Magnetization Dynamics in Ferromagnetic Thin Films

Evaluation of Different Contributions to Damping in Co$_2$FeAl and FeCo Film Structures

SERKAN AKANSEL
Static and dynamic magnetic properties of Co2FeAl and Fe65Co35 alloys have been investigated. Co2FeAl films were deposited at different temperatures and the deposition parameters were optimized with respect to structural and magnetic properties. As a result, a film with B2 crystalline phase was obtained without any post-annealing process. A lowest magnetic damping parameter of was obtained for the film deposited at 573K. This obtained low value is comparable to the lowest values reported in research literature. After optimizing the deposition parameters of this alloy, different seed layers and capping layers were added adjacent to the Co2FeAl layer and the effect of these layers on the magnetic relaxation was investigated. In addition to adding nonmagnetic layers to Co2FeAl, the dependence of the magnetic damping parameter with respect to the thickness of Co2FeAl was investigated by depositing films with different thicknesses. A temperature dependent study of the magnetic damping parameter was also performed and the measured damping parameters were compared with theoretically calculated intrinsic Gilbert damping parameters. Different extrinsic contributions to the magnetic damping, such as two magnon scattering, spin pumping, eddy-current damping and radiative damping, were identified and subtracted from the experimentally obtained damping parameter. Hence, it was possible to obtain the intrinsic damping parameter, that is called the Gilbert damping parameter.

In the second part of the thesis, Fe65Co35 alloys were investigated in terms of static and dynamic magnetic properties. Fe65Co35 films were deposited without and with different seed layers in order to first understand the effect of the seed layer on static magnetic properties of the films, such as the coercivity of the films. Then the films with seed layers yielding the lowest coercivity were investigated in terms of dynamic magnetic properties. Fe65Co35 films with different rhenium dopant concentrations and with ruthenium as the seed and capping layer were also investigated. The purpose of this study was to increase the damping parameter of the films and an increase of about ~230% was obtained by adding the dopant to the structure. This study was performed at different temperatures and after subtraction of the extrinsic contributions to the damping, the experimental values were compared with theoretically calculated values of the Gilbert damping parameter. During the thesis work, magnetic looper and superconducting quantum interference device magnetometers set-ups were used for static magnetic measurements and cavity, broadband in-plane and broadband out-of-plane ferromagnetic resonance set-ups were used for dynamic measurements.

Keywords: spintronics, Gilbert damping parameter, magnetization dynamics, ferromagnetic resonance, Heusler alloys, magnetic thin films

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For everything in this world, for civilization, for life, for success, the truest guide is science. To seek a guide other than science is a mark of heedlessness, ignorance, and aberration.

Mustafa Kemal Atatürk

Aileme
List of Papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.


III Akansel, S., Kumar, A., Behera, N., Husain, S., Brucas, R., Chaudhary, S., Svedlindh, P. Thickness dependent enhancement of damping in Co$_2$FeAl/$\beta$-Ta thin films (Submitted)


V Akansel, S., Venugopal, V. A., Kumar, A., Gupta, R., Brucas, R., George, S., Neagu, A., Tai, C-W., Gubbins, M. A., Andersson, G., Svedlindh, P., Effect of seed layers on dynamic and static magnetic properties of Fe$_{65}$Co$_{35}$ thin films (Submitted)


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Papers not included in the thesis


6. Concluding Remarks.................................................................64
6.1. Summary .................................................................................64
6.2. Outlook...................................................................................65

7. Sammanfattning .......................................................................66

Acknowledgments.........................................................................69

Bibliography .................................................................................71
## Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>CPW</td>
<td>Coplanar waveguide</td>
</tr>
<tr>
<td>FFT</td>
<td>Fast fourier transform</td>
</tr>
<tr>
<td>FMR</td>
<td>Ferromagnetic resonance</td>
</tr>
<tr>
<td>( g )</td>
<td>Landé g-factor</td>
</tr>
<tr>
<td>GIXRD</td>
<td>Grazing incidence XRD</td>
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<tr>
<td>GMR</td>
<td>Giant magnetoresistance</td>
</tr>
<tr>
<td>( H_c )</td>
<td>Coercivity</td>
</tr>
<tr>
<td>( H_u )</td>
<td>Uniaxial anisotropy field</td>
</tr>
<tr>
<td>( H_{\text{eff}} )</td>
<td>Effective magnetic field</td>
</tr>
<tr>
<td>( H_r )</td>
<td>Resonance field</td>
</tr>
<tr>
<td>HRTEM</td>
<td>High resolution TEM</td>
</tr>
<tr>
<td>LL</td>
<td>Landau Lifshitz</td>
</tr>
<tr>
<td>LLG</td>
<td>Landau Lifshitz Gilbert</td>
</tr>
<tr>
<td>( M_{\text{eff}} )</td>
<td>Effective magnetization</td>
</tr>
<tr>
<td>MRAM</td>
<td>Magnetoresistive random access memory</td>
</tr>
<tr>
<td>( M_s )</td>
<td>Saturation magnetization</td>
</tr>
<tr>
<td>MOKE</td>
<td>Magneto-optic Kerr effect</td>
</tr>
<tr>
<td>MPMS</td>
<td>Magnetic properties measurement system</td>
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<tr>
<td>MTJ</td>
<td>Magnetic tunnel junction</td>
</tr>
<tr>
<td>PMA</td>
<td>Perpendicular magnetic anisotropy</td>
</tr>
<tr>
<td>PPMS</td>
<td>Physical properties measurement system</td>
</tr>
<tr>
<td>RBS</td>
<td>Rutherford backscattering</td>
</tr>
<tr>
<td>RT</td>
<td>Room temperature</td>
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<tr>
<td>SQUID</td>
<td>Superconducting quantum interference device</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>XRR</td>
<td>X-ray reflectivity</td>
</tr>
<tr>
<td>( \alpha_{\text{eddy}} )</td>
<td>Eddy-current contribution to damping</td>
</tr>
<tr>
<td>( \alpha_{\text{eff}}, \alpha_{\text{total}} )</td>
<td>Total damping parameter including both intrinsic and extrinsic contributions</td>
</tr>
<tr>
<td>( \alpha_G, \alpha_{\text{int}}, \alpha_{\text{red}} )</td>
<td>Intrinsic Gilbert damping parameter which remains after extrinsic contributions are subtracted from total damping</td>
</tr>
<tr>
<td>( \alpha_{\text{rad}} )</td>
<td>Radiative contribution to damping</td>
</tr>
</tbody>
</table>
\( \alpha_{sp} \)  Spin pumping contribution to damping
\( \Delta H \)  Full width half maximum of the FMR absorption linewidth
\( g_{\uparrow \downarrow}^{\dagger} \)  Effective spin mixing conductance
1. Introduction

The word magnetism comes from a city called Manisa, which is today on the west coast of Turkey. The magnetic material magnetite was first found in this city by the ancient Greeks and this material was named after the Greek tribe “the Magnetes” who lived in the city Magnesia (Manisa), which belonged to the Greeks at that time. The source of the inspiration for one of today’s the most important scientific fields “magnetism” is magnetite. Most of the technological devices that we use today in our daily life are based on magnetism and magnetic materials. The word dynamics comes from the Greek word “dynamikos” which means “powerful”. However, in physics it refers to properties of a system which change over time. The research field magnetization dynamics is based on the change of magnetic properties of materials over a period of time and this change is mostly dependent on the applied external magnetic field. Magnetization deals mostly with the dynamic properties of a quantum property of the electrons called “spin”, which refers to the field known as spin dynamics. Spintronics is a relatively new aspect of technology which developed from spin dynamics.

Conventional electronics technology was based on the charge of electrons. However, spintronics integrates the spin properties of the electrons into electronics technology. Spin can basically be explained as the quantum mechanical angular momentum which is hard to understand through classical physical properties. Instead of using the charge of electrons in daily applications, replacing charge by spin and understanding and tailoring dynamic properties of the electron spins will lead to production of novel devices that work faster and in a more energy-efficient way.

The discovery of giant magnetoresistance is regarded as the starting point of the research field of spintronics. Two different research groups led by Fert and Grunberg independently discovered giant magnetoresistance (GMR)\(^1\_2\) and later in 2007 this discovery was awarded the Nobel Prize in physics. The word spintronics was introduced for the first time in 1996, to refer to the longer term SPIN Transfer electrONICS.\(^3\) It was expected that devices produced on basis of spintronics would be non-volatile in terms of data storage, processing the data at an increased speed and being more energy efficient compared with conventional data storage devices. In addition, they were expected to have increased integration densities.\(^3\) Today some of the aims have been achieved and spintronics structures are utilized commercially, especially in magnetic recording heads.
One of the most common structures known in spintronics is spin valves. The working principle of these structures is dependent on GMR. This is a sandwich structure where a ferromagnetic layer is coupled to an antiferromagnetic layer via exchange bias. This layer is called a pinned layer since a higher magnetic field is needed to rotate its magnetization, compared with the so-called free layer, which requires very small fields to get its magnetic moment orientation changed. There is a nonmagnetic conductor layer between these free and pinned layers. When the magnetic moments of the layer are aligned in a parallel way, the structure has a lower resistance. When the magnetization of these separate layers is anti-parallel aligned, they show a higher resistance. These high and low resistance states are considered as 1 and 0 digital states in digital data recording. When the nonferromagnetic conductor layer is replaced by an insulating layer, larger magnetoresistance is obtained at room temperature and these structures are called magnetic tunnel junctions (MTJ) and are the basis of the magnetoresistive random access memories (MRAMs). Beside used in MRAMs, GMR and MTJ are utilized for applications such as magnetic field sensors, read heads of hard drives and galvanic isolators as well.

This thesis was written as a comprehensive summary of six papers. In the work done for these six papers, two kinds of alloys, Fe$_2$CoAl and Fe$_{65}$Co$_{35}$ were investigated in terms of their magnetization dynamics.

Paper I is about optimization of deposition parameters of Co$_2$FeAl in order to obtain B$_2$ phase, with low damping parameter, and as a result a damping parameter that is comparable with the lowest values reported so far was obtained.

Paper II and III are about adding different seed and capping layers and investigating the effect of these layers on the magnetic relaxation properties of Co$_2$FeAl films. In addition to adjacent layers, the effect of thickness was also investigated.

Paper IV is about running temperature dependent FMR measurements on Co$_2$FeAl films, extracting different extrinsic contributions to magnetic relaxation and comparing the experimentally obtained intrinsic damping parameter with theoretically calculated values.

Paper V and VI are about investigating the effects of different seed layers and Re doping on both static and magnetic properties of Fe$_{65}$Co$_{35}$ films. Different extrinsic contributions were extracted on the magnetic damping parameter of Re doped films, and temperature dependent damping parameters were obtained and compared with theoretical calculations.
2. Experimental Techniques

2.1. Structural Characterization

2.1.1. X-ray diffraction

X-ray diffraction (XRD) is a widely used technique for the structural characterization of thin films. This powerful technique reveals information about the grain size, crystalline structure, lattice parameters of the thin films and the phase of the material. It is also possible to investigate the stress and strain effect on the lattice by using this technique. It is a non-destructive technique, so when the x-ray interacts with the sample it does not change the chemical or physical properties of the sample. It basically works on the principle that when an electromagnetic wave interacts with a part of the crystal structure of the same scale as its wavelength, destructive and constructive interference occur and the constructive interference creates a spectrum which is unique for each different material. A phase difference occurs when x-rays from different atomic planes interact. Observing constructive interference or not depends on the distance between the planes. The necessary condition for observing a constructive interference pattern is defined by Bragg’s law, given as

\[ 2d\sin\theta = n\lambda, \]

where \( n \) is an integer, \( d \) is the distance between the crystallographic planes, \( \lambda \) is the wavelength of the x-rays, \( \theta \) is the angle between the crystal plane and the incident and reflected x-rays. In this study all measurements were done with a CuK\( \alpha \) x-ray source, where \( \lambda \) is 1.54Å. A schematic of the XRD set up is given in figure 1.
For this thesis work, the grazing incidence XRD (GIXRD) technique was used. For this technique the x-ray source is kept fixed during the whole scan, and the x-rays hit to the film with a very small angle. By using this technique a larger area on the film plane is illuminated; hence more intense counts are detected by the detector even though the investigated sample is a thin film.

In this thesis work GIXRD was used for the phase analysis of the films, extracting the crystallite size and lattice parameter. To extract the crystallite size $\tau$, Scherrer’s\(^5\) formula was used which can be given as

$$\tau = \frac{k\lambda}{\beta \cos \theta}. \quad (2)$$

Here $\lambda$ is the wavelength of the x-ray, $\theta$ is the Bragg angle, $\beta$ is the full width half maximum of the diffraction peak. $k$ is a dimensionless shape factor which is kept constant at 0.9 for all calculations. Although this value of the shape factor is defined for spherically shaped crystallite structures with cubic unit cells,\(^6\) traditionally it is used even for differently shaped structures. This value can vary slightly due to different shape and unit cell structure of the material, however since Scherrer’s equation is not a very precise way of determining crystallite size it will not affect the result drastically. Besides that, the unit cell structure of the films investigated in this thesis work is cubic so we assumed taking the shape factor as 0.9 is a good enough estimation.

In addition to crystallite size, the lattice parameter is also extracted by using the XRD data. To extract the lattice parameter the $d$ value is first calculated from equation 1. The crystalline structures of the films investigated in this thesis are cubic. For cubic crystals the relation between $d$ and the lattice parameter $a$ is given as\(^5\)

$$d_{hkl} = \frac{a}{\sqrt{h^2+k^2+l^2}}, \quad (3)$$
where \( h, k \) and \( l \) are the miller indices. Depending on equation 3, lattice parameters were extracted for the different films.

2.1.2. X-ray reflectivity

X-ray reflectivity (XRR) is a very powerful and widely used technique to obtain the thickness of different layers in thin film stacks as well as their density and roughness, both at the surface and the interfaces. The same geometry as in GIXRD technique is used for XRR. However for XRR the reflected and refracted X-rays are detected and evaluated instead of the diffracted X-rays. Using this technique the intensities of the reflected and refracted X-rays are measured with respect to the incident angle of the x-ray beam on the sample surface at very small angles.

