Materials-based Control of Ultrafast Relaxation in Ferromagnetic Thin Films

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ABSTRACT

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As data rates in magnetic information storage approach 1GHz and above, strategies to control the magnetization dynamics in films become a more pressing need. Materials-based techniques to control relaxation can offer a straightforward implementation for this purpose.

Strategies to both increase and decrease the damping constant in ferromagnetic thin films are described in this thesis. By doping rare earth elements, the damping constant of Ni$_{81}$Fe$_{19}$ (Permalloy) can be greatly increased. The increased damping correlates well to the magnetic states of the rare earths. Precessional frequency can also be tuned independently in this system.

Fe has the lowest damping constant of all elemental ferromagnets. We demonstrate that by doping V into pure Fe, the damping constant can be further reduced. The decrease in the damping constant is closely related to the reduced magnetic anisotropy in the system.

The results of this thesis will provide more freedom in engineering the GHz response of the magnetoelectronic devices.
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Chapter 1

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Chapter 2

Introduction

2.1 Magnetization dynamics

When a magnetic field is applied to magnetic material, the magnetization of the material will align with the applied field. How does magnetization behave in this process? What controls the rate of magnetization change? Magnetization dynamics study this process and answer these questions.

At equilibrium, the direction of magnetization \( M_s \) in ferromagnetic material is always parallel to that of the effective field \( H_{\text{eff}} \) applied to the material. If \( H_{\text{eff}} \) suddenly changes its direction, there will be a torque \( T \) acting on \( M_s \),

\[
T = M_s \times H_{\text{eff}} \quad (2.1)
\]

which will cause the change in the momentum \( L \),

\[
\frac{dL}{dt} = T \quad (2.2)
\]
There is a relationship between $\mathbf{M}_s$ and $\mathbf{T}$,

$$\mathbf{M}_s = -\gamma \mathbf{L}$$  \hfill (2.3)

$$\frac{d\mathbf{M}_s}{dt} = -\gamma \mathbf{T}$$  \hfill (2.4)

in which $\gamma = g \frac{e}{2m_e}$ is the gyromagnetic ratio. $g$ is the spectroscopic splitting factor. For $g = 2$, we have $\gamma = 1.758 \times 10^7$ Oe$^{-1}$s$^{-1}$. From (2.1)-(2.4) we can obtain the equation of motion,

$$\frac{d\mathbf{M}_s}{dt} = -\gamma \mathbf{M}_s \times \mathbf{H}_{\text{eff}}$$  \hfill (2.5)

Eq. (2.5) shows that with the torque $\vec{\tau}$ acting on $\mathbf{M}_s$, $\mathbf{M}_s$ will precess around the axis of $\mathbf{H}_{\text{eff}}$, as shown in Fig. 2.1 (a). This process is called Larmor precession.

![Figure 2.1: The precession of $\mathbf{M}_s$ (a) without damping (Larmor precession); (b) with damping.](image)

Eq. (2.5) describes the magnetization precession without damping, which has the Larmor angular frequency $\omega_0 = \gamma \mathbf{H}_{\text{eff}}$. $\mathbf{M}_s$ rotates around $\mathbf{H}_{\text{eff}}$ eternally. In reality this case will never happen because damping always exists in ferromagnetic material, which causes the energy of the precession dissipated. The precessional frequency will be less than the Larmor frequency and the angle between $\mathbf{M}_s$ and $\mathbf{H}_{\text{eff}}$ will become smaller and smaller.
The precession ends after a number of cycles when $\mathbf{M}_s$ aligns in the direction of $\mathbf{H}_\text{eff}$. Fig. 2.1 (b) shows the magnetization precession with damping.

### 2.2 Ferromagnetic dynamical equations

There are two widely known equations describing the magnetization precession with damping. The older form is the Landau-Lifshitz equation\[^1\]:

$$\frac{d\mathbf{M}_s}{dt} = -\gamma \mathbf{M}_s \times \mathbf{H}_\text{eff} - \frac{\lambda}{M_s^2} \mathbf{M}_s \times (\mathbf{M}_s \times \mathbf{H}_\text{eff})$$  \hspace{1cm} (2.6)

$\lambda$ is the damping parameter, called relaxation rate, which has a unit of s$^{-1}$. The more recent one is the Landau-Lifshitz-Gilbert equation\[^2\]:

$$\frac{d\mathbf{M}_s}{dt} = -\gamma \mathbf{M}_s \times \mathbf{H}_\text{eff} + \frac{\alpha}{M_s} (\mathbf{M}_s \times \frac{d\mathbf{M}_s}{dt})$$  \hspace{1cm} (2.7)

$\alpha$ is the damping constant (with no dimension). Both equations are termed phenomenological since the damping terms do not follow from basic principles. Fig. 2.2 (a) and (b) shows the vector terms in Eq. 2.6 and 2.7 respectively, which are visualized on the surface of the sphere of radius $|\mathbf{M}_s|$. On the right side of both 2.6 and 2.7, the first term (azimuthal component) corresponds to undamped precession and the second term corresponding to the ”damping torque” $T_D$, which makes $\mathbf{M}_s$ move towards the direction of $\mathbf{H}_\text{eff}$. The difference is that in Landau-Lifshitz form, the damping is purely a polar component while in Gilbert form, it contains both azimuthal and polar components.

The separation into orthogonal azimuthal and polar parts in Landau-Lifshitz form greatly facilitates mathematical analysis and is partly responsible for its popularity. In this case, Gilbert form is less convenient. However, in the limit of very large $\lambda$, Landau-Lifshitz form predicts that the $\frac{d\mathbf{M}_s}{dt}$ increases and thus the switching time decreases as $\lambda$
increases, which is a physically implausible situation\cite{3, 4}. It is thus concluded that the Landau-Lifshitz form is only valid in the small damping case. Gilbert form, on the other hand, predicts that low damping ($\alpha \ll 1$) produces almost pure precession with many cycles, so the switching time is long; high damping ($\alpha \gg 1$) almost results in no precession, but the switching time is long too because the "friction" to magnetization motion is too large. The intermediate damping ($\alpha = 1$) gives the fastest switching. This condition is called critical damping condition. A time constant $\tau$ is used to describe the relaxation time, which can be seen later in Eq. \ref{eq:2.20}. The larger $\tau$ is, the longer the relaxation will take. Fig. \ref{fig:2.3} plots the change of $\tau$ as a function of damping constant $\alpha$, which reveals the minima in $\tau$ at an intermediate $\alpha$ value\cite{5}.

The observed damping constant $\alpha$ for magnetic materials is generally small, in the range between 0.01 and 0.1\cite{6}. In this regime ($\alpha \ll 1$), Eq. \ref{eq:2.7} is approximately equivalent to Eq. \ref{eq:2.6} with $\alpha = \frac{1}{7M_s}$\cite{7}.
2.3 Magnetization dynamics in thin films

2.3.1 Fast switching in thin films

One of the most important requirements of magnetoelectronic devices is a very fast switching time. Geometry becomes a key consideration in this matter because of the extreme importance of demagnetizing energy in the magnetic reversal process. Magnetization reversal by sequential process of domain wall motion is time-consuming, which can be avoided in thin films[4]. The required character is obtained by introducing an internal uniaxial anisotropy by magnetic anneal. The film becomes a single domain oriented in the annealing field direction and simultaneous reversal of the magnetization of the film is anticipated. It has been reported that the switching time of thin film metal films is very close to the theoretical limit $10^{-9}$ sec as long as the available field is at most a few oersteds[5].

A typical application for this fast switching is the magnetic random-access memory (MRAM). Current MRAM prototypes rely on magnetic field pulses to switch between two stable states of the magnetic device[8]. The strong out-of-plane demagnetizing field causes the magnetization to rotate, mostly in plane, between opposite stable states of the spin valve.
Recently an ultrafast switching was demonstrated in small spin valves using a subnanosecond field pulse applied along the magnetization hard axis. The process requires a large enough field and short enough rise time for the magnetization to overshoot the hard axis during the application of the pulse. During the first oscillation cycle, the short duration field pulse, which turns off while the magnetization is near the opposite end of the spin valve, will cause the device to switch. A slightly longer field pulse will allow the magnetization to rotate back towards the initial state, resulting in a no-switch event after the field turns off.

Kittel equation describes the effects of specimen shape on resonance frequency. For an applied field of $H(x, y, z)$, the internal magnetic field $H^i$ in the sample with magnetization $M(x, y, z)$ is

$$
H^i_x = H_x - N_x M_x,
$$
$$
H^i_y = H_y - N_y M_y,
$$
$$
H^i_z = H_z - N_z M_z,
$$

where $N(x, y, z)$ is the demagnetizing factor of the sample. Suppose the applied field is applied in $\hat{z}$ direction ($H_z = H$). To the first order approximation, we have $M_z = 0$ and $M_z = M$. According to the equation of motion (2.5), we have

$$
\frac{dM_x}{dt} = -\gamma(M_y H^i_z - M_z H^i_y) = -\gamma M_y [H + (N_y - N_z)M],
$$
$$
\frac{dM_y}{dt} = -\gamma(M_z H^i_x - M_x H^i_z) = -\gamma M_x [H + (N_x - N_z)M].
$$

(2.9)

Solutions to Eq. dMx-dMy with time dependence $exp(-i\omega t)$ exist only if

$$
\begin{vmatrix}
  i\omega & -\gamma [H + (N_y - N_z)M] \\
  \gamma [H + (N_x - N_z)M] & i\omega
\end{vmatrix} = 0
$$

(2.10)
The ferromagnetic resonance frequency $\omega_p$ is extracted as

$$\omega_p^2 = \gamma^2 [H + (N_y - N_z)M][H + (N_x - N_z)M] \quad (2.11)$$

If $H$ is parallel to the plane of a thin film, $xz$ plane, with $N_x = N_z = 0$ and $N_y = 4\pi$, $\omega_p$ becomes

$$\omega = \gamma [H(H + 4\pi M)]^{1/2} = \gamma (H^2 + 4\pi HM)^{1/2}, \quad (2.12)$$

which is the well-known Kittel equation.

### 2.3.2 Dynamic equations in spherical coordinates

If a spherical coordinate system is chosen in which $\phi$ describes the rotation of the magnetization in the plane of the film and $\theta$ describes the rotation of the magnetization normal to the plane of the film ($\theta = 0$ for in-plane magnetization and $\theta = \frac{\pi}{2}$ for magnetization completely out of plane), the Landau-Lifshitz-Gilbert (LLG) equation has the form

$$\frac{d\theta}{dt} = \frac{\gamma}{1 + \alpha^2} (H_{\phi} + \alpha H_{\theta}),$$

$$\frac{d\phi}{dt} = \frac{\gamma}{\sin \theta (1 + \alpha^2)} (\alpha H_{\phi} - H_{\theta}), \quad (2.13)$$

where $H_{\phi}$ and $H_{\theta}$ are the components of $H_{\text{eff}}$ in the spherical coordinate system.

We can also express the LLG equation by the free energy density $U$ instead of $H_{\phi}$ and $H_{\theta}$. Recall that

$$\mathbf{T} = \mathbf{M} \times \mathbf{H} = \hat{r} \times (-\nabla U) = \frac{1}{\sin \theta} \frac{\partial U}{\partial \phi} \hat{\theta} - \frac{\partial U}{\partial \theta} \hat{\phi} \quad (2.14)$$

The LLG equation then becomes[10]

$$\frac{d\theta}{dt} \cos \theta = \frac{\gamma}{M_s} \frac{\partial U}{\partial \phi} + \alpha \frac{d\phi}{dt} \cos^2 \theta,$$
\[- \frac{d\phi}{dt} \cos \theta = \frac{\gamma}{M_s} \frac{\partial U}{\partial \theta} + \alpha \frac{d\theta}{dt} \quad (2.15)\]

With the same process, the Landau-Lifshitz has the form

\[\frac{d\theta}{dt} = \gamma (H_\phi + \frac{\lambda}{M_s} H_\theta), \]
\[\frac{d\phi}{dt} = \frac{\lambda}{M_s \sin \theta} H_\phi - \frac{\gamma}{\sin \theta} H_\theta, \quad (2.16)\]

or[4]

\[- \frac{d\phi}{dt} \cos \theta = \frac{\gamma}{M_s} \frac{\partial U}{\partial \phi} - \frac{\lambda}{M_s^2} \frac{\partial U}{\partial \theta} \cos \theta, \]
\[- \frac{d\phi}{dt} \cos \theta = \frac{\gamma}{M_s} \frac{\partial U}{\partial \theta} + \frac{\lambda}{M_s^2} \frac{\partial U}{\partial \phi} \cos \theta \quad (2.17)\]

The free energy density \( U \) includes the magnetic anisotropy energy (MAE), the Zeeman energy, and the demagnetizing energy. For a magnetic thin film, because of the large demagnetization field in out-of-plane direction, the magnetization will be mostly suppressed within the film plane, and we can make approximation that \( \sin \theta = \theta \) and \( \cos \theta = 1 \). We can also assume the applied field \( \ll 4\pi M_s \). Then Eq. [2.17] can be written as[11, 12]

\[\frac{d\phi}{dt} = \frac{\gamma}{4\pi M_s} \theta - \frac{\lambda}{4\pi M_s^2} \frac{\partial U}{\partial \phi}, \quad (2.18)\]

For thin ferromagnetic film where \( M_s \gg H_k \) ("soft" ferromagnet) and \( \lambda \ll \gamma M_s \) ("underdamped" oscillator), by differentiation and substitution within Eq. [2.18] a second-order differential equation in \( \phi \) alone can be obtained as[4, 13]

\[\frac{d^2\phi}{dt^2} + 4\pi \frac{\lambda}{M_s} \frac{d\phi}{dt} + 4\pi \gamma^2 \frac{\partial U}{\partial \phi} = 0 \quad (2.19)\]
Figure 2.4: The oscillation of magnetization angle with time in ferromagnetic thin films under step-field pulses.

Eq. 2.19 leads to a simple solution for switching response. For thin films under step-field pulses in the film plane, solving Eq. 2.19 leads to an exponentially damped sinusoidal solution\cite{11, 14, 15}

\[
\phi(t) = \phi_0 + \beta_0 \sin(\omega_p t + \varphi)e^{-t/\tau},
\]

where \(\phi_0\) is the equilibrium angle, \(\beta_0\) and \(\varphi\) are the parameters, and \(\omega_p\) is the precessional frequency. \(\tau\) is the switching time, same as shown in Fig. 2.3. It can be related to the relaxation rate as \(\lambda = \frac{1}{2\pi\tau}\). Fig. 2.4 shows the oscillation of magnetization angle with time. A field step is applied at \(t=0\), which rotates the magnetization direction to a relatively large angle. After some damped sinusoidal oscillations, the magnetization stops at a new equilibrium angle \(\phi_0\).

2.4 Observation and measurement of magnetization dynamics

One key to better understanding of magnetization dynamics is to observe the magnetization reversal experimentally and make a meaningful correlation with that deduced from the phenomenological equations of motion of the magnetization. There have been different methods to achieve this goal.
2.4.1 Inductive method

Since the 1950s, the reversal time has been measured directly by induction method[16] [12] [17]. The thin films are first magnetized in a certain direction, then a magnetic field as a step pulse is applied in the opposite direction. When the direction of magnetization in the thin film reverses, a voltage pulse is induced in a pick-up coil, which is amplified and observed on an oscilloscope. The switching time is defined as the time interval which begins when the switching field has increased to the coercive force and ends when the induced voltage has diminished to 10% of its peak value. The pulse rise time is in nanoseconds range in these measurements. In the 1960’s, Dietrich, Proebster, and Wolf first measured switching speeds of 1 ns using the inductive technique[18] [19]. The field impulses were applied transverse to the easy axis of the film and the pulse widths were less than 350 ps.

As densities and speeds of magnetic devices are pushed to the limit (as in high-speed ultrahigh density magnetic recording), the interest in physics of magnetization processes in nanostructured magnetic materials has become more intense than ever before. Direct experimentation on dynamic phenomena has become more advanced, with shorter pulse rise time and more detailed process tracing. High speed inductive techniques for the measurement of ultrafast magnetic phenomena in thin film materials have been advanced by Silva et al. Innovations in their work include modern high-speed sampling technology, lithographic waveguide fabrication, and digital signal processing[11] [20]. The technique has been named as PIMM (pulsed inductive microwave magnetometry). Fig. 2.5 presents a schematic representation of the arrangement.

A commercial solid-state pulse generator produces the magnetic field step with rise time of 50 ps. A lithographically patterned coplanar waveguide is used to deliver field pulses to the magnetic film, which is deposited and subsequently lithographically patterned onto the waveguide structure in the center of the waveguide. The new technique applies the time-domain transmission geometry instead of a separate inductive pick-up coil. The transmitted
pulse is detected by a high-speed sampling oscilloscope. A helmholtz coil pair is used to apply a variable longitudinal bias field to the sample. With the magnetic sample saturated by a large transverse field, a reference wave form in which no magnetic activity has occurred is acquired and subsequently subtracted from the previously acquired wave forms, allowing the observation of precessional effects. PIMM method is relatively simple and intuitive both in design and in use. For permalloy, switching times as short as 200 ps were measured by PIMM\textsuperscript{11}.

2.4.2 Time-resolved MOKE and SHMOKE

Another technique to study magnetization dynamics employs time-resolved magneto-optical Kerr effect (MOKE) microscopy, in which the combined picosecond temporal and submicrometer spatial resolutions allow one to directly study the time dependence of magnetic
excitations and acquire "snapshot" magnetic maps of the sample surface. The observed nonuniform spatial profiles are not easily expected from electrical measurements. The damping constant is estimated by fitting the time-domain experimental data into Landau-Lifshitz or Gilbert equation. The Gilbert damping constant $\alpha$ for permalloy is determined to be 0.008 by this method. The optical measurements in this technique are performed with picosecond pulses delivered by a synchronously pumped laser and the polar or transverse Kerr rotation monitored by a polarizing beamsplitter and differential diode detection scheme, which measures the linear MOKE signal (see Fig. 2.6). Another technique uses second-harmonic (SH) magneto-optic, whereby a sample is illuminated with light at frequency $f$ and generates light at $2f$, known as SHMOKE. The light reflecting off the sample is passed through two filters to block the fundamental beam and the second-harmonic light is detected. The nonlinear magneto-optical effects are limited to sites without inversion symmetry, and SHMOKE is therefore surface- or interface-sensitive. Due to its unique sensitivity, SHMOKE may be of importance in characterizing the magnetization dynamics at film surfaces and interfaces.

2.4.3 Ferromagnetic resonance

Another powerful technique to study magnetization dynamics is ferromagnetic resonance (FMR), where a small transverse microwave (rf) field excitation causes coherent precession of the magnetic moments around a constant effective field composed of internal and external field contributions. The energy of the rf field is absorbed when the rf-frequency coincides with the precession frequency. FMR technique was developed in 1940s by Griffiths and Kittel. Theoretically, the resonance frequency and the linewidth can be determined from Landau-Lifshitz or Gilbert equation, 

$$\omega_{\text{res}} = \frac{\gamma M_s}{\sin \theta} \left( \frac{\partial^2 U}{\partial \theta^2} \frac{\partial^2 U}{\partial \phi^2} - \frac{\partial^2 U}{\partial \theta \partial \phi} \right)^{1/2}$$

(2.21)
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Figure 2.6: Schematic layout of time-resolved MOKE (after Freeman et al.). A fast rise time magnetic field "pump" is used to excite the magnetization on a picosecond time scale and an ultrashort optical "probe" to sample the magnetization at an instant of time.

\[
\Delta \omega = \frac{G}{M_s^2} \left( \frac{\partial^2 U}{\partial \theta^2} + \frac{\partial^2 U}{\partial \phi^2} \frac{1}{\sin^2 \theta} \right)
\]

(2.22)

G is the Gilbert damping parameter, \( G = \alpha \gamma M_s \).

A simple diagram of the FMR spectrometer is shown in Fig. 2.7[30] [31]. The thin film sample is placed in a microwave cylindrical resonant cavity. Microwaves with certain frequency travel down a waveguide to the cavity. The microwave power is coupled into the cavity through a small hole in the upper end wall of the cavity and some of the energy is absorbed by the specimen and the cavity wall. The reflected microwave power is directed by a microwave directional coupler to a diode that is used to detect the FMR signal. The applied field is weakly modulated at a low frequency and a lock-in amplifier is used to detect and amplify the corresponding ac component of the diode voltage. The measured signal is
then proportional to the derivative of the reflected microwave electric field amplitude with respect to the sweeping field.

Figure 2.7: A block diagram of an FMR spectrometer (after Heinrich et al.). Microwaves of certain frequency travel down a waveguide to the cavity, inside which the thin film sample is located. Reflected power is directed by a microwave coupler and detected by a diode. A low frequency modulation and a lock-in amplifier are used to monitor the field derivative of the reflected power.

Previous studies of FMR linewidth in magnetic materials showed that the dependence of the FMR linewidth on the microwave frequency follows a linear dependence\[32, 33, 34]\n
\[
\Delta H = \Delta H_0 + 1.16 \frac{\omega}{\gamma M_s} G
\]

from which the Gilbert damping parameter can be determined. $\Delta H_0$ is the frequency independent linewidth which arises from the presence of magnetic inhomogeneities. FMR research indicates that Fe has the narrowest FMR linewidth and smallest relaxation rate for magnetic precession of any of the 3d transition metal magnetic materials. Values of
the Gilbert relaxation rate $G$ obtained from FMR linewidth reported for pure Fe at room temperature cover a relatively large range, though, from $0.4 \times 10^8$ to $1.3 \times 10^8$ Hz$^3$. The large uncertainty in the damping parameter arises from exchange conductivity analysis and associated uncertainties. FMR at 35 GHz on Fe single crystal film grown by MBE shows a linewidth of 45 Oe, which represents the narrowest 35 GHz ferromagnetic resonance linewidth ever measured in single crystal Fe. This value approaches the theoretical lower limit set by Landau-Lifshitz broadening in Fe$^3$. Recently, an FMR linewidth at 9.5 GHz as low as 15 Oe was obtained for polycrystalline Cu/Fe/Cu thin film structure, with the nonintrinsic contribution of only a few Oe$^3$.

The large magnetic field used in FMR causes the specimen to be magnetized as a single domain, and thereby simplifies the interpretation of the experimental results. FMR is particularly well suited to the study of thin films of conducting ferromagnetic materials because of the small skin depth of the microwave fields used in the experiments.

2.5 Damping mechanisms in ferromagnetic materials

Damping, like electrical resistivity, consists of intrinsic contributions, always present in a particular material, and extrinsic contributions, which can be suppressed through control of microstructure.

As seen in Fig. 2.8, both electrical resistivity and relaxation describe the return of electronic configurations to equilibrium. If an electric (magnetic) field is applied to a metallic system and switched off at $t = 0$, the electrical current (transverse moment) decays to zero as $\exp(-t/\tau)$, where $\tau^{-1}$ is the relaxation rate. $\tau^{-1}$ describes a scattering process out of one specified state and into any other.

The temperature dependence of resistivity $\rho(T)$ is well described by a linear relationship

$$\rho(T) = \rho_0 + \kappa T$$  (2.24)
where $\rho_0$ is a temperature-independent contribution to $\rho$, describing extrinsic scattering processes, and $\kappa$ is the constant of proportionality, describing intrinsic scattering processes. Extrinsic scattering can vary from one sample to another, depending upon preparation. It arises from microstructural imperfections or finite geometry. It can, in principle at least, be suppressed. Intrinsic scattering is present for a perfect single crystal at a given temperature; it comes about mostly through interaction with lattice vibrations (phonons). Intrinsic scattering cannot be suppressed.

An experimental technique to separate intrinsic from extrinsic damping parallels the temperature dependence of resistivity. Here, frequency $\omega$ takes the place of temperature, and scattering $\tau^{-1}$ is measured through the field-swept ferromagnetic resonance linewidth $\Delta H = \gamma^{-1}\tau^{-1}$.

As seen in Eq. [2.23], the linewidth broadening comes from two kinds of contributions. One is the inhomogeneous linewidth broadening, which is caused by magnetic inhomogeneities in the sample. These inhomogeneities come from structural defects and complex
geometrical features in the sample and they can in principle be avoided. Thus, damping from these contributions is called extrinsic damping. The other kind of damping comes from unavoidable contributions, which should be called intrinsic damping. For example, at finite temperatures phonons and magnons can not be avoided. The magnetic relaxation processes which involve the electron scattering with phonons and thermally excited magnons can be called intrinsic because they are integral part of the system\[38\].

2.5.1 Intrinsic damping mechanism

Microscopic theories have been developed for the intrinsic damping in metals. Existing treatments agree with each other in broad terms\[39, 40, 41, 38, 42\] in the sense that mobile electrons, combined with spin-orbit coupling, are the actors which first transfer energy out of the spin system. There has been a revival of interest in the theory of damping in the past several years\[29, 43, 44, 45, 46, 47\] due in part to its technological relevance in spin electronics.

Formal considerations

Many of the treatments are largely formal, starting with a particle picture for damping. Precession describes a spin wave with infinite wavelength (uniform mode). These spin waves are quantized and represented as magnons, bearing a single $\pm \mu_B$ of spin moment. The number of uniform-mode magnons excited determines, proportionally, the precessional cone angles $\theta$. Damping is the process where uniform mode ($q = 0$) magnons are destroyed, closing the cone angle to zero.

In a metal, many free electrons are available to collide with magnons; in certain configurations, the magnon is annihilated in scattering. Scattering has to conserve total (spin) angular momentum. The most basic damping process then involves the collision of a spin-up electron ($k, \uparrow$) with a $q = 0$ magnon, flipping its spin ($k, \downarrow$) and absorbing the energy of
the magnon \( \hbar \omega \) (4.1 \( \mu \text{eV/GHz} \)). In the absence of spin-orbit coupling, all selection rules which allow the scattering are not easily satisfied. Spin and \( \mathbf{k} \) are conserved, but an empty minority state must be available \( \hbar \omega \) above the initial majority state to allow energy to be conserved. This is usually impossible because the exchange splitting is \( \sim 1 \text{ eV} \).

Formal treatments agree that spin-orbit coupling enables relaxation, in ordinary (spin-conserving) scattering as well as in spin-flip scattering. Ordinary scattering is confined to electronic states within either \( \uparrow \) and \( \downarrow \) subbands; spin-flip scattering crosses from one subband to the other. The two types of process lead to separate temperature dependences for \( \lambda \).\[^{[40]}\]

Since 1970’s it has been shown that the intrinsic magnetic relaxation in metals is caused by incoherent scattering of electron-hole pair excitations by phonons and magnons. The electron-hole interactions involve three particle scattering, as shown in Fig.\[^{[2.9]}\][48]. The excitations are either accompanied by electron spin flip or the spin remains unchanged. Spin-flip excitations (\( \sigma = +\frac{1}{2}, \sigma' = -\frac{1}{2} \)) can be caused by the exchange interaction between magnons and itinerant electrons (s-d exchange interaction), during which the total angular momentum is conserved. The spin conserving scattering (\( \sigma = \sigma' \)) is caused by spin-orbit interaction which leads to a dynamic redistribution of electrons in the electron \( \mathbf{k} \)-momentum space. The Gilbert damping can be calculated using Fermi’s golden rule, which sums up all available states that satisfy the energy conservation. The incoherent scattering by phonons can be accounted for by broadening the energy conservation into a Lorenzian:

\[
\frac{\hbar}{\tau_{\text{eff}}} = \frac{\hbar/\tau_{\text{eff}}}{\left(\hbar \omega_q + \varepsilon_{k,\sigma} - \varepsilon_{k+q,\sigma'}\right)^2 + \left(\hbar/\tau_{\text{eff}}\right)^2}
\]

(2.25)

where the relaxation rate, \( 1/\tau_{\text{eff}} \), describes the incoherent scattering with phonons and magnons.

For the spin-flip excitations (subscript \( \text{eff} \) becomes \( \text{flip} \)) the difference in electron energy, \( \varepsilon_{k,\sigma} - \varepsilon_{k+q,\sigma'} \), is given by the exchange interaction energy which is much bigger than \( \hbar/\tau_{\text{flip}} \).
Figure 2.9: Spin wave with energy $\hbar \omega_q$ collides with an itinerant electron with energy $\varepsilon_{k,\sigma}$ ($\sigma$ represents the spin state), and creates an itinerant electron with momentum $k+q$ and spin orientation $\sigma'$.

In this case the second term in the denominator is negligible and Eq. 2.25 is proportional to $\tau_{\text{flip}}^{-1}$. The lifetime of the spin-flip electron-hole pair, $\tau_{\text{flip}}$, is enhanced compared to the orbital relaxation time, $\tau_{\text{orb}}$, because spin-orbit interaction is needed to flip the electron spin by phonons. Elliot [49] showed that $\tau_{\text{flip}} = \tau_{\text{orb}}/\Delta g^2$, where $\Delta g$ is the enhancement of the $g$ factor from the spin-orbit interaction. In this case the Gilbert damping is proportional to $\tau_{\text{orb}}^{-1}$ and consequently should be proportional to the sample's resistivity, which is proportional to $\tau_{\text{orb}}^{-1}$ and increases with increasing temperature. In reality, a large distribution of the energy gap $\varepsilon_{k,\sigma} - \varepsilon_{k+q,\sigma'}$ modifies the overall temperature dependence and the spin-flip excitations can be expected to be dependent on resistivity only at low temperatures [50]. At high temperatures $\hbar/\tau_{\text{flip}}$ becomes comparable to the energy gap, which results in a gradual saturation of the damping caused by spin-flip excitations with increasing temperatures.

For electron spin conserving collisions the change in the electron energy can involve a small wave number $q$ of the FMR magnon and can be neglected compared to $\hbar/\tau_{\text{orb}}$. Eq. 2.25 is proportional to $\tau_{\text{orb}}^{-1}$. In this case the Gilbert damping is proportional to the sample’s conductivity. It increases with decreasing temperature as the mean free path becomes longer. Kambersky made a model corresponding to this damping mechanism based on the observation that the Fermi surface changes with the direction of the magnetization [40].
As the precession of the magnetization evolves in time and space the Fermi surface also distorts periodically in time and space. The effort of electrons to repopulate the changing Fermi surface is delayed by a finite relaxation time of electrons and this results in a phase lag between the Fermi surface distortions and precessing magnetization. This is a typical scenario for frictional damping.

