NEW SOFT MAGNETIC MATERIALS FOR
HIGH FREQUENCY APPLICATIONS

by

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ABSTRACT

Soft magnetic materials are widely used in devices such as inductors, transformers, antennas, magnetic hard drives, etc. Some of those devices will benefit greatly from operating at high frequencies. Thus fundamental study on finding the materials that have better soft magnetic properties is essential for improving the performance of those devices. Fe alloys have been proved to be promising candidates for high frequency applications. In this dissertation, an extensive study of magnetic properties of FeAl, (FeCo)-Al and (FeCo)-Si alloy thin films and their dependence on the film thickness and growth temperature has been presented. These films have body-centered cubic structure and columnar growth morphology. It is shown that the thickness of the film, which has an influence on the stress inside the film, may affect the coercivity through the magnetic-elastic coupling. The same mechanism is observed in the growth temperature dependence study, where reduced stress caused by increased growth temperature leads to a decrease in coercivity. The effective damping parameter shows a huge increase at small thickness due to the spin pumping effect. In-plane rotation ferromagnetic resonance measurements unveil the existence of four-fold anisotropy in (FeCo)-Si films. In addition, a four-fold symmetry is observed in the FMR linewidth vs. in-plane angle plot, which indicates anisotropic damping caused by the two-magnon scattering contribution. The film thickness dependence of FMR linewidth caused by the two-magnon scattering suggests that the origin of the two-magnon scattering is not pure interfacial.
DEDICATION

To my dear parents and grandmother.
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>$A$</td>
<td>Exchange constant</td>
</tr>
<tr>
<td>$AC$</td>
<td>Alternating current</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>Gyromagnetic ratio</td>
</tr>
<tr>
<td>bcc</td>
<td>Body-centered cubic</td>
</tr>
<tr>
<td>CPW</td>
<td>Coplanar waveguide</td>
</tr>
<tr>
<td>DAQ</td>
<td>Data acquisition</td>
</tr>
<tr>
<td>DC</td>
<td>Direct current</td>
</tr>
<tr>
<td>$\Delta H$</td>
<td>FMR spectrum peak to peak linewidth</td>
</tr>
<tr>
<td>$\Delta H_0$</td>
<td>Inhomogeneous broadening linewidth</td>
</tr>
<tr>
<td>EDX</td>
<td>Energy-dispersive X-ray spectroscopy</td>
</tr>
<tr>
<td>FMR</td>
<td>Ferromagnetic resonance</td>
</tr>
<tr>
<td>GPIB</td>
<td>General Purpose Interface Bus</td>
</tr>
<tr>
<td>$H_c$</td>
<td>Coercivity</td>
</tr>
<tr>
<td>$H_{\text{eff}}$</td>
<td>Effective field</td>
</tr>
<tr>
<td>$K_2$</td>
<td>Uniaxial anisotropy constant</td>
</tr>
<tr>
<td>$K_4$</td>
<td>Four-fold anisotropy constant</td>
</tr>
<tr>
<td>LL</td>
<td>Landau-Lifshitz</td>
</tr>
<tr>
<td>LLG</td>
<td>Landau-Lifshitz-Gilbert</td>
</tr>
<tr>
<td>MOKE</td>
<td>Magneto-optical Kerr effect</td>
</tr>
<tr>
<td>$M_s$</td>
<td>Saturation magnetization</td>
</tr>
<tr>
<td>PSSW</td>
<td>Perpendicular standing spin wave</td>
</tr>
<tr>
<td>$t$</td>
<td>Film thickness</td>
</tr>
<tr>
<td>TEM</td>
<td>Transmission electron microscopy</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Description</td>
</tr>
<tr>
<td>-------------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>VSM</td>
<td>Vibrating sample magnetometer</td>
</tr>
<tr>
<td>XPS</td>
<td>X-ray photoelectron spectroscopy</td>
</tr>
<tr>
<td>XRD</td>
<td>X-ray diffraction</td>
</tr>
<tr>
<td>XRR</td>
<td>X-ray reflectivity</td>
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1 INTRODUCTION

1.1 Magnetization dynamics

1.1.1 Equations of Motion

In the classical limit, the magnetization dynamics can be described by the Landau-Lifshitz (LL) equation of motion [1]:

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} - \frac{\lambda}{M_s^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}}), \quad (1.1)$$

where $\gamma$ is the gyromagnetic ratio, $\mathbf{M}$ is the magnetization vector, $M_s$ is the saturation magnetization, $\lambda$ is the LL damping parameter. The effective field $\mathbf{H}_{\text{eff}}$ is given by the negative derivative of the Gibbs free energy with respect to the magnetization vector:

$$\mathbf{H}_{\text{eff}} = -\frac{\partial \mathbf{E}}{\partial \mathbf{M}}. \quad (1.2)$$

The Gibbs free energy $\mathbf{E}$ includes the exchange energy, the Zeeman energy, the magnetic anisotropy energy, and demagnetization energy. The first term on the right hand side of Eq. (1.1) represents the precessional motion of magnetization under an external magnetic field, while the second term describes the LL damping torque that drags the magnetization towards the direction of the field.

An alternative form of the LL equation called the Landau-Lifshitz-Gilbert (LLG) equation was proposed by Gilbert in 1955 [2]:

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \frac{\alpha}{M_s} \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t}. \quad (1.3)$$
Here the damping term in the LL equation is replaced by the Gilbert damping term, in which a dimensionless damping parameter $\alpha$ is used to represent the strength of damping. The LL equation and the LLG equation are mathematically equivalent as pointed out by Gilbert himself [3]. The LL equation can be rewritten in the form of Eq. (1.3) by substituting the equality $M \times H_{\text{eff}} = -\gamma^{-1}[\partial M/\partial t + (\lambda/M_s^2)M \times (M \times H_{\text{eff}})]$ into the righthand side of Eq. (1.1) [3]. The relationship between the gyromagnetic ratio ($\gamma$) and the damping parameter ($\alpha$ and $\lambda$) in the LL equation and the LLG equation is $\alpha_{\text{LLG}} = \lambda_{\text{LL}}/\gamma_{\text{LL}} M_s$, $\gamma_{\text{LLG}} = \gamma(1 + \alpha_{\text{LLG}}^2)$. In this dissertation, we choose to use the LLG form.

1.1.2 Ferromagnetic Resonance Condition

The LLG equation implies that the motion of magnetic moment in a static magnetic field will eventually diminish due to damping. However, if an additional microwave field that is perpendicular to the static magnetic field is applied, the precessional motion of the magnetic moment is able to sustain indefinitely by absorbing energy from the microwave...
counting the energy loss from damping. In addition, when the frequency of the microwave field and the amplitude of the static field satisfies certain condition, the precessional angle of the magnetic moment and the absorption of microwave energy will reach a maximum value. This condition is called the Ferromagnetic resonance (FMR) condition. The FMR condition can be derived using a macro spin approximation, where all magnetic moments in the system are assumed to precess in phase thus the system can be treated as a single domain. This has been formulated by Smit and Beljers in 1955 [4]:

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

(1.4)

Here \( f \) is the frequency of the microwave field, \( \gamma' = \frac{g \mu_B}{h} \) is the gyromagnetic ratio, \( \theta \) and \( \phi \) represents the polar and azimuth angle of \( M \). The second derivative of the Gibbs free energy \( E \) should be evaluated at the equilibrium angles \( \theta_0 \) and \( \phi_0 \), i.e., \( \frac{\partial E}{\partial \theta} \bigg|_{\theta_0} = \frac{\partial E}{\partial \phi} \bigg|_{\phi_0} = 0 \). The damping parameter \( \alpha \) is neglected in the derivation of Eq. 1.4 because it is usually \( \ll 1 \) [5, 6].

![Figure 1.2: Schematic of the coordinate system used for Eq. (1.5). \( \hat{e}_u(\hat{e}_4) \) represents the direction of the easy axis of the uniaxial (four-fold) anisotropy. \( \phi_u(\phi_4) \) stands for the azimuth angle of the easy axis of the uniaxial (four-fold) anisotropy. \( \phi_0 \) is the equilibrium in-plane angle of the magnetization.](image)

For thin films with tetragonal symmetry, the dispersion relation for in-plane geometry
$(\theta = \pi/2)$ is given by [6, 7]:

\[
\left( \frac{f}{\gamma'} \right)^2 = \left\{ H \cos(\phi - \phi_H) + \frac{K_4}{2M_s} [3 + \cos 4(\phi - \phi_4)] \right. \\
+ \frac{K_u}{M_s} [1 + \cos 2(\phi - \phi_u)] + 4\pi M_{\text{eff}} \right\} \cdot \\
\left\{ H \cos(\phi - \phi_H) + \frac{2K_4}{M_s} \cos 4(\phi - \phi_4) + \frac{2K_u}{M_s} \cos 2(\phi - \phi_u) \right\},
\]

where $K_4$ and $K_u$ is the in-plane four-fold and uniaxial anisotropy, respectively.

$4\pi M_{\text{eff}} = 4\pi M_s - \frac{K_\perp}{M_s}$ is the effective magnetization and and $K_\perp$ accounts for any perpendicular anisotropies present in the films. Dispersion relations for other conditions can be found in Ref. [6].

### 1.1.3 Spin Waves

![Sketch of spin waves](image)

Figure 1.3: Sketch of (a) the uniform precession mode and (b) a spin wave mode with finite wavelength $\lambda$. $\beta$ is the angle between the magnetic moment $\mathbf{M}$ and magnetic field $\mathbf{H}$. $\phi$ is the angle between the neighbor magnet moments' projections on the x-y plane. [8, 9]

The concept of spin waves as the lowest lying states above the ground state of a magnetic system was first introduced by Bloch[10]. If some spins deviate from their equilibrium orientation, their neighbors will be influenced through dipolar interaction and exchange interaction and thus deflected. These disturbances will propagate through the medium, creating a spin wave (see Fig. 1.3 (b)). Spin waves are the fundamental dynamic magnetic excitations of a magnetic system. They provide the basis to describe the spatial
and temporal evolution of the magnetization of a magnetic object. The uniform precession mode measured in FMR experiments can be treated as a spin wave that has zero wave vector as shown in Fig. 1.3(a). Besides the uniform mode, another commonly observed collective excitation in FMR experiments is the Perpendicular standing spin wave (PSSW) mode, which has a quantized wave vector perpendicular to the film plane (see Fig. 1.4). The non-zero wave vector spin wave mode can be excited by inhomogeneous microwave field or inhomogeneities inside the film itself [11, 12]. Kittel[12] further pointed out that only odd spin wave modes could be observed if spins are completed pinned by surface anisotropy on both surfaces of the film. However, the completely pinned condition is not always satisfied in experiments. It is possible to have partial pinned or even completely unpinned boundary conditions. Fig. 1.4 shows the mode profiles of the uniform mode \((n = 0)\) and first two PSSW modes. Here a completely unpinned boundary condition is used for both interfaces.

