( S \) at the FM|NM interface.

For small rf precession the induced rf magnetic moment in FM is nearly perpendicular to the saturation magnetic moment and consequently the rf accumulated spin density in linear approximation is also perpendicular to the saturation magnetization and will be described by \( s \). The expression \( n \times S \times n \) can be then replaced by \( s \) at the FM|NM interface.

Using Equations (6.7) and (6.12) to (6.15) the spin backflow at the FM|NM interface is

\[ I_{bf} = -\frac{1}{2} v_F s(x) \bigg|_{x=0}, \]  

(6.16)

where \( s(0) \) is the accumulated spin density at the FM|NM interface and \( v_F \) is the Fermi velocity in the NM layer adjacent to FM layer.

The Fermi surface in zero electric field must be symmetric about the origin. Equation (6.16) states that the accumulated spin density associated with \( k \)-vectors impinging on the FM|NM interface is perfectly absorbed by FM.

### 6.1.2 Spin pumping at the FI|NM interface

From the from of Equations (6.1) to (6.3), it is not particularly obvious that spin pumping should be possible from a ferromagnetic insulator (FI). This is because the carrier for the spins is the electron and at the FI|NM interface \( |r_{\uparrow,mn}| = |r_{\downarrow,mn}| = 1 \) and \( |t_{\uparrow,mn}| = |t_{\downarrow,mn}| = 0 \), as the ferromagnet is an insulator. However, when one considers that the reflection and transmission parameters are complex owing to the spin’s quantum mechanical nature, one can show that spin pumping is in fact possible from a FI.

Since the magnitude of the transmission coefficients is 0, one only needs to consider the reflection coefficients. First, one can separate the complex reflection coefficients into the product of their amplitude and their quantum mechanical phase,

\[ r_{\uparrow,mn} = |r_{\uparrow,mn}| e^{i\theta_{\uparrow,mn}} = e^{i\theta_{\uparrow,mn}} \]  

(6.17)

\[ r_{\downarrow,mn} = |r_{\downarrow,mn}| e^{i\theta_{\downarrow,mn}} = e^{i\theta_{\downarrow,mn}}. \]  

(6.18)

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where $\theta_{\uparrow, mn}$ and $\theta_{\downarrow, mn}$ are the phases of the reflected majority and minority spins. Since the FI has different potential barriers for the majority and minority spins, the phase of the reflected spins is spin dependent.

Inserting Equations (6.17) and (6.18) and $t_{\uparrow, mn} = t_{\downarrow, mn} = 0$ into Equation (6.2) yields the expression

$$g_{\uparrow \downarrow} = \frac{1}{2} \sum_{mn} \left[ (e^{i\theta_{\uparrow, mn}} - e^{i\theta_{\downarrow, mn}}) (e^{-i\theta_{\uparrow, mn}} - e^{-i\theta_{\downarrow, mn}}) \right]$$

$$= \sum_{mn} \left( 1 - \cos (\Delta \theta_{mn}) \right),$$

where $\Delta \theta_{mn} = \theta_{\uparrow, mn} - \theta_{\downarrow, mn}$ is the quantum mechanical phase difference of a NM electron in $n$th conduction channel reflected at the FM|NM interface. Equations (6.9) to (6.16) are still valid in the case of spin pumping from FI into a medium that is in the diffusive regime with respect to spin currents. Equation (6.8) is still valid when spin pumping into mediums where spin current is dephased in the ballistic electron transport regime. Theses different regimes will be discussed in Sections 6.2 and 6.3.

### 6.2 Spin diffusion theory

#### 6.2.1 Spin diffusion length

Consider an itinerant electron with charge and spin; this electron has both linear momentum and spin momentum. On average, the electron travels the mean free path $\lambda_m$ before a linear momentum scattering event occurs. When spin relaxation is mainly due to the spin-orbit interaction, the electron undergoes $N$ linear momentum scattering events before spin flip scattering in the time $\tau_{sf}$. When the propagation of the electron is diffusive, the average distance traveled perpendicular to the interface is the spin diffusion length $\delta_{sd} = \lambda_m \sqrt{N/3}$.

The itinerant electron is moving about at the Fermi velocity $v_F$, therefore the total distance travelled by the electron before a spin flip scattering is given by $N\lambda_m = v_F \tau_{sf}$. The spin diffusion length can therefore be written as $\delta_{sd} = \sqrt{\lambda_m v_F \tau_{sf} / 3}$. Using the relation $v_F = \lambda_m / \tau_m$, one can write the spin diffusion length in terms of the mean free path and the ratio of the spin flip scattering rate to the linear momentum scattering rate,

$$\delta_{sd} = \lambda_m \sqrt{\frac{1}{3} \frac{\tau_{sf}}{\tau_m}}.$$
6.2.2 Spin diffusion in metallic heterostructures

The propagation of accumulated spin density is described by spin diffusion theory which is similar to standard diffusion with the modification made to allow the relaxation of spin density on time scales of $\tau_{sf}$, leading to the spin diffusion equation [9],

$$\frac{\partial s_i(x)}{\partial t} = D_i \frac{\partial^2 s_i(x)}{\partial x^2} - \frac{s_i(x)}{\tau_{sf,i}}, \quad (6.22)$$

where $s_i$ is the accumulated spin density, $D_i = v_i^2 \tau_{m,i}/3$ is the spin diffusion constant, $v_i$ is the Fermi velocity, $\tau_{m,i}$ is the electron momentum scattering time, and $\tau_{sf,i}$ is the spin flip scattering time all in the $i$th layer. Since the magnetization precession times at our microwave frequencies are much longer than the spin flip relaxation times, Equation (6.22) can be approximated as time independent. The general solution is therefore of the form

$$s_i(x) = A_i e^{\kappa_i x} + B_i e^{-\kappa_i x}, \quad (6.23)$$

where the coefficients $A_i$ and $B_i$ are determined by appropriate boundary conditions given by the magneto-electronic equations and $\kappa_i = 1/v_i \sqrt{\tau_{m,i}\tau_{sf,i}/3}$ is the inverse spin diffusion length.

At the FM|NM1 interface the boundary conditions is, see cartoon in Figure 6.2,

$$- D_1 \frac{\partial}{\partial x}s_1(x) = I_{sp} - \frac{1}{2} v_1 s_1(x) \bigg|_{x=0}, \quad (6.24)$$

which states that the net forward flow of spin current at the FM|NM1 interface, $I_{sp}^{net} = -D_1 \partial / \partial x s_1(0)$, is given by the pumped spin current $I_{sp}$ plus the the backflow spin current $I_{bf}$ (note that $I_{bf}$ is defined as negative) evaluated at the FM|NM1 interface, see Equation (6.16). Explicitly, the relation is

$$I_{sp}^{net} = I_{sp} + I_{bf}. \quad (6.25)$$

The net spin current pumped into the NM is equal to the net spin momentum lost by the FM, i.e. the induced damping in the FM layer. For ultrathin FM,

$$\left( \frac{\partial M}{\partial t} \right)_{sp} = \alpha_{sp} \left[ M \times \frac{\partial \mathbf{n}}{\partial t} \right], \quad (6.26)$$

$$= \frac{\gamma}{d_{FM}} \left( I_{sp} - \frac{1}{2} v_1 s_1(0) \right). \quad (6.27)$$
The term before the brackets on the RHS of Equation (6.26) takes into account the conversion from spin dynamics to magnetization dynamics and the fact that for ultrathin FM the interface transfer of moment is shared equally throughout the volume of FM by the exchange interaction. This leads to the spin pumping induced damping in the FM

$$\alpha_{sp} = \frac{g\mu_B}{4\pi M_s} \tilde{g}_{1\uparrow\downarrow} \frac{1}{d_{FM}} \left[ \frac{1}{1 + \Upsilon} \right],$$

with the form of $\Upsilon$ determined by the material parameters and boundary conditions of the adjacent system.

**Spin diffusion in FM|NM bilayer**

![Figure 6.2: A schematic view of the FM|NM1 bilayer. A precessing magnetization in the FM layer leads to a spin current $I_{sp}$ in the adjacent NM1 structure. At the FM|NM1 interface, an accumulation of spin density leads to a backflow spin current $I_{bf}$ into the FM. The net spin current pumped into the system is therefore $I_{net}^{sp} = I_{sp} + I_{bf}$.](image)

In the case of a single NM layer of thickness $d_1$, see Figure 6.2, there is a perfect reflection of spin current at the NM1|vacuum interface. This leads to the second boundary condition (in addition to Equation (6.24))

$$- D_1 \frac{\partial}{\partial x} s_1(x) = 0 \bigg|_{x=d_1}. \quad (6.29)$$

The solution to Equations (6.22), (6.24) and (6.29) is

$$s_1(x) = \frac{I_{sp}}{\frac{v_1}{2} + D_1 \kappa_1 \tanh(d_1 \kappa_1)}. \quad (6.30)$$

Using Equations (6.26), (6.27) and (6.30) leads to $\alpha_{sp}$ of the FM|NM1 structure having dependence on the thickness of NM1 with $\Upsilon$ in Equation (6.28) taking the form

$$\Upsilon_1 = \frac{v_1}{2} D_1 \kappa_1 \tanh(d_1 \kappa_1). \quad (6.31)$$
When the thickness of NM1 is much smaller than the spin diffusion length in NM1, 
\( d_1 \ll \delta_{sd,1} \), NM1 is a very inefficient spin absorber; the pumped spin current is reflected 
at the vacuum interface and reabsorbed by FM. This process leads to no increase in the 
Gilbert damping of the FM.

**Spin diffusion in FM|NM1|FM2 trilayer**

![Figure 6.3: A schematic view of the FM|NM1 bilayer. A precessing magnetization in the FM layer leads to a spin current \( I_{sp} \) in the adjacent NM1|FM2 structure. The spin current reaching the NM1|FM2 interface is absorbed by FM2.](image)

In the case of a NM spacer of thickness \( d_1 \) between two FMs (FM at resonance, FM2 off resonance), see Figure 6.3, only the boundary condition at \( x = d_1 \) changes to that of perfect absorption of all spin current impinging on the NM1|FM2 interface (spin sink) [9]

\[
-D_1 \frac{\partial}{\partial x} s_1(x) = \left. \frac{1}{2} v_1 s_1(x) \right|_{x = d_1}.
\]  
(6.32)

The solution to Equations (6.22), (6.24) and (6.32) is

\[
s_1(x) = \frac{D_1 \kappa_1 \cosh(\kappa_1 (d_1 - x)) + \frac{v_1}{2} \sinh(\kappa_1 (d_1 - x))}{v_1 D_1 \kappa_1 \cosh(d_1 \kappa_1) + \left( (D_1 \kappa_1)^2 + \left( \frac{v_1}{2} \right)^2 \right) \sinh(d_1 \kappa_1)} I_{sp}.
\]  
(6.33)

Using Equations (6.26), (6.27) and (6.33) leads to \( \alpha_{sp} \) of the FM|NM1|FM2 structure
having dependence on the thickness of NM1 with \( \Upsilon \) in Equation (6.28) taking the form

\[
\Upsilon_2 = \frac{\frac{v_1}{2} D_1 \kappa_1 + \left( \frac{v_1}{2} \right)^2 \tanh(d_1 \kappa_1)}{\frac{v_1}{2} D_1 \kappa_1 + (D_1 \kappa_1)^2 \tanh(d_1 \kappa_1)}.
\]  
(6.34)

When the thickness of NM1 is much smaller than the spin diffusion length in NM1, 
\( d_1 \ll \delta_{sd,1} \), the forward spin current, \( I_{sp}^{net} = I_{sp}/2 \), is entirely absorbed at the NM1|FM2 interface. This process leads to the maximum increase in the Gilbert damping of FM. If both ferromagnets are on resonance at the same time and NM1 is \( d_1 \ll \delta_{sd,1} \), but thick
enough for FM and FM2 to not be static interlayer exchange coupled, then there will be negligible change in damping in both layers [75].

**Spin diffusion in FM|NM1|NM2 trilayer**

![Diagram of a schematic view of the FM|NM1|NM2 bilayer](image)

Figure 6.4: A schematic view of the FM|NM1|NM2 bilayer. A precessing magnetization in the FM layer leads to a spin current $I_{sp}$ in the adjacent NM1|FM2 structure. The boundary conditions at the NM1|NM2 interface are the continuity of accumulated spin density and continuity of spin current. The spin current reaching the NM2|vacuum interface is reflected.

In the case of the FM|NM1|NM2 system with NM1 thickness $d_1$ and NM2 thickness $d_2$, see Figure 6.4, the boundary condition at the FM|NM1 interface remains the same as Equation (6.24). At the NM1|NM2 interface, the boundary conditions are the continuity of accumulated spin density

$$s_1(x) = s_2(x) \bigg|_{x=d_1} \quad (6.35)$$

and the continuity of spin current across the interface

$$-D_1 \frac{\partial}{\partial x} s_1(x) = -D_2 \frac{\partial}{\partial x} s_2(x) \bigg|_{x=d_1}. \quad (6.36)$$

At the outer NM2|vacuum interface the boundary condition is that of perfect reflection of spin current

$$-D \frac{\partial}{\partial x} s_2(x) = 0 \bigg|_{x=d_1+d_2}. \quad (6.37)$$

The solution to Equations (6.22), (6.24) and (6.35) to (6.37) is

$$s_1(x) = \frac{D_1 \kappa_1 \cosh (d_2 \kappa_2) \cosh (\kappa_1 (d_1 - x)) + D_2 \kappa_2 \sinh (d_2 \kappa_2) \sinh (\kappa_1 (d_1 - x))}{Num.} I_{sp}, \quad (6.38)$$

$$s_2(x) = \frac{D_1 \kappa_1 \cosh (\kappa_2 (d_1 + d_2 - x))}{Num.} I_{sp}, \quad (6.39)$$
where
\[
\text{Num.} = D_1 \kappa_1 \cosh (d_2 \kappa_2) \left( \frac{v_1}{2} \cosh (d_1 \kappa_1) + D_1 \kappa_1 \sinh (d_1 \kappa_1) \right) + \\
D_2 \kappa_2 \sinh (D_2 \kappa_2) \left( D_1 \kappa_1 \cosh (d_1 \kappa_1) + \frac{v_1}{2} \sinh (D_1 \kappa_1) \right). \tag{6.40}
\]

Using Equations (6.26), (6.27), (6.38) and (6.39) leads to \( \alpha_{sp} \) of the FM|NM1|NM2 structure having dependence on the thickness of both NM1 and NM2 with \( \Upsilon \) in Equation (6.28) taking the form
\[
\Upsilon_3 = \frac{\frac{v_1}{2} D_1 \kappa_1 + \frac{v_1}{2} D_2 \kappa_2 \tanh (d_2 \kappa_2) \tanh (d_1 \kappa_1)}{D_1 \kappa_1 D_2 \kappa_2 \tanh (d_2 \kappa_2) + (D_1 \kappa_1)^2 \tanh (d_1 \kappa_1)}. \tag{6.41}
\]

It is important to note that there is a discontinuity in the evaluation of Equation (6.41) when setting \( d_1 = 0 \). This arises from the fact that both \( \tilde{g}_{\uparrow \downarrow} \) and \( v_1 \) need to be defined for the layer adjacent to the precessing FM. This is because the spin current is being pumped into this layer and then diffuses throughout the rest of the system. The appropriate result in this case, see Equation (6.31), can be had by letting \( v_1 \rightarrow v_2 \) in the numerator of Equation (6.41) as this term is a consequence of the backflow of spin current, see Equation (6.24).