In both cases, a direct dependence of damping on spin orbit coupling is predicted. Kambersky\textsuperscript{[40]} finds

$$\lambda \sim (g_{\text{eff}} - 2)^2$$  \hspace{1cm} (2.26)

where $g_{\text{eff}}$ is the effective $g$ factor, reflecting pure spin type magnetism for $g = 2$. Two regimes are found for the damping: for low temperature, controlled by the ordinary scattering and breathing Fermi surface mechanism, $\lambda \propto \Delta g^2 \tau_{\text{orb}}$. For high $T$, $\lambda \propto \Delta g^2 / \tau_{\text{orb}}$. At room temperature, both terms are of roughly equal weight.

The Kambersky theory is successful on two counts. First, it explains the broad minimum in $\lambda$ observed at room temperature, increasing at higher and lower temperatures. At low temperatures, the ordinary scattering dominates ($\propto \tau_{\text{orb}}$); at high temperatures, spin-flip scattering dominates ($\propto \tau_{\text{orb}}^{-1}$). Second, the theory explains why intrinsic relaxation is proportional to frequency (Gilbert type.) A number of electrons proportional to $\bar{\hbar} \omega$ can be involved in the scattering: initial states are available to $E_F - \hbar \omega$.\textsuperscript{[42]} Experimentally, in Ni the Gilbert damping was found significantly increased when approaching the cryogenic range of temperatures and saturated below 50 K\textsuperscript{[51]}. Another experiment showed that in high-purity single-crystal slabs of Ni the Gilbert damping below RT was well described by the two terms which were equal in strength and proportional to the conductivity and resistivity\textsuperscript{[52]}. This is in good agreement with the predictions from both spin-flip and spin conservation mechanisms.

The model has several failings, however. For example, the temperature dependence of $\lambda$
in Ni is given by a form \( \lambda(T) = A(T/300K)^2 + B(300K/T)^2 \).\(^{43}\) As the (phonon) scattering rate \( \tau_{\text{orb}}^{-1} \) is proportional to \( T \) at moderate temperature, the Kambersky model does not describe the correct power law. Secondly, a recent extension of this model using realistic band structure\(^{43}\) fails to predict the correct trend in \( \lambda \) between Fe, Co, and Ni.

**Eddy current**

In a ferromagnetic material which is electrically conducting, any change in the magnetization induces eddy currents which tend to oppose this change, and thus provide a damping mechanism. When the magnetization of a thin ferromagnetic film changes very rapidly (in GHz range), the field induced within the film is 90° out of phase with the magnetization in a direction opposing the rotation, and the field within the turn at distance \( x \) from the center is given by\(^{53}\)

\[
H(x) = 4\pi M_s \frac{d^2 - x^2}{c^2} \sigma \frac{d\phi}{dt} \tag{2.27}
\]

in which \( \sigma \) is the electrical conductivity, \( c \) is the velocity of light in free space, \( d \) is the film thickness, and \( \phi \) is the in-plane angle of the magnetization. The mean damping torque \( T_d \) per unit volume of the film is then given by

\[
T_d = \int_{-d/2}^{d/2} H(x) M dx = \frac{1}{12} \sigma c^2 (4\pi M_s)^2 \frac{d\phi}{dt} \tag{2.28}
\]

The effect of eddy current damping is to add to the left-hand side of Eq. 2.19 an additional term proportional to \( \frac{d\phi}{dt} \) given by Eq. 2.28 multiplied by \( 4\pi \gamma^2 \):

\[
\frac{d^2 \phi}{dt^2} + 4\pi [\lambda_0 + \frac{1}{12} \sigma c^2 (4\pi \gamma M_s)^2] \frac{d\phi}{dt} + 4\pi \gamma^2 \frac{\partial U}{\partial \phi} = 0 \tag{2.29}
\]
where $\lambda_0$ is the hypothetical intrinsic damping parameter not due to eddy currents. The damping due to eddy currents can therefore be described by an effective $\lambda_e$ given by

$$
\lambda_e = \frac{1}{12} \frac{d^2}{c^2} (4\pi \gamma M_s)^2.
$$

(2.30)

According to Eq. 2.30, $\lambda_e$ increases in proportional to $d^2$, which means eddy current damping will play an increasingly important role as film thickness increases. For ultrathin film thickness, the role of eddy current is negligible; $\lambda_e$ becomes comparable to the intrinsic damping without eddy current contribution at an intermediate thickness; in the thick film limit, the FMR line broadening by eddy current is proportional to $\sqrt{\sigma A}$, where $A$ is the exchange constant\cite{38,54}. The spatial distribution of the rf magnetization in the skin depth is altered by the surface magnetic anisotropy, which consequently affects the FMR linewidth in the thick film limit. The contribution of eddy current and exchange coupling to the FMR linewidth thus is also referred to as the exchange-conductivity mechanism.

**Direct magnon-phonon scattering**

The magnetization relaxation by a direct magnon-phonon scattering is another possible damping mechanism. Suhl analyzed the case for samples smaller than a domain wall thickness, in which the coupling to the lattice is by direct relaxation via magnetostriction into a lattice of known elastic constant. Calculations for the magnon-phonon Gilbert damping $G_{ph}$ shows\cite{29}

$$
\frac{G_{ph}}{\gamma} = 2 \frac{(1 + \nu)^2 B_2^2}{E^2} \eta \gamma
$$

(2.31)

where $\nu$ is the Poisson ratio, $B_2$ is a measure of the magnetoelastic energy, called magnetoelastic shear constant, $E$ is the Young’s modulus, and $\eta$ is the phonon viscosity. At 1 GHz, the sound wavelength should be of order of 1 $\mu$m, just about of order of the length at which wall formation becomes energetically feasible. For samples in excess of this size, both
sound propagation and nonuniform magnetization fields must be considered. In that case, the loss can occur as the result of twisting of the magnetization vector, as in domain wall motion. There have been some theoretical calculations on the effect of magnetoelasticity on the FMR linewidth, but experimentally this kind of phonon resonance is hard to establish in real samples.

2.5.2 Extrinsic damping mechanism

two-magnon mechanism

Two-magnon scattering is one very important source of relaxation in materials containing magnetic inhomogeneities. Surface roughness, grain boundaries and atomic disorder are potentially important sources of the scattering. The basic idea is that such inhomogeneities result in a coupling between the otherwise orthogonal uniform precession and degenerate spin-wave modes and that energy transfer out of the uniform precession to the degenerate modes is important in the initial stages of relaxation. The total number of magnons is unchanged since one magnon is annihilated and another is created. The interaction is sensitive to the nature of the inhomogeneity. As a general rule, the coupling is large for spin-wave wavelengths greater than the dimensions of the inhomogeneity.

In the idealized FMR experiment, a uniform mode is excited whose wave vector $k_{||}$ parallel to the surface is zero. For a simple film with magnetization $M_s$ and the dc applied field $H_0$ parallel to the surface, the frequency of this mode (in the absence of anisotropy) is given by Kittel equation

$$\omega_{FM} = \gamma [H_0(H_0 + 4\pi M_s)]^{1/2}$$

In the presence of dipolar couplings between spins, there will be short wavelength spin waves degenerate with the FMR mode. Defects in the film scatter energy from the uniform modes to these states, producing relaxation of dephasing character. The spin-wave frequency is
given by \[58\]

\[
\omega^2(k_{||}) = \omega_{FM}^2 - 2\pi\gamma^2 M_s k_{||}d(H_0 - 4\pi M_s \sin^2 \phi_{k_{||}}) + \gamma^2(4\pi M_s + H_0)Dk_{||}^2,
\]

where \(d\) is the film thickness, \(\phi_{k_{||}}\) is the in-plane angle of the magnon wave vector \(k_{||}\), and \(D\) is the exchange stiffness. The scattering process is illustrated schematically in Fig. 2.10, with the dispersion relation shown in Eq. 2.33. From both Fig. 2.10 and Eq. 2.33 we can see that in the ultrathin film limit, the dipolar energy generates a term in the dispersion relation linear in the wave vector. For finite thickness and a range of propagation angles, the initial slope in the \(\omega \sim k_{||}\) plot is negative. However, the exchange leads to the positive term quadratic in wave vector in Eq. 2.33, and thus at finite wave vectors, we have modes degenerate with the uniform FMR mode.

![Figure 2.10: Graphic representation of the scattering process from a uniform mode to a mode degenerate with it, of wave vector \(k^*_{||}\). (after Arias, et al.)](image)

In two magnon scattering the magnon momentum is not conserved due to sample inhomogeneities (loss of translational invariance), but the energy is conserved. The number of degenerate magnons is proportional to \(\omega^2\) when \(\omega \rightarrow 0\), and consequently the two magnon scattering eventually decreases linearly to zero with decreasing microwave frequency. For
both 2D and 3D spin-wave manifold there are no magnons degenerate with the FMR mode in the perpendicular configuration, hence the FMR linewidth should be smaller than that in the parallel configuration\[59\]. Experiments on amorphous metal films showed that the two magnon scattering did not have to affect the slope of Eq.\[2.23\] and the zero-frequency intercept observed in plots of FMR linewidth versus frequency is indeed caused by two magnon scattering\[60\]. A convincing evidence for two magnon scattering mechanism was obtained by investigating the dependence of FMR linewidth on the angle $\theta_H$ between the dc magnetic field and the sample plane, as shown in Fig.\[2.11\]. When the magnetization is inclined from the surface, the damping decreases significantly. The calculated FMR linewidth takes the intrinsic value of the Gilbert damping. The difference between the measured FMR linewidth and that expected for the intrinsic damping is caused by two magnon scattering. The peak in the FMR linewidth for $\theta = 78^\circ$ is caused by dragging of the magnetization behind the external applied field.

![Figure 2.11: FMR linewidth $\Delta H$ for 200Pd/30Fe/GaAs(001) thin film as a function of $\Theta_H$ between the dc magnetic field and the sample plane. The dots represent the measured data and the dashed line represents the calculated linewidth using the Gilbert damping.](image)

The structural defects in ultrathin multilayers are often caused by the lattice misfit between individual layers. It was shown that the onset of the self assembled network of misfit dislocations was accompanied by the presence of a strong extrinsic damping\[38\]. The lattice
defects decrease the local symmetry and create inhomogeneous magnetic anisotropies. Their angular dependence is given by the symmetry of defects. This means that the intensity of two magnon scattering can have an explicit dependence on the direction of the magnetization with respect to the axes of magnetic defects. In FeV superlattices it was assumed that defects were caused by surface steps. The line defects are the source of local uniaxial anisotropy. The uniaxial internal effective field disappears for the magnetization oriented close to 45° away from the uniaxial axis, which indicates that in this direction the uniaxial anisotropy does not affect the internal field and two magnon scattering is switched off.

**Dry friction and hysteresis loss**

The dissipation connected with the motion of macroscopic system sometimes results from dry friction. In this mechanism, an irregular spin can depart from the path of the uniform motion and may spiral down towards its local potential minimum. This provides the quick motion which occurs even when the uniform motion is arbitrarily slow. Since the spin is exchanged coupled to its neighbors in uniform motion, it will be dragged out of its well, which represents an energy loss to the uniform motion.

In ferromagnetic resonance, it was found that in the presence of dry friction, the shape of the observed resonance line depends on the amplitude of the driving field $h$. Below some critical value of $h$ the resonance is not possible, because the total torque acting on the magnetic moment is too small and the dry friction stops the precession of the magnetization. When $h$ is just large enough so that the magnetization can rotate, the resonance line is extremely flat: its width tends to be infinity while its height tends to be zero. If $h$ is further increased, the width of the resonance line will decrease towards the value coming from intrinsic Gilbert damping whereas the height is roughly proportional to the square of $h$. Dry friction is possible in rare-earth ions for a range of the ratio between the anisotropy and interatomic exchange coupling constants.
In the case of hysteresis the collapse of Bloch wall structures fulfills this condition. If the amplitude of magnetization is very small, the hysteresis loss depends on the amplitude of magnetic field. It becomes less important in the high frequency range because the wall displacement is mostly damped in this range and is replaced by the rotation magnetization.

2.6 Strategies to control magnetization dynamics

The precessional oscillation of magnetization is the natural result of ferromagnetic resonance, an intrinsic property of all ferromagnetic materials. As a result, the total observed time of magnetization rotation from one equilibrium state into another in response to an applied magnetic field is actually much longer than the initial magnetization risetime: Damping of the oscillations typically requires multiple precessional cycles. For applications such as magnetic recording write heads, read sensors, and magnetic random-access memory (MRAM), the “ringing” presents a serious impediment to realizing fundamentally limited data transfer rates. It is pressing to develop strategies to reduce both unwanted oscillations occurring during high-speed sense operations and chaotic behavior occurring during memory-element switching.

2.6.1 Temporal control strategies

One technique to suppress resonant oscillations is the temporal tailoring of the driving magnetic field pulse. The idea is based on the principle of destructive interference when adding two coherent signals, which could be the incident pulse and back-reflection of the pulse at the short end of a coplanar waveguide. The magnetization’s time response was measured at different observation points along the waveguide, which correspond to different time separations of the two signals. Fig. 2.12 shows data measured at three locations. The upper graph exhibits a typical time domain FMR, where the separation between the two signals is virtually nonexistent. The time required for the magnetization to reach the first
peak is approximately 350 ps. By applying two signals with the adjusted delay of 380 ps between them, the FMR oscillations are completely suppressed, as seen in the middle graph.

Figure 2.12: Dynamic response of magnetization for 50 nm thick permalloy to the two coherent signals. The upper, middle, and lower graphs were obtained at spots of 0.4, 23, and 37 mm, respectively, from the shorted termination. The equivalent time delays between the incident and reflection signals are 7, 380, and 620 ps, respectively.

The magnetization dynamics can also be controlled by two independently triggered field pulses with independent field strengths[65]. Arbitrary voltages can be applied to the pulse switches and an arbitrary time delay can be adjusted between the two pulses, which enables one to shape the in-plane magnetic field pulse.
2.6.2 Damping constant adjustment and materials-based control

Importance of magnetization dynamics in spintronics

Spin electronic ("spintronic") devices rely on ferromagnetic ultrathin films to control the resistance state of the device. As shown in Fig. 2.13, trilayer structures of two ferromagnets FM$_1$, FM$_2$, separated by a nonmagnetic spacer NM, have an electrical resistance that changes up to 100% with the magnetization state of M$_1$ and M$_2$. As in an optical analogy, resistance varies from high (antiparallel M$_1$, M$_2$, or "crossed polarizers") to low (parallel M$_1$ and M$_2$). In most devices, M$_1$ is fixed, and the resistance state depends entirely on M$_2$. Data are encoded in the resistance states: M$_2$ "up" (high R) encodes a "1", and M$_2$ "down" (low R) encodes a "0."

Figure 2.13: Optical analogy of spin-electronic phenomena (after Bailey).

The switching dynamics of the free magnetic layer control the attainable data rate of the device. Precession and relaxation parameters, frequency $\omega_p$ and damping $\alpha$, define how long it takes M$_2$ to rotate through 180°. This determines the response due to a (magnetic field)
excitation: does the spintronic device roll off below 1 GHz, above 1 GHz, or closer to 10 GHz? These questions need to be answered in the context of metallic ferromagnetic thin films, since these are the FM layers of interest in spintronics for the foreseeable future. Alloys of Fe, Ni, and Co show the highest moments (to $B_{sat}=2.4\, T$), highest Curie temperatures (> 1000 ° C) lowest switching fields (<1 mT, or 10 Oe), and highest remanence of any known materials; oxide or semiconductor alternatives have some attractive features, but are not competitive in all respects.

**GHz speed limits from precession**

A fundamental limit for the speed of magnetization rotation is set by the precessional frequency, $f_p$, on the order of 2-3 GHz for Fe under normal conditions. While the precession gives the speed of rotation, the damping of the motion determines when it finally settles. The damping $\alpha$ then gives a secondary speed limit. If the characteristic settling time $2/\gamma\mu_0 M_s \alpha$ is greater than $1/f_p$, the final state is not stable before the next pulse. In magnetic recording, data rates are beginning to push towards 500 Mb/s, and the speed limit from precession is less urgent than that from damping. We will review these barriers and describe ways to extend data rates into the mid GHz.

For thin films, Precessional frequencies are much higher than would be expected from the simple Larmor equation, since the actual magnetic fields felt by the sample are not just the applied fields $\mathbf{H}$. Any canting of $\mathbf{M}$ out of the film plane creates a strong and opposite demagnetizing field $\mathbf{H}_d = -M_z \hat{z}$, restoring $\mathbf{M}$ to the film plane. Kittel was the first to realize that for FMR of arbitrary ellipsoids, shape effects are important; the thin-film simplification also bears his name:

$$f_p = \frac{\gamma}{2\pi} \sqrt{M_s (H + H_K)} \quad \text{Kittel eqn.} \quad (2.34)$$

The precessional frequency is fundamentally important to magnetization dynamics. Like
any other rising edge in time, a rising edge of magnetization (risetime \( \tau \)) cannot be faster than \( \approx 0.3/f_{\text{max}} \), where \( f_{\text{max}} \) is the cutoff (highest) frequency of the system. A ferromagnet cannot respond at a higher frequency than \( f_{\text{p}} \). The higher the frequency, the faster the rotation.

A time-domain solution can be written for the LL or LLG equation, valid for small rotation angles and single domain behavior, under a step or impulse magnetic field:

\[
\phi(t) = \phi_0 + \beta_0 e^{-\lambda t/2} \sin (\omega_{\text{p}} t + \varphi)
\]  

(2.35)

where \( \phi_0, \beta_0, \) and \( \varphi \) are constants. The magnetization angle \( \phi(t) \) converges to the new equilibrium exponentially, with a characteristic time given by \( 2/\lambda \). The higher \( \lambda \), the faster the alignment.

The benefits of increased \( \lambda \) are expected to be great for patterned structures. Schrefl and coworkers have estimated, using computational micromagnetics, the effect of damping \( \alpha \) on the risetime at the pole tip of a perpendicular recording head. They find that the risetime is fastest for \( \lambda = 160 \) GHz (listed as \( \alpha = 1 \)), far faster than that calculated for a nanostructure with a more typical value of damping \( \lambda = 3\)GHz (\( \alpha = 0.02 \)). See Fig. 2.14.

Fast dynamic response of the data layer and of the soft underlayer (SUL) is also very important for achieving high performance in magnetic recording. It was found in perpendicular recording that varying the Gilbert damping parameter in the data layer can change the sweep time of the recording process. Fig. 2.15 demonstrates the influence of magnetic dissipation on the sweep time. The plot shows that increasing the damping constant of the SUL from 0.01 to 0.1 can lead to a significantly shorter sweep time.

Materials-based tailoring can offer a both fundamental and straightforward way to implement the adjustment of the damping parameter. Dilute concentrations (dopants) of the rare-earth element Tb have been found to be very effective in manipulating damping constant of \( \text{Ni}_{81}\text{Fe}_{19} \). Fig. 2.16 shows the effect of Tb on magnetization dynamics of
Figure 2.14: The effect of the damping constant of the head material on the flux rise time of the write field. The dashed line is the signal current. The three solid lines are the write fields corresponding to damping constants of $\alpha=0.02$, 0.1, and 1, respectively, as marked in the plot.

Ni$_{81}$Fe$_{19}$. The magnetic dynamics proceed from underdamped at 0%, to nearly critically damped at 2%, to overdamped at 4% Tb. By comparison with that doped with Gd, whose magnetic moment is completely due to spin, it was inferred that the coupling of the NiFe spin system to the local orbital moment of Tb might be responsible for the increased damping.

**Size limits and importance of decreased damping**

Increased damping (dissipation) $\alpha$ is beneficial for efficient motion in precession, and therefore a reduced settling time. However, high $\alpha$ implies also a higher noise (fluctuation) in $\mathbf{M}$. Neil Smith[67] has applied the fluctuation-dissipation theorem to estimate the white noise in nanoscale spintronic devices, and compared with the experimental noise in GMR read heads. A white noise contribution, expressed as the power spectral density of an equivalent fluctuating field ($S_H$, where $\sqrt{S_H}=A/m/\sqrt{Hz}$) is found as

$$S_H = \frac{4k_B T \alpha}{\gamma M_s V}. \quad (2.36)$$
CHAPTER 2. INTRODUCTION

Figure 2.15: Dynamic coercivity as a function of the sweep time for different Gilbert damping constants in the soft underlayer in the medium. Open symbols, $\alpha = 0.01$; closed symbols, $\alpha = 0.1$; diamonds, $T=0$ K; squares, 100 K; circles, 350 K. For a fixed dynamic coercivity, increasing $\alpha$ from 0.01 to 0.1 greatly reduce the sweep time.

For ambient temperatures, the noise power increases strongly with decreasing sample volume $V$ and increasing damping $\alpha$. For small sensors at high frequencies, on the order of $400 \times 400 \times 4$ nm, the ”magnetization noise” dominates the spectrum. In this limit, the SNR ratio is predicted even to be independent of the GMR ratio $\Delta R/R$, scaling only as $M_s V/\alpha$. Low damping is critical for high SNR of small structures.

Spin-momentum transfer (SMT) switching has been proposed as a promising means to switch magnetic random access memory (MRAM) elements. Extremely tight switching threshold distributions are not needed to eliminate writing errors for adjacent elements. After some development, SMT may be a part of the best solution to the question of high data rate operation. Gilbert damping is an effective torque which acts against the action of the spin polarized currents in SMT. Therefore, the critical current $i_{\text{crit}}$ needed to switch a
Figure 2.16: Magnetization dynamics of Tb-doped Ni$_8$Fe$_{19}$ thin films (after Bailey et al.). Damping constant $\alpha$ is extracted from Landau-Lifshitz-Gilbert fitting.

Ferromagnetic layer through SMT is proportional to the damping rate $G$. Slonczewski has derived a widely used formula\[68\]

\[
i_{\text{crit}} = \frac{e}{\hbar \epsilon} \left( 6.31a^2 G \frac{H_{\text{eff}}}{\gamma} \right),
\]

where $\epsilon$ is the dimensionless efficiency factor for conversion of electric current into torque, $t$ is the time, and $a$ is the radius within which the current flows. It is clear from Eq.\[2.37\] that low power SMT switching requires low $G$.

Ferromagnetic metal thin films (e.g. Fe) have been demonstrated recently as elements for compact and lightweight GHz telecommunication devices. Fe films on GaAs waveguides have been considered in fabrication of tunable bandstop filters because of their higher magnetization and superior process integrability compared to the more conventional ferri-magnetic insulators\[69, 70\]. According to the Kittel equation, higher magnetization leads
to a higher device operating frequency with large electronic tunability.

A disadvantage of ferromagnetic thin films is their comparatively greater dissipation of RF energy during magnetization motion. This dissipation, or loss, is expressed as a reduced real permeability and/or broadening in resonant frequency linewidth in FMR experiment. Minimum reported values of the 9.8 GHz linewidth are 0.15 Oe for ferrites\textsuperscript{71}, compared with 15 Oe for ultrathin film ferromagnets\textsuperscript{72}. Therefore, reducing damping in the ferromagnets will raise the possibility of the application of ferromagnetic thin films in the telecommunication devices.

However, the materials engineering capability to make low-damping ferromagnets, with $\alpha$ reduced from that of pure materials, is not known.

\section*{2.7 This thesis}

In this thesis, materials-based strategies to both increase and decrease damping constant will be presented. The thesis is divided into two parts.

Research Part I is described in Chapter 4, which involves research on the effect of rare-earth (RE) dopants on the magnetization dynamics in Ni$_{81}$Fe$_{19}$ thin films. Introduction to Part I is described in 4.1. Review of magnetic properties and development of Ni$_{81}$Fe$_{19}$ is presented. Materials-based control in magnetization dynamics as motivation of research in part I is explained. 4.2 describes the systematical effect of RE dopants on the magnetization dynamics of RE-doped Ni$_{81}$Fe$_{19}$. In Sm, Dy, Ho, and Tb doped Ni$_{81}$Fe$_{19}$ there are all big contribution of damping from the rare earth dopants, with minimal effect on the precessional frequency. In Eu doped Ni$_{81}$Fe$_{19}$ thin film there is a big boost in resonance frequency but damping is not changed. The contributed damping by rare earth dopants can be related to the magnetic states of the free-atom RE elements. The boost of the resonance frequency correlates well to the increase of the dynamic anisotropy.

Magnetization dynamics of RE-doped/undoped Ni$_{81}$Fe$_{19}$ bilayers are described in 4.3.
A bilayer method is demonstrated to tune the precessional frequency and damping parameter separately. In Eu-doped/undoped Ni$_{81}$Fe$_{19}$ bilayers, precessional frequency can be adjusted over a large range by changing the Eu-doped/undoped thickness ratio. Strongly enhanced anisotropy in the bilayers is important to explain the boost in the frequency. In Tb-doped/undoped Ni$_{81}$Fe$_{19}$ bilayers, magnetic damping can be widely tuned by changing the bilayer thickness ratio. The results are independent of the deposition order. The results found in 4.2 and 4.3 can offer greater flexibility in engineering the GHz response of magnetoelectronic devices.

X-ray magnetic circular dichroism (XMCD) characterization of RE-doped Ni$_{81}$Fe$_{19}$ is described in 4.4. Both total electron yield (TEY) and transmission modes are used in the measurements. By applying the sum rule, orbital, spin, and total magnetic moments are extracted for each element in the RE-doped films. XMCD characterization of Tb and Gd dopants reveals a large orbital moment fraction on Tb sites, but zero orbital moment on Gd. The results provide support for the idea that spin-orbit coupling through introduction of local orbital moments is important for controlled damping from lanthanide dopants.

Chapter 5 describes research Part II, the magnetization dynamics in MgO/Fe$_{1-x}$V$_x$ epitaxial thin films. Introduction to this part is presented in 5.1. Research in Fe thin films are briefly reviewed. Motivation of addition of V into Fe to reduce the Gilbert damping is explained. FMR research on the magnetic anisotropy and relaxation rate in Fe thin films are reviewed.

The structural characterization on the MgO/Fe$_{1-x}$V$_x$ epitaxial films is described in 5.2. XRD spectra are taken with different V concentrations and high quality epitaxial structure is confirmed. 5.3 presents the static magnetic property characterization. Hysteresis loops are taken with VSM and the saturation magnetization is determined from the loops. As V is added, the magnetization decreases, which is explained by the fact that magnetic moments in V align antiparallel to those in Fe. 1 T in the magnetization with 31% is about the level
of permalloy.

Spin dynamics of MgO/Fe$_{1-x}$V$_x$ characterized by FMR are summarized in [5.4]. Close to the narrowest FMR linewidth has been realized in the MgO/Fe epitaxial film. Angular dependent FMR shows clearly the fourfold symmetry in the resonance field. As V is doped in, the crystalline magnetic anisotropy constant decreases. Above 45% V, the easy axis and hard axis exchange their crystallographic directions. The addition of V greatly reduces the Gilbert damping parameter in the epitaxial Fe thin film. Field dependent FMR shows a decrease in the frequency linewidth as V is added, which confirms the decreased Gilbert damping in the MgO/Fe$_{1-x}$V$_x$ epitaxial thin films. The results will provide a materials-based strategy to reduce the microwave loss in ferromagnets, which represents an initiative research in this field. By applying this strategy, SNR could be improved in the potential spintronic devices. The conclusion for the part II research is summarized in [5.5].
Chapter 3

Experimental techniques

3.1 Preparation of thin film samples

3.1.1 Ion beam sputtering

RE-doped Ni$_{81}$Fe$_{19}$ thin films were deposited on the oxide side of Si(500 $\mu$m)/SiO$_2$(1 $\mu$m) substrate by ion beam sputtering in a load-locked, production-oriented chamber (Veeco Millatron). The scheme of the deposition chamber is shown in Fig. 3.1. The ion source is the broad-beam Kaufman gridded type source[73]. During deposition, Ar gas is fed into the discharge chamber where its molecules are ionized. The ions are then accelerated through a pair of fine-mesh grid screens. The resulting beamlets merge into a broad beam outside the discharge chamber which then bombard the surface of the target with energetic ions. The emitted target atoms traverse the high-vacuum ambient and deposit atomistically on the substrate to form a film.

The base pressure in the main chamber is $5 \times 10^{-8}$ torr. It was found that low sputtering pressure and high sputtering power produce optimal soft magnetic properties. $1 \times 10^{-4}$ torr is the lowest pressure for which the plasma can be sustained in our chamber and thus determined to be the working pressure during deposition. The RE dopants were introduced
CHAPTER 3. EXPERIMENTAL TECHNIQUES

Figure 3.1: The scheme of ion-beam sputtering chamber. Dopants were introduced by cosputtering with rare earth foils.

into the films by cosputtering with rare earth thin foils, which are mounted on a manipulator that can be positioned in and out of the beam. The dopant concentration $x$ in deposited $(\text{Ni}_{81}\text{Fe}_{19})_{1-x}\text{RE}_x$ thin film was adjusted by raising or lowering the rare earth foil out of or into the beam prior to film deposition. The film thickness is 50 nm, with 5 nm of Ta capping layer to prevent the film surface oxidation. The resulted film structure is, therefore, Si/SiO$_2$/$(\text{Ni}_{81}\text{Fe}_{19})_{1-x}\text{RE}_x(50\text{nm})$/Ta(5nm).

An electromagnet is attached on the back of the substrate, which applies a bias field of 20 Oe to the substrate during deposition to enhance the uniaxial anisotropy. The substrate is rotated during sputtering to eliminate the anisotropic film properties other than the induced magnetic anisotropy. To control for possible ex situ surface oxidation of the targets, both Ni$_{81}$Fe$_{19}$ target and RE foil were presputtered iteratively up to 10 h till consistent dynamical characteristics were obtained for the doped films.