Equations explaining the reflectivity properties of visible light, such as Snell’s law and the Fresnel equations, are also valid for reflected X-rays. When they are solved for the defined refraction index of an X-ray, which is slightly less than one, refractions and reflections of X-rays can be analysed.\(^7\)

Reflected x-ray beams from the surface of a thin film structure, from the interfaces between the layers in the structure and from the substrate interact, and interference occurs between these x-rays and due to these interferences the intensity of the detected X-ray beam oscillates. These oscillations depend on the thickness of the different layers in the film stack and since they were observed by Kiessig for the first time they are known as Kiessig fringes.\(^8\) The period of the oscillations depends on the layer thicknesses, and by analyzing the oscillations the layer thicknesses can be extracted. The determination of the thickness of the layers of a film stack is independent of the material type.\(^7,9\) Depending on the roughness of the surface and the interfaces in the film stack, the decrease rate of the oscillatory x-ray intensity varies. An increase in the roughness of the surface also increases the decaying rate of the detected reflected x-rays. By analyzing the decay rate of the x-ray intensity, the surface and interface roughness values can be calculated.\(^9\) When it comes to extracting the material density of different layers, necessary information is obtained from the position of the edge of total reflection in the X-ray intensity-reflection angle plot. This edge value is also known as the critical angle for the reflection. Since the absorption of x-rays is dependent on the density of the material, the shape and position of this edge provide the necessary information about the material density.\(^7,9\)

2.2. Magnetic Characterization

Magnetic films are characterized in terms of both static and dynamic magnetic properties. For static characterization, superconducting quantum interference device (SQUID) and looper setups were used. Dynamic characteriza-
tions were mainly based on ferromagnetic resonance (FMR) measurements. FMR measurements were done in two different ways. The first type of measurement was done in a resonant cavity at constant microwave frequency. The second type of measurement was done with a waveguide, where the microwave frequency was varied in the broad range.

2.2.1. Static Magnetization

Looper
A looper is a setup which measures hysteresis curves of magnetic flux density versus magnetic field strength, and also known as a $BH$ looper. Basically a $BH$ looper consists of two Helmholtz coils and two identical pick-up coils. An AC magnetic field is applied to the Helmholtz coils to change the flux and this may induce a voltage in the pick-up coils. To avoid this problem the pick-up coils are connected differentially. Since they are connected differentially, the induced voltage coming from the flux change due to the Helmholtz coils is cancelled out and the voltage induced by the time varying magnetization of the measured thin film positioned in the center of one of the pick-up coils is enhanced. The voltage induced by the thin film sample in the pick-up coil can be defined by Faraday’s law,

$$v = N \frac{d\phi}{dt} = NA \frac{dB}{dt}. \quad (4)$$

Here $A$ is the cross sectional area of the pick-up coil and $N$ is the number of turns. If $v$ is integrated over time, the value of $B$, flux density, is obtained. The magnetic field applied on the thin film by the Helmholtz coil is usually measured by mounting a Hall probe next to the pick-up coil. $BH$ loopers are generally used for obtaining $BH$ hysteresis curves since they make very quick measurements possible. However, they are limited when it comes to sensitivity; hence they are suitable for characterizing large samples$^{10,11}$. In this thesis work a looper was used for measuring the $BH$ hysteresis loops of the FeCo thin films, which were deposited on large substrates with a diameter of 20.32cm (8 inch). Since the applied field by a Helmholtz coil is limited in magnitude it was only possible to saturate the films when the field was applied within the film plane. So only hysteresis loops of in-plane measurements were obtained with this setup. During the measurements, the sample was rotated so the external field was applied both along the easy and the hard axis of magnetization. As a result of these measurements, in plane coercivity ($H_c$) both for the easy and the hard axis, and the saturation magnetization ($M_s$) were extracted. A schematic of the principle of operation of the $BH$ looper is given in figure 2.
When it comes to measuring magnetic flux, the SQUID is one of the most widely used and most sensitive techniques. A SQUID is basically a closed superconducting loop which includes one or two Josephson junctions, referred to as RF and DC SQUID, respectively. The working principle of a SQUID is based on the quantization of flux which is multiples of the constant flux quantum given as

$$\phi_0 = \frac{h}{2e} \approx 2.07 \times 10^{-15} \text{ Wb},$$

where $h$ is the Planck constant and $e$ is the charge of the electron. Since the SQUID is a magnetic flux to voltage transducer, exploiting such a small quantity as the flux quantum makes the SQUID to be an extremely sensitive measurement setup. In a DC SQUID the superconducting loop is biased with a DC current and this current passes through the Josephson junctions. Depending on the external magnetic flux the voltage across the Josephson junctions will change periodically with the period of one flux quantum. A change in this voltage is monitored in order to determine the magnetic flux that is coupled to the superconducting loop\textsuperscript{12,13}. A schematic of the working principle of a DC SQUID is given in figure 3. For this thesis work experiments were done by using an RF SQUID which means that there was only one Josephson junction in the superconducting loop and the loop was inductively coupled to an RF circuit. An RF SQUID with a sensitivity of $10^{-11} \text{ Am}^2$ was included in a Quantum Design magnetometer setup called the magnetic

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**Figure 2. Principle of a BH looper.**
properties measurement system (MPMS) which can apply fields up to 5T. Since the system has such a high sensitivity and can apply such a high field, it was possible to measure small pieces of thin films both with the applied field in-plane and out-of-plane with respect to the film surface.

![Figure 3. Schematic of a DC SQUID.](image)

### 2.2.2. Dynamic Magnetization

The magnetization dynamics of the thin films in this thesis work was characterized using frequency domain measurements. Different types of FMR setups were used. FMR is a phenomenon which defines different aspects of the resonant magnetization dynamics of the magnetic materials. When a static magnetic field is applied to a magnetic material, the magnetization vector of the material tends to align parallel to the applied field. At this point another external microwave excitation field, which is weak in comparison, is applied perpendicular to the static field and this microwave field excites a precessional motion of the magnetization vector around the static magnetic field. The magnetization precesses with a frequency called the Larmor frequency. When the frequency of the microwave excitation field matches the Larmor frequency of the material, resonance condition occurs and the maximum absorption of the microwave signal is observed. The relaxation of the dynamics often referred to as the damping of magnetization dynamics can be extracted by analyzing the absorption line shape of the FMR spectrum. There are different types of FMR setups in terms of measurement geometries and the way of transferring the microwave signal to the sample.

#### Cavity FMR

A resonant cavity FMR technique was used in this thesis to measure the FMR spectrum of the thin films. An Elexysys setup from Bruker was used for this thesis work. This setup consists of a microwave bridge, resonant cavity and an electromagnet. The microwave bridge is used both as a microwave source and a microwave detector. The microwave field is transferred to
the cavity via a waveguide. In case the microwave energy is stored in the cavity without any reflection, this condition is called resonance. This is also necessary condition to run a successful FMR measurement. One of the most important characteristic of the cavity FMR setup is quality factor $Q$ which is $Q = \frac{v_{res}}{\Delta \nu}$ where $v_{res}$ is the resonance frequency and $\Delta \nu$ is the linewidth of the resonance curve. Standing electromagnetic waves occur inside the cavity when there is resonance. This means that if the magnetic field component of the wave is maximum at a point in the cavity, then the electric field component should be minimum, or the other way round. During FMR measurements, the microwave magnetic field interacts with the magnetization of the film and microwave energy is absorbed at FMR condition, which means if the film is located at the point of microwave magnetic field maximum, the highest sensitivity can be obtained. In this system the cavity and the waveguide should be coupled which is provided by the iris. The iris is a structure which matches the impedances of the waveguide and the cavity simply by adjusting the amount of microwaves entering the cavity. This is done by changing its size with the help of a screw type structure. When a film is mounted inside the cavity and the magnetic field is swept, resonance microwave energy is absorbed by the sample. This absorption changes the Q factor and this change results in the reflection of the microwaves from the resonator and recorded as FMR signal. In the setup used during this work, the cavity is located between the poles of an electromagnet, which provides a uniform DC magnetic field. An X-band cavity with constant 9.8 GHz microwave frequency is used. The measurement is performed in field-swept mode, and when the magnitude of the DC magnetic field matches the necessary condition of FMR, absorption in the reflected microwave is observed by the detector at the microwave bridge. The cavity FMR setup is equipped with a goniometer which allows the sample to be rotated with respect to the DC magnetic field direction. Repeating the FMR measurement for different orientations (angles) of the sample with respect to the DC magnetic field direction allows us to extract the type of magnetic anisotropy and anisotropy fields. Two different types of quartz sample holder rods are used in the setup which allows the film sample to be rotated with the DC magnetic field either in or out of the film plane. This feature of the setup helps to observe different types of contributions to the magnetic damping. Although it is a drawback that one cavity can only run at a constant frequency and where you need different cavities to run measurements at different frequencies, cavity FMR is a more sensitive technique compared to other FMR techniques. It is useful when you for instance would like to perform measurements on ultrathin magnetic films. A schematic of a cavity FMR setup is given in figure 4.
In-plane broadband FMR

Beside single frequency cavity FMR measurements, broadband FMR measurements were done where the sample is put face-down on a coplanar waveguide (CPW) and FMR is measured in a wide frequency range. In these measurements a homemade setup was utilized. This setup consists of a microwave signal generator, SMF 100A from Rohde&Schwarz, coaxial cables, an electromagnet, Helmholtz coils, a coplanar waveguide with via holes, an RF diode and a lock-in amplifier for signal detection. A picture of the entire setup is given in figure 5. The signal generator can generate frequencies up to 43.5GHz and the signal is transferred to the CPW via coaxial cables. The CPW can handle frequencies up to 40GHz, so limits the setup in terms of frequency. The measurement is performed in field-swept mode with the DC magnetic field applied perpendicular to the microwave magnetic field. Absorption occurs when the magnitude of the DC magnetic field matches the FMR condition. When recording an FMR spectrum, the microwave frequency is set to a constant value and the DC magnetic field is swept. Basically absorption of the power is recorded when the DC magnetic field and the microwave frequency meet the resonant condition. To improve the sensitivity of the measurement and get rid of the noise in the signal, the magnetic field is low frequency modulated by an AC magnetic field with amplitude 0.25mT and frequency 211Hz applied via the Helmholtz coils to modulate the output FMR absorption signal. An RF diode is used to rectify the microwave signal to a low frequency modulated DC signal, which is then detected.
by the lock-in amplifier. It should be noted that the CPW used in this setup has via holes laid by the central conductor line, which connect the top and bottom layers of the CPW and act as a microwave wall. The created lateral walls in the CPW create another waveguide mode and stops surface waves inside the structure. In order to make it possible that CPW can handle higher frequencies, the distance between these via holes should be less than a quarter of the wavelength of the microwave signal.\textsuperscript{16} When this condition is fulfilled the array of via holes can act as a wall and the high frequency microwave signal can be transmitted among these walls without loss or absorption. This feature of the CPW distinguishes it from ordinary CPWs and makes it operable up to 40GHz. End launchers are also used for providing connection and transfer of microwaves between the CPW and coaxial cables. A photograph revealing the details of the CPW is given in figure 6.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{Photograph of the in-plane broadband FMR set-up. On the top an overall of the set-up is given. On the bottom a zoomed in photo in to the gap between electromagnet poles is given.}
\end{figure}
Out-of-Plane Temperature Dependent FMR

The FMR measurements in this geometry are done by applying the DC magnetic field perpendicular to the film plane at different temperatures by utilizing a custom built setup. The setup used for these measurements consists of a Quantum Design physical properties measurement system (PPMS), a multifunctional probe of the PPMS system, coaxial cables, which can transmit microwaves even at cryogenic temperatures, a CPW, which can also work at cryogenic temperatures and a PNA network analyzer, N5227A from Agilent Technologies. The network analyzer is used both as a source and detector for microwave signals, since it has two ports that measure all $S_{ij}$ parameters of the scattering matrix. The CPW is mounted at the lower end of the multifunctional probe and the sample film is mounted face down on the CPW. The network analyzer can apply microwaves up to 67 GHz. However the CPW can handle frequencies only up to 50GHz, so 50GHz is the limit of the setup in terms of frequency. The PPMS allows a DC magnetic field of up to 9T to be applied, which is applied perpendicular to the film surface. During the measurement, the frequency was set to a constant value on the network analyzer and the applied DC field was swept. The transmitted complex $S_{21}$ signal was measured by the network analyzer. The measurements are repeated at many different frequencies to be able to extract the details of the spin dynamics. Since the PPMS is used for these measurements it is possible to cool down the system with liquid helium and run measurements between 4 and 350K. Cables and waveguide were produced in such a way that they can handle cryogenic temperatures. Like the one used in in-plane measurements, the CPW used for out-of-plane measurements is also equipped with via holes laid next to the central conductor so it can work up to high frequencies. A photograph of CPW used is given in figure 7 and the schematic layout of out-of-plane FMR is given in figure 8.
Figure 7. Photograph of the CPW inserted in the PPMS chamber for out-of-plane FMR measurements.

Figure 8. Schematic of out-of-plane temperature dependent FMR setup.
3. Magnetization Dynamics

3.1. Magnetic Damping

As was explained in the definition of FMR, when a magnetic field is applied on a magnetic material the magnetic moment vector of the material starts precessing around the applied field, and after some time the precession dampens and the moment vector aligns with the applied field vector. Damping of precession and the mechanism behind the alignment of the magnetic moment vector of the material is related to both intrinsic and extrinsic factors that define the damping. Intrinsic damping is called Gilbert damping. However, apart from Gilbert damping, there are a number of extrinsic factors such as two magnon scattering (TMS), eddy current damping, spin pumping into a nonmagnetic metal, radiative damping, and contributions coming from the inhomogeneity and mosaicity of the sample. Understanding the damping properties of thin film materials is quite important since the damping mechanism defines how suitable they are for daily life applications. For some applications a low damping parameter is required since materials with a low damping parameter require low critical switching current for magnetic switching. However, a higher damping parameter is essential for faster switching of magnetization, especially for memory applications.

