Optical detection of the magnetization precession
– choreography on a sub-nanosecond timescale

PROEFSCHRIFT

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mert én vagyok az alany és a tárgy,
jaj én vagyok az omega s az alfa

1[...]the subject and the object. Heavy fate:
the alpha and the omega am I."
The Lyric Poet’s Epilogue, by Babits Mihály, transl. by Peter Zollman
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Chapter 1

Introduction

Ferromagnetism, although not identified nor scientifically described, was known and used for thousands of years, in form of permanently magnetized minerals such as magnetite. In the early years of technology, its usefulness was exhausted at the level of a few robust applications; a classic example is the compass, but we all might remember our grandmothers using a permanent magnet to collect pins from the carpet. In any case, the macroscopic attractive force exerted by these objects was applicable in the daily life without any in-depth knowledge of the phenomenon, the physics behind it. A few basic characteristics of magnetism were noted; e.g. the inseparable nature of the poles —even today, the quest for the magnetic monopole continues involving astrophysics and cosmology— and the curvature/closure of the field lines, easily imaged by a handful of iron dust and a sheet of paper.

The extensive studies of electricity quickly led to the discovery of the interaction between moving electric charges (i.e. electric current) and magnetic objects. The experiment of Rowland with the rotating charges, culminating in the Biot-Savart law and the induction described by Ampère allowed more sophisticated applications of magnetism: electric generators, motors, relays, transformers, particle accelerators and amplified music.

In the informational era of the XXth-XXIst century, the need for speed and capacity in recording/reading/transmitting data outgrew the limits of libraries and printing. One branch of data recording technology was therefore started around magnetism. The possibility of writing and rewriting data by reorienting the magnetization of a ferromagnetic particle is used both in analogue (audio- and video tapes) and digital (data recording tapes, magnetic arrays and disks) technology. Although there is an immense evolution from the first IBM RAMAC magnetic data recording unit to a modern hard disk drive with respect to capacity, speed and reliability, the essence did not change: the information is stored by switching the magnetization of a tiny ferromagnetic “bit”. The first RAM (random access memory) unit for computers, dating from the 1950s before thin film semiconductor technology, was also based on magnetism: it consisted of an array of induction coils with ferromagnetic cores switchable by electric current.

The magnetic RAM, slow and robust in those times, was dropped in favour of semiconductor-based volatile memory, and only in the last years was considered again
by industry, with a completely different approach. However, in case of long-term data recording magnetic disks still prevail. The limits of the technology were constantly pushed further by introducing microelectronics, thin film technology and magnetic engineering of multilayers, developing new recording materials, improving quality/purity of these, applying new effects from mesoscopic magnetism such as Giant or Tunnel Magnetoresistance (GMR/TMR) in the read-heads etcetera. Hybrid technologies e.g. magneto-optic recording (see schematic drawing in Fig. 1.1) were implemented, to improve portability and capacity of recording media while keeping it cost-effective, a very important factor in the industrial world. Spintronics — combining micromagnetism, the electron spin, with traditional semiconductor electronics, i.e. charge of the electron— is also very promising for applications such as the magnetoresistance-based new MRAM, and it became bread-and-butter of a number of research groups in the last years. Magnetic microsensors, on the other hand, are used nowadays in a plethora of environments. From the automobile industry till biology and health care, new ideas are turned into applications on a daily basis. These developments were impossible without detailed investigations of the magnetization dynamics on a nanosecond and micrometer scale.

Figure 1.1: New applications are made possible by combining magnetism with optics (e.g. MO recording), electronics with optics (optoelectronic devices, e.g. light detectors) and magnetism with electronics (spintronic devices - magnetic semiconductors, MRAM etc)

At this point, we can formulate the questions: What exactly happens with the magnetization on these (sub)nanosecond timescales, under external perturbations? Where are the fundamental limits of switching speed? How small can we make a magnetic structure, a “bit” before we lose control over its dynamics? What are the factors influencing the switching speed and stability of micromagnetic entities? The work presented in this thesis will try to address the above questions, keeping an eye on the possibilities for novel applications and, hopefully, giving birth to new questions in the mind of the reader.

1.1 Spin- and magnetization dynamics

The studies presented in this thesis are in majority local dynamic and static measurements of the magnetization of ferromagnetic thin layers and microstructures. Therefore, the term “magnetization” met through the text should be understood...
as the “local magnetization field” used in the Landau-Lifshitz and Gilbert terminology [1, 2, 3]. This space- (and time-) dependent physical quantity $\vec{M}(\vec{r})$ is the expectation value of the magnetic moment per unit volume yielded by the individual electron spins $\vec{s}$ averaged over a few lattice cells. The number of included cells is large enough to average out the atomic-level fluctuations (in magnitude and orientation) of the spins, but small enough not to include domain walls, vortices and other complicated magnetic structures. Note, that the contribution from the orbital motion of the unpaired electrons is already included in the gyromagnetic ratio $g$.

Averaging the “local magnetization fields” or the local magnetizations over the entire volume of the ferromagnetic structure would result in the so-called average magnetization $\bar{\vec{M}}$ of the studied sample. A schematic drawing of the spin-to-total-magnetization hierarchy is shown in Fig. 1.2.

![Diagram of spin-to-total-magnetization hierarchy]

An intermediate step between the individual spins and the magnetization is the so-called macrospin $\vec{S}$, consisting of spins that are completely identical in their magnitude, orientation and external influence, therefore dynamically they behave as one single vectorial entity.

Switching the magnetization direction of a macroscopic ferromagnetic structure looks on the timescale of microseconds uniform and instantaneous. However, at the speed of reading/writing data on a modern magnetic disc (tens of megabytes per second), e.g., hundreds of millions of switching cycles of the small “bits” happen in one second. This switching time approaches already the nanosecond timescale, where the dynamics of the magnetization vector are not so simple anymore. Additionally, the small sizes and extremely complicated nature of the multilayers used in practice necessitate in-depth study of the GHz regime local magnetization dynamics, both theoretically and experimentally.
Let us imagine one isolated electron spin (or, equivalently, a macrospin) $\vec{s}$, in an external DC magnetic field as shown in Fig. 1.3. The lowest energy state requires that the equilibrium direction of the spin is parallel with the magnetic field lines. A sudden change of the orientation of the external field (e.g. by switching on a second, perpendicular magnetic field) will define a new equilibrium direction, and the spin will try to realign with this new direction. Due to the misalignment, a torque will act on the spin perpendicular to the plane formed by the magnetic field and the spin itself. The dynamics of the spin under the action of the torque will be a precessional motion around the new equilibrium direction. The spin precession was described mathematically by Landau and Lifshitz in the 1950s [1]. Considering a coupling of the precessing spin to its environment (other spins, lattice, etc), a dissipation channel for the precessional motion will be present, and the precession will be damped, allowing the spin to finally arrive to the new equilibrium direction. Hence, a phenomenological damping was introduced by Gilbert [2] to explain the full dynamics including dissipation.

In the case of a 180° canting of the equilibrium direction (by reversing the direction of the external field), a similar damped precessional motion will lead to complete reversal (switching) of the spin. Naturally, the full switching time will be dictated by the maximum of the two characteristic timescales of this motion, the precessional period and the damping. These characteristic timescales will depend on the magnitude of the spin, the couplings to its environment and the external field, and will be discussed in chapter 2 of this thesis.

When talking about magnetization dynamics, one has to consider the spatial dependence of the magnetization vector as well. While the essence of the dynamics is the same — damped precessional motion around the equilibrium direction —, a multitude of local effects due to the spatial dependence and interaction with the environment complicate the motion of the magnetization vector. Experimental studies on complex GHz-range dynamics will be shown in chapters 4 and 6, for realistic ferromagnetic microstructures and thin (multi)layers.
1.2 Local pump-probe measurement technique

Magnetization dynamics on the nanosecond timescale was possible to study since decades, e.g. by the FMR (ferromagnetic resonance) or ESR (electron spin resonance) techniques. However, these methods rely on measuring the spectrum of the resonant absorption of energy by an entire ferromagnetic object, therefore the technique is global and in the frequency domain.

Since the advent of ultrafast (pico- and femtosecond) mode-locked lasers, a new technique is available for measuring the nanosecond and faster magnetization dynamics. The technique is exploiting an interaction between polarized light and ferromagnetic materials: the so-called Kerr effect. This effect consists of the following. A linearly polarized light reflected from the surface of a ferromagnetic sample experiences a rotation in its polarization axis (Kerr rotation) and, simultaneously, gains a non-zero ellipticity (Kerr ellipticity). In the limit of small perturbations, these effects are linear with the magnitude of the local magnetization within the studied region. Hence, by comparing the polarization state of the incident and reflected light, one can obtain information on the magnetization state of the sample.

![Figure 1.4: Schematic drawing of a time-resolved magneto-optic measurement (TRMOKE) setup. See text for details.](image)

A schematic drawing of a magneto-optic Kerr effect (MOKE) measurement setup is shown in Fig. 1.4. The main components of the setup are: an electromagnet EM to generate an external field, a laser, a polarizer P1, the ferromagnetic sample S, a second polarizer (analyzer) P2 and a detector D. The electric signal output of the detector is connected to a data acquisition device (e.g. personal computer) PC.

To obtain spatial resolution, the laser light can be focused on the sample by means of a high numerical aperture lens L. In our setup, a resolution of 1 µm was reached. The reflected light can be collected by the (same or another) lens and directed towards the detector.

Modulation techniques (e.g. polarization modulation and lock-in amplifiers) can be used to obtain a signal-to-noise ratio as good as necessary.

To measure the fast dynamics of the magnetization however, the experiment has to contain a fast perturbation at time zero, followed by a detection. The perturbation
can be of different natures; we used femtosecond laser heating for some experiments and external magnetic field pulses for others. The detection is done by the Kerr effect using a mode-locked pulsed laser of pulse length < 100 fs and repetition rate 80 MHz. The time delay $\Delta t$ between the perturbation (“pump”) and detection (“probe”) can be controlled by a delay generator marked DG in Fig. 1.4. This can be simple and accurate to the level of femtoseconds: a mirror in the path of the probe beam moved by a computer-controlled translation stage can make the optical path of the probe pulses shorter or longer. The experiment is stroboscopic: in every second, 80 million pump-probe cycles are executed, while the delay time between them is increased step by step. This way, a magneto-optic response to the external perturbation, proportional to the local magnetization, can be probed directly in the time domain.

Generally, the MO response (the Kerr rotation/ellipticity) represents a combination of the responses of all the local magnetization vectors within the probed region defined by the space vector $\vec{r}_0$ (the laser spot):

$$MO(\vec{r}_0,t) = \int F(\vec{r},\vec{r}_0,t)\vec{M}(\vec{r},t)d\vec{r}.$$ 

The factor $F$ is, in principle, unknown. Its time-dependence is discussed in [4] and it proves to be important in case of femtosecond dynamics. As to its spatial dependence, in case of a homogeneous thin film multilayer $F$ is not dependent on $\vec{r}_0$. Moreover, for the fundamental precessional mode $k = 0$, as we will see later, $F$ is also independent of the lateral dimensions ($x$ and $y$) of the probed region. The dependence on the “depth” $F(z)$ indicates that different layers within the probed volume could have different contributions to the final MO response. In case of laterally confined or strongly non-uniform layers, additional local variations in $F$ could disturb the $MO(M)$ linearity. Despite of these considerations, in our measurements the nonlinearity appears to be insignificant; we will see that the MOKE technique produces very useful and consistent results on the microscopic and (sub-)nanosecond scale.

1.3 This thesis

The work presented in this thesis focuses on the local magnetization dynamics of ferromagnetic structures on the nanosecond and subnanosecond timescale. Particular attention is given to the influence of sample structure, neighboring layers, size, external perturbations and couplings upon the precessional parameters (damping $\alpha$ and frequency $\omega$) with a close watch on the consequences for future applications.

After a brief introduction to the ultrafast (femtosecond) demagnetization phenomena, a theoretical description of the GHz precessional dynamics will be given in Chapter 2, including magnetic anisotropies and spin wave generation/localization in microstructures and thin films. The Landau-Lifshitz-Gilbert equation will be introduced as the main equation governing the precessional dynamics of a macrospin, and it will be extended to the space-dependent magnetization and effective magnetic field. The intrinsic and extrinsic contributions to the damping will be discussed as well.

In Chapter 3 several magnetization detection techniques will be discussed, with a complete description of our experimental setup, the time-resolved magneto-optic Kerr setup. The Kerr-effect as detection method for magnetization dynamics will
be described, and a vectorial version of the technique will be shown, developed to measure multiple spatial components of the magnetization simultaneously. Two ways of magnetization perturbation will be presented: pumping with external magnetic field and pumping with ultrafast laser heating. In the frame of the first pumping scheme, the problematics of generating sub-nanosecond magnetic field pulses will be treated, by means of an electronic waveguide design. Within the description of the second, optical pumping scheme, the anisotropy field pulse and its characteristics will be discussed. The technique will be demonstrated with some examples from our experimental results.

The experiments on local magnetization dynamics in Permalloy microstructures will be presented in detail in Chapter 4. A number of magnetic couplings important for such systems will be described. Then, starting with simple Py discs, elementary precessional dynamics will be shown, followed by the spin wave detection in antiferromagnetically coupled microscopic Py discs, so-called artificial spin chains. Finally, measurements on the Py elements of magnetic tunnel junctions (MTJ) will be presented. Within these, studies on localized spin waves, edge modes and a dynamic domain imaging technique will be shown. The results will be compared to theoretical predictions and micromagnetic simulations on similar structures, and conclusions will be drawn with respect to the importance of local effects. Measurements on large-angle precessional dynamics will be presented too, and the possibility of a precessional switching technique will be considered.

Chapter 5 will present a technique to back-trace magnetic field pump pulses generated in microscopic waveguide structures. The goal is to obtain a complete image of the field pulse profiles on the picosecond timescale, an important factor in fast micromagnetic applications such as MRAM. The method will be extended to the anisotropy field pulses generated by ultrafast laser heating, yielding interesting results both qualitatively and quantitatively.

In the last chapter the so-called spin pumping effect will be presented as possible additional damping channel for normal metal/ferromagnetic bi- or multilayers. Our experimental search for the effect will be shown via systematic measurements on several wedge-shaped NM/FM samples. The influence of the sample morphology on the effective damping will be discussed as well. Our results will be compared to similar experiments published recently in the literature, commenting on the discrepancies we have noticed.

At the end of Chapters 4, 5 and 6 a brief section is dedicated to the general conclusions on the presented work. In these sections, the relevance of the results to the current state of fundamental research and to future applications will also be addressed.
Chapter 2

An overview of fast magnetization dynamics

In this chapter we introduce the reader to the theoretical background of fast magnetization dynamics. After a brief description of the femtosecond demagnetization phenomena, we will focus on the effects subject to our experimental studies. These are effects in microscopic ferromagnetic entities, happening on the timescale of not more than a few nanoseconds, and they are governed by the so-called Landau-Lifshitz-Gilbert equation of magnetization motion. We will present the macrospin theory and the precessional dynamics, the Gilbert damping, the LLG-equation and its physical relevance. Some LLG-simulations will be included in order to create an easy-to-follow image of the dynamics that will be shown in the latter experimental chapters. Extrinsic damping terms will be discussed, as well as the generation of local precessional modes, i.e. lateral spin waves. Finally, the special case of perpendicular, standing spin waves in thin films will be presented.

2.1 Ultrafast demagnetization - mechanisms and time scales

The fastest effect encountered in our (all-optical type) measurements shown in this thesis is the laser-induced femtosecond demagnetization. It is well known, that ferromagnetism turns into paramagnetism above a certain temperature, called Curie temperature. The net magnetization of a ferromagnet decreases with increasing temperature due to the loss of long-range order. At the first glance, the reaction of magnetization to the increase of temperature seems to be instantaneous. However, the developments in the field of femtosecond pulsing lasers allow for magneto-optic studies of this ”ultrafast demagnetization”. A strong laser pulse of a duration below 100 fs, focused on a thin ferromagnetic sample, lead to sudden local heating of the sample (with 50 K or more); the reaction of the magnetization to this heating can be monitored by the second, weak laser pulse via the magneto-optic Kerr effect as described in chapter 3.

Microscopically, optical excitation leads to an instantaneous increase of the energy
CHAPTER 2. FAST MAGNETIZATION DYNAMICS

of the “electron sea” by absorption of the photons, creating a non-equilibrium distribution. On a 100 fs timescale, the electrons will thermalize and reach an equilibrium distribution at an elevated temperature. The heating of the crystal lattice follows on a longer timescale, up to 3 ps (in case of 6 monolayers of Ni on Cu(001), see [5]), via inelastic electron-phonon scattering and lowering the electron temperature. However, the magnetization is reduced by *spin scattering processes*. The question arises, whether the lattice is involved in the mechanism of magnetization loss or not. One method of answering this question, is to perform time-resolved measurements on the (sub-)picosecond timescale.

The first light-induced demagnetization experiments in the eighties, by Agranat et al. [6] and later by Vaterlaus et al. [7, 8], indicated a spin relaxation time of 100±80 ps (in Gd). This order of magnitude was in good agreement with the theoretical estimate of Hübner et al. [9] based on spin-lattice relaxation, yielding demagnetization times of hundreds of picoseconds with a large variation for different ferromagnets.

In 1996 however, Beaurepaire et al. performed time-resolved magneto-optical experiments on thin Ni films with laser pulses of 60 fs [10]. The result, surprisingly, showed that the magnetization of a 22 nm thick Ni film considerably dropped and reached a minimum already at 2 ps after the onset of the heating pulse (see Fig. 2.1). The conclusion was drawn based on a so-called “three-temperature model” for the dynamics: the spin-lattice interaction was assumed too slow to explain this very fast spin relaxation, therefore an efficient electron spin scattering was considered.

![Figure 2.1: The results of the pump-probe magneto-optic Kerr measurement performed by Beaurepaire et al. (1996) on a 22 nm thick Ni film using 60 fs laser pulses. The figure, taken over from Ref. [10], shows that the magnetization reaches a minimum within 2 ps, nearly three orders of magnitudes faster than predicted and measured previously.](image)

Shortly after Beaurepaire’s experimental demonstration, a number of research groups obtained demagnetization results on a variety of samples, using different methods of time-resolved magnetization detection [11, 12, 13, 5, 15, 4]. Most of these results indicated demagnetization timescales even faster than the original 2 ps value, down to 100 fs, challenging the experimental setup’s temporal resolution. Moreover, measurements on electron dynamics indicated an electron thermalization happening on the same 100 fs timescale.
2.2. PRECESSIONAL DYNAMICS

So far the magneto-optic effects were considered to be linear with the magnetization. However, serious doubts were rising when measurements of complete laser-induced magnetization loss in ferromagnets was reported on even shorter timescales by Conrad et al. [5, 14]. Additionally, in several experiments by Regensburger et al. [15] and Koopmans et al. [4] the dynamics was found to depend strongly on the measured polarization component, indicating that the correlation of magnetism and magneto-optics in the laser-excited state is very complicated. With respect to the experiments presented later in this thesis we have to remark, that on the picosecond timescale the MO effects still properly scale with the magnetization [4], therefore they can be used for slower magnetic processes such as precessional dynamics or switching.

The details and the latest models of the laser-induced ultrafast demagnetization phenomena are out of the scope of this thesis; these can be found in the references cited above as well as in [16] and [17]. Also, interesting recent results can be found on ultrafast creation of ferromagnetic ordering in [18, 19], considered also from the applications point of view.

2.2 Precessional magnetization dynamics

2.2.1 Premises

On a slower timescale, typically hundreds of picoseconds, the demagnetization process can be followed by a directional change of the magnetization - a precessional motion in space. Such a motion can be experimentally excited both magnetically (field pulse) and optically (laser heating), as we will show later in chapter 3. The main condition for the precessional dynamics is a nonzero angle between the magnetization equilibrium direction and its actual direction at the moment \( t_0 \).

To explain the onset of such a precession, let us consider the case of a single spin \( \vec{S} \) in a DC external magnetic field along the x-axis \( \vec{H}_{DC} \). If the spin forms a nonzero angle \( \theta \) with the direction of the field, a torque will act on its magnetic moment, perpendicular to the plane formed by the spin and the magnetization directions, that can be written as:

\[
\vec{T} = -|\gamma|\mu_0\vec{h}\vec{S} \times \vec{H}_{DC},
\]

where \( \gamma = \frac{g\mu_B}{h} \) is the gyromagnetic ratio, \( \mu_0 \) is the vacuum permeability and \( \vec{h}\vec{S} \) represents the angular momentum of the spin. The action of this torque can be derived both classically as presented by J. Miltat in [36] and via quantum mechanics; we will use only the latter. The Schrödinger equation describes the temporal evolution of the mean value of spin as operator:

\[
i\hbar\frac{d}{dt} <\vec{S}> (t) =< [\vec{S}, \vec{H}(t)]>,
\]

where \( \vec{H}(t) \) represents the Hamiltonian of a spin under the action of a magnetic field, i.e. the Zeeman term:

\[
\vec{H}(t) = -\frac{g\mu_B}{\hbar}\mu_0\vec{S} \cdot \vec{H}.
\]

Using the commutation rules, the three components of the right-hand side of the Schrödinger equation 2.2 can be written as

\[
[S_z, \vec{H}(t)] = -\frac{g\mu_B}{\hbar}\mu_0i\hbar(H_y(t)S_z - H_z(t)S_y);
\]
\[ [S_y, H(t)] = -\frac{g\mu_B}{\hbar}\mu_0 i\hbar (H_x(t)S_x - H_x(t)S_z); \]

\[ [S_z, H(t)] = -\frac{g\mu_B}{\hbar}\mu_0 i\hbar (H_x(t)S_y - H_y(t)S_x). \]

Therefore, eq. 2.2 can be rewritten containing a torque on the right-hand side:

\[
\frac{d}{dt} \langle \vec{S} \rangle (t) = -\frac{g\mu_B}{\hbar}\mu_0 (\langle \vec{S} \rangle \times \vec{H}(t)).
\]

(2.4)

This result is based on a general time-dependent \( \vec{H}(t) \), however it is valid for our special case, a DC external magnetic field as well. Denoting the magnetic moment of the spin as \( \vec{\mu} = -|\gamma|\hbar\vec{S} \), its dynamics under the effect of the torque 2.1 will be governed by the equation

\[
\frac{d\vec{\mu}}{dt} = -|\gamma|\vec{T}.
\]

(2.5)

Since we defined \( \vec{H}_{DC} = H_{DC} \cdot \hat{x} \), the above equation can be written as

\[
\begin{align*}
\frac{d\mu_x}{dt} &= 0; \\
\frac{d\mu_y}{dt} &= -|\gamma|\mu_0 (\mu_z H_x); \\
\frac{d\mu_z}{dt} &= -|\gamma|\mu_0 (-\mu_y H_x).
\end{align*}
\]

(2.6)

This set of differential equations has the trivial solution for the magnetic moment

\[
\begin{align*}
\mu_x(t) &= \mu \cos \theta = \text{const}; \\
\mu_y(t) &= \mu \sin \theta \cos(\omega t); \\
\mu_z(t) &= \mu \sin \theta \sin(\omega t);
\end{align*}
\]

(2.7)

yielding a precessional motion around the x-axis (the direction of the DC field) as plotted in Fig. 2.2, with the Larmor-frequency \( \omega = -|\gamma|\mu_x H_{DC} \). One can see that in this description, neglecting dissipation channels, the precessing spin will never align with the DC magnetic field, the angle \( \theta \) between the two vectors being kept constant.

![Figure 2.2: Macrospin precession: An ensemble of spins in a coherent precession (same phase, same frequency) under the influence of a uniform, isotropic external DC field. If no dissipation is considered, the amplitude of the torque T as well as the angle \( \theta \) is conserved through the precession. The magnetic moment of one single spin is denoted by \( \mu \).](image-url)
2.2. PRECESSIONAL DYNAMICS

2.2.2 Macrospin - coherent dynamics in finite sized structures?

Experimentally, we always measure the dynamics of an ensemble of spins. If these are precessing incoherently, with different frequencies and/or out of phase, the net measurement signal will be zero. However, it is possible to start a coherent precession of a large number of spins \( \vec{S}_i \) in a ferromagnetic sample, within the range of the measurement, the condition being an effective DC field over the whole region of interest (yielding a uniform Larmor frequency) as well as fixing a unique initial phase for the whole ensemble of spins, as drawn in Fig. 2.2. In this case, the precessing spins can be considered as one single (vectorial) entity called \textbf{macrospin}. The magnetic moments \( \vec{\mu}_i \) of the single spins add up to form the \textbf{magnetization vector} \( \vec{M} = \sum \vec{\mu}_i \).

Expanding the torque equation 2.5 from the moment of one spin to the full magnetization will result in a dynamics of precessional nature for the macrospin:

\[
\frac{d\vec{M}}{dt} = -|\gamma|\mu_0 (\vec{M} \times \vec{H}_{DC}). \tag{2.8}
\]

The macrospin precessional frequency \( \omega \) is the same Larmor frequency as in case of the single spins.

A condition to the existence of a macrospin state is to have a uniform effective magnetic field over the considered region. This field is built up from the applied external field, a set of anisotropy fields and an exchange field if the magnetization is spatially varying. The magnetic anisotropy represents preferential ("easy") magnetization directions in space; once aligned in these directions, the magnetic energy of the system reaches a local minimum. A model for the energy function was implemented by Stoner and Wohlfarth [20], valid for the single-domain regime. The effect of the anisotropies is similar to an additional applied field, therefore it can be treated as such; in general, an anisotropy field can be derived from the anisotropy energy through the relationship

\[
\vec{H}_{anis} = -\frac{1}{|\vec{M}|} \nabla E_{anis}(\vec{M}). \tag{2.9}
\]

**Anisotropies**

A number of anisotropies can coexist in a ferromagnetic sample, rising from the different interactions of the individual spins with each other and with their environment. \textbf{Shape anisotropy} is the result of long-range magnetic dipolar interaction and is strongly influenced therefore by the position of the sample boundaries. The anisotropy field associated to it can be written as

\[
\vec{H}_{shape} = -\overline{N}\vec{M}, \tag{2.10}
\]

where \( \overline{N} = \sum N_{ij}\hat{i}\hat{j} \) is the demagnetization tensor \((i, j \in \{x, y, z\})\). The most frequently met shape anisotropy in our measurements is the in-plane anisotropy of a thin film: all the tensor elements are zero, except \( N_{zz} = 1 \), leading to an unfavorable out-of-plane magnetization direction. For such a sample, typically an external field of hundreds of kA/m has to be applied to cant the magnetization significantly out of plane. An even more restricting anisotropy is the uniaxial one, where one single preferential axis is available for a stable magnetization direction.
Spin-orbit and dipole-dipole interactions can lead to a so-called \textbf{magneto-crystalline anisotropy} in, evidently, crystalline materials. This reflects the symmetry of the lattice, therefore it can be derived for each specific case from the lattice structure. The lattice plays a crucial role in these interactions, and any change to its parameters will lead to a modification of the magneto-crystalline anisotropy. Between others, a strong dependence on the local temperature has to be considered.

The magneto-crystalline anisotropy can be altered by adding a strain to the crystal lattice (e.g., by epitaxial growth of multilayers of different metals). This effect can be considered as an additional, \textbf{magneto-elastic} anisotropy. In some “sandwiched” ferromagnetic thin layers this strain-based anisotropy can create an out-of-plane preferential direction, directly competing with the shape anisotropy. The final easy-axis direction therefore will be decided by details of sample geometry, such as the thickness of the FM layer and lattice constants. On the other hand, sudden local heating of a thin ferromagnetic film will lead to a perpendicular expansion of its lattice resulting, again, in a strain over the lattice. This way a unidirectional magneto-elastic anisotropy field can be created by ultrafast laser heating. Together with the temperature-dependence of the magneto-crystalline anisotropy, the strain can be used to change the magnetization equilibrium direction on a picosecond timescale and launch a precession (see the experimental chapters of this thesis).

Besides the bulk component, magneto-crystalline anisotropy also has a surface component, i.e. we can talk about a \textbf{surface anisotropy} important in case of very thin films. At surfaces and interfaces where the lattice symmetry is broken, the surface anisotropy can be enhanced to a level of exceeding the bulk crystalline anisotropy by as much as two orders of magnitude \cite{22}. In case of several thin film systems, this leads to perpendicular anisotropy below a certain critical film thickness $d$ (typically a few monolayers) yielding a magnetization oriented out-of-plane. However, the surface contribution to the overall thin film magnetic anisotropy diminishes with $1/d$.

In case of realistic ferromagnetic samples, the torque equation for the macro-spin (eq. 2.8) is only valid if $H_{DC}$ contains all the anisotropy fields present in the region of interest. Some of these fields might be time-dependent directly or via their dependence on the magnetization of the film, therefore the “DC” suffix will be further omitted.

### 2.2.3 Inducing a gigahertz precession in magnetic micro-structures

We have seen from eq. 2.8 that a magnetic field applied in a nonzero angle versus the magnetization’s equilibrium direction will drive this into a precessional motion. The precessional axis will be the new equilibrium direction, defined by the applied magnetic field as well as all the fields existing inside the sample, rising from anisotropies, adjacent magnetic layers etcetera. In order to have a free precession, ideally the field has to be applied suddenly, as a step-edge. In experiments, a rising time shorter than the precessional period $T = 2\pi/\omega$ will suffice and can be reached with a number of techniques (see chapter 3 of this thesis).

Let us consider an example, the case of a thin ferromagnetic film with shape anisotropy only. We denote the demagnetization factors along the three axes with $N_{x,y,z}$; a DC field along the x-axis is present. From eq. 2.10 results that the spatial
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components of the effective magnetic field are

\[
H_x = H_x^{DC} - N_x M_x; \\
H_y = -N_y M_y; \\
H_z = -N_z M_z.
\]  

(2.11)

With a calculus similar to the case of a single spin starting with eq. 2.6, we can obtain the precession frequency of the magnetization in a sample dominated by shape anisotropy. The resulting formula is the so-called Kittel-formula:

\[
\omega = \gamma \mu_0 \sqrt{[(N_z - N_x)M_s + H_x^{DC}][(N_y - N_x)M_s + H_x^{DC}]},
\]

(2.12)

where \(M_s\) is the saturation magnetization and we consider a small-angle precession, where \(M_x = M_y = \text{const}\). This result shows, that the precession frequency depends not only on the DC field but on the properties of the ferromagnetic microstructure too.

The frequency of the precessional motion can be estimated from the Kittel-formula. For the case of a thin film transition metal sample, a typical example used in our experiments, the saturation magnetization \(M_s\) is in the range of 1000 kA/m and the demagnetization factors are \(N_x = N_y = 0\) and \(N_z = 1\). The gyromagnetic ratio \(\gamma = g \mu_B / \hbar \simeq 185 \text{ GHz/T}\) (with the Lande factor \(g = 2.1\)) and the vacuum permeability is \(\mu_0 = 4\pi \cdot 10^{-7} \text{Tm/A}\). Applying a DC field of, say, 10 kA/m results in a Larmor precession frequency \(\omega \simeq 23 \text{ GHz}\).

2.2.4 Introduction of the damping

The torque equation 2.8, derived from fundamental considerations, indicates a precessing magnetization without dissipation. The conservation of the precession angle becomes evident if we multiply this equation with the DC magnetic field \(\vec{H}_{DC}\) in the scalar manner, from the right-hand side. In reality however, the magnetization precession does relax to the new equilibrium direction, associated with an energy minimum, thus the torque equation has to be extended with a dissipation term.

This new term was first added to eq. 2.8 by Landau and Lifshitz [1] in the form of a mathematical construction of a torque component proportional to \(\vec{M} \times (\vec{M} \times \vec{H})\). This torque acts in the plane formed by \(\vec{M}\) and \(\vec{H}\), orthogonal to the precessional torque, forcing the magnetization to align with the field. The Landau-Lifshitz equation of magnetization motion has thus the form

\[
\frac{d\vec{M}}{dt} = -|\gamma|\mu_0(\vec{M} \times \vec{H}) - \frac{\lambda}{M_s^2}[\vec{M} \times (\vec{M} \times \vec{H})],
\]

(2.13)

with \(\lambda\) being a factor for the damping term and \(M_s\) the saturation magnetization.

The damping term of the Landau-Lifshitz equation is introduced mathematically, satisfying the need for a dissipation. However, physically there is no clear explanation behind this extension and the definition of the damping factor \(\lambda\) itself is, so far, vague.
2.2.5 The Landau-Lifshitz-Gilbert equation

Gilbert introduced the dissipation term in the torque equation with a more convenient form [2]. He interpreted the dissipation as an additional contribution next to the effective field proportional to and aligned with the variation of the angular momentum:

\[
\vec{H} \rightarrow \vec{H} + \frac{\alpha}{-|\gamma|\mu_0 M_s} \frac{d\vec{M}}{dt},
\]

(2.14)

where the constant \( \alpha \) is the so-called phenomenological Gilbert damping parameter. This substitution transforms the torque equation for the macrospin 2.8 into the so-called Landau-Lifshitz-Gilbert equation of magnetization motion:

\[
\frac{d\vec{M}}{dt} = -|\gamma|\mu_0 (\vec{M} \times \vec{H}) + \frac{\alpha}{M_s} \left( \vec{M} \times \frac{d\vec{M}}{dt} \right).
\]

(2.15)

In conclusion thus, the LLG equation of magnetization motion indicates that a suddenly applied magnetic field exerts a torque on the magnetization vector, resulting in a gigahertz precession with a damping. Qualitatively, the theory is valid for magnetization switching as well: after suddenly reversing the applied magnetic field, the damping of the precessional motion leads to a gradual alignment of the magnetization to its new equilibrium direction. Practically, the switching speed of a ferromagnetic structure is thus limited by the timescale of the damping. This fact elevates the Gilbert damping constant to a very high level of importance, igniting a large number of experimental studies on it.

Let us continue with our example of small excitation on a thin ferromagnetic layer’s magnetization, and extend the calculus with the Gilbert form of the damping. Similar to the procedure in section 2.2.3, where we calculated the precession frequency for a dissipation-free system, from the vectorial LLG equation we can obtain a system of the form

\[
\begin{align*}
\frac{dM_x}{dt} &= 0; \\
\frac{dM_y}{dt} &= \gamma\mu_0 \left( M_x + H_{Dx} \right) M_z - \alpha \frac{dM_z}{dt}; \\
\frac{dM_z}{dt} &= -\gamma\mu_0 H_{Dx} M_y + \alpha \frac{dM_y}{dt}.
\end{align*}
\]

(2.16)

Here we considered, again, that the perturbation is small and \( M_x \approx M_x = \text{const.} \) A solution for this system is

\[
\begin{align*}
M_y &= \cos(\omega t)e^{-t/\tau}; \\
M_z &= \epsilon \sin(\omega t + \phi)e^{-t/\tau},
\end{align*}
\]

(2.17)

with \( \epsilon \) representing the ellipticity of the precession – due to the strong in-plane anisotropy, a “flattening” of the precession has to be considered. Substituting the solution into eq. 2.16, we obtain the expressions:
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\[ \omega = \gamma \mu_0 \sqrt{AH_x^{DC}(H_x^{DC} + M_s)} - \alpha^2 M_s \left( \frac{2}{2(\alpha^2 + 1)} \right) ; \quad \tau = \frac{2(\alpha^2 + 1)}{\gamma \mu_0 \alpha (2H_x^{DC} + M_s)} ; \]

\[ \phi = \text{arcsec} \left( \frac{2\sqrt{H_x^{DC}(H_x^{DC} + M_s)}}{\sqrt{AH_x^{DC}(H_x^{DC} + M_s)} - \alpha^2 M_s} \right) ; \quad \epsilon = \sqrt{\frac{H_x^{DC}}{H_x^{DC} + M_s}}. \] (2.18)

When \( \alpha \ll 1 \), we can consider \( \alpha^2 \simeq 0 \), and the expressions will simplify considerably:

\[ \omega = \gamma \mu_0 \sqrt{H_x^{DC}(H_x^{DC} + M_s)} ; \quad \tau = \frac{2}{\gamma \mu_0 \alpha (2H_x^{DC} + M_s)} ; \]

\[ \phi = \frac{\alpha M_s}{\sqrt{AH_x^{DC}(H_x^{DC} + M_s)}} ; \quad \epsilon = \sqrt{\frac{H_x^{DC}}{H_x^{DC} + M_s}}. \] (2.19)

We can see, that introducing a damping term has no effect on the precessional frequency \( \omega \). Measuring one of the two magnetization components \( M_y(t) \) or \( M_z(t) \) will thus result in a sine-type oscillation with an exponential damping as shown in Fig. 2.3; a fit to the measurement with the corresponding function 2.17 will allow us to experimentally determine the factors \( \omega, \tau \) and \( \phi \). From the value of the characteristic damping time \( \tau \) we can calculate the Gilbert damping constant \( \alpha \). This method was used for the quantitative study of the damping through our precessional experiments (both the magnetic field pulse and the all-optical type), presented in chapters 4, 5 and 6.