Ex-situ compositional analysis was conducted by x-ray photoelectron spectroscopy (XPS), provided by a separate load-locked PHI 550 spectroscopy. XPS uses a soft X-ray source to
ionize electrons from the surface of a solid sample. The binding energy of these electrons are measured and are characteristic of the elements and associated chemical bonds (chemical state) in the top few atomic layers of the material. It probes the first 5-10 nm of a surface and provides identification of all elements (except H and He) present at concentrations 0.1 atomic %. Prior to analysis, a low-energy Ar sputter clean was applied in situ to remove the surface oxide of the Ni$_{81}$Fe$_{19}$:RE films.

Doping concentrations of some samples were measured by Rutherford Backscattering Spectroscopy (RBS). This popular thin-film characterization technique relies on the use of very high energy (MeV) beams of low-mass ions such as He$^+$. These have the property of penetrating thousands of angstroms or even microns deep into films or film/substrate combinations yet with negligible sputtering of surface atoms. The projectile ions lose their energy through electronic excitation and ionization of target atoms. By measuring the number and energy of backscattered He ions, information on the nature of the elements present, their concentration, and their depth distribution can be simultaneously acquired without appreciably damaging the specimen.

The film thickness was determined from deposition rate measured by crystal monitor. The monitor functions on the basis that a built-in quartz crystal oscillator has the characteristic of changing its frequency of oscillation as its mass changes. As the target material is deposited onto the quartz crystal, its mass goes up and the frequency of oscillation goes down. This change is more or less linear and it can be easily calibrated in terms of frequency vs. deposition thickness. To determine the tooling factor, which reflects the thickness difference between material deposited on the quartz sensor and that on the substrate, some samples with the nominal thickness were measured after deposition, thus the corrected tooling factor can be calculated by $Tooling_{preset} \times \dfrac{Thickness_{actual}}{Thickness_{nominal}}$.

The film thickness was measured by Dektak 3 profilometer, which is a microprocessor based contact stylus surface profiler used for making measurements on vertical features.
ranging in height from <100 angstroms to 655,000 angstroms. A diamond-tipped stylus is placed in contact with, and then gently dragged along the surface of the substrate. The vertical movement of the stylus are sensed by a transducer, digitized and stored in the memory for plotting and data manipulation.

3.1.2 DC magnetron sputtering

Epitaxial Fe$_{1-x}$V$_x$(100) thin films were deposited by DC magnetron sputtering. Fig. 3.2 shows the diagrammatic layout of the sputtering chamber. There are three sputtering sources and one evaporation source attached to the chamber, each controlled by a separate power supply. Comparing with the ion-beam sputtering system, this DC magnetron sputtering system can provide more flexible control of the sputtered alloy compositions and lower impurity content at UHV pressures. By cosputtering with separate targets, the doping range can be as broad as possible, even from 0 to 100%.

Figure 3.2: The diagrammatic layout of the DC magnetron sputtering chamber. There are three sputtering sources and one evaporation source attached.

The magnetron configuration is the planar one with parallel target and anode electrode surface. In this geometry, a typical DC electric field of $\sim$100 V/cm is impressed between the
target (cathode) and the substrate (anode). A circular ring permanent magnet is attached on the back of the target, which can create a strong magnetic field on the target front surface. Together with a strong central magnet to complete the magnetic circuit, electron motion is confined to a so-called "racetrack". Ionization of the Ar gas is most efficient above the racetrack and the plasma is most intense there\cite{73}. High strength magnets make the sources particularly well-suited for sputtering magnetic materials, as high susceptibility ($\mu$) of the target shields the racetrack and high moment permanent magnets are needed to average the shielding. The evaporation source is the miniature RADAK I, a resistance heated vacuum evaporator. It offers an excellent evaporation of certain materials up to 1500°C. It is particularly useful in vacuum chambers where space is limited. In the described research it has been used mainly as a source for noble metal (Au or Cu) cap layers.

The base pressure in the chamber is of $1 \times 10^{-9}$ torr. The pressure during deposition was $3 \times 10^{-3}$ torr, using Ar, purified at the inlet using a getting purifier. Substrate was transported into the main chamber through a load-locked chamber. Epitaxial Fe$_{1-x}$V$_x$(100) thin films were deposited by cosputtering from confocal Fe and V targets. See Fig. 3.3. The concentration of V can be adjusted by fixing the power of Fe gun and changing the power of V gun. The studied series span the concentration range $0 \leq x_V \leq 52\%$. The metallic thin films were grown onto the polished side of $10\text{mm} \times 10\text{mm} \times 0.5\text{mm}$ MgO(100) single crystal wafers. Some of the substrates were cleaned with a water-based etch which is composed of $1\text{NH}_4\text{OH}:1\text{H}_2\text{O}_2:100\text{H}_2\text{O}$ (volumetric) made with 30% solutions for the $\text{NH}_4\text{OH}$ and $\text{H}_2\text{O}_2$\cite{74}. XRD and FMR characterization on samples deposited on cleaned or as-received substrates did not show much difference. 2 nm Ti was deposited as cap layer from the third target to prevent the thin film from oxidation. 2 nm evaporated Cu was also tried, which showed a slightly broadened FMR linewidth, indicating a slight oxidation on sample surface.

During deposition, the substrates were heated using a UHV button heater that is capable
CHAPTER 3. EXPERIMENTAL TECHNIQUES

Figure 3.3: The schematic diagram of the epitaxial MgO/FeV film deposition. Thin films are deposited by cosputtering from confocal Fe and V targets. MgO(100) single crystal substrate is heated during deposition.

of 1200°C UHV operation. Temperature calibration was performed to the heater and the substrate with separate thermo-couples at UHV pressure, and the result is shown in Fig. 3.4. According to Fig. 3.4, 800°C on the heater corresponds to 200°C on the substrate since there is a ~3 mm gap between the heater and the substrate. The substrates were heated with temperatures raging from room temperature to 300°C during deposition. XRD and FMR characterizations on the samples with different deposition temperatures showed that 200°C deposition yielded the best crystalline quality, thus determined to be the deposition temperature for all the epitaxial thin films.

The film thickness ranges from 8 to 100 nm. Tooling factors (ratios of deposited to crystal-monitor recorded film thickness) were determined for both Fe and V to ensure accurate control of stoichiometry. Because the crystal monitor is located closer to the source than the substrate, the tooling factors are both less than 1. Step measurement method was used to determine absolute thickness of deposited samples and the tooling factor. In this method, a film with a nominal thickness of 100 nm or more is deposited onto a flat substrate.
Figure 3.4: The temperature on the substrate vs. the temperature on the substrate heater. The big difference between them is caused by the ∼3 mm gap between the substrate and the heater.

which has been marked in advance with a permanent marker pen. After deposition, a ink is lifted off and the depth of the "trench" measured by a profilometer is the actual film thickness. The ratio of actual to nominal thickness is the tooling factor.

To reduce contamination or oxidation during deposition, an Ultrapure 040 (mini) gas purifier was installed in the Ar gas line. After being activated at 450°C, it can be either operated at 450°C to reduce all the impurities, including H₂O, O₂, CO, and CO₂ to sub-ppb levels in the Ar flow or operated at room temperature for removal of H₂O, O₂, and CO₂ only. Samples deposited in both cases were examined by XRD, VSM and FMR, and the results did not show much difference. Therefore, the purifier was operated at room temperature during deposition of the studied thin films because of the operation convenience.
3.2 Characterization of the epitaxial structure

The structure properties of the Fe$_{1-x}$V$_x$ thin films were characterized by x-ray diffraction (XRD) with a Scintag X2 x-ray diffractometer in the conventional Bragg-Brentano ($\theta - 2\theta$) geometry. The accelerating voltage and current were 45 kV and 35 mA, respectively. Ni filter was attached to block Cu K$\beta$ x-ray. The resulted wavelength for CuK$\alpha$ x-ray is $\lambda = 1.5406$ nm. A long-range normal scan of $5 < \theta < 120^\circ$ was performed to make sure only the expected Fe$_{1-x}$V$_x$(200) reflection is present. Next, a small-range symmetric fine scan was performed with step 0.02$^\circ$ and scan rate 0.04$^\circ$/min to determine the exact peak position.

To determine the quality of the epitaxial structure, the detector was fixed at the 2$\theta$ position of the Fe$_{1-x}$V$_x$(200) peak and rocking curve (\omega scan) of the (200) plane was taken. The full width at half maximum (FWHM) was taken from the rocking curve spectrum. This determines the dispersion of the crystallographic direction (001) with respect to the sample normal and is characteristic of the crystalline quality.

3.3 Magnetization dynamics measurement

3.3.1 Pulsed inductive microwave magnetometry (PIMM)

PIMM measurements

Magnetization dynamics of the RE-doped Ni$_{81}$Fe$_{19}$ films was investigated at 10-20 GHz bandwidth by time-domain pulsed inductive microwave magnetometry (PIMM) with step pulse field excitation. The experimental setup of PIMM magnetometry in our lab is shown in Fig. 3.5.

The thin film sample, coated with photoresist as insulating layer, is placed on the microwave coplanar waveguide, with its easy axis along the longitudinal direction of the waveguide. The step pulse is generated by Picosecond Pulse Labs Model 10050 pulse generator,
Figure 3.5: The schematic diagram of the PIMM magnetometry showing fast pulse generator, high-speed sampling oscilloscope, orthogonal magnet subsystem, and coplanar waveguide with sample on the top.

which produces 10V pulses with 45ps risetime and duration of 10ns. When a step current pulse is produced and delivered to the waveguide, it produces a pulsed Biot-Savart field in the magnetic thin film, a fringing field which is orthogonal to the film easy axis. The pulse field is fairly small in magnitude. A static bias field up to 30 Oe generated by a Helmholtz pair is applied in the easy axis direction to enhance the single-domain structure of the film.

Coplanar waveguides were lithographically fabricated on 1.9 mm thick RT/Duroid 6010 substrates with 8 µm thick Cu electrodeposited on the top as received. RT/Duroid 6010 microwave laminates are ceramic-PTFE composite. They were chosen as waveguide substrate because of their high dielectric constant ($\epsilon_r=10.2$), low loss tangent, low moisture absorption, high resistivity, and good thermal mechanical stability. Fig. 3.6 shows the sketch diagram of the waveguide, which has a 500 µm center conductor width at the two ends for convenience of electrical connection, and the width tapers to 100 µm in the middle for a length of 6 mm. The total waveguide length is 20 mm. The waveguide was designed to be 50 Ω of characteristic impedance. According to conventional design criteria for coplanar
waveguides with finite-width ground planes [25],

\[
Z_l = \frac{30}{\sqrt{\epsilon_r/2}} \left[ \ln 2 + 2 \arctan \frac{1}{T} \right] 
\]

(3.1)

\[
T = \sqrt{\frac{1 - (w/b)^2}{1 - (w/d)^2}} 
\]

(3.2)

where \(Z_l\) is the characteristic impedance. \(w, b,\) and \(d\) are the dimensions shown in Fig. 3.6.

The actual measured impedance of the lithographically fabricated waveguide was \(Z_0 \sim 55 \Omega\).

![Figure 3.6: The sketch diagram of the waveguide used in PIMM measurement.](image)

The HP 54750A high-speed sampling digitizing oscilloscope has an 18 GHz bandwidth, maximum trigger signal is 2V, and the jitter time is less than 2 ps rms. The sampling time interval can be less than 8 ps, which can generate over 600 samples over 5 ns range. During measurement, we averaged up to 4096 sampled wave forms to reduce any possible noise effect. To avoid damage to the sensitive electronics of the instrument, the input from the pulse generator was reduced by 23 dB of high-bandwidth attenuators.

Another pair of Helmholtz coils are arranged to provide the saturation field of \(\sim 110\) Oe transverse to the sample easy axis. A reference wave form with the transverse field on was obtained in which no dynamic activity occurs. This waveform is subtracted from other
acquired ones with the transverse field off, allowing the observation of precessional effects. By this method, the inductive signal is

\[ V_p = \left( \frac{\mu_0 l \delta f(z, w)}{4} \right) \left( \frac{Z_0}{Z_0 + \frac{1}{2} R_{dc}} \right) \frac{dM_y}{dt} \]  

(3.3)

where \( \mu_0 = 4\pi \times 10^{-7} \text{ Wb/A⋅m} \) is the permeability of empty space, \( l \) is waveguide length, \( \delta \) is magnetic sample thickness, \( f(z, w) \) is space loss function, \( z \) is the distance above the waveguide, \( R_{dc} \) is the dc resistance of the waveguide, \( M_y \) is the component of magnetization transverse to the longitudinal direction of the waveguide. From Eq. (3.3) one can see that the net inductive signal is proportional to \( \frac{dM_y}{dt} \).

**Landau-Lifshitz simulation of PIMM data**

The dynamic response measured by PIMM was fitted to the Landau-Lifshitz (LL) equation. The LL equation in spherical coordinates for a magnetic thin film is shown in Eq. (2.18) and we have assumed \( \psi \ll 1 \). In magnetic thin films, the free energy \( U \) can be written as

\[ U = K \sin^2 \phi - H_B M_s \cos \psi \cos \phi - H_p M_s \cos \psi \sin \phi + 4\pi M_s^2 \sin^2 \psi, \]  

(3.4)

in which \( K \) is the anisotropy constant, \( H_B \) is the longitudinal bias field, and \( H_p \) is the transverse pulse field. The first term on the right side of Eq. (3.4) is anisotropy energy associated with the induced uniaxial anisotropy. The second and third terms describe the Zeeman energy, and the last term represents the demagnetizing energy. After substitution with Eq. (3.4) Eq. (2.18) was solved numerically with Runge-Kutta method[76]. The detected inductive voltage is proportional to \( \frac{dM_y}{dt} \), thus proportional to \( \frac{d\phi}{dt} \) for small displacement angle \( \phi(t) \ll 1 \). The experimental dynamic responses were fitted to the numerical solution. \( M_s \) was set to the literature value for undoped Ni_{81}Fe_{19}, 730 kA/m, \( H_p \) to an arbitrary value of 7 Oe. The PIMM signal is uncalibrated, and the choice of \( H_p \) does not affect results. Fit
parameters were constrained to the anisotropy field $H_k$, the LL relaxation rate $\lambda$, and the inductive coupling efficiency $\nu$, which scales only the magnitude of the observed voltage. LLG fits were carried out in comparison; we find that the two model predictions of $\psi(t)$ and $\phi(t)$ agree to better than 1% numerical accuracy up to $\lambda/4\pi = 2.5$ GHz ($\alpha = 0.3$), as expected in the regime $\alpha \ll 1$.

The dynamic precession can also be fitted to the exponentially damped sinusoidal function as shown in Eq. 2.20. Both precessional frequency $f_p = \omega_p/2\pi$ and relaxation rate $\lambda = 2/\tau$ can be determined from the fitting.

In addition to time-domain fitting, we may also determine the fundamental precession frequency by transforming the data to the frequency domain by a fast Fourier transform. Frequency spectra yielded the peaks which are not apparent from the time-domain fitting, and the peak characteristic of the coherent precession was identified.

### 3.3.2 Ferromagnetic resonance (FMR) characterization

**Traditional cavity-type FMR**

The magnetization dynamics of epitaxial MgO/Fe$_{1-x}$V$_x$ thin films were characterized by ferromagnetic resonance (FMR). Fig. 3.7 shows the diagram of the tradition cavity-type FMR setup in our lab. The cavity operates in the TE$_{01n}$ mode, which has the rf electrical field circulating about the cavity’s long axis and the rf magnetic field radially symmetric and covering both of the flat end-walls\[31\] [77]. For 10 GHz cavity operation, a Wiltron 6628A sweep generator generates a microwave with frequency adjustable between 8 and 12.4 GHz. The generated microwave travels down an E&M Laboratory waveguide which is operational between 8.2 and 12.4 GHz. The microwave power is coupled into the cavity through a small hole in the upper end wall of the cavity, which is located approximately halfway between the cavity axis and the cavity wall. Some of the energy is absorbed by the specimen and the cavity wall. The reflected microwave power is directed by an HP 10dB
microwave directional coupler to an HP negative type diode crystal detector which is used to detect the FMR signal. For the 10 GHz cavity, the actual resonance frequency is \( \sim 10.29 \) GHz. With different samples, this frequency will change slightly. A frequency sweep with zero applied field was performed before each FMR measurement to find the exact resonance frequency for the cavity.

![Block diagram of the FMR measurement setup](image)

**Figure 3.7:** A block diagram of the FMR measurement setup. The resonant frequency of the cavity is 10 GHz.

The dc applied field is supplied by a pair of Varian Associate electromagnets, which can provide magnetic field up to 1 Tesla in a 2” gap. The strength of the dc field was measured by a Lakeshore 421 Gaussmeter. FMR is measured by sweeping the DC field. At certain applied field, the precessional frequency of the magnetization in the sample coincides with the rf frequency and the microwave energy is absorbed the most. That field is the resonance
The relatively few ferromagnetic atoms present in the thin film and small surface area comparing with the total surface area of the cavity cause the power absorbed by the film to be much less than the power absorbed by the cavity walls. Thus, the variation in the diode voltage with sweeping DC field is small and superimposed on a large constant background. In order to emphasize the variations of diode voltage due to specimen absorption, the applied field is weakly modulated at a low frequency in the range of 100-200 Hz by a pair of Helmholtz coils attached to the pole pieces of the electromagnet. A Stanford Research System SR810 DSP lock-in amplifier is used to detect and amplify the corresponding AC component of the diode voltage. The output of the amplifier is fed into a data collection computer. Therefore, the FMR spectrometer measures a signal that is proportional to the derivative of the reflected microwave electric field amplitude with respect to the sweeping field. The Varian Associate magnets can be rotated up to 180°, which can provide out-of-plane angular dependent FMR measurement. The sample mounting stage in the resonant cavity can also be rotated through a manipulator, which can provide in-plane angular dependent FMR measurement, from which the anisotropy constant can be extracted.

"Waveguide type FMR"

In addition to the traditional cavity-type FMR measurement, the magnetic samples were also measured by "waveguide-type" FMR, which uses either a coplanar waveguide or a k-band directional coupler waveguide instead of the cavity. Fig. 3.8 shows the top view of the FMR spectrometer that uses a coplanar waveguide instead of the cavity, which can measure FMR in the range 10 MHz-18 GHz. A Wiltron 6668B sweep generator is used to generate low-power rf microwave in the range of 10 MHz-40 GHz. The rf microwave travels down to the coplanar waveguide. The magnetic sample is placed right on the top of the center of the waveguide. Some of the microwave energy will be absorbed by the sample and the
coplanar waveguide. The transmitted microwave power is detected by an Eclipse Microwave diode with operational range 0.01-18 GHz. If attaching a waveguide coupler between the RF source and the waveguide, the reflected microwave energy can also be measured.

The dc magnetic field is provided by a Varian Associates V2800 electromagnet, which is capable of generating 0.9 T fields in a 1” gap with the existing power supply. Cooling water for the magnet was kept on during FMR measurements. The strength of the dc field was measured by a Lakeshore 421 Gaussmeter and the Gaussmeter voltage was read by a digital voltmeter which was connected to the data collection computer. The dc field is weakly modulated and the AC component of the diode voltage is detected and amplified by a Stanford Research System SR830 DSP lock-in amplifier. FMR by this type of spectrometer can also be measured by fixing the DC applied field and sweeping the microwave frequency. When the frequency coincides with the resonance frequency of the magnetization in the sample, the rf microwave energy will be absorbed the most.

Higher frequency FMR in the range of 26-40 GHz can be measured by replacing the coplanar waveguide with a DE Mornay Bonardi K-band directional coupler waveguide. Fig. 3.9 shows the diagram of the coupler waveguide with the DC field magnets. The long-axis is vertical. The sample is directly placed on the bottom of the waveguide. The RF source is connected to the ”rf in” end to send in microwaves with frequency between 26 and 40 GHz. An Anritsu 75KC50 10 MHz-40 GHz RF diode is connected to the ”rf out” end to detect the reflected microwave energy. Again, a small ac field was applied to modulate the dc field and the ac component of the diode voltage was detected and amplified by the lock-in amplifier.

Both coplanar waveguide type and K-band directional coupler waveguide type were used to measure frequency dependent FMR for the MgO/Fe$_{1-x}$V$_x$ epitaxial films, from which g factor, anisotropy field, and damping terms including extrinsic and intrinsic terms as well as the damping parameter can be extracted.
Figure 3.8: A top view block diagram of the "waveguide type" FMR spectrometer, which uses a coplanar waveguide instead of the resonant cavity. It can measure FMR at the range of 0.01-18 GHz.

3.4 Static magnetic property characterization

3.4.1 Magneto-optical Kerr effect (MOKE) magnetometry

The static magnetic properties of most RE-doped Ni$_{81}$Fe$_{19}$ thin films are investigated by magneto-optical Kerr effect (MOKE) magnetometry. Some early-stage Fe$_{1-x}$V$_x$ thin films are also measured by MOKE. Easy and hard axis hysteresis loops are taken from the measurements. Coercivity $H_c$ can be extracted from easy-axis loop and anisotropy field $H_k$ can be estimated from the hard axis loop.

Fig. 3.10 shows the experimental arrangement of the MOKE magnetometer (longitudinal
mode) in our lab. In the longitudinal mode the magnetization $M_s$ is in the plane of the film surface and parallel to the plane of optical incidence. Kerr effect can occur for radiation incident in either the P-plane (electrical field $E$ parallel to the plane of incidence) or the S-Plane ($E$ perpendicular to the plane of incidence). What happens is that radiation incident in either of these linearly polarized states is, on reflection, converted to elliptically polarized light. The major axis of the ellipse is often rotated slightly with respect to the principal plane and this is referred to as the Kerr rotation. The sign and magnitude of these effects are proportional to $M_s$ and its direction[78].

Figure 3.9: A diagram of the K-band directional coupler waveguide with the DC field magnets. It can be used to measure FMR up to 40 GHz.
Figure 3.10: A diagram illustrating the MOKE setup, based on the phenomenon that linearly polarized light rotates in the polarization axis upon reflection of a surface with finite magnetization.

3.4.2 Superconducting quantum interference device (SQUID) magnetometry

The magnetic moment $\mu_s$ of the RE-doped Ni$_{81}$Fe$_{19}$ films is measured by SQUID. Saturation magnetization $M_s$ is calculated by $M_s = \frac{\mu_s}{V}$. $V$ is the volume of the film, which is calculated from the production of the profilometric film thickness and the surface area of the sample.

SQUID measurements were performed in Prof. Uemura's laboratory. The SQUID magnetometer may be the most sensitive measurement device known to man, according to John Clarke, one of the developers of the concept[79]. It can measure extremely tiny magnetic fields, as small as $10^{-10}$ Oe. SQUID consists of two superconductors separated by thin insulating layers to form two parallel Josephson junctions, as shown in Fig. 3.11. When there is a voltage $U \neq 0$ V applied to the two superconductors, a current begins to flow because of the quantum mechanical tunnelling effect. If a constant biasing current is maintained in the SQUID device, the measured voltage oscillates with the changes in phase at the two junctions, which depends upon the change in the magnetic flux. Counting the oscillations
allows you to evaluate the flux change which has occurred. Liquid helium is needed to
remain performance of the superconductors.

Figure 3.11: A schematic diagram illustrating the SQUID magnetometer formed by the
parallel circuit of two Josephson junctions.

3.4.3 Vibrating sample magnetometry (VSM)

The static magnetic properties of most Fe\(_{1-x}V_x\) thin films were characterized by vibrating
sample magnetometry (VSM). Hysteresis loops were taken along both Fe\(_{1-x}V_x\) [100] and
[110] directions, which correspond to easy and hard axes, respectively.

Fig. 3.12 shows the diagram of the VSM magnetometer in our lab. In this diagram,
the sample is attached to the end of a rod, the other end of which is fixed to a loudspeaker
cone (mechanical vibrator). Current through the loudspeaker vibrates the rod and the
sample, which induces a magnetic flux change in the pick-up coils, and the coils will pick
up a voltage corresponding to the flux change. The pick-up voltage is proportional to the
magnetic moment of the sample. With calibration with a specimen of known \(M_s\), the value
of magnetic moments of samples will be extracted.

Hysteresis loops along both easy and hard axes in the epitaxial MgO/Fe\(_{1-x}V_x\) thin
films were taken. Hysteresis loops were also taken for a bare MgO substrate and they
were subtracted from those of MgO/Fe\(_{1-x}V_x\). A good quality single crystal Fe thin film,
Figure 3.12: The schematic diagram of a VSM magnetometer. Vibration of a magnetic sample will induce a magnetic flux change in the pick-up coils which will generate a voltage in the coils. The induced voltage is proportional to the magnetic moment of the sample.

confirmed by XRD and FMR measurements, was selected as the sample for calibration, and the magnetization of this sample is assumed to be 1715 emu/cc, the documented $M_s$ value for bulk Fe at room temperature[80].

3.5 X-ray magnetic circular dichroism

X-ray magnetic circular dichroism (XMCD) spectroscopy is a relatively new technique that probes atomic magnetic properties utilizing the absorption of circularly polarized x-rays.
XMCD can be understood most easily as a two-step process. If we take Fe as an example, in the first step, 2p core levels (L edge) acts as a source for electrons with spin polarized along the direction of x-ray polarization. The L₃ and L₂ absorption edges correspond to opposite electron spin orientations \((l \pm s)\), with different total angular momentum \((2p_{1/2} \text{ and } 2p_{3/2})\), respectively. Right-handed (RH) and left handed (LH) polarized x-rays can transfer \(+\hbar\) or \(−\hbar\) of angular momentum, respectively, to photoelectrons originating from these states; some part of the angular momentum can be transferred to the spin, through the spin-orbit interaction. At the L₃ edge, since \(l\) and \(s\) are parallel, RH polarized light generates mostly electrons with spin along the polarization axis; at the L₂ edge, where \(l\) and \(s\) are antiparallel, RH polarized light produces more electrons with spin opposite to the polarization direction.

The magnetization dependence arises in the second step, due to the spin-split electronic 3d states in 3d-ferromagnets, as depicted in Fig. 3.13. Only the minority electrons (right) have an appreciable density of empty d-states, which are the holes to scatter electrons into, in these ferromagnets. X-ray absorption does not occur unless a final state is available for the photoelectron. Thus only spin-down photoelectrons are scattered strongly, and the dominant absorption can be considered in the minority (spin-down) channel only. Dichroism then arises, opposite in sign, at the L₂ and L₃ edges: absorption is present for only RH or LH photons at either of these edges.

The uniqueness of XMCD lies in its ability to provide the elemental specificity and direct and independent measurement of spin and orbital magnetic moments. The high element selectivity is especially useful for identifying the magnetism from different specific atoms in alloys, multilayers, etc. It is an ideal tool to characterize the magnetic characters of both Ni and Fe and RE dopants. Fig. 3.14 shows the diagrammatic sketch of XMCD measurement.

In XMCD technique, a circularly polarized soft x-ray was delivered to a magnetic thin
Figure 3.13: Principle of X-ray magnetic circular dichroism (XMCD). Left: Circularly polarized x-rays are absorbed in core-to-valence transitions; the absorption energy (illustrated here for L$_{23}$ transitions) provides elemental resolution. Right: x-ray absorption spectroscopy $\alpha(E)$ is produced by transitions from filled core levels to empty valence states; the helicity dependence of the absorption $\Delta\alpha(E)$ (dichroism) provides a measure of the elemental magnetic moment. The hypothetical case of a ferromagnet with enormous orbital moment $m_{\text{orb}} = 2\mu_B/\text{atom}$ is shown. After Stöhr [81].

film. A soft x-ray absorption spectroscopy (XAS) was obtained by measuring the photon flux transmitted through or reflected from the thin film by a sensitive photodiode. For MCD measurement, a magnetic field was applied in the sample plane. The MCD data were taken by alternating between two opposite directions of a saturation electromagnetic field at each photon energy level. If there is a nonzero spin or orbital moment in the valence band, these two absorption spectra will be different from each other. This difference is the MCD spectrum. MCD spectra can also be obtained from the difference of absorption of the right-handed circularly polarized light and the left-handed light at resonance absorption edges. By integrating both XAS and MCD spectra and applying sum rule [82, 83], both spin and orbital magnetic moments can be extracted.

The XMCD measurements were taken in both total electron yield mode (TEY) and
transmission mode at the UV ring of the National Synchrotron Light Source (NSLS), Brookhaven National Laboratory, Beamline U4B. Spectra were taken for fixed circular photon helicity, 75% polarization, with pulsed magnetization switching (H=± 300 Oe) at the sample; photon incidence was fixed at 45° with respect to the sample normal. Photon energy could be varied continuously in the experiments from 500-1350 eV using a grating monochromator, with a general drop in beam intensity towards higher photon energies. The samples were mounted with magnetic easy axis along the applied field direction; U4B at NSLS does not have the capability to reverse the photon helicity in a simple way, which prevents the collection of all four combinations of positive and negative fields and helicities which are optimum to avoid the presence of XMCD artifacts. Thus, XMCD measurements with H⁺ (M⁺) measured first and H⁻ (M⁻) measured second were averaged with measurements taken in reversed order (“duplex mode”), to correct for any drift in the monochromator which might lead to derivativelike artifacts in XMCD.

Samples for TEY mode were deposited on Si substrate. The TEY currents were nor-
malized to TEY currents measured at a reference grid, located ahead of the sample, to correct for any variation in beam intensity over the measurement. High photon energies (1130-1320 eV) were calibrated using electron yield signals from inline Eu$_2$O$_3$ and Dy$_2$O$_3$ powder references, taking RE edge positions for the oxides reported before[84]. A +3kV extraction voltage was applied near the sample surface, which was found to be important for reproducible MCD spectra.

Samples for transmission mode were grown on Si/Si$_3$N$_4$ membrane substrates. Background XMCD spectra from the bare membrane were recorded for each energy interval applied and later subtracted from the sample spectra.

