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Towards an all-optical magnetization switch, based on ultrafast laser pulse induced spin-transfer-torque

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Towards an all-optical magnetization switch, based on ultrafast laser pulse induced spin-transfer-torque

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Abstract

A new and exciting way to manipulate magnetization on the nanoscale is laser-induced spin-transfer-torque in a non-collinear magnetic bilayer. Using a femtosecond laser pulse, a hot-electron spin current is generated in an out-of-plane magnetized spin injection layer, which flows through a non-magnetic spacer layer into an in-plane magnetized spin absorption layer. The spin current exerts a torque on the in-plane layer within a few hundreds of femtoseconds, thereby canting the magnetization towards the surface normal. This results in precessional motion of the in-plane layer around the surface normal, eventually leading to a switch of the in-plane magnetization. Up to date, switching by laser-induced spin-transfer-torque is not reported in literature. In this thesis, laser-induced spin-transfer-torque switching is investigated by varying the compositions of the spin injection layer and the spin absorption layer.

To study the feasibility of using the non-collinear magnetic bilayers as a two-state memory element, a linear equation between the required canting angle for a switch of the in-plane layer and its damping factor was derived. This equation was confirmed by simulations of the magnetization after optical excitation. A factor $R_{CA}$ was introduced to quantify the quality of a magnetic bilayer in terms of switching by optical induced spin-transfer-torque. In this context, a minimum condition for switching is that $R_{CA} \geq 1$. A reference sample, of which the composition was based on literature, had a $R_{CA}$ factor of $0.45 \pm 0.05$. In order to increase the $R_{CA}$ factor above one, several optimizations of the reference sample have been investigated.

It was estimated that the absorption of transverse spins in a cobalt layer occurs within four monolayers. Furthermore, by decreasing the cobalt layer thickness, the $R_{CA}$ factor was increased to $0.69 \pm 0.08$. Decreasing the saturation magnetization of the spin absorption layer with a nickel layer resulted in an increase of the canting angle and consequently in a $R_{CA}$ factor of $0.8 \pm 0.1$. By optimizing the composition of the cobalt/nickel out-of-plane layer in the reference sample, an increase to $R_{CA} = 1.2 \pm 0.1$ was accomplished. With such a sample, the required demagnetization of the spin injection layer is still much larger than typical observed. It is predicted that a further optimization of spin injection - spin absorption combination will lead to the first experimental observation of all-optical switching by ultrafast spin-transfer-torque.
# Contents

1 Introduction  
1.1 A fast journey through magnetic data storage .......................... 1  
1.2 Laser-induced ultrafast demagnetization .............................. 3  
1.3 Ultrafast optical spin-transfer-torque .................................. 5  
1.4 This thesis ................................................................. 6

2 Ultrafast laser-induced spin-transfer-torque  
2.1 Magnetism ................................................................. 9  
2.1.1 Ferromagnetism ......................................................... 10  
2.1.2 Magnetic anisotropy ..................................................... 11  
2.1.3 Hysteresis ............................................................... 12  
2.2 Demagnetization .......................................................... 14  
2.2.1 Microscopic three temperature model (M3TM) ....................... 14  
2.2.2 Superdiffusive spin transport ......................................... 17  
2.3 Current induced spin-transfer-torque (STT) ............................ 19  
2.3.1 Spin current density flux and STT .................................. 20  
2.3.2 STT induced by a free electron current ............................ 21  
2.4 Precession of the IP layer in an IP field ............................... 23  
2.4.1 Equilibrium angle of the OOP layer ............................... 23  
2.4.2 Precession of the IP layer .......................................... 24  
2.5 All-optical switching by laser-induced optical spin-transfer-torque .... 26

3 Methodology ................................................................. 29  
3.1 Sample fabrication ....................................................... 29  
3.2 Magneto-optical Kerr effect (MOKE) .................................. 31  
3.2.1 Physics of MOKE ...................................................... 31  
3.2.2 Static MOKE .......................................................... 32  
3.2.3 Time-Resolved MOKE (TR-MOKE) ................................ 33
3.3 Vibrating sample magnetometry (VSM) ........................................... 35
3.4 Anomalous Hall effect (AHE) .................................................. 36

4 Experiment & data analysis ....................................................... 39
4.1 The experiment ................................................................. 39
  4.1.1 General geometry of the samples & the experiment ................. 39
  4.1.2 Key parameters: efficiency & canting angle ......................... 41
4.2 Data analysis routine ......................................................... 41
4.3 Discussion of the data analysis routine .................................. 48

5 Study of the in-plane layer using simulations with the LLG equation .... 51
5.1 Geometry and the LLG equation for Matlab .............................. 51
5.2 Field dependent frequency of precession .................................. 52
5.3 Switching by optical induced spin-transfer-torque ....................... 53
  5.3.1 Required canting angle without uniaxial IP anisotropy ............. 54
  5.3.2 Required canting angle with uniaxial IP anisotropy ............... 56
5.4 Time scale of a complete switch (the switching time) ................. 57
5.5 Canting angle as function of spin current .............................. 58
5.6 Conclusions ................................................................. 61

6 Investigating ultrafast laser-induced spin-transport-torque ............... 63
6.1 Geometry of the reference sample ....................................... 63
6.2 Static magnetic properties of the reference sample .................... 64
  6.2.1 Saturation magnetization of the spin injection layer .............. 65
  6.2.2 Faraday subtraction of static MOKE hysteresis loops ............ 65
  6.2.3 Static MOKE hysteresis loop ....................................... 66
6.3 Magnetization dynamics of the reference sample ....................... 67
6.4 Helicity and fluence dependency ......................................... 69
  6.4.1 Helicity of the pump beam ......................................... 69
  6.4.2 Fluence of the pump beam ......................................... 70
6.5 Discussion of the reference sample ..................................... 71
6.6 Investigating the spin injection layer ................................... 73
  6.6.1 Increase of the Co:Ni ratio with constant Co dusting layer .... 74
  6.6.2 Co dusting layer thickness in the Ni/Co/Cu interface ............ 76
  6.6.3 Summary .......................................................... 78
6.7 Cobalt spin absorption layer wedge ..................................... 79
  6.7.1 Static magnetic properties ....................................... 80
  6.7.2 Magnetization dynamics ........................................... 80
  6.7.3 Canting angle of the Co spin absorption layer .................... 82
6.8 Nickel spin absorption layer .............................................. 84
6.9 Discussion of the spin injection and absorption layer ............... 86

7 Conclusions &
  Recommended studies .......................................................... 87
  7.1 Physics of laser-induced spin currents ............................... 88
  7.2 Optical two-state memory element .................................... 88
  7.3 Recommended studies .................................................. 89
    7.3.1 Physics of laser-induced spin currents ......................... 90
7.3.2 Optical two-state memory element ........................................... 90

References ................................................................. 93

A Solutions of the LLG equation for the IP layer .......................... 101
   A.1 Steady precession around a fixed field .................................. 101
   A.2 Damped precession around a fixed field ................................. 102

B Switching of the IP layer by the demagnetization field .............. 105
   B.1 Critical canting angle as function of Gilbert damping factor .......... 105
   B.2 Time scale of switching ............................................... 109

C LLG equation for modelling .............................................. 111

D Effect of tantalum seeding layer on PMA of the OOP layer ........... 113
In the present world it is hard to find someone who is not connected to the digital universe. The amount of online data is currently estimated at about 9 zettabytes ($10^{21}$ bytes) and is predicted to increase towards 44 zettabytes in 2020 \cite{1}. These numbers do not even include the data stored on local drives. These huge numbers demonstrate the urgency for ever increasing data densities on storage devices and along with it, the need for faster processing of this data. Future solutions to these demands are promised by spintronic devices: devices using electron spin instead of electron charge as the information carrier. In this chapter, a fast journey through magnetic data storage is provided, after which a promising new way of processing magnetic data is introduced: optical manipulation of magnetization. Finally, the contribution of this thesis in the search for all-optical memory devices is discussed.

1.1 A fast journey through magnetic data storage

The first company to introduce commercial devices using magnetic data storage was IBM. The first hard disk ever was the IBM 350, which had a data density of 2 kbit/in$^2$. Writing a magnetic bit was done by applying a current through a coil, inducing a magnetic field. This field forces the magnetization of a magnetic bit along the desired direction. Reading is done in the reverse way: the magnetic field of a magnetic bit induced a current through the coil, which is then measured to determine the state of the magnetic bit. A revolutionary improvement towards nowadays data density records above Tbit/in$^2$ came in the end of the eighties. Fert \cite{2} and Grünberg \cite{3} separately discovered the giant magnetoresistance (GMR), for which they were granted the Nobel prize in physics in 2007. The GMR can be explained as follows. A current flowing through a ferromagnet is separated in a spin-up channel and a spin-down channel. Since the density of states at the Fermi level in a ferromagnet is different for spin-up and spin-down electrons, these two current channels experience different resistances. Consider a simple electric circuit, containing two ferromagnets separated by a non-magnetic metal, as drawn in figure 1.1. As is calculated in the figure, the total resistance of the circuit is different for parallel and anti-parallel alignment of the magnetization in the ferromagnetic elements. The difference in resistance $\Delta R/R$ is called GMR and is given by:
\[
\frac{\Delta R}{R} = \frac{(R_H - R_L)^2}{4R_H R_L}, \tag{1.1}
\]

where \(R_H\) and \(R_L\) are the high and low resistances due to the ferromagnets. The use of GMR in read heads of hard disk drives led to an increase of the data density to 3 Gbit/in\(^2\) in the first commercial hard disk drive using GMR. A further improvement was made by Seagate, the first company to use tunnelling magnetoresistance (TMR) in hard disks. TMR is the equivalence of GMR, but in TMR the two ferromagnets are separated by a thin insulator, resulting in much larger magnetoresistance as compared to GMR, increasing possible data densities.

\[\frac{1}{R_P} = \frac{1}{R_{\uparrow}} + \frac{1}{R_{\downarrow}} = \frac{R_H + R_L}{2R_H R_L}\]

\[\frac{1}{R_{AP}} = \frac{1}{R_{\uparrow}} + \frac{1}{R_{\downarrow}} = \frac{2}{R_H + R_L}\]

Figure 1.1: Electronic circuit used to measure GMR. The total resistance \(R\) depends on the orientation of the magnetization in the two ferromagnets.

However, write heads still consist of coils inducing a magnetic field to write a magnetic bit. This makes it difficult to scale memory storage to the nanoscale. The fact that hard disks require moving parts makes it difficult to embed it on chips. A promising device being investigated by both academia and industry is the so-called spin-transfer-torque magnetic random access memory (STT-MRAM). The physics behind spin-transfer-torque will be explained in detail in chapter 2. Briefly explained, a spin polarized current flowing through a ferromagnetic layer with free magnetization cant that magnetization towards the polarization of the spin current. A schematic drawing of the STT-MRAM device is shown in figure 1.2. The magnetic bits are made of tunnelling junctions: one fixed ferromagnetic layer and one free ferromagnetic layer, separated by a thin insulator. Attached below and above it are bit and word lines at right angles with each other. By sending a large current density (of the order of \(10^7\) A/cm\(^2\)) through the corresponding bit and word line, a single magnetic bit can be selected. The current gets polarized along the fixed magnetization, tunnels through the tunnel barrier and exerts a STT on the free ferromagnetic layer to write the magnetic bit. To read the bit, the bias voltage over the bits is smaller, which enables read out of the magnetic bit using TMR. The STT-MRAM is advantageous over other RAM technologies in that it is non-volatile (its bits are preserved after switching off the device), it has low power consumption and with a typical operating time of 2 to 30 ns it is quite fast.
CHAPTER 1. INTRODUCTION

A final technology being investigated by many researchers and worth mentioning is the racetrack memory proposed by Parkin [5]. In this memory, a current is sent through a wire containing magnetic bits. The current applies a spin-transfer-torque on the magnetization in the wire, effectively moving the magnetic bits towards a read and write head at a fixed location. In this way no moving parts are required, increasing the stability of the memory device.

1.2 Laser-induced ultrafast demagnetization

The processing times of the STT-MRAM devices introduced in the previous section are of the order of nanoseconds [6]. Exploration of faster time scales started with the discovery by Beaurepaire et al. of ultrafast demagnetization of a nickel layer upon femtosecond laser excitation [7]. This ultrafast demagnetization is shown in figure 1.3a. The figure shows the normalized magnetization as a function of time after laser excitation. The graph shows a clear demagnetization of almost 50% within a few hundreds of femtoseconds. This was explained by thermal arguments as follows. The electronic system heats up by the laser pulse and exchanges energy with the lattice and spins, as schematically drawn in figure 1.3b. Due to heating of the spin system, the magnetization reduces on an ultrafast time scale, because it is temperature dependent. This model was extended by Koopmans et al. to explicitly include the conservation of angular momentum [8]. This extended model (the microscopic three-temperature model) will be explained in more detail in chapter 2. The dissipation channel for angular momentum in the (microscopic) three-temperature model is local.

Although many experiments seemed to support the theory of heating to explain ultrafast demagnetization [9–12], other theories were evolving to include non-local demagnetization effects: the transport of spin away from the laser spot. One means of diffusive spin transport is due to the spin dependent Seebeck effect. The Seebeck effect induces a current in a thermal gradient. This thermal gradient is created by heat transport from the laser pulse into the structure. In a ferromagnetic material the Seebeck coefficient, which characterizes the rate of current generation, is spin dependent. The result is a spin polarized current. Evidence for this mechanism was observed by Choi et al. [13]. By comparing the spin-transfer-torque on a CoFeB layer exerted by a spin current generated in a Co/Ni multilayer versus a Co/Pt multilayer, they concluded that the spin current was generated by the spin-dependent Seebeck effect. Another spin current generation mechanism is the excitation of hot electrons in a ferromagnet with differences in lifetimes of spin-up and spin-down electrons. This superdiffusive spin current was studied

Figure 1.2: Schematically drawn example of a STT-MRAM. The working principle of the STT-MRAM is explained in the main text. Courtesy to Schellekens [4].
Figure 1.3: (a) Observation of ultrafast demagnetization in a nickel film. Courtesy to Beaurepaire et al. [7]. (b) Schematically drawn explanation of the ultrafast demagnetization on thermal grounds. The system is divided in three subsystems. The electronic system is heated by a laser pulse and exchanges energy with the other systems.

Theoretically by Battiatio et al. [14, 15]. The principles behind creation of superdiffusive spin currents will be addressed in chapter 2. The model of ultrafast demagnetization based on spin currents was supported by many publications [16–19]. On the other hand, experimental results against such a demagnetization mechanism was reported as well [11, 12, 20].

One of the first publications to provide evidence of spin transfer by spin currents was by Malinowski et al. in 2008 [16]. By comparing the demagnetization characteristics of two Co/Pt layers separated by a conductive or an insulating spacer, they showed that the demagnetization of these Co/Pt layers was dependent on their mutual magnetization alignment if the spacer layer was conductive, whereas the demagnetization was mutual independent with an insulating spacer. More evidence of spin currents was observed by Rudolf et al. [17], who studied demagnetization characteristics in an iron/ruthenium/nickel structure. For anti-parallel alignment of the magnetization in the iron and nickel layers, demagnetization was observed of both layers. In contrast, for the parallel alignment an enhancement of the iron magnetization was observed, even to values above the saturation magnetization. This was explained by the absorption of extra electrons from the nickel layer. In the recent years, more evidence for spin transfer mediated demagnetization was observed. Eschenlohr et al. [19] investigated a nickel layer buried below a thick gold layer. By laser excitation of the gold layer, a superdiffusive hot-electron current created in the gold layer flows through the nickel layer. In the nickel layer, spins aligned with the magnetization experience large inelastic scattering, whereas spins anti-parallel to the magnetization experience small scattering. The result is a net accumulation of spins parallel with the magnetization in the nickel. Since spin and magnetization are related to each other in an anti-parallel fashion, this resulted in demagnetization of the nickel layer.
As is clear from the discussion above, the scientific community had been in strong disagreement whether ultrafast demagnetization of a ferromagnetic layer results from thermal effects or spin transport effects. However, recently it is argued that the demagnetization process is mediated by both thermal effects and spin transport [18, 21]. Convinced by the presence of hot-electrons spin currents, new ways for the ultrafast application of spin-transfer-torque in for example STT-MRAM devices are being investigated, as will be discussed in the next section.

1.3 Ultrafast optical spin-transfer-torque

In 2014 Schellekens et al. introduced a new way of all-optical control of the orientation of the magnetization in a thin film in a non-collinear magnetic bilayer, using excitation of superdiffusive spin currents [22]. After their publication, this was also done by Choi et al. using spin currents generated by the spin-dependent Seebeck effect (see previous section). The relevant layers of the investigated structures are drawn schematically in figure 1.4. The core of the structure consists of a perpendicularly magnetized layer and an in-plane magnetized layer, separated by a conductive layer, as is shown in figure 1.4a. When the structure is excited by a laser pulse, both ferromagnetic layers demagnetize. Along with the demagnetization, a superdiffusive spin current is generated from the perpendicular magnetized layer to the in-plane magnetized layer and vice versa, as is shown in figure 1.4b. The spin current from the bottom layer to the top layer exerts a spin-transfer-torque on the magnetization in the in-plane layer, resulting in a canted state of the magnetization, as drawn in figure 1.4c. By applying a magnetic field along the in-plane axis, the canted magnetization starts precessional motion. This oscillation can be measured using time-resolved magneto-optical Kerr effect measurements, as will be explained in chapters 3 and 4.

![Figure 1.4: Cartoon of the experiment performed by Schellekens et al. [22]. (a) Before laser excitation, the bottom layer is perpendicularly magnetized and the top layer is magnetized in-plane. The ferromagnetic layers are separated by a conductive spacer layer. (b) The laser pulse hits the sample and generates superdiffusive spin currents between the two ferromagnetic layers. (c) The spin current from the bottom layer to the top layer exerts a spin-transfer-torque on the top layer, canting its magnetization towards the surface normal.](image-url)
In figure 1.5 results are shown of the magnetization-related Kerr rotation as function of time after laser excitation [22]. The oscillating signal caused by the precessing magnetization is clearly visible. The signal shows a double-frequency oscillation. One oscillation comes from the bottom layer, excited by another excitation mechanism than spin-transfer-torque. This mechanism is not relevant for the experiments performed through this thesis. It will be briefly discussed in section 4.3. The relevant frequency for this thesis is the spin-transfer-torque induced oscillation in the top layer. By analysing the data, precessional characteristics were deduced, proving that this frequency was resulting from a laser-induced spin-transfer-torque on the in-plane layer. Some important conclusions of relevance for this thesis were drawn. Firstly, it was shown that the spin-transfer-torque was exerted by laser-induced superdiffusive spin currents on an ultrafast time scale of several hundreds femtoseconds. A contribution from the spin-dependent Seebeck was excluded by calculations, proving that it is indeed a superdiffusive spin current which exerts the spin-transfer-torque. Secondly, it was concluded that the number of spins transported to the in-plane layer was approximately 2% of the spins participating in the demagnetization process of the bottom layer. Finally, the spin-transfer-torque resulted in a canting angle of the top layer of 60 mdegrees. In this thesis, the efficiency and the canting angle will be increased.

Figure 1.5: Double-frequency signal measured using the time-resolved magneto-optical Kerr effect. Courtesy to Schellekens et al. [22].

1.4 This thesis

Motived by the results published by Schellekens et al., this thesis will continue the search to possibilities of ultrafast laser-induced spin-transfer-torque in data storage applications. It will be shown that the number of spins transported to the top layer can be increased by optimizing the non-collinear magnetic bilayer. This leads to an enhanced canting angle of the magnetization of the top layer. Simulations of the time evolution of the magnetization in the top layer will provide insight in the required canting angle to switch the top layer. It will be argued that ultrafast laser-induced spin-transfer-torque is a promising technology for application in a two-state memory element. This thesis is constructed of the following chapters:

- Chapter 2: Ultrafast laser-induced spin-transfer-torque. The important concepts needed to understand the experiments are introduced. A brief introduction into magnetism is given. The two models explaining ultrafast demagnetization are introduced in more detail. A brief introduction of spin-transfer-torque and its effects on a ferromagnetic layer are introduced.
• **Chapter 3: Methodology.** The magneto-optical Kerr effect (MOKE) and the experimental setup to measure the MOKE are introduced. It is explained how static magnetic properties of ferromagnetic layers are measured and how the samples investigated in this thesis are fabricated.

• **Chapter 4: Experiment & data analysis.** Since the analysis of the data is far from trivial, this chapter is devoted to a discussion on the data analysis and how important quantities are deduced.

• **Chapter 5: Study of the in-plane layer using simulations with the LLG equation.** Simulations of the magnetization dynamics are done to investigate the characteristics of switching the in-plane layer.

• **Chapter 6: Investigating ultrafast laser-induced spin-transfer-torque.** This chapter provides the results of the experiments performed throughout this thesis. It will be shown that the physics of laser-induced superdiffusive spin currents is highly dependent on the specific composition of the magnetic bilayer, especially the morphology of its interfaces. It will be shown that the non-collinear magnetic bilayer can be optimized towards a switch of the in-plane layer.

• **Chapter 7: Conclusions & recommended studies.** The main conclusions of this thesis are drawn. Further suggestions towards switching the in-plane layer are proposed.
In this thesis, the ultrafast optical excitation of magnetization dynamics is investigated. In this chapter the theory behind the ultrafast magnetization dynamics is discussed. First, the origin of magnetism is described. Secondly, two models describing ultrafast, optically induced, demagnetization are introduced. These models are the microscopic three temperature model and the superdiffusive spin transport model. Finally, it is discussed how the superdiffusive spin current can cause an ultrafast spin-transfer-torque, which results in precessional motion of the in-plane magnetization in non-collinear magnetic bilayers.

2.1 Magnetism

Materials can exhibit different kinds of magnetism. Most elements do not have a spontaneous magnetization at room temperature. When these materials are placed in an external magnetic field, a magnetization is induced by the field. The behaviour of the induced magnetization is characterized by its magnetic susceptibility $\chi$. Diamagnetic materials have a negative susceptibility, meaning that the induced magnetization aligns itself anti-parallel to the field. Paramagnetic materials have a positive susceptibility, meaning that the induced magnetization aligns itself parallel to the field. The magnetic materials used throughout this thesis are ferromagnets, which exhibit a spontaneous magnetization up to a characteristic temperature, called the Curie temperature $T_C$. Above these this temperature the spontaneous magnetization disappears and the materials becomes paramagnetic. The ferromagnetic elements used in this thesis are cobalt (Co) and nickel (Ni). The Curie temperature of these elements is above room temperature: 1388 K for Co and 627 K for Ni [23], well above room temperature.
2.1.1 Ferromagnetism

To understand the origin of ferromagnetism (FM), consider an electron with electron mass \( m_e \) and charge \(-e\), orbiting a nucleus [23]. The electron carries an orbital angular momentum \( \vec{L} \), which carries an orbital magnetic moment. Besides the (classical) orbital angular momentum, electrons carry a quantum physical spin angular momentum \( \vec{S} \) as well. This spin angular moment gives the electron a spin magnetic moment \( \vec{\mu}_S \) of:

\[
\vec{\mu}_S = \gamma_e \vec{S},
\]

(2.1)

where \( \gamma_e = -g_e \frac{e}{2m_e} \) is the gyromagnetic ratio and \( g_e = 2 \) the Landé g-factor. The two magnetic moments described above add up to a total electron magnetic moment of \( \vec{\mu}_e = \vec{\mu}_L + \vec{\mu}_S \). The magnetization \( \vec{M} \) of a material is defined as the total magnetic moment per unit volume \( V \). In 3d transition metals like Ni and Co the orbital moment is highly quenched [24]. To account for the small orbital moment, \( g_e \) is increased, for Co \( g_e = 2.18 \) and for Ni \( g_e = 1.21 \). This means that the magnetization in these materials can be calculated by the spin magnetic moments, using the adjusted Landé g-factor:

\[
\vec{M} = \frac{\sum \vec{\mu}_e}{V} = n_S \vec{\mu}_S,
\]

(2.2)

where \( n_S \) is the spin density. The fundamental concept explaining ferromagnetism is called the exchange energy, which aligns the spins in a FM material without the presence of an external field [25]. Coulomb interaction between two electrons increases the energy of the electron system due to the charge repulsion when electrons approach each other. According to the Pauli exclusion principle the total wave function of two identical fermions (such as electrons) has to be antisymmetric under exchange of the two fermions. The total wave function of an electron system is the product of a spin part and an orbital part. In the case of parallel spins, the spin wave function is symmetric. In order to turn the total wave function antisymmetric, the orbital part has to be antisymmetric. This antisymmetry in orbital wave function is achieved by separating the electrons in different orbitals. As a result, the Coulomb energy decreases as compared to the electron configuration with anti-parallel spin alignment, where the electrons are closer to each other in the same orbital. The exchange energy \( E_{ex} \) is defined as the energy difference between the states with parallel and anti-parallel spins. Thus the exchange energy acts to align the spins parallel, creating spontaneous magnetism.

At zero temperature, the spontaneous magnetization is saturated with a value of \( M_{sat,T=0} \). The thermal energy of the FM material is given by \( k_B T \), where \( k_B \) is the Boltzmann constant and \( T \) temperature. If the thermal energy exceeds the exchange energy, the coupling between spins breaks down and the spins become randomized. The temperature associated with this randomization is called the Curie temperature. Above the Curie temperature, the spontaneous magnetization disappears and the material becomes paramagnetic. Using mean-field theory, the maximum spontaneous magnetization \( M_{sat} \) can be calculated as a function of temperature [23]. In figure 2.1 a schematic graph of the temperature dependence of the \( M_{sat} \) is shown. This graph will be used in section 2.2.1 to understand ultrafast demagnetization.
2.1.2 Magnetic anisotropy

One of the most widely used properties of magnetism is magnetic anisotropy [26]. A magnetic material can exhibit one or more directions along which the electron-spin system is in a lower energy state than it is along another direction. As a result, the magnetization aligns itself along this preferential direction. The axis along the preferential direction is called an easy axis. The axis in which the system is in a high energy state is called a hard axis. In general, two mechanisms are responsible for anisotropy, being the dipole-dipole interactions and the spin-orbit coupling. For thin films as used in this thesis the total energy density arising from both mechanisms can be combined in a single magnetic anisotropy energy density $U_{\text{ani}}$, which is given by:

$$U_{\text{ani}} = -K_{\text{ani}} \sin^2 (\theta_M).$$  \hspace{1cm} (2.3)

Here, $K_{\text{ani}}$ is the anisotropy constant characterizing all anisotropies arising from dipole-dipole interactions and spin-orbit coupling. The angle $\theta_M$ is defined by the magnetization and the film surface. For negative $K_{\text{ani}}$ the lowest energy state of the system is an in-plane (IP) magnetized film. This is called in-plane magnetic anisotropy (IMA). If $K_{\text{ani}}$ is positive, the system favours out-of-plane (OOP) magnetization alignment along the surface normal. In this case, the system has perpendicular magnetic anisotropy (PMA).

The top layer of the structures investigated in this thesis (see section 1.3) has in-plane magnetic anisotropy. IMA is mainly caused by dipole-dipole interactions. At the boundaries of a layer, free magnetic dipoles exist. These dipoles induce a so-called demagnetization field, which acts to align the spins in the layer along the in-plane axis. The associated anisotropy is called shape anisotropy with an anisotropy constant $K_{\text{shape}}$:

$$K_{\text{shape}} = -\frac{1}{2} \mu_0 M_{\text{sat}}^2.$$  \hspace{1cm} (2.4)

The bottom layer of the investigated structures has perpendicular magnetic anisotropy, which is caused by spin-orbit coupling. Spin-orbit coupling is a relativistic effect which arises because
of the high velocities of electrons orbiting a positive nucleus. The orbital motion is influenced by the lattice via the Coulomb potential $V_C$. In the reference frame of such an electron, the electric field arising from $V_C$ translates to a magnetic field. The magnetic moment of the electron spin interacts with this magnetic field and hence the spin is coupled to the electron’s orbital motion. At the interface between a ferromagnetic layer and a non-magnetic layer, the orbitals of the ferromagnetic layer have hybridisation with the orbitals of the non-magnetic layer. Due to spin-orbit coupling this results directly in magnetocrystalline anisotropy. This magnetocrystalline anisotropy can result in PMA by choosing an interface with a large bonding of the ferromagnetic layer with the adjacent layer.

