Optical control of ultrafast spin-wave relaxation by magnetic anisotropy in a ferromagnet

Kevin James Smith
College of William & Mary - Arts & Sciences

Follow this and additional works at: https://scholarworks.wm.edu/etd

Part of the Electromagnetics and Photonics Commons, and the Physics Commons

Recommended Citation
Smith, Kevin James, "Optical control of ultrafast spin-wave relaxation by magnetic anisotropy in a ferromagnet" (2010). Dissertations, Theses, and Masters Projects. Paper 1539623568.
https://dx.doi.org/doi:10.21220/s2-2jqz-0v96
Optical Control of Ultrafast Spin-wave Relaxation by Magnetic Anisotropy in a Ferromagnet

Kevin James Smith

Richmond, Virginia

Bachelor of Science, The College of William and Mary, 2003
Master of Science, The College of William and Mary, 2005

A Dissertation presented to the Graduate Faculty of the College of William and Mary in Candidacy for the Degree of Doctor of Philosophy

Department of Physics

The College of William and Mary
May 2010
This Dissertation is submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy

Kevin James Smith

Approved by the Committee, April 2010

Professor Gunter Lüpke, Applied Science
The College of William and Mary

Professor William J. Kossler, Physics
The College of William and Mary

Professor R. A. Lukaszew, Applied Science, Physics
The College of William and Mary

Assistant Professor Irina Novikova, Physics
The College of William and Mary

Professor Mark K. Hinders, Applied Science
The College of William and Mary
This thesis presents an investigation of the damping of spin waves in ferromagnetic Au(3 nm)/Ni(10 nm)/MgO(001) thin films using the time-resolved Magneto-optical Kerr Effect (TR-MOKE) and ferromagnetic resonance (FMR) techniques. In the optical measurements, a 150 fs, 800 nm laser beam pulse is split into pump and probe components. The pump pulse, containing most of the beam energy, thermally excites coherent spin precession. The weaker probe pulse, time-delayed by a variable beam path, captures the magnetization dynamics via the polar MOKE effect, and oscillations are observed as a function of external field amplitude and direction. The extracted precession frequency is consistent in both the optical and resonance techniques; however, additional damping is observed in the TR-MOKE measurements that is strongly correlated to the orientation of the magnetization with respect to the magnetic anisotropy. The damping is identical in TR-MOKE and FMR only when the external field is applied near the easy axis of magnetization. The enhanced damping in TR-MOKE is shown to be a consequence of pump-induced inhomogeneous broadening in the presence of magnetic anisotropy, a result of differing temperature recovery profiles for the magnetization and magnetic anisotropy. Finally, a simple model is developed which explains the anisotropic damping: mode broadening occurs in regions where the magnetization changes rapidly with respect to changes in the external field, as determined by the curvature of the magnetic free energy. We thus introduce a novel damping effect in TR-MOKE: pump-induced anisotropic damping (PIAD).
This work is dedicated to my family.
# Table of Contents

Abstract iii  

Acknowledgements xii  

1 Introduction 1  
   1.1 Magnetoelectronics ................................................. 1  
      1.1.1 Optical Excitation ........................................... 5  
      1.1.2 Spin Wave Relaxation ....................................... 6  
   1.2 Overview of Thesis .............................................. 7  

2 Theoretical Considerations 9  
   2.1 Ferromagnetism ................................................... 9  
   2.2 Magnetic Anisotropy .............................................. 14  
      2.2.1 Magnetocrystalline Anisotropy ............................... 15  
      2.2.2 Shape Anisotropy ............................................ 17  
      2.2.3 Exchange Bias ............................................... 19  
   2.3 Spin-Wave Dynamics .............................................. 20  
   2.4 Damping .......................................................... 23
2.4.1 Intrinsic Damping

2.4.2 Extrinsic Damping

3 Samples and Experimental Methods

3.1 Sample Growth and Characterization

3.2 Magneto-Optical Kerr Effect Technique

3.2.1 Static MOKE

3.2.2 Time-Resolved MOKE

3.3 Ferromagnetic Resonance

4 Results and Discussion

4.1 Static MOKE

4.2 TR-MOKE

4.2.1 In-plane TR-MOKE

4.2.2 Out-of-plane TR-MOKE

4.3 Ferromagnetic Resonance

4.4 Pump Induced Anisotropic Damping

5 Conclusion

5.1 Summary

5.2 Outlook

5.2.1 Unidirectional Anisotropy

5.2.2 Meta materials

5.2.3 Half Metals
List of Tables

2.1 Magnetocrystalline Energy (MCA) of various crystals. In the following expressions, $\alpha_1$, $\alpha_2$, $\alpha_3$ represent the directional cosines; $\theta$ is the polar angle, and $\phi$ is the azimuthal angle in spherical polar coordinates.

Reproduced from [1]. ............................................ 16

3.1 Fresnel coefficients. In the below expressions, $\alpha_1 = \cos \theta$, where $\theta$ is the angle between the ray and the film normal. $\alpha_2 = \sqrt{1 - \sin^2 \theta/n^2}$. $Q_p, Q_t, Q_l$ are the polar, transverse, and longitudinal projections of the Voigt vector, $Q$, which is proportional to the magnetization. ........ 34
List of Figures

1.1 Representation of the GMR [2, 3] effect in a spin valve. ............... 3

2.1 Schematic of density of states for a transition metal ferromagnet ... 10

2.2 The spontaneous magnetization, $M(T)$ as calculated graphically from equations 2.4 and 2.6 for Ni. ................................. 13

2.3 Isotropy of the exchange interaction ................................. 14

2.4 Anisotropy in a cubic crystal ................................. 15

2.5 Typical relative temperature dependence of the magnetocrystalline anisotropy, as calculated from Eq. 2.9 for the case of cubic anisotropy $(n = 4)$. ................................. 17

2.6 H Field of a Prolate Spheroid leads to demagnetization ............... 19

2.7 Schematic of a spin wave with $k \neq 0$ propagating through a lattice of spacing $a$. ................................. 20
2.8 Precession of a magnetic moment, \( \mathbf{m} \), in the presence of an external field, \( \mathbf{H}_{\text{ext}} \). The equation of motion (Eq. 2.20) contains two torque terms: the first (yellow) causes the moment to precess with frequency \( \omega \); the second (pink arrow) causes the magnetization to relax to the equilibrium direction (red arrow), determined by the effective field, \( \mathbf{H}_{\text{eff}} \).

2.9 Scattering between magnons, electrons and phonons results in the transfer of energy and damping.

2.10 Schematic of the two-magnon scattering process. In general, the dispersion relation (blue and green lines) will depend on the direction of \( \mathbf{M} \). When \( \mathbf{M} \) is in plane (green line), there is a degeneracy, and scattering from points \( k = 0 \) (A) to \( k \neq 0 \) (B) can occur (dashed red line). If, however, \( \mathbf{M} \) is tilted significantly out of the sample plane (blue line), the degeneracy is broken and scattering does not occur.

3.1 Two equivalent representations of the MOKE effect.

3.2 MOKE geometry.

3.3 Schematic of the static MOKE experiment.

3.4 Schematic of two color pump-probe TR-MOKE experiment.

3.5 A typical pump-probe TR-MOKE scan.

3.6 Schematic of a FMR experiment.

3.7 A typical FMR scan.

4.1 Coercivity vs. applied field angle, \( \phi_H \) as measured by SMOKE.

4.2 TR-MOKE: \( \phi_H = 0, \theta_H \approx 90 \) vs. \( \mathbf{H}_{\text{ext}} \).

4.3 TR-MOKE: \( \phi_H = 45, \theta_H \approx 90 \) vs. \( \mathbf{H}_{\text{ext}} \).
4.4 TR-MOKE: $\omega_{\text{MO}}$ vs. $H_{\text{ext}}$ for $\phi_H = 0^\circ, 10^\circ, 20^\circ, 30^\circ, 40^\circ, 45^\circ, \theta_H \approx 90^\circ$. 49
4.5 TR-MOKE: $\tau_{\text{MO}}$ vs. $H_{\text{ext}}$ for $\phi_H = 0^\circ, 10^\circ, 20^\circ, 30^\circ, 40^\circ, 45^\circ, \theta_H \approx 90^\circ$. 50
4.6 TR-MOKE: $\phi_H = 0^\circ, \theta_H \approx 0^\circ$ vs. $H_{\text{ext}}$ ................................. 52
4.7 TR-MOKE: $\omega_{\text{MO}}$ vs. $H_{\text{ext}}$ for $\theta_H \approx 0^\circ$ ................................. 53
4.8 TR-MOKE: $\tau_{\text{MO}}$ vs. $H_{\text{ext}}$ for $\theta_H \approx 0^\circ$ ................................. 53
4.9 FMR: $H_0$ vs. $\theta_H$ for Easy and Hard Axes ................................. 56
4.10 FMR: $\Delta H$ vs. $\theta_H$ for Easy and Hard Axes ................................. 57
4.11 The extracted decay time, $\tau_{\text{FMR}}$ vs. $\theta_H$ for easy (blue squares) and hard (red circles) Axes. The rapid decrease at $\theta_H \approx 35^\circ$ is due to the onset of Two-Magnon scattering [4, 5]. ................................. 57
4.12 The equilibrium direction of magnetization, $\theta^*$, vs. applied field direction, $\theta_H$, as calculated for the Easy (blue line) and Hard (red line) Axes. The direction of the magnetization vector changes rapidly for $\theta_H < 35^\circ$, and is defined as the direction that minimizes the magnetic free energy (Eq. 4.1). ................................. 58
4.13 Comparison of the temperature dependence of $M$ and $K$ ................................. 60
4.14 Pump-induced anisotropic damping for easy, hard, and normal geometries 61
4.15 FMR vs. TR-MOKE measured decay time for $\omega = 60.6$ GHz. FMR data (easy axis: $<$; hard axis: $*$) yields the characteristic Two-Magnon Scattering curve [4, 5] (solid line is guide to the eye) as the external field is swept normal to the film plane. TR-MOKE data yields similar results to FMR for the in-plane easy axis ($\triangle$), yet is inconsistent with FMR data for the hard ($\triangledown$) and normal ($\square$) axes. Error bars represent estimated standard error of the mean. ................................. 62
4.16 TR-MOKE dynamics for in-plane easy (Δ) in-plane hard (▽), and normal (□) geometries. Panel (a): observed decay time vs. external field magnitude. Panel (b): observed decay time vs. center frequency $\omega_{\text{MO}}$. Solid lines are fits to data, as discussed in the text. Panel (c): center frequency vs. the external field. Error bars represent one standard error of the mean. .............................................. 65

4.17 TR-MOKE decay time vs. in-plane external field angle for $B = 73$ mT. The solid line is calculated from the magnetic anisotropy, as discussed in the text. .............................................. 67
Acknowledgements

I am grateful to the many people who have directly or indirectly supported me during the completion of this work. First and foremost, I am especially indebted to my adviser, professor Gunter Lüpke, for his guidance during the years that I have been a member of his research group. He has always provided enthusiasm and perspective for the research process, and this work would not be possible without his support and guidance.

I am particularly grateful for the generosity of professor Ale Lukaszew and the members of her research group, Mr. J. R. Skuza, Dr. C. Clavero, and Mr. K. Yang. Their hard work provided the samples and the sample characterization necessary for this thesis, and they were a constant source of helpful and constructive feedback. Additionally, I would like to thank professor Lukaszew for the encouragement and advice that was so critical to the completion of this project.

I am similarly grateful for the kindness of professor Anne Reilly, Dr. Keoki Seu, and professor Haibin Zhao. They were ever sources of immediate, positive, and constructive encouragement; I am grateful for their support and advice.

I am indebted to Professor Natalia Noginova and Mr. Osei Amponsah of Norfolk State University for generously providing access to their FMR apparatus, and for their useful advice and input regarding the interpretation of the data.

I would also like to thank the other members of the Lüpke research group – especially Mr. Erik Spahr, Mr. Wei Zheng, Mr. Y. Fan and Mr. A. Petersen – for fostering an enjoyable and productive work environment. Additionally I am grateful to Mr. Petersen and Mr. Fan for their help with data collection.

I would also like to thank the committee members for their participation, comments, and questions in the review of this dissertation.

Finally, I wish to express my sincere gratitude to my friends and family; their constant love, encouragement and support are the source of my strength and motivation.
Chapter 1

Introduction

1.1 Magnetoelectronics

Modern society is to a large degree the product of a technological revolution precipitated by scientific experiments on electricity and magnetism conducted in the seventeenth and eighteenth centuries [6]. Likewise, the subsequent discovery of quantum mechanics and charge quantization led to the present information revolution through the development of semiconductor electronics. While this enormous progress yielded fundamental inventions such as the light bulb, the computer, and the microchip, engineers have only until very recently utilized the electric charge (or current). It is well known, however, that in addition to charge, electrons possess an intrinsic angular momentum (spin) and a corresponding magnetic moment. While current technology is based on only one half of the electron’s properties, one very promising emerging technology – spintronics – aims to utilize the best of both of these fundamental characteristics to construct spin-dependent electronic transport (spintronic) devices [7, 8].
This melding of standard microelectronics with spin-dependent effects opens the door to other revolutionary new technologies, such as quantum computation and quantum communication [7, 9, 10, 11, 12].