6.2.3 Spin diffusion theory applicability

It is worth expanding on the consequences of using spin diffusion theory outside of its derivation. Spin pumping theory was first derived assuming that the spin current from FM is pumped into a NM reservoir modeled as a perfect spin sink [78]. In a perfect spin sink, it is assumed that the spin current leaves the interface or decays fast enough that spin accumulation does not build up, i.e. there is no backflow spin current. In this case, the spin pumping is governed by the bare spin mixing conductance \( g_{\uparrow \downarrow} \), as in Equation (6.8).

In the diffusive NM system, one has to include the accumulated spin density, backflow spin currents, and the renormalized spin mixing conductance, see Equations (6.9) and (6.16). The ratio
\[
\epsilon = \frac{\tau_m}{\tau_{sf}} \tag{6.42}
\]
is an important parameter for determining whether a particular material can be treated as a diffusive NM with respect to spin currents.

To illustrate this fact, let us consider first the FM|NM1|FM2 system, with FM at resonance, NM1 as a diffuse spin scatterer, and FM2 as a perfect spin sink. Letting the thickness of the NM1 layer approach 0 leads to \( \lim_{d_1 \rightarrow 0} \Upsilon_2 = 1 \). In this limit, the term \( [1 / (1 + \Upsilon_2)] \) in Equation (6.28) reduces to 1/2. Multiplying this term by the renormalized spin mixing conductance \( \tilde{g}_{\uparrow \downarrow} (\approx 2g_{\uparrow \downarrow}) \) recovers the bare spin mixing conductance \( g_{\uparrow \downarrow} \), the original result.
of FM pumping directly into a perfect spin sink, i.e. the maximum spin pumping one can achieve.

Now consider the FM|NM system, where for very thick NM,

\[ \lim_{d_1 \to \infty} \Upsilon_1 = \left( \frac{4}{3} \epsilon \right)^{-\frac{1}{2}}. \]

(6.43)

If in this case \( \epsilon > 3/4 \), then the term \( \left[ 1 / (1 + \Upsilon_1) \right] \) in Equation (6.28) is greater than 1/2 and this yields a result of spin pumping that is greater than the maximum; this is not physically acceptable. Therefore, materials with \( \epsilon > 3/4 \) should not be treated as diffusive NMs with respect to Equation (6.22).

### 6.3 Spin decoherence theory

In metallic materials with strong spin correlation effects, such as Pd and Pt, thermally excited fluctuations of local spin moment known as paramagnons lead to dephasing of spin currents on much shorter length scales than in NM systems. In this thesis, I will refer to these materials as spin fluctuating metals (SFM). In SFMs, the spin pumping contribution to the Gilbert damping, \( \alpha_{sp} \), saturates at a thickness shorter than the mean free path of electrons. Using Equations (6.21) and (6.42) one can show that when \( \delta_{sd} < 2\lambda_m/3 \), then \( \epsilon > 3/4 \) and the spin diffusion model no longer holds. For this reason, Foros et al. [1] introduced the ballistic spin decoherence theory to describe spin transport in these systems.

#### 6.3.1 Ballistic spin decoherence

The model presented by Foros et al. [1] is based on ballistic transport through the SFM with exponential spin relaxation. The model was originally proposed to describe the \( \alpha_{sp} \) in Fe|Pd structures. Since the transport was taken to be ballistic, the concept of accumulated density is not needed. Without the accumulated density, the backflow due to accumulation is not present in this model, see Equation (6.16), and the pumped spin current is governed by the bare spin mixing conductance as in Equation (6.8). The spin current generated by the precessing FM at the FM|SFM interface propagates across the SFM layer and gets dephased due to exchange interaction with fluctuating paramagnons. The pumped spin current is dephased on the length scale \( \lambda_{dec} \).

For sufficiently thick SFMs, all the spin current pumped into the SFM layer is absorbed in the FM|SFM system. For thinner SFMs in the FM|SFM system, the spin current is reflected at the SFM|vacuum interface leading to the reflected spin current[1]

\[ I_{sp}^{ret} = I_{sp} e^{-2d_{SFM} / \lambda_{dec}}, \]

(6.44)
where $d_{\text{SFM}}$ is the thickness of the SFM layer. The net spin current absorbed by the SFM layer in the FM|SFM system is therefore

$$I_{\text{sp,dec}}^{\text{net}} = \frac{\hbar}{4\pi} g_{\uparrow \downarrow} \left[1 - e^{-\frac{2d_{\text{SFM}}}{\lambda_{\text{dec}}}}\right] \left[n \times \frac{\partial n}{\partial t}\right]. \quad (6.45)$$

This leads to

$$\alpha_{\text{sp,dec}} = g\mu_B \frac{g_{\uparrow \downarrow}}{4\pi M_s} \frac{1}{d_{\text{FM}}} \left(1 - e^{-\frac{2d_{\text{SFM}}}{\lambda_{\text{dec}}}}\right). \quad (6.46)$$

### 6.3.2 Modified spin decoherence

Burrowes and Heinrich [92] introduced a modified spin decoherence model that allowed for backflow of accumulated density at the FM|SFM interface in the FM|SFM bilayer. In this case the renormalized spin mixing conductance must be used and $I_{\text{sp}}$ is given by Equation (6.9) and the backflow due to accumulation $I_{\text{bf}}$ is given by eq. (6.16). In addition the spin current that reaches the SFM|vacuum interface is reflected back and is reabsorbed by the FM

$$I_{\text{sp}}^{\text{ref2}} = \frac{1}{2} v_{\text{SFM}} s_{\text{SFM}}(x)e^{-\frac{2d_{\text{SFM}}}{\lambda_{\text{dec}}}}, \quad (6.47)$$

which results in net forward spin current

$$I_{\text{sp, mdec}}^{\text{net}} = \frac{1}{2} v_{\text{SFM}} s_{\text{SFM}}(x) \left(1 - e^{-\frac{2d_{\text{SFM}}}{\lambda_{\text{dec}}}}\right). \quad (6.48)$$

The solution of Equations (6.9), (6.16), (6.25) and (6.48) leads to

$$\alpha_{\text{sp, mdec}} = g\mu_B \frac{2g_{\uparrow \downarrow}}{4\pi M_s} \frac{1}{d_{\text{FM}}} \left(\frac{1 - e^{-2d_{\text{SFM}}/\lambda_{\text{dec}}}}{2 - e^{-2d_{\text{SFM}}/\lambda_{\text{dec}}}}\right). \quad (6.49)$$
Chapter 7

Spin pumping and spin transport in
GaAs(001)|17Fe|d_{Au}Au|50Pd|20Au

In this chapter, spin injection by means of spin pumping at the ferromagnetic metal (Fe)/normal metal interface (Au) and the subsequent spin transport in Au|Pd heterostructures as studied using ferromagnetic resonance is presented. The spin pumping induced damping in Fe|Pd was greatly suppressed by the addition of a Au spacer layer in the structure Fe/Au/Pd. It is shown experimentally that this reduction of damping is not due to the removal of magnetic proximity effect induced damping at the Fe|Pd interface. Instead, it is shown that its origin is from a reflection of spin current at the Au|Pd interface; this indicates Pd cannot be treated as an ideal spin sink. In this chapter, three models of spin transport in the heterostructure are presented: (a) spin diffusion in Au and Pd, (b) spin diffusion in Au/modified spin decoherence in Pd, and (c) spin diffusion in Au/ballistic spin decoherence in Pd. Since the Pd layer represents a strongly spin correlated system, model (a) leads to a spin flip relaxation time that is nearly equal to the electron momentum scattering time in Pd; thus, it is concluded that spin diffusion is a non-physical model for Pd. Models (b) and (c) take into account the spin current decoherence due to interaction with fluctuating paramagnons in Pd. However, model (b) becomes material independent for thick Pd. Model (c), resolves this issue by introducing a material dependent reflection of spin current at the Au|Pd interface. Furthermore, oscillations in the amplitude of spin pumping induced damping were observed in Fe|Au|Pd as a function of Au thickness when the electron mean free path in the bulk Au was larger than the Au layer thickness. This indicates a formation of quantum well states in the Au spacer that affect pure spin currents.
7.1 Motivation

The generation and transport of spin currents in normal non-magnetic (NM) metals, such as Au, Ag, Cu, has been extensively studied by ferromagnetic resonance (FMR) in FM|NM and FM|NM|FM (FM stands for a ferromagnetic layer) structures and is well described by the standard spin pumping and spin diffusion model [10, 59, 82, 93–96]. Spin pumping leads to interface damping at the FM|NM interface that can be described by Gilbert phenomenology. There are two alternative theories of spin pumping at the FM|NM interface, the theory by Tserkovnyak et al. [78] based on time dependent scattering matrix formalism [97] and the theory by Simánek and Heinrich [79] based on time retarded response of the interlayer exchange coupling.

The interlayer exchange coupling between the magnetizations of two ferromagnets in FM|NM|FM2 structures was found experimentally in 1986 [98–100]. The 1988 discovery of the giant magnetoresistance [101, 102] stimulated a large interest, both experimentally and theoretically, into the study of multilayer FM and NM thin film structures. The simplest form of the interlayer exchange coupling is the so called bilinear interlayer exchange coupling which takes the form

$$E = -J_{ex} n_1 \cdot n_2,$$

(7.1)

where $J_{ex}$ is the bilinear exchange energy per unit area and $n_1$ and $n_2$ are the directions of the magnetization for FM1 and FM2, respectively. Positive values of $J_{ex}$ lead to lower energy for parallel alignment of FM1 and FM2, whereas negative values favor anti-parallel alignment.

Experiments in the late 1980s and early 1990s showed that the sign of $J_{ex}$ oscillates as a function of the NM spacer thickness in FM|NM|FM structures [98, 103, 104]. After this discovery, numerous theories were presented to explain the oscillatory behavior, see ref. [105] for an overview of the early theories. In 1991, Edwards et al. [106] developed a theory to describe the oscillations in the interlayer exchange coupling that was based on an analogue of the de Hass-van Alphen (dHvA) effect. This theory showed that the periodicity of the oscillations were given by the properties of the NM Fermi surface, mainly that the critical spanning vectors determine the periodicity of the oscillations. A critical spanning vector is a vector that connects two sheets on the Fermi surface, at $\pm k_F$, that are parallel, see Figure 7.1. In 1991, Bruno and Chappert [107] calculated two periods of oscillations for the Fe|Au(001) interface, 2.51 and 8.60 AL, corresponding to the critical spanning vectors based on the results of dHvA and cyclotron-resonance measurements [108]. In 1994, Unguris et al. [109] used scanning electron microscopy with polarization analysis (SEMPA) to measure the
oscillatory periods of the interlayer exchange coupling in Fe|Au|Fe(001) to be $2.48 \pm 0.05$ and $8.6 \pm 0.3$ AL, in great agreement with the periods predicted by Bruno and Chappert.

In 1993, Bruno [111] and Stiles [112] presented similar models of interlayer exchange coupling that provided a means to unify the previous models. While the previous models were able to show the oscillation period and its relation to the critical spanning vectors of the NM, they were not good at predicting the oscillation amplitude and phase. The models of Bruno and Stiles were also similar to that of Mathon et al. [113], in that they showed that spin dependent reflectivity at the FM|NM interfaces was the origin of the interlayer exchange coupling and that quantum well states (or resonances) are formed due to the spin polarized reflection. The parameters required to explain the interlayer exchange coupling involve the complex reflection parameters $r^\uparrow$ and $r^\downarrow$ at the FM|NM interfaces and the thickness of the NM spacer layer, which are also the parameters that are involved with spin pumping and spin diffusion. For more details on interlayer exchange coupling, see the review chapter by Stiles [105].

Photoemission and inverse photoemission have shown that the density of states at the Fermi level raises and lowers periodically with increasing NM thickness in FM|NM structures. The increase in density of states is associated with new quantum well states in the NM crossing the Fermi level [114, 115]. At the NM|vacuum interface there is perfect reflection and at the FM|NM interface there is a spin dependent potential step that makes the reflectivity spin dependent. The period of the oscillations in intensity in photoemission studies of FM|NM structures matches the period of oscillation in the interlayer exchange
coupling in FM|NM|FM structures when FM and NM are the same materials as studied in interlayer exchange coupling [114–117].