The anisotropy constant $K_{ani}$ as introduced in equation 2.3 can be split into a volume term $K_V$ and a surface term $K_s$. The volume term is caused by the dipole-dipole interactions and is equal to $K_{shape}$. The surface term is mainly caused by the spin-orbit coupling. Considering the sign conventions introduced with equation 2.3, $K_V$ is always negative. $K_s$ is negative if it favours IP alignment and positive if it favours OOP alignment. For an ultrathin FM layer sandwiched between two non-magnetic (NM) layers, the overall anisotropy constant $K_{ani}$ can be calculated by:

$$K_{ani} = K_V + \frac{K_{s,1} + K_{s,2}}{t_{FM}},$$

(2.5)

where $K_{s,1(2)}$ stands for the surface anisotropy constant of the first (second) FM/NM interface and $t_{FM}$ is the thickness of the FM layer. Consider a $K_s$ which favours OOP alignment. Since $K_V < 0$ and $K_{s,1(2)} > 0$ in that case, there is a thickness for which $K_{ani} = 0$. This thickness is called the critical thickness $t_{FM,crit}$ and can be calculated by:

$$t_{FM,crit} = -\frac{K_{s,1} + K_{s,2}}{K_V}.$$  

(2.6)

From a physical point of view, this means that the FM layer changes from an OOP magnetized layer to an IP magnetized layer at a thickness of $t_{FM,crit}$. For quantitative calculations and simulations the anisotropy can be regarded as an anisotropy field $H_{ani}$, written as:

$$H_{ani} = \frac{2K_{ani}}{\mu_0 M_{sat}} \sin (\theta_M).$$

(2.7)

### 2.1.3 Hysteresis

The static behaviour of ferromagnetic layers is investigated by measuring the magnetization as function of an external magnetic field. The resulting curve is called a hysteresis loop. Hysteresis means that the behaviour of the FM layer depends on the historical state of the magnetization. According to the Stoner-Wohlfarth theory \[27\], the direction of the magnetization in an arbitrary external applied field is determined by the balance of the anisotropy energy density (given by equation 2.3) and the Zeeman energy density. The Zeeman energy density $U_M$ originates from an external applied field and is calculated by:

$$U_M = -\mu_0 \vec{M} \cdot \vec{H}_{eff},$$

(2.8)
where $H_{\text{eff}}$ is the effective field, consisting of the external applied field and anisotropy fields. The total energy density $U_{M,\text{total}}$ of the magnetic system of a magnetized layer is determined by the sum of these two densities:

$$U_{M,\text{total}} = -K_{\text{ani}} \sin (\theta_M)^2 - \mu_0 H_{\text{app}} M_{\text{sat}} \cos (\theta_M - \phi_H),$$

(2.9)

where $\phi_H$ is the angle of the field $H_{\text{app}}$ with the surface plane. In this thesis, the field is aligned along the surface normal when examining the magnetostatic behaviour: $\phi_H = \pm \pi/2$. For PMA layers this results in an easy-axis loop, since the field is applied along the magnetization’s easy axis. For IMA layers the loop is called a hard-axis loop, since the field is applied along the magnetization’s hard axis in that case.

Consider the hysteresis loop for a PMA layer in an OOP field as schematically shown in figure 2.2a. For a PMA layer the global minima of function 2.9 are $\theta_M = -\pi/2$ for negative fields and $\theta_M = +\pi/2$ for positive fields. Starting at large negative field, the magnetization is thus in a minimum energy state for $\theta_M = -\pi/2$. The magnetization saturates therefore in the spin-down state as indicated by blue arrow (a). Increasing the field to a positive value, the new state with minimum energy becomes $\theta_M = +\pi/2$. However, at small field strengths the state with $\theta_M = -\pi/2$ is still a local minimum because of the anisotropy energy. The anisotropy energy acts as an energy barrier to be overcome to flip to the magnetization’s new global minimum of energy. The magnetization stays therefore in the spin-down state (blue arrow (b)). If the field is increased to the coercivity $+H_c$, the Zeeman energy is large enough to overcome the energy barrier of the anisotropy energy and the magnetization switches to its (saturated) spin-up state as indicated by blue arrow (c). Starting at large positive field the magnetization stays in this spin-up state (blue arrow (d)) until the field is decreased to the reverse coercivity $-H_c$. At this field the magnetization flips to its spin-down state.

In the case of an IP layer in an OOP field, the field is applied along the material’s hard axis. The minima of equation 2.9 are now changing as function of the field strength. This results in a magnetization angle which is gradually varying as function of field strength. A typical hysteresis loop for an IP layer in an OOP field is shown in figure 2.2b. At zero field the magnetization is aligned along the easy axis as indicated by blue arrow (a). At large negative field the magnetization cant slowly to the negative OOP direction as indicated by blue arrow (b). At larger fields the magnetization is increasingly canted until it is saturated in the OOP spin-down state for the saturation field $-H_{\text{sat}}$ (blue arrow (c)). Sweeping the field from $-H_{\text{sat}}$ to zero field the magnetization gradually recovers to the IP direction. For positive fields the magnetization is canted towards the positive OOP direction until it is saturated in the OOP spin-up state as indicated by the blue arrows (d) and (e). The saturation field is the field strength when the applied field opposes the anisotropy field. The saturation field is given by equation 2.7, considering that the angle $\theta_M = \pm \pi/2$, for the magnetization saturated along the surface normal.
Figure 2.2: Schematically drawn hysteresis loops. (a) Easy-axis loop for an OOP FM layer in an OOP field. The magnetization flips at a field strength of $\pm H_c$. (b) Hard-axis loop for an IP FM layer in an OOP field. The magnetization saturates in the OOP spin-down state for large applied negative field and slowly rotates to the IP direction at small fields. At positive field it rotates to the OOP spin-up state.

2.2 Demagnetization

Interaction of laser light with the spin system of a FM results in a decreased magnetization magnitude. The interaction between laser light and spin system occurs due to absorption of photon energy by the electron system of the FM layer. As was introduced in chapter 1, the scientific world is still in debate whether the disordering of the spin system is a result of thermal processes or transport processes. In this section both models are described.

2.2.1 Microscopic three temperature model (M3TM)

To describe the ultrafast quenching of the magnetization in a nickel film, Beaurepaire et al. introduced the phenomenological three temperature model (3TM) [7]. In this model, the magnetized system is divided in three subsystems: the electron subsystem at temperature $T_e$, the lattice subsystem at temperature $T_l$ and the spin subsystem at temperature $T_S$. Each subsystem $i$ has its own heat capacity $c_i$ and exchanges energy with the other subsystems $j$, which can be quantized with the coupling constants $g_{ij}$. A schematic overview of the three subsystems and their interactions is given in figure 2.3a. The pulsed laser excitation quickly heats the electron system with a few hundreds of Kelvins. Due to the exchange of energy, thermal equilibration takes places between the three subsystems. During this equilibration process the lattice and spin subsystems are heated. A heated spin subsystem results in a reduced magnetization. This is shown in figure 2.3b, where the temperature dependence of the magnetization is drawn (as
was introduced in section 2.1.1). Before laser excitation the spin subsystem has temperature and magnetization of the first blue dot (a). During equilibration the temperature of the spin subsystem changes with an amount of $\Delta T$. Due to this temperature increase the magnetization decreases, as indicated for the second blue dot (b).

![Figure 2.3: (a) Schematic drawing of the three subsystems in 3TM with their coupling constants. The three subsystems exchange energy due to the laser heated electron system. The spin subsystem is heated, which results in a decreased magnetization. (b) Temperature dependence of the magnetization of a FM, as introduced in section 2.1.1. The magnetization decreases with increasing temperature, according to the law of Curie.](image)

The 3TM explains demagnetization solely by considering exchange of energy between the three subsystems. The energy is exchanged due to inelastic electron-electron or electron-lattice scattering. Conservation of angular momentum was not explicitly considered in the 3TM. Conservation of angular momentum dictates that the total angular momentum of the system $\vec{J}_{\text{tot}} = \vec{L}_{\text{electron}} + \vec{S}_{\text{electron}} + \vec{L}_{\text{phonon}} + \vec{L}_{\text{photon}}$ is constant, where $\vec{L}_i$ and $\vec{S}_i$ refers to the orbital angular momentum and spin angular momentum respectively. The angular momentum transfer from the absorbed photon to the electron or spin system is negligible [28]. To account for conservation of angular momentum, Koopmans et al. [8] included a spin-flip with probability $a_{sf}$ at electron-lattice scattering events. This type of Elliot-Yafet scattering arises because of the relativistic spin-orbit interaction which was introduced in section 2.1.2. Furthermore, in M3TM heat diffusion from the ferromagnet to the ambience is included. In the microscopic three temperature model (M3TM) the time derivatives of the temperatures of the electron subsystem and the lattice subsystem are given by two differential equations, describing energy exchange between the laser heated electron subsystem and the lattice subsystem. The magnetization dynamics for $m = M/M_{\text{sat}}$ are governed by a differential equation describing the Elliot-Yafet type of spin-flip scattering. Combining the thermal equilibration process and spin-flip scattering, M3TM contains three differential equations:

$$c_e (T_e) \frac{dT_e}{dt} = P(t) + \nabla_z [\kappa \nabla_z T_e] + g_{el} [T_l - T_e]$$  \hspace{1cm} (2.10a)
\[
\frac{dT_1}{dt} = g_{el} [T_e - T_1] \quad (2.10b)
\]

\[
\frac{dm}{dt} = Rm \frac{T_1}{T_C} \left[ 1 - m \coth \left( \frac{mT_C}{T_1} \right) \right], \quad (2.10c)
\]

where \(\kappa\) is the electronic thermal conductivity and \(R\) is a measure of the demagnetization rate, which depends on \(\alpha_{sf}\). The term \(P(t)\) at the right-hand side of equation 2.10a describes electron subsystem heating due to the laser excitation. The second term describes thermal diffusion out of the ferromagnet. The schematic temporal time evolutions of the electron and lattice subsystems are shown in figure 2.4 for a nickel system (figure a) and a gadolinium system (figure b), along with the resulting demagnetization.

Figure 2.4: (a) Time evolution of the temperatures of the electron (red) and the lattice (blue) subsystems. The resulting demagnetization is shown with a green curve. The large value of \(R\) for this system (nickel) results in a one-step demagnetization. (b) Time evolution of the temperatures of the electron (red) and the lattice (blue) subsystems. The resulting demagnetization is shown with a green curve. The small value of \(R\) for this system (gadolinium) resulted in this case in a two-step demagnetization. Courtesy to Koopmans et al.[8].

Comparing figure 2.4a with figure 2.4b, it is seen that the demagnetizations of a nickel system and gadolinium proceed in a different fashion. The figure of merit is the demagnetization rate \(R\). Based on the value of \(R\), two regimes can be classified. For small \(R\) as in gadolinium, demagnetization is not finished before electron-lattice equilibration, resulting in a two-step demagnetization curve. However, the ferromagnets investigated in this thesis are classified in the regime of large \(R\). This means that the demagnetization is relatively quick and finishes before electron-lattice equilibration has been completed, resulting in a one-step demagnetization curve. For Co and Ni layers the typical demagnetization times are of the order of a few hundred femtoseconds. After demagnetization the magnetization recovers slowly to its equilibrium state.
2.2.2 Superdiffusive spin transport

Although the M3TM could explain the experimental observed demagnetization, it does not take into account the loss of angular momentum by spin transport. Since excited electrons travel at speeds of the order of nm/fs, it is evident that spin transport can be a source for demagnetization for structures on the nanoscale. Before introducing the physical model as derived by Battiato et al. [14, 15], an intuitive picture is presented to understand the concept of the generation of laser-induced spin currents in a FM layer.

Consider the spin-dependent density of states (DOS) of a non-magnetic (NM) metal as drawn in figure 2.5a. An example of such a metal is copper (Cu). For a normal metal the density of states is equal for electrons which have spin-up (↑) and electrons which have spin-down (↓). In the ground state all energy levels up to the Fermi level $E_F$ are filled. Consider the excitation of a spin-up electron in a $3d$ band at the Fermi level to a $4sp$-like band above the Fermi level. This electron absorbs the photon energy $E_{ph}$ and is excited to a state with energy $E = E_F + E_{ph}$ (event (i) in the figure). From this state it scatters back to a state with smaller energy (event (ii) in the figure). The available states for this inelastic scattering process are the states with energy between $E_F + E_{ph}$ and $E_F$. These states construct the phase space, which is depicted with the light-blue area A in figure 2.5a. Eventually the electron scatters to a state with an energy below the Fermi level (event (iii)), where it resides. An excited electron in the spin-down band experiences the same processes. The phase space for spin-down electrons to scatter back into after laser excitation is depicted by the light-blue area B in figure 2.5a. In a non-magnetic metal the phase spaces of spin-up and spin-down electrons are equal. This means that the current generated by optical excitation of electrons has no spin polarization in a non-magnetic metal.

Now consider the density of states for a ferromagnetic metal as schematically drawn in figure 2.5b. In a ferromagnetic metal the bands with spin-up and spin-down electrons are shifted with respect to each other. In the figure, the spin-up electron band is shifted downwards and the spin-down electron band is shifted upwards. The number of electrons with spin-up is therefore larger than the number of electrons with spin-down, as can be seen by the larger number of filled states below $E_F$. In this case, spin-up electrons are called majority electrons and spin-down electrons are called minority electrons. Consider again an electron which is excited to a state in the $4sp$-like band with an energy of $E = E_F + E_{ph}$. The largely filled majority electron band results in a small number of available states for scattering between energies $E_F + E_{ph}$ and $E_F$. The phase space for relaxation of excited electrons to a state below the Fermi level is thus small. The number of empty states above $E_F$ in the minority band is larger than the number of empty states above $E_F$ in the majority band. The phase space for hot electron relaxation is therefore larger for minority electrons than for majority electrons. This means that there are more available states for the minority electrons for inelastic scattering. The result is a larger scattering probability for minority electrons, resulting in shorter lifetimes as compared to majority electrons. The mean free path $\lambda_{mfp}$ is the product of the electron’s speed $v_e$ and lifetime $\tau_e$:

$$\lambda_{mfp} = v_e \tau_e. \quad (2.11)$$

Due to a smaller $\tau_{e,\downarrow}$, $\lambda_{mfp}$ is smaller for minority electrons. The small mean free path for minority electrons means that minority electrons travel a short distance after being excited by the laser. In contrast, the large mean free path of majority electrons means a large travel
Figure 2.5: (a) Density of states of a normal metal. (b) Density of states of a ferromagnetic metal. For both figures, the blue area shows the DOS for majority electrons, whereas the red area shows the DOS for minority electrons. Considering the process of electrons scattering back from their excited state (event (i)) to the Fermi level (events (ii) and (iii)), the phase space of majority electrons is indicated with a light-blue area A and the phase space of minority electrons is indicated with a light-blue area B. The phase space is different for majority and minority electrons in FM metals, as can be seen in figure b.

...
can be calculated by considering the continuity equation of the electron density \( n_e \) [15]:

\[
\frac{\partial n_e}{\partial t} + \frac{n_e}{\tau_e} = \left[ -\frac{\partial}{\partial z} \hat{\phi}_e + \hat{I} \right] S_e,
\]

(2.12)

where \( \hat{I} \) is the identity operator, \( \hat{\phi}_e \) the flux operator and \( S_e \) the total electron source. By considering the electron density for majority and minority electrons, the magnetization dynamics can be calculated by:

\[
M = 2\mu_B (n_\uparrow - n_\downarrow).
\]

(2.13)

Figure 2.6: (a) Geometry for the calculation of demagnetization due to a spin polarized current. The figure shows the scattering of majority and minority electrons. After an electron is excited by the laser (1), it starts its motion in a random direction on a straight trajectory until it suffers scattering with other electrons (2 and 3). The scattering event changes momentum and thus direction of the trajectory. Eventually the electron could pass the FM/NM interface (4). Some electrons experience scattering at the interface, from which they return to the FM layer or pass the spacer layer (5). After some scattering events the electron is scattered back to a state below \( E_F \) and stops its hot-electron like motion (6). Finally, during a scattering event the electron can flip its spin (7). Figure adapted from [14].

The transport of spin described by this equation belongs to the superdiffusive regime, meaning that the majority electrons experience only a few scattering events on their trajectory. For results of numerical simulations with this equation the reader is referred to references [14, 15]. In their numerical results they showed that indeed a decrease in the magnetization of the FM layer was calculated and an increase of the magnetization in the adjacent NM layer.

### 2.3 Current induced spin-transfer-torque (STT)

In section 2.2 it was explained how the excitation of hot electrons in a FM layer generates a spin polarized current in that FM layer. In the experiments performed in this thesis, the spin current is excited in an OOP layer and absorbed by an IP layer. When this spin current is absorbed by the IP layer, the spins in the spin current rotate to align themselves along the local magnetization, meaning that a torque is exerted by the magnetization of the IP layer. Due to conservation of angular momentum, this means that the spins of the spin current must exert an equal and opposite torque on the IP magnetization. In this section a theoretical background to calculate the STT exerted by the spin current on the IP magnetization is introduced. In the
calculation of this section the spin current density is quantized by the known tensor \( \vec{Q}_S \), which has components in both spatial and spin space. This spin current density exerts a STT on the magnetization in the IP layer \( \vec{M}_{IP} \). The calculations presented in this section can be found to a more detailed extend in reference [29].

### 2.3.1 Spin current density flux and STT

In order to calculate the STT, consider the spin density \( \vec{m} \) in the IP layer. The spin density can be changed by \( \vec{Q}_S \) entering the IP layer, spin-flip scattering of the spin current with the lattice \( (\delta \vec{m}/\tau_{\downarrow\uparrow}) \) and any external applied torque densities \( (\vec{\tau}_{\text{ext}}) \). In this section only the current-induced torque is investigated. It is furthermore assumed that spin-flip scattering of the spin current is zero. The time derivative of the spin density becomes then:

\[
\frac{\partial \vec{m}}{\partial t} = -\vec{\nabla} \cdot \vec{Q}_S = \vec{\tau}_{\text{current}}.
\]  

(2.14)

The equation shows that the divergence of \( \vec{Q}_S \) in a volume of FM material determines the torque exerted by the spin current on the magnetization in that volume. In figure 2.7 the geometry for the following discussion can be found. The NM/FM interface is situated at \( z = 0 \) and the spin current density flows along the \( z \)-axis. The magnetization is aligned along the \( y \)-axis. In general, the spin current density can be split in three components in spin space. Two of these components, \( Q_{s,xx} \) and \( Q_{s,zz} \) in figure 2.7, are transverse to the magnetization of the IP layer. In this notation the first index \( x, y, \) or \( z \) represent the spin polarization of the current in spin space and the second index \( z \) belongs to the travel direction of the spin current density in spatial space. Using the divergence theorem, integration of equation 2.14 shows that a current-induced torque on the magnetization of the IP layer arises from the net flux of spin current through the surfaces that enclose the IP layer. This means that the net flux of angular momentum is absorbed by the magnetization of the IP layer. The torque induced by the spin current density can thus be calculated by the total flux \( \Phi_{\text{STT}} \) of the spin current. The spin current density consists of an incident density \( \vec{Q}_{s,\text{in}} \), reflected density \( \vec{Q}_{s,\text{refl}} \) and transmitted density \( \vec{Q}_{s,\text{trans}} \). The STT exerted by the spin current density is then calculated by:

\[
\vec{\tau}_{\text{total}} = \left[ \vec{Q}_{s,\text{in}} + \vec{Q}_{s,\text{refl}} - \vec{Q}_{s,\text{trans}} \right] \cdot A_{\text{int}} \hat{z},
\]  

(2.15)

where \( A_{\text{int}} \) is the surface of the interface. Note that the STT is a vector in spin space because it is contracted with the unit vector along spatial \( z \).
2.3.2 STT induced by a free electron current

In the following discussion the STT of a spin current induced by free electrons will be considered. Before taking into account an ensemble of electrons, a single free electron is considered. The spin of the electron defines an angle $\theta_Q$ with the magnetization direction of the FM layer. The free electron is travelling from the NM layer to the IP layer along the $z$-axis, with wave vector $k \hat{z}$, where $k = \sqrt{2m_eE_e}/h$ and $h$ is Planck’s constant. Inside the IP layer the potential landscape for majority electrons $V_{\uparrow}$ and minority electrons $V_{\downarrow}$ differs with the exchange energy, as shown in figure 2.7b. The wave vectors of majority electrons $k_{\uparrow}$ and minority electrons $k_{\downarrow}$ are therefore different. In the following discussion it is assumed that the potential step for majority electrons is zero, $V_{\uparrow} = 0$ and $k_{\uparrow} = k$. For the minority electrons the potential step is thus $V_{\downarrow} = E_{ex}$ and $k_{\downarrow} = \sqrt{2m_e(E_e - E_{ox})}/h$. The wave functions for the incoming, reflected and transmitted electron can be calculated using general quantum mechanic principles [29]. The spin current densities associated with the incoming, reflected and transmitted single free electron are given by:

\[ \vec{Q}_{s,\text{in}} = \frac{\hbar^2 k}{2m_e} \left[ \cos(\theta_Q) \hat{y} + \sin(\theta_Q) \hat{z} \right] \]  

(2.16a)

\[ \vec{Q}_{s,\text{refl}} = \frac{\hbar^2 k_{\downarrow}}{2m_e} \left[ \frac{k - k_{\downarrow}}{k + k_{\downarrow}} \right]^2 \sin \left( \frac{\theta_Q}{2} \right)^2 \hat{y}. \]  

(2.16b)
\[ \vec{Q}_{s,\text{trans}} = -\frac{\hbar^2 k}{2m_e} \sin(\theta_Q) \sin ([k - k_\downarrow] z) \hat{x} + \frac{\hbar^2}{2m_e} \left[ k \cos \left( \frac{\theta_Q}{2} \right)^2 - k_\downarrow \left[ \frac{2k}{k + k_\uparrow} \sin \left( \frac{\theta_Q}{2} \right)^2 \right] \right] \hat{y} + \frac{\hbar^2 k}{2m_e} \sin(\theta_Q) \cos ([k - k_\downarrow] z) \hat{z} \] (2.16c)

It follows from these equations that \( \vec{Q}_{s,\text{in}} + \vec{Q}_{s,\text{refl}} - \vec{Q}_{s,\text{trans}} = 0 \) for the spin component along the IP layer’s magnetization. Applying equation 2.15 to a volume in the vicinity of the NM/FM interface, it is seen that only a transverse spin component exerts a spin-transfer-torque on the IP layer. The non-zero net flux of transverse spin current arises from the precession of spins inside the IP layer as predicted by equation 2.16c. This equation shows that in any material with exchange energy (resulting in \( k \neq k_\downarrow \)), the transmitted transverse spins (\( x \) and \( z \) components) precess around the magnetization. The spatial period \( L_{\text{prec}} \) of this precession is given by:

\[ L_{\text{prec}} = \frac{2\pi}{k - k_\downarrow}. \] (2.17)

This period extends over only a few atomic spacings for Co and Ni. In the experiments performed throughout this thesis spin-transfer-torque in an IP layer is investigated. Electrons of the spin current excited in the OOP layer have travelled different path lengths to any arbitrary point in the IP layer, because they are incident on the interface at different angles. As a result of summing over these electrons, the transverse spin components add destructively as a result of the aforementioned precession, resulting in spin dephasing. This cancelling of the transverse components can be regarded as an absorption of the angular momentum of the spin current density by the IP layer. The spin-transfer-torque can thus be calculated by:

\[ \vec{\tau}_{\text{STT, total}} = A_{\text{int}} \frac{\hbar^2 k}{2m_e} \sin(\theta_Q) \hat{z}. \] (2.18)

This equation shows that the STT exerted by the spin current is mainly determined by the electron energy and the spin polarization angle \( \theta_Q \). Increasing the angle between the spin polarization of the spin current and the magnetization increases the torque. Furthermore, the equation shows that the STT is independent of the exchange energy of the IP layer. However, for decreasing \( M_{\text{sat}} \) the period given by equation 2.17 increases, thereby increasing the spin dephasing distance inside the IP layer. Decreasing \( M_{\text{sat}} \) therefore increases the absorption depth of the spin current density by the IP layer. Furthermore, the saturation magnetization is a crucial parameter for the canting angle of the IP layer. An increased saturation magnetization means a smaller canting angle, since the ratio between the absorbed magnetization and the saturation magnetization is smaller in that case.

The discussion was based on electrons with larger energy than the potential step for minority electrons at the interface. For real interfaces, electrons can have smaller energies than the potential step, leading to evanescent states of the electrons inside the IP layer. These states alter the wave functions in the vicinity of the NM/FM interface, which influences the absorption depth of the transverse spins in the IP layer.
2.4 Precession of the IP layer in an IP field

In the experiments performed throughout this thesis, a spin current is excited in the OOP layer and is absorbed by the IP layer (see section 1.3). As described in the previous section the spin current in the IP layer exerts a spin-transfer-torque on the IP layer. As a result, the IP layer is canted towards the spin polarization direction of the spin current. In this section, it is shown how the IP layer behaves on longer time scales after its magnetization is canted.

The starting point of the discussion is the Landau-Lifshitz-Gilbert (LLG) equation \[ \dot{\mathbf{M}} = -\gamma_e \mu_0 \mathbf{M} \times \mathbf{H}_{\text{eff}} + \alpha \mathbf{M} \times \frac{Q_S}{M_{\text{sat}}^2 t_{\text{FM}} \mu_0} \mathbf{M} \times \left[ \mathbf{\sigma} \times \mathbf{M} \right], \] (2.19)

where \( \mathbf{\sigma} \) is the Pauli matrix for spin polarization, \( \alpha \) the Gilbert damping factor, \( \mathbf{H}_{\text{eff}} \) the effective field and \( Q_S \) the magnitude of the spin current density. The spin current density is derived from the charge current density \( J_c \) by counting the number of electrons in \( J_c \) and multiplying by the angular momentum per electron \( \hbar/2 \):

\[ Q_S = \frac{\hbar}{2} \frac{J_c}{e}. \] (2.20)

The first term of equation 2.19 accounts for the torque applied by the effective field consisting of the external applied field and any field representing anisotropy. The second term represents Gilbert damping, which damps the precessional motion caused by the torque of the effective field. The result of this Gilbert damping is that the magnetization aligns itself along the effective field. The last term describes spin-transfer-torque exerted by a spin current density \[ J_c \] by counting the number of electrons in \( J_c \) and multiplying by the angular momentum per electron \( \hbar/2 \).

The equilibrium state of the OOP layer will be briefly discussed, whereafter precessional characteristics of the IP layer are derived.

2.4.1 Equilibrium angle of the OOP layer

Before each experiment the OOP layer is aligned along the surface normal. Due to the applied IP field during a measurement, the magnetization of the OOP layer is canted towards the sample. This defines an equilibrium angle \( \theta_{\text{OOP}} \) between \( \mathbf{M} \) and the surface normal, as shown in figure 2.8. The effective field contains the applied field, the shape anisotropy and a surface anisotropy causing PMA [32]:

\[ \mathbf{M} = \begin{bmatrix} 0 \\ M_{\text{sat}} \sin(\theta_{\text{OOP}}) \\ M_{\text{sat}} \cos(\theta_{\text{OOP}}) \end{bmatrix}, \quad \mathbf{H}_{\text{eff}} = \begin{bmatrix} 0 \\ \frac{H_{\text{app}}}{2 K_{s,OOP} \mu_0 M_{\text{sat}}} \\ -1 \end{bmatrix} M_z, \] (2.21)

where \( K_{s,OOP} \) is the effective surface anisotropy constant. To find the equilibrium state of the OOP layer, the LLG equation is solved for \( d\mathbf{M}/dt = 0 \). In the static case there is no damping or
spin current: $\alpha = 0$ and $Q_S = 0$. For large applied fields the magnetization is aligned in-plane and the solution of equation 2.19 is:

$$\theta_{\text{OOP}} = \frac{\pi}{2}. \quad (2.22)$$

For small fields as used in this thesis, the OOP magnetization is only canted with a small angle towards the surface. The equilibrium angle for such small fields is given by:

$$\theta_{\text{OOP}} \approx \frac{H_{\text{app}}}{H_{\text{ani,OOP}}}, \quad (2.23)$$

where $H_{\text{ani,OOP}} = 2K_s,\text{OOP}/(\mu_0 M_{\text{sat}}) - M_{\text{sat}}$ is the total anisotropy arising from surface anisotropy and shape anisotropy. As will be shown in chapter 6, $H_{\text{app}}/H_{\text{ani,OOP}} \approx 0.1$ and therefore the equilibrium angle only deviates slightly from the surface normal (for maximum applied field about 6 degrees). Since $M_z/M_{\text{sat}} \approx 1$ it is assumed that the bottom layer is aligned along the surface normal during the experiments.

![Figure 2.8: Geometry for deriving the equilibrium angle of the OOP layer. The equilibrium angle is determined by the balance of the applied field, the demagnetization field and the surface anisotropy field. The equilibrium angle with maximum applied field in this thesis is about 6 degrees.](image)

**2.4.2 Precession of the IP layer**

Consider the geometry drawn in figure 2.9. In this figure an optical spin-transfer-torque (OSTT) has canted the IP layer magnetization towards the surface normal. The angle $\theta_M$ is defined as the angle between the magnetization and the surface, created by the OSTT. An external field is applied along the positive $y$-axis. This field results in a precessional motion of the magnetization around the field. Surface anisotropy is accounted for by a field quantified by $K_{s,\text{IP}}$ and equation 2.7. Furthermore, the laser pulse duration is of the order of a few hundreds of femtoseconds. Since the OSTT is a direct effect of the laser pulse, the OSTT is exerted within a few hundreds of femtoseconds, which can be approximated to be instantaneous as compared to the typical precession period due to the applied field (which is of the order of hundred picosecond) The last term in equation 2.19 can thus be excluded: $\vec{Q}_s = 0$ and the canting angle is taken as a fixed parameter. In the absence of damping ($\alpha = 0$) the LLG equation can be solved to give the
Kittel relation for the frequency $f_{IP}$ of precessional motion (see appendix A) [33]:

$$f_{IP} = \frac{\gamma_e}{2\pi} \sqrt{\mu_0 H_{app} \left[ \mu_0 H_{app} + \mu_0 M_{sat} - \frac{2K_{s,IP}}{M_{sat}} \right]}, \quad (2.24)$$

The initial canting angle of the IP layer is determined by the magnitude of the OSTT and determines the amplitude of the precessional motion.

Without damping the result of an OSTT and the applied field is steady a precessional motion of the magnetization around the applied field. Due to Gilbert damping the magnetization experience relaxation back towards the surface. Throughout this thesis measurements are performed of magnetization components perpendicular to the surface. For data analysis only the $z$-component of $\vec{M}$ is of importance. Due to the damped precession the magnetization of the $z$-component is described by sine describing precessional motion and an exponential decaying function describing Gilbert damping:

$$M_{z,IP} = A_z \sin \left( 2\pi f_{IP} t + \varphi \right) e^{-\frac{t}{\tau_d}}, \quad (2.25)$$

where $\tau_d$ is the characteristic time scale of damping and $\varphi$ is the phase of the precession. The Gilbert damping can be determined from $\tau_d$ by (see appendix A):

$$\alpha = \frac{1}{\gamma_e \tau_d \left[ \mu_0 H_{app} + \frac{\mu_0 M_{sat}}{2} - \frac{2K_{s,IP}}{M_{sat}} \right]}, \quad (2.26)$$

The spins of the spin current are aligned along the majority electron’s spin of the OOP layer. Consequently, the angle of the magnetization of the OOP layer with the surface normal determines the phase $\varphi$ of the precession of the IP layer. If the magnetization of the OOP layer is aligned along the positive $z$-axis, $M_{z,IP}$ starts precessional motion in a maximum and $\varphi = +\pi/2$. If the magnetization of the OOP layer is aligned along the negative $z$-axis, $M_{z,IP}$ starts precessional motion in a minimum and $\varphi = -\pi/2$. 