3.1.1. Gilbert Damping

As mentioned before, to observe the damping a magnetic field should be applied on the magnetic thin film. However, when it comes to the applied static magnetic field, the term effective field $H_{\text{eff}}$ should be introduced first. Total magnetic energy density of a magnetic thin film consists of different energy contributions such as exchange energy, Zeeman energy, demagnetizing energy, cubic anisotropy energy, uniaxial anisotropy energy, perpendicular anisotropy energy and surface anisotropy energy. In order to make the system stable and get to an equilibrium state some of these energies should be minimal. When the derivative of the total energy $\varepsilon_{\text{tot}}$ is calculated with respect to the magnetization vector $\mathbf{M}$, we get the effective magnetic field $H_{\text{eff}}$, as given in equation,

$$H_{\text{eff}} = -\frac{1}{\mu_0} \nabla_M \varepsilon_{\text{tot}}$$ (5)
When a static magnetic field is applied on a magnetic film, the magnetization vector tends to align parallel with $H_{\text{eff}}$, and when a small microwave field is applied perpendicular to the $H_{\text{eff}}$, the magnetization vector starts a precessional motion around $H_{\text{eff}}$. This precessional motion is defined by the Landau-Lifshitz (LL) equation of motion\textsuperscript{21,22}.

$$\frac{dM}{dt} = -\gamma \mu_0 M \times H_{\text{eff}} \quad .$$  \hfill (6)

Here $\gamma$ is the gyromagnetic ratio, where $\gamma = g|e|/2m_e$. $g$ is the Landé-g factor, $m_e$ is the mass of the electron and $e$ is the charge of the electron. However, the precessional movement of the $M$ vector does not last forever, and after a period of time the $M$ vector again tends to align with $H_{\text{eff}}$. This behavior is called damping. In the LL equation the damping term is missing and when a term which defines damping is introduced, the equation is called the Landau-Lifshitz-Gilbert (LLG) equation since the damping term was first introduced by Gilbert\textsuperscript{23}. The LLG equation is given as,\textsuperscript{21,22,24}

$$\frac{dM}{dt} = -\gamma \mu_0 M \times H_{\text{eff}} + \frac{\alpha}{M_s} \left( M \times \frac{dM}{dt} \right) \quad .$$  \hfill (7)

Here $\alpha$ is the dimensionless Gilbert damping constant, which defines how fast the precessional motion dampens and the $M$ vector aligns with $H_{\text{eff}}$. It should also be mentioned that the $M$ vector given in this equation keeps the magnitude conserved during this damping process. The Gilbert damping is an intrinsic property of the films. The origin of this damping is basically spin orbit coupling.\textsuperscript{25,26} In addition to spin orbit coupling it is possible to explain intrinsic damping by scattering of electrons by phonons and magnons. In the case of phonon scattering the process is mediated by spin orbit interaction however in the magnon scattering case angular momentum relaxes as scattered electrons repopulate the magnetization-direction-dependent Fermi volume. Relaxation of magnon modes occurs through exchange interaction with a conduction electron. Afterwards the spin of conduction electron relaxes to the lattice through spin orbit interaction.\textsuperscript{27}

3.1.2 Two Magnon Scattering

Two magnon scattering (TMS) is one of the extrinsic contributions to magnetic damping which increases the linewidth of the FMR absorption, hence the value of damping. When FMR condition occurs, quantized spin waves, known as magnons, may be excited, which precess as a wave defined by the wavenumber $k$. When the precession is uniform, $k = 0$. Then, these uniform spin waves can be scattered at scattering centers, to degenerate, non-uniform states where $k \neq 0$ and dissipate energy into the lattice. This process is called TMS.\textsuperscript{28–30} In the magnetic films TMS can occur depending on the defects in the structures, grains and grain boundaries.\textsuperscript{28–33} As well as affect-
ing the extrinsic relaxation by creating scattering centers for the TMS process, grains can also affect the increase of the damping parameter. Larger contributions of TMS to the relaxation can be observed depending on the grain size and anisotropy field. The TMS process is highly dependent on the measurement geometry. As mentioned in the section 2.2.2, broadband FMR measurements were run in two different geometries where the DC magnetic field was applied within the film plane and out of the film plane. The TMS process is angle dependent when the DC field is applied within the film plane, which may be used to extract information about the TMS contribution to the measured damping parameter. The TMS contribution can also be revealed by studying the frequency dependence of the FMR absorption linewidth, which should exhibit a nonlinear behavior if the measurements have been performed in a wide enough frequency range. However, when the FMR measurement is done by applying the DC field out of the film plane the TMS effect is suppressed. Therefore, out-of-plane FMR measurements are preferable in order to get rid of the TMS contribution and get a damping value which is closer to the intrinsic Gilbert damping parameter.

3.1.3. Eddy-Current Damping

Eddy-current damping is another extrinsic contribution to damping which comes from the interaction of the measured film with the high frequency microwave magnetic field. Eddy-current damping can be explained as a relaxation process caused by screening of the electromagnetic microwave field by the conduction electrons of the investigated film. A more macroscopic explanation is that an AC voltage is induced when a conducting material is exposed to a time varying magnetic flux created by the microwave magnetic field and the precessing magnetic moment. This fundamental phenomenon is known as Faraday’s law. AC currents are induced both in the CPW and in the ferromagnetic thin film when the spins precess in the conducting film. Energy dissipation due to these induced eddy currents is known as eddy-current damping. It should be mentioned that eddy-current damping is a thickness dependent contribution to the relaxation. If a film is thin enough eddy current damping is negligible but thick films, on the other hand, are exposed to a higher eddy-current damping contribution. The critical thickness required for a conducting film in order to induce a significant eddy current damping is defined by the skin depth, which can be explained as the depth below the surface of the film where the microwave magnetic field and the density of the current drops to 1/e of the their values at the surface. When the thickness of the film is comparable to or larger than the skin depth, then eddy-current damping becomes significant.
3.1.4. Radiative Damping

Radiative damping is a contribution to the relaxation which works in a similar way to eddy-current damping and comes from inductive coupling of the CPW and the measured thin film.\textsuperscript{39} Basically, as explained in the eddy-current damping section 3.1.3 precessing spin waves induce eddy-currents which dissipate energy. The contribution to the magnetic damping caused by eddy-currents induced in the CPW is known as radiative damping.\textsuperscript{36}

3.1.5. Spin Pumping

When the thin film structure is a stack of layers which includes a ferromagnetic layer and adjacent nonmagnetic metallic layers, it is possible to observe an enhanced damping parameter in such structures. Since spin currents are generated at the interface between a ferromagnetic layer and a nonmagnetic metallic layer due to the precessing magnetization, spin angular momentum can be injected into the nonmagnetic adjacent layer and relaxed in that layer. This relaxation enhances the damping in such structures.\textsuperscript{22,34} This phenomenon is observed when there is a stronger spin orbit coupling in the nonmagnetic layer and it is also thickness dependent, so if the nonmagnetic layer thickness is less than the spin diffusion length, then spin pumping contribution is negligible.\textsuperscript{22,40}

3.1.6 Mosaicity Contribution

Mosaicity in the thin film structure could also be a source of enhanced relaxation. If the crystallite orientations, internal fields and thickness of the film vary from one region to another within the film structure, then all of these individual regions can have slightly different resonance fields. The overall FMR signal will include a superposition of these local absorption contributions and will thus result in a broader absorption FMR linewidth, which means larger damping.\textsuperscript{41–43}

3.1.7. Inhomogeneity Contribution

Finally, it is also well-known that there is an extrinsic contribution to the magnetic relaxation due to inhomogeneity in the film structure, which is a frequency independent contribution. This inhomogeneity is caused by local variations of the magnetization and internal magnetic fields. This contribution is also defined in terms of inhomogeneity of local demagnetizing fields.\textsuperscript{44–46} In some cases this contribution can also be thickness dependent due to dispersion of the anisotropy in the films which increases by decreasing of the film thickness.\textsuperscript{44}
3.2. Equations for Analyzing Magnetization Dynamics

3.2.1. Cavity FMR Analysis

In cavity FMR analysis the absorption of the microwave power is measured with respect to the applied DC field, and the signal recorded by the equipment comes in the shape of the field derivative of the absorption. The reason for this is that the DC magnetic field is modulated by a low frequency AC magnetic field. The recorded raw data can therefore be fitted to the equation

\[
\frac{dA}{dH} \propto \frac{2(H-H_r)\frac{\Delta H}{2}}{\left[\left(\frac{\Delta H}{2}\right)^2+(H-H_r)^2\right]^2} - \frac{\left(\frac{\Delta H}{2}\right)^2-(H-H_r)^2}{\left[\left(\frac{\Delta H}{2}\right)^2+(H-H_r)^2\right]^2},
\]

where \(\frac{dA}{dH}\) is the magnetic field derivative of the microwave absorption. The resonance field \(H_r\) and the full width half maximum of the FMR absorption linewidth \(\Delta H\) were extracted from the raw data by fitting the data to this function. Hence these parameters were used as fitting parameters for the fitting. Since cavity FMR measurements are angle-resolved measurements, plotting angle dependent \(H_r\) and \(\Delta H\) values gives further understanding of the magnetic properties of the thin film samples.

When the \(H_r\) values were extracted with respect to angle from in-plane measurements, they were fitted by using the equation

\[
f = \frac{Y\mu_0}{2\pi} \left[\left(H_r \cos(\phi_H - \phi_M) + \frac{H_c}{2} \cos 4(\phi_M - \phi_C) + H_u \cos 2(\phi_M - \phi_u)\right) \times \left(H_r \cos(\phi_H - \phi_M) + M_{\text{eff}} + \frac{H_c}{8} (3 + \cos 4(\phi_M - \phi_C)) + H_u \cos^2(\phi_M - \phi_u)\right)\right]^{1/2},
\]

In equation (9) \(f\) is the constant frequency of the setup which is 9.8GHz and \(H_r\) is the resonance field. \(\phi_H, \phi_M, \phi_u, \phi_C\) are the directions of the magnetic field, magnetization, uniaxial and cubic anisotropy, respectively, with respect to the \([100]\) direction of the thin film substrate. \(H_u\) and \(H_c\) are the uniaxial and cubic anisotropy fields, respectively, where \(H_u = \frac{2K_u}{\mu_0 M_s}\) and \(H_c = 4K_c - \frac{4K_u}{\mu_0 M_s}\). Here \(K_u\) and \(K_c\) are the uniaxial and cubic magnetic anisotropy constants. \(M_{\text{eff}}\) is the effective magnetization and is defined as \(M_{\text{eff}} = M_s - H_k^+\), where \(H_k^+\) is the perpendicular anisotropy value. When angle-
resolved $H_r$ data were fitted to equation (9), $H_c$, $H_u$ and $M_{eff}$ were used as fitting parameters. Extracted $H_u$ and $H_c$ values, in conjunction with the pattern of the angle resolved $H_r$ plot, clearly reveal the type of dominant in-plane anisotropy in the film and the angle-resolved cavity FMR analysis is thus a very useful technique in this sense.

3.2.2. In-plane Broadband FMR analysis

FMR absorption spectra were recorded via the lock-in detection technique for in-plane broadband FMR measurements; hence the recorded microwave power versus magnetic field pattern comes in the shape of a derivative function. Since we have a derivative function shape, the same equation (8), as used in the extraction of $H_r$ and $\Delta H$ for the raw data recorded in cavity FMR measurement, was used here as well. Once the $H_r$ resonance fields are extracted for different frequencies, an $H_r$ versus frequency graph was plotted and fitted to an equation which is valid for the in-plane measurement geometry. The equation used for the in-plane measurement case can be modified depending on whether the DC magnetic field is applied along the easy axis or the hard axis of the film. In the equation set given below first equation is written for the easy axis and the second one is written for the hard axis.

\[
 f = \frac{\mu_0 Y}{2\pi} \sqrt{(H_r + H_u)(H_r + H_u + M_{eff})} \text{ (easy axis),}
\]

\[
 f = \frac{\mu_0 Y}{2\pi} \sqrt{(H_r - H_u)(H_r + M_{eff})} \text{ (hard axis).} \tag{10}
\]

$M_{eff}$ and $H_u$ were used as fitting parameters and the corresponding values were extracted.

Apart from the $H_r$ values, extracted $\Delta H$ values were also plotted versus frequency and this plot was fitted to equation

\[
 \mu_0 \Delta H = \frac{4\pi \alpha}{\gamma} f + \mu_0 \Delta H_0, \tag{11}
\]

where $\alpha$ is the total damping parameter, including both intrinsic and extrinsic contributions, and $\Delta H_0$ is the extrinsic frequency independent contribution to the linewidth of the absorption spectra. The main purpose of this fitting was to extract the total damping parameter of the film and distinguish the frequency independent contribution to the linewidth.

3.2.3 Out-of-plane Broadband FMR analysis

Out-of-plane FMR data were recorded by using a network analyzer instead of the lock-in technique used for the in-plane measurements. The transmitted complex $S_{21}$ parameter was detected via the network analyzer and the raw data were fitted to the equation set given below.

29
\[ S_{21}(H,t) = S_{21}^0 + Dt + \frac{\chi(H)}{\tilde{\chi}_0}, \]
\[ \chi(H) = \frac{M_{\text{eff}}(H-M_{\text{eff}})}{(H-M_{\text{eff}})^2 - H_{\text{eff}}^2 - i\Delta H(H-M_{\text{eff}})}, \quad (12) \]

where \( H_{\text{eff}} = \frac{2\pi f}{\gamma\mu_0} \). Here \( S_{21} \) is the complex transmission parameter of the microwave signal where \( S_{21}^0 \) is the nonmagnetic contribution to \( S_{21} \). \( \tilde{\chi}_0 \) is a frequency and film thickness dependent imaginary function, and \( \chi(H) \) is the complex magnetic susceptibility of the film. \( D \) is a phenomenological complex coefficient which is used for correction of the drift, depending on the time, coming from the measurement electronics. \( H_r \) and \( \Delta H \) were used as fitting parameters and extracted from the fitting of raw data to equation (12). Extracted \( H_r \) values were plotted versus frequency and this graph were fitted to the linear function

\[ \mu_0 H_r = \frac{2\pi f}{\gamma} + \mu_0 M_{\text{eff}}, \quad (13) \]

which enabled the extraction of \( M_{\text{eff}} \) and the Landé \( g \)-factor. \( \Delta H \) values versus frequency were plotted and fitted to equation (11) in the same way as in the in-plane analysis and the damping parameter and frequency-independent extrinsic linewidth contribution were extracted.

### 3.2.4. Subtracting Some Extrinsic Relaxation Contributions in Out-of-plane Measurements

**Eddy current damping**

Eddy current damping in the system can be calculated and subtracted from the total damping value of the film by using the equation,

\[ \alpha_{\text{eddy}} = \frac{C\gamma\mu_0^2M_s\delta^2}{16\rho}, \quad (14) \]

where \( C \) is a coefficient which defines the eddy-current distribution in the thin film structure. A decrease in the value of \( C \) means larger localization of eddy currents in the film. \( \rho \) and \( \delta \) are the resistivity and thickness of the film, respectively. The ideal way of obtaining \( C \) is to subtract all other extrinsic parameters from the total damping for samples with different thicknesses and to measure the resistivity of the films for different thicknesses and then plot the remaining damping parameter versus \( \delta^2 \) and extract the \( C \) value from the linear fit.
Radiative damping
The radiative damping contribution in the system can be calculated and subtracted from the total damping by using the expression,\(^{36,39,40}\)

\[
\alpha_{rad} = \frac{\eta \mu_0^2 M_s \delta l}{2Z_0 \omega},
\]

where \(\eta\) is a dimensionless parameter that accounts for the FMR mode profile, \(\delta\) and \(l\) are the thickness and length of the sample on the waveguide, respectively, \(\omega\) is the width of the center conduction line of the CPW and \(Z_0\) is the impedance of the CPW.