Joly et al. [118, 119] have shown spin polarized electron reflection experiments that exhibit quantum well state induced oscillations of the electron spin motion. The electron spin motion refers to a precession about or a rotation towards the magnetization vector of a spin polarized electron. In ref. [119], the system studied is Au|Co(0 0 1). In the experiments, a spin polarized beam of electrons impinges on the Au side of the sample at an angle of 45° to the film normal with the polarization perpendicular to the in-plane magnetization of the Co (which has been remanently magnetized along an easy [1 1 0] direction, which is defined as the z-direction). For an incident electron beam polarized in the x-direction (perpendicular to magnetization) the spin wave function can be expressed as a superposition of majority |↑⟩ and minority |↓⟩ eigenstates of the magnetization,

\[ \chi_i = \frac{1}{\sqrt{2}} (|↑⟩ + |↓⟩) e^{i\beta}, \] (7.2)

where \( \beta \) is the arbitrary phase. After spin dependent reflection, the spin wave function is

\[ \chi_r = \frac{1}{\sqrt{2}} \left[ |r_↑| e^{i\theta_↑} |↑⟩ + |r_↓| e^{i\theta_↓} |↓⟩ \right] e^{i\beta}, \] (7.3)

where \(|r_↑(↓)|\) and \(\theta_↑(↓)\) are the moduli and phases of the spin majority (minority) reflection amplitude. The transverse spin components of the reflected electron beam are measured using a Mott detector. The spin motion is given by

\[ \epsilon_s = \theta_↓ - \theta_↑ \] (7.4)

and

\[ \phi_s = \arctan \left( \frac{|r_↑|^2 - |r_↓|^2}{2P_i |r_↑| |r_↓|} \right), \] (7.5)

where \( \epsilon_s \) is the precession angle and \( \phi_s \) is the rotation angle of the polarized electron beam with respect to the Co magnetization and \( P_i \) is the degree of polarization of the electron beam, see Figure 7.2. Both \( \epsilon_s \) and \( \phi_s \) were shown to oscillate with the Au overlayer thickness between 0 and 50 AL. Again the parameters governing the spin motion are the same as those governing the spin pumping. This can be seen by expressing the real part of the spin mixing conductance as \( g_{↑↓} = (1/2) \sum_{mn} |r_↑|^2 + |r_↓|^2 - 2 \cos (\theta_↑ - \theta_↓) \) (see Section 6.1). However, in this case the electrons have energies above the Fermi level in Au and the beam impinges on the surface under an angle of 45° to the surface normal, as opposed to spin pumping and interlayer exchange coupling that involve electrons at the whole Fermi surface.
Figure 7.2: Schematic of the two angles of spin motion of the polarization vector: a precession about the magnetization $M$ by an angle $\epsilon_s$ and a rotation by an angle $\phi_s$. $P_i$ and $P_r$ represent the polarization of the incident and reflected beams.

Considering that the interlayer exchange coupling and spin pumping are a result of spin dependent reflectivity at the FM|NM interface and spin pumping also generates an accumulated spin density in NM [120, 121], it is not unreasonable to expect that in some structures the spin pumping contribution to the interface damping can in principle show some degree of oscillatory behavior as a function the NM layer thickness. This has not been previously reported, which is not that surprising for FM|NM systems because in FMR with a small angle of precession the spin current moment is fully absorbed at the FM|NM interfaces as the FMs act as ideal spin sinks [75, 120, 121]. Since no oscillatory behavior has been found in FM|NM systems using simple normal metals such as Au, Ag, and Cu, we decided to investigate spin pumping in heterostructures FM|NM1|NM2 where the second NM2 layer differs significantly in spin transport behavior from the NM1 layer.

In Pd, thermally excited fluctuations of local spin moment known as paramagnons [122, 123] lead to the absorption of spin current in Fe|Pd structures in a different manner than expected from using a simple spin diffusion model based on spin-orbit interaction relaxation alone. Fe|Pd structures have shown a large increase in the Fe layer interface damping that saturates with the Pd layer thickness around 10 nm. This is a significantly shorter length scale than that observed for Au layers where the spin diffusion length in Au was found to be $\sim 60$ nm [124, 125]. In fact, the spin decoherence length (5.6 nm) in Pd was actually found
to be shorter than the electron mean free path (∼9 nm) in the studied Fe|Pd structures [1]. This means that the decoherence of the spin current is happening in the ballistic electron transport regime, rather than the diffusive transport regime. For these reasons spin current propagation in Pd layers was interpreted using a concept of spin decoherence [1, 92]. In this respect Fe|Au|Pd represents an interesting heterostructure in which the Au layer behaves as a simple NM with spin moment relaxation provided by the spin-orbit interaction and in the Pd layer the spin current relaxation is dominated by exchange interaction with fluctuating paramagnons. Therefore, Pd layers represent a spin system which behaves in between that of a FM and a NM and will be referred to as spin fluctuating metal (SFM).

The goal of this work was to investigate the spin transport in heterostructures of the form Fe|Au(NM)|Pd(SFM)|Au. The thickness of the Pd layer was larger than the spin decoherence length in Pd, and therefore the returning spin current from the Pd|vacuum interface was only a small fraction of that entering the Pd layer. In this case, the study was primarily directed towards the effectiveness of spin pumping as a function of the Au interlayer thickness, allowing one to examine the role of the Au(NM)|Pd(SFM) interface in spin pumping and spin current transport.

7.2 Sample selection and preparation

For this study, single crystal GaAs(001)|17Fe|dAu50Pd20Au samples were prepared by means of molecular beam epitaxy (MBE), where the integers refer to the layer thickness in atomic layers (AL) along the [001] direction (1 AL of bcc Fe = 0.143 nm, 1 AL of fcc Au = 0.204 nm, 1 AL of fcc Pd = 0.195 nm). Commercially available, epi-ready GaAs(001) substrates were cleaned for 20 minutes by H· cleaning at 400° C followed by 3 hr of 650 V Ar+ sputter etching at room temperature. The samples were then annealed to 585° C resulting in a 4x6 surface reconstruction; annealing was monitored by means of reflection high energy electron diffraction (RHEED). The film thicknesses were monitored by means of oscillations in the intensity of the specular spot at an anti-Bragg RHEED reflection in conjunction with a quartz crystal thickness monitor, see Figure 7.3. The RHEED oscillations are used to calibrate a quartz crystal thickness monitor for determining the thicknesses of the Fe, Au, and Pd layers. For metallic films, the amplitude of the RHEED oscillations eventually decays due to a decreasing surface atomic terrace size when the adatoms are attached to existing atomic steps; in this case, the surface is laterally invariant to RHEED. For more detail, see Section 2.2. The rms roughness of grown films is of one atomic layer[126, 127].

These materials were selected for this experiment due to:

1) They are well lattice matched, allowing growth that is epitaxial and crystalline, which leads to sharp interfaces and fewer lattice defects. The GaAs has a zinc-blende structure
Figure 7.3: Reflection high energy electron diffraction (RHEED) anti-Bragg specular peak intensity oscillations illustrating epitaxial growth for the sample GaAs|17Fe|87Au|50Pd, for (top) the entire growth of the 17Fe layer and the initial growth of the (mid.) 87Au and (bot.) 50Pd layers. The nonoscillatory behavior of the Au and Pd layers for time > 1000 s is omitted.
with a lattice constant \( a_{\text{GaAs}} = 5.6535 \, \text{Å} \). The lattice constant for bcc Fe is \( a_{\text{Fe}} = 2.865 \, \text{Å} \) meaning that \( a_{\text{Fe}} \) is 1.4% larger than the half unit cell of GaAs. For GaAs(001)/Fe, the inplane epitaxial orientations is \([100]_{\text{Fe}}||[100]_{\text{GaAs}}\). The lattice constant for fcc Au is \( a_{\text{Au}} = 4.080 \, \text{Å} \), which represents a less then 1% mismatch with the Fe(001) diagonal. The inplane epitaxial orientations is \([100]_{\text{Au}}||[110]_{\text{Fe}}\), representing a 45° rotation of the Au(001) layer with respect to both the GaAs(001) and Fe(001) layers. The lattice constant for fcc Pd is \( a_{\text{Pd}} = 3.88898 \, \text{Å} \), which is 4.6% smaller than the Au. The Pd has the same orientation as the Au.

2) Spin pumping and spin transport in GaAs\(|\text{Fe}|d_{\text{Au}}\text{Au}\) [128], GaAs\(|\text{Fe}|d_{\text{Au}}\text{Au}|\text{Fe}\) [121], and GaAs\(|\text{Fe}|d_{\text{Pd}}\text{Pd}\) [1] structures has been well studied and understood. Thus the spin dynamic properties of the individual layers are well understood. This allows one to explore new behavior at the Au\(|\text{Pd}\) (NM\(|\text{SFM}\) interface.

The Fe layer thickness of \( d_{\text{Fe}} = 17 \, \text{AL} \) was chosen for the following reasons based on the results presented in Section 5.4: 1) The spin pumping contribution to the damping \( \alpha_{\text{sp}} \) is inversely proportional to the Fe film thickness \( d_{\text{Fe}} \), so the film must be thin to obtain good sensitivity to spin pumping. 2) there is an interface contribution to the damping at the GaAs\(|\text{Fe}\) interface that is also nearly inversely proportional to the Fe film thickness \( d_{\text{FM}} \). For \( d_{\text{Fe}} > 14 \, \text{AL} \), the dependence is already slow. 3) For \( d_{\text{Fe}} \geq 25 \, \text{AL} \), the GaAs\(|\text{Fe}\) lattice mismatch leads to the onset of misfit dislocations that lead to two magnon scattering contributions to the damping [46, 129].

### 7.3 Experimental details

FMR measurements were carried out in a multi-mode microwave cavity in a field swept, field modulated configuration, as detailed in Section 4.2. The cavity allowed four resonance frequencies at \( f \simeq 27.2, 31.2, 35.7, \) and 40.4 GHz. The FMR measurements were performed along the GaAs \([1\bar{1}0]\) crystallographic axis (corresponding to the hard axis of Fe). The measured FMR data was mainly described by a Lorentzian lineshape due the absorption contribution \( \chi'' \) to the complex transversal magnetic susceptibility \( \chi = \chi' + i\chi'' \), see Figure 7.4. A small contribution of \( \chi' \) was fit to the FMR data as described in Section 3.4.

The FMR linewidth was well described by Gilbert like damping,

\[
\Delta H(\omega) = \alpha \frac{\omega}{\gamma} + \Delta H(0),
\]

where \( \omega \) is the microwave angular frequency, and \( \Delta H(0) \) is the zero frequency line broadening due to long range magnetic inhomogeneities [38, 44], see Figure 7.5. For the Fe layer \( M_s = 1710 \, \text{G} \) and \( g = 2.09 \) [54, 55], leading to \( \gamma = 1.84 \times 10^7 \, \text{Hz/Oe} \).
Figure 7.4: Example of ferromagnetic resonance data. The colored dots are the raw data and the solid line is theoretical fit. (a) 35.7491 GHz data GaAs|17Fe|20Au with $H_{\text{FMR}} = 7287.8$ Oe and $\Delta H_{\text{FMR}} = 48.1$ Oe. (b) 35.7426 GHz data for GaAs|17Fe|3Pd|20Au with $H_{\text{FMR}} = 6944.7$ Oe and $\Delta H_{\text{FMR}} = 49.4$ Oe. (c) 35.7463 GHz data for GaAs|17Fe|75Au|50Pd|20Au with $H_{\text{FMR}} = 7258.0$ Oe and $\Delta H_{\text{FMR}} = 76.2$ Oe. (d) 35.6580 GHz data for GaAs|17Fe|50Pd|20Au with $H_{\text{FMR}} = 6684.7$ Oe and $\Delta H_{\text{FMR}} = 85.6$ Oe.
The contribution to the damping due to spin pumping is simply given by $\alpha_{sp} = \alpha_{tot} - \alpha_{bulk}$, where $\alpha_{tot}$ is the value of the measured Gilbert damping parameter $\alpha$ in Equation (7.6) and $\alpha_{bulk}$ includes all damping contributions from the Fe layer and the GaAs|Fe interface. Three sample growths of GaAs|17Fe|20Au, where the Au capping layer was sufficiently thin not to contribute to spin pumping induced damping, were measured to have $\alpha = 3.56 (\pm 0.02) \times 10^{-3}$. This value is used to define $\alpha_{bulk} \equiv 3.56 \times 10^{-3}$. It is interesting to note that $\alpha_{bulk}$ is in very good agreement with the fit of the in-plane damping of the samples GaAs|$d_{Fe}$Fe|20Au as a function of $d_{Fe}$ presented in Section 5.4, where using Equation (5.11) and the best fit parameters yeilds $\alpha = 3.57 \times 10^{-3}$. Additionally, all of the samples had small zero frequency offsets $\Delta H(0)$, indicating a high level of sample quality and the absence of two magnon scattering contributions to the damping.

Figure 7.6 illustrates the two main results of this work. First, $\alpha_{sp}$ is found to rapidly decrease from the 17Fe|50Pd|20Au value with the insertion of the Au spacer layer in the 17Fe|$d_{Au}$Au|50Pd|20Au structures. The 50Pd layer was thick enough that SFM might be
expected to behave as a spin sink [1]. If an SFM behaved like an ideal spin sink then a FM|NM|SFM structure would be analogous to FM1|NM|FM2. Surprisingly, however, a rapid decrease in damping was observed in the thickness range of Au, where the Au spacer thickness was substantially smaller than the spin diffusion length of \( \sim 290 \text{ AL (60 nm)} \) [124, 125]. In this thickness range, one could expect that the spin current generated at the Fe|Au interface is only affected by the spin relaxation process in the Au layer when changing \( d_{\text{Au}} \), and one would expect a more gradual decrease in \( \alpha_{\text{sp}} \) as shown by the upper line in Figure 7.6. Second, \( \alpha_{\text{sp}} \) exhibited an oscillatory like behaviour as a function of \( d_{\text{Au}} \). It is interesting to note that the spacing of the peaks is \( \sim 10 \text{ AL} \). This is very close to the long oscillatory period of the interlayer exchange coupling \( \sim 8.6 \text{ AL} \) in Fe|Au|Fe(001) structures [107, 109]. This is strong evidence that spin pumping is also affected by quantum well states in the Au layer.