---

**Figure 2.9:** Geometry for precessional motion of the magnetization of the IP layer around a field along the in-plane axis. The magnetization is canted by an OSTT and an external field is applied along the IP axis.
2.5 All-optical switching by laser-induced optical spin-transfer-torque

In this thesis, all-optical switching of the IP layer without the need of an external field is investigated. The driving force of switching is precessional motion around the demagnetization field, which results from OSTT induced canted state. The Gilbert damping factor determines the time of precession and is thereby a key parameter for the possibility of switching the IP layer. The purpose of this section is to derive an equation between the required canting angle for a switch and the Gilbert damping factor \( \alpha \), assumed that the canting angle is small. The complete derivation can be found in appendix B. In figure 2.10 the geometry considered in this section is schematically drawn. The magnetization is initially aligned along the positive \( y \)-axis and is canted by an OSTT towards the surface normal with an angle \( \theta_M \), which results in a demagnetization field along the negative \( z \)-axis. The angle \( \phi_M \) is defined as the angle between the projection of \( \vec{M} \) in the \( x,y \)-plane and the \( y \)-axis.

![Figure 2.10: Geometry for precessional motion of the IP layer around its demagnetization field. The magnetization is canted due to a laser-induced STT, exciting a demagnetization field of \( \vec{H}_d = -M_z \hat{z} \). The magnetization defines the angles \( \theta_M \) and \( \phi_M \).](image)

In cylindrical coordinates the magnetization is determined as function of \( M_r \), \( \phi_M \) and \( M_z \):

\[
\vec{M} = \begin{bmatrix} M_r \sin(\phi_M) \\ M_r \cos(\phi_M) \\ M_z \end{bmatrix},
\]

where \( M_r \approx M_{\text{sat}} \) since the canting angle \( \theta_M \) is small. The effective field and the time derivative of \( \vec{M} \) are given by:

\[
\vec{H}_{\text{eff}} = \begin{bmatrix} 0 & 0 \\ 0 & \frac{2K_{\text{IP}}}{\mu_0 M_{\text{sat}}^2} - 1 \end{bmatrix} \frac{d\vec{M}}{dt} = \begin{bmatrix} M_{\text{sat}} \cos(\phi_M) \dot{\phi}_M \\ -M_{\text{sat}} \sin(\phi_M) \dot{\phi}_M \end{bmatrix}.\]

(2.28)
Inserting these vectors in equation 2.19 with $Q_S = 0$ yields the following equations for $\dot{\phi}_M$ and $\dot{M}_z$:

$$\dot{\phi}_M = \gamma e \mu_0 M_z \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right] + \frac{\alpha}{M_{sat}} \dot{M}_z,$$

$$\dot{M}_z = -\alpha M_{sat} \dot{\phi}_M. \tag{2.29a}$$

Solving this system it is found that $M_z$ decays exponentially, as can be expected due to Gilbert damping:

$$M_z = M_{sat} \sin(\theta_M) e^{-\frac{t}{\tau_d}}, \tag{2.30a}$$

$$\tau_d = \frac{1 + \alpha^2}{\alpha \gamma e \mu_0 M_{sat} \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right]}, \tag{2.30b}$$

where $\theta_M$ is the initial canting angle determined by the magnitude of the laser-induced STT. Inserting an exponential damped $M_z$ and $\dot{M}_z$ in equation 2.29a yields:

$$\dot{\phi}_M = \frac{\gamma e \mu_0 \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right]}{1 + \alpha^2} M_{sat} \sin(\theta_M) e^{-\frac{t}{\tau_d}}. \tag{2.31}$$

To find the final angle $\phi_{M,\text{final}}$ at which the magnetization ends precessional motion, equation 2.31 is integrated from $t_0 = 0$ to $t_\infty = \infty$. The magnetization starts precessing at $\phi_{M,t=0} = 0$ and that it stops precessional motion at $\phi_{M,t=\infty} = \phi_{M,\text{final}}$. The complete integration can be found in appendix B. The end result of the integration is:

$$\phi_{M,\text{final}} = \sin(\theta_M) \frac{\alpha}{\alpha \phi_{M,\text{final}}}. \tag{2.32}$$

For small canting angles $\sin(\theta_M)$ can be approximated by $\theta_M$. The final result is:

$$\theta_M = \alpha \phi_{M,\text{final}}. \tag{2.33}$$

It is concluded that the initial canting angle required to achieve a rotation of $\phi_{M,\text{final}}$ in the $x,y$-plane is proportional with $\alpha$. The constant of proportionality is $\phi_{M,\text{final}}$. The required initial canting angle is independent of the interface anisotropy constant caused by interaction at the FM layer’s interfaces. For a complete switch, meaning that $\phi_{M,\text{final}} = 180$ degrees, the canting angle of the IP layer should be exactly equal to $\alpha \cdot 180$ degrees. The canting angle needed for a complete switch is 3.6 degrees for Co ($\alpha \approx 0.02$) and 7.2 degrees for Ni ($\alpha \approx 0.04$). Furthermore, a critical canting angle is defined as the canting angle for which the magnetization has a negative component along the IP axis. This means that $\phi_{M,\text{final}} = 90$ degrees. For Co the critical canting angle $\theta_{M,\text{crit}}$ is 1.8 degrees and for Ni $\theta_{M,\text{crit}}$ is 3.6 degrees.

Equation 2.33 shows that the position of the magnetization after precessional motion is independent of $M_{sat}$. However, since a smaller saturation magnetization means a smaller demagnetization...
field, the frequency of precession is smaller. The frequency can be calculated to be (see appendix B):

$$\omega = \gamma e \mu_0 \left[ M_{\text{sat}} - \frac{2K_{z,IP}}{\mu_0 M_{\text{sat}}} \right] \theta_M. \quad (2.34)$$

This formula assumes no damping of $M_z$. Damping of $M_z$ decreases the demagnetization field during the precessional motion and therefore the frequency decreases during precession. The frequency is thus time dependent. However, equation 2.34 shows that the frequency decreases for decreasing $M_{\text{sat}}$. To derive the time scale of switching, consider the rotation of the magnetization in the $x,y$ plane from $t = 0$ to $t = t$:

$$\phi_M = \omega t. \quad (2.35)$$

The time scale to end in the state with $\phi_M = \phi_{M,\text{final}}$ is given by (again damping is neglected):

$$t_{\text{switch}} = \frac{M_{\text{sat}}}{\gamma e \mu_0 \alpha \left[ M_{\text{sat}}^2 - \frac{2K_{z,IP}}{\mu_0} \right]^\frac{1}{2}}. \quad (2.36)$$

This equation shows that the denominator grows faster with $M_{\text{sat}}$ than the numerator. This implies that the time needed to switch the IP layer increases with decreasing $M_{\text{sat}}$. Furthermore, an interface contribution to the anisotropy decreases the denominator, which increases the switching time scale. This is to be expected, since a larger anisotropy constant $K_{z,IP}$ decreases the effective field, thereby decreasing the frequency of the magnetization.
To investigate ultrafast laser-induced spin-transfer-torque, non-collinear magnetic multilayer structures are fabricated. The structures are fabricated using a sputtering technique able to control the thickness of the layers on the nanometer scale. The measurements are performed using the time-resolved magneto-optical Kerr effect (TR-MOKE) setup, as will be explained in the second section of this chapter. Finally, to measure the saturation magnetization and the anisotropy constant of the FM layers in the structures used throughout this thesis, vibrating sample magnetometry (VSM) and the anomalous Hall effect (AHE) are used respectively and will be discussed in the final part of this chapter.

3.1 Sample fabrication

The structures studied in this thesis are magnetic bilayers with layer thicknesses on the nanometer scale. These bilayers are deposited using direct-current (DC) magnetron sputtering. In this section the sputtering technique, the sputtering system, the deposition of wedge-shaped samples and sample selective sputtering are introduced.

Substrate preparation and sputter system

The substrate is cut from a Si:B wafer, has a typical surface of mm² and a thickness of 500µm. The substrate is cleaned in an ultrasonic bath with acetone and subsequently with isopropanol, whereafter it is transported into the sputter system. The sputter system contains three chambers separated with a valve: the load-lock, the sputter chamber and the oxidation chamber. To avoid contamination of the sample during sputtering, the sputter chamber is held on a constant ultra-high vacuum (UHV) of the order of $10^{-7}$ mbar. The load-lock is used for fast transport of the sample to be sputtered from outside the system into the sputter chamber. Because of the valve separating the load-lock and the sputter chamber, the load-lock can be vented or pumped without affecting the UHV of the sputter chamber. This reduces the time needed to transport the sample into the sputter chamber and keeps the UHV as clean as possible. Before sputtering the required layers on the substrate, the substrate is cleaned using vacuum oxidation for 600 s. After oxidation cleaning the required layers are sputtered, whereafter the sample is cleaned again in an ultrasonic bath with acetone and subsequently with isopropanol.
Sputtering technique
The material to be sputtered is inserted as a target disk. Inside the sputter chamber six targets are available for sputtering. The targets of relevance for the typical structures investigated in this thesis are copper (Cu), nickel (Ni), cobalt (Co), platinum (Pt) and tantalum (Ta). In figure 3.1a a schematic drawing of the DC sputtering technique is presented. Argon (Ar) gas is lead into the sputter chamber between an anode and the target disk, which acts as a cathode. A high potential difference is applied across cathode and anode to accelerate electrons from the target to the anode. These electrons scatter with the Ar atoms, creating a plasma of Ar$^+$ ions. In the magnetron sputtering technique, a magnet is placed behind the target to increase the stability of the plasma by confining the electrons in the plasma cloud. The positively charged Ar$^+$ ions are attracted to the negative charged target disk, where they hit the surface atoms of the target disk. These target atoms are detached from the target and eventually reach the substrate below the plasma cloud. In this way a layer of target atoms is deposited on the substrate. Inside the sputter chamber the substrate is placed on a rotation table, which places the substrate below a selected target. In this way multiple layers of various materials can be deposited on the substrate, creating a multilayer structure. During deposition the pressure inside the sputter chamber is of the order of $10^{-2}$ mbar. After sputtering a layer the argon atoms are pumped away and the UHV is restored. The sputtering power of the argon plasma can be controlled and determines the growth rate of the layer. At low power the growth rate of the layer is low. This increases the quality of the interfaces between adjacent layers, which increases spin transport through these interfaces. For the experiments performed in this thesis a large spin transport is required for a large STT effect. Hence the sputtering powers should be as small as possible. The growth rates of the magnetic layers (with a sputtering power of 10 W) used in this thesis (Co and Ni) are between 0.5 Å/s and 1.0 Å/s.

Wedge-shaped samples
Thickness dependent measurements are performed ideally on one single sample. This excludes sample variations caused by slightly, unintended, changed growth conditions and reduces the number of samples to be fabricated. Thickness dependent measurements can be achieved by using a sample in which the thickness of a layer is varied along its length or width. If the probe is local (the measured area being much smaller than the layer surface), thickness dependent measurements can be performed with one single sample. To create a layer with varying thickness, a wedge mask can be inserted between the target disk and the substrate, as drawn in figure 3.1b. By inserting this mask, the part of the substrate below the mask is excluded from deposition. The mask can be retracted from a start position to a defined end position. This will vary the part of the substrate that is excluded from deposition, changing the deposited layer thickness between the mask’s start position and the mask’s final position. In this way a layer is deposited with a varying thickness along the wedge retraction axis.

Selection mask
Another mask selects a specific sample to be sputtered at. In this way, samples where only one layer is different can be fabricated in the same batch. The selection mask is a metallic plate with a hole. Placing the hole above the sample to be sputtered at, the layer is grown on this sample and the other samples below the metallic plate are masked from deposition.
3.2 Magneto-optical Kerr effect (MOKE)

In 1845 Michael Faraday discovered a rotation of the polarization axis of linear polarized light upon transmission of that light through a magnetized medium. Later in 1876, John Kerr discovered the same effect for reflected light, which is called the magneto-optical Kerr effect (MOKE) \[34\]. This effect can be used to measure the magnetization of a reflecting FM material. In this section the physics behind MOKE is discussed, whereafter the experimental setups for measuring the static MOKE and the time-resolved MOKE are introduced.

3.2.1 Physics of MOKE

To understand the origin of the MOKE, consider the dielectric tensor $\epsilon$ of an isotropic material, which describes the optical response of the material to an electric field \[35\]. Only the off-diagonal elements $\epsilon_{ij}$ are dependent on the magnetization, being the magnetization component along $\hat{i} \times \hat{j}$. In this thesis the polar MOKE configuration is used for measurements. This means that the light is incident along the surface normal and only the magnetization component along the surface normal is measured. The final form of $\epsilon$ becomes then:

$$\epsilon = \begin{bmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ -\epsilon_{xy} & \epsilon_{xx} & 0 \\ 0 & 0 & \epsilon_{xx} \end{bmatrix}. \tag{3.1}$$
The two eigenstates of this tensor construct left-hand circular polarized (LHCP) and right-hand circular polarized (RHCP) light with eigenvalues $\epsilon = \epsilon_{xx} \pm i\epsilon_{xy}$. These two eigenstates result in a difference in refractive index for LHCP light ($n_-$) and RHCP light ($n_+$). A linear beam incident on a FM layer is constructed of both a LHCP and a RHCP component of equal magnitude. In figure 3.2 the physics behind MOKE is schematically shown. A linear polarized light beam is incident on an OOP FM layer. As a result of the difference in refractive index of the two circular polarized components inside the FM layer, the components travel with different velocities through the FM layer. This introduces a phase difference between LHCP and RHCP in the reflected light. Consequently, reflection of the beam rotates the axis of polarization with the Kerr rotation angle $\vartheta_{\text{Kerr}}$. A second effect of the difference in refractive index is that the two circular polarized components experience different absorption rates. The result of this effect is that one of the two components is decreased more than the other component, which results in an ellipticity $\varepsilon_{\text{Kerr}}$ of the reflected light. The Kerr rotation angle and ellipticity can be written as one single complex rotation angle $\Theta_{\text{Kerr}}$:

$$\Theta_{\text{Kerr}} = \vartheta_{\text{Kerr}} + i\varepsilon_{\text{Kerr}}. \quad (3.2)$$

![Figure 3.2: The physics of MOKE. A linearly polarized beam is incident on a FM layer. The reflected light beam changes polarization angle and gains ellipticity due to a difference in refraction index of LHCP light and RHCP light inside the ferromagnetic layer.](image)

3.2.2 Static MOKE

Before measuring magnetization dynamics it is important to know the static magnetic properties of the investigated sample. By measuring the static properties it can be seen if the multilayer structure behaves as expected. Furthermore, from static MOKE measurements the ratio of magneto-optical signal between the IP FM layer and the OOP FM layer in non-collinear magnetic multilayer structures can be determined. A schematic drawing of the setup is shown in figure 3.3. The incoming probe beam is linearly polarized by a polarizer and is focused on the sample. The beam which is reflected from the sample is polarized again by an analyzer and measured with a photodiode detector. By tuning the angle between the polarizer and the
CHAPTER 3. METHODOLOGY

analyzer the signal can be optimized to include only minor non-magnetic contributions. A photo-elastic modulator (PEM) is used for a further increase in the signal-to-noise ratio. By inserting the PEM, polarization dependent effects are filtered out. The PEM introduces a phase shift between electromagnetic waves of different polarizations. Therefore, the incoming probe beam is alternating polarized in LHCP light and RHCP light with frequency $\Omega_{\text{PEM}} = 50\,\text{kHz}$. If the signal is measured at a frequency of $\Omega_{\text{PEM}}$, only magnetization induced contributions to the signal are measured. The frequency dependent measurement is achieved by using a lock-in with the PEM frequency as a reference signal. Using the Jones formalism it can be shown that the first harmonic signal measures the induced ellipticity and the second harmonic signal measures the Kerr rotation of the reflected light [35]:

$$I_{\text{DC}} = R_{\text{sam}} \left( \frac{1}{2} + \vartheta_{\text{Kerr}} J_0 (A_{\text{PEM}}) \right)$$  \hspace{1cm} (3.3a)

$$I_{\text{1h}} = \varepsilon_{\text{Kerr}} R_{\text{sam}} J_1 (A_{\text{PEM}}) \cos (\Omega_{\text{PEM}} t)$$  \hspace{1cm} (3.3b)

$$I_{\text{2h}} = \vartheta_{\text{Kerr}} R_{\text{sam}} J_2 (A_{\text{PEM}}) \cos (2\Omega_{\text{PEM}} t)$$  \hspace{1cm} (3.3c)

where $J_n$ is the n-th spherical Bessel function, $A_{\text{PEM}}$ is the amplitude of phase shift introduced by the PEM and $R_{\text{sam}}$ the reflectivity of the sample. This means that by measuring at the first (second) harmonic frequency, $\varepsilon_{\text{Kerr}}$ ($\vartheta_{\text{Kerr}}$) is measured. In this way the PEM increases the signal-to-noise ratio because it isolates the magnetic signal from the DC signal.

![Experimental setup of the static MOKE setup. The laser beam is polarized in circular polarization components by a linear polarizer and a PEM. After reflection off the sample the light undergoes rotation of the polarization axis and gains ellipticity, which are measured by a detector and lock-in, referenced to the PEM frequency. For the sake of clarity, the angle of incidence of the probe beam is not drawn as parallel to the surface normal. In the real setup, the probe beam has near normal incidence at the sample to measure the polar MOKE.](image)

3.2.3 Time-Resolved MOKE (TR-MOKE)

In this thesis magnetization dynamics are measured on an ultrafast (femtosecond) time scale. To achieve such time resolution, an all-optical pump-probe setup is used. A pump beam
demagnetizes the FM layers and generates a superdiffusive spin current, whereafter a probe measures the induced magnetization dynamics as function of time. The time scale of these dynamics is in the picosecond time scale. By controlling the optical path difference $\Delta s_{op}$ between the pump and the probe beams, this ultrafast time scale becomes accessible in measurements. A time interval $\Delta t$ between probe arrival and pump arrival at the sample is introduced by $\Delta s_{op}$ and the speed of light $c$:

$$\Delta t = \frac{\Delta s_{op}}{c}.$$  \hfill (3.4)

Realizing that the speed of light is about $3 \times 10^8 \text{ m/s}$, it is clear that only a small optical path difference of 0.3 mm is needed to achieve a time resolution of 1 ps. The static MOKE is expanded with a pump beam by inserting a beam splitter after the output beam of the laser, as is schematically drawn in figure 3.4. While the probe beam is directed as in the static MOKE, the pump beam is directed to a movable Bragg reflector mounted on a delay line and then focussed on the sample. After reflection, the pump beam needs to be blocked to prevent it from hitting the detector and disturbing the signal of the probe beam. To block the pump beam the pump and probe beams are focussed by the lens as shown by the inset of figure 3.4. By directing the incoming pump beam in a vertical plane through the middle vertical axis of the lens, the reflected pump beam is travelling through the same vertical plane. The incoming probe beam is directed in a horizontal plane parallel through the middle axis of the lens. The reflected pump beam is thus separated from the probe beam and can be blocked without blocking the probe beam.

Figure 3.4: Experimental setup to measure the TR-MOKE. The laser beam is split in a probe and pump beam with a power rate of 1:20. The probe beam is directed as described in the static MOKE setup. The travel path of the pump can be adjusted for the proper delay time between pump and probe.
Further improvement of the signal is achieved by chopping the pump beam [35]. The chopper is a rotating disk at a frequency of 60 Hz that blocks the pump beam half of the time. The resulting signal is schematically drawn in figure 3.5. A high frequency signal is observed due to the PEM modulated probe beam and a low frequency signal is observed coming from the chopper modulated pump. The quantity $\delta V$ is the change in Kerr effect induced by the pump. This signal can be measured by redirecting the output signal of the static lock-in (see the signal of 'lock-in 1' in the figure) to the input of a second lock-in and locking this second lock-in to the frequency of the chopper. The resulting double modulation scheme increases the signal-to-noise ratio for both pump and probe, since only the signals at the frequencies of both PEM (probe) and chopper (pump) are measured by the two lock-ins.

![Figure 3.5: MOKE signal in a double modulated setup. A high frequency signal arises because of the PEM modulated probe beam and a low frequency signal due to the chopper modulated pump beam. The first lock-in measures the static (equilibrium) Kerr rotation and the second lock-in measures the magnetization induced variation of the Kerr rotation. Courtesy to Kuiper [36].](image)

In this thesis, the sample is excited by a Ti:Sapphire mode-locked laser with a central wavelength of 790 nm (photon energy of 1.57 eV) and a spectral width at the full-width-at-half-maximum of 10 nm. The pulses which are produced by the laser have a temporal FWHM of 75 fs and a repetition rate of 80 MHz. The samples are excited by the pump beam with a pulse energy of 2 nJ. The pump beam is focussed on the sample with a spot size of 7.2 $\mu$m (radius at 1/e$^2$ of the maximum energy), resulting in a fluence of 1.23 mJ/cm$^2$. The fluence of the probe beam is 0.06 mJ/cm$^2$. During the measurements an external field up to 150 mT can be applied at angles ranging from 0 degrees to 20 degrees with respect to the sample.

### 3.3 Vibrating sample magnetometry (VSM)

It is complicated to extract the absolute value of the magnetic moment of a sample from MOKE measurements. In order to determine the absolute value of the saturation magnetization, vibrating sample magnetometry is used [37]. The sample is placed in a magnetic field applied by two magnets which can reach a field up to 2 T. When the sample is vibrated, the magnetization
of the FM layers in the sample creates a changing magnetic flux through four detection coils attached around the sample. This causes induction currents in the coils, which are proportional to the magnetic moment of the vibrating sample. Two detection coils measure the magnetic moment of the magnetization perpendicular to the sample. These are shown in the schematically drawn front view of the VSM device in figure 3.6. The two other detection coils measure the magnetic moment parallel to the sample. The IP and the OOP layers of the samples can thus be separately measured. The saturation magnetization of one layer can be extracted by dividing the measured total magnetic moment of that layer by the volume of the layer.

Figure 3.6: Schematic drawing of the front view of a VSM device. A sample in an applied field is vibrating which causes induction currents in the detection coils. The drawing shows the two detection coils which measure the perpendicular magnetization component. The induced current is proportional to the magnetic moment.

3.4 Anomalous Hall effect (AHE)

To measure the magnetic anisotropy of the OOP magnetized layer, the anomalous Hall effect is used [38]. The anomalous Hall effect is the occurrence of a potential difference perpendicular to a current flowing through a FM layer. The effect occurs due to spin-dependent scattering of the electrons perpendicular to the current, by which majority electrons are scattered in the opposite direction of the minority electrons. Besides the anomalous Hall effect, an ordinary Hall effect (OHE) is present if the sample is placed in a magnetic field. The OHE occurs because the Lorentz force resulting from the magnetic field bends the trajectories of the electrons to a direction perpendicular to the current. The Hall resistivity $\rho_{\text{HE}}$ perpendicular to the current due to the OHE and the AHE is given by:

$$\rho_{\text{HE}} = \mu_0 H_z R_{\text{OHE}} + \mu_0 M_z R_{\text{AHE}},$$

where $R_{\text{OHE}}$ and $R_{\text{AHE}}$ are the resistivity coefficients for respectively the ordinary and the anomalous Hall effect. By measuring the potential difference perpendicular to a current flowing
CHAPTER 3. METHODOLOGY

through a FM layer, the magnetization component along the surface normal can thus be measured. The measurements are performed using wire-bonded samples as drawn in figure 3.7a. A current flows from left to right. The electrons, which flow from right to left, are scattered in the FM layer. In the figure majority electrons (blue) are scattered to the bottom of the figure and the minority electrons (red) are scattered to the top of the figure. As a result of the rearranged electrons a potential difference sets up across the sample, which is a measure of the perpendicular magnetization component, as was explained above.

The PMA constant can be measured by considering the geometry as drawn in figure 3.7b. The magnetization is initially aligned along the surface normal. If a field is applied at a certain angle $\theta_H$ with the surface normal, the magnetization cants towards the surface, defining an angle $\theta_{OOP}$. This angle can be calculated by considering that the measured magnetization component along the surface normal is equal to $M_{sat} \cos(\theta_{OOP})$. The total energy density of the system $U_{M,HE}$ is given by the Zeeman energy density and the anisotropy energy density:

$$U_{M,HE} = -K_{ani} \cos(\theta_{OOP})^2 - \mu_0 H_{app} M_{sat} \cos(\theta_H - \theta_{OOP}).$$

(3.6)

By minimizing this energy density the equilibrium angle for $\theta_{OOP}$ is obtained as function of $H_{app}$, $\theta_H$ and $M_{sat}$. By measuring the magnetization at different field strengths and field angles, various measurements of the equilibrium angle can be performed. Since $M_{sat}$ is assumed to be known and $\theta_H$ and $H_{app}$ can be set by an external magnet, for every measurement the value of $K_{ani}$ can be calculated from the measured value of $\theta_{OOP}$. The final step is to perform a least square curve fitting routine, estimating the value of $K_{ani}$ with the smallest deviation from the theoretical value determined by minimizing equation 3.6 for various values of $H_{app}$ and $\theta_H$. 

Figure 3.7: (a) Wire-bonded samples to measure the AHE. The electrons which flow from right to left are bended by the anomalous Hall effect. The resulting potential difference perpendicular to the current is measured to measure the perpendicular magnetization component of the sample. (b) Geometry used to explain the measurement routine for estimating $K_{ani}$. The magnetization is canted towards the surface plane by an applied field. Figures are reproduced from [38].
In this chapter the data analysis routine is discussed. The graphs in this chapter are presented in order to illustrate the analysis routine. A detailed discussion of the results will be provided in chapter 6. First, the experiment is explained in detail. The important quantities to be extracted from the measurements will be introduced. Secondly, the routine used to analysis the data is discussed. Since the measurements are performed in the polar configuration of the MOKE setup, the magnetization along the surface normal is measured.

4.1 The experiment

In section 1.3 the experiment was briefly introduced. In this section a detailed discussion of the typical experiment performed in this thesis will be discussed. The important sample characteristics needed to investigate the effect of optimizing the sample structure are introduced.

4.1.1 General geometry of the samples & the experiment

In figures 4.1a the three important layers for the experiment are shown: the OOP FM layer, the spacer layer and the IP layer. The spacer layer is a non-magnetic metallic layer for good electron transport. A laser pulse penetrates the sample from the top of the sample and induces an ultrafast demagnetization of the FM layers, as depicted in figure 4.1b. Since the magnetization is measured in the polar configuration, only the component along the surface normal is measured. Only a contribution to the demagnetization signal of the OOP layer is therefore observed if the field is applied at an angle of zero degrees with the sample. A typical demagnetization curve is shown in the left-hand side of figure 4.1d. The figure shows the measured Kerr rotation \( \theta_{\text{Kerr}} \) as function of time after laser pumping. The total demagnetization is given by \( A_{\text{demag}} \). Absorption of laser light excites hot electrons, which form a spin current in both the OOP and the IP layers. The spin current flows through the spacer layer from the OOP layer to the IP layer and vice versa (see figure 4.1b). The result of this spin transport is a spin-transfer-torque. The experiments of this thesis investigate the spin-transfer-torque exerted on the IP layer by the spin current generated in the OOP layer. Because of this spin-transfer-torque the magnetization
of the IP layer is canted towards the surface normal, creating an angle of $\theta_M$ between the magnetization and the sample, as drawn in figure 4.1c. By applying an external magnetic field at zero degrees with the sample, the magnetization of the IP layer starts a precessional motion around the IP axis. This precession is measured up to 500 ps after laser excitation, as shown in the right-hand side of figure 4.1d. The amplitude of the observed oscillation $A_{osc}$ is a measure for the perpendicular magnetization component of the IP layer and is a measure of the exerted spin-transfer-torque and therefore a measure for the spin absorption in the IP layer.

Figure 4.1: Description of the experiment and example of measured magnetization dynamics. (a) Before the experiment the bottom layer is aligned OOP and the top layer is aligned IP. (b) Excited hot electrons induce a spin current in the OOP layer, which flows through the spacer layer into the IP layer. Hot electrons exited in the IP layer induce a spin current to the OOP layer, but no explicit evidence of this current is observed in the experiment. (c) Due to a spin-transfer-torque applied by the spin current, the magnetization of the IP layer is canted towards the surface normal. The field is applied at an angle of zero degrees with the sample. (d) Example of measured magnetization dynamics. The curve shows a demagnetization and remagnetization of the OOP layer at short time scale and an oscillation of the IP layer at long time scale.
4.1.2 Key parameters: efficiency & canting angle

The experiments performed throughout this thesis investigate the generation of spin currents in the OOP layer and the absorption of this current in the IP layer. To quantify the magnetic moment lost by the OOP layer which is lost by transport, consider the spin transfer efficiency $\eta$. This efficiency is defined as the ratio between absorbed magnetic moment by the IP layer and the lost magnetic moment by the OOP layer:

$$\eta = \frac{\Delta M_{IP} t_{IP}}{\Delta M_{OOP} t_{OOP}},$$

where $t_{IP}$ and $t_{OOP}$ are the thicknesses of respectively the IP layer and the OOP layer. The spin transfer efficiency is a fingerprint of the number of spins which are transported out of the OOP layer. The total spin transport is larger, since a part of the spin current is not transmitted at the spacer layer / IP layer interface. However, by investigating the efficiency of various structures, the effect of a change in the structure on the spin current generation can be investigated.