It is also possible to determine the radiative damping contribution in the system experimentally. When a glass spacer with large enough thickness is placed between the CPW and the sample, the radiative damping contribution becomes negligible.\(^{36,39,40}\)

Spin pumping
The contribution to magnetic relaxation due to spin pumping through interfaces within the film can be described by the equation,\(^{39}\)

\[
\alpha_{sp} = \frac{g \mu_B g_{eff}^\uparrow \downarrow}{4\pi M_s \delta},
\]

where \(g_{eff}^\uparrow \downarrow\) defines the effective spin-mixing conductance. It should also be mentioned that equation (16) is given just for the simplest case of one interface in the film structure. It is also possible to have a sandwich type structure with two identical interfaces. Then a coefficient of 2 should be added to the equation.\(^{39}\) The ideal way to extract \(g_{eff}^\uparrow \downarrow\) is to first measure the damping parameter of identical films with different thicknesses and then subtract all other extrinsic damping contributions from the total damping parameter and then plot the remaining spin-pumping contribution versus film thickness and fit that plot to equation (16) and extract \(g_{eff}^\uparrow \downarrow\) as a fitting parameter.
4. Dynamic Properties of Co$_2$FeAl Thin Films

This thesis is mainly structured around investigations of two different types of magnetic alloys in terms of the dynamic magnetic properties. One of these alloys is Co$_2$FeAl, which is also known as a full Heusler alloy, and the other is Fe$_{65}$Co$_{35}$. In this chapter material properties, the structural and magnetic characterization of Co$_2$FeAl are covered and the next chapter is about Fe$_{65}$Co$_{35}$. This chapter is based on the work done in collaboration with Sajid Husain and Sujeet Chaudhary from Indian Institute of Technology Delhi, whom contributed for paper I, II, III and IV.

4.1. Material Properties

Today, intensive fundamental research is being done on Heusler alloys and in particular on Co-based Heusler alloys and the specific composition Co$_2$FeAl. These alloys possess unique properties, such as high Curie temperature ($T_C = 1000K$), full spin polarization at Fermi level, which is also interpreted as half metallicity and high magnetization all of which are favorable properties for future spintronic applications in terms of producing faster and more energy efficient devices.\(^{50-57}\)

Co$_2$FeAl belongs to the structure family known as full Heusler alloys and has an $X_2YZ$ structure with three different phases. These three phases are labelled L$_{21}$, B$_2$ and A$_2$, which are fully ordered, partially ordered and fully disordered, respectively. In the L$_{21}$ phase, the different types of atoms occupy the sites which are individually assigned for them. This structure depicts the full spin polarization at the Fermi level and is known as a half-metallic structure.\(^{58}\) In the B$_2$ phase, Y and Z atoms, Fe and Al particularly in our study, share their sites randomly. A$_2$ phase is the name of the structure where all sites are randomly occupied by all different types of atoms.\(^{53,58-60}\) Physical properties of the films, and especially the half-metallicity of the produced thin films, are strongly dependent on the organization of the atoms within the crystalline structure. Half-metallicity or full-spin polarization is obtained when all atoms are fully ordered, as in the L$_{21}$ phase. Disorder in the crystal structure ends up adding extra states at the Fermi level, which reduces the spin polarization, hence the half-metallicity.\(^{50,61}\) Reduced spin polarization results as in increased value of the Gilbert damping parameter.
In this thesis work, the ion-beam sputtering technique was used in order to deposit all Co$_2$FeAl thin films as explained in paper I.\textsuperscript{52} Firstly, different films were deposited on Si(100) substrate in order to investigate the effects of deposition temperature on film structure. These samples deposited at different temperatures were used for the first part of the study. The substrate temperature was fixed at 300K, 573K, 673K and 773K. Co$_2$FeAl films were deposited at these temperatures with a nominal thickness of 53 nm and then capped with Ta with a nominal thickness of 2nm. Further details about the film deposition technique are explained in paper I. Structural analysis was done on these films in order to determine the crystallographic phase of the films, and then magnetization dynamics was investigated in order to understand the relation between the crystallographic phase and damping parameter.

Further study was done when the growth conditions were optimized. As a second part of the study, the effect of different seed layers and capping layers were investigated in terms of magnetization dynamics in order to understand the interfacial effects on magnetic damping. Information extracted from these studies would be useful for producing structures that make use of spin-transfer torque in spintronic devices. In this part, firstly, thin films of Co$_2$FeAl were deposited on a Si(100) substrate where a seed layer of Ta with nominal thickness of 10nm was added between Co$_2$FeAl and Si. The nominal thickness of the Co$_2$FeAl was kept constant at 8.4nm for all samples. One of the Co$_2$FeAl films was kept without a capping layer and two of them were capped with MgO and Ta with nominal thicknesses of 2nm and 4nm, respectively. During the deposition process the Ta seed layer was annealed at 673K for 30 minutes before the deposition of Co$_2$FeAl. After the deposition was completed all samples were annealed at 523K for one hour.

Beside studying structures with MgO capping and Ta seed, Co$_2$FeAl films were deposited with different thicknesses and Ta capping just to understand the effect of varying thicknesses and Ta interface on magnetic damping. For this part of the study Co$_2$FeAl films were deposited on Si(100)/SiO$_2$ with a capping layer of Ta. The nominal thickness of the Ta was 5nm and the nominal thickness values of Co$_2$FeAl were 8, 10, 12, 14, 16, 18 and 20 nm. Again the substrate temperature was kept constant at 573K since it is the optimal growth temperature determined in the initial part of the study. Results of this investigation revealed mainly the effect of the spin pumping contribution to the magnetic relaxation which occurs at the interface of Co$_2$FeAl and Ta. Apart from this the trend in behavior of magnetic damping with respect to Co$_2$FeAl thickness was determined.

In addition to the investigation of the effects of different interfaces of nonmagnetic layers, the magnetization dynamics of Co$_2$FeAl films were studied in a temperature-dependent way. For this part of the work Co$_2$FeAl films were again deposited on a Si(100) substrate where the substrate temperature was fixed at 573K, 673K and 773K. As was understood from the
first part of the study, substrate temperature during film deposition is the key factor which determines the crystal phase of the film. The reason for depositing samples at different temperatures was to study the temperature dependence of the magnetic damping for films with different degree of structural order. These Co$_2$FeAl films were deposited with a nominal thickness of 50 nm and were capped with Al with 4 nm nominal thickness. In order to get the fully disordered phase of Co$_2$FeAl, one film was deposited at 300K and it was capped with Ta. Related information about deposition parameters used in order to obtain a fully disordered phase is explained elsewhere.$^{63}$ Detailed analysis of the temperature-dependent damping profile of Co$_2$FeAl films with different crystal phases revealed the temperature dependent trend of magnetic damping. In this part of the study extrinsic magnetic relaxation contributions such as eddy-current damping and radiative damping are discussed in detail. Beside experimental studies, theoretical calculations about how intrinsic Gilbert damping behaves with respect to temperature were performed in this part of the study. By subtracting the extrinsic relaxation contributions, intrinsic Gilbert damping values were obtained experimentally and they were compared with calculated values.

As a final part of the investigation of Co$_2$FeAl, Co$_2$FeAl films were deposited with ultrathin thickness in order to investigate the effects of interface anisotropy on the magnetic properties and to observe perpendicular magnetic anisotropy. It is known that perpendicular magnetic anisotropy (PMA) can be obtained in ultrathin layers of a 3$d$ ferromagnetic element or alloy in direct contact with an oxide.$^{64}$ The origin of the PMA is hybridization between the ferromagnetic 3$d$ orbitals and the oxygen 2$p$ orbitals of the oxide, and this is therefore referred to as interfacial anisotropy. The hybridization between these orbitals makes the energy for 3$d$ orbitals pointing towards the interface (3$d_{xy}$, 3$d_{xz}$ and 3$d_{z^2}$) smaller than the energy for orbitals with planar symmetry (3$d_{xy}$ and 3$d_{x^2-y^2}$), resulting in a strong PMA. Systematic studies have shown that the PMA requires optimally oxidized stoichiometry of the oxide,$^{64}$ implying that both under- and over-oxidized stoichiometry of the oxide will prevent the magnetic layer from exhibiting PMA. Since interfacial PMA has been shown to exist for ultrathin layers of Co$_2$FeAl in direct contact with MgO,$^{65}$ we decided to make a detailed study of PMA in Co$_2$FeAl /MgO structures. Prior to deposition, the Si(100) substrate was treated with HF solution to remove the native SiO$_2$ layer and then immediately loaded into the high vacuum chamber of the ion-beam sputtering deposition system. A series of thin film Si/Ta(10 nm)/Co$_2$FeAl (t)/MgO(2 nm)/Cr(2-3 nm) structures were prepared, with the nominal thickness of Co$_2$FeAl varying the range 1 ≤ t ≤ 1.8 nm. The growth steps were as follows: Ta was room temperature (RT) deposited and then annealed at 400 °C for 30 minutes; Co$_2$FeAl was grown at RT; MgO was grown at RT and then the trilayer stack was annealed at 280 °C for 60 minutes; and the structure was completed by a Cr cap deposited at RT.
4.2. Analysis and Results

In the initial part of the study samples were first characterized in terms of their structural properties. Since the magnetization dynamics of these films are closely related to the crystal phase, this study was necessary before analyzing the magnetic properties of the samples. When the data coming from measurements related to structural analysis were interpreted and film deposition parameters were optimized then this information was used for further parts of the study. XRD and XRR were the mostly used methods when it comes to structural characterization in this thesis work. First of all, XRD measurements were done on films deposited at 300K, 573K, 673K and 773K in order to resolve whether the crystal phase of the film was A$_2$, B$_2$ or L$_{21}$. In the XRD pattern a peak from (200) diffraction was accepted as evidence of the formation of B$_2$ phase (paper I). However, it is necessary to observe diffraction peaks from (111) and (311) planes as evidence of the L$_{21}$ phase. XRD spectra of the films are given in figure 9.

![XRD Spectra](image)

*Figure 9. XRD spectra of the films prepared with stacking of Si/Co$_2$FeAl/Ta which were deposited at different temperatures (paper I) (the figure was taken from the article “Growth of Co$_2$FeAl Heusler alloy thin films on Si(100) having very small Gilbert damping by ion beam sputtering”, S. Husain *et al.*, *Scientific Reports* 6, 28692 (2016)).*

Appearance of the (200) peak for all films deposited at different temperatures is clear evidence of the presence of B$_2$ phase in all films. As well as (200), diffraction from (220), (400) and (422) planes were observed as well. The appearance of these peaks is because of the polycrystalline structure of the prepared films (paper I). When it comes to distinguishing the L$_{21}$ phase, it is difficult to resolve whether the structure is B$_2$ or L$_{21}$ by just relying on XRD analysis. However, the degree of B2 ordering can be calculated by
employing the Webster model along with the approach of Takamura et al.\textsuperscript{40,67} According to this approach the degree of B\textsubscript{2} ordering can be given as

\[ S_{B2} = \sqrt{\left(\frac{I_{200}}{I_{220}}\right) / \left(\frac{I_{200}^{\text{full order}}}{I_{220}^{\text{full order}}}\right)}, \]

(17)

where \( I_{200} / I_{220} \) is the intensity ratio of diffraction from (200) and (220) planes, obtained experimentally. \( I_{200}^{\text{full order}} / I_{220}^{\text{full order}} \) is the intensity ratio for the (200) and (220) peaks, which is theoretically calculated for the B\textsubscript{2} phase.\textsuperscript{68} \( S_{B2} \) values, for samples investigated in the temperature-dependent study part of the thesis, which were deposited at 573K, 673K and 773K are \(~90\%\), \(~90\%\) and \(~100\%\), respectively.\textsuperscript{68} The ratio of \( I_{200} / I_{400} \) was extracted as \(~30\) for all three samples deposited, which is comparable with the theoretical value for perfect B\textsubscript{2} order.\textsuperscript{69,70} When it comes to the L\textsubscript{21} phase, extracting the value of ordering parameter \( S_{L21} \) is also dependent on the ordering degree of the B\textsubscript{2} phase. It is possible to calculate the ordering degree of the L\textsubscript{21} phase depending on the intensity ratio of (111) and (220) peaks, given as \( I_{111} / I_{220} \), in conjunction with the \( S_{B2} \) parameter.\textsuperscript{67} However, a diffraction peak coming from the (111) planes was not obtained in the XRD spectra for any of the samples. This can be due to the fact that the theoretical intensity of the (111) peak is just 3\% of the principal (220) peak. In this manner it is very difficult to analyze the L\textsubscript{21} phase just depending on the XRD analysis. Since magnetic properties are strongly dependent on the crystalline ordering of the Co\textsubscript{2}FeAl films further discussion will be done in the coming part of this chapter on the structure depending on the magnetic analysis of the films. Beside films deposited at 573K, 673K and 773K, the film deposited at 300K and capped with Ta, did not reveal any (200) diffraction peak \textsuperscript{63}, which can be interpreted as this film exhibits the A\textsubscript{2} crystalline phase.

In addition to XRD analysis, the XRR technique was also used to analyze the surface and interface roughness of the films, as well as to obtain accurate thickness values of the different layers within the film stacking. For the films prepared for the initial part of the study, which have a stacking of Si/Co\textsubscript{2}FeAl/Ta, an oxidized surface layer of Ta with Ta\textsubscript{2}O\textsubscript{5} structure was observed. The surface roughness of the Co\textsubscript{2}FeAl films varies in the range between 0.41(±0.03) and 1.23(±0.03)nm, where the roughness increases with increasing deposition temperature. Full detailed information, including surface roughness, density and thickness of individual layers within the film stack is given in paper I. In the second part of the study, XRR was only done for the stacking of Si/Ta/Co\textsubscript{2}FeAl/MgO where there is a surface roughness of 0.10 nm. XRR measurements performed on the Si/Co\textsubscript{2}FeAl/Ta stacking again revealed that there is an oxidized Ta\textsubscript{2}O\textsubscript{5} layer on top of the Ta capping layer. The surface roughness of the Co\textsubscript{2}FeAl layer varies between 0.34(±0.06) and 0.60(±0.06) nm.
Regarding the magnetic properties of the deposited films, first Co$_2$FeAl films deposited at different temperatures were investigated by using the magneto-optic Kerr effect (MOKE) technique. Normalized hysteresis loops for samples deposited at different temperatures are given in paper I. Perfect square-shaped hysteresis curves were observed when MOKE measurements were done along the easy axis of the samples, which reveals that samples are almost defect free. When the measurement was varying the in-plane azimuthal angle, a uniaxial anisotropy was observed. $H_c$ values of the films were extracted via MOKE measurements. $M_s$ values of the samples were measured using the vibrating sample magnetometer (VSM) option of the PPMS setup. A presentation of $M_s$ and $H_c$ values with respect to deposition temperature is given in paper I. The extracted $M_s$ values are in reasonable agreement with the 5.0µB/f.u $M_s$ value of bulk Co$_2$FeAl. The increase in $M_s$ and decrease in $H_c$ by increasing temperature can be attributed to the improved ordering.

