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Optical magnetic switching of ferromagnetic Co/Pt thin films

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Award date:
2015

Link to publication
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August 27, 2015

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Abstract

The field of ultrafast magnetization dynamics is an exciting new area of nanomagnetism and a lot of fundamental research has been carried out since the first discovery of sub-ps magnetization dynamics induced by intense ultrashort laser pulses. Of special (technological) interest is the phenomenon of All-Optical Switching (AOS), in which only a single fs laser pulse is sufficient to switch the magnetization of a material. This phenomenon has a very high potential as a fast and efficient data writing technique for future memory devices. Nevertheless, the mechanisms behind it are poorly understood. This is demonstrated by the fact that AOS was thought to be exclusive to ferrimagnets, but it was recently also demonstrated in ferromagnets. Therefore in the present work the optical switching properties in ferromagnetic Co/Pt ultrathin films are studied, with both modelling and experiments.

The microscopic three-temperature model is used to describe the optical magnetic switching with ultrashort laser pulses. The switching is assumed to be driven by a magnetic field pulse that is induced by the inverse Faraday effect. The results of the simulations with this model show that AOS can be achieved for realistic experimental parameters. Recent experimental results of AOS in ferromagnets were successfully reproduced for the first time.

Experimentally, the demagnetization and reduction in coercivity caused by ultrashort laser pulses are demonstrated. Switching experiments under an applied magnetic field are performed, and the results correspond well with predictions from the simulations. However, the assistance of an applied field is required for optical switching in experiments performed for multiple samples and experimental parameters. All optical switching is thus not observed. This is likely caused by slight differences between the used experimental parameters and those reported in literature, and it shows that AOS is not a trivial and easily achievable process.

Cover image: An optical writing experiment, in which domains are written with a magnetization opposite to the rest of the film using ultrashort laser pulses. In this case the name of the TU/e was written on a micrometer scale.
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Chapter 1

Introduction

1.1 Magnetic data storage

Data is around us everywhere: in the computer on our desktop, the smart phones in our pockets and even flying through the air in the form of wireless communication signals. This was unthinkable in the 1950’s when the first computers were invented. Since then tremendous technological progress has been made to make computers smaller, faster and more energy efficient.

One of the main technologies that made this possible is the development of magnetic data storage. In 1956 IBM first introduced magnetic data storage in the form of a Hard Disk Drive (HDD). This IBM 350 could store 3.75 MB of data on fifty large disks with a total volume of 2 m$^3$. A huge improvement came with the discovery of the Giant Magneto Resistance (GMR) effect by Fert and Grünberg in 1988 [1], which made it possible to accurately read out magnetic data on a much smaller scale. The data storage density (and speed and efficiency) has kept increasing exponentially since, leading to the current record of 500 Gbit/in$^2$, almost a billion times denser than the IBM 350.

However, the current HDD technology is reaching its physical limits. Since 2010 the increase in storage density has slowed down from the historical 40% per year to only 10%, while the transistor density on chips still keeps growing according to Moore’s law. Since the amount of digital data in our society is ever increasing, there is still a need for better storage and processing technology. This is why academia as well as industry are now researching novel and more efficient data processing and storage strategies.

Traditionally, data is stored magnetically, processed and computed electronically, and transported optically. The conversion to and from electronic data is one of the main bottlenecks in this scheme. Ideally, this step would be eliminated in future high-speed devices, integrating photonics and magnetism into a hybrid device. One way to achieve this is by crossing a so-called magnetic racetrack memory with a photonic waveguide, at
1.2 A magneto-photonic memory device

the intersection of which a conversion between optical and magnetic bits takes place.

A description of such a hybrid magneto-photonic device will be given in section 1.2. The transfer of data between optics and magnetism can occur by using ultrashort laser pulse to optically write data into the magnetic memory. To explain this optical switching, we shall first have a look at ultrafast magnetization dynamics in section 1.3. Then the all-optical writing technique, which is the main focus of this thesis, will be discussed in section 1.4. The goal of this thesis, and an overview of its contents will be given in section 1.5.

1.2 A magneto-photonic memory device

Our proposed magneto-photonic device consists of three main components. Data enters the device in the form of optical bits from the waveguide, such as an optical fibre. From this waveguide the optical bits will be converted into magnetic bits. We discuss this writing of data at the end of this section. The magnetic bits are stored and moved in a magnetic racetrack memory which will be discussed now.