Using the optical induced spin-transfer-torque on the IP layer, the IP layer is canted towards the surface normal and a demagnetization field is excited. For large enough canting angle the strength of this demagnetization field is large enough to switch the IP layer. This will be studied in chapter 5. It is therefore interesting to investigate the canting angle of the IP layer. The canting angle of the IP layer is calculated by:

$$\theta_M = \arcsin \left( \frac{\Delta M_{IP} t_{IP}}{M_{sat,IP} t_{IP}} \right).$$

4.2 Data analysis routine

In this section the routine used to analyse the data is presented. The routine begins with determining the total demagnetization of the OOP layer. Secondly, the characteristics of the IP layer oscillations are extracted. Finally, the efficiency and canting angle are calculated using the demagnetization of the OOP layer and the oscillation characteristics of the IP layer. For the sake of clarity, in this section the subscript $z$ will be omitted: $M_z = M$.

**Step one: measuring the demagnetization of the OOP layer**

The change in magnetic moment of the OOP layer between two fixed pump-probe delays is measured using three hysteresis loops. A hysteresis loop gives a measure of the total magnetic moment in the probed region. In order to observe a switch of the OOP layer, the field is applied at 20 degrees with the sample. The three hysteresis loops with different pump conditions are shown in figure 4.2. The figure shows a hysteresis loop without pumping (black curve), at a fixed negative pump-probe delay $\Delta t_{neg}$ (blue curve) and at a fixed positive pump-probe delay $\Delta t_{pos}$ (red curve). Note that with a negative pump-probe delay the probe arrives at the sample before the pump, whereas the probe arrives after the pump for positive pump-probe delay. A clear step is observed at a field of $\mu_0 H_{app} \approx \pm 30 \text{ mT}$, representing a switch of the OOP layer. A smaller step is observed at a field of $\mu_0 H_{app} \approx \pm 7 \text{ mT}$, which represents a switch of the IP layer.
A linear background is observed, which originates from contributions of both the IP and the OOP layer. The hysteresis loops can be fitted by two error functions representing the switch of the OOP layer and the IP layer and a linear background function for the background:

\[
\vartheta_{\text{Kerr,tot}} = A \cdot \text{erf} \left( \frac{\mu_0}{w_{\text{OOP}}} [H_{\text{app}} \pm H_{c,\text{OOP}}] \right) + C \cdot \text{erf} \left( \frac{\mu_0}{w_{\text{IP}}} [H_{\text{app}} \pm H_{c,\text{IP}}] \right) + D \cdot \mu_0 H_{\text{app}},
\]

where \( D \) is the slope of the linear background, \( A \) (\( C \)) is the amplitude of the switch of the OOP (IP) layer and \( w_{\text{OOP}} \)(\( w_{\text{IP}} \)) is the width of the OOP (IP) layer switch. The switching amplitude of the OOP layer at negative pump-probe delay (\( A_{\text{neg}} \)) is larger than the switching amplitude at positive pump-probe delay (\( A_{\text{pos}} \)), since at negative pump-probe delay the magnetization is probed before it is excited by the pump pulse. The switching amplitude is related to the total perpendicular magnetization of the OOP layer \( M_{\text{OOP}} \). The difference between \( A_{\text{neg}} \) and \( A_{\text{pos}} \) is a measure of the demagnetization of the OOP layer at \( \Delta t_{\text{pos}} \), which can be calculated as a fraction of the equilibrium magnetization at negative pump-probe delay by:

\[
\Delta M_{\text{OOP, pos}} = \frac{A_{\text{neg}} - A_{\text{pos}}}{A_{\text{neg}}},
\]

At negative pump-probe delay a small demagnetization can be observed, which indicates the presence of heat accumulation between two pulses. \( A_{\text{np}} \) is the switching amplitude of the OOP layer without pumping. The small demagnetization for negative pump-probe delays can be calculated by:

\[
\Delta M_{\text{OOP, neg}} = \frac{A_{\text{np}} - A_{\text{pos}}}{A_{\text{np}}},
\]

Figure 4.2: Hysteresis loops measured to calculate the demagnetization of the OOP layer. The black, blue and red curves depict the hysteresis loops for respectively measurements without pump pulsing, at fixed negative pump-probe delay and at fixed positive pump-probe delay.
Step two: determining the total demagnetization

Using the demagnetization between two pump-probe delays as determined in step one, the total demagnetization of the OOP layer can be determined. A typical demagnetization curve with an applied in-plane field of $\mu_0 H_{app} = 120 \text{ mT}$ is shown in figure 4.3a. In order to reduce the noise of the signal, demagnetization curves measured with various in-plane fields will be averaged. In figure 4.3b the demagnetization curves with various IP applied field strengths are shown. The figure shows that the demagnetization of the various measurements is of similar magnitude. In chapter 6 it will be quantified that the total demagnetization of these measurements is indeed consistent with each other. The curves can therefore be averaged to obtain a better estimation of the total demagnetization of the OOP layer. The resulting curve is shown in figure 4.3c. The red arrow indicates the maximum change of Kerr rotation $\Delta \vartheta_{\text{Kerr, max}}$. To norm the demagnetization curve to the equilibrium magnetic moment of the OOP layer at negative pump-probe delays, the norming factor $R_{\text{norm}}$ is calculated by:

$$R_{\text{norm}} = \frac{\Delta M_{\text{OOP, pos}}}{\Delta \vartheta_{\text{Kerr, max}}},$$

where $\Delta M_{\text{OOP, pos}}$ is calculated by equation 4.4. The averaged and normed demagnetization curve is shown in figure 4.3d. This curve can be fitted with a solution of the 3TM [35]:

$$1 - \left[ A_1 \frac{1}{\sqrt{1 + \frac{t}{\tau_0}}} - \frac{A_2 \tau_e - A_1 \tau_m}{\tau_e - \tau_m} e^{-\frac{t}{\tau_0}} - \frac{[A_1 - A_2] \tau_e}{\tau_e - \tau_m} e^{-\frac{t}{\tau_0}} \right] H(t) + A_3 \delta(t) * \Gamma(t),$$

where $A_1, A_2$ and $A_3$ respond to the demagnetization amplitude after electron-phonon-spin scattering, the demagnetization amplitude by the initial electron temperature rise and the amplitude of state filling effects. State fillings effects mean that the pump blocks certain electron excitations for the probe beam on temporal overlap between pump and probe [39]. The time constants $\tau_e$ and $\tau_m$ are the electron-phonon relaxation time and the spin relaxation time. The time constant $\tau_0$ is accounting for diffusion of heat into the substrate. Furthermore, $H(t)$ and $\delta(t)$ represent the Heaviside step function and a convolution with a Gaussian laser pulse. The maximum demagnetization $\Delta M_{\text{OOP, max}}$ can be determined by the minimum value $M_{\text{min}}$ of a fit of the graph in figure 4.3d with equation 4.7:

$$\Delta M_{\text{OOP, max}} = M_{\text{min}} - 1.$$  

(4.8)

The maximum lost magnetic moment of the OOP layer between two laser pulses can be calculated by the total magnetic moment of the OOP layer ($M_{\text{OOP,tOOP}}$) and equation 4.8:

$$\Delta M_{\text{OOP,tOOP}} = M_{\text{OOP,tOOP}} \cdot \Delta M_{\text{OOP, max}}.$$  

(4.9)

The magnetic moment at negative pump-probe delay deviates from the total magnetic moment without pumping $M_{\text{OOP}}$, caused by a small demagnetization due to heat accumulation. The
total magnetic moment of the OOP layer is therefore substituted by:

\[ M_{\text{OOP}} \rightarrow M_{\text{OOP}} \cdot \Delta M_{\text{OOP,neg}}, \]  

(4.10)

where \( \Delta M_{\text{OOP,neg}} \) is measured as a fraction of the magnetization without pumping, using equation 4.5.

Figure 4.3: Examples of demagnetization curves. Graphs a and b show the raw data of measured demagnetization curves with various applied IP fields. Graph c shows the demagnetization curve resulting from averaging the curves with various IP field strengths. Graph d shows this averaged curved normed to the magnetization of the OOP layer at negative pump-probe delay \( M_{\text{OOP}} \) and fitted with equation 4.7.
Step three: extracting the oscillation characteristics

Using $R_{\text{norm}}$ calculated in step two, the oscillations measured at longer time scale can be normed to the magnetic moment of the OOP layer. The precessional motion can be fitted with equation 2.25. However, during the precessional motion the remagnetization of the OOP layer is not finished. A contribution of this remagnetization process is observed in the signal. The remagnetization is accounted for by an exponentially decaying function describing remagnetization at short time scale, an exponentially decaying function describing remagnetization at long time scale and a constant $C_{\text{osc}}$. The oscillation are fitted with an exponential decaying sine, as was introduced with equation 2.25. The function used to fit the oscillations is given by:

$$M_{\text{IP}}t_{\text{IP}} = A_{\text{osc}} \sin \left(2\pi f_{\text{IP}} \left(t - t_0\right) + \varphi\right) e^{-\frac{t-t_0}{\tau_{\text{d}}}} + A_1 e^{-\frac{t-t_0}{\tau_1}} + A_2 e^{-\frac{t-t_0}{\tau_2}} + C_{\text{osc}},$$  \hspace{1cm} (4.11)

where $t_0$ corrects for a shifted temporal pump-probe overlap due to alignment of the setup and $A_{\text{osc}}$ is the amplitude of the oscillation. $A_1$ and $\tau_1$ are the amplitude and time scale of the fast exponential background function, whereas $A_2$ and $\tau_2$ are the amplitude and time scale of the slow exponential background function. During the oscillation of the IP layer, the IP layer might still be in the process of remagnetization. Since the oscillation frequency depends on the magnetization, the frequency might be changing during the oscillation. However, it is assumed that the deviation of the magnetization from its equilibrium value is small during the oscillation. The oscillation is therefore fitted with a time-independent frequency. An example of a normed oscillation curve is shown in figure 4.4a, where the time evolution of $M_z/M_{\text{OOP, neg}}$ is shown for an IP field of 120 mT. The pump-probe delay for the first data point in this oscillation was 12 ps. There is therefore no demagnetization observed, since the demagnetization occurs within 1 ps. The oscillation is fitted with equation 4.11 to extract the amplitude, frequency and damping time of the oscillation.

Figure 4.4: (a) Normed measurement of an oscillation of the IP magnetic moment. The applied (in-plane) field was 120 mT. The oscillation can be fitted with equation 4.11. (b) Kittel fit of the field-dependent frequencies extracted from the oscillations.

The saturation magnetization of the IP layer can be extracted using the Kittel formula (equation 2.24). The fitted field-dependent frequencies are plotted as function of the applied field in figure 4.4b with black symbols. The red curve shows the Kittel formula to extract the saturation
magnetization of the IP layer $M_{\text{sat,IP}}$. The value of $M_{\text{sat,IP}}$ will be used to determine the ratio between the magneto-optical (MO) sensitivities of the OOP layer and the IP layer in the next step.

**Step four: magneto-optical sensitivity ratio between the ferromagnetic layers**

The amplitude of the oscillation is normed to the magnetic moment of the OOP layer. To extract the real magnetic moment of the IP layer from the oscillation amplitude, the ratio between the MO-sensitivities of the OOP layer and the IP layer has to be determined. The laser light decays exponentially in the multilayer structure, meaning that the IP layer, which is placed on top of the structure, absorbs more light than the OOP layer, which is placed in the bottom of the structure. The signal of the IP layer will therefore be larger than the signal of the OOP layer. The ratio between the MO-sensitivities is determined by measuring a static hysteresis loop of a sample with a field applied perpendicular to the sample. The resulting hysteresis loop consists of an easy-axis loop observed from the OOP layer and a hard-axis loop observed from the IP layer. A static hysteresis loop is shown in figure 4.5a. This hysteresis loop can be fitted with equation 4.12 to extract the switching amplitude $A$ of the OOP layer and the slope $D$ of the linear contribution of the IP layer. The fit is shown in the figure with a red curve.

$$\vartheta_{\text{Kerr,tot}} = A \cdot \text{erf} \left( \frac{\mu_0}{w_{\text{OOP}}} [H_{\text{app}} \pm H_{\text{c,OOP}}] \right) + D \cdot \mu_0 H_{\text{app}}. \quad (4.12)$$

![Figure 4.5: (a) Hysteresis loop of a non-collinear magnetic multilayer, measured with the static MOKE setup in a field perpendicular to the sample. The hysteresis loop consists of an easy-axis loop of the OOP layer and a hard-axis loop of the IP layer. (b) The measured hysteresis loop of figure a, expanded with the theoretically extrapolated hard-axis loop of the IP layer for negative fields (blue curve). The IP layer is saturated along the surface normal at a field of $\mu_0 H_{\text{app}} = 2K_{\text{ani}}/M_{\text{sat}}$.](image)
\[ \mu_0 H_{ani} = 2K_{ani}/M_{sat}, \]

where \( K_{ani} \) contains the anisotropy constant of shape anisotropy and interface anisotropy \( K_{s,tot} \) resulting from the adjacent layers. The signal of the total IP layer magnetic moment (\( C \) in figure 4.5b) is calculated by multiplying the saturation field with the slope extracted by a fit with equation 4.12 of the measured hysteresis loop. This can be regarded as the switching amplitude of the IP layer. The MO-sensitivity ratio \( R_{MO} \) between the signals coming from the saturated OOP layer (\( A \) in figure 4.5b) and the saturated IP layer is given by:

\[ R_{MO} = \frac{A}{C}, \] (4.13)

In step three the oscillation of the IP layer was normed to the magnetic moment of the OOP layer. Therefore, the MO-sensitivity ratio has to be determined in units of IP magnetic moment per OOP magnetic moment. This factor \( R_{MM} \) can be calculated by:

\[
R_{MM} = \frac{A}{M_{sat,OOP} t_{OOP}} = \frac{A}{D} \cdot \frac{M_{sat,IP} t_{IP}}{M_{sat,OOP} t_{OOP}}.
\] (4.14)

The saturation magnetization of the OOP layer is measured by VSM measurements and the saturation magnetization of the IP layer is determined by a Kittel fit (shown in figure 4.4b). The factor \( R_{MM} \) gives the conversion factor for the real magnetic moment of the IP layer from the OOP layer:

\[
M_{IP} t_{IP} = A_{osc} \cdot M_{OOP} t_{OOP} \cdot R_{MM}.
\] (4.15)

Step five: calculating the efficiency and canting angle

The last step is to calculate the spin transfer efficiency and the resulting canting angle of the IP layer. First the magnetic moment absorbed by the IP layer along the surface normal (\( \Delta M_{IP} t_{IP} \)) has to be calculated. For negative pump-probe delay the IP layer has its magnetization completely in the surface plane, it has no perpendicular magnetic moment. The amplitude of the oscillation in figure 4.4a caused by the laser-induced spin-transfer-torque is therefore equal to the absorbed perpendicular magnetic moment: \( \Delta M_{IP} t_{IP} \propto A_{osc} \). As was explained in step three, the oscillation amplitude is normed to the magnetic moment of the OOP layer. The true total magnetic moment absorbed by the IP layer is calculated by equation 4.15. The canting angle \( \theta_M \) of the IP layer is calculated by the ratio between \( \Delta M_{IP} t_{IP} \) and the total magnetic moment of the IP layer:

\[
\theta_M = \arcsin \left( \frac{A_{osc} \cdot M_{OOP} t_{OOP} \cdot R_{MM}}{M_{sat,IP} t_{IP}} \right).
\] (4.16)

The spin transfer efficiency shows how much perpendicular magnetic moment lost by the OOP layer is absorbed by the IP layer. This gives a quantification of the spin current reaching the IP.
layer as a percentage of the spins participating in the demagnetization of the OOP layer. The spin transfer efficiency $\eta$ in percentage is calculated by:

$$\eta = \frac{\Delta M_{IP} t_{IP}}{\Delta M_{OOP} t_{OOP}} = \frac{A_{osc} \cdot M_{OOP} \cdot t_{OOP} \cdot R_{MM}}{M_{OOP} t_{OOP} \cdot \Delta M_{OOP, max}}. \quad (4.17)$$

### 4.3 Discussion of the data analysis routine

The analysis routine presented here is subject to some assumptions and fitting routines. First, the wavelength dependence of the MO-sensitivity ratio will be discussed. Secondly, the analysis of demagnetization curves is discussed. Finally, some remarks about fitting the oscillations will be made.

#### Wavelength dependence of the MO-sensitivity ratio

In step four the MO-sensitivity ratio between the OOP layer and the IP layer is measured using the static MOKE setup. This setup uses a wavelength of 633 nm. The central wavelength used during time-resolved measurements is 790 nm. It is assumed that the MO-sensitivity ratio between the OOP and the IP layer is equal for both cases. One could argue that the hysteresis loops used to measure the demagnetization of the OOP layer (see figure 4.2) may be used to extract the MO-sensitivity ratio. However, these hysteresis loops are measured using a field applied at an angle of 20 degrees with respect to the sample. The linear slope is not solely originating from the IP layer in this case. A detailed analysis of the wavelength dependence of the MO-sensitivity ratio between the OOP layer and the IP layer is beyond the scope of this thesis.

#### Demagnetization curves

The hysteresis loops measured to determine the OOP layer demagnetization are measured with the field at an angle of 20 degrees with the sample. With the field applied at this angle a contribution of the IP layer can be observed, considering that the IP layer is forced into a canted state at an angle of $\sim 2$ degrees with the sample. Since the MO-sensitivity of the IP layer is larger than the MO-sensitivity of the OOP layer, this could lead to a substantial contribution of the IP layer to the demagnetization curve. Furthermore, the IP layer absorbs much more light, resulting in a larger demagnetization, increasing its contribution in the signal even more. The demagnetization curve to extract the total demagnetization is therefore measured with the field at 0 degrees with respect to the sample. Measuring the polar MOKE does not contain an IP layer contribution in that case. In figure 4.6a demagnetization curves measured with the field applied at 20 degrees (black curve) and with the field applied at 0 degrees with respect to the sample (red curve) are shown. The figure shows that the maximum signal with the field applied at 0 degrees is reduced to 72% of the maximum signal observed with a field applied at 20 degrees. The observed difference is a measure of the demagnetization of the IP layer.
The magnetic signal in measured demagnetization curves can be extracted from demagnetization curves with positive and negative fields. A demagnetization curve with a field of $\mu_0 H_{\text{app}} = +62 \text{ mT}$ (black curve) and a demagnetization curve with a field of $\mu_0 H_{\text{app}} = -62 \text{ mT}$ (red curve) is shown in figure 4.6b. The fields were canted at 20 degrees with the sample. The magnetic contribution and non-magnetic contribution in the signal are determined by respectively the difference and the sum of the two demagnetization curves [35]:

$$\vartheta_{\text{Kerr, mag}} = \frac{\vartheta_{\text{Kerr, H}} - \vartheta_{\text{Kerr, H}}^{+}}{2}$$  \hspace{1cm} (4.18a)$$

$$\vartheta_{\text{Kerr, nonmag}} = \frac{\vartheta_{\text{Kerr, H}}^{+} + \vartheta_{\text{Kerr, H}}^{-}}{2}.$$  \hspace{1cm} (4.18b)

The magnetic and non-magnetic contributions in the demagnetization curves are shown in figure 4.6c with respectively a red curve and a blue curve, along with the demagnetization curve measured with a negative field (black curve). The curves are normed to the maximum signal of the negative field curve. In the blue curve, a small signal with a maximum of 10% of the maximum negative field signal is observed for short pump-probe delays <0.4 ps. As is shown by the green vertical line, at the pump-probe delay of maximum demagnetization, no non-magnetic contribution is observed. A measurement with either negative or positive field is thus sufficient to determine the total demagnetization of the OOP layer. Furthermore, the black and the red curves shows that the demagnetization curves for the negative field measurement and the magnetic contribution calculated by equation 4.18a are consistent with each other. Since a switch of the OOP layer requires a perpendicular field component, demagnetization curves with opposite fields can not be measured using an IP field. Using that non-magnetic contributions are negligible in demagnetization curves, the norming factor calculated by equation 4.6 will be determined from a demagnetization curve with positive IP field.

Figure 4.6: (a) Demagnetization curves measured with the field at 20 degrees (black curve) and 0 degrees (red curve) with the sample. The curves show that an IP layer contribution can be observed for a field at 20 degrees. (b) Demagnetization curves measured with opposite fields applied at 20 degrees with respect to the sample. (c) Magnetic (red) and non-magnetic contributions (blue) in demagnetization curves, as calculated with equations 4.18 and using the measurements shown in figure 4.6b. The black curve shows a demagnetization curve measured with negative field.
Fitting the oscillations

Equation 4.11 has a large number of fitting parameters. To decrease the number of free fitting parameters, certain parameters can be fixed using the following reasoning. It was calculated by Schellekens et al. that the typical measured canting angles of the spin absorption layer are too small to originate in a spin-dependent Seebeck effect [22]. The STT is caused by superdiffusive spin currents, which flow with very fast velocities (in the order of nm/fs). During the experiments it can therefore be assumed that the STT is an instantaneous effect compared to a precession period (which is typical in the order of $10^2$ ps). As a result, the magnetization component of the IP layer along the surface normal starts in a maximum or a minimum, resulting in a phase of $\varphi = +\pi/2$ (maximum start) or $\varphi = -\pi/2$ (minimum start) assuming that the oscillation is fitted with a sine function as in equation 4.11. The actual phase depends on the orientation of the OOP layer. The offset time $t_0$ corresponds to the instant of a maximum (or minimum) in $M_{\text{IP}}$ at $t_{\text{IP}}$. Because the STT occurs on a fast time scale, for fitting purposes it can be approximated by an instantaneous effect occurring at the instant of temporal pump-probe overlap. Therefore $t_0$ can be deduced from measured demagnetization curves, by determining the start of the demagnetization. The background is empirically determined. The background function is determined by two exponential decay functions to account for a fast remagnetization process at short time scale and for a slower, more long lasting, remagnetization process at the longer time scale. It is believed that this background function fits the background quite well. However, it needs to be stressed that changing the background function changes the fitted amplitude of the oscillations by a considerable amount. For example, fitting the oscillation with only one exponential background function decreased the amplitude for all measurements (up to 10% systematically) and in most cases decreases the quality of the fit as compared to the data. To compare the various samples investigated throughout thesis, all measured oscillations were fitted with a background function consisting of two exponential functions and a constant, as introduced in equation 4.11.

Absence of a second frequency in the magnetization dynamics

The Kerr rotation measurements introduced in step three shows a single-frequency oscillation. The sample used as a reference in this thesis will be introduced in section 6.1 and showed such a single-frequency oscillation. In section 1.3 it was introduced that in the bilayers investigated in reference [22] a double-frequency oscillation was observed. The second frequency was attributed to an oscillation of the OOP layer caused by a laser-induced anisotropy pulse [40]. The amplitude of this oscillation decreases if the perpendicular magnetic anisotropy of the OOP layer is increased. However, in reference [22], the structures were fabricated without a Ta seeding layer. The addition of a Ta seeding layer in the magnetic bilayers investigated in this thesis increases the PMA, which suppresses the oscillation of the OOP layer. In appendix D, AHE measurements are shown on samples with and without Ta seeding layer, supporting the reasoning that Ta increases the PMA in the OOP layer. This leads to the observation of only one frequency.
The structures introduced in chapter 4 could be used as a two-state memory element. The IP layer of these structures (see figure 4.1) could define the first state as the magnetization along the positive in-plane axis and the second state as the magnetization along the negative in-plane axis. The two states are excited in an all-optical way without the need for an external magnetic field. In this chapter important parameters which influence the properties of such a memory element are studied using numerical simulations of the LLG equation with Matlab. First, a rewritten version of the LLG equation is derived for use in Matlab. This equation will be verified by simulating an IP magnetization in an external IP field. The frequencies for various field strengths and small canting angles should lead to the Kittel relation (see equation 2.24). Secondly, it is studied for which canting angles resulting from an optical induced spin-transfer-torque (OSTT) the IP magnetization switches as function of the Gilbert damping factor $\alpha$. It is studied how the switching time scale depends on $\alpha$ and $M_{\text{sat}}$. Finally, simulations have been performed to study the dependency of the canting angle on the properties of a spin current which flows into the IP layer.

5.1 Geometry and the LLG equation for Matlab

The simulations in this chapter investigate the behaviour of the magnetization of a cobalt and a nickel IP layer, as used in the experiments throughout this thesis. The geometry used in the simulations is drawn in figure 5.1. Initially, the magnetization is aligned along the positive $y$-axis, $\vec{M} = +M_{\text{sat}} \hat{y}$. Due to an OSTT the magnetization is canted towards the surface normal (green arrow), defining an angle $\theta_M$ between the magnetization and the $x,y$-plane. Unless stated otherwise, the canting angle means the initial canting angle due to OSTT. Due to shape anisotropy a demagnetization field $\vec{H}_d = -M_z \hat{z}$ (black arrow) results from the canted state. The demagnetization field creates a precessional motion of the magnetization around the $z$-axis. The angle of the magnetization projection in the $x,y$-plane with the positive $y$-axis is defined as $\phi_M$. The value of this angle after precessional motion is defined as $\phi_{M,\text{final}}$. Unless stated otherwise, the saturation magnetization of the IP layer is $1422 \text{ kA/m}$, equal to the saturation
magnetization of bulk cobalt at room temperature [23]. To verify the model, a field is applied
along the y-axis in the simulations of section 5.2 (red arrow). The LLG equation (see equation
2.19) describes the evolution of $\vec{M}$ as a function of time. The equation takes into account the
torques exerted on the magnetization by the effective field $\vec{H}_{\text{eff}}$, Gilbert damping with factor
$\alpha$ and the OSTT term, quantized with the spin current density $Q_S$. To implement the LLG
equation in Matlab, equation 2.19 has been rewritten to (see appendix C for a derivation):

$$\frac{d\vec{M}}{dt} = -\frac{\gamma e \mu_0}{1 + \alpha^2} \vec{M} \times \frac{\vec{H}_{\text{eff}}}{M_{\text{sat}}} - \frac{\alpha Q_S}{M_{\text{sat}} t_{\text{FM}}} \vec{M} \times \left[ \sigma \times \vec{M} \right].$$

To verify the implementation in Matlab of equation 5.1, precessional motion around an applied
field $\vec{H}_{\text{app}}$ along the in-plane axis is simulated. The initial canting angle was fixed at 1 degree.
For this canting angle the demagnetization field is small. Therefore the magnetization precesses
around the applied field and there is no precession around the z-axis. The surface anisotropy
constant was $K_s = 0$. Simulations are performed for 750 ps with various field strengths of the
applied field. A typical simulated curve for $M_z/M_{\text{sat}}$ can be seen in figure 5.2a, where the field
strength was 60 mT. As argued in section 2.4.2, the result is damped precessional motion with
damping time $\tau_d$ and amplitude $M_{z0}$. A fit with an exponential damped sine (equation 2.25)
gives a frequency of 9.3 GHz, which was to be expected from the Kittel formula (equation 2.24).

The simulations are performed for $\mu_0 H_{\text{app}} = -150$ mT up to $\mu_0 H_{\text{app}} = +150$ mT, in steps
of 10 mT. In figure 5.2b the frequencies for these simulations are plotted as function of field

5.2 Field dependent frequency of precession

To verify the implementation in Matlab of equation 5.1, precessional motion around an applied
field $\vec{H}_{\text{app}}$ along the in-plane axis is simulated. The initial canting angle was fixed at 1 degree.
For this canting angle the demagnetization field is small. Therefore the magnetization precesses
around the applied field and there is no precession around the z-axis. The surface anisotropy
constant was $K_s = 0$. Simulations are performed for 750 ps with various field strengths of the
applied field. A typical simulated curve for $M_z/M_{\text{sat}}$ can be seen in figure 5.2a, where the field
strength was 60 mT. As argued in section 2.4.2, the result is damped precessional motion with
damping time $\tau_d$ and amplitude $M_{z0}$. A fit with an exponential damped sine (equation 2.25)
gives a frequency of 9.3 GHz, which was to be expected from the Kittel formula (equation 2.24).

The simulations are performed for $\mu_0 H_{\text{app}} = -150$ mT up to $\mu_0 H_{\text{app}} = +150$ mT, in steps
of 10 mT. In figure 5.2b the frequencies for these simulations are plotted as function of field
CHAPTER 5. STUDY OF THE IN-PLANE LAYER USING SIMULATIONS WITH THE LLG EQUATION

Figure 5.2: (a) Typical simulated temporal curve for $M_z/M_{sat}$. For this particular simulation $\mu_0 H_{app}$ was 60 mT and the fitted frequency was 9.3 GHz. (b) Simulated frequencies as function of applied field. The Kittel relation for $M_{sat} = 1422$ kA/m is shown in red. As can be seen, the fitted frequencies follow the Kittel relation for the simulated value of $M_{sat}$.

strength. As can be seen in the figure, the fitted frequencies follow the Kittel relation for $M_{sat} = 1422$ kA/m, which was to simulated value of $M_{sat}$. It is therefore concluded that the model of equation 5.1 is correctly implemented in Matlab.