The dispersion relation for PSSW modes is slightly different from that of the FMR mode due to the additional exchange interaction. The dipolar interaction is ignored here because it is much smaller comparing to the exchange field in thin films [13]. For thin films with tetragonal symmetry, whose dispersion relation for the FMR mode can be described by Eq. (1.5), the dispersion relation for PSSW modes that are assumed to have perfect pinning condition at both surfaces is given by [7, 14]:

\[
\left( \frac{f}{\gamma} \right)^2 = \left\{ \frac{H}{M_s} \cos(\phi - \phi_H) + \frac{K_4}{2M_s} \left[ 3 + \cos 4(\phi - \phi_4) \right] + \frac{K_u}{M_s} \left[ 1 + \cos 2(\phi - \phi_u) \right] + 4\pi M_{\text{eff}} + H_{\text{ex}} \right\} \cdot \left\{ H \cos(\phi - \phi_H) + \frac{2K_4}{M_s} \cos 4(\phi - \phi_4) + \frac{2K_u}{M_s} \cos 2(\phi - \phi_u) + H_{\text{ex}} \right\},
\]

(1.6)

with the exchange field \(H_{\text{ex}} = \frac{2A}{M_s} \left( \frac{n\pi}{t} \right)^2\), where \(A\) is the exchange constant, \(n\) is the spin wave mode number and \(t\) is the film thickness. Thus, the investigation of PSSW modes in
FMR experiments provides a method of estimating the exchange constant. This will be utilized in chapter 5 to determine the exchange constant of (FeCo)-Si alloy thin films.

![Diagram of spin wave modes](image)

**Figure 1.4:** Shapes of the perpendicular standing spin wave modes \((n=0,1,2)\) for unpinned boundary condition. \(t\) is the film thickness. The horizontal distance between the solid lines and dashed lines represents the precession angle of the spins. Illustrative sketches of the precession angle of the spins along the thickness dimension are shown for \(n = 1\) mode. \[8, 9\]

### 1.2 Magnetization Relaxation

Although the phenomenological damping term introduced in the LLG equation was very successful in describing the magnetization dynamics. The physical origin of the damping is still an active research field. Figure 1.5 shows some relaxation processes. The uniform precession excited by the microwave can transfer its energy directly into the lattice system. Alternatively, it can also transfer its energy to an intermediate states within the spin system, such as the degenerated magnons or thermal magnons. Eventually, the degenerated magnons and thermal magnons will transfer their energy to the lattice system.

In the next few paragraphs, a few relaxation mechanisms will be discussed.

Multilayer thin films are very common in magnetization relaxation studies. If a metal layer is adjacent to a ferromagnetic layer in a multilayer thin film, the precessing magnetization in the ferromagnetic layer near the ferromagnetic/metal interface will pump a spin current into the adjacent metal layer, which results in a damping contribution known as spin pumping\[15–17\]. Spin accumulation will happen in the metal layer due to the injected spin current. The accumulated spins can either create a spin current which
flows back to the ferromagnetic layer or relax by spin-flip scattering. Higher spin-flip rate will lead to higher damping contribution. For a good spin sink, the contribution of spin pumping scales with the inverse film thickness[16].

Precessing magnetic moments, which create time dependent magnetic fields, will induce eddy currents inside metallic films [18, 19]. The damping contribution from eddy current has the form of the damping term in the LLG equation. For films thinner than the skin depth, the damping caused by the eddy current is proportional to square of the film thickness, \( t^2 \), and the conductivity \( \sigma \) [20, 21]. Thus the eddy current contribution to the damping in ultrathin film studies is usually small enough to be safely ignored. For films thicker than the skin depth, the linewidth broadening caused by the eddy current is proportional to \( \sqrt{\sigma A} \) [20]. This expression indicates the involvement of the exchange interaction. Thus this contribution to the damping is usually referred to as exchange conductivity mechanism [18].

Two-magnon scattering is a process through which the uniform precession mode with zero wave vector scatters into degenerate magnons with finite wave vector within the film plane, which is a relaxation mechanism of the uniform precession mode. The existence of both the scattering centers and the degenerate magnons is required for the two-magnon scattering to be operative. Degenerate magnons only exist when the angle between the wave vector of the degenerate magnon and the saturation magnetization is below a critical...
angle[22]. The defects naturally occur at the interfaces and surfaces of films are the most common scattering centers. To calculate the contribution of the two-magnon scattering, a quantitative model of the defects is needed. Mills and Arias developed an expression for the linewidth contribution of the two-magnon scattering assuming defects have rectangular shape[23]. The expression shows a nonlinear frequency dependence which distinguishes the two-magnon scattering contribution from other Gilbert like damping contributions. The strength of the two-magnon scattering caused by surface roughness is expected inversely proportional to square of the film thickness.

1.3 Experimental Setup

A fully automated FMR setup has been used to characterize the magnetization dynamics of thin films. The schematic diagram of the setup is shown in Fig. 1.6. The microwave field generated by a microwave source is transmitted through a CPW, where the sample is mounted. The reason of using a CPW instead of a cavity resonator or shorted waveguide is that a CPW can operate over a very wide frequency range. Traditionally, FMR measurements were only performed at one or very few frequencies, which results in large error margins of the measurement results. The CPW allows us to perform a broadband FMR measurement which will minimize the error margins. In addition, the flat surface of CPW is a perfect mounting stage for thin film samples, which results in an easier sample handling. The microwave power transmitted through the CPW is measured as voltage by a microwave diode and then detected by the lock-in amplifier. The lock-in amplifier has a built-in frequency synthesizer, which is used to modulate the quasi-static magnetic field via the modulation coils. The measured lock-in signal is proportional to the amplitude of the modulation voltage and the derivative of the transmitted energy with respect to the quasi-static field. The measured signal is then multiplied by the reference signal and the result will go through a low-pass filter, which filters out almost all AC signals and leaves only the DC signal. This step allows the lock-in amplifier to reject noise
Figure 1.6: Schematic diagram of a fully automated FMR setup. A pair of electromagnets is used to provide a DC magnetic field. The DAQ card that is connected to the power supply of the electromagnets is used to control the magnitude of the generated DC magnetic field. A pair of modulation coils is attached to the poles of the electromagnets. The modulation field is defined by the reference signal generated by the lock-in amplifier. An amplifier is used between the modulation coils and the lock-in amplifier in order to enhance the amplitude of the reference signal. The CPW between the electromagnets serves as both the transmission line of the microwave and the sample mounting stage. Next to the CPW is a Hall probe connected to the Gauss meter, which is used to measure the DC magnetic field. The DAQ card, the lock-in amplifier, the microwave source, and the Gauss meter are all controlled by computer via GPIB interface, which is shown in blue lines.
that is not around the reference frequency. The application of the lock-in technique\[24, 25\] greatly improves the signal to noise ratio. This is essential especially for high frequency measurements, where the signal is usually extremely weak.

Figure 1.7: Photo of (a) the room temperature FMR setup and (b) the low temperature FMR setup.

The room temperature FMR setup (shown in Fig. 1.7(a)) is able to operate in the frequency range of 2 to 66 GHz. The maximum DC magnetic field provided by the electromagnets is 1.7 T. The two attached rotation motors, which can control the in-plane angle of the sample and the elevation angle of the stage, provide the capability of 0° to 360° in-plane angle dependent measurements and 0° to 90° (90° corresponds to the in-plane configuration and 0° corresponds to out-of-plane configuration) elevation angle dependent measurements. The low temperature FMR setup (shown in Fig. 1.7(b)) shares most of the design of the room temperature setup. The major difference is that the CPW in the low temperature setup is inside a cryostat, which can be set to a temperature between 10 K to 300 K. Because of the inclusion of the cryostat, the upper frequency limit is 40 GHz and only elevation angle dependent measurements are possible using the low temperature setup.

Fig. 1.8 shows a typical FMR spectrum measured at a fixed frequency. As mentioned
earlier, the signal measured by the lock-in technique is proportional to the derivative of the transmitted energy with respect to the quasi-static field. The absorption line of FMR is known has a Lorentzian line shape. Thus, the spectrum can be fitted to the derivative of Lorentzian line shape using the following expression [26, 27] 

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

(1.7)

where \( y \) is the FMR signal, \( H \) is the applied quasi-static magnetic field, \( H_{\text{res}} \) is the resonance field, \( \Delta H_{\text{pp}} \) is the peak to peak linewidth and \( a \) and \( b \) are amplitudes of absorption and dispersion signals, respectively. In an ideal situation where the power loss is purely due to the ferromagnetic resonance of the magnetic sample, only the absorption signal will be observed. However, conductive and dielectric loss usually exist in FMR measurements as well, which will add a dispersive contribution to the line shape[28]. Thus it is essential to add the dispersion part in the line shape analysis for a precise resonance field determination.

The formula of peak to peak linewidth is given by [25, 29–31]:

\[
\Delta H_{\text{pp}} = \frac{1}{\sqrt{3}} \frac{\alpha}{|d^2 H_{\text{res}}/dH|^3} M_s \left[ \frac{\partial^2 E}{\partial \theta^2} + \frac{1}{\sin^2 \theta} \frac{\partial^2 E}{\partial \phi^2} \right] 
\approx \frac{1}{\sqrt{3}} \frac{\alpha f}{\gamma' \cos \beta}.
\]  

(1.8)

Here \( \beta \) is the angle between the magnetization and external DC magnetic field. Because the \( \cos \beta \) term is in the denominator, it will always broaden the linewidth. This linewidth broadening caused by the misalignment between the magnetization and external static field is thus called field dragging. However, if the external field is applied along the easy or hard direction of the magnetization and has a large enough magnitude, the magnetization will
Figure 1.8: A typical FMR spectrum measured by a field sweep at a fixed frequency of 40 GHz. The black line represents the raw data and the red line represents the fit to the data using Eq. (1.7)
be parallel to the external static field. Then Eq. (1.8) becomes:

$$\Delta H_{pp} \approx \frac{1}{\sqrt{3}} \alpha \gamma f. \quad (1.9)$$

A zero-frequency offset $\Delta H_0$ is usually added to Eq. (1.9) to take the linewidth caused by sample inhomogeneities into account, resulting in:

$$\Delta H_{pp} = \Delta H_0 + \frac{1}{\sqrt{3}} \alpha \gamma f. \quad (1.10)$$

When $H_{res}$ and $\Delta H_{pp}$ are determined for all frequencies in a broadband FMR measurement. One can plot $f$ vs. $H_{res}$ and fit it with the dispersion relation, which can be used to determine magnetic properties such as the gyromagnetic ratio, the effective magnetization and anisotropies. In addition, the $\Delta H_{pp}$ vs. $f$ plot provides information of the relaxation of the material. If a linear relationship is observed in the $\Delta H_{pp}$ vs. $f$ plot, one can fit the data with Eq. (1.10) and obtain a Gilbert damping parameter $\alpha$.

1.4 Study of Fe Alloys

In the 1930s, a magnetic metal powder called Sendust was invented[32]. Sendust consists of approximately 85% iron, 9% silicon and 6% aluminum. Its high permeability, low coercivity, low loss and good temperature stability make it very good soft magnetic material for applications such as inductors. Since then, various studies on Fe based alloys such as FeAl, FeCoAl, FeCoSi, FeCoSiAl were carried out[33–39]. However, the study on the magnetization dynamics is not very common in the literature. Since the understanding of the magnetization dynamics is essential in developing soft magnetic materials for high frequency applications, we utilize FMR to investigate the properties of (FeCo)-Al and (FeCo)-Si thin films. Other experimental techniques such as X-ray diffraction (XRD), X-ray reflectivity (XRR), X-ray photoelectron spectroscopy (XPS), Transmission electron microscopy (TEM), Energy-dispersive X-ray spectroscopy (EDX), Vibrating sample
magnetometer (VSM), and Magneto-optical Kerr effect (MOKE) are also used to characterize the structural and other magnetic properties of those thin films.