![Figure 2.3: Time-resolved measurement of the out-of-plane magnetization component (5 nm thick NiFe layer), using a 0.6 ns magnetic field pulse, see chapter 4. After the pulse is over, the magnetization follows a free precession towards the equilibrium, and can be fitted with a function of type 2.17 to obtain the precessional frequency \( \omega \) and damping \( \alpha \).](image)

**Mechanism of the intrinsic Gilbert damping**

The mechanism considered for the Gilbert damping in metallic ferromagnets is the scattering of conduction electron spins. Some of these scattering processes involve phonons and spin-orbit interactions. However, a more important role is played by the scattering events caused by exchange interaction between the free electrons and the local magnetic fields. The scattering happens in this case due to the small-scale inhomogeneities of the field, such as defects, domain walls, vortices etc. It is common to associate the *magnon* as a quasiparticle to these spin-scattering interactions. It is experimentally observed, that a single-crystalline ferromagnetic sample always presents...
a lower Gilbert damping parameter, proving the importance of the electron-magnon scattering process.

Theoretically, the phenomenological damping parameter \( 0 \leq \alpha \leq \infty \). We talk about critically damped precessional dynamics, when the magnetization arrives to its new direction in a time comparable or shorter than a precessional period, case described by the relationship \( \omega \tau \simeq 1 \). For thin ferromagnetic films with \( M_s \) in the range of 1000 kA/m and small effective fields of \( \simeq 10 \) kA/m, using the formulae 2.19 we find that the precession is already critically damped for \( \alpha \simeq 0.2 \). However, in the case of our experiments on (continuous or patterned) thin layers of transition metals we only encounter an underdamped precession: \( \alpha \) lies between 0.01 and 0.03. Within these limits, the damping shows a variation influenced by a number of sample parameters and the geometry of the experiment. Details about the additional damping mechanisms will be given after the introduction of spin waves, as well as at the discussion of the experimental results in chapter 4.

### 2.2.6 LLG simulations on damped precession

Based on the Landau-Lifshitz-Gilbert equation, the precessional motion of a slightly perturbed macrospin can be predicted. In the following paragraphs, we will show a set of numerical simulations, built on the knowledge of the material parameters \( \alpha, M_s \) and \( \gamma \) as well as the effective magnetic field \( \vec{H}(t) \).

The Landau-Lifshitz form of the equation of magnetization motion 2.13 is more convenient to use, since it can be linearized and used in an iterative calculus of the magnetization at time \( t \). To keep the Gilbert damping parameter \( \alpha \) in the calculus, the Landau-Lifshitz equation can be rewritten as

\[
(\alpha^2 + 1) \frac{d\vec{M}}{dt} = |\gamma|\mu_0(\vec{M} \times \vec{H}) + \frac{\alpha|\gamma|\mu_0}{M_s}[\vec{M} \times (\vec{M} \times \vec{H})],
\]

(2.20)

where both \( \vec{M} \) and \( \vec{H} \) are time-dependent.

![Figure 2.4: Geometry of the LLG-simulation. \( t = 0 \): magnetization in equilibrium aligned with the bias field. \( t > 0 \): new equilibrium direction defined by the effective field, magnetization starts to move.](image)

We applied the model to the magnetization dynamics of a thin Permalloy film with the parameters \( \alpha = 0.01 \) and \( M_s = 900 \) kA/m, values obtained from literature [80, 99]. The geometry of the sample, fields and magnetization is plotted in Fig. 2.4. A DC external bias field \( H_{bias} = 2.4 \) kA/m is applied in-plane, along the x-axis, creating an initial equilibrium direction for the macrospin. A second external field \( H_p(t) \) is applied, also in-plane but perpendicular to the DC field, in form of a step-edge, respectively pulse of a well-defined length and maximum amplitude of 0.44 kA/m. The sample is considered to have a strong in-plane anisotropy, characterized by \( N_x = \)
2.2. PRECESSIONAL DYNAMICS

$N_y = 0$ and $N_z = 1$, the resulting demagnetizing field being oriented out-of-plane: $H_{demag}(t) = -N_z M_z(t) = -M_z(t)$. The time-dependent effective field acting on the macrospin is then

$$\vec{H}(t) = (H_{bias} \hat{x}, H_p(t) \hat{y}, -M_z(t) \hat{z}).$$

(2.21)

The length of the magnetization vector is kept constant and considered fully aligned with the bias field at the moment $t = 0$. Formulae 2.19 yield a characteristic damping time $\tau = 0.95$ ns and a precessional frequency $\omega = 10.8$ GHz. The calculated $M_y$ and $M_z$ magnetization components are plotted in Fig. 2.5; their values are normalized to the saturation magnetization $M_s$, while the time scale is normalized to the characteristic damping time $\tau$. First, the effect of a step-edge is shown: the inplane $M_y$ component is oscillating around a nonzero value, indicating a new equilibrium direction for the magnetization. The $M_y$–$M_z$ plot helps visualizing the movement (damped precession) of the magnetization vector projected to the $yz$-plane. In the second pair of plots (Fig. 2.5 c, d) a 1.5 ns long magnetic field pulse is applied instead of a single step-edge. This case is more interesting from the point of view of our stroboscopic experiments, where using a step-edge would not be possible. On the $M_y$–$M_z$ plot two distinct precessional axes can be seen, corresponding to the onset and offset of the field pulse.

Varying the length of the magnetic field pulse, the amplitude of the second precession (around the x-axis) can be controlled. If, e.g., the falling edge of the pulse is coincident with the maximum $M_y$-value within that period (see Fig. 2.5 d, right

Figure 2.5: Simulated magnetization dynamics when applying a step-edge magnetic field (a,b) and a 1.5 ns long pulsed field (c,d). The shift of the equilibrium direction and the precession around it is revealed.
a large torque will act on the magnetization vector yielding a precession of increased amplitude. If the damping $\alpha$ is small enough compared to the precessional frequency $\omega$, there can be different moments during the precession when the values for $M_y$ and, respectively $M_z$ are the same, yielding intersections or “touching” points of the $M_y - M_z$ curves as indicated by the left arrow. The ellipticity of the precession $\epsilon = 0.051$ meaning, that the magnetization stays very close to the in-plane orientation (note the different scales used in the $M_y$ and $M_z$ plots). This distortion is due to the strong in-plane anisotropy of the film.

2.3 Spin waves

So far we have discussed the case of the macrospin dynamics, where all the spins in the region of interest are uniformly excited and precessing under the action of a uniform effective field as one single vectorial entity. If we allow the magnetization $\vec{M}$ as well as the effective magnetic field $\vec{H}_{eff}$ to vary through the region of interest, we can formulate a non-local extension of the LLG equation having a form similar to the local version 2.15:

$$\frac{d\vec{M}(\vec{r}, t)}{dt} = -|\gamma|\mu_0[\vec{M}(\vec{r}, t) \times \vec{H}_{eff}(\vec{r})] + \frac{\alpha}{M_s}(\vec{M}(\vec{r}, t) \times \frac{d\vec{M}(\vec{r}, t)}{dt}). \quad (2.22)$$

In Fig. 2.6 we plotted a basic representation of the uniform macrospin precession paired with a precession of a one-dimensional spin system where the motion of the individual spins is out of phase (albeit the frequency is the same). Such a case can be experimentally realized e.g. if the perturbation is localized, thus non-uniform over the spin system; a propagating magnetic excitation will be generated, called spin wave. Spin waves can be launched with a uniform perturbation too, if there is an implicit phase difference from the start. The initial orientation of the spins can be non-uniform due to a dispersion in the effective magnetic fields, sample morphology, magnetic domain structure and others. Finally, if the precessional frequency varies in space for whatever reason, then the precessing spins will dephase after some time, even if the dynamics were started with the same phase.

As any proper wave, spin waves can be described by a wavelength $\lambda$ and a wave vector $k$ (sometimes also denoted with $\vec{q}$ in literature). The uniform precessional
mode for infinitely large systems is in fact a special case of spin waves where the wave vector (and the phase dispersion) is zero and the wavelength is infinite, called therefore the fundamental mode.

If the length of the magnetization vector can be considered locally constant, we can extend the torque equation 2.8 to be valid for the spin system in the following way. A variable magnetization $\vec{m}(\vec{r}, t)$ has to be added to the saturation magnetization to form the total magnetization vector:

$$\vec{M} = \vec{M}_0 + \vec{m}(\vec{r}, t).$$

In the same time, the effective magnetic field $\vec{H}_{eff}$ in the torque equation includes all the possible local fields resulting from interactions in the magnetic system, and is therefore space- and time-dependent as well (thus it is not a DC field anymore), $\vec{H}_{eff} = \vec{H}_{eff}(\vec{r}, t)$.

In the limits of a small perturbation (small-angle precession, $|\vec{m}| << |\vec{M}_0|$), the variable magnetization can be expanded in a series of plane magnetization waves with the wave vector $\vec{k}$, representing the spin waves we have introduced above:

$$\vec{m}(\vec{r}, t) = \sum_{k} \vec{m}_k(t)e^{i\vec{k}\vec{r}}.$$  

The wave component with $k = 0$ corresponds to the fundamental mode; by annihilating the exponent in the above formula, this mode is only time-dependent. In the following paragraphs we will discuss a variety of thin film precessional geometries and the spin waves commonly associated with them.

2.3.1 Lateral spin waves in microscopic ferromagnets

In a thin ferromagnetic film, the spin waves generated in a region can travel laterally, in the film plane (denoted as the $xy$-plane) or perpendicularly into the depth of the film, along the $z$-axis. The former types will have their wave vector $\vec{k}$ in the film plane and will show up in literature as lateral spin waves, or lateral magnetostatic modes. Based on the orientation of the saturation magnetization $\vec{M}_0$, identical to the precessional axis of the individual spins, we can make distinction between three special cases of lateral spin waves.

If $\vec{M}_0$ is in-plane and perpendicular to the wave vector $\vec{k}$, the spin wave will be the so-called magnetostatic surface mode (MSSM), as shown in Fig. 2.7. This mode was first described by Damon and Eschbach [37] and is therefore also called Damon-Eschbach mode.

If the saturation magnetization $\vec{M}_0$ and the wave vector $\vec{k}$ are parallel in the film plane, the magnetostatic backward volume mode (MSBVM) will exist. As explained by Demokritov et al. in [36], this mode has a negative dispersion: the group velocity is antiparallel to the wave vector. This type of spin wave is sketched in Fig. 2.8.

The third case is when the film is magnetized out-of-plane, that is, $\vec{M}_0$ is aligned perpendicular to the film plane and to the wave vector as well. Such a case can be experimentally obtained by overcompensating for the in-plane anisotropy field (via other anisotropies or an external bias field). The resulting spin wave will carry the name magnetostatic forward volume mode (MSFVM), and it is shown in Fig. 2.9.
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Figure 2.7: Magnetostatic surface mode (MSSM) or Damon-Eschbach mode, generated in a thin ferromagnetic film extended in the $xy$-plane.

Figure 2.8: Magnetostatic backward volume mode (MSBVM)

Experimentally, the orientation of the saturation magnetization can be modified by the constant and isotropic (internal or external) magnetic fields acting on the sample. On the other hand, the orientation of the wave vector, as mentioned earlier, depends on the phase dispersion of the precessing spins, which can be controlled by
the perturbation method, by a field gradient or by sample morphology.

If we consider a micrometer sized magnetic thin film sample, the value of the in-plane wave vector will be quantized: \( k = n\pi/w \), where \( w \) is the lateral dimension and \( n \) is integer. This way, a number of localized spin waves can be generated in the sample, with distinct frequencies \( \omega_n \) corresponding to the values of \( n \). Since the lateral dimensions – the edges – of the sample can play a key role in the generation of such spin waves, these are often called **edge modes**. These modes were already observed by Jorzick et al. [23, 40] since 1999, as well as by Park et al. [69] in 2002. In our experiments presented in chapter 4 of this thesis, the long-range dipole interaction and the stray field from the underlying CoFe layer results in an inhomogeneous internal effective field within the studied microscopic NiFe sample. The inhomogeneity gives rise to localized spin waves with \( \vec{k}||\vec{M}_0 \), corresponding to the MSBVM geometry. These localized modes show up in the scanning type time-resolved measurements together with the fundamental precessional mode.

### 2.3.2 Perpendicular standing spin waves in a thin magnetic film

We briefly mentioned the possibility that the spin wave travels perpendicular to the sample surface. In a very thin (sub-nanometer) sample, this is practically very difficult to realize due to the need of having a phase difference between the spins along the \( z \)-direction. On the other hand, if the sample is not a thin film but a bulk centrosymmetric ferromagnet, there is no conceptual difference between the absolute orientations of \( \vec{k} \), only the relative orientation of \( \vec{k} \) versus \( \vec{M}_0 \) is important.

In the range of sample thickness of nanometers, however, a perpendicular spin wave can be realized by a number of methods. One of them involves the all-optical pump-
probe technique used by ourselves and described in chapter 3. Briefly, this technique is based on inducing perpendicular strain anisotropy by ultrafast laser heating the ferromagnetic layer in one spot. Since the penetration depth of light in metals is limited to the range of 10-15 nm, the spin system in a ferromagnetic thin film sample of say 30 nm will be non-uniformly perturbed, more intensely in a region close to the surface where most of the light absorption takes place. Therefore, a dispersion in the precessional amplitude for the individual spins is realized in the perpendicular direction. The very thin nature of the sample in the same time imposes a restriction on the values of the wave vector: just as discussed before, this will be quantized as \( k = n\pi/d \) with \( d \) the sample thickness. Hence, besides the fundamental precessional mode \((n = 0)\), a first order perpendicular standing spin wave is also induced in the thin film. With open boundary conditions, this wave has a single node in the middle of the layer and it is free at the surfaces, therefore it can be detected with the MOKE technique.

![Figure 2.10: Perpendicular standing spin wave (PSSW) in a 40 nm thick polycrystalline Ni film. Measurement done with the all-optical time-resolved setup. The first higher order precessional mode \((n=1)\) can be seen next to the fundamental mode \((n=0)\) as a faster oscillatory component of the \( M_z \).](image)

An experimental result on a 40 nm thick polycrystalline Ni layer is plotted in Fig. 2.10. The graph shows the response of the perpendicular magnetization component normalized to the saturation magnetization, plotted versus time. Note that the detection, just as the excitation, happens only down to a depth of 13 nm from the top of the sample. The result is at first glance chaotic, however it is reproducible and can be accurately fitted with two exponentially damped oscillations. The detection of the zeroth and the first order standing spin wave with two distinct precession frequencies \( \omega_{0,1} \) was possible thus with this technique. Higher order modes were so far not observed, a fact that can be attributed to the less efficient excitation and detection of...
2.4. EXTRINSIC GILBERT DAMPING

these modes. Detailed information about the technique and the measurement results on perpendicular standing spin waves can be found in [16, 88].

The advantage of the local excitation technique lies in the possibility of examining a multitude of spin wave phenomena in thin films. As an example, spin wave stiffness can be determined based on such experimental data. In the same time, bulk techniques such as microwave FMR are limited in the detection of spin waves since a nonzero net change of the magnetic moment over the entire volume of the sample is necessary. With FMR, the above mentioned first order mode with open boundary conditions cannot be detected.

A special case of perpendicular standing spin waves will be presented in chapter 4, section 4.2.1. We performed time-resolved measurements on so-called artificial spin chains, consisting of pillars of ten ferromagnetic discs separated by an insulator. The discs are antiferromagnetically coupled via their stray fields acting upon each other. Here, the precessional phase dispersion is present due to the dispersion of the effective magnetic field along the z-axis. Even with a uniform excitation thus, a series of higher order standing wave can be generated in the system.

2.4 Extrinsic Gilbert damping

Besides the intrinsic Gilbert damping mechanism discussed previously, a number of extrinsic mechanisms contribute to the further increase of the effective damping parameter. First of all, an important damping mechanism has to be considered, that does not need the scattering of conduction-band electrons. Especially in very thin films (see quantization of the spin waves in previous sections), a so-called two-magnon scattering is present. The inhomogeneities of the ferromagnetic film will intermediate a transfer of energy between the different modes of the spin precession without involving electrons. Namely, the uniform precessional mode, having the longest wavelength as spin wave, can lose some of its energy by scattering, energy that will be absorbed by shorter wavelength spin waves. These, on their turn, can (and will) lose their energy to the lattice via the common spin-orbit and exchange interactions, as a next step. Detailed aspects of the two-magnon processes and their contribution as extrinsic damping parameter are discussed in [24, 25, 26].

In the case when the excitation of spin waves has a strongly localized nature – e.g. excitation by an intense, focused laser pulse –, we can talk about a radiational damping mechanism. The higher-order, finite-wavelength spin waves are emitted into the unperturbed regions of the sample, waves that can gain energy from the fundamental mode by two-magnon processes. This additional damping was noticed in our all-optical experiments presented in chapter 6. The comparison of our effective damping to the literature values, as well as to the measurements with uniform excitation (e.g. $\alpha_{eff} = 0.014$ versus 0.011 for 10 nm NiFe) consistently yielded a larger value for the all-optical case.

It has been proposed, that yet another extrinsic contribution to the damping results from the so-called spin pumping effect [97]. The drift of a spin current from a ferromagnet into a non-magnetic adjacent metal yields a loss of momentum of the precessing magnetization of the ferromagnet itself. This will manifest macroscopically as an increase in the effective damping. The experimental detection of the additional damping is the subject of chapter 6 of this thesis.
Finally, experiments always have a finite resolution, and there can be a dephasing of the precessing spins within the measurement region. This yields a fast drop of the net magnetic moment, thus an apparent damping that can be very high compared to the intrinsic one. As shown by our measurements in chapter 4, in case of strong non-uniformities of the magnetic field within the measurement region, the dephasing can lead to an effective damping of an order of magnitude higher than the intrinsic value.
Chapter 3

Experimental

3.1 An update on today’s measurement techniques

Since the first suspended magnetite mineral used for navigation purposes, science seems to never exhaust the possibilities locked into magnetism. The number of methods for characterization and imaging of the magnetic properties of materials is continuously increasing as well. Today we have countless methods and devices to measure the magnetization of a sample of any size, shape or nature - down to the atomic level.

There are several directions in which the methods for magnetization measurement developed. Some of the most sensitive techniques to measure the global magnetic response of a distinct sample to external magnetic fields are the SQUID (Superconducting Quantum Interference Device), VSM (Vibrating Sample Magnetometer) and AGFM (Alternating Gradient Force Magnetometer). Other methods help us studying the local magnetic properties of materials on microscopic/nanoscopic scales, like detecting magnetic domain walls and their movement under external influence, “mapping” of magnetic domains of sub-micrometer size, and others. These are magneto-optic microscopy, MFM (Magnetic Force Microscopy), scanning Hall microscopy, TEM/SEM (Transmission/Scanning Electron Microscopy) with magnetic contrast, SPSTM (spin-polarized scanning electron microscopy) and others.

The techniques that allow us to probe the fast/ultrafast behaviour of magnetic properties, i.e. the evolution of magnetization on the (sub)nanosecond timescale, stand closest to the subject of this thesis. Therefore we will briefly present some of them in the following paragraphs. First we will introduce two methods that work in the frequency domain, that is, probe the magnetization dynamics based on spectroscopic measurements. The other option is to probe the changes in magnetization versus time directly; three different time-domain techniques will be presented, of which one is the base of the experimental part of this thesis.

Combination of the time- or frequency domain methods with nanoscopic spatial resolution represents nowadays a challenge for the research laboratories worldwide. However, the results can be rewarding by their contribution to the development of miniature magnetic devices with increased sensitivity and high reliability (sensors, long-term data storage, memory arrays and others).
3.1.1 Ferromagnetic resonance (FMR)

As we have seen before, an external perturbation on a (macro)spin like an external field pulse can lead to a dynamic excitation. In case of a coherent precession (same phase, same frequency) of individual spins confined in a region, we will have the macroscopic magnetization vector engaged in a damped precessional motion called uniform mode. This can be well described by the Landau-Lifshitz-Gilbert equation. In case of a multitude of spins precessing with the same frequency but different phase, we can talk about a propagating excitation in form of a wave - a spin wave.

Ferromagnetic resonance is, in simple terms, an effect based on exciting the uniform mode of the magnetization precession in a forced, resonant way, i.e. compensating for the Gilbert damping in each period of the precession to maintain a constant precessional amplitude. We apply a static, uniform external field $\vec{H}_{DC}$ to define the initial direction of the magnetization and then change the magnetization equilibrium state with a weak, high (usually radio) frequency alternating magnetic field perpendicular to the static field, of the form $\vec{H}_{RF}(t) = \vec{H} \cdot e^{i\omega t}$. The equilibrium direction of the magnetization will be determined by the vectorial sum of the two fields at any moment; the magnetization vector, obeying the Landau-Lifshitz governed dynamics [1], will start a precession perpendicular to the plane of the two fields. Obviously, in the case of a thin magnetic film, internal fields can arise due to anisotropies, and they will add up to the external applied field to form an effective local field. If we denote the spatial components of the static effective field as $H_y$ (in-plane component) and $H_z$ (orthogonal to the film plane), the Kittel equation 2.12 can be written in the form

$$\omega = \gamma \mu_0 \sqrt{H_y \cdot H_z}.$$  \hspace{1cm} (3.1)

This equation yields us the precession frequency $\omega$ as a function of the static field, for a known gyromagnetic ratio $\gamma$ and the vacuum’s magnetic permeability $\mu_0$. For an in-depth discussion of the theory of ferromagnetic resonance absorption, see the original work of Kittel [27].

The RF field, when using the same frequency as the precession has, constantly “pumps” energy into the resonant precession and takes care that the magnetization never reaches a static equilibrium state. For a certain precession frequency we see that there is a correlated external static field that leads to resonance. The absorption of energy at the resonance happens in an absorption line that is centered around $H_{DC}$; its finite line width is correlated to the relaxation [29, 30] in form of the Gilbert damping $\alpha$ via the formula

$$\Delta H = \frac{\alpha \omega}{\gamma \mu_0}.$$  \hspace{1cm} (3.2)

A device measuring the absorbed RF power by a magnetic sample in a microwave cavity is called a conventional FMR spectrometer (explained in details in [31]). The amplitude of the static applied field is tunable so that a resonant precession can be generated, which will show up as a Lorentzian line shape on the absorption spectrum (see Fig. 3.1). The power absorption is proportional to the imaginary part of the magnetic susceptibility of the sample. The position of the line on the absorption spectrum will indicate the resonance field amplitude $\mu_0 H_{DC}$, while its width $\mu_0 \Delta H$ can be used via formula 3.2 to calculate the intrinsic damping $\alpha$. 

3.1. MEASUREMENT TECHNIQUES

To increase sensitivity, the applied bias field is usually slightly modulated in amplitude, resulting in a spectrum that is the absorption derivative, instead of the absorption itself. Being able to calculate the precession frequency and the Gilbert damping means that the uniform precessional mode is fully determined. However, in the case of real magnetic samples magnetic inhomogeneities, defects and local fields will cause a decoherence in the spin precession over the sample volume, thus an extrinsic damping has to be considered besides the Gilbert one [33, 34, 35]. In an FMR spectrum, this will show up as an artificial broadening of the experimental FMR linewidth, distorting it from the Lorentzian shape:

\[
\Delta H^2_{\text{exp}} = \Delta H^2_{\text{intr}} + \Delta H^2_{\text{extr}}.
\]

(3.3)

3.1.2 Brillouin Light Scattering (BLS)

Similar to phonons, spin waves (or more generally, magnetic excitations, magnons) can also be involved in inelastic scattering of photons (called Brillouin Light Scattering). The interaction between an incident laser light and the magnetic excitations in a ferromagnetic material results in scattered light at a shifted frequency. A simple (classical) explanation of the process relies on the fact that a spinwave creates a phase-grating in the material, propagating with the phase velocity of the spinwave. The incident light is reflected (Bragg reflection [21]) from this grating and, according to the Doppler effect [21], it suffers a frequency shift by the spin-wave frequency. An in-depth treatment of the effect, as well as experimental BLS studies on particular cases such as arrays of microstructures are presented in [36] by Demokritov et al, chapter “Spinwaves in Laterally Confined Magnetic Structures”.

A BLS spectrometer (see Fig. 3.2) is built from a frequency-stabilised laser, a Fabry-Perot interferometer and a photodetector connected to a data acquisition/analyzing system. The light from the laser is focused to the sample by a lens and, after scattering, collected and sent to the interferometer (passing the etalons several times to detect the weak inelastic signals). The frequency of the interferometer is scannable; the light transmitted is then absorbed by the photodetector. Recording the voltage generated by the inelastic scattered light on the photodetector as a function of the selected frequency yields a BLS spectrum. Spatial filters are used in the BLS setup to filter out the background noise.

The main advantage of a BLS spectrometer against the traditional FMR method...
lies in the fact that the BLS setup uses focused light (typically 30 µm), allowing for spatially resolved measurements. This can be useful in case of microscopic patterned magnetic structures and detection of localized spin waves in thin ferromagnetic films. Additionally, Brillouin light scattering occurs at thermally excited spin waves as well. Thus, excitation of spin waves with high wave vectors is not required.

BLS spectrometers have proven to be very powerful tools for studying the characteristics of magnetostatic waves (MSW) and their propagation. The theory of MSW excitations in magnetic films was already investigated by Damon and Eschbach in the early sixties [37]; microwave signal processing devices (delay lines, filters, resonators, signal-to-noise enhancers etc.) were at that moment relying on theoretically deduced properties of such waves. However, the BLS technique is capable of measuring directly the dispersion and intensity of magnetic excitations in situ in actual device configurations [38].

Several results have lately been reported [39, 40] on the local measurement of so-called dipolar Damon-Eschbach modes as well as spatially localized spin waves in ferromagnetic thin film elements, relying on the BLS technique and its ability of measuring on a microscopic scale. Real time observation of spin wave propagation, spin wave tunnelling, reflection at a barrier etc. are topics of major interest.

The FMR and BLS techniques both work in the frequency domain. There are also possibilities to probe the magnetization dynamics directly in the time domain; in the following chapter, representatives of such techniques will be presented.
3.1. MEASUREMENT TECHNIQUES

3.1.3 Time-resolved photoelectron emission spectroscopy (TR-PES)

Since the discovery of photoemission of electrons from solids (1887), the effect was investigated extensively. Its first useful application was probing the electronic structure of solids (Berglung, Spicer, Eastman and others). Although electron spin polarization (ESP) analysis was proposed already in 1930 by Fues and Hellmann, the experiments to detect ESP of electrons emitted from ferromagnets (Fe, Ni, Co) failed for decades. The first successful ESP measurements based on photoelectron emission were obtained only in 1969 by Busch et al. [41], two years after gadolinium was proposed as ferromagnetic sample [42]. A well written description of the experimental setup used in the 1970s, including the ESP analyzer (a Mott scatterer) can be found in [43].

Following these first results, using the light of a mercury-xenon arc lamp focused on the photocathode to a spot of 1-2 millimeters, the technique went through a fast development. Today, several research groups around the world use the PES with soft x-rays (from synchrotron facilities, e.g.) to achieve high magnetic contrast imaging of microstructures (X-ray photoelectron emission microscopy, XPEEM). Besides the increased (sub-micrometer) lateral resolution, this method allows for chemical selectivity and high surface sensitivity.

Concerning the dynamic magnetization studies, in the early 1990s Vaterlaus et al. successfully measured spin-lattice relaxation (demagnetization) in gadolinium [7, 8]. The orientation of the magnetization on a microscopic level is also studied with this technique. In 2003 and 2004 the first successful time-resolved XPEEM experiments were presented [44, 45, 46]. Their temporal resolution reaches the nanosecond range; moreover, Schneider et al., in stroboscopic XPEEM studies on Permalloy microstructures at the European Synchrotron Radiation Facility (ESRF) Grenoble, reports a resolution of 130 ps [47]. This level of accuracy allowed the researchers to observe sub-nanosecond incoherent magnetization rotation processes, leading to transient stray-field formation at the edges of the ferromagnetic microstructures. These results are in good agreement with our studies, presented in Chapter 4.

3.1.4 Time-resolved Magneto-Optic Kerr Effect (TRMOKE)

A time-domain magnetization probe with an even higher spatial and/or temporal resolution than the BLS is a probe relying on stroboscopic measurement of the magneto-optic Kerr effect. The MOKE is the linear response of a magnetic surface illuminated by a light beam with a well-defined polarization. In short, linearly polarized light reflected off a magnetic medium will suffer a rotation of the polarization axis and will have a nonzero ellipticity in its polarization state as well. These are called Kerr rotation and Kerr ellipticity, and can be considered of being directly proportional to the magnetization itself. Thus, measuring their change on external influence (magnetic field, temperature, etc.) can be correlated to the change of the magnetization itself. Using ultrafast laser pulses (100fs or less) as perturbation and/or probe, focused on spots below 10 µm, yields a very convenient time-domain probe of the local magnetization’s dynamics.

The TRMOKE technique forms the base of the experimental studies of this thesis,
therefore it will be presented in full detail in section 3.2 of this chapter.

3.1.5 Magnetization induced second harmonic generation (MSHG)

Time-resolved magnetization induced second harmonic generation is a technique that stands close to the one we used for the experiments presented in this thesis, TRMOKE (see next section). However, there are several important differences in the concept and in the results obtained with it.

Second harmonic generation refers to the nonlinear response of a magnetic medium illuminated by a (laser) light beam with a very high intensity. This response manifests itself as a relatively large rotation of the polarization plane of the generated second harmonic light [48], due to changes in the longitudinal and/or polar components of the sample's magnetization vector at its surface. The variation of the transverse component of $\mathbf{M}$ will result, on the other hand, in intensity changes of the generated second harmonic. The signal is very weak in intensity; special filters and a sensitive detection method (such as a photomultiplier tube) are needed for its detection. Although nonlinear magneto-optics is considered a new technique, it was born from the “marriage” of magnetics research and nonlinear optics developed in the early 1960s (the “golden age” of magnetics research and Bloembergen’s development of magneto-optics) [49].

The power of the MSHG technique lies in four main aspects, namely:

- the incident laser light is focused to spots of diameter 10 µm or less, therefore it can be used as local probe of the magnetization;
- using pulsed laser beams in a stroboscopic way, it can be combined with pump-probe type experimental setups, to achieve time-resolved magnetization dynamics measurements [48, 50];
- polar, longitudinal and transversal magnetization components all can be measured to a certain extent;
- the sensitivity can be restricted to the sample surface or any buried magnetic interface [51].

The first two aspects are shared with the linear TRMOKE technique, as mentioned in the previous section.

Let us investigate the last characteristic of the MSHG technique. Using a high intensity incident light beam, that is, an incident electric field $\mathbf{E}(\omega)$ with a high amplitude, the linear dipole approximation $\mathbf{P} = \chi^{(1)} \cdot \mathbf{E}$ does not hold anymore. Thus the induced polarization $\mathbf{P}$ will have a term that represents the second harmonic, that is, $\mathbf{P}(2\omega) = \chi^{(2)} \cdot \mathbf{E}(\omega) \cdot \mathbf{E}(\omega)$. Here $\chi^{(i)}$ represents the susceptibility tensor of order $i$ and rank $i+1$. Similarly, we can add terms representing generation of higher harmonic signals to further extend the approximation.

However, in a centrosymmetric lattice (such as bulk metallic lattice can be) the potential wells for the electrons are also centrosymmetric. Since the induced polarization results from the motion of electrons with respect to the atomic lattice, the form of the harmonic terms defined above indicates that only odd harmonics (first, third etc.) can be present in the reflected bulk signal [52]. A detailed explanation is
beyond the scope of this brief review of the technique; however, we note that the same argumentation is valid for the magnetic component too. In other words, studying a sample with two different layers that are both centrosymmetric (including the case of metal-air), the second harmonic signal will be resulting \textit{from the interface region only}, where the inversion symmetry is broken. This gives a powerful tool for studying magnetic effects at interfaces, without a disturbing background signal generated in the bulk of the sample.

3.2 Our approach: TRMOKE

The experiments presented in this thesis rely on measuring the so-called time-resolved magneto-optic Kerr effect (TRMOKE). Our choice for this technique was motivated by the need to measure vectorially the magnetization dynamics of patterned microstructures on a nanosecond (precessional dynamics) and picosecond (demagnetization phenomena) timescale. Building an experimental setup around this idea proved to be a great learning experience, therefore we devote the rest of this chapter to the full description of the technique and equipment.

3.2.1 Pumping schemes for the spin dynamics

As introduced in the first chapter, our experimental setup is based on the pump-probe technique (called also stroboscopic in literature). In general, the pump-probe setup for magnetization measurements consists of a repetitive perturbation source (called pump), a magnetic sample (usually in form of a thin layer) and a probing method sensitive to small changes in the magnetization of the sample, repetitive as well. The pump and the probe are frequency synchronized and preferably at a high repetition rate (MHz regime) for a sufficient magnetic sensitivity; however, one has to make sure that the magnetization has enough time (typically ten nanoseconds or less) to relax to equilibrium during each pump-probe cycle.

The “time-resolved” feature of the setup is obtained through a variable time delay between the pump and the probe. Further in this thesis, “zero delay” corresponds to the case when they arrive to the sample at the same time, “positive delay” stands for the probe arriving after the pump and, similarly, “negative delay” refers to probing the magnetization before the moment of perturbation.

In our experiments, we implemented two different solutions in order to pump the sample, that is, to launch the magnetization dynamics. First we will present the magnetic field pump scheme (see the schematic drawing in Fig. 3.3), followed by the description of the all-optical pump-probe scheme (shown in Fig. 3.5).

Pumping with magnetic field

This scheme uses short magnetic field pulses generated locally, at the sample position, to perturb the spin system in a (more or less) uniform way through the whole microscopic sample. Note that we used this type of perturbation only for lithographically patterned thin film samples of diameter 30 \(\mu\)m and below. The magnetic field pulses we use have a typical rise/fall time of 200 to 300 ps, a length of 600 to 1500 ps and an amplitude of no more than 1.2 kA/m. The pulses have a repetition rate of 82 MHz,
modulated into pulse trains with a 10 kHz modulation frequency. They are generated by simple induction around a microscopic stripline deposited in the vicinity of the magnetic sample to study; the stripline connected to a commercial electric pulsegenerator (model AVTECH AVN-1-CP), that can generate electric pulses up to 5 V over a 50 Ω load. More about the generation of the electric and magnetic field pulses will follow soon in the section 3.3.

The probing is done by the magneto-optic Kerr effect (see description later in this chapter) using laser pulses of approximately 100 fs duration, generated by a Tsunami (Spectra Physics) mode-locked Ti:Sapphire laser. The laser pulses have a repetition rate of 82 MHz and are frequency synchronized with the pump (field) pulses. The wavelength we use is around 750 nm; the energy compressed in one pulse is approximately 10 pJ (yielding an average laser power of 1 mW). We have to use such low laser power in order to make sure that the probe pulse does not influence the evolution of the magnetization dynamics. The focusing of the laser pulses is done with a high aperture lens (either a high aperture laser objective of NA = 0.38 or a microscope objective of NA = 0.65) in order to obtain good lateral resolution. The smallest spotsize we achieved was of 1 µm diameter, allowing us to probe local magnetization dynamics in various regions within structures of typically 10 µm lateral dimensions. The reflected laser light travels backwards through the same focusing lens, is collimated and deflected towards a photodetector. We use a crossed polarizer-analyzer configuration to measure the polarization difference between the incident and the reflected beams. A schematic drawing of our field-induced MOKE setup is shown on Fig. 3.3. To detect the magnetization components in multiple directions,
we developed a special detection scheme, presented in detail in section 3.5.1.

