Magnetization Dynamics in NiFe Thin Films

by

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B.Sc., University of Western Ontario, 2008

A Dissertation Submitted in Partial Fulfillment of the Requirements for the Degree of

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ABSTRACT

The morphology, composition, and magnetic properties of NiFe thin films were characterized. Films with thicknesses up to 137 nm were deposited in an RF induction evaporator at high vacuum ($\sim 10^{-8}$ mbar). Time resolved magneto-optic Kerr effect microscopy (TR-MOKE) was used to measure the Gilbert damping constant, an important dynamic magnetic property with applications to magnetic data storage. The composition of each film was measured with energy-dispersive X-ray (EDX) microscopy and used to determine the weight percent of Ni and Fe in each film.

A trend of increased damping with increased thickness was found, in agreement with published results. Magnetic properties and roughness were found to differ significantly from previous films grown in the same vacuum chamber by Rudge, and are attributed to different growth modes produced by differing deposition conditions. However, the weight percent of Ni in each film was found to be inconsistent, deviating by up to 7% from the Ni$_{80}$Fe$_{20}$ evaporation source. Inconsistent composition, caused by the inability to control deposition parameters, prevents insight into Gilbert damping from being drawn from the analysis.
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DEDICATION

To my parents, whose love and support have fostered my curiosity for nature.
Chapter 1

Motivation

The transistor is not solely responsible for the remarkable advances in computing over the past 50 years. Though Moore’s Law, which states that the number of transistors that can be affordably incorporated in a chip will double every two years, has stayed true during this time period, the usefulness of increased computing power depends on the growth of related technologies. [23] Advances in both system architecture design and data storage are equally important in order to maximize data processing throughput.

Hard disk drives have been the primary data storage device used in commodity computers and servers to date. These mechanical devices store information on a rotating magnetic platter by writing to and reorienting magnetic domains present in the platter using a write head floating above it. Increases in on-chip transistor densities have been closely matched by growth in data storage densities in hard drives, and have been essential to the development of faster computers. Importantly, the speeds at which data can be stored and retrieved have also increased. However, as daily computing shifts towards mobile platforms, other data storage devices are becoming more popular.

With the rise of smartphones and tablet computers, manufacturers are increasingly conscious of power consumption in order to maximize battery life of consumer devices. Though there is a shift towards solid-state NAND flash storage due to higher reliability and lower power consumption, a recent report by a semiconductor industry consortium predicts that magnetic RAM will have the fastest write speed (< 0.5 ns) of any emerging data storage device within the next 15 years. [5, 31] Thus, the shift back towards magnetic storage devices will be driven by speed.

In order to improve data write speeds in hard drives and future solid state magnetic
devices, a better understanding of the fundamental physics involved is required. The write speed in a hard drive is constrained by the magnetization dynamics that occur in the ferromagnetic thin film that coats the platter. When the magnetization in an area of the film has been switched (e.g., when writing a bit), there is a period of time needed for the magnetization to settle. Understanding the physics behind this settling time is essential in order to create faster hard drives because it limits the speed at which data can be written. The physical origin of this settling time, known as the Gilbert damping constant, has been intensively studied, but mechanisms that dissipate magnetic energy are not well understood.

Characterization of magnetic thin films offers an opportunity to study the material properties that contribute to Gilbert damping. Results obtained from experiments and characterization can help constrain existing theories and illuminate factors that contribute to damping. It is hoped that faster hard disk drives or solid state magnetic storage devices will be manufacturable in the future due a better understanding of Gilbert damping.
Chapter 2

Background

2.1 Ferromagnetism and Domains

In magnetostatics, the origin of the magnetic force is the magnetic dipole moment. The magnetic dipole moment determines the force that will be exerted on a body by an external magnetic field. In a material, magnetic moments can interact to create a bulk magnetization, and the rich physics that result has been the study of more than a century of research.

In 1907, Weiss used the concept of an internal magnetic field in an attempt to explain the origin of spontaneous magnetization in ferromagnets. [22] Weiss postulated that a uniform molecular field originating from molecular magnetic dipole moments permeated a ferromagnet. As a result, ferromagnets spontaneously magnetize below the Curie temperature \((T_C)\), but in order to explain how demagnetization below the Curie temperature works, Weiss proposed the formation of magnetic domains inside a ferromagnet.

Domains are small areas in a ferromagnetic material with a uniform magnetization. In each domain, the magnetization is saturated, but the magnetization direction may only be slightly aligned with that of neighbouring domains unless the entire material is considered to be magnetized to saturation. When the domains are randomly oriented, the material is said to be unmagnetized. If the domains are aligned in some capacity, the material will have a net magnetization \((\mathbf{M})\). If an external magnetic field is applied, the domains will align preferentially along the field (with increasing field) either by growth or domain wall rotation until there is complete alignment, at which point the material has reached its saturation magnetization \((\mathbf{M}_s)\). If the field is
removed, the magnetization that persists is known as the remanence.

Though Weiss was correct in his prediction of magnetic domains, the physical origin of the molecular field was not understood. Today, it is known that domains form because of a competition between the demagnetizing field and the exchange interaction, as explained in Sec 2.3.

2.2 Stoner-Wohlfarth Model

Magnetic hysteresis describes the magnetization of a ferromagnet in response to an external field (in equilibrium). Hysteresis is characterized by plotting the component of the magnetization $\mathbf{M}$ that lies along an external field $\mathbf{H}$. When the field is cycled to some maximum in both directions along the same axis, two separate branches of the magnetization emerge, and the resulting plot is characterized by several quantities. When the external magnetic field is reduced to zero, the magnetization that remains is called the remanence. When the magnetization is at a maximum, it has reached saturation. When the magnetization is reduced to zero, the applied field is equal to the coercivity of the ferromagnet. In other words, the coercivity is the field required to reduce the remanent magnetization in the material to zero. These quantities provide a basis for comparing the static magnetic properties of ferromagnets.

In 1948, Stoner and Wohlfarth published a model for the magnetic hysteresis behaviour of heterogeneous alloys. The model considers the magnetic moment of an ellipsoidal, single-domain ferromagnetic particle that is governed by a uniaxial anisotropy along the long axis and an external magnetic field, and does not include dynamics or thermal effects. The motivation for this model was that understanding the magnetization behaviour of small magnetic particles could lead to a better understanding of alloys consisting of a matrix of strongly ferromagnetic particles embedded in a less ferromagnetic matrix.

This discussion closely follows a review in Ref. [35]. The uniaxial anisotropy energy of the particle is given by $E_A = K\sin^2(\theta)$, where $K$ is an anisotropy constant, and the magnetostatic (Zeeman) energy of the particle due to the external field is $E_Z = -\mathbf{M} \cdot \mathbf{H}$. Therefore, the energy of the particle is

$$E = E_A + E_B = K\sin^2(\theta) - H_M\cos(\theta - \phi),$$

(2.1)

where $\theta$ is the angle between the anisotropy axis and the magnetization, and $\phi$ is the
angle of the external magnetic field. Because of the finite dimensions of the particle, a demagnetizing energy is added to the anisotropy so that the total anisotropy energy is

\[ K_{\text{eff}} = (K + 2\pi M_s^2(N_\perp - N_\parallel))\sin^2\theta, \quad (2.2) \]

such that

\[ E = E_A + E_B = K_{\text{eff}}\sin^2(\theta) - H_{M_s}\cos(\theta - \phi), \quad (2.3) \]

where \( N_\parallel \) and \( N_\perp \) are the demagnetizing coefficients parallel and perpendicular to the long axis. The direction that the magnetization will take depends on the uniaxial anisotropy and the applied external field. The energy has reached an extrema when

\[ \frac{\partial E}{\partial \theta} = \sin(\theta)\cos(\theta) + h\sin(\theta - \phi) = 0 \quad (2.4) \]

and the extrema is a stable equilibrium point when

\[ \frac{\partial^2 E}{\partial \theta^2} = \cos^2\theta + h\cos(\theta - \phi) \geq 0, \quad (2.5) \]

where \( h = \frac{H}{H_k} \) and \( H_k = \frac{2K_{\text{eff}}}{M_s} \), and the state transitions from stable to unstable when \( \frac{\partial^2 E}{\partial \theta^2} = 0 \). These conditions can be combined to determine the critical field values where \( M \) will switch by eliminating \( \theta \) and considering components of \( H \) parallel and perpendicular to the easy axis. Doing so yields the Stoner-Wohlfarth asteroid,

\[ \left( \frac{H_\perp}{H_k} \right)^{\frac{2}{3}} + \left( \frac{H_\parallel}{H_k} \right)^{\frac{2}{3}} = 1, \]

shown in Fig. 2.1.