In chapter 2, an extensive investigation of sputter-deposited FeAl and (FeCo)-Al thin films is presented. The influence of Al and Co on the magnetic properties of the films is studied. In chapter 3 and chapter 4, the thickness and growth temperature dependence of both the quasi-static and dynamic magnetic properties of (FeCo)-Al thin films are reported, respectively. Chapter 5 focuses on the dynamic magnetic properties of (FeCo)-Si thin films. Furthermore, the exchange constant, the anisotropies and the relaxation mechanism of (FeCo)-Si thin films are discussed. Chapter 6 summaries the findings in chapter 2-5 and discusses possible future studies. I carried out all the FMR measurements and all the FMR data analysis in this dissertation. Sample preparation, XRD, XRR, XPS, TEM, EDX, VSM, and MOKE measurements were carried out by Isao Kanada, Yusuke Ariake, Kyotaro Abe and Yoshimoto Tanaka from TDK.
2 SOFT MAGNETIC PROPERTIES AND DAMPING PARAMETER OF (FeCo)-Al ALLOY THIN FILMS

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For high frequency device applications, a systematic study of the soft magnetic properties and magnetization dynamics of (FeCo)-Al alloy thin films has been carried out. A low effective damping parameter eff of 0.002 and a high saturation magnetization of about 1,800 emu/cc are obtained at $y = 0.2 \sim 0.3$ for $(\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2$ alloy thin films deposited onto fused silica and MgO(100) at an ambient temperature during deposition. Those films are of the bcc structure with the $\langle 110 \rangle$ orientation normal to the film plane. They possess a columnar structure, grown along the film normal. The column width is found to be about 20 nm for $y = 0.25$. It is concluded that the (FeCo)-Al thin films with a damping parameter as low as 0.002 and high saturation magnetization of about 1,800 emu/cc have been successfully fabricated, and that they are potential for future high frequency device applications.
2.1 Introduction

For soft magnetic materials for future high frequency device applications, a further improvement for high saturation magnetization, high permeability, low coercivity and low damping parameter must be realized. Among various candidates, Fe-based alloys are very attractive because there are still opportunities to improve the soft magnetic properties mentioned above. One of the Fe-alloys for commercially utilized is the Fe$_{49}$Co$_{49}$V$_2$ alloy, called “V-permendur”, which has the very high saturation induction (2.4T) and the high permeability (~20,000) [1]. The Fe-Al alloys have also been extensively studied for their magnetic properties. The rapidly quenched amorphous ribbons of Fe-16wt.%Al were reported to exhibit very low coercivity $H_c$ of 16 A/m [2]. Fe-1wt.%Al and Fe-5wt.%Al via power metallurgical process were also reported to exhibit high permeability $\mu$ of 4,250 and 4,367 and low $H_c$ of 90 A/m and 64 A/m, respectively [3, 4]. However, very few works on the soft magnetic properties including the damping parameter in Fe-Al thin films can be found in literature. Therefore, a systematic study has been carried out to examine the magnetic properties and structure of (FeCo)-Al thin films in order to explore a novel soft magnetic material for future high frequency device applications.

2.2 Experimental

Multilayers of $[\text{Fe}(d_1)/\text{Al}(d_2)] \times N$, and $[\text{Fe}(d_1)/\text{Fe}_{64}\text{Co}_{34}(d_3)/\text{Al}(d_2)]$ were sputter-deposited at an ambient temperature by using DC magnetron sputtering in Ar atmosphere of 4 mTorr. Here, $d_1$, $d_2$, $d_3$ and $N$ are the layer thickness for Fe, Al, Fe$_{64}$Co$_{34}$, and the number of the layer-repetition, respectively, which were varied from 0 to 1.8 nm for $d_1$, $d_3$, from 0 to 0.3 nm for $d_2$, and from 20 to 27 for $N$. The substrates were fused silica glass and MgO(100) single crystals. The 5 nm thick Ru layer was over-coated for protection. No post-annealing was performed. The base pressure during deposition was better than $2 \times 10^{-7}$ Torr. The deposition rates for Fe, Fe$_{64}$Co$_{34}$ and Al were 0.16, 0.17 and
0.04 nm/sec, respectively.

The film compositions for all samples under consideration were analyzed by X-ray photoelectron spectroscopy (XPS). In the present paper, the results of both $\text{Fe}_{100-x}\text{Al}_x$ and $(\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2$ thin films are presented, where $x$ and $y$ are from 0 to 11, and from 0 to 0.35, respectively. The total thickness of each sample is about 50 nm.

The film thicknesses were determined by X-ray reflectivity. Structural analyses were performed by X-ray diffraction (XRD) with Cu (K$_\alpha$) radiation, high resolution transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDX). Measurements of the quasistatic magnetic properties were carried out by using a vibrating sample magnetometer in fields up to 10 kOe and magneto-optical Kerr effect measurements in fields up to 1 kOe. In order to evaluate the magnetization dynamics ferromagnetic resonance experiments were performed over a frequency range from 10 to 70 GHz.

### 2.3 Result and discussion

#### 2.3.1 Fe-Al

Figure 2.1 shows the XRD patterns for Fe-Al films with the various compositions thus fabricated onto (a) fused silica glass and (b) MgO substrates. The samples for silica glass substrates exhibit rather broad and weak diffraction peaks of $\langle 110 \rangle_{\text{bcc}}$. No clear indication of the presence of the multilayer structure is found by low angle XRD. On the other hand, since the diffraction peaks of $\langle 110 \rangle_{\text{bcc}}$ for the samples onto MgO(100) overlapped with the $(200)$ peak of MgO(100), its structure was not confirmed.

Figure 2.1(c) shows the lattice constant, $a$ as a function of the Aluminum content $x$ for the $\text{Fe}_{100-x}\text{Al}_x$ films deposited onto silica glass, together with the bulk values. The lattice constant increases with Al content from 2.87 ($x = 0$) to about 2.94 Å ($x = 10.7$). The increase in lattice constant, $a$ with $x$ is not linear but seems to level off with $x$. It is noted that the present values of lattice constant, $a$ for the films are significantly larger (about 1.8% at $x = 10.7\%$) than those reported for bulk specimen [5].
Figure 2.1: The XRD patterns for Fe-Al films with the various compositions fabricated onto (a) fused silica glass and (b) MgO substrates. (c) The lattice constant, \( a \) as a function of \( x \) for the Fe\(_{100-x}\)Al\(_x\) films deposited onto silica glass, together with the bulk values [5].

Figure 2.2(a) shows EDX element mapping images of Fe, Al and Ru for the cross section of the Fe\(_{100-x}\)Al\(_x\) films with \( x = 0, 1.3 \) and 3.9, fabricated onto MgO(100) substrates. The distribution of Fe and Al are found to be homogeneous over the cross section of all samples.

Figure 2.2(b) are TEM cross-sectional images for the corresponding samples of Figure 2.2(a). A columnar growth morphology is clearly evident for all samples. The columnar growth direction is normal to the film-plane, and the width decreases with \( x \) from around 50 nm (\( x = 0 \)) to 10 ~ 20 nm (\( x = 1.3 \)). It is noted that an amorphous layer near the interface between the film and substrate is observed for all samples. The thickness of this amorphous layer increases with \( x \) from about 3 nm for \( x = 0 \) to about 6 nm for both \( x = 1.3 \) and 3.9. The EDX images in Fig. 2.2(a) indicate little evidence for any segregation of Fe and Al elements near the interface, implying that the amorphous layer consists of the same composition as the interior of the sample.

Figure 2.3 shows the saturation magnetization \( M_s \) and coercivity \( H_c \) determined by VSM as a function of \( x \) for the Fe\(_{100-x}\)Al\(_x\) films deposited onto silica glass and MgO(100) substrates. The \( M_s \) values for both the films deposited onto silica glass and MgO(100)
Figure 2.2: (a) EDX element mapping images of Fe, Al and Ru for the cross section of the Fe$_{100-x}$Al$_x$ films with $x = 0$, 1.3 and 3.9, fabricated onto MgO(100) substrates. (b) TEM cross-sectional images for the corresponding samples of FIG. 2.1(a). (c) Diffraction patterns for the corresponding samples.

Figure 2.3: The saturation magnetization $M_s$, together with the bulk values [5] and coercivity $H_c$ measured by VSM as a function of $x$ for the Fe$_{100-x}$Al$_x$ films deposited onto silica glass and MgO(100) substrates.
decreases monotonously with \( x \), which is consistent with dilution models \([6]\), and also in reasonable agreement with the bulk values for \( x \) up to about 6. Beyond \( x = 6 \), the \( M_s \) values for both the samples start to deviate from that for bulk \([7]\). According to the work by Sucksmith \([7]\), the \( M_s \) for bulk decreases drastically with \( x \) beyond \( x = 20 \), and the present result is at variance with the bulk data. The reason for this deviation is not understood at present. The coercivity \( H_c \) is found to drastically decrease with \( x \) from 20 ~ 30 Oe (\( x = 0 \)) to 3 ~ 5 Oe (\( x = 1.3 \)), and then gradually increases with \( x \) up to around 10 Oe at \( x = 10.7 \). The drastic decrease of \( H_c \) with increasing Al content may be associated with the change of the columnar growth morphology observed in Figure 2.2(b). It is also conjectured that since the addition of Al in the Fe-Al alloys increases the magnetostriction \([8, 9]\), the increase in \( H_c \) with \( x \) beyond about \( x = 5 \) could be associated with the stress induced magnetic anisotropy, leading to a higher \( H_c \) value.

Figure 2.4: (a) The dependence of FMR linewidth \( \Delta H \) on resonance frequency \( f_{res} \) for Fe\(_{100-x}\)Al\(_x\) alloy films fabricated onto fused silica and MgO(100) substrates. (b) The effective damping parameter \( \alpha_{eff} \) as a function of \( x \).

Figure 2.4(a) shows the dependence of FMR linewidth \( \Delta H \) on resonance frequency \( f_{res} \) for Fe\(_{100-x}\)Al\(_x\) alloy films fabricated onto MgO(100) substrates. It is found that the films with \( x = 1.3, 3.9 \) and 6.2 exhibit linear relationships between \( \Delta H \) and \( f_{res} \) with nearly the
same slope. On the other hand, for \( x = 0 \) and 10.7, the \( \Delta H \) vs \( f_{\text{res}} \) curves exhibit a complicated nonlinear behavior. This behavior may be associated with the morphology in those samples, leading to spin-wave excitations and inhomogeneous line broadening.

Based on the linear relationship between the \( \Delta H \) and the resonance frequency \( f_{\text{res}} \), the effective damping parameter \( \alpha_{\text{eff}} \) was estimated for the samples with \( x = 1.3, 3.9 \) and 6.2. Here, the equation,\(^{[10, 11]}\) \( \Delta H = \Delta H_0 + (4\pi/\sqrt{3})\alpha_{\text{eff}} f_{\text{res}}/\gamma \) is used, where \( \gamma \) is the gyromagnetic ratio. The effective damping parameter \( \alpha_{\text{eff}} \) thus estimated is shown in Figure 2.4(b).

As shown in Fig. 2.4(b), effective damping parameters of around 0.003 ∼ 0.004 are obtained for the samples with \( x = 1.3, 3.9 \) and 6.2 both on silica glass and MgO(100) substrates. These values are comparable to that reported for Fe \(^{[12]}\). There have been few reports in literature about the damping parameter in the Fe-Al alloy thin films. It is further noted that the inhomogeneous linewidth contributions \( \Delta H_0 \) for \( x = 1.3 \) and 3.9 are as small as 5 ∼ 10 Oe. Based on the results for the Fe\(_{100-x}\)Al\(_x\) films, further studies are worthwhile to improve the soft magnetic properties as well as saturation magnetization of Fe-Al thin films by addition Co. Therefore, experiments on Fe(Co)-Al thin films, discussed below have been performed.