The frequency synchronization between the pump and probe pulses is achieved by connecting the “monitor” electronic output (a somewhat distorted 82 MHz sine wave, of amplitude +/- 500 mV) of the Tsunami to the trigger input of the AVTECH pulsegenerator. This way, for every laser probe pulse a current pulse will be sent through the stripline, which will induce a magnetic field pulse through the sample.

A modulation is introduced in the setup, and a lock-in amplifier that is able to measure the photodetector signal with the frequency of this modulation, in order to increase our signal-to-noise ratio. The modulation device included in the pump scheme is an electronic switch (see schematics and details on Fig. 3.4), based on a PIN diode with a breakthrough voltage of ∼+7V. The diode was switched on and off with a 10 kHz sine wave, produced by a function generator.

![Figure 3.4: Schematic drawing of the electronic switch. The 82 MHz trigger signal is modulated with an external 10 kHz sine wave, that switches the PIN diode on and off. To block any crosstalk between the high- and the low-frequency signals, a pair of capacitors (C = 6.8 nF) and a pair of inductances (L = 1 mH) was included in the circuit.](image)

Sending the fast current pulses through such a device would have had a detrimental effect on the electric pulse quality (the high frequency components would have been blocked, resulting in low amplitude and very broad pulses), therefore we inserted the electronic switch between the Tsunami monitor output and the AVTECH trigger input. The result was (checked on an oscilloscope screen) a sine wave trigger signal of 82 MHz “chopped” into packages of 10 kHz (or whatever the modulation frequency was set to). The pulsegenerator emitted therefore 10 kHz pulse trains of undisturbed shape, with the Tsunami’s repetition rate of 82 MHz. The TTL output of the modulation source was connected to the trigger in of the lock-in amplifier to amplify selectively the 10 kHz (thus, pump-related) component of the photodetector signal.

A controllable time delay between the pump and the probe pulses was introduced by changing the optical path length of the probe laser pulses in comparison with the magnetic field pump pulses. After exiting the laser, the light pulses were reflected from the surface of a beam splitter into a retroreflector that was mounted on a motor-driven translation stage. The beam splitter is used to reduce the power of the laser beam by a factor of ∼25 (the intense transmitted beam was blocked). The retroreflector is a simple glass tetrahedra with its 3 facets perpendicular to each other (called also a “corner cube” being similar to a cut-off corner of a glass cube). Any beam falling near-perpendicular to the retroreflector’s base plane will be reflected parallel to the direction of incidence; the separation is dictated by the distance between the point of incidence and the center of the base. This is a very convenient solution for our need - we obtain a reflected beam that does not change its direction if we move
the retroreflector parallel with the incident beam, it can be focused to a small sample without having to worry about misalignment caused by shortening/lengthening of the optical path. And, at the same time, we have a spatial separation between the incident and reflected beams. The translation stage used to control the position of the retroreflector, thus the time delay between pump and probe, was a computer-controllable M-531.PD model manufactured by Physik Instrumente (PI) with a range of 306 mm and a resolution of 0.5 \( \mu \)m. This means a variable optical path of 0 to 612 mm for the probe beam, corresponding to a time delay of maximum 2 nanoseconds. For a longer time delay, we double-folded the beam along the translation stage (by sending the exit beam back to a second retroreflector on the stage) thus the total controllable delay time went up to 4 nanoseconds. In the same time, the resolution decreased four times as well, yielding 2 micrometers (equivalent to 6.6 femtoseconds) that is still more than enough for our experiments. The power of using an optical delay line as described above lies, in fact, within the very precise and practically jitter-free control of such a device.

The 82 MHz repetition rate of the pump-probe pulses yields a 12 ns period. Since this is out of reach for the optical delay line, a coarse adjustment of the pump-probe delay time was done by choosing the correct length for the coax cable transmitting the trigger signal for the electric pulsegenerator. The “zero delay” was placed this way close to the short end of the optical delay line, allowing for a few hundred pico-second “negative delay time” and a \(~4\) nanosecond “positive delay time” for our measurements. In some cases, two 4 ns timesweps with shifted zero delay positions were combined, “stitched” one after the other, to obtain a measurement of the magnetization dynamics over a 8 ns time domain.

**Optical pumping**

The second pump scheme sketched on Fig. 3.5 is using, instead of magnetic field pulses, an intense ultrafast laser pulse (1 nJ/100 fs pulse) to perturb the spin system on the surface of a ferromagnetic layer, by locally increasing the temperature \( T \). Depending on our goals, it can be applied in order to change the saturation magnetization \( M_s \) (partial demagnetization, \( M_s = f(T) \)) or start a precessional motion by changing the anisotropy constant \( K \ (K = f(T)) \) of the thin layer.

The pump pulses originate from the same Tsunami mode-locked laser as the probe pulses; we simply use a beam splitter (BS) to split each laser pulse into a strong (pump) and a weak (probe) pulse with a power ratio of 25:1. The pump pulse properties are thus, aside their energy, the same as the probe pulses previously described. After passing through the BS, the pump beam is following a different optical path than the probe, with fixed length, arriving in the end to the same lens which focuses the probe beam too. In this setup (named “all-optical”) we use a high aperture laser objective of NA = 0.38 with a physical entrance pupil of approximately 20 mm. The wide lens allows for physical separation of the two ingoing and two reflected beams: “dividing” the lens into four quarters, we use two (top left, bottom left) to focus the parallel laser beams on one spot on the sample (diameter of 8 to 10 \( \mu \)m) and two to collect the reflected beams (see Fig. 3.6). The beams having a diameter below 2 mm, it is relatively easy to align the setup in such a way that the focused spots are overlapping, while the reflected part of the pump beam, after exiting the lens, can
be blocked not to disturb our measurement. The reflected probe beam (and only the probe beam) is then directed towards the photodiode.

The path of the probe beam, including the delay line, is almost the same as in the previous, magnetic field pump configuration: beam splitter, retroreflector(s), polarizer, focusing lens, sample, focusing lens, analyzer and photodetector.

The control of the delay time between the pump and probe is in this case simpler. The pump and probe are split from the same original laser pulse, thus the repetition frequencies are synchronized from the start. By measuring and adjusting the optical path of the two beams, we can define the coarse position of the “zero delay” and position it close to the short end of the motorized delay line. In case of a good pump-probe spatial overlap on the sample, the reflectivity (due to the pump pulse’s effect on the electron temperature) will change on a 100 fs timescale, which, by slowly scanning the delay line, will show us the exact position of the temporal overlap of the two pulses, thus the “zero delay” (see Fig. 3.7). In some experiments,
a different model of translation stages was used - one with a short range of 100 mm (≈660 ps) only, but with increased resolution of 0.025 μm, corresponding to ≈0.1 fs. This increased resolution was helpful in the case of ultrafast demagnetization measurements, where the interesting effects happen on a timescale of a few hundred femtoseconds.

**The modulation technique**, obviously, could not have been relying on an electronic switch, since in this setup we did not use the electric pulsegenerator. We used instead a technique/device well-known from static Kerr effect measurements (MOKE): a photo-elastic modulator (PEM). This device consists of a compressible crystal and an electronic unit that can apply a 50 kHz signal to compress the crystal in one direction. A linearly polarized light beam sent through the crystal, perpendicular to the axis of compression, will become alternatingly a left- and right-handed circularly polarized beam. The effect of the PEM on the MOKE setup will be explained in the end of this chapter (section "Detection of magnetization based on Magneto-Optic Kerr Effect"); for now, we mention only the fact, that the oscillating polarization of the probe beam combined with the polarizer-analyzer will result in a signal on the photodetector that has a 50 kHz component proportional to the Kerr ellipticity and a 100 kHz one proportional to the Kerr rotation, induced by the magnetic sample. The lock-in amplifier is triggered by the TTL output of the PEM to amplify only this high frequency component.

In order to exclude any effect on the sample other than the pump pulse, we use a **double-modulation technique**: besides the probe modulation described above, the pump beam is also modulated. This is realized by a chopper - a rotating blade with holes, periodically blocking the pump beam (thus, creating optical pulse trains) with a frequency of 60 Hz. The signal outputted from the 50 kHz-locked amplifier serves as an input for a second lock-in amplifier, triggered by the 60 Hz TTL signal of the chopper. This way, the final measurement signal will be an amplified result of the 50 (100) kHz and 60 Hz component of the original voltage on the photodetector, carrying information about the change of polarization state of the probe beam due to the pump beam on the magnetic sample at a certain moment. The introduction of the second modulation into the setup significantly increased the signal to noise ratio. Since most ferromagnetic materials (except some garnet films) produce Kerr rotation angles of millidegrees, instabilities of some optical components and small
misalignment of the delay line would lead to considerable signal drift in time; the second modulation takes care to cut down this type of systematic noise too.

### 3.2.2 Bias field

A ferromagnetic thin layer naturally will have smaller or bigger magnetic domains separated by domain walls, the magnetization directions being different in these domains. Since the sizes of these domains are usually below the resolution of a time-resolved MOKE setup (our smallest probing spot size was approximately 1 µm in diameter), the measurement will represent an average of the magnetization dynamics over several grains. Due to the random static orientation of the magnetization vector in these domains, the precessional dynamics will be dephased, thus no precession will be seen over the entire measurement area. In addition to the grain structure, in patterned microscopic samples other effects might locally influence the phase of the precession, such as missing dipoles at the edge, stray fields from adjacent magnetic layers etcetera (some of these presented in detail in the chapter "Measurements of the field-induced precession"). Therefore, one has to align with a static magnetic field the magnetization of at least the whole region below the probing laser spot, prior to the fast perturbation. At the same time, a static field will also define the direction around which the precession will happen.

In most of our experiments we applied a static field externally. In the case of the field-induced measurements, the sample (Permalloy) needed a very small (typically 0.5 kA/m) in-plane external field to saturate its magnetization over its whole volume. At the same time, the low amplitude of the field pulses (below 2.1 kA/m) does not allow for a large external DC field. This field is applied perpendicular to the pulse field direction (both in the sample plane); the vectorial sum of them will define the new equilibrium direction of the magnetization, around which the precessional motion will start. If the DC field is much larger than the pulse field, the equilibrium direction due to the pulse field will not change considerably, resulting in a precessional motion of a very small angle that is hard to detect. As an example, the permalloy sample of the MTJ stack we studied had a coercivity field of 0.5 kA/m; with a DC field of 5 kA/m and a perpendicular pulse field of amplitude 2.1 kA/m, we detected an elliptical magnetization precession of approximately 24° (the largest angle deviation).

Since there was no need for a strong DC field, we used a pair of Helmholtz coils with an opening of 18 cm to generate a homogeneous field in its center up to 8.2 kA/m. The lack of a soft magnetic core had two advantages: the sample was much more accessible (electrical connections and optical access were both needed) and there was no remanent field from the cores, that is, zero electric current through the electromagnet results in zero external magnetic field. Additionally, an extra electromagnet with small poles mounted on a long arm and attached to the core was used for some measurements, together with the Helmholtz coils, to generate a DC field parallel or antiparallel with the pulse field.

The case of the all-optical setup was different. Here the external bias field had two roles: one to saturate the magnetization of the layer, removing magnetic domain walls from the region of interest and another one, to cant the magnetization of the layer out of the sample plane. Since these samples (polycrystalline Ni, permalloy
CHAPTER 3. EXPERIMENTAL

and CoFe thin layers) all had an in-plane anisotropy, the magnetization naturally was lying in the sample plane. Neither polar MOKE measurements for ultrafast demagnetization, nor precessional dynamics by laser pumping can be started with a fully in-plane magnetization. However, the demagnetization field for thin layers is of the order of a tesla (e.g. 400 kA/m for 10 nm Ni and 1400 kA/m for 10 nm Co). Therefore, to cant the magnetization just slightly out of plane, a large external field was needed. Obviously, our Helmholtz coil cannot produce such a field; we used therefore an electromagnet with soft magnetic cores positioned very close to each other, yielding a homogeneous field between the poles of 240 kA/m or less (see also Fig. 3.5). One of the poles had a hole parallel to the field lines, large enough to fit the lens in it, which was used to focus the pump and probe beams on the sample. The field was oriented in an angle of approximately $11^\circ$ (for some measurements, even $35^\circ$) versus the normal of the film plane. This configuration, with a field amplitude set to 200 kA/m, yielded an in-plane DC field $H_{\text{DC}}^x \simeq 40$ kA/m and a perpendicular component $H_{\text{DC}}^z \simeq 200$ kA/m, resulting in an equilibrium magnetization angle large enough to have a measurable dynamics of the polar component $M_z$.

The external DC field in the all-optical measurement setup provided a great tool to separate magnetization dynamics from the dynamics of electron reflectivity due to the ultrafast heating - just by measuring the dynamics twice, with opposite DC field directions and identical amplitude. More about this method will follow in section 3.4.

The alignment of the magnetization vectors of the different domains does not necessarily require applying an external bias field. It can be done by multiple means, such as building internal fields inside the sample by shape anisotropy or growth in magnetic field; depositing extra magnetic layers on or below the one in study to use the different couplings that can arise (Neel coupling; stray field in case of patterned microstructures; exchange bias field in case of using antiferromagnetic layers) and others. Most practical (micro)magnetic devices (as the MTJ stack we studied with the field-induced measurement setup) rely in fact on these type of fine tuned local fields, making external bias fields obsolete.

3.3 Generation of sub-nanosecond field pulses – striplines and waveguides

As mentioned in the description of the traditional FMR technique, there are options of applying a large scale (i.e. over the whole sample) “pumping” with a high frequency magnetic field. Another possibility is to apply rotating magnetic fields [53], used mostly for studying domain wall displacement, a somewhat slower magnetization dynamics.

However, for our time-resolved pump-probe type measurements local and fast magnetic field pulses were needed. At this moment, the most straightforward way to produce magnetic field pulses is, to use induction by a fast electric current pulse travelling through a microscopic “wire” in the vicinity of the sample. In the case of DC and low frequency currents, this is relatively easy to do; however, when field pulses of nanosecond duration or faster are needed, several details have to be taken care of. The generation of the electric current pulses that will travel through the conductor can be problematic as well. We will discuss consecutively these two technical problems,
beginning with the induction of field pulses around a narrow conducting wire.

In a microscopic sample configuration, it is convenient to deposit the current pulse wire as a narrow and thin metallic stripline before the deposition of the sample. On a well chosen substrate (GaAs or other non-metallic substrates with high thermal conductivity would be the first choice) the striplines can be patterned lithographically and grown using conventional lift-off techniques. After the stripline was made, there is the option of growing the ferromagnetic sample directly on the same substrate, e.g. on the top of the stripline with a thin electric insulating layer in between, or a mechanical positioning of a sample grown on a different substrate. We manufactured the sample following the first option, having the advantage of good alignment of the microscopic structures (stripline and sample), ease of optical accessibility and having in mind “real” samples for industrial applications such as MTJ stacks, etc. The second option is, however, more flexible; the stripline can be used for generation of field pulses through a set of samples by replacing only the ferromagnetic sample on the top. Although the need for precise alignment complicates measurements on microstructured samples, measurements on uniform thin layers represent no problem [54].

For the case of thin film samples, where striplines of sub-micron thickness and a width of several micrometers are used, the magnetic field above the central region of the stripline can be considered spatially homogeneous and isotropic. If the ferromagnetic sample is positioned here, it will “feel” a field that is in-plane (perpendicular to the direction of the current pulse through the stripline) and having an amplitude defined by the formula

\[ H_p \approx \frac{V}{2Zw}. \]  

(3.4)

Here \( V \) stands for the amplitude of the voltage pulse applied on the stripline, \( Z \) is the impedance and \( w \) is the physical width of it. This formula is derived from Ohm’s and Biot-Savart’s law for an infinitely thin current conductor (thickness \( \ll \) width), and it is valid for any height above the stripline as long as the height is much smaller than the width. Our samples have striplines with width of 30 to 50 \( \mu \)m and thicknesses of 300 nm and below. The separation between the ferromagnetic thin layer and the stripline is also very small (the largest being 200 nm, see sample description for details in the beginning of Chapter 4). The above formula is therefore a good way to estimate the field pulse amplitudes we are able to produce with our striplines.

The generation of the *current pulses* of such a short duration and high amplitude needs some attention as well. There are several methods used in research, such as optical switches built into the stripline close to the sample position, triggered by laser pulses [50]; pumping Schottky diodes with ultrafast laser pulses [57, 58] and external pulsegenerator devices [80, 73, 74]. Commercial pulsegenerators in the GHz range are based on a special electronic component called "step recovery diode" (SRD). This diode, if a sufficiently high amplitude fast sinusoidal voltage is applied on it, has an abnormal behaviour for the negative region of the sine: it conducts until the very fast, step-like recombination of all the charge carriers that were generated during the positive half period of the sine. The result is an intense voltage pulse generated with the repetition rate of the sinusoidal’s frequency.

In our experiments, we used an electronic pulsegenerator with a maximum amplitude of 5V, a repetition rate of 50 to 250 MHz, a pulse duration of approximately 300 ps to 2 ns and a shortest rise/fall time of 120 ps each. Three major aspects had to be
considered prior of choosing the model. One is, obviously, the time duration and the steepness of the rising and falling edges of the pulses. For a high temporal resolution, one needs field pulses rising/falling as fast as possible (without any afterpulsing), to let the magnetization evolve in an undisturbed way. The 120 ps rise/fall times proved to be satisfactory. The second factor is the amplitude. We needed a voltage pulse of at least several volts/50Ω to get field pulses that can start a magnetization precession of a measurable amplitude (see formula 3.4). The third factor was dictated by the stroboscopic aspect of our experiments. Since we use a laser with a repetition rate of 82 MHz, we needed a pulsegenerator that can work at this speed. Reducing the repetition rate by means of a pulse picker for the Ti:sapphire laser would have been another possibility, however, that would have meant reducing the average power of the probe beam with the same factor. Since we rely on the collective effect of millions of pulses to measure one single data point, this would have resulted in a decrease of the signal-to-noise ratio of the experiment. Commercial pulsegenerators are usually a compromise between these three parameters: they can produce very fast pulses (down to 30ps) with a high repetition rate of several GHz, but the amplitude will be far below 1V, or they can produce voltage pulses up to 50V in the nanosecond time domain, but with a repetition rate of 100 kHz or less.

The parameters of our pulsegenerator mentioned above are valid for an ideal voltage pulse applied on a 50 Ω impedance load using perfect connections. As an example, on Fig. 3.8 we included screenshots of a 12.5 GHz sampling oscilloscope showing a voltage pulse as close to the ideal as possible. Also, the formula 3.4 stands for DC current and pulses travelling without any loss through the stripline. In the case of sub-nanosecond current pulses, this is impossible to realize in practice.

To optimize the transmission of the pulse between the generator and the stripline, a waveguide design is necessary. Coplanar waveguides, as illustrated on Fig. 3.9, are (micro- or macroscopic) structures with a well defined impedance for the desired
3.3. GENERATION OF FIELD PULSES

Figure 3.9: Schematic drawing of a ground-signal-ground (GSG) type coplanar waveguide deposited on a dielectric (d) and having a gap width w. The field rising in-plane inside a microstructure on top of the signal line can be calculated with the formula 3.4.

frequency region, allowing the electric field wave to travel through the structure without losses and enter/exit without reflections. They usually consist of a thin central stripline with a certain width and ground plates on both sides, separated by a gap of air, the whole structure being deposited on a dielectric. In order to obtain the needed impedance of the waveguide and as small losses through the substrate as possible, a strict relationship between the width of the central strip, the gap between the ground planes and the dielectric constant of the substrate has to be respected. Some extra restrictions were present in our case: we needed optical accessibility from the top, good heat conductance of the substrate and lateral dimensions that allowed a stable electric contact on both (entrance and exit) sides.

Leaving the details for a section dedicated to our samples, we mention here that two types of striplines were used in our experiments. One type was a true Au coplanar waveguide of parameters central strip width $s = 50 \, \mu m$, strip to ground plane gap width $w = 100 \, \mu m$ and GaAs substrate with a dielectric constant $\epsilon = 13.5$. The impedance of this waveguide appeared to be optimal for the coupling with the output of the pulsegenerator; however, a 20 mm long waveguide structure was needed for the optical accessibility between the contact pins (see also Fig. 4.3). This increased the total DC resistance to 120$\Omega$. The resulting magnetic field pulses, presented in Chapter 5, show a total loss of 20% on the pulse amplitude, compared to the amplitude of 0.4 kA/m calculated with formula 3.4.

In the case of the magnetic tunnel junction (MTJ) type samples we obtained from Philips Research, the stripline was a simple Cu wire of width $30 \, \mu m$ and thickness $\simeq 17$ nm without ground planes, with two large ($200 \, \mu m$ wide) contact pads on both ends, deposited on a Si/SiO$_2$ substrate. The DC resistivity of the structure was adjusted by its dimensions to exactly 50 $\Omega$, while the high-frequency impedance was not optimized at all, yielding a mismatch with the 50 $\Omega$ output impedance of the pulsegenerator. The resulting field pulse (due to the thinner strip and lower DC resistance) was much higher than in the case of the previous sample, but the losses/reflections due to the impedance mismatch increased, and the pulse profile is also more distorted (see again for concrete results Chapter 5).

These results suggest that major improvements in the amplitude and quality of the magnetic field pulse could be obtained with impedance matched waveguides based on a central strip as narrow as possible, and special care should be taken that the DC resistance is kept low as well. Since a good electric coupling needs contacting pads of $\simeq 30 - 50 \, \mu m$ width, for further high-frequency experiments a tapered waveguide
structure should be implemented [55, 56].

3.4 All-optical pump-probe technique

As mentioned in the previous sections, the all-optical technique to measure magnetization dynamics relies on using an intense heat (laser) pulse that suddenly changes the local temperature of the ferromagnetic layer. With certain well-defined equilibrium conditions, the sudden temperature change will launch magnetization dynamics on the sub-nanosecond timescale that we can experimentally detect with a second, weaker and time-delayed probe pulse. But what exactly happens that causes the magnetization to start a precessional motion?

3.4.1 The “anisotropy field pulse”

It is well known that the amplitude of the magnetization vector in case of ferromagnetic metals strongly depends on the temperature. An ultrafast, intense laser pulse (<100 fs, 1 nJ/pulse) focused on a spot of diameter of 10 µm or less, can locally increase the temperature with tens of degrees. In Chapter 2, we briefly presented the ultrafast demagnetization phenomena based on laser heating; experimental studies are well described by van Kampen et al. [4, 16]. However, in some experiments it was noticed that, several picoseconds after the equilibrium magnetization amplitude is restored, i.e. the heat is diffused out from the studied sample region, the measured magneto-optic signal continued to increase. On a longer (hundreds of picoseconds) timescale, this manifested itself as an oscillation of the MO signal centered on the zero value (the value before the laser pulse arrived). For a typical experiment showing such an oscillation of the z (out-of-plane) component of the magnetization, see Fig. 3.10. The oscillating effect is not related to the amplitude of the magnetization anymore, but to its orientation that follows a damped precessional motion (LLG-type behaviour) around the equilibrium direction at any moment in time, until it arrives at the equilibrium again. So, apparently without applying any external fast magnetic field pulse, we started precessional magnetization dynamics.

The key to this dynamics lies in the external bias field, canting the magnetization out of its preferred equilibrium direction defined by the anisotropies in the sample. As an example, a thin ferromagnetic film with in-plane anisotropy will have no out-of-plane magnetization component, unless we apply an out-of-plane DC magnetic field of considerable amplitude. The equilibrium direction for the magnetization will then be governed by the balance between the external bias field orthogonal to the sample surface and the counteracting anisotropy field:

\[ \vec{H}_{DC} = \vec{H}_{appl.} + \vec{H}_{ani}(T), \]

where the anisotropy field is built up from different type of anisotropies (shape, magneto-crystalline, magneto-elastic) and it is temperature-dependent. Keeping the bias field constant and heating up the sample changes the anisotropy of the sample leading to a new equilibrium direction for \( \vec{M} \). If we use an ultrafast laser heating, the magnetization will start a very fast precession around the new (time-dependent) equilibrium direction due to the torque rising from the angular difference between the
3.4. ALL-OPTICAL PUMP-PROBE TECHNIQUE

Figure 3.10: Typical all-optical measurement on a sample consisting of Al(2nm)/Py(15nm)/Ta(10nm)/SiO$_2$/Si. Negative time delay means probing before the perturbation ($\Delta M_z(t) = 0$). After the ultrafast demagnetization (200 - 300 fs), the magnetization amplitude almost completely recovers ("remagnetization") during the first picoseconds. The precessional dynamics are observed as an oscillating magnetization component $\Delta M_z$ on the sub-nanosecond timescale.

magnetization and effective field directions. The precession will start, however, on an elliptical trajectory tangential to the sample plane, thus, in the first picosecond it will not result in a measurable change of the out-of-plane magnetization component $\Delta M_z$. As a next process, the heat will diffuse from the small spot hit by the laser pulse into the rest of the sample, thus the equilibrium direction will drift back to its original orientation, simultaneously with the remagnetization process. However, the magnetization vector forms now an angle with the equilibrium direction, therefore it will continue its precessional motion on a timescale of typically hundreds of picoseconds, governed by the Gilbert damping parameter $\alpha$. This will show up in a $\Delta M_z(t)$ time-resolved measurement as an oscillation of the signal. The precessional motion in three dimensions is illustrated in Fig. 5.5; its projection on the $z$-axis is shown, in form of a real $\Delta M_z(t)$-measurement, in Fig. 3.10.

Observing the similarity between the precession started by the sudden change of anisotropy due to fs laser heating and the precession started by an external magnetic field pulse, we can introduce the concept of "anisotropy field pulse" as a new way of perturbing the spin system. The shape, amplitude and properties of this anisotropy field pulse will be studied in depth in section 5.3.

3.4.2 Optically induced magnetization precession - local and fast

After introducing a new measuring technique in general, the question inevitably pops up: What are its advantages and drawbacks versus the conventional methods?

Spatial resolution is a considerable advantage against any bulk-type magnetization probing technique, such as SQUID, VSM and FMR. By focusing the pump and probe laser beams to small spots (in principle, down to the sub-micrometer diffraction limit), we can measure the magnetization dynamics locally. This allows us to study
the local effects like spin wave generation and propagation, change of anisotropies, magnetic domain formation and others. Besides this direct advantage, there are some special uses of the high spatial resolution provided that we mount the sample on a controllable translation stage. One of these would be the possibility to scan the surface of a thin film ferromagnetic microstructure and monitor the magnetization dynamics of the whole sample. In the case of magnetic structures of very reduced dimensions, the dynamics and the switching mechanism can be greatly affected by the local effective fields, the shape of the structure, the edges, the neighboring layers etcetera. To understand and make use of these effects, measuring the magnetization dynamics with a high spacial resolution is of ultimate importance. Another “trick” making use of the spatial resolution is measuring on wedge-shaped samples, similar to the studies presented in chapter 6 of this thesis. By growing a thin film sample in a controlled wedge shape and measuring on different positions, we can obtain information about the magnetization dynamics as a function of sample thickness on one single sample. One can see that this allows for consistent thickness-dependent measurements where determining the layer’s thickness with a sub-nanometer precision is well within the reach of the experimentalist.

**Temporal resolution** is the main advantage of the all-optical time-resolved measurements. While similar or better spatial resolution can be reached with other local techniques (such as Kerr microscopy or MFM), the femtosecond temporal resolution of the laser pumping technique is, so far, unique. Additionally, splitting the original laser pulse into pump and probe pulses automatically removes any problems related to jitter of electronic devices, frequency/phase synchronization or shift of frequency in time. The high speed of light in air allows us to vary the temporal delay between the pump and probe pulses in steps of a femtosecond by relatively simple means of a motorized translation stage. Moreover, due to the same reason, the repeatability of the measurements is very high as well. In our experiments, laser pulses of 100 fs and shorter were used (the lower limit being 50 fs, achieved just at the exit of the resonant cavity of a Tsunami mode-locked Ti:sapphire laser). The generation of the time delay between pump and probe pulses, implemented by means of the motorized translation stages as described earlier, allowed resolutions down to the sub-femtosecond range.

**The power** of the heating pump pulse (typically 1 nJ per pulse, or 100 mW average laser power) has a very strong effect on the material’s anisotropy. The fact that we use the equilibrium between the strong anisotropy field (hundreds of kA/m) and an external magnetic field of similar amplitude, leads to very large torques on the magnetization in the moment of canting the equilibrium direction. This allows for a large inplane bias field without pinning the magnetization (a problem persisting in the case of field-induced measurements, where the external fast field pulse has an amplitude limited to a few kA/m). Therefore, the precession frequency and the whole precessional magnetization dynamics will be accelerated to a sub-nanosecond timescale, comparable to the traditional FMR-type experiments in the gigahertz frequency domain.

**The signal-to-noise ratio** of the all-optical pump-probe experiments is much improved by the double modulation technique. The fact that we measure the differential of the rotation / ellipticity signal increases the experiments’ sensitivity by two orders of magnitude when compared to static MOKE measurements. The measurement’s results are further improved by the elimination of systematic errors of
non-magnetic origin (small optical misalignments, wobbling of the translation stage and others) via substracting measurements at opposite magnetic fields, presented in the following paragraphs.

3.4.3 A useful side-effect: the dynamics of electron reflectivity

When an intense laser pulse is used to pump the magnetization of a thin layer, the sudden change of temperature leads to spin dynamics and to electron dynamics simultaneously. The electron distribution function in the vicinity of the Fermi level is modified by the femtosecond heating of the electron population. An observable consequence will be the change in the absorption spectrum of the sample. The electron dynamics will thus show up as a transient reflectivity change observed in the measurement together with the change of magnetization; however, it is independent of the external bias field. Studying transient electron dynamics via the time-resolved reflectivity or transmission is vastly treated in literature \[59, 60, 61, 62\] and therefore, its detailed treatment is not within the scope of this thesis. We mention it briefly as a side-effect of the time-resolved magnetization dynamics measurements that can be used to get a feeling of the electron temperature changes in the sample. To obtain the correct magnetization dynamics it has to be excluded from the all-optical measurements.

Knowing that, in first order approximation, the reflectivity should not depend on the external (or any existing) magnetic field, its separation from the magnetization dynamics proves to be rather easy. We perform a set of two consecutive measurements with changing only the sign of the applied bias field in between. This results in a spatially inverted magnetization \(M'_z = -M_z, M'_x = -M_x\) as on Fig. 5.5) while its amplitude and all the other parameters remain the same. The resulting data will have a symmetric contribution from the “reflectivity dynamics” but an antisymmetric one from the magnetization dynamics. Adding the two measurements will therefore yield a dataset representing the double of the reflectivity change versus time, while subtracting them will yield the pure magnetization dynamics.

Every all-optical magnetization measurement presented in this thesis is already processed with the above described simple method, thus it has no contribution from the electron dynamics.

3.5 Detection of magnetization based on the Magneto-Optic Kerr Effect

In all the experiments presented in this thesis, the detection of magnetization dynamics was based on the standard Magneto-Optic Kerr Effect technique. In this section, we will briefly explain how the subtle changes in the magnetization can be detected via the reflection of a polarized laser beam.

Linearly polarized light can be considered a combination of left- and right-handed circularly polarized light. In a cylindrical system of coordinates, this has the form of (for example) \(\hat{x} = 1/\sqrt{2}(\hat{e}_+ + \hat{e}_-)\), where \(\hat{e}_+\) and \(\hat{e}_-\) represent the left- and right-
handed circularly polarized electric fields respectively. These can be written as
\begin{align}
\hat{e}_- &= \frac{1}{\sqrt{2}}(\hat{x} - i\hat{y}), \\
\hat{e}_+ &= \frac{1}{\sqrt{2}}(\hat{x} + i\hat{y}), \\
\hat{e}_z &= \hat{z}.
\end{align}

In cubic ferromagnets (these being materials with broken symmetry only due to the magnetization), the dielectric tensor reads
\begin{equation}
\bar{\epsilon} = \begin{pmatrix}
\epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\
-\epsilon_{xy} & \epsilon_{xx} & \epsilon_{yz} \\
-\epsilon_{xz} & -\epsilon_{yz} & \epsilon_{xx}
\end{pmatrix},
\end{equation}
where the cubic symmetry and the Onsager relations yield the identities
\begin{align*}
\epsilon_{yx} &= -\epsilon_{xy}, \\
\epsilon_{zx} &= -\epsilon_{xz}, \\
\epsilon_{zy} &= -\epsilon_{yz}, \\
\epsilon_{yy} &= \epsilon_{xx}, \\
\epsilon_{zz} &= \epsilon_{xx}.
\end{align*}

If we consider the case when the magnetization is parallel to the z-axis, the dielectric tensor becomes
\begin{equation}
\bar{\epsilon} = \begin{pmatrix}
\epsilon_{xx} & 0 & 0 \\
0 & \epsilon_{xx} & 0 \\
0 & 0 & \epsilon_{xx}
\end{pmatrix}.
\end{equation}

In a first-order approximation, the off-diagonal elements \( \pm \epsilon_{xy} \) are the only ones dependent on the magnetization \( M \). At the same time, due to these tensor elements, an electric field along \( \hat{x} \) induces a finite polarization in the \( \hat{y} \)-direction. In cylindrical coordinates, assuming wave propagation along \( \hat{z} \), the dielectric tensor has the diagonalized form
\begin{equation}
\bar{\epsilon}_c = \begin{pmatrix}
\epsilon_{xx} - i\epsilon_{xy} & 0 & 0 \\
0 & \epsilon_{xx} + i\epsilon_{xy} & 0 \\
0 & 0 & \epsilon_{xx}
\end{pmatrix}.
\end{equation}

From the formulae 3.6 and 3.9 we can define nonequal dielectric constants for the left- and right-handed circularly polarized light, namely
\begin{equation}
\epsilon_{\pm} = \epsilon_{xx} \pm i\epsilon_{xy}.
\end{equation}

Left- and right-handed circularly polarized light beams have thus different reflection coefficients for a ferromagnetic material, the difference being dependent on the magnetization. As a net effect, linearly polarized light will experience a rotation in its polarization axis (Kerr rotation) and will gain an ellipticity (Kerr ellipticity) upon reflection from such a material. Measuring the polarization change of the light along the direction of the magnetization (\( z \)-direction in the above case) yields thus a complex rotation \( \psi \) proportional to the magnetization itself:
\begin{equation}
\psi = C \cdot M_z,
\end{equation}
with \( C \) as a complex constant. With the aid of the (complex) dielectric tensor elements \( \epsilon_{xx} \) and \( \epsilon_{xy} \) and using the Fresnel reflection coefficients, this can be written as
\begin{equation}
\psi = \psi_r + i\psi_e = \frac{\epsilon_{xy}}{\sqrt{\epsilon_{xx}(\epsilon_{xx} - 1)}},
\end{equation}

\( \psi_r \) being the real part and \( \psi_e \) the imaginary part of the polarization change.
where $\psi_r$ represents the rotation of the polarization axis and $\psi_e$ is the ellipticity of the reflected light.

For a completely arbitrary magnetization orientation and angle of incidence, it can be shown [63] that $\psi$ is a sum of the complex rotations of type 3.11 for the three directions $x, y$ and $z$, with different constants $C$ each.

Microscopically, the Kerr effect can be understood as follows. For an arbitrary sample, dipole selection rules are governing the MO Kerr effect. In the simplest case of spherical symmetry and no spin-orbit (SO) coupling, the rules only allow for photon-electron interactions that lead to $\Delta l = \pm 1$; the photons couple to the orbital momentum of the electrons. If SO coupling is present however, like in a transition metal ferromagnet, the selection rules apply for the quantum number $j$ instead of $l$. Due to the coupling, the total orbital momentum $\vec{L}$ is correlated to the spin momentum $\vec{S}$, allowing for measurements on the spin system through the photons’ selective interaction with the orbital momentum.

### Detection of Kerr rotation and ellipticity

In ferromagnetic transition metals, the orbital momentum $\vec{L}$ of the conduction-band electrons is reduced, yielding very small changes in the polarization of the reflected light (tens of millidegrees for Ni, as an example). To increase signal-to-noise ratio, traditionally a modulation of the light beam is implemented in the setup. A very efficient signal amplification can be obtained by using a birefringent crystal that can be compressed in one direction periodically (in the kHz range), just before the polarized light reaches the sample. This device, called photo-elastic modulator (PEM), induces thus a periodically changing phase difference between the polarization components along and perpendicular to the axis of compression. A lock-in amplifier is triggered on the same frequency as the PEM (50 kHz in our case); from the measured 50 kHz voltage and its higher harmonics, the Kerr ellipticity and/or rotation can be calculated.