In addition to the magnetometer measurements, films were also investigated in terms of magnetic dynamics in order to decide which deposition temperature was the optimum in terms of obtaining the most favorable dynamic properties. In this part of the work, FMR measurements were done by applying the static magnetic field within the film plane. The measurements were performed in field sweep mode for frequencies between 6 and 12 GHz in steps of 1 GHz. Recorded raw FMR spectra were fitted to symmetric and antisymmetric derivative Lorentzian functions, as given in paper I, in order to extract $H_f$ and $\Delta H$ values. Extracted $H_f$ values were plotted versus frequency and this plot was fitted to the Kittel equation, and as a result of this fit the effective magnetization $M_{eff}$ and the in-plane uniaxial anisotropy field $H_u$ were extracted. Depending on the deposition temperature, $H_u$ varies between 1 and 25 Oe. As explained in section 3.2.1, $M_{eff}$ is dependent on $M_s$ and $H_u$. In this thickness regime, perpendicular anisotropy is not expected in the films, hence it is expected that the $M_{eff}$ values to be similar to the $M_s$ values. As given in paper I, $\mu_0M_s$ varies between 1.2 and 1.29 T whereas $\mu_0M_{eff}$ varies between 1.34 and 1.4 T. The small offset between $M_s$ extracted from magnetometry and the $M_{eff}$ extracted from dynamic measurements can be attributed to difficulties and errors in determining the exact magnetic volumes of the films used in the magnetometer measurements. $\mu_0\Delta H$ values were plotted with respect to frequency and fitted to equation (11). By this fit, an $\alpha$ value of 0.0015±0.0001 was obtained for the film deposited at 573K, and this value is comparable to the lowest values obtained for this alloy. It should be mentioned that both of these studies in literature include a post-annealing process of the as deposited films and both of the films were deposited on MgO substrates. However, in our case, post-annealing was avoided and the films were deposited on a Si substrate, which is a commonly used material in industrial applications. In addition to the $\alpha$ value, equation (11) includes a frequency independent linewidth term.
due to sample inhomogeneity, and this value was extracted as 1.64(±0.25) and 6.46(±0.09) mT for films deposited at room temperature and 573K, respectively. It was observed that the linewidth due to sample inhomogeneity is larger for the film deposited at 573K, which has the lowest damping constant. The $\mu_0\Delta H_0$ value does not vary systematically with deposition temperature and it has the lowest value for film deposited at room temperature and the largest at 573K. Since these FMR measurements were done with in-plane measurement geometry, it could be expected that damping parameter may include a TMS damping contribution. To clear this issue, necessary fittings of $H_r$ and $\Delta H$ plots were done to relevant equations (details are given in paper I) and the TMS corrected $\alpha$ was found as 0.00139(±0.00013). Extracted damping parameters with and without the TMS contribution are given in paper I. As can be seen from the figure 9 in the paper, the TMS does not give a very large contribution to the $\alpha$ values. When the $\Delta H$ values were plotted with respect to frequency, the shape of the curve was linear, although a non-linear curve is expected when there is a significant contribution from TMS. Hence it can be concluded that the contribution coming from TMS was small in this part of the study.45

In the second part of this chapter, films with a stack of Si/Ta/Co2FeAl were prepared by ion-beam sputtering, where one sample was kept without capping and the other two were capped with MgO and Ta. In the literature it is reported that a decrease in the $\alpha$ value is expected due to spin accumulation at the ferromagnetic layer/MgO interface and a back-flow of spin current creating an anti-damping spin torque.72 The $M_s$ value of 9.65(±0.25)×105 A/m was obtained for the MgO capped film, which is comparable to the $M_s$ of bulk Co2FeAl. $H_c$ values were extracted by utilizing MOKE and the values for uncapped, Ta-capped and MgO capped films were 0.54, 0.71 and 0.88 mT, respectively. The reason for higher coercivity in the MgO-capped film could be interfacial pinning centers. As in the previous part of the study, in-plane FMR measurements were done in field sweep mode at different frequencies. Obtained raw FMR spectra were fitted to the derivative function given in paper I, and $H_r$ and $\Delta H$ values were extracted. $H_r$ versus frequency curves were fitted to the Kittel equation as given in the paper II.73 By this fitting, $\mu_0M_{eff}$ values of 1.211(±0.003), 1.170(±0.002) and 1.201(±0.007) T were obtained for the MgO capped, Ta capped and uncapped samples, respectively. In addition, $\mu_0H_u$ values were extracted as 4.92(±0.02), 2.87(±0.03) and 4.98(±0.02)mT for MgO capped, Ta capped and uncapped samples, respectively. Beside analysis of the frequency dependent resonance field, $\Delta H$ versus frequency were fitted to equation (11) and the $\alpha$ parameter was extracted. Obtained $\alpha$ values for different stacks of films are given in figure 10. Among the films prepared for this part of the study, the lowest $\alpha$ value of 0.0075(±0.00014) was obtained for the MgO-capped film. $\alpha$ was increased when the film was capped with Ta, which can be attributed to higher spin-pumping contribution to the magnetic relaxation.
coming from the Co2FeAl/Ta interface. This possible effect is investigated further for Co2FeAl films in the coming part of this chapter. XRR revealed that there is an oxide layer on top of the Co2FeAl structure for the uncapped film, which could be a possible reason for the highest obtained $\alpha$ among the different film stackings. In addition to $\alpha$, fitting to equation (11) revealed frequency-independent linewidth contributions to the FMR spectra, which were $0.12(\pm 0.013)$, $1.95(\pm 0.013)$ and $0.84(\pm 0.013)$ for MgO capped, Ta capped and uncapped films, respectively. It can be concluded that the film with MgO capping exhibits the best quality, by depending on its almost negligible frequency independent linewidth contribution.

![Figure 10. Extracted $\alpha$ values for different film stacks. (taken from paper II)](image)

After analyzing the interface effects of Ta seed and Ta and MgO capping on Co2FeAl films, detailed investigation was also done on the effect of thickness variation and interface contribution to magnetic relaxation. Ultrathin magnetic electrode layers are required for spin transfer torque devices, however which can lead to an increase in magnetic damping due to interface effects. Since surface and interface effects of the films tend to increase by decreasing film thickness, it is expected that the effect on magnetic relaxation will be increased by decreasing film thickness.75 Besides the effect of film thickness, its known that precession of the magnetization in the ferromagnetic layer transfers spin angular momentum into the nonmagnetic layer.76 Due to this transfer, magnetic relaxation may occur in the nonmagnetic
layer and as a result damping parameter is enhanced in such ferromagnetic/nonmagnetic stacks. Since it is common to use ferromagnetic/nonmagnetic stacking of different materials in spin transfer torque devices understanding of this spin pumping effect is critical in order to improve favorable materials for such devices. The spin pumping effect is observed when a ferromagnetic layer is in direct contact with a nonmagnetic layer with a large spin orbit coupling. Enhancement in magnetic relaxation can be expected when nonmagnetic materials of heavier elements with $p$ and $d$ electrons in the conduction band are used in such stackings. Enhancement of the damping parameter is not expected when either lighter elements or heavier elements with only $s$ electrons in their conduction band are used.

For this part of the study, film stacks of Si/Co$_2$FeAl/Ta were deposited by ion-beam sputtering with Co$_2$FeAl thicknesses of 8, 10, 12, 14, 16, 18 and 20 nm and 5 nm of $\beta$-Ta capping. The magnetization dynamics was analyzed by using angle resolved cavity FMR technique, in-plane and out-of-plane broadband FMR techniques. Out-of-plane FMR technique was utilized, since, as mentioned in section 3.1.2, the TMS contribution is avoided in this measurement geometry. When films are measured in out-of-plane geometry, a contribution to magnetic relaxation is expected only to come from intrinsic and extrinsic effects, such as thickness dependent spin pumping at the Co$_2$FeAl/Ta interface. Comparing in-plane and out-of-plane measurement results is quite enlightening in terms of understanding the effect of TMS. The angle resolved cavity measurement reveals the type of in-plane anisotropy and anisotropy fields. In-plane hysteresis curves recorded via SQUID measurements for 8, 14 and 20nm thick films are given in figure 11. An out-of-plane hysteresis curve for the 20nm thick film is also given in the same figure. According to our SQUID measurements, a rectangular hysteresis curve was observed for all samples, revealing also a weakly thickness-dependent $\mu_0H_c$ variation in the range of 0.65 to 1.1mT. The $\mu_0M_s$ value was estimated from the saturation field in the out-of-plane measurement, indicating a weakly thickness-dependent saturation magnetization with a value of about 1.10T.
Figure 11. Normalized in-plane hysteresis curves for Co$_2$FeAl films with different thicknesses. Out-of-plane normalized hysteresis curve for the 20nm thick Co$_2$FeAl film is shown in the inset. (where $t_{CFA}$ is the Co$_2$FeAl thickness)

Figure 12. $H_r$ versus $\phi_H$ for films with different thickness. Open circles are measurement points and the solid line is the fit to equation (9).

Regarding dynamic characterization, raw data obtained from cavity FMR measurements were first fitted to equation (8) in order to extract $H_r$ and $\Delta H$ values. Then the $H_r$ values were plotted versus angle, which is defined as the orientation of the external DC magnetic field with respect to the [100] direc-
tion of the silicon substrate. This plot was fitted to equation (9), as given in figure 12, in order to extract the $M_{eff}$, uniaxial and cubic anisotropy fields. For this fitting, the $g$ value was kept constant at 2.05 and $\mu_0M_{eff}$ was found to be in the range 1.03 to 1.10T, with a small variation depending on the film thickness. Extracted $\mu_0M_{eff}$ values are comparable with the $M_s$ values obtained from magnetometry measurements, implying that the perpendicular anisotropy is negligible for the investigated films. In addition, the extracted $\mu_0H_u$ indicates a decreasing trend with increasing film thickness, with extracted values in the range of 1.85 to 3.25mT. The cubic anisotropy field values are less than one tenth of $H_u$ values proving that uniaxial anisotropy is dominant in these films. When it comes to $\mu_0\Delta H$ values extracted from cavity FMR measurements, it should be mentioned that the linewidth includes contributions from sample inhomogeneity, spin pumping, film mosaicity, TMS and intrinsic Gilbert damping. As explained in detail in paper III, TMS contributions appear when the FMR measurement is performed in in-plane geometry as was done here and it is an angle dependent contribution. Details of the origin of TMS is explained in paper III and a thickness dependent TMS contributions to $\Delta H$ were extracted in the same paper by fitting the angle dependent $\Delta H$ values to equations describing the linewidth contribution from TMS. Extracted TMS contributions vary from 2.6 mT to 4.5 mT for films with thickness in the range from 20 to 8 nm.

Figure 13. a) $\mu_0\Delta H$ versus frequency for films with different thickness. Solid lines are fits for equation (11). b) $\alpha_{eff}$ versus film thickness. (where $t_{CFA}$ is the Co$_2$FeAl thickness)

In addition to cavity measurements, broadband in-plane FMR measurements were done by applying the magnetic field along the easy axis of the films and the obtained raw data were fitted to equation (8) to extract $H_r$ and $\Delta H$ values. When $H_r$ values were fitted to equation (9), by setting $g$ to 2.05, the extracted $\mu_0M_{eff}$ values show a small variation with thickness in the range from 1.10 to 1.15 T. When $g$ is used as a free parameter it takes a value of about 2.10, where $\mu_0M_{eff}$ slightly decreases to the range from 1.00 to 1.05
The extracted $\mu_0\Delta H$ values were plotted versus frequency, as given in figure 13(a), and fitted to equation (11); extracted $\alpha_{eff}$ values are given in figure 13(b) versus film thickness. It should be mentioned that the extracted $\alpha_{eff}$ from in-plane measurements includes contributions from both spin pumping towards the Co$_2$FeAl/Ta interface and TMS. It was observed that $\alpha_{eff}$ decreases with increasing film thickness, and the inhomogeneous linewidth broadening varied between 1.2 and 2.5 mT, where the smaller values were observed for films with a thickness less than 12 nm. Apart from in-plane geometry measurements, broadband FMR measurements were done with out-of-plane geometry. The data obtained from these measurements were fitted to equation set (12) to extract $H_r$ and $\Delta H$ values. Plots of frequency versus $\mu_0H_r$ and $\mu_0\Delta H$ versus frequency are given in figure 14 (a) and (b), respectively. The frequency versus $\mu_0H_r$ plot was fitted to equation (13) as given in figure 14(a) in order to extract $\mu_0M_{eff}$ and $g$. The $\mu_0\Delta H$ versus frequency plot was fitted to equation (11) as given in figure 14(b) in order to extract $\alpha_{eff}$. $\mu_0M_{eff}$ and $g$ values vary in the range from 1.15 to 1.20 T and 2.07 to 2.13, respectively. Extracted $\alpha_{eff}$ values are given in figure 15(a), which follows an increasing trend with decreasing thickness of films as obtained from in-plane FMR measurements. Here, the obtained $\alpha_{eff}$ includes contributions both from intrinsic Gilbert damping $\alpha_G$ and extrinsic spin pumping $\alpha_{SP}$, and can thus be written as $\alpha_{eff} = \alpha_G + \alpha_{SP}$. The spin-pumping mechanism towards the nonmagnetic layer is restricted to metals with a ratio of the spin conserved to spin-flip scattering times (spin flip probability) $\epsilon = \tau_{el}/\tau_{SP} = (\lambda_{el}/\lambda_{SP})^2/3 \gtrsim 10^{-3}$, where $\lambda_{el}$ and $\lambda_{SP}$ are the mean free path and spin-diffusion length, respectively. $\epsilon \gtrsim 10^{-2}$ should be fulfilled for a nonmagnetic material in order to be an efficient spin sink. For a ferromagnet/Ta interface, derived values for $\lambda_{el}$ and $\lambda_{SP}$ are 0.5 nm and 2.5 nm, respectively. If spin-flip probability is calculated by using these values, $\epsilon = 1.3 \times 10^{-2}$ is obtained. This value of $\epsilon$ indicates that Ta can act as an efficient spin sink in our system.