### 2.3.2 (FeCo)-Al

The XRD patterns for \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) alloy thin films with various \( y \) values from 0 to 0.35, fabricated on fused silica substrate at an ambient temperature are shown in Figure 2.5(a). The films fabricated on MgO(100) are not shown because the peaks from the films overlap with those of MgO(100) substrate, as in the case of Figure 2.1(a). The (110)\(_{\text{bcc}}\) peaks are observed for all the films. Little evidence of the presence of the multilayer structure is found by low angle X-ray diffraction. Figure 2.5(b) shows the lattice constant, \( a \) for the \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) alloy thin films. It is found that the lattice constant \( a \) decrease monotonously with \( x \). The change of the lattice constant is very small because the atomic radiuses for Fe and Co are close to each other.
Figure 2.5: (a) The XRD patterns for \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) alloy thin films with various \(y\) from 0 to 0.35, fabricated onto fused silica substrate at an ambient temperature. (b) The lattice constant, \(a\) for the \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) alloy thin films fabricated onto fused silica substrate at an ambient temperature as a function of \(y\).
Figure 2.6: (a) EDX element mapping images of Fe, Co, Al and Ru for the cross section of the \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) films with \(y = 0.25\) fabricated onto MgO(100) substrates. (b) TEM cross-sectional images for the corresponding samples of FIG. 2.6(a).

Figure 2.6 shows (a) EDX and (b) TEM images for the cross-section of the sample with \(y = 0.25\). It is seen that the all the elements of Fe, Co and Al are uniformly distributed, indicating little evidence of atomic segregation. The TEM images show columnar structures elongated along the film normal. The average width of the columns is about 20 ~30nm, similar to the Fe-Al case of Figure 2.2. It is noted that the initial layer (about 1 nm thickness) near the interface between the film and MgO substrate is present. The TEM diffraction pattern shown in the inserted figure reveals the growth direction of the film is \(\langle 110 \rangle\), consistent with the X-ray diffraction pattern.

Figure 2.7(a) shows the saturation magnetization \(M_s\) as a function of \(y\) for the \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) alloy thin films deposited onto silica glass and MgO(100) substrates. \(M_s\) increases monotonously with \(y\), reaching a maximum at around \(y = 0.3\). The saturation magnetization at \(y = 0.3\) is about 1,800 emu/cc, about 15% larger than that at \(y = 0\).

Figure 2.7(b) shows the dependence of \(H_c\) on \(y\) for the \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) alloy thin films. The coercivity increases with \(y\) up to \(y = 0.25\). It is known [7] that the addition of Co to the Fe-Co alloys decreases the magnetocrystalline anisotropy constant \(K_1\) to be nearly zero.
Figure 2.7: (a) The saturation magnetization $M_s$ as a function of $y$ for the $(\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2$ alloy thin films. (b) The dependence of $H_c$ on $y$ for the $(\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2$ alloy thin films.

at about Co at 45% from $5 \times 10^5$ erg/cc for bcc Fe. On the other hand, magnetostriction ($\lambda_{100}$) increases with Co by an order of the magnitude from bcc Fe to about Co at 45%. The increase in $H_c$ observed in the present study is therefore likely due to the stress induced magnetic anisotropy through the magneto-elastic coupling [9].

Figure 2.8: The damping parameter $\alpha_{\text{eff}}$ for the $(\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2$ alloy thin films deposited onto silica glass and MgO(100) substrates.

Figure 2.8 shows the effective damping parameter $\alpha_{\text{eff}}$ evaluated by FMR measurements
described in section 2.3.1 Fe-Al for the \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) alloy thin films deposited onto silica glass and \(\text{MgO}(100)\) substrates. The \(\alpha_{\text{eff}}\) at \(y = 0\) is about 0.004 and decreases with \(y\), becoming minimum 0.002 at \(y = 0.25\) and then increases to reach about 0.004 at around \(y = 0.3\). The present result of the decrease of \(\alpha_{\text{eff}}\) with \(y\) is consistent with earlier works [13].

2.4 Summary

For high frequency device applications, a systematic study of the soft magnetic properties and damping parameter of (FeCo)-Al alloy thin films has been carried out. A low effective damping parameter \(\alpha_{\text{eff}}\) of 0.002 and a high saturation magnetization of about 1,800 emu/cc are obtained at \(y = 0.2 \sim 0.3\) for \((\text{Fe}_{1-y}\text{Co}_y)_{98}\text{Al}_2\) alloy thin films deposited onto fused silica and \(\text{MgO}(100)\) at an ambient temperature during deposition. Those films are of the bcc structure with the \(\langle 110 \rangle\) orientation normal to the film plane. They possess the columnar structure, grown along the film normal. The column width is found to be about 20 nm for \(y = 0.25\). It is concluded that the (FeCo)-Al thin films with a damping parameter as low as 0.002 and high saturation magnetization of about 1,800 emu/cc have been successfully fabricated, and that this material is an excellent candidate for future high frequency device applications.

2.5 Acknowledgement

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2.6 References


THE THICKNESS DEPENDENCE OF SOFT MAGNETIC PROPERTIES of (FeCo)-Al ALLOY THIN FILMS

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A systematic study of the film thickness dependence of soft magnetic properties and magnetization dynamics of Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films has been carried out. These films have columnar structures grown along the film normal, with the average width of the columns of about 30 nm. The saturation magnetization $M_s$ is about 1,550 emu/cc, which remains nearly unchanged with film thickness over a range from 10 to 120 nm. The coercivity $H_c$ is found to increase with film thickness from 15 to 64 Oe over a range of 11 to about 50 nm, and then slightly decrease with further increase of thickness. The effective damping parameter $\alpha_{\text{eff}}$ measured by ferromagnetic resonance over a frequency range from 12 to 66 GHz is found to decrease with film thickness, becoming 0.0004 and 0.0006, for the films deposited onto fused silica and MgO (100) substrate, respectively.
3.1 Introduction

For soft magnetic materials for future high frequency device applications, high saturation magnetization, high permeability, low coercivity, low eddy current loss and low damping parameter are indispensable. Among many choices for materials, Fe-based alloys are attractive as represented by Alperm (Fe\textsubscript{85}Al\textsubscript{15}) which exhibits high permeability, while the alloy possesses relatively high magnetostriction constants with increasing Al content [1]. However, there have been few reports found in literature regarding the soft magnetic properties of Fe-Al thin films, especially for the dependence of damping parameter on thickness and alloy composition [1–4].

A recent work on Fe\textsubscript{73}Co\textsubscript{25}Al\textsubscript{2} thin films showed an effective damping parameter with saturation magnetization of 1,800 emu/cc, suggesting a candidate for future high frequency device application [5]. This result is consistent with the work reported Fe\textsubscript{75}Co\textsubscript{25} thin film [6]. An extrinsic portion of an effective damping parameter is sensitive to inhomogeneity in film morphology such as grains, surface roughness, thickness and so on. In terms of the thickness dependence of damping parameter, it was reported that the damping parameter for yttrium iron garnet thin films decreased with thickness [7] and permalloy thin films [8], whereas it increased with thickness for epitaxial iron thin films [9] and epitaxial Fe\textsubscript{3}O\textsubscript{4} thin films [10].

In order to shed light on the correlation between morphology and observed damping parameter, a systematic study of soft magnetic properties and a damping parameter as a function of film thickness of Fe\textsubscript{73}Co\textsubscript{25}Al\textsubscript{2} alloy thin films has been performed.

3.2 Experimental

Multilayers of [Fe(0.47 nm)/Fe\textsubscript{66}Co\textsubscript{34}(1.4 nm)/Al(0.042 nm)] \times N were sputter-deposited by using DC magnetron sputtering, where N was varied from 5 to 53, corresponding to the total thickness of 11 to 120 nm. Deposition was carried out in Ar
atmosphere of 4 mTorr at an ambient temperature. An in-plane field of 50 Oe was applied during deposition in order to induce a uniaxial magnetic anisotropy of the films thus fabricated. The substrates were fused silica glass and MgO (100) single crystals. A 5 nm thick Ru layer was over-coated for protection. No post annealing was performed. The base pressure prior to deposition was better than $2 \times 10^{-7}$ Torr. The deposition rates for Fe, Fe$_{66}$Co$_{34}$ and Al were 0.16, 0.17 and 0.042 nm/s, respectively.

The film thicknesses were estimated by X-ray reflectivity. Structural analyses were performed by X-ray diffraction (XRD) with Cu K$_\alpha$ radiation, high resolution transmission electron microscopy (TEM), and energy-dispersive X-ray spectroscopy (EDX). The composition of these samples was estimated as Fe$_{73}$Co$_{25}$Al$_2$ by EDX. Measurements of magnetic properties were carried out by a vibrating sample magnetometer (VSM) in fields up to 10 kOe and the longitudinal magneto-optical Kerr effect (MOKE). The magnetization dynamics was evaluated by ferromagnetic resonance (FMR) over a frequency range from 12 to 66 GHz.

### 3.3 Results and discussion

Fig. 3.1 shows the XRD patterns for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films with the various thicknesses deposited onto fused silica substrate. The samples with 48, 85 and 120 nm thicknesses show the relatively broad diffraction peaks of (110)$_\text{bcc}$ at $2\theta = 44.8^\circ$, suggesting the nanocrystalline structure. The lattice constant was estimated as $a = 0.286$ nm, in agreement with the value of bulk Fe$_{75}$Co$_{25}$ (0.286 nm) [11]. On the other hand, the 11 nm thick sample does not show a (110)$_\text{bcc}$ peak, implying an amorphous phase or a nanocrystalline structure. For the samples deposited onto MgO (100) substrate, the peaks from the substrate overlapped, thus preventing their detection, and therefore the data of MgO (100) substrates are not given here.

Fig. 3.2 shows the TEM cross-sectional images for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films with the thicknesses of 11, 48, 85 and 120 nm deposited onto fused silica substrate. A columnar type
Figure 3.1: XRD patterns for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica substrate with thicknesses of 11, 48, 85 and 120 nm.

Figure 3.2: TEM cross-sectional images for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica substrate with thicknesses of 11, 48, 85 and 120 nm.
growth structure along the direction normal to the film plane is present for all the samples. Although the width of such a columnar structure varies from place to place, the average width of the columns is about 30 nm. An amorphous layer near the interface between the film and substrate is also observed for all the samples. The thickness of this amorphous layer is about 1 nm for the samples with 48, 85 and 120 nm, whereas about 2-3 nm for the 11 nm thick sample. Comparing the TEM cross-sectional images of film deposited onto fused silica with that onto MgO substrate, little difference is found in morphology.

![EDX element mapping images of Fe, Co, Al and Ru for the cross section of Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica substrate with thicknesses of 11, 48, 85 and 120 nm and their diffraction patterns.](image)

Figure 3.3: EDX element mapping images of Fe, Co, Al and Ru for the cross section of Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica substrate with thicknesses of 11, 48, 85 and 120 nm and their diffraction patterns.