![Figure 2.1: The Stoner-Wohlfarth asteroid.](image-url)
Switching behaviour can be determined by analysing tangents to the asteroid curve. For a given field value, the tangents to the curve that are closest to the easy axis \((H_{\parallel})\) will determine the stable magnetization directions. For field values that lie outside the asteroid, there will only be one tangent (closest to the easy axis) that intercepts that point, which means there is only a single energy minimum. For field values that lie inside the asteroid, two tangents closest to the easy axis will intercept it, representing two energy minima. However, the energy minimum that the magnetization direction chooses can be easily switched by magnetization dynamics or perturbations, so these are not considered stable minima. Only field values that lie inside the asteroid will reliably switch the magnetization direction, within this model.

Understanding switching behaviour is important because magnetic moments form the basis for data storage in hard disk drives. The Stoner-Wohlfarth model is a starting point for understanding the influence of static external fields on magnetization and how magnetic anisotropy causes hysteresis.

\section{2.3 Energy Contributions in Ferromagnetism}

The magnetic moment of the electron, or \textit{spin}, has many interactions in a material which are characterized by their energies. In transition metals like Ni and Fe, ferromagnetism arises because of unpaired electron spins in the d orbital. This results in a small net magnetic moment at each atom in the metal, which then interact to yield collective phenomena like domain formation. However, understanding how these small magnetic moments interact to produce bulk magnetic phenomena requires investigating the energies involved. [9][15]

\subsection{2.3.1 Zeeman Energy}

A magnetic dipole moment \(\mu\) in an external magnetic field \(H\) has a potential energy

\[ W = -\mu_0 \mu \cdot H. \quad (2.6) \]

This magnetostatic energy describes the tendency for a magnetic moment to align with an external field, and is maximized when the magnetic moment is perpendicular to the external field. In other words, it is the energy required in order to rotate the magnetic moment away from being parallel with the field. It is important to note that
this energy does not take into account any interaction between dipoles in a material. It describes only the interaction of each individual dipole with the field.

2.3.2 Demagnetization Energy

The demagnetization energy is caused by the coupling of between magnetic moments and the magnetic field inside the material. Since each magnetic moment contributes to the internal magnetic field, the demagnetization energy can be thought of as the dipole-dipole interaction between all of the magnetic moment in a sample. This presents a challenging problem because in order to calculate the effective magnetic field applied on any magnetic moment, the direction of every other magnetic moment must be known. The demagnetization energy $W_D$ is given by

$$W_D = -\frac{1}{2} \int \vec{M}(\vec{r}) \cdot \vec{H}_D dV,$$

where $\vec{M}$ is the magnetization and $\vec{H}_D$ is the demagnetization field, which generally points opposite to $\vec{M}$. The demagnetizing field attempts to demagnetize a given sample by minimizing free poles at the surface of the material, and reduces the internal effective field.

2.3.3 Anisotropy Energy

The anisotropy energy in materials is due to the different energy densities present in different directions. This anisotropy results in ferromagnets having easy and hard axes, or a preferred magnetization plane. There are many different mechanisms that can cause this, with perhaps the simplest being the magnetocrystalline anisotropy. Magnetocrystalline anisotropy is caused by the energy densities varying with direction in crystal structures. The primary physical cause for this is the spin-orbit interaction. Uniaxial magnetocrystalline anisotropy is caused by a coupling between electron spins and their orbits within a crystal lattice, and this energy is given by

$$W_u = K_u \sin^2(\theta),$$

where $K_u$ is the anisotropy constant and $\theta$ is the angle between a spin and the preferred magnetization direction. However, because NiFe films are polycrystalline, the net anisotropy is negligible in these systems.
Shape anisotropy is caused by the demagnetizing field being unequal in all directions, also contributes to the anisotropy energy. The demagnetizing field is anisotropic for particles which are not spherically symmetric.

### 2.3.4 Exchange Energy

The exchange energy arises from quantum mechanical interaction between spins in an ensemble. Unlike the effect of the demagnetizing field, the most energetically favourable configuration for the exchange interaction is when adjacent spins are aligned. In quantum mechanics, this interaction is described by the Heisenberg Hamiltonian,

$$ H = -J(r) \vec{S}_1 \cdot \vec{S}_2, $$

(2.9)

where $\vec{S}_1$ and $\vec{S}_2$ are neighbouring spins, and $J(r) = E(r)_{\uparrow \downarrow} - E(r)_{\uparrow \uparrow}$ is the exchange energy. In general, the Hamiltonian must be taken to be a sum over all pairs of spins, but a nearest neighbour approximation can be made,

$$ -\sum_{i,j} J_{i,j} \vec{S}_i \cdot \vec{S}_j \rightarrow -JS^2 \sum_{\text{neighbours}} \cos(\phi_{ij}) $$

where $\phi_{ij}$ is the angle between spin $i$ and $j$. For neighbouring spins, $\phi_{ij}$ is small and the approximation $\cos(\phi) \approx 1 - \frac{1}{2} \phi^2$ can be made. The angle can therefore be written as,

$$ \phi_{ij}^2 \approx |\vec{m}_i - \vec{m}_j|^2 \approx |\vec{r}_{ij} \cdot \nabla \vec{m}|^2 $$

$$ = \nabla m_x^2 + \nabla m_y^2 + \nabla m_z^2 $$

The exchange energy can finally be written as

$$ W_{\text{ex}} = \frac{JS^2}{a} C_{\text{neighbour}}(\nabla m_x^2 + \nabla m_y^2 + \nabla m_z^2), $$(2.10)

where $C_{\text{neighbour}}$ is the number of nearest neighbours and $\frac{JS^2}{a} \equiv A$, which is known as the exchange or stiffness constant.

Magnetic domains form due to a competition between the demagnetization energy and the exchange energy. The formation of domains allows the demagnetization energy to decrease at the expense of a high exchange energy along the boundary between domains. [25, 2]Because the demagnetizing field has a stronger interaction per
unit volume, situations can arise where there is magnetic alignment of neighbouring domains microscopically but no net magnetization macroscopically.

2.4 Magnetization Dynamics

When studying magnetization, one can derive an effective magnetic field from the energies discussed in Section 2.3 as

$$H_{\text{eff}} = -\frac{\partial W}{\partial M},$$  \hspace{1cm} (2.11)

where $W$ is the energy density and $M$ is the magnetization. To look closer at the dynamics of magnetization, the equation of motion for a magnetic dipole $\mu$ in a magnetic field $H_{\text{eff}},$

$$\frac{1}{\gamma} \frac{d\mu}{dt} = \mu \times H_{\text{eff}},$$  \hspace{1cm} (2.12)

serves as a starting point. ($\gamma$ is the gyromagnetic ratio.) Rewritten in terms of magnetization, this equation is

$$\frac{dM}{dt} = \gamma M \times H_{\text{eff}}.$$  \hspace{1cm} (2.13)

In other words, the rate of change of magnetization is always be perpendicular to both the magnetization direction and effective field, which describes Larmor precession of a dipole in a field. However, due to Zeeman coupling, one expects the magnetization to relax to an equilibrium direction along the magnetic field, so a phenomenological damping term $\vec{R}(\vec{M}, \vec{H})$ is added:

$$\frac{dM}{dt} = \gamma M \times H_{\text{eff}} + \vec{R}(M, H)$$  \hspace{1cm} (2.14)

In 1935, Landau and Lifshitz suggested an explicit form for $\vec{R}$ [18] which allows one to rewrite Eqn. 2.14 as

$$\frac{dM}{dt} = \gamma M \times H_{\text{eff}} - \frac{\lambda}{M_s^2} M \times (M \times H_{\text{eff}}),$$  \hspace{1cm} (2.15)

where $\lambda$ is the relaxation frequency and $M_s$ is the saturation magnetization. This equation is known as the Landau-Lifshitz equation, but it is in only valid in the limit of small damping. In 1955, Gilbert introduced a new phenomenological damping term
that removed this limit,

$$\vec{R} = -\frac{\alpha}{M_s} \vec{M} \times \frac{d\vec{M}}{dt}, \quad \text{(2.16)}$$

where $\alpha$ is a dimensionless damping parameter. There are several different mechanisms which can contribute to damping, including lattice vibrations, crystal defects, and domain walls. Inserting this new expression into Eqn. 2.14 yields the Landau-Lifshitz-Gilbert (LLG) equation,

$$\frac{d\vec{M}}{dt} = \gamma (\vec{M} \times \vec{H}_{\text{eff}}) - \frac{\alpha}{M_s} (\vec{M} \times \frac{d\vec{M}}{dt}). \quad \text{(2.17)}$$

The LLG equation is the starting point for micromagnetic modeling. It describes the dynamics of magnetization inside a material that is governed by some effective magnetic field $\vec{H}_{\text{eff}}$. Depending on the situation, $\vec{H}_{\text{eff}}$ can include additional energy terms in order to model the impact of certain phenomena such as eddy currents. Once the form of $\vec{H}_{\text{eff}}$ is determined on paper, one can then proceed to solve the LLG computationally.