Figure 14. a) frequency versus $H_r$ and b) $\Delta H$ versus frequency for films with different thicknesses. Symbols are measurement points and solid lines are fits to equation (13) and equation (11), respectively.
Now equation (16) should be used in order to extract the contribution of spin pumping to the total magnetic relaxation. XRR measurements revealed that there is a Ta₂O₅ layer on top of Ta capping and both of these layers are in close proximity to the Co₂FeAl layer, which means that $g_{\text{eff}}^{\uparrow\downarrow}$ will be a function of the conductance at both interfaces. Extracted $\alpha_{\text{eff}}$ values from out-of-plane measurements are given in figure 15(a) and this plot was fitted to equation (16) in order to obtain $g_{\text{eff}}^{\uparrow\downarrow}$ and a value of $g_{\text{eff}}^{\uparrow\downarrow} = (2.90 \pm 0.1) \times 10^{19}$ m² was extracted, which is comparable to the value found for the Pd/CoFe/Pd structure.⁷⁹ Since $g_{\text{eff}}^{\uparrow\downarrow}$ is obtained it is possible to calculate $\alpha_{\text{SP}}$ and extract $\alpha_G$ from the effective damping parameter. A value of $\alpha_G = 1.1(\pm 0.2) \times 10^{-3}$ was obtained for $\alpha_G$, which is in agreement with the values determined in previous studies (paper I and IV). $\alpha_{\text{eff}}$ values are given together with $\alpha_{\text{SP}}$ and $\alpha_G$ in figure 15(b).

Figure 15. a) $\alpha_{\text{eff}}$ versus $1/t_{\text{CFA}}$ (where $t_{\text{CFA}}$ is the Co₂FeAl thickness), where symbols are measurement results and the solid line is the fit to equation (16). b) $\alpha_{\text{eff}}$, $\alpha_{\text{SP}}$ and $\alpha_G$ values versus $t_{\text{CFA}}$.

After investigating the Co₂FeAl in terms of thickness-dependent relaxation, further investigation was carried out on temperature-dependent relaxation. Since sample deposition was optimized in the first part of the study, the same ion beam deposition technique was used with the same parameters and films were deposited at 573, 673 and 773 K (which are labeled as LP573K, LP673K and LP773K in the figures, respectively), with a stacking of 50nm Co₂FeAl layer on top of a Si(100) substrate and capped with a 4nm thick Al layer. An Al₂O₃ layer with a thickness of 1.5 nm was formed over the Al layer. In addition to these samples, which were optimized to yield the $B_2$ phase or a mixed phase of $B_2$ and $L_21$, a film with $A_2$ structure was also deposited at 300K (labeled as HP300K in the figures) with 100 W ion-source power and capped with a Ta layer, as explained in previous studies.⁶³ Before
performing dynamic magnetization experiments, the Co$_2$FeAl structure was investigated by our collaborators in terms of theoretical calculations, and theoretically expected magnetic properties, including the temperature dependent Gilbert damping parameter, were provided by them. During this part of the study we had a chance to compare the theoretical values with the experimental results that were obtained. First of all, in-plane cavity FMR measurements were performed and the collected data were fitted to equation (9) yielding values of $H_u$ and $M_{eff}$. Cubic anisotropy fields were extracted as well, and for all samples they were less than 0.22 mT. $H_u$ is 3.12, 1.56, 1.97 and 1.78 mT for samples deposited as 300, 573, 673 and 773 K, respectively. $g$ was extracted as 2.01, 2.06, 2.05 and 2.05 for samples deposited at 300, 573, 673 and 773 K, respectively. After cavity measurements, out-of-plane broadband FMR measurements were carried out versus temperature in the 50 to 300 K range. As before, the FMR measurements were performed in field sweep mode. The collected data were fitted to equation set (12) and as a result $H_r$ and $\Delta H$ were extracted. Frequency versus $\mu_0 H_r$ plots at different temperatures are given in figure 16. All these plots were fitted to the out-of-plane Kittel equation, as given in paper IV and, as a result of this fitting, temperature dependent $\mu_0 M_{eff}$ values were extracted, as given as insets in figure 16. Extracted $\mu_0 M_{eff}$ results are close to the $\mu_0 M_s$ values (given in paper IV) obtained from magnetometer measurements, which evidences that the perpendicular anisotropy is negligible in these films. The extracted out-of-plane $g$ factor is equal to 2.0 for all samples, which indicates that $g$ factor dependent changes in Gilbert damping is negligible. Discrepancy between $g$ factors extracted from in-plane and out-of-plane FMR measurements can be due to the limited frequency range of the out-of-plane measurements.
Figure 16. Frequency versus $\mu_0 H_r$ for samples deposited at different temperatures. Temperature dependent $\mu_0 M_{eff}$ and $\mu_0 \Delta H_0$ are given as insets (taken from paper IV)

Extracted $\mu_0 \Delta H$ values were plotted versus frequency and fitted to equation (11), as given in figure 17, and temperature-dependent $\alpha$ values were extracted, as given as inset in the same figure. The extracted $\alpha$ values include contributions from intrinsic effects as well as extrinsic effects, such as radiative damping $\alpha_{rad}$ and eddy-current damping $\alpha_{eddy}$. Here TMS is out of our concern since the measurement was carried out out-of-plane. Regarding the contribution from interfacial spin pumping, since the films are capped with Al and since the spin-pumping contribution in low spin-orbit coupling materials with a thickness less than the spin diffusion length is negligible, no subtraction is done in terms of the spin-pumping contribution in Al capped samples. However, the film deposited at 300K is capped with Ta and a spin pumping enhanced $\alpha$ is expected in this sample. To subtract this contribution, the $\alpha$ value of a film with capping was compared with $\alpha$ of a film without capping and a difference of $1 \times 10^{-3}$ was obtained. To obtain the intrinsic Gilbert damping, $\alpha_{int}$, all extrinsic contributions should be subtracted from the total damping parameter.
Figure 17. $\mu_0 \Delta H$ versus frequency for samples deposited at different temperatures. $\alpha$ versus temperature is given as inset for each sample (taken from paper IV)

$\alpha_{rad}$ can be obtained in two possible ways, either by using equation (15), or by experimental techniques. $\alpha_{rad}$ values extracted by using equation (15) are given in figure 18. When it comes to obtaining $\alpha_{rad}$ experimentally, a 200µm thick glass spacer was placed between the film and the CPW and the radiative damping was in this way avoided as explained in the literature.\textsuperscript{36} By using this method $\alpha_{rad}$ can be obtained as the difference between the damping parameters obtained from measurements done without and with a spacer. The experimentally obtained $\alpha_{rad}$ value for a sample deposited at 773K is $0.79(\pm0.22) \times 10^{-3}$, which matches well with the calculated value of $0.78 \times 10^{-3}$ by using equation (15).
Figure 18. a) $\mu_0 \Delta H$ versus frequency for film deposited at 773 K. Measurements done with and without placing a glass spacer between the film and the CPW. Red lines are fits to equation (11). b) Temperature dependent $\alpha_{rad}$ values extracted by using equation (15), the lines are a guide to the eye. c) $\alpha - \alpha_{rad}$ versus $\delta^2$, the red line is fit to equation (14). d) Temperature dependent $\alpha_{eddy}$ extracted by using equation (14), lines are a guide to the eye. (taken from paper IV). 40

Frequency dependent $\mu_0 \Delta H$ values for measurements with and without spacer, and extracted temperature dependent $\alpha_{rad}$ values from equation (15) are given in figure 18. Another extrinsic contribution $\alpha_{eddy}$ can be calculated by the use of equation (14). In order to use this equation, $\alpha_{rad}$ should be subtracted from the total $\alpha$ and then the remaining part of the $\alpha$ value should be plotted with respect to square of film thickness. This plot should then be fitted to equation (14) in order to obtain the $C$ value in equation (14). This fit is given in figure 18 for a film deposited at 573K. When $C$ has been obtained it is possible to calculate $\alpha_{eddy}$ from equation (14). Extracted temperature dependent $\alpha_{eddy}$ for all samples is given in figure 18.
As seen in figure 19(b), the theoretical calculations revealed that the Gilbert damping parameter increases by decreasing temperature for L\textsubscript{21} phase, whereas it decreases with decreasing temperature for the B\textsubscript{2} and A\textsubscript{2} structures. It can be observed that the film deposited at 673K shows a Gilbert damping increasing with decreasing temperature, which can be taken as evidence of a fully ordered L\textsubscript{21} structure.

In theory, temperature-dependent Gilbert damping has two contributions.\textsuperscript{80,81} The first one is intraband scattering and in this mechanism the band index number is always conserved and it is linearly dependent on the electron lifetime. It is a conductivity like scattering and in the low temperature regime this term increases rapidly. The other contribution is coming from interband scattering. This is a resistivity like scattering and has an inverse dependence on electron lifetime. It increases with increasing temperature. For the L\textsubscript{21} phase a non-monotonic temperature dependence of Gilbert damping is expected due to a contribution which is the sum of both intraband and interband electron scatterings, whereas only interband scattering is expected for the A\textsubscript{2} and B\textsubscript{2} phases, which results in a monotonic increase of Gilbert damping with increasing temperature.

As a final part of the studies on Co\textsubscript{2}FeAl, films with PMA were investigated. First magnetometry measurements were done with the SQUID magnetometer. Figure 20(a) shows the normalized in-plane and out-of-plane magnetization versus magnetic field results for the sample with 1 nm nominal Co\textsubscript{2}FeAl thickness, clearly indicating that the Co\textsubscript{2}FeAl film exhibits interface PMA. The saturation field for the in-plane magnetization is 0.2 T at 150 K, while the switching of the magnetization for the out-of-plane case is very sharp. However, as shown in figure 20(b), the switching field for the out-of-plane magnetization is strongly temperature dependent, indicating a likewise strong temperature dependence for the PMA. It can be seen from figure 20(b) that coercivity decreases by increasing temperature.
To investigate the temperature dependence of the PMA, we performed out-of-plane angle dependent FMR measurements at different temperatures using the cavity FMR setup. Using the approach described by Fu et al.,\textsuperscript{82} PMA can be described by first and second order uniaxial anisotropy terms, so that the energy density in a magnetic field can be written as,

\[ F = -\mu_0 H M_S \cos(\theta_M - \theta_H) + B_1 M_S \frac{\sin^2(\theta_M)}{2} + B_2 M_S \frac{\sin^4(\theta_M)}{4}, \tag{18} \]

where \( \theta_M \) is the out-of-plane angle of the magnetization vector, \( \theta_H \) is the out-of-plane angle of the magnetic field and \( B_i \) are the anisotropy fields; \( B_1 = \mu_0 M_s + 2K_1/M_s \) and \( B_2 = 4K_2/M_s \). The angular dependence of the resonance field using \( F \) and the resonance condition derived from the LLG equation can be written as,

\[ 2\pi f = \gamma \sqrt{W_X W_Y}, \tag{19} \]

where \( f \) is the cavity resonance frequency and \( \gamma = \frac{g\mu_B}{h} \), and \( W_X \) and \( W_Y \) are stiffness fields defined by

\[ W_X = \mu_0 H_r \cos(\theta_M - \theta_H) - B_1 \sin^2(\theta_M) - B_2 \sin^4(\theta_M) \quad \text{and} \quad \tag{20} \]

\[ W_Y = \mu_0 H_r \cos(\theta_M - \theta_H) + B_1 \cos(2 \theta_M) + B_2 (3 \sin^2(\theta_M) \cos^2(\theta_M) - \sin^4(\theta_M)), \tag{21} \]

The equilibrium angle of the magnetization is calculated by minimizing \( F \) with respect to \( \theta_M \), yielding

\[ -2\mu_0 H \sin(\theta_M - \theta_H) = \sin(2\theta_M) (B_1 + B_2 \sin^2(\theta_M)) \tag{22} \]
Using equations (20)-(22) to numerically fit the experimental results for $H_r$ versus $\theta_H$, it is possible to extract the values for the anisotropy fields. Perpendicular magnetic anisotropy is obtained when $B_1 < 0$ and $|B_1| > |B_2|$, while easy-cone anisotropy will occur when $B_1 < 0$ and $|B_1| < |B_2|$

Figure 21 shows $\mu_0 H_r$ versus $\theta_H$ for the Si/Ta (10nm)/Co$_2$FeAl (1nm)/MgO (2nm)/Cr (2nm) and Si/Ta (10nm)/Co$_2$FeAl (1.8nm)/MgO (2nm)/Cr (2nm) samples together with fits using equations (20)-(22). Together with the results for $B_1$ and $B_2$ versus temperature shown in figure 22, all samples with Co$_2$FeAl thickness in the range $1 \leq t \leq 1.8$ nm exhibit PMA in the investigated temperature range. It can, however, be concluded from these results that the $t = 1.8$ nm sample approaches an easy-cone anisotropy at the highest temperature.

The results presented here for ultrathin Co$_2$FeAl layers represent ongoing work that will be completed once the study has been extended to include thicker Co$_2$FeAl layers.

Figure 21. Resonance field versus out-of-plane angle of the magnetic field. a) $\mu_0 H_r$ versus $\theta_H$ for Si/Ta (10nm)/Co$_2$FeAl (1nm)/MgO (2nm)/Cr (2nm) and b) $\mu_0 H_r$ versus $\theta_H$ for Si/Ta (10nm)/Co$_2$FeAl (1.8nm)/MgO (2nm)/Cr (2nm).
Figure 22. Anisotropy fields versus temperature for Si/Ta (10nm)/Co₂FeAl (t)/MgO (2nm)/Cr (2-3nm) samples with nominal Co₂FeAl thicknesses 1 nm, 1.4 nm and 1.8 nm. a) $B_1$ and b) $B_2$ versus temperature. Lines are a guide to the eye.
5. Dynamic Properties of Fe$\text{65Co}_{35}$ Thin Films

As mentioned in the introduction of section 4, in addition to Co$_2$FeAl alloys, Fe$_{65}$Co$_{35}$ alloys were also investigated during this thesis study, in particular for their magnetization dynamics properties.