Magnetoelectronics has already begun to revolutionize information technology through advances in magnetic data storage. The capacity of magnetic hard disk recording drives increased exponentially over the past two decades - from 0.1 GB in 1990 to 1000 GB in 2010. This advance is a direct consequence of the discovery of the spin-dependent giant magnetoresistance (GMR) effect [2, 3], which was first discovered in the late 1980s. In this effect, spin dependent transport manifests an increase in the resistivity of a multilayer stack (consisting of a normal metal sandwiched between two conducting ferromagnetic layers) as the magnetization of the ferromagnetic layers is switched from parallel to anti-parallel alignment, as shown schematically in Fig. 1.1.

GMR is relevant to the magnetic storage industry because of its application in a practical device, the spin valve, which acts as a magnetic sensor. In this device, the magnetization of one of the ferromagnetic layers is fixed, or “pinned” (an antiferromagnetic layer and the exchange bias effect, discussed in section 2.2.3 are commonly used [13]). The other ferromagnetic layer, constructed out of a material that is easily magnetized, is free to align in a local external field. In magnetic hard disk drives, information is stored as magnetic bits on a track and the spin valve is placed above the bit track with the unpinned layer in close proximity to the magnetic recording medium. This “free” layer will align with the magnetic state of the underlying bit; by rotating the medium and monitoring the resistance through the valve, one can read out the recorded magnetization states.
1.1 Magnetoelectronics

Figure 1.1: Representation of the GMR [2, 3] effect in a spin valve. Top: A non-magnetic conducting layer (white) is sandwiched between two ferromagnetic layers (grey), which are aligned parallel (left) or anti-parallel (right). A current (consisting of “up” and “down” spins) is sent through the valve; scattering is more likely to occur when the spin is anti-aligned with the magnetic layer. Bottom: schematic of an equivalent circuit, where \( R_1 < R_2 \). The parallel alignment (left) results in a lower resistivity \( (R_{\text{parallel}} = \frac{2R_1R_2}{R_1+R_2}) \), than the anti-parallel (right, \( R_{\text{anti}} = \frac{1}{2}(R_1 + R_2) \)) case.

Modern drive heads employ magnetic tunneling junctions (MTJs) [14], which are spin valves in which the spacing layer is made thin enough (typically a few nm) that electrons can tunnel from one ferromagnetic layer to the other. Recent advances in the fabrication of MTJ structures have put the relative resistivity ratio at nearly 600% at room temperature [15] for Fe/MgO/FeCo(001).

Another, more recent application of MTJs is in integrated microcircuits as magnetic random access memory (MRAM) [7, 16, 17]. Like hard disk drives, MRAM
stores information as magnetic states and thus does not require a constant power source - a written state is stable until “flipped” by a magnetic field [18]. MRAM thus combines the stability of hard disk recording with the fast readout time of random access memory (reports of access times have been as much as 10000 times faster than that of hard disk drives [7]). Furthermore, there is the possibility of incorporating spin-dependent transport effects to reduce the power needed to read and write e.g., Spin-Torque Transfer (STT) [19], which enables a spin-polarized current to flip the magnetization state.

One bottleneck in the development of magnetoelectronics devices is a lack of a complete understanding of magnetization switching and control, since the spin relaxation time governs the ultimate timescale at which these devices can manipulate information [20]. There are two facets to this problem: first, the identification of novel mechanisms to excite spin waves, or spin switching, and second, the understanding of pathways for spin relaxation or damping. Thus an understanding of magnetization dynamics is of critical importance as devices move into the GHz regime.

To these ends, there has been a wealth of activity in utilizing ultrafast lasers to excite and probe magnetization dynamics, [21, 22, 23, 24, 25, 26, 27] since one can access the dynamics on the femtosecond timescale with these tools. Additionally, the possibility of coupling magnetoelectronics to magneto-optics or optoelectronics suggests additional avenues of exploration. Furthermore, the development of compact ultrafast lasers [28] suggests that optical sources might supersede current [29, 30] or field [31, 32, 33] pulses for spin control.
1.1.1 Optical Excitation

In addition to the above mentioned technological benefits, identifying the fundamental limit of magnetization switching is also an intriguing problem in the theory of magnetism [34, 35, 36, 37, 38, 39, 40]. While general work in this field dates back nearly a half a century, femtosecond laser pulses were first used to alter the magnetization state in transition metal ferromagnets in the late 1990s [41, 42, 43, 44, 45, 46]. In these experiments, it was found that the optical pulse generated thermal heating and reduced magnetic order on the timescale of ≈ 2 ps, which is much faster than what was initially expected from traditional near-equilibrium experiments [20]. Of major concern was whether the observed demagnetization was the result of a direct coupling between the photon field and the spins, or an indirect consequence of thermal heating [46, 47]. Th. Rasing et al. [48] demonstrated the use of circularly-polarized femtosecond laser pulse to control magnetization switching directly in DyFeO$_3$ through the inverse Faraday effect [49, 50], which couples the angular momentum of the laser light to the magnetic medium. They found that state reversal occurs for times less than ≈ 200 fs. This experiment showed that the magnetization state switches linearly, i.e., without ringing oscillations. Very recent work [51] showed that this method can be used to store and read information within 30 ps for a 5 μm domain. While these results are very promising, a much larger Faraday constant is needed for linear reversal than what is characteristic of transition metal ferromagnets. Nevertheless, other very recent work [52, 53] found that the field of a single femtosecond optical pulse coherently couples to the spins in transition metal ferromagnets (such as Ni) beyond the standard spin-orbit interaction within the first few femtoseconds, prior to thermalization.
1.1 Magnetoelectronics

1.1.2 Spin Wave Relaxation

There has been a similar wealth of work regarding the use of femtosecond laser techniques to probe "long time" (≈ ns) magnetization dynamics: M. Van Kampen, et al. [54] demonstrated that an optical pump pulse excites coherent uniform spin precession, i.e. the ferromagnetic resonance (FMR) mode, by thermally altering the magnetic anisotropy and measuring the subsequent dynamics in the time domain using the time-resolved magneto-optical Kerr effect (TR-MOKE). The details of this excitation mechanism are discussed in sections 3.2.2 and 4.4. Initial work showed that the thermal heating caused by the pump pulse is sufficient to perturb the system to far from equilibrium for times as long as a few nanoseconds. In this regime, the magnetization magnitude may not be conserved. Nevertheless, for sufficiently low laser fluences [55], the Landau-Lifshitz Gilbert (LLG) formalism (section 2.3), in which the magnetization is conserved, captures the precession dynamics very well [56], and can be used to identify the precession frequency in a manner similar to FMR.

Later work was concerned with exploring other magnetization relaxation channels [57, 58, 59, 60]. One example is the non-local transport of angular momentum via Spin-Torque Transfer [61], important in MRAM applications, as described above. Additional work investigated anisotropy in the observed damping, which was thought to have its origin in magnon-magnon scattering at defects [62].

Despite this progress there is still an incomplete understanding of the active relaxation channels in the optical pump-probe technique, since reports have shown that TR-MOKE often yields stronger damping than the traditional FMR technique [61, 63, 59, 64, 65]. It is thus important to identify and understand the dominant damping mechanism present in ultrafast pump-probe optical experiments.
1.2 Overview of Thesis

The goal of this dissertation was to use pump-probe TR-MOKE to explore and identify the spin-wave damping mechanisms present in a ferromagnet. This study led to the discovery of a pump-induced anisotropic damping (PIAD) effect, in which the optical pulse couples to the magnetic anisotropy to broaden the excited mode distribution as a function of the angle between the external field and the easy axis, resulting in enhanced damping. This thesis will detail these findings in the subsequent chapters.

Chapter two reviews the concepts relating to magnetism and magnetization dynamics which form a relevant theoretical background: ferromagnetism, magnetic anisotropy and spin wave dynamics. Finally, various mechanisms for spin-wave relaxation are reviewed.

Chapter three is a presentation of the samples and experimental methods used for this thesis. Sample growth and characterization is first presented, followed by discussions of the pump-probe technique, the magneto-optical Kerr effect, and the apparatus used in this thesis. Finally, a complementary technique, Ferromagnetic Resonance, is detailed.

Chapter four comprises the heart of this work: the results of the experiments and a detailed description of the novel damping mechanism. The enhanced anisotropic damping is found to be unique to TR-MOKE through contrasting the results of the optical experiments with FMR. The pump-induced anisotropic PIAD effect is shown to follow the symmetry of the underlying magnetocrystalline anisotropy. A simple model is presented that links the damping to the anisotropy by coupling the free energy to the excited mode distribution.
1.2 Overview of Thesis

Chapter five concludes this thesis by summarizing the findings of the previous chapters and suggesting future avenues for research.
Chapter 2

Theoretical Considerations

This chapter introduces the concepts that are important to the rest of this work; magnetization, as defined in itinerant ferromagnetism, is discussed briefly within the Stoner model. Next, various sources of magnetic anisotropy are introduced, and finally, a framework for magnetization dynamics is presented, with an emphasis on the current understanding of magnetization relaxation. The following resources may be helpful for further reading: ref. [66] contains a basic survey of magnetism and magnetic materials; ref. [67] is comprehensive, with an emphasis on material properties; and ref. [68] provides a generous and detailed discussion of the microscopic origin of magnetism.

2.1 Ferromagnetism

Magnetism is a consequence of three primary interactions: the exchange interaction between spins, responsible for spin alignment; the spin-orbit interaction, which indirectly couples the spins to the lattice and generates magnetic anisotropy; and the
Figure 2.1: Schematic of spin densities of states for 3\textit{d} electrons in a transition metal ferromagnet. A shift in the density of states due to the exchange interaction results in a net spin polarization. Here spins are shown quantized relative to the direction of an external field.

Zeeman interaction, which governs material behavior in an external field, \textbf{H}. For the transition metal ferromagnets, Fe, Ni, and Co, the simultaneous occupancy of both localized and delocalized electronic states precludes a simple model for room temperature ferromagnetism, and a band-theory approach is adopted, as first applied by Mott [69], Slater [70, 71] and Stoner [72, 73]. Pauli exclusion, in combination with bond hybridization and the Coulomb interaction, results in an exchange interaction that promotes one spin orientation of the 3\textit{d} band over the other, with a rigid shift in the density of states by a factor of the Stoner parameter, \textit{I}. If the density of states is such that a transfer of electrons from a spin-opposite to spin-parallel state is energetically favorable, a net spin polarization will manifest itself, as shown in Fig
2.1 Ferromagnetism

Stoner showed [73] that this process will occur if

\[ \text{IN}(E_f) > 1, \]  

(2.1)

where \( N(E_f) \) is the non-magnetic density of states evaluated at the Fermi energy. For Fe, Co, and Ni, \( \text{IN}(E_f) = 1.43, 1.70 \) and \( 2.04 \), respectively. The induced magnetization, \( M \), is simply the difference of the number of electrons in each state:

\[ M = \mu_B (N_+ - N_-), \]

(2.2)

where \( N_+ \) and \( N_- \) represent the number of spin-up and spin-down electrons, respectively.

In the above picture, the Stoner exchange energy (i.e., the spin flip energy) is the energy required to reorient one spin of a single 3d electron, and is on the order of 1 eV [68]. Macroscopically (averaged over many spins), the exchange energy can be thought of as manifesting as a molecular field that permeates the sample. This macroscopic picture was developed by Weiss and scales the intensity of the molecular field present in the sample to the sample magnetization: \( H_w = CM \).

The Weiss field is useful for determining the temperature dependence of the magnetic susceptibility, \( \chi \). In the Curie-Weiss formalism, \( \chi \) suffers a singularity at the Curie temperature, \( T_c \), the point at which thermal effects overcome long-range ordering:

\[ \chi = \frac{M}{H} \propto \frac{1}{T - T_c}. \]

(2.3)

Above \( T_c \), thermal effects destroy the "intrinsic" spin polarization, and the sample
behave as a paramagnet; the internal magnetization is simply proportional to the external field.