### 7.4 Magnetic proximity effect

The first few atomic layers of Pd at the Fe|Pd interface have an induced ferromagnetism [130], a phenomenon known as the (magnetic) proximity effect. This leads to an effective ferromagnet of Fe + FM Pd. One might expect the change in the effective ferromagnetic layer to have a change in the effective damping field \( (\Delta H = \alpha \omega / \gamma) \) of the system of the form [38, 131],

\[
\frac{\alpha_{\text{tot}}}{\gamma_{\text{tot}}} = \frac{\alpha_{\text{Fe(Pd)}}}{\gamma_{\text{Fe}}} \frac{\mu_{\text{Fe}}}{\mu_{\text{Fe}} + \mu_{\text{Pd}}} + \frac{\alpha_{\text{Pd}}}{\gamma_{\text{Pd}}} \frac{\mu_{\text{Pd}}}{\mu_{\text{Fe}} + \mu_{\text{Pd}}},
\]

where \( \alpha_{\text{Fe(Pd)}} \) and \( \gamma_{\text{Fe(Pd)}} \) are the damping parameter and gyromagnetic ratio in the individual Fe and Pd layers and \( \mu_{\text{Fe(Pd)}} = d_{\text{Fe(Pd)}} M_{\text{Fe(Pd)}} \) is the total moment per unit area of each individual layer. Furthermore, one might expect a large \( \alpha_{\text{Pd}} \) owing to the larger spin orbit interaction in Pd compared to Fe.

To determine if the proximity effect could lead to additional damping in the ferromagnetic layer in our structures, the sample GaAs|17Fe|3Pd|20Au was prepared. This sample was found to have \( \alpha_{\text{tot}} = 3.58 \times 10^{-3} \), which is very close to that of the GaAs|17Fe|20Au samples \( (\alpha_{\text{tot}} = \alpha_{\text{bulk}} \equiv 3.56 \times 10^{-3}) \), see lower lines in Figure 7.5. Therefore, the ferromagnetic Pd on its own does not contribute noticeably to damping; in the 17Fe|\( d_{\text{Au}} \)Au|50Pd structure as compared to the 17Fe|50Pd system, the rapid suppression of \( \alpha_{\text{sp}} \) with the insertion of the Au layer cannot be attributed to the removal of magnetic proximity effect damping.
Figure 7.6: Main: Spin pumping induced damping $\alpha_{\text{sp}}$ multiplied by the Fe thickness for the samples GaAs(001)|17Fe|$d_{\text{Au}}$Au|50Pd|20Au as a function of the Au spacer layer thickness $d_{\text{Fe}}$. The upper line is calculated using spin diffusion theory for 17Fe|$d_{\text{Au}}$Au|FM (ideal spin sink) and the lower line for 17Fe|$d_{\text{Au}}$Au (perfect reflection at Au|vacuum). Two important effects can be seen: (1) $\alpha_{\text{sp}}$ rapidly decreases upon the insertion of the Au spacer layer. (2) An oscillatory behavior is observed in $\alpha_{\text{sp}}$ when $d_{\text{Au}} < 100$ AL. The dashed grey line is an eye guide bisecting the oscillatory data. **Top inset:** A closer view of the oscillatory dependence of $\alpha_{\text{sp}}$. **Bottom inset:** (●) The raw $\alpha_{\text{sp}}$ measurements as a function of $d_{\text{Au}}$ and (○) $d_{\text{Fe}}\alpha_{\text{sp}}/d_{\text{Fe}}^{\text{mean}}$ show that the oscillatory dependence is not a result of small deviations in $d_{\text{Fe}}$. 
7.5 Spin transport models for the Fe|Au|Pd

In this section the non-oscillatory dependence of $\alpha_{sp}$ will be modeled. Since the dependence of $\alpha_{sp}$ cannot be explained by treating the thick Pd layer as an ideal spin sink, the spin transport in Pd will be modeled using three different transport theories: spin diffusion in Section 7.5.1, modified spin decoherence in Section 7.5.2, and ballistic spin decoherence in Section 7.5.2. Spin diffusion theory is shown to not be valid in the Pd layer. However, the consequence of its application is still explored as this is quite common in the literature.

In the GaAs(001)|17Fe|d Au|50Pd|20Au system, the Au layer represents a simple NM; therefore the transport of spin current in the Au layer will be described using spin diffusion theory for all models. For the Au layer, $g_{↑↓} = 9.5 \times 10^{14} \text{ cm}^{-2}$ was determined from the difference in damping between the reference samples (17Fe|20Au) and the sample 17Fe|20Au|40Fe|20Au using Equations (6.28), (6.31) and (6.34). In the following analysis, the spin diffusion parameters used in the Au layer are $\tau_{m,\text{Au}} = 2.3 \times 10^{-14} \text{ s}$ and $\tau_{sf,\text{Au}} = 23.2 \times 10^{-14} \text{ s}$, in agreement with the values measured in refs. [125, 132]. The Fermi velocity in Au is taken to be $v_{F,\text{Au}} = 1.4 \times 10^8 \text{ cm/s}$.

As previously mentioned, the Pd layer represents a strongly spin correlated system with interactions of spin current with fluctuating paramagnons, referred to as a SFM. Therefore it is not clear which is the most appropriate model to describe spin transport in Pd layer. In the following discussion the Foros et al. [1] data on 16Fe|d Pd is fit using three models of spin transport: spin diffusion, modified spin decoherence, and ballistic spin decoherence, see Figure 7.7. The parameters determined by this analysis will then be used in the Pd layer of the 17Fe|d Au|50Pd system, with the Au parameters mentioned above describing the d Au Au layer.

7.5.1 Spin diffusion in Au and Pd

In order to apply the spin diffusion model for both the Au and Pd layers in the system 17Fe|d Au Au|50Pd, the spin diffusion parameters $\tau_{m,Pd}$ and $\tau_{sf,Pd}$ need to be determined for the Pd layer. Foros et al. [1] measured the sheet resistance of 50 AL thick sheet of Pd to be $\rho = 18.2 \mu\Omega \text{ cm}$, which allows the independent measurement of $\tau_{m,Pd}$ using the relation

$$\frac{1}{\rho} = \frac{e^2N}{m^*}\tau_{m},$$

(7.8)

where $e$ is the fundamental charge, $N$ is the carrier density, and $m^*$ is the effective mass of the electron. We take the values of the $\Gamma$-sheet as reported by Dye et al. [133]. The number of conduction electrons per Pd atom was found to be 0.3749, which results in the carrier density $N = 2.53 \times 10^{22} \text{ cm}^{-3}$. The effective electron mass was found to be $m^* = 2.05 \text{ m}$,
Figure 7.7: Fitting of $\alpha_{sp}$ data, originally publish by Foros et al. [1], in the system GaAs(001)$|16Fe| d_{pd}$Pd using spin diffusion theory (Dif$1,2,3,4$), modified spin decoherence theory (Dec$1$), and ballistic spin decoherence (Dec$2$). The fitted parameters for the data presented in this figure are listed in Table 7.1.
where \( m \) is the free electron mass. Using these values, one arrives at \( \tau_{m,Pd} = 1.58 \times 10^{-14} \) s. Using the Fermi velocity of Pd, \( v_{Pd} = 5.71 \times 10^7 \) cm/s (Γ sheet)[133], one finds the electron mean free path to be \( \lambda_{el,Pd} = v_{Pd} \times \tau_{m,Pd} = 9.02 \) nm.

**Dif1:** With \( \tau_{m,Pd} (= 1.58 \times 10^{-14} \) s) independently determined, one can fit the \( \alpha_{sp} \) data for 16Fe|d_{Pd}Pd of Foros et al. [1] as

\[
\alpha_{sp} = \frac{g\mu_B}{4\pi M_s} \tilde{g}_{\uparrow\downarrow} \frac{1}{d_{Fe}} \left[ \frac{1}{1 + \frac{v_{Pd}}{2} D_{Pd}\kappa_{Pd} \tanh(d_{Pd}\kappa_{Pd})} \right],
\]

which is given by inserting Equation (6.31) into Equation (6.28). The resulting fit is shown as Dif1 in Figure 7.7. The resulting fit parameters are \( \tau_{sf,Pd} = 1.58 \times 10^{-14} \) s and \( g_{\uparrow\downarrow} = 8.74 \times 10^{14} \) cm\(^{-2}\); the spin diffusion length is therefore \( \delta_{sd,Pd} = 1/\kappa_{Pd} = 5.6 \) nm.

The entirely spin diffusion model of spin transport in the 17Fe|d_{Au}Au|50Pd system gives the damping dependence

\[
\alpha_{sp} = \frac{g\mu_B}{4\pi M_s} \tilde{g}_{\uparrow\downarrow} \frac{1}{d_{Fe}} \left[ \frac{1}{1 + \Upsilon_{Au,Pd}} \right],
\]

where

\[
\Upsilon_{Au,Pd} = \frac{\frac{v_{Au}}{2} D_{Au}\kappa_{Au} + \frac{v_{Pd}}{2} D_{Pd}\kappa_{Pd} \tanh(d_{Pd}\kappa_{Pd}) \tanh(d_{Au}\kappa_{Au})}{D_{Au}\kappa_{Au} D_{Pd}\kappa_{Pd} \tanh(d_{Pd}\kappa_{Pd}) + (D_{Au}\kappa_{Au})^2 \tanh(d_{Au}\kappa_{Au})}.
\]

Equations (7.10) and (7.11) are obtained by inserting Equation (6.41) into Equation (6.28) with layer 1 = Au and layer 2 = Pd. With the parameters in the Pd layer in the previous paragraph and the previously mentioned Au parameters in the Au layer; the calculated dependence of \( \alpha_{sp} \) is shown in Figure 7.6 as Dif1.

Dif1 leads to \( \delta_{sd,Pd} < \lambda_{el,Pd} \) and \( \epsilon = 0.85 \). Interestingly, Dif1 qualitatively accounts for the rapid reduction of \( \alpha_{sp} \) and represents the \( d_{Au} > 200 \) AL data. However, for \( d_{Au} < 200 \) AL, Dif1 does not follow the general trend of the data and does not bisect the oscillatory behavior.

**Dif2:** One can estimate the consequence of considering \( \tau_{m,Pd} = 2.66 \times 10^{-14} \) s from the bulk value (\( \rho = 10.8 \) µΩ cm [134]) and fitting the Foros et al. [1] data, see Dif2 in Figure 7.7. The resulting spin diffusion fit parameters in the Pd layer are \( \tau_{sf,Pd} = 1.50 \times 10^{-14} \) s and \( g_{\uparrow\downarrow} = 7.51 \times 10^{14} \) cm\(^{-2}\); therefore, \( \delta_{sd,Pd} = 6.6 \) nm. The calculated dependence of \( \alpha_{sp} \) given by Equation (7.10) with the preceding parameters, is shown in Figure 7.6 as Dif2.

Using the bulk \( \tau_{m,Pd} \) estimate does not describe the 17Fe|d_{Au}Au|50Pd system well, agreeing in the asymptotic behavior for \( d_{Au} \geq 400 \) AL. However, it also quantitatively
shows a reduction in $\alpha_{sp}$ as compared to treating the SFM Pd as an ideal spin sink. Also in this case, one finds $\delta_{sd,Pd} < \lambda_{el,Pd}$ and $\epsilon = 1.77$.

**Dif3:** It is interesting to compare the values of the determined $g_{\uparrow\downarrow}$ for the Fe$|$Pd interface to that of an estimate using a spherical approximation for the Pd Fermi surface. Here we take the value of the $k$ vector of the $\Gamma$-centered sheet reported by Dye et al. [133], $k_{F,Pd(\Gamma)} = 1.01 \times 10^8$ cm$^{-1}$. Therefore we can use the estimation, see Equations (6.7) and (6.11),

$$g_{\uparrow\downarrow} = g_{Sh} = \frac{k_{F,Pd(\Gamma)}^2}{4\pi} = 8.14 \times 10^{14}$ cm$^{-2}$.

(7.12)

One can see that this value lies between the values for $g_{\uparrow\downarrow}$ shown in Table 7.1 under Dif1 and Dif3.

Furthermore, one can use this value of $g_{\uparrow\downarrow}$ for the Fe$|$Pd interface and fit the Foros et al. [1] data with the free parameters being $\tau_{m,Pd}$ and $\tau_{sf,Pd}$, see Figure 7.7. The resulting spin diffusion parameters are $\tau_{m,Pd} = 1.95 \times 10^{-14}$ s, $\tau_{sf,Pd} = 1.67 \times 10^{-14}$ s, $\epsilon = 1.17$, and $\delta_{sd,Pd} = 6.0$ nm. Using these parameters for the Pd layer in the 17Fe$|$d$_{Au}$Au$|$50Pd system leads to the $\alpha_{sp}$ dependence, given by Equation (7.10), shown as Dif3 in Figure 7.8. Dif3 describes the data when $d_{Au}$ is thicker than $\lambda_{el,Au}$ as well as bisects the oscillatory data.

![Figure 7.8: Modelling of $\alpha_{sp}$ data in the system 17Fe$|$d$_{Au}$Au$|$50Pd using spin diffusion theory (Dif1,2,3), modified spin decoherence theory (Dec1), and ballistic spin decoherence (Dec2) in Pd and spin diffusion in Au.](image-url)
Table 7.1: Pd layer parameters. The superscripts are as follows: $f$ is for values used as fitting parameters, $m$ is for values determined by separate measurements, and $d$ are values for parameters that can be defined in terms of other parameters. $\tau_{\text{dec},\text{Pd}} = \lambda_{\text{dec},\text{Pd}}/v_{\text{F},\text{Pd}}$ is defined here for comparison with the $\tau_{\text{sf},\text{Pd}}$ in the spin diffusion model. The entries for $\tau_{m,\text{Pd}}$ for the models Dec1 and Dec2 were taken from the Pd bulk conductivity because in this case the spin transport in Pd is assumed to be ballistic.