Fig. 3.3 shows the EDX element mapping images of Fe, Co, Al and Ru and the electron diffraction patterns for the samples with the various thicknesses. It is found that Fe, Co and Al are homogeneously distributed in the films, except for the sample with 11 nm thickness, where there is a signature of a layered structure for Co. The electron diffraction patterns show the ring patterns corresponding to the (110), (200) and (211) of bcc structure for the samples with 48, 85, and 120 nm thicknesses. For the 11 nm thick sample, the halo rings are observed, implying an amorphous phase, consistent with the result of XRD.
Figure 3.4: M-H curves for the easy axis direction in Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica substrate with thicknesses of 11, 48, 85, and 120 nm measured by VSM.
Fig. 3.4 shows the M-H curves for the easy axis direction in Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica substrate with the thicknesses of 11, 48, 85, and 120 nm measured by VSM. All the M-H curves exhibit a high squareness. The results of saturation magnetization $M_s$ and coercivity $H_c$ are summarized in Fig. 3.5. As shown, the $M_s$ is about 1,550 emu/cc, for all the samples under consideration. The coercivity $H_c$ increases with film thickness from 15 Oe (11 nm) to 64 Oe (48 nm), and then slightly decreases. It is found that there is little difference in both $M_s$ and $H_c$ between the fused silica and MgO (100) substrates. This result is qualitatively consistent with the structural data, as mentioned above. The trends of the $H_c$ with film thickness is consistent with the case for polycrystalline Fe thin films, where $H_c$ was reported to increase with film thickness up to about 50 nm, and then decrease [12]. The maximum of $H_c$ is an indication of the transition of a wall structure from Néel to Bloch wall with increasing film thickness. The main reason for this high $H_c$ values is believed to result from the stress induced magnetic anisotropy through magneto-elastic effect, since the magnetostriction constant is of the order of $10^{-5}$, and also since the present samples were fabricated onto the substrates kept at an ambient temperature during deposition, thus leading to a residual stress in the films [1–4]. One way to lower this effect may be to increase substrate deposition temperature, as in the present study, it was the ambient temperature.

Fig. 3.6 shows resonance frequency as a function of resonance field for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films with the thicknesses of 11, 48, 83, 85, and 120 nm obtained by FMR measurements. All the samples exhibit a very similar behavior to each other. The gyromagnetic ratio $\gamma$ and the effective magnetization $M_{\text{eff}}$ were determined by fitting these data to the Kittel equation

$$f_{\text{res}} = \frac{\gamma}{2\pi} \sqrt{H_{\text{res}} (H_{\text{res}} + 4\pi M_{\text{eff}})}.$$  (3.1)

Figure 3.7 shows the FMR linewidth $\Delta H$ as a function of resonance frequency for
Figure 3.5: Thickness dependence of saturation magnetization $M_s$ and coercivity $H_c$ for the easy axis direction in Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica and MgO (100) substrates, respectively.

Figure 3.6: Resonance frequency $f_{\text{res}}$ as a function of resonance field $H_{\text{res}}$ for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica substrate with thicknesses of 11, 48, 83, 85, and 120 nm.
Fe\textsubscript{73}Co\textsubscript{25}Al\textsubscript{2} alloy thin films deposited onto fused silica substrate. It is found that the 11 nm thick sample exhibits a linear relationship over a range from 12 GHz to 66 GHz. On the other hand, the other samples exhibit a nonlinear behavior at lower frequencies, and a linear relationship can only be found at frequencies higher than approximately 40 GHz. Therefore, in the present paper, the effective damping parameters were estimated based on the linear relationship over a frequency range from about 40 to 60 GHz, as shown by a dotted line, and by using the following equation:

\[
\Delta H = \Delta H_0 + \frac{4\pi \gamma \alpha_{\text{eff}}}{\sqrt{3} f_{\text{res}}}. \tag{3.2}
\]

In Fig. 3.8, the effective damping parameters $\alpha_{\text{eff}}$ thus estimated are plotted as a function of film thickness for the samples deposited onto fused silica and MgO (100) substrates. The $\alpha_{\text{eff}}$ values are found to decrease with film thickness up to about 83 nm, and then increase. The minimum value of 0.0004 and 0.0006 of $\alpha_{\text{eff}}$ for the samples deposited onto fused silica substrate and MgO (100) substrate were found at around $d = 83$ nm, respectively. Since the morphology is found to be insensitive to the type of substrate in the present study, it is reasonable to find that the values of effective damping parameter are nearly the same within an accuracy for both the substrates cases. For the thinner samples ($d < 83$ nm), the contribution from surface roughness to the linewidth is expected to become more important than that for the thicker samples. In contrast, for thicker films ($d > 83$ nm), contributions reflecting the microstructure of the films are expected to be more dominant. For these samples, the columnar structure of the films may play a key role for the relaxation mechanism. In the present study, it is not obvious to assess the correlation between the columnar structure and the effective damping parameter, and a further study is needed to clarify this correlation.
Figure 3.7: Linewidth $\Delta H$ as a function of resonance frequency $f_{\text{res}}$ for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto fused silica substrate with thicknesses of 11, 48, 83, 85, and 120 nm. A thin dotted line combining the data point at 66 GHz and the origin for the 120 nm thick sample is also shown in order to estimate the maximum error bar, as shown in Fig. 3.8.
3.4 Conclusion

A systematic study of the thickness dependence of soft magnetic properties and effective damping parameters of Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films has been carried out. These films have columnar structures grown along the film normal, with the average width of the columns of about 30 nm. The saturation magnetization $M_s$ is about 1550 emu/cc, which remains nearly unchanged with film thickness. The coercivity $H_c$ is found to increase with film thickness from about 15 to 64 Oe over a film thickness range from 11 to 50 nm up, and then to decrease. It is noted that the effective damping parameter $\alpha_{\text{eff}}$ is found to become minimum at around a film thickness of 83 nm, where the $\alpha_{\text{eff}}$ values of 0.0004 and 0.0006 are obtained for the films fabricated onto fused silica and MgO (100) substrates, respectively.

3.5 Acknowledgment

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3.6 References


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The soft magnetic properties and effective damping parameters of Fe73Co25Al2 alloy thin films are discussed. The effective damping parameter $\alpha_{\text{eff}}$ measured by ferromagnetic resonance for the 10 nm thick sample is nearly constant ($\approx 0.004 \pm 0.0008$) for a growth temperature $T_s$ from ambient to 200 °C, and then tends to decrease for higher temperatures and $\alpha_{\text{eff}}$ is 0.002 ± 0.0004 at $T_s = 300$ °C. For the 80 nm thick sample, the $\alpha_{\text{eff}}$ seems to increase with $T_s$ from $\alpha_{\text{eff}} = 0.001 \pm 0.0002$ at $T_s = \text{ambient}$ to $\alpha_{\text{eff}} = 0.002 \pm 0.0004$. The $\alpha_{\text{eff}}$ is found nearly constant ($\alpha_{\text{eff}} = 0.004 \pm 0.0008$) over a temperature range from 10 to 300 K for the 10 nm films with the different $T_s$ (ambient, 100 and 200 °C). Together with an increasing non-linearity of the frequency dependence of the linewidth at low $T_s$, extrinsic contributions such as two-magnon scattering dominate the observed temperature dependence of effective damping and linewidth.
4.1 Introduction

For future high frequency device applications of soft magnetic thin films, further improvement in saturation magnetization ($M_s$), permeability ($\mu$), coercivity ($H_c$), eddy current loss and damping parameter ($\alpha_{\text{eff}}$) is indispensable [1]. Among many candidates of materials to choose, FeCo-based alloy thin films are attractive because they exhibit high $M_s$ and low $\alpha_{\text{eff}}$ [2–5]. A recent work on Fe$_{73}$Co$_{25}$Al$_2$ thin films reported the thickness dependence of effective damping parameter $\alpha_{\text{eff}}$ and showed the values of $\alpha_{\text{eff}} = 0.0004$ at about 85 nm, indicating an attractive candidate as soft magnetic materials for future high frequency device applications [5]. However, since the coercivity for those films was still high for any practical use, lowering coercivity is desirable. The present paper describes a systematic study of the growth and measurement temperature dependences of soft magnetic properties and damping parameter of Fe$_{73}$Co$_{25}$Al$_2$ thin films.

4.2 Experimental

Multilayers of $[\text{Fe (0.45 nm)}/\text{Fe}_{66}\text{Co}_{34} (1.3 \text{ nm})/\text{Al (0.038 nm)}] \times N$ were sputter-deposited onto MgO (100) by DC magnetron sputtering in Ar atmosphere of 4 mTorr, where $N = 5$ and 37, corresponding to the total thickness of 10 and 80 nm, respectively. The substrate-deposition temperatures ($T_s$) were varied from ambient to 350 °C. In order to induce a uniaxial magnetic anisotropy, an in-plane field of 50 Oe was applied during deposition. A 5 nm thick Ru layer was over-coated at ambient temperature for protection. The base pressure prior to deposition was better than $2 \times 10^{-7}$ Torr. The deposition rates for Fe, Fe$_{66}$Co$_{34}$ and Al were 0.15, 0.17 and 0.038 nm/s, respectively.

The film thicknesses were estimated by X-ray reflectivity. Structural analyses were performed by X-ray diffraction (XRD) with Cu $K_{\alpha}$ radiation, high resolution transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDX). The composition of these samples was estimated as Fe$_{73}$Co$_{25}$Al$_2$ by EDX [5]. Measurements of
magnetic properties were carried out by a vibrating sample magnetometer (VSM) in fields up to 10 kOe. Two different types of measurements for magnetization dynamics were carried out by ferromagnetic resonance (FMR); namely over a frequency range from (i) 12 to 66 GHz at room temperature and (ii) 10 to 40 GHz for a temperature range from 10 to 300 K.

4.3 Results and discussion

4.3.1 Growth temperature dependence

Figure 4.1: XRD patterns for Fe\textsubscript{73}Co\textsubscript{25}Al\textsubscript{2} alloy thin films deposited onto MgO (100) substrate. Low angle XRD patterns for the films deposited at 100 °C are shown in the inserted figure where blue line shows the pattern of (200)\textsubscript{FeCoAl} and red line shows of (200)\textsubscript{MgO}.

Figure 4.1 shows the XRD patterns for 80 nm thick Fe\textsubscript{73}Co\textsubscript{25}Al\textsubscript{2} alloy thin films deposited at various substrate temperatures onto MgO (100) substrate. The samples
deposited at higher than 100 °C show the diffraction peaks of (200)bcc at 2θ at around 64.8 degree. (The diffraction of (110)bcc can’t be seen since the peak from the MgO (100) substrate overlaps.) It was found that the low angle XRD pattern for the 10 nm thick samples deposited above 100 °C showed the four peaks of (200)bcc separated at every 90 degree. In addition, it was also found that these peaks were shifted by 45 degree with respect to the peaks of (200)MgO, indicating that the film is a single crystalline film with \langle 100 \rangle_{\text{FeCoAl}} \parallel \langle 110 \rangle_{\text{MgO}}.

Figure 4.2: TEM cross-sectional image and the electron diffraction pattern for Fe_{73}Co_{25}Al_{2} alloy thin films deposited at ambient onto MgO (100) substrate [3].

Figure 4.2 shows the TEM cross sectional image, together with the diffraction patterns for 50 nm thick Fe_{73}Co_{25}Al_{2} alloy thin films deposited at ambient temperature onto MgO (100) substrate [3]. The TEM image shows the columnar structure grown along the film normal with the average width of about 20-30 nm. The diffraction patterns indicate that the film is polycrystalline film and its growth direction is \langle 110 \rangle, which is different of the films deposited above 100 °C.