The temperature dependence of the magnetization below the Curie temperature is derived from statistical mechanical arguments [67]. The thermodynamic average of $M$ in a net field $H_{\text{eff}} = H_{\text{ext}} + H_w = H_{\text{ext}} + CM$ is given by the Brillouin function, $B_J(\alpha)$ [67]:

$$M = M_0 B \left[ \frac{2J + 1}{2J} \coth \left( \frac{2J + 1}{2J} \alpha \right) - \frac{1}{2J} \coth \left( \frac{\alpha}{2J} \right) \right]$$

$$= M_0 B J(\alpha),$$

(2.4) (2.5)

where $\alpha = \frac{M_0 (H_{\text{ext}} + CM)}{kT}$ is a dimensionless parameter. In the above expression, $J = \frac{1}{2}$, $M_0$ is magnetization at $T = 0$, and $k$ is the Boltzmann constant. Since the equation for $\alpha$ also involves $M$, one can rewrite it as

$$M = \frac{kT}{MC} \alpha - \frac{H_{\text{ext}}}{C}.$$  

(2.6)

The parameter $\alpha$, present in both eq. 2.4 and eq. 2.6, is eliminated by solving these two equations simultaneously, obtaining $M(T)$. Figure 2.2 contains a plot of $M$ vs $T$ for Ni.
Figure 2.2: The spontaneous magnetization, $M(T)$ as calculated graphically from equations 2.4 and 2.6 for Ni.
2.2 Magnetic Anisotropy

Figure 2.3: If spins are subject to exchange only, system (a) is energetically equivalent to system (b); there is no preferred magnetization direction.

In the Heisenberg model of magnetism [67], the Coulomb repulsion between electrons and the Pauli exclusion principle are modeled as a spin-spin exchange energy:

\[ E_{i,j} = -2JS_i \cdot S_j = -2JS^2 \cos \phi, \]

where \( \phi \) is the angle between the spins \( S_i \) and \( S_j \), and \( S \) is the spin magnitude. This energy is minimized (for \( J > 0 \), which is the case for ferromagnets) if all spins are parallel. Since there is no overall preferred direction of the magnetization (Fig. 2.3), the exchange interaction is isotropic. Nevertheless, real ferromagnetic materials often exhibit a preferred direction of magnetization, and an external field is required to orient the magnetization in an arbitrary direction. Furthermore, shape is also an important factor. Magnetic anisotropy is an important factor that determines the utility of magnetic materials, and is critical in many applications, such as spin valves and magnetic data storage. This section will outline the more important aspects of anisotropy.
2.2 Magnetic Anisotropy

2.2.1 Magnetocrystalline Anisotropy

The coupling of spins to their orbital angular momenta is responsible for the energy required to reorient the magnetization from the easy axis to the hard axis. This change in energy is referred to as the magnetocrystalline anisotropy (MCA) energy, as it reflects the underlying crystalline symmetry [74]. In cubic systems, such as Ni and Fe, the MCA energy can be expressed as a power series of the directional cosines \( \alpha_1, \alpha_2, \alpha_3 \) of the magnetization with respect to the cubic axes; the high degree of symmetry results in many simplifications (e.g. inversion symmetry precludes any

Figure 2.4: Schematic of Anisotropy Energy. The relative magnetocrystalline energy of an FCC crystal (black dots represent atom centers) with \( K_1, K_2 < 0 \) is plotted on a unit sphere. An easy axis corresponds energy minima (blue); conversely, a hard axis corresponds to energy maxima (red). For this case, the global easy axis is along \((111)\).
2.2 Magnetic Anisotropy

terms odd in $\alpha$):

$$E = K_1 \left( \alpha_1^2 \alpha_2^2 + \alpha_1^2 \alpha_3^2 + \alpha_2^2 \alpha_3^2 \right) + K_2 \left( \alpha_1^2 \alpha_2^2 \alpha_3^2 \right) + O(\alpha^6), \quad (2.8)$$

where $K_1$ and $K_2$ are the cubic anisotropy constants. For Nickel at 296 K, $K_1 = -5.7 \times 10^3 \text{ J m}^{-3}$, and $K_2 = -2.3 \times 10^3 \text{ J m}^{-3}$. Figure 2.4 plots this energy on a unit sphere, showing the location of the easy and hard axes (along (111) and (001) directions, respectively). Table 2.1 contains a compilation of the MCA energy for various other symmetry types.

Table 2.1: Magnetocrystalline Energy (MCA) of various crystals. In the following expressions, $\alpha_1$, $\alpha_2$, $\alpha_3$ represent the directional cosines; $\theta$ is the polar angle, and $\phi$ is the azimuthal angle in spherical polar coordinates. Reproduced from [1].

<table>
<thead>
<tr>
<th>Crystal class</th>
<th>example</th>
<th>Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cubic</td>
<td>Fe, Ni</td>
<td>$K_1 \left( \alpha_1^2 \alpha_2^2 + \alpha_1^2 \alpha_3^2 + \alpha_2^2 \alpha_3^2 \right) + K_2 \left( \alpha_1^2 \alpha_2^2 \alpha_3^2 \right) + \ldots$</td>
</tr>
<tr>
<td>Tetragonal</td>
<td>Ni, Fe, Co/Cu(001)</td>
<td>$-\frac{1}{2} K_2 \alpha_3^2 - \frac{1}{2} K_{44} \alpha_1^4 - \frac{1}{2} K_{4\parallel} \left( \alpha_1^4 + \alpha_3^4 \right) + \ldots$</td>
</tr>
<tr>
<td>Hexagonal</td>
<td>Co</td>
<td>$K_2 \sin^2 \theta + K_4 \sin^4 \theta$ $+ K_{6\parallel} \sin^6 \theta + K_{6\parallel} \cos^6 \phi \sin^6 \theta + \ldots$</td>
</tr>
<tr>
<td>Trigonal</td>
<td>$\alpha - \text{Fe}_2\text{O}_3$</td>
<td>$K_2 \sin^2 \theta + K_4 \sin^4 \theta + K_{6\parallel} \sin^6 \theta$ $+ K_{4\parallel} \sin^3 \theta \cos \theta \cos^3 \phi + K_{6\parallel} \cos^6 \phi \sin^6 \theta$ $+ K_{6,3} \sin^3 \theta \cos^3 \theta \cos^3 \phi + \ldots$</td>
</tr>
<tr>
<td>Orthorhombic</td>
<td>Fe$_3$O$_4$</td>
<td>$\sin^2 \theta \left( k_1 \cos^2 \phi + k_2 \sin^2 \phi \right)$ $+ \sin^4 \theta \left( k_3 \cos^2 \phi + k_4 \sin^2 \phi \cos^2 \phi + k_2 \sin^4 \phi \right)$ $+ \sin^2 \theta \cos^2 \theta \left( k_6 \cos^2 \phi + k_7 \sin^2 \phi \right) + \ldots$</td>
</tr>
</tbody>
</table>

The MCA energy is more responsive to temperature than the sample magnetization for $T \ll T_C$, as it is influenced by the temperature dependence of the lattice, the valence states, and thermal excitation of the spins [67]. In general, the anisotropy
2.2 Magnetic Anisotropy

Figure 2.5: Typical relative temperature dependence of the magnetocrystalline anisotropy, as calculated from Eq. 2.9 for the case of cubic anisotropy ($n = 4$).

Constants follow a power law [75]:

$$K^{(n)} \propto M_s^{n(n+1)/2},$$  \hspace{1cm} (2.9)

where $n$ denotes the order of the anisotropy constant, e.g., for cubic crystals, $n = 4$.

2.2.2 Shape Anisotropy

When a sample is placed in an external magnetic field, surface magnetic free poles will generate an internal field opposing the sample magnetization (Fig. 2.6). This field is known as the demagnetization field, $H_d$, and is proportional to the magnetic
free pole density (and hence the magnetization):

\[ H_d = -\frac{N}{\mu_0} \mathbf{M}, \]  

(2.10)

where \( N \), the demagnetization factor, is a tensor that depends only on the sample geometry. The true field inside the sample is less than the external field:

\[ \mathbf{H} = \mathbf{H}_{\text{ext}} - \frac{N}{\mu_0} \mathbf{M}. \]  

(2.11)

Osborn [76] found the form of \( N \) for a general ellipsoid; in the special case of an infinite, thin slab (i.e., a thin film), where the \( z \)-axis corresponds to the film normal,

\[ N = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \]  

(2.12)

and the magnetic energy corresponding to the demagnetization field is

\[ E_d = -H_d \cdot \mathbf{M} = \frac{M^2}{\mu_0} \cos^2 \theta, \]  

(2.13)

where \( \theta \) represents the angle between the magnetization and the film normal. The demagnetization field can be large; real materials reduce their total energy by forming magnetic domains. In this case, it is possible to achieve a net magnetization of zero while the magnet is locally saturated, as required by the Curie-Weiss law for temperatures below \( T_c \). When a sample is placed in a magnetic field, domains parallel to the external field grow at the expense of others; if the external field is large enough
2.2 Magnetic Anisotropy

Figure 2.6: The surface magnetic free poles induced by $H_{\text{ext}}$ generate a field (broken lines). The internal field, $H_d$ opposes $M$ and acts to demagnetize the sample.

to overcome all of the domains, the sample magnetization will saturate at $M_s$. Ref. [67] provides a detailed discussion of domain formation and displacement.

2.2.3 Exchange Bias

When a ferromagnet is layered on an antiferromagnet, exchange at the interface leads to a unidirectional anisotropy in the FM layer:

$$E_{\text{exb}} = -M \cdot H_{\text{exb}} = -M_s H_{\text{exb}} \cos \theta, \quad (2.14)$$

where $\theta$ represents the angle between the magnetization and the exchange bias field, $H_{\text{exb}}$. The addition of a unidirectional anisotropy results in a shift in the hysteresis $M$ vs $H$ loop by $H_{\text{exb}}$ in the direction opposing the field, and is important in magnetic devices, such as magnetic memories, since a ferromagnetic layer can be “pinned” by the exchange coupling at the AFM/FM interface.
2.3 Spin-Wave Dynamics

Spin-Waves (magnons) are collective excitations of spins that carry momentum and energy (figure 2.7). Their existence can be inferred from the time evolution of the spin Hamiltonian, \( H \):

\[
\dot{S}_i = i \hbar \frac{\partial S_i}{\partial t} = [\hat{S}_i, H].
\]  

(2.15)

However, the equation of motion for a collection of spins can be motivated by considering the angular momentum of a magnetic moment, \( \gamma L = m \), where \( \gamma \) is the gyromagnetic ratio:

\[
\gamma = \frac{g \mu_B}{\hbar} = \frac{g |e|}{2mc} = g \pi 2.79924916 \text{ MHz Oe}^{-1}.
\]  

(2.16)

The torque acting on \( m \) in the presence of a background field, \( H_{\text{eff}} \), gives the equation of motion

\[
N = m \times H_{\text{eff}} = \frac{\partial N}{\partial t} = \frac{\partial m}{\gamma \partial t}
\]  

(2.17)

which indicates that \( m \) will rotate about \( H_{\text{eff}} \). In real ferromagnets, \( m \) will eventually relax into the direction of \( H_{\text{eff}} \), which can be described by adding a phenomenological viscous damping term. Averaging over a collection of moments yields the equation of
2.3 Spin-Wave Dynamics

motion, known as the Landau-Lifshitz Gilbert (LLG) equation for the magnetization

\[ \frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \frac{\alpha_G}{|\mathbf{M}|} \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t}, \]  

(2.18)

where \( \alpha_G \) is a dimensionless constant that represents the damping.

One immediate consequence of equation 2.18 is that \( \mathbf{M} \cdot \frac{\partial \mathbf{M}}{\partial t} = 0 \), or that \( |\mathbf{M}| = M_s \) is a constant of motion. Taking the cross product \( \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t} \), and collecting terms yields

\[ \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}}) - \alpha_G M_s \frac{\partial \mathbf{M}}{\partial t} \]  

(2.19)

substituting this equation into equation 2.18 yields another form of the LLG equation:

\[ \frac{\partial \mathbf{M}}{\partial t} = -\gamma \frac{1}{1 + \alpha_G^2} \mathbf{M} \times \mathbf{H}_{\text{eff}} - \frac{\gamma \alpha_G}{M_s (1 + \alpha_G^2)} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}}). \]  

(2.20)

The vectors corresponding to each term are shown in Fig. 2.8; the first term (yellow arrow) acts as a torque that generates precession about \( \mathbf{H}_{\text{eff}} \), and the second term (purple arrow) causes relaxation into \( \mathbf{H}_{\text{eff}} \). The Gilbert damping parameter is typically much smaller than unity (\( \alpha_G \ll 1 \)) and is the subject of the discussion of section 2.4.