### 3.6 Resistivity measurement

To study the effect of eddy current on the damping, the resistivity of the epitaxial films was measured by an in-line four-point probe. A schematic of a four point probe is shown in Fig. 3.15. In this diagram, four wires (or probes) have been attached to the test sample. A current $I$ is made to flow the length of the sample through probes labelled 1 and 4 in the figure. This can be done using a current source or a power supply as shown. If the sample has any resistance to the flow of electrical current, then there will be a drop of potential (or voltage) as the current flows along the sample, for example between the two wires (or probes) labelled 2 and 3 in the figure. The voltage drop between probes 2 and 3 $V$ can be measured by a digital voltmeter. Thus the resistance between probes 2 and 3 $R$ is measured as

$$R = \frac{V}{I}. \quad (3.5)$$

The effects of finite sample dimensions on resistance were corrected using standard tech-
Figure 3.15: The schematic of four-point probe. 1 and 4 are the current probes and 2 and 3 are the voltage probes.

Techniques, according to which the resistivity of the measured thin film is

\[ \rho = \frac{V}{I} \frac{\pi}{\ln 2} t, \]

where \( t \) is the film thickness. Excellent agreement (5\%) was found with resistance measurements in our samples.
Chapter 4

Control of damping and precessional frequency by RE dopants in Ni$_{81}$Fe$_{19}$ thin films

4.1 Introduction

Ni$_{81}$Fe$_{19}$ (Permalloy) is a magnetic alloy with excellent soft ferromagnetic properties. Because of its small anisotropy and magnetostriction, low coercivity, high permeability, and high-saturation magnetization, permalloy can provide excellent writing performance and thus has been used throughout the data storage industry for recording heads for more than 40 years\[86\] [87].

It has been known since 1950’s that fast switching speeds could be achieved in thin films of Ni$_{81}$Fe$_{19}$\[4\]. The preparation and magnetic properties of vacuum-evaporated Ni$_{81}$Fe$_{19}$ thin films ($\sim$ 1000 A thick) were first reported by Blois in 1955\[88\]. When the film is deposited in the presence of a dc magnetic field in the plane of the film, the film develops a preferred magnetic axis (induced anisotropy) in the field direction. Under small applied fields ($\sim$ 5
CHAPTER 4. CONTROL OF DAMPING AND PRECESSIONAL FREQUENCY BY RE DOPANTS IN Ni₈₁Fe₁₉ THIN FILMS

G), it is possible to attain close to a single domain state for the sample. The films have rectangular hysteresis loops along the direction of inducing field and nearly hysteresis-free loops in a direction at right angles to the inducing field. With the application of sufficiently large switching fields, the films reverse their magnetization by a coherent rotation process. In early 1960’s the switching time of <1 ns was measured in Ni₈₁Fe₁₉ thin films by Dietrich, Proebster, and Wolf, and the reversal signals were successfully explained by Landau-Lifshitz equation\[19][89].

As the areal density of hard disk data storage systems increases with roughly fixed platter-rotation speeds, a faster rate of magnetic recording becomes more and more important. Extrapolation of current trends would project data rates in the 1-2 GHz range in magnetic recording channel in 5 years\[90]. In this regime, precessional dynamics, expressed by free magnetic oscillations, become a prominent phenomenon. Measurements of time-resolved response of sub-micron spin-valve devices, with 5 nm thick Ni₈₁Fe₁₉ as free layer, showed that while the magnetization reverses rapidly (<0.5 ns), it took several nanoseconds for the energy to be removed from the magnetic system\[91]. The switching energy was stored in short wavelength magnetic fluctuations that could dramatically affect the re-reversal process 1-2 ns after the first reversal. This ”ringing” in the dynamic responses is becoming an increasingly important phenomenon which limits data transfer rates in magnetoelectronic devices.

Some efforts have been made to reduce the unwanted oscillations during high-speed sense operations through ”coherent control” of applied fields. Opposite-sign magnetic field pulses, staggered precisely by \(\frac{1}{4}\) of the precessional period \(\tau_p\) (\(\sim 250\) ps), cause the response to be suppressed after the second pulse. Staggered step excitation has been performed to cancel the FMR oscillations in Ni₈₁Fe₁₉ film 50 nm thick grown by dc magnetron sputtering on a Si substrate 100 \(\mu\)m thick\[64]. As described in 2.6.1 two-pulse-technique was also demonstrated to control the precessional motion of the magnetization in 8 nm Ni₈₁Fe₁₉ thin
Both techniques apply the temporal tailoring of the driving magnetic field pulses. A more straightforward approach to eliminate "ringing" lies in the control of the damping parameter. As described in 2.5, a large contribution to the damping is intrinsic, arising from processes specific to the material composition at a given temperature. Damping, and the precessional response, can therefore be controlled through the composition of the ferromagnetic thin film. Integrated magnetic recording simulations showed that the damping constant of the head material considerably influences the write field\(^9\). The higher the damping constant, the shorter the flux rise time.

In previous work, Bailey et al. found that low concentrations (2-10\%) of Tb in 50 nm Ni\(_{81}\)Fe\(_{19}\) films, formed by low-power cosputtering of a Tb target with high-power sputtering of a Ni\(_{81}\)Fe\(_{19}\) target, can increase the Gilbert magnetic damping parameter over two orders of magnitude without great effect on the soft magnetic properties (as shown in 2.6.2). The dynamical response can be adjusted from underdamped to critically damped and even to overdamped situation. Gd dopants, by contrast, showed no effects on damping constant. The difference between the two dopants - adjacent in the periodic table - was attributed qualitatively to the magnetic character of the 4f shell, which, in isolation, is known to describe the magnetic properties of lanthanides well. Tb possesses an orbital moment, which can couple the spin moment to the lattice. Gd, on the other hand, does not, and thereby does not contribute damping.

The motivation for this section is twofold. First, we would like to develop further correlation between RE magnetic states and contributed relaxation. Second, in doing so, we expect to determine whether dopants exist which are more effective than Tb in enhancing relaxation. Since the magnetic states change systematically across the lanthanide series, it will be worthwhile to research the effect of lanthanide series dopants on the dynamical response of Ni\(_{81}\)Fe\(_{19}\) systematically and find out if the contributed damping from the rare earth elements can be related to their magnetic states.
CHAPTER 4. CONTROL OF DAMPING AND PRECESSIONAL FREQUENCY BY RE DOPANTS IN Ni_{81}Fe_{19} THIN FILMS

Table 4.1: Outer shell electronic configurations and the magnetic states of free lanthanide rare earth ions.

<table>
<thead>
<tr>
<th>Ln</th>
<th>La</th>
<th>Ce</th>
<th>Pr</th>
<th>Nd</th>
<th>Pm</th>
<th>Sm</th>
<th>Eu</th>
<th>Gd</th>
<th>Tb</th>
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<th>Ho</th>
<th>Er</th>
<th>Tm</th>
<th>Yb</th>
<th>Lu</th>
</tr>
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<tbody>
<tr>
<td>Config.</td>
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<td>(4f^1)</td>
<td>(4f^2)</td>
<td>(4f^3)</td>
<td>(4f^4)</td>
<td>(4f^5)</td>
<td>(4f^6)</td>
<td>(4f^7)</td>
<td>(4f^8)</td>
<td>(4f^{10})</td>
<td>(4f^{11})</td>
<td>(4f^{12})</td>
<td>(4f^{13})</td>
<td>(4f^{14})</td>
<td></td>
</tr>
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<td>2</td>
<td>(\frac{11}{2})</td>
<td>1</td>
<td>(\frac{13}{2})</td>
<td>0</td>
</tr>
<tr>
<td>L</td>
<td>0</td>
<td>3</td>
<td>5</td>
<td>6</td>
<td>5</td>
<td>3</td>
<td>0</td>
<td>3</td>
<td>5</td>
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<tr>
<td>J</td>
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<td>(\frac{3}{2})</td>
<td>4</td>
<td>(\frac{5}{2})</td>
<td>4</td>
<td>(\frac{7}{2})</td>
<td>0</td>
<td>(\frac{5}{2})</td>
<td>6</td>
<td>(\frac{12}{2})</td>
<td>8</td>
<td>(\frac{14}{2})</td>
<td>6</td>
<td>(\frac{16}{2})</td>
<td>0</td>
</tr>
</tbody>
</table>

In this part, a systematical research on the magnetization dynamics of rare-earth (RE) doped Ni_{81}Fe_{19} thin films were conducted. The studied dopants span the central portion of the lanthanide series, which include Sm, Eu, Tb, Dy, and Ho.

4.2 Systematical effect of RE dopants on the magnetization dynamics of Ni_{81}Fe_{19} thin films

Hund’s rules describe the ground state of a multi-electron atom. The three Hund’s rules are: (1) The state with maximum spinmomentum \(S\) has the lowest energy level; (2) for a given \(S\), the state with the largest value of orbital angular momentum \(L\) has the lowest energy; (3) for atoms with less than half-filled shells, the level with the lowest value of total angular momentum \(J = |L - S|\) lies lowest in energy; otherwise, if the outermost shell is more than half-filled the level with highest value of \(J = |L + S|\) is the one with the lowest energy.

Table 4.1 lists the electronic configuration of the outer shell of the free ions of the lanthanide rare earths. The values of \(S\), \(L\), and \(J\) are calculated according to the Hund’s rules and are also listed in the table. From Table 4.1 we can see that the orbital momentum \(L\) is zero for Gd and the maxima lie in Ho, Er, Nd and Pm. There is also a systematical change in the total angular momentum \(J\), with \(J = 0\) lying in Eu.
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4.2.1 Damping contributions from RE dopants

Films of (Ni$_{81}$Fe$_{19}$)$_{1-x}$RE$_x$ (RE=Sm, Eu, Tb, Dy, and Ho) were prepared by ion-beam sputtering (see Section 3.1.1). The RE dopant concentration $x$ ranges from 0 to 15%, as measured by ex situ XPS. Magnetization dynamics of the thin films were characterized by PIMM, and the experimental measurements were fit to the numerically integrated full-angle LL equation (see Section 3.3.1). Fit parameters including the anisotropy field $H_K$ and the LL relaxation rate $\lambda$ are extracted.

![Figure 4.1: Inductive wave forms for undoped Ni$_{81}$Fe$_{19}$ thin films measured by PIMM with different bias fields $H_B$. The voltage signal is proportional to the component of $M$ transverse to sample easy axis direction.](image)

Several fully processed inductive wave forms of the undoped Ni$_{81}$Fe$_{19}$ thin film measured by PIMM are shown in Fig. 4.1. The five waveforms were acquired with different dc bias
fields $H_B$ applied in the easy axis direction of the film. $H_B$ ranged from 0 to 30 Oe. The dynamic response data exhibit free inductive oscillations. The oscillations last almost 5 ns in the case of 30 Oe bias field. The existence of multiple inductive oscillations is indicative of underdamped gyromagnetic precession. As bias field decreases, the free oscillations also decrease. In fact, the waveform for zero bias field shows only minor oscillations, which is indicative of nearly critical damping.

![Figure 4.2: Inductive waveforms for a series of Dy-doped Ni$_{81}$Fe$_{19}$ thin films (0 < $x_{Dy}$ < 6%) with 20 Oe bias field. LL/LLG fits and estimated values of $\lambda/4\pi$ are shown.](image)

Representative experimental measurements of magnetization dynamics are shown, with LL/LLG fits, in Fig. 4.2 for the case of the (Ni$_{81}$Fe$_{19}$)$_{1-x}$Dy$_x$ system, 0 < $x_{Dy}$ < 6%. The longitudinal bias field is 20 Oe. Full-angle, numerically integrated LL/LLG fitted values of $\lambda$, expressed as $\lambda/4\pi$, are listed in the fits; these can be compared visually with the "ring-down time" $\tau$, in which the damping term in $V_{ind}(t)$ takes the form $e^{-t/\tau}$, and $\lambda = 2/\tau$. Note that the damping parameter $\lambda$ has SI units. In CGS it is $\lambda_{cgs} = \lambda/4\pi$. Agreement
between the model and experiment is close except in the case of undoped Ni$_{81}$Fe$_{19}$, in which some evident shift in the frequency appears at longer times (3 ns). $H_k$ is set to 5-6 Oe in all fits except Eu according to $\omega_p$ ($H_B$) measurements (to be discussed).

It is evident from Fig. 4.2 that the Dy doped series, formed by ion-beam deposition, spans underdamped to nearly critically damped dynamics. This range of moderate effects is of great interest for applications, and had not been attainable through cosputtering elemental targets of Tb and Ni$_{81}$Fe$_{19}$. Compositional analysis of the most heavily Dy-doped film, with $\lambda/4\pi = 550$ MHz (bottom right in Fig. 4.2, by XPS yields $x_{Dy} = 5.2 \pm 1.5\%$.

Figure 4.3: Dynamic responses and LL/LLG fits of representative members of the Eu, Sm, Tb, and Ho doped Ni$_{81}$Fe$_{19}$ series, with composition characterized by XPS. Strong effects on damping are observed for all dopants except Eu.

To yield an estimate of contributed damping per atomic percentage of the RE dopant, we make two assumptions. First, relaxation rates of the undoped film are assumed to sum with the contributed effects of the dopants. This approach can be justified through different temperature-dependent relaxation behavior [$\alpha = \alpha(T)$] found for undoped and 2%
Tb-doped films\textsuperscript{14}: weak and positive for undoped films, and strong and negative for Tb-doped films. Secondly, we assume that the contributed damping is proportional to atomic percentage of RE in the dilute doping regime, as found previously for Tb. We then find $(\lambda/4\pi)/(%_{Dy})=(550-180) \text{ MHz/(5.2}\pm1.5\%)$, or $\lambda/(%_{Dy})=(890\pm260) \text{ MHz}$.

This analysis has been applied additionally to Sm, Eu, Tb, and Ho. Analogous effects are shown except in the case of Eu, in which no damping is contributed. The data and fits are shown in Fig. 4.3 still with $H_B=20 \text{ Oe}$. Good agreement is again found between model and experiment, although the fit quality is poorer for the case of Sm- and Ho-doped films.

![Contributed damping to Ni\textsubscript{81}Fe\textsubscript{19} as a function of lanthanide dopant atomic number $Z-57$ (La=57). The angular momentum states of the isolated dopants are shown for comparison.](image)

Figure 4.4: Contributed damping to Ni\textsubscript{81}Fe\textsubscript{19} as a function of lanthanide dopant atomic number $Z-57$ (La=57). The angular momentum states of the isolated dopants are shown for comparison.

The comparison of contributed relaxation rate across the central portion of the lanthanide series, $\lambda/(%_{RE})$ ($Z$), is presented in Fig. 4.4. We find a roughly V-shaped dependence of contributed damping for these elements, with zeroes corresponding to Eu and Gd. The theoretical orbital and total angular momentum numbers $L$ and $J$ are plotted for
comparison. The zero in contributed damping corresponds to zeroes in L and J expected for the $4f$ shell. The result indicates that the magnetic states of free RE elements might control the magnetization dynamics in ferromagnets.

4.2.2 Precessional frequency boost in $\text{Ni}_{81}\text{Fe}_{19}$ by Eu dopant

Eu doping (3%) increases the resonant frequency of the system without contributing relaxation. Comparing the response of undoped (Fig. 4.2, top left) with that of Eu-doped (Fig. 4.3, top left) $\text{Ni}_{81}\text{Fe}_{19}$ reveals six maxima in the latter case and five in the former for $H_B=20$ Oe. More quantitative measurement of resonant frequency $f_p (=\omega_p/2\pi)$ is found through a Fourier transform of the data, appropriate for the low relaxation rates of $\lambda/4\pi=160$ and 180 MHz, found respectively for Eu-doped and undoped $\text{Ni}_{81}\text{Fe}_{19}$.

![Kittel plot for undoped, Tb-doped, and Eu-doped $\text{Ni}_{81}\text{Fe}_{19}$, with longitudinal bias field. Inset, left: $\omega_p$ versus bias field $H_B$. Table, right: extracted values of dynamic $H_k$ and $g$ factor. Note the 9 Oe enhancement of dynamic $H_k$ provided by Eu.](image)

Fig. 4.5 shows the bias field dependence of $f_p$, plotted as $f_p^2(H_B)$ to compare with the
Kittel relation\[93],

\[ f_p^2 = \left( \frac{\omega_p}{2\pi} \right)^2 = \frac{\gamma^2 \mu_0^2 M_s}{4\pi^2} (H_k + H_B), \]  

for in plane ferromagnetic resonance (FMR) in a thin film, where the prefactor evaluates to 0.07199\((g_{\text{eff}}/2)^2\) GHz\(^2\)/Oe. We use this relationship to extract dynamic anisotropy fields \(H_k\) and the \(g_{\text{eff}}^2 M_s\) product for the films. Marked differences are found in the dynamic anisotropy field \(H_k\): (5\(\pm\)1) Oe for undoped and Tb-doped films, but (14\(\pm\)1) Oe for 3\% Eu-doped. This corresponds to a boost in resonant frequency by 480 MHz without increase in loss. The result found for Eu-doped Ni\(_{81}\)Fe\(_{19}\) is significant to the extent that other materials techniques are not well known which can enhance \(\omega_p\) without increasing dissipation.

Figure 4.6: MOKE hard-axis hysteresis loops for undoped and Eu-doped Ni\(_{81}\)Fe\(_{19}\); dynamic \(H_k\) values are shown for comparison.

Essentially constant values of \(g_{\text{eff}}^2 M_s\) are found within experimental error. Assuming that \(M_s\) remains constant at 730 kA/m, we have \(g_{\text{eff}} = 2.16\pm0.05\) for Eu 3\%, 2.15\(\pm\)0.05 for Tb 9\%, and 2.12\(\pm\)0.05 for Ni\(_{81}\)Fe\(_{19}\), consistent with the value of 2.07 found previously\[11\]. We note furthermore that the fitted value of damping constant \(\alpha\) is essentially constant.
for applied field $H_B > 5$ Oe for Eu-doped and $H_B > 10$ Oe for undoped or Dy-doped films, providing some justification for the use of the Gilbert damping form. Comparison of measured dynamic $H_k$ with static $H_k$, as measured by MOKE (Fig. 4.6 and Fig. 4.7), shows good agreement for undoped and Tb-doped Ni$_{81}$Fe$_{19}$, but a much larger dynamic $H_k$ for the Eu-doped case by nearly a factor of two.

Figure 4.7: MOKE hard-axis hysteresis loop for Tb-doped Ni$_{81}$Fe$_{19}$; dynamic $H_k$ value is shown for comparison.

The MOKE measured easy axis hysteresis loops of undoped, Eu-doped, and Tb-doped Ni$_{81}$Fe$_{19}$ are shown in Fig. 4.8. Magnetization is normalized to 1 in all the cases. All samples show well-defined squareness along the easy axis. The coercivity for undoped Ni$_{81}$Fe$_{19}$ is 1.1 Oe, and that for Eu and Tb doped samples is a little bit bigger. The high coercive squareness along the easy axis and near-zero remanence along the hard axis (Fig. 4.6 and Fig. 4.7) show the good soft magnetic properties in these samples.
Figure 4.8: MOKE measured easy-axis hysteresis loops for undoped (left and right), Eu-doped (left) and Tb-doped (right) $\text{Ni}_{81}\text{Fe}_{19}$, all showing well-defined squareness and small coercivity.

### 4.2.3 Comparison of dynamics in RE-doped $\text{Ni}_{81}\text{Fe}_{19}$ with that in RE-substituted YIG

Some of our observations on dynamics of RE-doped $\text{Ni}_{81}\text{Fe}_{19}$ are consistent with 1960s’ observations on FMR of RE-substituted YIG (Yttrium iron garnet, a ferrimagnetic material widely used in various microwave and optical-communication devices and other applications). RE species are thought to contribute damping in YIG by coupling the magnetization to the lattice through the orbital moment\[^{57}\]. Negligible contributed damping in Eu:YIG and Gd:YIG had been attributed to the nonmagnetic ground state of Eu ($J_{\text{Eu}}=0$) and the absence of an orbital moment in Gd ($L_{\text{Gd}}=0$), respectively\[^{94}\]. A strong, growth-induced anisotropy has also been observed in substituted europium iron garnet, absent in gadolinium iron garnet\[^{95}\]. These similarities suggest that the properties of doped films arise from the magnetic properties of the isolated RE elements.
4.3 Magnetization dynamics control in RE-doped/undoped Ni$_{81}$Fe$_{19}$ bilayers

4.3.1 Precessional frequency tuning in Ni$_{81}$Fe$_{19}/$(Ni$_{81}$Fe$_{19}$)$_{1-x}$Eu$_x$ bilayers

In the previous section we found that dilute concentrations of rare-earth elements can be used to tune the dynamic response of ferromagnetic thin film magnetization. A systematic variation in strength has been observed across the lanthanide series. Among these elements, Eu is special in that it does not contribute damping to the system, but increases the precessional frequency by roughly 0.1 GHz per atomic percentage. The dopants can thus be used to raise a basic speed limit to precessional motion.

In this chapter, we examined the effect of Eu-doped thickness fraction on the precessional frequency of Ni$_{81}$Fe$_{19}/$(Ni$_{81}$Fe$_{19}$)$_{1-x}$Eu$_x$ bilayers. The Eu-doped Ni$_{81}$Fe$_{19}$ is a high $f_p$ layer and Ni$_{81}$Fe$_{19}$ is a lower $f_p$ layer; bilayers exhibited intermediate values. This bilayer structure could be useful in magnetoelectronics, e.g. giant magnetoresistive (GMR) or tunneling magnetoresistive (TMR) spin valves. Our motivation is to remove dopants from ferromagnetic/nonmagnetic interfaces, preserving spin transport properties, but acting on magnetization dynamics through the exchange interaction. We present a study of the dynamic properties including relaxation rate $\lambda$, precessional frequency $f_p$, spectroscopic splitting factor $g$, as well as static properties of the bilayers, as a function of Eu-doped thickness fraction.

The bilayer structure SiO$_2$/Ni$_{81}$Fe$_{19}$(1-y)50 nm)/Ni$_{81}$Fe$_{19}$:Eu$_x$([y]50 nm)/Ta was prepared by ion-beam sputtering. The Eu content in Eu-doped layer was fixed at $x = 3\%$. The total thickness is 50 nm. The thickness fraction $y$ of the Eu-doped region varied as $0 \leq y \leq 1$. 
Magnetization dynamics and the frequency tuning

Fig. 4.9 shows inductive waveforms of Ni$_{81}$Fe$_{19}$/Ni$_{81}$Fe$_{19}$:Eu$_x$ bilayers with 5 Oe bias field. As the thickness fraction of the Eu-doped layer $y$ increases, we see more oscillations in the waveform over a fixed period of time, indicating higher precessional frequency. The full width at half maximum (FWHM) of the primary inductive wave is 250 ps for $y = 1$, as compared with 330 ps for $y = 0.5$ and 350 ps for $y = 0$. We performed a simple average between the signals for $y = 1$ and $y = 0$, and the result is also shown in Fig. 4.9, which is quite different from $y = 0.5$. We can see that the response from bilayers is not simply the superposition of the two single layers. This illustrates the strong exchange coupling between the two layers. Similar measurements were completed with different dc bias fields applied along the easy-axis direction.

Figure 4.9: Inductive waveforms of Ni$_{81}$Fe$_{19}$([1-$y$/50nm])/Ni$_{81}$Fe$_{19}$:Eu$_x$([$y$/50nm]) bilayers, acquired with 5 Oe of transverse bias field. Average from $y = 0$ and $y = 1$ is also shown for comparison.
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The time-domain data were transformed to the frequency domain by fast Fourier transform, as shown in the inset of Fig. 4.10. The frequency of the lowest spectral peak (n=1) is almost the same as that extracted from the sinusoidal fitting. This frequency is believed to approach the standard FMR result for weak damping$^{11}$ and was studied here. Fig. 4.10 shows the extracted precessional frequency $f_p$ as function of bias field $H_B$ for the bilayers with different thickness fraction $y$. For the series studies, the processional frequency has been increases by as much as roughly 0.3 GHz across the series in the bilayers.

![Figure 4.10: Precessional frequency versus bias field of Ni$_{81}$Fe$_{19}$/(1-y)50nm)/Ni$_{81}$Fe$_{19}$:Eu$_x$((y)50nm) bilayers, thickness fraction 0 ≤ y ≤ 1. Inset shows Fourier transform of the time-domain inductive waveform for y = 0.5 with $H_B$=20 Oe.](image)

**LL and sinusoidal fits and the extracted dynamic parameters**

Magnetization dynamics measurements were fitted to both exponentially decaying sinusoidal function and the numerically integrated full-angle LL equation to extract the relaxation rate $\lambda$. Both methods give similar results. Fig. 4.11 shows $\lambda$ for bias field $H_B$=20 Oe as a function
of thickness fraction. By both methods, as thickness fraction \( y \) changes from 0 to 1, there is no significant change in the damping parameter. The relaxation rate \( \lambda/4\pi \) remains \( \sim 150 \) MHz for LL simulation and \( \sim 140 \) MHz for sinusoidal function fit. The same analysis was also applied to different dc bias fields and consistent results were found.

![Figure 4.11: Relaxation rate \( \lambda \) of the bilayers, simulated from both exponentially decaying sinusoid and numerically integrated LL equation.](image)

From Kittel equation, plotted as \( f_p^2 \) versus \( H_B \), we can extract both \( g \) factor and dynamic anisotropy field \( H_k \). Fig. 4.12 shows the dynamic \( H_k \) as function of thickness fraction \( y \). The inset shows the linear relationship between \( f_p^2 \) and \( H_B \) for the sample with thickness fraction \( y = 0.5 \). \( g \) factor was extracted as 2.26 from the slope and dynamic \( H_k \) 6.12 Oe from the interception with \( H_B \). As the thickness fraction of the Eu-doped layer increases, dynamic \( H_k \) also increases. The corresponding \( f_p^2 \) is also shown in the figure for comparison.

We find that the boost of the precessional frequency from Eu dopants corresponds well to an enhancement of anisotropy. Present results suggest that identity between dynamic
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Figure 4.12: Dynamic anisotropy field $H_k$ of the bilayers, extracted from the interception with $H_B$ in Kittel plot. The corresponding $f_p^2$ for $H_B = 20$ Oe is also shown for comparison. The inset is the Kittel plot for $y = 0.5$.

and static anisotropy field (see below).

$g$ factor extracted from both LL simulation and Kittel plot are shown in Fig. 4.13. In both methods, the $g$ factor changes linearly as the thickness fraction of Eu-doped layer increases. $g$ factor values obtained by $f_p^2(H_B)$ fits, taking the FFT-derived values of $f_p$, yield $g$-factor values increasing from 2.0 to 2.3. Estimates taken by the full-angle LL fit are $\sim 0.1$ higher, due in part to some deviation from the simulation at longer times. The obtained values for undoped Ni₈₁Fe₁₉ of 2.0 and 2.1 are close to the $g = 2.05$ measure by Silva et al.[11]. The inset in Fig. 4.13 shows the slope of Kittel plot, $(\mu_0\mu_B/h)^2y^2M_s$, versus thickness fraction $y$. As $y$ increases, there is a slight increase in the slope, about 2% more for $y = 1$ than for $y = 0$. As later we will show that magnetization $M_s$ decreases with $y$,
this result indicates that the increasing rate of $g^2$ is slightly faster than the decreasing rate of $M_s$. Since $g$ is directly related to the ratio of orbital and spin moment $^{96,97}$,

$$\frac{\mu_L}{\mu_S} = \frac{g}{2} - 1,$$

the increase of $g$ across the series suggests that fraction of the total momentum contributed by orbital motion compared with that contributed by spin motion is strengthened as the Eu-doped layer thickness is increased. However, at present, we have not been able to verify this by XMCD; Fe edges of the Ni$_{81}$Fe$_{19}$ show $\mu_L/\mu_S$ ratios which are constant and independent of Eu doping, and only negligible XMCD signals have been observed on Eu.

Figure 4.13: $g$ factor of the bilayers, extracted from LL simulation and Kittel plot, separately. Inset: slope in Kittel plot, $(\mu_0\mu_B/h)^2g^2M_s$, versus thickness ratio $y$. 

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Static magnetic properties of the bilayers

The easy and hard axis hysteresis loops were measured by SQUID magnetometry for the whole series, shown in Fig. 4.14. Loops for $y = 0$, Ni$_{81}$Fe$_{19}$ single film are shown to the upper left, and those for $y = 1$, Eu-doped Ni$_{81}$Fe$_{19}$ single film are to the lower left. For the series tested, the squareness along the easy axis is very well defined. As thickness fraction of Eu doped layer increases from 0 to 1, coercivity $H_c$ increases from 1.2 to 2.8 Oe, and anisotropy field $H_k$ increases from 2.7 to 11.6 Oe. The static anisotropy field agrees well with the dynamic values. This observation disagrees with previous finding that dynamic $H_k$ is larger than the static value in Eu-doped Ni$_{81}$Fe$_{19}$ thin films (measured by MOKE) (as in 4.2.2); we presume that the improved resolution of the SQUID has given more reliable data.

Saturation magnetization $M_s$ was calculated from the magnetic moment measured by SQUID. The volume of the measured sample was measured through a combination of profilometric film thickness and the surface area of the film or substrate. To the right of Fig. 4.14 shows $M_s$ as function of $y$. As $y$ increases, $M_s$ decreases. This consistent with previous data for Gd and Tb doped films[14] and the idea that rare earth moments align antiparallel with transition metal moments.

In conclusion, a bilayer method was demonstrated to tune the precessional frequency in Ni$_{81}$Fe$_{19}$ thin films with minimal effect on the damping parameter. The measured dynamic data were fitted to both the Landau-Lifshitz equation and exponentially decaying sinusoid and the extracted parameters agree well with each other. Strongly enhanced anisotropy in the films (2.7 to 11.6 Oe) is important to explain the boost in the resonance frequency (of 300 MHz).
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Figure 4.14: Static magnetic properties measured by SQUID. Upper left: easy and hard axis hysteresis loops for Ni$_{81}$Fe$_{19}$ single films; lower left: hysteresis loops for Eu-doped Ni$_{81}$Fe$_{19}$ single films; right: saturation magnetization of the bilayers. Note the difference in the scale of applied field between the two loop figures.

