INVESTIGATION OF MAGNETIC RELAXATION MECHANISMS AND DYNAMIC MAGNETIC PROPERTIES

IN THIN FILMS USING FERROMAGNETIC RESONANCE (FMR) TECHNIQUE

by

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A THESIS

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ABSTRACT

Investigating the damping processes and the behavior of dynamic magnetic properties in ferromagnetic thin films has been an important key towards design and fabrication of different microwave and magnetic recording devices. This thesis discusses the dynamic magnetic properties and also the physics behind different relaxation mechanisms in ferromagnetic thin films using comprehensive experimental investigations by means of broadband ferromagnetic resonance (FMR) technique. In chapter one the basics of ferromagnetic resonance technique and the experimental features of the FMR setup used in this study are discussed, also the FMR data analysis is explained. Chapter two is devoted to the study of the interfacial perpendicular magnetic anisotropy (PMA) and damping parameter in Co2FeAl thin films. In chapter three broadband temperature dependent FMR measurements were carried out on Ni80Fe20/Gd thin films to investigate the behavior of ferromagnetic relaxation, and gyromagnetic ratio as the system goes through the Curie temperature of Gd. In chapter four, the ferromagnetic relaxation mechanisms in ferrites are discussed. The low loss Nikel Ferrite and Lithium ferrite single crystal, thin and ultra-thin films were characterized by detailed FMR measurements to investigate the effect of microstructural defects on the magnetization relaxation. A comprehensive study on the interlayer exchange coupling strength in Co90Fe10/Ru/Co90Fe10 multilayers is the subject of chapter five, in which the mutual spin pumping is discussed as a recently discovered channel for relaxation in exchange coupled multilayers.
DEDICATION

To

My wife, Elahe Saeidi

My beloved mother, Shekoofe Parvizi
**LIST OF ABBREVIATIONS**

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Full Form</th>
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<tbody>
<tr>
<td>AC</td>
<td>Alternative current</td>
</tr>
<tr>
<td>AF</td>
<td>Anti-ferromagnetic</td>
</tr>
<tr>
<td>BTIBD</td>
<td>Biased target ion beam deposition</td>
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<tr>
<td>CFA</td>
<td>Cobalt Iron Aluminum</td>
</tr>
<tr>
<td>CPW</td>
<td>Coplanar waveguide</td>
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<tr>
<td>CVD</td>
<td>Chemical vapor deposition</td>
</tr>
<tr>
<td>DAQ</td>
<td>Data Acquisition card</td>
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<tr>
<td>DC</td>
<td>Direct current</td>
</tr>
<tr>
<td>DLI</td>
<td>Direct liquid injection</td>
</tr>
<tr>
<td>DOS</td>
<td>Density of states</td>
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<tr>
<td>FM</td>
<td>Ferromagnetic</td>
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<tr>
<td>FMR</td>
<td>Ferromagnetic resonance</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full width at half maximum</td>
</tr>
<tr>
<td>GMR</td>
<td>Giant Magneto Resistance</td>
</tr>
<tr>
<td>IEC</td>
<td>Interlayer exchange-coupled</td>
</tr>
<tr>
<td>LFO</td>
<td>Lithium Ferrite</td>
</tr>
<tr>
<td>LL</td>
<td>Landau-Lifshitz</td>
</tr>
<tr>
<td>LLG</td>
<td>Landau-Lifshitz Gilbert</td>
</tr>
<tr>
<td>MMIC</td>
<td>Monolithic microwave integrated circuits</td>
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<tr>
<td>MTJ</td>
<td>Magnetic tunnel junctions</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Description</td>
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<tr>
<td>--------------</td>
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<tr>
<td>NFO</td>
<td>Nickel Ferrite</td>
</tr>
<tr>
<td>PC</td>
<td>Personal computer</td>
</tr>
<tr>
<td>PLD</td>
<td>Pulsed laser deposition</td>
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<tr>
<td>PSSW</td>
<td>Perpendicular standing spin wave</td>
</tr>
<tr>
<td>PMA</td>
<td>Perpendicular magnetic anisotropy</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning electron microscopy</td>
</tr>
<tr>
<td>SLK</td>
<td>Slater Loudon and Kittel</td>
</tr>
<tr>
<td>STTR-MRAM</td>
<td>Spin transfer torque magnetic random access memory</td>
</tr>
<tr>
<td>TEM</td>
<td>Transmission electron microscopy</td>
</tr>
<tr>
<td>TMS</td>
<td>Two magnon scattering</td>
</tr>
<tr>
<td>VSM</td>
<td>Vibrating sample magnetometer</td>
</tr>
<tr>
<td>XRD</td>
<td>X-ray diffraction</td>
</tr>
<tr>
<td>YIG</td>
<td>Yttrium iron garnet</td>
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</table>
ACKNOWLEDGMENTS

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1. INTRODUCTION

I. Ferromagnetic resonance

There is a historical evolution in the form of phenomenological equations that have been introduced to represent the magnetization dynamics in ferromagnetic systems [1,2,3,4]. Among all, the following equations known as Landau-Lifshitz (LL) and Landau-Lifshitz Gilbert (LLG) equations have a better agreement with the physical characteristics of ferromagnetic systems

\[
\text{LL: } \frac{d\vec{M}}{dt} = \gamma \vec{M} \times \vec{H}_{\text{eff}} - \frac{\alpha \gamma}{M_s} \left( \vec{M} \times \vec{M} \times \vec{H}_{\text{eff}} \right) \tag{1.1}
\]

\[
\text{LLG: } \frac{d\vec{M}}{dt} = \gamma \vec{M} \times \vec{H}_{\text{eff}} - \frac{\alpha}{M_s} \left( \vec{M} \times \frac{d\vec{M}}{dt} \right) \tag{1.2}
\]

where $\vec{M}$ is the magnetization vector, $\vec{H}_{\text{eff}}$ is the effective internal magnetic field, $\alpha$ is the damping parameter, $\gamma$ is the gyromagnetic ratio and $\gamma$ is equal to $\frac{\gamma}{1 + \alpha^2}$. $\vec{H}_{\text{eff}}$ is the sum of quasi static, anisotropy and microwave magnetic fields. In both equations the first term represents the precessional motion and the second term describes the ferromagnetic dissipation or damping. The LLG and LL equations are mathematically equivalent.

The LLG differential equation can be solved to determine the ferromagnetic resonance (FMR) frequency, while neglecting the second term on the RHS of the LLG equation (damping term) as its magnitude is much smaller than the first term representing processional motion. For a
bulk ellipsoidal shape ferromagnetic sample with principal axes along x,y and z directions and cubic crystallographic symmetry, the ferromagnetic resonance (FMR) frequency is found to be [5]

\[ \dot{f} = \gamma \sqrt{\left[H_{res} + 4\pi M_s (N_{yy} - N_{zz})\right] \left[H_{res} + 4\pi M_s (N_{xx} - N_{zz})\right]} \]  

(1.3)

Where \( H_{res} \) is the resonance field, \( M_s \) is the saturation magnetization and \( N_{xx}, N_{yy} \) and \( N_{zz} \) are the diagonal elements of the demagnetization tensor for ellipsoidal geometry.

\[ \bar{N} = \begin{pmatrix} N_{xx} & 0 & 0 \\ 0 & N_{yy} & 0 \\ 0 & 0 & N_{zz} \end{pmatrix} \]  

(1.4)

In the case of a thin film (figure 1.1) \( N_{zz} = 1, N_{xx} = N_{yy} = 0 \). Also, due to the different contributions from the demagnetization field to the \( \bar{H}_{eff} \) two geometries, which are in-plane and out-of-plane, are considered. In the in-plane geometry the quasi static or external magnetic field lies in the plane of the thin film, while the out-of-plane geometry represents the case in which the external magnetic field is applied perpendicular to the plane of the film. In the FMR equations of thin films a quantity called effective magnetization is used, that is given by [6,9]

\[ 4\pi M_{eff} = 4\pi M_s - \frac{2K_u}{M_s} \]  

(1.5)

and contains two contributions, which are the demagnetization field \( 4\pi M_s \) and the uniaxial perpendicular magnetic anisotropy \( \frac{2K_u}{M_s} \). The demagnetization field and a uniaxial perpendicular anisotropy have the same functional dependence on the angle of the magnetization with the film normal. Therefore, these two terms cannot be separated in an FMR experiment.
In thin films the LLG equation can be linearized for both geometries using the fact that the time dependent components of the magnetization and effective magnetic field are much smaller than the corresponding static components [6,7], to find the solutions of the LLG equations. The FMR frequencies or the solutions to the LLG equation for in-plane and out-of-plane geometries can also be found using the Smit-Beljers energy minimization approach [8,9], in which the resonance frequency is given by the following equation

\[
\left(\frac{f}{\gamma}\right)^2 = \frac{1}{M_s^2 \sin^2 \theta} \left[ \frac{\partial^2 E}{\partial \theta^2} \frac{\partial^2 E}{\partial \phi^2} - \left( \frac{\partial^2 E}{\partial \theta \partial \phi} \right)^2 \right]
\]

where \(E\) is the magnetic free energy density and \(\theta\) and \(\phi\) are the azimuthal and polar angles of the magnetization. The second derivatives are determined at the equilibrium of the magnetization \((\theta_0, \phi_0)\), which minimize the free energy and thus the first derivatives vanish.

Regardless of the adopted approach the following equations are found for the resonance frequency in the tetragonal symmetry which is relevant in the thin films [6,9](see figure 1.1).

\[
f_{ip} = \gamma \sqrt{\left( H_{res} + \frac{2K_4}{M_s} (\cos 4\phi)(4\pi M_{eff} + H_{res} + \frac{K_4}{2M_s}(3 + \cos 4\phi)) \right)}
\]

\[
f_{oop} = \gamma (H_{res} - 4\pi M_{eff})
\]

where \(K_4\) is the four-fold in-plane anisotropy constant. The equations (1.7) and (1.8) are valid for the in-plane and out-of-plane geometries, respectively. In both cases it has been assumed that the magnetization vector is perfectly aligned with the external magnetic field, i.e. \(\theta = \theta_H\) and \(\phi = \phi_H\) (see figure 1.1)
Figure 1.1: Schematic showing the tetragonal symmetry which is usually valid for the magnetic thin films. \( \theta (\theta_H) \) and \( \phi (\phi_H) \) are the azimuthal and polar angles of the magnetization (external magnetic field).

In a typical FMR measurement, at a fixed microwave frequency, the external magnetic field is swept through the resonance field of the sample. By measuring the \( H_{res} \) vs. \( f \) and fitting to the correct functional dependence (equations (1.3), (1.7), (1.8)) gyromagnetic ratio \( \gamma \), effective magnetization \( M_{eff} \) and a wide verity of magnetic properties such as crystallographic magnetic anisotropies, interface magnetic anisotropy, exchange stiffness and exchange coupling field can be determined. These properties will be discussed extensively in chapters 4 and 5 and partly in chapters 2 and 3.

The peak-to-peak FMR linewidth \( \Delta H \) (defined in section III) on the other hand contains information about relaxation mechanisms in ferromagnetic systems. According to the Gilbert phenomenology the FMR signal linewidth is related linearly to the FMR frequency and damping.
\[ \Delta H = \Delta H_0 + \frac{2}{\sqrt{3}} \frac{\alpha f}{\gamma} \]  

where \( \Delta H_0 \) represents the inhomogeneous linewidth broadening due to the imperfections, defects or other microstructures that can locally modify the internal magnetic field. However, as will be discussed in this thesis not all relaxation mechanisms obey the Gilbert phenomenology.

Comprehensive FMR measurements containing a combination of broadband frequency, angle and temperature dependent measurements are used to investigate the relaxation mechanisms in ferromagnetic systems such as Co\textsubscript{2}FeAl and NiFe\textsubscript{2}O\textsubscript{4} thin films, Ni\textsubscript{80}Fe\textsubscript{20}/Gd heterostructure and Co\textsubscript{90}Fe\textsubscript{10}/Ru/Co\textsubscript{90}Fe\textsubscript{10} trilayers in chapters 2-5.

**II. Experimental setup**

A schematic of the FMR setup used in the measurements presented in this thesis is shown in figure 1.2. A Coplanar Waveguide (CPW) is used to send a microwave signal to the place of the sample. The width of the central conductor and the dielectric gap of the CPW are designed to match the microwave cable impedance of 50 Ohms, thus avoiding back reflection at the microwave cable-CPW junction.
Figure 1.2: Schematic of the broadband FMR setup. Digital and Analog (DC/AC) connections are shown with dashed and solid lines, respectively. The end launchers at CPW are connected to the microwave source and diode using microwave cables. DAQ card is used to translate the digital signal (provided by the PC) to a DC signal and thus set the current field in the electromagnet using a power supply. An ac signal with a specific reference frequency is provided to the modulation coils by the Lock-in amplifier and the transmitted microwave signal is fed into a microwave diode and the signal oscillating with the reference frequency is detected by a lock-in amplifier.

Using CPW enables carrying out FMR measurements in a broad frequency range. There are usually large systematic error margins in the reported values of FMR properties arising from the limited number of frequencies available for the measurements, which is minimized by using CPW in a broad-band setup [10].
The signal transmitted across the sample placed on the CPW is fed into a microwave diode, and a lock-in [11,12] amplifier is used to detect the microwave diode signal. The use of lock-in technique together with applying a small ac magnetic field, oscillating with the lock-in reference frequency on top of the external magnetic field, enables the detection of the small microwave absorption of the sample at ferromagnetic resonance.