This racetrack memory, as introduced by Parkin et al. [2] at IBM, is shown schematically in Figure 1.1a. Data is stored in a magnetic nanowire in a similar way as in a conventional HDD: the ferromagnetic material consists of domains with opposite magnetization direction. One magnetization direction represents a ‘0’, the other a ‘1’. In a HDD, the storage medium is a very thin disk which spins around underneath a read and write head to be able to access different parts of the disk. In the racetrack memory the data is moved through the nanowire instead of moving the nanowire itself, by sending a spin-polarized current through the wire (based on an effect called the spin transfer torque, see [3]). This eliminates the need for mechanically moving parts, thereby greatly increasing the speed and reliability of data access. Another advantage is that the nanowire can be folded into the third dimension, increasing the storage density.

Currently, data is being written on a magnetic (racetrack) memory using a locally applied magnetic field. This requires the conversion of the optical data to an electric current that generates the desired magnetic field. Furthermore, the switching time is typically in the order of a nanosecond, and is fundamentally limited to a couple of hundreds of picoseconds. This is too slow to keep up with the desired optical data transfer rate of $>$20 GHz.

A new scheme to write data would be to convert the optical bits into ultrashort laser pulses that can reverse the magnetization on a sub-picosecond time scale, which addresses both of these issues. An additional advantage is that this switching process is thought to be more energy efficient than the current writing technique. Nanometer scale domains can be written by focusing the laser beam using plasmonic antennas (50 nm domains have been reported [4]).

A complete sketch of our magneto-photonic memory is schematically shown in Figure 1.1b.
1.3 Ultrafast magnetization dynamics

In 1996, Beaurepaire et al. [5] performed the first experiments with ultrashort laser pulses on ferromagnetic materials with a sub-ps time resolution. They used an intense 60 fs laser pulse to excite a Ni thin film, and measured the magnetization as a function of time using delayed probe pulses. Surprisingly, they found that the Ni film demagnetizes within one ps (see Figure 1.2), much faster than previously assumed.

That a ferromagnet demagnetizes due to heating by a high power laser pulse is to be expected, and the effect was phenomenologically modelled using a three-temperature model (3TM). This model describes the energy flow between three temperature baths, being the electron, phonon and spin system. However, the mechanisms thought to be involved, such as spin-lattice relaxation, have much longer time scales than the observed sub-ps demagnetization.
1.4 An all-optical approach to magnetic switching

Beaurepaire’s experiment triggered a entire new field of research within spintronics: the ultrafast manipulation of magnetization, also called femtomagnetism. The main goal in this field has been to uncover the microscopic mechanisms behind these ultrafast dynamics. Many experiments were done on a variety of magnetic materials, and several theoretical models were proposed. And even though progress has been made, the underlying mechanism is still a matter of debate (see section 2.2).

Furthermore, a lot of research has focussed on exploiting these ultrafast dynamics for technological applications. One of the most technologically interesting questions is the following: would it be possible to reverse the magnetization on a much faster time scale than traditional field induced switching?

1.4 An all-optical approach to magnetic switching

The above question was answered when Stanciu et al. [6] showed in 2007 that it was possible to deterministically switch the magnetization using only an ultrashort pulse, the so-called All-Optical Switching (AOS). The direction of the switch of the GdFeCo ferrimagnetic alloy was purely determined by the helicity of the light, as shown in Figure 1.3, giving rise to the term All-Optical Helicity Dependent Switching (AOHDS). The AOHDS was explained as an interplay of two effects: heating by the absorbed laser pulse, which causes the quenching of the magnetization, and a symmetry breaking magnetic field. This magnetic field was
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thought to be caused by the Inverse Faraday Effect (IFE), which states that an optical pulse in a magnetized material induces a magnetic field, the direction of which is dependent on the helicity of the light.

Figure 1.3: The first time AOS was observed, reproduced from [6]. Starting out with a sample with multiple magnetic domains, a linear ($L$), left-handed ($\sigma^-$) or right-handed ($\sigma^+$) circularly polarized beam was swept across the sample. AOS occurs depending on the polarization of the light.

Further research showed that this AOS occurs on the time scale of several tens of ps, so indeed much faster than field induced switching. More experiments quickly followed and the effect was also found in other Rare-Earth Transition Metal (RE-TM) ferrimagnetic compounds. In 2014, Mangin et al. [7] showed that AOS was possible in a much broader range of ferrimagnets. They even showed AOS in synthetic ferrimagnets, which are layers of thin films that are antiferromagnetically coupled to mimic a ferrimagnet.

