ANISOTROPIES AND SPIN DYNAMICS IN ULTRATHIN MAGNETIC MULTILAYER STRUCTURES.

by

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Abstract

High quality magnetic films were prepared by Molecular Beam Epitaxy (MBE) using Thermal Deposition (TD) and Pulse Laser Deposition (PLD) techniques. Ferromagnetic Resonance (FMR) and Mössbauer studies have shown that the Fe films prepared by PLD exhibited a more intermixed interface lattice structure than those prepared by TD. Dramatic decrease of the in-plane interface uniaxial anisotropy for the PLD films compared to those prepared by TD has shown that the in-plane uniaxial anisotropy is caused by magnetoelasticity driven by the Fe/GaAs(OOI) interface lattice shear.

Magnetization dynamics of the ultrathin Fe/Au,Ag/Fe films was studied using Time-Resolved Magneto-Optical Kerr Effect (TRMOKE) and FMR in the frequency range from 1 to 73 GHz. The Gilbert damping was studied in the Au/Fe/GaAs(001) structures as a function of the Fe and Au layer thickness, respectively. The observed increase in magnetic damping in the Fe film covered with thick Au capping layers was explained by spin pumping at the Fe/Au interface accompanied by spin relaxation and diffusion of the accumulated spin density in the Au layer. The spin diffusion length in Au was found to be 34 nm at room temperature. Significant increase of the Gilbert damping was observed in the Au/Fe/GaAs structures with decreasing Fe film thickness. Its origin lies in the additional damping at the Fe/GaAs interface.

Direct detection of the spin current propagating across the Ag spacer in Fe/Ag,Au/Fe/GaAs(001) structures was carried out with stroboscopic TRMOKE measurements. The Fe layer grown on GaAs served as a spin pumping source and the Fe layer grown on the Au,Ag spacer was used as a probe for detection of the spin current propagating across the Au and Ag spacers. The experimental results were interpreted using selfconsistent solution of the Landau Lifshitz Gilbert (LLG) equations of motion with the spin diffusion equation for the accumulated spin density in the Au and Ag spacers. The spin diffusion length in Ag was found to be 150 nm.
To my wife Monika and my sons, Kacper and Victor.
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to my fellow graduate colleagues and friends. Many thanks to Dr. Radek Urban and Dr. Georg Woltersdorf who taught me to master MBE techniques and thin films growth. Especially, I wish to thank to my colleague and friend Dr. Oleksandr (Sasha) Mosendz for the great company and time we spent together at University and in Vancouver. I always valued discussions about our experiments with him and enjoyed spending the long hours in the lab performing experiments and preparing samples.

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Chapter 1

Introduction

In nanomagnetism ultrathin (few atomic layers) films are incorporated into multilayer structures. In the eighties it was found that their magnetic and transport properties are qualitatively and quantitatively different from those in the bulk magnetic materials. The success in this field came with the development of Molecular Beam Epitaxy (MBE) techniques. Ultra high vacuum removed the problems related to chemical contamination and *in situ* structural and chemical techniques allowed one to perform reliable experiments with multilayer structures with well defined interfaces and controlled magnetic properties. Many researchers have recognized that these modified magnetic properties are attractive to applications. It was already in the 1990’s that the new magnetic sensors and memories had been created and became attractive to the magnetic high density memory media and magnetic RAM (MRAM). The discovery of several new magneto-electronic effects such as Giant Magneto Resistance (GMR) \[1, 2\], Tunneling Magneto Resistance (TMR) \[3, 4\] and spin-injection/filtering \[5, 6, 7\] set in motion the research in electronics coined by spintronics where the spin of the electron rather than its charge is exploited and manipulated.

The physics and applications in spintronics have been increasingly using ultrathin structures where the interface properties play a major role. The research emphases in the recent years have been on the fast magnetization switching and rf magnetization dynamics. The work in this thesis was directed towards understanding the underlying physical processes governing the interface magnetic properties and spin dynamics.

Fe grown on GaAs(001) template (Fe/GaAs(001)) was used as the basic system in this thesis. Fe/GaAs(001) system allows one to grow high quality crystalline structures
allowing to investigate their magnetic properties without being affected by spurious
effects caused by structural defects. Fe has a good lattice match (1.4%) to the GaAs(001)
template which fulfils the main requirement for a good epitaxial and crystalline growth.
In addition the metallic Fe structures represent a robust ferromagnet (F) with the Curie
temperature way above the room temperature (RT) even for the films which are as thin
as 4 atomic layers. The structural defects can lead to interesting properties but again
in this case one has to be able to create them in a controlled way which was achieved in
this thesis by using the lattice mismatch between the Fe(001) and Pd(001) templates.

During the course of this thesis several important steps and variety of techniques
were employed. The single-crystalline magnetic heterostructures were grown using two
alternative MBE techniques: Thermal deposition (TD) and Pulsed Laser deposition
(PLD). The surface morphologies were characterized using vacuum compatible and
monolayer-sensitive analysis techniques: Reflection High Energy diffraction (RHEED),
Scanning Tunneling Microscopy (STM), X-ray Photoelectron Spectroscopy (XPS), and
Auger spectroscopy. The structural and chemical properties of GaAs/Fe(001) interfaces
were investigated by Mössbauer conversion electron spectroscopy (CEMS). The magnetic
anisotropies and relaxation processes of ultrathin Fe films were studied by two comple­
mentary magneto-dynamic techniques: (i) Angular-resolved Ferromagnetic Resonance
(FMR) in a wide range of frequencies from 9-73 GHz where the rf precession of the
magnetic moment was excited by a continuous microwave magnetic field. The resonance
field $H_{FMR}$ and linewidth $\Delta H$ (related to magnetic damping) were investigated as a
function of the microwave frequency and the angle between the magnetization and the
crystallographic axes. (ii) Sophisticated time (ps) and lateral ($\mu m$) resolved Magneto
Optical Kerr Effect (TRMOKE) system where the time evolution of the magnetization
was measured in response to ultrashort magnetic field pulses.

The thesis is organized as follows: Chapter 2 provides a theoretical description of
physical concepts employed during the tenure of this thesis. Section 2.1 describes the
magnetic free energy of thin films including Magnetic Anisotropy Energy (MAE). Sec­
tion 2.2 introduces Landau-Lifshitz-Gilbert (LLG) equation of motion. Ferromagnetic
Resonance (FMR) equations for the in-plane and out-of-plane configurations are pro­
vided in section 2.3. Section 2.4 gives an overview of several mechanisms responsible for
the magnetic relaxation in thin Fe films. They can either be intrinsic or extrinsic. The
intrinsic ones arise either from the bulk of the crystal (bulk Gilbert damping) or from
the ferromagnet/normal metal interface (spin pump effect). The two-magnon scattering described in the section 2.4.3 is an example of extrinsic contribution to the damping. The last section of Chapter 2 gives a theoretical description of the parameters used in Mössbauer spectroscopy.

Chapter 3 is divided into three major sections. Section 3.1 describes the Ultra High Vacuum (UHV) system and tools used in surface analysis. Section 3.2 gives a detailed overview of the measurement techniques used during the course of this thesis.

Chapter 4 is devoted to the detailed studies of ultrathin films growth and their structural analysis. Section 4.1 describes the GaAs substrate preparation. Two different deposition techniques were employed in the Fe growth during sample preparation: Thermal deposition (TD) and Pulsed Laser Deposition (PLD). They are described in section 4.2.

Chapter 5 consist of two parts. Section 5.1 provides the list of the single layer Fe/GaAs(001) structures prepared by TD and PLD which were employed in the Mössbauer measurements and FMR studies. The structural differences between the Fe/GaAs(001) interfaces in the TD and PLD samples were identified by CEMS using $^{57}$Fe as a probe layer at the Fe/GaAs(001) interface. Sections 5.2 and 5.2.2 cross correlate the Fe/GaAs(001) interface structure in the TD and LPD samples with the corresponding interface magnetic anisotropies obtained from FMR measurements. Magnetic anisotropies for Fe grown on Au and Ag templates are listed in section 5.2.3.

Chapter 6 addresses the dynamic properties (magnetic damping) of Fe single and double layer structures. It will be shown that the magnetic damping in ultrathin films can be caused by various factors. Some of them are intrinsic and cannot be avoided while others are extrinsic and are caused by structural defects. It will be shown in section 6.1 that the intrinsic damping in the ultrathin Fe/GaAs(001) films can be described by the Gilbert damping mechanism which consists of both the bulk and interface terms. The extrinsic contribution to the damping will be illustrated in section 6.2 using the Fe/Au/Pd/Fe/GaAs(001) structures. A large lattice mismatch between the Pd to Fe lattice meshes resulted in a self assembled network of misfit dislocations. This leads to a strong two-magnon scattering which resulted in an effective magnetic damping that can exceed at low microwave frequencies ($\sim 10$ GHz) the intrinsic damping by more than one order of magnitude. This opens new possibilities for a selective design of magnetic damping in multilayer structures. It will be shown that the bulk damping is caused by
noise in spin orbit interaction and the interface damping is caused by spin pumping/sink effects at the Fe/normal metal interface.

Chapter 7 presents the studies of non-local spin transport in NM/Fe/GaAs(001) and Fe/NM/Fe/GaAs(001) structures, where NM stands for the Au and Ag layers, respectively. Spin diffusion theory including the spin pumping/sink effects was self-consistently formulated in section 7.2 allowing one to determine the spin diffusion length in Au and Ag. The Au(001) and Ag(001) lattice meshes are well matched to the Fe(001) mesh (> 0.6%) with the crystallographic axes [110]_Fe || [100]_Au (i.e. rotated 45°). This allows one to grow high quality crystalline samples. In magnetic single Fe/NM and double Fe/NM/Fe layer structures the spin current generated by spin pumping at the F/NM interface leads to the accumulated spin density \( m_{NM} \) inside the NM spacer layer. The accumulated spin density in NM is transported by itinerant electrons. The motion of itinerant electrons in NM is affected by momentum scattering (leading to resistance) and spin flip scattering (leading to loss of spin momentum). The loss of spin momentum at the F/NM interface is compensated by an additional Gilbert-like damping contribution which arises from the non-local spin transport.

In section 7.3 FMR measurements of Gilbert damping in single nAu/16Fe/GaAs(001) layer structures as a function of the Au thickness allowed to obtain the spin diffusion length of Au. Finally in section 7.4 it will be shown that in magnetic Fe1/NM/Fe2 double layer structures one can directly measure the propagation of pure spin currents across NM using TRMOKE stroboscopic method. The Fe1 layer provided spin pumping and the layer Fe2 served as a probe for the detection of spin currents propagating across NM.

The summary of important results and conclusions can be found in Chapter 8.

Additional informations are presented in appendices. A detailed description of the UHV system is provided in Appendix A. Appendix B provides a "quick reference" for the crystallographic orientation of standard GaAs(001) wafers.
Chapter 2

Theoretical background

2.1 Magnetic free energy

In the phenomenological description used in experimental work the crystallographic direction of the magnetization is determined by the minimum of the free-energy density $F$, which in an external magnetic field can be expressed as the sum [8]:

$$F_{\text{tot}} = F_{\text{ex}} + F_{\text{Zee}} + F_{\text{an}} + F_{\text{magel}} + F_{\text{d}}$$  \hspace{1cm} (2.1)$$

with $F_{\text{ex}}$ being the energy of the exchange interaction, $F_{\text{Zee}}$ the energy of the magnetized sample in the externally applied magnetic field, $F_{\text{an}}$ the crystallographic magnetic anisotropy energy, $F_{\text{magel}}$ the energy due to the magneto-elastic interaction, and $F_{\text{d}}$ the energy of the demagnetizing field of the sample due to the dipole-dipole interaction. The phenomenological expression for $F$ is usually based on symmetry considerations. In the following I will describe the cubic and tetragonal symmetries as these are widely found in thin films systems and are relevant to this thesis. Also, only the ultrathin limit for $F$ will be considered. This assumes that there is no variation of the magnetization across the film thickness. The particular contributions to the total magnetic free energy term $F$, will be discussed in separate sections.

2.1.1 Exchange interaction

In ferromagnetic materials, the exchange energy favors parallel alignment of the magnetic moments (spins). The ultrathin limit is well satisfied for films which are thinner than the
the exchange length $\sigma_{ex}$. From a simple magnetostatic argument $\sigma_{ex}$ is given by [9, 10]:

$$\sigma_{ex} = \sqrt{\frac{A}{2\pi M_S}}. \tag{2.2}$$

$A$ is the exchange constant and $M_S$ is the saturation magnetization. For Fe, $A = 2 \times 10^{-6}$ erg/cm and $M_S = 1710$ emu/cm$^3$. This yields $\sigma_{ex} = 33$ nm (23 atomic layers ML). At this length scale the exchange interaction is dominant over the demagnetizing energy. The moments are locked together across the film thickness and can be treated as a macrospin.

### 2.1.2 Zeeman Energy

The presence of an external magnetic field introduces the Zeeman energy density term:

$$F_{Zee} = -\vec{M} \cdot \vec{H}_{dc}. \tag{2.3}$$

where $\vec{M}$ is the saturation magnetization vector and $\vec{H}_{dc}$ is the external dc magnetic field vector. In principle, the direction of the magnetization $\vec{M}$ can be different from the direction of the external field $\vec{H}_{dc}$. This effect is particularly pronounced when the Zeeman energy density term is comparable to the anisotropy energy density terms in Eq. 2.1. When $\vec{H}_{dc}$ is not oriented along a magnetic easy axis the magnetization points somewhere in between the direction of the easy axis and the direction of the field $\vec{H}_{dc}$. This effect is often referred to as “magnetization dragging”.

### 2.1.3 Magnetic Anisotropy Energy (MAE)

Magnetic anisotropy describes the fact that the energy of a magnetic system depends on the direction of the magnetization with respect to the crystallographic axes. The easy magnetization directions, in the absence of the external field, are oriented along the magnetic energy minima. By analogy the hard magnetic axes are oriented along the magnetic energy maxima. The difference of the free energy $F$ with the magnetization pointing along the hard and easy magnetization axes is called the Magnetic Anisotropy Energy (MAE) which is equal to the work ($W_{MAE}$) required to rotate the the magnetization $\vec{M}$ (at constant temperature) between these two directions. There are fundamentally two sources of MAE: (a) Spin-orbit interaction (LS) and (b) the magnetic dipole-dipole
interaction [11, 12]. The MAE term, therefore, can refer to the magnetic free energy density which includes the magneto-crystalline and magneto-elastic contributions resulting from the LS coupling, and the dipole-dipole contributions. In particular, for zero applied external magnetic field \( F_{\text{exter}} = 0 \) the total magnetic free energy density \( F \) from Eq. 2.1 can be rewritten in a simpler form:

\[
F = F_{\text{an}} + F_{\text{magnet}} + F_d
\]  

(2.4)

The exchange interaction energy \( F_{\text{ex}} \) is assumed to be isotropic and therefore does not contribute to the MAE.

**Crystalline anisotropy**

Ultrathin films of cubic materials grown along the [001] crystallographic direction have in general a tetragonal symmetry. The corresponding free energy density term can be written in the form:

\[
F = -\frac{1}{2}K_{1\parallel}^4(\alpha_X^4 + \alpha_Y^4) - \frac{1}{2}K_{1\perp}^4\alpha_Z^4 - K_u^4\alpha_Z^2,
\]  

(2.5)

where \( \alpha_X \), \( \alpha_Y \) and \( \alpha_Z \) are the directional cosines of the magnetization vector \( \vec{M} \) with respect to the principle crystallographic axes (100), see Fig 2.1. \( K_{1\parallel} \) and \( K_{1\perp} \) are the in-plane and perpendicular to the film plane four-fold anisotropy coefficients, respectively. \( K_u^4 \) is the perpendicular uniaxial anisotropy coefficient.

\[
\begin{align*}
\alpha_X &= \sin \theta_{M} \cos \varphi_{M}, \\
\alpha_Y &= \sin \theta_{M} \sin \varphi_{M}, \\
\alpha_Z &= \cos \theta_{M},
\end{align*}
\]  

(2.6)

where \( \theta \) and \( \varphi \) are the polar angular coordinates, see Fig 2.1. In the literature \( K_1 \) and \( K_{1U} \) are sometimes labeled \( K_4 \) (fourth order) and \( K_2 \) (second order) anisotropy constants, respectively.

For a pure cubic anisotropy energy density Equation 2.5 can be written

\[
F_{\text{an}}^{\text{cub}} = K_1(\alpha_X^2\alpha_Y^2 + \alpha_X^2\alpha_Z^2 + \alpha_Y^2\alpha_Z^2),
\]  

(2.7)

since in the absence of tetragonal symmetry \( K_{1\perp} = 0 \) and \( K_{1\parallel} = K_1^4 \), and

\[
\alpha_X^4 + \alpha_Y^4 + \alpha_Z^4 = 1 - 2(\alpha_X^2\alpha_Y^2 + \alpha_X^2\alpha_Z^2 + \alpha_Y^2\alpha_Z^2).
\]  

(2.8)
CHAPTER 2. THEORETICAL BACKGROUND

Figure 2.1: The laboratory coordinate system is described by $X, Y, Z$ axes parallel to the principal crystallographic directions. The $Y - Z$ plane is perpendicular to the $X$ axis.

In thin films, surfaces and interfaces also break the symmetry of the crystal lattice. Any uniaxial distortion normal to the film plane leads to inequivalent in-plane and out-of-plane axes and result in an anisotropy term $\sim \alpha_Z^2$. When a uniaxial distortion of the crystal is present throughout the volume it contributes to the volume anisotropy. The uniaxial perpendicular anisotropy is then given by

$$F_{\text{uni,} \perp}^{\text{vol.}} = -K_u \alpha_Z^2,$$

(2.9)

In many cases a uniaxial anisotropy was also observed having its axis in the film plane and can be described by

$$F_{\text{uni,} \parallel}^{\text{vol.}} = -K_u \left( \hat{n} \cdot \hat{M} \right)^2 = -K_u \sin^2 \theta \cos^2(\varphi),$$

(2.10)

where $K_u$ is the in-plane uniaxial anisotropy constant and $\hat{n} = (\cos \varphi, \sin \varphi, 0)_{xyz}$ is the unit vector in the direction of the in-plane uniaxial axis. $\varphi = (\varphi_M - \varphi)$ is the in-plane angle between the saturation magnetization and the $K_u^\parallel$ axis. The origin of the in-plane uniaxial anisotropy still remains a controversial topic. A possible mechanism explaining a large $K_u^\parallel$ with the magnetic axis along the [110] crystallographic direction is based on the magneto-elastic energy term $B_2$ which is caused by an in-plane lattice shear. Recent experiments seem to support this view (see Ref.[13] and sec. 5.2 in chapter 5.).
CHAPTER 2. THEORETICAL BACKGROUND

**Volume and surface contribution**

A description of the magnetic properties of ultrathin magnetic structures can be simplified significantly compared with that for the bulk materials. All atomic moments across the film thickness are parallel due to the strong interlayer exchange interaction. This allows one to treat an ultrathin film like a giant magnetic molecule with a total magnetic moment given by a simple algebraic sum of all its constituent atomic moments. Ultrathin films have magnetic properties very different from those characteristic of bulk materials. The main reason for this difference lies in the interfaces. The presence of the interfaces changes the energetics of the system and must be taken into account. This means that the interface magnetic properties are shared together with the bulk magnetic properties resulting in typical 1/d contributions to the overall magnetic anisotropies and their effective fields: here d is the film thickness \[14, 15\]. The free energy \( F \) in ultrathin films can be written as a sum of volume and surface terms:

\[
F = f^V \cdot V + f^S \cdot S .
\]  

(2.11)

The energy per volume of a thin film becomes:

\[
\frac{1}{V} F = E \equiv f^V + \frac{1}{d} f^S .
\]  

(2.12)

By analogy the anisotropy constants can be written:

\[
K_i = K_i^V + \frac{(K_i^A + K_i^B)}{d} ,
\]  

(2.13)

where the index subscript \( i \) refers to the uniaxial and four-fold anisotropy coefficients. The superscripts A and B distinguish the contributions from the top and bottom (film/substrate) interfaces. Since in most measurements one is not able to distinguish the contributions from individual interfaces the term \((K_i^A + K_i^B)\) is often replaced by \(2K_i^S\).

**Demagnetizing energy**

In thin uniform magnetic films the presence of the surface develops magnetic pseudo charges leading to the dipole-dipole interaction. When the magnetization lies uniformly in the plane magnetic poles are avoided and this corresponds to the lowest magneto-static energy configuration. Any out-of-plane tilt of the magnetization by an external
magnetic field creates a magnetic surface charge density and results in a demagnetizing (restoring) energy density which has a form corresponding to a uniaxial anisotropy

\[ F_d = \frac{1}{2} \times 4\pi DM^2 \cos^2 \theta_M = 2\pi DM^2_{\perp}, \]  

(2.14)

where \( D \) is the effective demagnetizing coefficient which is very close to 1 for films thicker than a few atomic layers. \( M_{\perp} \) is the magnetization component perpendicular to the film surface and \( \theta_M \) is the angle of the magnetization with respect to the film normal as is illustrated in Fig. 2.1. The dipolar demagnetizing field is usually described in terms of a magnetic continuum in which the demagnetizing field is given by \( 4\pi MS \). This approach is however incorrect for films consisting of several atomic layers [16, 17, 18, 19]. The dipolar field depends on the number of atomic layers involved due to the discreetness of the magnetic moments that are localized around each atomic site. The average dipolar field inside the film decreases strongly as the thickness approaches the monolayer limit. The reduction of the dipolar field can be written as a reduced demagnetizing coefficient which for a bcc(001) Fe film is given by \( D = 1 - \frac{0.425}{N} \) where \( N \) is the number of atomic planes. For sufficiently thick films the approximation \( D = 1 \) is valid. In fact the thickness dependent part of the dipolar energy contributes to the perpendicular uniaxial surface energy.

Magneto-elastic contributions

Magneto-elastic anisotropies are created by lattice strain and the spin orbit interaction. The experimentally obtained magneto-elastic constants are significantly larger than the crystalline anisotropy constants [20, 21]. Consequently, even small strains may give rise to important anisotropy contributions. In epitaxial structures considerable strains may result from a film grown on a substrate having a different lattice parameter. At the initial stages of growth the lattice misfit can be accommodated by introducing a tensile strain in the substrate surface layer and a compressive strain in the film layers thereby adopting in this way the same in-plane lattice parameter. This situation occurs for thicknesses less than some critical thickness \( d_c \) (coherent regime). For thicknesses greater than \( d_c \) it becomes energetically more favorable to introduce misfit dislocations which partially accommodate the lattice misfit and thus reduce the strain (incoherent regime). The magneto-elastic magnetic anisotropy energy density \( F_{magel} \) in cubic systems
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can be written as follows [20]

\[
F_{magel} = B_1(\epsilon_{XX}\alpha_x^2 + \epsilon_{YY}\alpha_y^2 + \epsilon_{ZZ}\alpha_z^2) \\
+ 2B_2(\epsilon_{XY}\alpha_x\alpha_y + \epsilon_{YZ}\alpha_y\alpha_z + \epsilon_{ZX}\alpha_x\alpha_z) + ... 
\]

(2.15)

(2.16)

where \(B_i\) are the magnetoelastic coupling coefficients and \(\epsilon_{i,j}\) \((i,j = X,Y,Z)\) are the strain tensor components along axes that are perpendicular to each other, see the coordinate system defined in Fig. 2.1. Measured volume values of \(B_i\) for 3d ferromagnets can be found in Ref. [20]. In Eq. 2.15 if \(i = j\) the strain is directed along the cubic \(\langle 100 \rangle\) axes and leads to a change in volume of the unit lattice cell. For \(i \neq j\) the strain is directed along the \(\langle 110 \rangle\) axes and is equivalent to an in-plane shear strain which keeps the volume of the unit cell constant.

2.2 Landau-Lifshitz equation of motion

The magnetization dynamics for a film in the classical limit can be described by the well-known Landau-Lifshitz (LL) equation of motion [22, 14].

\[
\frac{1}{\gamma} \frac{\partial \vec{M}}{\partial t} = - \left[ \vec{M} \times \vec{H}_{\text{eff}} \right] - \frac{\lambda}{\gamma M_S^2} \left[ \vec{M} \times \vec{M} \times \vec{H}_{\text{eff}} \right],
\]

(2.17)

where \(\gamma = g | e | / 2mc\) is the absolute gyromagnetic ratio and \(\lambda = 1/\tau\) is the phenomenological damping constant equal to the inverse relaxation time. The first term on the right-hand side represents the precessional torque, and the second term represents the LL damping term. The free energies discussed in the previous section enter the equation of motion via the effective fields, \(H_{\text{eff}}\), given by the magnetization derivatives of the magnetic free energy density \(F_{\text{tot}}\) (see Eq. 2.1);

\[
\vec{H}_{\text{eff}} = - \frac{\partial F}{\partial \vec{M}}.
\]

(2.17)

In the static case, the magnetization vector \(\vec{M}\) has to be parallel to the effective field.

In the LL equation with \(\lambda \to \infty\) the damping term eventually becomes dominant over the precessional torque \(\vec{M} \times \vec{H}_{\text{eff}}\). One can view this term as an increasingly fast relaxation to the lattice meaning that the system is always in a quasi-equilibrium state. This behavior is unrealistic. In 1955 Gilbert introduced a different relaxation torque [23]
instead of the LL damping term and this results in the Landau-Lifshitz-Gilbert (LLG) equation of motion;

\[
\frac{1}{\gamma} \frac{\partial \vec{M}}{\partial t} = - \left[ \vec{M} \times \vec{H}_{\text{eff}} \right] + \frac{G}{\gamma^2 M_S} \left[ \vec{M} \times \frac{\partial \hat{n}}{\partial t} \right] \tag{2.18}
\]

\[
= - \left[ \vec{M} \times \left( \vec{H}_{\text{eff}} - \frac{\alpha}{\gamma} \frac{\partial \hat{n}}{\partial t} \right) \right] \tag{2.19}
\]

where \( G \) is the Gilbert damping in units of \([s^{-1}]\) and \( \hat{n} = \vec{M}/M_S \) is the magnetization unit vector. \( \alpha = \frac{G}{\gamma M_S} \) is a popular representation of the the dimensionless Gilbert damping parameter. Note that the damping term in Eq. 2.19 can be expressed using an effective damping field \( H_{\text{eff}}^G \)

\[
H_{\text{eff}}^G = - \frac{G}{\gamma^2 M_S} \frac{\partial \hat{n}}{\partial t} \tag{2.20}
\]

With increasing \( \alpha \) the second term on the right hand side of Eq. 2.18 eventually becomes dominant and \( \frac{\partial \vec{M}}{\partial t} \to 0 \). For \( \alpha \to \infty \) the magnetic system approaches equilibrium at an infinitely slow rate. For \( \alpha \ll 1 \) the Gilbert and LL damping terms are identical. Physically the magnetic damping leads to a loss of spin angular momentum and the relaxation term can be written as \( 1/\tau (\vec{m}/\chi_{\perp} - \vec{h}_{\text{eff}}) \), where \( \chi_{\perp} = M_S/H_{\text{eff}} \) is the transverse susceptibility, and \( \vec{m} \) and \( \vec{h}_{\text{eff}} \) are the rf magnetization and effective field components, respectively.

### 2.3 Ferromagnetic resonance

In ferromagnetic resonance (FMR) experiments a small microwave magnetic field \( \vec{h}_{\text{rf}} = \vec{h}_{\text{rf}} \sin \omega t \) excites the magnetization at a fixed frequency \( \omega = 2\pi f \). At the same time an external magnetic field is applied that changes the precessional frequency. The sample undergoes FMR when the precessional frequency equals the microwave frequency. The resonance is accompanied by an increase in microwave losses. In FMR the rf field \( h_{\text{rf}} \) in ultrathin films is equal to the external driving field created in a microwave cavity assuming that the driving field is symmetric, equal on both sides of the film. Gilbert damping is strong in ferromagnetic samples so that only small deviations of the magnetization \( \vec{M} \) from its static direction \( \vec{M}_S \) are usually excited. In that case the LLG equation of motion can be solved in the small angle approximation; \( |\vec{m}| \ll M_S \), where \( \vec{m} \) represents the rf
component of the magnetization vector. For small precessional angles equation 2.18 can be linearized by looking for solutions having the form:

\[ \vec{M} = \vec{M}_S + \vec{m}, \quad (2.21) \]

where \( \vec{M}_S \) and \( \vec{m} \) are the longitudinal and transverse components of the total magnetization \( \vec{M} \). The total effective field \( \vec{H}_{\text{eff}} \) entering the torque equation 2.18 is given by the sum of all fields

\[ \vec{H}_{\text{eff}} = (\vec{H}_{\text{dc}} + \vec{h}_{\text{rf}})_{\text{ext}} + \vec{H}_{\text{an}} + \vec{H}_d + \vec{H}_{\text{magel}}. \quad (2.22) \]

It is convenient to carry out this linearization in an \((x, y, z)\) magnetization coordinate system in which the \( x \) axis points along the direction of \( \vec{M}_S \) and the \( y \) and \( z \) axes lie in the plane perpendicular to \( \vec{M}_S \). The driving rf field \( \vec{h}_{\text{rf}} \), see Fig. 2.1, is parallel to the \( y \) axis, therefore it is assumed to be perpendicular to \( \vec{M}_S \).

### 2.3.1 In-plane configuration

In the parallel configuration the dc magnetic field \( H_{\text{dc}} \) and \( M_S \) lie in the plane of the film; \( \theta_M = \theta_H = 90^\circ \). The evaluation of the effective field can be solved by rotating the \( X, Y, Z \) coordinate system, related to the crystallographic directions, by an angle \( \phi_M \) around the [001] direction, where \( \phi_M \) is the angle between \( \vec{M}_S \) and the [100] direction. The rotation matrix can be written as

\[
\begin{pmatrix}
\cos \phi_M & \sin \phi_M \\
-\sin \phi_M & \cos \phi_M
\end{pmatrix}
\]

The saturation magnetization in the new \( x, y, z \) coordinate system is parallel to the \( x \)-axis

\[ \vec{M} = (M_S, m_y, m_z). \quad (2.23) \]

The resulting effective fields using magnetization derivatives of the free energy density terms defined in a previous section are given by

\[ \vec{H}_{\text{see}} = (H_{\text{dc}}(\cos (\varphi_M - \varphi_H), h_{\text{rf}} + H_{\text{dc}}(\sin (\varphi_M - \varphi_H)), 0), \quad (2.24) \]

\[ \vec{H}_d = (0, 0, -4\pi Dm_z), \quad (2.25) \]

\[ \vec{H}_{\text{magel}} = \left( 0, 0, \frac{-2B_1}{M_S^2} (e_\perp - e_\parallel) \right) \quad (2.26) \]
\[ \tilde{H}_{\text{an}} = \left( \frac{K_{\|}}{2M_S} \right) \left( 3 + \cos 4\varphi_M + 3\frac{m_y}{M_S} \sin 4\varphi_M \right) \frac{K_{\|}}{M_S} \left( 1 + \cos 2\varphi + \frac{m_y}{M_S} \sin 2\varphi \right), \]
\[ \frac{K_{\|}}{2M_S} \left( \sin 4\varphi_M + 3\frac{m_y}{M_S} (1 - \cos 4\varphi_M) \right) + \frac{K_{\|}}{M_S} \left( \sin 2\varphi + \frac{m_y}{M_S} (1 - \cos 2\varphi) \right), \]
\[ \frac{2K_{\|}}{M_S^2} m_z. \]

(2.27)

\( e_{\perp} \) and \( e_{\parallel} \) in Eq. 2.27 are the strains perpendicular and parallel to the film plane respectively. The LLG equations of motion for the time dependance \((e^{i\omega t})\) and using the effective fields evaluated above become a system of two coupled equations for \(m_y\) and \(m_z\):

\[ i\omega m_y + \left[ H_{\parallel} + 4\pi M_{\text{eff}} + A_z + i\frac{\omega}{\gamma} \right] m_z = 0, \]
\[ -i\frac{\omega}{\gamma} m_z + \left[ H_{\parallel} + A_y + i\frac{\omega}{\gamma} \right] m_y = M_S h_{\text{rf}}, \]

where

\[ A_z = \frac{K_{\|}}{2M_S} (3 + \cos 4\varphi_M) + \frac{K_{\parallel}}{M_S} (1 + \cos 2\varphi), \]
\[ A_y = \frac{2K_{\parallel}}{M_S} \cos 4\varphi_M + \frac{2K_{\parallel}}{M_S} \cos 2\varphi. \]

(2.28)

(2.29)

(2.30)

(2.31)

\( A_z \) and \( A_y \) represent the effective anisotropy fields in the plane perpendicular to \(M_S\). \( H_{\parallel} \) is the projection of the external dc field \(H_{\text{dc}}\) into the direction of the saturation magnetization and the effective demagnetizing field is the sum of all perpendicular uniaxial anisotropy contributions defined by

\[ 4\pi M_{\text{eff}} = 4\pi D M_S + \frac{2B_1}{M_S} (e_{\perp} - e_{\parallel}) - \frac{2K_{\parallel}}{M_S} \]

(2.32)

Using Eq.2.28 to eliminate \(m_z\) from Eq. 2.29 yields an expression for the rf transversal susceptibility \(\chi_{\perp} = \frac{m_y}{h_{\text{rf}}}\) as a function of the applied field \(H_{\text{dc}}\)

\[ \chi_{\perp} \equiv \chi_{\perp} + i\chi_{\perp}'' = \frac{M_S \left( H_{\parallel} + 4\pi M_{\text{eff}} + A_z - i\frac{\omega}{\gamma} \right)}{\left( H_{\parallel} + 4\pi M_{\text{eff}} + A_z - i\frac{\omega}{\gamma} \right) \left( H_{\parallel} + A_y - i\frac{\omega}{\gamma} \right) - \left( \frac{\omega}{\gamma} \right)^2}. \]

(2.33)

Ferromagnetic resonance occurs when the imaginary part of the susceptibility is at maximum and corresponds to a maximum power absorption by the film. The resonance
condition therefore is satisfied when the real part of the denominator is zero. In that case

\[
\left( \frac{\omega}{\gamma} \right)^2 = (H_{FM_R} + 4\pi M_{eff} + A_z)(H_{FM_R} + A_y) = B\mathcal{H},
\]

(2.34)

where \( B \) and \( \mathcal{H} \) are both evaluated at the resonance condition \( (H_{dc} = H_{FM_R}) \).