### 2.5 Gilbert Damping

The Gilbert damping constant, $\alpha$, was introduced into the Landau-Lifshitz formula by adding a dissipative term to the Lagrangian equations of motion for a macrospin vector. [8] In other words, the Gilbert damping constant is phenomenological, and encompasses the rate at which magnetic energy leaves the system. The physical origin and mechanisms that contribute to Gilbert damping have been the focus of much research over the past 40 years, but are still not entirely understood.

An effective damping constant is commonly split into two contributions as,

$$\alpha_{\text{eff}} = \alpha_{\text{int}} + \alpha_{\text{ext}}; \quad \text{(2.18)}$$

where $\alpha_{\text{int}}$ is called the intrinsic contribution to damping, and $\alpha_{\text{ext}}$ is the extrinsic contribution. Intrinsic damping is a homogeneous material property which includes factors like crystal structure, while extrinsic damping depends on impurities, grain size, and inhomogeneities in the geometry of the material.

These two contributions to damping can also be described by their physical mechanisms instead of their dependent measurable quantities. In 1998, Suhl explained
that there are two pathways for energy to transfer out a uniform precessional mode of the average magnetization. [34] In direct damping, energy is dissipated directly into motion of the lattice or conduction electrons in metal. Suhl argued that in samples smaller than a domain wall thickness, only direct damping can occur due to spin wave creation being energetically unfavourable.

Indirect damping involves energy loss through the creation of non-uniform precessional modes (spin waves), which then later lose their energy to the lattice or conduction electrons. Possible mechanisms for indirect damping involve scattering of spin waves off impurities, domain walls, and by thermal agitation. Indirect damping provides a theoretical basis for the observed extrinsic damping contributions.

However, the body of experimental evidence that supports these descriptions is neither systematic nor complete. Platow et al. observed an increase in effective damping as film thickness decreased in Co, Fe, and Ni films on Cu(001), which may have been due to a change in crystal structure at lower thicknesses (intrinsic), or due to an increase in extrinsic damping from surface roughness. It was also found that intrinsic damping can be anisotropic in certain ferromagnetic monolayer films. [28]

In 2005, Kuanr et al. used a two-magnon scattering model to explain ferromagnetic resonance (FMR) linewidths found in NiFe thin films, which was justified because of small grain inhomogeneities present in their samples. [17, 20, 21] As discussed by McMichael et al., a spatially varying anisotropy field due to surface voids, pits, or grain structure results in a spin-wave spectrum that varies locally, which facilitates two-magnon scattering. While this provides a potential mechanism through which surface roughness can enter damping, neither surface roughness nor the effect of film thickness were studied.

There is an incomplete understanding of the thin film and magnetic properties that contribute to Gilbert damping, and further progress is impeded by the lack of common measured parameters in existing experimental studies. Continued research on Gilbert damping is motivated by these aspects of current literature.

2.6 Magneto-Optic Kerr Effect

A useful tool for probing the magnetization of materials is the magneto-optic Kerr effect. Discovered in 1876 by John Kerr, the effect causes a polarized beam of light to rotate after reflection off a magnetized material.

In classical electrodynamics, the Kerr effect in a thin film is described by a change
in the dielectric permittivity tensor

\[ \epsilon(M, \omega) = \begin{bmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ -\epsilon_{xy} & \epsilon_{yy} & 0 \\ 0 & 0 & \epsilon_{zz} \end{bmatrix}, \quad (2.19) \]

cauised by the presence of the magnetization \( M \). The dielectric permittivity tensor \( \epsilon(M, \omega) \), which is complex in general, describes the response of a material to an electric field (real part), as well as the effect of the material back on the electric field (imaginary part). In a material, the dielectric tensor relates the electric field \( \mathbf{E} \) to the electric displacement field \( \mathbf{D} \) by the relation,

\[ \mathbf{D} = \epsilon(M, \omega) \cdot \mathbf{E}. \quad (2.20) \]

The Kerr effect can be illustrated by considering a linearly polarized beam of light incident normal to the surface of a material that has a dielectric tensor with off-diagonal terms, such as Eqn. 2.19. The electric field of a linearly polarized beam propagating in the \( \hat{z} \) direction can be written as

\[ \mathbf{E} = E_x \hat{x}, \quad (2.21) \]

where \( E_x \) is assumed to be time dependent. Across an interface between media, the parallel components of the electric fields \( E_A^\parallel \) and \( E_B^\parallel \) on either side of the interface are continuous, therefore

\[ E_A^\parallel - E_B^\parallel = 0 \quad (2.22) \]

and similarly for the electric displacement,

\[ D_A^\parallel - D_B^\parallel = 0. \quad (2.23) \]

If the first medium is linear, isotropic, and homogeneous, the dielectric tensor can be reduced to a scalar, \( \epsilon_A \). The second medium will have an anisotropic dielectric tensor \( \epsilon_B = \epsilon(M, \omega) \). Assuming no electric polarization in the media, combining Eqn. 2.23 and 2.20 yields the equations

\[ \epsilon_A E_x^A = \epsilon_{xx} E_x^B + \epsilon_{xy} E_y^B \quad (2.24) \]

\[ \epsilon_A E_y^A = -\epsilon_{xy} E_x^B + \epsilon_{yy} E_y^B. \quad (2.25) \]
However, the electric field outside the material, $E^A$, must be decomposed into components from the incident ray and reflected ray:

$$E^A_x = E^i_x + E^r_x$$  \hspace{1cm} (2.26)

$$E^A_y = E^r_y.$$  \hspace{1cm} (2.27)

Combining these equations with Eqn. 2.24 and 2.25 yields the result

$$E^r_x = \frac{\epsilon_{xx}}{\epsilon_A} E^B_x + \frac{\epsilon_{xy}}{\epsilon_A} E^B_y - E^i_x$$

$$E^r_y = -\frac{\epsilon_{xy}}{\epsilon_A} E^B_x + \frac{\epsilon_{yy}}{\epsilon_A} E^B_y.$$  \hspace{1cm} (2.28)

These equations encompass the phenomenology of the Kerr effect. Reflecting a linearly polarized beam with an electric field purely along the $\hat{x}$ direction off of a material with a dielectric tensor with off-diagonal terms results in a reflected beam with a polarization that has rotated towards the $\hat{y}$ direction. Therefore, there has been a rotation of the plane of polarization in the reflected beam.

The microscopic origin of this effect is not explained by classical electromagnetic theory, and quantum mechanics must be invoked. In certain insulators, the permittivity tensor can be derived using the Kubo formula and these insulators are shown to have different transition rules depending on whether incident light is left or right circularly polarized. [30] In general, the effect is explained as a change in band structure that occurs below the Curie temperature in ferromagnetic materials, which leads to these different transition rules. [11, 32]

It has been shown that these changes in the band structure cause the reflectivity of the material to change linearly with $M$. Therefore, the change in polarization of a reflected beam due to the Kerr effect can be used to probe the magnetization of a material. By measuring the change in polarization of a beam reflected off a magneto-optically thin film as an external magnetic field is swept, the magnetization of the sample can be measured as a function of applied field.
Chapter 3

Experiment

A series of experiments were carried out in order to study the effects of film thickness on extrinsic Gilbert damping in Ni$_{80}$Fe$_{20}$ thin films. In order to create Ni$_{80}$Fe$_{20}$ thin films to study, an RF induction evaporator was designed and built such that characterization of these films could be done in-situ (though ultimately all characterization was done ex-situ). An atomic force microscope was used to measure the thickness of each film and characterize the surface roughness, which may contribute to extrinsic damping. Static magnetic properties were measured using the magneto-optic Kerr effect technique to ensure each film was ferromagnetic. This experiment was then extended into the time domain using the pump-probe technique, and the response of the magnetization to a 4 ns field pulse was measured with 20 ps resolution in order to determine the Gilbert damping constant of each film. Lastly, energy-dispersive X-ray spectroscopy was used to measure the composition of each sample, to ensure each deposition resulted in the same NiFe alloy. Verification and control of alloy composition is essential because composition will change intrinsic damping.