5.3 Switching by optical induced spin-transfer-torque

In this section, switching of the IP layer by all-optical spin-transfer-torque is investigated. No external field is added in the simulations of this section. The OSTT results in a certain canting angle. This canting angle is a fixed parameter in the simulations of this section. Furthermore, the magnetization starts in the $y,z$-plane, meaning that initially $M_y > 0$. The demagnetization field, resulting from the canted state, is the driving force of a precessional motion around the $z$-axis. Equation 2.33 was derived to give a relation between the position of the magnetization after precessional motion as function of $\theta_M$ and $\alpha$. This relation only holds for small canting angles. In this section the position of $M_y/M_{sat}$ after precessional motion (further written as $M_y,_{final}$) has been simulated for a range of small to large canting angles and damping factors. In the following discussion a switch is defined as any state for which $M_y,_{final} < 0$ and a complete switch is defined for the state $M_y,_{final} = -1$. The critical canting angle is defined as the angle for which $M_y,_{final} = 0$ and $\phi_{M,y,final} = \pi/2$. The critical canting angle is thus a measure for the minimum canting angle for a switch. In order to determine the values of the canting angle as function of $\alpha$ for which the magnetization has a switch, a phase diagram is constructed, in which $M_y,_{final}$ is shown as function of $\theta_{M}$ and $\alpha$. Simulations with only shape anisotropy have been performed at first. Secondly, an uniaxial anisotropy is added along the $y$-axis to align the complete magnetization along the $y$-axis, $\hat{M}_{final} = \pm M_{sat}\hat{y}$, thereby studying an easier way of switching the in-plane layer.
5.3.1 Required canting angle without uniaxial IP anisotropy

A typical simulation result without uniaxial anisotropy is shown in figure 5.3a. The selected damping factor was 0.04, which is a typical measured value for the cobalt IP layer in the experiments throughout this thesis. The figure shows that $M_{y,\text{final}}$ oscillates as function of $\theta_M$. The oscillation arises from the precessional motion around the demagnetization field, as can be argued as follows. The frequency increases with increasing canting angle. Since $\alpha$ was fixed, the time of precessional motion does not change. Increasing the frequency means increasing the angular velocity and therefore increasing $\phi_{M,\text{final}}$. As a result $\phi_{M,\text{final}}$ is a continuous function of $\theta_M$, which results in a continuous $M_{y,\text{final}}$ as function of $\theta_M$.

The values of the canting angle are divided in bands for which $M_{y,\text{final}} > 0$ and bands for which $M_{y,\text{final}} < 0$. The first five bands for $\alpha = 0.04$ are indicated in figure 5.3a. The odd numbered bands contain the canting angles for which $M_{y,\text{final}}$ is positive. The canting angles located in the even numbered bands result in negative $M_{y,\text{final}}$. The critical canting angle for $\alpha = 0.04$ is 3.6 degrees and is indicated with a red circle in figure 5.3a. The angles for which a complete switch is achieved are all located in the even numbered bands. The canting angle for a complete switch in the second band is 7.2 degrees and is indicated in figure 5.3a with a blue circle. The required canting angle for a switch is dependent on the damping factor as was shown by equation 2.33. The simulation as described above is performed for several values of $\alpha$. The required canting angle for switching the in-plane layer as function of $\alpha$ is plotted in figure 5.3b with a red curve. The blue and the green curves show the dependency of the required canting angles for a complete switch in the second band and fourth band respectively. For small canting angles the curves can be fitted with the linear model of equation 2.33. The red, blue and green curves are fitted with a linear function between $\theta_M = 0$ degrees and $\theta_M = 5$ degrees. The table shows the fitted slopes. The values of these fitted slopes deviate with maximum 0.5 % from the expected slope based on equation 2.33, which is equal to $\phi_{M,\text{final}}$. For small canting angles, the linear model derived in equation 2.33 is therefore correct. The linear fits are extended to compare them with the temporal evolution of the simulation. For large canting angles the linear model starts to deviate from linearity, as can be seen in figure 5.3b.

These simulations have been done for $\alpha = 0.002$ up to $\alpha = 0.400$ in steps of $\Delta \alpha = 0.002$. At every value of $\alpha$, the initial canting angle was varied from $\theta_M = 0$ degrees to $\theta_M = 90$ degrees. In this way it is possible to draw a phase diagram of the final position of $M_y$ as function of $\theta_M$ and $\alpha$. The phase diagram is shown in figure 5.4. Values of $\theta_M$ and $\alpha$ for which $M_{y,\text{final}} = +1$ (no switch) are indicated by red. The values of $\theta_M$ and $\alpha$ for which $M_{y,\text{final}} = -1$ (complete switch) are indicated by blue. Intermediate values for $M_{y,\text{final}}$ are given by the colours as depicted by the legend. The black lines show the lines on which $M_{y,\text{final}}$ switches sign. The first of these lines indicates the critical canting angle. The phase diagram shows that $M_{y,\text{final}}$ is more sensitive to changes in $\theta_M$ at small $\alpha$ and more sensitive to changes in $\alpha$ at large $\theta_M$. This can be explained by considering the change in $\phi_{M,\text{final}}$. This angle is determined by the product of angular frequency $\omega$ and precessional time $t_{\text{pres}}$:

$$\phi_{M,\text{final}} = \omega t_{\text{pres}}. \quad (5.2)$$

A change of $\phi_{M,\text{final}}$ is given by:

$$\Delta \phi_{M,\text{final}} = \omega \Delta t_{\text{pres}}, \quad (5.3a)$$

54
CHAPTER 5. STUDY OF THE IN-PLANE LAYER USING SIMULATIONS WITH THE LLG EQUATION

Figure 5.3: (a) Simulated $M_y$ as function of $\theta_M$ for $\alpha = 0.04$. The critical angle is 1.8 degrees and a complete switch is achieved for a canting angle of 3.6 degrees, as indicated by a red and blue circle respectively. (b) Graph of the required $\theta_M$ as function of $\alpha$. Three curves are plotted. The red curve shows the critical canting angle. The slope of the fit is 79.8 degrees as is to be expected from the model (see equation 2.33). The blue curve shows the first $\theta_M$ for a complete switch ($\phi_{M,\text{final}} = 180$ degrees) and the green curve shows the second $\theta_M$ for a complete switch ($\phi_{M,\text{final}} = 540$ degrees). The extension of the fitted linear functions show that at larger canting angles the linear model starts to deviate.

\[
\Delta \phi_{M,\text{final}} = t_{\text{pres}} \Delta \omega, \tag{5.3b}
\]

where equation 5.3a depicts a change in $\phi_{M,\text{final}}$ due to a change in $t_{\text{pres}}$ and equation 5.3b depicts a change in $\phi_{M,\text{final}}$ due to a change in $\omega$. The angular frequency is determined by the canting angle and the precessional time is determined by $\alpha$. A large canting angle gives a high angular frequency, which causes large sensitivity of $\phi_{M,\text{final}}$ (and $M_y$) to changes in $\Delta t_{\text{pres}}$, as shown by equation 5.3a. A small $\alpha$ gives a long precessional time, which causes large sensitivity of $M_y$ to changes in $\omega$, or equivalently to changes in $\theta_M$. Using a large $\alpha$, the canting angle of a two-state memory element to switch between the two states can therefore be tuned with less precision as compared to a memory element with small $\alpha$. An IP layer with a large $\alpha$ is thus preferable if the tuning precision of the canting angle is considered. However, increasing $\alpha$ also increases the required canting angle, as is shown in figure 5.4 by the first black line. At small $\alpha$ the end position of the magnetization is quickly oscillation as function of the canting angle. This is attributed to the discretization of the steps in $\alpha$ and $\theta_M$. Decreasing the step size or tolerance for solving the LLG equation in Matlab would give a better resolution of the phase diagram. However, these options increase the simulation time and for the qualitative conclusions drawn in this section the current resolution is well enough.

In this section switching the in-plane layer of a two-state memory element by an optical induced spin-transfer-torque was studied by simulations using the LLG equation. It is concluded that a large $\alpha$ is preferable over a small $\alpha$ because this makes it easier to tune the canting angle for a switch. On the other hand, a large $\alpha$ increases the required canting angle for a switch. The most ideal material is thus a balance determined by the possibility to excite a certain canting angle by the optical excitation mechanism and the specific ability of the optical excitation mechanism...
to tune the canting angle.

Figure 5.4: Phase diagram of switching with shape anisotropy. The diagram shows $M_y$, final as function of $\theta_M$ and $\alpha$. Bands are visible where $M_y < 0$ and $M_y > 0$ for the corresponding values of $\theta_M$ or $\alpha$. The separation of these bands is indicated with black lines, at which $M_y$, final = 0.

5.3.2 Required canting angle with uniaxial IP anisotropy

In the absence of an IP anisotropy, $M_y$, final is a continuous function of $\theta_M$ and $\alpha$. The addition of an IP anisotropy along the $y$-axis causes the magnetization to align along the $y$-axis after precessional motion, $M_{\text{final}} = \pm M_{\text{sat}} \hat{y}$. This means that the magnetization either ends in its initial position or has a complete switch. Intermediate positions are not allowed. In this way the structure could in principle be used as a two-state memory element. Figure 5.5a shows a phase diagram of $M_y$, final as function of $\theta_M$ and $\alpha$ with an added in-plane anisotropy. The simulated IP anisotropy was 10 mT, corresponding to an IP anisotropy constant of $K_y = 7.11 \times 10^3$ J/m$^3$ (using equation 2.7). As can be seen, the magnetization indeed has either a complete switch (blue areas) or no switch at all (red areas). Comparing the phase diagram with figure 5.4 it is seen as well that the critical canting angle increases. For small canting angles the demagnetization field is dominated by the anisotropy field. This results in an effective field which is pointing closely along the positive $y$-axis. Therefore the magnetization ends precessional motion along the positive $y$-axis. The magnetization switches when the torque arising from the demagnetization field is larger than the torque arising from the anisotropy field. The torque of the anisotropy field can be seen as an additional barrier to be overcome by the demagnetization field and is the cause of the increased critical canting angle.

In figure 5.5b a phase diagram is shown with increased IP anisotropy of 50 mT, corresponding to an IP anisotropy constant of $K_y = 3.56 \times 10^4$ J/m$^3$. Because of the larger torque arising from the anisotropy, the critical canting angle is further increased. Furthermore, it is seen that the
CHAPTER 5. STUDY OF THE IN-PLANE LAYER USING SIMULATIONS WITH THE LLG EQUATION

Figure 5.5: (a) Phase diagram of switching with shape anisotropy and an IP anisotropy constant of $K_y = 7.11 \times 10^3 \text{J/m}^3$. The diagram shows $M_y,\text{final}$ as function of $\theta_M$ and $\alpha$. (b) Phase diagram of switching with shape anisotropy and an IP anisotropy constant of $K_y = 3.56 \times 10^4 \text{J/m}^3$.

The effect of changing anisotropy is decreased for large $\alpha$, which can be explained by the following. For a large damping factor the required canting angle for a complete switch increases. The demagnetization field is stronger for these required canting angles as compared to the case with small damping factor. The torque arising from the demagnetization field is therefore increased and less dominated by the anisotropy field. As a result the anisotropy field has a smaller effect on the canting angles for which $M_y,\text{final}$ changes sign.

In this section it was shown that the addition of an anisotropy along the in-plane axis makes it easier to tune the canting angle for a switch of the in-plane layer. The anisotropy should be as small as possible, since it increases the required canting angle for a switch.

### 5.4 Time scale of a complete switch (the switching time)

The results presented in the previous section showed the possibility of a two-state memory element by switching the IP layer using optical induced spin-transfer-torque. This section determines the behaviour of the switching time by studying $M_z$ as function of $M_{\text{sat}}$ and $\alpha$. The driving force for switching is the demagnetization field, which depends on $M_z$. The precessional motion of the magnetization stops when the demagnetization field is zero, and consequently $M_z = 0$. A complete switch was defined for $M_y,\text{final} = -1$. A definition of the switching time could be the first time step at which $M_z = 0$ and $M_y,\text{final} = -1$. However, since $M_z$ decays exponentially (see section 2.5), $M_z$ never exactly takes the value of zero in the simulation. For this definition an arbitrary tolerance condition is thus needed. Since only a qualitative study of the dependency of the switching time on $\alpha$ and $M_{\text{sat}}$ will be done, it is more convenient to consider the behaviour of the exponential damping time of $M_z$, which was shown by equation 2.30b to be dependent on $M_{\text{sat}}$ and $\alpha$.

Simulations of the canted IP layer are done for values of $\alpha = 0.004$ up to $\alpha = 0.2$. At every value of $\alpha$ the saturation magnetization was varied from $M_{\text{sat}} = 30 \text{kA/m}$ up to $M_{\text{sat}} = 1500 \text{kA/m}$. The simulation was evaluated for 5 ns. The initial canting angle was fixed at the required canting...
angle for a complete switch, which was shown with equation 2.33 to be $\phi_{\text{M, final}} = \alpha \cdot 180 \, \text{degrees}$. From the curve of $M_z$ as function of time, the damping time $\tau_d$ was extracted. The extracted damping times for all simulations are shown in a phase diagram in figure 5.6. The damping times are plotted with a logarithmic scale against the Gilbert damping factor $\alpha$ and the saturation magnetization. A green area means a small damping time (fast switching), whereas a red area means a large damping time (slow switching). For small $\alpha$ and small $M_{\text{sat}}$, a red area is seen, meaning that the damping time is large and switching is therefore slow. In contrast, the damping times at large $\alpha$ and large $M_{\text{sat}}$ show a large green area, meaning small damping time and fast switching. Considering the switching time, $\alpha$ or $M_{\text{sat}}$ should therefore be as large as possible for fast switching between the two states of a two-state magnetic memory element.

### 5.5 Canting angle as function of spin current

In this section the canting angle is studied as a function of the properties of the spin current which causes the spin-transfer-torque. First, the canting angle is studied as function of the magnitude of the spin current and as function of the number of electrons carrying the spin current. Secondly, the effect of decreasing the spin polarization of the spin current is studied. By decreasing the spin polarization, the magnitude of the transverse spin is decreased, which should decrease the canting angle of the in-plane layer. The spin current is excited by a laser pulse in the experiments, meaning that the spin current density $Q_S$ in equation 5.1 is time dependent. Since the spin current is a direct effect of laser excitation, the time dependence is assumed to follow the Gaussian time profile of the laser pulse. The applied current is modelled as Gaussian function:

$$Q_S = \frac{J_c \hbar}{2e} e^{-\left(\frac{t-100 \cdot 10^{-15}}{2\tau^2}\right)^2 \cdot 8 \ln 2},$$  \hspace{1cm} (5.4)
where $J_c$ is the amplitude of charge current density and $\tau_p = 75$ fs is the FWHM. The FWHM is chosen to equate the FWHM of the experimental laser pulse, which is 75 fs. $Q_S$ is assumed to be spin polarized along the positive $z$-axis. To study the resulting spin-transfer-torque, simulations of $M_z$ have been evaluated for 300 fs. After 300 fs, the spin current density is approximately zero, meaning that the effect of a STT is zero. The time interval of 300 fs is thus long enough to cant the magnetization to its maximum canting angle, but on the other hand short enough to neglect the torque of the demagnetization field, which pulls the magnetization back to the surface on a longer time scale. The simulation are evaluated from $t = 0$ fs to 300 fs. To include all electrons in the spin current on this time interval, the current is shifted by 100 fs. The results of the simulations are shown in figure 5.7, where $\theta_M$ is plotted as function of $J_c$. The black curve shows the canting angle of a Ni IP layer with $M_{\text{sat}} = 484$ kA/m. The red curve shows the canting angle of a Co IP layer with $M_{\text{sat}} = 1422$ kA/m. It is clearly seen in the graph that decreasing the saturation magnetization increases the canting angle at fixed spin current, as argued in section 2.3.

![Figure 5.7: Canting angle as function of charge current density. The charge current density is related to the spin current density by equation 5.4. For relatively small currents the canting angle is approximated by a linear function. For large $J_c$ the canting angle converges to the spin polarization direction of the spin current. For the large graph this was 90 degrees and for the inset this was 70 degrees. The top axis of the large graph shows the number of electrons in a excitation spot of $\pi \cdot 7.2 \mu m^2$, corresponding to the charge current density of the bottom axis.](image-url)
For large spin current the canting angle saturates to 90 degrees, aligning $\vec{M}$ along the spin polarization of the spin current. Equation 2.18 showed that the STT depends with a sine on the angle between the magnetization and the spin current polarization ($\theta_Q$). At large values of $J_c$ the magnetization is canted towards the surface normal by a considerable amount. The result is that $\vec{M}$ is not longer approximately aligned along the in-plane axis during the exerted STT, thereby reducing the angle $\theta_Q$. At a certain number of spins, $\theta_Q \approx 0$ and the STT term given by equation 2.18 is zero. For small $J_c$ the canting angle can be approximated by a linear fit. The linear fit (for 0 degrees < $\theta_M$ < 20 degrees) gives a slope of $1.79 \times 10^{-9}$ degrees/(A/cm$^2$) for the Ni IP layer. The Co IP layer is fitted with a slope of $0.61 \times 10^{-9}$ degrees/(A/cm$^2$). The spin current density needed to achieve a complete switch ($\theta_M = 7.2$ degrees for $\alpha = 0.4$) is thus very large, of the order of $10^{10}$ A cm$^{-2}$. The top axis of the large graph shows the number of electrons which carry the spin current, for a pump spot size with a diameter of 7.2 µm. A linear fit for small canting angles gives a slope of $7.51 \times 10^{-10}$ degrees per excited electron. This means that for a canting angle of 7.2 degrees the number of electrons to be excited is $9.59 \times 10^9$. Considering that the structures in this thesis are pumped with a photon energy of 1.57 eV, this means that a pulse energy of 2.41 nJ is required. This energy is in the same order of typical laser fluences used in the experiment performed in this thesis.

However, the simulations assume a spin polarization of 90 degrees, meaning that all excited electrons in the spot size are polarized transverse to the IP magnetization and along the same direction. In the real experiments, electrons spin polarized along the opposite direction are excited as well and not all excited electrons are absorbed by the IP layer. Furthermore, in the experiments the spin current is originating from an OOP layer buried at the bottom of the structures. The absorbed pulse energy is therefore <10% of the total pulse energy. The real pulse energy required to excite a switch of the IP layer is therefore at least a factor of 10 larger in the real experiment.

The inset of figure 5.7 shows the canting angle for a spin current which is spin polarized at an angle of 70 degrees with the in-plane. The figure shows that the canting angle for a given current is smaller than the canting angle achieved with a spin current which is complete spin polarized along the surface normal. This is caused by the fact that the sine in equation 2.18 has a lower value for a spin polarization of 70 degrees as compared to a spin polarization of 90 degrees. For large $J_c$ the canting angle saturates to 70 degrees, the value at which $\theta_Q = 0$. For this angle, the torque on the magnetization exerted by the spin current becomes zero.

The results in this section show that the current density to be excited in the experiments to induce a switch of the in-plane layer is very large due to their short time scale nature. On the other hand, because of their short lifetime, the spin-transfer-torque exerted by these spin currents is very fast, within 300 fs. In the experiments the spin current is originating from the OOP layer. Increasing the equilibrium angle of the OOP layer with the surface normal decreases the angle between the OOP layer and the IP layer. Consequently, the spin polarization of the spin current excited in the OOP layer is decreased. The equilibrium angle of the OOP layer with the surface is thus one of the parameters which determines the OSTT characteristics and should be as small as possible for fast switching with a current density as low as possible.
5.6 Conclusions

In this chapter the influence of $\alpha$, $M_{\text{sat}}$, $K_y$ and $Q_S$ on the effect of a spin-transfer-torque on the IP layer is studied. As was shown in section 5.3.1, for a complete switch in the absence of an IP anisotropy, the canting angle needs to be tuned very precisely. However, adding an IP anisotropy aligns the magnetization along the $y$-axis as was shown in section 5.3.2. This causes either a complete switch or no switch at all and makes it easier to tune the canting angle for switching the IP layer. Adding an in-plane anisotropy thus facilitates the possibility of a two-state memory element. The required canting angle for a switch is influenced by the value of the IP anisotropy constant. This influence is decreased for large Gilbert damping. Furthermore, the canting angle needs to be tuned with less precision for large $\alpha$. An increased $\alpha$ means a shorter switching time of the IP layer. However, increasing $\alpha$ increases the required canting angle for a switch as well. The canting angle for a given spin current can be increased by decreasing $M_{\text{sat}}$, but this increases the switching time. Concluding, the final memory element should consist of a material with well-balanced magnetic properties. Furthermore, a quantification was derived for the required pump pulse energy to excite a complete switch of the IP layer. In the simulation for $\alpha = 0.04$ and $M_{\text{sat}} = 1422 \text{kA/m}$, typical values for a cobalt layer, it was derived that the required pulse energy absorbed by the OOP layer is $2.41 \text{nJ}$. The pulse energy used throughout this thesis is $2 \text{nJ}$, meaning that the absorbed pulse energy by the OOP layer is below $0.2 \text{nJ}$. In the simulation it was assumed that all spins of the current are aligned along the positive surface normal and the simulation only accounts for angular momentum absorbed by the IP layer. In the real experiments, pulse energy is lost to the excitation of minority electrons and interface scattering of electrons. Considering these arguments, it is expected that a complete switch can not be observed with a cobalt layer in the experiments performed in this thesis. However, adding an anisotropy along the in-plane axis was shown to decrease the required canting angle for a complete switch. This anisotropy should be as small as possible, considering that it does increase the critical canting angle.
Investigating ultrafast laser-induced spin-transport-torque

In the previous chapter simulations were done to characterize the properties of a two-state memory element using laser-induced spin-transfer-torque. In this chapter the results are provided of measurements on various non-collinear magnetic bilayers. First, a reference sample will be introduced. Secondly, it will be investigated how helicity and pump fluence affect the optical induced spin-transfer-torque. Thirdly, it will be shown that changing the Co: Ni ratio in the Co/Ni multilayer increases the excitation of spin currents and increases the canting angle. Fourthly, the penetration depth of transverse spins in a cobalt layer is investigated using a wedge-shaped in-plane layer. Finally, the saturation magnetization of the IP layer is decreased to increase its canting angle.

6.1 Geometry of the reference sample

The first sample investigated will be used as a reference sample for the other investigated samples. This sample will be referred to as sample 1. The geometry of the reference sample is based on the non-collinear magnetic bilayer investigated by Schellekens et al. [22] and is shown in figure 6.1a. The magnetic bilayer is grown on a boron-doped silicon substrate (Si:B), which is a good heat conductor. This is required to transport heat out of the sample, preventing it from being burned by accumulation of heat after pulsed laser excitation. A seed layer of 2 nm Ta is grown to provide a smooth growth. The out-of-plane layer (OOP layer) consists of a sequence of 4 × Co[0.2]/Ni[0.6], with a Co[0.2] dusting layer between the fourth Ni layer and the spacer layer. A buffer layer of 4 nm Pt below the OOP layer induces PMA in the Co/Ni multilayer. The spin current flows from the OOP layer through the spacer layer, which is a 5 nm thick Cu layer. The in-plane layer (IP layer) of the reference sample is a 3 nm thick Co layer. To prevent oxidation of the IP layer, the sample is capped with a 1 nm thick Pt layer.

The sample is excited by a femtosecond laser pulse from the top. Part of the laser light is transmitted through the capping layer and is absorbed by the IP and the OOP layer. Due to laser excitation hot electrons are created in these layers, as drawn in figure 6.1b. As a result, a
hot-electron spin current is generated from the OOP layer to the IP layer and vice versa. In this thesis, the spin current generated in the OOP layer is investigated, which exerts an optical induced spin-transfer-torque (OSTT) on the IP layer. The OOP layer is therefore referred to as the spin injection layer and the IP layer is therefore referred to as the spin absorption layer. As a result of the OSTT, the spin absorption layer magnetization is canted towards the surface normal, defining an angle of $\theta_M$ between the magnetization and the sample, as drawn in 6.1c. Furthermore, the spin current from the IP to the OOP layer exerts an OSTT on the OOP layer. However, the signal of this OSTT is negligible small in the measurements due to the smaller sensitivity of the magneto-optical signal to the OOP layer.

6.2 Static magnetic properties of the reference sample

Before investigating laser-induced magnetization dynamics of the reference sample, some static magnetic properties are discussed in this section. These properties will be used to quantify the efficiency of spin transport and to quantify the canting angle of the spin absorption layer. First, the saturation magnetization of the spin injection layer is determined using VSM measurements. Secondly, it is shown that a linear Faraday background effect is observed in the static hysteresis loops. Since the slope of the linear signal in static hysteresis loops is used to calculate the magneto-optical (MO) sensitivity ratio between the spin injection and the spin absorption layer, the Faraday background effect has to be eliminated.
6.2.1 Saturation magnetization of the spin injection layer

To calculate the MO-sensitivity ratio between the spin injection and absorption layer as discussed in section 4.2, the value of the saturation magnetization of the spin injection layer ($M_{\text{sat,OOP}}$) is required. The saturation magnetization is measured with VSM measurements (see section 3.3). VSM measurements result in a hysteresis loop of the absolute magnetic moment of the investigated sample. The field was applied perpendicular to the sample, resulting in an easy-axis loop for the spin injection layer. The field strength was varied from $\mu_0 H_{\text{app}} = -1$ T up to $+1$ T. The hysteresis loop is shown in figure 6.2. A clear switch of the spin injection layer is observed, together with a linear background originating from the Si:B substrate and the spin absorption layer. To extract the saturation magnetization of the spin injection layer, the measurement is fitted with equation 4.12. The fitted switching amplitude was $6.36 \times 10^{-5}$ emu, which translates to a saturation magnetization of $M_{\text{sat,OOP}} = 6.5 \pm 0.4 \times 10^2$ kA/m. The saturation magnetization calculated by an weighted average of cobalt and nickel was $7.6 \times 10^2$ kA/m. This value assumes perfectly sharp interfaces and complete ultrathin layers within the Co/Ni multilayer. The experimental value is smaller, since sputtering creates rough interfaces and no complete separate Co and Ni layers. The saturation magnetization of $6.5 \pm 0.4 \times 10^2$ kA/m is used in the calculation of the MO-sensitivity ratio between the spin injection and absorption layers.

![Hysteresis loop of the perpendicular magnetic moment of a non-collinear magnetic bilayer measured with VSM. The field was applied perpendicular to the sample, resulting in an easy-axis loop for the spin injection layer.](image)

Figure 6.2: Hysteresis loop of the perpendicular magnetic moment of a non-collinear magnetic bilayer measured with VSM. The field was applied perpendicular to the sample, resulting in an easy-axis loop for the spin injection layer.

6.2.2 Faraday subtraction of static MOKE hysteresis loops

The Faraday effect was briefly introduced in section 3.2 as a rotation of the polarization axis of linearly polarized light, transmitted through a medium in a magnetic field. In this section it will be shown that the Faraday effect is largely independent of the layers in the investigated samples.

In figure 6.3a measurements of the Kerr rotation on various materials are shown, measured with the static MOKE setup. The field was applied perpendicular to the sample and its strength ranged from $\mu_0 H_{\text{app}} = -300$ mT to $+300$ mT. Four measurements were performed: two measurements on a metallic mirror (green and red curves), one measurement on a bare Si:B
substrate (blue curve) and a measurement on a sample containing an OOP layer (black curve). These measurements were fitted with a linear function, of which the slopes were of similar values. The differences in these slopes for various materials are only small, the difference in the smallest slope and the largest slope is only 5%. In section 6.2.3 it will be shown that this is a negligible variations. The Faraday effect is therefore mainly caused by the optical components in the setup and by the air through which the laser light travels. The Faraday effect can thus be regarded as a background effect, to be determined with a sample which does not contain a spin absorption layer. In such a sample there are only negligible sample-related linear effects. That measurement can thus be used to measure the Faraday effect and subtract it as a background contribution from static MOKE loops.

Figure 6.3: (a) Kerr rotation measurements of various materials. The linear contribution is originating from the Faraday effect, rotating the polarization of light travelling through air and the optical components of the setup. (b) Static hysteresis loop of the reference sample. The difference of the linear contribution to the hysteresis loop with and without Faraday effect subtraction is clearly visible.

6.2.3 Static MOKE hysteresis loop

In figure 6.3b the static hysteresis loop of sample 1 measured with the static MOKE setup in an OOP field is shown. The black curve shows a static hysteresis loop for sample 1 as-measured. The data after Faraday subtraction is shown with a red curve. Comparing the black and the red curves, the slope of the linear contribution is significantly changed by subtracting the Faraday effect. The linear slope of the static hysteresis loop is used to calculate the signal sensitivity of the spin absorption layer and the ratio between MO-sensitivity between the spin injection layer and the spin absorption layer. It is therefore important to subtract the Faraday effect from the data, to avoid an overestimation of the spin absorption layer MO-sensitivity. The blue curve shows a fit of the data with equation 4.12, which resulted in a ratio $A_{OOP}/D$ of $170.1 \pm 0.1 \text{ mT}^{-1}$. This ratio is used to calculated the MO-sensitivity ratio between the spin injection layer and the spin absorption layer, as was discussed in section 4.2. This ratio $R_{MM}$ was $0.23 \pm 0.01$ for sample 1.
6.3 Magnetization dynamics of the reference sample

In the previous section some static magnetization properties of sample 1 were investigated, required for the MO-sensitivity ratio between the spin injection and the spin absorption layer. In this section the laser-induced magnetization dynamics of sample 1 are discussed, measured in the TR-MOKE setup by pumping the non-collinear magnetic bilayer with a femtosecond laser pulse. In this way the generation and absorption of spin currents is investigated.

As was described in step one of the analysis routine (see section 4.2), an estimation of the demagnetization of the spin injection layer is measured using three hysteresis loops. The measured hysteresis loops for sample 1 are shown in figure 6.4a. The field was applied at an angle of 20 degrees with respect to the sample. The black curve shows a measurement without pumping, the blue curve shows a hysteresis loop for a pump-probe delay of $\Delta t_{\text{neg}} = -1 \text{ ps}$ and the red curve shows a hysteresis loop measured with a pump-probe delay of $\Delta t_{\text{pos}} = 0.5 \text{ ps}$. Comparing the hysteresis loop without pumping with those with pumping, a large reduction of the coercivity is observed when the sample is excited by the laser. This reduction is caused by the temperature-dependence of the anisotropy of the spin injection layer [41] and is equal for both negative and positive pump-probe delays [42]. A second observation is the slanted switching of the magnetization when it is laser pumped. This indicates a non-uniform heating of the sample. This creates local variations in the anisotropy inside the laser spot. These variations cause a spread in coercivity for the local magnetization in the sample, resulting in a more slanted switching hysteresis loop.