Figure 4.3(a) are the M-H curves for the 10 nm thick samples deposited at various T_s. These curves were measured along the direction of \langle 100 \rangle_{\text{FeCoAl}} and \langle 110 \rangle_{\text{FeCoAl}} by VSM. The shape of the curve measured along the direction of \langle 100 \rangle_{\text{FeCoAl}} is significantly changed from the sample with the T_s of 100 °C to 200 °C, where the remanence M_r becomes much higher with higher T_s. The values of M_s, H_c and M_r/M_s for the samples with the thicknesses of 10 and 80 nm deposited at various T_s are summarized in Figure 4.3(b). It is seen that M_s remains unchanged (1,600 emu/cm^3) with T_s. On the other hand, H_c measured along the direction of \langle 100 \rangle_{\text{FeCoAl}} changes with T_s, becoming minimum at
Figure 4.3: (a) M-H curves and (b) growth temperature $T_s$ dependence of saturation magnetization $M_s$, coercivity $H_c$ and $M_r/M_s$ ratio for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto MgO(100) substrate. The $<110>$FeCoAl and $<100>$FeCoAl indicate the direction of applied field during the measurement, corresponding to the direction of $<100>$MgO and $<110>$MgO, respectively.
around 100 and 200 °C for the 10 and 80 nm thick samples, respectively. The observed decrease of $H_c$ is probably caused by reducing a residual stress in the films which induces magnetic anisotropy through magneto-elastic effect. The $M_r/M_s$ measured along the ⟨110⟩FeCoAl decreases with $T_s$, becoming minimum at $T_s = 200$ °C. The $M_r/M_s$ along the ⟨100⟩FeCoAl, on the other hand, increases with $T_s$ and it becomes higher than that along the ⟨110⟩FeCoAl above $T_s$ of 150 °C.

Figure 4.4: (a) FMR linewidth $\Delta H$ as a function of resonance frequency $f_{res}$ and (b) growth temperature $T_s$ dependence of effective damping parameter $\alpha_{eff}$ for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto MgO (100) substrate. The reported result in Ref. [5] is shown as the red square.

Figure 4.4(a) shows the FMR linewidth $\Delta H$ as a function of resonance frequency $f_{res}$ for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited at various $T_s$. The linewidth $\Delta H$ tends to decrease with $T_s$, and the linearity of these relationships is improved for higher $T_s$ over a wide range of frequency for the 10 nm thick sample. However, for the 80 nm thick film, the nonlinear frequency dependence of $\Delta H$ is found for $T_s = \text{ambient}$ and 100 °C. As $T_s$ goes higher, the crystallinity becomes improved, as found by XRD and TEM. Therefore, one would expect
the contributions of crystalline anisotropy field to the linewidth broadening which varies from grain to grain [6]. For the 10 nm thick sample fabricated at $T_s = $ ambient temperature, the nonlinear linewidth evolution can be attributed as the characteristic of two-magnon scattering due to the boundary of Ru/FeCoAl, as shown by Lenz et al [7].

The $\alpha_{\text{eff}}$ was estimated based on the linear relationship, as shown by a dotted line, fitted to the following equation:

$$\Delta H = \Delta H_0 + \frac{4\pi}{\sqrt{3}Y} \alpha_{\text{eff}} f_{\text{res}}.$$  \hspace{1cm} (4.1)

The result of $\alpha_{\text{eff}}$ as a function of $T_s$ is shown in Figure 4.4(b). It is found that the $\alpha_{\text{eff}}$ for the 10 nm thick sample is nearly constant ($\approx 0.004 \pm 0.0008$) for $T_s$ from ambient to 200 °C, and then tends to decrease for higher temperatures and $\alpha_{\text{eff}}$ is $0.002 \pm 0.0004$ at $T_s = 300$ °C. For the 80 nm thick sample, although there is much scatter in the data points, the $\alpha_{\text{eff}}$ seems to increase with $T_s$ from $\alpha_{\text{eff}} = 0.001 \pm 0.0002$ at $T_s = $ ambient to $\alpha_{\text{eff}} = 0.002 \pm 0.0004$. It should be pointed out that the sample fabricated at $T_s = 150$ °C has $\alpha_{\text{eff}} = 0.0007 \pm 0.0002$, in agreement within an error with the value reported [5]. As pointed out by Li et al. [8], there can be a contribution from eddy current even in relatively thin films.

An estimation of damping parameter contribution of eddy current loss in a 80 nm thick cobalt film is about 0.001, and therefore in the present study its contribution to the measured effective damping parameter may not be negligible for the 80 nm thick samples. However, due to the quadratic dependence on the film thickness this contribution is negligible for the 10 nm thick film. Angle dependent measurements of the $H_{\text{res}}$es and $\Delta H$ were also performed, showing a four fold symmetry for the resonance field, which is consistent with the in-plane XRD measurements, with the easy axis along the $\langle 100 \rangle_{\text{FeCoAl}}$.

### 4.3.2 Measurement temperature dependence

Figure 4.5 shows the temperature dependence of $\alpha_{\text{eff}}$ for the 10 nm thick films with different growth temperatures $T_s$, together with the data of permalloy [9] and YIG [10] thin films reported. For all the samples under consideration, the $\alpha_{\text{eff}}$ are nearly constant.
Figure 4.5: Measurement temperature dependence of effective damping parameter $\alpha_{\text{eff}}$ for Fe$_{73}$Co$_{25}$Al$_2$ alloy thin films deposited onto MgO (100) substrate at ambient, 100 and 200 °C with 10 nm thickness. Also shown are the results of permalloy thin films \cite{9} (□) and of YIG thin films \cite{10} (△).
(\(\alpha_{\text{eff}} = 0.004 \pm 0.0008\)) over a temperature range from 10 to 300 K for the three different \(T_s\) (ambient, 100 and 200 °C). On the other hand, the permalloy and the YIG thin films exhibit the decrease with decreasing temperature, although the permalloy film shows a slight increase for a range from 100 to 50 K. The present result of \(\alpha_{\text{eff}}\) vs. \(T\) is at variance with those results. It is noted that in the present study an increasing non-linearity of the frequency dependence of the linewidth at low growth temperatures was observed, therefore it is likely that extrinsic contributions such as two-magnon scattering dominate the observed temperature dependence of effective damping and linewidth [6]. Although further studies are necessary, the present result of the \(\alpha_{\text{eff}}\) which is insensitive to temperature suggests that a thin (around 10 nm thick) Fe\(_{73}\)Co\(_{25}\)Al\(_2\) alloy thin film may be useful for high frequency device applications.

### 4.4 Summary

The growth- and measurement-temperature dependences of soft magnetic properties and effective damping parameters of Fe\(_{73}\)Co\(_{25}\)Al\(_2\) alloy thin films are discussed. The saturation magnetization \(M_s\) is about 1,600 emu/cm\(^3\), independent of thickness and the substrate deposition temperature \(T_s\). Coercivity \(H_c\) is found to decrease with \(T_s\) up to around 100 ∼ 200 °C, which is probably caused by reducing the stress in the film.

The effective damping parameter \(\alpha_{\text{eff}}\) measured by ferromagnetic resonance (FMR) over a frequency range from 12 to 66 GHz at room temperature and over a temperature range from 10 to 300 K. For the 10 nm thick sample the effective damping parameter is nearly constant (\(\approx 0.004 \pm 0.0008\)) for \(T_s\) from ambient to 200 °C, and then tends to decrease for higher temperatures and \(\alpha_{\text{eff}}\) is 0.002 ± 0.0004 at \(T_s = 300\) °C. For the 80 nm thick sample, the \(\alpha_{\text{eff}}\) seems to increase with \(T_s\) from \(\alpha_{\text{eff}} = 0.001 \pm 0.0002\) at \(T_s = \text{ambient}\) to \(\alpha_{\text{eff}} = 0.002 \pm 0.0004\). As pointed out by Li et al. [8], there can be a contribution from eddy currents even in relatively thin films. However, due to its quadratic thickness dependence eddy currents do not contribute significantly to the linewidth of the 10 nm film.
The temperature dependence of $\alpha_{\text{eff}}$ obtained for a frequency range from 10 to 40 GHz for the 10 nm films with the different growth temperatures $T_s$ (ambient, 100 and 200 °C) shows that the $\alpha_{\text{eff}}$ is nearly constant ($\alpha_{\text{eff}} = 0.004 \pm 0.0008$) over a temperature range from 10 to 300 K. Together with an increasing non-linearity of the frequency dependence of the linewidth at low growth temperatures, extrinsic contributions such as two-magnon scattering are likely responsible for the observed absence of a temperature dependence of the effective damping and linewidth.

4.5 Acknowledgements

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4.6 References


Fe$_{69}$Co$_{26}$Si$_5$ alloy thin films of various thicknesses were deposited on MgO(100) single crystal substrates by magnetron sputtering at 230 °C substrate temperature. The thickness dependence of the dynamic magnetic properties was investigated using broadband ferromagnetic resonance (FMR). The X-ray diffraction (XRD) results indicate that all films are of the bcc structure with an in-plane epitaxial alignment of [100]$_{\text{FeCoSi}}$ // [110]$_{\text{MgO}}$. The exchange constant was determined from the field shift between the uniform precession FMR mode and the first order perpendicular standing spin wave (PSSW) resonance mode in the FMR spectra. The effective damping parameter decreases dramatically with increasing film thickness up to 16 nm due to the decrease of the spin pumping contribution and then remains relatively constant as the film thickness increases. In-plane angle dependent FMR measurements reveal that the in-plane anisotropy of these films are dominated by a four-fold magnetic anisotropy, which increases sharply with increasing film
thickness up to 16 nm and then shows a slightly decreasing trend as the film thickness increases. In-plane angular dependence of the FMR linewidth shows a strong two-magnon scattering contribution.

5.1 Introduction

High frequency applications such as magnetic recording head and antenna require soft magnetic materials with high saturation magnetization, high permeability, low coercivity and low damping parameter. Fe-Co based alloys such as Fe-Co-Al, Fe-Co-Si and Fe-Co-Si-Al were investigated by several groups both in bulk and thin films because of their high saturation magnetization and low coercivity.\cite{1-5} So far, very few studies on the magnetization dynamics of these materials can be found in the literature. However, understanding of the magnetization dynamics of soft magnetic thin film is essential for improving their high frequency performance. Both quasi-static and dynamic magnetic properties of (FeCo)-Al alloy thin films as functions of Al content, film thickness and growth temperature have been studied.\cite{6-8} An effective damping parameter as low as 0.0004 was found for an 83 nm thick Fe$_{73}$Co$_{25}$Al$_2$ thin film, which is in good agreement with the value reported by Schoen et al.\cite{9} for Fe$_{75}$Co$_{25}$ alloy thin film. In this paper, a systematic study on the thickness dependence of the dynamic magnetic properties of Fe$_{69}$Co$_{26}$Si$_5$ alloy thin films is presented, additional information about the quasi-static properties of these films can be found in reference \cite{10}.

5.2 Experimental Method

Multilayers of [Fe(0.3 nm)/Fe$_{66}$Co$_{34}$(1.0 nm)/Si(0.1 nm)] $\times$ N were deposited on MgO(100) single crystal substrates using a DC magnetron sputtering system. The chamber was pumped down to a base pressure lower than $2 \times 10^{-7}$ Torr and the films were sputtered using 4 mTorr of Ar pressure. The deposition rates for Fe, Fe$_{66}$Co$_{34}$, and Si were 0.12, 0.15, and 0.026 nm/s, respectively. The substrate temperature during the deposition was
230 °C. A 5 nm thick Ru capping layer was deposited for protection purposes. A small in-plane magnetic field of 50 Oe was applied during the deposition along the (110) direction of Fe$_{69}$Co$_{26}$Si$_5$.