Because many different properties of NiFe thin films may contribute to Gilbert damping, consistent film composition and structure are important so that the influence of each property on extrinsic Gilbert damping can be clearly separated and measured. Previous work by Rudge suggests that surface roughness may be the dominant contribution to extrinsic damping for ultra-thin films.[29] As a continuation of that work, studying Gilbert damping over a wider range of film thicknesses could provide further insight into damping mechanisms.

Initially, in-situ magnetization characterization was attempted in order to reduce the likelihood of oxidation destroying the ferromagnetism of the samples. However, in-situ measurements were hampered by high noise caused by a lengthy optical path
coupled with an insufficiently damped vacuum chamber. Additionally, each film studied showed no loss in ferromagnetism after being exposed to dry air for a period of weeks.

3.1 Deposition and Atomic Force Microscopy

A set of NiFe thin films with different thicknesses were grown using a home-made RF induction evaporator constructed as part of this project. The evaporator consisted of a water-cooled RF coil mounted inside an ultra-high vacuum chamber, connected to a Lepel Model T-5-3 RF generator. Permalloy (Ni$_{80}$Fe$_{20}$) pellets were placed inside an alumina crucible mounted inside the RF coil.

For each evaporation, a glass slide patterned with a gold microcoil was mounted 20 cm above the evaporation source. The microcoils had a wire width of 12.5 µm and a gap of 30 µm. (Fig. 3.2) Each microcoil was patterned by optical UV lithography at the University of British Columbia as part of a prior project by Rudge.[29] The plane of the glass was oriented at normal incidence to the source. Evaporation times and base pressures are listed in Table 4.1.

The thickness of the resulting NiFe films was measured ex-situ using a contact-mode Nanosurf easyScan Atomic Force Microscope (AFM). An AFM operates by dragging a cantilever, which has a tip that narrows down to several nm in width at the end, across a surface. The tip is raster scanned across a sample using piezoelectrics, and the force between the surface and the tip results in a deflection of the cantilever. The deflection of the cantilever is detected by a laser beam that reflects off the top cantilever onto a position-sensing photodiode. With proper calibration, the raster scan produces a topological image of the sample.

Though the AFM is primarily used for obtaining topological information, film thickness can be measured by finding an edge of the film, where a clear flat region of both the film and substrate are present in a single image. By calculating the difference between the average AFM tip height over the film and substrate, one can measure film thickness. This can be a challenging process because cleanly cut film edges can be difficult to locate and poor adhesion at the edges can result in peeling. However, with patience, this method yields consistent thickness measurements.
3.2 Static Magneto-Optic Kerr Effect (MOKE)

Magnetic hysteresis loops were measured using the magneto-optic Kerr effect in the in-plane geometry. A red diode laser was directed through a polarizer to produce a linearly polarized beam, focused by an objective lens, and reflected off a thin film sample at near 45° incidence. The reflected beam was passed through a second polarizer (analyser) before terminating on a Si photodiode. An optical chopper was placed in the beam path to modulate the laser with a reference signal.

A ferrite-core electromagnet was placed around the sample, with the magnetic field oriented along the plane of the film. The magnetic field was swept from -90 to 90 Oe at a rate of approximately 0.5 Hz, for 100 cycles. The photodiode voltage (proportional to detected intensity) was amplified by 50x, passed into the lock-in amplifier, and demodulated from the chopper reference signal. As the magnetic field was swept, a small ellipticity in the polarization of the reflected beam was introduced due to the magneto-optic Kerr effect, as discussed in Sec. 2.6, and a voltage proportional to the in-plane magnetization was recorded from the lock-in amplifier. Hysteresis loops measured from each sample are shown in Fig. 4.7.

3.3 Time Resolved Magneto-Optic Kerr Effect (TR-MOKE)

The static magneto-optic Kerr effect measurement can be extended using the stroboscopic technique to provide a temporal measurement of the magnetization with picosecond resolution. The stroboscopic technique requires that a system be perturbed at a fixed frequency and a measurement be performed on that system in a much shorter time. In this case, the magnetization must be perturbed at a fixed frequency, and the dynamic response of the magnetization must be measured within picoseconds.

The stroboscopic technique can be illustrated in simple terms by considering a leaky faucet that is dripping at a rate of 4 drops per second. If this faucet were in a completely dark room, an observer would see nothing. If a strobe light that flashed briefly at the same rate of 4 Hz in sync with the faucet was added to the room, the observer would see the water droplet appear to be in the same place during each flash. The droplet would appear to be frozen in time, and shifting the phase between the
dripping water and the strob e would result in the droplet appearing to move back and forth in time. The ability to repeatedly measure quick events occurring at a fixed frequency and control the phase between the event and the measurement is the purpose of the stroboscopic technique. This technique is also often referred to as the “pump-probe technique”, where the pump is the act that excites the sample, and the probe is the measurement of that sample’s response.

Now, replace the faucet with a magnetic thin film and the drip with a 4 ns magnetic field pulse. After the pulse occurs, the magnetization in the sample will respond by tilting away from its initial direction and precessing as it decays back to equilibrium, sometimes in less than 1 ns. This is the quick event that TR-MOKE measures. The analogy is completed by replacing the strob e light with a fs-pulse laser and the observer with optics to detect a magneto-optic Kerr signal. [19, 6]

The first studies of magnetization dynamics on this time-scale were done in the early 1960s, using inductive loops placed around NiFe thin film.[4, 38] However, the optical approach to measurement offers the advantages of higher temporal resolution and the ability to perform spatial imaging of the magnetization as well.

![TR-MOKE apparatus optical path](image)

Figure 3.1: TR-MOKE apparatus optical path.

The TR-MOKE apparatus used this in work started with a mode-locked Ti-Sapphire fs-pulse laser (Spectra Physics 3941-M1BB Tsunami) with a nominal output of 1 mW, consisting of 100 fs pulses at 800 nm with a repetition rate of 800 kHz. The laser beam was split into two branches, the first of which was sent through a 1500 Hz optical chopper and terminated on a fast photodiode. (Fig. 3.1) The output pulse
from the fast photodiode was the trigger for the pump in this experiment, and caused the excitation field pulses to be synchronized with pulses from the laser.

A delay pulse generator (SRS DG535) used the fast photodiode pulse to trigger another pulse, after a delay time of between 948 and 452 ns. The delayed pulse was 1µs-long, and acted as a trigger for a high-current picosecond pulse generator (Picosecond Pulse Labs 2600C). For each trigger pulse, the picosecond pulse generator output a 45 V / 1.0 A pulse that was sent through a microcoil beneath the magnetic thin film under investigation. This fast current pulse induced a magnetic field pulse directed out of the plane of the sample, and acts as the pump. (Fig. 3.2)

**Figure 3.2: Microcoil and excitation field geometry (schematic).**

A simple calculation gives an estimate of the strength of the magnetic field pulse in this setup. Treating the microcoil wire as a 12.5 µm wide infinitely thin sheet of current and using Ampere’s law yields a field strength of 400 Oe at the edge of the wire. Though the field strength in the centre of the microcoil will be weaker, the measurements in this work were carried out at the edge of the microcoil wire (see Ch. 4).

The second branch of the beam was directed through a linear polarizer in order to ensure the polarization state of the beam did not fluctuate. The beam was then passed through a 50-50 beam splitter in order to obtain normal incidence with the sample. The beam was focused by a 10x microscope lens and reflected off the sample at normal incidence, which then causes a rotation in polarization angle proportional to the magnetization of the sample due to the Kerr effect. The average magnetization
was probed over a beam spot size of \( \sim 10 \mu m \).