4.3.2 Control of high-speed damping in Tb-doped/undoped Ni$_{81}$Fe$_{19}$ bilayer thin films

For reversals of magnetization, the characteristic time for transient motion to decay out of the system is given by $2/\lambda$, where $\lambda$ is the relaxation rate in s$^{-1}$. Therefore moderate enhancement of $\lambda$ shortens the allowed time between pulsed reversal of magnetization. In this chapter, we show that the damping of magnetic thin films can be controlled using a coupled bilayer structure where the dopants are confined to one layer. Tb-doped Ni$_{81}$Fe$_{19}$
with higher damping parameter was deposited before or after the deposition of Ni$_{81}$Fe$_{19}$ with lower damping parameter. By controlling relative thickness between two layers, the damping parameter of the bilayer is systematically changed.

Bilayer thin films of Ni$_{81}$Fe$_{19}$:Tb5%/Ni$_{81}$Fe$_{19}$ were prepared by ion beam sputtering. Relative thickness ratio of TbNiFe layer, $\frac{TbNiFethickness}{bilayerthickness}$, was changed while total bilayer thickness was fixed at 50 nm. By changing the deposition order of TbNiFe and NiFe, we tested possible effects of any lattice mismatch or different growth mode between two layers. All films were capped with 2-5 nm Ta layer to prevent oxidation. The overall film structure is, therefore, Ni$_{81}$Fe$_{19}$:Tb5%[x nm]/Ni$_{81}$Fe$_{19}$[(50-x) nm].

Dynamic magnetic properties were characterized by PIMM measurement. Measured inductive signals were fitted using exponentially decaying sinusoidal function. Spin relaxation rate $\lambda$ was approximated by this method. Similar results were given by numerical fitting using the Landau-Lifshitz equation.

Figure 4.15: Inductive signals of selected bilayer samples, Ni$_{81}$Fe$_{19}$:Tb5%[x nm]/Ni$_{81}$Fe$_{19}$[(50-x) nm]. The Tb-doped thickness fraction (x/50) is shown on each graph (0, 0.2, 0.6, and 1). Plots are offset for comparison. Open circles are the experimental data and lines are the fitted data using exponentially decaying sinusoidal function.
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Magnetization dynamics in TbNiFe/NiFe bilayers

Fig. 4.15 shows selected results of dynamic measurements for TbNiFe[x nm]/NiFe[(50-x) nm] bilayers. Thickness ratio of TbNiFe to the total bilayer, x/50, is shown on each graph. As TbNiFe thickness increases, the oscillation and the intensity of inductive signal decrease, which indicates the enhancement of damping.

The behavior is more clearly shown in Fig. 4.16. Damping parameter used for fitting, spin relaxation rate $\lambda/4\pi$, is shown in the figure as a function of TbNiFe thickness ratio in the bilayer structure. It should be noted that wide variation of damping parameter, actually in the full range between two end values of NiFe and TbNiFe single films, can be obtained simply by changing the thickness ratio in the bilayer structure.

![Graph showing relaxation rate $\lambda/4\pi$ vs. doped thickness fraction](image)

Figure 4.16: Relaxation rate $\lambda/4\pi$, obtained from Fig. 4.15, as a function of doped thickness fraction.

Furthermore, the deposition order of each layer is not important for the damping of bilayers. Two samples with reverse deposition orders show the same inductive signals in Fig. 4.17. This result is suggestive in that growth mode or microstructure is not very important in determining the damping for these films.
Figure 4.17: Comparison of inductive signals for two bilayers of reversed deposition orders. Both show the same dynamic waveforms.

**Static magnetic properties of the bilayers**

Static magnetic properties were measured by MOKE and are shown in Fig. 4.18. Coercivity $H_c$ extracted from easy axis loop and anisotropy field $H_k$ from hard axis loop are summarized separately. As shown in the figure, $H_c$ and $H_k$ do not depend strongly on the bilayer structures. Their average values are 1.1 Oe and 5.2 Oe, respectively. The fact that static magnetic properties are roughly conserved will foster application prospects for the future, as high-speed damping can be adjusted independently.

From the results of TbNiFe/NiFe bilayers, only the damping is significantly changed, while other properties remain roughly the same. In our bilayers, the damping parameter of TbNiFe is much higher than that of NiFe. This implies that the magnetic moment of the TbNiFe layer is more sluggish, while the magnetic moment of the NiFe layer is more mobile under the dynamic external field. In the limit of infinitely strong exchange coupling between the layers, we would expect a proportional relationship between damping $\lambda$ and doped thickness fraction analogous to the proportionality between $\lambda$ and rare earth.
concentration described in 4.2 as no canting angle could develop and the bilayer would act as a single film. The bilayer damping, however, does not change linearly with thickness fraction, and is even discontinuous as seen in Fig. 4.16. This suggests more complicated or incomplete coupling, and further investigation is needed to understand it.

In conclusion, it is shown that the magnetic damping of thin films can be controlled systematically using bilayer structure with two different damping parameters. By changing the bilayer thickness ratio, damping parameter is widely tuned between two end values of each single layer. The behavior is independent of the deposition order. The results for the control of damping using bilayer structure will give greater flexibility to design magnetoelectronic devices.
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4.4 XMCD characterization of RE-doped Ni$_{81}$Fe$_{19}$

Sub-nanosecond magnetization dynamics play an important role in determining the >1 GHz data rate of spin electronic devices. Rare-earth dopants can provide control over both the rotational speed and damping of magnetization motion, parameterized as the precessional frequency $f_p$ and Landau-Lifshitz(-Gilbert) damping $\lambda_{LL}$ or $\alpha$, respectively, in Ni$_{81}$Fe$_{19}$. Eu dopants in Ni$_{81}$Fe$_{19}$ have been found to enhance $f_p$ by 0.1 GHz/%. Terbium (Tb) adds roughly two orders of magnitude to $\lambda_{LL}$ over the concentration range 0-10%; the contributed damping is general for the lanthanides Sm-Ho, $\Delta\lambda_{LL}/\%$ of dopant scaling roughly with the nominal orbital moment $<\hat{L}_{RE}>$ of the dopant.[98]

X-ray magnetic circular dichroism (XMCD) is an ideal probe of the atomistic basis of magnetization dynamics in doped films, due to its ability to separate orbital and spin moments $m_l$ and $m_s$ on individual atomic sites[99]. Using XMCD, many rich phenomena have been identified in magnetic heterostructures previously, including induced spin and orbital moments on "nonmagnetic" atoms. Induced Au orbital moments, observed by XMCD[100], have explained the perpendicular magnetic anisotropy in Co/Au/Co, of present technological interest in perpendicular recording media.

4.4.1 Fe orbital moment in RE-doped Ni$_{81}$Fe$_{19}$

The dopant orbital moment appears to be important for producing effects on GHz magnetization dynamics. In this study, we ask how localized this effect is to the lanthanide dopant sites, and whether some induced orbital character is present in the (adjacent) transition metal sites. We present an XMCD investigation of the magnetic character of Fe in Ni$_{81}$Fe$_{19}$(50nm) doped with rare earths Eu, Gd, Tb, and Dy, characterizing the orbital-to-spin moment ratio as a function of doping. We wish to understand whether the spin-lattice coupling thought to be enhanced by the dopants is localized to RE sites, or distributed partially around the dopants and borne by transition metal neighbors.
Films were deposited and magnetization dynamics were measured using methods described in ref. [98]. RE-doped Ni$_{81}$Fe$_{19}$ (50 nm) thin films, along with a reference pure Fe(50 nm) film, were prepared using ion beam deposition in a load-locked, multitarget chamber (Veeco Millatron) with base pressure of 6x10$^{-8}$ Torr, on thermally oxidized Si substrates. Uniaxial anisotropy was induced with 20 Oe deposition field.

The RE concentrations of the Gd doped and Tb doped films were measured at 1.2% and 2.7%, respectively, by Rutherford Backscattering (RBS) at SUNY Albany. The top surfaces of the films were protected by a 20 Å Ta cap layer. This thickness was found to be somewhat optimal since uncapped Ni$_{81}$Fe$_{19}$ exhibited a distorted XAS / XMCD Fe spectrum characteristic of oxidation, and a 50 Å cap attenuated the signal too strongly.

Magnetization dynamics of the thin films were characterized using a time-domain pulsed inductive technique. Measured waveforms are proportional to $\partial\phi/\partial t(t)$, where $\phi$ is the in-plane angle of the magnetization. A bias field $H_B = 20$ Oe is applied along the magnetic (induced) easy-axis direction, orthogonal to the pulsed field. See refs [11, 14] for details. Magnetization dynamics were measured within one day of deposition; samples were stored in a dessicator for less than a week before transfer to UHV at NSLS.

XMCD measurements were taken in total electron yield mode (TEY) at the UV ring of the National Synchrotron Light Source (NSLS), Brookhaven National Laboratory, Beamline U4B[101]. XMCD was measured for fixed circular photon helicity, 75% polarization, with pulsed magnetization switching ($\pm$ 300 Oe) at the sample. The photon incidence (helicity $\sigma$) was fixed at 45° with respect to the sample normal. Thus as the magnetization was switched, the projection of $\mathbf{M}$ along the helicity direction $\sigma$ was reversed, and drain currents $I_+$ and $I_-$ ($\sim$100 pA or less) were measured. Magnetic fields were applied along the induced easy axis of magnetization.

Sample TEY currents were normalized to TEY currents measured at a reference grid ($I_o$), located ahead of the sample, to correct for any variation in beam intensity during the
measurement. A +3kV extraction voltage was applied near the sample surface; this was found to be important for reproducible XMCD difference spectra.

The Fe edge electron yield XMCD data were processed minimally to extract the $m_l$ to $m_s$ ratio. A constant offset was applied to the data before integration of the EY difference $(I_+(h\omega) - I_-(h\omega))$, in order to correct for drift of the integrated signal over the post-edge region.

Sub-nanosecond magnetization dynamics measurements, taken by pulsed inductive microwave magnetometry (PIMM), are shown in Fig. 4.19. The responses of five films are shown: undoped Ni$_{81}$Fe$_{19}$ and Eu-, Gd-, Tb-, and Dy-doped Ni$_{81}$Fe$_{19}$. The y-scale, corresponding to mV of inductive response, is preserved in the plots.

Figure 4.19: Comparison of 20 GHz PIMM response, (50 nm) undoped, and RE doped, RE=(Eu, Gd, Tb, Dy).

For small rotation angles ($\phi, \theta$) and low damping of a single-domain thin film under a stepped transverse field excitation, the Landau-Lifshitz equation can be solved in closed
form, giving a time derivative of in-plane magnetization angle:

\[
\left( \frac{d\phi(t)}{dt} \right) \propto e^{-\lambda t/2} \cos \left( \omega_p t + \delta \right) \tag{4.3}
\]

which is an oscillatory response with frequency \( \omega_p \) and exponential decay envelope with characteristic time \( 2/\lambda \). The PIMM response is proportional to \( \left( \frac{d\phi(t)}{dt} \right) \); \( \lambda \) can be estimated from the decay envelope of the signal, and \( \omega_p \) from the frequency of oscillation.

We find that for the films investigated by XMCD, there is an appreciable variation in both \( \lambda \) and in \( \omega_p \). Thus if the orbital moment of Fe plays a strong role in producing the engineered magnetic properties, its change may be seen by XMCD. Consistent with previous results, we find that Gd adds no relaxation, Eu adds no relaxation but enhances \( \omega_p \), and Tb and Dy increase \( \lambda \). For the films considered, the concentration of Dy increases \( \lambda \) from 1.6 GHz to 9.4 GHz. The concentration of Tb apparently increases \( \lambda \) by a substantially greater amount; fits using the simple sinusoidal form fail in this case.

The Fe orbital to spin moment ratios were determined using the technique demonstrated by Chen et al in ref. [102]. \( m_t/m_s \) can be found from \( L_{2,3} \) edges of 3d transition metals through the formulae

\[
\frac{m_{\text{orb}}}{m_{\text{spin}}} = \frac{2}{3} \left[ \frac{|A| - |B|}{|A| + 2|B|} \right] = \frac{2q}{9p - 6q}. \tag{4.4}
\]

\( |A| \) and \( |B| \) are the integrated XMCD intensities over \( L_3 \) and \( L_2 \) edges, respectively.[81] The \( L_3 \) XMCD peak is negative: \( |A| = -A \), so the orbital moment is proportional to the total integrated XMCD. To correct for possible variations in background, it is useful to consider total XMCD integrals \( f(E) = \int_{E_0}^{E} [I_+(\hbar\omega) - I_-(\hbar\omega)] d(\hbar\omega) \) starting from far below the \( L_3 \) edge; \( E_0 = 686 \) eV in our experiment. \( p \) and \( q \) are then defined as \( p = f(E_1) \) and \( q = f(E_2) \), where \( E_1 \) is immediately before the \( L_2 \) edge (\( E_1 = 715 \) eV) and \( E_2 \) is after both \( L_3 \) and \( L_2 \) edges (\( E_2 = 740 \) eV).[102] In eq. [4.4] the two expressions are redundant since \( p = -A, q = B - A \). The relative magnitude of the orbital moment is given proportional to
Fig. 4.20 shows smoothed XMCD difference data \([I_+ \hbar \omega) - I_- \hbar \omega])\) for Fe in the various films. XMCD for Fe in pure Fe(50nm) is shown on top, followed by Fe in undoped Ni_{81}Fe_{19}, then Eu-, Gd-, Tb- and Dy-doped Ni_{81}Fe_{19}. Since Fe has a majority of its moment coming from spin, the two peaks (negative at \(L_3\) and positive at \(L_2\)) are roughly equally weighted. It can also be appreciated visually that the relative weights of \(L_3\) and \(L_2\) XMCD peak areas do not change visibly. The shape of Fe XMCD in pure Fe is somewhat different from that in the Ni_{81}Fe_{19} samples, however; it is broader in \(L_3\) and has a low-energy shoulder at \(L_2\).

![Diagram of XMCD for Fe in pure Fe(50nm), Ni_{81}Fe_{19}(50nm), and RE-doped Ni_{81}Fe_{19}, where the dopant is Eu, Gd, Tb, and Dy. Scales are normalized to \(L_3\) peaks and offset for visual comparison of shapes.](image)

Figure 4.20: XMCD for Fe in pure Fe(50nm), Ni_{81}Fe_{19}(50nm), and RE-doped Ni_{81}Fe_{19}, where the dopant is Eu, Gd, Tb, and Dy. Scales are normalized to \(L_3\) peaks and offset for visual comparison of shapes.

The procedure for extracting the \(m_l/m_s\) ratio is illustrated in Fig. 4.21, taking Fe in undoped Ni_{81}Fe_{19}(50nm) as an example. The integrated XMCD signal \(f(E)\) is plotted; reading of \(p\) and \(q\) is illustrated. It is evident from the figure that \(|q| < |p|\), thus the negative-going peak at \(L_3\) and the positive-going peak at \(L_2\) balance each other for the most part, illustrating the relatively low orbital moment of Fe in the samples considered. We measure \(p = 0.0976\) and \(q = 0.0212\) for an orbital to spin moment ratio \(m_l/m_s = 0.056\).
using eq. \[4.4\], the error of ±0.01 considers uncertainties in the estimate of background.

Figure 4.21: Illustration of numerical integration procedure to determine \(m_l/m_s\) for Fe in the various films, shown for undoped Ni\(_{81}\)Fe\(_{19}\)(50nm).

Measurements of \(m_l/m_s\) data are summarized in Fig. \[4.22\]. We find constant values of \(m_l/m_s\) for Fe in all environments within experimental error. Pure Fe(50nm), undoped Ni\(_{81}\)Fe\(_{19}\)(50nm), and Eu, Gd, Tb, and Dy doped Ni\(_{81}\)Fe\(_{19}\)(50nm) all show \(m_l/m_s = 0.049 \pm 0.01\). The values are in good agreement with the pure Fe value of 0.043 determined by Chen et al through transmission experiments in ref[102].

We find essentially constant values of \(m_{orb}/m_{spin}\) for Fe moments in RE-doped Ni\(_{81}\)Fe\(_{19}\)(50nm). Controlled Ghz dynamical properties are realized in the same films with a frequency enhancement \(\Delta\omega_p = 300\) MHz for Eu doping, damping enhancement \(\Delta\lambda = 8\) GHz for Dy doping, and a yet larger enhancement for Tb doping. We have treated Fe moments in the study because of their higher moment per atom in the alloy, 2.5 \(\mu_B/\text{at}\) compared with 1.6 for Ni \(\mu_B/\text{at}\), and consequent higher XMCD signal. Ni moments have been examined less completely; we find constant values of \(m_l/m_s = 0.11 \pm 0.01\) for the several films on which
Figure 4.22: Extracted spin to orbital moment ratios, $m_{\text{orb}}/m_{\text{spin}}$ of Fe in pure Fe (left), RE-doped Ni$_{81}$Fe$_{19}$ (50 nm), and undoped Ni$_{81}$Fe$_{19}$ (50 nm) (right), by XMCD analysis as shown in Figure 4.21.

statistics allowed application of sum rules.

The results show that the engineered dynamical properties in $\omega_p$ and $\lambda$ produced by lanthanide dopants, are not accompanied by a strong induced orbital character on the transition metal moments. Damping is understood to be closely related between the coupling between spin moments and the lattice; orbital moments mediate the coupling. This implies that the location of the orbital moments identifies the sites for precessional relaxation, where energy is transferred out of the spin system. In a separate XMCD study\cite{103}, we have located these sites on Gd and Tb dopants; $m_l/m_s = 0.0$ for Gd and $0.75 \pm 0.15$ for Tb, respectively. The absence of effect on Fe orbital moments localizes the sites for relaxation to those of the dopant atoms themselves.
4.4.2 XMCD characterization of RE dopants in Ni$_{81}$Fe$_{19}$

Dilute concentrations of rare earth (RE) dopants have been found to be effective in contributing damping in Ni$_{81}$Fe$_{19}$ in 4.2 and 4.3. The effectiveness of the damping contribution scales roughly with the nominal orbital moment $<L_{RE}>$ of the dopant 4f shell. Consistent with this idea, and with early measurements of FMR linewidth in RE-substituted YIG, Gd dopants have shown no significant effect on damping.

Direct evidence has not been available previously to link RE impurity magnetization states to precessional damping, either in YIG or in modern thin-film magnetic systems. The rare earth magnetization is usually approximated as that of an isolated 4f shell, occupied as 4f$^{Z-57}$, where $Z$ is the atomic number of the RE impurity; spin, orbital, and total moments ($S, L, J$) are calculated using Hund’s rules. This approach yields moments which agree well with experimental moments except for metallic lanthanides near Eu; measurement of the magnetic character of rare earths in a metallic alloy is therefore worthwhile.

X-ray magnetic circular dichroism (XMCD) is an ideal tool to characterize the magnetic character of RE dopants. Orbital and spin moments $<L>$ and $<S>$ can be measured separately on individual atomic sites using sum rules. High resolution XMCD data have been measured previously in Tb single crystals at M$_{4,5}$ edges and in Gd single crystals at the M$_5$ edge. To our knowledge, however, sum rules have not yet been applied to the dilute RE impurities in transition metal ferromagnets relevant for controlled damping.

**TEY mode XMCD characterization of Gd and Tb dopants in Ni$_{81}$Fe$_{19}$**

In this section, we use XMCD to measure spin to orbital moment ratios of Gd and Tb (2%) in Ni$_{81}$Fe$_{19}$ (50 nm). We show that for these elements, the calculated $L/S$ ratios from a 4f$^{Z-57}$ shell are verified. A fivefold enhancement in GHz relaxation rate from Tb dopants is accompanied by a large orbital moment fraction on the Tb site, indicating that spin-lattice coupling is decisive in enhancing relaxation.
Films were deposited by ion beam sputtering. Doping concentrations were measured as 1.7% of Gd and 1.8% of Tb using Rutherford Backscattering Spectroscopy (RBS). Magnetization dynamics were measured within one day of deposition; samples were stored in a desiccator for less than a week before XMCD measurement. XMCD measurements were taken in total electron yield mode (TEY) at the UV ring of the National Synchrotron Light Source (NSLS), Brookhaven National Laboratory, Beamline U4B\[101\]. XMCD was measured for fixed circular photon helicity, 75% polarization, with pulsed magnetization switching \(H = \pm 300\, \text{Oe}\) at the sample; photon incidence was fixed at 45° with respect to the sample normal. To correct for the non-grazing incidence and incomplete circular polarization, XMCD measurement were divided by the factor \(0.75 \cos 45°\).

XMCD characterization of the local magnetization states on RE dopants is shown in Fig. 4.23. X-ray absorption spectra for Gd and Tb are shown in the top panel; XMCD measurements are shown in the middle panel (dots). The XMCD data have been smoothed using a polynomial fit with variable window position (lines); the smoothed data, integrated numerically over energy, are shown in the bottom panel.

The nominal XAS peak positions at \(M_5\) and \(M_4\) correspond within ±0.7 eV to published values for elemental Tb and Gd samples\[105,106,107\]. Main peaks are seen for Gd at 1184.7 eV (\(M_5\)), 1213.8 eV (\(M_4\)) and Tb at 1238.7 eV (\(M_5\)), 1271.7 eV (\(M_4\)). Additionally, Gd exhibits a small shoulder on the high energy side of \(M_5\) and a split peak in \(M_4\) consistent with previous findings.

XMCD characteristics are quite different for Gd and Tb. Gd shows a positive and negative peak at \(M_5\) and \(M_4\) respectively. Tb shows a net positive peak at \(M_5\), with a small negative peak on the low energy side, and a small disturbance at \(M_4\) on the threshold of experimental error. The positive MCD signal at the \(M_5\) edge implies that magnetic moments of both Gd and Tb are antiferromagnetically coupled to transition metals, which is confirmed in ref. [14] and consistent with our experimental results that saturation magnetization
Figure 4.23: XMCD characterization of RE dopant magnetism in Ni$_{81}$Fe$_{19}$:RE(2%), RE=Gd, Tb. Top: X-ray absorption spectra (XAS); middle: XMCD data (circles) with polynomial smoothing fit (lines); bottom: energy integral of XMCD spectra. The $M_5$ XMCD integral $A$ and $M_4$ XMCD integral $B$ are indicated.

decreases as concentration of doped rare earth increases. Tb observations are consistent with elemental Tb XMCD spectra found by van der Laan et al.\cite{105}; here the negative peak on $M_5$ and small positive and negative peaks at $M_4$ are roughly 15% and 7% the heights of the large positive peak at $M_5$.

The bottom panel shows the energy integrals of the two XMCD spectra, following the method applied by Chen et al.\cite{102}. $A$ and $B$ are the integrals over $M_5$ and $M_4$ XMCD, respectively. Orbital to spin number ratios $<L>/<S>$ obtained at $M_{4,5}$ edges can be
CHAPTER 4. CONTROL OF DAMPING AND PRECESSIONAL FREQUENCY BY RE DOPANTS IN Ni$_{81}$Fe$_{19}$ THIN FILMS

Table 4.2: Orbital to spin moment ratios $L/S$ on RE dopants RE=(Tb, Gd) in Ni$_{81}$Fe$_{19}$:RE(2%)(50 nm), as measured by XMCD and predicted by Hund’s rules. The range of $L/S$ estimate for Tb depends on the sum rule used; see text for details.

<table>
<thead>
<tr>
<th></th>
<th>Tb</th>
<th>Gd</th>
</tr>
</thead>
<tbody>
<tr>
<td>XMCD experiment</td>
<td>1.5 ± 0.3</td>
<td>0.0 ± 0.1</td>
</tr>
<tr>
<td>Hund’s rule, 4$f^{Z-57}$</td>
<td>1</td>
<td>0</td>
</tr>
</tbody>
</table>

determined from the formula\cite{82,83}

$$\frac{<L_z>}{<S_z>} = 2 \frac{|A| - |B|}{|A| + \frac{3}{2}|B|} \left(1 + \frac{<T_z>}{<S_z>}\right) \quad (4.5)$$

where $T_z$ is the magnetic dipole operator. We extract, for d, $A = B = 0.027 \pm 0.004$, and for Tb, $A = 0.026 \pm 0.002$, $B = 0.000 \pm 0.002$. $<T_z>$ and $<S_z>$ for all lanthanides, except Eu, have been estimated according to atomic calculations, yielding $<T_z/S_z>_{\text{Gd}} = -0.009$ and $<T_z/S_z>_{\text{Tb}} = -0.08$\cite{108}. Our estimates of orbital to spin number ratios (twice the magnetic moment ratios) are given in Table 4.2. We find $<L_z/S_z>_{\text{Gd}} = 0.0 \pm 0.1$, and $<L_z/S_z>_{\text{Tb}} = 1.5 \pm 0.3$. These values are in qualitative agreement with Hund’s rule estimates. Gd (Z=64, 4$f^7$) has $S = \frac{7}{2}$, $L = 0$ ($L/S = 0$), and Tb (Z=65, 4$f^8$) has $S = 3$, $L = 3$ ($L/S = 1$).

XMCD measurements show a great difference between the magnetic character of dilute Gd and Tb in Ni$_{81}$Fe$_{19}$. Gd is found to be pure spin type ($S$-state) and dilute Tb is found to have roughly equal parts spin and orbital moment. The approximation of the isolated 4$f^{Z-57}$ moment is broadly validated for these two RE dopants in Ni$_{81}$Fe$_{19}$, although the Tb $L/S$ value is roughly 50% higher than that found through calculation. Alternate handling of the spin sum rule, such as the conventional neglect of the $<T_z>$ term, increases the disagreement. Based on the conclusion of Teramura et al\cite{108}, the validity of the spin sum rule is not seriously in question for these two elements, although it may not hold for the lighter lanthanides.

The XMCD measurement verifies an important criterion for an atomistic basis of con-
distributed damping in RE-doped Ni$_{81}$Fe$_{19}$. It has been proposed that the presence of spin-orbit coupling is essential for the damping of uniform precession by electronic excitation\cite{39, 40}, which can ultimately be absorbed by phonon and dissipated as heat. Rare earth elements can provide local centers for spin-orbit coupling: the orbital moment of the RE can couple to the Fe, Ni spin system through the RE spin moment. A necessary criterion for this mechanism is the presence of an orbital moment on RE sites which enhance the damping. We have validated its presence in Tb and absence in Gd.

In conclusion, we have seen that XMCD characterization of Tb and Gd dopants in Ni$_{81}$Fe$_{19}$ reveals a large orbital moment fraction on Tb sites, accompanied by a large increase in precessional damping, but zero orbital moment on Gd, with no effect on precessional damping. The results provide support for the idea that spin-orbit coupling, through introduction of local orbital moments, is important for controlled damping from lanthanide dopants.

**Transmission-mode XMCD of magnetic moment alignment in Tb-doped Ni$_{81}$Fe$_{19}$**

Total electron yield (TEY) XMCD has been used in \cite{4.4.2} to confirm the nominal orbital-to-spin moment ratios of Tb and Gd dopants in Ni$_{81}$Fe$_{19}$ using sum rules. However, total projected moments could not be extracted by TEY, as is typical, due to the surface-sensitivity of the measurement and possible influence of an oxidized surface.

Transmission-mode XMCD is known to be a superior technique for quantitative extraction of total element-specific moments due to its minimal influence from surface oxidized layers, critical for RE species, its absence of yield saturation effect and absence of field-induced background drift\cite{102}. In this section, we have carried out transmission-mode XMCD measurements on Si/Si$_3$N$_4$(membrane)/(Ni$_{81}$Fe$_{19}$)$_{1-x}$Tb$_x$(100nm)/Au(3 nm) samples. We determined that the projected moment fraction of Tb is roughly 8% that of Ni and Fe, indicating noncollinear moment alignment of the rare earth species.
Thin film samples were grown by confocal sputtering from Ni$_{81}$Fe$_{19}$ alloy and elemental Tb targets, by UHV DC magnetron sputtering, onto Si/Si$_3$N$_4$ membrane substrates at a base pressure of 10$^{-9}$ Torr. A magnetic field of 20 Oe was applied to the substrate during deposition to induce the uniaxial anisotropy. Tb-doped Ni$_{81}$Fe$_{19}$ thin films (100 nm), with doping level 0% and 6%, were investigated in the study. All thin films were capped with a thermally evaporated 3 nm Au layer to prevent oxidation. Film thickness and alloy compositions were monitored by a quartz crystal microbalance and ex situ profilometry.

XMCD measurements were taken in transmission mode at the same beamline described in 4.4.2 with different facility. XMCD were measured for fixed circular photon helicity, 70% polarization, with pulsed magnetization switching ($H = \pm 40$ Oe) at the sample. The measurements were divided by the factor $0.70 \cos 45^\circ$ to correct for nongrazing incidence and incomplete circular polarization. Background spectra from the bare membrane were recorded for each energy interval and applied in the background subtraction.

Selected results of XMCD characterization of the local magnetization states on Fe, Ni, and Tb are shown in Fig. 4.24 and 4.25. The L$_2$ and L$_3$ edge MCD and summed XAS spectra and their integrals for Fe and Ni in the undoped Ni$_{81}$Fe$_{19}$ thin film are shown in Fig. 4.24, which correspond to the strong 2$p \to 3d$ excitations in transition metal ferromagnets. The M$_4$ and M$_5$ edge MCD and summed XAS spectra and their integrals for Tb in the 6% Tb-doped Ni$_{81}$Fe$_{19}$ are shown in Fig. 4.25, which correspond to the region of the 3d levels, due to $3d^{10}4f^n \to 3d^{9}4f^{n+1}$ transitions. Compared to the MCD and summed XAS spectra obtained in the TEY mode, higher quality spectra are obtained here with transmission mode. The Tb MCD spectra show a net positive peak at M$_5$ edge with a small negative peak on the low energy side and a small disturbance at M$_4$ edge. This is consistent with TEY mode result and the elemental Tb MCD spectra found by van der Laan et al [105].