The actual microwave absorption signal is very small for thin films due to their small total magnetic moment, therefore a signal amplification is much desired. A typical AC signal amplifier has a large bandwidth and thus the noise level even impedes the amplified signal. This problem is resolved by lock-in amplifier which operates as an amplifier followed by a narrow-band filter [13].

![Image](image1.png)

Figure 1.3: (a),(b) Room temperature FMR setup. Stepper motors are used to change the in-plane or out-of-plane angles, the sample placed on a CPW is irradiated with a microwave signal and the transmitted power is measured. (c) A closed cycle cryostat is used to carry out low temperature FMR measurements.

Pictures of the broadband FMR setup used in this thesis can be seen in figures 1.3 (a-c) and 3. The setup operates in the frequency range of 1 to 65 GHz and the temperature range of 10K to 300 K. The angle between field and the direction normal to the thin film and also the in-plane angle between the field and sample sides can be changed automatically. The interface
between the PC and all other equipment is handled with Labview codes to form a fully automated broadband FMR setup.

A similar custom designed setup is used with a closed cycle cryostat for temperature dependent broadband FMR measurements as shown in figure 1.3 (c), enabling measurements from 10K to 300K. In many cases knowing the temperature dependent FMR properties is helpful to further understand and investigate the dynamic magnetic properties and relaxation mechanisms in magnetic thin films.
III. Data Analysis

In the FMR measurements, the component of an incident microwave transverse to the direction of external magnetic field is used to excite the uniform magnetic precession mode in a ferromagnetic sample, which is also subjected to a quasi-static external magnetic field. At a fixed microwave frequency, the external magnetic field is swept through the resonance field of the sample. The microwave loss at the resonance condition can be detected by measuring the microwave power transmitted through the sample, in which case the FMR appears as a reduced transmission with a Lorentzian lineshape. Because of the lock-in technique used to detect this small change in transmission the derivative of this Lorentzian lineshape with respect to the magnetic field, is recorded as shown in figure 1.4. Working with a derivative of the microwave loss has the benefit of eliminating any possible offset in the signal, which translates into a better signal to noise ratio. This derivative of the Lorentzian lineshape or the raw FMR signal is fitted with the following model which is a sum of dispersive and absorptive terms

\[
y = \frac{a \left( \frac{H_{res} - H}{\Delta H} \right) + 9b - 3b \left( \frac{H_{res} - H}{\Delta H} \right)^2}{\left( \frac{H_{res} - H}{\Delta H} \right)^2 + 3}^2
\]

where \(a\) and \(b\) are the absorption and dispersion amplitudes [14]. The resonance field \(H_{res}\) and the peak-to-peak linewidth \(\Delta H\) are the fit parameters used throughout this thesis. In a simple picture where power loss is solely due the ferromagnetic resonance, one would expect to observe just an absorptive lineshape. However, in addition to the FMR or magnetic loss
there exist dielectric or conductive losses which would contribute to the experimental FMR signal as dispersive or asymmetric lineshapes.[15,16]

Figure 1.4: The derivative of transmitted power with respect to the magnetic field (blue line) is fitted (red line) using equation (1.11) to determine $H_{res}$ and $\Delta H$. data Corresponds to a 5nm Co$_2$FeAl sample measures at 20 GHz.
Figure 1.5: (a) FMR spectrum measured at three different applied modulation fields. Inset shows the $R^2$ as a function of modulation field which indicates the FMR spectrum fit quality decreases with the increase of modulation field. (b) FMR linewidth as a function of modulation field amplitude. (c) FMR signal absorption amplitude as a function of modulation field amplitude. Data correspond to a 35 nm Ni$_{80}$Fe$_{20}$ thin film. One can also numerically calculate the dependence of the amplitude and measured peak-to-peak FMR linewidth on the modulation field, for details see T. Mewes et al. [17].

By increasing the modulation field one can increase the amplitude of the measured signal, see figure 1.5(a) and (c). However, if the amplitude of the applied modulation field is comparable or larger than the linewidth the lineshape will be distorted. This as shown in the figure 1.5(b) for a 35nm Ni$_{80}$Fe$_{20}$ sample will lead to an overestimation of the real physical linewidth, and also as shown in the inset of figure 1.5 (a) will result in a lower quality for the fit (equation 1.10). Here the goodness of the fit is described by $R^2$ which has a value between zero and one. An $R^2$ value of 1 means a perfect agreement between the experimental data and the fit
function. Therefore, one needs to choose the appropriate modulation field which increases the FMR signal while not overmodulating and thereby distorting the lineshape. This becomes particularly important when measuring samples with very small FMR signals. For all the measurements presented in this study, appropriate modulation fields were used.
2. \textbf{Co}_2\text{FeAl THIN FILM}

Half metallic ferromagnets are promising candidates for spintronic devices, on account of their high spin polarization and low Gilbert damping parameter [18,19, 20]. However, for applications such as Spin Transfer Torque MRAM (STT-MRAM), in addition to the above mentioned characteristics, thin film structures with a strong perpendicular magnetic anisotropy (PMA) are also desired [21,22,23]. As will be discussed in this chapter, the Co$_2$FeAl Heusler thin films capped with MgO exhibit a significant interfacial PMA and also a low Gilbert damping parameter, which makes them a material of choice for STT MRAM applications. The results discussed in this chapter have already been published in Applied Physics Letters [24] and Journal of Applied Physics [25]

1. \textbf{Sample preparation}

Biased Target Ion Beam Deposition (BTIBD) technique was used by collaborators at the University of Virginia to synthesize epitaxial Co$_2$FeAl films [24,25]. The use of BTIBD technique provides the opportunity of investigating the effect of chemical ordering on the damping parameter and PMA in Co$_2$FeAl(CFA) thin films. Two sets of CFA samples were deposited to investigate different magnetic properties using broadband FMR characterization. The Cr(40 nm)/ Co$_2$FeAl(t)/ MgO(2.3 nm)/ Ru(5 nm) structures were deposited on MgO(001) substrates to study the behavior of interfacial PMA as a function of heat treatment and the
effective thickness of the CFA layer. Rapid thermal annealing for 30s at 350-450 °C was used for the heat treatment. The aforementioned stack structure is referred as series-1 throughout this chapter.

Co$_2$FeAl(50 nm)/ Al(5 nm) structures were deposited on MgO(001) substrates, the substrates were annealed at 350-450 °C for 1 hour and pre-cleaned for 5 minutes using argon/oxygen plasma to improve the epitaxiality. These structures of CFA thin films are referred as series-2, the effect of chemical ordering on the damping parameter was investigated for this series.

**II. Perpendicular magnetic anisotropy**

Broadband ferromagnetic resonance is a suitable tool to investigate the perpendicular magnetic anisotropy in thin film structures. Figure 2.1(a) shows the FMR signal at 20 GHz and figure 2.1 (b) represents the broadband ferromagnetic resonance frequency vs. resonance field for the Cr(40 nm)/ Co$_2$FeAl(1.33 nm)/ Mg (0.7 nm)/MgO(2.3 nm)/ Ru(5 nm) sample. Note that the thin Mg layer was used to avoid the over-oxidation of the Co$_2$FeAl layer by MgO. As the FMR measurements were done in the out-of-plane geometry the experimental data shown in figure 2.1(b) are fitted using the out-of-plane Kittel formula (equation 1.8) to determine FMR properties such as gyromagnetic ratio $\gamma$ and effective magnetization $M_{eff}$. The effective magnetization is defined as

$$4\pi M_{eff} = 4\pi M_s - \frac{2K_u}{M_s}$$

where the first term on the right hand side of the equation (2.1) represents the demagnetization field and the second term is the PMA field.
Figure 2.1: (a) FMR raw spectra at 20 GHz for the Cr(40 nm)/ Co2FeAl(1.33 nm)/ MgO(2.3 nm)/ Ru(5 nm) (RTA 10s at 400°C) sample, red line is the fit to experimental data (equation 1.10) (b) Resonance frequency vs. resonance field or the Kittel plot for the same sample, red line shows the out-of-plane fit to the experimental data (equation 1.8). FMR measurements were carried in the out-of-plane geometry.

According to equation 2.1 a negative value for $M_{eff}$ means that the PMA field exceeds the demagnetization field and the sample will magnetize perpendicular to the film plane, whereas a positive value for $M_{eff}$ indicates that the magnetization prefers to stay in the plane of the film.
Figure 2.2: Effective magnetization (a) as a function of thickness for the Cr(40 nm)/Co2FeAl(0.57 nm)/MgO(2.3 nm)/Ru(5 nm) (RTA 10s at 350°C) (b) as a function of annealing temperature. The sample is magnetized out-of-plane at 0.57 nm and 1.33 nm, while at the thickness of 1.90 nm the magnetization lies in the plane of the film. The highest PMA (larger negative $M_{\text{eff}}$) is achieved at 350°C, with further increase of annealing temperature PMA decreases.

The effective magnetization is shown as a function of Co$_2$FeAl layer thickness for the series-1 samples in figure 2.2(a). While figure 2.2(b) shows the annealing temperature dependence of $M_{\text{eff}}$ for the 0.57 nm thick CFA sample. It is noteworthy to mention that by “thickness” in this study, exclusively the effective thickness is meant considering the presence of a magnetically dead layer in the thin film stack [24].

The perpendicular magnetic anisotropy (PMA) in thin films is the sum of two different contributions which are the interface PMA and the bulk PMA [26,27]. The bulk PMA includes crystallographic anisotropy, while the interface PMA can arise from the spin orbit interaction between the magnetic and non-magnetic atoms at the interface and can be modified by surface
roughness [23,27]. The PMA can thus be written as follows

\[ K_u = K_V + \frac{K_i}{t} \]  

(2.2)

Where \( K_V \) is the bulk and \( K_i \) is the interfacial PMA anisotropy constant and \( t \) is the thickness of the magnetic layer. The increase in the contribution of interface magnetic anisotropy with decreasing the thickness of Co2FeAl layer pulls the magnetization out of the film plane and forms an out-of-plane easy axis below a certain thickness, which can be seen from in figure 2.2(a).

Provided that the MgO layer is properly oxidized, at the interface with a Co/Fe based ferromagnetic layer, an interfacial perpendicular anisotropy is created. This phenomenon has been explained theoretically [28] and reported experimentally [29,30]. The hybridization within Co/Fe-3d orbits and between Co/Fe-3d and O-2p orbits caused by weak spin-orbit interaction (SOI) alters the degeneracy of the electronic states at the Fermi level. As a result, the energy level related to the out-of-plane orientation of the magnetization becomes the preferable ground-state and thus the interfacial perpendicular anisotropy is created [28]. One cannot see a significant interface PMA in the as-deposited sample (not shown) since the protective 0.7nm Mg capping layer might have been oxidized which could cause Co/Fe oxidization at the interface, which on the other hand can lead the elimination of the energy splitting and thus suppresses the interface PMA [28,29,30].

The PMA has its extremum (most negative \( M_{eff} \)) value at 350 °C. This is to be expected considering the increase in the ordering parameter with increasing annealing temperature [24]. However, at temperatures higher than 350 °C as shown in figure 2.2(b), the interface PMA
decreased with increasing annealing temperature, which could be related to the reduction of Co/Fe-O bound at the interface [30].

III. Relaxation

The effect of annealing treatment on the Gilbert damping parameter was investigated for the series-2 samples, i.e. the MgO/Co$_2$FeAl(50 nm)/Al(5 nm) structures. Conventional furnace annealing (1 hour) was used for the heat treatment of the series-2 samples with the temperature range of 350 °C to 600 °C. The crystalline structures of the films were measured using a high resolution X-Ray diffractometer with Cu Kα radiation [25].

![Figure 2.3](image)

Figure 2.3 : (a) 2θ scan of the Co$_2$FeAl film annealed at 500, 550 and 600 °C for 30 (b) The ordering parameter $S$ which is proportional to the ratio between (002) and (004) peaks [25], as a function of annealing temperature

The 2θ of XRD sans are shown in figure 2.3. The observed (004) peak is attributed to the cubic symmetry of the Co$_2$FeAl thin film, while the (004) peak corresponds to the additional B2 ordering [25]. Thus, a long range ordering parameter $S$, was defined to quantify the B2 ordering
in the samples [25, 31]. All the fabrication processes and microstructural characterizations of the thin films were carried out at the University of Virginia.

Figure 2.4: (a) The FMR spectrum for the MgO/Co$_2$FeAl(50 nm)/ Al(5 nm) sample annealed 350 °C for one hour. The low field mode signal is a PSSW mode and the high field signal represents the FMR mode. (b) Linewidth vs. frequency data for the FMR mode, an upper limit for the Gilbert damping parameter can be estimated using the highest frequency data.

Figure 2.4 (a) shows a typical FMR spectrum for the MgO/Co$_2$FeAl(50 nm)/ Al(5 nm) sample where two peaks are observed. The low field mode is a perpendicular standing spin wave (PSSW) mode, whose characteristics will be discussed in detail in chapter 4, and the high field mode represents the FMR signal. Figure 2.4(b) shows the broadband frequency dependence of the FMR mode linewidth for the same sample.

An upper limit for the Gilbert damping parameter was estimated using the highest frequency data and assuming no offset for the linewidth at zero frequency (see equation 1.9). As will be discussed in the next chapters the nonlinear frequency dependence of the linewidth shown in figure 2.4(b) is most likely caused by the two magnon scattering mechanism.
As shown in figure 2.5 the B2 ordering parameter increases while the damping parameter decreases with increasing the annealing temperature. Similar behavior has also been previously reported for Heusler Co$_2$FeAl and Co$_2$Fe$_x$Mn$_{1-x}$Si alloys [32,33] and Co$_{1-x}$Fe$_x$Ge thin films [34].