Linearizing the LLG equation for small-angle perturbations [211] about the spherical coordinates \( \theta \) and \( \phi \) gives the frequency, \( \omega \) of the collective-mode excitation (Kittel mode):

\[ \omega = \sqrt{\frac{\gamma}{M \sin \theta_M} \left( E_{\theta_M \theta_M} E_{\phi_M \phi_M} - E_{\theta_M \phi_M}^2 \right)}, \]  

(2.21)

where \( E_{\theta_M \theta_M}, E_{\theta_M \phi_M}, E_{\phi_M \phi_M} \), represent the second partials of the magnetic free energy with respect to the angles \( \theta_M \) and \( \phi_M \), evaluated at equilibrium.
Figure 2.8: Precession of a magnetic moment, $\mathbf{m}$, in the presence of an external field, $\mathbf{H}_{\text{ext}}$. The equation of motion (Eq. 2.20) contains two torque terms: the first (yellow) causes the moment to precess with frequency $\omega$; the second (pink arrow) causes the magnetization to relax to the equilibrium direction (red arrow), determined by the effective field, $\mathbf{H}_{\text{eff}}$. 
2.4 Damping

Magnetic damping is the transfer of energy from a collective spin excitation (i.e., a magnon) to some other mode (Fig. 2.9) and is represented by the Gilbert damping term, $\alpha_G$, in the Landau-Lifshitz-Gilbert equation of motion (equation 2.20). Intrinsic processes, such as spin-spin, spin-electron, or spin-lattice collisions, are unavoidable properties of the ferromagnetic material and place a fundamental upper limit on the relaxation time [77]. In contrast, extrinsic processes (such as two-magnon scattering, described in section 2.4.2) depend on sample geometry and preparation. The Gilbert parameter can therefore vary over a wide range; in Ni, the intrinsic parameter (described in section 2.4.1) is $\alpha_G = 0.045$ [78, 79], however, the novel mechanism described in this thesis results in $\alpha_G \approx 0.6$. This section will first outline the origin of the intrinsic damping by showing that relativistic corrections to the spin-wave hamiltonian couple the spins to the electric field. A description of the two-magnon scattering process, which is commonly invoked to explain anisotropic damping in thin films of transition metal ferromagnets, is then presented.

There may, however, be additional sources of damping. Dry magnetic friction [80, 81] is a result of magnetic inhomogeneities caused by the magnetic anisotropy having locally irregular directions; this is active for rare-earth magnetic ions, in which the anisotropy energy is a significant portion of the exchange energy, however, it is unlikely that this damping source is dominant in 3d transition metals [82].

Damping can also occur through the (non local) transfer of energy and angular momentum to adjacent layers, since the magnetization torque in the ferromagnetic layer generates a spin-polarized current at the interface between a ferromagnet and a normal metal. Transport theory [83, 84, 85, 86] is applied in this case, and a
large damping enhancement is observed for the case where the thickness of the non-magnetic metal layer is a couple of times larger than the spin coherence length. The material thickness dependence explains, for example, the damping enhancement found in Fe(16 nm)/Pd(25 nm) compared with Fe(16 nm)/Au(20 nm) [87].

### 2.4.1 Intrinsic Damping

Recently, Hickey and Moodera showed [88] that intrinsic Gilbert damping has its origin in spin-orbit coupling by examining spin-dependent corrections to a non-relativistic Hamiltonian for an electron with momentum \( \mathbf{p} \) subject to an electric field \( \mathbf{E} \) and magnetic field \( \mathbf{B} \):

\[
H^S = -\frac{\hbar}{2m_0c^2} \beta \mathbf{S} \cdot \mathbf{B} - \frac{\hbar}{4m_0^2c^2} \mathbf{S} \cdot \mathbf{E} \times \mathbf{p} - \frac{i\hbar^2}{8m_0^2c^2} \mathbf{S} \cdot (\nabla \times \mathbf{E}),
\]  

(2.22)
2.4 Damping

where $e$ and $m_0$ are the charge and rest mass of an electron, and the operators $S_i$ are given by

$$
\hat{S}_i = \begin{pmatrix}
\sigma_i & 0 \\
0 & \sigma_i
\end{pmatrix}
$$

for the Pauli spin matrices, $\sigma_i$, and

$$
\beta = \begin{pmatrix}
1 & 0 \\
0 & -1
\end{pmatrix}
$$

Using Maxwell's equation, $\nabla \times \mathbf{E} = -\frac{d\mathbf{B}}{dt}$, and the relationship $\mathbf{B} = \mu_0(\chi^M_1 + 1)\mathbf{M}$, the curl term in 2.22 can be rewritten in terms of $\frac{\partial \mathbf{M}}{\partial t}$:

$$
H^S = -\frac{e\hbar}{2m_0c^2} \beta S \cdot B - \frac{e\hbar}{4m_0^2c^2} S \cdot E \times \mathbf{p} - \frac{i\hbar^2\mu_0}{8m_0^2c^2} \mathbf{S} \cdot (1 + \chi^-_M) \cdot \frac{\partial \mathbf{M}}{\partial t}.
$$

In the above, the first two terms are identified as the time-independent Hamiltonian and define the ground state of the system; the last term is treated as a time-dependent perturbation in the Schrödinger picture. The magnetization observable is given by $\mathbf{M} = \sum_i \frac{q_i}{V} \text{Tr} \rho \hat{S}_i$, from which the time derivative and the equation of motion are obtained:

$$
\frac{d\mathbf{M}}{dt} = \sum_i \frac{q_i}{V} \sum_j \left( \frac{d\rho}{dt} \langle \psi_j | S_i | \psi_j \rangle + \rho \left( \frac{d}{dt} \langle \psi_j | \right) S_i | \psi_j \rangle + \rho \langle \psi_j | S_i \left( \frac{d}{dt} | \psi_j \rangle \right) \right),
$$

where in the Schrödinger picture, $\frac{d\mathbf{S}}{dt} = 0$. Using the quantum Liouville theorem,

$$
\frac{d\rho}{dt} = -\frac{1}{i\hbar} \left[ \rho, H \right],
$$

25
2.4 Damping

equation 2.26 simplifies to

\[
\frac{dM}{dt} = \sum_i \frac{g\mu_B}{V} \text{Tr} \{ \rho[S_i, H] \}.
\]  

(2.28)

This trace is computed with some algebra and the help of the commutation relations

\[ [S_i^a, S_j^b] = i\hbar \epsilon_{ijk} S_k^c \delta_{ab} \]  

[89, 88]; the final result has the same form as the LLG equation

\[
\frac{dM}{dt} = -\gamma M \times H - \frac{i\hbar \mu_0}{8m^2c^2} (1 + \chi_M^{-1}) \cdot M \times \frac{dM}{dt}
\]  

(2.29)

and the (intrinsic) Gilbert damping parameter is identified as

\[
\alpha_G = \frac{i\hbar \mu_0 M}{8m^2 \gamma c^2} \left( 1 + \chi_M^{-1} \right).
\]  

(2.30)

2.4.2 Extrinsic Damping

While the Gilbert damping parameter, \( \alpha_G \), in metals has its origin in the spin-orbit interaction of the itinerant electrons [90], studies have shown that the damping also depends on extrinsic factors, such as the sample growth method [82] and morphology [91, 83, 92]. One important source of damping is scattering from the uniform mode (for which \( k = 0 \)), into nonuniform modes \( (k \neq 0) \) at sample defects, a process known as Two-magnon scattering (TMS). This process was first proposed in 1961 by Kittel et al. [93] to explain an enhanced FMR linewidth in yttrium iron garnet spheres; however, more more recent work has focused on this mechanism in metallic thin films [87, 94, 95, 96], after it was shown [5, 4] that the spin-wave dispersion relation favors this mechanism for certain orientations of the magnetization; i.e., for certain directions of \( M \), there are \( k \neq 0 \) modes degenerate with the \( k = 0 \) mode, and
scattering occurs. A schematic of this process is shown in Fig. 2.10.

![Schematic of the two-magnon scattering process. In general, the dispersion relation (blue and green lines) will depend on the direction of $\mathbf{M}$. When $\mathbf{M}$ is in plane (green line), there is a degeneracy, and scattering from points $k = 0$ (A) to $k \neq 0$ (B) can occur (dashed red line). If, however, $\mathbf{M}$ is tilted significantly out of the sample plane (blue line), the degeneracy is broken and scattering does not occur.](image)

In the thin film limit, the boundary conditions for collective spin excitations (magnons) are such that only modes with a $\mathbf{k}$ parallel to the sample surface are allowed. In this case, the dispersion relation is [4]

$$
\omega^2(k) = \omega_0^2 - 2\pi \gamma M_d k d ([\sin^2 \theta_M \\
- \cos^2 \theta_M \cos^2 \phi_k] [H_{\text{ext}} \cos(\theta_H - \theta_M) - H_d \cos^2 \theta_M] \\
- \sin^2 \phi_k [H_{\text{ext}} \cos(\theta_H - \theta_M) + H_d \sin(2\theta_M)]) \\
+ \gamma^2 D k^2 (2H_{\text{ext}} \cos(\theta_H - \theta_M) + H_d (1 - 3 \cos^2 \theta_M)),
$$

27
2.4 Damping

where $H_d$ is the effective demagnetization field, $D$ is the exchange stiffness, and $\phi_k$ is the angle between $k$ and $M_s \sin \theta_M$ (the in-plane component of $M$). The zero mode, $\omega_0$ is given by equation 2.21. For scattering to occur, the term linear in $k$ in the above equation must be positive, since this implies that a region of the dispersion relation will pass through a minimum (green line, Fig. 2.10). There are two cases where this can happen [5]:

$$\sin^2 \phi_k < \frac{H_{\text{ext}}}{H_{\text{ext}} + H_d},$$

and

$$\theta_M < \pi/4.$$  

The second condition implies that the TMS mechanism shuts off as the magnetization is pulled normal to the sample.

This type of relaxation is a mode conversion; energy is pumped into other modes which subsequently dephase with the resonant ($k = 0$) mode. Defects at the sample surfaces contribute to local variations in the surface anisotropy field and act as scattering centers. Momentum transfer occurs and translational invariance is broken at defects. The exact expression for the decay rate thus depends on the symmetry properties of the defects [5]. This decay mechanism thus leads to an azimuthal angular dependence in the damping, which has been observed in FMR measurements on Pd/Fe(001)/GaAs(001) [96].

28
Chapter 3

Samples and Experimental Methods

This chapter will describe the samples and primary techniques employed in this thesis to investigate the damping of magnetization precession. First, sample selection, growth and characterization is discussed. Next, the principle of the time resolved pump-probe MOKE experiment is presented, followed by a discussion of the experimental setup employed in this work. Finally, a complementary method for investigating magnetization dynamics, Ferromagnetic Resonance (FMR), is introduced and experimental parameters detailed.

3.1 Sample Growth and Characterization

To study magnetization damping mechanisms, Nickel was selected as a common transition metal ferromagnet. Nickel and Nickel alloys are of great interest in magneto-electronic applications, as these materials can be designed to exhibit low coercivity
3.1 Sample Growth and Characterization

and layered structures can exhibit significant magnetoresistance [67, 66]. Studies of
the dynamical properties of these materials have been ongoing, and understanding
the damping has been particularly challenging. Early FMR measurements placed the
gilbert damping parameter, $\alpha_G$ at 0.02 [79], however, in 1975, Patton et al. [97] found
that the FMR line width in 10 nm thin film permalloy varies linearly with the reso-
nant frequency for frequencies above 10 GHz, corresponding to $\alpha_G = 0.005$, and the
discrepancy was attributed to extrinsic factors. Intrinsic damping was investigated
theoretically as a consequence of spin-orbit coupling [77, 88], however, extrinsic dam-
ping sources [98, 90], such as two-magnon scattering [5, 4] and spin-transport [61]
are still active areas of research. In particular, TR-MOKE measurements often yield
stronger damping than the FMR technique [59, 61, 62]. Furthermore, FCC Ni has
a fourfold magnetocrystalline anisotropy with a well-characterized temperature de-
pendence [75], which turns out to be important in the observed optically induced
damping mechanism, described below.

The samples were provided and characterized by professor Ale Lukaszew and her
group at the College of William and Mary. Nickel films 10 nm thick were grown on
MgO(001) substrates via magnetron sputtering using an Ar pressure of $6 \times 10^{-3}$ Torr
in an ultra high vacuum (UHV) system with a base pressure in the low $10^{-9}$ Torr.
In this deposition process, a cathode containing the target material (Ni) is held at a
low potential with respect to an anode containing the substrate (MgO). Argon ions
are accelerated by a potential difference, and bombard the target, ejecting material
which collects as a thin film on the substrate. To prevent oxidation at the surface, a
3 nm thick Au capping layer is deposited.

Ni(001) epitaxial growth is demonstrated by X-ray symmetric diffraction scans.
3.2 Magneto-Optical Kerr Effect Technique

A strong lattice mismatch between the MgO and Ni (16%) causes additional strain
anisotropy and defects at MgO/Ni interface [99].