It can be shown that for the saturated case \( (\varphi_M = \varphi_H \text{ and } H_{||} = H_{dc}) \) the imaginary part of the susceptibility, Eq. 2.33, is given by an almost perfect Lorentzian function [14]

\[
\Im \chi_\perp \equiv \chi_\perp'' = M_S \frac{B}{B + \mathcal{H}} \times \frac{1}{\Delta H} \frac{1}{1 + \left( \frac{H_{dc} - H_{FM_R}}{\Delta H} \right)^2},
\]

(2.35)

where

\[
\Delta H = \frac{\omega}{\gamma} = \frac{\omega}{\gamma} \frac{G}{M_S}
\]

(2.36)

is the half width at half maximum (HWHM) linewidth and \( H_{FM_R} \) is the field corresponding to the center of the absorption line.

### 2.3.2 Perpendicular configuration

In the out-of-plane configuration the external dc magnetic field and magnetization are oriented strictly parallel to the film normal \( (\theta_M = \theta_H = 0^\circ) \). The external field and magnetization vectors are defined by

\[
\vec{H}_{dc} = (0, h_{rf}, H),
\]

(2.37)

\[
\vec{M} = (m_x, m_y, M_S).
\]

(2.38)

The FMR resonance condition and FMR linewidth are derived in the same way as for the parallel configuration by solving equations 2.18 for a small deviation from equilibrium. They are found to be

\[
\left( \frac{\omega}{\gamma} \right)^2 = \left( H_{FM_R} - 4\pi M_{eff} + \frac{2K_1^\perp}{M_S} \right) \left( H_{FM_R} - 4\pi M_{eff} + \frac{2K_1^\perp}{M_S} - \frac{2K_u}{M_S} \right)
\]

(2.39)

\[
\Delta H = \frac{\omega}{\gamma} \frac{G}{M_S}
\]

(2.40)

where \( K_1^\perp \) is the four-fold perpendicular anisotropy.
For both the parallel and perpendicular configurations, the FMR linewidth follows the same expression regardless of the ellipticity of the rf precession [15]. Within the Gilbert phenomenology, the FMR linewidth is strictly proportional to the angular microwave frequency $\omega$ and inversely proportional to the saturation magnetization $M_s$.

### 2.4 Spin dynamics and relaxation mechanisms

Spin dynamics in ferromagnetic films includes a relaxation mechanism which can be divided into two categories: a sample dependent damping (extrinsic damping) or a sample independent damping (intrinsic damping). As pointed out in sec. 2.3 magnetic relaxation is described by the second term on the right hand side of the LLG equation of motion, Eq. 2.18. Experimentally the magnetic damping parameter $\alpha$ is usually extracted from the frequency dependence of the FMR linewidth ($\Delta H$) using equation Eq. 2.36. $\Delta H$ arising from an extrinsic damping mechanism can be caused by structural defects and sample inhomogeneities. These can be avoided in principle through the preparation of high quality crystalline films. On the other hand, there are physical processes which are unavoidable, such as electron scattering by phonons and magnons. These processes are called intrinsic and determine the Gilbert damping parameter. For thick metallic layers one cannot also avoid energy dissipation by eddy currents. From the experimental point of view the intrinsic value of the magnetic relaxation is given by the smallest measured value of $\Delta H$ that satisfies Eq. 2.36. In ultrathin films the Gilbert damping is caused by the spin-orbit interaction and contains both bulk and interface contributions. The interface contribution cannot be avoided and therefore it is also considered to be intrinsic. Physical concepts, which describe the intrinsic damping mechanisms, will be discussed in the next section. Extrinsic relaxation mechanisms lie outside the scope of this thesis and therefore they will not be discussed in detail. Experimental evidence for extrinsic contributions to $\Delta H$ will be presented in Chapter 5. A more complete list of references for extrinsic damping in ultrathin films can be found in [24, 25].

#### 2.4.1 Gilbert damping in ferromagnets

The Gilbert damping coefficient $G$ in metals is caused by incoherent scattering of electron-hole pair excitations by phonons and magnons. The electron-hole pair excitations involve
both spin-flip and spin conserved scattering.

**Spin-flip scattering.**

The spin-flip model of magnetic relaxation was originally proposed by Heinrich et al [26]. It is based on the s-d exchange interaction. One should consider two groups of electrons. Those that are localized (d-electrons) and those that are itinerant (s-electrons). In reality itinerant electrons are hybridized states of s-p and d electrons. In this model the interaction can be obtained by integrating the s-d exchange density functional [27]

\[
E_{s-d} = \sum_{j} \int_{V} J(\mathbf{r}_{j} - \mathbf{r}) S_{j,d} S_{S}(\mathbf{r}) d\mathbf{r}^{3},
\]

where \( J(\mathbf{r}_{j} - \mathbf{r}) \) is the s-d exchange interaction between the s-p electron spin density \( S_{S} \) and the localized d-electron spin density \( S_{j,d} \), where \( j \) is the lattice site. The Hamiltonian which involves the rf component of magnetization is described by a three-particle collision term

\[
H_{s-d} = \sqrt{\frac{2S}{N}} \sum_{k} J(\mathbf{\bar{q}}) a_{k,\uparrow} a_{k+\mathbf{q},\downarrow}^{\dagger} b_{\mathbf{q}} + (h.c.),
\]

where \( N \) is the number of atomic sites, \( S \) is the spin of d-electrons, \( J(\mathbf{\bar{q}}) \) is the s-d exchange constant, \( a \) and \( a^{\dagger} \) are annihilation and creation operators, respectively for the wave-vectors \( \mathbf{k} \) and \( \mathbf{k} + \mathbf{\bar{q}} \) with the appropriate spin (\( \uparrow, \downarrow \)). \( b \) annihilates the magnon with wave vector \( \mathbf{q} \). The graphical representation of Eq. 2.42 is shown in Fig. 2.2. The electron-hole pair is created or annihilated due to scattering with a magnon.

The total spin angular momentum for this process is conserved, and therefore the annihilation of a magnon requires that the electron spin flip in the scattering process. The scattering of electron-hole pairs does not lead to magnetic damping for long wavelength spin waves (\( \mathbf{q} \to 0 \)). One needs to include the finite life-time (noise) of the electron-hole pairs (\( a_{k,\uparrow} a_{k+\mathbf{q},\downarrow}^{\dagger} \)) due to incoherent scattering by thermally excited phonons and magnons. This can be done by introducing an imaginary term in the electron-hole pair energy [26]

\[
\Delta \epsilon_{\mathbf{k},\mathbf{k}+\mathbf{\bar{q}}} = \epsilon_{\mathbf{k}+\mathbf{\bar{q}},\downarrow} - \epsilon_{\mathbf{k},\uparrow} + i\frac{\hbar}{\tau_{sf}},
\]

where \( \tau_{sf} \) is the effective lifetime of the electron-hole pair excitation. \( \tau_{sf} \) is caused by incoherent electron scattering by phonons and magnons which flips the electron spin.
Figure 2.2: A graphical representation of a three-particle collision. A magnon with an energy $\hbar \omega_q$ collides with an itinerant electron of energy $\varepsilon_k$ and spin $\uparrow$. This results in annihilation of the magnon and creation of an electron-hole pair. The excited electron carries the energy $\varepsilon_{k+q}$ and spin $\downarrow$. Note that the spin angular momentum is conserved in this process.

The spin flip process requires the presence of the spin orbit interaction. The Gilbert damping can be calculated by evaluating the rf susceptibility using the Kubo Green function formalism in the Random Phase Approximation (RPA) [28, 29]. The parameter $\alpha$ can be expressed as the effective damping field due to spin-flip scattering, $H_{eff}^{sf}$ and is given by the imaginary part of the Green function

$$H_{eff}^{sf} = \alpha = \frac{\omega}{\gamma} = \frac{2\overline{S}}{N g \mu_B} \hbar \omega_k \sum_k |J(q)|^2 \left| \frac{dn}{d\varepsilon} \right| L,$$  \hspace{1cm} (2.44)

where the summation is carried out over all available states at the Fermi surface. $\overline{S} = M_S(T)/M_S(0)$ is the reduced spin $S$, and $n$ is the density of states (DOS). The factor $g \mu_B = \gamma \hbar$ was used to convert energy into an effective field. The probability of a scattering event in which energy is conserved ($\hbar \omega_k = \varepsilon_{k+q,\uparrow} - \varepsilon_{q,\uparrow}$) is represented by the Lorentzian factor $L$ in Eq. 2.44

$$L = \frac{\frac{\hbar}{\tau_{sf}}}{(\hbar \omega_k + \varepsilon_{q,\uparrow} - \varepsilon_{k+q,\uparrow})^2 + \left(\frac{\hbar}{\tau_{sf}}\right)^2}.$$  \hspace{1cm} (2.45)

Equation 2.44 can be simplified further by noticing that the $(\hbar/\tau_{sf})^2$ term can be neglected when compared to the $(\hbar \omega_k + \varepsilon_{q,\uparrow} - \varepsilon_{k+q,\uparrow})^2$ term. Assuming a Fermi distribution of the form

$$\left| \frac{dn}{d\varepsilon} \right| = \delta(\varepsilon_k - \varepsilon_F),$$  \hspace{1cm} (2.46)
where $\delta$ is the Dirac delta function and $\varepsilon_F$ is the Fermi energy, the Gilbert damping field can be rewritten as

$$H_{\text{eff}}^{sf} = \frac{2S}{Ng\mu_B} \sum_k |J(q)|^2 \delta(\varepsilon_k - \varepsilon_F) \hbar \omega_q \frac{\hbar}{\tau_{sf}} \frac{1}{(\hbar \omega_k + \varepsilon_{q,1} - \varepsilon_{k+q,1})^2} \tag{2.47}$$

In FMR the sample response is given by the nearly homogenous mode, $q \to 0$. The difference in electron energies in a spin-flip process is dominated by the exchange energy:

$$\hbar \omega_k + \varepsilon_{q,1} - \varepsilon_{k+q,1} \approx \varepsilon_{q,1} - \varepsilon_{k+q,1} = -2S J(0). \tag{2.48}$$

Assuming unit volume and using $N_S g \mu_B = M_S$, $H_{\text{eff}}^{sf}$ can be expressed as

$$H_{\text{eff}}^{sf} = \frac{\hbar^2 \omega}{2M_S \tau_{sf}} \sum_k k \delta(\varepsilon_k - \varepsilon_F). \tag{2.49}$$

Note that Eq.2.49 is directly proportional to $\omega$ and inversely proportional to $M_S$. This is the signature of Gilbert damping, see Eq. 2.20. Comparing Eq.2.49 and Eq. 2.20 one can use the expression for the Gilbert damping parameter $G$

$$G = \frac{\chi_P}{\tau_{sf}} \Rightarrow \alpha = G = \frac{\chi_P}{\gamma M_S} = \frac{\chi_P}{\gamma M_S \tau_{sf}}; \tag{2.50}$$

where $\chi_P$ is the Pauli susceptibility of the itinerant electrons and can be calculated by integration over an appropriate Fermi surface sheet.

$$\chi_P = \left(\frac{\hbar \gamma}{2\pi}\right)^2 \int k^2 \delta(\varepsilon_k - \varepsilon_F) dk = \mu_B^2 n(\varepsilon_F), \tag{2.51}$$

where $n(\varepsilon_F)$ is the density of states at the itinerant electron Fermi level.

**Spin-flip relaxation time**

The spin relaxation time $\tau_{sf}$ is enhanced compared to the electron momentum relaxation time, $\tau_{el}$. The reason for the longer relaxation time is that one needs to invoke the spin-orbit interaction in order to flip the electron spin during the momentum scattering process. For normal metal electron scattering by 3d impurities the ratio between the spin flip scattering and non-spin flip cross section is $\sim 10^{-2}$ [30]. A good estimate of $\tau_{sf}$ can be obtained by evaluating the spin diffusion length $l_{sd}$ which can be obtained from the Current Perpendicular to Plane Giant Magnetoresistance (CPP GMR):

$$l_{sd} = \sqrt{\frac{\lambda_{FM}^{FP} \mu_F \tau_{sf}}{6}}, \tag{2.52}$$
where \( v_F \) is the Fermi velocity and \( \lambda_{FM}^* \) is the effective momentum mean free path in the ferromagnet [31] defined by

\[
\frac{1}{\lambda_{FM}^*} = \frac{1}{2} \left[ \frac{1}{\lambda_{\uparrow}} + \frac{1}{\lambda_{\downarrow}} \right],
\]

(2.53)

where \( \lambda_{\uparrow,\downarrow} \) is the mean free path for majority and minority electrons. In the free electron model \( \lambda_{FM}^* \) can also be estimated from the formula

\[
\lambda_{FM}^{\text{star}} = \frac{m v_F}{n e^2 \rho^*}
\]

(2.54)

where \( \rho^* = \rho^{DC}(1 - \beta^2) \). Here \( \rho^{DC} \) is the measured resistivity of the ferromagnet and \( \beta \) is a bulk spin asymmetry coefficient. Experimental values of \( l_{sd} \) can be found in the references [32, 33, 34]

**Non-spin-flip scattering**

In the seventies Kambersky proposed a model for the relaxation mechanism in metals using a spin orbit Hamiltonian [35, 36]. Kambersky invoked in his model an explicit dependence of the electron energy levels on the magnetization direction, \( \hat{n} = \hat{M}/M_S \). This is caused by the spin orbit interaction and has to be included using relativistic electron band calculations. As the magnetization precession evolves in time and space the Fermi surface also changes periodically in time and space. This is often referred to as the *breathing Fermi surface*. The electron population number \( n_{k,u} \) relaxes towards the instantaneous equilibrium value by means of an electron-phonon scattering process. The magnetic damping arises from the phase lag between the actual electron population and the population corresponding to the instantaneous magnetization direction [35, 37].

The variation of the total electron energy density

\[
\mathcal{E} = \Omega^{-1} \sum_{k,u} \varepsilon_{k,u} n_{k,u}
\]

(2.55)

with the magnetization direction can be described by an effective field

\[
\mathcal{H} = -\frac{1}{M_S \Omega} \sum_{k,u} n_{k,u} \frac{\partial \varepsilon_{k,u}}{\partial \hat{n}}
\]

(2.56)

where \( \Omega \) is the volume of the sample and \( \varepsilon_{k,u} \) is the energy of the appropriate electron state. The non-equilibrium populations \( n_{k,u} \) are approximately given by Boltzman's
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equation

\[ n_{k,u} = f_{k,u} - \tau_{k,u} \frac{\partial f_{k,u}}{\partial t} \]  \hspace{1cm} (2.57)

where \( f_{k,u} = f(\varepsilon_{k,u}) \), \( f(\varepsilon) \) is the Fermi function and \( \tau_{k,u} \) is the lifetime of the appropriate electron state. Since \( f_{k,u} \) depends on \( t \) through \( \varepsilon_{k,u}[\hat{n}(t)] \), the out-of-phase part of the effective field is obtained from the chain rule using Eq. 2.57

\[ \mathcal{H} = -\frac{1}{M_S} \sum_{k,u} \tau_{k,u} \left( \frac{\partial f(\varepsilon_{k,u})}{\partial \varepsilon_{k,u}} \right) \frac{\partial \varepsilon_{k,u}}{\partial \hat{n}_j} \frac{\partial \hat{n}_j}{\partial t} \]  \hspace{1cm} (2.58)

Equation 2.58 simplifies further assuming that \(-\frac{\partial f(\varepsilon_{k,u})}{\partial \varepsilon_{k,u}} \approx \delta(\varepsilon_F - \varepsilon_{k,u})\). Using the same effective lifetime \( \tau \) for all the states one can obtain an expression for the damping parameter \( \alpha \)

\[ \alpha = \frac{\tau \gamma}{M_S} \sum_{k,u} \delta(\varepsilon_F - \varepsilon_{k,u}) \left( \frac{\partial \varepsilon_{k,u}}{\partial \hat{n}_j} \right)^2 \]  \hspace{1cm} (2.59)

Since there is no spin flip scattering involved, the electronic lifetime \( \tau \) is equal to the momentum electron relaxation time \( \tau_{ed} \).

2.4.2 Spin transport and relaxation at a ferromagnet/normal metal interface. Spin-pump effect

The role of interface Gilbert damping in thin ferromagnetic films has been experimentally studied by [38, 39, 40, 41]. In ultrathin ferromagnetic films \( \alpha \) was in some cases found to be quite large in comparison with the bulk intrinsic value \( (\alpha_b) \), and dependent sensitively on the substrate and capping layer materials and their thicknesses. Tserkovniak, Bratas and Bauer [42] introduced a theory which quantitatively explained these experimental results. Their theory was based on a new physical picture, according to which the ferromagnetic damping could be understood as an adiabatic pumping of spins into the adjacent normal metals. In the following description the spin transfer is governed by the reflection and transmission matrices of electrons in the normal metal films adjacent to the ferromagnetic film. They used Brouwer’s dynamic scattering theory [43]. The complete derivation of the spin pumping theory is rather complex, and a full description will be omitted in this thesis. It can be found in review articles by Tserkovniak et. al [44, 45] and Simanek [46]. Therefore this section will highlight only the main aspects of the spin
pumping theory and the related spin relaxation mechanism. The results presented below generate quantitative predictions for the magnetization damping in ferromagnet-normal metal systems that can be tested by experiment.

**Precession induced Spin Pumping**

One can consider a ferromagnet (F)-normal metal (NM) junction (NM/F/NM) as shown in Fig. 2.3. Without a voltage bias, no spin or charge currents flow when the magnetization of the ferromagnet is static. When the magnetization direction starts to precess the spin current $I_S$ is pumped out of the F/N interfaces [42]. This spin current from the F to the N layers depends on the complex parameter $A = A_r + iA_i$ (the "spin-pumping conductance") and is given by

$$i_{S}^{\text{pump}}(t) = \frac{\hbar}{4\pi} A_r \left( \hat{n} \times \frac{\partial \hat{n}}{\partial t} \right) - \frac{\hbar}{4\pi} A_i \frac{\partial \hat{n}}{\partial t}, \quad (2.60)$$

where $\hat{n}(t)$ is the unit vector along the magnetic moment $\vec{M}$ assuming a single-domain magnet (a macroscopic spin system) having a spatially uniform magnetization at all times. The scattering matrix of the ferromagnetic film and is given by:

$$A(r) = \frac{1}{2} \sum_{mn} |r_{mn}^r - r_{mn}^l|^2 + |r_{mn}^r - t_{mn}^l|^2, \quad (2.61)$$
\[ A(i) = Im \sum_{mn} [r_{mn}^\dagger (r_{mn}^\dagger)^* + t_{mn}^\dagger (t_{mn}^\dagger)^*]. \]  

(2.62)

\( r_{mn}^\dagger \) are reflection and \( t_{mn}^\dagger \) are transmission coefficients for the spin-up (spin-down) electrons on the normal-metal side, see Fig.2.3. The indexes \( m \) and \( n \) label the modes (channels) corresponding to \( \vec{k}_{F,\perp} \) wave vectors (parallel and perpendicular to the interface) at the Fermi energy. The coherent spin pumping occurs at the ferromagnet surface up to the depth of thickness defined by:

\[ \pi/(k_F^1 - k_F^1), \]  

(2.63)

where \( k_F^1 \) are the spin-dependent Fermi wave vectors in \( F \). The spin pumping matrix is usually defined per unit area:

\[ \frac{A}{S} = g_{11}^i + ig_{11}^{11}. \]  

(2.64)

Substituting Eq. 2.64 into Eq. 2.60 one can obtain an expression for the resulting magnetic moment per unit area that is generated by spin pumping:

\[ \vec{j}_{m}^{\text{pump}} = -\frac{g_\mu_B}{4\pi} \left[ g_{11}^i \hat{n} \times \frac{\partial \hat{n}}{\partial t} + ig_{11}^{11} \frac{\partial \hat{n}}{\partial t} \right]. \]  

(2.65)

By conservation of the total angular momentum the spins ejected by the spin current create a torque on the ferromagnet, \( \vec{\tau} = -\vec{j}_{S}^{\text{pump}} \). If one disregards spin-flip processes at the interface \( \vec{\tau} \) is entirely transferred to the magnetization precession. The spin dynamics can then be described by a generalized Landau-Lifshitz-Gilbert equation:

\[ \frac{\partial \vec{M}}{\partial t} = -\gamma \left[ \vec{M} \times \vec{H}_{\text{eff}} \right] + \alpha_0 \left[ \vec{M} \times \frac{\partial \hat{n}}{\partial t} + \frac{1}{d_F} \vec{j}_{m}^{\text{pump}} \right], \]  

(2.66)

where \( \alpha_0 = \frac{G}{\gamma M_s} \) is the dimensionless bulk Gilbert damping parameter and \( \gamma = \frac{g_{\mu_B}}{\hbar} \) is the gyromagnetic ratio. \( \vec{M} \) is the vector of instantaneous saturation magnetization of the ferromagnet, and \( d_F \) is the film thickness.

**Additional Gilbert damping due to a spin pump**

The real part of the mixing conductance contributes to the intrinsic magnetic damping, while the imaginary part of the mixing conductance contributes to the gyromagnetic coefficient. The additional damping \( \alpha^{\text{op}} \) can be observed in FMR spectra as an enhanced FMR linewidth [39, 47]. \( g_r \gg g_i \) and therefore spin pumping contributes in FMR mostly
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to the FMR linewidth (damping) and can be neglected in the resonance field \cite{44}. Using
equations 2.65 and 2.66 one can obtain an expression for the dimensionless damping
parameter due to spin pumping
\[ \alpha^{sp} = \frac{g\mu_B}{4\pi M_S} g^{\text{II}} \frac{1}{d_F}, \]
(2.67)
where \( g \) is the spectroscopic g-factor coefficient and \( \mu_B \) is the Bohr magneton. \( d_F \) is
the thickness of the ferromagnetic layer and \( g^{\text{II}} \) is evaluated for a unit interfacial area.
\( g^{\text{II}} \) is proportional to the effective interface mixing conductance (in units of \( e^2/h \)) which
to a very good approximation is given by the number of electron transverse channels
impinging upon the normal metal F/N interface,
\[ g^{\text{II}}_r = \frac{k_F^2}{4\pi} \approx 0.76n^{3/2}, \]
(2.68)
where \( n \) is the density of itinerant electrons per spin and \( k_F \) is the Fermi wave vector
in the normal metal (N). For simplicity the subscript \( r \) will be disregarded in further
discussions and \( g^{\text{II}} \) will refer to real part of the spin mixing conductance.

Schep et al. \cite{48} found that the interface conductances have to be renormalized in
terms of the interface resistance \( 1/g^{\text{II}} \) by studying transport through transparent inter­
faces in a diffuse environment. He found that the bulk resistances, which are proportional
to the layer thicknesses, are in series with the interface resistances.
\[ \frac{1}{g^{\text{II}}} = \frac{1}{g^{\text{II}}} - \frac{1}{2} \left( \frac{1}{N_{NM}} \right), \]
(2.69)
where \( N_{NM} \) is the number of modes (channels) of the normal metal (NM). Physically, the
second term on the right hand side in Eq. 2.69 represents the resistance between NM and
the ohmic lead, see Fig. 2.3, which is equal to 1/2 of Sharvin resistance. Thus, inverse
mixing conductance has to be corrected for the normal-metal Sharvin contribution only:
\[ \frac{1}{\tilde{g}^{\text{II}}} = \frac{1}{\tilde{g}^{\text{II}}} - \frac{1}{2g_{NM}^{\text{Sh}}}, \]
(2.70)
where \( g_{NM}^{\text{Sh}} = k_F^2/4\pi \) is the Sharvin conductance (number of transport channels \( N_{NM} \) of
the normal metal). Using the approximation \( g_{NM}^{\text{Sh}} \approx g^{\text{II}} \) \cite{44} and Eq. 2.70 one arrives at
the simple conclusion;
\[ \tilde{g}^{\text{II}} = 2g^{\text{II}}. \]
(2.71)
The values of the interface mixing conductance can be obtained experimentally by mea­
suring the additional damping parameter \( \alpha^{sp} \) due to spin pumping. The calculated values
of \( g^{\text{II}} \) and \( g_{NM}^{\text{Sh}} \) for various intermetallic interfaces have been tabulated in Ref. [44].
2.4.3 Extrinsic damping - two-magnon scattering.

Two-magnon scattering model

It was shown in the section 2.3 (see Eq. 2.36 and Eq. 2.40) that intrinsic damping results in an FMR linewidth which is linearly proportional to the microwave frequency. Experimentally, however, the linewidth is often found to have a linear frequency dependence with an extrapolated non-zero frequency offset ($\Delta H(0)$). Consequently, the measured linewidth versus frequency is often interpreted by using the following expression

$$\Delta H(\omega) = \Delta H(0) + \alpha \frac{\omega}{\gamma}. \quad (2.72)$$

The linear term in Eq. 2.72 is assumed to be a measure of the intrinsic damping. The frequency offset $\Delta H(0)$ can be appreciable and in some cases can reach several hundred Oe [49]. The frequency offset approaches zero for the best Fe samples [39]. Clearly $\Delta H(0)$ depends on the film quality. This implies that $\Delta H(0)$ is an extrinsic linewidth contribution and is caused by magnetic defects.

The extrinsic contribution to the FMR linewidth can be explained by a two magnon scattering mechanism. A uniform magnon, $q \sim 0$, driven by the rf field at FMR, scatters into a $q \neq 0$ magnon. Energy conservation requires that the scattered magnon have the same energy as the resonant uniform magnon. However the momentum conservation ($q$ wave vector conservation) is not required due to the loss of translational invariance. The two-magnon scattering mechanism was proposed in the 1960’s to explain the extrinsic FMR line broadening in YIG spheres [50]. The same mechanism was successfully used to describe the extrinsic damping mechanism in ferrites [51, 52, 53, 54, 55] and in metallic films [56].

The two magnon scattering matrix is proportional to components of the Fourier transform of the magnetic inhomogeneities

$$A(\vec{q}) = \int \Delta U(\vec{r}) \exp^{-i\vec{q}\vec{r}} \, d\vec{r}, \quad (2.73)$$

where $\Delta U(\vec{r})$ stands symbolically for variations of the local magnetic anisotropy energy. In ultrathin films the magnon $\vec{q}$ vectors are confined to the film plane.

In 1999 Arias and Mills [57] introduced a theory of two-magnon scattering that applies to ultrathin films. For an in-plane magnetization oriented along the external field direction, and neglecting magnetocrystalline anisotropies, the spin wave manifold
can be written in the form:

\[ \left( \frac{\omega_{\text{f}}}{\gamma} \right)^2 = [H + 4\pi M_{\text{eff}} - 2\pi M_S \bar{q} \bar{d} + Dq^2] \times [H + 2\pi M_S \bar{q} \bar{d} \sin^2 \psi_{\bar{q}} + Dq^2], \quad (2.74) \]

where \( \bar{q} \) is the wavevector of the scattered magnon, and \( \psi_{\bar{q}} \) is the angle between the in-plane projection of the static magnetization and the propagation direction of the spin wave with the wavevector \( \bar{q} \). The FMR mode corresponds to a nearly homogeneous mode with \( \bar{q} = 0 \). The third term of the first bracket on the right hand side of eq. 2.74 is negative and proportional to \( \bar{q} \), and therefore it lowers the resonance frequency, see Fig 2.4. For large \( \bar{q} \) vectors, the fourth term in the first bracket (representing the exchange field) increases the magnon energy. Consequently, the magnon dispersion curve crosses the energy of the homogeneous precession at a magnon wave vector \( \bar{q}_0(\psi_{\bar{q}}) \). This means that the homogenous FMR mode is degenerate with the \( \bar{q}_0(\psi_{\bar{q}}) \) mode and can be involved in two-magnon scattering. The magnitude of \( \bar{q}_0(\psi_{\bar{q}}) \) decreases with increasing \( \psi_{\bar{q}} \) and disappears for a critical angle between \( \bar{q} \) and the magnetization \( \vec{M} \). Therefore no degenerate modes are available for \( \psi_{\bar{q}} \) larger than \( \psi_{\text{max}} \) [57]:

\[ \psi_{\text{max}} = \arcsin \left( \frac{H_{\text{FMR}}}{H + 4\pi M_{\text{eff}}} \right)^{1/2}, \quad (2.75) \]

where \( H_{\text{FMR}} \) is the field at FMR. A more general theory which includes out-of-plane orientations of the magnetization can be found in [25] and [58] and shows that there are no degenerate modes when the film magnetization is oriented with an angle larger than \( \pi/4 \) from the film plane. Therefore the two magnon scattering mechanism is absent in the perpendicular FMR configuration, (see also Fig 2.4).

Two-magnon scattering formally enters the transverse rf susceptibility [57] as an additional energy term \( R \):

\[ \chi'' = \frac{M_S B_{\text{eff}}}{B_{\text{eff}} H - \left( \frac{\omega_{\text{f}}}{\gamma} \right)^2 + i(H + B_{\text{eff}})\alpha \frac{\omega}{\gamma} + [\Re R + i\Im R]}, \quad (2.76) \]

where \( B_{\text{eff}} = H + 4\pi M_{\text{eff}} \). The anisotropy terms are neglected for simplicity. The real part, \( \Re R \), leads to a shift in the FMR field and the imaginary part \( \Im R \) provides additional damping. Both contributions must satisfy the symmetries required by the magnetic defects. The static magnetic properties measured using FMR can depend upon frequency due to the \( \Re R \) term. For samples having a two magnon scattering
contribution the static and dynamic measurements do not, in principle, have to provide the same magnetic anisotropies; however, any differences are usually small especially at low microwave frequencies.

The strength of two-magnon scattering varies as the static magnetization changes its orientation with respect to the in-plane crystallographic axes and is given by the imaginary part of \( R \), \( \Im R \). The imaginary part of the two-magnon contribution to the susceptibility is proportional to the square of the Fourier components that describe the inhomogeneous magnetic scattering potential:

\[
\Im R \sim 2 \int_{-\psi_{\text{max}}}^{\psi_{\text{max}}} I(q_0, \psi_q, \varphi_M) \frac{q_0 d\psi_q}{dq} (q_0, \psi_q), \tag{2.77}
\]

where \( \psi_q \) is the angle between the spin-wave wave vector \( \vec{q} \) and the magnetization vector, \( \varphi_M \) is the angle between the dc magnetization and the [100] crystallographic axis, and \( I(q_0, \psi_q, \varphi_M) \) is the square of the effective two-magnon scattering potential.

The two magnon scattering linewidth in the films affected by a two dimensional network of misfit dislocations [58, 25] can be well described by taking \( I(q_0, \psi_q, \varphi_M) \) to
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have the following form:

\[ I(q_0, \psi_q, \varphi_M) \sim \cos^2(2\varphi_M)\cos^4(2\varphi_q), \]  \hspace{1cm} (2.78)

The first term in Eq. 2.78 represents the uniaxial character of the magnetic defects caused by the misfit dislocation network and the second term represents the additional lattice defect symmetry. The misfit dislocation network in Fe/Pd(001) systems is oriented along the \(<100>_<Fe>\) crystallographic directions of Fe(001). The glide planes of the misfit dislocations create local uniaxial fields. The local uniaxial anisotropy contribution does not shift the FMR field when the magnetization is oriented 45° away from its axis and therefore this two-magnon scattering is absent for the magnetization oriented parallel to the \(<110>_Fe\) directions, see eq 2.78. This means that the two-magnon scattering intensity can have an explicit dependence on the direction of the magnetization and can exhibit a strong anisotropic contribution. An anisotropic extrinsic damping (measured along the \([100]\) and \([110]\) axes) has been observed for FeV(001) superlattices by Lindner et al. [59] and by Woltersdorf et al. [25] using GaAs/Fe/Pd/Fe(O01) structures. Linder et al. used the Arrias-Mills theory to interpret their results. The Arrias-Mills theory predicts an isotropic extrinsic contribution to two magnon scattering. In order to explain their angular \(\Delta H\) variations, they postulated an additional angular term \(\cos^2(2\varphi_M)\) without any explanation. A more general analysis of two magnon scattering in GaAs/Fe/Pd/Fe(O01) systems was carried out by G. Woltersdorf and B. Heinrich [25, 58].