Figure 2.5: The dependence of B2 ordering and damping parameters on annealing temperature for the MgO/Co$_2$FeAl(50 nm)/Al(5 nm) system heat treated for 1 hour using conventional furnace annealing.

Dependence of the damping parameter on annealing temperature and thus on the B2 chemical ordering parameter, can be understood by the Kamberský torque-correlation relaxation mechanism. The Kamberský relaxation mechanism is the dominant Gilbert-like damping process in the transition metals. In a simple picture the angular momentum can be transferred from the spin system to the free electrons through the weak spin-orbit coupling (SOC) interaction and finally quenched due the itinerancy of free electrons[35].
According to the torque correlation model the spin-orbit contribution to the Gilbert damping parameter is given by [36,37]

$$\alpha = \frac{\mu_0 g^2 \mu_B^2}{\hbar} \frac{\pi^2}{2\Omega_{at}} \left( \sum_{m,n} |\Gamma_{mk,nk}|^2 W_{mk,nk} \right)_k$$

(2.4)

Where $g$ is the Landé factor, $\mu_0$ is the permeability of free space, $\Omega_{at}$ is the atomic volume, $\mu_B$ is the Bohr magneton, $n$ and $m$ are band indices, $k$ is the electron wave vector and $<>_k$ denotes the average over the first Brillouin zone. The $\Gamma_{mk,nk}$ are the transition matrix elements that represent the spin-orbit interaction between states $m$ and $n$. The $W_{mk,nk}$ is the spectral overlap function that measures the overlap between the average densities of states (DOS), which themselves depend on electron-lattice scattering life time $\tau$. Therefore the Gilbert-like damping parameter is proportional to the density of states at the Fermi level.

As explained by Y. Miura et al. the DOS near the Fermi level can change as the crystal structure goes from L2$_1$ phase into the B2 phase [37]. As reported previously, since the B2 ordering parameter of Co$_2$FeAl increases from 0 to 1, the DOS at the Fermi level is reduced to one third of its initial value[32]. This reduction of the DOS with increasing ordering parameter can explain the decrease of the Gilbert damping parameter seen in figure 2.4.
3. Ni\textsubscript{80}Fe\textsubscript{20}/Gd HETEROSTRUCTURES

I. Introduction

In some cases, it is very helpful to measure the broadband ferromagnetic resonance properties of thin film structures at different temperatures to further understand and investigate the relaxation mechanisms. In this chapter, the temperature dependent FMR properties of Ni\textsubscript{80}Fe\textsubscript{20}/Gd bilayer structures are discussed. Considering the low Curie temperature of Gd thin films\cite{38,39}, the effect of the paramagnetic-ferromagnetic phase transition in the Gd layer on the magnetization dynamics of the Ni\textsubscript{80}Fe\textsubscript{20}/Gd system is studied experimentally.

The SiO/ Ni\textsubscript{80}Fe\textsubscript{20} (5nm)/Gd(3nm)/Ru(3nm) and SiO/ Ni\textsubscript{80}Fe\textsubscript{20} (10nm)/Gd(3nm)/Ru(3nm) structures studied here were synthesized using magnetron sputtering deposition technique\cite{40}. All the FMR measurements were carried out in the in-plane geometry.

II. Temperature dependence of gyromagnetic ratio and effective magnetization

The gyromagnetic ratio is an important dynamic magnetic property in ferromagnetic systems as it provides valuable information about spin orbit interaction in materials. As shown recently by Shaw et al.\cite{41}, the broadband experimental determination of gyromagnetic ratio can significantly reduce the experimental error margins in measuring the gyromagnetic ratio.
Figure 3.1: FMR spectrum for the Ni$_{80}$Fe$_{20}$(5nm)/Gd sample at 20 GHz for 1 for temperatures 250K, 125K and 65K.

The FMR spectrum of the Ni$_{80}$Fe$_{20}$ (5nm)/Gd sample at 20 GHz is shown figure 3.1 for temperatures 250K, 125K and 65K. As one can see the FMR signal broadens significantly and shifts towards lower fields as temperature decreases. As will be discussed in more detail in the following, the shift in the position of the FMR signal is mainly caused by the increase of the gyromagnetic ratio and the broadening is due to the increase of the effective damping parameter with decreasing of temperature. The FMR linewidth broadening at low temperatures significantly deteriorates the signal to noise ratio. Therefore, the lowest temperature for which FMR properties were determined are 65K for the 5nm film and 50K for the 10nm Ni$_{80}$Fe$_{20}$ film. Figure 3.2 shows the broadband frequency vs. resonance field experimental data measured in the temperature range of 65K-300K for the 5nm Ni$_{80}$Fe$_{20}$film. The gyromagnetic ratio

$$\gamma' = \frac{\gamma}{2\pi} = \frac{g\mu_B}{h}$$

and the effective magnetization $M_{\text{eff}}$ at each temperature were determined by
fitting the frequency vs. resonance field experimental data with in-plane Kittel formula.

\[ f = \gamma \sqrt{H_{\text{res}} \left(4\pi M_{\text{eff}} + H_{\text{res}}\right)} \]

![Graph showing broadband frequency versus resonance field experimental data, at different temperatures for the Ni_{80}Fe_{20} (5nm)/Gd sample. As an example, the fits to Kittel’s equation at 65K and 300K are shown (blue and red lines respectively).](image)

Figure 3.2: Broadband frequency versus resonance field experimental data, at different temperatures for the Ni_{80}Fe_{20} (5nm)/Gd sample. As an example, the fits to Kittel’s equation at 65K and 300K are shown (blue and red lines respectively).

The Kittel fit results indicate that the gyromagnetic ratio undergoes a significant increase with the decreasing temperature as shown in figure 3.3. This is similar to the increase of gyromagnetic ratio seen in rare earth containing transition metals near the compensation point [42, 43, 44], which will be explained in the following.
Figure 3.3: Temperature dependence of the gyromagnetic ratio for 5nm and 10nm Ni\textsubscript{80}Fe\textsubscript{20} films. Experimental data were fitted using Kittel’s equation to determine the gyromagnetic ratio at each temperature, see figure 3.2.

Assuming an antiferromagnetic coupling between Gd and Py (Ni\textsubscript{80}Fe\textsubscript{20}) layers at temperatures lower than the Curie temperature of Gadolinium, and using the mean-field theory, the effective g-factor can be expressed by [42]

\[
\gamma'_{\text{eff}} = \frac{m_{\text{Py}} - m_{\text{Gd}}}{m_{\text{Py}}/g_{\text{Py}} - m_{\text{Gd}}/g_{\text{Gd}}} 
\]

(3.1)
A qualitative understanding of the expected dependence of the effective g-factor in this model can be obtained assuming temperature independent g-factors for Ni$_{80}$Fe$_{20}$ and Gd based on literature ($g_{Py} = 2.1^8$ and $g_{Gd} = 2.0^9$). The temperature dependence of the effective g-factor is then a consequence of the strong temperature dependence of the Gd saturation magnetization around its Curie temperature. If one assumes that Ni$_{80}$Fe$_{20}$ and Gd are completely intermixed, then the formula (3.1) could be used to estimate the effective g-factor. Which would have a singularity for

$$\frac{M_{Py} \times t_{Py}}{g_{Py}} = \frac{M_{Gd} \times t_{Gd}}{g_{Gd}}$$

(3.2)

Where $t_{Py}$ and $t_{Gd}$ are the Ni$_{80}$Fe$_{20}$ and Gd layers thicknesses respectively. If one assumes $M_{Py}(T=0) = 900$ [emu/cm$^3$], $Tc$- Ni$_{80}$Fe$_{20} = 870[^oK][45]$ and $M_{Gd}(T=0) = 2017$ [emu/cm$^3$], $Tc$-Gd = $75[^oK][38]$(for a 3nm Gd layer) one obtains a singularity at $T\sim 40[^oK]$ for the 5nm thick Ni$_{80}$Fe$_{20}$ film. Given the simplicity of the model and the crude assumptions made the agreement with our experimental data is reasonable, see figure 3.4.
Figure 3.4: Effective g factor as a function of temperature; simulated data for 10nm sample (black line), simulated data for 5nm sample (green line), experimental data for 10nm sample (black diamonds), experimental data for 5nm sample (green open circles)
Figure 3.5 shows the behavior of experimentally determined $M_{\text{eff}}$ as a function of temperature for both 5 and 10nm samples. The difference between effective magnetization values of the two samples is most probably caused by the creation of an interface PMA, with the thinner sample having a stronger interface PMA (smaller positive value for $M_{\text{eff}}$). Both samples exhibit effective magnetization values which are smaller than the typical saturation magnetization in Ni$_{80}$Fe$_{20}$ thin films [46,47], which is also consistent with the existence of an interface PMA (see equation 1.5).

There is an approximate increase of ~150 emu/cm$^3$ in the value of $M_{\text{eff}}$ with decreasing temperature from 300 to 50K for both samples, which is similar to the previous experimental
reports on the temperature dependence of the saturation magnetization in Permalloy thin film [44,47]. A sudden decrease of effective magnetization occurs for both Ni$_{80}$Fe$_{20}$ (5nm)/Gd and Ni$_{80}$Fe$_{20}$ (10nm)/Gd thin films at the same temperature regime where the change in gyromagnetic ratio occurs, which most probably can be related to the phase transition and ferromagnetic ordering in Gd layer around the Curie temperature [48, 49].
III. Temperature dependence of the effective damping parameter

As explained in chapter 1, the frequency dependence of the FMR linewidth in the Gilbert model for magnetization relaxation is described by equation 1.9

\[ \Delta H = \Delta H_0 + \frac{2}{\sqrt{3}} \frac{\alpha_{\text{eff}}}{\gamma'} f \]

This equation was fit to the experimental data to determine the effective damping parameter \(\alpha_{\text{eff}}\) at each temperature, as shown in figure 3.6, for the Ni\(_{80}\)Fe\(_{20}\) (5nm)/Gd sample. Furthermore, the experimental results indicate that the inhomogeneous broadening \(\Delta H_0\) is negligible for these samples (see figure 3.6).

Figure 3.6: Broadband frequency dependence of FMR linewidth for the Ni\(_{80}\)Fe\(_{20}\) (5nm)/Gd sample. The lines indicate fits to the experimental data using equation 1.9 to determine effective damping parameter at each temperature.
Figure 3.7 shows the temperature dependence of the effective Gilbert damping parameter for both 5 and 10nm Ni$_{80}$Fe$_{20}$ thin films capped with 3nm Gd. The effective damping parameter significantly increases with the decrease of temperature. As will be discussed in the following, this observation can be explained by the change in the spin pumping contribution to the ferromagnetic relaxation as temperature is lowered towards the Curie temperature of Gd capping layer.

Spin pumping is an extrinsic relaxation mechanism in ferromagnetic thin films, given that the ferromagnetic layer has an interface with a normal metal or another ferromagnetic film.[50,51] As shown schematically in figure 3.8, the uniform precessing magnetization vector in the ferromagnetic layer can generate a spin polarized current that is pumped into the metallic...
capping layer, which can act as an extra channel for the magnetic damping. In other words, the precessing magnetization transfers angular momentum to the capping layer or the “spin sink” by emitting a spin current.

Figure 3.8: Schematic representation showing the contribution of spin pumping to the effective Gilbert damping parameter in NiFe/Gd system.

The theory developed by Tserkovnyak et al. [50] describes the effective damping parameter of a ferromagnetic film adjacent to a spin sink by

\[ \alpha' = \alpha_0 + \alpha' \]  \hspace{1cm} (3.3)

Where \( \alpha_0 \) is the intrinsic Gilbert damping parameter of the ferromagnetic layer and \( \alpha' \), which is the spin pumping contribution to the effective damping, is given by [50]

\[ \alpha' = \left[ I + g^{\uparrow \downarrow} \beta \right]^{-1} \frac{\hbar \gamma g^{\uparrow \downarrow}}{4\pi \mu} \]  \hspace{1cm} (3.4)

\[ \mu = M_s St \]  \hspace{1cm} (3.5)

where \( \mu \) is the film total magnetic moment; \( M_s \) is the saturation magnetization; \( S \) is the cross section area; \( t \) is the film thickness; \( \gamma \) is the gyromagnetic ratio; the \( g^{\uparrow \downarrow} \) is the interfacial spin mixing conductance; and \( \beta \) the back reflection factor given by [50]
\[
\beta = \frac{\tau_{SF} \delta_{SD}}{\hbar \tanh(L/\lambda_{SD})}
\] (3.6)

Here \( L \) is the thickness of the normal metal; \( \tau_{SF} \) is the spin flip time, or the average time between two collisions leading to a spin flip; \( \delta_{SD} \) is the effective energy-level spacing of the specific states that are involved in the spin-flip scattering; and \( \lambda_{SD} \) is the spin diffusion length of the spin sink layer. Here, the spin diffusion length is the maximum distance that a spin current can penetrate into a spin sink layer before losing its spin polarization [52]. However, in this case, since the Gd layer undergoes a paramagnetic-ferromagnetic phase transition at its Curie temperature, the spin diffusion length changes considerably. Based on the work by Bass et al., the spin diffusion length of a normal metal or a metallic ferromagnet is given by [52]

\[
\lambda_{SD} = \sqrt{\left(1 - \beta_F \right) \frac{\lambda_{SF} \lambda_r}{6}}
\] (3.7)

where \( \lambda_r \) is the mean free path and \( \lambda_{SF} \) is the spin flip length which is defined by \( \lambda_{SF} = v_r \tau_{SF} \).