An estimation of the demagnetization at $\Delta t_{\text{pos}}$ can be made by comparing the switching amplitude of the spin injection layer at negative and positive pump-probe delays. Using the ratio of these switching amplitudes, the measured demagnetization curve can be normalized. In figure 6.4b the normalized demagnetization curve is shown, averaged over six measurements.
with various IP field strengths. The curve shows the temporal evolution of the perpendicular magnetization, as a fraction of the total perpendicular magnetization measured for negative pump-probe delay \((M_{z,\text{neg}})\). The total demagnetization of the spin injection layer was \(8.3 \pm 0.4\%\). The table in the figure shows the total demagnetizations measured for the six measurements with various in-plane field strengths. The error bar in the demagnetization is estimated at 5\% of the total demagnetization, based on the error of fitting the hysteresis loops and the spread in demagnetization values of separate field-dependent measurements.

![Image](image.png)

Figure 6.5: Laser-induced magnetization dynamics of sample 1. On short time scale the demagnetization curve with an applied in-plane field of 82 mT is shown. At longer time scale oscillations are observed, measured with an in-plane field of respectively 82 mT (black curve) and 120 mT (blue curve). The inset shows a Kittel fit of the field-dependent frequencies.

To explore the effect of changing the IP field strength on the OSTT, consider figure 6.5. In the left-hand side the demagnetization curve with an IP field of 82 mT is shown. As is indicated for negative pump-probe delays, the accuracy of the signal is about 0.5\% of \(M_{z,\text{neg}}\). The standard deviation of the demagnetizations measured with various IP field strengths is 0.4\%, well within the accuracy of the measured signal. It is therefore justified to average the demagnetization curves measured with various IP field strengths. On longer time scale, a single-frequency oscillation is observed, as is shown in the right-hand side of figure 6.5 with a black curve. The oscillation is originating from an OSTT exerted on the spin absorption layer by the generated spin current. A fit with equation 4.11 gives a precession frequency of 9.92 ± 0.04 GHz. The blue curve in the right-hand side of figure 6.5 shows a measurement with an increased in-plane field of 120 mT. A fit of this measurement depicted an increased frequency of 11.96 ± 0.04 GHz. This increase is caused by the increased field strength. The oscillations were measured with six IP field strengths and for every field strength the oscillation frequency was extracted. The field-dependent frequencies are plotted versus the field in the inset of figure 6.5. A fit with the Kittel relation (equation 2.24) resulted in \(M_{\text{sat,IP}} = 1198 \pm 6\text{ kA/m}\). The bulk value for the saturation magnetization reported in literature is 1422 kA/m [23]. Such a deviation of \(M_{\text{sat}}\) from the bulk value was reported before for thin film magnetic layers [38].

As was argued in section 2.4.2, the OSTT can be regarded as an instantaneous effect, compared with the precession period of the spin absorption layer for the applied fields used in this
thesis. The OSTT determines the amplitude of the oscillation of the spin absorption layer. The amplitudes of the oscillations measured with various IP field strengths should therefore similar, meaning that these amplitudes can be averaged. Using equation 4.17, the efficiency of sample 1 is $4.4 \pm 0.4\%$, meaning that $4.4 \pm 0.4\%$ of the magnetic moment lost by the spin injection layer is absorbed by the spin absorption layer. The resulting canting angle of the spin absorption layer was $1.3 \pm 0.1 \times 10^2$ mdegrees, resulting in a canting angle of $15 \pm 2$ mdegrees per $1\%$ demagnetization of the spin injection layer. The efficiency and canting angle are a factor of two larger than the ones measured in reference [22] (see section 1.3). This is attributed to the factor that in that publication, the static MOKE data was not corrected for the Faraday effect. As was shown in section 6.2.3, this results in different MO-sensitivity ratio between the spin injection layer and spin absorption layer. The values for the efficiency and canting angle determined for sample 1 will be the reference values to investigate the effect of the remaining experiments performed in this thesis.

6.4 Helicity and fluence dependency

In this section the dependency of the laser-induced spin-transfer-torque on the properties of the laser light is discussed. In the first subsection helicity dependent measurements will be performed. Secondly, the dependency of the optical induced spin-transfer-torque (OSTT) on laser fluence will be discussed. The measurements are performed on a sample with an equal structure as sample 1. This sample was grown in a separate batch than sample 1 and will be referred to as sample 2.

6.4.1 Helicity of the pump beam

The laser light used in the experiments of section 6.3 was linearly polarized, meaning that the amount of left-handed circularly polarized light (LHCP) light and right-handed circularly polarized light (RHCP) light is equal. LHCP photons carry a momentum of $-\hbar$ and RHCP photons carry a momentum of $+\hbar$. The conservation of angular momentum dictates therefore that the two different helicities excite electrons with opposite spin. Since the spin currents in this thesis are generated by optical excitation of electrons, this implies a possible helicity dependency of the optical induced spin-transfer-torque. However, in transition metals the selection rules are not so strict due to spin-orbit coupling [43], which could lead to helicity-independent generation of spin currents. The linear polarization of the pump beam is converted to circularly polarized light by a linear polarizer and a quarter-wave plate (QWP). The angle between their axis determines the light polarization: 0 degrees gives linearly polarized light and $\pm 45$ degrees gives circularly polarized light with opposite helicities.

The results are shown in figure 6.6. In the left-hand side of the figure the demagnetization curves for QWP axis rotations of 0 degrees (black curve), $+45$ degrees (red curve) and $-45$ degrees (blue curve) are shown. In the right-hand side of the figure, the OSTT-induced oscillations of the spin absorption layer are shown in the same colours. The phase of the oscillations is $+\pi/2$ for all three QWP axis rotations. This means that in all three measurements the OSTT on the spin absorption layer is exerted by electrons with spins pointing along the same direction. The spin polarization direction of the optical generated spin current is thus helicity-independent. Furthermore, the inset of the figure shows that the canting angle for the three QWP configurations was not
significantly changed. The pump fluence and the pump-probe overlap were constant during
the three measurements. The constant canting angle means that the net absorbed magnetic
moment in the spin absorption layer is equal for the three QWP axis rotations. The equal spin
polarization direction and the constant canting angle indicate that the optical generation of
hot-electron spin currents is helicity-independent.

Figure 6.6: Demagnetization and oscillations of sample 2 with linearly polarized pumping (black curve)
and circularly polarized pumping (blue and red curves). The graph shows no indication of a helicity-
dependent spin current generation. The inset shows that the canting angle of the spin absorption layer
was not significantly changed.

6.4.2 Fluence of the pump beam

To investigate the behaviour of an optical induced STT as function of demagnetization, mea-
surements are performed with varying pump fluences. This is done by inserting a half wave
plate (HWP) and a linear polarizer in the pump beam path. By fixing the linear polarizer axis
and rotating the half wave plate axis, the fluence of the pump beam can be varied with a fixed
polarization. The pump fluence at the sample was varied from 0.26 mJ/cm² to 0.85 mJ/cm².

Figure 6.7a shows the total demagnetization of the spin injection layer as function of laser fluence.
The demagnetization is approximately linear dependent on the pump fluence with a slope of
6.1 ± 0.3 % per mJ/cm². The linear fit depicted an intercept with the y-axis of 0.71. This could
be caused by a misalignment of the pump-probe overlap. Furthermore, the demagnetizations at
a pumping fluence of 0.70 mJ/cm² and 0.62 mJ/cm² were almost equal. Apart from a possible,
not intended, change in pump-probe overlap, no physical explanation could be provided for this.
Because of the offset and the equal demagnetization for two fluences, it is difficult to analyse the
generation of spin current as function of pump fluence. However, the generation of OSTT can
be investigated as function of demagnetization of the spin injection layer. It will be explained in
this section that this facilitates the comparison of data between various measurements.

For every laser fluence the canting angle of the spin absorption layer is calculated using equation
4.16. The canting angle is plotted against the demagnetization of the spin injection layer.
CHAPTER 6. INVESTIGATING ULTRAFAST LASER-INDUCED SPIN-TRANSPORT-TORQUE

Figure 6.7: (a) Demagnetization of sample 2 as function of laser fluence. The demagnetization could be fitted with a non-zero intercept through the y-axis, which could not be explained. (b) Canting angle of the spin absorption layer as function of demagnetization of the spin injection layer. The canting angle could be fitted with a linear function with a slope of $14.1 \pm 0.2$ mdegrees/% demagnetization.

in figure 6.7b. The canting angles are fitted with a linear curve, resulting in a slope of $14.1 \pm 0.2$ mdegrees/%. The intercept of the fit was fixed at the origin, since a demagnetization of 0% means that no spin is transported and no OSTT is exerted on the spin absorption layer. The linear dependency makes it possible to compare various samples using the canting angle per 1% demagnetization of the spin injection layer. Since the typical spot sizes of pump and probe are of the order of microns, the alignment of the pump-probe overlap is a very delicate task. Comparing the canting angles of different measurements is therefore highly sensitive for small misalignments of the pump and probe spots. By comparing the canting angle per 1% demagnetization of the spin injection layer, effects created by pump-probe overlap differences between measurements are eliminated.

The linear dependency of the canting angle on the demagnetization was measured for fluences up to 0.85 mJ/cm$^2$. It is predicted that the spin currents are saturated for large laser fluences [14, 15]. The saturation of the spin currents at large pump fluences will result in a saturated canting angle of the spin absorption layer. It is therefore expected that the linear relation between the spin injection layer demagnetization and the spin absorption canting angle only holds for a certain range of pump fluences, which is the case for the measurements presented in this thesis. The measured canting angles of the experiments throughout this thesis can thus be compared by using the canting angle per 1% demagnetization of the spin injection layer.

6.5 Discussion of the reference sample

The results of sample 1 show a clear demagnetization of the spin injection layer of typically <10%. The demagnetization is partly caused by a spin current, which flows through the spacer layer from the spin injection layer into the spin absorption layer. This spin current cants the magnetization
of the spin absorption layer towards the surface normal. The magnetization dynamics on long
time scale are governed by a single-frequency oscillation, caused by laser-induced spin-transfer-
torque on the spin absorption layer. It was shown that the canting angle resulting from a
laser-induced spin-transfer-torque is linear with demagnetization for the fluences used in this
thesis. This makes it possible to eliminate effects induced by variations in pump-probe overlap
between measurements.

In chapter 2, it was shown that the required canting angle for an all-optical switch of the spin
absorption layer is proportional to the Gilbert damping factor $\alpha$ of that layer (see equation
2.33). To examine the quality of a sample in terms of switching possibility, consider the ratio
$\theta_M/\theta_{M,\text{crit}}$ between the measured canting angle and the critical canting angle (see section 2.5).
In section 6.4 it was shown that the canting angle is proportional with pump fluence. Assuming
full demagnetization, an upper limit of the maximum achievable canting angle is thus $100\theta_{M,1\%}$, where $\theta_{M,1\%}$ is the canting angle of the spin absorption layer with 1% demagnetization of the
spin injection layer. Combining this with equation 2.33, a quality factor $R_{CA}$ in terms of a
switching possibility of the spin absorption layer is defined:

$$R_{CA} = \frac{100\theta_{M,1\%}}{\theta_{M,\text{crit}}} = \frac{100\theta_{M,1\%}}{90\alpha}, \quad (6.1)$$

This factor is valid for $\phi_{M,\text{final}} = 90$ degrees, the angle for which the magnetization has a small
component opposite to its initial direction. This was defined in section 5.3 as a switch of the
spin absorption layer. If $R_{CA} < 1$, the spin injection layer is fully demagnetized before the spin
absorption layer is canted enough towards the surface normal to result in a switch. A sample
with $R_{CA} < 1$ will therefore never be able to switch the spin absorption layer by laser-induced
spin-transfer-torque. It is thus desirable that $R_{CA} \geq 1$. However, the $R_{CA}$ factor assumes a linear
relation between the canting angle of the spin absorption layer and demagnetization of the spin
injection layer. As was discussed in section 6.4, the linear relation between demagnetization and
canting angle does not hold for large fluences. Furthermore, the required canting angle in the $R_{CA}$
factor is calculated under the condition that the canting angle is small. The condition $R_{CA} \geq 1$
is therefore only a minimum condition for the possibility of switching the spin absorption layer
by OSTT.

In section 6.3 the canting angle $\theta_{M,1\%}$ of sample 1 was determined to be $15 \pm 2$ mdegrees. The
damping factor of sample 1 was $0.037 \pm 0.002$, resulting in a quality factor $R_{CA} = 0.45 \pm 0.05$.
Since this is below unity, it is not possible to switch the spin absorption layer using optical
induced spin-transfer-torque in a magnetic bilayer containing the layers of sample 1. In the
following experiments the magnetic bilayer of sample 1 is optimized to induce an increase of the
quality factor $R_{CA}$.

The number of electrons which carry the absorbed magnetic momentum was calculated to be
$1.3 \times 10^8$. The resulting canting angle of this number of electrons is consistent with the simulation
results provided in section 5.5. The total number of excited electrons in the spin injection
layer is $5.9 \times 10^8$, meaning that 22% of the excitation energy is converted to a laser-induced
spin-transfer-torque.

Furthermore, it is estimated that approximately 40% of the excitations in nickel occur in the
majority electron band [44]. Assuming such a percentage for the Co/Ni layer, this means that
approximately 55% of the excited majority electrons exert a spin-transfer-torque on the spin
absorption layer. Considering that the total pulse energy was $2 \text{nJ}$, the power efficiency of the reference sample is $1.6\%$.

### 6.6 Investigating the spin injection layer

In the first experiment towards optimization of the canting angle, the spin injection layer was investigated. In this section the Co:Ni ratio will be changed by increasing the amount of cobalt in the Co/Ni multilayer. Based on the density of states, this should increase the spin polarization of the spin current generation in the Co/Ni spin injection layer, as can be argued as follows. In figure 6.8 the density of states for Ni (figure a) and Co (figure b) is shown as function of the energy $E_e - E_F$. The blue dashed line indicates the energy of the laser photons, meaning that all excited electrons take an energy state between the Fermi level and the blue dashed line. As was explained in section 2.2, the spin current is generated by the asymmetry between majority and minority phase space for scattering of excited electrons. In the figure, the green area shows the phase space of the majority electrons and the red area shows the phase space for minority electrons. The ratio between them depicts the asymmetry in the phase space. Inspecting the DOS for both Ni and Co, it is seen that the phase space asymmetry for energy states between the Fermi level and the pumping level is larger for cobalt. Based on the DOS, the spin polarization of a current generated in a cobalt layer should therefore be larger. Theoretical studies show that the asymmetry between majority and minority lifetimes is indeed larger for Co than for Ni [45]. Increasing the Co:Ni ratio in the injection layer should therefore increase the canting angle of the spin absorption layer. However, in figure 6.8c experimental data found in literature of the ratio between majority and minority lifetimes is shown. The black symbols depict the measured lifetime ratios of Co and the red symbols depict the measured lifetime ratios of Ni. The data are taken from three references: Aeschlimann et al. [46], Knorren et al. [45] and Goris et al. [47]. As can be seen, the lifetime ratio of nickel is larger than the lifetime ratio of cobalt for most excitation energies below $1.2 \text{eV}$. In this thesis, the electrons are excited with a photon energy of $1.57 \text{eV}$. This could result in a larger asymmetry between majority and minority electrons for cobalt as compared to nickel. The difference between the calculated and the experimental values is caused by spin mixing effects like elastic scattering and spin-orbit mediated spin-flips.

Furthermore, the light absorption in Co is larger than the light absorption in Ni. This results in a larger light absorption of the spin injection layer and subsequently in a larger spin current generation in layers with increased Co:Ni ratio. In this section, three samples will be discussed to investigate the effect of increasing the amount of cobalt in the spin injection layer. The compositions of these samples are summarized in table 6.1, along with the calculated light absorption of the spin injection layer. To calculate the light absorption, a Matlab script based on the transfer matrix method was used, which is available from reference [49]. The Co:Ni ratio of the reference sample was 1:2.4. In the first sample, sample 3a, the Co:Ni was increased to 1:0.9. In sample 3b the Co:Ni is further increased to 1:0.3. In sample 3c the Co/Ni multilayer was similar to the one in sample 3a, with an increased Co dusting layer at the Ni/Co/Cu interface.
Figure 6.8: Density of states for nickel (a) and cobalt (b). The DOS is taken from reference [48]. The figures show that the asymmetry in phase space (ratio between the red and green areas) for cobalt is larger than for nickel. (c) Experimental data of measured spin-dependent lifetime ratios of Co and Ni, taken from references [45–47].

Table 6.1: Overview of the samples used to investigate the effect on spin current generation by changing the spin injection layer. In each sample, the Co:Ni ratio in the Co/Ni multilayer is adjusted.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Spin Injection layer (4+ Co/Ni)</th>
<th>Spin absorption layer</th>
<th>Light absorption</th>
<th>Sample</th>
<th>Spin Injection layer (4+ Co/Ni)</th>
<th>Spin absorption layer</th>
<th>Light absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Co[0.2]Ni[0.6] + Co[0.2]</td>
<td>Co [3]</td>
<td>7.4%</td>
<td>3b</td>
<td>Co[0.6]Ni[0.4] + Co[0.2]</td>
<td>Co [3]</td>
<td>9.0%</td>
</tr>
<tr>
<td>3a</td>
<td>Co[0.4]Ni[0.6] + Co[0.4]</td>
<td>Co [3]</td>
<td>7.9%</td>
<td>3c</td>
<td>Co[0.4]Ni[0.4] + Co[0.2]</td>
<td>Co [3]</td>
<td>8.3%</td>
</tr>
</tbody>
</table>

6.6.1 Increase of the Co:Ni ratio with constant Co dusting layer

In sample 3a, the spin injection layer was changed to a sequence of 4 × Co[0.4]/Ni[0.4] with a Co[0.2] dusting layer, increasing the Co:Ni ratio to 1:0.9. A measurement of the magnetization dynamics is depicted in figure 6.9. In the left-hand side, the red curve shows the demagnetization of the spin injection layer, which was $5.7 \pm 0.3\%$. The black curve shows the demagnetization curve of sample 1, which had a total demagnetization of $8.3 \pm 0.4\%$. The pump-probe overlap was kept constant during the measurements, thereby eliminating a difference in demagnetization caused by a change in pump-probe overlap. The reduction in demagnetization could be caused by either a thermal effect or a spin transport effect.

In the case of a thermal effect, a reduced demagnetization can be caused by an increase of the Curie temperature, as can be explained by considering the inset in the right-hand side of figure 6.9. The magnetization curve as function of temperature for sample 1 is schematically drawn with a black curve. The magnetization curve for a sample with increased Curie temperature, such as sample 3a, is schematically drawn with a red curve. The green lines show the tangent lines of the magnetization curves for the initial temperature $T_0$ before laser excitation of the sample. These tangent lines show that the derivative at $T_0$ is smaller for sample 3a, meaning that sample 3a has a smaller demagnetization rate upon laser heating. Increasing the Co:Ni ratio is expected to increase the Curie temperature of the Co/Ni multilayer [50, 51]. This results in a smaller demagnetization rate for sample 3a as compared to sample 1.
For the case of a spin transport effect, a reduction in demagnetization can be caused by a spin transport effect if the net transport of magnetic moment out of the spin injection layer is decreased. The magnetic moment absorbed by the absorption layer can be calculated with equation 4.15. In the right-hand side of figure 6.9 the red curve shows the oscillation of the spin absorption layer of sample 3a with an IP field of 100 mT. Using the amplitude of this oscillation, the magnetic moment absorbed by the spin absorption layer in sample 3a ($1.2 \pm 0.1 \times 10^{-12} \text{emu}$) was not significantly changed, as compared to the magnetic moment absorbed in sample 1 ($1.3 \pm 0.1 \times 10^{-12} \text{emu}$). It is therefore concluded that the generation of spin current is not significantly decreased, meaning that the smaller demagnetization can not be caused by an increase of generated spin currents in the spin injection layer. However, the magnetic moment lost by the spin injection layer in sample 3a ($2.6 \pm 0.2 \times 10^{-11} \text{emu}$) was not significantly changed as compared to sample 1 ($3.0 \pm 0.2 \times 10^{-11} \text{emu}$) as well. Since the total magnetic moment of the spin injection layer is increased by increasing the Co:Ni ratio, the lost magnetic moment is a smaller percentage of the total magnetic moment, resulting in the apparent smaller demagnetization rate. Based on the presented results, it is non-conclusive whether a decrease in the demagnetization is occurring at all and if so, whether it is mainly caused by a thermal or a spin transport effect.

To examine the effect of the Co:Ni ratio on the spin polarization of the spin current generated in sample 3a, consider the light absorption of the spin injection layer. In table 6.1 it was shown that the light absorption in sample 3a is increased by 6% with respect to sample 1. The excitation of hot electrons is directly related to the light absorption, meaning that the number of hot electrons, and subsequently the transported magnetic moment, should be increased by 6% in sample 3a. It is likely that the change in absorbed magnetic moment was less than 6%, implying a decrease in spin polarization of the generated spin current. This indicates that the lifetime ratio between majority and minority electrons is decreased for the increased Co: Ni in sample 3a, as was predicted by the experimental data found in literature (see figure 6.8c).
The efficiency of sample 3a was 4.4 ± 0.5%. The efficiency of spin generation and transport in sample 3a is thus not significantly changed as compared to sample 1, which had an efficiency of 4.4 ± 0.4%. The canting angle of the spin absorption layer of sample 3a was 19 ± 2 mdegrees per 1% demagnetization of the spin injection layer. This indicates an increase in the canting angle (sample 1: \( \theta_{M,1\%} = 15 \pm 2 \) mdegrees), attributed to the decrease of the demagnetization. The Gilbert damping factor \( \alpha \) of sample 3a was 0.040 ± 0.002, resulting in a \( R_{CA} \) factor of 0.53 ± 0.07. For sample 1 the \( R_{CA} \) factor was 0.45 ± 0.05. This indicates that increasing the Co:Ni ratio results in a non-significant increase in the \( R_{CA} \) factor. However, for both samples \( R_{CA} < 1 \), meaning that in both samples the spin injection layer is fully demagnetized before the spin current required to switch the spin absorption layer is generated.

To investigate a further increase of the Co:Ni ratio, the Co/Ni ratio was increased to 1:0.3 in a second sample (sample 3b). The static hysteresis loop of this sample is shown in figure 6.10a. The hysteresis loop shows that both the spin injection layer and the spin absorption layer exhibit in-plane magnetization. The shape anisotropy of the spin injection layer in this sample is stronger than the surface anisotropies, caused by the increased magnetization of that layer. This results in an in-plane alignment of the spin injection layer. Consequently, the spin polarization of the spin current is parallel to the spin absorption layer's magnetization, leading to the absence of an OSTT in this sample.

![Figure 6.10: (a) Static hysteresis loop of sample 3b. The hysteresis loop indicates that the magnetization of both FM layers in this sample is aligned in the surface. (b) Static hysteresis loop of sample 3c. The slanted switching indicates the existence of a multi-domain state with a spread in anisotropies.](image)

### 6.6.2 Co dusting layer thickness in the Ni/Co/Cu interface

In the third sample, sample 3c and fabricated in a new batch, the thickness of the 0.2 nm Co dusting layer in the Ni/Co/Cu interface was increased to 0.4 nm. The other layers in the spin injection layer were equal to sample 3a (see table 6.1). The static hysteresis loop of the sample is shown in figure 6.10b. The hysteresis loop shows a slanted-like switching of the magnetization. This indicates that the spin injection layer is in a multi-domain state at low fields. The anisotropy is slightly varying over the different domains. The result is that the domains having small...
anisotropy switch at smaller fields than the ones with large anisotropy, causing the slanted-like switching behaviour. The spin injection layer thickness in this sample is 3.6 nm. The slanted switching indicates that the domain length of this spin injection layer is smaller than its thickness. However, before measuring the ultrafast magnetization dynamics, a field is applied to align all spins of the injection layer approximately along the surface normal. The strength of this field is 120 mT, well above the coercivity of the largest-anisotropy domains. After applying this field there is a collective alignment of the spins along the surface normal. It is therefore assumed that the multi-domain state at low fields does not affect the optical induced spin-transfer-torque.

The magnetization dynamics of sample 3c are shown in figure 6.11 with a blue curve, along with the magnetization dynamics of samples 1 and 3a for the sake of clarity. The demagnetization curve is shown in the left-hand side of the figure. The total demagnetization was decreased from 5.7 ± 0.3 % for sample 3a to 3.6 ± 0.2 % for sample 3c. The pump-probe overlap was kept constant during the measurements of samples 1, 3a and 3c. The decrease in lost magnetic moment for sample 3c was at least 25 % with respect to sample 3a, meaning that indeed more magnetic moment was lost in sample 3c. As in the comparison of sample 1 with sample 3a, the change in demagnetization between samples 3a and 3c could by caused by either thermal effects or spin transport effects. However, the absorbed magnetic moment in the spin absorption layer of sample 3c was increased by at least 14 % with respect to sample 3a. The increased absorbed magnetic moment indicates a larger generation of spin current in sample 3c. This should increase the demagnetization of the spin injection layer in sample 3c, whereas a decrease in the demagnetization was observed. It is therefore concluded that the decrease in demagnetization for sample 3c is dominated by thermal effects.

The oscillations for samples 1, 3a and 3c with an IP field of 100 mT are compared in the right-hand side of figure 6.11. The frequency of the oscillations observed in the three samples was of similar magnitude, confirming that the oscillation is originating from the unchanged in-plane layer. The efficiency was increased from 4.4 ± 0.5 % for sample 3a to 9.7 ± 0.9 % to sample 3c. This means that the generation mechanism for spin currents is more efficient in sample 3c. This can be explained as follows. As will be shown in section 6.8, a Ni/Cu interface blocks the transmission of the greater part of the incident spins. Since the atomic diameter of Co is 3.04 Å [52], the dusting layer of 0.2 nm is not fully covering the last Ni layer of the Co/Ni spin injection layer. This results in local Ni/Cu interfaces in the Ni/Co/Cu interface, which block the transmission of spins. Increasing the dusting layer to 0.4 nm increases the Co coverage of the Ni layer, thereby increasing the transmission of spins through the Ni/Co/Cu interface. The result is an increase in the magnetic moment transported away from the spin injection layer and absorbed by the spin absorption layer. This results in the observed increase of the efficiency to 9.7 ± 0.9 % for sample 3c.

The increased efficiency implies that the canting angle of the spin absorption layer was increased. For sample 3c, the canting angle $\theta_{M,1\%}$ was $43 \pm 5$ mdegrees, which is indeed a significant increase as compared to both samples 1 and 3a. The Gilbert damping factor of sample 3c was 0.040 ± 0.002, resulting in a quality factor of $R_{CA} = 1.2 \pm 0.1$. For sample 3c the $R_{CA}$ is thus larger than unity. As was argued in section 6.5, this means that a switch of the spin absorption layer does not require a full demagnetization of the spin injection layer in this sample.

The condition $R_{CA} \geq 1$ assumes a linear relation between the canting angle and the demagnetization. It is expected that this proportionality is not valid for large demagnetization rates. Furthermore, demagnetization rates of the spin injection layer in the experiments performed
throughout this thesis are below 10%. A switch of the spin absorption layer is therefore not observed in the experiments of this thesis. However, the increase in the $R_{CA}$ shows the positive effect of increasing the Co dusting layer in the Ni/Co/Cu interface of the spin injection layer.

6.6.3 Summary

In this section the spin injection layer was investigated using three samples. The Co:Ni ratio was increased from 1:2.4 for sample 1, to 1:0.9 for sample 3a and to 1:0.3 to sample 3b. It was shown that a Co:Ni ratio of 1:0.3 increases the shape anisotropy enough to force an in-plane alignment of the spin injection layer. Increasing the amount of Co decreased the demagnetization of the spin injection layer, which was concluded using sample 3c to originate from an increase of the Curie temperature. It was furthermore shown that increasing the dusting layer of Co from 0.2 nm to 0.4 nm increases the generation of spin currents in the spin injection layer, which was attributed to a larger Co coverage of the last Ni layer in the Co/Ni multilayer. The important results of investigating the spin injection layer are shown in table 6.2.