The initial explanation of an interplay of heating and a helicity dependent effective field was abandoned when it was discovered that AOS was also possible with just linearly polarized light. A new theory was developed in which heating alone was sufficient for switching [8]. This was possible due to the different demagnetization rates and Curie temperatures of the sub-lattices of the ferrimagnet and their exchange interaction. The helicity dependence was ascribed to different absorption of the laser pulse for different helicities [9].

But then Lambert et al. [10] recently discovered AOHDS in ferromagnetic materials such as simple Co/Pt layers. These materials consist of alternating thin Co and Pt layers, and they exhibit perpendicular magnetic anisotropy (PMA), meaning that the magnetization is directed out of the plane. These materials are even more technologically interesting than the above mentioned ferrimagnets, since they are already widely used as magnetic recording media. But the currently accepted theory for AOS does not hold in this case since there are no antiferromagnetically coupled sub-lattices.

The experiments of Lambert et al. have not been explained up to now. The authors argued that maybe the IFE, though previously ruled out, is still playing a role in these
Another symmetry breaking contribution might come from dipolar fields from the film surrounding the switched area. Nevertheless, these results give a new impulse to the already exciting field of AOS, as further research is required to understand the mechanism behind this phenomenon.

1.5 This Thesis

From the above discussion it should be clear that AOS is still not properly understood, especially in ferromagnetic materials. But to be able to exploit this exciting phenomenon in memory devices, it is crucial to understand the mechanisms behind it. In this thesis we shall therefore investigate the optical switching properties of Co/Pt layers.

Chapter 2 provides the background and theory that is necessary to understand our simulations and experiments. We shall first look at the mechanisms behind the ultrafast demagnetization induced by fs laser pulses. The Microscopic 3 Temperature Model (M3TM) is then derived to describe these dynamics, and it is adapted to model the optical switching experiments carried out in this thesis. Finally the different hypotheses that try to explain the AOS phenomenon will be discussed.

The methodologies used in this project are described in Chapter 3. The growth procedure and structure of the researched Co/Pt samples are explained first. These samples are investigated using the Magneto Optical Kerr Effect which will be briefly introduced. The Time Resolved MOKE (TR-MOKE) measurement setup is specifically designed to study the ultrafast magnetization dynamics of the samples, and it is discussed more extensively. The chapter is concluded with the experimental parameters and procedure.

In Chapter 4 the results of the simulations using the M3TM will be presented. The key parameters in the model will be discussed and a standard ultrafast demagnetization experiment is simulated. We continue by simulating optical switching, first with an externally applied field, and then with a field pulse caused by the IFE. Finally, to understand the mechanisms involved in AOHDS in (ferro)magnetic materials, the results of the experiments of Lambert et al. [10] on Co/Pt layers will be reproduced.

Chapter 5 presents the experimental results of this project. The samples are first characterized using static MOKE. Then the ultrafast demagnetization traces obtained by TR-MOKE will be discussed. Next to this demagnetization, the laser pulse also causes a reduction in coercivity, which allows us to perform field assisted switching experiments. Finally, helicity dependent (all)-optical switching is discussed.

This thesis will be concluded in Chapter 6, which also includes an outlook onto future research and applications. A final note to the reader: there are several acronyms that will be used frequently throughout this thesis. Each of them will be explained at their first occurrence, but an overview can be found in Appendix A for reference.
Chapter 2

Background and Theory

In this chapter, we will discuss the relevant concepts and theories that are needed to understand the experimental and simulation results on optical switching. We start off with a brief introduction to the basic concepts of thin film magnetism, for the readers who are not familiar with the subject. Then we shall describe the current theoretical understanding of ultrafast magnetization dynamics. Subsequently the M3TM will be derived, which we use to describe these ultrafast dynamics. Finally, we present the different theories that are used to explain AOS, and the M3TM will be adapted to describe AOS.

2.1 Nanomagnetism

2.1.1 Magnetic anisotropy

Ferromagnetic materials like Co have a spontaneous magnetization below the Curie temperature \( T_C \). This means that the spins in the material are aligned in the same direction, without the presence of an external magnetic field. The mechanism behind this ordering is the direct exchange interaction, which is a result of the Pauli exclusion principle. The magnetization \( \vec{M} \) that is formed this way has no intrinsic direction.