It is important to distinguish between the spin flip length \( \lambda_{SF} \) and the spin diffusion length \( \lambda_{SD} \) as \( \lambda_{SD} \) is the distance an electron travels with diffusive motion during the spin flip time \( \tau_{SF} \).

The asymmetry coefficient \( \beta_F \) in equation (3.7) describes the asymmetric spin-flip scatterings for conducting electrons with majority and minority spin polarizations while diffusing in a ferromagnetic material. Thus for a normal metal as there is no difference in the scattering rates of spin up and spin down channels, \( \beta_F \) is zero, while for a ferromagnet it has a value between zero and one [52]. Therefore according to (3.4), (3.6) and (3.7) as Gd goes through the paramagnetic-ferromagnetic phase transition, its spin diffusion length decreases and consequently the contribution of spin pumping to the effective damping increases.
The maximum possible enhancement of the effective Gilbert damping parameter caused by the reduction of spin diffusion length in a spin sink layer can be estimated simply by assuming no back reflection

\[ \alpha'_{\text{max}} = \frac{\hbar \gamma \mu B}{4 \pi \mu} \]  

(3.8)

However, using the typical values reported in the literature for the spin mixing conductance [53,54], the experimental enhancement of the effective Gilbert damping parameter at low temperatures observed in our experiments is notably larger than the above estimate. For instance using the value of \( \frac{g^{\uparrow \downarrow}}{S} = 20 \text{nm}^{-2} \), that is almost three times larger than the reported value for Py/Cu/Pt system [54], results in \( \alpha'_{\text{max}} \approx 0.037 \) which is significantly smaller than the experimentally observed enhancement of damping parameter (see figure 3.7). Even consideration of the enhanced value of the g-factor caused by the increase of the gyromagnetic ratio at low temperatures cannot significantly reduce the gap between the estimation in (3.8) and the observed experimental enhancement in the effective damping parameter.

Recently Ohnuma et al.[55] showed that the spin mixing conductance can enhance significantly for a spin sink layer temperature nears its Curie temperature. In the Ohnuma et al. study, the spin mixing conductance is shown to be proportional to the momentum sum of the imaginary part of the dynamical transverse spin susceptibility in the spin sink layer

\[ g^{\uparrow \downarrow} \propto \sum_k \text{Im} \chi^R_k(\omega_{rf}) \]  

(3.9)

where \( \omega_{rf} \) is the microwave frequency and \( \chi^R_k \) is the dynamical transverse spin susceptibility.
It has been shown experimentally [56,57] that the magnetic susceptibility of Gd increases considerably near its Curie temperature and consequently the spin mixing conductance increases according to equation (3.9). This relative increase in the value of spin mixing conductance can be as large as ten times [55], which can explain the difference between experimentally observed enhancement of the effective damping parameter and the estimation given in equation (3.8).

In summary, it was shown that the effective Gilbert damping parameter and gyromagnetic ratio significantly increase in the vicinity of Curie temperature of Gd in Ni$_{80}$Fe$_{20}$/Gd bilayer. As discussed in this chapter, three mechanisms contribute to the temperature dependence of the effective Gilbert damping parameter: These mechanisms are the increase of gyromagnetic ratio with decreasing temperature, the decrease of spin diffusion length in Gd with decreasing temperature, and the increase of spin mixing conductance with decreasing temperature.
4. FERRITES

I. Introduction

Ferrites are insulating magnetic materials which have wide ranging applications in different branches of electronics and microwave engineering industry. Ferrite thin films are also promising candidates for the future generation of spintronic devices. Their low eddy current loss and low intrinsic Gilbert damping parameter together with a high dynamic permeability make them the material of choice for these applications. Ferrite materials are found in three crystallographic structures, which are spinel, garnet and hexagonal. In this chapter, dynamic magnetic properties and relaxation mechanisms in lithium ferrite (LFO) spinel single crystals, nickel ferrite spinel (NFO) thin films and Yttrium Iron Garnet (YIG) thin films are investigated using broadband FMR.

Ferrite materials with spinel structure are being explored for their applications in microwave devices such as isolators, phase shifters, gyrators and circulators [58,59,60]. Spinel ferrites are also promising candidates for the next generation of Monolithic Microwave Integrated Circuits (MMIC) devices. Understanding ferromagnetic relaxation mechanisms in spinel ferrites is very important for design and engineering of microwave or spintronic devices. For example growth of spinel ferrite thin films with low FMR linewidth is much desired for microwave applications as it indicates small intrinsic and extrinsic losses attributed to the magnetization relaxation. Therefore understanding the physical mechanisms contributing to the relaxation would enable engineers to control the microwave loss in such devices. The magnetization relaxation in spinel and hexagonal ferrite materials can be explained by different
physical mechanisms. Based on the growth conditions, defects and microstructures, stoichiometry and surface morphology the contribution of each mechanism can be made pronounced or negligible. Among all these relaxation mechanisms the two-magnon scattering [61,62], fast and slow relaxing impurities [63], valence exchange [64] and eddy current dissipation [65,66] are well verified in experimental and theoretical studies. While the intrinsic damping parameter in ferrite systems can be related to the magneto-elastic coupling between spin waves and phonons in a recent theoretical study[67], its experimental observation is very challenging as it is usually influenced by other mechanisms. Also inhomogeneous linewidth broadening or the zero frequency linewidth (see equation 1.9) is often so large that impedes the experimental determination of the intrinsic damping parameter in the spinel ferrite thin films.

Different Spinel ferrite systems are studied in sections I, II and III. In section I, the broadband determined gyromagnetic ratio and the effective Gilbert damping parameter of LiFe\(_5\)O\(_8\) single crystals are reported and the effect of B-site ordering on the FMR properties is verified. Section II mainly discusses the ferromagnetic relaxation in chemical vapor (CVD) deposited Ni\(_{0.8}\)Fe\(_{2.2}\)O\(_4\) thin films on (100) MgAl\(_2\)O\(_4\) substrates using detailed broadband temperature dependent and in-plane angle dependent FMR characterizations. FMR studies on NiFeO\(_4\) thin films grown on single crystal MgGa\(_2\)O\(_4\) substrates discussed in section III reveal excellent microwave properties as the FMR linewidth is significantly reduced. The observed value of the FMR linewidth is one order of magnitude lower than the value reported in section II, which itself is comparatively much lower than the values reported in the literature.
II. Lithium ferrite (LiFe$_5$O$_8$) single crystal

There are very few ferromagnetic resonance studies on the Lithium ferrite (LiFe$_5$O$_8$) in both thin film and single crystal formation. All the previous reported ferromagnetic resonance properties resulted from FMR measurements at one or two frequencies [68,69]. While, such experimental investigations suffer from large error margins in determination of dynamic magnetic properties such as gyromagnetic ratio and effective damping parameter [70]. Here broadband FMR measurements were carried out to determine dynamic magnetic properties of LiFe$_5$O$_8$ single crystals.

Standard Flux-melt grown technique was used to grow the lithium ferrite single crystals. The as-grown sample was further polished using air milling until it formed a 0.5 mm diameter sphere. This was done to minimize the irregularities in the shape of the sample, thus minimizing the inhomogeneous linewidth broadening. As the irregularities in the shape of the single crystals would cause the demagnetization field to be inhomogeneous across the surface of the sample leading to inhomogeneous FMR linewidth broadening. Furthermore, by polishing the sample to form a spherical shape, one can minimize the contribution of two magnon scattering mechanism to the FMR linewidth. As will be discussed in detail in the section II, the two magnon scattering process requires small scale inhomoginities in the effective magnetic field. Consequently using the air milling a minimized FMR linewidth can be obtained. The as-grown spherical lithium ferrite single crystal was further annealed at 600°C for 2 hours in presence of oxygen. The XRD analysis on the annealed samples showed additional superlattice peaks which were not present in the diffraction pattern for the as grown sample [71]. These extra peaks
indicate a cation ordering for the annealed samples, which was further confirmed by TEM and Raman spectroscopy characterizations [71]. The as grown and annealed spherical LFO single crystals are labeled as $S_1$ and $S_2$ in the following while discussing the FMR properties.

![FMR spectrum](image)

Figure 4.1: FMR spectrum for lithium ferrite single crystal ($S_2$ sample) with linewidth of 26 Oe, measured at 30 GHz. Red line shows the fit to the raw data.

Figure 4.1 shows an FMR spectra at 30 GHz for the $S_2$ sample, which indicates a clean signal with now distortion. The linewidth value which results from the fit (red line) is as low as 26 Oe. Considering the spherical shape of the samples, the demagnetization factors along any arbitrary Cartesian coordinate are equal, therefore the Kittel formula for a bulk sample (equation 1.4), is simplified as

$$f = \gamma' H_{res} \tag{4.1}$$

Therefore, the broadband frequency dependence of the resonance field shown in figure 4.2 for $S_1$ and $S_2$ samples, is fitted using 4.1 to determine the gyromagnetic ratio $\gamma'$. The experimentally
obtained values for \( \gamma' \) are \( 2.768 \pm 0.003 \text{ GHz/kOe} \) and \( 2.784 \pm 0.006 \text{ GHz/kOe} \) for \( S_1 \) the ordered \( S_2 \) samples respectively. While the reported error margins contain only statistical errors, systematic errors in measuring \( \gamma' \) are minimized by carrying out broadband measurements [41].

There is no previous report on the experimental or theoretical determination of gyromagnetic ratio for lithium ferrite. However, one can compare the values reported in this study to the experimentally obtained gyromagnetic ratio of \( 2.8 \text{ GHz/kOe} \) for barium ferrite single crystal [72] and \( \text{Ba}_2\text{Zn}_2\text{Zr}_x\text{Fe}_{12-x}\text{O}_{22} \) single crystal [73] and \( 2.84 \text{ GHz/kOe} \) for YIG films [74], which are not far from the \( 2.784 \text{ GHz/kOe} \) reported here for lithium single crystal.

![Figure 4.2](image)

Figure 4.2: \( H_{\text{res}} \) vs. frequency data for \( S_1 \) and \( S_2 \) LFO single crystals. The experimental data of both samples are too close to be distinguished from the figure.

The linewidth vs frequency experimental data for both \( S_1 \) and \( S_2 \) lithium ferrite single crystals are fitted using equation (1.9) to determine the effective Gilbert damping parameter \( \alpha \), see figure 4.3.
Figure 4.3: FMR linewidth vs. frequency data for both S1 and S2 samples. The Gilbert damping parameter is measured to be equal to 0.0021 after fitting the experimental data with equation (1.7). Red line shows the fit for S2.

The fit results show the same value of $\alpha = 0.0021 \pm 0.0001$ for both as deposited ($S_1$) and annealed ($S_2$) single crystals. Which indicates that the damping parameter does not change with the change in cation ordering.

To summarize, the broadband ferromagnetic resonance properties of lithium ferrite single crystals were measured and reported for the first time. It was shown that the dynamic magnetic properties such as gyromagnetic ratio and damping parameter do not vary with the additional ordering induced by annealing.
III.  Ni$_{0.8}$Fe$_{2.2}$O$_4$ thin films grown on MgAl$_2$O$_4$ by chemical vapor deposition

Growth of NiFe$_2$O$_4$ ferrite thin films with low FMR linewidth is much desired for microwave applications as a low linewidth can indicate a low intrinsic magnetization loss. However, in order to design low loss ferrite-based devices, one needs to deeply understand the physical characteristics of the ferromagnetic relaxation mechanisms in such materials. In this section, the results of a detailed broadband, angle and temperature dependent FMR study is presented to investigate the magnetization relaxation in nickel ferrite thin films.

Direct liquid injection chemical vapor deposition (DLI-CVD) technique was used to grow NFO thin films on (100) MgAl$_2$O$_4$ substrates at three different growth temperatures (500°C, 600°C and 700°C) and with different stoichiometries (Ni$_x$Fe$_{3-x}$O$_4$, where $x = 0.6$, 0.8 and 1) [75]. The best FMR results were obtained for a 700 nm thick Ni$_{0.8}$Fe$_{2.2}$O$_4$ film deposited at 600°C. Broader FMR linewidth or multiple resonance signals were observed for the films with different stoichiometry, thickness or deposition temperature.

Figure 4.4(a) shows a partial 2θ- ω XRD scans of the Ni$_x$Fe$_{3-x}$O$_4$ films deposited at 600 °C using CVD. Figure 4.4(b) shows the rocking curve of the 700 nm thick Ni$_{0.8}$Fe$_{2.2}$O$_4$ film deposited at 600°C with a narrow FWHM=0.43° that indicates the high structural quality of the film. Furthermore, the XRD Φ scan of the Ni$_{0.8}$Fe$_{2.2}$O$_4$ sample shown in figure 4.4(c) is consistent with the cubic spinel crystallographic structure that is expected for the NiFe$_2$O$_4$ thin films.
Figure 4.4: (a) 2θ-ω XRD scan around of Ni\textsubscript{x}Fe\textsubscript{3-x}O\textsubscript{4} films grown on (100)-oriented MgAl\textsubscript{2}O\textsubscript{4} substrate at 600°C of and different stoichiometries (x=0.6, 0.8 and 1.0 for) using CVD. (b) Rocking curve of Ni\textsubscript{0.8}Fe\textsubscript{2}O\textsubscript{4} (400) reflection with a narrow FWHM=0.43°, indicating high structural quality of the film, and (c) Φ scan of Ni\textsubscript{0.8}Fe\textsubscript{2}O (220) reflection confirming the epitaxial growth and the fourfold symmetry of spinel structure.
Figure 4.5: (a),(b) Plan view SEM images for the 700 nm NFO thin film deposited at 600 °C, using CVD, (b) has a higher resolution. (c) Plan view SEM image for the 700 nm PLD deposited NFO film at 600°C.