3.2 Magneto-Optical Kerr Effect Technique

3.2.1 Static MOKE

In general, the polarization components of light scattered from a magnetized medium
will mix depending on the state of magnetization, resulting in a modified ellipticity.
For incident light that is linearly polarized, this process is known as either the Faraday
or Magneto-optical Kerr effect, depending on if it occurs in transmission or reflection.
The microscopic origin of this effect is spin orbit coupling; light generates a potential
gradient $\nabla V$ that couples local spins, $\hat{S}$, with momenta, $p$, adding a term $(\nabla V \times p) \cdot \hat{S}$
to the Hamiltonian [100]. In the macroscopic picture, the dielectric tensor, $\epsilon$, of
an isotropic magnetized medium contains off-diagonal elements that depend on the
magnetization [101, 100, 102]:

$$\epsilon = n^2 \begin{pmatrix}
1 & iQ_z & -iQ_y \\
-iQ_z & 1 & iQ_x \\
iQ_y & -iQ_x & 1
\end{pmatrix},$$

(3.1)

where $n$ is the average refractive index, and $Q(\alpha_1, \alpha_2, \alpha_3)$ is the Voigt vector contain-
ing the directional cosines $\alpha_1, \alpha_2, \alpha_3$ of $\mathbf{M}$. The Voigt vector is a phenomenological
parameter that relates magnetism and optics and is proportional to the magnetiza-
tion [67]. Normal modes derived from Maxwell’s equations with a dielectric tensor of
3.2 Magneto-Optical Kerr Effect Technique

![Diagram of MOKE Effect](image)

Figure 3.1: Two equivalent representations of the MOKE effect.

The form of 3.1 are right- and left-handed circularly polarized light with indicies of refraction [103]

\[
\begin{align*}
    n_L &= n \left( 1 - \frac{1}{2} Q \cdot \hat{k} \right) \\
    n_R &= n \left( 1 + \frac{1}{2} Q \cdot \hat{k} \right).
\end{align*}
\]  

(3.2) \hspace{1cm} (3.3)

The right- and left- handed components of propagating light "see" differing complex indicies of refraction and travel at separate velocities and attenuation rates. Since linearly polarized light is a combination of right- and left-handed polarized light with equal amplitudes, reflected light will exhibit a change in polarization (see figure 3.1).

When considering the Kerr effect, p- and s- polarized plane waves (Fig. 3.2), defined in terms of the plane of incidence (POI as shown in Fig. 3.2), provide a convenient decomposition scheme. Reflection is represented by

\[
\begin{pmatrix}
    E_{out}^s \\
    E_{out}^p
\end{pmatrix} =
\begin{pmatrix}
    r_{ss} & r_{sp} \\
    r_{ps} & r_{pp}
\end{pmatrix}
\begin{pmatrix}
    E_{in}^s \\
    E_{in}^p
\end{pmatrix},
\]

(3.4)

where $E_{in}^p$, and $E_{in}^s$ are the s- and p-components of the incoming light, and $E_{out}^p$ and
3.2 Magneto-Optical Kerr Effect Technique

Figure 3.2: MOKE geometry. Linearly polarized light (red line) reflects off of the sample, forming the plane of incidence (POI), and the s- and p- polarization axes. Similarly, the vector $\mathbf{M}$ (blue arrow) is decomposed into transverse and longitudinal components, which are orthogonal projections on the plane of the sample (POS), and a polar component, which is normal to the sample.

$\mathbf{E}_{\text{out}}$ are the corresponding outgoing components. The components of the scattering matrix, $\mathbf{r}$, depend on the direction of $\mathbf{M}$. In practice, $\mathbf{M}$ is decomposed in a basis that exploits experimental geometry, facilitating sensitivity (see figure 3.2). The longitudinal component is the intersection of the plane of the sample (POS) with the plane of incidence. The transverse component is also in the POS, but is perpendicular to the POI. Finally, the polar component is perpendicular to the POS. The Fresnel reflection coefficients of the $\mathbf{r}$ matrix are given in table 3.1 for various geometries.

For the special case when incident light is p- or s-polarized, the reflected polariz-
### 3.2 Magneto-Optical Kerr Effect Technique

Table 3.1: Fresnel coefficients. In the below expressions, $\alpha_1 = \cos \theta$, where $\theta$ is the angle between the ray and the film normal. $\alpha_2 = \sqrt{1 - \sin^2 \theta/n^2}$. $Q_p, Q_t, Q_l$ are the polar, transverse, and longitudinal projections of the Voigt vector, $Q$, which is proportional to the magnetization.

<table>
<thead>
<tr>
<th>Component</th>
<th>$r_{pp}$</th>
<th>$r_{ps} = -r_{sp}$</th>
<th>$r_{ss}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polar</td>
<td>$n\alpha_1 - \alpha_2$ (n\alpha_1 + \alpha_2)</td>
<td>$Q_p n\alpha_1$ (i(n\alpha_1 + \alpha_2)(\alpha_1 + n\alpha_2))</td>
<td>$\alpha_1 - n\alpha_2$ (\alpha_1 + n\alpha_2)</td>
</tr>
<tr>
<td>Longitudinal</td>
<td>$n\alpha_1 - \alpha_2$ (n\alpha_1 + \alpha_2)</td>
<td>$Q_l n\alpha_1 \tan \theta_2$ (i(n\alpha_1 + \alpha_2)(\alpha_1 + n\alpha_2))</td>
<td>$\alpha_1 - n\alpha_2$ (\alpha_1 + n\alpha_2)</td>
</tr>
<tr>
<td>Transverse</td>
<td>$n\alpha_1 \sqrt{1-Q_t^2/\alpha_2^2 - \alpha_2 - \alpha_2 \sin Q_l \alpha_1 \tan \theta_2}$ (n\alpha_1 \sqrt{1-Q_t^2/\alpha_2^2 + \alpha_2 - \alpha_2 \sin Q_l \alpha_1 \tan \theta_2})</td>
<td>0</td>
<td>$\alpha_1 - n\alpha_2$ (\alpha_1 + n\alpha_2)</td>
</tr>
</tbody>
</table>

When $p$-polarized light is reflected from a magnetized medium, it will gain a small $s$-component. If a polarizer is placed after reflection and set to a small angle, $\beta$ near-extinction, the intensity as measured by a photodetector will be [104]:

$$I = R(\beta^2 + 2\beta \kappa + \kappa^2 + \epsilon_k^2),$$

(3.7)

where $R = |r_{ss}|^2$ is the reflectivity. Taking the partial yields

$$\Delta I(t) = 2R \beta \Delta \kappa(t) + \beta^2 \Delta R(t),$$

(3.8)
3.2 Magneto-Optical Kerr Effect Technique

where $|\beta| \gg |\kappa + i\epsilon_k|$, which is generally true for transition-metal films [104]. A detector is thus sensitive to both the pump induced change in $\kappa$ and to the reflectivity, $\Delta R$, so care must be taken to identify the magnetic contribution to the pump-probe MOKE signal. The MOKE effect thus provides a powerful tool for observing the pump-induced change in the magnetization.

![Diagram of the static (longitudinal) MOKE experiment]

Figure 3.3: Schematic of the static (longitudinal) MOKE experiment. A HeNe laser is sent through a p-polarizer (P), the sample (S), and an s-polarizer (A). The sample is located between the poles of an electromagnet on a stage that allows azimuthal rotation.

Static longitudinal MOKE measurements are used to investigate the angular dependence of the sample’s anisotropy, which, in combination with the dynamic information obtained from TR-MOKE (section 3.2.2) and FMR (section 3.3), yield the anisotropy constants. A schematic of this experiment is shown in Fig. 3.3. To obtain this angular dependence, the sample is mounted inside an electromagnet (GMW 3470) on a computer-controlled rotary stage. A He-Ne laser ($\lambda = 633$ nm) is used for the optical source, and is modulated at $f = 300$ Hz with an optical chopper. The beam is first sent through a Glan-Taylor polarizer (Newport 10GL08) with an extinction
3.2 Magneto-Optical Kerr Effect Technique

ratio of $10^5 : 1$ to isolate the p-polarization component. The beam is then reflected off the sample at angle of roughly $60^\circ$ from the normal and directed through a second, identical polarizer, which is set to near-extinction (s-polarization) to isolate the longitudinal MOKE signal. Finally, the light is detected by a photodiode (Thorlabs DET110). The detector output and chopping frequency are sent to a lock-in amplifier (SRS SR530), which is used to increase the signal to noise ratio.

### 3.2.2 Time-Resolved MOKE

![Figure 3.4](image)

Figure 3.4: Schematic of two color pump-probe TR-MOKE experiment. A pulsed Ti:Sapphire laser is split into pump and probe arms and recombined at the sample (S). A BBO crystal doubles the frequency of the pump beam ($\lambda = 400$ nm). The probe beam ($\lambda = 800$ nm) is time-delayed by a computer-controlled delay stage and sent through a p-polarizer (P), the sample (S), and an s-polarizer (A). The sample is located between the poles of an electromagnet on a stage that allows azimuthal rotation.

Figure 3.4 contains a schematic of the pump-probe MOKE experiment employed in this thesis. The optical source is an amplified pulsed laser system. The seed for the amplifier is a pulsed Ti:Sapphire laser (Spectra-Physics Tsunami) with $\lambda =$
3.2 Magneto-Optical Kerr Effect Technique

Figure 3.5: A typical pump-probe TR-MOKE data set. Several scans are averaged and fit to a damped exponential, as discussed in the text, to obtain the center precession frequency, \( \omega_0 \), and the decay time, \( \tau_{\text{MO}} \).

800 nm, a pulse width \( (w_{\text{seed}}) \) of \( w_{\text{seed}} \approx 150 \) fs, and a repetition rate, \( r_{\text{seed}} = 50 \text{ MHz} \). The regenerative amplifier (a Ti:Sapphire Spectra-Physics Spitfire) is pumped with a Nd:YLF (Spectra-Physics Evolution) \( (\lambda_{E_v} = 527 \text{ nm}, w_{E_v} = 150 \text{ ns}, r_{E_v} = 1 \text{ kHz}) \) for an energy gain of \( \approx 10^6 \). The final pulse train employed in the experiments has \( \lambda_{\text{exp}} = 800 \text{ nm}, w_{\text{exp}} = 150 \text{ fs}, r_{\text{exp}} = 1 \text{ kHz} \).

The beam is split into pump and probe arms (figure 3.4). To limit cross-noise from the pump beam in the probe detection circuit, a two-color scheme is used: the pump beam is frequency doubled to \( \lambda_{\text{pump}} = 400 \text{ nm} \) using a \( \beta - \text{BaB}_2\text{O}_4 \) (BBO) doubling crystal and sent to the sample at normal incidence. The spot diameter as measured at the sample using standard techniques \([105]\) is \( 350 \pm 20 \mu\text{m} \). The pump fluence, unless otherwise noted, was 2 mJ/cm\(^2\).

The time-resolved MOKE setup is employed to measure the magnetization dynamics. A computer-controlled movable delay sets the delay time of the probe beam. To isolate the polar MOKE component, two Glan-Taylor polarizers (Newport 10GL08,
extinction ratio = $1 : 10^5$) are placed immediately before (P) and after (A) the sample. The P-polarizer sets the incident light to p-polarization; the A-polarizer is set at $1 - 2\degree$ from extinction. The spot size of the probe beam, measured at the sample, is $215 \pm 20 \mu m$ with a fluence of $0.25 \pm 0.05 \text{mJcm}^2$.

A mechanical chopper is used to modulate the pump beam at $f \approx 300 \text{Hz}$. A color filter placed after (A) isolates the $\lambda = 800 \text{nm}$ component of the probe beam, limiting noise caused by scattering from the pump. A photodiode (Thorlabs DET110) is employed to measure the intensity of the probe beam, and fed to a lockin-amplifier (SRS SR530).

Two geometries are employed to measure the dynamics as a function of magnetic field. For the case where the field is applied in the plane of the sample (as shown in 3.4), the sample is placed between the poles of a GMW 3470 electromagnet on a computer-controlled motor stage which allows azimuthal angle control. For the case where a field is applied normal to the sample, a split-coil superconducting magnet (Oxford TL) is employed, and the optical beams are sent through one of the coils. Unless otherwise noted, all measurements are performed at room temperature.

A typical set of raw data is shown in Fig 3.5. To obtain the oscillatory component of the MOKE signal, the data is fit to $A \exp(-t/\tau_{mo}) \cos(\omega_0 t + \delta) + B \exp(-t/\tau_2)$. The second term is employed to capture a slow thermal background (from the MgO) component. The frequency $\omega_0$ and oscillatory damping $\tau_{MO}$ correspond to the frequency and decay time of the Kittel oscillation mode, defined in equation 2.21. External control parameters included the field amplitude $H_{ext}$ and direction $\theta_H, \phi_H$ (defined relative to the magneocrystalline anisotropy, as in figure 2.8, temperature, $T$, and pump power, $P$. 

38
3.3 Ferromagnetic Resonance

Figure 3.6: Schematic of a FMR experiment. A waveguide couples microwaves into a resonance cavity containing the sample. An external field, $H_{ext}$ is swept, and a detector measures the absorbed power differential, $\frac{dP}{dH_{ext}}$.

Similar to electron spin (ESR) and nuclear magnetic resonance (NMR), Ferromagnetic Resonance (FMR) spectroscopy measures the absorption of electromagnetic radiation by unpaired spins to determine the magnetic response of a system. This technique is now nearly a century old [106], and there is abundant literature discussing the technique and its application to a variety of material systems (see, e.g., the reviews in [107, 108, 109, 20, 1]). This section will briefly outline the important aspects of this technique.