In the Jones formalism [21], the PEM can be represented by the 2x2 matrix

$$M_{PEM} = \begin{pmatrix} 1 & 0 \\ 0 & e^{iA \cos(\omega t)} \end{pmatrix},$$

where $\omega$ is the modulation frequency and $A$ is the maximum retardation between the two polarization components. Knowing the Jones vector for a linearly polarized light (Exiting the first polarizer), the resulting polarization vector after passing through the PEM in a 45° angle versus the axis of compression becomes $\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ e^{iA \cos(\omega t)} \end{pmatrix}$. For a retardation $A$ set to $\pi/2$, the result represents light with a polarization state modulated between fully left- and right-handed circular polarization of form $\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm i \end{pmatrix}$.

Next, the light is incident on the magnetic sample, with the Jones-matrix

$$M_{sample} = \begin{pmatrix} r_s & r_{ps} \\ r_{sp} & r_p \end{pmatrix},$$

with $r_s$ and $r_p$ the complex reflection coefficients for s- and p-polarized light. The magnetization of the sample generates the off-diagonal components proportional to
\( \epsilon_{xy} \). After the reflection from the sample and the second polarizer ("analyzer"), the Jones formalism yields a voltage \( V \) on the detector containing the Kerr rotation and ellipticity:

\[
V \simeq \frac{\alpha E^2}{4} (|r_s|^2 + |r_p|^2) \{ 1 + 2\psi_e \sin[A \cos(\omega t)] + \sin 2(\psi_r + \phi) \cos[A \cos(\omega t)] \}, \tag{3.15}
\]

where \( E^2 \) is the light intensity, \( \alpha \) describes the sensitivity of the detector to light (working in the linear regime of a semiconductor photodiode), \( |r_s|^2 \) and \( |r_p|^2 \) are the sample's Jones reflection coefficients and \( \phi \) is the angle between the analyzer and the incident light’s polarization axis.

Observing that the prefactor \( \frac{\alpha E^2}{4} (|r_s|^2 + |r_p|^2) \) is independent of the modulation frequency \( f = \omega/2\pi \) and contains the parameters of the laser, the sample and the detector, we denote it with \( V_0 \). Also, the terms \( \sin[A \cos(\omega t)] \) and \( \cos[A \cos(\omega t)] \) can be expanded into harmonic series of \( \omega t \), having \( J_n \) the \( n \)-th order Bessel function:

\[
\sin[A \cos(\omega t)] = 2J_1(A) \cos(\omega t) + 2J_3(A) \cos(3\omega t) + ..., \]

\[
\cos[A \cos(\omega t)] = J_0(A) + 2J_2(A) \cos(2\omega t) + ... \]

therefore, Eq. 3.15 becomes

\[
V = V_0 [1 + J_0(A) \sin 2(\psi_r + \phi) + 4J_1(A) \psi_e \cos(\omega t) + 2J_2(A) \sin 2(\psi_r + \phi) \cos(2\omega t)], \tag{3.16}
\]

Separating the above result into terms containing the zeroth, first and second order harmonics in \( \omega t \) (and neglecting the higher ones), we obtain

\[
V = V_{static} + V_{1f} + V_{2f}. \tag{3.17}
\]

Since the rotation angle \( \psi_r \) is very small and \( \phi \) can be reduced by setting the analyzer to reduce the offset, \( \sin 2(\psi_r + \phi) \approx 2(\psi_r + \phi) \ll 1 \) and the zeroth order term \( V_{static} = V_0 [1 + J_0(A) \sin 2(\psi_r + \phi)] \approx V_0 \).

The measured 1f-signal is thus proportional to the Kerr ellipticity \( \psi_e \):

\[
V_{1f} = V_0 4J_1(A) \psi_e \cos(\omega t), \tag{3.18}
\]

while the 2f-component of the signal indicates the Kerr rotation \( \psi_r \):

\[
V_{2f} = V_0 4J_2(A) (\psi_e + \phi) \cos(2\omega t). \tag{3.19}
\]

The advantage of using a PEM is that we obtain amplified 1f- and 2f-signals purely proportional to the Kerr effect; minor offsets due to small misalignments in the setup can be corrected by tuning the orientation of the analyzer.

### 3.5.1 Vectorial detection scheme

Depending on the experimental configuration, we can distinguish three different types of MOKE measurements, as sketched in Fig. 3.11. **Polar MOKE** represents the case where the out-of-plane component of a thin film’s magnetization is measured. **Longitudinal MOKE** would be a measurement of a magnetization component in the sample
3.5. DETECTION BASED ON MOKE

plane, when the magnetization is in the plane of incidence of the light as well. Although much smaller in amplitude, there is a third type of MOKE geometry, where the magnetization component is again in the sample plane, but perpendicular to the plane of incidence of the laser light - this is called transversal MOKE.

As we have shown previously (eq. 3.11), if we ignore the very small transversal component, a fully perpendicular incident laser beam (\(\alpha = 0\) angle of incidence versus the normal to the sample surface) measures only the polar Kerr effect and is completely insensitive to the in-plane magnetization components. Similarly it can be proven that, although experimentally difficult to implement, an angle of propagation of \(\alpha = 90^\circ\) of the light inside the material would yield a measurement sensitive to the inplane magnetization (longitudinal MOKE) and insensitive to the out-of-plane magnetization. Evidently, any laser beam reflected on the sample surface in an angle \(\alpha \in (0, 90^\circ)\) will carry information about both the inplane and out-of-plane magnetization components (if there are any). Practically this means that, for a precessing magnetization, a laser beam falling oblique on the sample surface can be used to measure two of the three spatial components of the magnetization vector. However, separating the longitudinal and polar MOKE contributions in such a measurement would be very complicated if not impossible; the angle of incidence, the different sensitivities to the two types of Kerr effects, the complex constants \(C\) in eq. 3.11 correlating the Kerr effect and the actual magnetization and other parameters all mix up to generate the signal on the detector.

The easiest way of separating the longitudinal and polar Kerr contributions can be accomplished from realizing the following. If a second laser beam is incident on the same spot symmetrically against the sample normal (\(\alpha' = -\alpha\)) but in the same plane of incidence, having the same intensity and same polarization state as the first beam, we will have two reflected beams to measure the out-of-plane magnetization \(M_z\) with the same overall sensitivity \(K_p\) (\(p\) denotes “polar” type of MOKE). At the same time, the in-plane magnetization component \(M_y\) will contribute to the two detector signals with opposite sign, \(K'_l = -K_l\) (\(l = \)”longitudinal” MOKE). Note that the factors \(K_{p,l}\) represent the proportionality between the magnetization and the final voltage signal on the detector, including the constants \(C\) in eq. 3.11 as well as the factors in eq. 3.18 and 3.19. Measuring time-resolved with two such beams simultaneously, and adding the two data sets, we will obtain the double of the polar

![Figure 3.11: The three MOKE measurement geometries, drawn schematically. i = incident laser beam, r = reflected beam, FM = ferromagnetic structure, \(\vec{M}\) = magnetization vector.](image-url)
MOKE, that is, the dynamics of the out-of-plane magnetization component $2K_y M_z$, while subtracting them yields the longitudinal MOKE, proportional to the in-plane magnetization component $2K_l M_y$. Rotating the plane of incidence with $90^\circ$ (or using a second pair of laser beams) we can in the same way measure the out-of-plane $M_z$ and the second in-plane component $M_x$.

Since the incident and reflected beams would fully overlap, one can see that experimentally it would be very difficult to set up the above measurement. Hence we implemented a one-beam configuration based on a wide beam strongly focused on the sample with a high aperture objective, as shown schematically in Fig. 3.12. **With a probe beam incident perpendicular to the surface** (in the figure, for the ease of illustration the beam is drawn not completely perpendicular to the sample surface) and the reflected beam divided into four quarters, each quarter beam can be considered as a separate laser beam following exactly the above presented symmetrical path. The effective angle of incidence $\alpha$ for these quarter beams will depend on the focus distance $f$, the beam width $d$ and the power distribution across the beam, considered to be Gaussian. A beam splitter was used to deflect the reflected beam towards the four-quadrant detector. The relationships between the detector signals (A,B,C and D) and the three magnetization components $M_{x,y,z}$ are shown in the following table:
In our experiments that were based on pumping with magnetic field pulses, the vectorial scheme was relatively easy to implement, since only one laser beam had to be focused on the sample. We used a commercial microscope objective with a numerical aperture NA = 0.65, resulting in an effective angle of incidence $\alpha = 15^\circ$ for each quadrant beam. Since in case of small-angle excitations the $M_x$ component hardly changes during the precessional dynamics, we were unable to detect any effect of it. The setup was therefore simplified to using only two opposite quadrants of the detector and measuring $-K_lM_y + K_pM_z$ only.

The alignment of the probe beam on the optical elements, especially on the optical axis of the microscope objective, the sample normal and on the two quadrants of the detector is of course crucial. Any misalignment will induce errors in the assumption that the two opposite quarter beams have the same $K_l$ coefficient with the sign change. First of all, the DC power should be the same on both quadrants of the detector, an indication of good alignment on the detector. This is rather easy to realize. Any remaining crosstalk will be mainly due to the small misalignment of the beam versus the optical axis of the lens, which cannot be fully excluded. However, in the end results the crosstalk will show up in the two signals proportional to $M_y$ and $M_z$, calculated from the raw detector signals. Ideally, the phase difference between $M_y$ and $M_z$ should be $\pi/4$ (when the magnetization vector is at its maximum y-deviation, its z-deviation should be zero and vice-versa). This fact can be used to define two correction factors $c_A$ and $c_B$ for the two quadrants used (A,B) before we do the calculus of summation and subtraction. The total signal amplitude has to be preserved, not to affect the $M_z$-measurement. Therefore, we modify the calculus in the following way:

$$M_y \sim c_A \cdot A - c_B \cdot B;$$  \hspace{1cm} (3.20)

$$M_z \sim c_A \cdot A + c_B \cdot B;$$  \hspace{1cm} (3.21)

The correction procedure has thus the effect of setting the sensitivity to the in-plane magnetization component equal in both quadrants. In practice, the two correction factors proved to be very small, in order of 0.95 and 1.05 respectively. We note here, that once these correction factors were determined, they are always valid unless alignment changes are made in the measurement setup. Concrete examples of the vectorial MOKE setup in use will be shown in the next chapter.

In the case of the all-optical measurements, implementing the vectorial detection scheme presents some difficulties that will be addressed in detail in section 5.3.
Chapter 4

Precessional dynamics of micromagnetic structures

Lateral confinement of a ferromagnetic layer to structures of (sub)micrometer dimensions is an important goal of today’s recording and sensor technology. However, the reduced size manifests strongly in the fast magnetization dynamics of the structure, giving rise to a number of interesting (useful or unwanted) phenomena. This chapter is dedicated to the experimental study of the magnetization precession and spin wave phenomena in such confined structures, keeping in mind the consequences for possible applications. We will start with introducing some of the magnetic couplings and local (edge) effects that play an important role in the magnetization dynamics of a microstructure. We will describe the samples manufactured to study these effects and present the experimental results we obtained on them. First, a simple, almost ideal system of individual thin film Permalloy discs will be discussed, followed by experiments on a pillar of coupled identical ferromagnetic discs (“spin chain”). Finally, a set of measurements on Magnetic Tunnel Junction (MTJ)-type multilayered samples will be presented. The importance of coupling effects as local bias field in such structures will be discussed, and the practical approach of switching magnetization by large angle precessional dynamics will be introduced as well.

4.1 Local magnetic effects in microstructures

As we have explained in chapter 2, local inhomogeneities in the magnetic field through a ferromagnetic layer can lead to higher-order spin wave generation, to localized (standing) spin waves with shifted frequencies and to increase of damping. The inhomogeneities of the local magnetic field can result as much from the shape/size/growth properties of the studied ferromagnetic structure itself, as from interactions with nearby magnetic layers or structures.

Dipole-related edge effects

Let us consider a microscopically structured ferromagnetic layer of non-ellipsoidal shape, such as the top layer FM1 on Fig. 4.1, placed into a uniform, constant external
bias field of small amplitude that creates a preferential direction for the magnetization (aligns the dipoles).

One of the important sources of an inhomogeneous effective field through FM1 rectangle is the **demagnetization field** at the edges of the sample. Considering the sample as a surface, at these edges the spins in one half plane are “missing”, thus symmetry is broken. This results in a different coupling of the dipoles (weaker or stronger) compared to dipoles in the inner region. The change in the internal field caused by the missing dipoles at the edge is described by a demagnetization field, gaining more and more importance with reducing the sample size. This demagnetization field is thus the result of the missing dipole fields, \( H_{\text{demag}} = -H_{\text{dipole}} \). At edges parallel with the external bias field, the demagnetization field will be parallel with the bias (case depicted in Fig. 4.1), while at edges perpendicular to the bias field, the local dipole field will counteract the bias, resulting in a decrease of the effective field in this region. In the case of no external bias field, this demagnetization field can dominate the magnetic equilibrium and lead to domain formation within the microscopic sample, as we will show later in this chapter.

![Figure 4.1: Magnetic microstructure consisting of two ferromagnets FM1, FM2 separated by a thin insulator of rough surface. The magnetization of FM2 is considered fixed. The dipole field, the orange-peel interlayer coupling and the stray field of FM2 acting upon FM1 are indicated.](image)

**Interlayer couplings**

In case of applications, it is common to use a multilayered magnetic structure consisting of magnetic layers in contact or separated by non-magnetic metals or dielectrics. These magnetic layers may interact in many ways, influencing each others' static and dynamic magnetic properties.

Let us consider a simple microstructure consisting of two ferromagnets FM1 and FM2 separated by a thin insulator, see again Fig. 4.1. Such a structure resembles the active region of a magnetic tunnel junction (MTJ) stack, and will be discussed in the section “Samples” of this chapter. The bottom ferromagnetic layer FM2 is considered to have its magnetization direction fixed (e.g. through exchange bias by an antiferromagnet). Being very close to each other, coupling effects can appear between the two ferromagnets. One type of interlayer coupling is a consequence of the **stray field** \( H_{\text{stray}} \) of FM2 protruding into the top ferromagnet. Based on the geometry of the field line closure through the top layer, we can regard this effect as a
4.1. SIZE EFFECTS AND COUPLINGS

weak antiferromagnetic coupling between FM2 and FM1. The strongest effect of the stray field will manifest at the edges of the structure that are perpendicular to the direction of the magnetization of FM2, as presented on Fig. 4.1.

Competing with the stray field coupling, a ferromagnetic coupling can arise between the two layers, if the insulator between them is thin (\(\sim\) nanometers) and has a rough surface. This coupling is generally called orange peel - coupling and originates from the magnetic field lines of the FM2 layer that penetrate the barrier and close through FM1, as illustrated in, again, Fig. 4.1. If the thickness of the top ferromagnet is low, as it is in case of many applications, this coupling can be considered as acting uniformly over the whole FM1 structure, biasing its magnetization parallel with the underlying FM2 (thus opposite to the stray field).

A special case of repetitive stray field coupling of antiferromagnetic geometry can arise in a multilayered thin film structure of reduced dimensions. Let us consider \(n\) identical ferromagnetic structures without domain walls or vortices, vertically separated by thin, smooth insulating layers. If the coupling is dominated by the dipolar stray field of each ferromagnet, this field will align the magnetization of the nearest-neighboring structures antiparallel with each other, forming a so-called (artificial) spin chain. Dynamic magnetic excitation of such a spin chain (e.g. by the all-optical method described in the previous chapter) will lead to generation of standing spin waves in the multilayer, resembling the perpendicular waves in case of thin films (PSSW, see section 2.3.2). However, these are \(n\) discrete modes of order \(0\) to \(n - 1\) coupled via the stray field, each individual structure being a possible node. The quantization of the waves is not a result of the finite thickness but of the finite number of precessing macrospins. Therefore, the modal frequencies are determined not by the thickness of the multilayer structure but by the parameters of the individual discs and of the couplings between these.

Consequences of the field inhomogeneities and couplings

In case of static magnetization measurements such as hysteresis loops, the above described two coupling effects will manifest as a bias field acting on the magnetization, measurable both by local techniques (e.g. MOKE) and global ones (like SQUID).

The inhomogeneities in the effective local field will inherently lead to magnetization dynamics with a dispersion in its various parameters such as precessional frequency and damping. The different precessional frequencies will contribute to the full magnetic response of the sample in form of additional, localized modes. These modes, called intuitively edge modes [69], are expected to mostly manifest besides the uniform mode within a few micrometers from the sample’s edge.

Concerning magnetization dynamics over the entire ferromagnetic structure (important first of all for applications e.g. MRAM or magnetic sensors), the onset of the edge modes with a frequency shift will yield a dephasing of the precession from the central region towards the periphery, thus a non-uniform behaviour of the sample. Moreover, the effective damping, as explained in chapter 2, increases considerably in the measurement region due to the dephasing and the coexistence of multiple modes. These effects, together with the detection of higher-order spin waves and edge modes, will be the main purpose of the experiments presented in the next sections.
4.2 Samples

4.2.1 Permalloy disks on a coplanar waveguide

The first sample, fabricated to test our experimental setup and technique, is shown on Fig. 4.2 in a top view. It consists of a GaAs wafer substrate ($\epsilon_r = 13.5$), with several microscopic coplanar waveguides and an array of magnetic microstructures on it. The GaAs was chosen first of all for its convenient dielectric constant; a coplanar waveguide structure of 50 Ω impedance with $\epsilon_r = 13.5$ can be realized using signal leads and gaps of width in the order of 50–100 $\mu$m, easily accessible both optically and for electrical microcontacting (see further on in this section). Also, the good heat-absorbing properties of the GaAs were important, for absorption of the heat generated by the current through the structure. The waveguide itself was designed to give a 50 Ω impedance to minimize the electronic losses in the GHz range when the external electronic pulses are coupled into it. Therefore, an array of parallel, identical Au striplines of width 50 $\mu$m, thickness 200 nm and center-to-center distance 150 $\mu$m was used. The striplines were covered with an insulating SiN$_x$ layer (200 nm thick), to prevent a current flow through the magnetic structures on the top of the striplines. The magnetic elements consist of sputter deposited Ni$_{81}$Fe$_{19}$ (Permalloy, Py) discs of 4 to 50 $\mu$m diameter, with a thickness of 30 nm. Note that on the sample micrograph shown in Fig. 4.2, non-circular magnetic structures can also be seen. These being deposited in-between the striplines, were not used in our measurements.

To create the microstructures including the striplines and the Permalloy elements, UV lithography in combination with a conventional lift-off technique was applied. Two separate masks were used for the stripline array and for the array of Py dots, respectively. The sample was fabricated in collaboration with E. Smalbrugge from the

![Figure 4.2: Micrograph of a region on sample A (top view). The full sample contains 60 Au striplines and an array of Py “dots” with a diameter varying from 4 to 50 $\mu$m.](image)

In the experiments, three striplines were connected at once, the central one being chosen to have the Py discs on it, while the two adjacent lines were grounded. At the sample edges, the striplines were not covered with the insulating layer, allowing for touch-contact with a microscopic set of pins on both sides (see Fig. 4.3 for a schematic drawing). The central ("signal") line was connected to the two grounded lines via 50 Ω terminating resistors on the exit side, in order to facilitate the exit of the electronic pulses and reduce reflections back into the central line. Although the dc resistance of the stripline and the connectors was rather high (114 Ω), current pulses with a rise time of 200 ps could be sent through the central line. An upper limit of the field pulse amplitude can be estimated using formula 3.4: with $V = 5$ Volt electric field pulse applied by the generator, $w = 50\mu m$ width of the central line and a 114 Ω resistance, one obtains an amplitude limit of $H_{\text{pulse}} = 0.4$ kA/m. As it will be shown in chapter 5, the measured value is approximately 0.3 kA/m for the sub-nanosecond field pulses.

In our measurements, we concentrated ourselves on the 30 $\mu m$ Py dots. This choice had practical reasons - due to sample configuration, these dots proved to be the easiest accessible, both optically (focusing the laser beam) and electronically (contacting the corresponding striplines). In the same time, they are small enough to allow us considering the external field pulses uniform throughout the whole disc. Further in this work, we will refer to this sample as "sample A". Measuring the static hysteresis loop, the sample has shown a coercivity of 0.4 kA/m, with a clear in-plane preferential magnetization direction, but without any uniaxial anisotropies.

### 4.2.2 Pillars of coupled magnetic discs

To study the precessional dynamics and the effect of stray field coupling in a multilayered microstructure, a set of samples was produced by R. Brucas (Institute of Physics, Uppsala) and O. Kazakova (Chalmers University of Technology, Göteborg, Sweden), a so-called "artificial spin chain" ("sample B") shown in Fig. 4.4. First, an $\text{Al}_2\text{O}_3/\text{Ni}_{81}\text{Fe}_{19}$ multilayer was deposited on an oxidized Si(001) substrate by dc and rf sputtering of permalloy and $\text{Al}_2\text{O}_3$ targets. Then, a $1\times1\text{ mm}^2$ array of pillars of 300 and 550 nm diameter was created by electron beam lithography and ion-beam (Ar$^+$)
milling of the uniform multilayered film. Center-to-center distance between pillars was 0.6 and 1.0 µm, respectively. Every pillar consisted of ten Py discs of thickness 3 nm, separated by an 1.8 nm thick aluminum-oxide barrier. The pillars were grown on, and capped with, amorphous Al₂O₃ to ensure polycrystalline growth (low anisotropy) and prevent oxidation. As explained in the beginning of this chapter, due to the stray fields of the Py discs and the separating dielectric layer, an antiferromagnetic coupling was present between the adjacent layers within each pillar.

Figure 4.4: Schematic drawing of sample B: array of Al₂O₃/Ni₈1Fe₁₉ pillars on a Si substrate. For type 1, φ = 300 nm, d = 600 nm; for type 2, φ = 550 nm, d = 1µm. Layer thicknesses are given in nanometers between brackets.

The laser spot used in our TRMOKE measurements covered several of the pillars and, due to the reduced thickness of the Py discs, the sensitivity was extended to several layers below the top one.

Although in this case the measurements were done with the all-optical setup, not having an electronic waveguide built into the sample, the physics of the system were similar to the data obtained on the permalloy discs of sample A. The extra complexity of the dynamics resulted from the multilayered aspect of the sample and the dynamic coupling between the layers.

4.2.3 Rectangular magnetic structure within a MTJ-type microscopic cell

In the frame of a collaboration with H. Boeve at Philips Research Eindhoven, a set of micromagnetic samples ("sample C") were fabricated and measured by our field-induced MOKE technique. The goal of these measurements was studying the coupling- and size-related effects on the magnetization dynamics within a magnetic microstructure that is feasible for future applications. Hence these samples consist of a more complicated layer structure (a magnetic tunnel junction, MTJ), similar to a MRAM cell, on which static magnetic measurements were previously performed. On top of a Si/SiO₂ substrate, a copper layer was deposited and patterned into an array of striplines with contact electrodes by means of conventional lift-off techniques. As shown in Fig. 4.5, for each sample unit two contact pads of 100x400 µm² are interconnected by a stripline of 25 µm width. The thickness of the Cu structure (17 nm) was chosen to match the 50 Ω dc resistance as good as possible. At the central region of the stripline, a pile of several layers was grown to create a magnetic tunnel junction: Ta[3.5]/NiFe[3]/IrMn[10]/CoFe[4]/Al₂O₃[1]/NiFe[5]/Ta[3.5], where
4.2. SAMPLES

Figure 4.5: Micrograph of sample C, top view. On the left hand side, we see several of the MTJ-type structures on the substrate. On the right hand side, a magnified region of interest is shown: the contact pads, the Cu stripline and the MTJ pile. The pulse field rises along the y-axis; the effective DC field composed of external bias, interlayer coupling and anisotropy fields acts along the x-axis.

numbers between brackets represent thickness in nm. The Ta together with the NiFe on the bottom is a seed layer for the antiferromagnetic IrMn that exchange biases the 3 nm NiFe layer on it. The thin Al₂O₃ forms a tunnel barrier between the bottom CoFe and top NiFe magnetic layers. The whole stack is capped with Ta to prevent oxidation. The junctions have a rectangular shape and the lateral dimensions vary from 9x70 µm² to 5x5 µm². The exchange bias was predefined during growth in the direction perpendicular to the Cu stripline, however later it was rotated along the Cu line for the convenience of dynamic magnetization measurements.

The static magnetic properties of the sample were studied by SQUID. Prior to an annealing treatment (required for our experiments), we have found that the CoFe layer is strongly biased by the antiferromagnet, with an exchange bias field of 22 kA/m. On the other hand, the "free" upper Py layer is not exactly free - it has a slight coupling to the CoFe with an interlayer coupling field of 2.4 kA/m. The Py coercivity was determined to be 0.7 kA/m.

To improve the definition of the switching fields, as well as to rotate the exchange bias direction with 90 degrees, we annealed some of the samples for 30 minutes, in an

Figure 4.6: SQUID measurement on the MTJ-type magnetic microstructure, sample C, after annealing. The interlayer coupling field between the two FM layers \( H_{\text{int}} \simeq 5 \) kA/m and the exchange coupling to the antiferromagnet \( H_{\text{ex}} \simeq 31 \) kA/m are indicated.
external field of approximately 600 kA/m and at a temperature of 400K. The SQUID measurement data on the annealed sample are plotted in Fig. 4.6 and indicate an increased exchange coupling field (31 kA/m) and interlayer coupling field (5 kA/m). Simultaneously, the Py coercivity was reduced to 0.5 kA/m.

The high-frequency electrical connection was realized by slightly modifying the micropin connector used in the case of sample A. The two 100x400 µm² contact pads were used to transmit a current pulse; obviously, for the HF regime this connection scheme is not ideal, since there was no true waveguide structure involved. Due to the lower impedance (50 Ω) and smaller width of the stripline, formula 3.4 gives a theoretic magnetic field pulse amplitude of 2.1 kA/m, five times higher than in case of sample A. However, due to the lack of waveguide and proper impedance matching, the power loss and the reflections at the contacts are much more important in this case (see also chapter 5 for details).

4.3 Dynamics induced in small ferromagnetic discs

4.3.1 Uncoupled, individual Permalloy discs

When the vectorial measurement setup was built, for the “test” measurement we chose to use the ferromagnetic discs of sample A. When measured in an applied external magnetic field, these discs were expected to behave close to “ideal”, namely the magnetization precession was expected to be uniform and coherent at least within the central region of the 30 µm discs, due to the simple in-plane anisotropy geometry and lack of adjacent magnetic structures. Moreover, a real 50Ω impedance-matched waveguide structure was available to generate the magnetic field pulses as short and clean as possible. The only disadvantage of sample A consisted in the wider induction stripline and higher DC resistance, yielding a field pulse amplitude somewhat decreased but still sufficient to observe small-angle precession.

The High-Aperture Laser Objective (HALO) of NA = 0.38 was used to focus the probing laser beam on the sample (spot size being 6 µm in diameter). In order to increase the light intensity on the detector, the laser beam was sent off-axis into the HALO and the reflected beam was entirely deflected by a mirror towards the analyzer/detector. (This setup was later on altered using an on-axis incident beam, a microscope objective of higher numerical aperture and a beam splitter instead of the mirror - to gain resolution for the much smaller structures of sample C.) Since for such a thick sample, the Kerr ellipticity is much more sensitive to the dynamics of the in-plane magnetization component, a λ/4 retardation plate was inserted between sample and analyzer.

As explained in section 3.5.1, two quadrants of a four-quadrant photodetectors were used to measure the magneto-optic (MO) signal correlated with the field pulses. In order to saturate the magnetization in-plane and create a uniform magnetization without domain walls all over the Py disc, an in-plane DC bias field of 2.4 kA/m was applied perpendicular to the pulsed field direction. The field pulses were approximately 600 ps long and 0.3 kA/m high. The resulting MO signals, measured on the central region of the disc and amplified by the two lock-ins, are plotted in Fig. 4.7 a).
4.3. DYNAMICS INDUCED IN SMALL FERROMAGNETIC DISCS

Figure 4.7: Time-resolved measurement on 30 µm wide Py discs, \( t_{\text{pulse}} \approx 0.6 \) ns, \( H_{\text{bias}} = H_{\text{DC}}^{\perp} = 2.4 \) kA/m. a) Raw signal registered on the quadrants A and B of the photodetector, normalized to the DC voltage. b) \( M_y \) and \( M_z \) magnetization components calibrated to \( M_s \), calculated from the raw measurements using formulae 3.20 and 3.21.

Although not very large, there is a difference between the two signals, indicating that they have different contributions from the \( M_y \) and \( M_z \) components.

Based on the formulae 3.20 and 3.21, the dynamics of one in-plane (\( M_y \)) and the out-of-plane (\( M_z \)) magnetization component can be deduced; to remove the crosstalk between the quadrants, the correction factors were calculated to be \( c_A = 0.95 \) and \( c_B = 1.05 \). Since they result from the measurement geometry only, these correction factors are valid for all the measurements within the same unchanged optical alignment. The resulting magnetization components, calibrated to the saturation magnetization (\( m_{y,z} = M_{y,z}/M_s \)), are plotted in Fig. 4.7 b). The phase (\( \pi/4 \)) and amplitude (22:1) difference between the \( m_y \) and \( m_z \) signals is revealed; the latter indicates that the precessional motion is, as expected, strongly elliptical with a very small deviation out-of-plane. Interestingly, the subtraction used for the calculus of the in-plane component efficiently removes all correlated noise between the two raw signals from Fig. 4.7 a), yielding a very smooth \( M_y/M_s \) graph. Based on the arguments presented in section 3.5.1, the ratio between the Kerr measurements' sensitivities to \( m_y \) and \( m_z \) are calculated to be 1:90. The reason for this is a combination of two factors: on one hand, the angle of incidence of the laser beam is close to perpendicular, thus in our geometry we are more sensitive to the polar component of the Kerr effect; on the other hand, the polar Kerr rotation itself is in principle more sensitive to the magnetization, than the in-plane one [21]. This value is much larger than the 22:1 ratio between the amplitudes, therefore one can understand why the out-of-plane component is dominating the raw signals of Fig. 4.7 a) (and all the others that will be shown in this chapter).

For easier visualization of the magnetization precession, we plot in Fig. 4.8 the out-of-plane data against the in-plane data obtaining the projection of the precessional motion on the (yz)-plane. We can see how the equilibrium magnetization direction (the axis of precession) is canted out from the (0,0) position (the x-axis, in three dimensions) by the field pulse, to the position marked with a cross, followed by its drift back towards the original orientation. Simultaneously, the precession is dampened as
well. The correction factors $c_A$ and $c_B$ make sure that the long axes of the precessional ellipses are aligned horizontally, along $M_y$. It is clear from the figure, that the applied magnetic field pulse is shorter than the precession period; the magnetization precession happens almost completely around the original equilibrium direction, the x-axis.

Fitting any of the two curves in Fig. 4.7 b), excluding the time domain where the field pulse is still present, is possible with an exponentially dampened sine wave function of form

$$f(t) = Ae^{-\frac{t}{\tau}} \sin \left(2\pi \frac{t - t_0}{T}\right).$$

The fitting parameters $\tau$, $t_0$ and $T$ allow a determination of the precessional frequency $\omega = 2\pi/T$ and the Gilbert damping parameter $\alpha \approx \frac{2}{\tau \gamma \mu_0 M_s}$, considering that $H_x^{DC} \ll M_s$. For the 2.4 kA/m applied field, the calculus yields $\omega = 1.7$ GHz and $\alpha = 0.011$, values within the range of our expectations and also corresponding to the literature values $0.007$ [64] – $0.013$ [65].

A more interesting case of magnetization precession can be measured vectorially when the field pulse duration exceeds the precessional period $T$. Such a measurement is shown in Fig. 4.9, having the same external bias field of 2.4 kA/m but a field pulse of 1.5 ns duration and 450 A/m amplitude. On the two horizontal scales we plot the corrected and normalized magnetization components $m_y$ and $m_z$; the vertical scale is the time scale of the measurement. The plot, just like predicted by the LLG-simulations on Fig. 2.5, shows the two distinct precessional axes, corresponding to the magnetization equilibrium directions during and after the external field pulse.

Considering that the amplitude of the magnetization is kept constant, via the equation $m_x^2 + m_y^2 + m_z^2 = 1$ the $m_x$ component can be calculated, and thus a complete image of the magnetization dynamics can be obtained. Although studies have been reported addressing the possibility that the magnetization amplitude does not conserve after excitations, in our case of small perturbation with a field pulse we do not see a reason for a considerable change.
4.3. DYNAMICS INDUCED IN SMALL FERROMAGNETIC DISCS

Figure 4.9: Time-resolved measurement on 30 µm wide Py discs, \( t_{\text{pulse}} \approx 1.5 \text{ ns} \), \( H_{\text{bias}} = H_x^{DC} = 2.4 \text{ kA/m} \). Composite of two measurement of 4 ns duration each. \( M_y \) and \( M_z \) magnetization components are calibrated to \( M_s \), calculated from the raw measurements using formulae 3.20 and 3.21. The measurement is vertically extruded using the original timescale.

As explained in section 3.2.2, the constant bias field has a crucial importance in the LLG-type magnetization dynamics. A set of experiments was performed to investigate the influence of the external bias field amplitude on the precession frequency. The measurement points can be fitted well with the Kittel-formula 2.12 (adapted to our geometry),

\[
\omega = \gamma \mu_0 \sqrt{H_x^{DC}(H_x^{DC} + M_s)},
\]

with \( M_s \approx 900 \text{ kA/m} \), as shown in Fig. 4.10. At bias fields below 0.8 kA/m, due to less coherent precession of the spins across the laser spot (domain formation), the signal-
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to-noise decreases dramatically, therefore a precessional motion of the magnetization can hardly be detected. Note, that adding a small anisotropy term to the Kittel-formula next to the DC field, the curve gets shifted towards left; in fact, the lowest-field measurement point indicates that such an anisotropy might be acting (the fit is not very accurate for this point).

Based on the results presented above, we can conclude that the central region of the 30 µm wide, 30 nm thick Py discs behaves magnetically as a simple continuous thin film. Having only an in-plane anisotropy without preferential directions, its magnetization seems to be uniform and isotropic, yielding a coherent precession of all the spins with precessional parameters conform to the theoretic expectations. Domain formation and loss of coherence is easily prevented with a low (0.8 kA/m) external in-plane bias field.

Drastically reducing the size of such structures, as well as building them into a multilayered structure together with other (magnetic) layers can lead to completely different dynamics, as it will be shown in the following sections.

4.3.2 Artificial spin chains

A way of introducing interlayer coupling between ferromagnetic microstructures is to fabricate a multilayered structure as sample B, with permalloy discs in vertical columns – “pillars” – separated by a thin aluminum-oxide layer. The coupling between the discs is dominated by the dipolar stray field through the insulator, therefore an antiferromagnetic ground state along each pillar is favoured as shown in Fig. 4.11, perpendicular to the surface of the individual discs. (A metallic separation between the Py discs would additionally lead to an RKKY-type coupling.) Magnetic and structural characterization, experimentally and by simulation, of the sample is described in detail in [81]. Important for us is, that an AF coupling was shown – at zero field, there is no net magnetic moment over the array – and a 60 mT saturation field was found for the 300 nm wide discs. Also, the magnetization of the discs lies completely in-plane but there was neither uniaxial anisotropy nor a vortex state within the discs observed. In case of sub-micron, very thin Py discs such single-domain structure is also predicted by simulations of N. Dao et al., presented in [66]. Each individual disc can be considered therefore as a macrospin. Due to the dipolar coupling, the resulting structure is a chain of spins $S_i, S_j$ coupled by a Heisenberg-type interaction, $H = -J(d_{ij})\vec{S}_i \cdot \vec{S}_j$.

In a thin, single-layer structure numerical methods (for large excitations) or lin-
earizing the LLG equation (small excitations) allow for calculating the precessional frequency as a function of effective magnetic field, yielding a Kittel-type relationship \cite{16, 27}. Introducing the dipolar coupling between the discs, the calculus can be done to determine all the modal frequencies in spin chains as well, again, as a function of field \cite{81}. The coupling is expected to influence the magnetization dynamics in form of splitting it into a set of 10 discrete spinwave excitations, each corresponding to the moment of one disc at a certain “depth” within the pillar, with a well-defined coupling to the rest. (Coupling between pillars is neglected in the calculus.) The frequency vs. field curves yielded by the calculus are plotted in Fig. 4.12. Due to the negative dispersion of the spin waves\cite{67, 68}, the fundamental mode (called also “acoustical mode” and marked “a” on the figure) has the highest frequency. The “optical modes” i.e. higher order modes however, have frequencies that are intermixing for most field values, as the figure shows.