5.1. Material Properties

Soft magnetic alloys are of interest due to their possible application in devices such as spin valves, MTJs, spin injectors and magnetic recording write heads. The performance of such spintronic and memory devices is highly dependent on the magnetic damping parameter of the alloy used in device fabrication. In some cases, when low critical switching current is desired, a low damping parameter is necessary, whereas it is the opposite for a device which requires fast magnetic switching. In the latter case higher magnetic damping is essential. It is known that the thermal noise limit of the write head is also related to Gilbert damping. The thermal noise limit is simply given by the probability of random magnetic fluctuations occurring when the head element size is reduced, and it provides a limiting factor for the recording density. When it comes to magnetic recording or producing high frequency spintronic materials, it is favorable to have high $M_s$, high permeability, thermal stability and high resistivity. FeCo alloys are promising in this case since they possess all of these properties. Today, Fe$_{1-x}$Co$_x$ alloys, where 0.3<$x$<0.4, are known to have the highest $M_s$ values. Among the FeCo compositions, extra attention was paid to Fe$_{65}$Co$_{35}$, which has an $M_s$ value of 2.39T. Although it has favorable properties, one drawback of Fe$_{65}$Co$_{35}$ can be its high $H_c$ value. As shown in previous studies and confirmed by our own work presented in paper V, this problem can be solved by adding a seed layer between the substrate and the Fe$_{65}$Co$_{35}$ film. Adding a seed layer between Fe$_{65}$Co$_{35}$ and the substrate not only affects the static magnetic properties, but also the dynamics of the magnetization. The effect of the seed layer on the dynamic properties should be understood in order to be able to tailor the properties to make the magnetic film suitable for spintronic device applications. As mentioned above, a low damping parameter is not desirable in all spintronic devices and memory storage applications. Depending on the purpose, a high damping parameter could be required in order to produce devices where high switching speed is...
essential. In the literature FeCo alloys have so far been doped with \(\text{Yb}^{101}\), \(\text{Dy}^{102}\), \(\text{Gd}^{103}\) and \(\text{Si}^{104}\), and in all cases the damping parameter increased. Apart from doping, an ultrathin layer of a rare earth element, with large orbital moments, deposited next to NiFe or CoFe layers has also been shown to increase the damping parameter.\(^{105}\)

In the first part of the study, as explained in detail in paper V, Fe\(_{65}\)Co\(_{35}\) films with a nominal thickness of 30 nm were deposited on Si(100) substrates with a native oxide layer by using DC magnetron sputtering. Thereafter, Fe\(_{65}\)Co\(_{35}\) films were deposited on Si/SiO\(_2\) substrates using seed layers of Ru, Ni\(_{82.5}\)Fe\(_{17.5}\), Rh, Y and Zr, where the nominal thickness was 1nm for all seed layers. None of the films were capped and all films were deposited at room temperature. Films were deposited on two separate substrate wafers where one wafer was annealed at 200°C for 3 hours. Magnetometric characterization was performed using the looper in order to analyse the effect of seed layer on the \(H_c\) values of the films. It was observed that magnetically softer films were obtained by the addition of Ru, Ni\(_{82.5}\)Fe\(_{17.5}\) and Rh as seed layer, while the \(H_c\) values obtained using Y and Zr as seed layer were higher than these. It was therefore decided to continue with further structural and magnetic characterization of the films using Ru, Ni\(_{82.5}\)Fe\(_{17.5}\) and Rh as seed layers. Structural characterization was done using techniques such as XRD, XRR and transmission electron microscopy (TEM) in order to understand the effect of the seed layers on the crystalline structure of the Fe\(_{65}\)Co\(_{35}\) films. After structural characterization, the magnetization dynamics was characterized by using cavity FMR, in-plane and out-of-plane broadband FMR techniques.

In the second part of the study as explained in detail in paper VI, Fe\(_{65}\)Co\(_{35}\) was deposited on Si/SiO\(_2\) substrates with an addition of a Ru seed layer by DC magnetron sputtering. All films were capped with a Ru layer. The nominal thickness of both Ru seed and cap layer was 3nm. The films Fe\(_{65}\)Co\(_{35}\) were deposited as two batches with nominal thickness of 20 and 40 nm. In addition, the Fe\(_{65}\)Co\(_{35}\) layers were doped with Re by co-sputtering from a Re target, varying the nominal dopant concentration from 0 to 10.23 at%. Rutherford back scattering (RBS) measurements were performed in order to determine the exact Re concentration within the films. XRR and XRD analysis were done as well to understand the crystalline structure, effect of the Re dopant on the structure and to determine the thickness and roughness of the layers in the stack. After structural characterization, static magnetic properties were characterized by a SQUID magnetometer and dynamic magnetic properties were characterized by cavity FMR, in-plane and out-of-plane broadband FMR techniques. Beside experimental studies, theoretical calculations were also done to investigate the effect of Re doping on structural as well as magnetic properties, and a systematic study which compares theoretical and experimental results was performed.
5.2. Analysis and Results

In the first part of the study, static magnetic properties were investigated by the looper, followed by structural characterization. The looper results are given later in this chapter, but first the structural properties are explained. GIXRD scans were done on Fe$_{65}$Co$_{35}$ films deposited with Ru, Ni$_{82.5}$Fe$_{17.5}$ and Rh seed layers and without a seed layer. Measured GIXRD spectra are given in figure 23(a).

![Figure 23](image)

*Figure 23.* a) XRD 0-20 scans for films deposited with and without seed layers. a.p. refers to as prepared samples and an. to annealed samples. b) Crystallite size of the films deposited with and without seed layer. Red circles show the results for annealed films and black squares show results for as prepared samples. Lines are a guide to the eye.

XRD scans revealed that all films have a bcc structure and there was no indication of impurity phases or the formation of new phases induced by annealing. The crystallite size of the films was calculated using the Scherrer equation and calculated values are given in figure 23(b). Adding a seed layer decreased the crystallite size, consistent with results from former studies, and as expected, annealing resulted in increased crystallite size. The crystallite size of the film without a seed layer is about 18 nm, whereas the size is around 14 nm for films with seed layer. After annealing, the crystallite size increased to 16 nm for films with a seed layer. The lattice parameter did not vary dramatically by the addition of a seed layer or annealing the films and the lattice parameter for all films is around 0.285 nm.

In addition to XRD, XRR measurements were done in order to determine the thickness and roughness of the layers in the film stack. Detailed results of this analysis are given in paper V. The surface roughness of the Fe$_{65}$Co$_{35}$ films, both with and without a seed layer, varied in the range of 0.8 ($\pm 0.043$) to 1.3 ($\pm 0.1$) nm. The roughness between SiO$_2$ and the Fe$_{65}$Co$_{35}$ layer for the film deposited without a seed layer and the roughness between SiO$_2$ and the seed layer for films deposited with a seed layer varied in the range of 0.3 ($\pm 0.006$) to 0.8 ($\pm 0.11$) nm, where the interface is rougher for the sample deposited without a seed layer. The interface roughness between seed and Fe$_{65}$Co$_{35}$ layers for as deposited films are 0.26($\pm 0.01$) and 0.31($\pm 0.004$) nm.
for Ru and Rh seed layers, respectively. The XRR results for the Ni$_{82.5}$Fe$_{17.5}$ seed layer was not reliable due to the fact that the electron densities of CoFe and NiFe structures are very similar and hence the X-ray contrast between these structures is very low, which in turn leads to high uncertainty in the results. Since there is a small variation in the roughness values of the surfaces/interfaces between the samples, the variation in magnetic properties can not be traced back to the roughness values of the film stacks. As mentioned above the films were not capped with a protective layer and as a result an oxide layer was observed on top of the films as confirmed by the XRR analyses.

As well as XRD and XRR, further structural characterization was done by high resolution TEM (HRTEM) imaging. HRTEM images of the films with different seed layers and without seed layer both for as prepared and annealed samples are given in figure 24.

*Figure 24.* HRTEM images for Fe$_{65}$Co$_{35}$ films with different seed layers and their corresponding Fast Fourier Transforms (FFT) as insets. The white arrows indicate the growth direction from the SiO$_2$ substrate (arrow’s end) to the surface of the films (arrow’s head) where a thin oxide layer is found. In the case of the films with Ni$_{82.5}$Fe$_{17.5}$ as seed this thin layer could not be imaged due to the fact that the atomic masses of Ni, Fe and Co are very close so no mass contrast is visible in the TEM.

FFT images reveal that all films have a polycrystalline structure. Regarding the morphology of the films, it was observed that there is a preferential columnar growth for the films deposited on seed layers, whereas for the film deposited without a seed layer, crystallites have large width along the film lateral direction. The crystallite width varies in the range of 28 to 50 nm for the film deposited without a seed layer, whereas it varies in the range of 5 to 21 nm for the as deposited films with a seed layer. Detailed numerical values for crystallite widths and thickness of films with and without seed layers and as prepared and annealed films are given in paper V. It was observed that,
when it comes to film thickness, there is a discrepancy between the TEM and XRR results. However, this was expected, since TEM cannot distinguish the surface roughness of the films precisely and gives thickness values corresponding to the sum of the surface roughness and film thickness. Hence, thickness values coming from TEM imaging were always larger than values extracted from the XRR results.

In addition to films deposited on different seed layers, films deposited on a Ru seed layer with different Re dopant concentrations were characterized in terms of their compositional and structural properties. In order to determine the exact composition and amount of Re in the alloy structure, RBS measurements were done on doped films with 20nm nominal thickness. These measurements revealed that the different films were doped with Re concentrations of 3.0±0.1 at%, 6.6±0.3 at% and 12.6±0.5 at%. In addition, depending on the results of the Fe and Co atomic fractions within the films, it can be concluded that there is no preferential replacement with Re and the two elements are replaced according to their respective concentration. GIXRD spectra of the films and extracted lattice parameters from GIXRD results and calculated theoretical lattice parameters are given for comparison in figure 25.

As was shown in the inset of figure 25(a), a systematic shift depending on the Re concentration was observed for the (110) peak and for other peaks as well. This systematic shift of the lattice parameter can be accepted as experimental evidence of an increasing amount of Re dopant within the alloy structure. The peaks were shifted to lower 2θ values, hence the lattice parameter increases with increasing Re concentration. As can be seen in figure 25(b), the estimated theoretical lattice parameters are larger than the experimentally obtained values. This is not unexpected since the approximation used in the theoretical calculations for the exchange-correlation potential has a tendency to overestimate the lattice parameter. In addition to

Figure 25. a) GIXRD spectra for Fe₆₅Co₃₅ films with different Re concentration. The inset shows the shift of the (110) peak with Re concentration. b) Extracted lattice parameters from GIXRD versus Re concentration and the corresponding theoretical values calculated for Re concentrations in the range 0-10 at%.
GIXRD, XRR measurements were also performed on these films; however surface roughness of Fe₆₅Co₃₅ is less than 1nm which is not expected to affect the magnetic properties much.

After the structural properties were investigated, magnetic characterization was done starting with the seed layer films. First of all, looper measurements were performed on as prepared Fe₆₅Co₃₅ films deposited on Ru, Ni₈₂.₅Fe₁₇.₅, Rh, Y and Zr seed layers and on the film deposited without a seed layer.

Normalized hysteresis curves of films with Ru, Ni₈₂.₅Fe₁₇.₅ and Rh seed layers and the film without a seed layer are given in figure 26.

![Figure 26. a) Normalized hysteresis curves for Fe₆₅Co₃₅ films deposited on Ru, Ni₈₂.₅Fe₁₇.₅ and Rh seed layers. b) Comparison of hysteresis curves of Fe₆₅Co₃₅ films deposited on a Ru seed layer and deposited without a seed layer.](image)

As can be clearly observed from figure 26(b), the hysteresis curve of Fe₆₅Co₃₅ deposited without a seed layer is more rounded when compared with the one deposited on the Ru seed layer. Beside its rounded shape the $\mu_0 H_C$ value extracted from this curve was around 12.5 mT, which is very large when compared with the $\mu_0 H_C$ of the films deposited on different seed layers. As can be seen in figure 26(a), Fe₆₅Co₃₅ films deposited on Ru, Ni₈₂.₅Fe₁₇.₅ and Rh seed layers have more square shaped hysteresis curves with $\mu_0 H_C$ values around 1.25 mT. When it comes to saturation fields it can be observed from the figures that films with seed layers saturate at applied fields of 2 mT whereas the film deposited without a seed layer saturates at about 50mT, which gives evidence that films with seed layers have better aligned easy axes than the film deposited without a seed layer. The $\mu_0 H_C$ values extracted from looper measurements for films both with and without seed layer are given in figure 27 for comparison. The $\mu_0 H_C$ values obtained for Y and Zr seeds are 6.5 and 8.5mT, respectively. These values decreased by annealing but they still cannot compete with the values obtained with Ru, Ni₈₂.₅Fe₁₇.₅ and Rh as seed layer. Annealing did not much affect the $\mu_0 H_C$ values of the films with Ru, Ni₈₂.₅Fe₁₇.₅ and Rh as seed. Since low coercivity is essential for memory applications, further characterization both in terms of
structure and magnetic properties were performed on films deposited with Ru, Ni$_{82.5}$Fe$_{17.5}$ and Rh as seed layer. For comparison purposes, the film deposited without seed was also included in the continued study.

![Graph](image)

**Figure 27.** $\mu_0 H_c$ values from looper measurements for Fe$_{65}$Co$_{35}$ films deposited with and without a seed layer. Black squares show results for as prepared samples (including sample with no seed) and red circles show results for annealed samples. Lines are a guide to the eye.

The random anisotropy model can be helpful in terms of explaining the correlation between the decreased crystallite size and the decreased $H_c$ values. This model predicts that coercivity should be proportional to $D^6$, where $D$ is the crystallite size, when $D$ is smaller than the exchange length of the material, which is the case for Fe$_{65}$Co$_{35}$ films in the present situation. However, the annealed films do not follow this model. We expected to find higher coercivity values for annealed samples since their crystallite sizes were larger than for as prepared films, but according to experimental observations, the coercivity of the annealed films was even slightly smaller than as prepared films. This result indicates that beside the crystallite size, the coercivity values could be dependent on the structure and homogeneity of the films.