The stimulation of magnetic precession by the absorption of electromagnetic radiation in the presence of an external field can be described by the macroscopic LLG formalism discussed in chapter 2, taken in combination with Maxwell’s equations for a magnetic medium [110]. Resonance occurs in the microwave regime for a ferromagnet when the applied radiation (r.f.) frequency coincides with the precession frequency: $\omega_0 = \gamma H_{eff}$, where $H_{eff}$ is the local static magnetic field. The power of the radiation absorbed is given by [110] $P = \frac{1}{2} \omega \text{Im}(\chi) H_r^2$, where $\chi$ represents the complex magnetic
3.3 Ferromagnetic Resonance

rf susceptibility, and $H_{rf}$ is the magnitude of the radiation field. Hence, monitoring the absorbed power yields the dynamic response, $\text{Im}(\chi)$ of the magnetic system.

In practice it is easier to measure $\frac{P}{dH_0} \propto \frac{d\text{Im}x}{dH_0}$ [1] for a variety of field amplitudes $H_0$ and directions, $\theta_H, \phi_H$ (defined similarly to that in Fig. 2.8). The sample is placed in or at the end of a microwave cavity with an eigenfrequency $\omega_0$, and the DC field is swept through resonance. Line spectra yield important information; the external field at which the maximum energy is observed is the resonant field, i.e., $\frac{dx}{dH_0}|_{H_r} = 0$. The sets $[H_r]$ vs. $[\omega_0], [\phi_H]$ and $[\theta_H]$ are substituted into the Kittel formula (equation 2.21) to determine the magnetic anisotropy constants and the gyroscopic ratio, $g$. While the resonance condition yields information about the precession frequency, the line shape and peak-to-peak linewidth, $\Delta H_{pp}$, yield information about the homogeneity and damping.

The linewidth is considered in terms of homogeneous and inhomogeneous contributions

$$\Delta H = \sqrt{3}\Delta H_{pp} = \Delta H_{\text{hom}} + \Delta H_{\text{inhom}}$$ (3.9)

Gilbert-like damping of the form in the LLG equation of motion (eq. 2.20) is of the form $\Delta H_{\text{hom}}$ and can be written as [1]

$$\Delta H_{\text{hom}}(\theta_M, \phi_M) = \frac{2}{\sqrt{3}} \left| \frac{\partial \omega_0}{\partial H_{\text{res}}} \right|^{-1} \frac{\alpha \gamma}{M_s} \left( \frac{1}{\sin^2 \theta_M} \frac{\partial^2 F}{\partial \theta_M^2} + \frac{1}{\partial \phi_M^2} \right)$$ (3.10)

Where, as in equation 2.21, $F$ is the magnetic free energy, $\theta_M, \phi_M$ are the polar and azimuthal angle of the magnetization, $M$, and $\alpha$ is the gilbert damping parameter. In the above, the known anisotropy of the intrinsic gilbert parameter [111, 88] is not included, however, for the experiments described in this thesis other, non-intrinsic
3.3 Ferromagnetic Resonance

contributions to the Gilbert parameter dominate: \( \alpha = \alpha_{\text{eff}} > \alpha_{\text{intrinsic}} \). For Ni, \( \Delta H_{\text{hom}} \approx 90 \) Oe for measurements at \( \omega_0 = 56 \) GHz [1].

Sample inhomogeneities broaden the FMR linewidth, \( \Delta H_{\text{inhom}} \), and cause deviations from a pure-Lorentzian or Gaussian lineshape. Obtaining a direct expression for the lineshape is greatly complicated in this case, and contributions to damping from sources such as Two-magnon scattering (section 2.4.2) must be included. Additionally, for directions far from the principle anisotropy axes, there can be a large difference between the directions of \( \mathbf{M} \) and \( \mathbf{H} \); this “dragging” of the magnetization with respect to the field causes a corresponding asymmetry in the line spectrum.

The FMR and TR-MOKE techniques are complementary; FMR is a frequency-domain technique, while TR-MOKE measures the dynamics in the time domain. To link the linewidth to the average decay time, the following expression is used:

\[
\frac{2}{\tau} = \Delta \omega = \left| \frac{\partial \omega}{\partial H} \right| \Delta H
\]

the expression \( \frac{\partial \omega}{\partial H} \) is calculated implicitly from the Kittel formula (equation 2.21) and depends on the anisotropy parameters and \( \theta_H, \phi_H, \theta_M, \phi_M, \omega, \) and \( H_r \).

FMR experiments for both easy- and hard-axes were conducted at Norfolk State University with the assistance of professor Natalia Noginova and Mr. Osei Amponsah. A sample scan is shown in figure 3.7.
3.3 Ferromagnetic Resonance

Figure 3.7: A typical FMR scan. The external field is swept through resonance, as discussed in the text. The resonance field is the zero-point crossing, and $\Delta H = \sqrt{3} \Delta H_{pp}$ is the peak-to-peak linewidth. The blue line represents the raw data; the red line is a smoothed average. The large spikes about $H_0 = 3500$ are due to electron spin resonance of the sample substrate.
Chapter 4

Results and Discussion

This chapter contains a discussion of the results of the TR-MOKE and FMR experiments on Ni/MgO(001). Details of these experimental methods can be found in chapter 3. The key result of these experiments is that while the frequency of oscillation spectra are consistent in both TR-MOKE and FMR, and are well-determined by the magnetic anisotropy, the pump pulse in the optical technique causes additional damping, depending on the orientation of $\mathbf{M}$ relative to the magnetic anisotropy. This novel relaxation channel, pump-induced anisotropic damping, is discussed in detail in section 4.4.

4.1 Static MOKE

To understand the anisotropy symmetry present in the system, we first examined the static longitudinal MOKE. Hysteresis loops were measured as a function of in-plane field direction, $\phi_H$, from which the coercive field, $H_c$, was obtained. This field is defined as the intensity of the applied magnetic field required to reduce the
magnetization to zero. The azimuthal dependence of $H_c$ indicates a four-fold in-plane anisotropy (Fig. 4.1), which we incorporate in fits to the FMR and TR-MOKE data. As in ref. [99], we notice the presence of four “spikes” located between the easy ($\phi_M = 0^\circ, 90^\circ, 180^\circ, 270^\circ$) and hard ($\phi_M = 45^\circ, 135^\circ, 225^\circ, 315^\circ$) axes of magnetization. The TR-MOKE frequency data, however, does not reflect this additional symmetry, and since it is strongly localized, we omit an additional four-fold anisotropy term in the free energy, considering only the dominant four-fold term.

Figure 4.1: Coercivity vs. applied field angle, $\phi_H$ as measured by SMOKE. The estimated error bars are roughly the size of the data point.

Studies have shown [112, 99] that on thicker Ni films (30 nm) on MgO, annealing
induces an exchange bias (defined in section 2.2.3) at the interface by the formation of NiO at the Ni/MgO boundary. Previously, Seu et al [113] used a pump pulse to induce a pinning state in exchange-biased FeMn/Co. In our sample, however, we did not detect exchange bias, as the hysteresis loops were symmetric about $H_{\text{ext}} = 0$ both before and after the TR-MOKE experiments, suggesting that the beam fluence ($< 2 \text{ mJ/cm}^2$) was not enough to anneal the sample.

4.2 TR-MOKE

4.2.1 In-plane TR-MOKE

The TR-MOKE apparatus generally employs a large set of experimental parameters with which to access the spin dynamics and the damping: the external field amplitude and direction ($H_{\text{ext}}, \phi_H, \theta_H$), the sample temperature ($T$), and the laser power ($P$). In the following set of experiments, the sample temperature and laser power were fixed, and the dynamics were observed as a function of field direction. The principle parameters of the TR-MOKE spectra are the precession frequency, $\omega_{\text{MO}}$, and the decay time, $\tau_{\text{MO}}$, which are extracted by fitting the magnetic component with a damped sine wave, as described in chapter 3. The sample geometry is shown in Fig. 2.8; angles are defined in spherical coordinates relative to the surface normal ($\theta_H = 0^\circ$) and the in-plane easy axis ($\phi_H = 0^\circ$). Two general TR-MOKE experiments were conducted: in the first, the external field was applied near-parallel ($\theta_H \approx 90^\circ$) to the sample plane; in the second, the field was applied normal ($\theta_H \approx 0^\circ$).

For the in-plane experiments, the external field was applied near the plane of the sample ($\theta_H \approx 97^\circ$) at a fixed angle from the easy axis, $\phi_H$. The slight tilt of $H_{\text{ext}}$
4.2 TR-MOKE

out of the sample plane is necessary to excite oscillations when the magnetization is aligned along axes of high crystalline symmetry. The $\mathbf{M} \times \mathbf{H}_{\text{eff}}$ term in the LLG equation of motion (Eq. 2.18) vanishes for the case where the magnetization vector, external field, and effective field are co-linear (i.e. when the sample is magnetized along the easy or hard axes). For these special cases, the pump-induced anisotropy change does not provide the initial torque necessary to excite oscillations, since $\mathbf{H}_{\text{eff}}$ does not change direction when the anisotropy is "turned off" by heating from the pump pulse, and consequently, $\mathbf{M} \times \mathbf{H}_{\text{eff}} = 0$ for all $t$. Tilting the external field at a slight angle out of plane provides the needed initial perturbation, since, for this case, the components of $\mathbf{H}_{\text{eff}}$ are not co-linear; nevertheless, the relatively strong shape anisotropy ensures that $\theta^* \approx 90^\circ$ for the external field values employed in this thesis, and the magnetization vector thus relaxes in the plane of the sample. The requirement that $\mathbf{M} \times \mathbf{H}_{\text{eff}} \neq 0$ at $t = t_0$ ultimately broadens the excited mode distribution, leading to enhanced damping in TR-MOKE experiments, as discussed later in this chapter.

Oscillations were observed as a function of field amplitude for $\phi_H = 0^\circ, 10^\circ, 20^\circ, 30^\circ, 40^\circ$ and $45^\circ$. The data for the easy ($\phi_H = 0^\circ$) and hard ($\phi_H = 45^\circ$) axes are presented in Fig. 4.2 and Fig. 4.3, respectively. Typically several scans were taken for each field value; the averaged data were smoothed with a floating average filter with a step size of less than a quarter-period (effectively a low-pass filter) and fit to damped sine wave on a exponentially decaying background (represented as red lines): $A e^{-t/\tau_{\text{MO}}} \sin(\omega_{\text{MO}} t + \delta) + B e^{-t/\tau_2}$. The fit parameters were not fixed, and the extracted error bars represent 90% confidence intervals of the parameters obtained from weighted, non-linear least-squares fits to the data. The frequency, $\omega_{\text{MO}}$, and damping, $\tau_{\text{MO}}$, are the principal fit parameters and are discussed below. Both were found
to be robust relative to the estimates of the other fit parameters. The background term is a combination of magnetic (MOKE) and thermal (reflectivity) signals, and is highly sensitive to the setting of the analyzer in TR-MOKE experiments employing a crossed-polarizer geometry [104]; in the TR-MOKE experiments of this thesis, the ratio of the amplitude of the background term to that of the oscillatory component \((A \sin \delta)\) is typically approximately 0.5.

As expected from the theory of magnetization dynamics (Chapter 2), the precession frequency (Fig. 4.4) varies weakly with the orientation of the magnetization
vector, and strongly with the external field amplitude. The error in the estimation of $\omega_{MO}$ is typically within 1% for fields above saturation ($\approx 10$ mT for the easy axis and $\approx 20$ mT for the hard axis). The solid lines in Fig. 4.4 were generated from the Kittel formula (Eq. 2.21) using the anisotropy parameters obtained from fits to FMR data (section 4.3). At very low fields, and for orientations near the hard axis, $\omega_{MO}$ differs from the model. This discrepancy is caused by the excitation mechanism; the pump pulse temporarily heats the sample, destroying $M$ within the first few ps. At low fields ($H_{ext} < 20$ mT), the recovering ferromagnet is no longer saturated, and
M and $\omega_{MO}$ are not uniformly well-defined across the beam diameter. Thus, the LLG formalism (and therefore the Kittel model) may not apply for fields below the saturation magnetization when the field is significantly away from the easy axis. For high fields, there is close agreement between the model and $\omega_{MO}$.

Figure 4.4: Precession frequency, $\omega_{MO}$, extracted from the data for $\phi_H = 0^\circ, 10^\circ, 20^\circ, 30^\circ, 40^\circ, 45^\circ$, and $\theta_H = 90^\circ$. The lines are predictions from the Kittel formula (Eq. 2.21) using the anisotropy parameters obtained from FMR.