In order to further investigate the effect of deposition technique on the magnetic properties of the NFO thin film, Pulsed Laser Deposition (PLD) technique was used to fabricate a NFO thin film at the same deposition temperature and similar thickness[75]. The SEM images of the CVD deposited film are shown in figures 4.5(a) and 4.5(b), which indicate the formation of pyramid-like microstructures at the film surface with a fourfold symmetry. In contrast to this, the film surface of the PLD deposited NFO film is very smooth and no significant anisotropy of the surface roughness is observed.
Figure 4.6: (a) Raw FMR spectra at 10 GHz for the NiFe$_2$O$_4$ (solid red), Ni$_{0.8}$Fe$_{2.2}$O$_4$ (solid black) and Ni$_{0.6}$Fe$_{2.4}$O$_4$ (solid blue) thin films, the fitted values of linewidth are 750 Oe, 448 Oe and 554 Oe, respectively. The fits to the experimental data are shown in dotted lines. (b) Raw FMR spectra at 10 GHz for the Ni$_{0.8}$Fe$_{2.2}$O$_4$ thin films deposited at 500°C (solid blue), 600°C (solid black) and 700°C (solid red), the fitted values of linewidth are 860 Oe, 448 Oe and 902 Oe. The fits to the experimental data are shown in dotted lines.

The room temperature in-plane FMR spectra at 10 GHz for the NiFe$_2$O$_4$ (Fe:Ni=2), Ni$_{0.8}$Fe$_{2.2}$O$_4$ (Fe:Ni=2.75) and Ni$_{0.6}$Fe$_{2.4}$O$_4$ (Fe:Ni=4) thin films deposited at 600°C, and the Ni$_{0.8}$Fe$_{2.2}$O$_4$ thin films deposited at 500 °C, 600 °C and 700 °C are shown in Figs. 4.6 (a,b). The lowest in-plane FMR linewidths at all frequencies were observed for the 600 °C deposited Ni$_{0.8}$Fe$_{2.2}$O$_4$ sample, equaling 458 Oe which is less than the so far lowest reported values of 550 Oe for nearly stoichiometric NiFe$_2$O$_4$ films in the in-plane geometry [76]. It is important to mention that in all of these films the FMR linewidths are largely influenced by extrinsic contributions such as inhomogeneities and various defects present in the film, therefore it is difficult to draw any systematic correlation between the composition (e.g., Fe$_2$+/Fe$_3$+ ratio) and intrinsic linewidth or damping in the films.
The out-of-plane FMR spectrum for all the samples exhibit multiple-line resonances. Figure 4.7 shows a three resonance peak spectrum observed for the 600 °C deposited Ni$_{0.8}$Fe$_{2.2}$O$_4$ sample at 10 GHz with linewidth values varying between 98Oe to 140 Oe. The separation between the three modes along the field axis decreases with decreasing frequency, therefore the formation of the multiple peaks in the out-of-plane geometry can likely be attributed to sample inhomogeneities that result in local variations of the internal field. The experimental data that corresponds to the signal with the largest absorption amplitude (in Fig. 4.7) and of linewidth 98 Oe, is considered as the out-of-plane FMR mode.

![Figure 4.7: Raw FMR data for the 600 °C deposited Ni$_{0.8}$Fe$_{2.2}$O$_4$ sample at 10 Ghz. Three resonance peaks are observed. Linewidth values vary between 98Oe to 140 Oe](image)

Broadband FMR measurements (1-40 GHz) were done at different temperatures (300-10 °K), for both in-plane and out-of-plane geometries. The frequency dependence of the in-plane resonance field was fitted at to determine the effective magnetization, $M_{eff}$ at each temperature.
The temperature dependence of the effective magnetization \(4\pi M_{\text{eff}} = 4\pi M_s - \frac{2K_u}{M_s}\) and the temperature dependence of the saturation magnetization \(M_s\) obtained from VSM measurements shown in figure 4.7 indicate that the thin film has no significant out-of-plane anisotropy \(K_u\), as the two values agree with each other within their error margins.

Figure 4.8: \(M_s\) (red squares) and \(M_{\text{eff}}\) (blue triangles) vs. temperature, obtained from VSM and broadband FMR measurements. The exemplary error margins are shown for data points at 50K.

The broadband temperature dependent response of the FMR linewidth in both in-plane and out-of-plane configurations are shown in figures 4.9 (a) and 4.9 (b). Note that the overall value of the linewidth is smaller in the out-of-plane than the in-plane configuration, which as will be discussed in the following is consistent with the two magnon scattering (TMS) relaxation mechanism.
Figure 4.9: The temperature dependent broadband FMR linewidth of the CVD deposited NFO film in the (a) in-plane and (b) out-of-plane geometries

In-plane angle dependent measurements were carried out at room temperature to further study the behavior of the resonance field and linewidth as a function of the in-plane angle. As shown in figure 4.10(a) a fourfold symmetry is observed for resonance field $H_{res}$. The observed symmetry in the resonance field is expected based on the spinel structure of Ni$_{0.8}$Fe$_{2.2}$O$_4$, as the magnetic anisotropy should follow the crystallographic symmetry. Furthermore, an interesting observation is the fourfold symmetry seen in FMR linewidth, figure 4.10(b).
Figure 4.10: In-plane angle dependent measurement results at 40 GHz for the CVD deposited NFO film (a) resonance field (b) linewidth. Magnetic easy axis corresponds to the [011] crystallographic direction.

The in-plane angle dependent measurements were repeated for the PLD deposited NFO thin film. While the in-plane fourfold anisotropy in the $H_{\text{res}}$ is again observed for the PLD deposited film, no clear symmetry is seen for the linewidth, figures 4.11(a) and 4.11(b). These observations imply that the anisotropy observed in the resonance field has an intrinsic nature as it is maintained in the Ni$_{0.8}$Fe$_{2.2}$O$_4$ films regardless of the deposition technique. Because the intrinsic magnetic anisotropy in magnetic materials stems from the anisotropy in the spin-orbit coupling interaction due to the crystallographic symmetry [77, 78, 79]. However, the fourfold symmetry of the linewidth observed in the CVD deposited film should be of extrinsic origin as it vanishes when PLD growth is used.
The broadband temperature and in-plane angle dependence of the FMR linewidth in the CVD deposited Ni$_{0.8}$Fe$_{2.2}$O$_4$ film can be explained using the two magnon scattering (TMS) relaxation mechanism. Two magnon scattering is an extrinsic relaxation mechanism which tunes the magnetization relaxation in ferromagnetic thin films. In a two magnon scattering process the incoming uniform precession mode magnon (with $k = 0$) is scattered to another magnon with $k \neq 0$ [80, 81] in the proximity of a scattering inhomogeneity. The inhomogeneities or scattering centers leading to the two magnon scattering relaxation can be in different forms such as voids, pores, misfit dislocations, surface roughness or a combination of these defects. This way as graphically shown in figure 4.12(a), the FMR mode is relaxed to the magnon with $k \neq 0$ in the scattering process. The initial and final magnons should be degenerate in energy (frequency).
while their linear momentums (k vectors) are not equal, as the change in linear momentum is transferred to the scattering defect.

The dispersion relation of spin waves or so called magnons for both in-plane and out of plane FMR geometries is shown in Fig. 4.12(b) [80, 81]. In the out-of-plane geometry when the static external field and magnetization vector are normal to the plane of the film, no degenerate spin wave state is available. Therefore the two magnon scattering relaxation mechanisms is absent which typically leads to narrower linewidths in the out of plane geometry. This is in agreement with our results, since the broadband out-of-plane linewidth is narrower than the broadband in plane linewidth at all temperatures, figures 4.9(a) and 4.9(b).

Figure 4.12: (a) Schematic presentation of k=0 and k≠0 spin wave modes (b) Spin wave dispersion relation for the in-plane and out-of-plane thin film geometries, note the FMR frequency.

According to the theory of two magnon scattering developed by Slater Loudon and Kittel (SLK) [82], the contribution of TMS to the FMR linewidth should scale with the saturation magnetization $M_S$. This is consistent with the experimental observations in this study, as the in-plane linewidth increases for all frequencies with decreasing temperature (see figure 4.9 (a)), following the experimental increase of $M_s$ and $M_{eff}$ with decreasing temperature, see figure 4.8.
Furthermore, much less variation is observed for the out-of-plane linewidth with the change of temperature which is also consistent with the theory, figure 4.9 (b).

Moreover, the anisotropic behavior of the in-plane FMR linewidth observed for the DLI-CVD deposited thin film (see figure 4.9(b)) can be explained by the anisotropic pyramid-like microstructures, figures 4.5(a) and 4.5(b). Here the fourfold anisotropy of the surface microstructures causes the TMS relaxation and thus the linewidth to have the same symmetry. This is further confirmed, as the observation of smooth surface for the PLD deposited NiFe$_2$O$_4$ thin film is accompanied by an isotropic in-plane linewidth behavior, figures 4.5(c) and 4.11 (b).

It is noteworthy to mention that there is always an intrinsic contribution causing anisotropic symmetry in the in-plane angle dependent FMR Linewidth due to field dragging effect [83,84]. The field dragging linewidth broadening is caused by the small misalignment between the external magnetic field and magnetization vectors that vanishes along easy and hard axes and is maximum of an intermediate angle. One can quantitatively describe the field dragging effect on the FMR linewidth from the following equation [83,84]

$$\Delta H_{pp} = \frac{2}{\sqrt{3}} \frac{\alpha f}{\gamma \cos \beta}$$

(1)

Where $\Delta H_{pp}$ is the peak-to-peak FMR linewidth, $\alpha$ is the damping parameter, $\gamma$ is the gyromagnetic ratio, $f$ is the frequency and $\beta$ is the angle between the magnetization vector and the external magnetic field. Having the experimental values for gyromagnetic ratio, saturation magnetization and magnetic anisotropy, the change in $\beta$ as the direction of external magnetic field is changed can be simulated using energy minimization.[8] As shown in figure 4.13, the $1/\cos \beta$ term remains very close to unity as the external magnetic field direction is changed. Which implies that field dragging effect leads to a very weak contribution to the FMR linewidth,
which furthermore has an eightfold symmetry.

Figure 4.13: Stimulated data showing the asymmetry of $1/\cos \beta$ as the direction of external magnetic field changes, the value of $1/\cos \beta$ stays very close to unity causing a negligible eight fold anisotropy in the FMR linewidth. Due to its small amplitude this contribution is experimentally not observable.
IV. NiFe$_2$O$_4$ thin films grown on MgGa$_2$O$_4$ by pulsed laser deposition

In section III Ni$_{0.8}$Fe$_{2.2}$O$_4$ thin films were grown on MgAl$_2$O$_4$ substrate leading to a 3% lattice mismatch between film and substrate [75]. Also, other substrates such as MgO [85] and SrTiO$_3$ [86] used in previous studies on NFO thin film suffer from relatively large lattice mismatches. Here the single crystal MgGa$_2$O$_4$ substrate with a similar spinel structure [87] to that of NiFe$_2$O$_4$ is used. This results in a small lattice mismatch of 0.6% for the MgGa$_2$O$_4$/NiFe$_2$O$_4$ structures, which were grown by Pulsed Laser Deposition (PLD). This small lattice mismatch, as will be discussed in the following, results in a low linewidth of the NiFe$_2$O$_4$ thin films. NiFe$_2$O$_4$ films were deposited on (100) MgGa$_2$O$_4$ substrates at 700 °C using pulsed laser deposition. A single resonance profile free of deterioration was observed for a 444nm thick sample, figure 4.15(a). Thinner samples exhibited multiple resonance peaks which prevented precise determination of FMR resonance field and linewidth. In order to further understand the effect of film-substrate lattice mismatch on the microstructural and FMR properties, experimental results are compared to the case of 700nm Ni$_{0.8}$Fe$_{2.2}$O$_4$ thin film deposited on MgAl$_2$O$_4$ substrate (discussed in previous section).

The bulk lattice parameters of NiFe$_2$O$_4$, MgGa$_2$O$_4$ and MgAl$_2$O$_4$ spinel structures are 0.8333 nm [88], 0.828 nm [87] and 0.809 nm [76] respectively. The $2\theta - \omega$ XRD scan on the MgGa$_2$O$_4$/NiFe$_2$O$_4$ thin film is shown in figure 4.14(a), which indicates the much smaller separation between substrate and films (400) peaks as compared to the MgAl$_2$O$_4$/Ni$_{0.8}$Fe$_{2.2}$O$_4$, see figure 4.4(a). Moreover, the XRD $\Phi$ scan shown in figure 4.14(a) indicates that the in-plane lattice parameter of NiFe$_2$O$_4$ is exactly matching the MgGa$_2$O$_4$ substrate (0.828 nm) that causes the measured out-of-plane lattice parameter (0.840 nm) to be slightly larger than the bulk value.
However, in the case of MgAl$_2$O$_4$/Ni$_{0.8}$Fe$_{2.2}$O$_4$ thin film the in-plane lattice parameter is smaller than the bulk value and therefore the film thin film is relaxed.