In the definition of $R_{CA} \geq 1$, a switch was defined as a small magnetization component along the opposite direction of the initial magnetization alignment axis, after precessional motion around the demagnetization field. If the structures investigated in this thesis are to be used as an all-optical memory element, the spin absorption layer should be driven into a complete switch. This means that the rotation angle $\phi_{M,\text{final}}$ should be 180 degrees. For a complete switch the condition is therefore $R_{CA} \geq 2$. In the next section it will be investigated how optimization of the spin absorption layer changes the $R_{CA}$ factor towards a switch of the spin absorption layer.
### Table 6.2: Table with the summarized results of the samples used to investigate the spin injection layer.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Spin injection layer (4x Co/Ni)</th>
<th>Spin absorption layer</th>
<th>η (%)</th>
<th>$\theta_{\text{M,13%}}$ (mdegrees)</th>
<th>$R_{\text{CA}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Co[0.2]/Ni[0.6] + Co[0.2]</td>
<td>Co [3]</td>
<td>4.4 ± 0.4</td>
<td>15 ± 2</td>
<td>0.45 ± 0.05</td>
</tr>
<tr>
<td>3a</td>
<td>Co[0.4]/Ni[0.4] + Co[0.2]</td>
<td>Co [3]</td>
<td>4.4 ± 0.5</td>
<td>19 ± 2</td>
<td>0.53 ± 0.07</td>
</tr>
<tr>
<td>3b</td>
<td>Co[0.6]/Ni[0.2] + Co[0.2]</td>
<td>In-plane spin injection layer</td>
<td>In-plane spin injection layer</td>
<td>In-plane spin injection layer</td>
<td>In-plane spin injection layer</td>
</tr>
<tr>
<td>3c</td>
<td>Co[0.4]/Ni[0.4] + Co[0.3]</td>
<td>Co [3]</td>
<td>9.7 ± 0.9</td>
<td>43 ± 5</td>
<td>1.2 ± 0.1</td>
</tr>
</tbody>
</table>

#### 6.7 Cobalt spin absorption layer wedge

By measuring the dependence of an optical induced spin-transfer-torque on the thickness of the spin absorption layer, the penetration depth of transverse spins in the Co spin absorption layer can be determined. Furthermore, the term representing STT in the LLG equation is inversely proportional with spin absorption layer thickness. It is therefore expected that the canting angle increases if the thickness is decreased. To examine the penetration depth of transverse spins in a Co layer, a wedge of the Co layer was fabricated using the wedge mask in the sputtering equipment (see section 3.1). This sample will be referred to as sample 4. The geometry of the sample is shown in figure 6.12a. The spin injection layer consisted of a sequence of 4 × Co[0.2]/Ni[0.6], with a Co[0.2] dusting layer (similar to sample 1). The spin absorption layer consisted of a wedge-shaped Co layer, ranging from 0 nm to 8 nm. The MO-sensitivity ratio between the spin injection and the spin absorption layers changes with increasing thickness of the spin absorption layer. Before discussing the magnetization dynamics, static magnetic properties are therefore discussed. Secondly, demagnetization curves and oscillations curves are shown. Finally, the penetration depth of the transverse spins in the cobalt layers and the resulting canting angle is discussed.
6.7.1 Static magnetic properties

The light absorption of both the spin injection and the spin absorption layers change when the thickness of the latter is increased. The MO-sensitivity ratio between the two layers consequently becomes thickness dependent. In figure 6.12b static hysteresis loops are shown for measurements with a spin absorption layer thickness of 3 nm (black curve) and 7 nm (red curve). The switching amplitude of the spin injection layer in the 7 nm measurement is clearly decreased as compared to the 3 nm measurement. In contrast, the slope of the linear contribution originating from the spin absorption layer is larger for the 7 nm measurement as compared to the 3 nm measurement. Assuming a similar saturation field of the spin absorption layer in both measurements, this means that the MO-sensitivity of the spin absorption layer is increased. Combining the decreased MO-sensitivity to the spin injection layer and the increased MO-sensitivity to the spin absorption layer, the sensitivity ratio between the spin injection and the spin absorption layer is decreased.

6.7.2 Magnetization dynamics

In figure 6.13 the magnetization dynamics of the measurements with a 3 nm spin absorption layer thickness (black curve) and with a 7 nm thickness (red curve) are compared. In the left-hand side of the figure the demagnetization curves of both measurements are shown. It is clearly visible that a 7 nm thick spin absorption layer results in a much smaller demagnetization of the spin injection layer. The smaller demagnetization for the 7 nm measurement is caused by a decreased light absorption by the spin injection layer. Magnetization dynamics measurement for longer time scale showed a clear oscillation for both thicknesses, as is shown in the right-hand side of figure 6.13. The field was applied in-plane and had a strength of $\mu_0 H_{app} = 100 \text{ mT}$. A small increase in the frequency is observed for the 7 nm measurement as compared to the
3 nm measurement, caused by the thickness dependent anisotropy constant (see equation 2.5). Furthermore, a thickness dependency of the saturation magnetization is observed, as is shown in the inset in the right-hand side of the figure. As was shown in the Kittel formula (equation 2.24), a thickness-dependent saturation magnetization results in a thickness-dependent frequency. This reduction in saturation magnetization could be resulting from interactions of the spins in the cobalt layer with spins in the spacer layer or capping layer and from a demagnetization which has not fully recovered to the equilibrium magnetization. However, a thorough investigation of the thickness dependency of the saturation magnetization is beyond the scope of this thesis.

The efficiency of spin transport between the spin injection and the spin absorption layer is a fingerprint of the absorbed spins in the spin absorption layer. To investigate the penetration depth of transverse spins in the Co spin absorption layer, the efficiency is therefore inspected as function of thickness. Consider the penetration depth $\lambda_{tr}$, for which all transverse spins of a spin current are absorbed. For $t_{Co} > \lambda_{tr}$ all transverse spins entering the spin absorption layer are absorbed. The efficiency, which reflects the number of absorbed spins, should be a constant for $t_{Co} > \lambda_{tr}$. For $t_{Co} < \lambda_{tr}$ the absorption of the transverse spins should increase with increasing layer thickness. In this case, the efficiency should increase with increasing spin absorption layer thickness.

The efficiency is plotted as function of the Co spin absorption layer thickness in figure 6.14a. For a Co layer thickness of zero no magnetic moment should be absorbed and the efficiency should be zero. For thicknesses $t_{Co} > \lambda_{tr}$ it was explained that the absorption and the efficiency should be constant. Furthermore, the absorption of transverse spins should have an exponential dependency on the thickness of the spin absorption layer [53]. The plotted efficiency is therefore fitted with $\eta_{\text{max}} \left[ 1 - e^{-t_{Co}/\lambda_{tr}} \right]$, where $\eta_{\text{max}}$ is the efficiency with total transverse spin absorption and $\lambda_{tr,e}$ is the thickness with an efficiency of $\eta_{\text{max}}/e$. As can be seen, the measurements for thicknesses >5.4 nm seem to result in an efficiency smaller than would be expected. This could be caused by a local structural variation in the spin absorption layer. However, the exponential fit depicted a
\( \lambda_{tr,e} \) of 0.73 ± 0.08 nm and a maximum efficiency of 4.37 ± 0.09 %, which is drawn with a dashed blue line in the figure. At a thickness of 1.7 nm, 90% of the spins are absorbed. In chapter 2, equation 2.17 was derived for an estimation of the spatial spin dephasing period in the spin absorption layer. For cobalt, this spatial dephasing period should result in a penetration depth of approximately 6 nm. However, this equation was derived using a free electron description of spin transport. Due to spin relaxation processes not accounted for by the free electron model, the measured penetration depth is smaller: approximately 1.7 nm. This result is consistent with earlier reported values of the penetration depth of transverse in spins [53, 54].

![Figure 6.14](image)

**Figure 6.14**: (a) Measurements of the efficiency of the laser-induced STT as function of the Co spin absorption layer thickness. The maximum efficiency was 4.37 ± 0.09 %. (b) Canting angle of the spin absorption layer as function of the product \( M_{\text{sat}} t_{\text{Co}} \). The red line is a guide to the eye.

### 6.7.3 Canting angle of the Co spin absorption layer

The canting angle resulting from OSTT is inversely proportional with the spin absorption layer thickness, as can be derived from the LLG equation (equation 2.19). The fact that the penetration depth of transverse spins in cobalt is smaller than 3 nm should therefore lead to a larger canting angle for thicknesses below 3 nm. Figure 6.14b shows the canting angle of the spin absorption layer as function of layer thickness. To eliminate the variation of the saturation magnetization, the canting angle is plotted as function of \( M_{\text{sat}} t_{\text{Co}} \). The graph shows a reciprocal dependency of the canting angle on \( M_{\text{sat}} t_{\text{Co}} \), as can be expected from the LLG equation (equation 2.19). A red dashed line provides a guide to the eye, which is the product of a reciprocal function and an exponential function describing thickness-dependent spin absorption. The maximum canting angle \( \theta_{M,1\%} \) was achieved with a spin absorption layer thickness of 1 nm and was 38 ± 4 mdegrees.

Due to the inverse proportionality of the canting angle on layer thickness, the canting angle can be increased by decreasing the thickness. However, in figure 6.15a the thickness dependency of the Gilbert damping angle is depicted as function of \( t_{\text{Co}} \). It is seen that \( \alpha \) is inversely proportional to \( t_{\text{Co}} \). This is indicative for a spin pumping effect. Due to this effect spins are
transported out of the spin absorption layer, caused by the precession of the magnetization in the spin absorption layer [55]. The spin pumping effect is effectively a channel for spin loss and manifests itself therefore as an increase of the Gilbert damping factor. The spin diffusion length of Cu is 350 nm at room temperature [56]. It is therefore expected that the Cu layer of 5 nm does not absorb any spins. The spins flowing out of the spin absorption layer are thus absorbed by the spin injection layer and the Pt capping layer. The large enhancement of the Gilbert damping indicates that these layers are good spin sinks. If the spin injection layer would not be a good spin sink, the spin current reaching this layer would be reflected and flow back into the spin absorption layer. The enhancement of the Gilbert damping would in that case be suppressed. Note that the oscillation causing the spin pumping effect occurs on a time scale where the laser-induced STT is already exerted. The spin currents caused by the spin pumping effect are not related to the laser-induced spin currents and do not affect the magnitude of the laser-induced STT. However, they do decrease the possibility of switching the spin absorption layer due to the Gilbert damping enhancement.

Due to the increase in Gilbert damping it is not necessarily advantageous to decrease the Co layer thickness in order to increase the canting angle. In figure 6.15b the quality factor $R_{CA}$ is plotted as function of $t_{Co}$. The figure shows that the quality factor is proportional with the spin absorption layer thickness with a negative slope. This linear dependency shows that a smaller thickness increases the possibility for a switch of the magnetization. The linear fit of $R_{CA}$ depicted an intercept with the $y$-axis of $R_{CA} = 0.72 \pm 0.02$, which is an upper limit for $R_{CA}$ using sample 4. The maximum value of $R_{CA}$ is thus less than unity for a cobalt spin absorption layer, using the Co/Ni multilayer as in sample 4. It is concluded that it is not possible to switch the spin absorption layer of sample 4 using optical induced spin-transfer-torque.
6.8 Nickel spin absorption layer

The previous results showed that the maximum canting angle using a cobalt spin absorption layer is too small for all-optical switching of the spin absorption layer. Decreasing the saturation magnetization of the spin absorption layer increases the ratio between absorbed magnetic moment and the total magnetic moment. Decreasing $M_{\text{sat,IP}}$ increases therefore the canting angle of the spin absorption layer. To investigate the effect of decreasing the saturation magnetization on the canting angle, the Co spin absorption layer is substituted by a Ni layer. The spin injection layer was a sequence of $4 \times \text{Co}[0.2]/\text{Ni}[0.6]$, with a Co[0.2] dusting layer, similar to sample 1.

![Figure 6.16: Magnetization dynamics of a structure with a 3 nm Ni spin absorption layer on a Cu spacer layer (black curve). The red curve shows the magnetization dynamics of a Ni spin absorption layer with a Cu/Co/Ni interface, where the Co dusting layer was 0.2 nm thick. The grey curve shows the oscillation of the reference sample.](image)

In figure 6.16 the magnetization dynamics are depicted with a black curve for the sample with the Ni absorption layer. The thickness of the Ni absorption layer was 3 nm. On the left-hand side a clear demagnetization of the spin injection layer is observed. The total demagnetization was $5.4 \pm 0.3 \%$. In the right-hand side of figure 6.16 the magnetization dynamics on longer time scale are shown (again with a black curve). The in-plane field $\mu_0 H_{\text{app}}$ was 100 mT. No clear evidence of an oscillation of the spin absorption layer was measured. This indicates that the generated spin current in the spin injection layer is blocked somewhere in the structure. In this sample there is a Cu/Ni interface between the spacer layer and the spin absorption layer. It was discussed before that this interface blocks spin transport and this will be shown in this section. Spin blocking at the Cu/Ni arises because of the high solubility of Ni in Cu. Consequently, there is a small diffusion layer with a mixture of Ni and Cu atoms, which reduces the magnetic moment of the Ni atoms due to the low Curie temperature of Ni. The result is a shift of the majority electron band structure which now does not match with the Cu band structure. This results in a large scatter probability of both minority and majority electrons at the Cu/Ni interface, effectively blocking any spin transport through the interface.

The spin blocking effect can be overcome by inserting a dusting layer of Co between the Cu and Ni layers. In figure 6.16 the magnetization dynamics are shown of a sample with a dusting
layer of 0.2 nm Co with a red curve. This sample will be referred to as sample 5. The total demagnetization was 5.2 ± 0.3%. The demagnetization curve shows that the demagnetization proceeds faster than the demagnetization of the sample without the Co dusting layer. The demagnetization of the latter was measured with the field applied at an angle with the sample of 20 degrees. The demagnetization curve is a sum of the demagnetization of the spin injection layer and the spin absorption layer. The demagnetization of sample 5 was measured with an in-plane field and contains therefore only the demagnetization of the spin injection layer. This could explain the observed difference in demagnetization time. However, the feature of interest is the remagnetization curve, as shown with a red curve in the right-hand side of figure 6.16. It is clearly visible that inserting the Co dusting layer leads to spin transmission through the new Cu/Co/Ni interface. To compare the oscillation observed in sample 5 with the oscillation observed using a Co layer, the latter is added with a grey curve, normalized to a demagnetization of 5.2%. A clear reduction in the frequency of the oscillation in sample 5 is observed. Inspection of the Kittel relation (equation 2.24) depicts that this can be attributed to a reduction in saturation magnetization of the spin absorption layer of sample 5, which was 215 ± 3 kA/m.

The efficiency of sample 5 was 4.6 ± 0.4%. The efficiency is consistent with the efficiency of sample 1 (which was 4.4 ± 0.4%). This means that for equal demagnetization of the spin injection layer, the spin absorption is equal in both samples. The results of the Co wedge (section 6.7) showed that the reference sample had maximum spin absorption achievable with a Cu/Co interface. This indicated that the transmission of spins through the Cu/Co interface was maximum for sample 1. Since local Cu/Ni interfaces block spin transport, it can be expected that the maximum spin transmission through a Cu/Co/Ni interface is determined by the maximum transmission of spins through the Cu/Co interface. The consistency between the efficiencies of sample 5 and sample 1 shows that a dusting layer of 0.2 nm is already large enough to reach maximum spin transmission through the Cu/Co/Ni interface.

The canting angle in sample 5 was 84 ± 9 mdegrees. Compared to the reference sample (sample 1: θM,1% = 15 ± 2 mdegrees), this is an increase with a factor of 5.5. The Kittel fit for the field dependent frequencies of sample 5 depicted a saturation magnetization of 215 ± 3 kA/m for Ni layer. For sample 1 the saturation magnetization was 1198 ± 6 kA/m, resulting in a ratio $M_{\text{sat},1}/M_{\text{sat},5} = 5.6$. From the LLG equation (equation 2.19) it can be shown that the canting angle is inversely proportional to the saturation magnetization, which is indeed the case in sample 5. Furthermore, this confirms that the 0.2 nm Co dusting layer is thick enough for maximum spin transmission.

The Gilbert damping factor of the spin absorption layer in sample 5 was 0.116 ± 0.006. Since the increase in α is smaller than the increase in canting angle, the quality factor $R_{\text{CA}}$ is increased. The $R_{\text{CA}}$ factor was 0.8 ± 0.1 for sample 5. This is too small for a switch of the spin absorption layer, which requires $R_{\text{CA}} \geq 1$. However, the results presented in this section show that decreasing the saturation magnetization increases the $R_{\text{CA}}$ factor towards a switch of the spin absorption layer.
6.9 Discussion of the spin injection and absorption layer

The reference sample had a $R_{CA}$ factor of maximum 0.50. The wedge-shaped Co spin absorption layer showed that the combination of the reference spin injection layer with a cobalt spin absorption layer results in a maximum $R_{CA}$ of 0.72, which is too small for all-optical switching. It was shown that increasing the Co dusting layer between the spin injection layer and the spacer layer increases the $R_{CA}$ to at least 1.1. With such a sample an all-optical switch of the spin absorption layer is in principle possible, but requires a full demagnetization of the spin injection layer. Furthermore, the condition $R_{CA} \geq 1$ assumes that the proportionality between canting angle and demagnetization holds. This might not be correct.

For a further increase of the quality factor, the Co spin absorption layer was substituted by a Ni layer. This resulted in an increased canting angle and $R_{CA}$ of at least 0.7. Combining the spin injection layer of sample 3c with the Ni spin absorption layer, a $R_{CA}$ of 2.13 could be possible. Furthermore, if the thickness dependency of the quality factor is equal in Ni as compared to Co, a further increase of the $R_{CA}$ to 3.26 is expected. Inspecting the definition of the quality factor (equation 6.1), the required demagnetization for a switch is given by $2/R_{CA} = 2/3.26 = 61\%$. Such a demagnetization rate could be possible in a new laser system currently being aligned in the lab. It is therefore predicted that a further optimization of the spin injection layer and the spin absorption layer will lead to the first experimental observation of ultrafast optical induced spin-transfer-torque.
In this thesis, laser-induced spin-transfer-torque was investigated in non-collinear magnetic bilayers. Fundamental insights about the generation and absorption of spin currents were gained. Furthermore, the possibility of switching the spin absorption layer was investigated. The most important results of the samples are shown in table 7.1. Key parameters for investigating all-optical switching using spin-transfer-torque are the canting angle and quality factor. The canting angle $\theta_{M,1\%}$ depicts the canting angle of the spin absorption layer, assuming a demagnetization of the spin injection layer of 1%. The quality factor $R_{CA}$ is the ratio between the canting angle and required canting angle for a switch. A minimum condition for all-optical switching is $R_{CA} \geq 1$. In this chapter the conclusions of optimizing the investigated magnetic bilayers are provided. It will be argued that in principle, all-optical switching by optimizing the magnetic bilayers is feasible. Experiments to be performed towards such a switch will be suggested.

Table 7.1: Table of the most important results of the experiments performed throughout this thesis.

<table>
<thead>
<tr>
<th>Sample (reference)</th>
<th>Spin injection layer (4x Co/Ni)</th>
<th>Spin absorption layer</th>
<th>$\eta$ (%)</th>
<th>$\theta_{M,1%}$ (mdegrees)</th>
<th>$R_{CA}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Co[0.2]/Ni[0.6] + Co[0.2]</td>
<td>Co [3]</td>
<td>4.4 ± 0.4</td>
<td>15 ± 2</td>
<td>0.45 ± 0.05</td>
</tr>
<tr>
<td>3a</td>
<td>Co[0.4]/Ni[0.4] + Co[0.2]</td>
<td>Co [3]</td>
<td>4.4 ± 0.5</td>
<td>19 ± 2</td>
<td>0.53 ± 0.07</td>
</tr>
<tr>
<td>3c</td>
<td>Co[0.4]/Ni[0.4] + Co[0.4]</td>
<td>Co [3]</td>
<td>9.7 ± 0.9</td>
<td>43 ± 5</td>
<td>1.2 ± 0.1</td>
</tr>
<tr>
<td>4</td>
<td>Co[0.2]/Ni[0.6] + Co[0.2]</td>
<td>Co [1]</td>
<td>3.1 ± 0.3</td>
<td>38 ± 4</td>
<td>0.69 ± 0.08</td>
</tr>
<tr>
<td>4</td>
<td>Co[0.2]/Ni[0.6] + Co[0.2]</td>
<td>Co [7]</td>
<td>4.3 ± 0.4</td>
<td>6.7 ± 0.7</td>
<td>0.40 ± 0.05</td>
</tr>
<tr>
<td>5</td>
<td>Co[0.2]/Ni[0.6] + Co[0.2]</td>
<td>Ni [3] + Co [0.2]</td>
<td>4.6 ± 0.4</td>
<td>84 ± 9</td>
<td>0.8 ± 0.1</td>
</tr>
</tbody>
</table>
7.1 Physics of laser-induced spin currents

Laser-induced ultrafast demagnetization of a ferromagnetic layer is partly caused by a hot-electron spin current, which transports angular momentum away from that layer. In this thesis, these transported spins were detected by measuring the spin-transfer-torque they exert on a second ferromagnetic layer, perpendicular magnetized to the first ferromagnetic layer. The first ferromagnetic layer is referred to as the spin injection layer, whereas the second ferromagnetic layer is referred to as the spin absorption layer. The spin transport efficiency is a fingerprint of the number of spins transported out of the spin injection layer. Table 7.1 depicts the efficiency for the relevant samples.

Using thickness dependent measurements on a cobalt spin absorption layer, it was estimated that all transverse spins entering a cobalt layer are absorbed within four monolayers. It was indicated that sample 1 had maximum spin absorption (compare samples 1 and 4-7 nm in table 7.1). Substituting the Co spin absorption layer by a nickel spin absorption layer showed that a 0.2 nm Co dusting layer at the Cu/Ni interface is thick enough to ensure maximum spin transmission through the new Cu/Co/Ni interface. Without the Co dusting layer, no evidence of a spin-transfer-torque was observed. This illustrates the importance of the morphology of the interfaces in the non-collinear magnetic bilayer.

The importance of the Ni/Cu interface in the spin injection layer was shown by increasing the Co dusting layer in this interface from 0.2 nm to 0.4 nm. The efficiency was increased from 4.4 ± 0.4% to 9.7 ± 0.9% (compare the efficiencies of samples 1 and 3c in table 7.1). Furthermore, the effect of changing the Co:Ni ratio in the spin injection layer was investigated. An increase of the ratio from 1:2.4 to 1:0.9 indicated that the spin polarization of the induced spin current was decreased. This was caused by a smaller lifetime asymmetry between majority and minority electrons in cobalt, as compared to nickel.

In summary, it is concluded that the role of superdiffusive spin currents in ultrafast demagnetization of the spin injection layer depends on the specific composition of that layer. In this thesis, the part of the ultrafast demagnetization caused by spin transport was increased to a maximum of at least 8.8%.

7.2 Optical two-state memory element

The non-collinear magnetic bilayers investigated throughout this thesis could be used as a two-state memory element with all-optical excitation of the states. Injecting a laser-induced spin current in the spin absorption layer resulted in a canted state of the spin absorption layer. Simulations using the LLG equation showed that it is principally possible to switch the magnetization of the spin absorption layer using its demagnetization field. The simulations showed that important parameters influencing the canting angle are the spin polarization of the induced spin current, the magnitude of the spin current and the saturation magnetization of the spin absorption layer. A linear equation was derived to show that the required canting angle for a switch depends linearly on the Gilbert damping factor for small canting angles. This equation was confirmed by the simulations based on the LLG equation.

A quality factor $R_{CA}$ was introduced to quantify the possibility of switching the spin absorption layer. A minimum condition for all-optical switching by laser-induced spin-transfer-torque is
$R_{CA} \geq 1$. In magnetic bilayers with a factor $R_{CA} < 1$ the required canting angle for an all-optical switch requires a demagnetization of the spin injection layer larger than 100%. This is physically impossible. The $R_{CA}$ factor is therefore an important figure of merit. The reference sample (sample 1 in table 7.1) had a $R_{CA}$ factor of $0.45 \pm 0.05$. One way to increase the $R_{CA}$ factor was by decreasing the thickness of the spin absorption layer. This resulted in an improved $R_{CA}$ factor of $0.69 \pm 0.08$ for a Co thickness of 1 nm. It was shown that the $R_{CA}$ factor is negatively proportional to the spin absorption layer thickness and the combination of the Co/Ni spin injection layer in sample 1 with the Co spin absorption layer is not feasible for switching by laser-induced spin-transfer-torque, since the maximum $R_{CA}$ is smaller than unity. 

A next step towards $R_{CA} \geq 1$ was by changing the composition of the Co/Ni spin injection layer. Table 7.1 shows that the $R_{CA}$ factor was increased to $0.53 \pm 0.07$ (sample 3a) by increasing the Co:NI ratio. This improvement was mainly attributed to the increased total magnetic moment by increasing the Co:NI ratio. As a result, the fractional demagnetization is smaller, whereas the generated spin current is constant. This means that the ratio between generated spin current and demagnetization is smaller, indicating a larger $R_{CA}$. Increasing the Co dusting layer in the Ni/Co interface improved the generation of spin currents and decreased the thermal demagnetization. Both effects contribute positively to the $R_{CA}$ factor, which was determined to be larger than unity: $R_{CA} = 1.2 \pm 0.1$. This demonstrates that the non-collinear magnetic bilayer can be optimized towards an all-optical switch of the spin absorption layer.

By substituting the spin absorption layer for a nickel layer, the canting angle was increased by a factor of 5.6 as compared to the reference sample. The increased canting angle was attributed to the decreased saturation magnetization of nickel. The resulting $R_{CA}$ factor was $0.8 \pm 0.1$. This was an improvement with a factor of approximately 1.8 as compared to the reference sample (see samples 1 and 5 in table 7.1).

The results in this thesis show that certain combinations of spin injection and spin absorption layers can in principle be used as a two-state memory element. The excitation of the two states is all-optical by ultrafast laser-induced spin-transfer-torque on the spin absorption layer. However, a switch was defined as any state in which the spin absorption layer has a negative component along the axis along which it is initially aligned. A complete switch, meaning that the magnetization is completely aligned opposite to its initial direction, is desirable for a two-state memory element. This implies that $R_{CA} \geq 2$. Simulations with an uniaxial anisotropy showed that the anisotropy reduced the required canting angle for a complete switch. By creating uniaxial anisotropy in the spin absorption layer, the minimum condition for switching can is reduced to $1 \leq R_{CA} \leq 2$, depending on the strength of the in-plane uniaxial anisotropy. This reduces the required demagnetization of the spin injection layer, increasing the possibility of switching the spin absorption layer.

### 7.3 Recommended studies

In this section, some experiments will be proposed for a further investigation of the physics behind the laser-induced spin currents. Secondly, it will be argued that all-optical switching in non-collinear magnetic bilayers is possible by optimizing the structures investigated in this thesis. Some suggestions will be proposed towards a demonstration of optical induced ultrafast switching using laser-induced spin-transfer-torque.
7.3.1 Physics of laser-induced spin currents

The wedge-shaped cobalt layer to investigate the penetration depth of transverse spins in cobalt ranged from 0 nm to 8 nm and measurements were performed in steps of 0.4 nm. With the knowledge that all spins are absorbed within four monolayers, measurements should be performed in a smaller range with a smaller resolution. In that way the evolution of the efficiency for small thicknesses can be investigated, resulting in a better estimation of the penetration depth. A nickel wedge could provide more insights in the absorption of the transverse spin for small thicknesses as well. Since most interfaces between nickel and metallic layers have anisotropy favouring in-plane alignment, a nickel layer can be made thinner than a cobalt layer before the magnetization aligns itself along the surface normal. It is therefore expected that a laser-induced spin-transfer-torque in nickel is observed for smaller thicknesses than for the cobalt layer.

Another interesting experiment would be the extension of the fluences for power-dependent measurements to the high-fluence regime. As was mentioned before, the spin currents are predicted to saturate at high fluence. The demagnetization caused by spin transport should be constant for fluences above the saturation fluence. By measuring the demagnetization dependency on laser fluence above the spin current saturation fluence, some means of quantifying the thermal part and spin transport part of the ultrafast demagnetization of the spin injection layer should be possible.

Finally, the density of states is energy dependent. This is especially interesting regarding the phase space for scattering of minority electrons. Up to a certain energy, the phase space asymmetry of the minority electrons grows faster than the phase space of the majority electrons. This results in an increase of the asymmetry between minority and majority electron lifetimes for larger excitation energies. It would therefore be interesting to study the generation of spin current as function of excitation energy, which should reflect a change in spin polarization.

7.3.2 Optical two-state memory element

The non-collinear magnetic bilayers investigated in this thesis can in principle be used as a two-state memory element. However, a switch of the in-plane was not yet observed in the samples studied in this thesis. A next step towards switching the spin absorption layer is to increase the demagnetization of the spin injection layer. This increases the generation of spin currents. A larger demagnetization could be accomplished by pumping the structure from the substrate side. Calculation with the transfer matrix method predicts an increase of the light absorption and demagnetization of the spin injection layer by 50% as compared to pumping the structure from the capping layer side.

The next structure to be investigated should combine all the results obtained in this thesis. Such a sample would consist for example of a $4 \times \text{Co}[0.4]/\text{Ni}[0.4] + \text{Co}[0.4]$ spin injection layer and a 3 nm Ni absorption layer. Inspection of table 7.1 predicts that such a sample could result in a quality factor of $R_{CA} = 2.13$, assuming that the effects of changing the spin injection and the spin absorption layer are independent of each other. For a cobalt spin absorption layer the $R_{CA}$ was increased by 1.53 times by decreasing the thickness to 1 nm. A further increase of $R_{CA}$ can thus be expected by increasing the Ni spin absorption layer of the fictitious sample of this section.
Assuming that the Gilbert damping enhancement for small film thickness in nickel is comparable to the damping enhancement in cobalt, a $R_{CA}$ factor of 3.26 can be anticipated for such a sample. Note that this assumes equal spin absorption ratios between 3 nm Co and Ni layers versus 1 nm Co and Ni layers. Assuming for now that $R_{CA} = 3.26$, the required demagnetization of the spin injection is $2/3.26 = 61\%$ to observe a complete switch of the spin absorption layer, which is still much larger than the typical demagnetization rates observed in this thesis. However, as was shown with simulations, the creation of an uniaxial anisotropy in the surface plane of the spin absorption layer could result in the requirement $R_{CA} = 1$ instead of $R_{CA} \geq 2$ for a complete switch, implying that only a demagnetization of $1/3.26 = 31\%$ would be needed. The creation of an uniaxial anisotropy in Ni films is possible, as was reported by various groups [59, 60]. Furthermore, demagnetization rates of approximately 50\% in Co films are observed in a new laser system currently being aligned in the TR-MOKE setup. It is therefore predicted that all-optical switching by ultrafast laser-induced spin-transfer-torque will be observed for the first time by optimizing the investigated structures in this thesis.
References


93


REFERENCES


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In this appendix several solutions of the LLG equation are provided. First the steady (undamped) precession around a fixed field is discussed. After undamped precession, Gilbert damping is included.