In previous experiments, the beam was typically focused on the region of the sample that lies between the gap of the microcoil because that was where the out-of-plane magnetic field pulses would be the most uniform and most likely to give a strong signal. However, in the current work, no precession was observed in any sample if the beam was focused perfectly in the middle of the coil. However, strong signals were observed by placing the beam on the edge of the microcoil structure, resulting in measurements where the excitation field was in the plane of the film as opposed to out of plane in the previous geometry.

The reflected light from the sample was collected by a pair of Si photodiodes. A beam-splitting polarizer was placed in the beam path such that each photodiode measured an orthogonal component of the polarization. A half-wave plate was placed before the beam-splitting polarizer and rotated such that the measured intensities of the two orthogonal components were equal. The photodiode signals were subtracted with an SRS SR560 low-noise preamplifier, which doubled the signal strength of any detected change in polarization. This signal was processed by an SRS SR830 lock-in amplifier using the 1500 Hz signal generated by the optical chopper as a reference. The lock-in amplifier dramatically increased the signal-to-noise ratio by multiplying the photodiode signal by the reference signal and integrating it over a period of 300 ms, and then low-pass filtering the resulting DC signal. Finally, the filtered output from the lock-in amplifier was captured by a National Instruments data acquisition card in a PC and a custom LabVIEW virtual instrument written for this project.

As the PC read from the lock-in amplifier, the delay time on the delay pulse generator was changed in order to sweep the timing between the pump and the probe, and this allowed a time-resolved picture of the average magnetization within the beam spot to be recorded with 20 ps resolution.

A limitation of this approach is that only relative changes in the polarization are measured, so the initial magnetization state cannot be determined.

### 3.4 Scanning Electron Microscopy and Energy-Dispersive X-Ray Spectroscopy

In order to probe features smaller than optical wavelengths, scanning electron microscopy (SEM) is a valuable tool. A scanning electron microscope accelerates a
beam of electrons towards a sample and analyses the scattered electrons to construct an image of the sample. Delicate electron optics are required in order to focus the beam tightly, and several steps of manual focusing are required in order to resolve a good image. [27]

The scattered electrons detected by the SEM were both secondary electrons ejected from near the sample surface and back-scattered electrons ejected from a deeper volume in the samples. Secondary electrons are produced as a result of ionization of species near the sample surface and contain topographical information. Back-scattered electrons are the result of elastic collisions with species within a deeper volume of the sample, and because the scattering cross-section is highly dependent on the atomic number Z, they carry information about composition. In practice, back scattered electrons provide higher contrast when looking at samples of varying composition, and a mix of between both secondary and back-scattered electron signals can be most useful for imaging.

SEM also requires conductive samples, otherwise charging will occur and produce blurred or low-contrast images. In order to reduce charging, the samples under examination were mounted to an aluminium SEM stub using conductive graphite paint with a strip painted across one edge of the sample surface. Painting a strip across one edge was necessary in order to ground each NiFe thin film to the stub because the sample substrate was non-conductive glass. This was found to significantly reduce charging under the high accelerating voltages and probe currents required to perform EDX.

Energy-Dispersive X-Ray (EDX) Spectroscopy is an elemental analysis technique that works within an SEM by analysing the X-rays emitted by a sample under electron bombardment. [36] When an accelerated (primary) electron strikes an atom, it may eject an inner, strongly bound electron from the atom. An outer electron will fill the resulting vacancy and an X-ray will be emitted to balance the energy difference between the states. These transition energies are specific to each atomic species, and so the energies of the resulting X-rays can be used to characterize the elemental composition of a sample.

SEM images were obtained using a Hitachi S-4800 FESEM with 5 nm resolution at the University of Victoria Advanced Microscopy Facility. EDX spectra were measured using a Bruker Quantax EDS module attached to the same Hitachi SEM. Additional SEM images were obtained using a Raith 50 SEM with 50 nm resolution at the University of Victoria Nanofabrication Facility. EDX spectra were integrated over
100 s at an accelerating voltage of 15 kV, which yielded enough counts to determine the ratio of Ni to Fe by weight with less than 1% error.
Chapter 4

Results

The NiFe films grown were found to have thicknesses of 130 nm, 17 nm, and 34 nm. The complete set of measured deposition parameters are presented in Table 4.1. The large difference in evaporation rates can be attributed to the temperature of the NiFe source, which was not possible to measure during evaporation. Throughout 11 different evaporation processes, it was observed that the speed with which the source material melted appeared to depend on the geometry and mass of the source material. Because the NiFe was melted inductively, these factors likely played a large role in determining the rate of energy transfer from the induction coil into the NiFe source material. In the case of the 17 nm film, it was not certain whether the NiFe source material melted or had instead deposited by sublimation. Such limitations are inherent to the design of the thermal evaporator built, at the high temperatures required to melt NiFe.

The thin film surface topology was imaged using an AFM. The images have been drift corrected and median difference line corrected using Gwyddion to improve presentation, but all quantitative analysis was performed on data which was only plane-subtracted (when appropriate) using the Nanosurf easyScan software. [16]

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sample</th>
<th>Sample</th>
<th>Sample</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
<td>C</td>
</tr>
<tr>
<td>Base Pressure (mbar)</td>
<td>$3.6 \times 10^{-8}$</td>
<td>$9.8 \times 10^{-8}$</td>
<td>$1.2 \times 10^{-7}$</td>
</tr>
<tr>
<td>Evaporation Time (s)</td>
<td>1050</td>
<td>60</td>
<td>30</td>
</tr>
<tr>
<td>Thickness (nm)</td>
<td>17</td>
<td>34</td>
<td>130</td>
</tr>
<tr>
<td>RMS Roughness (nm)</td>
<td>2</td>
<td>4</td>
<td>5</td>
</tr>
<tr>
<td>Evaporation Rate (nm/s)</td>
<td>0.016</td>
<td>0.57</td>
<td>4.3</td>
</tr>
<tr>
<td>Composition</td>
<td>Ni$<em>{81}$Fe$</em>{19}$</td>
<td>Ni$<em>{86}$Fe$</em>{14}$</td>
<td>Ni$<em>{88}$Fe$</em>{12}$</td>
</tr>
</tbody>
</table>

Table 4.1: NiFe film parameters
Figure 4.1: AFM scans of the NiFe thin films showing surface topology.

(a) Sample A (17 nm)

(b) Sample B (34 nm)

(c) Sample C (137 nm)
Figure 4.2: 3D view of a Sample C AFM scan at the film edge. The NiFe film is the raised area (left), while the depressed region is glass (right). Peeling is also seen at the film edge (middle).

Thickness measurements were also carried out using the AFM (Table 4.1). Clear cut edges of the film where both the film and clean glass were identified, and the film thickness was measured by taking the difference of the mean height in each region. A 3D view of one of the films can be seen in Fig. 4.2, which shows a clear difference in height at the film edge.

The RMS area roughness of each film was calculated using the AFM images as

$$S_q = \sqrt{\frac{1}{MN} \sum_{k=0}^{M-1} \sum_{l=0}^{N-1} [z(x_k, y_l) - \mu]^2},$$

where $M$, $N$ are the image dimensions, $z$ is the height of the current pixel, and $\mu$ is the average height. [7] The RMS area roughness was measured both inside the microcoil and directly on top of the microcoil and found to be consistent across these regions of the film. (Table 4.1)
Scanning electron micrographs were obtained of each sample in order to verify the continuity of the NiFe thin film and to resolve more information about the micro-scale structure of both the NiFe film and the Au microcoil. Fig. 4.3 shows scanning electron micrographs of the microcoil on each sample. Fig. 4.3b is unique in that the microcoil from Sample B displays a significant ripple throughout it. It is unclear whether the NiFe or the Au layer has buckled, but this inhomogeneity may have contributed to the differences in the TR-MOKE data observed for that sample.

Figure 4.3: Scanning electron micrographs of the Au microcoil structures, after deposition of a NiFe thin film across the entire substrate.

The thin film structure observed in Sample B by AFM (Fig. 4.1b) was verified with SEM (Fig. 4.3b). The structure of Sample C was also verified.