2.4.4 Eddy currents

In metallic films the magnetic damping can be affected by eddy currents and can be considered an intrinsic contribution to the material’s property. Eddy current damping starts to play a role when the ferromagnetic layer has greater than a certain thickness. An important parameter in metallic films is the skin depth

\[ \delta = \frac{c}{\sqrt{2\pi \sigma \omega}} \]  \hspace{1cm} (2.79)

where \(c\) is the velocity of light and \(\sigma\) is the conductivity of the material in CGS/esu units. Assuming a full penetration of the rf magnetic field across the sample thickness and a circularly polarized precession the effective eddy current damping (in Gauss’s units) can
be expressed as [24]:

\[ \alpha_{eddy} = M_S \gamma \left( \frac{4\pi}{c} \right) 3\sigma d^2, \]  

where \( d \) is the film thickness. For \( d = 70 \) nm the damping contribution to the damping parameter is \( \alpha_{eddy} \approx 0.002 \). This value is comparable to the spin-orbit contribution to the intrinsic damping parameter \( \alpha \) in Fe. One can notice also from Eq. 2.80 that \( \alpha \sim d^2 \) and therefore decreases rapidly with the film thickness. For Fe films such that \( d < 10 \) nm (70 atomic layers for Fe) \( \alpha_{eddy} \) is negligible and can be ignored.

### 2.5 Mössbauer spectroscopy

In Mössbauer spectroscopy studies one needs to use a nuclide which exhibits the Mössbauer effect. The most common isotope used is \(^{57}\)Fe. This enables Mössbauer spectroscopy to be used for magnetic studies of ferromagnetic Fe films. By depositing a small amount of \(^{57}\)Fe inside the Fe film or at the surface interface one can obtain valuable information about the chemical and magnetic properties of ultrathin Fe structures. In this section the parameters that are obtainable from Mössbauer spectroscopy will be described. The theory of the Mössbauer spectroscopy used in this chapter is primarily based on the book edited by U. Gonser [60].

#### 2.5.1 Mössbauer parameters

The hyperfine interaction consist of interactions between a nuclear moment at an atomic site and appropriate electronic states. One can write a Hamiltonian which contains a number of terms representing the different kinds of interactions between the nuclear moment and lattice electric and magnetic fields.

\[ \mathcal{H} = \mathcal{H}(e0) + \mathcal{H}(m1) + \mathcal{H}(e2) + \ldots \]  

The first term stands for the Coulomb interaction between the nucleus and the electrons at the nuclear site. The second term refers to the coupling between the nuclear magnetic moment and the effective magnetic field at the site of the nucleus. The third term expresses the interaction of the electric quadrupole moment with the components of the diagonalized electric field gradient tensor (EFG) at the nucleus. In Mössbauer
spectroscopy only these three kinds of interaction have to be taken into account. Interactions of higher orders can be neglected since their energies are several orders of magnitude smaller. \((e0)\) interactions shift the nuclear energy levels without altering their degeneracies giving rise to the so-called isomer shift (chemical shift). The \(m1\) and \(e2\) terms are responsible for the number of components and intensities of the Mössbauer line multiplet. The first two parameters in Eq. 2.81 yield chemical information. The magnetic hyperfine interaction \((m1)\) is very useful for analysis of metals, alloys and compounds. Usually the Mössbauer spectrum consists of all three of these contributions. Therefore the analysis of the spectra can be quite complicated. In the following paragraphs each interaction will be described separately. They are shown schematically in Fig. 2.5

Figure 2.5: Hyperfine splitting of the \(^{57}\text{Fe}\) Mössbauer nuclear energy levels. Schematic Mössbauer spectra are shown at the bottom of the figure. The excited state of Energy \(E_e\) in the Source decays to the ground state energy \(E_g\). This transition results in the emission of a 14.4 keV \(\gamma\) ray which is absorbed by the \(^{57}\text{Fe}\) Absorber. The magnetic dipole splitting is shown in the absence of the quadrupole perturbation \((\mathcal{H} \neq 0, \text{EFG} = 0)\).
Isomer shift.

The isomer shift arises from the fact that an atomic nucleus occupies a finite volume, and s-electrons have the ability to spend a fraction of the time at the nucleus site. The s-electron density at the Mössbauer nuclei in the source and in the absorber in most cases are different and vary due to different chemical environment in the absorber and source. The nuclear charge interacts electrostatically with the s-electron charge cloud. As a result the ground ($E_g$) and excited ($E_e$) states of the nuclear energy of the absorber ($E_A$) and source ($E_S$) are changed, See Fig. 2.5. In Mössbauer experiments the difference between the energies ($E_S$) and ($E_A$) results in an energy shift $\delta$ which can be expressed in the non-relativistic approximation by

$$\delta = C \frac{\delta R}{R} (\left| \psi_A(0) \right|^2 - \left| \psi_S(0) \right|^2),$$

(2.82)

where $C$ is a constant for a given isotope containing nuclear parameters [61] and $\delta R/R$ is the relative change of the nuclear radius between excited and ground states. The term in the brackets represents the difference in the electron density between absorber and source isotopes evaluated at the nucleus. Mössbauer spectroscopy is a relative method in which the source spectrum is related to the absorber spectrum by motion using the Doppler energy shift (see section 3.2.3). Therefore one of the two has to be known in order to determine the other. Usually one uses a standard source and looks at various absorbers for a given Mössbauer nucleus. In this case $\left| \psi_S(0) \right|^2 = \text{const}$ and the isomer shift is a linear function of the electron density $\left| \psi_A(0) \right|^2$ in the absorber. Also the ratio $\delta R/R$ is known (not to a great degree of accuracy) for many Mössbauer nuclides [62]. The shift can be positive or negative. For instance $\delta R$ has a negative sign for $^{57}\text{Fe}$ indicating that the nuclear radius of the excited state is smaller than the radius of the ground state. From Eq. 2.82 a positive isomer shift implies a decrease of the electron density at the nucleus. The parameter $\delta$ can be obtained for different Mössbauer isotopes from Mössbauer effect database resources [63].

Quadrupole splitting

Discussion of the Coulomb interaction assumed that the nuclear charge has a uniform distribution of spherical symmetry. In many cases the nuclear charge distribution deviates from spherical symmetry. Furthermore the deviation may be different for each state
of excitation. The electric quadrupole moment \( eQ \) is the measure of this deviation from spherical symmetry. It is a tensor quantity

\[
Q_{ij} = \int \varrho_n(r) x_i x_j dV ,
\]

where \( \varrho_n(r) \) is the nuclear charge density at a point \( r \) and \( x_i, x_j \) are the cartesian coordinates corresponding to \( r \). The sign of \( Q \) indicates the shape of the distorted nucleus. \( Q \) is negative for a flattened nucleus and positive for an elongated one. Any nuclear state that possesses a non zero quadrupole moment can interact with an inhomogenous electric field gradient (EFG) at the nucleus. The EFG is given by following expression:

\[
\text{EFG} = \nabla \vec{E} = -\nabla^2 V = - \begin{bmatrix} V_{xx} & V_{xy} & V_{xz} \\ V_{yx} & V_{yy} & V_{yz} \\ V_{zx} & V_{zy} & V_{zz} \end{bmatrix},
\]

where \( V_{ij} = \frac{\partial^2 V}{\partial x_i \partial x_j} \), \( i, j = x, y, z \). The quadrupole interaction may be expressed by a Hamiltonian which, in general, has the form

\[
\mathcal{H}(\varepsilon 2) = \hat{Q} \cdot (\nabla \vec{E})
\]

\( \hat{Q} \) denotes the quadrupole moment operator, and \( \nabla \vec{E} \) is the EFG tensor operator. \( Q \) is constant for a given Mössbauer nuclide in different compounds (\( Q = 0.21 \times 10^{-24} \text{cm} \) for \( ^{57}\text{Fe} \) in the excited state, \( I = 3/2 \)). Therefore changes in the observed energy can arise only from changes in the EFG. In order to understand differences in the quadrupole splitting one has to know how EFG varies with different chemical and physical environments: this subject lies outside the scope of this thesis. A detailed discussion can be found in [60]. It is important for further discussion to introduce a few properties of the EFG. The EFG tensor can be described by only two independent parameters: (1) \( V_{zz} = \frac{\partial^2 V}{\partial z^2} \) where \( V_{zz} \) is usually called the principal components of the digonalized EFG. One can show that the EFG can be defined in a coordinate system where the off-diagonal components vanish and the diagonal components are ordered in such a manner that \( V_{zz} \) has the largest contribution; (2) The asymmetry parameter which is defined by \( \eta = (V_{xx} - V_{yy}/V_{zz}) \) and is restricted to \( 0 \leq \eta \leq 1 \). In the special case when \( \eta = 0 \) the EFG is called axially symmetric. There are two fundamental sources which can contribute to the total EFG and therefore that are responsible for observable quadrupole splitting in Mössbauer spectra:
• **ligand/lattice contribution** - charges on distant atoms or ions surrounding the Mössbauer atom in non-cubic symmetry;

• **valence electron contribution** - non-cubic symmetry of the electron distribution in partially filled valence orbitals of the Mössbauer atom.

The eigenvalues of Eq. 2.85 are

\[
E_Q = \frac{eQV_{zz}}{4I(2I-1)} \left[ 3m_I^2 - I(I+1) \right] (1 + \eta^2/3)^{1/2},
\]

where \(m_I = I, I-1, ..., -I\) are the nuclear magnetic spin quantum numbers corresponding to the nuclear spin value \(I\). For \(^{57}\text{Fe}\) \(I = 3/2\) in the excited state and \(I = 1/2\) in the ground state. The nuclear ground state is not split, because \(Q = 0\). The excited state is split into two doubly degenerate sub-states \(|3/2, \pm 3/2\rangle\) and \(|3/2, \pm 1/2\rangle\). The splitting of the lines (with respect to the center line) are equal for both sub-states and therefore the center is not shifted (see Fig. 2.5). Since \(\gamma\) transitions are possible between the ground state and both substates one can observe two resonance lines of equal intensities. The separation between these lines (\(\Delta\)) is called the *quadrupole splitting* and corresponds exactly to the energy splitting

\[
\Delta E_Q = \frac{eQV_{zz}}{2}.
\]

**Magnetic Dipole interaction; Hyperfine splitting**

A nucleus with spin \(I > 0\) possesses a magnetic dipole moment \(\vec{\mu}\) that can interact with a magnetic field \(\vec{H}\) at the nucleus. This interaction can be described by the Hamiltonian

\[
\mathcal{H}(m_I) = -\vec{\mu} \cdot \vec{H} = -g_N\beta_N\vec{I} \cdot \vec{H},
\]

where \(g_N\) is the Landé splitting factor and \(\beta_N = e\hbar/2Mc\) is the nuclear Bohr magneton. \(M\) is the mass of nucleus. The eigenvalues of \(\mathcal{H}(m_I)\) using first-order perturbation theory are found to be

\[
E_m(m_I) = \frac{-\mu H m_I}{I} = -g_N\beta_N H m_I.
\]

Equation 2.89 shows that the magnetic dipole interaction splits the nuclear state into \((2I + 1)\) equally spaced sub-states characterized by the \(m_I\) (nuclear Zeeman effect). For \(^{57}\text{Fe}\) with \(I = 1/2\) ground state and \(I = 3/2\) excited state, each of these states has
a magnetic dipole moment and therefore will be split by the hyperfine interaction. \(\gamma\) transitions between the sublevels of the ground state and those of the excited states are subject to selection rules; only transitions with \(\Delta l = 1, \Delta m = 0, \pm 1\) are allowed. This gives one six equally spaced spectral lines as shown in Fig 2.5. The relative line intensities depend on the angle \((\theta_m)\) between the direction of the hyperfine magnetic field at the nucleus and the propagation direction of the \(\gamma\)-rays. For \(\theta_m = 0^\circ\) and \(\theta_m = 90^\circ\) the relative line intensities are 3:0:1:0:3 and 3:4:1:1:4:3, respectively. For randomly oriented hyperfine fields the distribution of radiation is isotropic and integration over all directions results in the relative intensities 3:2:1:1:2:3. The integers are given by the squares of the Clebsh-Gordan coefficients. From the relative intensities one can deduce right away the magnetization direction from the propagation direction of the \(\gamma\)-rays (usually normal to the plane of the sample). Since the Coulomb interaction (isomer shift) is always present one expects that the centroid of the resonance sextet may be shifted from the zero position on the velocity scale in the Mössbauer spectrum. Furthermore, the nuclear state may be simultaneously perturbed both by the magnetic dipole and the electric quadrupole interactions. In that case the sublevels of the \(I = 3/2\) state of \(^{57}\text{Fe}\) will no longer be equally spaced. Assuming that \(\mathcal{H}(e2) \ll \mathcal{H}(m1)\), the quadrupole coupling may be treated as a first-order perturbation of the nuclear Zeeman effect. Depending upon whether \(V_{zz} > 0\) or \(V_{zz} < 0\) the sublevels \(|+3/2\rangle\) and \(|-3/2\rangle\) will be raised (lowered), and the sublevels \(|+1/2\rangle\) and \(|-1/2\rangle\) of the excited state will be lowered (raised) by the energy \(E_Q(\pm m_I) = \Delta/2\). This opens the possibility of determining the quadrupole coupling and hence the sign of the EFG. If the EFG is axially symmetric and if its principal axis makes an angle \(\phi\) with the direction of the magnetic field the general expression for the eigenvalues of the asymmetric Zeeman effect is given by

\[
E = -g_N\beta_N H m_I + (-1)^{m_I+1/2} \left( \frac{eQV_{zz}}{8} \right) \left( 3\cos^2\phi - 1 \right) .
\]

(2.90)

2.5.2 Mössbauer spectrum analysis

The quadrupole interaction is not encountered in measurements using bulk metallic Fe. An example of such a spectrum using a bcc-Fe foil having a hyperfine magnetic field of \(H_{hf} = 330\) kOe is shown in Fig 2.6. This spectrum is often used as a calibration spectrum for \(^{57}\text{Fe}\) Mössbauer spectroscopy. This spectrum simplifies the data analysis
Figure 2.6: Mössbauer transmission spectrum of an α-Fe foil used as a calibration sample. The energy splitting is determined by the α-Fe field magnitude, 330 kOE. The integers which label individual peaks in the spectrum refer to the transitions between sublevels of the ground and excited states, see Fig 2.5 of Mössbauer spectra in ultrathin Fe films.

Obtaining quantitative information on the magnetic states in the measured specimen requires detailed fitting of the measured Mössbauer spectrum line. Many fitting models have been developed over the years [64, 65]. Proper interpretation of the data is based on choosing the correct model in which the fitting parameters provide a good fit to the measured data.
Chapter 3

Experimental Methods

In this chapter the experimental systems which are relevant to the work presented in this thesis will be described. Ultrathin magnetic multilayers were prepared in an ultra-high vacuum (UHV) system which was equipped with tools to monitor the sample preparation process: Auger electron spectroscopy (AES), X-ray photoemission spectroscopy (XPS), reflection high energy electron diffraction (RHEED), scanning tunneling microscope (STM) and atomic force microscope (AFM). The magnetic properties were studied by means of ferromagnetic resonance (FMR), magneto-optical Kerr effect (MOKE) systems and Mössbauer spectroscopy.

3.1 Ultra High Vacuum system

The highest quality crystalline films were produced by means of molecular beam epitaxy (MBE). Epitaxy refers to the growth of one film on top of either a template or another film. The word epitaxy comes from Greek words epi, meaning "on" and taxis, meaning arrangement. MBE is essentially an evaporation technique under ultrahigh vacuum conditions (with pressures $\sim 10^{-10}$ Torr) allowing one to create uncontaminated multilayer film structures having controllable interfaces and lattice structures. One can create highly ordered crystalline film structures where the individual layer thickness can range from a few atomic layers to hundreds of nm. Such low pressures ensure that the atoms produced from thermal sources are ballistically deposited onto a crystalline substrate while maintaining a high purity of the deposited materials. This makes MBE particularly suitable for growing ultrathin film structures. MBE atomic sources require a special
construction which, in the case of 3d transition metals, limits the growth rate to 2-10 atomic layers of material per minute.

### 3.1.1 MBE system configuration

Epitaxial growth of ultrathin film structures at the Surface Science Laboratory, Simon Fraser University, was carried out using the MBE system shown in Fig. 3.1. The system consists of several interconnected ultrahigh vacuum chambers: the intro chamber, analysis chamber, intermediate chamber, and the growth chamber. Each chamber is constructed of stainless steel with separate pumping units (see below) isolated by gate valves. The pumping arrangement with detailed description of the types of pumps used is described in Appendix A. All UHV preparatory and characterization facilities (e.g., sputtering, AES, XPS, RHEED) are housed in these chambers. The main arm with the sample holder at the end allows one to move the sample along the MBE system and put it in place for the required operations. Additionally the sample holder includes a high precision goniometer for polar and azimuthal angle adjustments. The sample therefore can be oriented at arbitrary angles in all of the chambers. The temperature of the sample can be varied from $-180^\circ$C to $+700^\circ$C. All system components are able to withstand a bake out temperature at $200^\circ$C.

### 3.1.2 Electron spectroscopy

Auger electron spectroscopy (AES) and x-ray photoemission spectroscopy (XPS) allow one to measure the elemental and chemical composition of the deposited samples. In these techniques either an incident primary electron beam (AES) or an incident x-ray beam (XPS) results in emission of electrons from the measured specimen. Fig. 3.2 shows a schematic energy level diagram which is valid for a conducting sample (left-hand side). The energy of the electrons is measured from the Fermi level which is assumed to be zero. In XPS and AES the binding energies are measured with respect to the Fermi level of the electron energy analyzer [66].

Figure 3.2 shows the sequence of events following an ionization of the core level K. The incident electron energy $E_p$ has to be greater than the binding energy $E_K$. Subsequently the hole in the K level can be filled via a transition from the outer level $L_1$ as shown
Figure 3.1: A 3D realistic rendering of the MBE system at the Surface Physics Laboratory, Simon Fraser University
Figure 3.2: A schematic diagram of XPS and Auger processes. The x-ray photon gives rise to an emission of a photo-electron with the kinetic energy $KE_{XPS}$ shown in the left diagram. In the AES process the incident electron creates a hole in the core level $K$ by ionization. The hole in the $K$ shell is filled by an electron from the $L_1$ level, releasing the energy $(E_K - E_{L_1})$ which can be given up to another electron (the Auger electron). In this example the initial energy shell of the Auger electron is in the $L_{2,3}$ levels. The kinetic energy of the ejected Auger electron is then given by $(E_K - E_{L_1} - E_{L_{2,3}})$. The electron analyzer is shown on the right hand side. In both XPS and AES the binding energies are measured with respect to the Fermi level of the electron analyzer.

In this example. The energy difference $(E_K - E_{L_1})$ becomes available and can be used by the ionized atom in two ways. It can appear as a characteristic X-ray photon at that energy (X-ray fluorescence) or it can be given to another electron whereupon the second electron is ejected (Auger emission). The energy of the ejected Auger electron in this example is

$$KE_{KL_{2,3}} = E_K - E_{L_1} - E_{L_{2,3}} - \phi_A,$$  \hspace{1cm} (3.1)

where the subscripts denote the binding energies (BE) in the solid. One can see that the Auger energy expressed by equation 3.1 is a function only of the atomic energy levels so that for each element from the Periodic table there is a unique set of Auger energies.
allowing one to quickly identify the elemental composition of the sample. In AES the initial ionization of the core level is achieved either by means of high energy electrons from the primary electron beam $E_p$ or by means of an X-ray beam. Optimally $E_p$ should be greater than $5 \times E_K$. In our case we used a primary energy of $E_p=3 \text{ keV}$.

The XPS technique is based on the photo-electric effect discovered by Hertz in 1887 in which an X-ray beam is used to excite the core level electrons into vacuum. The energy of such a photo-electron is given by

$$KE_{XPS} = h\nu - E_B - \phi_A,$$

where $KE$ is the kinetic energy of the photo electron, $h\nu$ is the energy of the exciting radiation, and $E_B$ is the binding energy (BE) in the solid. In our case we used Al and Mg targets for the generation of the X-ray radiation. For Al-K$_\alpha$ $h\nu = 1486.6\text{eV}$, and for Mg-K$_\alpha$ $h\nu = 1253.6\text{eV}$. The advantage of having two different energies for the primary X-ray beams becomes transparent when comparing equations 3.1 and 3.2. The (KE) of an Auger electron is independent of the energy of the exciting radiation. If however the x-ray source is changed from Mg-K$_\alpha$ to Al-K$_\alpha$ all photo-electron peaks increase in KE by 233eV while the Auger peaks remain unchanged. Since the Auger electron peaks are present in XPS a change in the target allows one to separate the Auger and XPS spectra. XPS can be used to identify different chemical states due to different chemical shifts of the XPS peak positions. The energy spectrum of emitted electrons was measured by means of a hemispherical capacitive analyzer (SCA) [67] equipped with a "Physical Electronics" (PHI) digital analyzer system (PHI 5400) which is suitable for both AES and XPS measurements with a maximum instrumental resolution of 0.85eV. The 100 – 1000 eV electrons analyzed in typical AES and XPS experiments have a mean free path ranging from 1 to 3nm [68]. The sensitivity of either technique allows one to detect a fraction of a single atomic layer. Additionally, using the UHV goniometer, the angle between the analyzer and the sample surface can be changed which further allows one to separate the bulk and surface contributions to the signal. The AES and XPS techniques were essential in determining the level of substrate cleanliness: the substrate was usually contaminated by various oxides and Carbon compounds. Sputtering using low energy Ar$^+$ ions and H atom cleaning was employed to prepare substrates clean of contaminants.
3.1.3 Reflection High Energy Diffraction (RHEED)

A standard tool mounted in the MBE system is a reflection high energy diffraction system, (RHEED). RHEED consists of an electron beam having an energy, typically of tens of keV, arranged to fall at a glancing incidence on the sample - the beam makes an angle of a few degrees with respect to the sample surface. The diffracted beams illuminate a phosphorescent screen and a CCD camera is typically used to record the diffraction pattern. Due to the shallow angle of incidence very little momentum perpendicular to the surface is available and hence the primary electron beam interacts mainly with atoms confined to the sample surface. The small wavelengths of the electrons (10 keV-0.122Å in our system) is typically an order of magnitude smaller than the atomic spacing in solids, and they are therefore sensitive to the atomic arrangements of the surface atoms. In order to understand RHEED diffraction patterns it is useful to use the Ewald sphere construction, see Fig. 3.3. The tip of the incident $k_i$ wave vector is drawn at the origin of the reciprocal space of a surface lattice mesh. A rotation of the incident wave vector about the origin defines the Ewald sphere and gives all possible scattered wave vectors satisfying the conservation of energy. The reciprocal space rods are the consequence of limiting the electron diffraction only to surface atoms. The wave vectors $k_f$ of the diffracted electron beams are determined by the intersections of the reciprocal lattice rods with the Ewald sphere. The difference between the diffracted and incoming wave vectors $K = k_f - k_i$ is given by the in-plane reciprocal lattice vector $G$. The electron energy, 10 keV, is chosen such that the Ewald sphere is large enough to intercept several reciprocal lattice rods. The interpretation of the diffraction images is a difficult task and is complicated by surface roughness and inelastic scattering which leads to Kikuchi patterns. One usually limits the RHEED analysis to a simple kinematic interpretation which is sufficient to determine the in-plane lattice and the degree of surface roughness. The interpretation of absolute intensities is usually avoided because multiple electron scattering processes are involved in RHEED. Generally, the more perfect the surface the sharper are the intensity maxima and the closer they fall along Ewald's circle. Surface roughness tends to dim and broaden the diffraction maxima to produce diffraction streaks. A simple kinematic theory allows one to identify the main features of the surface distribution of atoms [69], see also a discussion of RHEED images in the text.

A substantial body of literature exists concerning RHEED. Theoretical treatments
Figure 3.3: The Ewald sphere construction and the RHEED diffraction geometry. The radius of the Ewald sphere is equal to the magnitude of the incident wave vector $k_i$. Primary electron beam impinges the sample surface with the angle $\theta$. The reciprocal lattice consists of rods with in-plane reciprocal wavewectors $G$ given by the two dimensional mesh of surface atoms. (a)- top view. (b)- 3D perspective view. The intersection of the reciprocal space rods with Ewald’s sphere specify the diffraction beams. The diffraction vectors, $k_f$, satisfy the surface lattice symmetry and energy conservation. The intersection of the diffracted beams with a fluorescent screen provides a diffraction pattern. For an ideal 2D surface the diffraction spots occur on the so-called Laue’s circle of radius $L_n$ which is centered at the point C of the fluorescent screen. The specular reflection $S$ is given by the intersection of the Ewald sphere with the (00) rod. Notice that the lower half of the luminescent screen is shadowed by the sample.
have reached an advanced degree of sophistication, and many aspects of RHEED have
been studied extensively and in great detail, however their complexity places them out-
side the scope of this thesis. For an extensive review of experimental and theoretical
RHEED methods, see Ref. [70].

3.1.4 Scanning Tunneling Microscope (STM)

The kinematic interpretation of RHEED comes about by comparing Fourier transforms
of a model distribution of the surface atoms with the observed RHEED patterns. This,
obviously, only provides limited information. Scanning Tunneling Microscopy (STM)
provides the real space surface topology. STM is based on a tunneling process. The
tunneling current can be calculated using a simplified picture in which the vacuum region
acts as a barrier between two metallic electrodes. These electrodes are the tunneling tip
and the nearest surface atoms. When the sample is biased by a negative voltage \((-V)\), its
Fermi level is raised with respect to the Fermi energy of the tunnelling tip. The electrons
will flow out of the filled states of the surface atoms, into empty states of the tip. This
situation is illustrated in Fig 3.4. Assuming a one dimensional rectangular vacuum
barrier and applying the WKB (WentzelKramersBrillouin) approximation method [71]
to the Schrödinger equation the calculated transmission probability through a barrier is
given by:

\[ |T|^2 = e^{-2\gamma}, \]  

(3.3)

where

\[ \gamma = \int_0^s \frac{2m\varphi}{\hbar^2} \, dx = \frac{s}{\hbar} \sqrt{2m\varphi}, \]  

(3.4)

where \( m \) is the mass of the electron, \( s \) is the width of the barrier (tip-sample separation),
and \( \varphi \) is the height of the barrier, which is given by the work functions of the tip and
sample. The tunnelling barrier can be measured by recording the tunnelling current as a
function of the tip-sample separation which decays exponentially with the barrier width
\( s \) as

\[ I_T \propto e^{-2s/\hbar\sqrt{2m\varphi}}, \]  

(3.5)

The tunneling current can decrease nearly an order of magnitude for a 1Å increase in the
separation between the tip and the sample surface. In practice the tip-sample separation
is usually maintained at $\sim 1\text{Å}$. Such a small separation requires very precise control of the tip position and limits vibrations to a scale less than one Ångstrom. The basic idea behind STM is illustrated in Fig. 3.5. A sharp metal tip is brought close enough to the sample surface so that electrons can tunnel through the vacuum barrier. The position of the tip is controlled accurately by means of piezoelectric drives in all three dimensions. The tip is scanned in a lateral direction while the feedback circuit constantly adjusts the tip height to keep the current constant. A constant current mode of operation allows one to image the surface topography. STM images were obtained at Simon Fraser University using an in-situ UHV Omicron system - LS SPM (Large Sample Scanning Probe Microscope) [72]. Images were also obtained during my stay in the Max-Planck Institute in Halle, Germany, using a similar Omicron STM system. For high resolution scanning probe microscopy a high quality vibration decoupling system is essential. The SFU Omicron SPM scanner is suspended on soft springs. The resonance frequency of the spring system is 2 Hz. Vibrations in this suspension system are suppressed using a non-periodic eddy current damping mechanism. The microscope offers a wide scan range.
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Figure 3.5: A schematic cartoon showing the principle of the operation of a Scanning tunnelling microscope (STM). A sharp metal tip is brought close enough to the sample surface so that electrons can tunnel through the vacuum barrier. The position of the tip is controlled accurately by means of piezoelectric drives in all three dimensions. The tip is scanned in lateral directions while the feedback circuit constantly adjusts the tip height to keep the current constant: this allows one to image the surface topography. The tunnelling current can be adjusted by varying the voltage difference between the tip and sample. For a given tunnelling current the lower the voltage the closer is the tip to the sample surface.

with coarse tip positioning within an area of 10 x 10 mm, and variable step sizes ranging from 20-200 nm. In order to achieve an atomic resolution the scanning tip has also to be of superior quality. Generally, the sharper the tip, the higher the resolution one can achieve down to atomic scales. Most common are tungsten tips (used at MPI, Halle) which are very robust but must be sputtered followed by in-situ annealing in order to remove an oxidation layer from the tip. Our SFU STM system did not have this option, therefore PtIr tips provided by the Agilent company were used. Chemically etched PtIr tips do not oxidize and one can use them right away after inserting them into the STM. In order to achieve atomic resolution some tip pre-conditioning is required and a great deal of patience. One is able to sharpen the tip in vacuum either by tapping the surface or by gently crashing the tip into the metal surface. Prepared in this way PtIr tips
3.1.5 Metallic film growth techniques

Metallic films were grown by means of molecular beam epitaxy (MBE) directly on GaAs(001). A detailed preparation and structural analysis of the GaAs(001) substrate will be presented in the next chapter, section 4.1. Two techniques of film deposition were used during the course of this thesis: a thermal deposition technique (TD) at Simon Fraser University and a pulsed laser deposition method (PLD) at the Max Planck Institute in Halle. In all samples the first layer grown on GaAs(001) was Iron. Body-centered cubic (bcc) Fe grows on GaAs(001) as a single crystalline film [73], [74] with a lattice spacing \(a_{\text{Fe}}^{\text{bcc}} = 2.87\) which is mismatched by only -1.4 % with half the lattice parameter of GaAs(001). An epitaxial Fe(001) seed layer served as a template for other epitaxially grown layers: Au(001), Ag(001), Pd(001). Both techniques were used under UHV conditions \((\sim 3 \times 10^{-10} \text{ Torr})\) with a usual growth rate of \(\sim 2\) monolayers (ML) per minute.
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Thermal deposition (TD) at SFU

Figure 3.7 shows the basic design of the evaporation sources used at SFU. A crucible filled with the desired material was placed inside a multilayer Ta radiation shield and heated resistively by means of a tantalum filament. All metal sources used an alumina crucible (Al₂O₃) except for Au, which used a pyrolytic boron nitride (PBN) crucible. PBN crucibles cannot be used for transition metals because they tend to react with PBN to form N₂ and nitrides when hot. All furnaces were surrounded by a liquid nitrogen cooled cryo shroud providing additional pumping due to the absorption of unwanted gases when the furnaces became hot during the film growth. Additionally the copper feed-troughs were water cooled. This furnace construction combined with a fast ion pump allowed one to achieve pressures of $3 \times 10^{-10}$ Torr during the deposition.

![Figure 3.7: The construction of the MBE thermal evaporation source used at SFU for the deposition of metals.](image)

Pulsed laser deposition (PLD) at MPI, Halle

Pulsed laser deposition (PLD), sometimes also called laser ablation, is another method for growing good crystalline metallic films and multilayers. A typical UHV system designed...
for both PLD and MBE methods of film growth is schematically shown in Figure 3.8. The PLD process involves focusing an intense laser pulse on a target thereby removing the target material in some volatile phases e.g. gas or plasma. A krypton fluoride (KrF) excimer laser \[75\] with Ne buffer was used at MPI-Halle. It produced light having a wavelength of 248 nm and a pulse energy of 325 mJ. The repetition rate of 5 Hz was controlled by a computer operated mechanical shutter accompanied by a simultaneous rotation of the target. The laser was focused on Fe and Fe\(^{57}\) targets of 99.99 \% purity resulting in a fluence of approximately 5 J/cm\(^2\). The GaAs(001) substrate was placed at a distance of 15 cm from the target.

### 3.1.6 RHEED oscillations

RHEED allows one to monitor the growth in real time. The RHEED oscillations can be observed during a layer-by-layer growth \[76\]. In this growth mode, also called Frank-van der Merve growth, one layer is essentially completed before additional material is added to the following layer. Figure 3.9 shows the specular spot intensity recorded as a function of time. This intensity oscillates with the film thickness. Each full period of the RHEED intensity oscillation corresponds to the formation of a new atomic layer. This is a consequence of a periodic change in the surface morphology as the impinging add-atom nucleate new atomic islands which eventually coalesce forming a new surface before the subsequent layer begins to grow. The strongest RHEED oscillations are observed if the electron beam interferes destructively (anti-Bragg condition) with the surface atoms of two sequential atomic layers. This means that the incident electrons reflected from the top layer and electrons reflected from the exposed second layer are out of phase. In most cases the growth is not ideal and the second layer nucleates prior to completion of the first layer. This type of growth is often referred to as a "quasi layer-by-layer" growth. For a good layer-by-layer growth one can observe many oscillations, see Fig. 3.9, which are eventually damped-out as the film thickens. The damping of the RHEED oscillations usually occurs on a timescale which is considerably longer than the RHEED oscillation period. This observation can be interpreted as an increase in the long-range roughness that approaches a steady state \[77\]. In the steady state case the atomic terraces are short enough that the deposited atoms reach the terrace step edges before nucleating an appreciable island formation on the existing terraces. The growth proceeds by lateral
Figure 3.8: A sketch of the MBE system equipped with a PLD apparatus used at MPI, Halle. The krypton fluoride (KrF) excimer laser [75] with Ne buffer produces light having a wavelength of 248 nm and a pulse energy of 325 mJ. The repetition rate of 5 Hz was controlled by a computer operated mechanical shutter accompanied by a simultaneous rotation of the target. The laser was focused on Fe and Fe$^{57}$ targets of 99.99 % purity resulting in an ablation of the Fe. The GaAs(001) substrate was located 15 cm from the target.
translation of the terrace distribution. This transition to step-flow growth happens when
the average terrace size becomes comparable to the mean nucleation distance.