Furthermore, the $K\alpha_1$ and $K\alpha_2$ peaks are observed well separated for the (400) MgGa$_2$O$_4$/NiFe$_2$O$_4$ film (figure 4.14(a), whereas for the MgAl$_2$O$_4$/Ni$_{0.8}$Fe$_{2.2}$O$_4$ film (figure 4.4(a)) a broad single (400) peak was found. This verifies that the growth quality is comparatively better on the MgGa$_2$O$_4$ substrate than the MgAl$_2$O$_4$ substrate.

![Figure 4.14](image-url)

Figure 4.14: (a) 2θ-ω XRD scan around of NiFe$_2$O$_4$ film grown on (100)-oriented MgGa$_2$O$_4$ substrate at 700 °C using PLD deposition. Distinct $K\alpha_1$ and $K\alpha_2$ (400) NiFe$_2$O$_4$ peaks are observed. (b) Φ scan of NiFe$_2$O$_4$ (440) and MgGa$_2$O$_4$ (440) peaks showing cube-on-cube geometry of the substrate and the film.

The high compressive strain for the MgGa$_2$O$_4$/NiFe$_2$O$_4$ is further confirmed by the TEM diffraction patterns, figure 4.15. Two distinguishable diffraction spots are observed for the MgAl$_2$O$_4$/Ni$_{0.8}$Fe$_{2.2}$O$_4$ thin film (labeled with arrow) compared to the MgGa$_2$O$_4$/NiFe$_2$O$_4$ case where the substrate and film diffraction spots are merged together.
Figure 4.15: TEM diffraction patterns (a) MgAl$_2$O$_4$ / Ni$_{0.8}$Fe$_{2.2}$O$_4$ sample, the substrate and thin film spots are distinguishable at high “g” vector points (marked with arrow) which indicates strain relaxation (b) MgGa$_2$O$_4$ / NiFe$_2$O$_4$ sample, the substrate and thin film diffraction spots are merged together indicating a highly strained film.
The VSM Magnetization reversal loop for the MgGa$_2$O$_4$/NFO thin film is shown in figure 4.16. The average value of saturation magnetization determined from the in-plane and out-of-plane loops is equal to 224 emu/cm$^3$. Furthermore, the saturation field $H_s$ for the hard-axis (out-of-plane) reversal is 15200 Oe.

Figure 4.16: M-H loop for the MgGa$_2$O$_4$/NiFe$_2$O$_4$ (444nm) thin film measured by VSM. The measured value of $M_s$ is 218 and 230 emu/cm$^3$ in the inplane and out-of-plane configurations, respectively. The saturation field $H_s$ that is measured in the out-of-plane geometry indicates the magnitude of the field needed to pull the magnetization out of the film plane.

Figure 4.17 shows exemplary in-plane FMR spectrums for both samples at 40GHz. By fitting the FMR spectrum of the MgGa$_2$O$_4$/NiF$_2$O$_4$ thin film (figure 4.17(a)) a low linewidth
value of 65Oe is found, which is an order of magnitude smaller than the value for CVD deposited MgAl2O4 / Ni0.8F2.2O4 sample (discussed in the previous section) at the same frequency (see figure 4.7(a)).

In order to determine the $M_{\text{eff}}$ and the gyromagnetic ratio $\gamma'$, multiple frequency FMR measurements were carried out along the hard and easy axes (figure 4.18) since the resonance field exhibits a fourfold in-plane symmetry, the inset of figure 4.18. The frequency vs. $H_{\text{res}}$ data along the easy ([011]) and hard axes were then fitted using the following combined fit [89]

$$f = \gamma' \sqrt{(H_{\text{res-hard}} - H_4)(H_{\text{res-hard}} + \frac{H_4}{2} + 4\pi M_{\text{eff}})}$$  \hspace{1cm} (4.2)

$$f = \gamma' \sqrt{(H_{\text{res-easy}} + H_4)(H_{\text{res-easy}} + H_4 + 4\pi M_{\text{eff}})}$$

Figure 4.17: FMR spectra for the (a) MgGa2O4 / NF2O4 (444nm) thin film deposited at 700C using PLD. The small signal on the low field side is a Perpendicular standing spin wave mode (b) MgAl2O4 / Ni0.8Fe2.2O4 (700nm) thin film deposited at 600C using CVD. The linewidth of the MgGa2O4 / NF2O4 (444nm) sample is one order of magnitude smaller.
in which the $M_{\text{eff}}$ and $\gamma'$, are the shared fit parameters and $H_4$ is the fourfold anisotropy field and $M_{\text{eff}}$ is given by equation (1.8)

$$4\pi M_{\text{eff}} = 4\pi M_s - \frac{2K_u}{M_s}$$

where $K_u$ is the out of plane uniaxial anisotropy constant.

Figure 4.18: Resonance frequency as a function of resonance field along the easy ($\Phi = 55^\circ$) and hard ($\Phi = 5^\circ$) axes for the MgGa$_2$O$_4$/NFO thin film. The inset shows in-plane angle dependent $H_{\text{res}}$ data at 30 GHz.

Thus, the experimentally determined values for the $M_{\text{eff}}$, $H_4$ and $\gamma'$ and $H_4$ are $1225\pm0.003$ emu/cm$^3$, $280\pm6$ Oe and $2.97\pm0.01$ GHz/kOe respectively.

The considerable difference between the experimentally determined values of $M_{\text{eff}}$ equaling 1225 emu/cm$^3$ and $M_s$ equaling 218-230 emu/cm$^3$ measured by FMR and VSM
techniques respectively, implies the existence of a large negative perpendicular anisotropy constant $K_u$ given the equation (1.8). The negative sign corresponds to an easy-plane anisotropy. This is further verified by comparing the values of out of plane saturation field $H_s = 15200$ Oe resulting from VSM measurements with the $4\pi M_{\text{eff}} = 15394$ Oe determined by FMR, as their values are reasonably close. One need In order to saturate the film in the out of plane geometry external magnetic field needs to overcome both the demagnetization $4\pi M_s$ and the negative out of plane anisotropy $H_u = \frac{2K_u}{M_s}$ fields, therefore the $H_s$ and $4\pi M_{\text{eff}}$ should be the same. Having experimentally measured values of $M_{\text{eff}}$ and $M_s$, one can calculate the perpendicular uniaxial anisotropy constant $K_u$ to be around $1409000$ erg/cm$^3$ and the perpendicular anisotropy field $H_u$ to be $12580$ Oe which is one order of magnitude larger than the intrinsic fourfold crystalline anisotropy.

The origin of the large negative out of plane anisotropy can be attributed to the significant compressive strain generated at the film substrate interface as verified by the TEM and XRD characterizations. Similar compressive strain-induced negative out of plane anisotropy has also been reported previously for Pr$_{0.67}$Sr$_{0.33}$MnO$_3$ thin films [90] and La$_{0.67}$Ca$_{0.33}$MnO$_3$ thin films grown on SrTiO$_3$ [91].

The ultra-low linewidth FMR signal observed for the MgGa$_2$O$_4$/NiF$_2$O$_4$ thin films, shows a close to linear frequency dependence as shown in the figure 4.19. Therefore, one can determine the damping parameter of NFO thin film by fitting the $\Delta H$ vs. $f$ data using equation (1.9) which results in the value of $\alpha = 0.0025 \pm 0.0001$ for the Gilbert damping parameter.
Figure 4.19: Broadband measured FMR linewidth. The experimental data are fitted (red line) with the equation (1.7) to determine the damping parameter.
5. **Co$_{90}$Fe$_{10}$/Ru/Co$_{90}$Fe$_{10}$ EXCHANGE COUPLED TRILAYER**

Interlayer exchange-coupled (IEC) structures in which magnetic layers are separated with a non-magnetic spacer have been extensively studied in the last decades [92,93,94,95,96]. In these systems the magnetic moments of the ferromagnetic layers are coupled through exchange interaction mediated by the free electrons of the spacer layer [95]. As the thickness of the spacer layer changes the coupling oscillates between ferromagnetic and anti-ferromagnetic [95]. The interlayer exchange coupling was first discovered in 1986 for Fe/Cr/Fe [97], Gd/Y/Gd [98] and Dy/Y/Dy[99] structures. A couple of years later, the discovery of Giant Magneto Resistance (GMR) effect in the interlayer exchanged-coupled Fe/Cr/Fe systems gave birth to the field of spintronics [100]. Interlayer exchange-coupled layers with a strong anti-ferromagnetic coupling in combination with an exchange-biased layer have also been extensively used in magnetic tunnel junctions (MTJ’s). Phenomena like spin dependent transport [101, 102], dependence of coupling strength on the spacer layer thickness and magnetic anisotropies [96,103] in IEC structures have been studied in numerous theoretical and experimental works. However, so far ferromagnetic relaxation in interlayer exchange-coupled systems has been much less studied.

In this chapter a comprehensive experimental investigation of the magnetization relaxation in the Co$_{90}$Fe$_{10}$(5 nm)/Ru(x)/Co$_{90}$Fe$_{10}$ (8nm) (0.8nm <x< 2.6 nm) trilayers is presented using broadband FMR characterization. Thus far, there are also very few experimental studies on the temperature dependence of the interlayer exchange coupling strength [104,105]. Here a detailed temperature dependent broadband FMR study on Ta(3nm)/Ru(2nm)/Co$_{90}$Fe$_{10}$ (5 nm)/Ru(x)/Co$_{90}$Fe$_{10}$ (8nm) is also provided and the experimental results are compared with the theoretical
models. The samples were fabricated using magnetron sputtering by collaborators at Western Digital (WD), Freemont.

I. FMR characterization of interlayer exchange-coupled trilayer systems

Ferromagnetic resonance is a powerful tool to study dynamic magnetic properties in interlayer exchanged-coupled systems. FMR has been used extensively to study exchange coupling and magnetic anisotropy in IEC structures in various experimental studies [103,104]. Solving the LLG equation for a trilayer structure leads to two different resonance modes referred as acoustic and optical modes [93,106]. Equation (5.1) shows the corresponding resonance frequencies of the two modes for the case of two identical ferromagnetic layers [93,106]

\[
 f = \gamma' \sqrt{(H_{res,ac} + H_{u-ip})(H_{res,ac} + H_{u,ip} + 4\pi M_{eff})} 
\]

\[
 f = \gamma' \sqrt{(H_{res,op} + H_{u,ip} - 2H_{ex})(H_{res,op} + H_{u,ip} - 2H_{ex} + 4\pi M_{eff})} 
\]

In this simplified configuration the quasi-static field is applied in-plane and \( H_{res} \geq 2H_{ex} - H_{u} \), where \( H_{ex} \) is the interlayer exchange coupling field and \( H_{u,ip} \) is the uniaxial in-plane anisotropy field for both layers. A positive value for \( H_{ex} \) shows a ferromagnetic (F) coupling, while negative values of \( H_{ex} \) indicate an antiferromagnetic coupling, moreover the optical mode is shifted by the interlayer exchange coupling. Note that the \( H_{res,ac} \) is identical to the resonance field of the individual films.
As shown in figure 5.1, for an interlayer exchange-coupled structure, the optical mode corresponds to the out-of-phase and the acoustic mode corresponds to the in-phase precessional motions of the magnetization vectors in the two layers [93,104,106]. When the two ferromagnetic layers are identical, the acoustic mode has the same energy as the FMR mode of a single layer, since the magnetic moments of the two layers stay parallel during the precessional motion [93,104,106]. While, the optical mode will have a higher (lower) energy with respect to the acoustic mode for a FM (AF) coupled system due to the exchange interaction [93,106,106].
As discussed in chapter 1 for the FMR measurements carried out in this experimental study at
each frequency the external field is swept to meet the FMR condition. Therefore, one would
observe the optical mode at lower (higher) field side of the acoustic mode for the FM (AF)
coupled trilayers. In the case of two identical ferromagnetic layers it is not possible to excite the
optical mode with a homogenous microwave field as the net transverse magnetic moment would
be zero for the optical mode [95,96]. The relative positions of the optical and acoustic modes
along the field axis can be seen in the experimental FMR spectrum of the Co$_{90}$Fe$_{10}$ (5
nm)/Ru(0.8nm)/Co$_{90}$Fe$_{10}$ (8nm) (figure 5.2(a)) and Co$_{90}$Fe$_{10}$ (5 nm)/Ru(2nm)/Co$_{90}$Fe$_{10}$ (8nm)
(figure 5.2(c)) which show anti-ferromagnetic couplings and Co$_{90}$Fe$_{10}$ (5nm)/Ru(1.4 nm)/
Co$_{90}$Fe$_{10}$ (8 nm) (figure 5.2(b)) and Co$_{90}$Fe$_{10}$ (5nm)/Ru(2.6 nm)/Co$_{90}$Fe$_{10}$ (8 nm) (figure 5.2(d))
samples which show ferromagnetic couplings.
Figure 5.2: (a),(c) Raw FMR spectra for the Co$_{90}$Fe$_{10}$(5 nm)/Ru(0.8 nm)/Co$_{90}$Fe$_{10}$(8 nm) and Co$_{90}$Fe$_{10}$(5 nm)/Ru(2 nm)/Co$_{90}$Fe$_{10}$(8 nm) AF coupled samples. (b),(d) Raw FMR spectra for the Co$_{90}$Fe$_{10}$(5 nm)/Ru(1.4 nm)/Co$_{90}$Fe$_{10}$(8 nm) and Co$_{90}$Fe$_{10}$(5 nm)/Ru(2.6 nm)/Co$_{90}$Fe$_{10}$(8 nm) FM coupled samples. Note that the optical mode appears on the low field side of the acoustic mode for the ferromagnetic coupling and on the high field side for the antiferromagnetic coupling.