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<th>$g_{t\perp}^\uparrow$ [$10^{14}$ cm$^{-2}$]</th>
<th>$\tau_{m,\text{Pd}}$ [$10^{-14}$ s]</th>
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### 7.5.2 Hybrid spin diffusion (Au)/ modified spin decoherence (Pd) theory

Since $\epsilon \geq 0.85$ for all the Pd spin diffusion models, the application of spin diffusion theory in Pd is rather dubious, see Section 6.2.3. The transport of spin currents in the system Fe|Au|Pd can modeled as FM|NM1|SFM2, where the numbers refer to the layer number. The two theories available to describe the spin transport in the SFM layer are ballistic spin decoherence and modified spin decoherence. The idea of modified spin decoherence was to be able to introduce the idea of backflow from an SFM while not using truly diffusive spin current propagation, see Section 6.3.2. In this system, spin transport in NM1 is given by spin diffusion and in SFM2 by using a modified spin decoherence theory. The boundary condition for the net flow of spin current from NM1 into SFM2 at the NM1|SFM2 interface is

$$- D \frac{\partial}{\partial x} s_1(x) = \frac{1}{2} v_1 s_1(x) - \frac{1}{2} v_2 s_2(x) \bigg|_{x=d_1}, \quad (7.13)$$

where the second term on the RHS represents the backflow of spin current from the SFM2 side of the interface due to spin accumulation in SFM2. The net absorption of spin current by the SFM2 layer, Equation (6.48), can then be set equal to the net forward flow of spin current from the NM1 layer, Equation (7.13), allowing one to rewrite the boundary condition Equation (7.13) in terms of $v_1$ and $s_1$ only,

$$- D \frac{\partial}{\partial x} s_1(x) = \frac{\eta}{2} v_1 s_1(x) \bigg|_{x=d_1}, \quad (7.14)$$
where $\eta = 1 - 1/(2 - e^{-2d_2/\lambda_{dec,2}})$. This leads to $\alpha_{sp}$ of the FM1|NM1|SFM2 structure having dependence on the thickness of both NM1 and SFM2 with $\Upsilon$ in Equation (6.28) taking the form

$$\Upsilon_4 = \frac{\eta}{2} D_1 \kappa_1 + \eta \left( \frac{\eta}{2} \right)^2 \tanh (d_1 \kappa_1) \frac{(d_1 \kappa_1)}{\eta D_1 \kappa_1} + (D_1 \kappa_1)^2 \tanh (d_1 \kappa_1).$$ (7.15)

Note for $2d_2 \gg \lambda_{dec,2}$, boundary condition Equation (7.14) becomes independent on the material properties of SFM2. The spin pumping induced damping in the Fe|Au|Pd system is therefore

$$\alpha_{sp} = \frac{g\mu_B}{4\pi M_s} \frac{2}{d_{Fe}} \left[ 1 + \frac{e_{sp} D_{Au} \kappa_{Au} + \eta (e_{sp})^2 \tanh (d_{Au} \kappa_{Au})}{\eta e_{sp} D_{Au} \kappa_{Au} + (D_{Au} \kappa_{Au})^2 \tanh (d_{Au} \kappa_{Au})} \right]^{-1},$$ (7.16)

with $\eta = 1 - 1/(2 - e^{-2d_{Pd}/\lambda_{dec,Pd}})$.

Dec1: The modified spin decoherence parameters for Pd obtained by fitting the Foros et al. [1] data using

$$\alpha_{sp,mdc} = \frac{g\mu_B}{4\pi M_s} \frac{2}{d_{FM}} \left[ 1 - \frac{e^{-2d_{SFM}/\lambda_{dec}}}{2 - \eta} \right],$$ (7.17)

see Section 6.3.2. The parameters are $\lambda_{dec,Pd} = 9.4$ nm and $g\gamma = 9.6 \times 10^{14}$ cm$^{-2}$ [92]. This value of $\lambda_{dec,Pd}$ and the Au parameters (see above) are used in Equation (7.16) to determine $\alpha_{sp}$ for the hybrid spin diffusion/modified spin decoherence model. The resulting dependence of $\alpha_{sp}$ in $^{17}$Fe|$d_{Au}$Au|$50$Pd is plotted as Dec1 in Figure 7.6. Note that this curve both describes the data when $d_{Au}$ is thicker than the mean free path in Au, $\lambda_{el,Au} = 190$ AL (38 nm), as well as bisects the oscillatory data.

### 7.5.3 Hybrid spin diffusion (Au)/ballistic spin decoherence (Pd) theory

In the $^{17}$Fe|$d_{Au}$Au|$50$Pd system, one can treat the spin decoherence in Pd in the same manner as Foros et al. [1] by replacing the pumped spin current $I_{sp}'$ at the Fe|Pd interface by the spin current from Au to Pd at the Au|Pd interface. Using ballistic spin decoherence in the SFM Pd layer is similar to treating the Pd as an efficient, but not ideal, spin sink. In order to describe the data the forward spin current needs to be taken as a fraction of the maximum spin current which is available from Au given by $\frac{1}{2}v_{F,Au}s_{Au}(d_{Au})$, see Equations (6.15) and (6.16).

The boundary condition at the NM1|SFM2 interface is therefore similar to that of an ideal spin sink, Equation (6.32), but the net forward spin current is reduced by a material
dependent reflection parameter $R$,

$$
-D \frac{\partial}{\partial x} s_1(x) = (1 - R) \left( 1 - e^{-2d_2/\lambda_{\text{dec},2}} \right) \times \frac{1}{2} v_1 s_1(x) \bigg|_{x=d_1},
$$

(7.18)

where the parameter $R$ describes the reduction of the maximum forward spin current from NM1 to SFM2 due to reflection of spin current at the NM1|SFM2 interface, $0 \leq R \leq 1$. The term $\left( 1 - e^{-2d_2/\lambda_{\text{dec},2}} \right)$ is needed in order to account for the backward spin current due to reflection at the outer interface of the SFM2 layer, see Equation (6.44), and assure that for very thin SFM2 layers ($d_2 \ll \lambda_{\text{dec},2}$) no spin current absorption occurs in SFM2.

Boundary conditions Equations (6.24) and (7.18) lead to $\alpha_{sp}$ of the FM|NM1|SFM2 structure having dependence on the thickness of both NM1 and SFM2 with $\Upsilon$ in Equation (6.28) taking the form,

$$
\Upsilon_5 = \frac{\frac{v_1}{2} D_1 \kappa_1 + \eta \left( \frac{v_1}{2} \right)^2 \tanh (d_1 \kappa_1)}{\eta \frac{v_1}{2} D_1 \kappa_1 + (D_1 \kappa_1)^2 \tanh (d_1 \kappa_1)},
$$

(7.19)

where $\eta = (1 - R) \left( 1 - e^{-2d_2/\lambda_{\text{dec},2}} \right)$.

The spin pumping induced damping in the Fe|Au|Pd system is therefore

$$
\alpha_{sp} = \frac{g \mu_B}{4 \pi M_s} g_{\uparrow \downarrow} \frac{1}{d_{\text{Fe}}} \left[ 1 + \frac{\frac{v_{\text{Au}}}{2} D_{\text{Au}} \kappa_{\text{Au}} + \eta \left( \frac{v_{\text{Au}}}{2} \right)^2 \tanh (d_{\text{Au}} \kappa_{\text{Au}})}{\eta \frac{v_{\text{Au}}}{2} D_{\text{Au}} \kappa_{\text{Au}} + (D_{\text{Au}} \kappa_{\text{Au}})^2 \tanh (d_{\text{Au}} \kappa_{\text{Au}})} \right]^{-1},
$$

(7.20)

with $\eta = (1 - R) \left( 1 - e^{-2d_{\text{pd}}/\lambda_{\text{dec},\text{pd}}} \right)$.

**Dec2:** Using the ballistic spin decoherence model,

$$
\alpha_{sp,\text{dec}} = \frac{g \mu_B}{4 \pi M_s} g_{\uparrow \downarrow} \frac{1}{d_{\text{FM}}} \left( 1 - e^{-2d_{\text{FM}}/\lambda_{\text{dec}}} \right),
$$

(7.21)

to fit the data in Figure 7.7 leads to $\lambda_{\text{dec},\text{pd}} = 5.5$ nm. Using Equation (7.20) with a value of $R \approx 0.52$ reproduces the original dependence in ref. [135] (Dec1 in Figure 7.6). One can introduce the corresponding transmission parameter $T = 1 - R$. It is interesting to note that setting $T = v_{\text{pd}}/v_{\text{Au}} = 0.41$, which is 18% less than the value $T = 0.48(R = 0.52)$ that reproduces the trend in Dec1. This suggests that the reflection parameter $R$ could have its origin in the mismatch in Fermi velocities between the NM Au and the SFM Pd layer.

As shown in Section 7.4, the rapid suppression of $\alpha_{sp}$ with the insertion of the Au layer cannot be attributed to the removal of magnetic proximity effect damping. The models of entirely spin diffusion, hybrid spin diffusion/modified spin decoherence, and hybrid spin diffusion/ballistic spin decoherence can explain this rapid suppression of $\alpha_{sp}$ and describe
the general (nonoscillatory) behavior of the \textit{17Fe}\textit{d}_{\text{Au}}\textit{Au}\textit{50Pd} data well\(^1\), as can be seen in Figure 7.6.

In the model using only spin diffusion, the rapid reduction of \(\alpha_{\text{sp}}\) can be seen as a consequence of the backflow of accumulated spin density in Pd owing to the reduced Fermi velocity in Pd as compared to Au. Since the electronic processes are all much faster than the magnetization dynamics, the backflow current acts as an effective reflection. In the hybrid spin diffusion/modified spin decoherence model the reduction of \(\alpha_{\text{sp}}\) can also be seen as due to backflow from the Pd layer. However, it is questionable if the concept of backflow currents is applicable when modeling the spin current propagation as primarily ballistic. In the hybrid spin diffusion/ballistic spin decoherence, the reflection of spin current is explicit.

Using only spin diffusion has the benefit of being more easily applied to multilayers; however, the resulting values of the parameter \(\epsilon\) for Pd, as listed in Table 7.1, are all in the range where spin diffusion theory is not applicable, \(\epsilon > 3/4\). In fact for \(d_{\text{Pd}} > \delta_{\text{sd,Pd}}\) in the Fe\textit{Pd} structure, the parameters imply that Pd is an even better spin current absorber than an ideal spin sink, which is physically invalid.

The hybrid spin diffusion/modified spin decoherence model has the advantage of describing the data well in the Au\textit{Pd} system by simply applying values of the respective models in Au and Pd alone. Its disadvantage is that the backflow spin current due to spin accumulation from Pd does not depend on the material parameters of Pd when the Pd layer is sufficiently thick.

The hybrid model using spin diffusion/ballistic spin decoherence describes the spin transport in Pd in a more physically appropriate manner than the entirely spin diffusion model. Its has two advantages over the hybrid spin diffusion/modified spin decoherence model: 1) For \(d_{\text{Pd}} \gg \lambda_{\text{dec,Pd}}\), Pd acts as a spin sink, i.e. no accumulated spin density and backflow. However, it is not an ideal spin sink as not all of the spin current impinging on the Au\textit{Pd} interface is transmitted into Pd. 2) It contains the phenomenological parameter \(R\) describing the reduction of spin current transfer across the Au\textit{Pd} interface, which in principle could be interface/material dependent.

7.6 Discussion of quantum well states and \(\alpha_{\text{sp}}\)

There currently does not exist spin pumping or spin transport theories that account for the oscillatory dependence of \(\alpha_{\text{sp}}\) on the thickness of the normal metal (Au) spacer layer. It is not obvious that the confined geometry of an ultrathin NM must lead to quantum size

\(^1\) It is important to remember, as noted in the end of Section 6.2, that \(\alpha_{\text{sp}}\) for \(d_{\text{Au}} = 0\) data points in \textit{17Fe}\textit{d}_{\text{Au}}\textit{Au}\textit{50Pd} is not given by either the entirely spin diffusion models (Dif1,2,3,4) or the hybrid models (Dec1,2). This sample is \textit{17Fe}\textit{50Pd}, and \(\alpha_{\text{sp}}\) is given by Equation (6.46) (or Equations (6.28) and (6.31) if one wishes to apply spin diffusion theory for \(\epsilon \sim 1\)
effects in spin pumping, considering that this is an irreversible effect. However, quantum size effects of ultrathin FM were predicted to lead to oscillations in the spin pumping efficiency in NM|FM|NM structures with changing FM thickness by both Mills [136] and Šimánek [137] using the dynamic version of the Ruderman-Kittel-Kasuya-Yosida (RKKY) theory of ref. [79]. Both of these theories, however, considered semi-infinite NMs adjacent to the precessing ferromagnet and thus did not have any insights into quantum size effects in the NM. However, Zwierzycki et al. [90] later showed how this effect would only be notable for extremely thin FMs, where the thickness was comparable to the transverse spin decoherence length of a few atomic layers. No oscillations were so far predicted with changing NM thickness, nor had they previously been observed experimentally.

At this point, one can only speculate on the origin of the oscillatory behavior of $\alpha_{sp}$ in the present study. One of the possible candidates is that $g_{\uparrow\downarrow}$ is modulated with $d_{Au}$ as $g_{\uparrow\downarrow}$ is given explicitly by spin dependent reflectivity. One possible explanation is that the terms $r_{\uparrow,nn}(r_{\downarrow,nn})^*$ in Equation (6.5) do fully cancel out in the presence of quantum well states, which would lead to a modulation of $g_{\uparrow\downarrow}$. However, other factors that could be influenced are the Sharvin conductance governing the spin current backflow the accumulated spin density $S$ as it depends on the density of states at the Fermi level in the NM. It is also possible that the reflection of spin current at the Au|Pd interface is affected by the quantum well states.