Figure 3.9: RHEED specular spot intensity oscillations observed for Fe grown on
GaAs(001). Each oscillation corresponds to the deposition of the equivalent of one
atomics layer. The many visible oscillations indicates a smooth epitaxial quasi layer-by-
layer growth.

3.2 Measurement techniques

3.2.1 Ferromagnetic resonance

Ferromagnetic resonance (FMR) operates in the range of microwave frequencies. The
FMR signal is measured by monitoring microwave losses in the studied sample as a
function of the applied external dc magnetic field $H_0$. FMR is suitable for measure-
ments of the static magnetic properties (saturation magnetization and magnetocryst-
taline anisotropies), and dynamic magnetic properties (damping, electron g-factor) of
ultrathin film magnetic materials.
Microwave spectrometer

Figure 3.10 shows a block diagram of the FMR setup used in our laboratory. Microwave reflex klystrons were used for the generation of 9, 14 and 72 GHz microwaves, and Gunn diodes (Gunn oscillators) were used for operations at 24 and 36 GHz. Both sources provide excellent power stability and will be described separately below.

Reflex klystrons contain a reflector plate (repeller). An electron beam is modulated by passing it through an oscillating resonant cavity. The feedback required to maintain oscillations within the cavity is obtained by reversing the electron beam thereby sending it back through the cavity grid. The electrons in the beam are velocity-modulated before the beam passes through the cavity a second time and gives up the energy required to maintain oscillations in the microwave cavity. The electron beam is turned back by the
electric fields associated with a negatively charged repeller. Three power sources are required for reflex klystron operation: (1) Filament power. (2) A positive resonator voltage (beam voltage) used to accelerate the electrons through the cavity grid. (3) A negative repeller voltage that is used to reverse the direction of the electron beam.

Gunn diodes are an important solid-state source of microwaves operating in the range of millimeter-wavelengths. They exhibit a low FM (Frequency modulation) noise and a wide operating frequency range. Essentially a Gunn diode consists only of N-doped semiconductor material, whereas most diodes consist of both P and N-doped regions. In the Gunn diode, three spatial regions exist: two heavily N-doped regions that are connected to the terminals, with a thin layer of lightly doped material in between. When a voltage is applied to the device, the electrical gradient will be largest across the thin middle layer. Eventually, this layer starts to conduct. In general, the current tends to rise with increasing voltage, but there is a region between the so-called peak voltage ($V_{th}$) and the valley voltage ($V_{valley}$), where the the current falls as the voltage is increased. For applied voltages less than $V_{th}$ the device acts as a passive resistance ($R = V/I$). However, for applied voltages greater than $V_{th}$ the electron velocity (current) decreases as the voltage increases. In practice, this means that a Gunn diode has a region of Negative Differential Resistance (NDR). The peak voltage, $V_{th}$ is often called the threshold voltage since it represents a threshold that must be reached in order to operate in the NDR region. This is the essential feature that leads to Gunn oscillations in an active device. The negative resistance region is due to the special conductance band structure of direct band gap semiconductors such as GaAs. A detailed description of how exactly the NDR phenomenon in GaAs results in Gunn-oscillations can be found in the article by J. B. Gunn, [78].

The negative differential resistance results in an RF relaxation oscillator when a suitable dc current flows through the device. Gallium arsenide Gunn diodes are available for frequencies up to 200 GHz. Gallium nitride Gunn diodes are available for frequencies up to 3 THz. Frequency tuning of the Gunn diode resonator can be accomplished either mechanically by inserting a tuning rod into the cavity, or by changing the Gunn oscillator frequency electrically using a varactor. The tuning rod should preferably be made from a low-loss dielectric such as sapphire having a dielectric constant $k = 9.3$. A varactor diode uses a p-n junction in reverse bias and the capacitance of the diode varies with the reverse voltage. A DC control voltage varies the capacitance of the varactor and thereby
changes the Gunn diode frequency so that the microwave frequency can be tuned to
the cavity resonance frequency. Our Gunn diodes (ZAX Millimeter Wave Corporation)
operating at frequencies of 24 and 36 GHz are equipped with both tuning methods and
provide a maximum tuning range of ±60 MHz. A recommended heat sink was used
in order to achieve a frequency and a power thermal stability of 1.5 MHz/°C and 0.01
dB/°C, respectively.

In FMR experiments the microwave absorption signal was measured by monitoring
the microwave losses in the films under investigation. When the magnetic sample
undergoes FMR the microwave losses are increased and the amplitude of the reflected
microwave electric field changes and this change is detected by means of a microwave
detector. The microwave frequency was kept constant while the external magnetic field
was swept over a suitable range. The magnetic field was measured using a Hall probe
which was calibrated using the NMR of protons in H₂O. The sample was usually in­
serted into a microwave cavity which significantly increases the rf driving field on the
sample. Since the quality factor (Q) of the resonant cavity is high (a Q of the order of
a few thousands) it was possible in our setup to detect an FMR signal from ultrathin
films as thin as 5 atomic layers. The microwave cavity was placed between the pole
pieces of a Varian 3800 electromagnet capable of producing magnetic field up to 29 kOe,
see Fig. 3.10. Most of our microwave cavities were of a cylindrical shape operating in
the “doughnut” TE₀₁₁ mode as shown in Fig. 3.11. This mode is particularily suitable
because it has a fairly high Q factor [79]. There is zero current flowing between the
end plates and the cavity cylindrical body. This enables one to use a tuning piston-like
plunger without affecting the Q factor of the cavity. In addition, a poor electrical contact
(gap) between the plunger and the cylindrical wall supresses the TMₘₙ mode which is
degenerate with the TE₀₁₁ mode. The microwave cavity is coupled to the spectrometer
waveguide through a hole in its upper end wall which is located approximately half way
between the cavity axis and a cavity side wall, see Fig 3.11. In order to keep the the
cavity tuned during the FMR measurements the microwave signal is slightly frequency
modulated at 70kHz. The 70 kHz error signal from the microwave diode was rectified
by a lock-in amplifier and the resulting dc output voltage was used as a feed back signal
for the microwave power control unit to shift the frequency slightly in order to keep
the cavity on resonance. In that case the reflected microwave amplitude at the detector
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Figure 3.11: A schematic drawing of a cylindrical cavity and samples. The $TE_{01}$ cylindrical cavity resonator is coupled via a hole in the end plate to the rectangular waveguide operating in the $TE_{10}$ mode. The force lines of the rf magnetic field inside the cavity are shown by the dashed lines. For an in-plane DC magnetic field the sample was mounted on the end plate in a position symmetrical with the coupling orifice. For a sample mounted on the cylindrical wall the DC magnetic field, and hence the magnetization, can be tilted out of the film plane allowing one to carry out FMR measurements in the out of plane configuration (including the perpendicular case). The $H$ in the right picture displays the external field direction, which can be rotated in the plane of the cavity base.

changes linearly with the absorbed power in the sample. The average absorbed power $P$ per unit volume is given by

$$ P = \frac{1}{2} \chi'' \omega h_{rf}^2 $$

(3.6)

where $\chi''$ is the imaginary part of the rf transversal susceptibility (see also Eq. 2.33).

In order to enhance the FMR signal to noise ratio the external dc field was given a small 100 Hz amplitude using a separate set of coils mounted on the magnet pole pieces. Finally, the FMR signal was detected at 100 Hz by means of another lock-in amplifier whose output signal was proportional to the magnetic field derivative of the absorption
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signal.

Typical FMR data

A typical FMR spectrum is shown in Fig. 3.12 (a). It contains two important pieces of information: The line position $H_{\text{res}}$ and the linewidth $\Delta H$. The angular dependence of $H_{\text{res}}$ allows one to determine magnetic anisotropies from the effective magnetization, $4\pi M_{\text{eff}}$. The linewidth is related to the magnetic damping using the equation 2.36, and its angular and frequency dependence provide information about the damping mechanisms. $\Delta H$ was defined as the half linewidth measured at half maxima of $\chi''$ (HWHM). Therefore the peak-to-peak linewidth $\Delta H^{PP}$ obtained using the field modulation is related to $\Delta H$ for a purely Lorentzian lineshape by the following formula:

$$\Delta H = \frac{\sqrt{3}}{2} \Delta H^{PP} \quad (3.7)$$

$H_{\text{FMR}}$ and $\Delta H$ were extracted from the spectra by fitting the FMR data to the derivative of an Lorentzian function given by Eq. 3.8

$$F(H) = \cos(\theta) \frac{\partial}{\partial H} \Im\chi_\perp + \sin(\theta) \frac{\partial}{\partial H} \Re\chi_\perp \quad (3.8)$$

where the imaginary (absorption) and real (dispersion) parts of the derivatives of $\chi_\perp$ are as follows:

$$\frac{\partial}{\partial H} \Im\chi_\perp = \frac{A}{\Delta H} \left( \frac{-2x}{(1+x)^2} \right) \quad (3.9)$$

$$\frac{\partial}{\partial H} \Re\chi_\perp = \frac{A}{\Delta H} \left( \frac{1-x}{(1+x)^2} \right) \quad (3.10)$$

where

$$x = \frac{H - H_{\text{FMR}}}{\Delta H} \quad (3.11)$$

and $A$ is the intensity of the of the Lorentzian absorption peak. $A'$ is related to the intensity of the derivative of the absorption peak, $A'$ (defined as the difference between maximum and minimum values), by the following relation:

$$A' = \frac{3\sqrt{3}}{4} \frac{A}{\Delta H} \quad (3.12)$$
Figure 3.12: (a) A typical FMR spectrum of $20\text{Au}/16\text{Fe}/\text{GaAs}$ sample measured at 24 GHz. (b) The FMR spectra for a $20\text{Au}/12\text{Fe}/900\text{Ag}/16\text{Fe}/\text{GaAs}$ (thin solid line) and $20\text{Au}/12\text{Fe}/450\text{Ag}/16\text{Fe}/\text{GaAs}$ (thick solid line) samples at 24 GHz. The thin solid line shows the FMR spectrum acquired for a sample deposited on a semi-insulating GaAs(undoped) wafer. The resonance fields for the individual layers are well separated due to the strong in-plane uniaxial anisotropy of the 16Fe film grown on GaAs. The FMR signal at the lower field corresponds to the 16Fe bottom layer and the high field signal to the 12Fe top layer. The sample was symmetrically driven on both sides of the sample by a uniform rf field ($h_{rf}$) inside the cavity. The thick solid line shows the FMR signal for a similar specimen but grown on semi-conducting GaAs(Si-doped). Clearly the signal from the 16Fe layer grown on GaAs(Si-doped) is much smaller than that obtained from the 16Fe layer grown on the undoped GaAs. This can be explained by the skin effect in the GaAs(Si-doped) substrate. (c) A schematic sketch of the uniform $h_{rf}$ distribution inside the specimen grown on insulating GaAs and shown by the dotted line rectangle. (d) A schematic representation of the inhomogeneous rf driving field in the sample grown on GaAs(Si-doped). The skin depth of $\sigma \sim 16\mu\text{m}$ leads to the attenuation of $h_{rf}$ inside the 0.375 mm thick GaAs. Also the impedance mismatch between the metallic layers and the GaAs substrate leads to a triangular distribution of the rf field inside the sample: The field is strongest at the top of the 12Fe film and decays linearly to a small value at the 16Fe/semiconductor interface.
Another parameter which can be obtained from this fitting is the mixing angle $\theta$ between dispersive and absorptive components of $\chi_{L}$. A small mixing angle was always present in the FMR data. The lock-in amplifier would usually pick up a component of $\partial \Re \chi_{L} / \partial H$ no matter how carefully the phase angle was set up. This situation comes from the fact that the rf component driving the sample is affected by multiple reflections of the microwave power inside the GaAs substrate.

Figure 3.12(b) illustrates the situation in which the distribution and symmetry of the rf magnetic driving field inside the sample is strongly affected by the type of GaAs substrate used. The thin solid line shows an FMR spectrum acquired at 24 GHz on the $20\text{Au/12Fe/900Ag/16Fe/GaAs(semi-insulating)}$ sample. The sample was mounted in the parallel configuration (see Fig. 3.11). The resonance fields for the individual layers are well separated due to the strong in-plane uniaxial anisotropy of the 16Fe grown on GaAs, see Eq. 2.34. The dc magnetic field ($H_{dc}$) was applied along the $(110)_{Fe}$ (easy direction). In that case the FMR signal at the lower field corresponds to the 16Fe bottom layer and the high field signal corresponds to the 12Fe layer. Both measured signals were symmetric derivatives of Lorentzian lines and their amplitude ratio $A_{16Fe}/A_{12Fe} \approx 1.3$ was identical to the thicknesses ratio of those layers. This means that the sample was symmetrically driven inside the cavity by a uniform rf field ($h_{rf}$) which was the same at both sides of the sample. A schematic sketch of the uniform $h_{rf}$ distribution inside the film is shown in Fig. 3.12(c) by the dotted line rectangle.

The situation described above was very different when a similar sample $20\text{Au/12Fe/450Ag/16Fe/GaAs}$ was deposited on a Si-doped (semi conducting) GaAs wafer. The FMR spectrum for this sample is shown by the thick solid line in Fig. 3.12(b). Clearly the signal from the 16Fe layer grown on GaAs(Si-doped) is much smaller than the signal from the 16Fe layer grown on GaAs(undoped). At the same time the signal from the 12Fe layer had the same strength for both substrates. This can be explained by differences in the resistivities of the two GaAs wafers that strongly affects the rf skin depth of the GaAs. One can make an simple estimation using the formula for the skin depth,

$$\sigma = \sqrt{\frac{2\rho}{2\pi f \mu_r \mu_0}}$$  \hspace{1cm} (3.13)

where $\rho$ is the resistivity in units of Ohm-meters. For the semi-insulating GaAs wafer, $\rho = 3.6 \times 10^7$ Ohm-cm (value taken from the manufacturer data sheet). Since $\mu_r \approx 1$
this leads to the value of the skin depth, $\sigma \sim 1m$. This value is significantly larger than the thickness of the GaAs substrate, $d_{GaAs} = 0.375$ mm, hence the skin effect can be neglected. For the Si-doped GaAs wafer the resistivity, $\rho = 2.5 \times 10^{-3}$ Ohms-cm is several orders of magnitude lower than in the case of the undoped wafer which leads to a value $\sigma \sim 16\mu m$ which is smaller than the thickness of the wafer. In this case the skin effect has to be taken into account and this results in the suppression of the $h_{rf}$ field inside the wafer. The resulting rf field distribution is shown schematically by the dotted triangle in Fig. 3.12(d). Since the rf driving field cannot penetrate the GaAs, the impedance mismatch between the metallic layers and the GaAs substrate leads to a triangular distribution of the rf field inside the sample: the field is strongest at the top of the 12Fe film and decays linearly to a small value at the 16Fe semiconductor interface. The strength of the field is given by Maxwell’s equations and the boundary conditions required to satisfy the impedance mismatch. This situation is shown in Fig. 3.12(d). As a consequence, the driving field at the 16Fe film will be significantly decreased. Additionally, the conducting GaAs substrate perturbs the cavity mode and causes an asymmetry of the FMR signal from the 16Fe layer.

The skin effect in the metallic magnetic films studied in this thesis can be disregarded because they are very thin compared with typical metallic skin depths.

3.2.2 Time Resolved Magneto-Optical Kerr Effect (TRMOKE)

The magneto-optical effect provides an important tool to investigate static and dynamic magnetic properties [80]. The magneto-optical effect is based on the off diagonal elements of the dielectric permittivity. It can be described using an analogy with the extraordinary Hall effect. The role of the current is played by the electric field component of the light and the magnetization provides an effective B field. The theoretical background of the magneto-optical Kerr effect (MOKE) can be found in [80, 81]. Linearly polarized light reflected from a magnetic material changes its polarization vector. The incident light having either s or p linear polarization after reflection from a magnetic sample becomes elliptically polarized. The polarization component perpendicular to the incident polarization can be thus characterized by rotation $\theta$ and ellipticity $\delta$ components which are proportional to the magnetization of the material. Magneto-optical effects can be used in time-resolved studies by employing pulses of light as a probe. The experimental
pulsed method employs time-resolved Kerr microscopy and is capable of measuring all three components of the magnetization vector in the picosecond temporal regime and with sub-micrometer spatial resolution [82]. It is one of the leading techniques used in the investigation of low-dimensional magnetic materials. The following section gives a detailed description of the TRMOKE system. This technique requires fast (ps) magnetic pulses in which the repetition rate is under precise optical control.

Experimental setup.

Figure 3.13 illustrates a complete TRMOKE system. It includes an ultrafast femtosecond source of 70fs laser pulses, optics and electronics for controlling the magnetic pulse generation, and signal detection.

Signal detection

Figure 3.14 shows a realistic 3D rendering of the current TRMOKE setup used in Mark Freeman’s laboratory, the University of Alberta, Edmonton. The Ti:Saphire laser (1) was used as the primary light beam. It provided 70 fs pulses of 800 nm wavelength (near infra-red) with a repetition rate of 80 MHz. The laser beam was split into pump and probe beams using a 50/50 beam splitter (88). The linearly polarized probe beam having a polarization vector perpendicular to the plane of incidence (s-polarized) was next directed (3) toward the sample and focused using the microscope objective lens (OBJ). The spatial resolution of the beam incident on the sample was determined using the numerical aperture of the objective lens. One was able to achieve a resolution of 1μm. In order to minimize heating of the sample the average power of the probe beam was attenuated to ~1.1 mW using the attenuator (2). Polarization analysis of the light reflected from the sample was carried out using a Thompson polarization beam splitter (7) which splits the light into two orthogonal polarization states. It was set to 45° with respect to the incident polarization plane. Consequently the two split light beams having approximately equal intensities were sent to the two quadrant photodiodes (8)(see Fig. 3.13). The outputs from the photodiodes were differentially subtracted resulting in the desired signal that was detected by the lock-in amplifier. If there is no polarization rotation in the absence of the Kerr effect the split beams will be of equal intensities and the differential subtraction will result in a zero output signal which is shown schematically
Figure 3.13: A schematic diagram of a TRMOKE system. A Ti:Saphire laser was used as a light source that provided 70 fs pulses. Optical mirrors and a beam splitter were used to direct the pump and probe beams onto a photoswitch (PS) and a sample respectively. Polarization analysis of the reflected light from the sample was carried out using a Thompson polarizing beam splitter which splits the light into two orthogonal polarization states. The Kerr signal from the sample was detected by means of two quadrant photodiodes (QPD). Their outputs were differentially subtracted and the resulting signal was measured using synchronous modulation of the magnetic excitations and lock-in detection. Modulation of the magnetic excitation was accomplished by modulating the ps magnetic pulses with a square wave envelope at 2.4 kHz. Finally, the MOKE signal was detected by means of a lock-in amplifier operating at 2.4 kHz. A detailed description of particular components of the TRMOKE set-up can be found further in the text.
in Figure 3.15 by the solid black arrows. A small polarization rotation $\theta$ induced by the Kerr effect (dotted grey arrows in Fig. 3.15) leads to different intensities of the split beams. One increases in intensity and the other decreases in intensity. The net signal at the output of the photodiodes is proportional to the sample magnetic moment. If the optical powers are measured by the output signals from the two detectors, the PD1 and PD2 planes in Fig. 3.15, the difference of their outputs will be [83]:

$$I_{PD1} - I_{PD2} = I_0[(\cos^2 \theta - \sin^2 \theta)r_{ss}^2] + I_0[4 \cos \theta \sin \theta r_{ss} r_{sp} \cos \delta]$$  \hspace{1cm} (3.14)

where $I_0 = 0.5E_0^2$ is the optical power of the incident beam, $r_{ss}$ is the ordinary metallic reflection coefficient, $r_{sp}$ is the Kerr reflection coefficient (the reflected p-wave amplitude for a unit amplitude incident s-wave). $\theta$ and $\delta$ are the Kerr rotation and ellipticity respectively. The rotational component $\theta$ of the Kerr polarization vector is in-phase with the reference polarization and the elliptical component $\delta$ is in quadrature with the reference polarization ($\pi/2$ shifted from the reference polarization). Therefore the output from the detection diodes is insensitive to the elliptical polarization and one is able to detect only the rotation angle $\theta$. The phase of the Kerr polarization with respect to the reference polarization can be adjusted using a 1/4 wavelength plate (6). In fact, the signal detection using a Thompson prism works as lock-in amplifier where the reference signal is the reflected light which is unaffected by the Kerr effect.

Quantitative information is contained in the $r_{sp}$ term which is proportional to the magnetization. This differential approach gains a factor of two in signal over a system using a single analyzer and detector set up. The first term in Eq. 3.14 contains information about surface imperfections which can cause some depolarization by multiple reflections. This term may overwhelm the second term which contains the Kerr magnetic information. This contribution could be eliminated by using synchronous modulation of the magnetic excitations and lock-in detection. Modulated synchronous magnetic excitation was carried out by superposing a square wave envelope at 2.4 kHz on the ps magnetic pulses. Finally the MOKE signal was detected by means of a lock-in amplifier operating at 2.4 kHz.
Figure 3.14: Photo realistic rendering of the TRMOKE experimental setup used at the University of Alberta, Edmonton. The linearly polarized laser beam from the Ti:Sapphire laser (1) is split into a pump and a probe beam using the 50/50 beam splitter (BS). The probe beam is attenuated using the attenuator (2) and is focused on the sample using the microscope objective (OBJ). The polarization of the reflected light is analyzed by the Thompson beam splitter (7). The polarization rotation is turned into an intensity shift inside the Thompson polarizer and the intensities of the split beams are detected using the two quadrant photodiodes (8). The optical delay line is used in order to synchronize the probe beam with the magnetic pulses. The time difference in travel for the pump and probe beams is computer-controlled using mirrors mounted on the slider (delay line) (4). Magnetic pulses are generated by the photo switch (PS) triggered by the pump beam itself. Samples are mounted on a slotted transmission line which is attached to the piezo-driven stage (5): this enables scanning of the sample surface and focusing of the probe beam by moving the objective lens. The raster scanning with a resolution of $\sim 1\mu m$ allows for precise positioning of the probe beam on the transmission line. Other components include the permanent magnet (9), a 1/4 waveplate (6), and a beam splitter (3).
CHAPTER 3. EXPERIMENTAL METHODS

Figure 3.15: A schematic diagram of the coordinate system used to describe the reflected light measured by the two quadrant photodiodes (PD). The S axis is perpendicular to the plane of incidence while the P axis is in the plane of incidence. These S and P axes correspond to s- and p-polarized light, respectively. The Thompson beam splitter was set at 45° with respect to the incident polarization plane and splits the s-polarized light reflected from the sample into two orthogonal polarization components indicated by the solid arrows drawn along the PD1 and PD2 lines. In this case in the absence of a Kerr effect equal intensities of light are sent to the quadrant photodiodes PD1 and PD2. Differential subtraction results in a zero output signal. A polarization rotation $\theta$ induced by the Kerr effect (dotted grey arrows) will produce an intensity shift on the photodiodes such that the intensity at PD1 increases and the intensity at PD2 decreases. This results in an increase of the PD1 signal and a decrease in the PD2 signal and thus a resulting difference signal proportional to the specimen magnetization. The Kerr effect creates a small p-polarized light component along the P axis which is $\pi/2$ rotated (in space) with respect to the reference polarization which is directed along the S axis.

Magnetic pulse generation

The magnetic field pulse generation was achieved by using a reverse biased GaAs photoconductive switch (PS) connected to a microwave transmission line (slotted line). A cartoon illustrating this set-up is shown in Figure 3.16. The focused laser beam (pump beam) closes the photo-conductive (PS) switch since the laser photons have an energy greater than the bandgap energy of GaAs. Thus a laser pulse frees up carriers between two biased, metallized regions in the GaAs semiconductor substrate so that the switch becomes conducting. The current pulses triggered by the Ti:Saphire laser pulses (80
Figure 3.16: A cartoon illustrating the geometry used for TRMOKE. The waveguide (slotted line) is connected to the biased \( V_B \) GaAs photoconductive switch (PS). The focused laser pump beam closes the photo switch (PS) resulting in a current pulse which travels down the waveguide and discharges the capacitance of the slotted line to generate a magnetic field pulse having a rise time of \( \sim 10 \) ps and a decay time of \( \sim 100 \) ps. Magnetic field force lines are shown by the solid line circles. The magnetic sample was placed on the slotted line with the magnetic film facing the line. The probe beam passed through the glass used to support the transmission line and was used to monitor the time evolution of the film magnetization. The probe beam was incident normal to the film plane and was focused between the conductors of the transmission line, and monitored the out-of-plane component of the magnetization (polar MOKE). In the double layer structures the two ferromagnetic (FM) layers were separated by a thick nonmagnetic (NM) spacer. For a sufficiently thick NM spacer the probe beam was fully attenuated inside the NM and the direct MOKE signal from the bottom FM layer grown on GaAs could not be observed.
3.16. The measured signal therefore was sensitive to the polar Kerr effect. Transmission lines were fabricated using lithographic techniques. In order to obtain \( \sim 20 \) Oe strong magnetic field pulses transmission lines were used that had small gaps of 3 and 5 \( \mu m \) between the conductors. The external dc magnetic field was applied in the plane of the sample surface. TRMOKE is a stroboscopic method which requires that temporal excitations of the system are triggered synchronously with the probing pulses. In our case such a synchronization was achieved by using the laser to trigger both the magnetic pulses and the probe pulses that monitored the magnetization dynamics. The response to many repetitive equivalent excitation events (having the same time delay between the magnetic pulse and probe) were averaged out and represented a single time-averaged event.

3.2.3 Mössbauer conversion electron spectroscopy (CEMS)

Mössbauer spectroscopy allows one to undertake simultaneously bulk and interface magnetic and structural studies. It is a non-destructive technique which can be applied \textit{in situ}. The isotope \( ^{57}\text{Fe} \) exhibits strong Mössbauer spectra which makes this technique suitable for investigation of the structural and magnetic properties of Fe films and their interfaces. Moreover the deposition of a few atomic layers of \( ^{57}\text{Fe} \) together with a deposition of either regular Fe (with only 2\% of \( ^{57}\text{Fe} \)) or of \( ^{56}\text{Fe} \) (no Mössbauer signal) makes this technique suitable for a depth selective analysis. The power of Mössbauer spectroscopy in the study of magnetic films, interfaces, and superlattices is well described by Korecki [84]. The high sensitivity of Conversion Electron Mössbauer Spectroscopy (CEMS) is particularly suited for applications using only a few atomic layers of \( ^{57}\text{Fe} \). An excellent review of the Mössbauer effect and various applications of this technique can be found in [60].

**Principles of CEMS**

Mössbauer spectroscopy makes use of \( \gamma \)-ray emission from an excited nucleus (source) bound in the solid and subsequent resonant absorption of this \( \gamma \)-ray in another nucleus (target) by the reverse process. The Mössbauer discovery (Nobel prize in 1961) relies on the fact that a \( \gamma \)-ray having the energy \( E_{\gamma} \sim 5\text{-}150 \text{ keV} \) when leaving the source does not transfer momentum to the source or target nucleus. This implies that the recoil
momentum is taken up by the whole crystal lattice and therefore there is a negligible recoil energy loss from the nucleus itself. The same process occurs in the resonance absorption. Consequently the nuclear transition energy $E_0$ is equal to the energy of the radiation, $E_\gamma$.

The most commonly used source in Mössbauer studies is $^{57}$Co. The half-life time of $^{57}$Co is 270 days which decays to the excited state ($I = 5/2$) of $^{57}$Fe by electron capture (EC). This excited state decays next to the 14.4 keV excited state ($I = 3/2$) of $^{57}$Fe with the emission of a 122 keV $\gamma$-ray, Figure 3.17,a). The subsequent decay of the 14.4 keV state to the ground state ($I = 1/2$) provides the $\gamma$ photons used for Mössbauer spectroscopy in $^{57}$Fe. In order to tune the energy of the $\gamma$ radiation with

![Figure 3.17: (a) Decay scheme for a $^{57}$Co in the CoRh alloy used in Mössbauer spectroscopy. (b) The inner conversion process in $^{57}$Fe. The 14.4 keV excited state of $^{57}$Fe can de-excite also by other means than direct photon emission. It can transfer its energy to one of the inner shell electrons (typically an s-electron). This process is called "Internal Conversion". The probability of such an event in $^{57}$Fe is nine time larger than the probability of emission of a 14.4 keV photon. De-excitations by internal conversion results in the ejection of electrons having a kinetic energy of 7.3 keV.](image-url)

the resonance fluorescence spectrum one needs to be able to continuously adjust the energy of the incoming photons. Various method have been developed. The main idea is based on the linear Doppler effect. The $\gamma$-ray of energy, $E_\gamma = 14.4$ keV, emitted from a nucleus moving with a velocity $v$ (with respect to the sample and along the propagation direction of the $\gamma$-ray) is shifted in energy ($\Delta E_D$) by the linear Doppler effect:
The Doppler velocity needed to obtain a spectrum has to be on the order of the line width or on the order of the hyperfine splitting. In the case of a $^{57}$Fe source a velocity of 1 mm/s varies the 14.4 keV $\gamma$-ray energy by $\Delta E_D = 4.79 \times 10^{-8}$ eV. This energy shift is small compared with the $\gamma$-ray energy and therefore it is suitable for the observation of changes in the nuclear hyperfine energy structure. When the energy of an emitted $\gamma$-ray coincides with a transition in the sample nuclei, some of the gamma quanta are absorbed resonantly.

The line broadening caused by thermal vibrations of the emitting nucleus almost disappear since the first order Doppler broadening is proportional to the square root of the thermal energy $E_K = (1/2)k_BT$ and the recoil energy $E_R$:

$$D_T = 2\sqrt{E_K E_R}$$

(3.16)

This eliminates the major source of $\gamma$ line broadening, and for high quality crystalline source and absorber the Mössbauer line width is close to the value given by the uncertainty principle $(\Delta E)(\Delta t) \geq \hbar$. Applying this principle to Mössbauer transitions one can estimate the natural linewidth $\Gamma$ of the absorption line. The mean life time $\tau$ of the excited state is related to the half-time, $t_{1/2} = \tau n2$. The uncertainty relation for this case is given by

$$\Delta E = \Gamma = \frac{\hbar}{\tau} = \frac{0.693\hbar}{t_{1/2}}$$

(3.17)

The first excited state of $^{57}$Fe (14.4 keV) is characterized by a half-time value $t_{1/2} \sim 10^{-7}$ s. Thus the energy broadening is $\Gamma \sim 5 \times 10^{-9}$ eV. This implies a resolution $E/\Delta E$ of approximately $10^{12}$, which explains the usefulness of Mössbauer spectroscopy in identifying small energy shifts in an $^{57}$Fe nucleus associated with specific atomic environments. In conventional Mössbauer spectroscopy the emitted $\gamma$ rays are transmitted through a thin absorber and subsequently detected either by scintillation or by proportional counters. When the resonant absorption occurs the number of detected $\gamma$ events decreases.

A typical absorption Mössbauer spectrum, is shown in Fig. 2.6. CEMS takes advantage of the fact that the 14.4 keV excited state of $^{57}$Fe can also de-excite by internal conversion, see above. The probability of internal conversion in $^{57}$Fe is nine time higher than
the spontaneous emission of a 14.4 keV photon. De-excitation by internal conversion results in the ejection of a conversion electron having a kinetic energy of 7.3 keV. Other de-exitation processes are shown in Fig. 3.17 b). In CEMS the backscattered electrons are detected rather than γ photons. One can apply this method to samples deposited on thick γ-ray nontransparent substrates such as 0.35 mm thick GaAs. CEMS is also more surface sensitive than the conventional Mössbauer transmission method because the electron escape depth is ~ 0.5 μm. In practice, a sensitivity to interface order can be achieved by depositing a few atomic layers of pure $^{57}\text{Fe}$ within a structure which was otherwise prepared using a regular Fe source. Regular Fe contains only 2% of $^{57}\text{Fe}$ and is practically insensitive to the Mössbauer effect if the equivalent amount of $^{57}\text{Fe}$ in the regular Fe is much less than the amount of $^{57}\text{Fe}$ in the probe layer.