The LLG equation can be solved numerically to determine the resonance frequencies of the optical and acoustic modes. Figure 5.3 shows the simulated frequency dependence of the $H_{res-acoustic} - H_{res-optical}$ for a symmetric Co$_{90}$Fe$_{10}$(5 nm)/Ru/Co$_{90}$Fe$_{10}$(5 nm) trilayer for both AF and FM coupling configurations. The same values of $\gamma$ and $M_{eff}$ were used for both layers, moreover $H_{ex}$ was fixed to an arbitrary value. As expected from equation (5.1) the results show a flat line.
with no change with the frequency. However, as will be discussed in the next section for asymmetric trilayers both the experimental and numerically simulated values of $H_{\text{res-\ acoustic}} - H_{\text{res-\ optical}}$ will depend on the microwave frequency.

Figure 5.3: The simulated frequency dependent $| H_{\text{res-\ acoustic}} - H_{\text{res-\ optical}} |$ data resulted from equation 5.2 for a symmetric Co$_{90}$Fe$_{10}$(5nm)/Ru/Co$_{90}$Fe$_{10}$ (5nm) trilayer with an assumption of $M_{\text{eff}}$ of 1400emu/cm$^3$ and $H_{\text{ex}}$ of 250 Oe.
II. Measuring coupling strength in the asymmetrical Co$_{90}$Fe$_{10}$ (5 nm)/Ru(x)/Co$_{90}$Fe$_{10}$ (8 nm) trailer system

As explained in section I, for two identical ferromagnetic layers the separation between the optical and acoustic modes along the magnetic field axis is proportional to the exchange coupling strength and the optical mode appears on the lower (higher) field side for a FM (AF) coupling. Therefore a positive (negative) value for the $H_{res-acoustic}-H_{res-optical}$ corresponds to a FM (AF) coupling. In this study the separation between two resonance modes of the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(x)/Co$_{90}$Fe$_{10}$ (8 nm) system is investigated over a broad frequency range. For all FMR measurements the quasi static field is applied in-plane.

In order to determine the exchange coupling strength using FMR measurements in asymmetrical trilayers where the thickness of ferromagnetic layers are not identical, a more complete dispersion relation than equation (5.1) should be considered. In this case, according to Zhang et al. the resonance frequencies of the optical and acoustic modes can be derived from the following equation [104]

$$\left(\frac{f}{\gamma'}\right)^4 - b\left(\frac{f}{\gamma'}\right)^2 + c = 0$$

(5.2)

Where b and c are multi-term constants that depend on the angles of magnetization in the two layers, thicknesses of the FM layers, saturation magnetization and the second derivatives of the energy with respect to the magnetization angles at equilibrium [104]. In general, in an asymmetrical configuration the two FM layers will have different contributions from the interface magnetic anisotropy in their effective internal magnetic field, can lead to different effective magnetizations for the two ferromagnetic layers.
Equation (5.2) was solved numerically in the 0-15000 Oe interval to derive the corresponding frequencies of the optical and acoustic modes at each field point. The simulations were carried out assuming a gyromagnetic ratio of 3.03 GHz/kOe. Moreover, the exchange coupling strength (H_{ex}) was fixed to different values for both AF and FM coupling configurations. The values of M_{eff} and \gamma were used in the simulations were chosen based on the experimental values of 1420 emu/cm\textsuperscript{3} and 3.02 GHz/kOe for a Co\textsubscript{90}Fe\textsubscript{10} (5nm)/Ru(0 nm)/Co\textsubscript{90}Fe\textsubscript{10} (8nm) single layer. Figures 5.4 show the results of numerical simulations for interlayer coupling values of -1125(a), 680 Oe (b), -230 Oe (c) and 95 Oe (d), which are chosen in analogy to the experimental results shown in figure 5.2. The negative sign corresponds to an antiferromagnetic coupling. In the numerical results shown in figure 5.4, a small interfacial perpendicular magnetic anisotropy was assumed, which leads to the different effective magnetizations of the 5nm (1300 emu/cm\textsuperscript{3}) and 8nm Co\textsubscript{90}Fe\textsubscript{10} (1400 emu/cm\textsuperscript{3}) layers according to equation (1.6). The deviations observed at low frequencies for the AF coupled samples (figures 5.4(a) and (c)) indicates that the samples are not saturated as the external magnetic field is not large enough to overcome the interlayer antiferromagnetic coupling. Note that the non-saturation state is moved to the higher fields for the sample with larger coupling strength. Figure 5.4 shows the simulated Kittel plots. The deviations observed at low frequencies for the AF coupled samples (figures 5.4(a) and (c)) indicates that the samples are not saturated as the external magnetic field is not large enough to overcome the interlayer antiferromagnetic coupling. Note that the non-saturation state is moved to the higher fields for the sample with larger coupling strength.
Figure 5.4: The simulated Kittel plots resulted from equation 5.2 with an assumption of $M_{\text{eff}}$ of 1330 emu/cm$^3$ and 1400 emu/cm$^3$ for the 5nm and 8nm Co$_{90}$Fe$_{10}$ layers. (a) Assumed Hex = -1125 Oe (b) Assumed Hex = 550 Oe (c) Assumed Hex = -250 Oe (d) Assumed Hex = 90 Oe. Negative sign corresponds to an antiferromagnetic coupling.

Figure 5.5 shows the value of the separation between the optical and acoustic modes determined from the simulated Kittel graphs for arbitrary coupling strengths, where the low frequency data of the AF-coupled samples corresponding to the non-saturation regions are omitted. Figure 5.5(a) shows the results for the case where the effective magnetizations of the 5nm and 8nm Co$_{90}$Fe$_{10}$ layers are assumed to be 1300 emu/cm$^3$ and 1400 emu/cm$^3$, respectively. While, figure 5.5(b)
shows the numerical results assuming both 5 and 10nm Co<sub>90</sub>Fe<sub>10</sub> layers have identical effective magnetizations of 1400 emu/cm<sup>3</sup>. The numerical results indicate that layers with different effective magnetizations result in a frequency dependent mode separation which is more noticeable for the weakly coupled samples. Moreover, this frequency dependence vanishes when identical values for effective magnetizations of the two Co<sub>90</sub>Fe<sub>10</sub> layers are assumed.

The experimental data were analyzed in the light of the obtained numerical results. Figure 5.6 shows the experimental Kittel graphs and figure 5.7 shows the exemplary frequency dependent experimental <i>H</i><sub>res-acoustic</sub>-<i>H</i><sub>res-optical</sub> data for the asymmetrical Co<sub>90</sub>Fe<sub>10</sub>/Ru/Co<sub>90</sub>Fe<sub>10</sub> samples with Ru thicknesses of 0.8, 1.4, 2 and 2.6 nm, both showing similar frequency dependencies as the simulated data. The data corresponding to the strongly coupled samples are fitted with a constant value over the whole frequency range to determine 2<i>H</i><sub>ex</sub> (a,b). On the other hand only the two lowest frequency data points are used in the fit to estimate the 2<i>H</i><sub>ex</sub> value for the weakly coupled
samples (c, d).

Figure 5.6: (a), (c) Experimental Kittel Plots for the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(0.8 nm)/Co$_{90}$Fe$_{10}$ (8 nm) and Co$_{90}$Fe$_{10}$ (5 nm)/Ru(2 nm)/Co$_{90}$Fe$_{10}$ (8 nm) AF coupled samples. (b), (d) Experimental Kittel Plots for the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(1.4 nm)/Co$_{90}$Fe$_{10}$ (8 nm) and Co$_{90}$Fe$_{10}$ (5 nm)/Ru(2.6 nm)/Co$_{90}$Fe$_{10}$ (8 nm) FM coupled samples. The data in magenta color correspond to the optical mode and black corresponds to the acoustic mode.
Figure 5.7: (a),(c) Experimental frequency dependent $H_{res\text{-acoustic}}$-$H_{res\text{-optical}}$ data for the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(0.8 nm)/Co$_{90}$Fe$_{10}$ (8 nm) and Co$_{90}$Fe$_{10}$ (5 nm)/Ru(2 nm)/Co$_{90}$Fe$_{10}$ (8 nm) AF coupled samples. (b),(d) Experimental frequency dependent $H_{res\text{-acoustic}}$-$H_{res\text{-optical}}$ data for the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(1.4 nm)/Co$_{90}$Fe$_{10}$ (8 nm) and Co$_{90}$Fe$_{10}$ (5 nm)/Ru(2.6 nm)/Co$_{90}$Fe$_{10}$ (8 nm) FM coupled samples. The data are fitted with a constant value (dashed line) over the whole frequency range to determine $2H_{ex}$ for the large coupling strengths (a,c) while only the two lowest frequency data points are used in the fit (dashed line) for the small coupling cases (b,d).

The same approach is used to determine the experimental value of $H_{ex}$ for the rest of the asymmetrical Co$_{90}$Fe$_{10}$ (5 nm)/Ru(x)/Co$_{90}$Fe$_{10}$ (8 nm) samples. Figure 5.8 shows the experimentally determined exchange coupling strengths as a function of Ru spacer layer thickness. Note that the strongest antiferromagnetic coupling is found at the thickness of 0.8 nm and the first transition from antiferromagnetic coupling to ferromagnetic coupling happens between 1 nm and 1.2 nm and the second transition from AF to FM coupling taking place between 2 nm and 2.4 nm. The strongest ferromagnetic coupling is observed at the thickness of 1.2 nm with
the second anti-ferromagnetic and ferromagnetic peaks occurring at 1.8nm and 2nm, and the coupling cross over from ferromagnetic to antiferromagnetic between 1.4nm and 1.8nm. Based on the results found in the numerical study which indicates a systematic uncertainty in the FMR determination of the $H_{ex}$ in asymmetrical trilayers an error margin of 5% is assumed for the experimentally determined data.

Figure 5.8: Experimentally determined $H_{ex}$ of the Co$_{90}$Fe$_{10}$/Ru/Co$_{90}$Fe$_{10}$ interlayer exchange coupled system as a function of Ru thickness. The strongest AF and FM couplings are observed at the Ru thicknesses of 0.8nm and 1.2nm. The coupling oscillates between AF to FM at 0.8nm-1nm, 2nm - 2.4nm and from FM to AF at 1.4nm-1.8nm. The data shown on the dashed line correspond to the samples where the optical and acoustic signals were merged together due to a very weak coupling, therefore the coupling strength was assumed to be equal to zero.
III. Ferromagnetic relaxation in the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(x)/Co$_{90}$Fe$_{10}$ (8nm) trilayer

Figure 5.4 shows the exemplary frequency dependent FMR linewidth data for the samples with Ru spacer layer thicknesses of 0.8nm, 1.4nm, 2nm and 2.6nm, respectively.

Figure 5.4: Shows the exemplary frequency dependent FMR linewidth data for the samples with Ru spacer layer thicknesses of 0.8nm, 1.4nm, 2nm and 2.6nm, respectively.

Figure 5.9: (a), (c) FMR linewidth as a function of frequency for the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(0.8nm)/Co$_{90}$Fe$_{10}$ (8nm) and Co$_{90}$Fe$_{10}$ (5 nm)/Ru(2nm)/Co$_{90}$Fe$_{10}$ (8nm) AF coupled samples, respectively. Fitted values of the damping parameter are $\alpha = 0.0070$ and $\alpha = 0.0073$ for the acoustic modes, and $\alpha = 0.0127$ and $\alpha = 0.0123$ for the optical modes respectively. (b), (d) FMR linewidth as a function of frequency for the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(1.4nm)/Co$_{90}$Fe$_{10}$ (8nm) and Co$_{90}$Fe$_{10}$ (5 nm)/Ru(2.6nm)/Co$_{90}$Fe$_{10}$ (8nm) FM coupled samples, respectively. Fitted values of the damping parameter are $\alpha = 0.0069$ and $\alpha = 0.0066$ for the acoustic modes and $\alpha = 0.0114$ and $\alpha = 0.0086$ for the optical modes respectively. The high frequency optical mode data are not included in the fit to determine the damping parameter for the Co$_{90}$Fe$_{10}$/Ru(2nm)/Co$_{90}$Fe$_{10}$ sample (7.9(c)) due to the low quality of the FMR spectrum fit (see the inset of 5.9 (c)).
The results in figure 5.9 show that the value of FMR linewidth for the optical mode is significantly larger than its acoustic mode counterpart at all frequencies. The frequency dependent data are fitted to determine the effective damping parameter for both modes which results in a considerable enhancement of the damping parameter for the optical mode.