The trends in the damping are complementary to those of the frequency; $\tau_{MO}$ varies weakly with field and strongly with angle. The mechanism for this behavior, pump-induced anisotropic damping, is developed in section 4.4.
Figure 4.5: Damping, $\tau_{MO}$, extracted from the data for $\phi_H = 0^\circ, 10^\circ, 20^\circ, 30^\circ, 40^\circ, 45^\circ$ and $\theta_H \approx 90^\circ$. 
4.2.2 Out-of-plane TR-MOKE

In the out-of-plane measurements, the external field was applied near-normal ($\theta_H \approx 0$) to the sample surface and oscillations were observed as a function of field amplitude. The raw data series is shown in Fig. 4.6, with fits to the data (red lines). We do not observe oscillations for field values above 450 mT, when the external field is strong enough to completely pull the magnetization normal to the surface; in this case, $H_{\text{ext}}$, $H_{\text{eff}}$ and $M$ become co-linear, and the torque term in the LLG equation vanishes. All TR-MOKE measurements for the normal geometry are thus observed for field values below the out-of-plane saturation (but well above the in-plane saturation, roughly 20 mT for the easy axis). The red lines in Fig. 4.6 represent fits to the raw data using the same method discussed above for the in-plane data. However, for field values above 450 mT, where no oscillations were present, the amplitude of the oscillatory term, $A$, was set to zero.

The TR-MOKE scans show only one oscillation with a frequency of approximately $\omega_{\text{MO}} = 14 \pm 3$ GHz (Fig. 4.7), and $\tau_{\text{MO}} = 0.1 \pm 0.05$ ns (Fig. 4.8); both estimates are independent of field amplitude within the error. The anisotropy parameters obtained from the FMR data were used to model the frequency, as described above for the in-plane data. The frequency data agree well with the Kittel model for $H_{\text{ext}} < 300$ mT if the external field angle is corrected to $\theta_H = 5^\circ$ in the model. This correction is reasonable given the uncertainty in $\theta_H$, which was approximately $8^\circ$. If the raw experimental value of $\theta_H = 0^\circ$ is used, the Kittel model underestimates the data by approximately 2 GHz. The data does not agree with the model for $H_{\text{ext}} = 375$ mT. This discrepancy is likely due to a poor estimate of the frequency, as for this and higher fields, the background signal begins to dominate, and the fits to the oscillatory
Figure 4.6: TR-MOKE data vs. $H_{\text{ext}}$ for $\phi_H \approx 0^\circ$, $\theta_H \approx 0^\circ$ (Normal). Red lines represent fits to the data, as described in the text.

component of the TR-MOKE data are poorly determined.
Figure 4.7: Precession frequency, $\omega_{MO}$, extracted from the data is independent within the error of field amplitude for the normal geometry, $\theta_H \approx 0^\circ$. The solid line represents the prediction from the Kittel formula (Eq. 2.21) using the anisotropy parameters obtained from FMR.

Figure 4.8: The decay time, $\tau_{MO}$, is independent of field amplitude within the error for the normal geometry $\theta_H \approx 0^\circ$. 
4.3 Ferromagnetic Resonance

FMR measurements were taken at $2\pi f = 60.6$ GHz for both the easy ($\phi_M \approx 0^\circ \pm 5^\circ$) and hard ($\phi_M \approx 0^\circ \pm 5^\circ$) axes as a function of the field angle normal to the sample surface, $\theta_H$. The raw data were fit to a Dysonian line shape, from which the resonant field $H_0$ and the line width $\Delta H$ were obtained. The Dysonian line shape was used to capture the asymmetry that is observed for cases where the external field is oriented significantly out of the plane of the sample. This asymmetry is caused by the “dragging” of the magnetization vector relative to the external field vector [1].

The resonance field data (Fig. 4.9) yields information about the anisotropy parameters when fit to the Kittel formula (Eq. 2.21). This equation involves the curvature of the magnetic free energy evaluated at the equilibrium angles of magnetization ($\theta^*, \phi^*$), which are defined as the angles which minimize the energy. The magnetic free energy in this system is:

$$E = -\mathbf{H}_{\text{ext}} \cdot \mathbf{M} + \frac{1}{2} \mu_0 M_s^2 \cos^2 \theta_M - \frac{1}{2} K_2 \cos^2 \theta_M - \frac{1}{2} K_{4\perp} \cos^4 \theta_M - \frac{1}{8} K_{4\parallel} (3 + \cos 4\phi_M) \sin^4 \theta_M$$

(4.1)

where $M_s$ is the saturation magnetization, and $K_2$, $K_{4\perp}$, and $K_{4\parallel}$ are energy constants.

Since only $\theta_H, \phi_H$ are known experimentally, an iterative approach must be used to determine the anisotropy constants. For the first iteration, the equilibrium angles are set to $\theta^* = \theta_H$ and $\phi^* = \phi_H$ (effectively ignoring all but the first term in the above equation), and the data fit to Eq. 2.21 to obtain a least-squares estimate of the anisotropy parameters. At the beginning of each subsequent iteration, new estimates of $\phi^*$ and $\theta^*$ are calculated from the previous anisotropy estimate, and the process is continued until the angle estimates converge. This method yields $K_2 = -108 \pm$
10 kJ/m$^3$, $K_{41} = -5.1 \pm 1$ kJ/m$^3$, and $K_{4||} = -2.4 \pm 0.5$ kJ/m$^3$. The solid lines in Figs. 4.9 and 4.4 are generated from Eq. 2.21 and the above anisotropy parameters. These parameters were also used to predict the frequency spectra of the TR-MOKE measurements, as discussed above.

The data trend in Fig. 4.9 is well-described by the Kittel formula and can be understood by considering the relative contribution of the magnetic anisotropy and the external field to the effective field, which determines the resonance frequency. When the sample is magnetized along the in-plane easy axis, there is a relatively large contribution to the effective field magnitude from the magnetic anisotropy, and a relatively low external field is needed to excite resonance. Conversely, when the sample is magnetized away from the easy axis, a higher external field is required to excite resonance. The resonance field, $H_{\text{res}}$, is thus at a maximum when the sample is magnetized along the normal ($\theta_H = 0$), and at a minimum when the sample is magnetized in plane, along the easy axis.

The linewidth (Fig. 4.10) varies strongly with $\theta_H$ for both the easy and hard axes. When the field is applied normal to the sample plane ($\theta_H = 0$), $\Delta H = 288 \pm 5$ G. The linewidth increases rapidly to $\Delta H = 491 \pm 5$ G, for the easy axis, and $\Delta H = 542 \pm 5$ G for hard axis when the external field is approximately $27^\circ$ from the normal, and decreases to $\Delta H = 343 \pm 5$ G and $\Delta H = 353 \pm 5$ G when the field is applied in-plane for the easy and hard axes, respectively. The large deviation from the intrinsic value $\approx 80$ G indicates that extrinsic damping is dominant in this sample, and the presence of defects at the Ni/MgO interface contributes to mode-conversion by the Two-magnon scattering process [5, 4].

The decay time, $\tau_{\text{FMR}}$, (Fig. 4.11) is calculated from Eq. 3.11, which involves both
4.3 Ferromagnetic Resonance

Figure 4.9: $H_0$ vs. $\theta_H$ for Easy (blue squares) and Hard (red circles) Axes. Error bars (not shown) are approximately the size of the data points. The solid lines represent a fit to the Kittel Mode (Eq. 2.21) from which the anisotropy parameters, $K_2, K_{4\perp}, K_{4\parallel}$ are obtained.

the anisotropy parameters and the equilibrium angles. The large variation between the easy (blue) and hard (red) axis estimates in the region $5^\circ < \theta_H < 35^\circ$ is likely caused by poor estimates of $\theta^*$, which is itself limited by the uncertainty in the fitted anisotropy parameters. The equilibrium direction of the magnetization is changing very rapidly over this range, as shown in Fig. 4.12; however, at the field normal $\theta_H = 0^\circ$ or for $\theta_H > 35^\circ$, the magnetization direction changes less rapidly, and the estimates of $\tau_{FMR}$ are more reliable.
4.3 Ferromagnetic Resonance

Figure 4.10: $\Delta H$ vs. $\theta_H$ for Easy (blue squares) and Hard (red circles) Axes. Error bars (not shown) are approximately the size of the data points.

Figure 4.11: The extracted decay time, $\tau_{\text{FMR}}$ vs. $\theta_H$ for easy (blue squares) and hard (red circles) Axes. The rapid decrease at $\theta_H \approx 35^\circ$ is due to the onset of Two-Magnon scattering [4, 5].
Figure 4.12: The equilibrium direction of magnetization, $\theta^*$, vs. applied field direction, $\theta_H$, as calculated for the Easy (blue line) and Hard (red line) Axes. The direction of the magnetization vector changes rapidly for $\theta_H < 35^\circ$, and is defined as the direction that minimizes the magnetic free energy (Eq. 4.1).
4.4 Pump Induced Anisotropic Damping

In this section, we demonstrate that an ultrafast pump pulse can control the effective damping in a ferromagnet by thermally altering magnetic anisotropy and that TR-MOKE and FMR are inconsistent for geometries in which the magnetization is pulled away from the easy axis. We therefore introduce a novel optically mediated decay mechanism: pump-induced anisotropic damping (PIAD).

In pump-probe TR-MOKE, the pump pulse temporarily heats the spin population of the irradiated region above or near the Curie temperature within the first few picoseconds, destroying or heavily modifying both the magnetic anisotropy and the magnetization [54]. As the pumped region cools in an applied field, the magnetization recovers rapidly with temperature in the form $|M| \sim \sqrt{1 - \frac{T^2}{T_c^2}}$, as described by the Curie-Weiss law. However, the magnetic anisotropy constants have a gradual, power-law temperature onset ($K \sim |\frac{M(T)}{M(0)}|^{10}$)[75], and recover less rapidly. The temperature dependence of the magnetization and anisotropy constant are compared in Fig. 4.13 for the temperature range of the TR-MOKE experiments of this thesis ($T > 300$ K). Consequently, spins in the pumped region recover within the first 100 ps in the direction of the external field. In contrast, spins in the unpumped region remain relatively unperturbed along the initial effective field direction (Fig. 4.14). As the system evolves, the magnetization, effectively an average over the moments in the probed region, precesses toward equilibrium; however, the non-uniform temperature profile (and hence effective field gradient) and exchange between perturbed spins broadens the excited spin wave mode distribution, $\Delta \omega$, about the center frequency, $\omega_{\text{MO}}$. This process generates inhomogeneous dephasing on the timescale $\Delta \omega^{-1}$, and a reduction of the measured decay time, $\tau_{\text{MO}}$. 

59
4.4 Pump Induced Anisotropic Damping

Figure 4.13: Temperature dependence of the magnetization (red line) and anisotropy constant (blue line) for $T = 300$ K to $T = 627$ K, reproduced from [75]. Both $M$ and $K$ are normalized to 1 at $T = 300$ K for easy comparison. In TR-MOKE, the pump pulse quickly heats the sample; $M$ thus recovers first, followed by $H_{\text{eff}}$, which contains contributions from the anisotropy.

We control the relative strength of this enhanced damping by tuning the direction of the external field relative to the magnetocrystalline anisotropy, as shown schematically in Fig. 4.14. As described in section 4.3, our system has a four-fold in-plane anisotropy symmetry with an in-plane easy axis along [110], and an in-plane hard axis along [100]. By applying an external field along the preferred direction of anisotropy, i.e., the in-plane easy axis, we minimize the mismatch between pumped and non-pumped spins, since, for this geometry, the pump beam modulates primarily the magnitude of the effective field during the excitation process. Thus, there is not a large angle between the pumped and non-pumped spins, and the excited mode distribution is mostly homogeneous. While there must always be a slight deviation between
4.4 Pump Induced Anisotropic Damping

Figure 4.14: Pump-induced anisotropic damping for easy (a), hard (b), and normal (c) geometries. The black arrow indicates the direction of the applied field. The pump beam (not shown) heats the irradiated region and generates inhomogeneities in the excited spin distribution (represented as arrows in the left panel) depending on the direction of the magnetization relative to the magnetic anisotropy (center). Subsequent dephasing manifests as effective damping observed in TR-MOKE (raw data for $B = 150$ mT shown in right panel) that increases for directions away from the easy axis. Solid lines represent fits to the data.

the external field direction and the easy axis to provide the initial toque necessary to excite oscillations (see Section 4.2.1), the PIAD effect does not dominate, and the TR-MOKE result corresponds within the error to the FMR result at the same frequency ($\omega_{MO} = 60.5$ GHz) (Fig. 4.15): $\tau_{MO} = 0.28 \pm 0.04$ ns; $\tau_{FMR} = 0.245 \pm 0.007$ ns. Despite the overlap of the error estimates, it is not surprising that FMR yields a shorter relaxation time than TR-MOKE, since FMR is a global technique, and some additional broadening is likely caused by inhomogeneities and edge defects. TR-MOKE, in comparison, has a spot diameter of 0.2 mm, and is thus less sensitive to global sample irregularities.
4.4 Pump Induced Anisotropic Damping

Figure 4.15: FMR vs. TR-MOKE measured decay time for $\omega = 60.6$ GHz. FMR data (easy axis: o; hard axis: *) yields the characteristic Two-Magnon Scattering curve [4, 5] (solid line is guide to the eye) as the external field is swept normal to the film plane. TR-MOKE data yields similar results to FMR for the in-plane easy axis ($\Delta$), yet is inconsistent with FMR data for the hard ($\triangledown$) and normal (☐) axes. Error bars represent estimated standard error of the mean.