Spectrometer setup

The basic setup to record Mössbauer spectra is fairly simple and can be split into two parts: (a) Detection unit and (b) the unit controlling the motion of the γ-ray source. Both units have to be synchronized. The block diagram for the CEMS spectrometer used at the University of Technology, Krakow, Poland is shown in Figure 3.18. The CEMS detector is essentially a gas-flow proportional counter [85] which was designed with a cylindrical geometry. Its diameter was adjusted to a minimum of 3.5 mm in order to achieve a high signal/noise ratio. The casing of the electron counter was made of Teflon and was mounted inside an aluminium housing. A tungsten wire (anode) of 10 μm diameter was placed in the center of the counter at an applied voltage of 800-1000 V. The detector was run with a constant gas flow of 90% He + 10% CH4. A 3.5 mm thick helium gas layer at 0.1 MPa has efficiencies of less than 0.02% for 6.4 keV x-rays and less than 0.002% for 14.4 keV γ-rays, therefore the electron counter detects practically only the conversion electrons. The sample was mounted inside the detector to obtain optimum efficiency for backscattered electrons. The front side of the CEMS detector was closed with a thin Al window which could absorb more than 80% of unfavorable Fe K x-rays from the source, while the reduction of the 14.4 keV γ-ray intensity was about 20%. The rear side of the CEMS detector provided a sample holder for easy mounting. The design of the detector can sometimes be very sophisticated and depends mainly on the system configuration, therefore various Mössbauer groups often use their own
Figure 3.18: A block diagram of the CEMS spectrometer used at the AGH-University of Science and Technology, Krakow, Poland
particular detector design which best suits their experiments. In the proportional gas­flow counter the conversion electrons were collected from the anode as a charge pulse. The pulse from the detector was then amplified using a pre-amplifier and amplifier, and accumulated by the single channel analyzer (SCA). The single channel analyzer is in principle an analog to digital converter. It converts the amplifier output pulse into a logic pulse for input into a counting device. A typical logic pulse height measured at the output of the (SCA) is $\sim 10^6 - 10^8$ counts. The source of $\gamma$-rays ($^{57}$Co) was Doppler shifted using a velocity transducer which followed a triangular waveform at a frequency of 10 Hz supplied from a signal generator. The electro-mechanical transducer used a feedback loop in order to achieve lowest possible error signal. The waveform generator was synchronized with a multichannel scaler so that every channel accepted the counts from the (SCA) at a particular transducer velocity in equal time intervals. The full spectrum was recorded in 256 channels using a standard PC computer. In CEMS one detects the conversion electrons which are backscattered from the sample, therefore at resonance the number of counts increases. In this case the spectrum will appear “upside down” to a typical Mössabuer spectrum shown in Fig. 2.6. This is another characteristic feature of the CEMS method. The resonances are peaks instead of valleys but the information contained is the same. Despite the high efficiency of the CEMS detectors and a very high signal/noise ratio a great deal of time has to elapse before enough counts are recorded to obtain clean spectra ($> 10^6$ counts/channel). In practice, the measurements of our thin films required at least 3 days of running time in order to obtain desirable counting statistics.
Chapter 4

Sample preparation

4.1 GaAs(001) Substrate preparation

All samples were grown on GaAs(001) wafers. Commercial epi-ready GaAs substrates (manufactured by American X-tal Technology) were always covered by a few nanometers of natural oxides. Since the GaAs wafers were exposed to air prior to loading into the MBE system they required outgassing which was always the first step in the preparation process. Outgassing was performed in the introductory chamber at an elevated temperature (~370°C) mainly to desorb water. This process usually took several hours until the pressure in the chamber reached ~ 3 × 10^{-10} Torr. In order to remove oxides from the surface and to obtain stoichiometric clean GaAs surfaces the following procedure was employed:

1. 30 - 40 minutes of Hydrogen cleaning
2. 3 hours of Ar$^+$ sputtering with azimuthal sample rotation
3. 40 - 50 minutes annealing at a temperature of ~ 600°C

Each of the above steps requires precisely controlled settings and they will be described separately in detail below.

4.1.1 Hydrogen cleaning

Surface cleaning using atomic hydrogen provides a fast method for removal of surface contamination. An atomic hydrogen (H$^+$) beam was generated using a hydrogen thermal
gas cracker (Oxford Scientific). The gas cracker passes molecular hydrogen (H$_2$) through a narrow 2000°C hot tungsten tube. H$_2$ molecules are dissociated into atomic radicals H* during collisions with the hot walls of the tube. This process provides a beam with approximately 15 degrees of divergence and a 90% dissociation rate. In order to obtain high purity H$_2$ a gas purifier and liquid nitrogen cold trap were placed between the gas tank and the hydrogen gun. The hydrogen cleaning process can be divided into two steps since the atomic hydrogen interacts differently with As and Ga oxides [86]. The first step reduces the As oxides, and can be represented by the following reaction:

$$\text{As}_2\text{O}_x + 2x\text{H}^* \rightarrow x\text{H}_2\text{O} \uparrow + \text{As}_2 \uparrow,$$

where $x = 1, 3, 5$ stands for various As oxides and (↑) represents the product removed from the surface during cleaning. The second step reduces the remaining Ga oxides;

$$\text{Ga}_2\text{O}_3 + 4\text{H}^* \rightarrow \text{Ga}_2\text{O} \uparrow + 2\text{H}_2\text{O} \uparrow,$$

The As oxides can be removed already at room temperature while the complete desorption of Ga$_2$O requires elevated temperatures (300-400 °C). Heating a GaAs wafer to ~400 °C does not result in the thermal desorption of As. Moreover it also limits the diffusion of H* into GaAs [87]. Therefore the GaAs substrate was held at 400°C in order to achieve a maximum reaction yield during the cleaning process.

After 30 min of hydrogen cleaning at a hydrogen pressure of 5 × 10$^{-7}$ Torr the AES spectrum showed a very weak oxygen signal comparable to the noise level. One should mention that the Carbon signal was also reduced to the noise level suggesting that the hydrogen cleaning further reduced Carbon contamination. This is in agreement with Schafer et al. [88] who used a similar hot-filament hydrogen cracking method.

### 4.1.2 Ar$^+$ sputtering

Ar$^+$ ion bombardment is another method commonly used to remove native oxide layers from the substrate surface and reach a reliable 4 × 6 reconstruction after annealing. At SFU Ar$^+$ sputtering was performed in the analysis chamber using a standard differential ion gun. The Ar gas flow was precisely controlled using a bleeder valve that maintained a pressure of 10$^{-4}$ Torr inside the gun. The Ar back-pressure inside the chamber was 3 × 10$^{-7}$ Torr during sputtering. The GaAs wafer was bombarded using 650eV Ar$^+$ ions at RT. In order to minimize surface damage the Ar beam was set up at an incident
grazing angle of 65° with respect to the surface normal. During sputtering the wafer was rotated around its normal axis and the ion beam was rastered over 10 × 10 mm². The samples prepared at SFU required 2.5 - 3 hours of sputtering to completely remove contaminants. The GaAs(001) substrates prepared at MPI in Halle required 7-8 hours of sputtering with the same ion energy and Ar⁺ dose (pressure): in their case hydrogen cleaning was not available. In both cases the cleanliness of the GaAs surface was verified by means of AES. It is important to point out that the sputtering allowed one to obtain a well defined 4 × 6 GaAs surface reconstruction after annealing whereas hydrogen cleaning after annealing resulted only in a weak 4 × 6 GaAs surface reconstruction.

4.1.3 Annealing and GaAs surface reconstruction

The GaAs compound semiconductor has a cubic zincblende structure that can be thought of as an equivalent to two interpenetrating fcc lattices, each occupied by Ga and As atoms. The fcc lattices are displaced along the lattice diagonal by \( \sqrt{3}a/4 \), where \( a = 5.64 \) Å is the lattice constant of GaAs, see figure 4.1). The As and Ga atom nearest neighbor environment has a tetrahedral coordination with sp³ hybridization of the atomic orbitals. The Ga-As bonds have predominantly a covalent character with some ionic contribution due to the different electronegativities of Ga and As. The \{100\} planes in bulk GaAs are occupied either by cations or by anions in an alternating fashion. An ideally terminated GaAs(001) surface should be therefore either Ga or As terminated with two dangling bonds (unpaired electrons) per surface atom. In fact this never happens and depending on predominant atomic species the surface is classified either as Ga rich or as As rich [89]. The GaAs(001) surface exhibits a whole range of reconstructions which involve significant rearrangements of the surface atoms. These reconstructions are driven by a large surface free energy associated with dangling bonds at the surface. The reconstructions on GaAs(001) are usually identified using the notation A×B where A and B represent the periodicities in the [11̅0] and [110] directions, respectively [90].

A few general principles have been derived from calculations and experiments which offer a good qualitative explanation for a large number of semiconductor surface reconstructions [91]. For the special case of tetrahedrally coordinated compound semiconductors those principles can be stated as follows:
1. Surface atomic geometry is determined primarily by a rehybridization-induced lowering of the surface-state bands associated with the (filled) anion dangling bond orbitals.

2. Surfaces tend to be autocompensated by requiring that no charge accumulate at the surface.

3. Surface structure observed will be the lowest energy structure which is accessible under the preparation conditions.

For GaAs(001) those principles have the following implications:

Principle 1 implies that the dimerization of adjacent surface atoms reduces the number of energetically unfavourable dangling bonds by a factor of 2 for an ideally terminated surface. According to the definition of the zincblende structure each Ga atom bonds to the As atoms in the [111], [111], [111] and [111] directions (see figure 4.1). Therefore the dangling bonds on Ga terminated GaAs(001) surfaces lie in the (110) plane whereas on As terminated surfaces they lie in the (110) plane. As the surface dimers are formed along
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the direction of the dangling bonds this will yield a surface periodicity 2× parallel to dimer’s directions; [110] on Ga terminated surfaces and [110] on As terminated surfaces.

Principle 2 determines the surface stoichiometry. Due to a big difference in energy between the Ga and As dangling bond states it is energetically favorable to transfer charge from the cation-derived surface states to the anion-derived ones. For GaAs the As dangling bond states lie below the valence-band maximum whereas the Ga dangling bond states are well within the conduction band. To achieve an uncharged semiconducting surface it is necessary to completely empty all of the Ga dangling bond states and to fill all the As dangling bond states (two electrons) as well as any AsAs and GaGa dimer bonds (two electrons) on the surface. Since such conditions are not possible on an ideal GaAs(001) surface, non-stoichiometric missing dimer surface reconstructions are formed which compensate any lack/excess of electrons in the first layer by transferring charge from/to exposed second layer atoms. This process of missing dimer formation (autocompensation) gives rise to ×N symmetry that is normal to the 2× symmetry, see above.

Principle 3 addresses the fact that the surface reconstruction depends crucially on the precise processing history. Each semiconductor surface corresponds to a local free energy minimum. Different energy minima are usually associated with different surface structures and stoichiometries and the energy difference between them is often very small. Therefore a slight modification of the preparation method can yield a significantly different surface reconstruction. For example Q. K. Xue et. al. [92] in his STM studies showed a number of reconstructions ranging from As-rich to Ga-rich surfaces. In order of decreasing As content and increasing annealing temperature the following reconstructions were reported: (4 × 4), (2 × 4), (1 × 6), (4 × 6), (4 × 2) and (8 × 2). Additionally Biegelsen et al. [93] found that the (4 × 6) reconstruction consisted of coexisting (4 × 2) and (2 × 6) domains referred to as a “pseudo” (4 × 6) reconstruction which is less Ga rich than the (4 × 2) reconstruction. Finally there is also a “genuine” (4 × 6) reconstruction which is believed to be more Ga rich than the (4 × 2) reconstruction [92].

At SFU and MPI-Halle the specimen was annealed as the last step in a substrate preparation. The substrate was moved to the growth chamber and the sample temperature was gradually raised either within 30-40 min to 590°C (SFU) or within 20 min to ~600°C (MPI). Throughout this annealing process the specimen was kept under RHEED observation until a well ordered (4 × 6) reconstruction was obtained. During annealing
usually one observed the surface periodicity along the [110] direction by monitoring the x6 diffraction spots with the RHEED beam oriented along the [110] crystallographic axis of GaAs. If the wafer was overheated the x6 diffraction spots changed to the x2 periodicity corresponding to the 4 x 2 reconstruction accompanied by Ga clusters on the surface. Figure 4.2 a) and b) show the RHEED diffraction patterns and corresponding STM images c) and d) for GaAs(001) samples prepared at SFU and MPI-Halle, respectively. In both cases a well reconstructed 4 x 6 surface was observed. However a further investigation using the STM clearly revealed that those reconstructions were not exactly the same; they possessed domains having different reconstructions. SFU prepared substrates consisted of a "pseudo" (4 x 6) reconstruction dominated mainly by (2 x 6) domains (Fig. 4.2 c)) while the substrates prepared at MPI-Halle exhibited a (4 x 6) reconstruction which was a combination of (2 x 6) and "genuine" (4 x 6) domains. This clearly demonstrates that a difference in preparation method (long sputtering and no H-cleaning for MPI samples) can lead to different surface topographies that are indistinguishable using RHEED observations.

4.2 Thin films growth

The preparation of Fe films was carried out by means of Molecular beam epitaxy (MBE). In order to obtain high quality crystalline films the substrate lattice net has to be well matched to the lattice of the deposited material. For Fe films, GaAs is a good candidate. The Fe(001) lattice mesh has a lattice constant that is 1.4% smaller than the size of half unit cell of GaAs(001), and the in-plane epitaxial relation is [100]Fe || [100]GaAs. For all samples, the Fe film was always deposited directly on the GaAs(001) at room temperature (RT). The Fe films were deposited using two methods: Thermal deposition (TD) and Pulsed Laser Deposition (PLD). Both methods resulted in high quality crystalline Fe films and will be described below in separate sections.

4.2.1 Thermal deposition (TD)

All metallic films at SFU were prepared by means of the TD method using thermal sources described in section 3.1.5. The base pressure during the growth was less than 3 x 10^{-10} Torr for deposition rates of ~ 2 ML/min (Monolayers/minute). The film
Figure 4.2: RHEED diffraction patterns and STM images of a (4 x 6) reconstructed GaAs(001) wafer. (a) and (b) show the RHEED diffraction patterns for GaAs(001) samples prepared at SFU and at MPI-Halle, respectively. The direction of the electron beam with respect to the crystallographic axes of GaAs(001) is indicated by the arrows. Corresponding 100 x 100 nm² STM images are shown in (c) for $V_{\text{bias}} = -2.2$ V and (d) $V_{\text{bias}} = -2.3$ V, respectively. Substrates prepared at SFU possess a so-called “pseudo” (4 x 6) reconstruction which is mainly dominated by (2 x 6) domains, while the substrates prepared at MPI-Halle show a (4 x 6) reconstruction that is a combination of (2 x 6) and “genuine” (4 x 6) domains.
thickness was monitored by means of RHEED intensity oscillations and a Quartz crystal microbalance (thickness monitor).

Growth of Fe on GaAs(001)

Figure 4.3: (a) RHEED specular spot intensity oscillations for 16ML of Fe grown on GaAs(001). The dramatic increase of the specular spot intensity and RHEED oscillations observed after deposition of an equivalent of three atomic layers indicates the formation of a continuous film. The period of the oscillations corresponds to the formation of an additional atomic layer; this was confirmed by the thickness monitor. (b) RHEED diffraction images taken after the deposition of the first 5 atomic layers of Fe on GaAs(001) for the electron beam oriented along the [110] crystallographic direction of Fe. A well-defined streaky pattern indicates the formation of a crystalline film.

Excellent UHV conditions and relatively slow deposition rates ensured pseudo layer-by-layer growth where the top surface consisted of a maximum of two unfinished atomic layers. RHEED oscillations were visible during the deposition of 16 atomic layers of the Fe film as shown in Fig. 4.3 a). During the initial stages of growth the oscillations were weak suggesting that the transition from the Fe/GaAs interface formation to a continuous Fe film occurred after the deposition of an equivalent of three atomic layers [94]. A dramatic increase of the specular spot intensity and the amplitude of the RHEED oscillations confirmed that the growth of Fe proceeded in a quasi layer-by-layer mode.
At the same time the RHEED patterns were sharply defined, see Fig. 4.3 b), and the separation between the RHEED streaks revealed that the Fe film grew in the bcc single crystal structure. The diffraction streaks were split indicating also that the atomic terraces were small and several nm across. This was confirmed by in situ STM images. Figure 4.4 a), b) shows the surface topography for Fe films 2 and 10 ML in thickness, respectively. The atomic terraces were small with an average terrace size of $\sim 3.5$ nm. The surface roughness was mainly confined to the two top atomic layers. The mean surface roughness (RMS) was found to be 0.18 nm and the maximum height difference $\delta h = 0.37$ nm i.e. 2.5 atomic layers for both the 2 and 10 ML thick Fe films.

Figure 4.4: 100 x 100 nm$^2$ STM images corresponding to the 2 ML (a) and 10 ML (b) thick Fe films grown on GaAs(001). In both cases the atomic terraces were small with an average terrace size of $\sim 3.5$ nm in (a) and $\sim 4.5$ nm in (b). The surface roughness was mainly confined to the two top atomic layers. The mean surface roughness RMS=0.18 nm and the maximum height difference $\delta h = 0.37$ nm were found to be the same for both the 2 and the 10 ML Fe films.

Growth of Au and Ag on Fe/GaAs(001)

Au and Ag are very good candidates for multilayer structures in which the Fe films are separated by nonmagnetic spacers. A thin Au cap layer was used to protect the samples from ambient conditions (oxidation). Ag could not be used as a capping layer because it tends to accumulate sulfur. Therefore 20 ML of Au was always used as a capping layer on all structures prepared for ex situ magnetic measurements.
Both Ag and Au grow epitaxially on Fe(100) with the in-plane crystal axes rotated by 45° with respect to the Fe surface mesh, $[100]_{\text{Ag,Ag}} || [110]_{\text{Fe}}$. This epitaxial orientation gives rise to a small lattice mismatch of 0.6% between the Fe and Au meshes, and a 0.8% mismatch between the Fe and Ag meshes. Figure 4.5 a) shows the RHEED intensity oscillations for Au grown on Fe/GaAs(001). The oscillations were visible for thicknesses up to 20 ML of Au indicating a layer-by-layer growth. The RHEED diffraction images of Au(001) grown directly on Fe/GaAs(001) at RT always showed a $2 \times 2$ reconstruction pattern as illustrated in Fig. 4.5 b) and c). This type of Au(001) reconstruction instead of the well known $5 \times 1$ reconstruction is attributed to the segregation of the As atoms on the Au surface [90]. X-ray Photoemission spectroscopy measurements (XPS) indicated that during the Fe deposition on GaAs, approximately 0.5 ML of As was segregated to the surface of the Fe film [74]. After the deposition of additional Au layers the same amount of As was found on the Au surface, and this implies that the As atoms act as a surfactant and no As stays inside the metallic layers. STM studies on Au grown on

![Figure 4.5: (a) The RHEED specular spot intensity oscillations observed for Au grown on Fe/GaAs(001). The maximum Au thickness is 20ML. The sample was prepared using standard thermal deposition techniques (TD). (b) The RHEED pattern observed for the 20Au/16Fe/GaAs(001) surface with the electron beam oriented along the $\langle 100 \rangle_{\text{Au}}$ and (c) $\langle 110 \rangle_{\text{Au}}$ directions, respectively. The additional streaks, indicated by the arrows between the zeroth and first order diffraction streaks, show the presence of a $2 \times 2$ reconstruction that is caused by the surfactant As atoms.](image-url)
Fe/GaAs(001) structures showed that the surface roughness is mainly confined to two atomic layers of Au with a maximum height difference of 4.8 Å and an average height of 2.3 Å (1 ML of Au) over a distance of 200 nm indicating a smooth quasi layer-by-layer growth, see Fig. 4.7 d). The STM image presented in Fig. 4.7 e) at an atomic resolution confirmed the $2 \times 2$ surface reconstruction observed using RHEED. The line profile presented in Fig. 4.7 f) shows that the maxima corresponding to the Au atoms are separated by 8 Å (double the Au lattice parameter) along the $\langle 100 \rangle$ direction (the directions are referenced to the Au lattice).

For Ag grown on Fe/GaAs(001) strong and streaky RHEED diffraction patterns developed after the deposition of 5 ML of Ag and showed no reconstruction. This indicates that As did not segregate to the Ag surface. This is in agreement with XPS studies by Ludeke et al. [95] who investigated the interfacial properties of Ag on GaAs(001). They showed that the interaction between the Ag atoms and the GaAs substrate was weak and no interdiffusion and surface segregation of either As or Ga was observed. Indeed the deposition of 20 ML of Au on Ag showed the typical $5 \times 1$ surface reconstruction with four well defined equally spaced streaks between the regular $[100]$ reciprocal lattice streaks, see the RHEED patterns in Fig. 4.6 b). For the electron beam oriented parallel to the $[100]$ crystallographic direction of Au the RHEED streaks were split showing a typical fan-out diffraction pattern, see Fig 4.6 a). This effect is caused by a rectangular network of surface steps oriented along the $\langle 110 \rangle$ directions [69, 96]. The fan-out effect is entirely absent when the electron beam is oriented along the $\langle 110 \rangle$ direction. Big area atomic resolution STM images of $20\text{Au}/1500\text{Ag}/16\text{Fe}/\text{GaAs}(001)$ (the integers correspond to the number of atomic layers) are shown in Fig. 4.7 a) and b). The $5 \times 1$ reconstruction consists of surface corrugations separated by 14.5 Å along the $\langle 110 \rangle$ direction, see the inset in Fig. 4.7 a). The terraces are roughly 50 nm long and 40 nm wide. The atomic resolution image revealed that the surface corrugations are 0.6 Å high and are caused by buckling of the surface atoms, see Fig. 4.7 b) and the line profile shown in Fig 4.7 c). The surface buckling is caused by the lattice mismatch between the fcc (100) surface and the hexagonal top layer [97]. The hexagonal top layer minimizes the Au surface energy. Notice that due to the absence of As on the surface of Au grown on Ag, the atomic terraces are larger, have square shapes, and no screw dislocations are present.
Figure 4.6: RHEED diffraction patterns of a (5 \times 1) reconstructed Au surface grown on Ag/Fe/GaAs(001) at RT by thermal deposition (TD). (a) The primary electron beam was oriented along the (100)_{Au} direction. The streaks are split showing a typical fan-out diffraction pattern that is caused by a rectangular network of surface steps oriented along the (110) directions. (b) The primary electron beam was oriented along the (110)_{Au} direction. Four well defined equally spaced streaks between the regular lattice streaks indicate the presence of a 5\times surface superlattice periodicity.

### 4.2.2 Pulsed laser deposition (PLD)

**Features of laser ablation of metals**

In a pulsed-laser deposition short laser pulses strike a target resulting in ablation of materials from the target. Once the laser pulse is absorbed in the target's surface region the electromagnetic energy is immediately converted into atomic and electronic excitations in the form of plasmons and unbound electrons. Within a few picoseconds the excited electrons then transfer their energy to the lattice via electron-phonon (e-p) coupling. For a laser pulse duration \( \tau \) longer than the e-p coupling time the material is ablated. This is caused exclusively via conventional heat deposition.

In the laser ablation process the focused light produces a large electric field sufficient to remove some of the electrons within the optical absorption depth of the material from their atoms by an ionization process. The optical absorption depth of most materials lies in the region of 10 nm, hence the affected volume is of the order of \( 10^{-8} \text{cm}^3 \) for a 1 mm\(^2\) laser focus. This volume may contain \( 10^{15} \) unbound electrons. [98]. The ablation plasma is seeded by these initially-created electrons within typically \( \tau \approx 10 \text{ ps} \) for ns-laser pulses. The electrons oscillate in the electromagnetic field of the laser light, and also
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Figure 4.7: STM images of Au grown on Ag/16Fe/GaAs(001) and on Fe/GaAs(001) templates using TD. (a) A 200 × 200nm² STM image of the Au capping layer grown on 1500Ag/16Fe/GaAs(001). The atomic terraces are square shaped and ∼ 40 nm wide. The inset shows a magnified terrace image that exhibits surface corrugations along the (110)Au direction indicating a 5× reconstruction. The atomic resolution STM image in (b) reveals that the surface corrugations are 0.6 Å high and are separated by 14.5 Å along the (110) direction (see also the line profile in (c)). The corrugations are caused by buckling of the surface atoms. (d) A 200 × 200nm² STM image of a 150 ML thick Au layer grown directly on Fe/GaAs(001). The formation of screw dislocations is clearly visible. (e) An atomic resolution STM image showing the 2 × 2 reconstructed surface of Au grown on Fe/GaAs(001). (f) The line profile corresponding to black line in figure (c). The maxima correspond to rows of Au atoms separated by 8 Å (double the Au lattice parameter) along the (100)Au direction. The images were processed using: “WSxM- free software downloadable at http://www.nanotec.es”.
collide with neighboring atoms or ions, thereby transferring some of their energy to the lattice within the surface region of the target. Once a sufficient density of free electrons has been generated, this primary process is followed by an inverse Bremsstrahlung effect driven by the remainder of the laser pulse. After the duration of the laser pulse a very hot cloud of vaporized material, typically at a temperature of 10,000 K or more is generated: this cloud is referred to as the ablation plasma, or plume.

In the case of metal targets, in which the optical absorption depth is smaller than the thermal diffusion length, the energy from the laser pulse is first transferred into the absorption layer, and then thermally transported into the depth corresponding to the thermal diffusion length; this length is proportional to $l_D = \sqrt{D\tau}$, where $D$ is the thermal diffusion constant and $\tau$ is the time needed to create an erosion plasma after the start of the laser pulse. For metals this depth is typically several tens of nanometers [99]. With a sufficiently strong laser fluency, (laser pulse energy/focal area), the transfer of electron-lattice energy allows significant surface vaporization to continue for about 100 ps after application of the initial nanosecond laser pulse. The plasma contains neutrals of a few eV in kinetic energy and ions that can have kinetic energies as great as a few hundred eV depending on the laser fluency. For metallic materials a large laser fluency of about 5 J/cm$^2$ is required [100, 101] to obtain sufficiently great ablation rates (on the order of 0.01 nm per pulse). Both the neutrals and ions contained in the ablated plume have a considerably larger kinetic energy than thermally deposited atoms using standard effusion cells ($\sim$0.1 eV). The difference in kinetic energy and in the large density of nucleation centres obtained using PLO compared with TD results in different growth morphologies of the deposited films.

**Laser preparation and PLD deposition of Fe**

A first step for the application of PLD was to outgass the Cu, Fe, and $^{57}$Fe targets. This was achieved by directing the KrF excimer laser beam onto the targets with a pulse repetition rate of 10 Hz until the base pressure in the growth chamber reached $\sim 10^{-10}$ Torr. In order to achieve the required thicknesses of the deposited Fe films the deposition rates were calibrated by growing Fe and $^{57}$Fe on Cu(100) single crystal substrates. RHEED oscillations were not observed in the PLD growth of Fe on GaAs(001). Most likely this was caused by technical difficulties associated with the sample holder adjustment required for obtaining the anti-Brag diffraction condition. RHEED oscillations were observed for
the PLD growth of Fe on Cu(100), see Fig. 4.8, thereby allowing one to calibrate the thickness monitor. However one has to keep in mind that Fe grows on Cu(100) in the fcc lattice structure compared with the Fe bcc structure for growth on GaAs(001), and therefore the deposition time $t_{\text{bcc}}$ for one ML of the bcc-Fe had to be rescaled by taking into account the number of atoms required to fill the bcc and fcc Fe(001) sites. The fcc-Fe(100) plane (on fcc Cu(001)) has a square unit cell $a_0 = 3.56 \, \text{Å}$ on an edge and contains two atoms. For the bcc-Fe(100) template the unit cell edge is $a_0 = 2.87 \, \text{Å}$ and contains one atom. Knowing the deposition time for 1 ML of fcc-Fe ($t_{\text{fcc}}$), see Fig. 4.8, one can obtain the deposition time $t_{\text{bcc}}$ required to grow 1 ML of bcc-Fe on GaAs(001) from the following relation:

$$t_{\text{bcc}} = \frac{6.3}{8.2} t_{\text{fcc}} \simeq 0.8 t_{\text{fcc}} \quad (4.1)$$

Since the deposition rates in PLD may vary over a long period of time and depends on

![Figure 4.8: RHEED specular spot intensity oscillations observed during the PLD growth of Fe on Cu(100).](image)

the alignment of the excimer laser onto the targets the calibration of the growth rates had to be performed before the preparation of a new sample. Therefore the growth of an Fe/GaAs(001) structure was performed immediately after the “calibration growth” on Cu. All Fe and $^{57}\text{Fe}$ films prepared by the PLD technique were deposited at RT. The PLD
deposition was carried out on 4 × 6 reconstructed GaAs(001) substrates prepared by the sputtering and annealing process described in sec 4.1. The substrates were attached to the sample holder with their surfaces oriented parallel with the target surface at a target-to-substrate distance of approximately 15 cm. In order to obtain a steady ablation rate the targets were rotated in the plane. Figure 4.9 a) shows the 30 keV RHEED diffraction pattern after a PLD deposition of 5 ML of Fe which consisted of 2 ML of $^{57}$Fe and 3 ML of natural Fe. Well developed RHEED streaks indicate a smooth PLD growth of Fe on GaAs(001), and they are similar to the RHEED patterns observed during TD (see Fig. 4.3 b)). The STM image of 2ML of $^{57}$Fe shown in Fig. 4.9 b), indicates that the GaAs(001) substrate was fully covered after the PLD deposition of 2 ML of Fe. The RMS roughness of 0.19 nm and the maximum height difference $\delta h = 0.37$ nm were nearly the same as those observed for TD films: this suggests that the PLD growth also proceeded in a quasi layer-by-layer manner. The average Fe terrace size obtained using PLD was somewhat smaller (3 nm) compared with films prepared using TD (3.5 nm). After the PLD deposition the Fe films were covered by a capping TD Au layer 20 ML thick. Surprisingly the RHEED and STM images of the Au capping layer, see Fig. 4.9 c), d), exhibited a 5 × reconstruction patterns with the electron beam oriented along the $[110]_\text{Au}$ direction; a pattern similar to that obtained for Au deposited on a Ag film, see Fig. 4.6, b). This means that during PLD the Arsenic atoms did not segregate to the surface contrary to the segregation observed for the TD Fe films. This can be explained on a qualitative level by taking into account so called preferential resputtering of atoms from the film surface which often happens in PLD [102, 103]. At a laser fluency of $\sim$5 J/cm$^2$, an ablated plume contains neutral atoms with an energy of about 5 eV and energetic ions (often more than 50% of all ablated particles) with an average kinetic energy of about 100 eV [104]. At these energies ions can be implanted as much as a few monolayers deep into the film thereby at the same time causing atoms from the film surface to be resputtered. A much higher probability for Arsenic to be removed from the Fe film rather than the removal of Fe itself can be understood by comparing the sputter yields, $Y$, for these elements. For 100 eV ions impinging on the surface the sputter yields for Fe and As are $Y_{Fe} \simeq 0.25$ and $Y_{As} \simeq 1.2$, respectively. These values were obtained from [105] using the Ar ion sputtering yields. The sputtering yield for Fe from ref. [105] is in good agreement with the sputtering yield $Y_{Fe} \simeq 0.2$ quoted in
Figure 4.9: (a) A 30 keV RHEED diffraction pattern after the PLD deposition of 5 ML equivalent of Fe on GaAs(001). The RHEED electron beam was oriented along [110]$_{Fe}$ crystallographic direction. (b) A 200 × 200nm$^2$ STM image of 2 ML PLD deposited $^{57}$Fe shows that the GaAs substrate was fully covered after the deposition of 2 ML of Fe. The RMS roughness was 0.19 nm, and the maximum height difference was $\delta h = 0.37$ nm. (c) A RHEED image with the electron beam oriented along [110]$_{Au}$. (d) The corresponding 30 × 30 nm$^2$ STM image of the 20Au$_{TD}$10Fe$_{PLD}$/GaAs structure. The subscripts correspond to the deposition technique. The 5x surface reconstruction of Au indicates that Arsenic does not segregate on the surface during PLD.

ref. [103] obtained for PLD under UHV conditions with a laser fluency of $\sim 4.5$ J/cm$^2$. This significant difference in the sputter yields for As and Fe (about 5 times higher for As) can explain the absence of surfactant As on the PLD films. In fact the PLD deposition of an equivalent of 2 ML of $^{57}$Fe on GaAs(001) plus an additional thermally deposited 8 ML of Fe and 20 ML of Au exhibited the 5x Au reconstruction. This leads to the conclusion that As removal from the Fe surface during PLD occurred relatively quickly within the deposition of the first 2 ML of Fe on GaAs.
Chapter 5

Properties of ultrathin Fe films

5.1 Fe/GaAs(001) CEMS interface studies

Conversion Electron Mössbauer Spectroscopy (CEMS) provides an excellent tool for identification of the electronic and magnetic states of Fe atoms, see section 2.5. CEMS experiments described in this section have been motivated by ongoing discussion of the structural and chemical properties of crystalline GaAs/Fe(001) interfaces [106, 107]. They are directed towards two goals: (a) Understanding of the chemical environment of Fe at GaAs/Fe(001) interfaces that were prepared by thermal deposition (TD) and pulse laser deposition (PLD) techniques. (b) Understanding the role of TD and PLD on the interface magnetic anisotropies (see Section 4.2.2).