In some of the previous experimental reports the enhancement of the linewidth of the optical mode compared to the acoustic mode has been attributed to inhomogeneous linewidth broadening caused by spatial variations in the value of $H_{ex}$ over the spacer layer [96, 107, 108]. However, the inhomogeneous broadening mostly contributes to the linewidth offset at zero frequency while the experimental results reported here show a significant increase in the slope the linewidth as a function of frequency for the optical mode linewidth and thus its effective damping parameter. This considerable enhancement observed in the damping parameter of the optical mode can be explained by the recently predicted mutual spin pumping effect [109, 110, 111]. Similar to the discussion in chapter 3 for the spin pumping effect in a single layer, the precessing magnetization vectors in each of the ferromagnetic layers generates a spin polarized current [51]

$$I_s = \frac{\hbar}{4\pi} g^\uparrow (1 - \beta g^\uparrow) \left( \vec{m} \times \frac{d\vec{m}}{dt} \right)$$  \hspace{1cm} (5.1)

which is pumped in to the Ru spacer layer and can be totally or in part absorbed by the other ferromagnetic layer depending on the spacer layer thickness and its spin diffusion length.
Figure 5.10: A Schematic which shows the mutual spin pumping effect in an interlayer exchange coupled trilayer. (a) Optical mode, the polarization of the pumped current from FM2 and FM1 are 180° out-of-phase which results in an enhanced loss of angular momentum at both FM layers. (b) Acoustic mode, the polarization of the pumped current from FM2 and FM1 are parallel, therefore the two spin currents cancel each other leading to almost no effect on the damping parameter.

As qualitatively shown in figure 5.10 when the magnetization vectors precess in phase (acoustic mode) the polarizations of the pumped spin currents are parallel and therefore each layer loses as much spin angular momentum as it gains, whereas in the case of out-of-phase precession (optical mode) the two spin currents have a 180° phase difference which leads to an enhanced loss of angular momentum through the mutual spin pumping [109]. Therefore the effective damping of the optical mode will be larger than for the acoustic mode. However, the mutual spin pumping effect will be significantly suppressed in case the thickness of spacer layer is comparable to the spin diffusion length as the spacer layer will act as a spin sink [111].
Figure 5.11 shows the experimentally determined effective damping parameters of both acoustic and optical modes as a function of Ru interlayer thickness. The data point at 10nm corresponds to a Co$_{90}$Fe$_{10}$(5nm)/Ru(10 nm)/Co$_{90}$Fe$_{10}$ (8nm) sample, where only a single resonance peak was observed in the FMR spectrum indicating a coupling close to zero.

With increasing Ru thickness the effective damping parameter of the optical mode tends to decrease in both FM and AF coupled samples, while the damping parameter of the acoustic stays almost constant.
IV. Temperature dependent exchange coupling strength in the Co$_{90}$Fe$_{10}$(5 nm)/Ru(x)/Co$_{90}$Fe$_{10}$(8nm) trilayer

Figure 5.13 shows the broadband temperature dependence of the separation between the two modes for the Co$_{90}$Fe$_{10}$(5 nm)/Ru(0.8nm)/Co$_{90}$Fe$_{10}$(8nm) (5.12(a)) and Co$_{90}$Fe$_{10}$(5 nm)/Ru(1.4nm)/Co$_{90}$Fe$_{10}$(8nm) (5.12(b)) samples. The frequency dependence of the field separation between the two modes remains very weak at all temperatures for both samples, therefore the experimental data are averaged over the available frequency range to determine the experimental value $2H_{ex}$ at each temperature. As seen in figure 5.12 the coupling strength increases with decreasing the temperature for both the ferromagnetically (Co$_{90}$Fe$_{10}$(5 nm)/Ru(1.4nm)/Co$_{90}$Fe$_{10}$(8nm)) and antiferromagnetically (Co$_{90}$Fe$_{10}$(5 nm)/Ru(0.8nm)/Co$_{90}$Fe$_{10}$(8nm)) coupled trilayers.

Figure 5.12: Broadband temperature dependence of the separation between optical and acoustic modes for (a) Co$_{90}$Fe$_{10}$(5 nm)/Ru(0.8nm)/Co$_{90}$Fe$_{10}$(8nm) and (b) Co$_{90}$Fe$_{10}$(5 nm)/Ru(1.4nm)/Co$_{90}$Fe$_{10}$(8nm) samples. The experimental data are fitted to a constant at each temperature to determine the value of $2H_{ex}$. 
Two different theoretical models have been proposed in the literature to explain the
temperature dependence of the interlayer-exchange coupling strength [112,113,114]. According
to the theoretical study by Edwards et al. [112] and Bruno et al. [113] one contribution to the
temperature dependence of the coupling strength is caused by the changes in the Fermi surface of
the spacer layer with the change of temperature as the free electrons of the spacer layer obey the
Fermi-Dirac statistics. Based on this model, which will be called Edwards-Bruno model in this
study, the coupling strength has the following dependence on the temperature

\[ H_{ex} = H_{ex,0} \frac{T}{T_0} \left( \sinh\left( \frac{T}{T_0} \right) \right)^{-1} \]  

(5.2)

where \( H_{ex,0} \) is the interlayer exchange-coupling strength at the absolute zero temperature and
the characteristic temperature \( T_0 \) is given by

\[ T_0 = \frac{\hbar v_f}{2\pi k_B d} \]  

(5.3)

where \( v_f \) is the Fermi velocity and \( d \) is the thickness of spacer layer. This model was later
modified in the experimental study by Zhang et al. [104] on the Co/Ru/Co trilayer structures as
they realized that a fit to the experimentally determined interlayer exchange coupling strength
using equation (5.2) does not follow the experimental data at temperatures higher than 100K.
Therefore, the equation (5.2) was modified as follows

\[ H_{ex} = (H_{ex,0} - H_{ex,inf}) \frac{T}{T_0} \left( \sinh\left( \frac{T}{T_0} \right) \right)^{-1} + H_{ex,inf} \]  

(5.4)

where \( H_{ex,inf} \) was introduced as the coupling strength at \( T = \infty \). However, this modification is a
phenomenological model and the physics behind the inconsistency between the theoretical
prediction (equation 5.2) and experimental data at high temperatures is not understood.
Figure 5.14 also shows the experimentally determined $H_{\text{ex}}$ values as a function of temperature for $\text{Co}_{90}\text{Fe}_{10} (5 \text{ nm})/\text{Ru}(0.8\text{nm})/\text{Co}_{90}\text{Fe}_{10} (8\text{nm})$ (5.13(a)) and $\text{Co}_{90}\text{Fe}_{10} (5 \text{ nm})/\text{Ru}(1.4\text{nm})/\text{Co}_{90}\text{Fe}_{10} (8\text{nm})$ (5.13(b)) samples. The solid and dashed lines show the best fits to the experimental data using the modified Edwards-Bruno and Edwards-Bruno models respectively. The solid and dashed red lines show the fits to the experimental data using the modified Edwards-Bruno and Edwards-Bruno models respectively. Blue line shows the fit to the experimental data using the $T^{3/2}$ model.

As expected due to the additional fitting parameter, the modified Edwards-Bruno model shows a better agreement with the experimental data. However as shown in table 5.1, the fits to the experimental data suggest very low values for the Fermi velocity of the Ru spacer layer, which are 1-2 orders of magnitude smaller than the typical values found for the nobel metals using the free electron gas model [115]. According to Zhang et al. this difference is caused by the fact that the effective $k_F$ used in equation (5.3) should be smaller than the $k_F$ of the electron gas based on the theory developed by Bruno and Edwards [112,113]. Note that there are large systematic error margins in the experimental value of $H_{\text{ex,in}}$ due to the lack of experimental data at higher temperatures than
300K. In contrast to the theory, which predicts the $T_0$ to increase with decreasing the thickness of spacer layer (equation 5.3), the experimentally determined $T_0$ is larger for the sample with thicker Ru spacer layer (see table 5.1). This can be due to the lower fit quality in the case of $\text{Co}_{90}\text{Fe}_{10}$ (5 nm)/Ru(0.8nm)/$\text{Co}_{90}\text{Fe}_{10}$ (8nm) sample leading to an underestimation of $T_0$.

According to the theoretical study by Almeida et al. the temperature dependence of the interlayer-coupling strength is also influenced by the thermal fluctuation of the spin waves near the ferromagnetic-space layer interface \[114,116\]. This model predicts a $T^{3/2}$ temperature dependence for the coupling strength

$$H_{ex} = H_{ex,0} \left(1 - \left(\frac{T}{T_C}\right)^{3/2}\right) \quad (5.5)$$

In an experimental work by Linder et al. it was shown that this model has a good agreement with the experimental data for Ni/Cu/Ni and Ni/Cu/Co trilayers [105].

The experimentally determined exchange coupling strength as a function of temperature for the $\text{Co}_{90}\text{Fe}_{10}$ (5 nm)/Ru(0.8nm)/$\text{Co}_{90}\text{Fe}_{10}$ (8nm) and $\text{Co}_{90}\text{Fe}_{10}$ (5 nm)/Ru(1.4nm)/$\text{Co}_{90}\text{Fe}_{10}$ (8nm) samples also fitted using the $T^{3/2}$ model (see Figure 5.14) and the fit results are shown in table 5.1 (c).

As can be seen in figure 5.13 and table 5.1 the agreement between the $T^{3/2}$ model and the experimental data is considerably weaker for the $\text{Co}_{90}\text{Fe}_{10}$ (5 nm)/Ru(0.8nm)/$\text{Co}_{90}\text{Fe}_{10}$ (8nm) as compared to the $\text{Co}_{90}\text{Fe}_{10}$ (5 nm)/Ru(1.4nm)/$\text{Co}_{90}\text{Fe}_{10}$ (8nm) trilayer structure. The fit quality of the $T^{3/2}$ model for the $\text{Co}_{90}\text{Fe}_{10}$ (5 nm)/Ru(0.8nm)/$\text{Co}_{90}\text{Fe}_{10}$ (8nm) sample is also significantly weaker compared to the modified Edwards-Bruno model.
Table 5.1: The results of fitting the experimental temperature dependent coupling strength with (a) Edwards-Bruno (b) Modified Edwards-Bruno and (c) the $T^{3/2}$ models. Note that the values are underestimated for the $\text{Co}_{90}\text{Fe}_{10}/\text{Ru}(0.8\text{nm})/\text{Co}_{90}\text{Fe}_{10}$ sample in Bruno models which can be due to lower fit quality or smaller $R^2$. Note that the values of $T_0$ found in this study are in an acceptable agreement for Edwards-Bruno model and relatively higher for modified Edwards-Bruno model as compared to the previous experimental results reported by Zhang et al. for similar Ru thicknesses in Co/Ru/Co structures [104].
6. CONCLUSION

To conclude the basics of ferromagnetic resonance was discussed and it was shown that the broad band ferromagnetic resonance technique is a very useful toll to deeply investigate magnetization dynamics and ferromagnetic relaxations in thin films. The comprehensive angle and temperature dependent FMR measurements can shed light into the on the complexities of spin dynamics in ferromagnetic systems.

It was shown that the PMA strongly depends on annealing conditions in Cr(40 nm)/Co2FeAl(t)/MgO(2.3 nm)/Ru(5 nm) structures. Furthermore, the damping parameter was found To decrease with increasing B2 ordering parameter for MgO/Co2FeAl(50 nm)/Al(5 nm).

The temperature dependent broadband FMR measurements showed that the gyromagnetic ratio and effective damping parameter noticeably change as the temperature is lowered towards the Curie temperature of Gd in NiFe/Gd heterostructures. The enhancement of the effective damping as the Gd goes through phase transition was found to be caused by the spin pumping effect.

A comprehensive FMR study on Nickel ferrite thin films showed that the ferromagnetic relaxation is strongly influenced by two magnon scattering for (100) MgAl2O4/ Ni0.8F2.2O4 samples deposited by CVD, while using (100) MgGa2O4 substrates resulted in much
better growth conditions and considerably lower FMR linewidth in PLD deposited MgGa$_2$O$_4$ / 
NiO.8F$_{2.2}$O$_4$ structures.

Using both numerical and experimental investigations, it was shown that there is a 
frequency dependence in the FMR mode separation of asymmetric 
Co$_{90}$Fe$_{10}$ (5 nm)/Ru(x)/Co$_{90}$Fe$_{10}$ (8nm) trilayers. Furthermore, it was verified that to reduce the 
large possible uncertainties in the experimental determination of coupling strength in asymmetric 
IEC systems using FMR technique it is very important to carry out broadband measurements. 
Also it was shown that both modified Edwards-Bruno and T$^{3/2}$ models have a reasonable 
consistency with the temperature dependence of the experimentally determined coupling strength 
for the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(0.8nm)/Co$_{90}$Fe$_{10}$ (8nm) sample, while the agreement between the 
models and the experimental data is weaker for the Co$_{90}$Fe$_{10}$ (5 nm)/Ru(1.4nm)/Co$_{90}$Fe$_{10}$ (8nm) 
sample. In general the thermal properties of the spacer layer electronic structure (Edwards-Bruno 
model) and the thermal fluctuations of spin waves in FM layers (T$^{3/2}$ model) can both contribute 
to the temperature dependence of coupling strength, however there is still controversy regarding 
which of the mentioned models is dominating. Further experimental evidence and theoretical 
study are needed to elucidate this phenomenon.
REFERENCES


[40] B. Khodadadi, J. B. Mohammadi, M. Jones, C. Mewes, T. Mewes, T. Eggers, C. Miller, M. Manno, C. Leighton (not published)


064420 (2005).


[70] C. K. A. Mewes and T. Mewes, “Relaxation in magnetic materials for spintronics,” in Handbook of Nanomagnetism-Applications and Tools (Pan


[75] N. Pachauri, B. Khodadadi, A. V. Singh, J. B. Mohammadi, R. L. Martens, P. R. LeClair, C. Mewes, T. Mewes, A. Gupta, “A comprehensive study of ferromagnetic resonance and structural properties of Ni_{0.8}Fe_{2.2}O_{4} (nickel ferrite) film grown by chemical vapor deposition” (in preparation)