An image of Sample C taken with the addition of the back-scatter detector (Fig. 4.3d) shows a significant difference in composition near the contact pads towards the left. During fabrication of the microcoils, an insulating layer of photoresist was left on in order to prevent the NiFe film from shorting out the Au microcoil. The bright areas in the image could either be areas where the photoresist or the Au itself had peeled off. However, EDX analysis on the brighter regions indicated 35% more gold was present compared to the darker regions. Additionally, the amount of Si detected from the glass substrate was 0% compared to 14% for the darker regions (due to more electrons
Figure 4.4: High magnification scanning electron micrograph of Sample B NiFe thin film at the outer edge of the microcoil.
<table>
<thead>
<tr>
<th>Sample</th>
<th>C</th>
<th>O</th>
<th>Si</th>
<th>Fe</th>
<th>Ni</th>
<th>Cu</th>
<th>Au</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.00</td>
<td>78.38</td>
<td>20.57</td>
<td>0.19</td>
<td>0.79</td>
<td>0.03</td>
<td>0.03</td>
<td>100.0</td>
</tr>
<tr>
<td>B</td>
<td>5.94</td>
<td>53.28</td>
<td>21.92</td>
<td>2.61</td>
<td>14.83</td>
<td>1.41</td>
<td>0.01</td>
<td>100.0</td>
</tr>
<tr>
<td>C</td>
<td>5.26</td>
<td>61.63</td>
<td>24.34</td>
<td>0.86</td>
<td>6.88</td>
<td>0.89</td>
<td>0.13</td>
<td>100.0</td>
</tr>
</tbody>
</table>

Table 4.2: Normalized atomic concentrations obtained by EDX spectroscopy for the NiFe thin films.

...penetrating through the metal layers). Both of these observations support the notion that at least some of the Au had peeled away, indicating fabrication issues that may have contributed to the absence of a TR-MOKE signal (due to faulty microcoils) in most of the samples studied.

Film composition was measured with EDX, and by comparing the weight percent of Ni to Fe, the NiFe thin film alloy composition was determined. (Table 4.1) However, a full spectral analysis revealed Cu contamination in the film in amounts comparable to the concentration of Fe in two of the samples. (Table. 4.2) The induction evaporator used to deposit the NiFe films was also frequently used for Cu evaporation, and it was observed that layers of Cu accumulated on the walls of the evaporator. The stress of repeatedly having Cu and NiFe layers deposited on the inside of the evaporator eventually caused these layers to peel off. Though it was never directly observed, it is plausible that Cu contamination could have occurred by peeling Cu falling into the hot crucible during a NiFe deposition.

The C, O, and Si detected was due to the probe electrons penetrating through the thin film and detecting the glass and carbon paint used to affix the sample to the SEM stub. The trace amounts of Au were likely residue beneath the NiFe from the original photolithography processing which created the microcoil pattern. Lastly, a composition map was produced of Sample C to verify NiFe film quality (Fig. 4.6). This was to ensure the NiFe films produced by the evaporator were homogeneous in composition over the beam spot size used for TR-MOKE measurements.

Hysteresis loops were obtained using the magneto-optic Kerr effect (Fig. 4.7) in the in-plane geometry. In-plane coercivities were calculated by averaging two sets of measurements taken at different areas of each film. Coercivities of 15 Oe, 14 Oe, and 5 Oe were found for Samples A, B, and C respectively. Though the general trend expected for magnetic thin films is increased coercivity with increased thickness, coercivity behaviour is strongly dependent on the weight percent of Ni in NiFe films. For example, the coercivity of Ni$_{80}$Fe$_{20}$ has been observed to drop by nearly a factor...
Figure 4.5: EDX spectrum of Sample C.
Figure 4.6: EDX composition map of Sample C illustrating uniform composition over the 10 µm area probed by TR-MOKE.
of 2 as electrodeposited film thickness grew from 50 nm to 100 nm. In the same study, the coercivity of Ni$_{88}$Fe$_{12}$ films were shown only increase by 6-9% in the same thickness range. [37] Thus, the coercivity of NiFe thin films has contributions from both thickness and composition, as well as imperfections that act as pinning sites, so a simple trend with either parameter is not expected.

The dynamic response of the magnetization to a 4-ns field pulse was probed using TR-MOKE. A typical data set obtained using TR-MOKE is presented in Figure 4.8. Initially ($t < 1.2$ ns), the magnetization is observed in a relaxed state that is aligned along the effective magnetic field direction in the film. After the onset of the pulse ($1.2$ ns $< t < 4.2$ ns), the magnetization precesses around the new effective field direction, and eventually damps out. After the magnetic field pulse ($t > 4.2$ ns), the effective field rotates back to the initial direction. However, if the magnetization was significantly displaced from the original equilibrium direction by the magnetic field pulse, then the relaxation back to this equilibrium direction can result in a second period of damped precession (as observed in Fig. 4.8).

The TR-MOKE experiment was repeated for each sample, in external bias fields of $\pm 75$, 150, 225, and 300 Oe. (Fig. 4.9) The area of each film being probed was approximately 10 $\mu$m in diameter. Because the laser beam was at normal incidence to the film, the resulting Kerr signal is proportional to the out-of-plane magnetization in the sample. The direction of the magnetic field pulse was also switched by inverting the electric pulse polarity.

In prior work by Rudge, the beam spot was aligned in the center of the microcoil, where there is a relatively uniform out-of-plane excitation field. However, in the present work, no precessional motion was ever observed by placing the beam spot near or in the centre of a microcoil. Precessional motion was only ever observed in the NiFe films studied by focusing the beam spot at the edge of the microcoil. This then leads to the question of which direction the magnetic field pulse pointed in this work.

For each sample, a combination of a positive field pulse and a positive bias field resulted in a highly suppressed excitation. Before excitation, the magnetization is thought to be aligned (in-plane) along an effective magnetic field direction inside the 10 $\mu$m spot being measured. If an excitation field is then also applied in-plane and parallel to the existing magnetization direction, then the excitation field would not change the effective field direction, and thus no precessional motion would be observed. If the excitation field was perpendicular to the magnetization direction,
Figure 4.7: In-plane hysteresis loops of the NiFe thin films, with the dashed line as a 3-point moving average.
then precessional motion would be expected to occur.

The difference in the TR-MOKE data of samples B and C is thus explained with this line of reasoning. The difference in dynamic behaviour in the data without a bias field is attributed to different angles between the initial magnetization direction and the excitation field pulse direction.

There is also a clear shift in the magnetization response time in some of the TR-MOKE data sets. The initial precession in Fig. 4.9f appears delayed as the bias field is increased, and the relaxation precession appears accelerated. These trends can be explained by considering the change in effective field during the onset and termination of the pulse field, recalling that the rise and fall times of the field pulse are fixed. The pulse field is directed opposite to the bias field, so when the bias field is increased, the net effective field is reduced. This means that the rate of change of the net magnetic field during the onset of the pulse is lower, resulting in a slower magnetic response. There is also a more significant shift in the precession observed in Sample C because, as discussed in Sec. 5.2, the field pulses were found to be about 150 Oe stronger in Sample A.
Figure 4.9: Out-of-plane component of the magnetization in response to a 4-ns magnetic field pulse, in various in-plane bias fields.
Chapter 5

Analysis

The various measurements taken reveal additional detail about the magnetic properties of each film, under further analysis, and permit a comparison of magnetic properties with film structure and composition. In particular, the Gilbert damping constant can be extracted from TR-MOKE data showing one component of damped magnetization precession, and a comparison with film thickness can further identify its contribution to extrinsic Gilbert damping.

5.1 Gilbert Damping

From the TR-MOKE data (Fig. 4.9), the Gilbert damping constant can be extracted for each film by fitting the Kerr signal (taken to be proportional to the out-of-plane magnetization) to the solution of a linearized of the LLG equation,

$$M_\perp = C \exp(-t/t_0) \cos(2\pi \frac{t}{t_0} + \phi), \quad (5.1)$$

where $M_\perp$ is the magnetization perpendicular to the equilibrium direction, $C$ is the amplitude, $t$ is the time, $t_0$ and $t_1$ are time constants, and $\phi$ is the phase. [12] Using $t_0$ as a fitting parameter yields the damping constant because

$$t_0 = \frac{2}{\alpha \gamma_0 M_s}, \quad (5.2)$$

where $\alpha$ is the Gilbert damping constant, $M_s$ is the saturation magnetization, and $\gamma_0 = \mu_0 |\mu_B| g$ where $g$ is the Landé g-factor and $\mu_B$ is the Bohr magneton.