Data analysis needed for the transmission spectra have followed the method of Chen et al [102]. Sum rules have been applied to extract projected orbital, spin, and total magnetic
Figure 4.24: L\textsubscript{2,3} edge XAS and MCD spectra measured by transmission-mode XMCD of Fe and Ni in undoped Ni\textsubscript{81}Fe\textsubscript{19} thin films. (a) and (c) are the summed XAS spectra and their integrals for Fe and Ni, respectively; (c) and (d) are the MCD spectra and their integrals for Fe and Ni, respectively.

moments of all the elements present.

In general, the magnitude of elemental moments is proportional to the magnetic circular dichroism (MCD, difference in absorption on magnetization/helicity switching) normalized to the x-ray absorption (XAS, averaged for magnetization switching). Spin and orbital moment ratios are given by the relative weight of MCD at the L\textsubscript{3} (Fe and Ni) or M\textsubscript{5} (Tb) edge (denoted as \( p \) in the figures) to that at the L\textsubscript{2}+L\textsubscript{3} (Fe, Ni) or M\textsubscript{4}+M\textsubscript{5} (Tb) edge (denoted as \( q \)). At the L\textsubscript{2,3} edges of transition metals (e.g., Fe, Ni), neglecting the \(< T_z > / < S_z > \) term, with angular integrals \( c = 1, l = 2 \) [83], orbital and spin moments
are given by

\[ m_{\text{orb}} = -\frac{4q(10 - n_{3d})}{3r} \]  (4.6)

\[ m_{\text{spin}} = -\frac{(6p - 4q)(10 - n_{3d})}{r} \]  (4.7)

where \( n_{3d} \) is the 3d electron occupation number of the specific transition metal atom. At the M_{4,5} edges of rare-earths (e.g., Tb), with angular integrals \( c = 2, l = 3 \), orbital and spin
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Table 4.3: Projected orbital and spin magnetic moments of Fe and Ni in the undoped Ni$_{81}$Fe$_{19}$ (0%) and those of Fe, Ni, and Tb in the 6% Tb-doped Ni$_{81}$Fe$_{19}$ (6%) thin films. For Fe and Ni, the experimental errors are estimated to be 5% for $m_{\text{orb}}/m_{\text{spin}}$ and 10% for $m_{\text{orb}}$ and $m_{\text{spin}}$. For Tb, the experimental errors are estimated to be 40% for $m_{\text{orb}}/m_{\text{spin}}$ and 50% for $m_{\text{orb}}$ and $m_{\text{spin}}$.

<table>
<thead>
<tr>
<th></th>
<th>Fe (0%)</th>
<th>Fe (6%)</th>
<th>Ni (0%)</th>
<th>Ni (6%)</th>
<th>Tb (6%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt; m_{\text{orb}} &gt; / &lt; m_{\text{spin}} &gt;$</td>
<td>0.05</td>
<td>0.02</td>
<td>0.08</td>
<td>0.014</td>
<td>0.54</td>
</tr>
<tr>
<td>$&lt; m_{\text{orb}} &gt;$ ($\mu_B/\text{atom}$)</td>
<td>0.08</td>
<td>0.01</td>
<td>0.06</td>
<td>0.002</td>
<td>0.06</td>
</tr>
<tr>
<td>$&lt; m_{\text{spin}} &gt;$ ($\mu_B/\text{atom}$)</td>
<td>1.78</td>
<td>0.51</td>
<td>0.73</td>
<td>0.15</td>
<td>0.11</td>
</tr>
<tr>
<td>$&lt; m_{\text{total}} &gt;$ ($\mu_B/\text{atom}$)</td>
<td>1.86</td>
<td>0.52</td>
<td>0.79</td>
<td>0.15</td>
<td>0.17</td>
</tr>
</tbody>
</table>

Moments are given by

$$m_{\text{orb}} = \frac{2q(14 - n_{4f})}{r}$$  \hspace{1cm} (4.8)

$$m_{\text{spin}} = \frac{(5p - 3q)(14 - n_{4f})(1 + 3\frac{<T_z>}{<S_z>})^{-1}}{r}$$  \hspace{1cm} (4.9)

where $n_{4f}$ is the 4f electron occupation number of the specific RE atom. Thus we have

$$\frac{m_{\text{orb}}}{m_{\text{spin}}} = \frac{2q}{5p - 3q}$$ for Fe and Ni, and

$$\frac{m_{\text{orb}}}{m_{\text{spin}}} = \frac{2q}{5p - 3q}(1 + 3\frac{<T_z>}{<S_z>})$$ for Tb.

For $n_{3d}$, we used values of 6.5 for Fe and 8.5 for Ni, as averaged from those values reported in recent theoretical calculations\[109,110,111\]. For $n_{4f}$, we used the elemental value of 8 for Tb according to $4f^{2-57}$. According to the atomic calculations done by Teramura et al\[108\], we have $< T_z > / < S_z > = -0.08$ for Tb. Using these assumptions, we can determine the $m_{\text{orb}}$ to $m_{\text{spin}}$ ratios, as well as the individual $m_{\text{orb}}$ and $m_{\text{spin}}$ values for Fe, Ni, and Tb. The results are listed in Table 4.3.

We find $m_{\text{orb}}/m_{\text{spin}}$ estimates of 0.05 for Fe, 0.08 for Ni, in the undoped Ni$_{81}$Fe$_{19}$ thin film; and of 0.54 for Tb in the 6% Tb-doped Ni$_{81}$Fe$_{19}$ thin film. The values of Fe and Ni are in reasonable agreement with those found previously\[112\]. The Tb result is close to the value of 0.5 from Hund’s rule; the difference can be attributed to the uncertainty in the value of $< T_z > / < S_z >$ in the alloy. Furthermore, we measured projected spin moments of 1.78 $\mu_B$/atom for Fe and 0.73 $\mu_B$/atom for Ni in the undoped case, close to the values of previous findings\[113,112,102\]. In the 6% doped film, these projected values are
reduced to roughly 25% of their undoped values, attributable to our inability to saturate the material at the available field of 40 Oe. The degree of alignment of Tb is far smaller, with only 0.11 $\mu_B$/atom for spin moment, that is, only roughly 8% of that of Fe and Ni at 6% doping level. The total projected moment of Tb is only 2% of its ideal saturated level of 9.0 $\mu_B$/atom\(^{[114]}\).

Low levels of dichroism and low projected moment observed on randomly distributed Tb dopants in polycrystalline Ni\(_{81}\)Fe\(_{19}\) can be interpreted as an accurate reflection of a small average projected moment from Tb sites in the alloy. Based on the experimental results of the low projected Tb moment, which nonetheless reverses with low applied fields, the magnetic state of the alloy thus appears to be sperimagnetic with a strong dispersion angle of Tb moments, nonetheless aligned net antiparallel to Fe and Ni moments. Much higher applied fields (to several Tesla) will be necessary to test whether saturation moments of Tb can be observed.

In conclusion, transmission mode XMCD has been used to characterize the alignment of elemental moments in Tb-doped Ni\(_{81}\)Fe\(_{19}\) thin films. Compared with TEY mode XMCD data, transmission mode give greatly improved Fe and Ni and somewhat improved Tb XMCD spectra. The low projected Tb moment, which nonetheless reverses with low applied fields, indicates a sperimagnetic alignment with respect to the Fe and Ni moment alignments in the cosputtered polycrystalline alloy.

4.5 Summary and discussion

4.5.1 Summary

A systematical study has been conducted on the magnetization dynamics of rare earth doped Ni\(_{81}\)Fe\(_{19}\) thin films. Of the studied rare earth dopants, Sm, Tb, Dy, and Ho all contribute an increase on the damping in Ni\(_{81}\)Fe\(_{19}\). The dynamic response spans from underdamped
to nearly critically damped, or even to overdamped situation.

Contributed damping per atomic percentage was calculated for each RE dopant. Sm, Dy, and Ho all increase damping in Ni$_{81}$Fe$_{19}$ more effectively than Tb does. The contributed damping from Ho is four times greater than that from Tb, and the effect is greater than seen in studies of Ingvaasson et al.\cite{115}. Recently, Ho doped Ni$_{81}$Fe$_{19}$ has been used by Seagate to suppress spin-momentum-transfer (SMT)-based noise in CPP (current perpendicular to the plane) spin valves.

The contributed damping of the RE dopants is compared with the theoretical orbital momentum of the free RE ions, and a good agreement is found between them. The greater the orbital momentum, the greater damping the element contributes. The agreement further proves damping is contributed as per atom basis, which means the effect can be scaled to small structures.

Eu does not contribute to the damping in Ni$_{81}$Fe$_{19}$. However, it boosts the resonance frequency over a large range (\(~500\) MHz). While $g^2M_s$ does not change much from undoped Ni$_{81}$Fe$_{19}$, $g$ factor increases from 2.0 to 2.3 for 3\% of Eu. Dynamic anisotropy is also enhanced in Eu-doped films. More thorough explanation for the frequency boost is still under investigation.

Undoped/RE-doped Ni$_{81}$Fe$_{19}$ bilayer structure was deposited and its magnetization dynamics was studied. Both the magnetic damping (RE=Tb) and the precessional frequency (RE=Eu) can be tuned over a large range in the bilayers. The result will benefit the magnetoelectronic device fabrication from the point of view that dopants are removed from ferromagnetic/nonferromagnetic interfaces so that spin transport properties remain; magnetization dynamics is controlled through the exchange interaction.

XMCD measurements on Tb and Gd doped Ni$_{81}$Fe$_{19}$ films reveal that a large orbital moment fraction resides on Tb sites, which is responsible for the large contribution to magnetic damping, but zero orbital moment on Gd, with no effect on damping. The results
provide proof that spin-orbit coupling, through introduction of local moments, is important
for controlling damping in Ni$_{81}$Fe$_{19}$.

4.5.2 Discussion

As described in 2.5.1, spin-orbit coupling dependence of magnetization relaxation is pre-
dicted by both spin-conserving scattering and spin-flip scattering mechanisms. In both
cases, there are $\lambda \sim (g - 2)^2$ and $\frac{\mu_L}{\mu_S} = \frac{g^2}{4} - 1$[40, 49]. From XMCD results, $\frac{\mu_L}{\mu_S}$ does scale
plausibly, with larger ratio corresponding to larger damping and zero corresponding to no
damping contribution from RE dopants. However, since we assume constant magnetiza-
tion in most of our experiments, exact $g$ factor values could not be obtained. More careful
measurement of $g$ factor is needed.

The difference between spin-conserving scattering and spin-flip scattering mechanisms is
that the former predicts $\lambda$ proportional to $1/T$, where $T$ is temperature, while the latter pre-
dicts $\lambda$ proportional to $T$. It was found that for low temperature, spin-conserving scattering
dommates, while at high temperature, spin-flip scattering dominates. At room temper-
ature, both terms are of roughly equal weight. To confirm this, temperature dependent
ferromagnetic resonance is necessary.

There are other damping mechanisms which might also be responsible for relaxation in
RE doped Ni$_{81}$Fe$_{19}$. For example, we could not exclude direct magnon-phonon scattering
mechanism (see 2.5.1). To clarify on this, characterization of magnetoelastic energy is
needed. Dry friction mechanism is also possible. FMR experiments with varying rf field
amplitude (microwave power) are helpful since the FMR lineshape including linewidth and
amplitude will change with varying rf field amplitude in dry friction mechanism.

Two-magnon mechanism, however, is not important in the magnetic damping of RE
doped Ni$_{81}$Fe$_{19}$. Recent FMR study on these samples show that the in-plane linear depen-
dence of the linewidth on the frequency remains even for very low frequencies[116].
Safonov proposed so-called, "slow-relaxing ion" and "fast-relaxing ion" mechanisms for magnetic relaxation\textsuperscript{[44]}. In the former mechanism, small magnetization oscillations modulate the impurity levels in the vicinity of thermal equilibrium. This energy modulation is absorbed by the lattice at an extremely fast rate. The latter mechanism is effective when the transverse relaxation rate of the impurity is so fast that the magnetization oscillations can excite direct transitions between impurity levels. Different equations of magnetization dynamics were derived for each mechanism. The damping parameter can be directly calculated from these microscopic impurity relaxation processes. If we carefully apply the calculation to our RE-doped Ni\textsubscript{81}Fe\textsubscript{19}, we can compare the calculated damping parameter with our experimental results and confirm these two damping mechanisms.

Other open issues include the origin of precessional frequency boost in Eu. The tendency of the boost seems to agree with the enhancement of $H_K$, which possibly arises from texture of growth. The interesting yet confusing aspect is the remarkably large value of $g$ factor (as large as 2.3) in Eu doped films with no effect on damping. It is noteworthy that Eu is the one RE for which valence is not always $4f^{Z-57}$ in the metallic state. Open issues also include the reason for the nonlinear response of damping constant vs. Tb concentration in undoped/Tb-doped Ni\textsubscript{81}Fe\textsubscript{19} bilayers. It can be clearly seen that there are three steps in Fig. 4.16. Perhaps single-sided excitation happens in the bilayers. Cavity FMR measurements should be carried out to see if different behavior shows up.
Chapter 5

Composition-based reduction of damping in MgO/Fe$_{1-x}$V$_x$ epitaxial thin films

5.1 Introduction

In this chapter, we ask whether impurities can have the opposite effect of reducing $\lambda$, particularly in iron (Fe.) Iron has the lowest $\lambda$ (to 57 MHz, or $\alpha = 0.0019$) of any known metallic ferromagnet; by doping this material, we seek lower damping than is known in any metal. The task is analogous (see Fig. 2.8) to finding a metal with room-temperature electrical resistivity than lower than that of copper. Pursuing the analogy with electronic resistivity, however, it does not seem obvious how dopants may be useful. A localized moment with high relaxation rate (as e.g. Ho in Ni$_{81}$Fe$_{19}$ in the previous chapter) clearly increases $\lambda$, but a localized moment with a lower relaxation rate should be ineffective, as the host could be expected to dominate $\lambda$.

To reduce the total intrinsic damping, larger impurity concentrations are likely to be
necessary. In some sense, the properties of the host need to be changed. Isostructural alloys can be expected, on the basis of the Slater-Pauling curve, to form magnetic materials with properties determined principally by the average atomic number $Z$. For compositions near $Z = 25$, one less than Fe ($Z = 26$), very old Einstein-de Haas measurements indicate that orbital moments and associated spin-lattice coupling can be nulled out. Such a material can be expected to have reduced damping $\lambda$, as all intrinsic relaxation models suppose some coupling between the spin system (magnetization) and the lattice (heat).

We have explored epitaxial thin films of BCC Fe-lower-$Z$ alloys, principally $\text{Fe}_{1-x}V_x$, to determine any influence of composition on reducing $\lambda$ in the alloy.

5.1.1 Epitaxial MgO/Fe, its application in magnetic recording, and the need for low-loss Fe alloys

As of even a few years ago, Fe thin films would not have been promising for applications in spin electronic information storage, their relaxation rate notwithstanding. Fe/MgO/Fe(001)-based magnetic tunnel junctions, despite theoretical predictions taking advantage of the favorable epitaxial structure available in this system, had shown tunneling magnetoresistance (TMR) ratios an order of magnitude lower than those based on Co or Ni$_{81}$Fe$_{19}$. This situation has changed very recently with the results of Parkin et al and Yuasa et al, demonstrating TMR in excess of 400% in Fe/MgO/Fe trilayers. In the future, a low-$\lambda$ Fe alloy, which can reduce thermal-fluctuation-based ”mag-noise” in Fe/MgO/Fe, could be highly valuable for magnetoresistive sensors or M-RAM elements.

Since the late 1990’s, Fe/MgO/Fe trilayers have been very interesting candidates for magnetic tunnel junctions, based on structural and theoretical considerations. Epitaxial ferromagnetic tunnel junctions are possible in this system, unlike in the more typical amorphous Al$_2$O$_3$-based junctions, due to a favorable lattice match (within 3.7%) between Fe and MgO. Structural aspects of the system are known from earlier studies\[117]\. Epitaxial rela-
tions between the bcc Fe layer and the MgO(100) rock-salt structure are Fe(001)//MgO(001) and Fe[100]//MgO[110]. The experimental Fe lattice constant is 2.866 Å, and the MgO lattice constant is about \( \sqrt{2} \) times that of the Fe. Therefore, the Fe lattice is rotated by 45° relative to the MgO lattice. Fe atoms are expected to be positioned on top of O atoms, based on calculations and low-energy electron diffraction (LEED) characterization\[118,119]. The Fe-O separation of 2.0 Å is considered most probable.

The translational symmetry present in epitaxial Fe/MgO/Fe has facilitated \textit{ab-initio} studies of magnetotransport. Butler \textit{et al.} calculated the tunnelling between Fe(100) electrodes separated by MgO tunnelling barriers by first principles electronic structure techniques\[120,121]. LEED studies show that the Fe-O distance is almost exactly equal to the distance between the neighboring MgO atomic planes\[122]. The electron population at the MgO interface plane is virtually the same as for the clean MgO surface, and Fe interface plane also behaves like a free Fe surface\[119]. Based on these, the tunnelling magnetoresistance (TMR) value is calculated numerically and the calculated value is very large, in excess of 1000% for perfect Fe/MgO/Fe junctions due to band structure effects.

Experimentally, Fe/MgO/Fe epitaxial MTJs had shown disappointingly low TMR values, until recently. A fully crystalline Fe/MgO/Fe(100) MTJs have been achieved by both MBE and pulsed laser deposition (PLD) of MgO and Fe on polished single crystal Fe(100) substrates and Fe whiskers\[123], but no TMR could be measured. Most recently, fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJs were fabricated by MBE and a MR ratio of 88% at room temperature (146% at T=20 K) was observed, which was the highest value yet reported\[124]. Recent results \[125,126\] have shown MR values in the 300-400% range.

As described in the first section, a low-\( \lambda \) (low-loss) ferromagnetic material is highly desirable for signal-to-noise (SNR) considerations in nanoscale spin electronic sensors. Near the ferromagnetic resonance (FMR) frequency, electronic noise will be dominated by thermal fluctuations of the magnetization, where the zeeman energy of the magnetic layer is
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comparable to thermal energy. SNR is predicted to be *inversely proportional to the damping*, and, remarkably, independent of \(\Delta R/R\) in this small size, high-frequency regime.[67]. If damping \(\lambda\) is decreased in epitaxial Fe-low Z alloys, it is easy to imagine e.g. Fe\(_{1-x}V_x\) as a ”drop-in” replacement for the free Fe layer in the structure.

It bears mentioning here as well that there are many potential applications for low-loss ferromagnetic thin films outside of magnetic information storage applications. Integrated, thin-film 0-40 GHz frequency-domain devices, which take advantage of the large microwave power absorption at FMR frequencies, have been proposed for epitaxial Fe on GaAs. However, the large frequency linewidth \(\Delta f \simeq \lambda\) for Fe has impeded the realization of integrated notch / band-pass filters, inductors, and circulators.

### 5.1.2 Prospects for reduced relaxation rates in Fe\(_{1-x}V_x\)

A principal reason to expect reduced relaxation rate \(\lambda\) in Fe\(_{1-x}V_x\) lies in very old (1950’s era) studies of magnetism through the Einstein de Haas effect. \(g\) was estimated directly through the mechanical torque imparted to bulk cylinders upon magnetization reversal. \(g\), and thus orbital moments \(\mu_L\) through the relationship \(g_{\text{eff}} \simeq 2 (1 - \mu_L/\mu_S)\), were found to be reduced to \(g \simeq 2\) for \(Z = 25\) transition metal alloys. The systematic variation of \(g\) with the number of electrons per atom, \(Z\), is shown in Fig. 5.1[127]. At this composition \(m\mu_L\) can be expected to be roughly nulled, and pure spin-type moments may be present. The \(g = 2\) value is found for \(Z \simeq 25\), the Mn composition, or for V \((Z = 23)\), at Fe\(_{67}V_{33}\). However, the Fe-V system itself has not been investigated, and some significant scatter is present among the alloy systems explored.

Recent *ab-initio* calculations of magnetocrystalline anisotropy energy (MAE) in Fe\(_{1-x}V_x\) support the view that alloying Fe with lower-Z elements can reduce spin-lattice coupling. In an ”ab-initio search for a high-permeability materials based on BCC iron,” MAE and linear magnetostriction, two quantities also linked closely with spin-lattice coupling, are
Figure 5.1: Landé $g$ factor as a function of electron concentration (alloying) in ferromagnetic metals. Alloying Fe with lower-Z element could result in the free-electron state. Predicted to be reduced drastically in a 10% V alloy. Calculations were carried out by the spin-polarized relativistic Korringa-Kohn-Rostoker method\cite{128}. The reduction of MAE and magnetostriction might result from a slight reduction in band filling and modest lattice expansion, although a clear physical conclusion cannot be located in the treatment.

Qualitatively, all models of intrinsic relaxation refer to coupling between spin moments and the lattice. This coupling can be effected strongly through orbital moments. If orbital moments are not present, spin-lattice coupling should be reduced. Explicitly, in the Kambersky model, Gilbert damping $G$ is proportional to $(g - 2)^2$, where $G = \alpha \gamma M_s$, $\gamma$ is the gyromagnetic ratio, $g$ is the Landé $g$-factor, and $g - 2$ is its departure from the free electron result\cite{40}. An identical expression was delivered by Eliot for electron spin resonance in semiconductors\cite{96, 97}. This is a measure of the spin-orbit coupling since $g - 2 \approx -\lambda/\Delta$, where $\lambda$ is the spin-orbit coupling energy, and $\Delta$ is the separation of nearly degenerate energy levels which it splits. Experimentally, the Kambersky relationship has been confirmed reasonably in FMR of Fe$_x$Co$_{1-x}$ thin film alloy system\cite{96}. A correlation was shown between
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$g$ and $G$, where $g$ values span a range from $2.09 \leq g \leq 2.14$, with $G$ spanning $0.5 \sim 2.5 \times 10^8$ s$^{-1}$.

Some prior work exists on Fe/V multilayers, which form a well-lattice-matched epitaxial system, one treatment of which contains FMR results (cite the Farle article). However, relaxation studies of Fe$_{1-x}$V$_x$ alloys are not to be found in the literature. From all the studies on Fe/V interfaces[129, 130], it is clear that there exists an induced magnetization at V interface (values from $0.3\mu_B$ to $1.5\mu_B$) antiferromagnetically coupled with the Fe substrate, and a decrease (5 - 20%) in the magnetization at the Fe interface atoms. However, there remains a strong controversy in two points: (1) the short- or long-range induced spin polarization in V and (2) the dependence of the magnetic profile of the Fe/V interfaces on the crystallographic orientation.

5.1.3 FMR measurements of Fe to determine the magnetic anisotropy and Gilbert damping

Magnetic anisotropy in Fe thin films

Ferromagnetic resonance (FMR) is one of the most powerful experimental techniques in the study of thin film magnetic properties. While the use of the FMR linewidth to estimate relaxation rates can be understood intuitively, FMR can also be quite useful for measurements of other fundamental quantities, such as magnetic moments $B_s$ and magnetocrystalline anisotropy (MAE). FMR has been used extensively to characterize anisotropies in systems of reduced dimension, particularly interface anisotropy in superlattices[131], but its application requires some explanation.

The in-plane angle dependent FMR resonance field can be simulated by expressing the free energy density with in-plane anisotropy constants in Eq. [2.21] and setting these constants as fitting parameters. As an example we consider the resonance condition in tetragonal symmetry, which occurs frequently in thin films, including Fe thin film. The free energy
density will be \[ U = MH \cos \theta_H \cos \theta + \sin \theta_H \sin \theta \cos(\phi - \phi_H) + 2\pi M^2 \cos^2 \theta + (K_{12} + K_{||4}) \sin^2 \theta \\
+ (K_{\perp 4} - \frac{7}{8} K_{||4}) \sin^4 \theta - K_{||2} \sin^2 \theta \cos \phi - \frac{1}{8} K_{||4} \sin^4 \theta \cos 4\phi, \] (5.1)

where \( M \) and \( H \) are the magnetization and applied static field, \( \phi \) and \( \phi_H \) are the in-plane angle of magnetization and applied filed with respect to the symmetry axis, \( \theta \) and \( \theta_H \) are the out-of-plane angles (\( \theta = \theta_H = 0^\circ \) if \( M \) and \( H \) are perpendicular to the plane and \( 90^\circ \) if parallel to the plane), \( K_{||2} \) and \( K_{||4} \) are the second- and fourth-order constants of in-plane magnetic anisotropy energy (MAE), and \( K_{12} \) and \( K_{||4} \) are the out-of-plane MAE constants. For cubic symmetry one has \( K_{\perp 4} = K_{||4}. \) The first term in Eq. (5.1) is the Zeeman energy, the second term is magnetostatic energy, and the following terms are the anisotropy energy.

A simple result can be seen for the perpendicular condition. In this configuration, not used in this thesis, the static applied field \( H \) and magnetization \( M \) are oriented perpendicular to the film plane. The solution in this case leads to

\[ \omega = H_{res} - 4\pi M + \frac{2(K_{12} + K_{||4})}{M}. \] (5.2)

In the parallel configuration, used in this work, where the static field \( H \) and magnetization \( M \) are oriented in the film surface, the resonance field \( H_{res} \) is given by \[ (\frac{\omega}{\gamma})^2 = [H_{res} \cos(\phi - \phi_H) + 4\pi M - \frac{2K_{||2}}{M} + \frac{K_{||4}}{2M}(3 + \cos 4\phi)][H_{res} \cos(\phi - \phi_H) + \frac{2K_{||4}}{M} \cos 4\phi]. \] (5.3)

Experimentally, one has a set frequency \( \omega / 2\pi \), assumed value of \( M_s \) known from other techniques, independent variable \( \phi \), and dependent variable of applied field magnitude \( H_{res} \) to match resonance. Anisotropies \( K_4 \) and \( K_2 \) are measured through a full fit of \( H_{res}(\phi) \). Note that the fit requires numerical solution of simultaneous equations, as \( \phi_M \), the magnetization
angle, can be solved only through minimizing the energy in Eq. 5.1, which is coupled to the choice of $K_4$ and $K_2$.

The in-plane angle dependent FMR was measured at 9.78 GHz for Fe single crystal films on GaAs (100) and the data were fitted into Eqs. 5.3 and 5.2. Assuming the magnetization does not change much with varying thickness and take it as 1600 Gs, the various anisotropy constants were determined by the fit. The results confirmed that the in-plane anisotropy shows a uniaxial symmetry when the film thickness is less than 12.7 ML and $K_{//2}$ is high. $K_{//2}$ decreases as film thickness increases due to lattice relaxation. Cubic magnetic anisotropy appears after 8.4 ML and its strength increases with film thickness, which makes fourfold symmetry seen clearly in the in-plane anisotropy. $K_{//4}$ reaches about 60% of the value of bulk Fe for 33 ML thickness. By measuring FMR with the dc magnetic field inclined to the film surface, one can identify the perpendicular uniaxial and fourfold anisotropy. $K_{\perp2}$ decreases and $K_{\perp4}$ increases with increasing film thickness. $K_{\perp4}$ gradually approaches the bulk cubic anisotropy constant.

**FMR linewidth in Fe thin films**

The FMR linewidth can be used to characterize the contribution of the intrinsic damping mechanism and determine the role of magnetic inhomogeneities. The peak-to-peak distance in FMR spectrum is the linewidth $\Delta H$. The studies of FMR showed that the linewidth on the microwave frequency follows a frequency-dependent relationship $\Delta H = \Delta H_0 + 1.46 \frac{C}{\gamma M_0} \omega$. $\Delta H_0$ describes the inhomogeneous linewidth broadening due to magnetic inhomogeneities. The second term is the contribution from intrinsic damping of the magnetization precession. It is a measure of the microscopic mechanism by which the absorbed microwave energy is dissipated from the spin system to the lattice vibrations. The intrinsic linewidth increases linearly with the frequency.

A high quality sample with less magnetic inhomogeneity is important in reducing the
linewidth broadening. Thick films in hundreds of nanometers also have broader linewidth due to the exchange conductivity mechanism; an inhomogeneous line broadening results from the spatial variation of the rf magnetization on a scale characterized by the microwave skin depth. MBE films can be grown with superior quality, which exhibit lower microwave losses than well prepared bulk samples. FMR at 35 GHz on a 13.6 nm thick Fe bcc single crystal film grown on fcc ZnSe (001) epilayers on GaAs (001) by MBE shows a linewidth of 45 Oe, which represents the narrowest 35 GHz ferromagnetic resonance linewidth ever measured in single crystal Fe. This value approaches the theoretical lower limit set by Landau-Lifschitz broadening in Fe\cite{36}. Recently, polycrystalline Cu/Fe/Cu were made by magnetron sputtering or electron beam deposition on polished Si wafers with native oxide surfaces\cite{37}. The frequency dependence of the FMR linewidth for the Cu1/Fe20/Cu1nm structure showed the nonintrinsic contribution was only a few Oe. FMR linewidth at 9.5 GHz was as low as 15 Oe.

There is a persistent question that concerns if Landau-Lifschitz (LL) and Gilbert relaxation terms is applicable in describing the intrinsic damping. LL and Gilbert models predict that the FMR linewidth in the parallel and perpendicular configuration have a strictly linear dependence on the microwave frequency, and the linewidth is identical for both configurations if the measurements are carried out along equivalent crystallographic directions. To test the validity of this prediction, one needs high quality single crystals with perfect surfaces to obtain convincing FMR results. Fe whisker prepared by chemical vapor transport provided such perfect systems. FMR linewidth on (100) planes of Fe whiskers were measured as a function of frequency over a wide range, from 20 to 100 GHz\cite{132}. The experimental data were fitted by calculations of LL equation of motion together with Maxwell equations. The results clearly showed that the intrinsic damping is well represented by LL damping, $\lambda_{Fe} = 5.72 \times 10^7$ s$^{-1}$. 

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5.2 Experiment: epitaxial structure characterization

The MgO/Fe\textsubscript{1−x}V\textsubscript{x} epitaxial thin films were deposited by DC magnetron sputtering (see 3.1.2). V concentration $x$ ranges from 0 to 52%. During deposition the substrate was heated with temperatures below 300°C.