The influence of the field on the modal frequencies has two main characteristics. First, a slight variation of 5° in the angle of the applied external field (see solid vs. dotted lines) results in a considerable change of the curve for most of the modes. This can be important when comparing calculus with experiments, with respect to the accuracy of setting the field orientation in the setup. Second, one can notice, that above a critical field value (around 200 mT for this sample) all the \( f(H) \) curves start

![Figure 4.12: Calculated and measured modal frequencies in sample B. The solid lines represent the modes computed with field applied in 15° versus the sample perpendicular; the dotted lines correspond to field in 10°. Measurement results are plotted by empty squares (acoustical mode) and full circles (one optical mode). Figure is courtesy of M. van Kampen.](image)
to converge towards one frequency value, the frequency of the acoustical mode. This can be understood, considering that a “large enough” in-plane field component will break the AF type coupling between the discs and align their magnetization parallel. The discs at the edge (top/bottom) of the pillar experience a less stiff coupling (a weaker effective field), therefore these can be aligned easier than the ones in the central region.

Experimentally we studied the dipolar coupling via the all-optical type pump-probe setup. An external field of 150 to 250 kA/m in several steps was applied at an angle of $\sim 10 \pm 3^\circ$ versus the sample perpendicular, canting the magnetization of the individual discs slightly out of plane (necessary to be able to measure polar MOKE). For each magnetic field value, we measured the response of the out-of-plane net magnetization component to the laser-induced excitation, i.e. a damped oscillation in its amplitude. Since both the excitation (laser pump pulse) and the detection (Kerr effect) is based on absorption in the top region, the magnetization precession of discs at different depth within a pillar is expected to contribute to the measured signal (the net magnetization) in a non-equal way. Practically, the magnetic behaviour of the discs furthest to the base of the pillars are not detected.

A resulting time-resolved Kerr ellipticity measurement on the sample with 300 nm wide discs is plotted as an example in Fig. 4.13. The measurement is done in a nearly-perpendicular angle of incidence, therefore it almost exclusively represents the out-of-plane magnetization components in the ten discs within a pillar. Note that the laser spot covers a number of such pillars simultaneously; however, the static measurements have shown no interpillar coupling of a magnitude that would affect the measurement.

The fit with two exponentially damped sine waves reveals the contribution of a higher-order standing mode (precessional frequency $\omega=3.8$ GHz, optical mode) excited simultaneously with the fundamental mode ($\omega=5.7$ GHz, acoustical mode). Excitations of higher order have, probably, a contribution to the signal below the noise level.

Similar fits to other measurements with different field values consistently yield two coexisting modes of distinct frequencies, as plotted in Fig. 4.12 with empty squares (acoustical mode) and full circles (one optical mode), respectively. The ratio of 1:0.55 between the Kerr ellipticity amplitudes of the two oscillation components is also consistent through all the measurements. In the limit of these experiments, the differen-
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The differential Kerr signal can be considered as directly proportional to the variation of the $M_z$ component, that is, the amplitude of the spin waves. Hence we can conclude, that the same ratio exists between the amplitude of the acoustical and first optical mode. The experimental data in Fig. 4.12, although they do not fully overlap, show a good accordance with the theoretical predictions. The slight shift can be a result of the error in determining the angle and amplitude of the magnetic field, the contribution of the in-plane magnetization component that we neglected, or combination of these.

This result indicates exactly the magnetic order that is expected to dominate such a spin chain. The sample proved to be a great “model system” for an experimental and theoretical study of the stray field coupling and its direct effect on the magnetization dynamics, i.e., the coexistence of higher order precessional modes. Disturbing effects that could rise within the individual discs, such as edge modes, lateral or perpendicular spin waves are eliminated via the uniform perturbation with the $10 \times$ larger laser spot. Also, the relatively large uniform effective field (applied + stray) does not allow domain or vortex-state formation, not even a considerable dispersion in the equilibrium spin configuration through the disc, resulting in a macrospin state. With respect to the measurement technique, the limited depth of detection and the heat related effects (shockwaves) do represent a problem in the detection of the additional modes.

4.4 Dynamics induced in a rectangular MTJ element

As explained in the section “Samples”, our special interest was related to precessional dynamics in practical micromagnetic structures feasible for future applications. To study the size- and coupling-related effects in the $9 \times 13 \, \mu m^2$ ferromagnetic element of the MTJ (sample C), the spatial resolution of the setup was increased to $\sim 1 \, \mu m$. This was realized via a microscope objective of $NA = 0.65$, used instead of the HALO; a sketch of the experimental setup is shown in Fig. 3.3 in chapter 3. A beam splitter in front of the objective was used to separate the incident and reflected beams, that followed the same path in this setup. Since the thickness of the Py layer (5 nm) used in our measurements is below the typical penetration depth of light (13 nm), the noise level increased considerably. To compensate for this, we chose to measure the Kerr rotation instead of the Kerr ellipticity, resulting in a larger sensitivity to the in-plane magnetization dynamics [54].

4.4.1 Central region, small angle local precession

First of all, we have performed a set of TRMOKE measurements on the central region of the $9 \times 13 \, \mu m^2$ Py element using “short” (0.6 ns) and “long” (1.5 ns) field pulses. These measurements were expected not to show any effect of the sample’s edges, the local effective field being governed by the orange-peel and stray field interlayer couplings only ($H_{eff} = H_{int}$). A typical “short pulse” result is shown in Fig. 4.14 a). The 0.6 ns current pulse launched by the generator is the same as in case of sample A, however, the lack of an impedance-matched waveguide structure and the lower static resistance resulted in different magnetic field pulse characteristics. No external bias
field was applied for the measurement; as shown in section 4.2.3, the couplings to the bottom ferromagnetic layer give rise to a 5 kA/m constant magnetic field directed along the x-axis (in-plane, perpendicular to the pulse field). The dynamics of the \(m_y\) and \(m_z\) magnetization components are calculated, again, from the raw signal of the two detector quadrants using formulae 3.20 and 3.21 and plotted in Fig. 4.14 b).

Comparing the result with a similar measurement on sample A, shown previously in Fig. 4.7, two differences are imminent. First, the oscillation amplitudes are doubled (indicating a more intense excitation); second, the in-plane \(m_y\) signal is noisier and full of nonuniformities, probably due to the thin sample and the non-ideal shape of the field pulse (afterpulsing with longer decay time). For a full analysis of the magnetic field pulses acting upon the spin system in sample C we refer to chapter 5.

![Figure 4.14: Time-resolved measurement on 9 × 13 µm² Py structures, part of a MTJ-type stack.](image)

Frequency

The pattern formed by the measurement points on Fig. 4.15 has the characteristics of a Kittel-type relationship with its symmetry axis shifted from \(H_{bias} = 0\) to \(H_{bias} = 5\) kA/m. This is expected from the static magnetic properties of the sample: the interlayer coupling field \(H_{int}\) acts locally on the Py structure, contributing to the total effective DC field. Additionally, the slight uniaxial anisotropy \(H_{ani} \sim 0.3\) kA/m) of the sample, shown previously also by the SQUID measurements in Fig. 4.6, can be noticed via the gap between the two branches of the curve. The fit to the measurement points is therefore possible by considering the Kittel-formula similar to the top left
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Figure 4.15: Bias field dependence of the precessional frequency and damping in the central region of the Py rectangle; the line is a fit with the modified Kittel-formula 4.1. The damping was extracted from a fit with an exponentially damped sine wave. The gray dots in the frequency plot indicate a second precessional mode, obtained from a fit with two exponentially damped sine waves.

The saturation magnetization value (570 kA/m) of the Py structure is lowered by the interface effects, known to have a considerable influence for layers thinner than 10 nm [54]. In the same time, the rectangular shape of the sample gives rise to an anisotropy field not bigger than 0.06 kA/m (demagnetization factors being \( N_{x,y} \approx \frac{2t}{\pi l_x l_y} \) [83] with layer thickness \( t \) and lateral dimensions \( l_x, l_y \) yielding \( N_x \approx 0.00035 \) and \( N_y \approx 0.00025 \)). Therefore, the observed 0.3 kA/m anisotropy field cannot be explained by shape anisotropy only; a contribution of growth-related crystalline anisotropy has to be considered.

A second set of frequencies is shown by gray dots in Fig. 4.15, having a very low or no dependence on the applied field values. This set was obtained by fitting some of the measured data at high negative bias fields with two exponentially damped sine waves, representing two precessional modes. Two of these datasets are plotted in Fig. 4.16, with \( H_{bias} = -4.3 \) kA/m and -7.2 kA/m, respectively.

We do not have a clear explanation for the appearance of a second mode in the center of the sample; the following effects might play a role. On one hand, the top ferromagnetic layer is thin (5 nm) and the thin insulator can be considered transparent, therefore a magnetic response of the bottom “pinned” (CoFe) layer could be detected by the laser beam. Quantitatively, this explanation has some discrepancies. The effective DC field (coupling + applied) through the CoFe layer, perpendicular to the field pulse, is in the order of 40 kA/m; considering, that the saturation magnetization of the CoFe should also be higher (\( \sim 1200 \) kA/m) than the one of the Py, the frequency of the first-order precessional mode in the CoFe should be three times as much as the value obtained from our fits and plotted in Fig. 4.15. Also, it should depend on the bias field, and it should be present at lower bias field values as well.

On the other hand, the pulse shape is not very regular in case of this sample (see chapter 5 for details). A strong afterpulsing, combined with a strong magnetic field, can result in the onset of a second precession at a certain moment. If the phase of

\[
\omega = \mu_0 \gamma \sqrt{(|H_{bias} - H_{int}| - H_{ani})(|H_{bias} - H_{int}| - H_{ani} + M_s)}.
\]
this precession is coincident with the phase of the original one, this could manifest as an amplification of the original precession, something similar to the measurement plotted in Fig. 4.16 a). If the second precession starts in antiphase with the original one, it might show up as a sudden amplitude decrease of the precession, which could be the case in Fig. 4.16 b). The quality of the double-frequency fits is not very high, indicating that the interpretation of the beating effect as two simultaneous oscillations might be wrong. However, the question arises why in the low bias field measurements the afterpulsing does not show such an effect.

Damping

The phenomenological damping parameter, extracted from the same fits to the data-sets, is also plotted in Fig. 4.15. For positive and small negative applied fields, the resulting $\alpha = 0.01 \pm 0.001$ corresponds to the values we have found for sample A, as well as the values given in literature. At negative applied fields stronger than -2 kA/m however, again an irregularity appears: the apparent damping decreases ($\alpha = 0.005 - 0.008$), a very surprising fact at first sight. A second precessional mode (or an artifact resulting from afterpulsing) can explain the low apparent damping of these measurements – the damping obtained from a single-exponential fit will suffer from the contribution of these features. Moreover, at very high negative fields (-5 kA/m and stronger, see the measurement plotted in Fig. 4.16 b) ) we were unable to determine a unique value for the damping.

From the central measurements on the Py microstructure we can conclude, that at applied fields above -2 kA/m the magnetization dynamics is completely dominated by the interlayer coupling, a small uniaxial anisotropy and the external applied field itself. The experiments show a regular GHz-range precession at a frequency in good accordance with the Kittel formula; the value of the determined Gilbert damping $\alpha = 0.01 \pm 0.001$ corresponds to values found for an uncoupled thin layer, such as sample A or many cases from the literature [64, 65]. At high external fields applied opposite to the interlayer coupling in the sample (-2 kA/m or more negative), a second precession was detected, with a frequency somewhat lower than the fundamental precession in
the Py; in the same time, a decrease of the apparent damping was noticed. This second precession might represent the onset of a magnetization precession within the “pinned” CoFe layer or an artifact resulting from the non-ideal shape of the magnetic field pulse, but we have no quantitatively correct explanation for the effect.

4.4.2 Lateral spin waves: detection of central and edge modes

It is well known from literature [40, 82], that microscopic ferromagnetic structures of non-ellipsoidal shape, biased by an external magnetic field, exhibit a dispersion in the local effective field. This leads to a dispersion in the precession frequencies of regions within the sample, giving rise to spin waves of higher order besides the fundamental precessional mode. Naturally, these extra modes rise to a considerable amplitude in regions where the symmetry of the sample is broken, e.g. the edge of the sample. Besides its frequency, the damping of the precessional dynamics is also influenced; the motion of the spins becomes incoherent within the sample, increasing the apparent damping. The transfer of energy between the central and edge spin modes can also lead to additional damping, difficult to distinguish from the intrinsic (Gilbert) damping parameter. The dispersion of effective fields, increase of damping and the rise of higher order spin modes in non-elliptical FM microstructures are studied extensively by Jorzick et al. (Brillouin light scattering experiments, [40]). A general introduction to lateral spin waves can be found in section 2.2.8 of this thesis.

In the case of MTJ type structures however, the interlayer couplings in addition to the edge effects lead to a further complication in the spectrum of the local magnetic fields. The physics of magnetization precession in a multilayered rectangular microstructure as the one drawn in Fig. 4.1 was discussed in section 4.1. of the current chapter, simultaneously with the prediction of the localized precessional modes.

![Figure 4.17](image)

Figure 4.17: Time-resolved measurements of the $M_z$-component at different positions across the sample; effective DC field was provided by the demagnetization at the edges and the interlayer coupling with the underlying CoFe structure. The field pulse was of 0.6 ns duration and 1.2 kA/m amplitude. The measurements show a clear difference between the central (C,B) and the edge (A) precessional modes.

In order to detect the higher order localized modes in the rectangular Py element of sample C, we performed several measurements on the edge of the structure. The geometry of the experiments was the so-called “magnetostatic backward volume modes” (MSBVM) geometry [40], explained in detail in the framework of the lateral
spin waves in chapter 2. An example of such a set of measurements, executed on the center and the edge of the $9 \times 13 \mu m$ Py rectangle is shown in Fig. 4.17. Note, that applying an external bias field was not necessary since the interlayer coupling between the Py and the CoFe provided a local magnetic field $H_{\text{int}} = 5 \text{kA/m}$. The frequency at the edge (position A, $4 \mu m$ from the center) is $7\%$ lower than in the center (C) due to the dipole demagnetizing field in the region and the stray field of the CoFe rectangle underneath the studied Py. In the same time, the apparent damping $\alpha_{\text{eff}}$ increases drastically towards the edge, nearly three times. We consider the following reasons for this increase: loss of coherence in the dynamics within the laser spot ($\alpha_{\text{deph}}$) and transfer of energy between the central and edge modes ($\alpha_{\text{prop}}$), resulting in two additional terms next to the intrinsic Gilbert damping: $\alpha_{\text{eff}} = \alpha_i + \alpha_{\text{deph}} + \alpha_{\text{prop}}$.

Mounting the sample on a 2-dimensional motorized translation stage, we performed a number of controlled “surface scans”, meaning sets of pump-probe measurements on different positions of the sample surface, with a resolution of $1 \mu m$. The results of such measurements are comparable with the ones presented by Park et

Figure 4.18: Top: Out-of-plane magnetization dynamics along lines parallel ($x$) and perpendicular ($y$) to the bias field, crossing the center of the $9x13 \mu m^2$ Py sample. Bottom: Fourier transformations of the same measurements, showing the shift of the precessional frequency $\omega$ at the sample’s edges. Middle inset: coordinates $x, y$ across the sample surface.
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al. [69]; however, in our case, the largest effect on the local magnetic fields is brought by the stray field of the underlying CoFe structure, with its pinned magnetization. As explained in the theoretical introduction of this thesis (section 2.2.8, Lateral spin waves), the quantization of the wave vector \( k = n\pi/w \) due to the reduced lateral dimensions \( w \) can lead to generation of backward volume spin waves [40].

As an example, the top two images in Fig. 4.18, marked (x) and (y), illustrate the space-dependent dynamic response of the out-of-plane magnetization component \( m_z \) to a 0.6 ns long magnetic field pulse. These measurements were done along lines crossing the center of the sample parallel (x) and perpendicular (y) to the effective bias field direction (x-axis) as shown in the middle inset of the figure. The amplitude of the oscillation, \( M_z/M_s \), is represented by the light and dark tones of the images, the values being in the same range as in Fig. 4.17, namely, between \(-0.75\%\) and \(+0.68\%\). The bottom two images are the corresponding Fourier-transformates, indicating the intensity of the frequency components as a function of the position. The curvature of the dark/bright stripes in the measurement plots represent a shift in the precessional frequency towards lower/higher values in the edge region, as already observed in Fig. 4.17. Additionally, the Fourier-transformation for the (x) linescan shows a breaking point of the main frequency line (1.85 GHz) at -4 and +4 \( \mu m \) distance from the center. This is an indication of a second, distinct localized mode at the sample’s top/bottom edge, generated in a different frequency domain (1.68 GHz). The frequency shift corresponds to a difference of 0.8 kA/m between the local effective fields, indicating the competition of the stray field and the demagnetization at the edges perpendicular to \( H_{bias} \).

The linescan-measurements plotted in Fig. 4.18 also show a much higher damping in the edge region. The measured damping values are plotted in Fig. 4.19 versus position along the x-axis. The result corresponds to our expectations from section 4.1: the dephasing precession within the measurement spot, as well as a transfer of energy between the fundamental and higher order spin waves result in an enhanced apparent damping, as already indicated by the three distinct measurements of Fig. 4.17. Similar but less dramatic increase of the damping is observed when scanning along the y-axis; the dephasing is smaller in this direction, since the stray field coupling is not contributing, only the edge demagnetization.

To get an image of the magnetization dynamics over the whole sample, we performed so-called raster scans, where the delay line (thus the pump-probe time delay) was fixed at different positions, while the sample was scanned under the laser spot. The measurement scheme being the vectorial one, both the \( M_y \) and the \( M_z \) com-
Figure 4.20: Dynamics of the out-of-plane magnetization component measured over the whole sample surface recorded in steps separated by 50 ps; the 0.6 ns long field pulse starts at frame 1. A strong dephasing can be seen during the free precession (frames 13-30). Top inset overlapping frame 2 shows the colour coding corresponding to the $M_z/M_s$ values.

The measurements were made, at time intervals of 50 ps. No external bias field was used (the effective field being, again, built by the local effects discussed previously). The field pulse was the one of 0.6 ns duration and 1.2 kA/m amplitude. The “image frames” put together as plotted in Fig. 4.20 highlight the interesting features of such a microscopic magnetic rectangle, built into the MTJ stack. While the magnetic field pulse is still on (frames 1-12), the measurements show a uniformly excited magnetization via a constant out-of-plane component $M_z/M_s$ over the whole surface. Starting with frame 13, the free precession of the magnetization is measured; one can immediately see a strong dephasing between the magnetization precession of the different regions due to
the differences in the local precession frequencies as explained previously. On frame no. 15 e.g., the top and bottom edge of the sample still presents a magnetization with a positive out-of-plane component (dark coloured); the middle of the element has its magnetization vector completely in-plane (mid-gray), while the brightness of the left-right edges indicate a negative $M_z/M_s$. The set of images is thus in accordance with the previously presented, continuous time-resolved measurements at different positions on the sample.

Concluding this section, we have shown experimentally that the coupling- and edge demagnetization effects strongly influence the local effective magnetic fields. The $H_{\text{eff}}(\vec{r})$ dependence yields, even with a uniform excitation over the whole sample, a shift of the magnetization precession frequencies and a strong damping enhancement close to the sample’s edges. On the other hand, the Fourier-transformations of the time-resolved “linescan” measurements show a distinct precessional mode localized at the edges (“edge mode”), generated in a frequency domain offset from the central mode.

Similar results were obtained on $3 - 6 \mu m$ wide, $15 \text{ nm}$ thick permalloy disks by Buess et al., using time-resolved polar Kerr microscopy [70, 71]. The geometry of these experiments was based on a microscopic coil instead of a stripline, surrounding the magnetic sample and generating an out-of-plane magnetic field pulse via induction. The electrical pulses were triggered optically. Magnetic eigenmodes up to third order were visualized with the aid of a Fourier transformation of a sequence of magnetic images taken at different delay times. However, these modes represent lateral standing waves within the discs generated from a vortex-type initial spin configuration, while in our case the central and edge modes start from a completely in-plane equilibrium state.

4.4.3 Numerical simulations, the effect of the stray field

So far we experimentally observed a dispersion in the precessional frequency over the sample surface, and we attributed it to the existence of a non-uniform effective field. This is built up by external fields, demagnetization fields along the edges, stray fields and orange-peel couplings to the bottom CoFe layer. In order to verify the influence of the couplings and local demagnetization effects, a set of numerical simulations were performed with the OOMMF program package [72]. The parameters (sizes, thicknesses, magnetization values, applied fields etc.) were chosen to match the geometry of the MTJ sample and the experiments in general.

In the case of no applied DC field as described in the previous section, the CoFe stray field at the position of the Py was calculated. Combining its effect with the orange-peel coupling and the demagnetization effect at the edges, the internal effective field $H_{\text{int}}$ was obtained to have a position-dependence as shown in Fig. 4.21. We have found that the stray field has the strongest influence at the edges of the sample parallel to the x-axis (perpendicular to the direction of the interlayer coupling), and it manifests itself in a strong suppression of the local effective field.

The resulting field distribution was included in a simulation of a time-resolved “line scan” experiment (already shown in Fig. 4.18), based on the Landau-Lifshitz-Gilbert equation and cell sizes of $25 \times 25 \text{ nm}^2$. The output of the simulation, together with the corresponding experimental result, is plotted in Fig. 4.22. The correspondence
between measurement (a) and simulation (b,c) becomes satisfactory only in the case when the stray field is included in the calculus of $H_{\text{int}}$ (b). In the measurement plot, a difference between the left-hand side and right-hand side edge frequencies is noticeable (1.15 and 1.3 GHz indicated by dotted lines). This is unexpected and can be explained with experimental uncertainty, as well as etching defects on the sample. In the case of the simulation with the stray field included, the distinct central and edge modes ( $f_c = 1.85$ GHz and $f_e = 1.31$ GHz, dotted lines) appearing in the fast Fourier transform are symmetric and very close to the experimental values, within the limits of the experimental precision. Moreover, in this simulation the continuous shift of the central frequency ($\Delta f_c \simeq 0.2$ GHz) is also visible, as well as the higher frequency FFT peak at 2.75 GHz that was vaguely noticed in the measurement. An additional peak ($f = 1.66$ GHz, dotted circle) can be seen at the edges in the FFT simulation (b) as continuation of the central mode, indicating the coexistence of the two distinct modes in this region. This peak is not clearly visible in the measurement (a), possibly due to its lower amplitude and the lower resolution of the experiment. The results of the simulation without stray field (c) fall short from reproducing the measurement both qualitatively —missing peaks at the edges— and quantitatively —$f_e = 1.59$ GHz versus the 1.15 – 1.3 GHz that we measured—.

We can conclude, that the stray field of the CoFe incorporated into the MTJ stack has a crucial role in the formation of multiple precessional modes of the Py magnetization. A correct model of the Py dynamics has to include the stray field coupling and its effect on the Py internal field. Further details on the simulations as well as the applied field dependence of the central- and edge modes in the Py rectangle are presented by Rietjens et al. in [73] and [74].

### 4.4.4 Dynamic domain imaging

So far we have shown measurements with a well-defined effective field over the whole Py rectangle, with the nonuniformities restricted to the edge regions. While the effective field was space-dependent, the initial (equilibrium) magnetization was considered...
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Figure 4.22: a) FFT of a “line scan” measurement along the x-axis, no external bias field (same as Fig. 4.18 (x)); different modes are indicated with dotted lines and circles, see text. b) Simulation of the same experiment including the stray field of the underlying CoFe rectangle; two distinct modes are clearly visible at the sample edges. c) Simulation ignoring the stray field; the central frequency is flatter and only one distinct precessional mode visible at edges. Figures courtesy of J. H. H. Rietjens.

to be uniform over the sample due to an overall coupling field of 5 kA/m. The question arises, what will happen with the dynamics of the magnetization in absence of such coupling, i.e. governed only by the local effects (stray field, demagnetization at edges and a slight uniaxial anisotropy). In this case both the effective field and the magnetization will be space-dependent, resulting in domain formation, vortex structures etc. On an ideal MTJ-type sample this is a given configuration, since no orange-peel coupling between the pinned and free ferromagnets is present. In our sample, one possible way to achieve this experimentally is to compensate for the interlayer coupling with an external bias field applied antiparallel to the coupling field.

“Raster” scans similar to the ones presented in the previous section were done using a 5.2 kA/m bias field, generated by a pair of Helmholtz coils. This DC field compensated the interlayer coupling almost completely, allowing the local magnetic fields within the rectangle to form a microscopic domain pattern. By doing space- and time-resolved measurements on different regions of the rectangle, we can probe the response of the local magnetization vector on the field pulses. Observing the frequency and the phase of the precessional motion, we can obtain information on the static domain structure of the moment previous to the arrival of the field pulse. As a
simple example, when the initial magnetization direction is parallel to the field pulse (along y-axis), according to the Landau-Lifshitz-Gilbert equation no excitation will occur since there is no torque acting on the magnetization vector. However, when the in-plane magnetization is initially perpendicular to the pulse field (thus aligned along the x-axis), a torque will create an oscillating nonzero \( M_z \) component. By looking at the phase of the oscillation at a certain moment, we can distinguish between an initial magnetization parallel or antiparallel with the x-axis.

![Image](image.png)

Figure 4.23: a) \( M_z \) component of the magnetization over the whole sample, 50 ps after the onset of the field pulse. The darker the shade the more positive the measured value. A compensating DC bias field of 5.2 kA/m was used. b) Initial orientation of the local magnetization pattern, deducted from the measurement in a) and the LLG torque equation.

A raster scan of the Py rectangle taken at a fixed time delay of 50 ps after the onset of the field pulse is shown in Fig. 4.23 a). A domain pattern is revealed by plotting the intensity of the \( M_z \) component in shades of gray; white indicates the positive extreme, black means the largest negative value for the out-of-plane magnetization. Based on this set of measurements, we can reconstruct the stable domain structure that was present within the microstructure before the field pulse disturbed it. An advantage of the dynamic measurements versus static MOKE microscopy is, that we detect the change of the magnetization in time, thus the measurement is differential, giving a contrast of approximately 2 orders of magnitude higher between signal and background [4]. Due to the differential nature of the measurements, topography-related noise is seriously reduced. However, in our case a strong precession was started even by a relatively weak field pulse due to the lack of pinning by an external field. In case of experiments with a very small amplitude precession the signal quality is expected to deteriorate.

A schematic drawing of the result is shown in Fig. 4.23 b); the stray field along the x-axis has a strong influence on the “horizontal” (top/bottom) edge regions, aligning the spins perpendicular to the edge. Due to the small uniaxial anisotropy, in the central region the spins align along the y-axis. Two vortices can be seen forming in the top right and bottom left corners.
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To put the above speculations on stronger feet, we decided to measure the full time-
response in different regions of the structure. The results confirmed the correctness of
the sketch in Fig. 4.23 b); as examples, three measurements are plotted in Fig. 4.24.
On the first graph (central region), an almost complete lack of dynamic response was
measured, indicating the lack of torque on the spins aligned along the y-axis. The
second and third graphs show peaks of opposite signs, resulting from two domains
with antiparallel magnetization vectors aligned at an angle versus the y-axis. Due
to the large apparent damping and the very low effective fields, no regular (sinusoidal)
oscillation can be seen in these measurements.

Figure 4.24: Three time-resolved measurements at different positions on the sample,
indicating different magnetization dynamics. The first graph shows almost no dynamic
response ($\vec{M} \parallel \vec{H}_{\text{pulse}}$); the other two show a start of precession in opposite directions,
representing domains with antiparallel magnetization vectors.

With the aid of the OOMMF package [72], numerical simulations were performed
on the sample, this time with compensated interlayer coupling. First, the spin system
was left to relax to equilibrium without any field pulse, using the specific parameters
of the experimental sample (stray field of the underlying CoFe, edge demagnetiza-
tion, uniaxial anisotropy, dimensions, saturation magnetization, exchange stiffness,
etc). Then, a field pulse was simulated along the y-axis, having the same shape
and amplitude as the experimental one. The LLG-based simulation was resulting in
“snapshot images” at different moments during the precessional motion of the local
magnetization vectors. The preliminary results are in accordance with the experi-
mental “image” we have shown. For the details of the simulation and the fine-tuned,
most recent results see references [73] and [74].

From the results of the raster scan type measurements with a space-dependent
magnetization vector we can conclude that, on one hand, we have a powerful tool of
recording both the magnetization dynamics and the equilibrium domain structure on
microstructured ferromagnetic samples. On the other hand, the results indicate the
increased importance of local magnetic effects (stray fields, edge demagnetization)
within multilayered structures with reduced dimensions. These can lead to mag-
netic domain and vortex formations, yielding an incoherent magnetization dynamics
through the structure. Hence, a fast and stable switching of the whole ferromag-
netic structure in complicated devices may present difficulties and deserves special
attention.
4.4.5 Large angle precession

In the precessional experiments with uncompensated interlayer coupling, the amplitude of the magnetic field pulse was relatively small compared to the DC effective field. The canting angle of the magnetization equilibrium direction measured to its static direction prior to the pulse onset was therefore small, resulting in a small-angle precession. Looking at the maximum values of $M_y/M_s$ and $M_z/M_s$ on Fig. 4.14 (0.3, respectively 0.015), we can estimate the precessional angle as $17^\circ$ in-plane and less than $1^\circ$ out-of-plane.

However, in the regime with bias field values between 3.3 and 6.7 kA/m, the difference between the bias- and interlayer coupling fields (thus the effective DC field in the center of the element) is below 1.5 kA/m. Consequently, the spins are less “pinned” in the x-direction. The magnetic field pulse with an amplitude of 1.2 kA/m becomes comparable or in some cases even exceeds the local DC field, resulting in a canting of $45^\circ$ or more for the magnetization equilibrium direction. A schematic drawing of such a case ($H_{\text{pulse}} > H_{\text{bias}}^{\text{DC}}$) is presented in Fig. 4.25. In the case of these measurements, we noticed that the $M_y(t)$ and $M_z(t)$ data were very difficult to fit with an exponentially damped sine wave. The reason for this was a large angle precessional motion, with a considerable change in the $M_x$ component. At bias field amplitudes close to the interlayer coupling field, the uniaxial anisotropy in the y-direction becomes important as well, yielding a very complicated precessional motion. Before we discuss switching dynamics based on precession (next section), let us spend some time on these large-angle precessional experiments.

As an example of such behaviour, a measurement done with a compensating DC bias field of 4.3 kA/m is shown in Fig. 4.26; this results in an overall DC bias field of approximately 0.7 kA/m. A 0.6 ns field pulse was used, with a maximum amplitude of 1.2 kA/m. The length of the magnetization vector (physically representing $M_s$) is considered to be constant during the measurement. From the top two graphs one can see that the initial magnetization vector is not fully aligned with the x-axis ($M_x/M_s < 1$); the relationship between the $M_x$ and $M_y$ components indicate an initial angle of $\phi_i = 29^\circ$. On the top graph, at approximately 1.5 ns, we can see the effect of the pulse’s falling edge: the oscillation started by the rising edge is disturbed. In all three graphs the free precession (started by the falling edge) shows a very low frequency and high (apparent) damping; after two periods, the precession is practically gone. This is attributed to the very low local fields and the strong dephasing of the spins within the measurement region, respectively. We have to note that, due to the strong dephasing, the consideration $|\vec{M}| = \text{const}$ in the large-angle...
Figure 4.26: Measurement of the three magnetization components during a large angle precession with a 4.3 kA/m DC field applied along the x-axis (\(H_{DC} \simeq 0.7 \text{kA/m}\)) and an 1.2 kA/m field pulse of 0.6 ns duration along the y-axis. The latter is also indicated on the (y) graph as calculated in Chapter 5. All values are normalized to the saturation magnetization of 570 kA/m.

In Fig. 4.27 we have plotted the same measurement data in the real (xyz) space, to give a better impression about the magnetization dynamics. The 3-D plot reveals some more peculiarities of such a “precession”. On one hand, the excitation angle is very large (\(\phi_e = 119^\circ\)), indicating a deviation of the equilibrium direction of almost 60\(^\circ\) due to the rising of the field pulse. On the other hand, the “final” magnetization direction (after 4 ns) is not the same as the initial one, that is, the magnetization does not relax to the initial equilibrium state but it rotates towards \(m_y = 0.96\), indicating a switch of \(\phi_s = 45^\circ\). Considering, that the initial magnetization direction had a nonzero angle (\(\phi_i = 29^\circ\)) with the x-axis, the two angles add up and the final direction comes very close to the y-axis that is, the anisotropy direction. During the last 1.5 ns of the experiment, the precessional motion is stopped by the large effective damping and a slow drift towards the original equilibrium direction \(\phi_i\) can be seen. This drift has to be completed during the following 8.5 ns, before the next magnetic field pulse arrives - otherwise a stroboscopic experiment would not be possible.
As a conclusion, we have been able to generate large-angle excitations in the range of 120° with the condition of a sufficiently suppressed interlayer coupling. Within these dynamics, the $M_x$ component changed drastically, reaching negative values. The magnetization switched temporarily towards the $y$-axis due to the slight uniaxial anisotropy present in the sample, however on the long timescale it returned to the original equilibrium direction. No 180° switch was achieved. A strong decoherence of the precessing spins due to local magnetic fields led to a very high effective damping, quenching the free precession after only two periods.

4.4.6 The ultimate goal: precessional switching

We have seen from the presented large-angle precessional measurements, that a sign reversal of the $M_x$ magnetization component is possible on a very short timescale. Practically, in half of a precessional period of $\approx 800$ ps even a relatively small pulse field (1.2 kA/m) can reverse the magnetization almost completely. In the ideal case, when the field pulse ($y$-axis) is much larger than the effective DC field ($x$-axis), with the onset of the pulse the new equilibrium direction will be along the $y$-axis. The torque acting on the magnetization will push this out of the sample plane into a precessional motion; however, due to the in-plane anisotropy, a strong out-of-plane demagnetization field will rise at this point ($H_{\text{demag}}(t)$, see Fig. 4.25). Generating a new torque this will take care of the fast rotation of the magnetization vector in a plane almost parallel to the sample plane. The result will be a precessional angle close to 180°. Hence, the $M_x$ component will perform a full sign reversal and after half
4.4. DYNAMICS IN A MTJ ELEMENT

For a period of precession, the magnetization will arrive close to the switched direction defined by the vector $\vec{M} = [M_x = -M_s, M_y \approx 0, M_z \approx 0]$. Once the magnetization vector is within this new potential well, the field pulse can be switched off. According to the torque equation, the lack of torque means the magnetization will stop precessing and remain in this new equilibrium orientation. This type of very fast magnetization switching is called precessional switching [78, 54]. In reality, due to Gilbert damping and small imperfections in the sample geometry, the magnetization is expected to “land” on the wall of the potential well, that is, to align imperfectly with the $-x$ direction, leading to a low-amplitude ringing before stabilization. The geometry of such experiments requires two potential wells, representing the two static equilibrium directions along $x$ and $-x$. This can be realized via uniaxial (shape) anisotropy along the x-axis, exchange biasing and other techniques. Finally, to perform experiments in a stroboscopic manner, one should make sure that the magnetization switches back to the positive x-direction within each period. Using two, non-symmetrical potential wells and/or a second, negative field pulse with the correct timing would solve this issue.