We increase the PIAD effect by rotating the direction of the applied field away from the easy axis, since this widens the angle between the magnetic anisotropy field and the external field (Fig. 4.14 b). The pump beam now alters both the magnitude and direction of the effective field, and the recovering spins in the heated region recover first in the direction of the external field. The non-uniform temperature profile results in inhomogeneous broadening of the excited spin distribution. The oscillation measured by the probe beam is thus more heavily damped. This effect increases until the near hard axis. For this case, the PIAD effect results in a shorter decay time in TR-MOKE than in FMR: $\tau_{MO} = 0.18 \pm 0.03$ ns versus $\tau_{FMR} = 0.246 \pm 0.007$ ns.
4.4 Pump Induced Anisotropic Damping

(Fig. 4.15).

We observe different dynamics between FMR and TR-MOKE for the case in which the field is applied normal to the film surface (Fig. 4.15) because the anisotropy becomes increasingly important as the magnetization is pulled out of the sample plane. Here TR-MOKE and FMR employ slightly different geometries, as discussed in sections 4.2.1 and 4.3; however, since the precession frequency is consistent in both TR-MOKE and FMR, the techniques are comparable. In the case of a field applied out-of-plane, direct comparison for FMR and TR-MOKE is possible for fields above which we saturate the magnetization out of plane (B > 0.45 T). In TR-MOKE, we are unable to observe oscillations at these field values since the magnetization becomes "pinned". Since both $\tau_{\text{MO}}$ and $\omega_{\text{MO}}$ remain independent with field amplitude, we extrapolate the TR-MOKE values in comparison with FMR in this geometry. For the normal geometry, our TR-MOKE results are very different from FMR, which we attribute to the role of shape anisotropy ($\sim \cos^2 \theta_M$) in the excitation mechanism; the slow recovery of the magnetocrystalline anisotropy is again the cause of the broadening of the excited spins, however, since the external field now points normal to the surface, the recovering spins are more sensitive to the angle out of plane. For this case, the oscillations of TR-MOKE are heavily damped (Fig. 4.14 b), with $\tau_{\text{MO}}$ independent of field amplitude within the error, between 0.07 ± 0.04 ns and 0.125 ± 0.04 ns (Fig. 4.15). For FMR, with the external field applied normal to the sample, we observe a decay time of $\tau_{\text{FMR}} = 0.55 \pm 0.03$ ns. In the case of TR-MOKE, local inhomogeneities in pumped vs. non-pumped regions manifest as deviations in the tilting angle of the magnetization. Thus the pump beam excites a very broad spectrum of spin-wave modes, resulting in a large deviation from the FMR picture about a poorly defined
center frequency and greatly enhanced damping through inhomogeneous dephasing.

The Two-Magnon Scattering (TMS) process [4, 5] accounts for anisotropic damping in most systems by providing a mechanism by which the FMR-mode decays into other, degenerate spin-wave modes via scattering at defects. This process manifests as an effective damping with a symmetry mirroring that of the defects. There is a strong lattice mismatch at the Ni(001)/MgO(001) interface (16% as measured with X-ray diffraction) which encourages defects with fourfold-symmetry. FMR scans (Fig. 4.15, open squares) reveal a characteristic TMS decrease in the decay time as the field is rotated out of plane, indicating the presence of TMS in this system. However, TMS does not account for the large reduction in \( \tau_{MO} \). There is a significant discrepancy between FMR and TR-MOKE for the hard axis. Furthermore, TMS shuts off in the normal geometry, since the degeneracy condition for scattering is broken; however, TR-MOKE scans yield a decay time nearly one third of the expected amount. This shows that TMS is neither the dominant decay mechanism nor the source of anisotropy in \( \tau_{MO} \).

The low frequency limit (Fig. 4.16) yields further evidence of the PIAD effect. Dominant in this limit, the magnetic anisotropy magnetizes the sample along the easy axis and precession oscillations approach a common frequency (Fig. 4.16 c), independent of the direction of the weak external field. However, pumped spins initially rotate in the direction of the external field, as described above. The subsequent inhomogeneity primarily depends on this degree of rotation, i.e., the direction of the external field relative to the anisotropy, and not the amplitude of the field itself. Thus, while the precession frequencies approach a common weak-field limit, the damping exhibits a heavy anisotropy (Fig. 4.16 b). To illustrate this effect, we expand the
observed TR-MOKE decay rate to first order in frequency: \( \tau_{\text{MO}}^{-1} = \tau_0^{-1} + C \omega_0 \), where \( \tau_0 \) denotes the effective decay time at zero-frequency, and \( C \) is a dimensionless constant that accounts for the field dependency. We obtain \( \tau_0 \) and \( C \) from a weighted least-squares fit to the data (solid lines in Fig. 4.16 b) for both the easy and the hard axes.

Figure 4.16: TR-MOKE dynamics for in-plane easy (\( \triangle \)) in-plane hard (\( \triangledown \)), and normal (\( \square \)) geometries. Panel (a): observed decay time vs. external field magnitude. Panel (b): observed decay time vs. center frequency \( \omega_{\text{MO}} \). Solid lines are fits to data, as discussed in the text. Panel (c): center frequency vs. the external field. Error bars represent one standard error of the mean.

The transient reorientation of the magnetization during the excitation process at low field determines \( \tau_0 \). Since the easy axis geometry minimizes the angle between the applied and the anisotropy fields, we expect the longest decay time for this case. Indeed, we observe \( \tau_0 = 0.37 \pm 0.05 \) ns, identical within the error to the intrinsic limit \([88, 77]\). The converse is true for the hard axis, for which we observe a 43%
4.4 Pump Induced Anisotropic Damping

reduction: \( \tau_0 = 0.21 \pm 0.05 \text{ ns} \). As the applied field increases, the effective field rotates away from the easy axis and generates greater perturbations in the excited spin distribution. This additional field dependency manifests in the parameter C, which increases 170\%, from 0.0302 \( \pm \) 0.0116 at the easy axis to 0.0812 \( \pm \) 0.0232 at the hard axis. The anisotropy in both \( \tau_0 \) and C indicate that the damping in TR-MOKE couples to the anisotropy through the excitation process unique to this technique.

Finally, we present a phenomenological model that captures the essential features of \( \tau_{\text{MO}} \) (shown for \( B = 73 \text{ mT} \) in Fig. 4.17 vs. azimuthal angle) by relating the anisotropy directly to the excited distribution of spins. Since the pump mechanism involves a temporary change in the anisotropy – effectively a switch – we postulate that the excited distribution roughly follows the rate of change of the effective field, i.e., the equilibrium direction, \( \hat{e}_M \), with respect to the external field direction, \( \hat{e}_H \):

\[
\tau_{\text{MO}}^{-1} \approx \left| \frac{d\theta_M}{d\theta_H} \right|.
\]

In the case where we apply the external field in the plane of the sample, \( \frac{d\theta_M}{d\theta_H} \approx \frac{d\phi_M}{d\phi_H} \). Here, \( \phi_M \) represents the angle that minimizes the magnetic free energy, \( E = -\mathbf{H} \cdot \mathbf{M} + \frac{1}{2} \mu_0 \mathbf{M}_s^2 \cos^2 \theta_M - \frac{1}{2} K_2 \cos^2 \theta_M - \frac{1}{2} K_{4\perp} \cos^4 \theta_M - \frac{1}{8} K_{4\parallel} (3 + \cos 4\phi_M) \sin^4 \theta_M \), where \( \mathbf{M}_s \) is the saturation magnetization, and \( K_2, K_{4\perp}, \) and \( K_{4\parallel} \) are energy constants. Using this model and the anisotropy parameters obtained from the FMR measurements (\( K_2 = -108 \pm 10 \text{ kJ/m}^3, K_{4\perp} = -5.1 \pm 1 \text{ kJ/m}^3, K_{4\parallel} = -2.4 \pm 0.5 \text{ kJ/m}^3 \)), we reproduce the measured evolution of the dephasing time as a function of the azimuthal angle for \( B = 73 \text{ mT} \) as shown in Fig. 4.17, thus demonstrating that the effective damping couples to the magnetic anisotropy.
4.4 Pump Induced Anisotropic Damping

Figure 4.17: TR-MOKE decay time vs. in-plane external field angle for $B = 73$ mT. The solid line is calculated from the magnetic anisotropy, as discussed in the text.
Chapter 5

Conclusion

5.1 Summary

We have investigated the damping of spin waves in ferromagnetic Au(3 nm)/Ni(10 nm)/MgO(001) using two techniques: pump probe TR-MOKE and ferromagnetic resonance. In the optical measurements, we used a pump beam pulse to thermally excite coherent spin precession. We measured the magnetization precession in the time domain as a function of field amplitude and direction. We found that the extracted precession frequency was well-characterized by the Kittel model, which incorporated fit parameters obtained from FMR. While the frequency response was similar in both TR-MOKE and FMR, we observed additional source of damping in the TR-MOKE measurements which exhibited strong anisotropy.

The enhanced damping in the optical measurements is a consequence of inhomogeneous broadening generated by the pump beam in the presence of magnetic anisotropy. This new damping mechanism, pump-induced anisotropic damping (PIAD), is a con-
sequence of the differing temperature recovery of the magnetization and anisotropy. After the sample is heated by the pump pulse, the magnetization recovers initially in the direction of the applied external field, as the magnetic anisotropy has a gradual temperature onset. If the external field is applied in a direction away from the global energy minimum, pumped spins will recover in a different direction than unpumped spins; this inhomogeneity results in damping through dephasing. The sensitivity of the broadening to the direction of the external field relative to the anisotropy axis results in the observed anisotropy in the damping. Comparison between FMR and TR-MOKE shows that the damping is consistent if the field is applied along the easy axis of magnetization; however, in the case when the field is applied normal to the sample, TR-MOKE results in critically damped oscillations, while the decay time as measured by FMR is a factor of five times longer.

Finally, we developed a simple model which links the anisotropy parameters to the excited mode distribution by considering that the deviation between pumped and non-pumped spins is large in regions where it is relatively easy to perturb the magnetization away from equilibrium. We thus predict that the decay time should correspond to the curvature of the free energy, which was calculated for the case where the magnetization is in plane using the anisotropy parameters obtained from FMR. Comparison with the data shows that the two are in agreement up to a scaling factor.
5.2 Outlook

There are additional avenues for research, which are briefly outlined below.

5.2.1 Unidirectional Anisotropy

A first step would be to conduct a study of the PIAD effect in the case where the dominant anisotropy is unidirectional, i.e., in an exchange-biased system. The presence of an unidirectional anisotropy should result in an enhanced unidirectional damping, which might be useful in spin transport experiments. Additionally, there is the possibility of using the optical pulse to alter the anisotropy, as evidenced by the previous work of Seu et al. [113] in which it was shown that an ultrafast pump pulse can induce a pinning state in exchange-biased FeMn/Co. Incorporating a pump-induced exchange bias might enable a way to simultaneously tailor two of the desired dynamic characteristics: the recovery state (as determined by the pinning field) and the relaxation time (as determined by PIAD). It thus might be possible to construct devices which can switch functionality depending on the application of an ultrashort optical pulse.

5.2.2 Meta materials

Meta materials provide an additional interesting direction for future work. Recent work by Muzenburg et al. [114] used pump-probe TR-MOKE to probe spin wave dynamics on patterned Ni films. They found that in this case, the pump beam excites a very broad spectrum of relatively long (ns) lived oscillations, which they attributed to local field inhomogeneities at pattern sites. By altering the dimensions
of the pattering matrix, it is possible to alter the persistence of the higher order spin wave modes. This would be a useful testing ground for future investigations of the PIAD effect.

5.2.3 Half Metals

All-optical pump-probe techniques have recently been employed to investigate the magnetization dynamics in the half metals [115, 116], which are particularly promising candidates for spintronic applications, as they simultaneously act as both conductors and insulators, depending on the transport spin band. Zhang, et al. [117] found a very slow demagnetization time relative to normal metals in TR-MOKE experiments. In half-metals, the population of the minority spin band limits Elliot-Yafet (spin-flip) scattering; thus the electron-spin channel in the excitation process is effectively blocked in these materials, and the demagnetization time represents a diagnostic parameter of half-metallicity [118]. It would be interesting to observe how the increased magnetization recovery time in these materials alters the observed damping, since in this case, the thermal lag between the pumped spins and the magnetic anisotropy is reduced.
Bibliography