A.1 Steady precession around a fixed field

Consider the geometry as introduced in section 2.4.2. This geometry is shown in figure A.1. The magnetization is IP and the external field is applied along the IP easy axis. The \( y \)-axis is the easy axis and the surface normal points along the \( z \)-axis. The magnetization dynamics are determined by the LLG equation without Gilbert damping and STT term [32]:

\[
\frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \vec{H}_{\text{eff}}.
\]  

(A.1)
If the magnetization is tilted towards the surface normal, it follows from solving equation A.1 for \( M_x \) or \( M_z \) that the magnetization starts a precessional motion around the applied magnetic field. For small canting angles \( M_y \approx M_{sat} \) and \( dM_y/dt \approx 0 \). A solution for the time-dependent magnetization and its time derivative is then given by:

\[
\vec{M} = \begin{bmatrix} A_x \sin(\omega t) \\ M_{sat} \\ A_z \cos(\omega t) \end{bmatrix}, \quad \frac{d\vec{M}}{dt} = \begin{bmatrix} \omega A_x \cos(\omega t) \\ 0 \\ -\omega A_z \sin(\omega t) \end{bmatrix}. \tag{A.2}
\]

The effective field is composed of the applied field, an interface anisotropy and the shape anisotropy:

\[
\vec{H}_{eff} = \begin{bmatrix} 0 \\ H_{app} \\ \frac{2K_{s,IP}}{\mu_0 M_{sat}} - 1 \end{bmatrix} A_z \cos(\omega t). \tag{A.3}
\]

From this it follows that the torque exerted by the effective field is:

\[
-\gamma_e \mu_0 \vec{M} \times \vec{H}_{eff} = \begin{bmatrix} \gamma_e \mu_0 \left[ H_{app} + M_{sat} - \frac{2K_{s,IP}}{\mu_0 M_{sat}} \right] A_z \cos(\omega t) \\ -\gamma_e \mu_0 A_z \cos(\omega t) A_x \sin(\omega t) \\ -\gamma_e \mu_0 H_{app} A_x \sin(\omega t) \end{bmatrix}. \tag{A.4}
\]

Inserting these equations in equation A.1 yields the following relations between \( M_x \) and \( M_z \):

\[
\omega A_x = \gamma_e \mu_0 \left[ H_{app} + M_{sat} - \frac{2K_{s,IP}}{\mu_0 M_{sat}} \right] A_z, \tag{A.5a}
\]

\[
\omega A_z = \gamma_e \mu_0 H_{app} A_x. \tag{A.5b}
\]

These relations can be solved to give the angular frequency of the precessional motion around the fixed applied field:

\[
\omega = \gamma_e \mu_0 \sqrt{H_{app} \left[ H_{app} + M_{sat} - \frac{2K_{s,IP}}{\mu_0 M_{sat}} \right]}. \tag{A.6}
\]

The frequency of the motion is given by \( f_{IP} = \frac{\omega}{2\pi} \). The frequency of the precessional motion is then given by:

\[
f_{IP} = \frac{\gamma_e}{2\pi} \sqrt{\mu_0 H_{app} \left[ \mu_0 H_{app} + \mu_0 M_{sat} - \frac{2K_{s,IP}}{M_{sat}} \right]} \cdot \tag{A.7}
\]

### A.2 Damped precession around a fixed field

The next step is to include damping in the precessional motion. As a result of Gilbert damping the oscillations of \( M_x \) and \( M_z \) decay exponentially with damping time \( \tau_d \). The steady motion
as discussed in section A.1 can be seen as the special case for which \( \tau_d \to \infty \). The purpose of this section is to derive a relation between the experimental observable \( \tau_d \) and the Gilbert damping factor \( \alpha \). Still assuming that the canting angle is small, \( M_y \approx M_{\text{sat}} \). Consider the general expressions for \( \vec{M} \), \( \frac{d\vec{M}}{dt} \), and \( \vec{H}_{\text{eff}} \) [32]:

\[
\vec{M} = \begin{bmatrix} M_x \\ M_y \\ M_z \end{bmatrix}, \quad \frac{d\vec{M}}{dt} = \begin{bmatrix} M_x' \\ M_y' \\ M_z' \end{bmatrix}, \quad \vec{H}_{\text{eff}} = \begin{bmatrix} 0 \\ H_{\text{app}} \\ \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}^2} - 1 \end{bmatrix} M_z.
\]  

(A.8)

In this case, the magnetization dynamics are determined by the LLG equation including Gilbert damping:

\[
\frac{d\vec{M}}{dt} = -\gamma e \mu_0 \vec{M} \times \vec{H}_{\text{eff}} + \alpha \frac{M_{\text{sat}}}{M_{\text{sat}}} \vec{M} \times \frac{d\vec{M}}{dt}.
\]  

(A.9)

Inserting the expressions for \( \vec{M} \), \( \frac{d\vec{M}}{dt} \), and \( \vec{H}_{\text{eff}} \) in equation A.9 gives:

\[
\begin{align*}
\dot{M}_x &= \gamma e \mu_0 \left[ H_{\text{app}} + M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] M_z + \alpha \dot{M}_z, \\
0 &= -\gamma e \mu_0 M_x M_z \left[ 1 - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] + \frac{\alpha}{M_{\text{sat}}} \left[ M_z \dot{M}_x - M_x \dot{M}_z \right], \\
\dot{M}_z &= -\gamma e \mu_0 H_{\text{app}} M_x - \alpha \dot{M}_x.
\end{align*}
\]  

(A.10)

To extract the damping time consider the motion along \( x \) and \( z \). To eliminate cross terms between \( M_x \) and \( M_z \), it is useful to examine the second derivatives of equations A.10a and A.10c:

\[
\begin{align*}
\ddot{M}_x &= \gamma e \mu_0 \left[ H_{\text{app}} + M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] \dot{M}_z + \alpha \ddot{M}_z, \\
\ddot{M}_z &= -\gamma e \mu_0 H_{\text{app}} M_x - \alpha \ddot{M}_x,
\end{align*}
\]  

(A.11)

from which it follows that (together with equation A.10a):

\[
\begin{align*}
\left[ 1 + \alpha^2 \right] \dddot{M}_z &= -\alpha \gamma e \mu_0 \left[ 2H_{\text{app}} + M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] \dot{M}_z \\
&\quad -\gamma e \mu_0^2 H_{\text{app}} \left[ H_{\text{app}} + M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] M_z.
\end{align*}
\]  

(A.12)

As introduced in the beginning of this section, the resulting magnetization motion will be an exponential decaying precession around the applied field. The magnetization for \( z \) is therefore given by:

\[
\begin{align*}
M_z &= A_z \cos(\omega t) e^{-\frac{t}{\tau_d}}, \\
\dot{M}_z &= -\omega A_z \sin(\omega t) e^{-\frac{t}{\tau_d}} - \frac{M_z}{\tau_d},
\end{align*}
\]  

(A.13)
\[ \dot{M}_z = -\omega^2 M_z + \frac{2\omega A_z \sin(\omega t) e^{-\frac{t}{\tau_d}}}{\tau_d} + \frac{M_z}{\tau_d}. \]  

(A.13c)

Inserting this in equation A.12 the equation to extract the Gilbert damping factor from the observed damping time is:

\[ \left[1 + \alpha^2\right] \left[ -\omega^2 \cos(\omega t) + \frac{2\omega \sin(\omega t)}{\tau_d} + \frac{\cos(\omega t)}{\tau_d} \right] = \alpha \gamma_e \mu_0 \left[ 2B_{\text{app}} + M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] \left[ \omega \sin(\omega t) + \frac{\cos(\omega t)}{\tau_d} \right] - \gamma_e \mu_0^2 H_{\text{app}} \left[ H_{\text{app}} + M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] \cos(\omega t). \]

(A.14)

The LLG equation should hold for any arbitrary time \( t \). Therefore the time can be chosen such that \( \cos(\omega t) = 0 \) and then \( \sin(\omega t) = 1 \). At that timestep all the terms in equation A.14 with a cosine are zero and the damping time \( \tau_d \) is then determined by:

\[ \frac{2 \left[1 + \alpha^2\right] \omega}{\tau_d} = \alpha \gamma_e \mu_0 \left[ 2B_{\text{app}} + M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] \omega \]

\[ \Rightarrow \tau_d = \frac{2 \left[1 + \alpha^2\right]}{\alpha \gamma_e \left[ 2B_{\text{app}} + \mu_0 M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{M_{\text{sat}}} \right]}. \]

(A.15)

In real ferromagnetic layers the damping factor is small, therefore \( \alpha^2 \rightarrow 0 \) and the damping factor is approximated by:

\[ \alpha \approx \frac{1}{\gamma_e \tau_d \left[ B_{\text{app}} + \frac{\mu_0 M_{\text{sat}}}{2} - \frac{K_{s,\text{IP}}}{M_{\text{sat}}} \right]} . \]

(A.16)
In this appendix precessional motion of the IP magnetization around its demagnetization field is investigated. The geometry is drawn in figure B.1. The magnetization is canted and creates an angle $\theta_M$ with the surface plane. The demagnetization field $\vec{H}_{\text{demag}} = -M_z \hat{z}$ is the driving force of precessional motion. The angle $\phi_M$ is the angle of the projection of $\vec{M}$ in the surface with the $y$-axis. However, in cylindrical coordinates the angle of the magnetization projection has to be defined by the projection with the $x$-axis: $\phi_x$.

Figure B.1: Geometry for precessional motion of the IP magnetization around its own demagnetization field. The magnetization is canted due to a laser-induced STT, exciting a demagnetization field of $\vec{H}_{\text{demag}} = -M_z \hat{z}$.

B.1 Critical canting angle as function of Gilbert damping factor

In this section the detailed derivation of the canting angle as function of Gilbert damping factor is shown. To investigate the magnetization dynamics in this case, the LLG equation with field
torque and Gilbert damping is used:

\[
\frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \vec{H}_{eff} + \frac{\alpha}{M_{sat}} \vec{M} \times \frac{d\vec{M}}{dt}.
\] (B.1)

In cylindrical coordinates the magnetization is determined by \(M_r, \phi_x\), and \(M_z\):

\[
\vec{M} = \begin{bmatrix} M_r \cos (\phi_x) \\ M_r \sin (\phi_x) \\ M_z \end{bmatrix}; \quad \frac{d\vec{M}}{dt} = \begin{bmatrix} M_r \cos (\phi_x) - M_r \sin (\phi_x) \dot{\phi}_x \\ M_r \sin (\phi_x) + M_r \cos (\phi_x) \dot{\phi}_x \\ M_z \end{bmatrix}. \tag{B.2}
\]

For small canting angles \(M_r \approx M_{sat}\) and \(dM_r/dt \approx 0\). Furthermore, the magnetization starts precessional motion above the \(y\)-axis. Therefore the angle \(\phi_M\) is required instead of the angle \(\phi_x\). The relation between the two is:

\[
\phi_x = \frac{\pi}{2} - \phi_M \\
\Rightarrow \\
\cos \left( \frac{\pi}{2} - \phi_M \right) = \sin (\phi_M) \\
\sin \left( \frac{\pi}{2} - \phi_M \right) = \cos (\phi_M). \tag{B.3}
\]

The magnetization and its time derivative reduce therefore to:

\[
\vec{\tilde{M}} = \begin{bmatrix} M_{sat} \sin (\phi_M) \\ M_{sat} \cos (\phi_M) \\ M_z \end{bmatrix}; \quad \frac{d\vec{\tilde{M}}}{dt} = \begin{bmatrix} M_{sat} \cos (\phi_M) \dot{\phi}_M \\ -M_{sat} \sin (\phi_M) \dot{\phi}_M \\ M_z \end{bmatrix}. \tag{B.4}
\]

The effective field is given by:

\[
\vec{H}_{eff} = \begin{bmatrix} 0 \\ 0 \\ \left[ \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} - 1 \right] M_z \end{bmatrix}. \tag{B.5}
\]

The torque applied by the effective field is then given by:

\[
-\gamma_e \mu_0 \vec{M} \times \vec{H}_{eff} = -\gamma_e \mu_0 \begin{bmatrix} \dot{x} \\ \dot{y} \\ \dot{z} \end{bmatrix} M_{sat} \sin (\phi_M) \ M_{sat} \cos (\phi_M) \ M_z \\
0 \ 0 \ \left[ \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} - 1 \right] M_z \\
\gamma_e \mu_0 M_{sat} M_z \cos (\phi_M) \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right] \\
-\gamma_e \mu_0 M_{sat} M_z \sin (\phi_M) \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right] \\
0 \end{bmatrix}. \tag{B.6}
\]
The Gilbert damping term is given by:

\[
\frac{\alpha}{M_{\text{sat}}} \vec{M} \times \frac{d\vec{M}}{dt} = \frac{\alpha}{M_{\text{sat}}}
\begin{vmatrix}
\hat{x} & M_{\text{sat}} \sin (\phi_M) & M_{\text{sat}} \cos (\phi_M) \\
\hat{y} & M_{\text{sat}} \cos (\phi_M) & -M_{\text{sat}} \sin (\phi_M) \\
\hat{z} & M_{\text{sat}} \cos (\phi_M) M_z + M_{\text{sat}} \sin (\phi_M) M_z \dot{\phi}_M \\
& -M_{\text{sat}} \sin (\phi_M) M_z + M_{\text{sat}} \cos (\phi_M) M_z \dot{\phi}_M \\
& -M_{\text{sat}}^2 \dot{\phi}_M
\end{vmatrix}
\]

Inserting these torques in equation B.1 the following relations are derived:

\[
\begin{align*}
M_{\text{sat}} \cos (\phi_M) \dot{\phi}_M &= \gamma_e \mu_0 M_{\text{sat}} \cos (\phi_M) M_z \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{\text{sat}}^2} \right] + \alpha \left[ \cos (\phi_M) \dot{M}_z + \sin (\phi_M) M_z \dot{\phi}_M \right], \quad (B.8a) \\
- M_{\text{sat}} \sin (\phi_M) \dot{\phi}_M &= -\gamma_e \mu_0 M_{\text{sat}} \sin (\phi_M) M_z \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{\text{sat}}^2} \right] - \alpha \left[ \sin (\phi_M) \dot{M}_z - \cos (\phi_M) M_z \dot{\phi}_M \right], \quad (B.8b) \\
\dot{M}_z &= -\alpha M_{\text{sat}} \dot{\phi}_M. \quad (B.8c)
\end{align*}
\]

The equations B.8a and B.8b can be simplified by multiplying equation B.8a with \(\cos (\phi_M) / M_{\text{sat}}\) and multiplying equation B.8b with \(- \sin (\phi_M) / M_{\text{sat}}\). The next step is to sum the two equations. The result is an equation for the time derivative of \(\dot{\phi}_M\):

\[
\begin{align*}
\cos (\phi_M)^2 \dot{\phi}_M &= \gamma_e \mu_0 \cos (\phi_M)^2 M_z \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{\text{sat}}^2} \right] + \alpha \left[ \cos (\phi_M)^2 \dot{M}_z - \sin (\phi_M) \cos (\phi_M) M_z \dot{\phi}_M \right], \quad (B.9a) \\
\sin (\phi_M)^2 \dot{\phi}_M &= \gamma_e \mu_0 \sin (\phi_M)^2 M_z \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{\text{sat}}^2} \right] + \alpha \left[ \sin (\phi_M)^2 \dot{M}_z + \sin (\phi_M) \cos (\phi_M) M_z \dot{\phi}_M \right]. \quad (B.9b)
\end{align*}
\]

The time derivative of \(\dot{\phi}_M\) follows by summing equations B.9a and B.9b:

\[
\dot{\phi}_M = \gamma_e \mu_0 M_z \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{\text{sat}}^2} \right] + \frac{\alpha}{M_{\text{sat}}} \dot{M}_z, \quad (B.10)
\]

where in the summation it was used that \(\cos (\phi_M)^2 + \sin (\phi_M)^2 = 1\). Substituting equation B.10 in equation B.8c it follows that \(M_z\) decays exponentially:

\[
\dot{M}_z = -\alpha \gamma_e \mu_0 M_{\text{sat}} M_z \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{\text{sat}}^2} \right] - \alpha^2 \dot{M}_z \quad (B.11a)
\]

\[
\Rightarrow M_z = M_{z,0} e^{-\frac{\dot{m}}{\alpha}}, \quad (B.11b)
\]
\[ \dot{M}_z = -\frac{M_{z,0}}{\tau_d} e^{-\frac{t}{\tau_d}}, \]  
(B.11c)

with \( \tau_d \) and \( M_{z,0} \) determined by:

\[ \tau_d = \frac{1 + \alpha^2}{\alpha \gamma_e \mu_0 M_{sat} \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right]} \]  
(B.12a)

\[ M_{z,0} = M_{sat} \sin (\theta_M), \]  
(B.12b)

where the last equation is determined by assuming that the STT is instantaneous. Inserting \( M_z \) and \( \dot{M}_z \) in equation B.10 yields:

\[ \dot{\phi}_M = \left[ \gamma_e \mu_0 \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right] - \frac{\alpha}{M_{sat} \tau_d} \right] M_{sat} \sin (\theta_M) e^{-\frac{t}{\tau_d}} \]  
(B.13)

To find the final angle \( \phi_{M,final} \) at which the magnetization ends precessional motion equation B.13 is integrated from \( t_0 = 0 \) to \( t_\infty = \infty \):

\[ \phi_{M,\infty} = \frac{\gamma_e \mu_0 \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right]}{1 + \alpha^2} M_{sat} \sin (\theta_M) \int_0^\infty e^{-\frac{t}{\tau_d}} dt \]  
(B.14)

\[ = -\frac{\gamma_e \mu_0 \left[ 1 - \frac{2K_{s,IP}}{\mu_0 M_{sat}^2} \right] \tau_d}{1 + \alpha^2} M_{sat} \sin (\theta_M) \left[ e^{-\frac{1}{\tau_d}} + C_i \right]_0^\infty, \]

where \( C_i \) is an integration constant. To find the final angle \( \phi_{M,final} \), it is assumed that at \( t = 0, \phi_{M,0} = 0 \) and at \( t = \infty, \phi_{M,\infty} = \phi_{M,final} \) and equation B.12a is inserted for \( \tau_d \):

\[ \phi_{M,final} = -\frac{\sin (\theta_M) \left[ e^{-\frac{\infty}{\tau_d}} - e^{-\frac{1}{\tau_d}} \right]}{\alpha} \]  
(B.15)

\[ = -\frac{\sin (\theta_M)}{\alpha} \left[ 0 - 1 \right] \]

\[ = \frac{\sin (\theta_M)}{\alpha}. \]

For small canting angles \( \sin (\theta_M) \approx \theta_M \). Substituting this in equation B.15, the final result for the canting angle as function of Gilbert damping factor is:

\[ \phi_{M,final} = \frac{\theta_M}{\alpha} \Rightarrow \theta_M = \alpha \phi_{M,final}. \]  
(B.16)
B.2 Time scale of switching

In this section the time scale to switch the IP layer is investigated. The geometry for this section is the same as in the previous section (see figure B.1). To calculate the frequency of the precessional motion of $\vec{M}$ around its demagnetization field, consider again the cartesian coordinates of $\vec{M}$. $\vec{M}$ is now given by (assuming that the canting angle is small):

$$
\vec{M} = \begin{bmatrix}
M_{\text{sat}} \sin (\omega t) \\
M_{\text{sat}} \cos (\omega t) \\
M_{\text{sat}} \sin (\theta_M) e^{-\frac{t}{\tau_d}}
\end{bmatrix}.
$$

The effective field is composed of the demagnetization field and an anisotropy field from interface anisotropy:

$$
\vec{H}_{\text{eff}} = \begin{bmatrix}
0 \\
0 \\
\frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}^2} \mu_0 M_{\text{sat}} - 1 \end{bmatrix} M_z.
$$

The torque applied by the effective field is:

$$
-\gamma_e \mu_0 \vec{M} \times \vec{H}_{\text{eff}} = \begin{bmatrix}
\gamma_e \mu_0 M_{\text{sat}}^2 \sin (\theta_M) \cos (\omega t) \left[1 - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}^2} \right] e^{-\frac{t}{\tau_d}} \\
-\gamma_e \mu_0 M_{\text{sat}}^2 \sin (\theta_M) \sin (\omega t) \left[1 - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}^2} \right] e^{-\frac{t}{\tau_d}} \\
0
\end{bmatrix}.
$$

For $dM_x/dt$ the following equation arises:

$$
\omega M_{\text{sat}} \cos (\omega t) = \gamma_e \mu_0 \sin (\theta_M) \cos (\omega t) \left[M_{\text{sat}}^2 - \frac{2K_{s,\text{IP}}}{\mu_0} \right] e^{-\frac{t}{\tau_d}} \Longrightarrow \omega = \gamma_e \mu_0 \left[M_{\text{sat}} - \frac{2K_{s,\text{IP}}}{\mu_0 M_{\text{sat}}} \right] \theta_M,
$$

where it is assumed that there is no Gilbert damping ($\tau_d \rightarrow \infty$) and $\sin (\theta_M) \approx \theta_M$. To find the time scale needed to reach the final state where $\phi_M = \phi_{M,\text{final}}$, consider the value of $\phi_M$ at timestep $t$:

$$
\phi_M = \omega t,
$$

$$
\Longrightarrow t = \frac{\phi_{M,\text{final}} M_{\text{sat}}}{\gamma_e \mu_0 \theta_M \left[M_{\text{sat}}^2 - \frac{2K_{s,\text{IP}}}{\mu_0} \right]} = \frac{M_{\text{sat}}}{\gamma_e \mu_0 \alpha \left[M_{\text{sat}}^2 - \frac{2K_{s,\text{IP}}}{\mu_0} \right]}.
$$

where equation B.16 was used. The frequency is time-dependent due to the time-dependent demagnetization field. This time-dependence was not considered here. However, the purpose of the derivation is to show that the time scale of switching depends on $M_{\text{sat}}$, as argued in section 2.5.
To model the LLG equation using Matlab, the time derivative on the right-hand side of the LLG equation (equation 2.19) has to be eliminated. In this appendix it is shown how the equation is rewritten. The starting point is the original equation:

$$\frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \vec{H}_{\text{eff}} + \frac{\alpha}{M_{\text{sat}}} \vec{M} \times \frac{d\vec{M}}{dt} + \gamma_e \mu_0 \frac{Q_s}{M_{\text{sat}}^2 FM \mu_0} \vec{M} \times [\vec{\sigma} \times \vec{M}] .$$  \hspace{1cm} (C.1)

The first contraction is to include the term coming from spin-transfer-torque in the effective field:

$$\vec{H}_{\text{new}} = \vec{H}_{\text{eff}} - \frac{Q_s}{M_{\text{sat}}^2 FM \mu_0} [\vec{\sigma} \times \vec{M}] .$$  \hspace{1cm} (C.2)

The original LLG equation then reduces to:

$$\frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \vec{H}_{\text{new}} + \frac{\alpha}{M_{\text{sat}}} \vec{M} \times \frac{d\vec{M}}{dt} .$$  \hspace{1cm} (C.3)

To eliminate the time derivative at the right-hand side of equation C.3, the cross product of $\vec{M}$ and $d\vec{M}/dt$ is calculated from equation C.3:

$$\vec{M} \times \frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \left[ \vec{M} \times \vec{H}_{\text{new}} \right] + \frac{\alpha}{M_{\text{sat}}} \vec{M} \times \left[ \vec{M} \times \frac{d\vec{M}}{dt} \right] .$$  \hspace{1cm} (C.4)

The second term on the right-hand side of equation C.4 can be rewritten as:

$$\frac{\alpha}{M_{\text{sat}}} \vec{M} \times \left[ \vec{M} \times \frac{d\vec{M}}{dt} \right] = \frac{\alpha}{M_{\text{sat}}} \vec{M} \left[ \vec{M} \cdot \frac{d\vec{M}}{dt} \right] - \frac{\alpha}{M_{\text{sat}}} \frac{d\vec{M}}{dt} [\vec{M} \cdot \vec{M}]$$

$$= -\frac{\alpha}{M_{\text{sat}}} \frac{d\vec{M}}{dt} [\vec{M} \cdot \vec{M}] = -\alpha M_{\text{sat}} \frac{d\vec{M}}{dt} ,$$  \hspace{1cm} (C.5)
where it was used that \( \vec{M} \) and its derivative are orthogonal and their dot product is therefore zero. Inserting equation C.5 in equation C.4 results in the cross product of \( \vec{M} \) and its derivative:

\[
\vec{M} \times \frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \left[ \vec{M} \times \vec{H}_{\text{new}} \right] - \alpha M_{\text{sat}} \frac{d\vec{M}}{dt}. \tag{C.6}
\]

Inserting this cross product as the Gilbert damping term in equation C.3 results in:

\[
\frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \vec{H}_{\text{new}} - \frac{\alpha \gamma_e \mu_0}{M_{\text{sat}}} \vec{M} \times \left[ \vec{M} \times \vec{H}_{\text{new}} \right] - \alpha^2 \frac{d\vec{M}}{dt}
\]

\[
\Rightarrow [1 + \alpha^2] \frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \left[ \vec{H}_{\text{new}} + \frac{\alpha}{M_{\text{sat}}} \vec{M} \times \vec{H}_{\text{new}} \right]
\]

\[
\Rightarrow \frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \vec{H}_{\text{new}} + \frac{\alpha}{M_{\text{sat}}} \vec{M} \times \vec{H}_{\text{new}}. \tag{C.7}
\]

Remembering that \( \vec{H}_{\text{new}} = \vec{H}_{\text{eff}} - \frac{Q_s}{M_{\text{sat}}^2 \gamma_F \mu_0} \left[ \vec{\sigma} \times \vec{M} \right] \), the end result is:

\[
\frac{d\vec{M}}{dt} = -\gamma_e \mu_0 \vec{M} \times \left[ \vec{H}_{\text{eff}} + \frac{\alpha}{M_{\text{sat}}} \vec{M} \times \vec{H}_{\text{eff}} - \frac{Q_s}{M_{\text{sat}}^2 \gamma_F \mu_0} \left[ \vec{\sigma} \times \vec{M} \right] - \frac{\alpha Q_s}{M_{\text{sat}}^2 \gamma_F \mu_0} \vec{M} \times \left[ \vec{\sigma} \times \vec{M} \right] \right]. \tag{C.8}
\]
In reference [22] measurements on a similar structure as the reference sample introduced in section 6.1 showed a double-frequency oscillation. The first frequency was shown to originate from a laser-induced STT on the IP layer, like the oscillations discussed in section 4.2. The second frequency observed in reference [22] was attributed to a laser-induced oscillation in the OOP layer. This oscillation was caused by an ultrafast change in the anisotropy of the OOP layer, caused by laser heating. For a detailed description of this optical anisotropy pulse induced oscillation can be found in reference [40]. However, the structure investigated in reference [22] contained no Ta seeding layer. In this appendix it will be shown that the addition of a Ta seeding layer increases the perpendicular magnetic anisotropy, thereby reducing the effect of an ultrafast anisotropy pulse.

Upon laser heating of the OOP layer, the effective anisotropy inducing PMA decreases. In an IP field this results in a new, low-anisotropy, equilibrium state of the OOP layer magnetization, canted more towards the surface as compared to the static, high-anisotropy, equilibrium state. At a longer time scale the anisotropy is recovered, but the magnetization is still in its new low-anisotropy equilibrium position. As a result, a precessional motion back to its old high-anisotropy state is excited. In the samples investigated throughout this thesis, a Ta seeding layer is included between the substrate and the PMA-inducing Pt buffer layer. This seeding layer was absent in the structures studied in reference [22]. Increasing the PMA decreases the effect of an anisotropy pulse. The addition of a Ta seeding layer increases PMA [61–64] and could therefore result in a non-observable amplitude of the anisotropy pulse induced oscillation in the OOP layer. The effect of the Ta seeding layer on the PMA in the investigated samples in this thesis is studied using two different samples. The first sample contains the same layers as the reference sample introduced in section 6.1, but the IP layer layer was excluded. In the second sample both the IP layer and the Ta seeding layer were excluded.

The effective anisotropy constant $K_{ani}$ is measured using the AHE. By measuring the perpendicular magnetization component using the AHE, the static equilibrium angle of the magnetization with the surface normal can be measured as function of an external field. As was explained in section 3.4, minimization of the total energy density (equation 2.9) results in the expected
equilibrium angle for given $K_{ani}$ as function of the external field. The field strength and the field angle with the surface normal are both varied. By comparing the measured equilibrium angles for these different field configurations with the equilibrium angle calculated by equation 2.9, the value for $K_{ani}$ with the least square error can be estimated. In figure D.1a measurements of $M_z$ for fields from 0.15 T up to 0.9 T are shown for the sample with a Ta seeding layer. The angle of the field with the surface normal was varied from 0 degrees to 60 degrees.

This routine to calculate the least square as explained above resulted in an effective anisotropy (including shape anisotropy) of $K_{ani} = 2.71 \times 10^5 \text{ J/m}^3$. The fits of the measured magnetization curves as function of field using this value of $K_{ani}$ are shown with red curves in figure D.1a. In figure D.1b similar measurements are shown for the sample without the Ta seeding layer, with the same scale along the vertical axis. The effective anisotropy of the sample without the Ta seeding layer with the least square was $K_{ani} = 1.06 \times 10^5 \text{ J/m}^3$. The smaller anisotropy of this sample results in a larger canting of the magnetization towards the field direction. This increases the equilibrium angle of the magnetization with the surface normal and subsequently decreases the perpendicular component. The latter is measured and depicted in figures D.1a and b. Comparing the two measurements, it is clearly seen that the magnetization of the sample without the Ta seeding layer cants much more towards the magnetic field direction than the sample with Ta seeding layer.