The film thicknesses $t$ were determined by X-ray reflectivity. The structural properties were characterized by x-ray diffraction (XRD) with Cu K$_\alpha$ radiation. The morphology of the films including the alloying of the stack was studied using cross-sectional TEM/EDX of the films.[10] The quasi-static magnetic properties were measured using a vibrating sample magnetometer (VSM) and a magneto-optical Kerr effect (MOKE) system and have been reported in reference [10]. The dynamic properties were determined using broadband ferromagnetic resonance (FMR) covering a frequency range from 12 to 66 GHz. In-plane angle dependent FMR measurements were carried out at 30 GHz to determine the in-plane anisotropy of the samples.

5.3 Results and Discussion

5.3.1 Structural and Quasi-Static Magnetic Properties

Figure 5.1 shows the XRD patterns for Fe$_{69}$Co$_{26}$Si$_5$ thin films with various thicknesses. The films show a broad (200) bcc peak around $\theta = 65^\circ$, the corresponding (110) bcc peak of the Fe$_{69}$Co$_{26}$Si$_5$ film and the (200) peak of the MgO substrate are so close that only one peak can be observed around $\theta = 43^\circ$. The low angle in-plane XRD pattern of the 60 nm thick sample shows four (200) bcc peaks separated by 90°. The (200) peaks of MgO substrate are shifted by 45° with respect to the (200) peaks of the sample film, which indicates that [110]$_{\text{FeCoSi}}$ // [100]$_{\text{MgO}}$.

The Hysteresis loops measured along [110], [100] or [110] directions are shown in Fig. 5.2(a). The observed hysteresis curves are consistent with a four fold in-plane anisotropy with easy axes along the (100) directions. The thickness dependence of coercivity $H_c$, saturation magnetization $M_s$ and the remanence to saturation magnetization ratio $M_r/M_s$ are shown in Fig. 5.2(b). $H_c$ significantly increases from 4 to 16 nm and decreases with
Figure 5.1: (Color online) XRD patterns for Fe$_{69}$Co$_{26}$Si$_{5}$ thin films with various thicknesses. Inset: low angle in-plane XRD patterns for the 60 nm thick film (black line in the top part) and MgO substrate (red line in the bottom part).
Figure 5.2: (Color online) (a) Hysteresis loops for Fe\textsubscript{69}Co\textsubscript{26}Si\textsubscript{5} thin films with various thicknesses measured by VSM. The magnetic field is applied in the [1\overline{1}0], [100] or [110] direction. (b) Thickness dependence of coercivity $H_c$, saturation magnetization $M_s$ and the remanence to saturation magnetization ratio $M_r/M_s$ for Fe\textsubscript{69}Co\textsubscript{26}Si\textsubscript{5} thin films. All the values are measured along [110]
further increasing thickness. $M_s$ on the other hand slightly increase from 4 to 16 nm and only slightly decreases with further increasing thickness. $M_r/M_s$ shows a slight decrease with increasing thickness across the whole thickness range. The details of the quasi-static magnetic properties are discussed elsewhere.[10]

5.3.2 Dynamic Magnetic Properties

5.3.2.1 FMR Theoretical Model

Figure 5.3: (Color online) Sketch of the in-plane angle dependent FMR measurements geometry. All magnetic fields and the magnetization $\mathbf{M}$ are in the film plane. The external field $\mathbf{H}$ is applied along the direction of angle $\phi_H$. The microwave field $\mathbf{h}_{mw}$ is perpendicular to the external field. $\hat{e}_u$ and $\hat{e}_4$ represent the directions of the easy axes of the uniaxial and four-fold anisotropy field, respectively. $\phi_u$ and $\phi_4$ stand for the in-plane angles of the easy axis of the uniaxial and four-fold anisotropy with the x-axis, respectively. The equilibrium in-plane angle of the magnetization is $\phi_M$.

The coordinates we use in this paper are shown in Fig. 5.3. When the external magnetic field is applied along an arbitrary in-plane angle $\phi_H$, the equilibrium in-plane angle of the magnetization is denoted as $\phi_M$. In our model we include a uniaxial anisotropy field $H_u = \frac{2K_u}{M_s}$ and a four-fold anisotropy field $H_4 = \frac{4K_4}{M_s}$ with easy axes along in-plane angles $\phi_u$ and $\phi_4$, respectively. $K_u$ and $K_4$ stand for the uniaxial and four-fold anisotropy constants, respectively. The dispersion relation for the uniform precession FMR mode in
such a system is given by \[11\]

\[
\left( \frac{f}{\gamma'} \right)^2 = H_a \cdot H_b,
\]

with

\[
H_a = H_{\text{res}} \cos(\phi_M - \phi_H) + \frac{H_A}{2} \cos 4(\phi_M - \phi_4)
+ H_u \cos 2(\phi_M - \phi_u),
\]

\[
H_b = H_{\text{res}} \cos(\phi_M - \phi_H) + \frac{H_A}{8} [3 + \cos 4(\phi_M - \phi_4)]
+ \frac{H_u}{2} [1 + \cos 2(\phi_M - \phi_u)] + 4\pi M_{\text{eff}},
\]

where \( f \) is the frequency of the microwave field, \( \gamma' = \frac{g \mu_B}{h} \) is the gyromagnetic ratio, \( H_{\text{res}} \) is the resonance field, \( 4\pi M_{\text{eff}} = 4\pi M_s - K_\perp \) is the effective magnetization and \( K_\perp \) accounts for any perpendicular anisotropy present in the films. We did our broadband FMR measurements along the \([1\bar{1}0]\) direction, which is the in-plane hard axis. To obtain a simpler equation for the analysis of the broadband FMR measurements, the uniaxial anisotropy is ignored \( (H_u = 0) \) since it is much weaker compared to the four-fold anisotropy. Because the external field \( H \) is applied along the hard axis and exceeds the saturation field in the broadband FMR measurements, we have \( \phi_M - \phi_H = 0 \), thus \( \cos 4(\phi_M - \phi_4) = -1 \).

Substituting the above conditions into Eq. (5.1), we arrive at a simplified equation for broadband FMR measurements along the hard axis as follows

\[
\left( \frac{f}{\gamma'} \right)^2 = \left( H_{\text{res}} - \frac{H_A}{2} \right) \cdot \left( H_{\text{res}} + \frac{H_A}{4} + 4\pi M_{\text{eff}} \right).
\]

Perpendicular standing spin waves (PSSW) are commonly observed in FMR experiments. The dispersion relation for spin waves can be written in a similar form as Eq. (5.1): \[12–14\]

\[
\left( \frac{f}{\gamma'} \right)^2 = (H_a + H_{\text{ex}}) \cdot (H_b + H_{\text{ex}}),
\]

with the exchange field \( H_{\text{ex}} = \frac{2A}{M_s} \left( \frac{n\pi}{T} \right)^2 \), where \( A \) is the exchange constant, \( n \) is the spin
wave mode number and $t$ is the film thickness. Similarly, Eq. (5.3) can be simplified to

$$
\left( \frac{f}{\gamma'} \right)^2 = \left( H_{\text{res}} - \frac{H_4}{2} + H_{\text{ex}} \right) \cdot \left( H_{\text{res}} + \frac{H_4}{4} + 4\pi M_{\text{eff}} + H_{\text{ex}} \right),
$$

when the external field is along an in-plane hard axis.

### 5.3.2.2 FMR Measurement Results

Figure 5.4: (Color online) (a) FMR spectrum at 40 GHz and (b) Kittel plot for the 82 nm thick Fe$_{69}$Co$_{26}$Si$_{5}$ thin film measured along [1\bar{1}0]. Red lines correspond to fits to the data using Eq. (5.4) with $n = 0$ and $n = 1$. (c) Estimation of exchange constant $A$ for Fe$_{69}$Co$_{26}$Si$_{5}$ thin films except the thinnest one.

A representative FMR spectrum of the 82 nm thick film measured at 40 GHz along the [1\bar{1}0] direction is shown in Fig. 5.4(a). Two resonances are visible in this spectrum. The
stronger one at 7.1 kOe is the FMR mode and the weaker one at 6.6 kOe is the first order PSSW mode. This PSSW mode can be observed for all samples except the thinnest one, where the expected large field separation and small signal strength of the PSSW mode prevented its detection. The field dependence of the resonance frequency for the 82 nm thick film is shown in Fig. 5.4(b). By fitting the two modes simultaneously using Eq. (5.4), we can obtain the exchange constant $A$. The extracted exchange constants range from 15 to 25 pJ/m (see Fig. 5.4 (c)), which is comparable with the reported values of Fe$_{65}$Co$_{35}$ (17 pJ/m)[15] and Co$_2$FeSi (31.5 pJ/m).[16] The variations in $A$ are probably due to the finite spin pinning at the surface caused by an additional surface anisotropy and magnetization inhomogeneity close to the interfaces of the film.[17, 18]

Figure 5.5: (Color online) (a) FMR linewidth as a function of resonance frequency $f$ for Fe$_{69}$Co$_{26}$Si$_5$ thin films with various thicknesses. Lines correspond to fits to the data. (b) Thickness dependence of effective damping parameter. The upper limits are determined from the highest frequency data points and the origin.

Figure 5.5(a) shows the linewidth as a function of microwave frequency, which for all samples shows a linear relationship. The linewidth of the thinnest sample is significantly larger than for the other four samples. The frequency dependencies of the linewidth in the figure are fitted using equation[19–22]

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

(5.5)
where $\Delta H_0$ is the inhomogeneous broadening and $\alpha_{\text{eff}}$ is the effective damping parameter. The extracted $\alpha_{\text{eff}}$ are shown in Fig. 5.5(b). The upper limit of the effective damping parameter, as indicated by the asymmetric error bars, can be determined by assuming that the linewidth at the highest frequency is caused solely by a Gilbert-like linewidth contribution. This approach provides a conservative estimate for the upper limit of the error margins of the effective damping parameter even for cases where two-magnon scattering or inhomogeneous broadening contribute significantly to the linewidth. The effective damping parameter decreases drastically from the 4 nm to the 16 nm thick film. Then it stays in the range between 0.002 and 0.003. The effective damping parameter we extracted here contains both intrinsic and extrinsic contributions such as two-magnon scattering,[23, 24] eddy-current damping,[25, 26] and radiative damping.[26, 27] The intrinsic damping will be lower than the effective damping, especially those of the thicker samples, which suffer from a larger eddy-current damping.

The significant increase of the effective damping parameter increase for the 4 nm thick film has two potential origins: two-magnon scattering and spin pumping. A rough estimation of the spin pumping contribution to the effective parameter for the 4 nm thick film using equation[28] $\alpha_{\text{sp}} = \frac{\gamma' \hbar}{2M_s^2} g_{\uparrow\downarrow}$ gives a value of $\alpha_{\text{sp}} \approx 0.01$. Here $\hbar$ is the Planck constant and $g_{\uparrow\downarrow}$ is the spin mixing conductance. In this estimate we have assumed that the 5 nm Ru cap layer is a perfect spin sink, which based on reported spin diffusion lengths[29] appears to be a reasonable assumption. So far no spin mixing conductance values have been reported for FeCoSi/Ru bilayer systems in the literature. However, the reported $g_{\uparrow\downarrow}$ values for similar ferromagnet/non-magnetic metal bilayer systems such as CoFeB/Pt ($4 \times 10^{15}$ cm$^{-2}$)[30] NiFe/Ru ($3.8 \times 10^{15}$ cm$^{-2}$)[28] and CoFe/Pt ($4.0 \times 10^{15}$ cm$^{-2}$)[31] are very close despite the material differences in each system. Thus we expect the spin mixing conductance of our system to be of the same order and used $g_{\uparrow\downarrow} \approx 4.0 \times 10^{15}$ cm$^{-2}$ in our estimate. Since the estimated spin pumping contribution of the 4 nm thick film is of the same order as the measured effective damping parameter, the
two-magnon scattering is unlikely to be responsible for the huge increase of the effective damping parameter of the 4 nm thick film. As will be shown in the following paragraphs, this interpretation is consistent with our results from in-plane angle dependent FMR measurements. In fact, the measurement direction for the broadband FMR investigations was chosen to minimize the two-magnon scattering contribution.