The Gilbert damping constant was determined to be 0.031, 0.013, and 0.021 for
the 130 nm, 17 nm, and 34 nm thick films respectively. These values are comparable to published results of 0.018 and 0.0128 for 204 nm and 30 nm thick NiFe sputtered films, and are plotted in Fig 5.1. [1, 17]

In the three films grown, the effective Gilbert damping constant increases with thickness. This is in agreement with results obtained by ferromagnetic resonance (FMR) from Youssef et al., in which the peak-to-peak zero-frequency FMR linewidth (which includes the extrinsic contribution to damping) increases with thickness. [1] However, Fig. 5.1 clearly shows the opposite trend in the data by Rudge obtained by TR-MOKE.

In the work by Rudge, it was concluded that the extrinsic contribution to Gilbert damping due to surface roughness is high for very thin (< 23 nm) and as thickness increases, the damping constant quickly decreases to its bulk value (because the surface roughness becomes insignificant). Youssef et al. explain the opposite trend in their data as being due to additional relaxation mechanisms such as two-magnon scattering and a spatially varying anisotropy field. However, the data by Rudge is in a different thickness regime (8 - 22 nm) than the data by Youssef (150 - 1000 nm), and this perhaps provides an important clue as to why their observations are so different.

There are two unique transitions that occur between these regimes. The first is a transition in the morphology of the film, as is evident by comparing AFM images of the 34 nm and 137 nm films grown. (Fig. 4.1b, 4.1c) The grain size in NiFe is known to vary between 10 - 100 nm depending of the deposition environment, and this gives a range in which the film morphology is expected to change. [24, 13] As a metal film grows towards the percolation threshold or grain size, coalescence of islands into a continuous layer and recrystallization will occur.[14] These processes will decrease surface roughness and bulk inhomogeneities, and may contribute to the trend of decreasing damping with increasing thickness observed by Rudge.

The second transition that occurs within this thickness regime is a change from Neel to Bloch wall structure in Permalloy, where domain walls will begin to have an out-of-plane magnetization component.[3] In a study by Chen et al., damping was found to be constant for sputtered films between 30 - 90 nm thick, but increased linearly with thickness above 90 nm. Because the Bloch domain wall width increases linearly with thickness, it was proposed that an increasing number of perpendicular magnetic moments caused by these walls enhances the scattering of $k = 0$ (uniform mode) to $k \neq 0$ (non-uniform mode) magnons by creating more scattering sites. This scattering mechanism is thought to have a large contribution to Gilbert damping.[34]
Figure 5.1: Gilbert damping vs. thickness in various NiFe thin films.
(a) Zero-frequency peak-to-peak ferromagnetic resonance linewidth (damping) vs. film thickness in NiFe thin films studied by Youssef et al. [1]

(b) Gilbert damping constant vs. film thickness in NiFe thin films studied by Chen et al. [3]

Figure 5.2: Results from existing studies showing increased damping with increased thickness.
Both of these transitions may contribute to the observed trends in Gilbert damping with thickness, in conjunction with the different thermal evaporation technique used by Rudge. Thus, it is proposed that the thin films from Rudge (< 23 nm) are below one or both of these transition regimes, which explains the trend of decreased damping with increased thickness. It follows that the films grown for the present thesis are above this threshold, as the trend observed for these films agrees with published works (Fig. 5.2).[1, 3] (The zero-frequency peak to peak linewidth obtained in ferromagnetic resonance studies has a contribution due to Gilbert damping, and is a measure of energy dissipation in a thin film.) The thicknesses at which these transitions occur are certainly sensitive to deposition conditions which may explain the differences in observed damping constants.

However, films grown by sputtering are generally considered to be much smoother than thermally evaporated films. The invariant damping found by Chen et al. below 90 nm could be indicative that in this regime, thickness no longer dominates extrinsic damping and because the films studied were sputtered, the roughness of each film may not have changed significantly. (This serves as good motivation for measuring surface roughness.) In the thermally evaporated films grown for this work and by Rudge, roughness did change significantly within the same thickness regime, and a significant change in damping was observed. (Fig. 5.3)

Unfortunately, the differences between each of these experiments precludes a comparison between them from yielding any definitive knowledge. The experiment by Rudge used a resistive heating element for depositon, which resulted in NiFe thin films which displayed an anomalous out-of-plane easy axis. AFM scans indicated a growth mode that differed significantly both qualitatively and in roughness compared to the present work. Additionally, no composition data was obtained and the TR-MOKE data was captured in-situ, so the effect of oxidation is another unknown. Due to the number of variables that were changed across each experiment and their relative contributions to Gilbert damping (both extrinsic and intrinsic) being unknown, no additional insight into Gilbert damping can be obtained.

5.2 Fourier Analysis

The uniform, damped magnetization precession that is apparent in the TR-MOKE data can also be visualized in the frequency domain using a Fourier transform. The change in precessional frequency observed under different bias fields depends on the
Figure 5.3: Gilbert damping constant vs. roughness in NiFe thin films.
saturation magnetization $M_s$ of the film, and this can be measured.

Discrete Fourier transforms were performed on each TR-MOKE data set where precession was observed. Sample B was omitted from this analysis due to a lack of data with a clear precessional frequency. Each data set was cropped so that the initial precession and relaxation precession periods were analysed separately. A Hamming window was applied to each data set before transforming to minimize side lobes in the spectrum. DC offsets were also subtracted in the time domain. Fig. 5.4a and 5.4b show an example of cropped time domain data from TR-MOKE and the corresponding spectrum. The power spectral density was computed and the peak frequency in each spectrum was plotted against the bias field. (Fig. 5.6 and 5.5)

The peak frequency in each spectrum is the ferromagnetic resonance frequency $f_r$ (the uniform mode), and is related to the applied bias field $H$ by the Herring-Kittel equation [10],

$$f_r = \frac{\mu_0 \gamma}{2\pi} \sqrt{(H + H_k + M_s)(H + H_k)}.$$  \hspace{1cm} (5.3)

Due to the weak uniaxial anisotropy of polycrystalline NiFe, $H_k$ is small compared to $M_s$ and can be dropped. By plotting the precessional frequency during the magnetic field pulse in various bias fields, and fitting with Eqn. 5.3, the saturation magnetization $M_s$ can be determined.

At first glance, Fig. 5.6a and 5.6b appears disagree with Eqn. 5.3 because $f_r$ is decreasing with increasing bias field. However, because the bias field is directed opposite to the excitation field pulse (both of which are in-plane), the total effective field actually decreases with increasing bias field. This is compensated for in the fit with Eqn. 5.3 by substituting $H \rightarrow -H$. When the bias field and pulse field were applied in the same direction, no precession was observed, so no fit to the Herring-Kittel equation could be performed. The second precession that occurs after the excitation pulse is removed can also be fit with Eqn. 5.3, and because the effective field is increasing with the removal of the excitation pulse, the plotted precessional frequencies increase with bias field as expected.

The field pulse $H_{\text{pulse}}$ must be taken into account when fitting the data taken during the initial precession period in Fig. 5.6 and 5.5, so 5.3 becomes modified as

$$f_r = \frac{\mu_0 \gamma}{2\pi} \sqrt{(H_{\text{pulse}} - H_{\text{bias}} + M_s)(H_{\text{pulse}}-H_{\text{bias}})}.$$  \hspace{1cm} (5.4)

Extracted values of $M_s$ and $H_{\text{pulse}}$ are given in Table 5.1.
(a) Initial precession cropped for Fourier analysis. The dashed line indicates the start of the field pulse.

(b) Power spectral density of cropped precession.

Figure 5.4: Example TR-MOKE data obtained for Sample C in response to a 4-ns field pulse in a -150 Oe bias field.
The accepted value of $M_s$ for sputtered Ni$_{80}$Fe$_{20}$ films is 1.0 emu/cm$^3$, [26] but the composition of the films studied varies (see Table 4.1). However, the film with composition Ni$_{81}$Fe$_{19}$ (Sample A) was found to have $M_s = 1.1$ emu/cm$^3$ when measured with a positive pulse field applied, which is in good agreement with the expected value.

Figure 5.5: Precessional frequency vs. bias field in Sample A fit to the Herring-Kittel equation.
Figure 5.6: Precessional frequency vs. bias field in Sample C fit to the Herring-Kittel equation.