It is worth discussing the possible reasons why such dependence has not yet been observed in simple FM|NM or FM1|NM|FM2 structures. For the case of the FM|NM structures the magnitude of $\alpha_{sp}$ is very small when the thickness of the NM is much less than the spin diffusion length. For example, one can consider the Fe|Au system. In the $d_{Au} < 50$ AL region, where the oscillatory behavior is prominent, the total modulation in $\alpha_{sp}$ is $\sim 20\%$ around the dashed line in Figure 7.6. In comparison to the Fe|Au|Pd structure, $\alpha_{sp}$ (lower line in Figure 7.6) in Fe|Au structures is 10 times smaller in this region. Assuming a similar modulation of $\alpha_{sp}$ ($\sim 20\%$) to that in the Fe|Au|Pd case, the oscillations would be 10 times smaller than those observed in our studies; the effect would be comparable to our error bars. This illustrates why such behavior has not been observed in the Fe|Au structure where there is a perfect spin current reflection at the Au|vacuum interface. Therefore, it seems that the partial reflection at the Au|Pd interface was not the only factor allowing us to observe oscillations in $\alpha_{sp}$. In addition, one needs a large enough spin current absorption in the corresponding spin pumping structure. For this reason it may also be challenging to use FM|NM1|NM2 structures where both NM1 and NM2 are simple NMs. The case of FM1|NM|FM2 is challenging for bringing up an argument that would support the presence of oscillations in spin pumping damping. In this system, there is no reflection of the
spin current at the NM|FM2 interface. Therefore the presence of oscillations would require to have the spin pumping parameter $g_{\uparrow\downarrow}$ to be affected directly by changing the electronic states in NM by the presence of quantum well states. This would be similar to the interlayer exchange coupling which depends on passing the quantum well states in NM through the Fermi level. However, if $g_{\uparrow\downarrow}$ is directly modulated by the quantum well states in NM, one would expect to find similar behavior as observed in this study. One reason such oscillatory behavior of $\alpha_{sp}$ has not been seen in this case is that the vast majority of measurements in the literature are using the FM|NM|FM2 structures to determine the spin diffusion length in the NM and for practical reasons the thickness steps in the NM are much larger than in this study. Careful studies with small thickness steps in NM in FM|NM1|FM2 structures will be needed to narrow down the origin of the oscillations.

7.7 Summary

Spin pumping in the GaAs(001)|17Fe|$d_{\text{Au}}$Au|50Pd|20Au system, where $d_{\text{Au}}$ is the thickness of the Au spacer in atomic layers, was studied by means of ferromagnetic resonance. 50 Pd atomic layers is the thickness at which spin decoherence nearly saturates and the Pd layer acts as an efficient spin sink when in direct contact with Fe. The spin pumping contribution to interface damping $\alpha_{sp}$ exhibits two interesting behaviors. 1) $\alpha_{sp}$ rapidly reduces with the introduction of the Au spacer layer and reaches an asymptotic behavior with a constant value equivalent to that of both the 17Fe|$d_{\text{Au}}$Au and 17Fe|$d_{\text{Au}}$Au|40Fe. 2) In the thickness range where the mean free path inside the Au layer is larger than the Au layer thickness $d_{\text{Au}}$, $\alpha_{sp}$ shows oscillatory behavior with an apparent periodicity that is close to the long wavelength periodicity in Au(001) of the static interlayer exchange coupling (Fermi surface spanning $k$-vectors). Additionally, it was shown experimentally that the proximity effect at the Fe|Pd interface plays at most only a very minor role in Gilbert damping; therefore, the removal of proximity effect induced damping at the Fe|Pd interface cannot be the cause of the rapid reduction in the damping due to the addition of the Au layer in the 17Fe|$d_{\text{Au}}$Au|50Pd structures.

Spin transport in the 17Fe|$d_{\text{Au}}$Au|50Pd structures was analyzed using three different models: spin diffusion to both the Au and Pd layers, hybrid spin diffusion (Au)/modified spin decoherence(Pd), and hybrid spin diffusion (Au)/ballistic spin decoherence(Pd). All the models were able to account for the rapid decrease in $\alpha_{sp}$ by inserting a few atomic layers of Au as well as bisecting oscillatory behavior of $\alpha_{sp}$. Spin diffusion in Pd requires a very short spin flip relaxation time; in fact, the parameter $\epsilon (= \tau_m/\tau_{sf}) > 3/4$. This would imply that Pd is an even better spin current absorber than an ideal spin sink, which is physically invalid. The spin diffusion model for materials with $\epsilon > 3/4$ is a mathematical
construct and one should realize that it is not physically meaningful. The hybrid spin diffusion/modified spin decoherence model resulted in the strange conclusion that it was independent of the Pd material parameters when Pd was sufficiently thick. The modified spin decoherence theory is an attempt to accommodate the spin accumulation in the SFM, but allow the dephasing of spin currents to happen on a shorter length scale than spin diffusion. However, the concept of spin accumulation is usually introduced in the presence of a diffusive spin scatterer. The hybrid spin diffusion (Au)/ballistic spin decoherence (Pd) model assumes fully ballistic spin transport in the Pd layer (no accumulated spin density in Pd) and it allows the introduction of the material dependent phenomenological parameter $R$ describing the reduction of spin current transfer across the Au|Pd interface. This model treats Pd as an efficient spin sink for spin currents transmitted across the Au|Pd interface entering the Pd layer.

The presence of the oscillatory behavior of $\alpha_{\text{sp}}$ represents the fundamentally new result of this work. We have observed the presence of quantum size effects affecting the time irreversible spin pumping induced damping process for the first time. The observed oscillatory dependence of $\alpha_{\text{sp}}$ is not within the phenomenological models presented or any other known theoretical models of spin pumping. However, the experimental evidence of such behavior should be of interest to theorists in the field and its treatment would further advance the understanding of spin pumping and spin transport in heterogeneous structures.
Chapter 8

Spin pumping from the ferrimagnetic insulator Yttrium Iron Garnet (YIG)

In this chapter, spin pumping from the ferrimagnetic insulator Yttrium Iron Garnet (YIG, Y\(_3\)Fe\(_5\)O\(_{12}\)) into Au|Fe|Au structures will be shown. Spin injection across the ferrimagnetic insulator (YIG)|normal metal (Au) interface was studied using ferromagnetic resonance (FMR). First, the spin mixing conductances \(g_{↑↓}\) for untreated YIG samples were determined by comparing the Gilbert damping of bare (as-grown) YIG films with those covered by a Au|Fe|Au structure. The Gilbert damping was determined from the slope of the FMR linewidth as a function of microwave frequency. The Fe layers in YIG|Au|Fe|Au structures acted as spin sinks, as displayed by an increased Gilbert damping parameter \(\alpha\) compared to that in the bare YIG. Next, the surface of the YIG films was treated with Ar\(^+\) sputtering and H\(^-\) cleaning. The spin pumping efficiency is increased by Ar\(^+\) sputtering to a limit, while H\(^-\) cleaning results in a complete suppression of spin pumping.

8.1 Motivation

Ferromagnetic insulators (FI) are attractive for spin current related experiments and devices as conventional charge currents cannot propagate in insulators. Yttrium Iron Garnet (YIG, Y\(_3\)Fe\(_5\)O\(_{12}\)) is particularly attractive as it has the lowest intrinsic damping of all known ferromagnets with \(\alpha \simeq 3 \times 10^{-5}\), which is two orders of magnitude lower than Fe. Due to this extremely small damping, YIG materials have been used in a broad range of microwave device applications [138]. Furthermore, YIG materials are highly attractive for potential use
in future spintronic applications, such as the generation of large spin currents and spinwave based devices (magnonics) [139].

Additionally, a newly emerging field called spin caloritronics addresses the creation of spin currents by thermal gradients [140, 141]. In 2010, Xiao et al. [7] described the theory of a magnon driven spin Seebeck effect. If there is thermal gradient in the lattice temperature in the FM then the magnon temperature is different from the local lattice temperature. In fact, it equilibrates between the highest and lowest temperature of the lattice gradient. It is shown that magnon temperature is nearly constant [7] and therefore is lower at the highest temperature end and is higher at the lowest temperature end of the FM. If a NM is adjacent to a region where the magnon temperature is higher than the lattice temperature, then the magnetic system can pump a spin current into the adjacent NM to lower the effective magnon temperature. In fact, if the direction of the thermal gradient is reversed, so is the spin current; this is a unique feature which is not possible with ferromagnetic resonance based spin pumping. Uchida et al. [8] showed that spin current could be produced using the spin Seebeck effect in ferrimagnetic insulator LaY$_2$Fe$_5$O$_{12}$ films in LaY$_2$Fe$_5$O$_{12}$|Pt heterostructures. Although in this study, the spin current was detected using the inverse spin Hall effect and quantitative data about the spin pumping efficiency was not reported. Slonczewski [142] has predicted that higher efficiency in spin transfer torque devices can be achieved using ferromagnetic insulator (FI)|normal metal (NM) structures.

Ferromagnetic resonance is one of the best tools to determine the spin pumping efficiency, governed by the spin mixing conductance. At the beginning of these studies, it was still debated whether one could use FMR to induce spin pumping at a FI|NM interface. However, when one considers that the precessional motion of the magnetization reduces the magnetization component along the equilibrium direction, one realizes that FMR effectively increases the magnon temperature and therefore spin pumping should occur. Sandweg et al. [143] first showed that one could spin pump using microwave excitation in YIG|Pt structures. The studies were performed at a single frequency and the spin pumping effect was determined using the inverse spin Hall effect. However, similar to the studies of Uchida et al. [8], the spin pumping efficiency was not given quantitatively. Jia et al. [12] showed theoretically that the spin pumping efficiency of YIG is still determined by the real part of the spin mixing conductance. The main motivation of the studies presented in this chapter was to identify quantitatively the spin mixing conductance at the YIG|Au interface.

8.2 Yttrium-Iron-Garnet (YIG) sample preparation

Yttrium-Iron-Garnet belongs to the family of ferrimagnets. While the garnet structure is bcc cubic, the unit cell is quite complex [138, 144]. YIG has a lattice constant of $a_{\text{YIG}} =$
12.376 ± 0.004 Å [145]. Each unit cell contains 8 chemical formula units \((Y_3Fe_5O_{12})[138]\). Per chemical formula unit, three of the \(Fe^{3+}\) ions occupy tetrahedral sites and two occupy octahedral cites, while the \(Y^{3+}\) ions occupy dodecahedral sites. The \(Y^{3+}\) has no magnetic moment as it is \(4d^0\). Each \(Fe^{3+}\) provides a magnetic moment of \(5 \mu_B\) as it is \(3d^5\). However, the tetrahedral sites and octahedral sites are antiferromagnetically coupled across the O, resulting in a net moment of \(5 \mu_B\) per chemical formula or \(40 \mu_B\) per unit cell.

The growth of the YIG films in this study was performed at Colorado State University by our collaborators Young-Yeal Song, Yiyan Sun, and Prof. Mingzhong Wu. YIG \((Y_3Fe_5O_{12})\) films with thicknesses of 9.0 and 5.0 nm were grown on single crystal Gadolinium Gallium Garnet \((Gd_3Ga_5O_{12}(1 1 1), \text{GGG})\) substrates by means of pulsed laser deposition (PLD). GGG has a lattice constant of \(a_{\text{GGG}} = 12.3829 \pm 0.0002 \text{ Å [146]}.\) The deposition was performed in high purity oxygen with the substrate at 790° C and the oxygen pressure held at 0.1 Torr. Right after the deposition, the YIG film was then annealed at the same temperature and oxygen pressure for 10 minutes. The thickness of the YIG films was determined by low angle X-ray diffraction. The saturation induction was determined by SQUID magnetometry and was found to be \(4\pi M_s = 1.31 \text{ kG}.\) The surface roughness of the YIG films as measured by atomic force microscopy (AFM) was 0.5 nm. The low angle X-ray diffraction, SQUID magnetometry, and AFM studies were also performed by our collaborators at Colorado State University.

### 8.3 Untreated YIG samples

In the SFU UHV system, the YIG films were characterized by X-ray photoelectron spectroscopy (XPS) and the Au and Fe films were deposited by molecular beam epitaxy (MBE) at pressures in the low \(10^{-10}\) Torr. The experiments on untreated (as-grown) YIG films involved 3 samples. UT1: 9.0YIG|2.0Au|4.3Fe|6.1Au, UT2: 9.0YIG|6.1Au|4.3Fe|6.1Au and UT3: 5.0YIG|6.1Au|2.9Fe|4.1Au, with the numbers indicating film thickness in nm. The XPS spectra indicated Fe was deficient at the YIG surface. The atomic ratio Fe/Y for both samples was only \(\simeq 0.6\), while according to stoichiometry it is expected to be \(1.7\). The atomic ratio O/Y was found to be 4 and 6 for for the samples UT1 and UT2, respectively. The expected ratio by the chemical formula is 4. This indicates that the oxygen concentration in S2 was higher than that in S1. The difference in chemical composition at the YIG surface compared to its bulk is caused by the surface chemistry during PLD and it is similar to thick YIG films. The Au and Fe films were polycrystalline.

FMR measurements were carried out in the in-plane configuration, see Section 3.3.1, in microwave cavities at frequencies \(f \simeq 10, 14, 24, \text{ and } 36 \text{ GHz}\) using an Anritsu microwave generator. External field modulation was used for lock-in detection of the FMR signal. The
samples inserted into the microwave cavities were approximately 3×4 mm$^2$. As-grown YIG films were well fit by two closely separated FMR lines, see Figure 8.1. This indicates that the as-grown films consisted of two regions having slightly different magnetic properties. The relative area (RA) each region occupies on the sample is given from the fitted amplitude $\mathcal{A}$ of each FMR line, see Equation (3.57) in Section 3.4.