The result of the quasi-static magnetization reversal and broadband FMR measurements suggest the presence of an in-plane anisotropy. In order to get a better understanding of the anisotropy in these films, in-plane angle dependent FMR measurements were carried out. Figure 5.6(a) exemplary shows the in-plane angular dependence of the resonance field and linewidth for the 82 nm thick film. The resonance field and the linewidth show a clear four-fold symmetry, which is consistent with the crystal symmetry of Fe₉₉Co₂₆Si₅ and its epitaxial growth on MgO. In angular dependent FMR measurements, minima of the resonance field indicate an easy axis, whereas maxima correspond to hard axes. Therefore, the easy axes of the four-fold anisotropy are along the ⟨100⟩ directions of Fe₉₉Co₂₆Si₅, consistent with the quasi-static magnetic properties, see figure 5.2 and reference [10]. Close inspection of the difference of the residual using a purely four-fold fit, i.e. the angular dependence of the measured resonance field and a fit using only a four-fold anisotropy term in Eq. (5.1), reveals the presence of a very small uniaxial anisotropy in the films. The easy axis of the small uniaxial anisotropy is not aligned with the easy axis of the four-fold anisotropy, furthermore the easy axis varies from sample to sample. Such a small uniaxial anisotropy can for example be caused by the presence of a slight inclination during deposition [32].

Angles where the resonance field has maxima are minima for the linewidth and vice versa. To quantitatively analyze the anisotropies, we fit the resonance fields of the FMR and PSSW modes using Eq. (5.3) with n = 0 and n = 1, respectively. The extracted values of $H_u$ and $H_4$ as a function of the film thickness are shown in Fig. 5.6(b). The uniaxial anisotropy fields are smaller than 25 Oe for all samples and do not show a systematic
thickness dependence. The dominating four-fold anisotropy increases sharply from 4 nm to 16 nm then shows a decreasing trend as the film thickness increases. This kind of four-fold anisotropy thickness dependency was also observed in epitaxial Fe(001) thin films and can be explained using the Néel’s pair model.[33] In this model the non-monotonous thickness dependence of the four-fold anisotropy is caused by an interfacial contribution due to the broken symmetry and lattice misfit strain that influences the anisotropy through magneto-elastic coupling.[33, 34] The differences between the anisotropy fields determined form the FMR mode and those determined from PSSW mode can be attributed to an inhomogeneous four-fold anisotropy across the film thickness. Due to the different mode profiles for the FMR and PSSW modes the two modes weigh the anisotropy distribution across the film thickness differently, leading to the observed differences in the measured anisotropy fields. It is worth pointing out that inhomogeneities of the crystalline anisotropy can also be a source of two-magnon scattering, that is not interfacial in nature, see for example references [20, 35]. A broad distribution of magnetic inhomogeneities, as indicated by the differences between FMR and PSSW modes, in turn will lead to a broad frequency range over which the two-magnon contribution to the linewidth is approximately linear with frequency, cp. Fig. 5.5.

The in-plane angular dependence of the FMR linewidth can be analyzed by considering the different contributions:[36–39] Gilbert damping, two-magnon scattering, linewidth broadening due to mosaicity, and inhomogeneous linewidth broadening. The inhomogeneous linewidth broadening caused by the fluctuation of the strength of the anisotropy fields is angle independent.[37, 40] The in-plane angular dependence of the linewidth contributions from the field drag effect and the linewidth broadening due to mosaicity for a four-fold anisotropy have an eight-fold symmetry.[36] This leaves two-magnon scattering as the source of the observed four-fold in-plane angular dependence of the linewidth. For two-magnon scattering, misfit dislocations [41] in the crystalline structures, inhomogeneities of the crystalline anisotropy[20, 35] and interfacial
Figure 5.6: (Color online) (a) In-plane angular dependence of the resonance field and linewidth for the 82 nm thick Fe$_{69}$Co$_{26}$Si$_{5}$ film at 30 GHz. The solid squares and open circles represent the FMR mode and PSSW mode, respectively. Red lines in the upper plot correspond to fits to the data using Eq. (5.3) with n = 0 and n = 1; red lines in the bottom plot correspond to fits to the data using Eq. (5.6). (b) Thickness dependence of the uniaxial anisotropy and four-fold anisotropy. (c) Thickness dependence of the anisotropic linewidth contribution from two-magnon scattering.
contributions due to roughness are all expected to reflect the crystal symmetry and hence show a four-fold in-plane angular dependence. Therefore, the in-plane angular dependence of the linewidth is fitted with[39]

\[
\Delta H = \Delta H_{iso} + \Delta H_{2m} \cos^2 \left[ 2 (\phi - \phi_{2m}) \right],
\]

(5.6)

where \( \Delta H_{iso} \) represents all isotropic contributions including inhomogeneous broadening, spin pumping and Gilbert damping; \( \Delta H_{2m} \) represents the anisotropic contribution from two-magnon scattering; \( \phi_{2m} = 45^\circ \) represents the in-plane angle where the strength of the two-magnon scattering is maximum. The extracted values of \( \Delta H_{2m} \) are plotted in Fig. 5.6 (c) as a function of the film thickness. From the figure, one can see that \( \Delta H_{2m} \) generally decreases with increasing film thickness. For a two-magnon scattering contribution of strictly interfacial origin, one expects this linewidth contribution to scale like the square of the inverse film thickness.[9, 18, 23, 26, 42] However, for this sample series, such a dependence is not observed. In particular, the thickest film exhibits a significantly larger \( \Delta H_{2m} \) than what one would expect based on the rest of the series. This suggest that in addition to the interfacial two-magnon scattering contribution, another contribution dominates the linewidth at larger film thicknesses. Two-magnon scattering from misfit dislocations[41] and inhomogeneities of the crystalline anisotropy[20, 35] are possible mechanisms that are not interfacial in origin and thus do not necessarily decrease with increasing film thickness. Both will have the same symmetry as the crystal lattice, consistent with the observed four-fold symmetry of the linewidth. In particular, one expects the density of misfit dislocations to increase with increasing film thickness above a critical film thickness \( t_{c} \)[43] consistent with our observation of a larger two-magnon contribution for the thickest film.
5.4 Summary

The quasi-static and dynamic magnetic properties of Fe$_{69}$Co$_{26}$Si$_5$ alloy thin films of various thicknesses were studied using VSM and broadband FMR. The quasi-static magnetic properties such as saturation magnetization, coercivity and the remanence to saturation magnetization ratio all show only a weak thickness dependence, for more details see reference [10]. The dynamic measurements using broadband FMR and in-plane angle dependent measurements unambiguously show that [100] and [110] are the easy and hard axes respectively of the dominant four-fold anisotropy. The FMR measurements further enabled us to quantify the strength of the four-fold anisotropy. We also observed a very small in-plane uniaxial anisotropy. Based on the field shift between the FMR and the PSSW modes, the exchange constant of these films is estimated to be 20 ± 5 pJ/m. A strong four-fold in-plane anisotropy and a weak uniaxial anisotropy are observed in in-plane rotation FMR measurements. Both the effective damping parameter and the four-fold anisotropy show strong thickness dependence when the film thickness is below 16 nm. Finally, a four-fold symmetry was also observed in the linewidth vs. in-plane angle plot, which can be attributed two-magnon scattering.

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5.6 References


6 CONCLUSION

This dissertation presents an extensive study of the magnetic properties of (FeCo)-Al and (FeCo)-Si alloy thin films. In chapter 2, a series of FeAl thin films with various Al percentage is studied first. It is found that these films have bcc structure and columnar growth morphology perpendicular to the film plane. The addition of Al decreases the coercivity, which is probably due to the reduced column width. The minimum value of coercivity is observed at 1.5% Al atomic percentage. To improve the soft magnetic properties, especially the saturation magnetization, of FeAl thin film, Co is added to the system and a study of (FeCo)-Al is carried out. The saturation magnetization increases as the atomic percentage of Co increases up to about 25% as expected. The maximum value is about 1800 emu/cm$^3$. In addition, the effective damping parameter decreases as the atomic percentage of Co increases and reaches a minimum value of 0.002 at around 25% Co atomic percentage as well. However, the coercivity significantly increases as the the atomic percentage of Co increases. The likely reason for this is the increase of stress induced magnetic anisotropy through magneto-elastic coupling due to the increase of magnetostriction constant.

The thin films in chapter 2 are all deposited at room temperature and have the same thickness. The thickness dependence study of the magnetic properties of Fe$_{73}$Co$_{25}$Al$_2$ thin films deposited on fused silica and MgO(100) substrates at room temperature is presented in chapter 3. The films show a similar columnar growth morphology as the films in chapter 2. The saturation magnetization shows no significant thickness dependence and is around 1550 emu/cm$^3$ for all films. On the other hand, both the coercivity and the effective damping parameter show a strong thickness dependence. The coercivity first increases with
increasing film thickness up to about 50 nm then slightly decreases. The increase of coercivity is again likely due to the stress induced anisotropy. The decrease beyond 50 nm indicates that a domain wall structure transition from Néel type to Bloch type occurs. The effective damping parameter show a non-monotonous dependence on the thickness and reaches minimum at 83 nm. The value of minimum effective damping parameter is 0.0004 and 0.0006 for film deposited on fused silica and MgO(100), respectively.

In chapter 4, 10 nm and 80 nm thick Fe\textsubscript{73}Co\textsubscript{25}Al\textsubscript{2} films deposited on MgO(100) at various growth temperatures are investigated. The saturation magnetization shows no growth temperature dependence. The coercivity, as predicted in chapter 2, decreases at higher growth temperature due to reduced stress in the films. The minimum coercivity is observed in the range between 100 and 200 °C. The effective damping parameter does not show a clear growth temperature dependence. An almost constant effective damping parameter is obtained in FMR measurements carried out over a temperature range from 10 K to 300 K for the 10 nm thick film series.

In chapter 5, the thickness dependence of the magnetic properties of (FeCo)-Si alloy thin films is studied. A strong four-fold anisotropy is revealed by in-plane angle dependent FMR measurements. Both the effective damping parameter and the four-fold anisotropy are highly dependent on the thickness when the film thickness is smaller than 16 nm. The thickness dependence of the four-fold anisotropy is caused by surface anisotropy and lattice misfit strain. The thickness dependence of effective damping parameter is mainly attributed to the strong spin pumping effect at small film thickness. Furthermore, two-magnon scattering induced four-fold symmetry is observed for the in-plane angle dependent linewidth. The origin of the two-magnon scattering is found to be not pure interfacial.

The results in this dissertation show the great potential for (FeCo)-Al and (FeCo)-Si as soft magnetic material candidates for future high frequency applications. Further exploration regarding the deposition conditions are needed to further optimized the magnetic properties in all different aspects. It is also of interest to investigate the influence
of sample microstructure on the magnetic properties since all the samples studied in this dissertation are single crystals. The study of (FeCo)-Si thin films in chapter 5 reveals many interesting physics in this system. Further theoretical and experimental studies on the anisotropic damping behavior may provide more insights on the relaxation mechanisms in such system.
REFERENCES