The advantage of precessional switching lies in its increased speed versus the traditional, damping-governed dynamics. The characteristic timescale of the Gilbert damping is given by the formula

$$\tau = \frac{2(1 + \alpha^2)}{\alpha \gamma \mu_0 (2H + M_s)},$$

where $H$ applied field and $M_s$ saturation magnetization, yielding typically over a nanosecond, corresponding to the approximate switching time possible within this technique. A material with large damping constant is favoured to make the ringing time as short as possible. In the same time, a precessional switching with an in-plane field pulse will happen on a timescale defined by the speed of the large-angle precessional motion:

$$\tau_s \simeq \frac{\pi}{\omega} = \frac{\pi}{\gamma \mu_0 \sqrt{H_{eff}(H_{eff} + M_s)}}.$$  

Here, as shown on Fig. 4.25, due to the in-plane anisotropy a strong out-of-plane demagnetization field is acting, dominating the effective field. The geometry of the dynamics is thus similar to the case of a strong magnetic field pulse applied perpendicular to the sample plane, generating an in-plane precession of the magnetization - however, practically a magnetic field pulse of this amplitude would be very hard to generate externally. For thin films of ferromagnetic layers, the magnitude of the out-of-plane demagnetization field can be estimated with the formula

$$H_{demag}(t) \simeq \frac{2K}{\mu_0} m_z(t),$$

where $K$ is the anisotropy constant of the material in the $z$-direction, yielding a field of the order of magnitude of $M_s$ or larger. The higher the material’s anisotropy the stronger the demagnetization field will be, thus, the faster the precessional switching will happen. The switching time of such a geometry can be therefore shorter than 100 ps. On the other hand, the applied field pulse has to be “tailored” to a duration...
correlated with the switching time — in the ideal case, it has to stop right in the moment when half of the precession is executed, or it has to be the odd multiple of \( \tau_s \). Finally, opposite to the damping-based mechanism, here a material with very low damping would be preferred, since after half a precession the magnetization is required to arrive as close to the switched state as possible and without any ringing that could drive it out from the final potential well.

The data recording technology would greatly benefit the exclusion of the slow Gilbert damping from the writing process. Several research groups have been reporting lately on precessional switching experiments in laboratory environment [50, 75, 76, 80, 78], with different approaches and degrees of success. The greatest challenges of the experiments seem to be the finite size effects such as spin waves, decoherence and increased apparent damping, as well as the correct timing and high quality (fast + strong) of the magnetic field pulse(s).

4.5 Conclusions

The time-resolved MOKE setup we have presented in the previous chapter proved to be successful for local measurements of the magnetization dynamics on simple and complex microstructures.

The measurements in the central region of simple Py discs indicated a fundamental (zeroth order) magnetization precession in accordance with the LLG theory.

Higher-order precessional modes (acoustical and first optical mode) were detected in antiferromagnetically coupled Py discs. The modal frequencies obtained experimentally and their dependence on the applied magnetic field showed a good overlap with the theoretical predictions in such systems.

Studying magnetization dynamics in a Py element part of an MTJ structure we have obtained evidence of distinct precessional modes at the edge of the sample (“edge modes”) coexisting with the fundamental mode. The coupling and edge demagnetization effects strongly influence the local magnetic fields resulting in frequency shifting and strong effective damping over the sample. Comparison with numerical simulations indicate that the stray field of additional magnetic layers within the MTJ have a crucial role in the formation of multiple precessional modes and the detected severe dephasing between the central and edge magnetization dynamics. We have shown that from dynamic measurements over the whole microscopic sample structure, with the aid of the LLG-theory the equilibrium domain pattern can be reconstructed. Finally, large angle precessional measurements were presented, achieved by suppressing the interlayer coupling fields. Temporal switch of the magnetization was obtained, together with strong decoherence of the precessing spins that led to very high effective damping. The concept of precessional magnetization reversal as possible fast magnetization switching technique was introduced.
Chapter 5

The magnetic field pulse profile reconstruction

As we have shown in Chapter 3, generating a fast and intense magnetic field pulse in a pump-probe geometry necessitates fine tuning of electronic and material parameters. This is especially valid for precessional switching techniques, where speed and stability are the main concern [77, 50, 78, 79, 80, 82]. However, a number of imperfections will always be present and distort the field pulse generated by the external electric pulse generator.

Most critical of these is the quality of the electrical contacts and the impedance matching between the generators output circuit, the cables and the waveguide. At the connections between high-frequency electronic components, in case of impedance mismatch, a part of the signal (the GHz electronic pulse, in our case) will be reflected. This can cause three problems of different nature. First, the amplitude of the current pulse will drop, therefore, the induced magnetic field pulse will get weaker. Neither in our experiments, nor in case of applications is a loss of power over the contacts affordable.

Second, the major reflections will be produced in the highest frequency domain, while the lower frequency components (like, the flat top of a pulse) can get through the contact without serious losses. This will result in a shift of the pulse frequency towards the lower region, i.e. broadening of the rise and fall times of the pulse. A too long fall time of the pulse can interfere with the precessional dynamics. Thus, the detected precession will be disturbed, which will complicate the extraction of damping parameter from the measured data.

Finally, in the case of bad contact and/or impedance mismatch at the exit of the waveguide, the reflected pulse will travel backward and can cause, depending on the distance it has to travel, distortion of the original pulse or afterpulsing. Both of these will deteriorate the quality of the measured data; however, in a clever geometry with an open-end stripline [50], one can make use of the total reflection for tailoring the shape of the original pulse by (partially) overlapping it with its reflection.

It is important thus, that displaying the ideal electric current pulse on an oscilloscope screen is not enough to have an accurate image over the intensity and shape of the magnetic field pulse. Directly measuring the field pulse at the position of the
micromagnetic entity under consideration, with a micrometer and picosecond resolution, would be very complicated if not impossible. Therefore, we have developed a method for back-tracing the field pulses from the pump-probe type magnetization dynamics measurements themselves.

In this chapter, the idea of the back-tracing routine will be presented, with the mathematical deduction of the formulae describing the temporal evolution of the pulse field in several geometries. We will apply these formulae to some in-plane and out-of-plane measurements that were obtained with the field-induced experimental setup, and we will extend the calculation to all-optical type measurements. By this, the idea of an ultrafast anisotropy field pulse generated by the local laser heating, gets direct proof and quantitative description. In principle, there is no direct obstacle for extending the model and calculus to more complex cases. Application of the method to other, less trivial pump-probe schemes can contribute to the development of novel type magnetic recording technologies.

5.1 Decomposing the LLG equation

The idea of the back-tracing technique is to revert the Landau-Lifshitz-Gilbert equation of magnetization motion to equations that contain as variables only the pulse field we investigate, well-defined measurement data on the precessing magnetization and derivatives/integrals of these two. Let us start from the simple vectorial form of the LLG equation,

\[
\frac{d\vec{M}(t)}{dt} = -\gamma\mu_0(\vec{M} \times \vec{H}_{eff}) + \alpha \frac{M_s}{M_s} \left( \vec{M} \times \frac{d\vec{M}(t)}{dt} \right),
\]

(5.1)

where \(\vec{H}_{eff}\) is the local effective field including the static and the pulse field, \(\vec{M}\) is the magnetization component provided by the measurement and \(M_s\) is the saturation magnetization of the magnetic layer.

Field-induced geometry

The characteristics of the geometry of our field-induced experiments are the following:

- the film plane is \(xy\);
- the DC bias field is applied in the film plane, along the direction \(x\):
  \(\vec{H}_{DC} = H_0 \cdot \hat{x}\);
- the field pulses rise in-plane, along \(y\), perpendicular to the bias field:
  \(\vec{H}_p(t) = H_p(t) \cdot \hat{y}\);
- the inplane anisotropy gives an out-of-plane demagnetizing field:
  \(\vec{H}_{demag} = -M_z \cdot \hat{z}\);
- the precessional angle is small, thus we consider a weak perturbation:
  \(M_x \simeq \text{const} \simeq M_s\).
5.1. DECOMPOSING THE LLG EQUATION

The question is thus, can we determine the original pulse field $H_p(t)$ knowing $M_y(t)$ and/or $M_z(t)$ at every moment $t$?

With the above assumptions, the LLG equation can be separated for the three directions $x, y, z$:

$$\frac{\Delta M_x}{\Delta t} = 0,$$

$$\frac{\Delta M_y}{\Delta t} = -\gamma \mu_0 (-M_x H_z + M_z H_x) + \frac{\alpha}{M_s} (-M_x \dot{M}_x - M_z \dot{M}_z), \quad (5.2)$$

$$\frac{\Delta M_z}{\Delta t} = -\gamma \mu_0 (M_x H_y - M_y H_x) + \frac{\alpha}{M_s} (M_x \dot{M}_y - M_y \dot{M}_x).$$

Substituting the components for the magnetization $M_{x,y,z}$ and field $H_{x,y,z}$ we obtain

$$\frac{\Delta M_y}{\Delta t} = -\gamma \mu_0 (M_s + H_0) M_z - \alpha \dot{M}_z, \quad (5.3)$$

$$\frac{\Delta M_z}{\Delta t} = -\gamma \mu_0 (M_t H_p(t) - M_y H_0) + \alpha \dot{M}_y.$$

We can normalize the above set of equations to $M_s$:

$$\dot{m}_y = -\gamma \mu_0 (M_s + H_0) m_z - \alpha \dot{m}_z, \quad (5.4)$$

$$\dot{m}_z = -\gamma \mu_0 (H_p(t) - M_y H_0) + \alpha \dot{m}_y,$$

where we introduced $m_i = M_i/M_s$. Note that without damping and after the field pulse is switched off ($\alpha = 0$, $H_p(t) = 0$) we have a trivial solution that is

$$M_y = \cos(\omega t),$$

$$M_z = \epsilon \cos(\omega t), \quad (5.5)$$

which, substituted into Eq. 5.4, yields $\epsilon = \sqrt{\frac{H_0}{M_s + H_0}}$ (the ellipticity of the precessional motion of the magnetization vector) and $\omega = \gamma \mu_0 \sqrt{H_0 (M_s + H_0)}$, the Landau precession frequency as known from the FMR-type studies.

By substitution and derivation, set 5.4 can be reduced to one single equation eliminating either $m_y$ or $m_z$. **In case we measure the orthogonal component $m_z$, we obtain the formula**

$$\dot{H}_p(t) = -\frac{\alpha^2 + 1}{\gamma \mu_0} \dot{m}_z - \alpha (M_s + 2H_0) \dot{m}_z - \gamma \mu_0 H_0 (M_s + H_0) m_z, \quad (5.6)$$

or, by integration,

$$H_p(t) = -\frac{\alpha^2 + 1}{\gamma \mu_0} \int m_z \, dt - \alpha (M_s + 2H_0) \int m_z \, dt - \gamma \mu_0 H_0 (M_s + H_0) \int m_z \, dt, \quad (5.7)$$

This equation is quite convenient, since in practice we are able to measure purely the perpendicular component $m_z$ of the magnetization vector; furthermore, the first derivative $\dot{m}_z$ of the dataset (as we will show later in this chapter) is reasonably smooth and the integration $\int m_z \, dt$ presents no problem. The rest of the terms are well known parameters.

In practice, $\alpha^2 \ll 1$, therefore it can be neglected. The only significant contribution of the damping will appear in the prefactor of the $m_z$. Any error in the considered
value of \( \alpha \) will result in an incorrect prefactor, which will change the contribution of this term in the final formula of the pulse field, 5.7. Thus, an oscillation in-phase with the \( m_z \) measurement will be seen, superposed on the reconstructed pulse profile, and can be easily corrected by fine-tuning the value of the damping.

In case we measure the in-plane component \( m_y \), we will have to eliminate the \( m_z \) components from the set 5.4. Here a complication arises: both the pulse field and its time derivative will appear in the final formula:

\[
H_p(t) + \frac{\alpha}{\gamma \mu_0 (M_s + H_0)} \dot{H}_p(t) = -\frac{\alpha^2 + 1}{\gamma^2 \mu_0^2 (M_s + H_0)} \ddot{m}_y + \frac{\alpha (M_s + 2H_0)}{\gamma \mu_0 (M_s + H_0)} \dot{m}_y + H_0 \cdot m_y.
\]

A quick calculus shows that, again for \( \alpha^2 \ll 1 \) and field pulses of hundreds of picoseconds rise/fall time, the very small factor in front of \( \dot{H}_p(t) \) will strongly reduce the contribution of this term, therefore we will consider it negligible. (Note that this approximation will not hold in the case of ultrafast pulses rising from laser heating, case presented later in this section.) An extra complication is caused by the second derivative of the measured dataset which shows up this time, instead of its integral. If we don’t pay enough attention to the signal-to-noise ratio of the measurement, this term can be very noisy and can make the reconstruction of the field pulse impossible. A certain amount of postprocessing (i.e. smoothing) of the data will be needed in some cases, as the one presented in the next section.

All-optical geometry

Before showing concrete examples of back-calculation (see section 5.2), let us discuss the case of the all-optical experimental configuration. As explained in section 3.4, in the all-optical experiments we use the influence of a femtosecond laser heat pulse on the equilibrium between an external field and the anisotropy field of a thin magnetic layer. Based on the similarity of the resulting dynamics, we can regard this type of pumping as creating a field pulse, that we previously called “anisotropy field pulse”.

In order to obtain the time-dependent formula for this field pulse we can start from the same decomposed form Eq. 5.2 of the LLG equation. However, it is clear that the geometry and the parameters of the all-optical type experiments are different from the field-induced case. The following points have to be emphasized thus:

- the film plane is, again, \( \pi y \);
- the DC bias field is applied almost perpendicular to the film plane, therefore it will have a considerable component along \( z \) (to cant the magnetization out of plane) and a smaller component along the easy-axis \( x \):
  \[
  \hat{H}^D C = H^D C \cdot \hat{x} + H_z^D C \cdot \hat{z};
  \]
- the field pulses rise perpendicular to the film plane, along \( z \):
  \[
  \hat{H}_p(t) = H_p(t) \cdot \hat{z};
  \]
- the in-plane anisotropy gives an out-of-plane demagnetizing field:
  \[
  \hat{H}_{dem} = -M_z \cdot \hat{z};
  \]
- therefore, the total field along axis \( z \) will be:
  \[
  H_z = H_z^D C - M_z + H_p(t);
  \]
5.1. DECOMPOSING THE LLG EQUATION

• the precessional angle is small, thus we consider a weak perturbation: 
  \[ M_x \simeq \text{const} \simeq M_s. \]

The orientation of the anisotropy field pulse can be parallel or antiparallel with the 
axis \( z \) – it depends on the way the temperature influences the anisotropy constant.
The anisotropy of the ferromagnetic layer being a complex function of crystalline, 
shape and other anisotropies, with increasing temperature its associated field \( H_{\text{ani}} \)
can either increase (\( H_p \) antiparallel to \( z \), thus shows up with negative sign; \( H_{\text{DC}}^z \)
being constant, the new equilibrium direction will be more in-plane) or decrease (\( H_p \)
parallel to \( z \), thus negative; equilibrium direction goes more out-of-plane). In the final 
formula, the result of this consideration will show up only as a global sign change.

With the above considerations, and with the normalization to the saturation mag-
netization \( M_s \), from Eq. 5.2 we arrive to the set

\[
\begin{align*}
\dot{m}_y &= \gamma \mu_0 H_p(t) + \gamma \mu_0 H_{\text{DC}}^z + \gamma \mu_0 (M_s - H_{\text{DC}}^x) m_z - \alpha \dot{m}_z, \\
\dot{m}_z &= \gamma \mu_0 H_{\text{DC}}^x m_y + \alpha \dot{m}_y,
\end{align*}
\]

which, with simple substitution and derivation, will yield an equation that contains
only the \( m_z \) component as measured data:

\[
\begin{align*}
\dot{H}_p(t) + \frac{\gamma \mu_0 H_{\text{DC}}^z}{\alpha} \cdot H_p(t) &= \frac{\alpha^2 + 1}{\alpha \gamma \mu_0} \cdot \dot{m}_z + \\
+(2 H_{\text{DC}}^z - M_s) \cdot \dot{m}_z + \frac{\gamma \mu_0 H_{\text{DC}}^z (H_{\text{DC}}^z - M_s)}{\alpha} \cdot m_z - \frac{\gamma \mu_0 H_{\text{DC}}^x H_{\text{DC}}^z}{\alpha}.
\end{align*}
\]

The above result is a first order differential equation linear in \( H_p(t) \) and \( \dot{H}_p(t) \), non-
homogeneous, of the form

\[
\dot{y} + p(t) \cdot y = r(t)
\]

with \( y \equiv H_p(t), p(t) = \text{const.} \) and \( r(t) = f(\dot{m}_z, \dot{m}_z, m_z) \). The exact analytical solution
will therefore have the final form [87]:

\[
\begin{align*}
H_p(t) &= B \cdot e^{-At} \cdot \int_{-\infty}^{t} e^{At} \dot{m}_z(t) dt + \\
&+ C \cdot e^{-At} \cdot \int_{-\infty}^{t} e^{At} \dot{m}_z(t) dt + \\
&+ D \cdot e^{-At} \cdot \int_{-\infty}^{t} e^{At} m_z(t) dt + \\
&+ E \cdot A + \\
&+ e^{-At} \cdot \text{const.}
\end{align*}
\]

The factors A to E are constant and fully determined by the sample and measurement parameters:
\[ A = \frac{\gamma \mu_0 H_{DC}^x}{\alpha}; \]
\[ B = \frac{\alpha^2 + 1}{\alpha \gamma \mu_0}; \]
\[ C = 2H_{DC}^x - M_s; \]
\[ D = \frac{\gamma \mu_0 H_{DC}^x (H_{DC}^x - M_s)}{\alpha}; \]
\[ E = -H_{DC}^z. \]

The term \(-H_{DC}^z\) indicates only the fact that the field pulse will be counteracting the external magnetic field along the z-axis and can thus be left out from the calculus. The constant in the last term of Eq. 5.12 is specific to non-homogeneous linear differential equations; however, from initial conditions \((H_p(t))_{t=0} = 0\), i.e. no field pulse before the laser pump pulse arrives) we conclude that \(\text{const.} = 0\).

A quick calculation with the parameters characteristic to our experiments \((\gamma \mu_0 = 2.321 \cdot 10^5 \text{ m/As}, H_{DC}^x = 35.7 \text{ kA/m} \text{ and } \alpha = 0.05)\) yields \(A \approx 4.1 \cdot 10^{11} \text{ s}^{-1}\). Up to a few picoseconds, this will mean that the exponential factors \(e^{-At} \approx e^{At} \approx 1\) which will considerably simplify the form of Eq. 5.12:

\[ H_p(t) \approx \frac{\alpha^2 + 1}{\alpha \gamma \mu_0} \cdot \dot{m}_z + (2H_{DC}^x - M_s) \cdot m_z + \frac{\gamma \mu_0 H_{DC}^x (H_{DC}^x - M_s)}{\alpha} \cdot \int_{-\infty}^{t} \dot{m}_z dt - \frac{\gamma \mu_0 H_{DC}^x H_{DC}^z}{\alpha} \cdot t. \]

The solution in this form much resembles the one for the perpendicular field-induced measurements’ case, Eq. 5.7; however, on the longer timescales the approximation does not hold anymore. In practice, the results of the calculations based on this simplified formula proved to be erroneous for \(t > 5 \text{ ps}\).

### 5.2 Back-tracing an external field pulse profile

After performing a vectorial time-resolved measurement, the numerical calculus of the field pulse profile involves the following steps:

- normalizing the measured data to the saturation magnetization:
  \[ m_i = M_i/M_s; \]
- calculating the precession frequency \(\omega\), either from the applied field \(H_{DC}^x\) and saturation magnetization values \(M_s\) using the formula 2.19, or from a fit to the data;
- calculus of the Gilbert damping parameter from a fit to the data:
  \[ \alpha = \frac{2}{\gamma \mu_0 \left(2H_{DC}^x + M_s\right) \cdot \tau} \approx \frac{2}{\gamma \mu_0 M_s \cdot \tau}. \]
where \( \tau \) is the characteristic decay time of the damped precessional motion and \( H_{2}^{DC} \ll M_s \);

- eventual smoothing of the measured data \( m_i \) to be able to perform the next two steps;

- differentiation of the (smoothed) data;

- second derivation of the (smoothed) data (only in the case when we measure and work with the component \( m_y \));

- integration of the dataset: \( \int_{-\infty}^{t} m_i dt \) (only in the \( m_z \) case);

- calculus of the factors that show up in the formulae 5.7 and 5.8, respectively;

- calculating the pulse shape \( H_p(t) \) with the formula 5.7 or 5.8;

- fine-tuning the value of \( \alpha \) until there is no remanent oscillation superposed on the pulse shape. In the case of an error in the calculated damping parameter, the remanent oscillation will be in-phase with the original data (\( m_z \) case) or its first derivative (\( m_y \) case).

For the accuracy of the calculus it is of utmost importance to have a measurement of purely one component of the magnetization. Any remaining inplane contribution to an out-of-plane measurement data or vice-versa will forbid using the formulae 5.8 and 5.7, respectively.

**Smoothing**

In the “recipe” for the calculus we mentioned the necessity of **smoothing the dataset** before differentiation. Excessive use of smoothing will obviously have a detrimental effect on the temporal resolution we stated above, and questions the reliability of the back-traced pulse profile. In the case of the measurements we will

![Figure 5.1: a) A raw \( M_Z \) measurement and its smoothed version (15 point adjacent-average method). b) Magnified section of the same plot. Noticeable distortion happens only for features shorter than 100 picoseconds, such as the local minimum of an oscillation – invisible in the full plot a).](image-url)
present here we used a simple adjacent averaging method over each 5 to 15 measurement points (the range depending on how good the signal-to-noise ratio of the measurement was). An example of the effect of smoothing on our measurements is shown in Fig. 5.1. Combining the smoothing rate with an original data density of 1 point per 5 ps, measurement details slower than 100 ps will be always conserved and these will carry their information over to the final curve representing the magnetic field pulse shape.

**Short pulse case**

Let us apply both of these formulae to measurement datasets we have obtained using a short (0.6 ns) field pulse on the sample with the coplanar waveguide (presented in chapter 4). The measurement itself is the “snail” measurement shown in Fig. 4.8. The original current pulse shape is shown by a 12.5 GHz oscilloscope in Fig. 5.2; the results of the back-tracing calculus (applied both to the $m_y$ and $m_z$ data) are overlapped to the current pulse in the figure. The two calculated curves overlap to a level of indistinguishability. Since these two curves result from the same measurement configuration but different formulae, their accordance indicates the correctness of our back-tracing procedure.

![Figure 5.2: "Short" (0.6 ns) magnetic field pulse generated by our waveguide, reconstructed from an $m_z$ and an $m_y$ measurement. The two back-traced curves almost perfectly overlap, indicating a high level of precision in the calculus. The original current pulse coupled to an ideal 50-ohm impedance, as shown by a 12.5 GHz oscilloscope, is overlapped with the calculus. The plotted vertical scale refers to the calculated field pulse amplitudes; the scale for the oscilloscope image is arbitrary.](image)

At a first glance it seems that the magnetic field pulse, reconstructed from the precessional measurements, is shorter than the electric field pulse used to induce it, which would be impossible to realize in our geometry. However, note that the flat plateau of the $\approx$500 ps long pulse is gone, the rise and fall times being much longer in the magnetic case. This might be related to the peculiarity mentioned in the
5.2. BACK-TRACING AN EXTERNAL FIELD PULSE PROFILE

introduction: the highest frequency components are harder to send through the non-ideal contacts, therefore the rise/fall times broaden considerably. There is no way to compare the amplitude of the two pulses, one being electric current in a 50 ohm probe and the other one, magnetic field pulse as plotted on the vertical scale of Fig. 5.2. Furthermore, the base of the two pulses are nearly identically wide. However, the much smaller value of the FWHM of the field pulse we cannot fully explain.

More important is the lack of strong afterpulsing and/or reflected pulses travelling backward that would disturb the measurement. The amplitude can be compared to the simple calculations based on the formula 3.4 that assumes a perfect coupling of the pulses from the generator to the waveguide, i.e. no losses at the connections, and a low DC resistance of the waveguide structure. The deviation indicates a loss of peak power of approximately 20% due to the non-ideal coupling.

Long pulse case

The similarity between the current pulse coupled to an ideal 50-ohm impedance, shown by an oscilloscope, and the back-traced magnetic field pulse shape in the real sample we studied, is even more striking in the case of longer pulses. The result of the calculus in the case of a 1.5 ns pulse is shown in Fig. 5.3. The dataset used for the back-tracing was, in this case, the perpendicular \( m_z \) component of the measurement shown in chapter 4, Fig. 4.9. For the ease of comparison, the calculated curve is overlapped to the original oscilloscope image; however, once again, the vertical scale is valid for the calculated magnetic field pulse only, the scale of the current pulse

![Figure 5.3: "Long" (1.5 ns) magnetic field pulse reconstructed from an out-of-plane \( m_z \) measurement dataset, overlapped with an image of an oscilloscope display showing the electric pulse, on the same timescale. The scale of the current pulse \( i_p \) is arbitrary, chosen to graphically match the calculated amplitude. The arrows point to fine details (negative baseline, pulse plateau features, afterpulsing) that can be found on both curves.](image)
(i_p) being arbitrary. Comparing the result with the predicted pulse amplitude, the coupling loss in the long pulses case seems to be even less than 20%.

As explained before, the higher frequency components are more suppressed by the non-ideal contacts, therefore the “corners” of the pulse shown by the oscilloscope screen are smeared out in the back-traced field pulse. This leads, again, to an effective broadening of the rise and fall times from 120 ps to approximately 300 ps each.

Interestingly, also some of the small details of the original pulse (e.g. plateau unevenness) can be found back in the calculated field pulse shape, making the overlap of the two curves surprisingly accurate. The smallest features resolved indicate a precision of the back-tracing method of approximately 100 ps.

Field pulses in the MTJ geometry

Finally, let us review the case of the measurements on the MTJ sample presented in section 4.3. The geometry of the experiments were similar to the previous case; however, the lateral sizes are smaller and the magnetic field pulse is induced by a "wire", not a true waveguide. From the quality of the experiments presented in chapter 4, we know that a reasonably fast and intense field pulse was induced at the sample position. The pulse generator was set to emit the same 600 ps "short" pulses like in the case presented above (the electric pulse is thus the one shown in Fig. 5.2 b). 

The results of the back-tracing, based on one in-plane (m_y) and the out-of-plane (m_z) component of the measurement shown in chapter 4, Fig. 4.14 b), however, do not look as good as for the case of the coplanar waveguide geometry. The two curves calculated with the formulae 5.7 (for m_z) and 5.8 (for m_y) are plotted in Fig. 5.4. One can immediately notice, that the two pulse profiles do not fully overlap. Although they both show a field pulse of approximately 600 ps with a fast rise time and a well-defined maximum value, the pulse calculated from the m_z data has a very

![Figure 5.4](image)

Figure 5.4: "Short" (600 ps) magnetic field pulse, reconstructed from an out-of-plane (M_z) and an in-plane (M_y) measurement on the 9 × 13 μm² MTJ sample.
long decay time (> 600 ps), while the one resulting from the $m_y$ data shows a sharp falling edge and strong afterpulsing. There are two main reasons that can cause this discrepancy, both related to the derivation of the formulae for $H_p(t)$:

- the “weak perturbation” ($M_x \approx \text{const} \approx M_s$) approximation is not valid anymore; the measurements show a deviation of the magnetization with an angle of 23.5° to the axis $x$ (this corresponds to a change of the $m_x$ magnetization component with 10% versus the 1% in the previous sample’s case);
- a weak uniaxial anisotropy of the ferromagnetic layer along $y$-axis was noticed, not considered in the derivation of the two equations.

The amplitude of the field pulse turns out to be 1.2 kA/m. Due to the lower DC resistance (50 Ω versus 112 Ω) and thinner current line (30 µm versus 50 µm), this value is almost 4 times larger than in the case of the sample using the true waveguide. This indicates that, by reducing the dimensions and taking care of the electrical properties of the structure, the efficiency of the experimental setup can be dramatically increased. In the same time, this value is still below the calculated 2.1 kA/m (based on the formula 3.4), which is in part due to imperfect contacts. Moreover, the lack of a true waveguide in the MTJ case leads to impedance mismatch and increased coupling reflections in the gigahertz frequency range.

5.3 Back-tracing the “anisotropy field pulse”

An all-optical polar measurement on a 7 nm polycrystalline Ni thin film (Si/SiO$_2$ substrate) is shown in Fig. 5.6. Laser pulses of $\approx$100 fs are used, and a bias field of 200 kA/m is applied at an angle of 35° with respect to the surface normal. The field induces a canted orientation of the magnetization vector $\vec{M}$ with an angle $\Theta$ compared to the in-plane orientation. In the same time, a sufficient in-plane field for saturating the magnetization is obtained as well. Due to the applied bias field that is considerably larger than in the presented field-induced measurements, the precession frequency is also higher ($\approx$10 GHz). We will apply the back-calculation procedure adapted to the all-optical geometry (formula 5.12) on this measurement and obtain the “anisotropy field pulse” that pushes the magnetization into the precessional motion. In order to do so, we have to remember that there are more effects than just the precessional motion of the magnetization vector (further we will call this “orientational effect”). Let us thus discuss the full $\vec{M}(t)$ behaviour.

At any moment $t$, the magnetization component along the z-axis (the out-of-plane component) can be written as

$$M_z(t) = |\vec{M}(t)| \sin \Theta(t),$$

(5.15)

where $|\vec{M}(t)|$ is the amplitude of the magnetization vector, that is, the saturation magnetization $M_s(t)$ (see also Fig. 5.5). In contrast with the experiments based on external pulse fields, here the magnetization amplitude also varies in time, via the temperature change (laser heating). Therefore, any change in the $M_z$ component we measure will have two contributions: the demagnetization/remagnetization related to
Figure 5.5: Sketch of laser-induced fast magnetization dynamics. In equilibrium, the magnetization $M$ is canted out of the sample plane $xy$ by a constant external field $H^{DC}$ at an angle $\theta$. At $t=0$, the laser pulse heats up the sample in a small spot - the anisotropy field $H_{ani}(t)$ is suddenly changed ($t=0-0.2$ ps) and the equilibrium direction for $M$ changes. A large torque will force the magnetization $M'$ into a very fast precessional motion along a large ellipse ($t=0.2-1$ ps), around point $C$ (the intersection of the new equilibrium direction with the plane $yz$). The laser pulse vanishes and the temperature begins to drop, the equilibrium direction drifts back to its original position while the precession of the magnetization $M''$ is still on ($t=1-100$ ps). The damping will finally align the magnetization ($t=100-1000$ ps) along the old equilibrium direction defined by $H_{z}^{DC}$ and $H_{ani}(t)$. Depending on the timescales of the damping and the heat diffusion, $M$ during its precession can arrive to directions with $\theta'<\theta$. Simultaneously with the LLG-type precessional motion, the amplitude of the magnetization $|M|$ changes: it suddenly drops due to the heat pulse and then restores to the full value with the heat diffusing out of the ferromagnetic sample ($|M| \rightarrow |M'| \rightarrow |M''| \rightarrow |M|$).
5.3. BACK-TRACING THE “ANISOTROPY FIELD PULSE”

Figure 5.6: All-optical measurement on Ni[7nm]/SiO$_{2}$/Si sample (gray line marked with circles). The thick full curve (FL) represents the fit on long timescale (0.2-450 ps) with formula 5.23, neglecting the third (exponential) term. The dashed curve FS+E is the remagnetization due to loss of heat; it is obtained from the short timescale fit (0.2-1 ps) with the non-LLG terms and extrapolating up to 600 ps. The top, star-marked black line is the difference between the measurement and the remagnetization curve, thus it shows the pure orientational (LLG-type) effect and will be used to back-calculate the anisotropy field pulse.

the temperature change, and the precession around the current equilibrium direction (described by the LLG equation):

$$\Delta M_z(t) = \Delta \hat{M}(t) \sin \Theta(0) + |\hat{M}(0)| \Delta \sin \Theta(t).$$

(5.16)

Here we note with $\Theta(0)$ the initial magnetization angle, identical to the equilibrium magnetization angle before the pump pulse arrives, and $|\hat{M}(0)| = M_s$, the saturation magnetization at the base temperature of the experiment. The separation of the LLG and non-LLG components of the magnetization dynamics is possible considering that on a sub-picosecond timescale the LLG dynamics are not yet involved in the measured signal. If we can mathematically describe the remagnetization effect, we can extrapolate it to the long timescale and eliminate it from the measurement, obtaining the “orientational effect” (the pure LLG dynamics).

Heat transfer and heat diffusion; remagnetization processes

After the ultrafast demagnetization (the signal drop on the 100 fs timescale as shown in Fig. 5.6), the amplitude of the magnetization will start to recover due to the exchange of heat between the electrons and the crystal lattice (spin temperature drops), effect described by an exponential law. The electron-phonon relaxation time is typically around 400 fs, hence it is not influencing the GHz magnetization precession. As a
next step, the metal lattice will transfer the heat to the substrate, reaching a temper-

ature equilibrium with this. The result will be a gaussian temperature profile building
up in the substrate, having its top temperature at the ferromagnet/substrate inter-

face. The metal layer’s temperature will therefore gradually drop following a \( 1/\sqrt{t} \)

law. Accordingly, we can write a phenomenological formula for the remagnetization

term as

\[
\Delta|\vec{M}(t)| = A_1 \cdot e^{-t/t_{cp}} + \frac{A_2}{\sqrt{t + t_0}},
\]

(5.17)

where \( A_1, A_2, t_0, t_{cp} \) are well-defined parameters (\( t_{cp} \) representing the electron-phonon

relaxation time, while \( t_0 \) defines the width of the “instantaneously” heated region

inside the substrate at the moment \( t = 0 \)). Note that, in Eq. 5.16, \( \sin \Theta_o = \frac{M_z(t=0)}{M_s} = m_z \) which is the normalized out-of-plane magnetization component at \( t = 0 \) (the initial equilibrium case).

In the above treatment, we considered that the heat is instantaneously transmitted

from the metal lattice to the interface region of the substrate. In practice, heat

transmission strongly depends on the substrate properties and the insulating material

in between (e.g. the thin oxide on a Si substrate), and the physics behind the effect

deserves a detour from our calculus. The transmitted heat and the temperature drop

of the ferromagnetic layer in time can be calculated from the two equations

\[
\frac{dQ}{dt} = \frac{k_i A}{L} \Delta T(t),
\]

(5.18)

\[
Q(t) = C_m(T_m(t) - T_m(0)) = C_s(T_s(t) - T_s(0)).
\]

(5.19)

Here \( k_i \) represents the heat conductivity of the insulating layer (which can be the

substrate itself), \( A \) is the laser spot size, \( L \) is the thickness over which the gaussian

temperature profile in the substrate is considered flat and \( \Delta T \) is the temperature

difference between the hot and cold sources (the laser-heated metal and the substrate

at base temperature) at any moment \( t \). \( C_m, C_s \) are the heat capacities of the metal

and substrate in the considered volume, while \( T_m, T_s \) are their temperature at the

moment \( t \).

The analytical solution of the above two equations can be obtained within two

limits. For \( t = 0 \) there’s no heat transferred yet (\( Q = 0 \)); for \( t = \infty \) the ferromagnet

and the substrate are in thermal equilibrium (\( \Delta T = 0 \)). The result will be a long
timescale exponential drop of the temperature difference between the two layers:

\[
\Delta T(t) = \Delta T_0 \cdot \exp \left[ -t \frac{k_i A C_m + C_s}{L C_m C_s} \right],
\]

(5.20)

where \( \Delta T_0 \) represents the initial temperature difference between the laser heated

region of the ferromagnetic layer and the underlying, cold substrate.

We will consider two limits for the real sample-substrate heat transfer, illustrated
also in Fig. 5.7:

1. Easy heat flow from the metallic layer into the substrate. The heat will be

quickly driven directly into the large heat-capacity substrate, leading to a con-

siderable temperature drop of the metal before a gaussian temperature

gradient would build up inside the substrate. The metallic layer’s tem-

perature behaviour (and the remagnetization process \( \Delta|\vec{M}(t)| \)) will be governed
thus by the exponential process 5.20 described above, with the parameters \( k_i, C_s \) of the substrate and \( C_m \) of the ferromagnetic layer.

2. Metal-substrate heat transfer blocked, e.g. by a thick insulating layer (oxide, nitride...) in between. The heat conductivity \( k_i \) in Eq. 5.20 is in this case parameter of the insulating layer, usually two orders of magnitude smaller than the Si heat conductivity, therefore the exponential decay of the hot source’s temperature will be too slow to compete with the heat diffusion inside the insulator/substrate. The low-\( k_i \) insulator will be heated up without any strong temperature drop of the metallic layer, and the further heat transfer will be governed by the Boltzmann-type heat diffusion within the thick insulating layer and the corresponding \( 1/\sqrt{t} \) law. The heat transfer from the insulator to the substrate will not play a role anymore — we measure the temperature drop of the metallic layer only.

In case of our samples, a thin oxide layer exists at the metal-substrate interface, therefore the remagnetization is expected to follow a combination of the exponential and gaussian decay functions.