The FMR linewidth (half width at half maximum) was described by Gilbert like damping,

$$\Delta H(\omega) = \frac{\omega}{\gamma} + \Delta H(0),$$

(8.1)

where $\omega (= 2\pi f)$ is the microwave angular frequency, $\gamma (= g\mu_B/h)$ is the absolute value of the gyromagnetic ratio ( $g$ is the Landé g-factor in the YIG film and $\mu_B$ is the Bohr magneton), and $\Delta H(0)$ is the zero frequency line broadening due to long range magnetic inhomogeneities [38, 44], see figs. 8.2 and 8.3. The YIG(111) films showed negligible in-plane anisotropy. Therefore, the effective perpendicular demagnetizing field $4\pi M_{\text{eff}}$ and the Landé g-factor in the YIG films were determined from the frequency dependence of the ferromagnetic resonance field, $H_{\text{FMR}}(f)$, using the ferromagnetic resonance condition,

$$H_{\text{FMR}} = \frac{-4\pi M_{\text{eff}} + \left((4\pi M_{\text{eff}})^2 + 4 \left(\frac{\omega}{\gamma}\right)^2\right)^{\frac{1}{2}}}{2},$$

(8.2)

see Equation (3.40). The Landé g-factor was found to be $g = 2.02 \pm 0.01$ for all the YIG films. $4\pi M_{\text{eff}}$ was found to be 1.89, 1.97, and 1.69 kOe for samples UT1, UT2, and UT3, respectively. For the 4.3Fe film, $4\pi M_{\text{eff}} = 10.6$ kOe.

Considering the structure FI|NM|FM, with small precessional angles of the magnetic moment in FI during FMR, the pumped magnetic moment is almost entirely transverse to the static magnetic moment and the FM layer will act as an ideal sink [121]. In YIG|Au|Fe|Au magnetic double layer structures, the FMR fields corresponding to the YIG and Fe films are separated by several kOe due to a large difference in $4\pi M_{\text{eff}}$. In this situation, the YIG and Fe films do not undergo FMR at the same field for a given frequency and are not involved at the same time in interchanging spin currents [75]. When the Au films covering YIG are much thinner than the spin diffusion length in Au ($\simeq 60$ nm [124, 125]), to a good approximation, neglect the loss of accumulated spin momentum in the Au layer. The spin current generated at the YIG|Au interface leads to an increased Gilbert damping in YIG, and in this approximation is given by

$$\alpha_{sp} = \frac{g\mu_B}{4\pi M_s} g \uparrow \downarrow \frac{1}{d_{\text{YIG}}},$$

(8.3)
where \( g_{\uparrow\downarrow} \) is the spin mixing conductance at the YIG|Au interface and \( d_{\text{YIG}} \) is the thickness of the YIG film, see Sections 6.1.2 and 6.2.2 for further details.

Spin pumping was measured in samples UT1, UT2, and UT3. The Gilbert damping contribution to the FMR linewidth in YIG, \( \alpha \) in Equation (8.1), had two contributions: (a) the bulk contribution, \( \alpha_{\text{bulk}} \) as measured in bare (UT1,UT2) or Au covered (UT3) YIG films and (b) the interface spin pumping contribution \( \alpha_{\text{sp}} \) given by Equation (8.3). The spin pumping contribution is therefore given as

\[
\alpha_{\text{sp}} = \alpha - \alpha_{\text{bulk}}. \tag{8.4}
\]

Figure 8.1: Example FMR data for the sample UT1: 9.0YIG|2.0Au|4.3Fe|6.1Au and corresponding bare YIG film. (a) \( f = 14.133 \) GHz, bare 9.0YIG: The sample had two closely spaced FMR peaks as shown in Figure 8.1 (a). After the deposition of 2.0Au|4.3Fe|6.1Au on 9.0YIG resulted in splitting the original two closely spaced FMR peaks into three well separated FMR absorption lines, see Figure 8.1 (b). This means that the 2.0 nm Au spacer was thin enough that the YIG and Fe layers were coupled by interlayer exchange coupling \( J_{\text{ex}} \). From the relative areas of the peaks listed in Figure 8.1, it is most likely that area 2 in the bare YIG split into areas 2 and
3 for UT1. The peak at the highest field remained very near of the FMR field for the bare YIG, but the other two peaks became visibly down shifted to lower resonance fields. These shifts in the FMR line indicated the presence of magnetic coupling. The layers are coupled by ferromagnetic interlayer exchange coupling $J_{ex} = + (0.08$ and $0.15) \text{ erg/cm}^2$ for the area (1) and (2), respectively. Even more importantly, the slope of the FMR linewidth as a function of microwave frequency, $\Delta H(f)$, increased for the coupled samples while the uncoupled sample showed no increase in this slope within measurement error, see Table 8.1 for damping parameters. This is in perfect agreement with an alternative theory of spin pumping that was proposed by Šimánek and Heinrich [79]. They have shown that spin pumping has its origin in the time retarded response of the interlayer exchange coupling, which is caused by $sd$-type interface exchange. In fact, Figs 8.1 and fig. 8.2 represent the first clear experimental demonstration of this relationship between the static interlayer exchange coupling and the spin pumping mechanism.

The sample UT2 (9.0YIG|6.1Au|4.3Fe|6.1Au) did not show any appreciable shift in the FMR field compared to that in the bare YIG; therefore the static interlayer exchange coupling for the 6.1Au spacer is negligible. However the slope of $\Delta H(f)$ clearly increased compared to that in the bare YIG, indicating that a spin pumping contribution to the damping is present, see fig. 8.3 (a) and Table 8.1. The sample UT2 showed 2 FMR lines in
Table 8.1: Magnetic parameters for the samples UT1: 9.0YIG|2.0Au|4.3Fe|6.1Au, UT2: 9.0YIG|6.1Au|4.3Fe|6.1Au, and UT3: 5.0YIG|6.1Au|2.9Fe|4.1Au and the corresponding bare (Au capped for UT3) YIG films.

<table>
<thead>
<tr>
<th>sample</th>
<th>$g_{↑↓}$</th>
<th>$\alpha_{sp}$</th>
<th>$\alpha$</th>
<th>$\Delta H (0)$</th>
<th>$\alpha_{bulk}$</th>
<th>$\Delta H_{bulk} (0)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>UT1 area (1)</td>
<td>1.2 ± 0.5</td>
<td>1.9 ± 0.8</td>
<td>2.2 ± 0.8</td>
<td>25 ± 7</td>
<td>0.3 ± 0.1</td>
<td>5 ± 1</td>
</tr>
<tr>
<td>UT1 area (2)</td>
<td>0.8 ± 0.2</td>
<td>1.4 ± 0.2</td>
<td>1.6 ± 0.2</td>
<td>16 ± 2</td>
<td>0.2 ± 0.1</td>
<td>7 ± 1</td>
</tr>
<tr>
<td>UT2</td>
<td>1.1 ± 0.3</td>
<td>1.7 ± 0.5</td>
<td>2.4 ± 0.4</td>
<td>4 ± 3</td>
<td>0.7 ± 0.2</td>
<td>8 ± 2</td>
</tr>
<tr>
<td>UT3</td>
<td>1.4 ± 0.2</td>
<td>4.1 ± 0.4</td>
<td>4.7 ± 0.4</td>
<td>13 ± 3</td>
<td>0.6 ± 0.1</td>
<td>15 ± 1</td>
</tr>
</tbody>
</table>

both the bare YIG and the full structures. The portion with the larger relative area was found to have $g_{↑↓} = 1.1 (±0.3) \times 10^{14}\text{cm}^{-2}$, in agreement with areas 1 of UT1.

For the sample UT3 (5.0YIG|6.1Au|2.9Fe|4.1Au), FMR measurements were made on the bare YIG as well as following the growth of 4.3Au. The 5.0YIG/4.3Au sample was then placed back into the UHV system where a small oxidation of Au was removed by Ar$^+$ sputtering under oblique incidence. The remaining structure was then deposited and FMR measurements were made on the entire structure. The sample UT3 also displayed 2 FMR lines in all the measurements. The spin pumping contribution to the damping $\alpha_{sp}$ in UT3 was significantly greater than in UT1 and UT2, see Table 8.1. However, this is expected according to Equation (8.3) as $d_{YIG}$ was much smaller. The portion with the larger relative area (0.8) was found to have $g_{↑↓} = 1.4 (±0.2) \times 10^{14}\text{cm}^{-2}$, in agreement with the results carried out using the 9.0YIG films, UT1 and UT2, showing that spin pumping from YIG is indeed an interface effect.

The spin mixing conductance at the YIG|Au interface observed for the untreated YIG films $g_{↑↓} \simeq 1 \times 10^{14}\text{cm}^{-2}$ is one order of magnitude smaller than the spin mixing conductance typically observed at the FM|NM interface $g_{↑↓} \simeq 1 \times 10^{15}\text{cm}^{-2}$. Theoretical predications of Jia et al. [12] suggest that the spin mixing conductance could be larger for an ideal YIG interface, $g_{↑↓} \simeq 7 \times 10^{14}\text{cm}^{-2}$. However, these predictions are for YIG films without the Fe deficiency at the interface observed in these samples.

### 8.4 Surface treated YIG samples

As the spin pumping mechanism is an interface effect, it is interesting to see if the spin pumping efficiency can be enhanced by means of surface treatment. In this section the surface treatment of the YIG films by Ar$^+$ (sputter) etching and atomic hydrogen (H·) etching is explored. The same FMR analysis was performed on the surface treated YIG
Figure 8.3: FMR linewidth as a function of microwave frequency for: (a) The sample UT2: 9.0YIG/6.1Au/4.3Fe/6.1Au (●) and the corresponding bare YIG film (○). (b) The sample UT3: 5.0YIG/6.1Au/4.3Fe/6.1Au (●), 5.0YIG/4.3Au (○), and the bare (as-grown) 5.0YIG (■). The deposition of 4.3Au on the 5.0YIG film had negligible effect on the damping on 5.0YIG film.
samples as for the untreated YIG samples above in Section 8.3. For these samples, the GGG|YIG wafer was cut into smaller pieces prior to the surface treatment and deposition of the Au|Fe|Au overlayers. This was done as we had limited YIG samples and wanted to allow a number of surface treatments to be performed. The sample was attached to the sample holder by a Mo wire clip. The sample under the clip was Ar\(^+\) etched by using grazing incidence of sputtering and rotating the substrate. After Ar\(^+\) etching, the 6.1Au|4.3Fe|6.1Au structure was deposited on the samples. During Au and Fe deposition the clip covered a small portion the sputtered area as the deposition beams are nearly perpendicular to the sample face and the sample holder did not rotate.

XPS spectra were used as a supplemental tool to qualitatively evaluate the chemical changes of the YIG surface under the various surface treatments. XPS analysis was done following the method presented by Šimša and Zemek [147]. However, as we did not have calibration samples for the individual Fe states, the effects of asymmetry and shake up modes are neglected. The main 1s O peak corresponds to the O\(^{2-}\) chemical state and is defined to have a binding energy at 530.1 eV. Due to sample charging, the XPS spectra shifted in energy. To account for the charging, the XPS spectra for each sample were shifted to have the main oxygen peak at 530.1 eV as Šimša and Zemek [147] showed that the O\(^{2-}\) line is the most stable in position and can be well taken as a reference energy for other spectral lines in similar garnets including YIG. The analysis of the Fe line is done in a very simple model and is intended to give qualitative information. The binding energies of the 2p\(_{3/2}\) Fe lines in the garnet are 707, 709, 711, 712.8 eV for metallic Fe, Fe\(^{2+}\), Fe\(^{3+}\), and Fe\(^{4+}\) respectively [147]; the fitted Fe valence peaks are constrained to \(\pm 0.5\) eV from these positions such that peak positions do not overlap in range.

The first surface treatment performed was 0.8 kV Ar\(^+\) etching for 10 min at 400\(^\circ\) C on the as-grown YIG sample. The sample is labelled ST1 (9.0YIG/6.1Au/4.3Fe/6.1Au) in Figures 8.4 and 8.5 and table 8.2. XPS spectra of ST1 for the 1s O and 2p\(_{3/2}\) Fe XPS lines for the as-grown YIG surface are shown in Figure 8.5 (a) and (d), respectively, and after surface treatment in Figure 8.5 (b) and (e). The as-grown YIG films show two 1s O lines, the dominant O\(^{2-}\) at 530.1 eV and a second line with binding energy near 532 eV, which the authors of ref. [147] attribute to adsorbed hydroxyl species OH\(^-\). With Ar\(^+\) etching, the 532 eV line is reduced in intensity. The 2p\(_{3/2}\) Fe XPS spectra also show two lines corresponding to Fe\(^{3+}\) and Fe\(^{4+}\) with concentrations at the ratio of Fe\(^{3+}/Fe^{4+}\) increasing after the treatment. The 3 d\(_{5/2}\) Y line was found near 158 eV (for all samples). In the following the relative surface concentrations for Y, Fe, and O will be quoted as Y\(_x\)Fe\(_y\)O\(_z\) normalized such that \(x + y + z = 20\) for comparison to stoichiometry in the bulk. For ST1 before treatment, the surface concentration were found to be Y\(_{2.8}\)Fe\(_{1.7}\)O\(_{15.5}\) compared the
expected bulk values of $Y_3Fe_5O_{12}$, indicating the surface is Fe deficient and O rich. After Ar$^+$ etching, the surface stoichiometry was found to be $Y_{3.1}Fe_{2.0}O_{14.9}$, showing that both the Fe deficiency and O richness were reduced.

Figure 8.4 (a) shows the 35.9926 GHz FMR spectrum for ST1. Two FMR peaks can be seen, one wider peak corresponding to the full 9.0YIG/6.1Au/4.3Fe/6.1Au structure and one narrower line corresponding to the surface treated YIG film only (portion masked by the clip). The frequency dependence of the the linewidth $\Delta H (f)$ is shown in Figure 8.4 (b) for ST1 (●) and the corresponding YIG only portion (○). The damping in the full structure was found to be $\alpha^{ST1}_{\text{bulk}} = 6.5 (\pm 0.1) \times 10^{-3}$. The damping for the uncovered portion was $\alpha^{ST1}_{\text{sp}} = 0.4 (\pm 0.3) \times 10^{-3}$, which is similar to the uncovered portions for the untreated samples, see Table 8.1. Therefore, the spin pumping induced damping is $\alpha^{ST1}_{\text{sp}} = 6.1 (\pm 0.3) \times 10^{-3}$. Using Equation (8.3), the spin mixing conductance is found to be $g^{ST1}_{\uparrow\downarrow} = 3.8 (\pm 0.2) \times 10^{14}$ cm$^{-2}$, which is a significant increase from the untreated samples.