5.2.1 XRD characterization of MgO/Fe epitaxial structure

The structural quality of the epitaxial films was characterized by x-ray diffraction. Fig. 5.2 presents high-angle diffraction data taken from MgO/Fe(50nm) samples grown at 200°C. The scan was taken with a fixed-slit detector, fix-slit tube diffractometer and Cu K\textsubscript{α} radiation. The XRD spectrum was identified and indexed by comparing with the ICDD PDF database. The spectrum shows only one diffraction peak associated with Fe, Fe (200) peak. The second peak with the strongest intensity corresponds to the MgO substrate, MgO.

![Figure 5.2: High-angle specular XRD scan from MgO/Fe epitaxial thin film. Note the intensity cut on MgO (200) peak. The spectrum shows only (100) related features for both Fe and MgO, indicating the film has a single growth orientation.](image-url)
(200), witnessing the epitaxial growth orientation. The third peak, Al (111), comes from the aluminum stage on which the sample was placed.

The lattice constant of BCC Fe is 0.287 nm, while that of rocksalt MgO is 0.412 nm, which is about $\sqrt{2}$ times of that of Fe. There have been several studies of the growth of Fe on MgO or MgO on Fe and of the interface between the iron and MgO\cite{120, 133, 121}. It was found that the lattice mismatch between Fe and MgO is accommodated by a 45° rotation as Fe(100)[110]//MgO(100)[100], with Fe atoms sitting atop the O atoms when Fe is deposited onto a clean MgO (100) surface. A diagram of the interfacial structure is shown in Fig. 5.3. The in-plane alignment was not investigated in this thesis.

![Diagram of interfacial structure](image)

Figure 5.3: Interface for MgO(100)/Fe(100) epitaxial structure. Large atoms are iron. Darker atoms above iron atoms represent oxygen. Small light atoms represent magnesium.

The effect of deposition temperature on the structural quality of the epitaxial thin films is shown in Fig. 5.4. Fig. 5.4(a) shows the XRD Fe (200) peaks taken from MgO/Fe (50nm) epitaxial thin films with growth temperature between room temperature and 300°C. No (200) diffracted intensity is present when the film was grown at room temperature. For those grown at 200°C and 300°C, both spectra show a sharp, well-defined (200) peak. A
simple measure of the structural quality of a thin film is the width of a rocking curve. Fig. 5.4 (b) shows the rocking curves taken at the maximum intensity of the spectral Fe (200) diffraction for the films deposited at 200° and 300°. The full widths at half-maximum (FWHMs) are very small in both cases, 0.67° for 200°C deposition and 0.68° for 300°C deposition, which indicate the structural quality of these samples is indeed high. However, the easy axis hysteresis loop measured by VSM demonstrated a smaller coercivity for the sample deposited at 200°C. This will be described later. Therefore, we determined that 200°C is the optimal growth temperature for our epitaxial deposition.

Figure 5.4: XRD characterization of epitaxial Fe thin films deposited at different temperatures. (a) symmetric scan around Fe (200) peak for RT, 200°C and 300°C depositions; (b) rocking curves on Fe (200) diffraction for films deposited at 200°C and 300°C, respectively.

Fig. 5.5 shows the XRD characterization on MgO/Fe epitaxial thin films which were deposited with gas purifier on and with cleaned substrate, respectively. Both were grown at 200°C. Fig. 5.5 (a) and (b) are the symmetric scan and rocking curve for the film deposited with gas purifier on; (c) and (d) are those for the film deposited on the substrate cleaned with 1NH₄OH:1H₂O₂:100H₂O etchant. In both cases, the symmetric scan shows a
sharp, well-defined Fe (200) peak and the rocking curve demonstrates very small FWHM around Fe (200) diffraction, 0.64° in (b) and 0.62° in (d). Comparing with the sample deposited with gas purifier off and on as-received MgO substrate, there is some, although finite, improvement in the crystal quality.

![Figure 5.5](image)

Figure 5.5: XRD characterization of epitaxial Fe thin films deposited with gas purifier on and on cleaned substrate, respectively. (a) symmetric scan, (b) rocking curve for the sample deposited with gas purifier on; (c) symmetric scan, (d) rocking curve for the sample deposited on cleaned MgO.

5.2.2 Effect of V alloying on the crystal structure of Fe$_{1-x}$V$_x$ epitaxial thin films

MgO(100)/Fe$_{1-x}$V$_x$(100) epitaxial thin films is shown in Fig. 5.6. Symmetric scans around Fe$_{1-x}$V$_x$ (200) peak are shown in the left; rocking curves on the (200) diffraction are shown in the right. All samples show sharp and well-defined (200) peak in the symmetric scan and small FWHM in the rocking curve, indicating the presence of a high degree of structural perfection in the epitaxial thin films. This is in agreement with the fact that both Fe and
V have bcc crystal structure and there is only 5% lattice mismatch between Fe and V, which provides the high solid solubility and good crystalline quality of the epitaxial thin film. As V concentration increases, the (200) peak position in the symmetric scan shifts to lower angles, indicating that the lattice parameter becomes larger from the V addition. The FWHM on the (200) diffraction slightly broadens as V is introduced into Fe. Intensity decreases in both the symmetric scan and the rocking curve as V concentration increases. The decrease in the intensity and the slight broadening might be caused by the possible slight disorder or structural defects caused by addition of V into Fe.

![Figure 5.6](image_url)

**Figure 5.6:** XRD characterization of MgO(100)/ epitaxial thin films. Left: symmetric scan around Fe$_{1-x}$V$_x$ (200) peak; right: rocking curve on Fe$_{1-x}$V$_x$ (200) diffraction.

Fig. 5.7 shows the extracted Fe$_{1-x}$V$_x$ (200) peak position and FWHM of the rocking curves from Fig. 5.6. The plot on the left shows the dependence of the (200) peak position on V concentration $x_V$; The one on the right shows the dependence of FWHM of the rocking
curve on \( x_V \). As V concentration changes from 0 to 52\%, the (200) diffraction peak position shifts leftward by 1.26\(^\circ\), and the FWHM of the rocking curve increases from 0.67\(^\circ\) to 1.12\(^\circ\). It is clear that the broadening of rocking curve FWHM (less perfect crystalline structure) accompanies the expansion of the lattice.

Figure 5.7: Dependence of Fe\(_{1-x} V_x\) (200) peak position and FWHM of rocking curve on V concentration \( x_V \). Left: (200) peak position vs. \( x_V \); right: FWHM of rocking curve vs. \( x_V \). The lines are for the guidance of the eye.

From the peak position, we can calculate the lattice constant of the BCC Fe\(_{1-x} V_x\) structure according to the Bragg’s Law,

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

(5.4)

in which \( \lambda = 1.5406\,\text{\AA} \) is the wavelength of the incident Cu K\(\alpha_1\) source beam, \( d \) is the spacing between Fe\(_{1-x} V_x\) (200) planes, which is equal to \( \frac{1}{2} \) of the lattice constant of the BCC crystal structure. The extracted lattice constants for BCC Fe\(_{1-x} V_x\) crystalline structure are shown in Fig. 5.8. As V concentration increases, the lattice constant also increases. This is in agreement with the fact that the lattice constant of V (0.302 nm) is slightly (5\%) larger than that of Fe. Also plotted in the figure is the dashed line, the values calculated from Vegard’s law [134], which is an approximate empirical rule that states a linear relation
exists, at constant temperature, between the crystal lattice constant of an alloy and the concentrations of the constituent elements. We can see that the concentration dependence of lattice constant in $\text{Fe}_{1-x}\text{V}_x$ epitaxial structure exhibits an apparent deviation from Vegard’s law.

![Graph showing lattice constant vs. V concentration](image)

Figure 5.8: Lattice constant of $\text{Fe}_{1-x}\text{V}_x$ crystalline structure extracted from Fig. 5.7 as a function of V concentration. The solid circles are the extracted lattice constants, the dashed line is the calculation from Vegard’s law, and the solid line is the empirical relation calculation from Shiga.

There are different explanation for the notable deviation of Fe-V alloy lattice constant from Vegard’s law. Hanneman and Mariano explained the deviation as the result of stronger V-Fe bonds than Fe-Fe or V-V bonds\[135]. Shiga proposed an empirical relation between the magnetic moment and the lattice constant for 3$d$ transition metal alloys\[136]. This relation was applied to Fe-V alloys and an excellent agreement with experiments was obtained\[137]. We also plotted the relationship in the figure and found a good agreement between our experimental data and the proposed relation. According to Shiga, A lattice expansion could happen due to formation of localized magnetic moments. The large negative deviation from
Vegard’s law could originate from the collapse of Fe localized moments by alloying vanadium.

5.3 Static magnetic properties of Fe$_{1-x}$V$_x$ epitaxial thin films

5.3.1 Hysteresis loops of MgO/Fe epitaxial films

Figure 5.9: In-plane hysteresis loops for 50 nm Fe films deposited at different conditions, with magnetic field applied along [100] easy axis. (a) and (b) are for the samples grown at 200°C and 300°C, respectively; (c) and (d) are for the samples deposited with gas purifier on and on cleaned substrate, respectively, both deposited at 200°C.

To test the deposition conditions, hysteresis loops for 50nm Fe films grown at different conditions were measured by VSM. Fig. 5.9 shows the in-plane hysteresis loops for these samples, with the magnetic field applied along the [100] easy axis. Fig. 5.9 (a) and (b) are for samples grown at 200°C and 300°C, respectively; (c) and (d) are for samples deposited with gas purifier on and on cleaned MgO substrate, respectively, both grown at 200°C. Hysteresis loops were also taken along the [010] direction and they are similar to those in Fig. 5.9, showing the clear four-fold symmetry. All loops demonstrate a well defined squareness and
small coercivity, which spans 2.0 to 5.5 Oe. All these results indicate the agreement with the [100] easy axis characters expected in bcc Fe. Coercivity for the sample deposited at 300°C is clearly bigger than that in other samples, which indicates that deposition temperature greatly affects the magnetic properties in these epitaxial thin films.

Hysteresis loops were also measured along the [110] direction, which is the hard axis direction in bcc Fe. Fig. 5.10 shows a representative loop for 50 nm epitaxial Fe films. The curve demonstrates a gradual decrease in magnetization from approximately 500 Oe followed by an abrupt switching at about -10 Oe. This feature has been observed either by MOKE or SQUID before\cite{138,139}. If a coherent rotation process is assumed for the magnetization along the [110] hard axis, the cubic anisotropy $H_k = 2K_1/M_s$ of the films is about 500 Oe, which is about the same as that of the bulk bcc Fe, $H_k = 550$ Oe. Hysteresis loop was also taken along [110] direction, which shows similar features with those in Fig. 5.10, again indicating the four fold magnetic anisotropy in bcc Fe.

![Figure 5.10: The in-plane hysteresis loop for 50 nm Fe film deposited on the cleaned substrate, with magnetic field applied along [110] hard axis.](image)

Magnetic properties are well known to depend sensitively on film structure. The dis-
tinctly anisotropic hysteresis loops in Fig. 5.9 and 5.10 suggest that high quality bcc Fe films with well defined magnetic properties have been stabilized on the MgO(100)-1 \times 1cm^2 substrates.

### 5.3.2 Hysteresis loops and magnetization of Fe\textsubscript{1−x}V\textsubscript{x} epitaxial thin films

![Hysteresis Loops](image)

Figure 5.11: The in-plane hysteresis loops for Fe\textsubscript{1−x}V\textsubscript{x} epitaxial thin films, with magnetic field applied along [100] direction. V concentrations and the voltage changes in the hysteresis loops are shown in the plots.

Representative VSM measured hysteresis loops for Fe\textsubscript{1−x}V\textsubscript{x} epitaxial thin films are shown in Fig. 5.11 as a function of V concentration. The applied field was applied to [100] direction in all the plots. We can see for samples with V concentration up to 31%, the squareness are good and the coercivity is small, which indicate an easy axis characters. For 42% and 52% V, however, the squareness becomes worse defined and the coercivity
Table 5.1: The magnetization of Fe$_{1-x}$V$_x$ epitaxial thin films, extracted from the VSM measured hysteresis loops.

<table>
<thead>
<tr>
<th>$x_V$</th>
<th>0</th>
<th>10</th>
<th>19</th>
<th>23</th>
<th>31</th>
<th>42</th>
<th>52</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4\pi M_s$ (T)</td>
<td>2.20</td>
<td>1.97</td>
<td>1.49</td>
<td>1.35</td>
<td>1.00</td>
<td>0.79</td>
<td>0.39</td>
</tr>
</tbody>
</table>

gets bigger. This is where magnetic anisotropy gets close to zero and the easy axis and hard axis begin exchanging their directions, that is, [100] will become the hard axis and [110] will become the easy axis. Later we will confirm this by the angular-dependent FMR measurements.

The difference between the highest and the lowest voltages in the hysteresis loop is proportional to the saturation magnetic moment in the sample. From the above magnetic property characterization and the XRD results shown in 5.2, we presume that the Fe (50nm) deposited on MgO(100) substrate is of high quality bcc structure and possesses the magnetization of bulk bcc Fe. Thus we assume the magnetization $M_s$ is 1750 emu/cc and the Fe film was set as the calibration material for VSM measured magnetization. As all the Fe$_{1-x}$V$_x$ films were deposited on the same size (1 x 1 cm$^2$) MgO substrates and all the films were 50 nm thick, the volume of the film, $\Omega$, will be constant in all the samples. We have $M_s = \mu_s/\Omega$, where $\mu_s$ is the saturation magnetic moment. Therefore, the voltage difference in the hysteresis loop is directly related to the magnetization in the sample. By timing \( \frac{1750}{3.70 \times 10^{-5}} \), where 3.70 $\times$ 10$^{-5}$ is the voltage difference for the Fe (50nm) film, the voltage difference was translated into the magnetization $M_s$ in all the Fe$_{1-x}$V$_x$ thin films. Table 5.1 lists the extracted $4\pi M_s$ values. At 31\% V, the magnetization is 1 T, which is about the level of permalloy (Ni$_{81}$Fe$_{19}$) and is still far greater than that of ferrites.

The extracted magnetization of Fe$_{1-x}$V$_x$ is plotted as a function of the V concentration, $x_V$, as in Fig. 5.12. The experimental data were fitted into a linear relation, and the fitted line is also plotted. By comparison, the theoretical Slater-Pauling curve is also plotted in the figure, which displays the ground-state ferromagnetic properties of the transition metals and

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their alloys in the form of saturation magnetization per atom as a function of the number of valence electrons\cite{140, 141}. The commonly used mean atomic moment in Bohr magnetons, $\mu_B$/atom, has been transformed into T. The equivalent atomic volume of Fe$_{1-x}$V$_x$ is taken as the linear fractional combination of Fe and V atoms. Note that $1\mu_B = 9.27 \times 10^{-24}$ A.m$^2$ and 1A/m=1$\times$10$^3$ emu/cc.

![Figure 5.12](image.png)

Figure 5.12: The VSM measured magnetization for Fe$_{1-x}$V$_x$ epitaxial thin films as a function of V concentration $x_V$. The solid circles are the experimental data; the solid line is the linear fit to the data; the dashed line is the Slater-Pauling curve.

As seen in Fig. 5.12, as V concentration increases, the magnetization of Fe$_{1-x}$V$_x$ decreases linearly. The linear fit is in good agreement with the Slater-Pauling curve. It is well known from neutron-diffraction studies and electronic structure calculations that V atoms, when dissolved in Fe, acquire a sizable induced magnetic moment: $\sim -1\mu_B$ in the dilute limit, with respect to the Fe moments in the host metal\cite{142, 143}. It was found that the average magnetic moment of Fe-V alloys vanishes at about 70% at V concentration\cite{144, 137}, which agrees with our linear fit shown in Fig. 5.12 within the experimental errors if we extend the line to 70% V. The V magnetic moments are aligned antiparallel to those in the
Fe. It has been confirmed by XMCD measurement in Fe/V multilayers, where the MCD signal is negative on $L_3$ absorption edge, while that for V is positive\cite{15,16}.

Different models have been put forward to explain magnetism in Fe-V alloys. The first model assumes a regular decrease, to zero, of Fe moment as V concentration increases, supported by spin-polarized, self-consistent Korringa-Kohn-Rostoker coherent-potential approximation (KKR-CPA) calculation\cite{133}. In the second model, the Fe moment vanishes when an Fe atom is surrounded by four or more V atoms at any V concentration\cite{137}. The third model proposed a randomly oriented, but finite Fe moment is retained at any V concentration\cite{138,139}. The last model has been confirmed by XMCD in Fe/V multilayers and (001) oriented Fe-V alloys up to 20\% V\cite{145,146}. It was found that the Fe moments are hardly changed from their bulk values. However, the Fe orbital-to-spin moment ratio is reduced due to the hybridization of the Fe and V wave functions where the electrons associated with Fe acquire some of the character (orbital moment) associated with the V electrons. On the other hand, the neutron study of polycrystalline Fe-V alloys showed that Fe moment exhibits a slight maximum near 5\% V and then decreases with V concentration, while the V moment keeps the value of about -1\,\mu_B at the studied concentration range\cite{129}. It was found that the elemental magnetic moment also depends on the lattice distortion\cite{150}, which might explain the discrepancy found in epitaxial and polycrystalline Fe-V alloys.

5.4 FMR study of MgO/Fe$_{1-x}$V$_x$ epitaxial films

5.4.1 FMR linewidth and its thickness dependence in MgO/Fe epitaxial films

FMR linewidth in MgO/Fe

Peak-to-peak linewidth of the FMR resonant signal is a very sensitive measure of the sample’s structural and magnetic quality. It is generally observed that the narrowest FMR peak-
to-peak linewidth exists for samples which have the best structural quality. MgO/Fe(50 nm) epitaxial films deposited at different conditions have been measured by cavity-type FMR spectrometer in the X-band (10.29 GHz). The external dc magnetic field was applied in the film plane parallel to the [110] direction. Fig. 5.13 shows the measured spectra for Fe deposited on as-received substrate with gas purifier off during deposition, on as-received substrate with gas purifier on, and on cleaned substrate with gas purifier off in (a), (b), and (c), respectively. All samples were deposited at 200°C. As described in ??, the measured signal is the derivative of the absorbed microwave energy power with respect to the applied dc field (dP/dH) as a function of the dc field.

![Figure 5.13: 10GHz FMR measurements of MgO/Fe(50 nm) epitaxial films deposited at different conditions: (a) on the as-received substrate with gas purifier off during deposition; (b) on the as-received substrate with gas purifier on; (c) on the cleaned substrate with gas purifier off. The dc field is applied parallel to the in-plane [110] direction. The peak-to-peak linewidth is also shown in each plot.](image)

The FMR signal lineshape in Fig. 5.13 is almost perfectly symmetric and can be expressed as the first derivative of a Lorentz function. The good signal-to-noise ratio is observed in all the three spectra, which allows the precise determination of the peak-to-peak linewidth, $\Delta H$. $\Delta H$ can be determined either by directly reading the fields corresponding
to the maxima and the minima in $dP/dH$ and taking the difference between them or by fitting the data into the derivative of a Lorentz function,

$$\frac{dP}{dH} = -\frac{2A(H - H_0)}{(H - H_0)^2 + B^2},$$

where $H$ is the applied DC field, and $A$, $B$, and $H_0$ are the fitting parameters. $\frac{2B}{\sqrt{3}}$ is equal to the peak-to-peak linewidth $\Delta H$. $H_0$ is the resonance field where the maximum absorption happens in the microwave energy. For symmetric FMR spectra like those shown in Fig. 5.13, both ways should give similar results. Determined $\Delta H$ values are shown in Fig. 5.13, which demonstrate similar results in the three spectra, 26 Oe in (a), 27 Oe in (b) and (c). This similarity in $\Delta H$ indicates that magnetic homogeneity in the epitaxial films is not affected by whether or not the substrate is further cleaned and the gas purifier is turned on during deposition. In another word, the purity of the inlet Ar is high enough and the substrate is clean enough for the samples of FMR research within the accuracy range of the measurements.

The narrowest reported $\Delta H$ in Fe is $\sim 15$ Oe at 10 GHz, which is also the minimum ferromagnetic linewidth for a metal\[72,151\]. The relatively large linewidth in our samples is partly caused by eddy currents (conductivity effects) which give additional broadening in relatively thick films. As later demonstrated, smaller linewidths will be realized in thinner Fe films.

FMR signal is sensitive to the crystallographic directions in the epitaxial structure. Both the resonance field and the linewidth change as the dc field is applied along different crystallographic directions in the epitaxial film. To confirm that FMR of all the MgO/Fe samples has been measured and compared at the exactly same crystallographic direction, namely, the [110] hard axis direction, the in-plane angular dependent FMR measurement has been performed around the nominal [110] direction in the film. Fig. 5.14 shows the typical FMR spectra taken at 10.29 GHz with dc field applied along the nominal [110] direction,
3.6° away, and 7.2° away from the [110] direction. It is clear that as the applied field leaves the [110] hard axis direction, the resonance field shifts to the lower values. According to the Kittel equation, the resonance frequency is proportional to the sum of the magnetic anisotropy field and the resonance field. In bcc Fe, the anisotropy field is the largest along the [100] easy axis direction and smallest along the [110] hard axis direction. Therefore, the resonance field will change in the opposite way, that is, the maximum resonance field happens when the dc field is applied along the [110] hard axis direction.

Figure 5.14: Typical FMR spectra of MgO/Fe(50 nm) epitaxial film with the external dc field applied parallel to the nominal [110] direction, 3.6° away, and 7.2° away from the [110] direction. Note the shift to lower resonance fields with the dc field away from the [110] hard axis direction.

Angular dependent FMR has been measured for all the Fe films studied and it was confirmed that the maximum resonance field corresponds to the nominal [110] crystallographic direction in the samples. The FMR linewidth also depends on the crystallographic directions in the epitaxial films. $\Delta H$ in the [100] easy axis is always larger than that in the [110] hard axis. However, the angular dependence is more complicated than that of the resonance field and will be discussed later.
CHAPTER 5. COMPOSITION-BASED REDUCTION OF DAMPING IN MGO/FE\textsubscript{1−X}V\textsubscript{X} EPITAXIAL THIN FILMS

Thickness dependence of FMR linewidth in Fe

![Graph showing the variation of FMR linewidth with Fe film thickness](image)

Figure 5.15: FMR linewidth of Fe films with different thicknesses, measured at 10 GHz, with the external dc field applied parallel to [110] direction. Smaller linewidth exists in thinner Fe films.

FMR spectra have been taken at 10 GHz for Fe films of different thicknesses, ranging from 8-100 nm, with dc field applied along the [110] hard axis by the waveguide-type FMR spectrometer. Fig. 5.15 shows the variation of the FMR linewidth with respect to the Fe film thickness $t_{Fe}$. As the Fe film thickness increases, the FMR linewidth also increases. This dependence is in agreement with previous experimental result\textsuperscript{38}. In the limit of small thicknesses for an ideal sample (without any magnetic inhomogeneity), the FMR linewidth is given only by the Gilbert damping coefficient. In reality, for ultrathin film of thickness less than a critical value in which the lattice parameter of the Fe film reaches the bulk lattice parameter, there exists a relaxation due to the lattice mismatch at the film-substrate interface. In this range, the FMR linewidth increases with the decreasing film thickness. Our experimental result shows that the linewidth of Fe of 8 nm is about the same or even a little bit larger than that of 15 nm. Therefore, the minimum linewidth for our Fe films might be realized around 15 nm of the thickness. This critical thickness has also been found.
CHAPTER 5. COMPOSITION-BASED REDUCTION OF DAMPING IN MGO/FE\textsubscript{1–x}V\textsubscript{x} EPITAXIAL THIN FILMS

by Fermin et al.\textsuperscript{[152]}. $\Delta H$ for our Fe film of 15 nm thickness is equal to 16.6 Oe at 10 GHz, which is close to the narrowest linewidth ever reported in metals\textsuperscript{[37]}.

Figure 5.16: Frequency dependence of FMR linewidth measured along [110] direction of Fe films of 8 nm and 50 nm thick. Note the slope increase in 50 nm film with respect to the 8 nm film.

In metallic films the magnetic damping can be affected by eddy current. The role of eddy currents in thin films can be estimated by evaluating the effective Gilbert damping accompanying the magnetization precession in the presence of eddy currents. To evaluate how the eddy current affects the Gilbert damping in our Fe film, frequency dependent FMR was carried out for the Fe films with different thicknesses. Fig. 5.16 shows the frequency dependence of $\Delta H$ measured with the magnetic field applied along the [110] direction for films with $t_{Fe} = 8$ nm and 50 nm. This frequency dependence of $\Delta H$ can be used to characterize the contribution of the Gilbert damping mechanism and determine the role of magnetic inhomogeneities. The damping term in Landau-Lifshitz-Gilbert (LLG) equation leads to a linear dependence of the FMR linewidth on the microwave frequency, which is shown in Eq. 2.23. The solid lines represent the linear fits to the experimental data, in which the zero-frequency-intercept represents the inhomogeneous broadening $\Delta H_{\text{inhom}}$ and...
Table 5.2: The fitting parameter values for the frequency dependence of the FMR linewidths in Fe films of 8 nm and 50 nm thick.

<table>
<thead>
<tr>
<th>$t_{Fe}$ (nm)</th>
<th>$\Delta H_{inhom}$ (Oe)</th>
<th>$d(\Delta H)/df$ ($\times 10^{-4}$T/GHz)</th>
<th>$g$</th>
<th>$4\pi M_s$ (T)</th>
<th>$G$ (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>9.00 ± 1.14</td>
<td>1.0155 ± 0.0580</td>
<td>2.099</td>
<td>2.20</td>
<td>0.084 ± 0.005</td>
</tr>
<tr>
<td>50</td>
<td>15.5 ± 0.9</td>
<td>1.5573 ± 0.0546</td>
<td>2.099</td>
<td>2.20</td>
<td>0.128 ± 0.004</td>
</tr>
</tbody>
</table>

The slope $d(\Delta H)/df$ is equal to $1.16 \frac{2\pi G}{\gamma M_s}$.

We have $\omega = 2\pi \times 10$ GHz, $\gamma = 28\pi g = 28\pi \times 2.099$ GHz/T, and $4\pi M_s = 2.2$ T. With the slopes extracted from the fits, $(1.0155 \pm 0.058) \times 10^{-4}$ T/GHz for 8 nm and $(1.5573 \pm 0.0546) \times 10^{-4}$ T/GHz for 50 nm, we calculated Gilbert damping $G$ as $(0.084 \pm 0.005)$ GHz for 8 nm and $(0.128 \pm 0.004)$ GHz for 50 nm Fe films. The fitting and extracted parameters are shown in Table 5.2. The $G$ values in both films drop within the range of reported values of damping parameter obtained from FMR absorption linewidth, that is, from $0.4 \times 10^8$ to $1.3 \times 10^8$ Hz\cite{35}.

For thin films where the rf magnetization fully penetrates the film the contribution of eddy currents to the LLG equations of motion can be expressed by

$$G_{eddy} = \frac{1}{12} \sigma_{Fe} \left( \frac{t_{Fe}}{c} \right)^2 (4\pi M_s \gamma)^2$$  \hspace{1cm} (5.6)

where $\sigma_{Fe} = 9 \times 10^{16}$ s$^{-1}$ is the electrical conductivity and $c = 3 \times 10^8$ m/s is the velocity of light in free space. Thus we calculated $G_{eddy}$ as 0.0008 GHz, which is negligible, for 8 nm and 0.032 GHz for 50 nm Fe. It is apparent that eddy current plays an important role in the 50 nm thick Fe. This eddy current contribution is still smaller than the actual increase of the Gilbert damping observed in out 50 nm film with respect to the 8 nm film, which is 0.044 GHz. The reason for this discrepancy might arise in part from some inaccuracy in the nominal film thickness. More complicated damping mechanisms might also be needed to explain the discrepancy. From the knowledge of the author, no such quantitative discussion exists on the damping contribution from eddy current from experimental point of view.
5.4.2 Angular dependent FMR and the magnetic anisotropy in MgO/Fe$_{1-x}$V$_x$

Figure 5.17: FMR resonance field as a function of in-plane angle for MgO/Fe$_{1-x}$V$_x$ (50nm) epitaxial thin films, measured at 10.29 GHz. Different shapes of symbols represent the experimental data with different V concentrations. The solid lines represent the theoretical fitting with the in-plane resonance condition equations.

In order to characterize the magnetic anisotropy in MgO/Fe$_{1-x}$V$_x$ epitaxial thin films, we measured the FMR spectra as a function of in-plane angle between the applied dc field and the [100] easy axis direction in the samples. Fig. 5.17 shows the resonance field as a function of in-plane angle measured at 10.29 GHz by the cavity-type FMR spectrometer. The different shapes of symbols represent the data with different V concentrations. For the whole series, the angular dependence of the resonance field exhibits a fourfold symmetry, which is expected in bcc Fe, with the minima along [100] and [010] easy axes and the maxima along [110] and [1¯10] hard axes. This suggests that the structural relaxation due to the lattice mismatch between the film and the substrate has finished at this thickness. As V concentration increases, the resonance field also increases, but the magnetocrystalline anisotropy decreases. For the sample with 52% V, the crystallographic directions corre-
sponding to the maxima and minima in the resonance field exchange their directions. That is, [110] becomes the easy axis and [100] becomes the hard axis.