Having characterized the static magnetic properties of the Fe$_{65}$Co$_{35}$ films, we continued with dynamic characterization including cavity FMR, in-plane and out-of-plane broadband FMR measurements. Before discussing the results from different measurement techniques, it should be mentioned that the damping parameter could not be extracted for the film without a seed layer. Using the cavity FMR setup, the obtained $\Delta H$ value for the film without a seed layer was about 80 mT, whereas this value was about 3 mT for films deposited with a seed layer. Since $\Delta H$ was very large for the film without a
seed layer it was impossible to calculate the damping parameter. One possible explanation for such a case could be that the magnetic relaxation in the film without a seed layer is dominated by a large extrinsic contribution which screens out the intrinsic contribution to the damping.\(^{112}\)

First of all, cavity FMR measurements were done and FMR spectra were recorded for all as prepared and annealed films deposited on the seed layer. The recorded FMR spectra were fitted to equation (8) and from this fitting \(H_r\) and \(\Delta H\) values were extracted. The angle dependent \(H_r\) values (given in paper V) reveal a dominant twofold uniaxial anisotropy and the data were fitted to equation (9) to extract the uniaxial anisotropy field values. The uniaxial anisotropy field varies in the range of 2.62(±0.013) to 3.37(±0.018) mT for both as prepared and annealed films. All extracted uniaxial anisotropy values are given in paper V. After cavity FMR measurements, all films except the one without a seed layer were characterized by in-plane broadband FMR measurements. Again the recorded FMR spectra were fitted to the derivative function given in equation (8) and \(H_r\) and \(\Delta H\) values were extracted. The \(\Delta H\) values were fitted to equation (11) in order to extract \(\alpha\). Extracted \(\alpha\) values are given in paper V for all films, both for as prepared and annealed samples. It was observed that \(\alpha\) varies somewhat depending on the choice of seed layer material, but the more significant observation is that \(\alpha\), except for the Ru seed layer, decreases for annealed samples. After in-plane FMR measurements, out-of-plane measurements were done and recorded FMR spectra were fitted to equation set (12) in order to extract \(H_r\) and \(\Delta H\). Both of these values were plotted with respect to frequency and the data were fitted to equation (13) and (11), respectively, in order to extract the \(M_{eff}\), \(\alpha\), and \(g\) values. All obtained values are given in detail in paper V. Extracted \(M_{eff}\) values are all about 2.32 T, which is comparable to the \(M_s\) value of the Fe\(_{65}\)Co\(_{35}\) composition\(^{100}\) and implies that there is no significant contribution from perpendicular magnetic anisotropy in these films.

Comparing the \(\alpha\) values extracted from the in-plane and out-of-plane broadband FMR measurements, it was observed that \(\alpha\) values coming from in-plane measurements are 50-80% larger than the values extracted from out-of-plane results. This observation was expected due to the TMS contribution in in-plane measurements. However, the frequency dependent \(\Delta H\) plots for in-plane measurements depict a linear behavior which is unexpected for the case where frequency dependent TMS contributes to the magnetic relaxation. This can be explained by the limited frequency range in the measurements, which makes it difficult to resolve any deviation from linear behavior. A minimum \(\alpha\) value of 2.1×10\(^{-3}\) was reported in a former study, where a broadband out-of-plane FMR geometry was used, for the specific composition of Fe\(_{75}\)Co\(_{25}\).\(^{39}\) In this study, an investigation over different compositions of Fe\(_{1-x}\)Co\(_x\) was also performed and it was reported that \(\alpha\) increased with an increased amount of Co. The \(\alpha\) value extracted in this thesis study for the Fe\(_{65}\)Co\(_{35}\) composition was about 4×10\(^{-3}\), which is consistent
with the trend reported in former work, although it is a little larger in comparison.

In the second part of the study related to the Fe$_{65}$Co$_{35}$ alloy, Re doped films were investigated in terms of both static and dynamic magnetic properties. First of all SQUID measurements were performed in order to obtain hysteresis curves of the films with different dopant concentration. Normalized hysteresis curves are given in figure 28(a).

![Figure 28](image.png)

**Figure 28.** a) Normalized room temperature hysteresis curves for Fe$_{65}$Co$_{35}$ films with different Re dopant concentration. b) Low temperature $M_s$ values calculated for different dopant concentrations and extracted values from out-of-plane FMR measurements. Lines are a guide to the eye.

The coercivity for all films was about 2 mT and apart from for the 12.6at% doped film, all films have a rectangular shaped hysteresis loop. Low $H_C$ values are expected for these films since there is a seed layer between the substrate and the actual film, as explained in the first part of this study. Theoretically calculated and experimentally extracted $M_s$ values with respect to the dopant concentration are shown in figure 28(b). As expected, saturation magnetization decreases with increasing Re concentration.

After the magnetometer measurements, dynamic characterization was performed using cavity and out-of-plane broadband FMR measurements. The out-of-plane FMR measurements were performed at different temperatures. The FMR spectra recorded by cavity FMR were fitted to equation (8) and $H_r$ and $\Delta H$ values were extracted from this fit. The angle dependent $H_r$ values were fitted to equation (9) and from this fit $H_u$ values varying between 2.10 to 2.30 mT were obtained. The results clearly reveal that there is a twofold in-plane uniaxial anisotropy and that the anisotropy field $H_u$ is independent of Re concentration. After cavity measurements, broadband out-of-plane measurements were done in the temperature range of 50 to 300 K. All recorded FMR spectra were fitted to equation set (12) to extract $H_r$ and $\Delta H$ values. $\alpha_{total}$ was obtained by fitting the $\Delta H$ versus frequency data to equation (11) and the $H_r$ versus frequency data were fitted to equation (13) to extract the $M_{eff}$ and $g$ values. The obtained $g$ values were 2.064 and 2.075
for the undoped and 12.6 at% doped films, respectively, and similar values were obtained for all temperatures in the studied temperature range. The extracted $M_{eff}$ values are assumed to be equivalent to $M_s$ since these films are thick enough to make out-of-plane magnetic anisotropy negligible. As expected, $M_{eff}$ decreases with increasing dopant concentration. Extracted $\alpha_{total}$ values include both intrinsic and extrinsic contributions to the magnetic relaxation. Since theoretical calculations estimate the intrinsic damping, extrinsic contributions should be subtracted from $\alpha_{total}$ in order to make the comparison of experimental and theoretical results possible. There could be different types of extrinsic contributions such as TMS, eddy-current damping, radiative damping and spin pumping. Since the measurements were performed with an out-of-plane FMR set-up, the TMS contribution is avoided. The spin pumping contribution is expected to be negligible since we have low spin orbit coupling Ru as the capping and seed layer, with a thickness less than the spin diffusion length. Then the only expected contributions are eddy-current damping $\alpha_{eddy}$ and radiative damping $\alpha_{rad}$, hence $\alpha_{total}$ can be written as $\alpha_{total} = \alpha_{red} + \alpha_{rad} + \alpha_{eddy}$, where $\alpha_{red}$ is the intrinsic damping parameter. First of all, the temperature dependent $\alpha_{rad}$ was calculated by using equation (15). All of the calculated $\alpha_{rad}$ results are given in paper VI. After having calculated $\alpha_{rad}$, $\alpha_{eddy}$ was calculated by using equation (14). All of the calculated temperature dependent $\alpha_{eddy}$ values are given in paper VI. After the calculation of $\alpha_{rad}$ and $\alpha_{eddy}$, these values were subtracted from the total damping to yield the intrinsic $\alpha_{red}$. Comparisons of $\alpha_{total}$ and $\alpha_{red}$ for all concentrations are given in a temperature dependent way in figure 29. From this figure it can be seen that the damping parameter increases with an increasing amount of Re dopant. Since Re has a large orbital moment and the damping parameter is dependent on spin orbit coupling, this effect of Re was expected. Experimental $\alpha_{total}$ and $\alpha_{red}$ values are compared with the theoretical intrinsic damping parameter in figure 30 for the undoped and 12.6 at% Re doped films. Both the theoretically calculated damping parameter and the experimentally determined intrinsic damping parameter decrease with decreasing temperature, indicating that the origin of the intrinsic damping is interband electron scattering. As can be seen from the comparison shown in figure 30 there is a discrepancy between the theoretical and experimental intrinsic damping parameters. One reason could be that theoretical values were calculated for the experimental nominal doping concentrations, which are slightly smaller than the real concentrations measured by RBS. However, this is not expected to have a drastic effect on the comparison. A more reasonable explanation could be the incompleteness of the model used to calculate the theoretical damping parameter. This model ignores a few complex scattering processes which can be the reason for the observed discrepancy between theory and experiment.
Figure 29. $\alpha_{\text{tot}}$ versus temperature for Fe$_{65}$Co$_{35}$ thin films with different concentration of Re. Besides showing $\alpha_{\text{tot}}$, intrinsic $\alpha_{\text{red}}$ values are also plotted, obtained by subtraction of radiative and eddy current damping contributions from $\alpha_{\text{tot}}$. Error bars are given for measured $\alpha_{\text{tot}}$ (same size as symbol size).

Figure 30. $\alpha_{\text{tot}}$ versus temperature for Fe$_{65}$Co$_{35}$ thin films with 0 at% and 12.6 at% concentration of Re. Besides showing $\alpha_{\text{tot}}$, reduced $\alpha_{\text{red}}$ values are also plotted obtained by subtraction of radiative and eddy current damping contributions from $\alpha_{\text{tot}}$. In addition to experimental results, theoretically calculated intrinsic damping parameters are given for similar concentrations of Re. Error bars are given for measured $\alpha_{\text{tot}}$ (same size as symbol size).
6. Concluding Remarks

6.1. Summary

The main purpose of this thesis work was to understand fundamental properties of the magnetization dynamics in thin films, in particular films of Co$_2$FeAl and Fe$_{65}$Co$_{35}$ alloys. In the first four papers detailed investigations of properties of the full Heusler alloy Co$_2$FeAl were performed. These alloys are promising materials due to their low magnetic damping parameters. In paper I, in order to optimize magnetic properties in general and in particular the damping parameter controlling the relaxation of the magnetization, the deposition parameters were optimized and films with B$_2$ crystalline phase were obtained without any post annealing process, yielding one of the lowest damping parameter reported so far for Co$_2$FeAl. However, magnetic relaxation in thin films is a complicated issue that depends both on the intrinsic properties of the material, scattering of magnons by material defects and extrinsic contributions depending on the interaction of the material with the experimental setup. After optimizing the deposition, in papers II and III, further investigations were done in order to disentangle the effects of different seed and capping layers on the relaxation of the magnetization. Individually in paper III, the effect of the Co$_2$FeAl thickness on the damping was investigated. The results presented in papers II and III are imported in terms of providing the information necessary to enable integration of Co$_2$FeAl films in spintronic devices. To be more specific, MTJ structures, which are the basis of MRAM devices, are composed of stackings of metallic ferromagnetic and insulating nonmagnetic layers, where the nonmagnetic layer affects the magnetic relaxation of the ferromagnetic layer. Therefore, investigating and gaining understanding of the effects of different nonmagnetic layers on the magnetic relaxation is quite important for the development of novel spintronic devices. In paper IV, more fundamental research was performed by calculating the temperature dependence of the damping parameter and comparing the theoretical results with the corresponding experimental results. This comparison gave us the opportunity to understand the fundamental principles behind the magnetic relaxation. In addition, the comparison of calculated and measured results show how the crystalline phase of a Co$_2$FeAl film can be identified from temperature dependent measurements of the magnetization dynamics.
In addition to Co$_2$FeAl, Fe$_{65}$Co$_{35}$ alloys were investigated in papers V and VI. This alloy composition is promising for magnetic recording devices. In paper V, the static magnetic properties were tailored by the addition of different seed layers in the growth process of the Fe$_{65}$Co$_{35}$ films and the effects of the seed layers on magnetization dynamics were investigated. In paper VI, Fe$_{65}$Co$_{35}$ was doped with Re in order to increase the damping parameter of the material and to make it a favorable material for high data rate recording applications. In this study, temperature dependent theoretical calculations were also performed. Both the experimental and theoretical results show that magnetization decreases with increasing dopant concentration, while the intrinsic damping parameter at the same time increases. Moreover, the trend with temperature for the damping parameter is the same in both results; the damping parameter decreases with decreasing temperature, giving evidence that the mechanism behind the magnetic relaxation is interband electron scattering.

6.2. Outlook

With its low intrinsic damping parameter, Co$_2$FeAl is a promising material for spintronic applications. During the investigations for this thesis special attention was paid to the effects of different nonmagnetic adjacent layers on the relaxation behavior of Co$_2$FeAl. Since this kind of ferromagnetic/nonmagnetic stacking constitutes the basis of MTJ systems and hence memory applications, different materials could also be investigated in terms of understanding the effects on the magnetic relaxation of the Co$_2$FeAl layer. Since it is necessary to tailor magnetic relaxation properties in order to produce energy efficient and fast spintronic devices, these studies and the fundamental understanding gained from them are crucial for the development of novel spintronic devices.

In addition to the effects of adjacent layers, Co$_2$FeAl films with a thickness of less than 2nm were investigated. However, this part of the study is still ongoing and will require further experimental work to be completed. The continued work should focus on Co$_2$FeAl thicknesses where the layers will exhibit easy-cone anisotropy. The experimental work should continue to use the cavity FMR setup, but efforts should also be made to improve the sensitivity of broadband FMR measurements in the PPMS system to enable measurements at different microwave frequencies. The possibility of achieving easy-cone anisotropy will be beneficial to spintronic devices where the switching of the magnetization is accomplished by a spin polarized current.

fokuserar mest på den magnetiska dämpningsparametern som påverkar mekanismen bakom vändningen av magnetiseringen i spintronik.

Figure 31. Arbetsprincipen för MRAM


I denna avhandling har två olika legeringar studerats, skråddarsytts och inneboende egenskaper har separerats från externa faktorer. Ett av materialen har varit en Heusler legering med sammansättningen Co2FeAl. I början av arbetet optimerades filmdeponeringsparametrarna hos materialet med syftet att få en så låg dämpningsparameter som möjligt, eftersom dämpningsparametern är starkt beroende av kristallstrukturen hos materialet. Vi lyckades få dämpningsparametrar som är jämförbara med de lägsta rapporterade i forskningslitteraturen för detta material. Efter detta studerades det icke-magnetiska lagrets påverkan på dämpningsparametern, detta då man i spintronik tillämpningar har använder flerlagerstrukturer där magnetiska och icke-magnetiska lager ingår. Vi analyserade och lyckades separera bidragen från
dessa lager till dämpningsparametern. Vi studerade dessutom inverkan av tjockleken hos Co2FeAl filmen på dämpningsparametern. Slutfilen studerade och jämförde vi temperaturberoendet hos dämpningsparametern hos materialet med teoretiska beräkningar. När de externa faktorerna togs bort fick vi en chans att förstå fundamentet hos dämpningsmekanismen i dessa legeringar.

Förutom Co2FeAl så har Fe65Co35 studerats i termer av dämpningsparametrar, detta då denna legering är populär i tillämpningar för magnetisk lagring. På liknande sätt som för Co2FeAl studerade vi det icke magnetiska lagrets inverkan på dämpningsparametern genom att välja olika icke magnetiska lager. Efteråt studerade vi tillsatsen av olika mängd Rhenium (Re) med syfte att öka dämpningsparametern. Studierna visade att tillsatt Re kan öka dämpningsparametern med ~230 % hos en Fe65Co35 film. Detta kan ha teknologiska tillämpningar då höga dämpningsparametrar ibland eftersträvas. Temperaturberoendet hos dämpningsparametern för Fe65Co35 studerades dessutom och genom att subtrahera de yttre faktorernas inverkan kunde vi få resultat som är i linje med de teoretiskt framtagna värdena.
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I will keep the rest of the text in Turkish in order to thank some special people from Turkey.

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