For the sample ST2 (9.0YIG/6.1Au/4.3Fe/6.1Au), the Ar$^+$ etching energy was increased to 1.0 kV and the time was increased to 22 minutes at a temperature of 400$^\circ$C. The XPS spectra for the 1s O and 2p$_{3/2}$ Fe lines after are Ar$^+$ etching are shown in Figure 8.5 (c) and (e), respectively. For this treatment, the 532 eV 1s O line has been dramatically reduced. The 2p$_{3/2}$ Fe lines are shown in Figure 8.5 (f). The 2p$_{3/2}$ Fe XPS spectra contains the two lines corresponding to Fe$^{3+}$ and Fe$^{4+}$ with concentrations with the ratio Fe$^{3+}$/Fe$^{4+}$ increasing more than for ST1; however, a small peak corresponding to metallic Fe at 707 eV has emerged. After Ar$^+$ etching the surface concentrations were found to be $Y_{3.0}Fe_{2.4}O_{14.6}$, showing a further increase in Fe compared to ST1. $\Delta H (f)$ is shown in Figure 8.4 (b) for ST2 (■). The damping for the full structure was found to be $\alpha^{ST2}_{\text{bulk}} = 6.4 (\pm 0.3) \times 10^{-3}$ and for the uncovered portion was $\alpha^{ST2}_{\text{sp}} = 0.2 (\pm 0.1) \times 10^{-3}$, and therefore $\alpha^{ST1}_{\text{sp}} = 6.2 (\pm 0.3) \times 10^{-3}$. The spin mixing conductance, $g^{ST2}_{\uparrow\downarrow} = 3.9 (\pm 0.2) \times 10^{14}$ cm$^{-2}$, is found in agreement with ST1.

For the sample ST3 (9.0YIG/6.1Au/4.3Fe/6.1Au), the Ar$^+$ etching energy was 1.0 kV and the time was increased further to 32 minutes at 400$^\circ$C. The 1s O spectra was similar to that for ST2. The 2p$_{3/2}$ Fe spectrum showed a further increase in the metallic Fe concentration, as shown in Figure 8.5 (g). The surface concentrations after Ar$^+$ etching were $Y_{3.6}Fe_{2.3}O_{14.1}$, showing a similar Fe concentration as ST1 and ST2 and a large increase in relative Y concentration. $\Delta H (f)$ is shown in Figure 8.4 (b) for ST3 (▲). This sample showed a large increase in $\Delta H (0)$, indicating that longer Ar$^+$ treatment times lead to a reduction in sample quality. In fact, $\Delta H (0)$ increased with increasing Ar$^+$ treatment for all the samples ST1-ST3, see Table 8.2. The damping for the full structure was found to be $\alpha^{ST3} = 3.9 (\pm 0.9) \times 10^{-3}$. The damping for the uncovered portion increased, $\alpha^{ST3}_{\text{sp}} =
Figure 8.4: (a) 35.9926 GHz FMR data for the samples ST1: 9.0YIG/6.1Au/4.3Fe/6.1Au. (b) FMR linewidth as a function of frequency for ST1 (●) and the corresponding bare film (○), ST2 (■), and ST3 (▲).

1.3 (±0.4) × 10⁻³, showing that longer Ar⁺ etching lowered the quality of the YIG. The spin pumping induced damping $\alpha_{\text{sp}}^{\text{ST1}} = 3 (±1) \times 10^{-3}$, and the spin mixing conductance $g_{\uparrow\downarrow}^{\text{ST3}} = 1.6 (±0.6) \times 10^{14}$ cm⁻², were found to be lower compared to ST1 and ST2, which is expected with a reduction in quality of the YIG|Au interface.

Table 8.2: Magnetic parameters for the samples ST1: 10 min 0.8 kV Ar⁺, ST2: 22 min 10 kV Ar⁺, and ST3: 32 min 10 kV Ar⁺ surface treatment 9.0YIG films at 400°C in the structure 9.0YIG|6.1Au|2.9Fe|4.1Au and the corresponding uncovered, surface treated YIG films.

<table>
<thead>
<tr>
<th>sample</th>
<th>$g_{\uparrow\downarrow}$ [10¹⁴ cm⁻²]</th>
<th>$\alpha_{\text{sp}}$ [10⁻³]</th>
<th>$\alpha$ [10⁻³]</th>
<th>$\Delta H (0)$ [Oe]</th>
<th>$\alpha_{\text{bulk}}$ [10⁻³]</th>
<th>$\Delta H_{\text{bulk}} (0)$ [Oe]</th>
</tr>
</thead>
<tbody>
<tr>
<td>ST1</td>
<td>3.8 ± 0.2</td>
<td>6.1 ± 0.3</td>
<td>6.5 ± 0.1</td>
<td>9 ± 1</td>
<td>0.4 ± 0.3</td>
<td>15 ± 2</td>
</tr>
<tr>
<td>ST2</td>
<td>3.9 ± 0.2</td>
<td>6.2 ± 0.3</td>
<td>6.4 ± 0.3</td>
<td>21 ± 2</td>
<td>0.2 ± 0.1</td>
<td>9.6 ± 0.2</td>
</tr>
<tr>
<td>ST3</td>
<td>1.6 ± 0.6</td>
<td>3 ± 1</td>
<td>3.9 ± 0.9</td>
<td>48 ± 7</td>
<td>1.3 ± 0.4</td>
<td>10 ± 4</td>
</tr>
</tbody>
</table>

Two further surface treatments were carried out. In one case, the as-grown YIG was treated with atomic H⁺ etching for 20 minutes at a temperature of 400°C. The 1s O peak was very similar to that for the untreated YIG. The 2p₃/₂ Fe spectrum is shown in Figure 8.5 (h). In this case, H⁺ etching resulted in very large concentration of the metallic state of Fe at the YIG surface. The relative surface concentrations were in this case $Y_{4.3}Fe_{2.8}O_{12.9}$. 124
Figure 8.5: X-ray photoemission spectroscopy (XPS) data for YIG 1s O for (a) untreated (ST1, before Ar$^+$ surface treatment), (b) 10 min 0.8 kV Ar$^+$ treatment (ST1), and (c) 22 min 1.0 kV Ar$^+$ treatment (ST2). The blue dashed line is the O$^{2-}$ (530.1 eV) line, the red dashed line is the OH$^-$ (532 eV) line, the solid black line is the fitted line, the dashed black line is the Shirley background, and the blue line is the measured XPS spectrum. XPS data for YIG 2p$_{3/2}$ Fe for (d) untreated (ST1, before Ar$^+$ surface treatment), (e) 10 min 0.8 kV Ar$^+$ treatment (ST1), (f) 22 min 1.0 kV Ar$^+$ treatment (ST2), (g) 32 min 1.0 kV Ar$^+$ treatment (ST3), (h) 20 min H$^-$ treatment, and (i) an additional deposition of 0.1 nm Fe on the YIG surface. The blue dashed line is the Fe$^{4+}$ (712.8 eV) line, the red dashed line is the Fe$^{3+}$ (711 eV) line, the green dashed line is the Fe$^{2+}$ (709 eV) line, and the purple dashed line is the Fe (707 eV) line.
After H- etching, the 6.1Au|4.3Fe|6.1Au structure was deposited on the YIG. Surprisingly, FMR measurements of this sample showed no increase in damping due to spin pumping. The other surface treatment was the deposition of the equivalent of $\sim 0.1$ nm of Fe on the as-grown YIG surface. This also lead to an appreciable concentration of metallic Fe state at the surface, see Figure 8.5 (i). The surface stoichiometry after the Fe deposition was Y$_{2.6}$Fe$_{2.4}$O$_{15.0}$. After the Fe deposition, the 6.1Au|4.3Fe|6.1Au structure was deposited on the sample, and again FMR measurements showed no increase in damping compared to the bare YIG.

8.5 Summary

Using FMR measurement of Gilbert damping in nm thin, pulsed laser deposition grown YIG films and YIG|Au|Fe|Au structures, it was shown that the untreated YIG|Au interface exhibits a strong spin pumping conductance, $g_{t\uparrow\downarrow} \simeq 1 \times 10^{14}$ cm$^{-2}$, which is $\simeq 10\%$ of that observed in typical metallic FM|NM interfaces. The strength of the spin mixing conductance was found somewhat inhomogeneous across the as-grown YIG surface. This inhomogeneity is not surprising considering that the thin YIG film surface chemistry deviates from that of the bulk and the films exhibit a fairly large surface roughness.

Using Ar$^+$ etching to treat the surface of the as-grown YIG films, the spin pumping efficiency was increased substantially, $g_{t\uparrow\downarrow} \simeq 4 \times 10^{14}$ cm$^{-2}$, which is 60% of that calculated for ideal YIG surfaces [12]. Ar$^+$ etching resulted in a decrease of O concentration and an increase in the Fe concentration at the surface. Additionally, a larger percentage of the Fe after etching was found to be in the Fe$^{3+}$ state that is expected in bulk YIG. However, longer Ar$^+$ times lead to a decrease in sample quality and a reduction in the spin pumping. Both atomic hydrogen etching and the deposition of the equivalent of $\simeq 0.1$ nm of Fe on the YIG films lead to a large concentration of metallic Fe at the YIG surface and the complete suppression of spin pumping.
Chapter 9

Conclusions

High quality, ultrathin magnetic films were prepared by means of molecular beam epitaxy (MBE). Magnetization dynamics and anisotropies were studied by means of ferromagnetic resonance (FMR) in GaAs|Fe|Au structures as a function of the Fe layer thickness, allowing the determination of bulk and interface properties of magnetic anisotropies and dynamics. It was shown that one can tune the FMR field position by changing the Fe layer thickness, which is important when designing devices and experiments involving multiple ferromagnets. It was also shown that the GaAs|Fe interface leads to an appreciable interface damping and that for Fe layer thinner than 25 AL, one can avoid large extrinsic contributions to the damping due to two magnon scattering processes.

Spin pumping and transport was studied by means of ferromagnetic resonance in MBE grown GaAs|Fe|Au|Pd|Au and GaAs|Fe|Pd|Au structures. The results of the GaAs|Fe|Au studies were used to choose an Fe layer thickness that was thin enough to maximize sensitivity to spin pumping and avoid two magnon scattering processes. Three important results were found:

1. The spin pumping induced damping was quickly suppressed with the addition of the Au spacer layer as compared to GaAs|Fe|Pd samples. It was shown that the Pd layer can neither be treated as an ideal spin sink or as a simple normal metal with respect to spin currents and that the reduction in damping is due to a reflection of spin currents at the Au|Pd interface.

2. It was experimentally shown that the reduction in spin pumping induced damping with the insertion of the Au spacer layer is not related to the removal of a magnetic proximity effect induced damping at the Fe|Pd interface.

3. The spin pumping induced damping showed an oscillatory dependence on the Au spacer layer thickness when this layer’s thickness was less than the electron mean free
path. This effect is attributed to the formation of quantum well states in the Au layer. This represents the first time such a behavior was observed in the time irreversible process of spin pumping.

Magnetization dynamics were investigated in nanometer thick ferrimagnetic insulator Yttrium Iron Garnet (YIG, Y₃Fe₅O₁₂) grown by pulsed laser deposition. Ferromagnetic resonance was used to determine the spin pumping induced damping in YIG and YIG|Au|Fe|Au structures. In the YIG|Au|Fe|Au structures, the YIG acts as a spin pump and the Fe as a spin sink when the YIG layer undergoes ferromagnetic resonance. Comparing the damping in the YIG and YIG|Au|Fe|Au structures allows one to determine the efficiency of spin pumping at the YIG|Au interface given by the spin mixing conductance. The important results found in this work were:

1. It was shown that the untreated YIG|Au interface exhibits a strong spin pumping conductance, $g_{↑↓} \simeq 1 \times 10^{14}$ cm$^{-2}$, which is $\simeq 10\%$ of that observed in typical metallic FM|NM interfaces.

2. Using Ar$^+$ etching to treat the surface of the as-grown YIG films, the spin pumping efficiency was increased substantially, $g_{↑↓} \simeq 4 \times 10^{14}$ cm$^{-2}$, which is 60% of that calculated for ideal YIG surfaces [12]. However, longer Ar$^+$ times lead to a decrease in sample quality and a significant concentration of metallic state Fe at the YIG interface.

3. Both atomic hydrogen etching and the deposition of the equivalent of $\sim 0.1$ nm of Fe on the YIG films lead to a large concentration of metallic Fe at the YIG surface and the complete suppression of spin pumping.

Additionally, two ferromagnetic spectrometers were built to allow broadband measurements of FMR with a single sample mounting. These spectrometers replaced the older systems that required a different FMR spectrometer for each frequency measurement. One spectrometer makes use of a multimode cavity and rectangular waveguides and allows four frequencies to be measured between 27-40 GHz. The other spectrometer is based on coaxial cables and a coplanar waveguide and can operate from near DC to 40 GHz. It was shown that even though the coupling on the coplanar waveguide can lead to large mixing between the real and imaginary portions of the transverse magnetic susceptibility, the signal could be analyzed in a simple manner and lead to great agreement with the cavity measurements. The coplanar waveguide system also has the advantages of having a much easier learning curve for new users than the cavity based systems as well as being suitable for use of samples grown on Si substrates. In fact, the data on single magnetic layer Si|3
nm)Ta|(3.45 nm)Py|(d_{Ta})Ta|(3.6 nm)Au and double magnetic layer Si|(3.0 nm)Ta|(3.45 nm)Py|(d_{Ta})Ta|(2.0 nm Py)|(4.0 nm Fe)|(3.6 nm)Au samples shows that spin pumping is present at the Py|Ta interface and that the spin pumping saturates on the length scale of ∼ 1 nm.
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