In Mössbauer Spectroscopy an isotope $^{57}$Fe is usually used as a probe layer. For this reason the equivalent of two atomic layers of $^{57}$Fe was deposited using TD and PLD at the GaAs(001) interface and inside the Fe film. The studies were carried out using the following structures:

- $\text{GaAs/}^{57}\text{Fe}_{\text{PLD}}/8\text{Fe}_{\text{TD}}/20\text{Au(001)}$
- $\text{GaAs/}^{57}\text{Fe}_{\text{PLD}}/8\text{Fe}_{\text{PLD}}/20\text{Au(001)}$
- $\text{GaAs/}^{57}\text{Fe}_{\text{TD}}/8\text{Fe}_{\text{TD}}/20\text{Au(001)}$
- $\text{GaAs/5Fe}_{\text{TD}}/^{57}\text{Fe}_{\text{TD}}/3\text{Fe}_{\text{TD}}/20\text{Au(001)}$
- $\text{GaAs/7Fe}_{\text{TD}}/^{57}\text{Fe}_{\text{TD}}/7\text{Fe}_{\text{TD}}/20\text{Au(001)}$
where the integers represent the number of atomic layers, and the subscripts describe the deposition technique. The CEMS spectra taken at RT are shown in Figures 5.1 and 5.3. The thin solid lines represent the relative contributions of the components in the Mössbauer spectrum. The thick solid lines are the overall fits to the data.

**Data analysis**

The method used to fit the Mössbauer data was based on the assumption that the Mössbauer spectrum associated with an Fe atom depends only on the configuration of the nearest and the next-nearest neighbors. Each such configuration (component) was treated as a separate Fe site having its own hyperfine field \( H_f \), isomer shift \( I_s \), and intensity. For ideal infinite Fe crystal, an Fe atom in the bcc structure has eight nearest neighbors \( (n = 8) \) and six next-nearest neighbors \( (m = 6) \); this results in \( 9 \times 7 = 63 \) possible combinations of neighbors in the first two zones \( (n, m) \). This assumes that only the number of Fe atoms in the particular zone is important, not their actual arrangement. In addition further assumptions have been made to limit the number of fitting parameters:

1. Multiple impurity-neighbor effect were additive i.e.,

\[
H_f(n,m) = H_{f0} + \Delta H(n+m)
\]  

(5.1)

where \( H_f(n,m) \) is the reduced hyperfine field for the given number of impurities in \( n \) and \( m \) zones. The largest measured hyperfine field \( H_{f0} \) was fixed at 322 kOe obtained from the sample GaAs/5Fe_{TD}/2^{25}Fe_{TD}/3Fe_{TD}/20Au(001), (see discussion of Fig. 5.3). The hyperfine field \( H_f \) was expected to decrease by \( \Delta H = -23.5 \) kOe [64] on average for each replacement of the nearest neighbor Fe atom with either an As or a Ga atom, except for the smallest hyperfine field components where \( \Delta H \) was used as a free parameter in order to obtain the best fit, see Fig. 5.1.

2. The \((n,m)\) configurations having the same total number of impurities in the \( n \) and \( m \) zones had equal probability i.e., the configuration \((8,0)\) having eight As or Ga atoms as the nearest neighbors and zero atoms as the next-nearest neighbors would result in the same \( H_f \) decrease as the \((4,4)\) configuration. Using this assumption the number of possible combinations could be limited to total of 15.
3. A quadrupole admixture existed in order to obtain small asymmetry in the Mössbauer spectra.

4. The same linewidth was assigned to all peaks in the given spectrum.

5. The line shape was Lorentzian.

The so-called additive model described above was adapted from the work of Newkirk et al. [108] and Vincze et al. [64] who used it for the analysis of Mössbauer data for Fe-Ga and Fe-As alloys. The actual fitting was carried out using an iterative least-square procedure described by Bent et al. [65].

Discussion

All measured CEMS spectra were fitted with the fixed relative peak intensities 3:4:1:1:4:3 for each Zeeman hyperfine pattern. This means that the angle between the magnetization direction and the propagation of the γ-rays was 90°. This is satisfied because in our experiments the γ-ray propagation was normal to the film surface and all Fe films were magnetized in the film plane.

The CEMS spectra in Fig. 5.1 for the TD and PLD films showed a broad hyperfine field distribution for the 57Fe probe layer grown directly on the GaAs(001) template. 8-10 Mössbauer components were required to account for the measured spectra. The relative contributions of the spectral components and the corresponding hyperfine fields \( H_f \) have been summarized in Fig. 5.2 and Table 5.1. The main difference between the TD and PLD samples was observed at the lowest and the highest range of the \( H_f \) components. Otherwise the components between these extremes were remarkably similar.

In order to compare the interface quality of TD and PLD deposited films it is convenient to calculate the average number of the Fe atoms \( \langle N \rangle \) as a nearest neighbors with respect to the Fe in the bcc lattice. This can be achieved by summing all relative contributions of the Mössbauer spectra components multiplied by their n.n Fe occupation number (see Table 5.1). For the GaAs/\(^{57}\text{Fe}_{\text{TD}}/8\text{Fe}_{\text{TD}}/20\text{Au}(001)\) sample the
Figure 5.1: Conversion electron Mössbauer spectra (CEMS) for a) TD and b) PLD samples. The thin solid lines represent the individual components of the Mössbauer spectra corresponding to the different chemical environments for the $^{57}\text{Fe}$ atom. The thick solid line represents the overall fit to the data.

Calculations lead to the following result:

$$\langle N \rangle = (0.24 \times 8) + (0.23 \times 7) + (0.14 \times 6) + (0.14 \times 5) + (0.05 \times 4) \quad (5.2)$$

$$+ \quad (0.04 \times 3) + (0.03 \times 2) + (0 \times 1) \cong 5.4$$

Similar calculations for PLD deposited films using data from Table 5.1 resulted in $\langle N \rangle \cong 4$. It is interesting to note that if the $^{57}\text{Fe}$ formed 2 ideal monolayers on ideal GaAs surface, the maximum $\langle N \rangle = 6$. On the other hand, assuming that during the deposition 1 of 2 monolayers of $^{57}\text{Fe}$ becomes completely incorporated into the GaAs lattice, then $\langle N \rangle = 2$. Those are however two extreme situations which can be used as a indication of the perfect and rough interface formation. Clearly in the TD sample $\langle \langle N \rangle = \ldots \rangle$
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<table>
<thead>
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<th>nearest neighbor (n.n)</th>
<th>TD</th>
<th>PLD</th>
<th>Comments</th>
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<tr>
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<td>322</td>
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</tr>
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<tr>
<td>center component&lt;sup&gt;b&lt;/sup&gt;</td>
<td>85.6</td>
<td>38.1</td>
<td>$H_f$ free parameter</td>
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</tbody>
</table>

Table 5.1: Fitting results of the Mössbauer spectra from the data shown in Fig. 5.1. Relative contributions (RI) of the fitted Mössbauer spectra components in % and their magnetic hyperfine fields ($H_f$) correspond to the particular configuration of the As or Ga atoms as the nearest Fe neighbors (n.n) in the bcc Fe structure. Integers in the brackets show the number of substituted Fe atoms in the bcc lattice by either As or Ga atoms. (0.0) labels the configuration for ideal bcc Fe crystal with 8 Fe atoms as a n.n.

<sup>a</sup>$H_f = 322$ kOe was fixed for fitting. It decreased $\Delta H_f = -23$ kOe for TD and $\Delta H_f = -24$ kOe for PLD samples with adding next As or Ga atom as the nearest neighbor (n.n) of Fe atom.

<sup>b</sup>$H_f$ was a free fitting parameter in order to obtain the best overall fits. The lowest hyperfine field components (around zero mm/s in Fig. 5.1) correspond to the Fe site having a non-magnetic environment.

5.4) the Fe/GaAs interface is somewhat closer to ideal interface then this deposited by PLD method ($\langle N \rangle = 4$). This supports the view that high kinetic energy of PLD deposited Fe particles could lead to a stronger intermixing and incorporation of Fe into the subsurface of the GaAs. This argument can be further supported by observing big relative contribution of the lowest hyperfine field component for PLD film, see Fig. 5.1 a) and Table 5.1. For the PLD film the lowest $H_f \approx 38$ kOe, and represents 22 % in the relative intensity (RI); while for the TD film $H_f \approx 86$ kOe and RI=11 %. The lowest hyperfine field components in both TD and PLD samples correspond to Fe sites having a non-magnetic atomic environment.

The observed multitude of hyperfine field components in Fig. 5.1 clearly demonstrates a complex transition from the GaAs substrate to the Fe film unaffected by interface mixing. This is in agreement with the recent STM images [106] indicating that during...
the deposition some of the Fe atoms penetrate into the GaAs(001) substrate. These Fe atoms substitute for the Ga atoms in the second layer and displace them into an interstitial position. In addition, it was suggested that the Fe atoms inside the top As layer together with the top Fe layer could form Fe$_2$As compound seed crystals for the bcc Fe growth.

Fitting the CEMS spectrum for the TD GaAs/(5+2$^{57}$+3)Fe$_{TD}$/20Au(001) film was significantly improved by adding a second component with $H_f=304$ kOe and $R_I=28\%$ to the main fitted component with $H_f=322$ kOe and $R_I=72\%$ as shown in Fig. 5.3, a). The $H_f=304$ component could be attributed to a state with seven Fe atoms in the n.n. configuration. This state has not been observed in the growth of Fe on Ag(001) substrates prepared from a single crystal Ag boule [109]. There is an essential difference between these two structures. During the TD growth of Fe on GaAs(001) the interfacial As acts partly as a surfactant floating on top of the Fe film. Ag in the Ag/Fe(001) structure also acts as a surfactant but it is known that the Ag definitely is not incorporated into the Fe film. The component with $H_f=304$ kOe in the GaAs/(5+2$^{57}$+3)Fe$_{TD}$/20Au(001) sample therefore could indicate that some of the surfactant As was either left inside the Fe film or created some lattice defect sites. Considering that the shift in the hyperfine field is less than 23.5 kOe the later case is more probable.

In fact even for the GaAs/(7+2$^{57}$+7)Fe$_{TD}$/20Au(001) sample a two component
Figure 5.3: CEMS spectra for (a) GaAs/(5+2^{57}+3)/Fe\textsubscript{TD}/20Au(001) and (b) GaAs/(7+2^{57}+7)Fe\textsubscript{TD}/20Au(001) samples. Two component fits were required for both samples with the following hyperfine field ($H_f$) and relative intensities (RI) parameters: (a) GaAs/(5+2^{57}+3)Fe\textsubscript{TD}/20Au(001); 1. $H_f=322$ kOe, RI=72 %, 2. $H_f=304$ kOe, RI=28 %. (b) GaAs/(7+2^{57}+7)Fe\textsubscript{TD}/20Au(001); 1. $H_f=326$ kOe, RI=82 %, 2. $H_f=310$ kOe, RI=18 %.

Fit was required with the following parameters (see Fig. 5.3, b): (1) $H_f=326$ kOe and RI=82 %; (2) $H_f=310$ kOe and RI=18 %. Obviously the higher $H_f$ values measured for sample(b) were closer to the ideal bulk field ($H_f=331$ kOe). A somewhat lower values of $H_f$ at RT compared with the bulk value of $H_f$ is expected. Ultrathin films have a lower critical point than bulk Fe, and $H_f$ strongly depends on the film thickness.

5.2 Magnetic anisotropies in Fe multilayers

Magnetic properties in ultrathin magnetic structures can be described using the bulk and interface contributions, see section 2.1.3

5.2.1 Au/Fe/GaAs structures

The magnetic anisotropies of thin Fe layers grown directly on (4×6)-GaAs(001) substrates using TD were investigated by means of FMR. A series of magnetic single Fe layers ranging in thickness from 1.5 to 39 nm were prepared: 20Au/nFe/GaAs(001), where the
integers represent the number of atomic layers (ML) and \( n = 10, 16, 25, 30, 40, 70, 100, 270 \). The in-plane angular dependence of the FMR field, Fig. 5.4, was used to determine the in-plane four-fold and uniaxial anisotropies \( K_\parallel \) and \( K_U \), and the effective demagnetizing field perpendicular to the film surface \( 4\pi M_{\text{eff}} \). The effective magnetic anisotropy and

\[
\begin{align*}
&\quad \text{Figure 5.4: The angular dependence of the resonant magnetic field} \ H_{\text{res}} \ \text{at 24 GHz. (a)} \ 20\text{Au}/16\text{Fe}/\text{GaAs(001)}. \ (b) \ 20\text{Au}/70\text{Fe}/\text{GaAs(001)}. \ \text{Both specimens were grown using TD. The angle of the applied magnetic field,} \ \varphi_H, \ \text{was rotated in the plane of the film and measured with respect to the [100]}_\text{Fe} \ \text{crystallographic direction. The data are plotted using the open squares. The contributions of the four-fold} \ K_\parallel \ \text{and uniaxial} \ K_U \ \text{magnetic anisotropies are shown using the dashed and thin solid lines, respectively. The thick solid line is a computer fit to the data using the following magnetic parameters: (a)} \ 20\text{Au}/16\text{Fe}/\text{GaAs} - K_\parallel = 2.61 \times 10^5 \ \text{erg/cm}^3, \ K_U = -5.01 \times 10^5 \ \text{erg/cm}^3, \ 4\pi M_{\text{eff}} = 16.8 \ \text{kOe}; \ (b) \ 20\text{Au}/70\text{Fe}/\text{GaAs} - K_\parallel = 3.77 \times 10^5 \ \text{erg/cm}^3, \ K_U = -0.2 \times 10^5 \ \text{erg/cm}^3, \ 4\pi M_{\text{eff}} = 19.6 \ \text{kOe}. \ \text{The} \ g \ \text{factor was fixed for fitting at} \ g = 2.09 \ \text{and the hard axis of the in-plane uniaxial anisotropy lies along the [110] direction. Clearly the 70 ML thick Fe film and the 16 ML Fe film have different magnetic anisotropies.}
\end{align*}
\]

\( 4\pi M_{\text{eff}} \) values for all samples as a function of \( 1/d_{\text{Fe}} \) are shown in Fig. 5.5. The intercept with the y-axis corresponds to an infinite thickness and therefore represents the bulk anisotropy. The slope arises from the interface contributions, see Eq. 2.13. The saturation magnetization, \( 4\pi M_S \), can be obtained from the effective demagnetizing field
CHAPTER 5. PROPERTIES OF ULTRATHIN Fe FILMS

Figure 5.5: The effective demagnetizing field perpendicular to the film surface $4\pi M_{\text{eff}}$ (a) and the effective in-plane fourfold (cubic) $K_1^\parallel$ and uniaxial $K_1^\perp$ anisotropies (b) plotted as a function of $1/d$ for single Fe films, where $d$ is the film thickness. The Fe films were grown on GaAs(001) and covered by 20 ML of Au(001). All films were grown using the TD technique, except as noted. Solid lines are the linear fits to the data. The intercept with the y-axis corresponds to infinite thickness and represents the bulk ($B$) anisotropy. The slope is related to the interface contribution $(\text{int})$ of the anisotropy constants. The resulting values of $K_1^\parallel_B$, $K_1^\parallel_\text{int}$, $K_1^\perp_\text{int}$ and $4\pi M_S$ are reported in Table 5.2. The (o) symbols in (b) show the measured values of $K_1^\parallel$ for the 20Au/(7,10,15)Fe/GaAs(001) samples grown using the PLD method and were not included in the fitting, (see further discussion in the text).

extrapolated to infinite thickness (intersect with the y axis ).

Values for the bulk and interface anisotropies using the data from Fig. 5.5 are shown in Table 5.2. From Table 5.2 one can see that the magnetic anisotropies projected to infinite thickness are nearly equal to anisotropies found in bulk Fe ($K_1^\parallel = 4.7 \times 10^5$ erg/cm$^3$ [110]). The in-plane 4-fold bulk and interface anisotropies $K_1^\parallel_B$ and $K_1^\parallel_\text{int}$ were found to be nearly equal to those reported by Urban et. al [39] and Brockman et al. [111].

The measured values of $K_1^\parallel_\text{int}$, $K_1^\perp_\text{int}$ and $4\pi M_S$, see Table 5.2, deserve more comments and will be discussed in the following sections.
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Table 5.2: The measured values of the anisotropy constants for the 20Au/nFe/GaAs(001) samples prepared by means of thermal deposition (TD). The superscripts (int) and (B) represent the interface and bulk contributions to the magnetic anisotropies. The interface anisotropies include contributions from the Au/Fe and Fe/GaAs(001) interfaces.

<table>
<thead>
<tr>
<th>Fe thickness ML</th>
<th>$K_{11,B}^{|}$ ($10^5 \text{ erg/cm}^3$)</th>
<th>$K_{11,\text{int}}^{|}$ (erg/cm$^2$)</th>
<th>$K_{U}^{|,\text{int}}$ (erg/cm$^2$)</th>
<th>$K_{U}^{\perp,\text{int}}$ (erg/cm$^2$)</th>
<th>$4\pi M_S$ (kOe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10, 16</td>
<td>$4.2 \pm 0.1$</td>
<td>$-0.03 \pm 0.003$</td>
<td>$-0.10 \pm 0.01$</td>
<td>$0.7 \pm 0.3$</td>
<td>$20.4 \pm 0.2$</td>
</tr>
<tr>
<td>30, 40, 70, 100</td>
<td>$4.2 \pm 0.1$</td>
<td>$-0.03 \pm 0.003$</td>
<td>$-0.03 \pm 0.01$</td>
<td>$0.7 \pm 0.3$</td>
<td>$20.4 \pm 0.2$</td>
</tr>
<tr>
<td>270</td>
<td>$4.2 \pm 0.1$</td>
<td>$-0.03 \pm 0.003$</td>
<td>$-0.03 \pm 0.01$</td>
<td></td>
<td>$21.4 \pm 0.2$</td>
</tr>
</tbody>
</table>

Uniaxial perpendicular anisotropy and $4\pi M_S$

Fe(001) ultrathin films exhibit large uniaxial anisotropies in which the easy axis is perpendicular to the film surface, see Eq. 5.4. FMR measurements [112] and Figure 5.5 a), have shown that the effective demagnetizing field includes a term that is inversely proportional to the film thickness. These results clearly demonstrate that the uniaxial perpendicular anisotropy, $K_{U,\text{int}}^{\perp}$, in the bcc Fe structures is indeed an intrinsic effect caused by the broken symmetry at the interfaces and depends strongly on the interface composition (see Sec. 5.2.2 and 5.2.3 for a further discussion).

The saturation magnetization value, for measurements carried out in the thickness range from 10 to 100 ML (1.5-15 nm), was found to be $20.4 \pm 0.2$ kOe, see Fig. 5.5 a) and Table 5.2. This leads to $M_S = 1630 \text{ emu/cm}^3$ which is slightly lower than that expected for bulk Fe ($M_S = 1710 \text{ emu/cm}^3$, $4\pi M_S = 21.5$ kOe [113]). However for the 270 ML (40 nm) thick Fe film the $4\pi M_{\text{eff}}$ value, 21.4 kOe, was found indeed to be in agreement with an Fe bulk value of $21.4 \pm 0.1$ kOe, see Fig 5.5 a). Clearly another term had to be included in order to explain the lower value of $4\pi M_S$ for samples thinner than 15 nm. It has been shown by Gordon et al. [114, 115] using X-ray absorption fine structure measurements (XAFS) on ultrathin Fe/GaAs(001) structures that the Fe films grown on GaAs(001) are tetragonally distorted relative to bulk bcc Fe. The in-plane lattice spacing follows the GaAs(001) lattice mesh and consequently the Fe lattice is vertically expanded to keep the atomic volume constant. The $c/a$ ratio for a 10 ML thick Fe film was found to be $1.03 \pm 0.02$. Due to the magneto-elastic coupling this strain in the Fe film results in an additional bulk-like uniaxial anisotropy field perpendicular to the film.
surface that affects the effective demagnetizing field, [14, 114, 116].

\[
4\pi M_{\text{eff}} = 4\pi M_S - \frac{2K_{U}^{L,S}}{M_S d_{Fe}} + \frac{2B_1(e_- - e_\parallel)}{M_S}
\]  

(5.4)

where \(B_1\) is the magneto-elastic coupling coefficient (\(B_1 = -3.43 \times 10^7\) erg/cm\(^3\) for Fe [114]) and \(e_\parallel\) and \(e_-\) are the strains parallel and perpendicular to the Fe film plane, respectively. If one uses the values \(e_\parallel = (-0.011 \pm 0.004)\) and \(e_- = (0.022 \pm 0.010)\) taken from [114], the tetragonal distortion due to the lattice mismatch between the Fe and GaAs(O01) lattice meshes results in a magneto-elastic field of \(-1.3 \pm 0.5\) kOe.

The \(e_\parallel\) and \(e_-\) strains can be also evaluated according to elasticity theory [117, 118]. From the condition that the film must be stress free along the film normal the ratio of the out-of-plane to in-plane strain is given by

\[
\frac{e_-}{e_\parallel} = -\frac{2C_{12}}{C_{11}} = -1.212
\]  

(5.5)

where \(C_{11} = 2.41 \times 10^{12}\) ergs cm\(^{-3}\) and \(C_{12} = 1.46 \times 10^{12}\) ergs cm\(^{-3}\) are the known elastic constants [116]. An in-plane contraction of the Fe lattice \((a = 2.866\) Å) can be understood as an effort to match the Fe lattice to the underlying GaAs lattice \((a/2 = 2.827\) Å). Since \(e_\parallel = (a_{sub} - a_{film})/a_{film} = -0.0136\) Å then the out-of-plane expansion can be found using Eq. 5.5 which results in \(e_- = 0.0165\) Å. The calculated \(e_\parallel\), \(e_-\) and \(B_1 = -3.43 \times 10^7\) erg/cm\(^3\) results in a magneto-elastic field of \(-1.2\) kOe thereby decreasing the value of \(4\pi M_{\text{eff}}\) to 20.3 kOe. From the projected values of the effective \(4\pi M_{S,\text{eff}} = 20.37\) kOe, see Fig 5.5 a), one can conclude that this tetragonal distortion of the Fe lattice remains remarkably constant for the films less than 100ML (15 nm) thick.

**In-plane uniaxial anisotropy**

The data in Fig 5.4 have shown that ultrathin Fe(001) films grown on GaAs(001) exhibit a uniaxial anisotropy of strength \(K_U^{L}\) (thin solid line) with the hard axis parallel with the [110]$_{Fe}$ axis. This uniaxial anisotropy is superimposed on the cubic anisotropy \(K_U\) (dashed line) with the easy axes directed along the \(<100>_{Fe}\) crystallographic directions. By comparing the anisotropy constants for 16 ML and 70 ML of Fe grown on GaAs(001), see Fig 5.4 a) and b), it is evident that the relative strength of \(K_U^{L}\) compared to \(K_U\) decreased with the film thickness, see also Fig 5.5. However, a small residual \(K_U^{L}\) is still present in the 70 ML Fe film.
The occurrence of a uniaxial in-plane anisotropy has attracted considerable scientific interest, but a conclusive answer as to its precise origin is still missing [90]. One of the possible mechanisms suggested to explain the origin of $K_U^{\|}$ was coupling of the interface Fe electronic structure to the substrate dangling bonds (sp3 like bonding) [119]. The GaAs(001) lattice template has uniaxial symmetry due to dangling bonds associated with the $(2 \times 6)$ reconstructed surface in which As dimers run along the [110] direction. On the contrary, it was shown by Moosbühler [120] that the uniaxial anisotropy does not depend upon variations in GaAs surface stoichiometry associated with different reconstructions of the GaAs substrate. This suggests that the electronic structure at the interface can not explain the underlying mechanism for the observed in-plane uniaxial anisotropy. This result, is however, not in agreement with the recent work of Muermann et. al [121]. In their studies on Fe(110) and FeCo(110) grown on GaAs(110) they claim that the source of the interface uniaxial anisotropy is related to the orientation of Ga and As dangling bonds projected onto the GaAs(110) surface.

The data presented in Fig 5.5 b) by the solid squares revealed that the uniaxial in-plane anisotropy $K_U^{\|}$, appeared in the initial stages of the film growth, and from about 10 ML onwards its thickness dependence can be fit with a $1/d_F$ dependence. This clearly suggests its interfacial origin. $K_U^{\|}$ is caused by the Fe/GaAs(001) interface since neither the interface between Fe and vacuum nor the interface between Fe and Au or Ag have shown to yield an appreciable $K_U^{\|}$ (see Sec. 5.2.3). For the 10 and 16 ML thick Fe films the uniaxial anisotropy $K_U^{\|}$ is large. In this thickness range the interface in-plane uniaxial anisotropy $K_U^{\|,int} = -0.10 \pm 0.01$ erg/cm$^2$. It is interesting that the interface uniaxial in-plane anisotropy values, $K_U^{\|}$ quoted in Ref. [39] and [111], in the thickness range from 10 to 30 ML were about 25% larger than those listed in Table 5.2. Ar$^+$ ion sputtering of the GaAs substrate for several hours at RT was used by Urban et al. [39], and sputtering at an elevated temperature was used by Brockman et al. [111]. This indicates that $K_U^{\|}$ is sensitive to the details of the GaAs cleaning and preparation procedure. The bulk contribution to the uniaxial anisotropy $K_U^{\|,B}$ was found to be very small and therefore was assumed to be zero. This confirms the purely interfacial character of $K_U^{\|}$. The interface contribution to the in-plane uniaxial anisotropy dramatically decreases for Fe films thicker than 30 ML, see Fig. 5.5 (b). The corresponding surface uniaxial anisotropy $K_U^{\|,int} = -0.03 \pm 0.01$ erg/cm$^2$ is almost an order of magnitude smaller than that for the thinner films (see Fig. 5.5, b)). The negative sign of $K_U^{\|,int}$ indicates that the hard axis
remains oriented along the \([1\overline{1}0]_F\)Fe direction across the whole measured thickness range.

A significant decrease of the interface uniaxial in-plane anisotropy for thicker films suggests that the interface uniaxial anisotropy is not caused by purely chemical interactions between the Fe and GaAs as has been so far claimed [121]. This is further supported by a dramatic change in \(K_{U}^{\|,int}\) in Fe films covered by Cr, [122]. A Cr cover layer deposited far away from the Fe/GaAs(001) interface can completely remove the in-plane uniaxial anisotropy. The recent observation by Gordon et. al [107] of interface shear at the GaAs/Fe(001) interface suggests that the in-plane interface uniaxial anisotropy can be explained by a magneto-elastic term \(B_2\) due to the lattice shear. Their polarization-dependent XAFS studies on a 2 ML Fe film have shown that the Fe nearest-neighbor distances along the \([-110]\) direction is larger than that along the \((110)\) direction due to an interface lattice shear. This lattice shear has the right sign to produce a uniaxial anisotropy, \(K_{U}^{\|,int}\), with a hard axis oriented along \([-110]_F\) [123]. Further indirect evidence for an interface shear was found in the Fe films grown by PLD. For the 7,10 and 15 ML Fe films grown by PLD on GaAs(001), see Fig 5.5, b), the in-plane interface uniaxial anisotropy \(K_{U}^{\|,int}\) was found to be the same as that found for thick Fe films grown by TD, see Fig. 5.5 (b). Our recent Moessbauer studies have shown that Fe films prepared by PLD exhibit a more intermixed interface than those prepared by TD, see further details in Sec. 5.1. Consequently the Fe lattice in the PLD films can be expected to be clamped more strongly to the GaAs(001) lattice than that in films grown by TD. The significant decrease in the in-plane interface uniaxial anisotropy in the PLD films can then be explained by a decrease in the lattice shear. In fact the PLD deposition affected all Fe film magnetic anisotropies. This is described in the next paragraph.

5.2.2 TD vs. PLD

FMR measurements have shown that Fe films grown by TD and PLD have different anisotropies, see Table 5.3 and Fig. 5.6 The in-plane uniaxial anisotropy is significantly different for the PLD film compared with the TD films and the four-fold anisotropy for the PLD film even exhibits an overall negative \(K_1^{\|}\) even for a 10ML thick Fe film: the PLD in-plane 4-fold anisotropy parameter has the opposite sign compared with \(K_1^{\|,B}\) for bulk iron.
Figure 5.6: The FMR field, $H_{FMR}$, at $f=24$ GHz as a function of the in-plane angle, $\varphi_H$, of the external field with respect to the [100]$_{Fe}$ axis. GaAs(001)/10Fe/20Au(001) samples were prepared by (a) TD and (b) PLD depositions. The dashed and dotted lines are the calculated resonance field due to the anisotropies $K_1$ and $K_2$, respectively. The solid lines represent the overall fit. The insets show the corresponding angular dependence of the FMR linewidth, $\Delta H$.

$4\pi M_{eff}$ decreased by less than 1 kG in the GaAs/2Fe$_{PLD}$8Fe$_{TD}$/20Au(001) sample compared to the film grown entirely by TD, whereas in the GaAs/10Fe$_{PLD}$/20Au(001) structure $4\pi M_{eff}$ decreased by approximately 5 kG relative to the GaAs/10Fe$_{TD}$/20Au(001) structure. This means that the interface uniaxial perpendicular anisotropy was mostly increased at the Fe/Au(001) interface. The thickness dependence has shown that the total perpendicular uniaxial interface anisotropy was increased from 0.7 erg/cm$^2$ for the TD deposited films to 1.3 erg/cm$^2$ for the PLD films.
Sample & $K_1^\parallel$ & $K_U^\parallel$ & $4\pi M_{\text{eff}}$ \\
\hline
GaAs/25$^\text{Fe}_{\text{TD}}$/8Fe$_{\text{TD}}$/20Au(001) & 2.02 & -7.01 & 14.74 \\
GaAs/25$^\text{Fe}_{\text{PLD}}$/8Fe$_{\text{TD}}$/20Au(001) & 0.73 & -1.755 & 13.92 \\
GaAs/25$^\text{Fe}_{\text{PLD}}$/8Fe$_{\text{PLD}}$/20Au(001) & -0.43 & -2.25 & 9.60 \\
\hline

Table 5.3: The measured values of the effective anisotropy constants for the 10 ML Fe films prepared by thermal deposition (TD) and pulsed laser deposition (PLD). The subscripts represent the deposition method.

### 5.2.3 Au/Fe/Au and Au/Fe/Ag structures

The static magnetic properties of Fe depend on the substrate templates. The magnetic anisotropies for 12 ML Fe grown on Au(001) and Ag(001) templates are shown in Table 5.4. The in-plane uniaxial anisotropy $K_U^\parallel$ was found to be very small and to have the same value for Fe films deposited on both Au(001) and Ag(001) templates. The positive sign of $K_U^\parallel$ shows that the easy axis was oriented along [110]$_{Fe}$ i.e rotated -90 degrees with respect to the easy axis ([110]$_{Fe}$) for an Fe film grown on GaAs(001), see Fig. 5.4. The in-plane four-fold anisotropy parameter, $K_1^\parallel$, was larger for an Fe film grown on a Ag(001) template than for a film deposited on a Au(001) template. This suggests that the interface contribution, $K_1^{\parallel \text{int}}$, was smaller at the Ag/Fe interface than at the Au/Fe interface. The smaller value of $4\pi M_{\text{eff}}$ obtained for the Fe film grown on Ag indicated that the uniaxial interface perpendicular anisotropy $K_U^{\perp \text{int}}$ was increased at the Ag/Fe interface relative to the Au/Fe interface [14].

Table 5.4: The measured values of the anisotropy constants for a 12 ML Fe film grown on Au(001) and Ag(001) templates.
Chapter 6

Damping in thin magnetic films

Ferromagnetic Resonance linewidth ($\Delta H$) measured as a function of microwave frequency provides information about the magnetic damping in thin ferromagnetic films. In this chapter the intrinsic and extrinsic contributions to Gilbert damping will be discussed.

6.1 Intrinsic Gilbert damping

Magnetic single crystalline ultrathin films are suitable candidates for the study of the intrinsic magnetic damping. A set of 20Au/nFe/GaAs samples was prepared by means of MBE, where $n=8, 10, 13, 16, 20, 25, 30$. The integers represent the number of atomic layers. The FMR linewidth ($\Delta H$) as a function of microwave frequency ($f$) was used to extract the damping parameter $\alpha$, see Fig. 6.1 (a) and (b). $\Delta H(f)$ for the Fe film thickness in the range from 8 to 16 ML is shown in Fig. 6.1 (a). $\Delta H$ was found to be proportional to the frequency in the range of microwave frequencies from 9.1 to 73 GHz with no appreciable zero frequency offset ($\Delta H(0)$). This behavior is characteristic for Gilbert damping described by Eq. 2.18, the FMR linewidth $\Delta H$ is given by Eq. 2.36; $\Delta H = \alpha \frac{\omega}{\gamma} = \frac{\omega}{\gamma} \frac{G}{\gamma M_s}$.