For in-plane configuration where both the magnetization $M_s$ and the applied Field $H$ are parallel to the film plane, the free energy density is

$$U = -MH \cos(\phi - \phi_H) + \frac{1}{8}K_4(1 - \cos 4\phi)$$

(5.7)

in which $\phi$ is the angle between [100] and $M_s$, and $\phi_H$ is the angle between [100] and $H$. $K_4$ is the fourth-order term of the magnetic anisotropy energy (MAE) term. The first term on the right side of Eq. (5.7) is the Zeeman energy, and the second term is the MAE. The second-order term of the MAE, $K_2$ is omitted because it is too small (one order of magnitude smaller) compared with $K_4$ in Fe. Substituting $U$ in Eq. (2.21) with Eq. (5.7) we can easily obtain the in-plane angular dependence of the resonance condition,

$$\left(\frac{\omega}{\gamma}\right)^2 = [H_R \cos(\phi - \phi_H) + 4\pi M_s + \frac{K_4}{2M}(3 + \cos 4\phi)][H_R \cos(\phi - \phi_H) + \frac{2K_4}{M} \cos 4\phi].$$

(5.8)

The equilibrium angle of $M_s$ at resonance is given by the condition of minimum of the free energy density,

$$\frac{\partial U}{\partial \phi} = 0.$$  

(5.9)

Substituting $U$ in Eq. (5.9) with Eq. (5.7) we can obtain the equilibrium condition

$$K_4 \sin 4\phi + 2M_sH \sin(\phi - \phi_H) = 0$$

(5.10)

Combining Eq. (5.8) and (5.10), we can extract both the resonance field $H_R$ and the equilibrium angle of the magnetization $\phi$ giving a known $\phi_H$. $M_s$ values are taken from the VSM measurement result shown in Table 5.1. By numerical calculation by Mathematica with the fitting parameter $K_4$, a simulation was carried out to fit the calculated $H_R$ to
the experimental data in Fig. 5.17. The $K_4$ value which gave the best fit of $H_R$ to the experimental data was determined to be the anisotropy constant of the film. The solid lines in Fig. 5.17 represent the best fits for the different V concentrations.

The extracted $K_4$ values for $\text{Fe}_{1-x}V_x$ thin films are shown in Fig. 5.18 as a function of V concentration. The solid line is for guidance of the eye. The dashed line shows zero in $K_4$ value. The $K_4$ value for MgO/Fe is also marked in the figure. The extracted $K_4$ value in our experiment is in an excellent agreement with the value of bulk Fe ($4.87 \times 10^5$ erg/cc). As V is doped in, $K_4$ value decreases. At about 45% V, $K_4$ becomes zero, which indicates that the film is magnetically "soft" at this concentration. Energies of domain walls, if there are any such domain walls, are small, and changes to the overall magnetization would happen easily. Beyond this concentration, $K_4$ becomes negative values, which means that the previous easy axis, [100] direction, has become the hard axis and the previous hard axis, [110] direction, has turned into the easy axis.

Figure 5.18: Magnetic anisotropy constant $K_4$ of MgO/Fe$_{1-x}$V$_x$ (50nm) extracted from the fitting shown in Fig. 5.17 as a function of V concentration.

In Fe/V multilayer structures, the behavior of in-plane $K_4$ is explained by considering
the competition between two parts, a volume part favoring the [100] direction, similar to bulk Fe, and a surface part favoring the [110]. This competition has been reported to lead to an in-plane reorientation of the easy-magnetization axis from [100] to [110] in Fe films on various substrates with decreasing Fe-thickness.

Comparing with Fe/V multilayers, much less is known about the magnetic anisotropy of Fe-V alloys. A recent electronic structure density-functional calculation based on the spin-polarized relativistic Korringa-Kohn-Rostoker (KKR) method showed that the magnetic anisotropy of Fe can be reduced by reducing the band filling and increasing the volume. The result suggested that binary alloys Fe$_{1-x}$V$_x$ may lead to magnetic softening when band filling $Z_A < Z_{Fe}$ and $a_A > a_{Fe}$, which is well satisfied by doing V into Fe. Our experimental results provided a strong support to this theoretical calculation.

The equilibrium angle between magnetization and [100] direction was also calculated for each V concentration. Fig. 5.19 shows the equilibrium angle of the magnetization $\phi$ as a function of the angle of the applied dc field $\phi_H$ with respect to [100] crystallographic direction for each V concentration. By comparison, the lines for $\phi = \phi_H$ are also plotted, shown as the dashed lines in the figure. It is clear that the magnetization $M_s$ does not follow the direction of the applied field $H$, except that at easy axis and hard axis directions, $\phi = \phi_H$. For $H$ applied along directions other than easy and hard axes, because of the magnetic anisotropy, the equilibrium angle between $M_s$ and the easy axis is always smaller than that between $H$ and the easy axis.

For $x_V < 45\%$, the magnetic anisotropy field is along [100] easy axis direction. As V increases from 0 to 45\%, the magnetic anisotropy decreases, and the difference between $\phi$ and $\phi_H$ gets smaller, indicating the magnetic softening is experienced in the film. For $x_V > 45\%$, $K_4$ becomes negative and [110] becomes the easy axis direction. The magnetic anisotropy, which is along [110] direction, increases again with V concentration, and $M_s$ becomes closer to [110], instead of [100], direction than $H$ is.
Figure 5.19: The equilibrium angle of magnetization $\phi$ in MgO/Fe$_{1-x}$V$_x$ as a function of the angle of the applied field $\phi_H$ with respect to [100] direction. The dashed lines are $\phi = \phi_H$ for comparison.

Magnetic anisotropy is closely related to the damping in the system, both originated from the coupling of spin moment with orbital moment. When an external field tries to reorient the spin of an electron, the orbit of that electron also tends to be reoriented. But the orbit is strongly coupled to the lattice and therefore resists the attempt to rotate the spin axis. The energy required to rotate the spin system of a domain away from the easy direction, which we call the anisotropy energy, is just the energy required to overcome the spin-orbit coupling.

Dynamic effects which are mediated by the spin-orbit coupling are the magnon-phonon scattering in magnetically ordered systems. The approach of Kambersky for magnetic metallic systems assumes two different processes for the relaxation of the net magnetization, namely, the ordinary process and spin-flip process\cite{40}. The Gilbert damping $G$ for both processes are directly proportional to the square of the deviation of the $g$ factor from its
free spin-value, thus proportional to the square of the ratio of orbital to spin moment.

Spin-flip scattering: \( G = (\gamma \hbar / 2)^2 Z_F (g - 2)^2 / \tau \)

Ordinary scattering: \( G = (\gamma / 2)^2 Z_F \lambda_{SO}^2 (g - 2)^2 \tau. \)

\( Z_F \) is the density of states at the Fermi level, \( \tau \) is the electron scattering time, and \( \lambda_{SO} \) is the spin-orbit-coupling constant. Therefore, \( G \) value provides a very sensitive probe for the spin-orbit-coupling effects.

It has been demonstrated for thin films of FeCo alloy system that compositional changes can induce the modification of both spin and orbital moments, which leads to changes of the \( g \) factor and the Gilbert damping \( G^{[33]} \). In addition, the magnetocrystalline anisotropy in this system which is governed by the spin-orbit coupling displays systematic trends which are consistent with the changes of the Gilbert damping \( G \).

### 5.4.3 Frequency dependent FMR and the damping in MgO/Fe\(_{1-x}V_x\) epitaxial films

Frequency dependent FMR was measured on the MgO/Fe\(_{1-x}V_x\) series by the waveguide-type FMR spectrometer. Fig. 5.20 shows the FMR spectra of Fe\(_{81}V_{19}\) film measured at different frequencies. The external magnetic field is applied in the film plane parallel to the [110] direction. From these frequency dependent FMR spectra we extracted both the resonance field and the FMR linewidth as a function of the resonance frequency in Fe\(_{81}V_{19}\). Same process has been done to all the Fe\(_{1-x}V_x\) series for \( 0 \leq x_V \leq 52\% \).

**Frequency dependent resonance field and Kittel plot**

It is obvious from Fig. 5.20 that as the resonance frequency increases, the resonance field also increases. The relation between the resonance field and the resonance frequency can be described by the Kittel equation. With the external field applied parallel to the film plane,
the Kittel equation can be expressed as

$$ f^2 = \left(\frac{\gamma \mu_0}{2\pi}\right)^2 H(H + M_s) = 144g^2 \mu_0(H_r - H_k)\mu_0(H_r - H_k + M_s), \quad (5.11) $$

where $g$ is the spectroscopic factor, $H_r$ is the resonance field, and $H_k$ is the magnetic anisotropy field. Fig. 5.21 shows the experimental $f^2$ as a function of the resonance field $H_r$. We fit the data into the Kittel equation and extracted the anisotropy field $H_k$ and the $g$ factor. $M_s$ was determined from the VSM measurements. The extracted parameter values are listed in Table 5.3. $H_k$ in positive value means the easy axis is along [100] while that in negative value means the easy axis is along [110]. In bcc Fe, the magnetic anisotropy field can be expressed by $H_k = \frac{2K_4}{M_s}$. Thus $K_4$ values can be calculated from $H_k$ and they are also listed in Table 5.3. Within the experimental error, the extracted $H_k$ values is in good agreement with the $K_4$ values extracted from the in-plane angular dependent FMR measurements described in 5.4.2.

The determined $g$ factor for Fe corresponds to the value of bulk bcc Fe (2.099) very
well. However, the difference in the determined $g$ factors for different V concentrations is very small. In fact, the $g$ factors overlap with each other considering the experimental errors. Therefore, a precise determination of the dependence of $g$ factor on V concentration is difficult to obtain. Note that high frequency FMR beyond 18 GHz, which is measured by k-band waveguide type FMR spectrometer, is not available to all the samples. The reason for the bad quality in the FMR spectra at high frequencies is still under investigation. For very small frequencies, say, 1 or 2 GHz, there might be some problem related to the small fields used in the measurements, that is, domain effects may become important issues.

A systematic investigation of the expected $g$-shift due to changes in the orbital moment of thin films has not been reported in the literature. The $g$ factors reported in the literature were usually obtained as a by-product of a fitting procedure to extract the anisotropy parameters from FMR[26]. It turned out that an unambiguous determination of $g$ was hard
Table 5.3: The anisotropy field \( H_k \) and the \( g \) factor of Fe\(_{1-x}V_x\) extracted from the Kittel plot fitting.

<table>
<thead>
<tr>
<th>( x_V ) (%)</th>
<th>0</th>
<th>10</th>
<th>19</th>
<th>31</th>
<th>42</th>
<th>52</th>
</tr>
</thead>
<tbody>
<tr>
<td>( H_k ) (Oe)</td>
<td>584 ± 42</td>
<td>413 ± 28</td>
<td>347 ± 21</td>
<td>239 ± 23</td>
<td>−10.7 ± 19.5</td>
<td>−192 ± 43</td>
</tr>
<tr>
<td>( K_4 ) (erg/cc)</td>
<td>5.11 ± 0.37</td>
<td>3.04 ± 0.21</td>
<td>2.13 ± 0.13</td>
<td>1.06 ± 0.06</td>
<td>−0.031 ± 0.056</td>
<td>−0.28 ± 0.10</td>
</tr>
<tr>
<td>( g )</td>
<td>2.109 ± 0.015</td>
<td>2.110 ± 0.012</td>
<td>2.139 ± 0.021</td>
<td>2.094 ± 0.007</td>
<td>2.100 ± 0.011</td>
<td>2.100 ± 0.023</td>
</tr>
</tbody>
</table>

to be achieved.

**Extrinsic and intrinsic damping in Fe\(_{1-x}V_x\)**

The frequency dependent peak-to-peak linewidth was also determined from the FMR spectra obtained at different frequencies. Fig. 5.22 shows the FMR linewidth as a function of resonance frequency in Fe\(_{1-x}V_x\) series for \( 0 \leq x_V \leq 52 \). The external field was applied along [110] direction. The solid lines show the linear fits to the experimental data according to Eq. 2.23. The data points shown in the figure represent those taken from well defined and symmetric FMR spectra. As V concentration increases, the magnetization decreases, so does the signal recorded by the lock-in amplifier, which makes good FMR spectra more difficult to be obtained.

**Inhomogeneous broadening and the extrinsic damping**

The inhomogeneous FMR spectral broadening \( \Delta H_0 \) was extracted from the zero-frequency intercept of the linear fit. \( \Delta H_0 \) arises from the presence of magnetic inhomogeneities and is usually regarded as independent of frequency. The values of the extracted \( \Delta H_0 \) are shown in Fig. 5.23 as a function of the V concentration. The dashed line is for the guidance of the eye. \( \Delta H_0 \) for the Fe film (50 nm) is 15 Oe, which is about twice of that of Fe (8 nm), which means that Fe of 8 nm is structurally and magnetically more homogeneous. However, we
Figure 5.22: FMR linewidth as a function of the resonance frequency in Fe$_{1-x}$V$_x$ thin films. The symbols represent the experimental data and the solid lines represent the linear fits to the data. V concentration is marked in each plot.

could not obtain the FMR spectra of 8 nm series for $x_V > 20\%$ because of the too small magnetic moments of the films. For the 50 nm series shown in Fig. 5.23, we can see that as the V concentration increases, $\Delta H_0$ also increases, from 15 Oe in Fe to 75 Oe in Fe$_{48}$V$_{52}$.

The increase in the inhomogeneous linewidth indicates that the magnetic homogeneity gets worse as V is doped in Fe. According to the XRD results (5.2.2), the broadening of the rocking curve measured around the (200) peak increases with V concentration, which means that the epitaxial structure is worsen when V is doped into Fe. This structural
CHAPTER 5. COMPOSITION-BASED REDUCTION OF DAMPING IN MGO/Fe$_{1-x}$V$_x$ EPITAXIAL THIN FILMS

The inhomogeneous FMR broadening as a function of the V concentration in Fe$_{1-x}$V$_x$ thin films. The dashed line is for the guidance of the eye.

Inhomogeneity will result in the inhomogeneous broadening in the FMR spectra. The results shown in Fig. 5.23 correspond well to the XRD characterized results.

There are various mechanisms for extrinsic damping. For variations in magnetic properties which are far enough from each other, which cause the characteristic inhomogeneity field larger than interaction field, the FMR linewidth is given by superposition of the local resonances\cite{155}. The local inhomogeneous fields simply add to the applied field and one can get indeed a genuine zero frequency offset $\Delta H_0$. In our FMR measurements, however, at very low frequencies, the FMR linewidth is still a finite value instead of approaching zero. Therefore, this type of relaxation does not happen in our samples.

Large samples (mm scale) comparing with the $\mu$m scale coplanar waveguide can also cause the linewidth broadening, because the samples are exposed to an inhomogeneous rf field and that can result in the generation of magnetostatic modes separated by dipolar rf fields resulting in a broadening of the FMR peak\cite{156}. This contribution is particularly important at low GHz range of frequencies.

Inhomogeneous magnetic properties can result in scattering of magnons. The uniform
mode can get scattered to nonuniform modes. This scattering process is usually referred to as two magnon scattering. The validity of two magnon scattering contribution can be tested by using FMR measurements with the magnetic moment in perpendicular configuration where $\Delta H$ as a function of microwave frequency should be described by Gilbert damping\cite{38}. Unfortunately, the resonance field is usually very high and the fields generated by the magnets in our FMR spectrometer cannot reach such high values. Therefore, two magnon scattering could not be confirmed as the damping mechanism in our experiments.

**Frequency dependent linewidth broadening and the intrinsic damping**

![Graph showing Gilbert damping $G$ as a function of V concentration in Fe$_{1-x}$V$_x$ thin films. The dashed line is to guide the reader's eye.](image)

Figure 5.24: The Gilbert damping $G$ as a function of V concentration in Fe$_{1-x}$V$_x$ thin films. The dashed line is to guide the reader’s eye.

The phenomenological values of the Gilbert damping $G$ were extracted from the slope $\frac{d(\Delta H)}{df}$ of the linear fits in Fig. 5.22. $G$ was calculated from $\frac{d(\Delta H)}{df} = 1.16 \frac{2\pi G}{\gamma^2 M_s}$, $\gamma = \frac{2\mu_B}{h} \approx 28\pi g$. $g$ factor takes the value of Fe, 2.099, and $M_s$ values were taken from the VSM measurements. Fig. 5.24 shows the extracted Gilbert damping $G$ as a function of V concentration. As V concentration increases, the damping $G$ decreases. $G$ value is as low as $0.17 \times 10^8$ s$^{-1}$

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for $x_V = 52\%$, which is only 14% of the $G$ value of the 50 nm Fe film ($1.2 \times 10^8$ s$^{-1}$) and 34% of the lowest $G$ value ever reported on ferromagnetic metals ($0.5 \times 10^8$ s$^{-1}$).

As we mentioned previously, the Gilbert damping is closely related to the magnetocrystalline anisotropy in metallic ferromagnets, both originated from the coupling of spin moments with orbital moments. Comparing with the experimental results shown in 5.4.2, we found that the $V$-concentration dependent Gilbert damping $G$ displays systematic trend which is consistent with that of the magnetocrystalline anisotropy.

An analytical relation between the Gilbert damping $G$ and the $g$ factor could not be established because of the too small differences between $g$ values of different $V$ concentrations. In fact, a considerable deviation of theoretical $g$ factor from the experimental data has been found also in Fe-Co system$^{[33,157]}$. The origin of the deviation was believed to be related to the band structure effects, but the details still remain unexplained. frequency $dP/df$.

![Figure 5.25](image_url)

**Figure 5.25:** The Gilbert damping $G$ as a function of $V$ concentration in Fe$_{1-x}$V$_x$ thin films. The dashed line is to guide the reader’s eye.

To further confirm the observed decrease in the Gilbert damping $G$ as $V$ is doped into
Fe, a field dependent FMR was carried out on the epitaxial films. The FMR spectral was obtained by fixing the external dc field and sweeping the microwave frequency. Fig. 5.25 shows the typical FMR spectra for Fe film obtained at three different fields, which were applied along the [110] crystallographic direction. The voltage signal collected by the lock-in amplifier is proportional to the first derivative of the absorbed microwave energy with respect to the sweeping.

From Fig. 5.25 we can see that as the external dc field increases, the resonance frequency increases, which is equivalent to the trend displayed in Fig. 5.20 and can be explained by the Kittel relation. The peak-to-peak frequency linewidth $\Delta f$ was taken from the spectra. Fig. 5.20 shows $\Delta f$ as a function of the external dc field for both Fe and Fe$_{69}$V$_{31}$ epitaxial films. The symbols represent the experimental data and the dashed lines represent the approximate $\Delta f$ levels in the samples.

![Figure 5.26: The FMR frequency linewidth as a function of external dc field in Fe and Fe$_{69}$V$_{31}$ thin films. The dashed lines represent the approximate $\Delta f$ levels in the Fe and Fe$_{69}$V$_{31}$ films.](image)

For smaller external fields, $\Delta f$ displays a relatively large value. This is probably related to the domain issues. The magnetization in the films might experience the incoherent precession, which results in a linewidth broadening. For external field larger than 1 kOe,
the frequency linewidth becomes almost constant in both films. The approximate $\Delta f$ level for Fe is 270 MHz and that for $x_V = 31\%$ is 184 MHz. It was found that the frequency linewidth is closely related to the Gilbert damping in ferromagnets\cite{158}. The higher the Gilbert damping, the broader the frequency linewidth. The quantitative relation between them was not established. By comparison with the trend in the V-concentration dependent damping $G$ shown in Fig. 5.24, we found that the decrease in the $\Delta f$ corresponds to the decrease in the damping $G$ when V is added.

We noted that eddy current might still plays an important role in the intrinsic damping. As discussed in 5.4.1, the damping constant contributed by eddy current $G_{eddy}$ is about 0.032 GHz for 50 nm thick Fe film, which is about 25% of the phenomenological damping $G$ extracted from the frequency dependent FMR measurements. As V is doped into Fe, the conductivity as well as the magnetization decrease, which makes the damping contribution from the eddy current decrease. Fig. 5.27 shows the resistivity of the 50 nm thick Fe$_{1-x}$V$_x$ films decided from four-point probe measurements. The resistivity $\rho$ was calculated as

$$\rho = R_{sheet} \cdot t_{film} = R_{(V/I)} \cdot \frac{\pi}{\ln 2} \cdot t_{film}. \quad (5.12)$$

$R_{(V/I)}$ is the reading from the measurements and $t_{film}$ is the film thickness.

For 31\% V, the resistivity is 73.2 $\mu$Ω·cm and the conductivity is $\sigma = \frac{9 \times 10^{17}}{\rho} = 1.23 \times 10^{16}$ s$^{-1}$. According to Eq. 5.6, we calculated the damping contributed by eddy current as $G_{eddy} = 1.2$ MHz, which is about 7\% of the phenomenological damping $G$. At higher V concentrations, the eddy current contribution will be negligible.

However, we should exclude eddy current as the main mechanism for the decrease in the damping $G$, because even if we subtract the eddy current contribution $G_{eddy}$ from the phenomenological $G$ value, the trend with V concentration still remains same. Previous research on high quality crystalline samples showed that the intrinsic damping in metals is caused by the itinerant nature of the electrons and the spin-orbit interaction\cite{38}. XMCD
CHAPTER 5. COMPOSITION-BASED REDUCTION OF DAMPING IN MGO/FE$_{1-x}$V$_x$ EPITAXIAL THIN FILMS

![Graph showing resistivity vs. V concentration]

Figure 5.27: The resistivity obtained from the four-point probe measurements as a function of V concentration in Fe$_{1-x}$V$_x$ thin films. The symbols represent the experimental data and the line represents the linear fit.

study of Fe/V multilayers also revealed that while the sum of the Fe spin and orbital moments remain same as that of the bulk Fe, the orbital-to-spin moment ratio is reduced, which indicates the hybridization of the Fe and V wave functions and the electrons associated with the Fe acquires some of the character (orbital moment) associated with the V electrons[135].

5.5 Conclusion

In this chapter, a materials-based strategy was developed to tailor the magnetization dynamics in ferromagnets. By alloying V into Fe, the Gilbert damping $G$ was greatly reduced from that of pure Fe, which in the undoped case possesses the lowest damping parameter of all the metal ferromagnets.

V dopant was added by cosputtering with V and Fe targets onto MgO (100) single crystal substrate which was heated to 200°C, and the resultant film structure is MgO(100)/Fe$_{1-x}$V$_x$(100) epitaxial structure, with the crystallographic arrangement MgO(100)[100]/Fe$_{1-x}$V$_x$(100)[110]. The high quality of the epitaxy was confirmed by the small rocking curve broadening of XRD
characterization. The lattice constant slightly increases and the width of the rocking curve slightly broadens as V is doped in, which is well explained by the small mismatch between bcc Fe and V lattices.

As V concentration increases, the magnetization of the film decreases. The changing behavior corresponds to the Slater-Pauling curve very well. The reduced magnetization is due to the antiparallel alignment of V moments with Fe moments. For 31% V, the magnetization reduces to \( \sim 1 \) T, which is about the same level as permalloy.

FMR results demonstrated that for 15nm Fe film, the linewidth is 16.6 Oe, which is close to the lowest reported linewidth for Fe. As the film gets thick, eddy current plays an important role in the damping. For 50 nm Fe, the damping contribution from the eddy current is responsible for most of the increase of the Gilbert damping \( G \) comparing with the 8 nm Fe.

In-plane angular dependent FMR clearly shows the fourfold symmetry in the resonance field. As V is doped in, the magnetocrystalline anisotropy decreases. At \( \sim 45\% \) V, the anisotropy constant \( K_4 \) reaches zero. Below 45\%, the magnetic easy axis is along [100] and the hard axis is along [110]; beyond 45\%, the easy axis and the hard axis exchange their crystallographic directions.

From frequency dependent FMR measurements both resonance field and the linewidth were studied. Both magnetic anisotropy and \( g \) factor were extracted from the Kittel plot. The extracted magnetic anisotropy coincides well with that extracted from the angular dependent FMR measurements. The relation between \( g \) factor and V concentration could not be established because of the too small separation between the values.

Inhomogeneous and homogeneous linewidths were separated from the frequency dependent FMR measurements. As the V concentration increases, the inhomogeneous linewidth broadens, which is in agreement with the broadening in the width of the XRD rocking curve, indicating the extrinsic damping contributed by the crystalline defects.
Gilbert damping $G$ was extracted from the linear dependence of the homogeneous linewidth with frequency. As V concentration increases, the $G$ value decreases from 0.12 GHz for Fe to 0.017 GHz for 52% V, which is about 34% of the lowest Gilbert damping ever reported for the metal ferromagnets. The decreased damping is consistent with the decreased magnetic anisotropy in the films. The reduced damping was further testified by the frequency linewidth of the field dependent FMR measurements. Damping contribution from eddy current was studied for the Fe$_{1-x}$V$_x$ films and was excluded as the main mechanism for the concentration dependent reduced damping. The damping mechanism was regarded to be the spin-orbit interaction and the hybridization of the Fe and V wave functions.
Appendix A

Code for PIMM Simulation
#pragma rtGlobals=1  // Use modern global access method.

Function ExpDec(w,dt,ic)
//Just about the simplest implementation of a finite difference algorithm
Wave w
Variable dt,ic
variable i
i=1
w[i]=ic
do
w[i]=w[i-1]-0.05*w[i-1]*dt
i+=1
while (i<101)
end

Function Heaviside(t,tp)
variable t,tp
if (t<tp)
return 0
endif
if (t>tp)
return 1
endif
end

Function dudp(t,x,y,l,Ms,hb, hp,hk,tp,dtp)
variable t,x,y,l,Ms,hb, hp,hk,tp,dtp
variable muo
muo=1.25664e-6
return -muo*Ms*hpc*(Heaviside(t,tp)-Heaviside(t,tp+dtp))*cos(x)*cos(y)+muo*Ms*hp*sin(x)*cos(y)+muo*Ms*hk*sin(x)*cos(x)
end

Function dudy(t,x,y,l,Ms,hb, hp,hk,tp,dtp)
variable t,x,y,l,Ms,hb, hp,hk,tp,dtp
variable muo
muo=1.25664e-6
return muo*Ms*sin(y)*(hp*(Heaviside(t,tp)-Heaviside(t,tp+dtp)))*sin(x)+hb*cos(x)+8*3.14159*Ms*cos(y)
end

Function LLG_f(t,x,y,l,Ms,hb, hp,hk,tp,dtp,g)
variable t,x,y,l,Ms,hb, hp,hk,tp,dtp,g
variable muo
variable gam
muo=1.25664e-6
APPENDIX A. CODE FOR PIMM SIMULATION

\texttt{\textbf{function} \texttt{fft\_convert(a1,a2)}}
\texttt{wave a1,a2}
\texttt{variable b}
\texttt{duplicate/O a1 a2}
\texttt{fft a2}
\texttt{a2=abs(a2)}
\texttt{return b}
\texttt{end}

\texttt{\textbf{function} \texttt{LLG\_g(t,x,y,1,Ms,hp,hb,hk,tp,dtp,g)}}
\texttt{variable t,x,y,1,Ms,hp,hb,hk,tp,dtp,g}
\texttt{variable muo}
\texttt{variable gam}
\texttt{muo=1.25664e-6}
\texttt{gam=1.7593e11}
\texttt{return(-gam*g/Ms*cos(y)*2*dupd(t,x,y,1,Ms,hp,hb,hk,tp,dtp))}
\texttt{end}

\texttt{\textbf{function} \texttt{LLG\_Integrator(dMqdt,Mq,nu,1,Ms,hp,hb,hk,tp,dtp,g,phi0,timebas)}}
\texttt{//Integrates LLG given inputs}
\texttt{Wave Mq}
\texttt{Wave dMqdt}
\texttt{Variable nu,1,Ms,hp,hb,hk,tp,dtp,g,phi0,timebas}
\texttt{variable a}
\texttt{variable xn1,xn,tn,dt,yn1,yn,kn1,kn2,kn3,kn4,ln1,ln2,ln3,ln4,i,h}
\texttt{a=1}
\texttt{//Make O/N=1000 Mq}
\texttt{//SetScale/P x,0,timebas,"s",Mq}
\texttt{h=timebas}
\texttt{i=0}
\texttt{Mq[0]=phi0}
\texttt{dMqdt[0]=0}
\texttt{xn=phi0}
\texttt{yn=0}
\texttt{tn=0}
do

kn1=LLG_f(tn, xn, yn, 1, Ms, hp, hb, hk, tp, dtp, g)
ln1=LLG_g(tn, xn, yn, 1, Ms, hp, hb, hk, tp, dtp, g)

kn2=LLG_f(tn+h/2, xn+h/2*kn1, yn+h/2*ln1, 1, Ms, hp, hb, hk, tp, dtp, g)
ln2=LLG_g(tn+h/2, xn+h/2*kn1, yn+h/2*ln1, 1, Ms, hp, hb, hk, tp, dtp, g)

kn3=LLG_f(tn+h/2, xn+h/2*kn2, yn+h/2*ln2, 1, Ms, hp, hb, hk, tp, dtp, g)
ln3=LLG_g(tn+h/2, xn+h/2*kn2, yn+h/2*ln2, 1, Ms, hp, hb, hk, tp, dtp, g)

kn4=LLG_f(tn+h, xn+h*kn3, yn+h*ln3, 1, Ms, hp, hb, hk, tp, dtp, g)
ln4=LLG_g(tn+h, xn+h*kn3, yn+h*ln3, 1, Ms, hp, hb, hk, tp, dtp, g)

xn1= xn+h/6*(kn1+2*kn2+2*kn3+kn4)
yn1= yn+h/6*(ln1+2*ln2+2*ln3+ln4)

Mq[i]=xn1
dMqdd[i]=nu*h/6*(kn1+2*kn2+2*kn3+kn4)
tn=tn+timebas

xn=xn1
yn=yn1

i=i+1
while(i<1001)
Mq*=360/2/3.14159
return a
end

Proc AngleStyle() : GraphStyle
    PauseUpdate; Silent 1    // modifying window...
    ModifyGraph/Z gfsSize=18
    ModifyGraph/Z mode[0]=3
    ModifyGraph/Z marker[0]=8
    ModifyGraph/Z ISize=2
    ModifyGraph/Z rgb[1]=(16384, 28160, 65280)
    ModifyGraph/Z msize[0]=4
    ModifyGraph/Z mthk[0]=2
    ModifyGraph/Z grid=1
    Label/Z left "\symbol\text{"\text\text\text\deg}"
    Label/Z bottom "Time (ns)"
    SetAxis/Z left 0, 180
    SetAxis/Z bottom 0, 6e-09
EndMacro
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