The complete laser-induced dynamics

Let us come back now to the treatment of the full magnetization dynamics. The remagnetization formula 5.17 should be thus extended with a second exponential function, with a characteristic timescale \( t_h \) much longer than the one of the electron-phonon relaxation time, \( t_{ep} \):

\[
\Delta |\vec{M}(t)| = A_1 \cdot e^{-t/t_{ep}} + \frac{A_2}{\sqrt{t+t_0}} + A_3 \cdot e^{-t/t_h}. \tag{5.21}
\]

To arrive at the complete formula describing the dynamics of the \( z \)-component of the magnetization, we will add the orientational terms to Eq. 5.21. These terms reflect on one hand the dynamics of the equilibrium direction of the magnetization — its drift back towards the undisturbed orientation, due to cooling down. On the other hand, the damped precessional motion of the magnetization will also manifest itself, in form of an oscillation. The former is the result of the equilibrium between the temperature-dependent anisotropy and the external bias field thus it accurately follows the drop of the temperature. Its form is similar to the second and third terms in Eq. 5.21 but with different amplitudes \( A'_2 \) and \( A'_3 \). The latter, precessional term
is a sine function with an exponential decay. The orientational (or, LLG) component \( \Delta \sin \Theta(t) \) from Eq. 5.16 is thus described by the formula

\[
\Delta \sin \Theta(t) = A'_2 \sqrt{\frac{t}{t + t_0}} + A'_3 \cdot e^{-\frac{t}{t_h}} + A_4 \cdot e^{-\frac{t}{t_d}} \cdot \sin(\omega t + \phi_0),
\]

(5.22)

where \( \omega \) represents the precession frequency and \( t_d \) is the characteristic decay time of the precession. Evidently, the time constants \( t_0 \) and \( t_h \) in the above formula are the same as in Eq. 5.21. This allows us to contract the final formula for the full magnetization dynamics to

\[
\Delta M_z(t) = A_1 \cdot e^{-\frac{t}{t_{ep}}} + \frac{A_2 + A'_2}{\sqrt{t + t_0}} + (A_3 + A'_3) \cdot e^{-\frac{t}{t_h}} + A_4 \cdot e^{-\frac{t}{t_d}} \cdot \sin(\omega t + \phi_0).
\]

(5.23)

As detailed above, depending on the heat absorbing properties of the layers under the ferromagnet, we can have cases when either the second (square-root) or the third (exponential) term can be ignored. Satisfactory results were obtained in fitting the data measured on a Si/SiO\(_2\)[5nm]/Ni[7nm] sample ignoring the exponential term; the fit results are shown on Fig. 5.6. The fitting was done on almost the full timescale of the measurement, however the large number of fitting parameters require that special care is taken to make them arrive to values that can carry a physical meaning. The electron-phonon relaxation time \( t_{ep} \), e.g., is known from literature [4, 84] to be around 500 fs, while the precessional period is in the range of hundreds of picoseconds.

We should not forget, that the goal of the whole model and fitting is to be able to separate the remagnetization effects, related to the amplitude change of the magnetization vector, from the orientational effects. These will be used then to back-calculate the “anisotropy field pulse” that started the whole LLG-type magnetization dynamics.

The fit with the full dynamics formula 5.23, although permitting us obtaining many of the parameters, does not allow for full separation between the remagnetization and the LLG dynamics. In detail, the amplitudes \( A_2 + A'_2 \) and \( A_3 + A'_3 \) will still contain the remagnetization via \( A_2 \) and \( A_3 \), respectively. A second fitting procedure, on a short timescale, will solve this problem as will be shown in the following:

In the first few picoseconds after the laser pulse, the magnetization vector starts to precess around the new equilibrium direction. However, this precession will start as a motion of the \( \vec{M} \) almost completely parallel with the sample plane, thus, it will not contribute to the change in the \( M_z \) component (yielding \( A_4 = 0 \) in Eq. 5.23). Similarly, the flip of the equilibrium magnetization direction will not yet be represented in the real \( M_z \) magnetization component that is measured, thus \( A'_2 \) and \( A'_3 \) will be zero. On the timescale of 0 to 1.5 ps therefore, we can consider that our measurements only represent the demagnetization and remagnetization. This allows us to use the formula 5.21 with the parameters \( A_1, t_0, t_{ep}, t_h \) fixed at the values already obtained to fit the picosecond measurement data and obtain the remagnetization amplitudes \( A_2, A_3 \). We can use these parameters to extrapolate \( \Delta |\vec{M}(t)| \) to the full timescale of the measurement and obtain the pure remagnetization curve. We complete this curve with the ultrafast demagnetization (0-0.2 ps), to obtain the non-LLG component of the magnetization dynamics. This operation has been performed on the measurement shown on Fig. 5.6 and it is plotted with dashed line in the same figure.
5.3. **BACK-TRACING THE “ANISOTROPY FIELD PULSE”**

Subtracting the demagnetization/remagnetization $\Delta |\vec{M}(t)|$ from the original measurement $\Delta M_z(t)$, we obtain the pure orientational effect $\Delta \sin \Theta(t)$ - the magnetization precession around the drifting equilibrium direction. The result is also included in Fig. 5.6, dotted line. This is used then as an input $M_z$ in the formula 5.14 for deriving the effective pulse profile $H_p(t)$.

Thus, the “recipe” for the back-calculation in the all-optical case is in short:

- fit the measurement on its full timescale with the formula 5.23 to obtain the involved parameters;
- fit the measurement on the very short (0.2 to 1.1ps) timescale using the already obtained parameters, but considering that the LLG-dynamics are not yet in action (the ultrafast demagnetization between 0 and 0.2 ps has to be excluded from both fits);
- extrapolate the obtained remagnetization curve to the full time scale of the measurement and complete it on the sub-picosecond timescale with the demagnetization;
- subtract the demagnetization/remagnetization from the original measurement, to obtain the pure orientational effect governed by the LLG equation;
- calculate the precession frequency $\omega$ and the Gilbert damping $\alpha$ from the oscillation parameters;
- calculate the first and second derivative of the dataset $\dot{m}_z$, $\ddot{m}_z$ (smoothing on the long timescale might be needed) as well as the factors A, B, C, D, E defined by formulae 5.13;
- apply the exact formula 5.12 for computing the anisotropy field pulse, $H_p(t)$.

When we subtract the demagnetization/remagnetization curve from the measurement, the early region (the first picosecond) will appear as completely flat, see Fig. 5.6. This is a consequence of disregarding the out-of-plane component of the precession that just starts, as explained above. The result is equivalent to shifting the “zero delay” i.e. the arrival of the laser pump pulse, resulting in an artificial compression (narrowing) of the “anisotropy field pulse” which in reality does not happen. Since the precessional data on the longer timescale does not change with this compression, the torque on the magnetization needed to produce the full precession has to be “conserved” through the calculus. To fulfill this requirement, parallel with the artificial compression of the pulse, an increase of the delta-peak’s amplitude will be happening, hence the integral will be conserved. Taking the timescale for the second fit as short as possible will minimize the distortion of the final result, as shown in an example in Fig. 5.8.

A back-calculation result on a longer timescale is shown in Fig. 5.9; note the very narrow delta-like peak (FWHM = 2 ps) superposed on a step-like background with a long fall time.

On the short (0-5 ps) timescale, the pulse back-traced from the pure LLG-dynamics (plotted in Fig. 5.6) shows a very fast rising peak of a high amplitude. This is orders of magnitude higher than the field pulses we were able to produce with the waveguide
Figure 5.9: The resulting “anisotropy field pulse” profile for the all-optical measurement shown before. The full line represents the calculus based on fitting parameters, while the line marked with circles shows the result that relies on the real measurement data. Although very noisy, on the long timescale the latter is clearly scattered around the former.
5.4. CONCLUSIONS

We have presented a back-calculation method for magnetic field pulses that drive magnetization precession in ferromagnetic microstructures. Applying the method to field pulses of 0.6 and 1.5 ns duration generated by induction in a waveguide structure, the results are consistent and show features on the level of 100 ps. Comparison with the original electric pulses screened by an oscilloscope indicate a good accordance.

Applying the back-calculation to the field pulses rising in the MTJ structure yielded satisfactory results. A strong afterpulsing and dependence on the measurement conditions was noted. This can be attributed to the lack of a true waveguide in the structure, an uniaxial magnetic anisotropy and the slight but nonzero variation of the \( M_x \) magnetization component.

We have extended the method to the calculation of the anisotropy field pulse rising in all-optical measurement geometries. We have observed that the derived pulse shape depends slightly on the exact procedure used to separate the amplitude and orientation of the transient magnetization. However, independent of those details, a delta-like pulse always appears, with a value for its integrated area that seems to be conserved and a FWHM of \( \sim 2 \) ps. The question arises, what is the physics behind this “special” pulse width of 2 ps? We propose an explanation based on the nature of the temperature-dependent anisotropy constant \( K(T) \) involved in the
generation of the pulse. Being a parameter of the crystalline lattice, \( K(T) \) and the anisotropy field pulse profile itself follows the timescale of the \textit{lattice temperature variation}; in other words, \( T \) refers to the lattice temperature. Both the heating and most of the cooling of the metal lattice happens on the timescale of a picosecond, and it is also known, that the magnetic order in ferromagnets adapts to the lattice and electron temperature faster than 1 ps \[36\]. Thus, it is indeed possible that the magnetic anisotropy is induced within this timescale.

We would like to stress that both the pulse-compression artifact and the whole process of obtaining the pure orientational effect could be avoided by using a \textit{vectorial measurement setup}, similar to the one implemented in the field-induced experiments. Probing the dynamics of the out-of-plane and the in-plane magnetization components simultaneously, the orientational effects on \( M_z \) and \( M_y \) would be 90 degrees phase shifted to each other, with identical demagnetization/remagnetization curves, allowing for separation between the effects. Technically however, this setup presents some problems: the incident and reflected probe beam cone, focused by the strong objective, should be exactly perpendicular to the sample surface. This would make the separation of the pump and probe beams extremely difficult (no pump light is allowed to reach the detector). One solution would be, suggested here for further research, a two-colour experiment: to use a frequency doubler unit for the pump beam, and a dense red filter in front of the detector to filter the pump light out.
Chapter 6

Studying damping by spin pumping in FM/NM structures

A number of extrinsic effects contributing to the damping of the magnetization precession was presented in the previous chapters. Manipulating the effective damping via these effects is of high relevance for future applications, e.g. with respect to improving the switching speed of the magnetization of microstructures. A new possibility of increasing the effective damping, proposed recently by Bauer et al., will be investigated in this chapter. The effect, called “spin pumping”, results in loss of spin momentum (flow of spin current) of a precessional ferromagnetic system to an adjacent non-magnetic layer. The original paper shows that the flow of spin current leads to an additional damping of the precession, and estimates the order of magnitude of such an effect.

First of all, we will introduce the notion of “spin torque”. This will be a key element of the spin pumping mechanism, to detail in the next section. At this point we will propose an experimental method to detect any additional extrinsic damping due to the loss of spin momentum. Further, the correlation of the damping and the morphology of ferromagnetic thin layers will be discussed, illustrated with experimental data. In the second part of this chapter we will present our results obtained on various ferromagnetic/non-magnetic metallic systems, these measurements being performed with the goal of detecting the spin pumping effect. Finally, a comparison with recent data from the literature will be given.

6.1 Spin torque

As we introduced it in the second chapter, the Landau-Lifshitz equation predicts that a net spin of electrons aligned in a nonzero angle with a magnetic field will have a precessional motion, due to the torque of the field acting on it.

Let us now consider a group of electrons with a zero net spin passing through a ferromagnetic/metallic/ferromagnetic stack of thin layers (in form of an electric
Figure 6.1: Metallic structure consisting of two ferromagnets FM1, FM2 with magnetizations $S_1, S_2$ oriented in a relative angle $\theta$ separated by a normal metal NM. An ensemble of electrons enters the first ferromagnet with zero net spin $S_0$ and gets “spin-polarized” along the direction of $S_1$. Thick arrow = majority spins, thin arrow = minority spins. While passing through the second magnetic layer, a net spin moment is transferred to the magnetization of FM2. See text for details.

The physics happening around the spin-polarized current that travels through the rest of the structure was studied in the ballistic regime by Slonczewski [90] and Berger [91] in 1995-1996. First of all, he considered that the thickness of the non-magnetic metallic spacer $d \ll \lambda_s$, where $\lambda_s$ denotes the spin diffusion length. This means, that the spin polarization of the electron flux is (at least partially) conserved through the spacer and a net spin moment $S$ will be entering FM2, oriented in a non-zero angle $\theta$ versus the magnetization of the layer. This can be interpreted as...
6.1. SPIN TORQUE

a coherent superposition of spins with an unequal distribution between the spin-up and spin-down orientations, to be called majority and minority spins, as sketched in Fig. 6.1. In a system of coordinates \( \hat{x}\hat{y}\hat{z} \) where \( \hat{z} \) corresponds to the magnetization direction of FM2, the initial state of the polarized spins entering FM2 can be described by the spinor

\[
\psi_0 = (c_\uparrow, c_\downarrow) = \left( \cos \frac{\theta}{2}, \sin \frac{\theta}{2} \right).
\]

The \( \hat{z} \) will be thus the quantization direction of the spin moment. What happens to the spins in FM2 is determined by the Coulomb and Stoner exchange potential of the magnet \( V_{11}(\xi) \). This potential is dependent on the position \( \xi \) along the direction \( \hat{x} \) orthogonal to the surface of FM2 and, in the same time, the potential looks different for the spin-up and spin-down channels. Slonczewski, using the WKB approximation (in short, the spin-up and spin-down energy bands are considered as parabolic bands), deduces the spinor function for the electron spin at any position within FM2:

\[
\psi(\xi) = \left( \cos \frac{\theta}{2}, e^{i(k_\uparrow - k_\downarrow)\xi} \sin \frac{\theta}{2} \right)
\]

(6.2)

where \( k_\uparrow \neq k_\downarrow \) are the components along \( \hat{x} \) for the spin-up and spin-down wave vectors and \( \xi \) takes any value between zero and the full width of FM2.

This form of the resulting spinor function indicates that the spin, while passing through FM2, is precessing around the \( \mathbf{z} \)-axis, with the position-dependent phase \( \phi = (k_\uparrow - k_\downarrow)\xi \). The phase of the entering spins is, of course, zero (\( \xi = 0 \)). However, in a realistic case, due to fluctuations in the thickness \( w \) of FM2 and the random nature of the wave vectors of the spins, the exit phase \( \phi(w) \) may be assumed random, thus the spinor for the exiting ensemble of spins will be

\[
\psi(w) = \left( \cos \frac{\theta}{2}, 0 \right).
\]

This results in a net spin momentum \( \mathbf{S} \) reoriented completely along the magnetization direction of FM2, only possible if a torque was acting on it by FM2. In Fig. 6.1, this reorientation is indicated as a counterclockwise rotation of the vector \( \mathbf{S} \) representing the net exiting spin. Note, that during the precessional motion, the \( z \)-components \( s_z \) of the individual spin momenta \( \mathbf{s} \) are conserved, indicating the loss of the \( x \)-component of the net momentum \( S_x \). Considering \( hS_x \) as the total spin momentum per unit area of the ferromagnet FM2, the principle of conservation of angular momentum says that the momentum lost by the ensemble of spins travelling through FM2 will be taken over by the ferromagnet itself, following the formula

\[
\Delta S_z = S_{\text{final}} - S_{\text{initial}}.
\]

Since only the \( S_x \) component is lost, the momentum gained by FM2 will also be along the \( x \)-direction: \( \Delta S_x = \left( \sin \frac{\theta}{2}, 0, 0 \right) \), resulting in a torque on \( S_x \) acting in the opposite direction compared to the torque on travelling spins, \( \mathbf{S} \). This will manifest as a clockwise rotation of the magnetization of FM2, as indicated on Fig. 6.1. The process can be understood also via the principle of action and reaction: FM2 rotates the passing spin via a torque (action), but the latter will also exert a torque of opposite sign and equal amplitude on the magnetization of FM2.

In case of a half-metallic device, where the spin polarization by FM1 is 100%, the spins entering FM2 will all have identical spin orientation (corresponding to the
magnetization of FM1. The transmission for these electrons is \( \cos^2 \frac{\theta}{2} \); if using the minority-majority picture, the minority spins (spin-down on Fig. 6.1) are all reflected back into the normal metal. Interesting to note that these electrons re-entering the first ferromagnetic layer, suffer a rotation in their spin orientation and in the same time give rise to a torque on the magnetization of FM1, making it rotate in the same clockwise direction as FM2. Another interesting problem to study is reversing the current, that is, injecting the electrons from the opposite side into the junction. This will result in torques of opposite direction; the torque is thus an antisymmetric function of the electron flux.

However, the main goal of the Slonczewski paper was to calculate the exerted spin torque, respectively the exchanged spin momentum between the injected electrons and the ferromagnet, using realistic ferromagnets with polarization \( P < 1 \). In this case, not all the electrons entering FM2 will be oriented in the same angle \( \theta \) to \( \mathbf{S}_2 \). This complicates the calculus considerably; with extra approximations (identical ferromagnets; \( V_{NM} = V_{[FM2]} \)), the torque exerted on FM2 is calculated to be

\[
\dot{\mathbf{S}}_2 = \frac{g I_e}{e} \hat{s}_2 \times (\hat{s}_1 \times \hat{s}_2),
\]

where \( I_e/e \) represents the flux of electrons, \( \hat{s}_{1,2} \) are the unit vectors for \( \mathbf{S}_{1,2} \) and \( g \) is a scalar function of the polarization \( P \) and the angle \( \theta \) between \( \mathbf{S}_{1,2} \).

If we consider the existence of an effective DC magnetic field (external, anisotropy or other fields) \( \mathbf{H} \) through the FM/M/FM structure, the Landau-Lifshitz-Gilbert equation can be written for the magnetization of FM2 and extended with the spin torque term 6.3:

\[
\dot{\mathbf{S}}_2 = \hat{s}_2 \times \left( \gamma \mathbf{H} - \alpha \dot{\mathbf{S}}_2 + \frac{g I_e}{e} \hat{s}_1 \times \hat{s}_2 \right). \tag{6.4}
\]

Here we considered the absolute permeability \( \mu_0 = 1 \). One can recognize the last, spin-torque term as a kind of “damping” of \( \mathbf{S}_2 \) towards aligning with \( \mathbf{S}_1 \). By this new suggested dynamic phenomenon, a broad range of research was ignited, with possible applications in the future. As examples we can mention spin-current induced magnetization switching in layered structures [92, 93], DC-current induced gigahertz precession, domain wall motion controlled by current pulses [94] and manipulating precessional damping by spin pumping [97]. The latter will be discussed in detail in the next sections, together with our experimental results on this subject.

### 6.2 Spin pumping mechanism in FM/NM layers

**The mechanism**

We have seen that a spin-polarized current injected into a ferromagnetic thin layer can exert a torque on the magnetization of this, contributing to its LLG-type dynamic behaviour. In 2002, Bauer et al. proposed [97], that the opposite of the spin torque effect can happen in similar conditions: a moving magnetization vector loses angular momentum by emitting a spin current into an adjacent normal metal (a spin sink). In case of a magnetization precessing under the action of a torque, “losing angular momentum” means that the precession is attenuated, i.e. an additional damping occurs. The idea was born from a number of experiments on Cu/Co and Pt/Co
layers reporting on a Gilbert damping considerably larger than the bulk value for Co [93, 95, 96].

An intuitive picture of the spin pumping effect can be given using a formalism similar to the spin torque described in the previous section. Let us assume a ferromagnetic thin layer FM of thickness $d$ magnetized uniformly along the z-axis (the vector $\mathbf{S}(t)$ representing its net magnetic moment per unit volume), and an adjacent nonmagnetic metallic layer NM of thickness $d_{NM}$ as shown in Fig. 6.2. We can interpret the net moment $\mathbf{S}$ in equilibrium as a number of spins $s_\uparrow$ all oriented parallel with the z-axis, described by the spinor function $\psi = (1, 0)$. Using an arbitrary method, at $t=0$ we apply an external torque $\mathbf{T}$ on $\mathbf{S}(t)$ forcing it into a precessional motion around the z-axis. Evidently, a net angular momentum is pumped into the FM/NM system this way. Assuming zero Gilbert damping, at any moment $t>0$ the vector $\mathbf{S}(t)$ will be oriented at an angle $\theta$ against the z-axis. The canted $\mathbf{S}$ can be interpreted as a superposition of majority/minority spins $s_\uparrow s_\downarrow$ in FM, oriented parallel/antiparallel with the z-axis. Since the number of majority spins $s_\uparrow$ did not change by applying the torque, the absorption of the external momentum by $\mathbf{S}$ is equivalent to a volume injection of a spin current of minority spins $s_\downarrow$. If this spin current is allowed to leak into the normal metal (see second/third frame on Fig. 6.2), after some time $\Delta t$ the minority spins $s_\downarrow$ vanish from FM and $\mathbf{S}$ realigns with the z-axis, reaching its original equilibrium state. Energetically, we can understand this “leaking” as the minority spins having a higher energy in FM than the Fermi level in the NM, while the majority ones exactly the opposite. Therefore, a FM to NM transfer of the minority spins is favoured through a transparent interface.

Macroscopically, the loss of momentum to NM will manifest itself as losing the torque on $\mathbf{S}$, i.e. damping of its precessional motion, characterized by a Gilbert-type dimensionless damping constant $\alpha'$. Since a transfer of momentum to an adjacent nonmagnetic layer is involved, this damping will be of a non-local type. Considering also the conventional, local Gilbert damping $\alpha_0$ acting within the ferromagnetic layer as it happens in a real sample, the effective damping of the precession of $\mathbf{S}$ will be given as a sum of these two:

$$\alpha = \alpha_0 + \alpha'.$$

Bringing thus a non-magnetic metal in contact with the ferromagnetic layer will create
an additional channel for precessional damping.

Spin accumulation and blocking of the spin current

A spin current $I_s$ pumped into the NM from the FM layer will lead to a spin accumulation on the NM side of the interface region (see third frame on Fig. 6.2). Since in a non-magnetic metal, the densities at the Fermi level are equal for spin-up and spin-down states, such a spin imbalance represents a non-equilibrium situation resulting in a spin current flowing **backwards** into the ferromagnet. This will rapidly annihilate the spin pumping effect and therefore, no continuous loss of momentum will occur in FM thus, macroscopically no additional damping will be observable. To assure that no spin imbalance is built up, once transferred to the NM layer the "minority" spins have to decay and/or leave the interface region sufficiently fast (by spin-orbit scattering, e.g.). Using a NM layer of thickness $d_{NM}$ with a large spin-orbit coupling, assuring that its thickness is larger than the spin-flip length $d_{NM} \gg \lambda_{sf}$, is a possibility to obtain a continuous flow of the spin current during the precession.

The magnitude of the additional damping

The goal of the original paper by Bauer et al. [97] is to calculate the spin current pumped into the normal metal as well as the additional damping resulting from this loss of momentum. A rigorous mathematic formulation of the effect yields a current pumped into the non-magnetic metal consisting only of spin current (**no charge transfer** between the two metals), described by the vector

$$I_s = \frac{\hbar}{4\pi} \left( A_r S \times \frac{dS}{dt} - A_i dS \frac{dt}{dt} \right),$$  \hspace{1cm} (6.5)

where $A_{r,i} = f(r^{\uparrow\downarrow}, t^{\uparrow\downarrow})$ are interface parameters that can be expressed in terms of the reflection and transmission coefficients of the FM/NM interface. Evidently, in the steady state (no magnetization precession, $dS/dt = 0$) there will be no spin current pumped into the non-magnetic metal.

To quantitatively describe the increase of the precessional damping, the Landau-Lifshitz-Gilbert equation has to be extended with the effect of momentum transfer (loss of torque). This can be done by keeping its original form 2.15 but renormalizing the gyromagnetic ratio $\gamma$ and the Gilbert damping constant $\alpha$:

$$\frac{1}{\gamma} = \frac{1}{\gamma_0} \left( 1 + \frac{g_L A_i}{4\pi M} \right);$$  \hspace{1cm} (6.6)

$$\alpha = \frac{\gamma}{\gamma_0} \left( \alpha_0 + \frac{g_L A_r}{4\pi M} \right).$$  \hspace{1cm} (6.7)

Here the zero indices refer to bulk values, $g_L$ denotes the Landé-factor and $M$ is the total magnetic moment of the ferromagnetic layer expressed in units of $\mu_B$. Via these renormalizations, the imaginary part $A_i$ of the interface parameter affects the precessional frequency, while the real part $A_r$ increases the Gilbert damping. For a number of cases met in most experiments (ballistic and diffusive contacts, and even for nonmagnetic tunnel barriers between FM and NM), according to [98] $A_i$ vanishes.
6.2. SPIN PUMPING MECHANISM IN FM/NM LAYERS

or becomes very small, therefore the precession frequency is not modified \((\gamma \simeq \gamma_0)\) and the formula 6.7 simplifies to a simple addition of an extra damping factor \(\alpha'\) to the intrinsic damping \(\alpha_0\):

\[
\alpha = \alpha_0 + \frac{g_L A_r}{4\pi M}.
\]  

(6.8)

6.2.1 Damping versus FM thickness

By measuring the ferromagnetic linewidth of NM/Py/NM sandwiches through FMR experiments, Mizukami et al. [96] noticed a dependence of the damping parameter on the thickness of the Py layer as shown on Fig. 6.3. Since \(M\) in Eq. 6.8 refers to the total magnetic moment of the ferromagnet, a dependence on the thickness can be understood - the precession of a reduced total magnetization \(M\) is more sensitive to the loss of moment through the interface. Knowing the parameters in 6.8, the additional damping due to the spin pumping effect for Py is estimated by Bauer et al. [97] as function of the Py film thickness \(d\):

\[
\alpha'(d) = \frac{\lambda}{d} f_0 f(d),
\]  

(6.9)

with the factor \(\lambda = 1.1 \cdot 10^{-10}\) m calculated for Py. Besides the thickness-dependence, this formula considers a non-ideal interface with the number of scattering channels per atom \(0 < \kappa < 1\) and a reduced atomic magnetization \(f_0 / f(d)\) for thin films \(d \sim\) nm. Using the interface scattering \(\kappa\) as adjustable parameter, the formula 6.9 fits well the thickness dependence of the damping observed by Mizukami et al. in Pt/Py/Pt \((\kappa = 1.0)\), Pd/Py/Pd \((\kappa = 0.6)\) and Ta/Py/Ta \((\kappa = 0.1)\) sandwiches, measurements shown on Fig. 6.3. However, an analytical method to calculate \(\kappa\) is not yet available.

Even more interesting, within the same set of experiments it was noticed, that the damping parameter also depends on what kind of non-magnetic metal is used as “spin sink”. As we mentioned before, a NM metal with a strong spin-orbit coupling is necessary to assure a fast decay of the NM spin imbalance and, thereby, a continuous spin pumping effect. Generally, the heavier the metal the shorter the spin-flip length is \((\lambda_{sf} \sim Z^4)\). This reasoning is used by Bauer et al. [97] to explain the influence of the NM material within the Mizukami-results, namely that the Gilbert damping pa-

![Figure 6.3: Gilbert damping dependence on the thickness of Py in FM/NM/FM sandwiches, measured by FMR. Figure courtesy of Bauer et al. [97] data measured by Mizukami et al.](image-url)
rameter for the Cu/Py/Cu sample is considerably lower than in the case of Pt/Py/Pt, Pd/Py/Pd and Ta/Py/Ta, and it does not seem to depend on the Py thickness at all.

After the birth of the spin-pumping theory, Mizukami et al. performed a number of additional FMR measurements on Cu/Py/Cu/Pt samples [100] with variable Cu and Py thicknesses, to clarify the effect of spin diffusion through the Cu layer on the Gilbert damping. Recently, the same group performed time-resolved MOKE type experiments on Cu/Py/Cu and Pt/Py/Pt multilayers varying the Py thickness [101]. The behaviour observed was consistent with the previous, FMR-type measurements: the Py sandwiched by Cu layers presented a constant damping parameter, while the damping for the Pt/Py/Pt samples was enhanced as the Py became thinner.

6.2.2 Damping versus NM thickness, our experimental approach

Besides the Mizukami experiments, a number of additional measurements on FM/NM samples (see section “Results”) show that in the presence of the non-magnetic metal, the effective damping is increased. However, these samples consist of a large variety of materials deposited and measured in many different ways, not giving a clear consistent image of the NM influence on the precession.

A systematic study of the spin-pumping effect and its distinction from other effects in the FM/NM junction can be done using the $d_{NM} \gg \lambda_{sf}$ criterium, i.e. knowing that above a certain NM thickness the spin pumping should be continuous and thus the damping should increase according to Eq. 6.8. By keeping the FM thickness $d$ constant and varying the NM layer thickness as $0 < d_{NM} < 20 - 30$ nm, thus using a wedge shaped sample as shown on Fig. 6.4, a series of measurements on the FM precession can be done as a function of the NM thickness on one single sample. This way the deposition method, the material and interface quality etc. will be unique for the whole set of experiments. On the same wedge sample, to check the uniformity of the magnetic parameters and the overall sample quality, measurements on different positions with the same NM thickness can be done as well. Evidently, a global measurement technique such as FMR cannot be used; to measure the $\alpha(d_{NM})$ dependence on a wedge, a local technique will be necessary that can detect the magnetization precession at different lateral positions within the sample.

With the all-optical type time-resolved MOKE setup described in Chapter 3, we
should be able to detect and measure any additional damping due to the spin pumping effect in a local fashion. The resolution in measuring against the NM thickness is dictated by the laser spotsize (≃ 10 µm), the accuracy of positioning the spot on the sample versus the wedge’s position (∼ 10 – 20 µm) and the slope of the wedge (∼ 0.5 nm per 1 mm). This adds up as a resolution better than 0.1 nm. The detailed description of the samples will be given in the following paragraphs.

6.2.3 Samples; expectations

The samples we have fabricated consisted of a Si/SiO$_x$ or glass substrate, a non-magnetic metallic wedge, a thin FM layer and a capping Al layer to prevent oxidation. In some cases a thin Al layer was used on the substrate as wetting layer. The polycrystalline metallic layers were always sputter deposited. Based on the $\lambda_{sf} \sim Z^4$ relationship, as well as due to practical reasons, we decided to use Ta (Z=73) as non-metallic "spin sink" layer, expecting it to have a larger effect than lighter metals like Pd (Z=46) or copper (Z=29) would have. This is partially contradictory to the Mizukami-results shown on Fig. 6.3; for further discussion, see section “Results”.

Samples based on CoFe as ferromagnetic layer

Four batches of such samples were made, with the following specifications:

- Si/SiO$_x$/Ta[0-30nm]/Co$_{90}$Fe$_{10}$[5,10,15nm]/Al[2nm]. At the sample edge, the CoFe was grown directly on the SiO$_x$ (shown on the left side of the sample drawn on Fig. 6.4), with no NM layer in its vicinity, to allow determining the Gilbert damping without any spin pumping effect.

- Si/SiO$_x$/Al[15nm]/Ta[0-30nm]/Co$_{90}$Fe$_{10}$[5,10,15nm]/Al[2nm]. A wetting Al layer was introduced to improve the growth of the CoFe and Ta, respectively. Since the Al is very light metal with a spin flip length of orders of magnitude larger than Ta, it can be considered a very bad "spin sink", thus practically no spin pumping can occur due to the Al.

- Si/SiO$_x$/Al[15nm]/Co$_{90}$Fe$_{10}$[10,15nm]/Ta[0-30nm]/Al[2nm]. The growth order for the NM wedge and FM layer was reversed, to have a more uniform morphology for the CoFe over the whole wedge.

- Glass[80 µm]/Al[2nm]/Co$_{90}$Fe$_{10}$[10,15nm]/Ta[0-30nm]/Al[2nm]. A glass substrate and a thinner Al wetting layer was used, to probe the sample from the bottom, through the substrate. This way the Ta was not blocking the probe beam, permitting measurements in the thick region of the wedge.

The samples with only 5 nm CoFe showed a very noisy MOKE signal, unsuitable for precisely determining the Gilbert damping parameter. The best signal-to-noise ratio was given by the samples containing 15 nm thick CoFe layer, however, according to formula 6.9 the additional damping due to the spin pumping effect should be at least 1.5 times less compared to $\alpha'$ in a 10 nm thick CoFe.

The expected additional damping due to the Ta layer should increase from zero (at very thin Ta) to a value (at 30 nm thick Ta) that can be estimated using a formula of the type 6.9. Evidently, the factor $\lambda = 1.1 \cdot 10^{-10}$ m for Py used by
Bauer et al., based on 6.8, has to be recalculated with the parameters of the CoFe. The most important difference is a larger (bulk) saturation magnetization $M \simeq 1400$ kA/m against the 900 kA/m of the Py, resulting in a factor $\lambda = 0.7 \cdot 10^{-10}$ m. The relative magnetization $f_0/f(d)$ is close to 1 for thick FM films; it reaches 1.5 or higher for films thinner than 10 nm, in which case the presence of the non-magnetic metal also influences $f(d)$, as shown in the top inset of Fig. 2 in [97]. The value of the interface parameter $\kappa$ is, again, empirically determined; considering values of 0.1 and 0.3 as used for Ta-Py systems by Bauer et al. and Lagae et al. [54] respectively, the spin-pumping induced additional damping $\alpha'$ should be between 0.001 and 0.003 for the $d_{FM} = 10$nm samples and somewhat lower for the 15nm ones. The intrinsic Gilbert damping parameter we determined from precessional measurements on thin CoFe single layers was $\alpha_0 \simeq 0.03$, corresponding to values from the literature.

Samples using Permalloy

As an alternative to the previously presented CoFe samples, we fabricated wedge samples similar to the generic one shown on Fig. 6.4, consisting of Si/SiO$_x$/Al[15nm]/Ta[0-30nm]/Ni$_{80}$Fe$_{20}$[10,15nm]/Al[2nm] layers. The estimated value of the additional damping $\alpha'$ in the 10 nm thick NiFe (Permalloy) layer, caused by loss of momentum to the Ta, is within 0.0025 – 0.006, depending again on the value of $\kappa$. Compared to the intrinsic damping for Permalloy, known to be $\alpha_0 \simeq 0.01$, the increase should be detectable easier than in the case of CoFe.

One advantage of the Py used as the FM layer in our samples is, that the growth conditions do not strongly influence the magnetic structure of a thin Py film, in contrast with a CoFe one, as we will show later by experiments.

6.3 Results

We have performed two series of measurements on all samples. First of all we have measured full magnetization loops using a static Magneto-Optic Kerr measurement setup, on different positions across the sample, extracting parameters such as coercivity in the function of position. An example is shown in Fig. 6.5. We did this to acquire an idea about how uniform the magnetic structure is (domains, local fields and anisotropies) on a scale of $\simeq 30\mu$m. We did these measurements at positions along the direction of both constant and increasing Ta thickness (two perpendicular directions on the sample surface), in order to check the effect of the underlayers on the FM morphology.

Further we have measured the time-resolved MOKE signal with the all-optical pump-probe setup, and from the precessional data we have determined the effective damping constant $\alpha = \alpha_0+\alpha'$ as a function of, again, the Ta thickness. For this, we use the top right formula from 2.19 correlating the full damping $\alpha$ and the attenuation time $\tau$. An example of a time-resolved MOKE measurement is shown in Fig. 6.6, including the fit with an exponentially decaying sinusoidal that allows determining $\omega$, $\tau$ and $\alpha$ respectively.

For very thin or no Ta, the damping was expected to not have the spin pumping contribution. For thick Ta, up to tens of nanometers, the $\alpha'$ component should have increased and saturated around a maximum value, corresponding to the maximum
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Figure 6.5: An example of a static MOKE measurement on the Py/Ta wedge sample with the Ta thickness of 18 nm. The calculated coercivity is plotted.