The $\Delta H(f)$ dependence for the 20, 25, and 30 ML Fe samples with the magnetization oriented along the hard magnetic axis $[\overline{1}0\overline{1}]_F$ had to be analyzed with caution. A clear downturn in linewidth was observed for frequencies less than 24 GHz. The solid lines in Fig. 6.1(b) indicate that $\Delta H$ approaches zero at low microwave frequencies, see [25]. This nonlinear contribution to the frequency dependence of the damping is typical for
Figure 6.1: (a) and (b): FMR linewidth ($\Delta H$) as a function of the microwave frequency in the 20Au/nFe(001)GaAs(001) single Fe layers, where n=8,10,13,16,20,25, and 30. The dashed lines show linear fits to the data points. Notice that in b) the linear fit was carried out using only the linewidths at 24 and 73 GHz. $\Delta H(f)$ was measured for the applied magnetic field directed along the [1\bar{1}0]$_{Fe}$ hard magnetic axis. (c) Thickness dependence of the magnetic damping parameter $\alpha$ obtained from the data in (a) and (b). The (o) points correspond to the magnetization oriented along the [1\bar{1}0]$_{Fe}$ axis. The solid line represents a fit to the data using the equation: $\alpha = \alpha_b + \alpha_{int}(1/d_{Fe})$, where $d_{Fe}$ is the Fe film thickness. The dashed line shows the fit using the equation $\alpha = \alpha_b + \alpha_{int}(1/d_{Fe})$. The legend shows the asymptotic values ($d_{Fe} \to \infty$) of $\alpha = \alpha_b$ and the corresponding Gilbert relaxation times obtained from the fits.
two magnon scattering and will be addressed further in the following section 6.2. For the thick Fe layers, see Fig. 6.1(b), the intrinsic damping contribution was obtained from the slope of the dashed lines which were drawn between the frequencies 24 and 73 GHz. In this frequency range the contribution of two magnon scattering usually reaches a plateau and the frequency dependence of the FMR linewidth $\Delta H(f)$ is expected to be due only to Gilbert damping [58]. The increased value $\Delta H(0)$ is caused by the two-magnon scattering mechanism. The thickness dependence of $\alpha$ is shown in Fig. 6.1(c) by the ($\circ$) symbols. The solid and dashed lines show computer fits to the data assuming that $\alpha$ arises from the bulk and interface contributions. The solid and dashed lines were obtained from the fitting ansatz $\alpha(d) = \alpha_b + \alpha_{int}/d^2$ and $\alpha(d) = \alpha_b + \alpha_{int}/d$, respectively. The $\alpha_b$ and $\alpha_{int}$ represents the bulk and interface contributions to the damping, respectively.

The $\alpha$ damping parameters were obtained also for applied magnetic fields directed along the $[110]_{Fe}$ and $[100]_{Fe}$ crystallographic axes. The deviations of $\alpha$ along those directions were found to be within 5% showing that the angular dependence of the damping is weak and mostly dependent on the film thickness.

The experimental data could be well fit in the thickness range from 10 to 30 ML using both of the above fitting procedures, see Fig. 6.1(c). The fitting models however led to an appreciably difference in the values of $\alpha_b$: $\alpha_b = 2.5 \times 10^{-3}$ (solid line fit) and $\alpha_b = 1.2 \times 10^{-3}$ (dashed line fit), see also Fig. 6.1. This corresponds to bulk Gilbert damping rates of $G = 0.78 \times 10^8 s^{-1}$ and $G = 0.38 \times 10^8 s^{-1}$, respectively. The model which assumes a $1/d_{Fe}$ dependence leads to a very small extrapolated value for the bulk Gilbert damping parameter that is a factor of two smaller than that found in the case of the $1/d_{Fe}^2$ dependence. Such a small value for $G$ is even smaller than the smallest bulk damping parameter reported by Frait et. al [124] for pure Fe single crystal platelets, $G = 0.42 \times 10^8 s^{-1}$; it is also smaller than the damping parameter recorded for single crystal bulk Fe ($G = 0.58 \times 10^8 s^{-1}$) [38]. On the other hand, $G = 0.78 \times 10^8 s^{-1}$ is only $\sim 20\%$ larger than the value of the damping parameter obtained by Heinrich et. al [112] ($G = 0.66 \times 10^8 s^{-1}$) for ultrathin Fe films grown on Ag(001) substrates which were prepared from a high quality crystalline Ag boule. It is interesting that their measurements of the damping parameter in the same thickness range (7-40 ML of Fe) showed that the damping parameter $\alpha$ also included a $1/d_{Fe}^2$ term.
The origin of this rapid increase in the magnetic damping with decreasing film thickness is unknown at this time. The dependence on $1/d^2_{Fe}$ suggests that its origin can be ascribed to a two magnon scattering mechanism caused by an inhomogeneous interface perpendicular uniaxial field which scales as $1/d$. The two magnon contribution scales with the squared power of the inhomogeneous field which consequently can lead to the observed $1/d^2_{Fe}$ dependence of the damping parameter, see section 2.4.3. However this is not the only possible explanation. Safonov and Bertram [125] pointed out that lattice point defects can lead to an additional Gilbert-like damping term. The diffusive background in the RHEED patterns observed for the thinnest Fe samples indicate that point defects are present at the initial stages of the Fe growth. These defects are concentrated near the GaAs/Fe(001) interface and can be expected to play a role in damping that could lead to a $1/d$ thickness dependence. It is not possible at this point to conclude which thickness dependence is more realistic. More experimental points are required and FMR measurements have to be carried out for both the in-plane and perpendicular FMR configurations. The two-magnon scattering is absent in the perpendicular FMR and this can distinguish between $1/d$ and $1/d^2$ contribution to the damping, see sec. 2.4.3. This task however lies outside the scope of this thesis and will be pursued later on.

### 6.2 Extrinsic damping-two magnon scattering

The two-magnon scattering model discussed in chapter 2, sect. 2.4.3 can be used to interpret the strong extrinsic contribution to the magnetic damping (FMR linewidth) in Au/Pd/Fe/GaAs(001) multilayers. The Pd(001) in-plane lattice constant has a large 4.4% mismatch with respect to the Fe(001) template even when the Pd template is rotated by $\pi/4$ with respect to the Fe [100] crystallographic direction. Au grows over Pd with the in-plane cubic axes parallel to each other ([100]Au || [100]Pd). The Pd and Au lattice spacings are mismatched by 4.5%. The presence of Pd in crystalline Au/Pd/Fe(001) multilayers provides a large lattice strain. For Pd film thicknesses greater than 4 ML this strain is relieved by the formation of misfit dislocations. The misfit dislocations form a rectangular network of lattice defects which create magnetic inhomogeneities that strongly affect the spin dynamics. Previous studies [25, 126] have shown that for sufficiently thick Pd layers dislocations can propagate right through the underlying Fe layer. The formation of magnetic defects in Fe layers grown directly on
GaAs(001) requires either a thick Pd layer \((n > 130)\) or a combination of a thin Pd layer \((n \geq 9)\) and a thick Au layer \((n > 70)\). For an Fe layer deposited on the Pd template the onset of two-magnon scattering occurs right after a misfit dislocation network is formed in Pd. Our FMR studies were carried out on the magnetic bilayer structure 20Au/40Fe/140Au/11Pd/16Fe/GaAs(001).

In such a structure several damping mechanisms could be observed. Firstly, the misfit dislocations network introduces two-magnon scattering. Secondly, spin pumping leads to the loss of spin momentum in the Fe layers. The loss of spin momentum is caused by the spin sink mechanism in the adjacent Fe layers and spin lattice relaxation in the thick Au spacer, see further discussion in sections 6.2.2 and 7.3.

### 6.2.1 STM studies

Epitaxial growth for all layers in the 20Au/40Fe/140Au/11Pd/16Fe/GaAs(001) structure was confirmed by the observation of RHEED specular spot intensity oscillations. The STM images shown in Fig. 6.2 (a) revealed the presence of line defects after deposition of 40ML of Au on the 11Pd/10Fe/GaAs(001) sample. These lines are indicated by the white arrows and are oriented along the \((110)_{\text{Au}}\) crystallographic directions. They are separated by \(\sim 10 - 15\)nm. The rectangular network of line defects on the surface is caused by the nucleation and glide of misfit dislocation half-loops that enter the crystal surface during film growth. The Au surface otherwise consists of rectangular shaped terraces. This is different from the case of the Au/Fe/GaAs(001) surface for which round terraces were observed, see Fig 4.7. Figure 6.2 (b) shows an STM image of the Au surface for the 20Au/20Fe/130Au/11Pd/10Fe/GaAs(001) sample. In this case the dislocation lines are not clearly visible and this suggests that the defects did not propagate through the thick Au \((26\) nm) and the top Fe layer. It follows that the lattice defects did not fully develop in the top Fe layer. Since two-magnon scattering in this sample is associated with these lattice defects, one expects a significantly smaller extrinsic damping contribution to the damping in the Fe layer deposited on a thick Au film deposited on a Pd layer.
Figure 6.2: STM surface topography of the 40Au/11Pd/10Fe/GaAs(001) and 20Au/20Fe/130Au/11Pd/10Fe/GaAs(001) structures. The derivatives of the images are shown to enhance the contrast between the terraces (constant intensity) and the edges (bright or dark). (a) 150 × 150 nm² image of the 40Au/11Pd/10Fe/GaAs(001) sample. The dislocation lines oriented along the (110)_Au crystallographic directions are indicated by the white arrows. They can be distinguished as straight thin lines which run across the field of view of the image (a). These line defects are attributed to the glide planes of misfit dislocations that originate in the Pd layer due to the Pd/Fe and Au/Pd lattice mismatch. (b) STM image of the top Au surface in the 20Au/20Fe/(130Au/11Pd)/10Fe/GaAs(001) specimen. Dislocation lines were not observed in this case.

6.2.2 FMR studies

FMR measurements were carried out using two independent experimental setups. (1) A standard FMR spectrometer, using microwave waveguides, and operating at frequencies of 9.1, 24, 36 and 73 GHz, and (2) a network analyzer (NA) operating in the frequency range from 1 GHz to 24 GHz. NA was operated in the transmission mode and essentially functioned as a lock-in amplifier at variable microwave frequencies. NA was attached to a Coplanar Transmission line (CTL) having a central conductor width of 800 μm and the central and outer conductors were separated by 150 μm. The coplanar transmission line (CTL) plus the sample geometry are shown schematically in Fig 6.3. A detailed description of NA and the FMR setup can be found in greater detail in ref. [49].

Figure 6.4 (b) shows the dependence of the FMR resonance field, \( H_{FMR} \), on the
Figure 6.3: A schematic diagram of a coplanar transmission line (CTL). A magnetic sample is placed on the transmission line "face down" with the GaAs substrate facing the room. Only the part of the CTL covered by the sample is shown.

in-plane angle, $\varphi_H$, of the applied dc magnetic field for the 16Fe and 40Fe films in the 20Au/40Fe/140Au/11Pd/16Fe/GaAs(001) structure. Due to a large in-plane uniaxial anisotropy in the bottom 16Fe film grown directly on the GaAs(001) (open triangles) the resonance fields were well separated from the resonance fields for the top 40Fe film grown on Au (open circles). This allowed one to study the dynamic properties (linewidth) for each individual Fe layer separately. The magnetic anisotropies for the 16Fe and 40Fe films were found to be similar to those obtained for single Fe films grown on GaAs(001), see section 5.2 in chapter 5. The striking difference between the double and single layer films was observed in the FMR linewidth, see Fig. 6.4 (a). $\Delta H$ for the 40Fe film was practically independent of the angle between the magnetization and the crystallographic Fe axes. $\Delta H$ for the 16Fe film, however, exhibited a large angular dependence that followed a distinct four-fold symmetry. The minima in $\Delta H$ were observed when the applied magnetic field was oriented along the $\langle 110 \rangle_F e$ crystallographic directions of Fe, and the maxima in $\Delta H$ were observed when the field was oriented along the $\langle 100 \rangle_F e$ crystallographic directions of Fe. It is interesting that the minimum values of the FMR linewidths observed at 73 GHz for the 16Fe film ($\Delta H \sim 157$ Oe) and the 40Fe film ($\Delta H \sim 97$) were very nearly the same as those expected for the intrinsic Gilbert damping in the magnetic double layer including the spin pump contribution (dashed lines in Fig. 6.4(a)).
Figure 6.4: (a) The FMR linewidth $\Delta H$, at 73 GHz for the 16Fe (▼) and 40Fe (□) films in the 20Au/40Fe/140Au/11Pd/16Fe/GaAs(001) structure as a function of the in-plane angle $\varphi_H$ of the applied dc magnetic field. A large angular dependence for $\Delta H$ was observed for the 16Fe film. The narrowest lines occurred when the magnetic field was oriented along the (110) Fe crystallographic directions. The widest lines were observed when the magnetic field was oriented along the (100) Fe crystallographic directions. The horizontal dashed lines indicate the expected values of $\Delta H$ due to intrinsic Gilbert damping (including the spin pump contribution); $\Delta H \sim 157$ for the 16Fe film, and $\Delta H \sim 97$ for the 40Fe film.

(b) The FMR resonance fields, $H_{FMR}$, at 73 GHz for the 16Fe film (▼) and the 40Fe film (□) for the same sample as shown in (a) as a function of the angle $\varphi_H$. The 0° angle corresponds to the [100] Fe crystallographic direction. The [110] direction corresponds to the hard magnetic axis for both the 16Fe and 40Fe films.

In order to study the extrinsic contribution to the magnetic damping due to two-magnon scattering one needs to measure the FMR linewidth as a function of the microwave frequency, $\Delta H(f)$. The measurements of $\Delta H(f)$ along the (100)$_{Fe}$ and (110)$_{Fe}$ directions for the 16Fe and 40Fe films are shown in Fig. 6.5.

$\Delta H$ for 16Fe shown by (△) symbols along the (110)$_{Fe}$ hard magnetic axes exhibited the minimum value of $\Delta H$, see Fig. 6.4 (a), and was linearly dependent on $f$, see also Fig. 6.5. The zero frequency offset indicated no extrinsic contribution to the damping. The linear fit shown by the dashed line allowed one to obtain the effective intrinsic Gilbert damping parameter $\alpha_{eff} = 6.2 \pm 0.3 \times 10^{-3}$. This value turned out to be good in agreement with two contributions. The first contribution, $\alpha = 3.6 \times 10^{-3}$, was found for the 16Fe single layer films on GaAs(001), see Fig. 6.1, and is close to the bulk value $\alpha_b = 2.5 \times 10^{-3}$. 
Figure 6.5: The frequency dependence of the FMR linewidths for the 20Au/40Fe/140Au/11Pd/16Fe/GaAs(001) structure. The ▲ and ◇ symbols represent the frequency dependence of $\Delta H(f)$ for the 16Fe film measured along the [100]$_{Fe}$ and [110]$_{Fe}$ crystallographic directions, respectively; (see also Fig. 6.4(a)). The solid line is a spline fit to the data and is a guide for the eye. The dashed lines are the linear fits to the data for the 16Fe film in the range of microwave frequencies $f$ such that the slopes correspond to the intrinsic Gilbert damping. Notice that the slopes are nearly the same for both crystallographic directions. The effective Gilbert damping parameter obtained from the linear slopes is $\alpha_{\text{eff}} = 0.0062$. The (o) points show the frequency dependence of the linewidth, $\Delta H(f)$, for the 40Fe film. The linear slope (corresponding to the dashed line) resulted in the damping parameter $\alpha_{\text{eff}} = 0.0037$. The dotted line shows the variation of $\Delta H$ as a function of $f$ for the 40Fe layer calculated assuming an intrinsic Gilbert damping parameter given by the sum of the bulk parameter, $\alpha_b = 0.0026$, and the spin pump contribution, $\alpha^{sp} = 0.0011$ (see further details in the text).

The second contribution comes from the spin momentum relaxation inside the 150 ML thick Au/Pd spacer; its value was found to be $\alpha^{sp} = 2.6 \times 10^{-3}$, and will be discussed in section 7.3.

$\Delta H(f)$ for the 16Fe film was quite different when measured with the saturation magnetization along the ⟨100⟩$_{Fe}$ directions, see the (▲) symbols in Fig. 6.5. In this case $\Delta H$ cannot be described by a simple linear dependence on the microwave frequency. However, the slope of $\Delta H(f)$ for frequencies greater than 36 GHz was found to be equal to the slope expected for an effective intrinsic Gilbert damping parameter $\alpha_{\text{eff}} = 6.2 \pm 0.3 \times 10^{-3}$. Extrapolation of this slope to $f = 0$ resulted in an appreciable frequency...
offset $\Delta H(0) = 94$ Oe. For frequencies less than 36 GHz the frequency dependence showed a clear downturn. This $\Delta H(f)$ behavior is characteristic of the two-magnon scattering mechanism [57]. The slope $\Delta H(f)$ for $f \geq 36$ GHz indicates that the two-magnon scattering contribution to the damping was already saturated and was equal to $\Delta H(0) = 94$ Oe.

Interestingly two-magnon scattering was not pronounced in the 40Fe film in 20Au/40Fe/140Au/11Pd/16Fe/GaAs(001) structure. The $\Delta H(f)$ dependence on frequency is shown in Fig. 6.5 by the symbol (o), and was found to be linear in the frequency range between 3 and 73 GHz. The effective Gilbert damping obtained from a linear fit to the data resulted in $\alpha_{\text{eff}} = 3.7 \pm 0.3 \times 10^{-3}$. Again $\alpha_{\text{eff}}$ can be explained be two contributions in similar manner as for 16Fe film. The first contribution, $\alpha = 2.6 \times 10^{-3}$ is nearly equal to the $\alpha_b$, see Fig.6.1. The second contribution $\alpha^{sp} = 1.1 \times 10^{-3}$ (see details in section 7.2.1). The difference between the data points (o) in Fig. 6.4 and Fig. 6.5 and $\Delta H$ given by $\alpha_{\text{eff}} = 3.7 \times 10^{-3}$ is a consequence of long wavelength inhomogeneities in the 40Fe film.

The linear frequency dependence of $\Delta H(f)$ observed for the 40 Fe layer supports the view that the formation of magnetic defects was not fully developed in this layer. This implies that the strain induced by the Pd layer in 20Au/40Fe/140Au/11Pd/16Fe/GaAs(001) was not sufficient to propagate misfit dislocation glide planes through the thick 140Au layer. On the other hand, for the 16Fe layer located underneath the Pd layer the incorporation of lattice defects caused by the misfit dislocation network in Pd triggered the extrinsic damping. The value found for this extrinsic damping parameter is approximately 4 times larger than the Gilbert damping observed for bulk Fe single crystals.
Chapter 7

Spin transport in Au and Ag

Fe double layers separated by nonmagnetic spacers are dynamically coupled by spin momentum transfer. In the case of thick spacers the spin transport is affected by spin dependent scattering processes leading to dissipation of the accumulated magnetic momentum inside the normal metal spacer. The dissipation of the magnetic momentum was described using diffusion theory. This chapter will cover the study of spin diffusion in thick normal metal spacers (NM) of Au and Ag. Two experiments were carried out allowing one to estimate the spin diffusion length in Au and Ag: (a) Ferromagnetic Resonance (FMR) and (b) Time Resolved Magneto-Optical Kerr Effect (TRMOKE).

7.1 Diffusive spin momentum transfer

In magnetic single (F/NM) layer structures and in double (F1/NM/F2) layer structures the spin current generated at the F/NM interface leads to an accumulated spin density \( m_{NM} \) inside the NM spacer layer. The accumulated spin density in the NM is transported by itinerant electrons. The motion of the itinerant electrons in the NM is affected by momentum scattering, \( \tau_{el} \), (leading to electrical resistance) and spin flip scattering, \( \tau_{sf} \), (leading to the loss of spin momentum). In normal metals the spin-flip scattering is caused by a weak spin-orbit coupling. This means that only some of the momentum scattering events are accompanied by a spin flip event [127], and therefore \( \tau_{sf} \gg \tau_{el} \). The spin diffusion length \( l_{sd} \) determines the length scale for the spin momentum decay. For spacers significantly thinner than \( l_{sd} \) the loss of spin momentum is negligible and the spin current propagation can be considered to be in the spin ballistic limit. This
scenario has been widely studied by Heinrich et al. [24] and Urban et al. [39]. For thick NM layers one has to include spin dependent relaxation processes that remove the magnetic angular momentum from the spin current. At this point, within existing spin transport theories [44, 46, 128] one is able to treat quantitatively only the spin ballistic and the spin diffuse limits. The experimental studies in this thesis were carried out over a continuous range from the ballistic to the diffuse limits. At this time there is no non-local spin transport theory available for normal metals and therefore the experimental results were interpreted by means of spin diffusion equations along with appropriate boundary conditions (see next section 7.2 in this chapter). However, this theory provides correct results for spacer thicknesses $d \ll l_{sd}$ and $d \sim l_{sd}$. It thus provides a useful theoretical model for the interpretation of the experimental results reported in this Chapter.

7.2 Theory of spin transport in diffuse normal metal media

7.2.1 Magnetic single F/NM layer structures

The spin pumping creates an accumulated magnetic momentum density $m_{NM}$ in the adjacent NM layer. Since spin pumping carries no net electric charge the $m_{NM}$ in spin ballistic limit propagates equally in the forward and backward directions perpendicular to the F/NM interface and at the Fermi velocity $v_F$. For a small precessional amplitude the rf component of $m_{NM}$ is nearly transverse to the static magnetization vector and the backward flow is fully absorbed at the F/NM interface. In equilibrium the total flow of magnetic moment through the F/NM interface is given by [44]

$$J_{m}^{\text{net}} = J_{m}^{\text{pump}} - J_{m}^{\text{back}},$$

where $J_{m}^{\text{back}} = 0.5v_F m_{NM}$. The coefficient 0.5 represents half of the magnetic momentum propagating towards the F/NM interface.

For thick NM layers one has to include relaxation of the accumulated magnetic momentum density. The propagation of $m_{NM}$ in the NM can be described by the diffusion equation

$$i\omega m_{NM} = D \frac{\partial^2 m_{NM}}{\partial x^2} - \frac{1}{\tau_{sf}} m_{NM},$$

where $\omega$ is the angular frequency, $D$ is the diffusion coefficient, $\tau_{sf}$ is the spin-flip time of the NM, and $m_{NM}$ is the magnetic momentum density in the NM.
CHAPTER 7. SPIN TRANSPORT IN AU AND AG

where $\omega$ is the angular frequency, $\tau_{sf}$ is the spin flip relaxation time and $x$ is the coordinate in the direction perpendicular to the interface. The diffusion coefficient $D$ is defined by [45]:

$$D = \frac{v_F^2 \tau_{el}}{3},$$  \hspace{1cm} (7.3)

where $v_F$ is the Fermi velocity in the NM and $\tau_{el}$ is the electron momentum relaxation time.

![Figure 7.1: Spin transport by spin pump/sink in a Ferromagnet/Normal metal (F/NM) structure. The spin current creates an accumulated magnetic momentum density $m_{NM}$ in the adjacent NM layer. $m_{NM}$ propagates equally in the forward and backward directions perpendicular to the F1/NM interface at the Fermi velocity $v_F$. The conservation of total angular momentum for the F1 layer leads to an additional damping in the F1 layer. For a single magnetic layer structure F1/NM the boundary conditions are determined by the continuity of the spin current from the ferromagnet into the NM at $x = 0$ and a free magnetic moment condition at the outer boundary $x = L$. The solution of Eqs 2.66 and 7.2 requires boundary conditions. For a single magnetic F/NM layer structure the boundary condition at $x = 0$ is determined by the continuity of the spin current from the ferromagnet into the normal metal Fig. 7.1:

$$(x = 0) : \tilde{j}_m^{\text{net}} = \tilde{j}_m^{\text{pump}} - 0.5 v_F m_{NM} = -D \frac{\partial m_{NM}}{\partial x}. \hspace{1cm} (7.4)$$

where $\tilde{j}_m^{\text{net}}$ represents the net flow of the magnetic momentum across the F/NM interface. Note the $\tilde{j}_m^{\text{net}}$ equals the spin diffusion current ($-D \frac{\partial m_{NM}}{\partial x}$) at the interface. At the outer interface, $x = L$, one uses a free magnetic momentum condition

$$(x = L) : \frac{\partial m_{NM}}{\partial x} = 0. \hspace{1cm} (7.5)$$
Solving equation 7.2 with boundary conditions 7.4 and 7.5 allows one to determine \( j_{\text{net}}^m \) at the F/NM interface. The conservation of the total magnetic moment in F/NM structure, see Eq. 2.66, results in the interface damping due to spin diffusion:

\[
\alpha_{sp} = \left[ 1 - \frac{(Dk + e^{-2kd_{NM}}Dk)0.5v_F}{Dk(Dk + 0.5v_F) + e^{-2kd_{NM}}Dk(0.5v_F - Dk)} \right] \frac{g\mu_B}{4\pi M_s g_{11}} \frac{1}{d_F}, \tag{7.6}
\]

where \( d_{NM} \) is the thickness of the NM layer. \( k \) is the wave vector defined by [44]:

\[
k = \sqrt{1 + i\omega \tau_{sf}} \frac{1}{l_{sd}}, \tag{7.7}
\]

where \( l_{sd} \equiv \sqrt{D\tau_{sf}} \) is the spin diffusion length in the normal metal. For a precession frequency \( \omega \) smaller than the spin flip relaxation rate \( \omega \ll \tau_{sf}^{-1} \) one can use the approximation, \( k \approx 1/l_{sd} \). The spin-diffusion length which sets the scale for the spin accumulation penetration into the normal layer is:

\[
l_{sd} = v_F \sqrt{\frac{\tau_{sf}}{3}}. \tag{7.8}
\]

The prefactor on the right-hand side of Eq. 7.6 reflects the reduction effect on the Gilbert damping that is caused by spin diffusion back into the ferromagnet.

Equation 7.6 has two limits: (a) \( d_{NM} \ll l_{sd} \Rightarrow \alpha_{sp} \rightarrow 0 \). (b) For \( d_{NM} > l_{sd} \)

\[
\alpha_{sp} = \frac{g\mu_B}{4\pi M_s g_{11}} \frac{1}{d_F} \left[ \frac{\sqrt{\varepsilon/3}}{1 + 0.5\sqrt{\varepsilon/3}} \right], \tag{7.9}
\]

where \( \varepsilon = \tau_{sf}/\tau_{sf} \). Note that the saturation value of \( \alpha_{sp} \) for small values of \( \varepsilon \) is proportional to \( \sqrt{\tau_{sf}/\tau_{sf}} \). In fact the de-enhancement factor in Eq. 7.9 can never reach the value 0.5 which corresponds to a perfect sink in magnetic double layer structures, see the following subsection.

### 7.2.2 Magnetic double F1/NM/F2 layer structures.

In F1/NM/F2 structures, where the integers denote the ferromagnetic layers, the presence of two ferromagnetic layers can make spin pumping damping possible even for a thin NM layer. If one ferromagnet (e.g F1) is at resonance while the other is not at resonance, the film F2 acts as a sink for transverse spin currents pumped by the film F1. As discussed in the previous section the accumulated magnetic moment density in the
NM propagates across the normal metal and can be absorbed at the NM/F2 interface. In the ballistic spin limit the propagating spin current is entirely absorbed at the NM/F2 interface, and thus F2 acts like a perfect spin sink. One should note that only half of the spin current is absorbed by F2. The other half returns to F1 which also acts like an ideal spin sink, as indicated in Fig. 7.2. The pumped magnetic momentum from the F1/NM interface which is absorbed at the NM/F2 interface provides an effective spin brake for the precessing magnetic momentum in F1. The conservation of total angular momentum for the F1 layer leads to an additional damping which follows the phenomenology of Gilbert damping with a damping coefficient $\alpha$ given by Eq. 267. This description is strictly correct only for small precessional angles where the pumped spin momentum is perpendicular to the saturation magnetization and the static magnetic moments of F1 and F2 are collinear.

Magnetization dynamics in the spin ballistic limit can be described by the LLG equation of motion 2.66 with the additional spin-pumping/sink terms

$$\frac{\partial \vec{M}_i}{\partial t} = -\gamma \left[ \vec{M}_i \times \vec{H}_{\text{eff}} \right] + \alpha_0 \left[ \vec{M}_i \times \frac{\partial \vec{n}_i}{\partial t} \right] + \alpha_i^{sp} \left[ \vec{n}_i \times \frac{\partial \vec{n}_i}{\partial t} - \vec{n}_j \times \frac{\partial \vec{n}_j}{\partial t} \right], \quad (7.10)$$

where $\vec{n}$ are the unit vectors along the direction of the instantaneous magnetization vectors $\vec{M}_i$ in F1 and F2: $i, j = 1, 2$ and $i \neq j$. The additional Gilbert damping parameter
$\alpha_{i}^{sp}$ due to the spin pump/sink effect is given by Eq. 2.67 with the corresponding subscript 1 or 2 for a given layer and its thickness. The exchange of spin currents is a symmetric concept and the equation of motion for a given layer can be obtained by interchanging the indices $i \leftrightarrow j$. The last term on the right hand side in Eq. 7.10 represents the spin pump/sink contributions. The sign (+) (spin pumping) and (-) (spin sink) indicate the spin current directions.

The spin pump/sink theory in the spin diffusion regime can be obtained by an extension of the diffusion theory for F/NM structures which was shown above. For a magnetic double layer structure F1/NM/F2 the boundary conditions at the F1/NM interface are equivalent to Eq. 7.4. The boundary conditions at the NM/F2 interface are [45]

$$-D \frac{\partial \mathbf{m}_{NM}}{\partial x} = 0.5v_{F}\mathbf{m}_{NM}. \quad (7.11)$$

The boundary conditions in Eq. 7.11 are valid for the case when the layer F2 is off resonance and therefore contributes negligibly to spin pumping. The right hand side of Eq. 7.11 represents the magnetic current from the NM into F2 and acts as a driving torque for the magnetic momentum in F2. The interface Gilbert damping for the F1 layer due to the spin pumping is then given by:

$$\alpha_{i}^{sp} = \left[1 - \frac{(Dk + 0.5v_{F} + e^{-2kd_{NM}} (Dk - 0.5v_{F})) 0.5v_{F}}{(Dk + 0.5v_{F})(Dk + 0.5v_{F}) + e^{-2kd_{NM}} (Dk - 0.5v_{F})(0.5v_{F} - Dk)}\right] \times$$

$$\frac{g_{\mu_{B}}}{4\pi M_{S}} \frac{1}{g_{11}d_{F}}. \quad (7.12)$$

In the limit of vanishing spin flip rate in the NM, $\tau_{sf} \rightarrow \infty$, and $\varepsilon \rightarrow 0$ and Eq. 7.12 reduces to Eq. 2.67. The accumulated magnetic momentum density propagates without any scattering in the NM and the maximum of the spin current (one half) from the film F1 is absorbed by the film F2 acting as a perfect spin sink. This sets the upper limit for the Gilbert damping enhancement. This is achieved when $l_{sd} \gg d_{NM}$. With increasing $d_{NM}$ less spin pump momentum manages to diffuse across the NM. In the limit $d_{NM} \gg l_{sd}$ the accumulated magnetic momentum is relaxed to the lattice before it is able to reach the interface NM/F2. This situation is equivalent to a simple F/NM bilayer system with a thick NM layer.
7.3 FMR studies in $n \text{Au}/16\text{Fe}/\text{GaAs}$ films—spin diffusion length in Au

The studies in this section were directed towards determining the spin diffusion coefficient $D$ and the spin flip relaxation time $\tau_{sf}$ in Au. $D$ and $\tau_{sf}$ were determined using Ferromagnetic Resonance (FMR) to study single iron Fe/Au(001) magnetic structures. Similar experiments were carried out by Mizukami et al. [41] on Cu/permalloy/Cu/Pt films using a single microwave frequency, $f = 10$ GHz.

The following series of samples were grown: $n \text{Au}/16\text{Fe}/\text{GaAs}(001)$, where $n = 20, 80, 150, 200, 250, 300$: the integers represent the number of atomic layers. FMR studies were carried out using standard microwave spectrometers operating at 10, 24, 36 and 73 GHz, see the details in [15]. The bulk and interface Gilbert damping contributions to $\Delta H$ were found to be strictly linearly dependent on the microwave angular frequency $\omega$, $\Delta H = \alpha \omega$. The damping parameter $\alpha$ was extracted from the slope of linear fits to the measured $\Delta H$ vs $\omega$ data. The frequency dependence of $\Delta H(f)$ is presented in Fig. 7.3.