<table>
<thead>
<tr>
<th></th>
<th>$M_s$ (emu/cm$^3$)</th>
<th>$H_{\text{pulse}}$ (Oe)</th>
<th>Reduced $\chi^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample C, $H_{\text{pulse}} &gt; 0$</td>
<td>$1.47 \pm 0.16$</td>
<td>$487 \pm 35$</td>
<td>0.0312719</td>
</tr>
<tr>
<td>Sample C, $H_{\text{pulse}} &lt; 0$</td>
<td>$1.85 \pm 0.15$</td>
<td>$415 \pm 20$</td>
<td>0.0306726</td>
</tr>
<tr>
<td>Sample A, $H_{\text{pulse}} &gt; 0$</td>
<td>$1.1 \pm 0.14$</td>
<td>$325 \pm 15$</td>
<td>0.0822266</td>
</tr>
<tr>
<td>Sample A, $H_{\text{pulse}} &lt; 0$</td>
<td>$1.39 \pm 0.06$</td>
<td>$259 \pm 5$</td>
<td>0.00628782</td>
</tr>
</tbody>
</table>

Table 5.1: Parameters for fitting the Herring-Kittel equation to the initial precession data from Fig. 5.6 and 5.5.

A systematic discrepancy between the $M_s$ measured in positive and negative bias fields is seen in Table 5.1. Measurements made with a negative field pulse polarity
consistently yielded higher $M_s$ values along with reduced $\chi^2$ values closer to 0 indicating overfitting. Measurements with negative field pulses were always performed immediately after the positive polarity measurements, which may be a clue to the origin of the discrepancy. One possible explanation is that the bias field calibration was skewed over time as the bias field electromagnet heated and the coil resistance increased, however, this would have resulted in an increase in measured $M_s$.

The fitted values of $H_{\text{pulse}}$ are in reasonably good agreement with the earlier calculated theoretical value of 400 Oe. The drop in $H_{\text{pulse}}$ values with negative polarity are consistent with a 1 dB described in the manual of the picosecond pulse generator used. However, the significance of this data is in the technique itself. The magnetic field over a $\approx 10 \, \mu m$ area adjacent to a 12.5 $\mu m$ transmission line was successfully measured by exploiting the change in ferromagnetic resonance frequency due to the magnetic field induced by the transmission line. In other words, by coating a micro- or nano-structure in a ferromagnetic thin film and performing TR-MOKE measurements on that film with various applied bias fields, one can measure the magnetic field around that structure.
Chapter 6

Discussion

A thorough, systematic study of thickness or other extrinsic contributions to Gilbert damping will necessarily involve a number of challenges, which have contributed to the lack of such a study to date (this work being no exception). Though there were a number of inconsistencies discovered during analysis of the data obtained (which was made possible by thorough characterization), it is fortunate that these were found in order to improve future experiments. The main issues discovered relate to sample preparation and film growth.

The glass substrates were not chemically cleaned prior to the NiFe evaporation to preserve photoresist that capped the gold microcoil. This layer was intended to prevent shorting of the coil when the NiFe was deposited on top, but the necessity of this layer was cast in doubt both by the ability to make electrical connections to the microcoil through the top NiFe layer and SEM images showing possible flaking of the photoresist. Future work could be improved by adding a chemical cleaning step to remove dust and contamination from the glass substrate, without worrying about preserving the insulating photoresist layer.

Additional control during the fabrication of the microcoils could help ensure the more complete removal of gold and adhesion layer from the surface. Near the bottom of Fig. 4.4, there is a small region where the film morphology changes to be more homogeneous. This is due to a Ti adhesion layer being present in those regions, leftover from the fabrication of the microcoils. This visible difference in morphology due to an adhesion layer serves as motivation for future work using a Ti seed layer to control roughness.

It is unclear why focusing the laser spot in the middle of the microcoils did not once yield a TR-MOKE signal. The pulse field near the microcoil centre would have
much weaker than near the microcoil wire. Differing magnetic properties may explain why this technique did not work. The films studied earlier by Rudge were found to have an out-of-plane easy axis, which may have permitted a larger magnetic excitation with less pulse field. To contrast, the films in present work all showed an in-plane easy axis, and were shown to have been excited by an in-plane pulse field.

Improvements to the electromagnet configuration used to apply varying bias fields during TR-MOKE could be made. Real-time monitoring of the applied bias field should be used in the future to ensure the magnetic field not drift and that all electrical connections are working properly.

Most importantly, future work should avoid the use of RF induction evaporation, as characterization of the films produced by it have shown unwanted variation. Though the relationship between film thickness and Gilbert damping was the main focus of the analysis, film composition can also be compared with damping. The weight fraction of Ni was found to be different for each film, and though it is not well understood, this will contribute to intrinsic damping.

Unfortunately, it is not possible to separate the contribution of composition to intrinsic damping from the contribution of film thickness to extrinsic damping. Such differences in composition was not expected, and it suggests that thermal evaporation does not offer the control over film growth needed to carry out a systematic study on film properties that influence Gilbert damping. RF magnetron sputtering is preferred because it produces higher quality films with more consistency.

More generally, future work on Gilbert damping should not be performed unless it can be demonstrated beforehand that a single parameter of interest can be varied while holding all others fixed. While this may seem an obvious lesson, it is only made clear due to the thoroughness of the characterization performed in this work. Much of the body of existing work on Gilbert damping in NiFe thin films neglects characterization of composition and film structure, which complicates comparisons between studies.

Lastly, better thin film characterization techniques are needed. Although film thickness measurements were made successfully by AFM, a more reliable tool would be an in-situ quartz crystal growth monitor. Questions raised about the effect of film structure (such as grain size) on extrinsic damping could also be answered by studying these properties with transmission electron microscopy (TEM).
Chapter 7

Conclusion

Though it may be difficult to draw concrete conclusions about the nature of Gilbert damping from the quantitative analysis of the NiFe films studied, a great deal of practical knowledge about how to ensure future work is more systematic was gained.

An RF induction thermal evaporator was built and used to deposit three NiFe films on glass. Magnetic hysteresis curves and coercivities of these films were measured by MOKE. Time-resolved MOKE was then used to measure magnetization dynamics in each film by exciting the magnetization with a 4-ns field pulse coming from a microcoil patterned on each glass substrate. TR-MOKE measurements were made in varying bias fields, and the Gilbert damping constant (in zero field) and saturation magnetization of each film were extracted. The strength of the excitation pulse field was also measured, and was found to be in good agreement with theory. The thickness and roughness of each film was measured with AFM, and the composition was determined by EDX.

The Gilbert damping constant in NiFe thin films was found to increase with film thickness, but this trend cannot definitively be attributed to thickness alone because the weight fraction of Ni also increased in the films studied. A comparison with existing literature supports a strong dependence of Gilbert damping on thickness, but suggests that the internal structure (both magnetic and granular) of thin films between 5 - 500 nm must be better characterized. Lastly, to ensure more consistent film composition in future work, RF magnetron sputtering is recommended in place of RF induction evaporation.
Appendix A

Future Work

Future experiments to continue investigating the thickness contribution to extrinsic Gilbert damping can take advantage of the lessons learned in this work. Through some clever simplifications, a more effective experiment with better controls can be carried out.

One way to perform a similar experiment in the future could be to grow a single thick (eg. 500 nm) NiFe film, perform the full set of characterization experiments on that film, and then etch it using a plasma etcher to reduce the thickness. After removing some amount (eg. 20 nm) of material from the film, the characterization experiments would be performed again, and this cycle would be repeated until it a MOKE signal could no longer be detected from the sample. The key advantage of this technique would be composition being held fixed through the use of only a single film, unless there was significant preferential etching of Ni or Fe. Another advantage would be that etching could provide a consistent surface roughness for the film at different thicknesses (in contrast to the data in Table 4.1). Since only a single microcoil would be required, the time spent on sample preparation would also be reduced. Combined, these simplifications would drastically speed up future work while yielding better results due to tighter controls.

Future work should also investigate the use of transmission lines instead of microcoils for the production of the pump magnetic field. Using in-plane excitation fields from transmission lines should provide stronger fields and the increased reflectivity of having a gold underlayer may contribute to larger TR-MOKE signals.
Bibliography