Figure 6.6: An example of an all-optical TRMOKE measurement on the Py/Ta wedge sample (full circles) fitted with an exponentially decaying sinusoidal (solid line). The remagnetization phenomenon is approximated with a linear slope after the first 100 ps, where the fitting starts. The frequency, damping time and damping factor is calculated.
loss of momentum that can occur due to the spin current as estimated in the previous section of this chapter. The increase of the damping in function of the Ta thickness $d$ we expect to follow an exponential function of the form

$$\alpha' \sim \left[ 1 - \exp \left( \frac{-d}{\lambda_{sf}} \right) \right],$$

where $\lambda_{sf}$ is the spin-flip length in the non-magnetic metal. While in ferromagnetic transition metals the spin-flip length is very short, only 1-2 nm, in non-magnetic metals it can extend from tens to thousands of nanometers (becoming shorter for heavy metals). For Ta, we expect $\lambda_{sf} \simeq 10$ nm based on literature values [99].

### 6.3.1 CoFe measurements; morphology dependence of the damping

**CoFe directly on substrate**

The MOKE loop measurements on the CoFe/Ta sample without any wetting Al layer show a very large variation of the coercivity between 1 kA/m and 6.7 kA/m, as plotted on Fig. 6.7. This indicates a severe variation in the local anisotropies in the CoFe layer, attributed to the underlying Ta thickness and growth conditions. Only with Ta thicknesses of over 20 nm the coercivity begins to stabilize around a value that we consider “normal” for our 15nm CoFe layer.

The same trend is noticeable in the few damping values plotted on the same figure 6.7. In the regions where the Ta thickness was below 20nm, only a few time-resolved measurements were showing a regular damped sinusoidal, and even there the error in calculating the damping parameter was rather large. The far right region of

![Figure 6.7: Measurement: CoFe on Ta wedge. Left axis: coercivity vs Ta thickness, determined from MOKE loop measurements. Right axis: effective Gilbert damping parameter, determined from TRMOKE precessional measurements. In many cases, the precession was not regular enough to extract the damping parameter. The line connecting the data points is only a guide-to-the-eye.](image-url)
the curve shows a damping stabilized at just above the literature value of $\alpha = 0.03$ for thin CoFe single layers. However, the error in determining the damping parameter and its variation along the sample is an order of magnitude larger than the spin-pumping induced $\alpha'$ we expected to find.

The measurements on 5 and 10 nm CoFe layers show similar behaviour with even stronger position dependence, hence they are omitted from this thesis.

**CoFe grown on a wetting layer**

The situation gets more controlled with using a 15 nm thick Al wetting layer on the Si/SiO$_x$ substrate. The growth of both the CoFe layer and the Ta wedge improves when it is done not directly on the SiO$_x$ [102, 103]. Traditionally, a heavy metal is used as seed layer but in our case that would have interfered with the Ta thickness needed for the onset of the spin pumping mechanism. The results using Al as seed layer did make some improvements compared to the previous case. The extreme values for the coercivity and effective damping are not that high, however the dependence of these parameters on the Ta thickness is still consistent, even more than in the previous case. The results of the measurement series (15 nm CoFe on Ta/Al) are shown on Fig. 6.8. The most important improvement is though, that the noise in the precessional measurements was much smaller, yielding a low error on the values of $\alpha$. Interestingly, the damping in this case is stable around the 0.03 value for the region with thin Ta (<4 nm) and increases on the right side. The onset of the Ta wedge on the left side of the graph still has a clear influence on both the coercivity and the effective damping, indicating the delicacy of the CoFe morphology.

![Figure 6.8](image.png)

Figure 6.8: Measurement: CoFe on Ta wedge using 15 nm Al wetting layer. Coercivity (left) and effective Gilbert damping parameter (right) vs Ta thickness. The solid lines represent polynomial fits to the datasets.

The influence of the Ta thickness on the morphology of the CoFe layer can be explained assuming that it follows the so-called Stranski-Krastanov growth [102]. A very thin layer (monolayers) of sputter deposited Ta decreases the roughness of the Al layer surface and the CoFe growth will be smoother. As the thickness of the Ta
layer is increasing however, its surface forms islands and the CoFe roughness increases, giving birth to grains of various sizes.

Looking at Fig 6.8, besides their dependence on the Ta thickness, a striking resemblance between the behaviour of the coercivity and effective damping can be noticed; an almost linear relationship seems to exist between them. It is not clear, why the damping on the nanosecond timescale should be correlated with a static magnetic parameter like coercivity. However, the dependence of both of them on the CoFe morphology can be understood. With the increasing Ta thickness, the appearance of grains in the CoFe layer means appearance of domain walls within the measurement region, defining distinct magnetic domains. These might have different equilibrium orientations for the magnetization dictated by local anisotropies and interactions between them. If the domain sizes consistently vary from thin to thick Ta following the Ta surface roughness, the contribution of surface Co and Fe atoms to the magnetization of the domain as well as the influence of the neighbouring domains will also vary monotonously. This will influence the shape/crystalline anisotropy of each domain, and, on the scale of our measurement, it will be reflected in the coercivity and remanence of the ferromagnetic material.

As to the damping increase, a magnetic structure with a spread in the equilibrium magnetization directions and in the local fields will present a higher effective damping. The increase is mostly due to dephasing of the precessing magnetization vectors of individual domains within the probing laser spot, and possibly to local generation of higher-order spin waves, as explained earlier in Chapter 4, Section 4.3. This reasoning is supported by results similar to ours, reported by Rantschler et al. [104], on thin sputter deposited CoFe layers with or without a Cu underlayer. The grain size, measured by TEM, proved to be considerably smaller with a Cu wetting layer (9 nm vs 50 nm, compared to the exchange length in CoFe $L_{ex} = 3$ nm). In the same time, the FMR linewidth decreased 20 times with the introduction of the Cu layer, indicating an influence of the morphology on the extrinsic damping parameter. The linewidth for CoFe was dominated by the large extrinsic contribution in such a manner that the small intrinsic damping parameter was impossible to determine. Similar behaviour was reported in the same article [104] on thin FeTiN layers; gradually increasing the nitrogen content from 0 to 6%, the TEM measurements show decrease of the grain size from 28 nm to 9 nm, while the FMR linewidth also drops by a factor of 10.

We can conclude thus, that an additional damping $\alpha'$ due to any spin pumping effect is impossible to detect in these experiments, since the effect of the CoFe morphology on the damping is at least an order of magnitude larger.

Let us finally address the behaviour of the precessional frequency (the resonance frequency in the case of FMR measurements). If this frequency would change in function of Ta thickness, a Kittel-type relationship would indicate that the average saturation magnetization of the CoFe and/or the effective local magnetic fields within the measurement region change. In the samples with CoFe, we could not detect any consistent change of the precessional frequency following the thickness change of the Ta wedge. This is an indication thus, that while the damping is severely influenced by the ferromagnetic layer’s morphology, the precessional frequency is not, and therefore it is not a good parameter to characterize the growth quality of the layer. The FMR results of Mizukami et al. on the various NiFe/NM samples, although interpreted to the favour of the spin-pumping mechanism, reflect the
same behaviour: strong influence of the underlayer on the FMR linewidth, while the resonance frequency is always the same. Also, Rantschler et al. show that the introduction of the Cu layer does not considerably change the saturation magnetization of the CoFe ($M_s = 1847 \text{ kA/m vs } 1850 \text{ kA/m without Cu}$) but the treatment of the local anisotropies is omitted by the authors.

Reversing the growth order of the NM/FM layers

To obtain a uniform CoFe morphology independent of the Ta thickness, we reversed the growth order of the two metals. The measurements had to be done through the Ta wedge in this case. As shown on Fig. 6.9, the coercivity was found to be relatively constant under 15 nm Ta thickness; the quality of the MOKE measurements above 15 nm was deteriorated by the low laser power transmitted through the Ta layer. The dynamic measurements yielded an effective damping that has a point-to-point variation of $\approx 0.008$ over the sample. The two marked points on Fig. 6.9 measured both at 4 nm thick Ta but at different positions on the sample indicate, that this variation is not correlated to the thickness of the Ta layer. This “error level” still exceeds the predicted value for the spin-pump induced additional damping. No systematic increase of $\alpha$ versus the Ta thickness was observed.

![Figure 6.9: Measurement: Ta wedge on CoFe using 15 nm Al wetting layer. The CoFe coercivity (left scale) is flat up to 15 nm; the effective Gilbert damping parameter (right scale) still has a large spatial spread, as shown by the two points measured at 4 nm thick Ta. The solid line is only a guide-to-the-eye.](image)

The signal-to-noise ratio could be improved and made independent of the Ta thickness by probing the CoFe layer through a transparent substrate. We manufactured therefore a set of samples on a 120 $\mu$m thick glass substrate, using only 5 nm Al as wetting layer. The static MOKE measurements looked promising again, with respect to the CoFe morphology. However, the heat conductivity of glass is much lower than the Si/SiO$_x$ and during the pump-probe measurements, the sample was deteriorated by cumulative heating from the pump pulses. Even at very low incident pump power, the DC heating did not allow for any all-optical measurements on the magnetization dynamics of the CoFe layer.
6.3.2 Measurements on Permalloy

The static MOKE measurements on 10 nm Py, deposited on Ta wedge, yielded a stable coercivity at around 300 A/m, indicating a good quality, homogeneous thin magnetic layer. The values are plotted against the Ta thickness on Fig. 6.10(b), left scale.

![Figure 6.10](image)

Figure 6.10: Measurement: Permalloy on Ta wedge with 15 nm Al wetting layer. The Py coercivity (left scale) does not considerably depend on the Ta thickness; the effective Gilbert damping parameter (right scale), determined at four different positions for each chosen Ta thickness, does not indicate any systematic increase from left to right.

The dynamic measurements on this sample were performed for Ta thicknesses of 0, 2, 4, 6, 10, 14 and 18 nm. Four such measurements were done at different y-positions on the sample separated by 200 µm, for each chosen Ta thickness, as shown on Fig. 6.10(a). The calculated effective damping factors are plotted on Fig. 6.10(b), right scale. An average of the four points was calculated for each Ta thickness, represented by the solid line on the same figure. A few measurements were of a lower signal-to-noise ratio and in the same time, yielded an extreme value for α. We attribute these deviations to defects on the surface on the micrometer scale, hence the corresponding points, marked on the figure, were left out from the calculus of the average damping value.

As seen from the plot, the expected value of the additional spin-pump induced damping $\alpha' = 0.0025 - 0.006$ is well above the point-to-point variation of the measured effective damping. However, no systematic increase of α can be seen on the graph when the Ta thickness increases (in fact, there seems to be a slight decrease of the values towards the right of the graph). We can conclude thus, that in our experiments no spin current pumped into the Ta can be observed. A possible explanation of this result is not having access to the values of $\kappa$ in the formula 6.9; in case the real value for Py/Ta interfaces is much below 0.1, the additional damping becomes too small to be experimentally observable. Another explanation would be, that the spin-flip length in Ta is above 30 nm, and the spin current pumped into the Ta layer in all our measurements is blocked by the spin accumulation around the interface. This is however not very feasible, since the large spin-orbit coupling in
Ta should result, as we mentioned earlier, in a spin-flip length shorter than the Ta thicknesses we used.

The Gilbert damping values plotted on Fig. 6.10(b) are all offset by 0.003 compared to the expected value of $\alpha = 0.011$ considered normal for thin Py layers. We conjecture that this shift is due to a peculiarity of the all-optical technique: we focus the pumping laser beam to a spot of $10 \mu m$ diameter, producing a local excitation of spin precession. The generated spin waves will have higher order components quickly diffusing into the surrounding regions of the Py layer, leading to a loss of momentum in the original excitation center. Since we probe the dynamics of this region only, the momentum loss will manifest as an increase in the effective, measured damping $\alpha$. This increase is, of course, independent of the underlying Ta thickness, thus it does not disturb the measurements.

On Fig. 6.11 we plotted the behaviour of the precessional frequency against the Ta thickness. While the damping and the coercivity is unchanged, the frequency clearly shows a slight monotonous increase ($\sim 2\%$) towards thicker Ta layers. This could be a heat-related artifact: the thinner Ta absorbs less heat from the pumped layer thus the DC temperature of the Py is higher, resulting in a somewhat lower saturation magnetization and, therefore, lower precessional frequency. However, a DC temperature difference of over 10K would be needed for such a difference in the $M_s$ values. Hence, we have to consider the possibility that this increase is, again, morphology-related.

**Induction-based measurements**

We have sent our Py sample to the magnetization labs of National Institute of Standards and Technology (NIST, Boulder, CO, USA) where induction-based pump-probe measurements were done by Gerrits et al. to determine the effective damping parameter. The technique consists of uniform excitation of the whole sample by magnetic field pulses and probing the local dynamics by measuring the current induced by the precessing magnetization. The lower values (0.011 versus our 0.014) for the damping parameter obtained on the sample region without any Ta are consistent with our
previous conjecture: a uniform excitation is needed to eliminate the extra damping resulting from diffusion of spin waves. The offset described in the previous paragraph does not apply here.

The resulting values for the effective damping are plotted on Fig. 6.12 against the Ta thickness. Aside of the offset, a good correspondence exists between this plot and our all-optical results. Again, no systematic increase of the damping was found in this sample, indicating the lack of a detectable spin-pumping effect.

Comparison with literature

The first studies indicating an additional damping due to adjacent metallic layers (including Ta) were the ones published by Mizukami et al. [96], as we presented before. These formed the basis of the spin-pumping theory as elaborated by Bauer et al. [97]. The results, together with the ones on Cu/Py/Cu/Pt multilayers published by the same group in 2002 [100], were obtained from non-local FMR measurements on a number of samples with different compositions. The consistency of the FM layer’s morphology was, in these cases, verified by the unique FMR frequency for all the samples at a certain applied magnetic field, while the width of the FMR absorption spectrum served for calculating the additional damping. The damping increase was purely attributed to the spin-pumping effect. However, as we have shown in our time-resolved experiments, the precessional frequency (thus the ferromagnetic resonance frequency) is not related in a straightforward way to the granular structure and magnetic domain formation in the thin FM layer.

Recently, Mizukami et al. performed time-resolved local Kerr measurements using a magnetic field pulse as pump and laser as probe, on Cu/Py/Cu and Pt/Py/Pt sandwiches [101]. The results obtained this way show that the two non-magnetic metals have a different effect on the precession of the Py magnetization. This is in accordance with their earlier FMR type measurements and is attributed by the authors to the spin-pumping effect present in the Pt/Py/Pt samples but missing in the Cu/Pt/Cu ones. An etch-back technique was used to flatten the SiO\textsubscript{2} substrate, in order to eliminate the effects of the roughness on the magnetic properties; in the same time, the differences in the Py morphology and the distortion of the magnetic order
that might result from the presence of the Pt underlayer compared to the lighter Cu, are not addressed. Moreover, at weak bias fields a deviation from the LLG-predicted values is observed and attributed to the anisotropic dispersion of the magnetic layer.

Figure 6.13: The damping values in Py obtained by Lagae et al. (black symbols) compared to our results (gray line+stars).

Studies on a number of samples with Py, Py/Cu, Py/Ta and Py/Cu/Ta thin layers were done by Lagae et al. [54], using a pump-probe experimental setup: Py thickness was either 5nm or 15nm. The resulting Gilbert damping parameters are plotted on Fig. 6.13 together with our results on the Py[10nm]/Ta[0-30nm] bilayer shown before. An increase in the effective damping was noticed in the case of samples with Ta, even when its thickness was only 2nm: this was attributed to the spin-pumping mechanism. The additional damping corresponds to the value predicted by the formula 6.9, if the value for the Ta/Py interface scattering parameter $\kappa$ is fixed at 0.29. This value however is three times higher than the value used by Bauer et al. on Ta/Py/Ta sandwiches. A severe drop ($\sim$30%) of the saturation magnetization $M_s$ was noticed with the introduction of the Ta layer, but not with the introduction of the Cu layer. The loss of the magnetic moment was calculated to be equivalent to a magnetically dead layer of $\sim$1 nm and was attributed to the interdiffusion between Ta and NiFe during deposition. The Ta clearly has an effect on the NiFe properties in case of these samples, and this might not be restricted to interdiffusion and decrease of $M_s$. We believe that, prior to the interpretation of the damping increase as the manifestation of the spin pumping effect, a systematic testing of the NiFe morphology is needed.

6.4 Conclusions

We were unable to detect any indication of a spin-pumping effect in CoFe/Ta and NiFe/Ta bilayers, but we did notice severe influence of the layer morphology on the damping parameter. Therefore, two important notes have to be made:

- To give a firm ground to the formula 6.9 within the spin-pumping model of Bauer et al., the numeric values for the NM/FM interface scattering parameter $\kappa$ have to be more precisely determined. First-principle calculations will be needed in order to do this.

- The morphology of the thin ferromagnetic layer depends on the underlying NM
layers and the substrate, and it has a severe influence on its magnetic properties. This can lead to an increased effective damping parameter. To distinguish between the intrinsic Gilbert damping, the morphology-related extrinsic damping and the additional damping induced by a spin-pumping effect, a systematic study of the magnetic domain structure and the layer morphology is necessary. However, through our experiments we have found, that the precessional frequency—the FMR frequency— cannot be directly used to verify the quality and uniformity of the magnetic layer on different samples.
Bibliography


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Summary

This thesis presents a set of experimental studies focussed on fast magnetic phenomena in microscopic ferromagnetic structures and thin layers. Ferromagnets such as iron, cobalt, nickel and their alloys present a net magnetic moment without application of an external magnetic field, therefore called “spontaneous magnetization”. Technology today would be impossible without the microscopic applications of magnetism – to give an example, a car manufactured in 2005 has over 100 magnetic microsensors built into its engine, steering, break system etc. –, however in the field of magnetic interactions on the short timescale there are still many open questions. Quick reaction to changes of external magnetic field as well as fast switching of tiny magnetic structures (e.g. in case of magnetic recording on the hard disk drive of a computer) necessitate in-depth studies of magnetization dynamics on a very short timescale.

Changing the orientation of the magnetization of a sample by applying an external magnetic field happens on the nanosecond timescale, that is, in the billionth of a second (1 ns = 0.000000001 s). In our experiments, as described in Chapter 3, we used a pump-probe technique to study the dynamics on such a short timescale. The technique relies on a high number of perturbation (“pump”) – detection (“probe”) cycles, with a repetition rate of 80 MHz (80 million cycles in one second), and a controllable time delay between the perturbation and the detection. The perturbation (either a magnetic field pulse of hundreds of picoseconds duration or a heating laser pulse of 1000 times shorter) is aimed to quickly change the orientation of the equilibrium direction for the magnetization vector. The detection (a linearly polarized laser pulse reflected from the sample surface) is weak enough not to influence the dynamics of the magnetization. Controlling and recording the time delay between the pump and probe by a computer makes it possible to record temporal change in the direction and/or amplitude of the magnetization, with a resolution of picoseconds (magnetic field pump) to femtoseconds (laser heating pump).

Focussing the probe laser beam to a small spot, we have achieved a spatial resolution as high as 1 micrometer necessary for probing the local magnetization dynamics on the samples. We have developed a vectorial detection scheme (described in Section 3.5.1) that allows measuring the dynamics of two spatial components of the magnetization vector simultaneously. In case of constant magnetization amplitude, the third spatial component and the full vectorial dynamics can be determined from these measurements.

The spin of the electrons, the building block of a macroscopic magnetization, is modelled in simple terms as the angular momentum of a sphere-shaped particle.
rotating ("spinning") around a symmetry axis. This axis represents the orientation direction of the spin. If an external perturbation such as a fast rising magnetic field is applied non-collinear with the spin direction, a torque will act on the spin driving it into a precessional motion around the direction of the field or, more generally, the new equilibrium direction. On a macroscopic scale, the same happens inside a ferromagnetic structure: the effect of an external field on the magnetization creates a new equilibrium orientation and the magnetization vector will be precessing around this direction. As shown in Fig. 6.14, spin (and magnetization) precession is similar to the macroscopic precessional motion of a spinning top and to the precessional motion of the Earth’s rotational axis. The precessional frequency (or period) of the electron spin can be tuned by the applied external field as well as parameters of the magnetic material; in the case of our studies this precession was in the gigahertz range, a billion times faster than the spinning top. Ultimately, the interaction of the electron spin with its environment will make sure that the precessional motion is damped and the spin (magnetization) will reorient along the new equilibrium direction, a phenomenon happening on the scale of 0.1-10 ns. Thus it is clear, that the above mentioned time resolution of our experiments is well suited for studying the precessional magnetization dynamics.

In order to understand the GHz magnetization dynamics, we have performed a series of pump-probe experiments on simple and complex ferromagnetic samples. We have measured the uniform magnetization precession in the center of a single microscopic ferromagnetic disc. In stacks of coupled ferromagnetic structures we have detected higher-order precessional modes predicted by theoretical considerations. Finally, we made measurements on the local magnetization dynamics over the whole area of a multilayered microscopic sample. In Chapter 4 we presented a selection of these experiments with special relevance to possible future applications. We have found that the frequency of the precession is strongly influenced, besides the applied external field, by local magnetic fields rising from microscopic effects. These are related to symmetry breaking at the edges of the sample and to magnetic interactions with ferromagnetic structures in the vicinity of the sample. More precisely, the spin...
Summary

Precession shows a strong dephasing across the sample surface, and spin waves of higher order are launched simultaneously with the fundamental, uniform precessional mode. On the other hand, the damping of the precessional motion is also influenced by these extrinsic effects. The higher-order spin waves can efficiently take over momentum from the fundamental precessional mode yielding an apparent increase in damping; in addition, at the sample’s edge the dephasing of the spin precession within the studied region (the laser spot) results in an impressive increase of the effective damping. The experimental observation of such incoherent dynamics compared to numerical simulations indicates, that the sample edges and the stray magnetic fields play a crucial role in the global magnetic behaviour on the (sub)nanosecond timescale. Future applications (sensors, data recording media, magnetic memory cells) aiming to switch the magnetization of even smaller multilayered stacks will have to consider and deal with this precessional dephasing and increased damping in order to control the speed and stability of the switching process.

In many cases, especially for commercial applications that aim for fast switching, an external magnetic field pulse of sub-nanosecond duration is a preferred perturbation source for initiating magnetization dynamics. Precise knowledge of the parameters (amplitude, shape, duration) of such field pulses is crucial for controlling the dynamics. Therefore we implemented and presented a back-calculation method for these pulses in Chapter 5. The method starts from pump-probe measurements on the magnetization precession in a microstructure and calculates the original magnetic field pulse that crossed the sample and generated the precessional dynamics. The example calculations (shown in Section 5.2.) indicate, that the method is well suited to back-trace the fast magnetic field pulses and reveal details in the pulse shape with a precision of under 100 picoseconds.

We have extended the calculation method to the “all-optical” experiments where laser heating was used as pump source, instead of external magnetic field pulses. The idea in these experiments is that the sudden, local heating of the ferromagnetic layer by a focused intense laser beam generates a strong change in the anisotropy of the heated region and this so-called “anisotropy field pulse” (a strongly localized, high-amplitude magnetic field pulse) drives the precession of the magnetization. The back-calculation performed on all-optical measurements (example shown in Section 5.3.) yielded a delta-like anisotropy field pulse (FWHM ~ 2 ps, amplitude ~ 30 kA/m), superposed on a step-like background with a fall time of hundreds of picoseconds. The result is in good accordance with previous conjectures on the existence of such an anisotropy field pulse. This pulse is orders of magnitudes faster and stronger than the field pulses we were able to produce by induction with current passed through a stripline, and can be of relevance for future applications e.g. hybrid recording technology, where a fast intense field pulse has to be created locally.

In Chapter 6 we presented an experimental study on the precessional damping in thin ferromagnetic layers in contact with non-magnetic metallic layers. Theoretical calculations suggest the possibility of an increased damping in FM/NM multilayers as a result of loss of spin momentum from the ferromagnet to the normal metal. The momentum loss is supposed to be mediated by a spin-current pumped through the interface into the normal metal without any net charge transfer, the effect being called “spin-pumping effect”. From the applications point of view the effect has potential, both for generating a spin current through a normal metal and for controlling the
damping in thin ferromagnetic layers. We have performed systematic all-optical type measurements on samples with two different ferromagnetic materials (CoFe and NiFe) and a variety of thicknesses for the NM (0 to 30 nm Ta wedges). The results indicated that the increase of the damping parameter yielded by a spin-pumping effect was masked by large extrinsic effects related to the morphology of the FM layer. The growth conditions and the varying thickness of the Ta underlayer resulted in a granular structure for the ferromagnet, giving rise to magnetic domain formation. When checking the influence of the Ta thickness on the ferromagnet’s morphology, an almost linear relationship was found between the precessional damping parameter and the coercivity of the ferromagnetic layer. Our results compared with recent studies presented in literature indicate the absolute necessity to verify the quality and uniformity of the magnetic layer, before any damping increase could be unambiguously attributed to the spin-pumping effect.

The experiments presented in this thesis took us closer to understand and control the fast dynamics of a spin ensemble in complex multilayered microstructures, to help perform a magnetic choreography on a sub-nanosecond timescale.
Samenvatting

Dit proefschrift geeft een beschrijving van experimenteel onderzoek op het gebied van snelle magnetische processen in microscopische, ferromagnetische structuren en dunne lagen. Ferromagneten zoals ijzer, kobalt, nikkel en hun alliages vertonen een “spontane magnetisatie,” een netto magnetisch moment zonder dat daarvoor een extern magneetveld aangelegd hoeft te worden. De hedendaagse technologie zou niet meer zonder de microscopische toepassing van ferromagnetisme kunnen – een auto uit 2005, bijvoorbeeld, bevat meer dan 100 magnetische microsensoren in zijn motor, stuurinrichting, remsysteem, enz. –, maar op het gebied van de snelle magnetische interacties is nog erg veel onbekend. Zowel de snelle respons op veranderingen in extern magneetveld als het snelle ompolen van minieme magnetische structuren (zoals in het geval van het magnetisch wegschrijven van data op de vaste schijf van een computer) vraagt om diepgaand onderzoek naar magnetisatiedynamica op extreem korte tijdschalen.

De verandering in oriëntatie van de magnetisatie van een sample door een extern magneetveld, gebeurt op de tijdschaal van een nanoseconde, het miljardste deel van een seconde (1 ns = 0.000000001 s). In de experimenten die staan beschreven in hoofdstuk 3 is een zogenaamde pomp-probeertechniek gebruikt om de dynamica op een dergelijke korte tijdschaal te onderzoeken. Deze techniek is gebaseerd op een groot aantal excitatie- (“pomp”) en detectie- (“probe”) cycli met een herhalingsfrequentie van 80 MHz en een controleerbare tijdvertraging tussen excitatie en detectie. De excitatie, een kortstondig een magneetveld van enkele honderden picoseconden, dan wel een 1000 maal kortere verhitingslaserpuls, is bedoeld om op een snelle manier de evenwichtsrichting van de magnetisatievector te veranderen. De detectie, een lineair gepolariseerde laserpuls, die wordt gereflecteerd aan het sampleoppervlak, is zo zwak dat deze de magnetisatiedynamica niet beïnvloedt. Door nu de vertraging van de probepuls ten opzichte van de pompuls te variëren, kan de tijdsafhankelijkheid van de richting en/of amplitude van de magnetisatie worden gemeten met een resolutie van picoseconden in het geval met een magneetveld als pomp en van femtoseconden bij verwarming met een laserpuls.

Door de probelaserpuls te focussen op een klein oppervlak is een ruimtelijke scheidbaarheid van maar liefst 1 micrometer behaald, wat nodig is om lokaal de magnetisatiedynamica te bepalen. Er is een vectorieel detectieschema ontwikkeld (beschreven in paragraaf 3.5.1), dat het mogelijk maakt om de dynamica van twee ruimtelijke componenten van de magnetisatievector gelijktijdig te meten. In het geval van een gelijkblijvende amplitude van de magnetisatie kunnen zowel de derde component als de volledige vectoriële dynamica hieruit worden afgeleid.
De elektronenspin, de bouwsteen van macroscopische magnetische momenten, wordt in een eenvoudig model opgevat als het impulsmoment van een bolvormig deeltje dat draait (“spint”) om zijn symmetrieas. Deze as geeft de oriëntatie van de spin aan. In het geval van een externe verstoring zoals een snel toenemend magneetveld *in een andere dan de richting van de spin*, zal een moment op de spin ontstaan. Het gevolg is dat de spin gaat precederen om de richting van het veld, of, algemener, om de nieuwe evenwichtsrichting. Iets soortgelijks gebeurt op macroscopische schaal in een ferromagnetische structuur: een extern magneetveld leidt tot een nieuwe evenwichtsrichting van de magnetisatie, die vervolgens om dit nieuwe evenwicht zal gaan precederen. Zoals te zien is in figuur 6.15, is spin- (en magnetisatie-) precessie analoog aan het precederen van de rotatieas van de Aarde. De precessiefrequentie (of -periode) van de elektronenspin kan worden geregeld via het extern aangelegde magneetveld, maar ook via de eigenschappen van het magnetische materiaal; in het geval van de hier beschreven experimenten is de precessiefrequentie in het gigahertzbereik, een miljard maal sneller dan een spinnde tol. Uiteindelijk zal de interactie van de elektronenspin met zijn omgeving ervoor zorgen dat de precessie uitdempt en de spin (de magnetisatie) langs de nieuwe evenwichtsas komt te liggen, iets wat typisch 0.1-10 ns duurt. Het moge duidelijk zijn dat bovengenoemde tijdsresolutie ruim voldoende is om de precessieve magnetisatiedynamica te bestuderen.

Figure 6.15: Precessie van (a) de Aarde in het gravitatieveld van de Zon en de Maan met een precessieperiode van $T = 26.000$ jaar, (b) een tol in het zwaartekrachtsveld van de Aarde met $T \sim 1$ s en (c) de elektronenspin in een magneetveld met $T \simeq 1$ ns. De witte pijl geeft de rotatierichting aan, de zwarte pijl de precessierichting.

Om de GHz magnetisatiedynamica beter te kunnen begrijpen, is een reeks pomp-probe experimenten uitgevoerd op eenvoudige en meer complexe ferromagnetische samples. Zo is de uniforme precessie van de magnetisatie middein in een enkele, microscopische ferromagnetische schijf gemeten. Daarnaast zijn in multilagen van gekoppelde ferromagnetische structuren hogere-orde precessiemodes waargenomen die al waren voorspeld op grond van theoretische beschouwingen. Tenslotte zijn er metingen uitgevoerd van de ruimtelijke afhankelijkheid van de *lokale* magnetisatiedynamica op het gehele oppervlak van een multilaag met microscopische laterale afmetingen. In hoofdstuk 4 wordt een keur van deze experimenten gepresenteerd met nadruk op de relevantie voor mogelijke toekomstige toepassingen. Gebleken is dat de *precessiefrequentie* behalve door het extern aangelegde veld ook sterk wordt beïnvloed
Samenvatting

Door lokale magneetvelden ten gevolge van microscopische effecten. Deze hangen samen met het verbroken worden van de symmetrie aan de randen en met magnetische interacties met ferromagnetische structuren in de nabijheid van het sample. Preciezer geformuleerd gaat de precessie van spins in verschillende delen van het sample sterk uit fase lopen en gelijktijdig met de uniforme precessie ontstaan spingolven door het sample. Aan de andere kant wordt ook de uitdemping van de precessiebeweging beïnvloed door deze indirecte effecten. De hogere-orde spingolven absorberen op een efficiënte manier impuls uit de precessiebeweging, waardoor de uitdemping sneller schijnt. Bovendien leidt het uit fase lopen van de spins binnen het meetgebied (de laser spot) aan de randen van het sample tot een indrukwekkende versnelling van de effectieve uitdemping. De observatie van dergelijke incoherente dynamica geeft, tezamen met numerieke simulaties, aan dat de randen van het sample en magnetische strooivelden een cruciale rol spelen voor het magnetische gedrag van het gehele sample op (sub-) nanosecondetijdschaal. Toepassingen in de nabije toekomst (sensoren, media voor dataopname, magnetische geheugencellen), waarin de magnetisatie van nog kleinere meerlaagse structuren moet worden omgepoold, dienen rekening te houden met het uit fase raken van de precessie en met de toegenomen uitdemping om de snelheid en stabilitéit van het schakelproces te beheersen.

Vaak, met name voor commerciële toepassingen waarin magneten snel worden geschakeld, wordt de voorkeur gegeven aan een puls van een extern magneetveld met een duur van minder dan een nanoseconde om de magnetisatiedynamica op gang te brengen. Een gedetailleerde kennis van de pulsparameters (amplitude, vorm en tijdsduur) van dergelijke magneetveldpulsen is noodzakelijk om zo de dynamica te beheersen. Daarom is een rekenmethode ontwikkeld om de pulsen terug te rekenen uit de metingen, die wordt gepresenteerd in hoofdstuk 5. Startpunt voor deze methode zijn de pomp-probe-metingen van de precessie van de magnetisatie in microstructuren, waaruit de oorspronkelijke magneetveldpuls, die op het sample heeft gewerkt en zo de precessie op gang heeft gebracht, wordt berekend. De berekeningen die in paragraaf 5.2 als voorbeeld worden gepresenteerd, tonen aan dat deze methode geschikt is om de oorspronkelijke snelle magneetveldpulsen te herleiden en zo details in de pulsomgeving met een precisie van 100 picoseconden te bepalen.

Deze rekenmethode is uitgebreid voor geheel optische experimenten waarin verwarming met laserpulsen wordt gebruikt als de pomp, in plaats van de gepulste magneetvelden. Deze experimenten zijn gebaseerd op de verwachte anisotropieverandering in het gebied waar de sterk gefocuseerde, intense pomplaserpuls wordt geabsorbeerd. Deze “anisotropieveldpuls” (een krachtige en sterk gelokaliseerde magneetveldpuls) is dan de bron van de magnetisatiedynamo in precessie. Terugrekenen vanuit de resultaten van geheel optische metingen (zoals die in paragraaf 5.3) heeft geleid tot een op een delta-piek lijkende anisotropiepuls (FWHM ~ 2 ps, amplitude ~ 30 kA/m) die is gesuperponeerd op een stapvormige achtergrond die uitdempt met een degeningsstijd van honderden picoseconden. Het resultaat komt overeen met eerdere voorspellingen over het bestaan van zo’n anisotropieveldpuls. Deze puls is ordes van grootte sneller en sterker dan de pulsen die konden worden geproduceerd door middel van induktie ten gevolge van een stroom door een nabijgelegen strip. Zo’n anisotropieveldpuls kan dus van belang zijn voor toepassingen, zoals in de hybride-opnametechnologie, waarbij lokaal een snelle, intense magneetveldpuls nodig is.

Hoofdstuk 6 gaat over een experimenteel onderzoek naar de precessie-uitdemping.
in dunne ferromagnetische lagen in contact met niet-magnetische metallische lagen. Berekeningen suggereren dat uitdemping sneller gaat in FM/NM multilagen als gevolg van verlies van spinimpulsmoment naar het niet-magnetische metaal. Men veronderstelt dat het verlies van impulsmoment wordt veroorzaakt door een stroom van spins door het interface het niet-magnetische metaal in, hoewel geen netto-overdracht van lading plaatsvindt, het zogenaamde “spinpompeffect.” Dit effect biedt verscheidene mogelijkheden vanuit het perspectief van toepassingen, zowel voor het creëren van een spinstroom door een niet-magnetisch metaal, als voor het controleren van de uitdemping in dunne ferromagnetische lagen. Er is een systematische studie uitgevoerd met geheel optische metingen aan samples met twee verschillende magnetische metalen (CoFe en NiFe) en een heel bereik aan diktes van de niet-magnetische laag (0 to 30 nm Ta wiggen). Uit de resultaten bleek dat de toename van de dempingparameter als gevolg van het spinpompeffect werd gemaskeerd door grote secundaire effecten op de morfologie van de FM laag. De groeiomstandigheden en de variërende dikte van de laag Ta eronder leidden tot een granulaire structuur van de ferromagneet, waardoor magnetische domeinen ontstonden. Bij het onderzoek naar de invloed van de Ta-dikte op de morfologie van de ferromagneet is een bijna lineair verband gevonden tussen de dempingparameter van de precessie en de coerciviteit van de ferromagneteische laag. Vergelijking van de metingen met ander recent onderzoek uit de literatuur geeft de noodzaak aan om de kwaliteit en uniformiteit van de magnetische laag na te gaan, alvorens een toename van de demping eenduidig toe te schrijven aan het spinpompeffect.

De experimenten beschreven in dit proefschrift hebben ertoe bijgedragen dat we de snelle dynamica in meerlaagse microstructuren nu beter begrijpen en kunnen beheersen, zodat we nu een stap dichter zijn bij het schrijven van een magnetische choreografie op een sub-nanosecondetijdschaal.
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