From Fig 7.3 one can see that the Gilbert damping ($\Delta H$) initially increases with increasing thickness of the Au layer. This means that the accumulated spin density becomes increasingly attenuated with increasing Au thickness ($d_{NM}$). The spin momentum relaxation is described by the term $m_{NM}/\tau_{sf}$. This increase in $\Delta H$ saturates when the Au thickness becomes larger than the spin diffusion length, $d_{NM} > l_{sd}$. For $d_{NM} \ll l_{sd}$, $\Delta H$ is given only by the intrinsic Gilbert damping of the Fe layer. The measured spin pumping Gilbert damping coefficient $\alpha^{sp}$ was interpreted by Eq. 7.6. The results of this fitting procedure are shown in Fig. 7.4 which compares the measured values of $\alpha^{sp}$ as a function of Au layer thickness with the calculated curve. The following fitting parameters were used: $\alpha_0 = 3.5 \times 10^{-3}$, $g_{11} = 2.4 \times 10^{15}$ cm$^{-2}$, $\tau_{el} = 1.2 \times 10^{-14}$ s, $\tau_{sf} = 15 \times 10^{-14}$ s. The Fermi velocity was assumed to be $v_F = 1.4 \times 10^8$ cm/s. These parameters resulted in a spin diffusion length $l_{sd}^{Au} = 34$ nm. One should emphasize that the spin relaxation parameters $\tau_{sf}$ and $\tau_{el}$ required to fit the data are well defined. This is because the approach to saturation of $\alpha^{sp}$ as a function of thickness is determined by $l_{sd}$ which is proportional to the product of the spin and momentum relaxation times, $(\tau_{sf}\tau_{el})^{1/2}$, and the saturation value of $\alpha^{sp}$ is proportional to $(\tau_{el}/\tau_{sf})^{1/2}$, see Eq. 7.9. The interface
spin mixing conductance \( g_{11} \) was determined using a 12Fe/20Au/16Fe/GaAs(001) double layer structure where the spin transport in the Au layer is nearly ballistic [24]. For two Fe films such that their resonance fields (\( H_{FMR} \)) are very different the spin sink provides a maximum spin pumping contribution to the Gilbert damping. The effective damping coefficients measured at 73 GHz for the above double layer structure were as follows: \( \alpha_{\text{eff}} = 0.0091 \) (\( \Delta H = 229 \) Oe) for the 12Fe film and \( \alpha_{\text{eff}} = 0.0076 \) (\( \Delta H = 189 \) Oe) for the 16Fe film. The constant value of the intrinsic bulk Gilbert damping parameter \( \alpha_b = 0.0035 \) was subtracted from the above effective values of \( \alpha_{\text{eff}} \) to determine the corresponding values of \( \alpha^{sp} \). Using Eq. 2.67 leads to the spin pumping parameter \( \tilde{g}_{11} = 2g_{11} = 2.4 \times 10^{15} \) cm\(^{-2}\). This is in good agreement with the first principle calculations by Tserkovnyak et al [44] for the Fe\( _{\text{bcc}} \)/Au\( _{\text{fcc}} \) interface, \( g_{11} = 1.19 \times 10^{15} \).

It is interesting to note that Kurt et al. [129] studied the spin diffusion length from current perpendicular to the plane (CCP) giant magnetoresistance measurements (GMR) on polycrystalline Au/Cu spacers. They obtained \( l_{sd} = 35 \) nm which is very close to our result. The ratio \( \varepsilon = \tau_{el}/\tau_{sf} = 0.08 \) indicates that in our samples \( \tau_{sf} \) is one order of
Figure 7.4: The dependence of the additional damping by spin pumping, $\alpha^{sp}$, on the Au layer thickness $d_{Au}$ in the Au/16Fe/GaAs(001) samples. The (●) symbols represent the measured data. The error bars were determined from some degree of uncertainty determining the slope in the measured $\Delta H(f)$ as a function of the microwave frequency $f$, see Fig. 7.3. The solid line shows the resulting fit to the data using the spin pumping and diffusion theory with the following parameters: $\tilde{g}_{z1} = 2.4 \times 10^{15} \text{ cm}^{-2}$, $\tau_{el} = 1.2 \times 10^{-14} \text{ s}$, and $\tau_{sf} = 15 \times 10^{-14} \text{ s}$. The corresponding spin diffusion length was found to be $l_{sd}^{Au} = 35 \text{ nm}$.

Careful inspection of Fig. 7.4 shows that the diffusion theory does not fully reflect the measured value of $\alpha^{sp}$ for very thin Au layers. Notice that the interface Gilbert damping depends linearly on the Au layer thickness, see solid line in Fig. 7.4. In fact, in the thinnest measured sample 20Au/16Fe/GaAs, the spin pumping contribution was smaller than that predicted by the spin diffusion theory indicating that in this thickness range the theory does not describe well non-local spin transport.

At this point one can evaluate the effective spin diffusion length in 20Au/40Fe/140(28 nm)Au/11(2.1 nm)Pd/16Fe/GaAs(001). Here the situation is more complicated because the normal spacer consist of Au and Pd layers. The formula 7.6 was derived for a single NM spacer. There is no spin diffusion theory for heterogenous spacers, however a simple argument can be used to replace the 140Au/11Pd by a single Au spacer. The spin
diffusion length in Au is \( l_{sd} = 35 \) nm. The spin current decay length in Pd in our work was found to be 5.1 nm \[130\]. Assuming that the spin current is attenuated in both layers by simple exponential factors given for the Au spacer \( \exp(-28/35) \) and for Pd \( \exp(-2.1/5.1) \), then the effective thickness, \( d_{eff} \), of the single Au spacer can be estimated using the simple relationship:

\[
e^{-\frac{28}{35}} \times e^{-\frac{2}{5.1}} = e^{-\frac{d_{eff}}{35}}
\]

This leads to \( d_{eff} = 42 \) nm which is already in the saturated limit of spin pumping contribution to the damping, see Fig. 7.4. Therefore one can neglect the spin sink effect in these structures. Using the formula 7.6 leads to \( \alpha^{sp} = 2.6 \times 10^{-3} \) for 16Fe film and \( \alpha^{sp} = 1.1 \times 10^{-3} \) for 40Fe film.

### 7.4 Time Resolved studies of spin transport in Au and Ag

20Au/12Fe/Au,Ag/16Fe/GaAs(001) structures were employed in the study of the propagation of spin currents across NM films. Time-Resolved Magneto-Optical Kerr Effect (TRMOKE) measurements are an ideal tool for the investigation of the propagation of spin currents in these structures. Stroboscopic measurements of the magnetization precession in the 10 GHz frequency range were carried out with picosecond time resolution, and sub-micrometer spatial resolution, using a slotted transmission line carrying repetitive ps magnetic excitation pulses. After a magnetic pulse excitation, laser pulses of 100 fs duration, and at a delay time \( t_D \), were used to probe the perpendicular component of the precessing magnetization in the top 12Fe layer: see the detailed description in Chapter 3, sec. 3.2.2. The accumulated spin density generated by the precession of the bottom 16Fe layer propagated across either Au or Ag normal metal spacers and resulted in the rf excitation of the top 12Fe film.

The following magnetic parameters were obtained from the in-plane angular dependence of the FMR resonant fields at 24 GHz:

(i) 20Au/12Fe/300Au/16Fe/GaAs(001) sample; The bottom 16Fe grown on GaAs(001): In-plane uniaxial anisotropy parameter \( K_U^{\parallel} = -4.10 \times 10^5 \) erg/cm\(^3\) with the hard magnetic axis directed along the [110] crystallographic orientation of the GaAs(001), the
The investigation of the magnetization induced spin current signals requires a careful quantitative analysis. Since the maximum spin current contribution to the main (direct) signal is only of the order of a few percent the acquired TRMOKE spectra had to be processed in the manner shown in Fig. 7.5 (a–c). A dc bias field, $H_B$, was applied in the plane of the magnetic film. Fig. 7.5 (a) shows the time evolution of the magnetization in a 30Fe/150Au/16Fe/GaAs structure in response to repetitive applied magnetic field pulses. In order to obtain a large signal to noise ratio the data were averaged over 9 scans. A low frequency background signal was always present in the raw data. This background signal had to be subtracted prior to further analysis. The background signal was mostly caused by reflections generated in the transmission line and its contacts to the photo switch. The background profile was obtained using the FFT filtering method that removed Fourier components in the time domain spectrum having frequencies lower than:

$$f_c = \frac{1}{n\Delta t}$$  \hspace{1cm} (7.14)

where $n$ is the number of data points obtained for one oscillatory period and $\Delta t$ is the time spacing between two adjacent data points. The result of the background subtraction is shown in Fig. 7.5 (b). The solid line shows the background signal. Fig. 7.5 (c) shows the Fast Fourier Transform (FFT) of the time domain spectrum from (b). One can clearly distinguish two peaks in the frequency domain corresponding to the resonant frequencies.
Figure 7.5: Time Resolved MOKE signal measured using a 30Fe/150Au/16Fe/GaAs sample. The TRMOKE measurements were carried out in the presence of a static magnetic field $H_B = 600$ Oe applied along the [110]$_{Fe}$ crystallographic direction (easy magnetic plane for the 16Fe film). The data were averaged over 9 scans. Notice that a strong Kerr signal was observable even after the laser beam travelled back and forth through the 60 nm Au spacer. (a) The raw TRMOKE signal data. (b) The resultant signal after the background subtraction. The solid line shows the time dependent background signal obtained using the FFT filtering method, see details in the text. (c) FFT amplitude and phase plots calculated for the time domain spectrum from (b). Notice the two peaks in the frequency domain corresponding to the resonant frequencies of the two Fe layers. The high amplitude peak at lower frequency corresponds to the main Kerr signal from the top 12Fe layer and the other peak corresponds to the 16Fe bottom layer.
of the two Fe layers. The high amplitude peak at the lower frequency corresponds to the main Kerr signal from the top 12Fe layer and the other peak corresponds to the signal from the 16Fe bottom layer. They are \( \sim 4 \) GHz apart due to the different interface anisotropies of the Fe films grown on Au and on GaAs, respectively [39]. The ratio of the FFT resonance peak areas \( r = \frac{\text{Area}_{16\text{Fe}}}{\text{Area}_{30\text{Fe}}} \) resulted in the ratio \( r \sim 10\% \). This is appreciably larger than one would expect from the spinpump/sink theory, \( (r \sim 3\%) \). In this case one can attribute the smaller amplitude FFT component in Fig. 7.5 (c) to the direct FMR signal from the bottom 16Fe film. Indeed the series of TRMOKE measurements on the nAu/16Fe/GaAs \((n=150, 200, 250)\) samples confirmed the presence of the direct signal from the 16Fe film.

Additional useful information is provided by the phase of the FFT, see the upper part of Fig. 7.5 (c). In fact the phase of the FFT can be used as an important tool in determining the difference between the direct and pure spin current induced signals. Detailed FFT phase analysis and discussion is presented in the last section 7.4.3 of this chapter.

### 7.4.2 Pure spin current propagation and spin diffusion in Ag

In order to study the propagation of spin currents in silver it was necessary to acquire the rf dependent Kerr signal from the top 12Fe layer only. A 300 atomic layer Au spacer \((60 \text{ nm})\) was sufficient to suppress the signal from the bottom 16Fe film. No measurable TRMOKE signal was observed from the 300Au/16Fe/GaAs(001) single iron film sample. Therefore the 20Au/12Fe/300Au/16Fe/GaAs(001) structure was used to detect the spin current propagation in Au.

A difference in the magnetic crystalline anisotropies for the top 12Fe film and for the bottom 16Fe film was used to separate their resonant frequencies by \( \sim 4.5 \) GHz. A dc magnetic field, \( H_B = 500 \), was applied in-plane in order to overcome the anisotropy fields and saturate the sample. This dc magnetic field was applied along the \([110]_F\) direction: the easy magnetic axis for the 16Fe film and the hard magnetic axis for the top 12 Fe film. The top 12Fe layer has a small uniaxial in-plane anisotropy that is almost negligible compared with the cubic anisotropy field.

The time dependence of the ps resolved Kerr signal and its Fast Fourier Transform (FFT) are shown in Fig. 7.6 (a) and (b), respectively. The spin current was significantly
CHAPTER 7. SPIN TRANSPORT IN AU AND AG

Figure 7.6: (a) Time resolved magnetization oscillations for the 20Au/12Fe/300Au/16Fe/GaAs(001) sample. The bias field was \( H_B = 500 \) Oe; it was applied along the iron [110]\(_{Fe}\) crystallographic direction. The solid line shows the background signal which was subtracted from the raw data. (b) The Fast Fourier Transform (FFT) of the data from (a): The thick solid line corresponds to the FFT of the data from a). The thin solid and dotted FFT lines show computer simulations for a maximum spin current contribution (infinite \( l_{sd} \)) and that corresponding to \( l_{sd} = 34 \) nm, respectively. The inset in b) shows a magnified portion of the FFT spectrum that allows one to view the tiny spin current contribution to the TRMOKE signal. The measured spin current peak is unfortunately comparable to the noise, but one can see that the intensity is compatible with that expected for the spin diffusion length in Au which was determined from the FMR measurements on the single magnetic Au/Fe/GaAs(001) layers. (c) The phase of the FFT signal obtained from the measured data.
decreased by the loss of spin momentum in the 60nm Au spacer. The FFT transform of the measured TRMOKE data showed mainly the strong resonance peak corresponding to the 12Fe layer. The spin current induced signal generated by the bottom 16Fe layer was buried in the noise, see Fig. 7.6 (b) and the inset. For an applied magnetic field $H_B = 500$ the spin pumping signal occurred at $f_{res} = 10.6$ GHz. Self-consistent computer simulations of the ps resolved transient magnetization oscillations were carried out using the equation of motion for 16Fe and Eq. 7.2 with the boundary conditions 7.4 and 7.11 including a realistic shape for the magnetic pulse. The expression on the right hand side of Eq. 7.11 was used as an external torque for the equation of motion of 12Fe. Magnetic and spin diffusion parameters were obtained from the FMR measurements, see section 7.3, 7.4 and Fig. 7.4. The simulation of the signal for 12Fe produced very weak spin current contribution as compared with the signal corresponding to the main FMR peak, see Fig. 7.6(b). A detailed inspection of the measured and simulated FFT spectra suggests that the spin current contribution might be present in the measured data, see the inset to Fig. 7.6. However, this signal is comparable to the noise and is at best only marginal evidence for spin current excitations.

Ag has a significantly smaller spin orbit interaction than Au and therefore one expects a larger spin diffusion length than that in Au, and with that a noticeably increased spin current signal. For this reason the use of Ag as a spacer seems to be more suitable for the study of spin current propagation. In addition, the optical properties of Ag in the near-infrared are such as to enhance the attenuation of the direct signal from the bottom 16Fe layer compared to that of a Au spacer of equivalent thickness. The calculation of the light propagation through a 12Fe/300Au/16Fe/GaAs(001) structure [49] showed that the penetration depth is of the order of $\sim 100$ nm for Au. Similar calculations using the magneto-optical properties of Ag and Fe for light having an 800 nm wavelength [131, 132] resulted in a penetration depth of $\sim 70$ nm. Indeed Fig. 7.7(b) clearly shows a spin pump signal for the 300Ag spacer. The ratio $r$ of the FFT areas corresponding to the spin pump and to the main frequency components was found to be $r = 1.8 \pm 0.5\%$. In order to minimize the error in the quantitative analysis the well known Savitzky-Golay filtering method was employed. The main advantage of this approach is that it tends to preserve features of the distribution such as relative maxima, minima and width. Computer simulations allowing for the maximum spin current contribution (negligible
Figure 7.7: (a) Time resolved data for the 20Au/12Fe/300Ag/16Fe/GaAs(001) sample. The dc magnetic field, $H_B = 500$ Oe, was applied along the [110]$_{Fe}$ crystallographic direction. The solid line shows the background signal which is subtracted from the raw data. (b) The FFT spectrum. The thick solid line represents the FFT of the data shown in (a). The thin solid line represents the FFT of a theoretical simulation using following parameters: $\tau_{el} = 5 \times 10^{-14}$ s, $\tau_{sf} = 70 \times 10^{-14}$ s and $l_{sd} = 151$ nm. The inset provides a magnified part of the FFT spectrum for easy viewing. (c) The phase of the FFT obtained for the data of (a). Notice the $\pi/2$ phase shift of the two frequency components.
Ag spacer thickness) resulted in $r = 3\%$. This clearly indicates that the 300Ag spacer already decreased the spin current strength at the Ag/12Fe interface. In order to obtain a calculated signal that is comparable to the experimental result with $r = 1.8\%$, see inset of Fig. 7.7, it was necessary to use the following parameters; $\tau_{el} = 5 \times 10^{-14}$ s and $\tau_{sf} = 70 \times 10^{-14}$ s. This leads to a spin diffusion length in Ag of 150±20 nm.

7.4.3 Discussion

The results of the TRMOKE measurements from Figures 7.5 (c) and 7.7 (b), (c) lead to an interesting observation. It turns out that the phase shifts associated with the two frequency components in the FFT spectra are different. In fact they were shifted by $\pi/2$. In order to investigate this point more computer simulations were carried out designed to examine the relationship between the phases for the signals generated by the direct FMR signal and that driven by spin current propagation. The following calculations were carried out using the magnetic parameters corresponding to the 20Au/12Fe/300Ag/16Fe/GaAs(001) structure (see section 7.4):

1. The satellite signal generated by the 16Fe film was due to a spin current propagated across the Au spacer. The spin diffusion parameters were as follow: $\tau_{el} = 5 \times 10^{-14}$ s and $\tau_{sf} = 70 \times 10^{-14}$ s, and $l_{sd} = 150$ nm. This results in an intensity of the satellite peak comparable to the experimental measurement, $r = 1.8\%$.

2. The satellite signal was caused by the direct FMR Kerr signal from the 16Fe bottom layer. Its phase was assumed to be the same as that corresponding to the main FMR signal from the 12Fe film.

3. As in 2., but the phase of the FMR pick up signal was changed by $\pi$

Case 1. corresponds to Fig. 7.7. The effective intrinsic Gilbert damping (including a spin pump contribution) was found to be $\alpha_{12Fe}^{12Fe} = 0.008$ and $\alpha_{16Fe}^{16Fe} = 0.0068$ for the 12Fe and 16Fe films, respectively.

In 2. and 3. the propagation of spin momentum was disabled by setting the spin pump and sink at the F/NM interfaces equal to zero. In order to simulate similar damping and strength of the TRMOKE signals to that in experiment the value of $\alpha_b$ for both layers (i.e. $\alpha_b^{12Fe} = 0.008$, $\alpha_b^{16Fe} = 0.0068$) was set equal to the corresponding
Figure 7.8: Computer simulations of TRMOKE measurements corresponding to the 20Au/12Fe/300Ag/16Fe/GaAs(001) structure. Self-consistent calculations were obtained by using the LLG equations of motion, spin pump/sink theory, and the spin diffusion equation in Ag. FFT transforms are shown on the right hand side of the figure. (a), (b), and (c) are computer simulations corresponding to conditions 1, 2, and 3 (see text above). The calculations were carried out using $\tau_{el} = 5 \times 10^{-14}$ s, $\tau_{sf} = 70 \times 10^{-14}$ s, and $l_{sd} = 150$ nm. Notice that in (a) the relative phase shift between the two signals in the FFT is $\pi/2$ while for (b) and (c) the phase shift between the two signals are $0^\circ$ and $180^\circ$, respectively.
value of $\alpha_{\text{eff}}$ and the direct signal from the bottom 16Fe was scaled down to $r = 1.8\%$.

Computer simulations applying the full solution of the LLG equations of motion and diffusion equation are presented in Fig. 7.8. The FFT on the right hand side of the figure correspond to their time domain spectra on the left. A striking difference is visible in the phase of the two signals. In Fig. 7.8 (a) the relative phase change between two frequency components is $90^\circ$. The phase difference and even the field dependence clearly resembles the features observed in our experimental data, see Fig 7.7. In Fig. 7.8(b) and (c) the relative phase shift is $0^\circ$ or $180^\circ$, respectively. Notice also that the field dependence of the phase of the satellite signal has a peak like character which is in sharp contrast with the spin current driven phase having a dispersion like character.

The phase of FFT clearly identifies the origin of the satellite peak.

Since the spin current driven signal is proportional to $d\vec{m}/dt$, it follows that the direct FMR signal is phase shifted by $\pi/2$ from the spin current induced signal. In fact this feature was used to distinguish between the direct and spin current induced signals in similar studies by Woltersdorf et al. [133]. However their TRMOKE setup was completely different. In their case the studies were carried out using a continuous wave (CW) microwave rf driving magnetic field that allowed one to monitor the transverse rf susceptibility $\chi$ by means of TRMOKE. In that case the absorption peak of the main FMR signal was accompanied by a dispersive like signal for the spin current driven signal and vice versa.
Chapter 8

Conclusions

High quality crystalline ultrathin single and multilayer Fe structures were grown on GaAs(001) by means of Molecular Beam Epitaxy (MBE). Two MBE growth techniques were used: Thermal Deposition (TD) and Pulsed Laser Deposition (PLD). Structural properties of the films were analyzed in-situ by means of two surface sensitive techniques: Reflection high energy diffraction (RHEED) and Scanning Tunneling Microscopy (STM). Conversion electron Mössbauer spectroscopy (CEMS) was used to investigate the structural properties and chemical environment of the Fe/GaAs(001) interfaces. In CEMS two atomic layers of pure $^{57}$Fe isotope were deposited at the Fe/GaAs(001) interface. The magnetic properties of the Fe multilayer structures were measured using angular-resolved Ferromagnetic Resonance (FMR) in the range of microwave frequencies from 1 to 73 GHz. Time Resolved Magneto Optical Kerr Effect (TRMOKE) having a ps temporal and micrometer spatial resolution was used to investigate the propagation of the spin currents across the Au and Ag spacers.

The studies presented in this thesis led to several important results:

1. The FMR data revealed that a uniaxial in-plane anisotropy $K_U^\parallel$, with the hard magnetic axis directed along the [110] crystallographic axis of the GaAs(001) template, appeared in the initial stages of the Fe film growth. The $1/d_{Fe}$ dependence of $K_U^\parallel$, where $d_{Fe}$ is the film thickness, suggested the interfacial origin of the uniaxial anisotropy. Neither the interface between Fe and vacuum nor the interface between Fe and Au (or Ag) have shown an appreciable contribution to $K_U^\parallel$. The surface in-plane uniaxial anisotropy $K_U^{\parallel,int} = -0.10 \pm 0.01 \text{ erg/cm}^{-2}$ was found to be present in the TD samples for film thicknesses up to 16ML (2.2 nm). This surface uniaxial anisotropy decreased appreciably
CHAPTER 8. CONCLUSIONS

for thick Fe films \(d_{Fe} \geq 30 \text{ML}\): \(K_{int}^{Fe} = -0.03 \pm 0.01 \text{erg/cm}^2\). Interestingly, the same small value for \(K_{int}^{Fe}\) was found for all Fe films grown using PLD. (b) Mössbauer studies have shown that the Fe films prepared by means of PLD exhibited a more intermixed interface than films prepared using TD. Consequently the bcc Fe lattice in the PLD films can be expected to be clamped more robustly to the GaAs(001) lattice than the films grown using TD. A significant decrease in the in-plane interface uniaxial anisotropy for the films grown using PLD (with the hard magnetic axis long [110]) suggests that the in-plane uniaxial anisotropy originates in an interface lattice shear. The in-plane uniaxial anisotropy is caused by magnetoelastic B₂ term. Conceivably the observed decrease in the uniaxial anisotropy in the PLD films can be explained by a decrease in the lattice shear compared to that in the TD films. A lattice shear at the Fe/GaAs(001) interface has recently been positively identified and measured by Gordon and Crozier [107] using surface EXAFS. It is interesting to note that the sign of the interface shear in [107] would lead to a hard magnetic axis directed along the \((1\overline{1}0)\) axis as is observed in the FMR measurements. However, this shear would result in the uniaxial anisotropy being factor of 5 smaller than that measured by FMR. This quantitative discrepancy is not surprising because the value of B₂ for the complex Fe/GaAs(001) interface could be significantly different from that for the bulk iron.

This suggests that the in-plane interface uniaxial anisotropy can be explained by a magneto-elastic term due to the interface lattice shear.

(2) The FMR linewidth \(\Delta H\) as a function of microwave frequency \(f\) was used to extract the intrinsic damping parameter \(\alpha\) in the set of 20Au/nFe/GaAs samples prepared using MBE (TD method); \(n=8, 10, 13, 16, 20, 25, 30\), where the integers represent the number of atomic layers. The experimental data were fit in the thickness range from 10 to 30 ML using the following thickness dependence: (a) \(\alpha(d) = \alpha(bulk) + \alpha(int)/d^2\) and (b) \(\alpha(d) = \alpha(bulk) + \alpha(int)/d\). The thickness independent part represents the bulk Gilbert damping parameter. \(\alpha_{b} = 2.5 \times 10^{-3}\) for fits using formula (a) and \(\alpha_{b} = 1.2 \times 10^{-3}\) for fits obtained using (b). The corresponding bulk Gilbert relaxation rates are \(0.78 \times 10^8\text{s}^{-1}\) and \(0.38 \times 10^8\text{s}^{-1}\), respectively. The \(1/d_{Fe}\) dependence (formula (b)) leads to a very small bulk Gilbert damping parameter that is even smaller than the smallest bulk damping found by Frait et. al [124] for the pure Fe single crystal platelets: \(G = 0.42 \times 10^8\text{s}^{-1}\).
On the other hand, $G = 0.78 \times 10^8 \text{s}^{-1}$ was only $\sim 20\%$ larger than the smallest value, $G = 0.66 \times 10^8 \text{s}^{-1}$, obtained by Heinrich et al [112] for ultrathin Fe films grown on Ag(001) substrates. The $1/d$ fit suggests that the increase in damping is caused by the presence of an interface damping contribution, whereas the $1/d^2$ fit can be explained by a two magnon scattering mechanism due to an inhomogeneous perpendicular uniaxial anisotropy as proposed by Mills [57]. Further experiments are required to decide which mechanism is more realistic.

(3) FMR studies on Au/16Fe/GaAs films, with a Au cover layer of variable thickness, have shown that the FMR linewidth ($\Delta H$) is strictly linearly dependent on the microwave angular frequency, $\omega$, and therefore can be interpreted by Gilbert damping: $\Delta H = \alpha \omega \gamma$. $\Delta H$ increased initially with increasing Au layer thickness but eventually saturated at a value that was appreciably greater than $\Delta H$ for bulk iron. This dependence was explained by spin pumping. The accumulated spin density in the Au layer becomes attenuated with increasing Au thickness, $(d_{NM})$, and the corresponding non-local contribution to the damping eventually saturates when the Au thickness becomes comparable to the spin diffusion length. These results were quantitatively interpreted by the application of self-consistent calculations based on the spin pumping mechanism, the spin diffusion equation, and the Landu-Lifshitz Gilbert (LLG) equations of motion. The theoretical fit to the experimental data allowed one to determine the momentum relaxation time, $\tau_{el} = 1.2 \times 10^{-14}$ s, and the spin flip relaxation time, $\tau_{sf} = 15 \times 10^{-14}$ s in gold. The corresponding spin diffusion length in gold was found to be $l_{sd}^{Au} = 34$ nm.

(4) TRMOKE measurements on 20Au/12Fe/300Ag/16Fe/GaAs(001) structures were used for detection of the spin current propagating across the 300Ag spacer layer. The 16Fe layer acted as a spin pump and the top 12Fe layer was used to probe the spin current propagating across the Ag spacer. Self-consistent computer simulations of the transient magnetization precessional motion with a ps time resolution led to a good quantitative agreement with the experimental data and resulted in the following diffusion parameters: $\tau_{el} = 5 \times 10^{-14}$ s and $\tau_{sf} = 70 \times 10^{-14}$ s. This led to an estimated spin diffusion length in Ag, $l_{sd}^{Ag} = 150 \pm 20$ nm.
Appendix A

MBE Pumping arrangement

The SFU MBE apparatus is equipped with several types of pumps that are responsible for maintaining UHV environment. The pumping in our MBE system is performed by ion and turbomolecular pumps in addition these pumps are augmented by Ti sorption pumps and rotary pumps. Each of the various types of pumps has a characteristic working pressure range, in which it has a particular high efficiency. One can readily achieve the pressure of low $10^{-10}$ Torr during deposition allowing one to grow uncontaminated single crystal films.

The **rotary vane pump** is the primary mechanical pump. In general it consist of a cylindrical housing stator in which rotates slotted rotor. The rotor contains vanes which slide along the stator walls and thereby push forward the air drawn in at the inlet. Air is finally ejected through the oil above the outlet discharge valve. The pump currently used in our MBE system is the two-stage rotary pump. It has "High vacuum" stage and "Rough vacuum stage". The High vacuum stage is separated from the outer atmosphere which allow to obtain the ultimate pressure range for this type of pump. Consequently the lowest working pressure is about at the lower limit of the medium vacuum pressure range i.e. $\sim 10^{-2}$ Torr. These pumps are used as back up pumps for turbo pumps.

The **Turbomolecular pumps** are especially designed to work in the pressure region below $\sim 10^{-2}$ Torr. As shown in Figure A.1 the pumping speed increases at intake pressure lower than $10^{-2}$ Torr and it remains constant over the whole working pressure range. This threshold value marks also transition from the region of laminar viscous flow to the region of molecular flow (above $10^{-2}$ Torr) of gases. The compression ratio of turbomolecular pumps, see Fig A.1 is for physical reasons higher for heavy molecules.
and adequately lower for light molecules [134]. Due to its high compression ratio for heavy molecules turbomolecular pumps can be directly connected to vacuum chamber. As the hydrogen partial pressure attained by the rotary pump (backing pump) is very low the turbomolecular pump is capable of producing extremely low pressures in the $10^{-10}$ Torr range.

The construction of the pump is in general an axial flow compressor of vertical design, which consist of a stator and rotor. The stator and rotor have turbine blades around their circumference, and each rotor/stator pair of circular blade rows form one stage, so that the whole assembly is composed of a multitude of stages mounted in series. The pumped gas arrives through a large aperture of the inlet flange at the active pumping area (top of the blades). The gas captured by the top vacuum stages is transferred gradually to the lower compression stages having blades of shorter radial span. In the compression stages gas is compressed to the backing pressure and leaves the pump through the outlet port of rotary pump.

![Figure A.1: Pumping speed and compression ratio for turbomolecular pump currently used in MBE system at SFU, model “Varian-501 Navigator”](image)

**Figure A.1:** Pumping speed and compression ratio for turbomolecular pump currently used in MBE system at SFU, model “Varian-501 Navigator”

**Ion pumps.** In our MBE system three ion pumps are used as primary pumps maintaining UHV, see Fig 3.1. These pumps are a combination of a titanium sublimation pump and a sputter ion pump which are incorporated in one cryo-cooled package (cryoshroud). For that reason they are also called “Combo-Vac” pumps [135]. The cross section of a
Figure A.2: Cross section of the ion pump.

typical ion pump is shown in Fig. A.2. The main elements of ion pump are the vacuum envelope, external magnets and element consisting of multiple cylindrical anode cells and two cathodes. The positive voltage applied to the anode creates a plasma discharge. Ions are formed from the gas molecules present in the system. These ions are accelerated toward one of the cathodes. When they strike they can be absorbed deeply by ion implantation. In addition titanium is sputtered from the cathode creating a Ti film inside the pump where it acts as a getter adsorbing reactive gas particles ($N_2, H_2, O_2, H_2O, CO, CO_2$). High speed of this pump is achieved by freshly depositing titanium film for 60-90 seconds just prior to the MBE work.
Appendix B

Crystallographic orientation and GaAs(001) wafer flats

A discussion of the in-plane magnetic uniaxial anisotropy axis for Fe/GaAs(001) structures depends on one’s ability to identify the [110] and [1\(\bar{1}\)0] crystallographic directions. This can be achieved by examining the surface reconstructions of GaAs(001) via LEED or RHEED. However, this can sometimes be misleading. For instance, the 2\(\times\) and 6\(\times\) diffraction patterns along the two perpendicular orientations can indicate either (2\(\times\)6) or (6\(\times\)2) surface reconstructions. Thus, the in-plane crystallographic orientation of wafers as quoted by the supplier is an important reference.

This information is usually provided by so-called wafer “flats”. The wafer’s flats correspond to the particular crystallographic planes that are orthogonal to the main wafer surface. The relevant specifications for GaAs(001) are set out in the Standard “SEMI M9-Specifications for Polished Monocrystalline Gallium Arsenide Slices” issued by Semiconductor Equipment and Materials International (SEMI), 3081 Zanker Road, San Jose, CA 95134, USA.

The SEMI M9 standard is based on two orthogonal \{110\} flats of different sizes. The larger flat is referred to as the primary flat (OF) and the smaller flat as the secondary flat (IF). In order to describe the wafer’s crystallographic orientation precisely, SEMI M9 further specifies the etching type for the primary flat and the relative angular offset of the secondary flat (measured on the front (001) surface), see Fig. B.1. The only complication in assigning the crystallographic directions arises from the fact that SEMI M9 allows for the two complementary flat options as described below and in Fig. 4.1:
• **Dovetail** option; Also called sometimes **EJ** (European-Japanese). Two flats are orthogonal to the front (001) surface:

  * primary flat: (110) face of dovetail etching type
  * secondary flat: (1\bar{1}0) face of V-groove etching type, 90° clockwise from the primary flat as measured on the front surface.

• **V-groove** option; Also called sometimes **US**. Two flats are orthogonal to the front (001) surface:

  * primary flat: (1\bar{1}0) face of V-groove etching type
  * secondary flat: (110) face of the Dovetail etching type, 90° counterclockwise from the primary flat as measured on the front surface.
Dovetail
or EJ (European Japanese)

V-groove
or US

Figure B.1: V-groove (US) and dove-tail (EJ) flat options of GaAs(001) as defined in the Standard SEMI M9 - Specifications for Polished Monocrystalline Gallium Arsenide Slices. To illustrate the differences in crystallographic orientations, etch pits are indicated for the particular definitions of the flats.
Bibliography


BIBLIOGRAPHY