This direction is set by the magnetic anisotropy. This uniaxial anisotropy defines an easy axis along which the magnetization aligns. The energy (density) needed to pull the magnetization away from this axis by an angle \( \theta \) is in first order given by

\[
\epsilon_{\text{ani}} = K \sin^2(\theta),
\]  

(2.1)

where \( K [\text{J/m}^3] \) is the anisotropy constant. Because atoms near the interface of a magnetic film have a different environment than the atoms in bulk, the anisotropy constant is different as well. We therefore divide the anisotropy constant in a volume \( K_V [\text{J/m}^3] \) and
surface $K_S [J/m^2]$ contribution:

$$K = K_V + \frac{2K_S}{t},$$

with $t$ the thickness of the magnetic film. The factor two is a result from contributions of both the top and bottom surface.

The volume anisotropy is mainly caused by the so-called shape anisotropy, which is a manifestation of dipolar interactions between spins. In our case of thin films, it favours an in plane orientation of the magnetization. Conventionally this corresponds to a negative value of $K_V$, i.e. $\theta$ is defined as the angle between magnetization and the surface normal.

The surface anisotropy is due to magneto-crystalline anisotropy, in which spin-orbit coupling (SOC) plays an important role. Depending on the materials at the interface, $K_S$ can be positive or negative. In our case of Co/Pt, we have a positive surface anisotropy, which is rather large due the large SOC in Pt.

From Equation 2.2 it should be clear that the contribution of the surface anisotropy becomes larger when the magnetic film becomes thinner. Consequently our Pt/Co/Pt samples, with a large $K_S$, will have a positive $K$ as long as the film is thin enough. Their easy axis is thus out of plane, and they are said to have a perpendicular magnetic anisotropy (PMA).

### 2.1.2 Hysteresis

The above description of magnetic anisotropy shows us that a thin Co/Pt film has an easy axis along the surface normal, giving two energetic minima: magnetization $\vec{M}$ up and down. The magnetic anisotropy provides an energy barrier between these two states, preventing spontaneous switching. A magnetic field $\vec{H}$ can be applied to overcome this barrier. The energy associated with this process is called the Zeeman energy $\epsilon_Z = -\mu_0 \vec{M} \cdot \vec{H}$, which favours a parallel orientation of the magnetization and the applied field. If this field is applied along the out of plane easy axis, we then get for the magnetic energy density (including the anisotropy energy):

$$\epsilon_{\text{mag}} = -\mu_0 MH \cos(\theta) + K \sin(\theta),$$

with $\theta$ again the angle between the easy axis (but now also the applied field) and the magnetization. This simplified description is called the Stoner-Wohlfarth model [11]. Minimisation of Equation 2.3 gives the magnetization direction as a function of the applied field.

The result of this is a hysteresis loop as shown in Figure 2.1. For very high applied fields we find the two previously mentioned up and down states. The value of the magnetization is then called the saturation magnetization $M_S$. At zero applied field, the state of the
magnetic material is dependent on its history. A magnetic switch occurs when the field exceeds the coercive field or coercivity $H_C$. In this model, we find that

$$H_C = \frac{2K}{\mu_0 M_S} \quad (2.4)$$

Figure 2.1: A typical easy axis hysteresis loop, as calculated with the Stoner-Wohlfarth model. The arrows indicate the direction of the switching when sweeping the magnetic field.

2.1.3 Magnetic Domains

The above model assumes a zero temperature and an ideal homogeneous sample that behaves as one single macrospin. At finite temperatures, thermal fluctuations can overcome the anisotropy energy barrier before the applied field reaches $H_C$, effectively lowering the coercivity with increasing temperature. Magnetic switching is thus a thermally activated process [12].

Inhomogeneity of the sample (such as thickness variation, edges, contaminations, etc.) can cause local deviations in the anisotropy. Areas where the anisotropy is low will switch first when sweeping the magnetic field, a process called nucleation. This way magnetic domains with an opposite orientation of the magnetization are formed. In between magnetic domains there is a narrow region where the magnetization rotates from up to down, which is
2.2 Ultrafast magnetization dynamics

called the domain wall. These domain walls can propagate through the sample for example by applying a magnetic field, eventually switching the entire sample (see Figure 2.2). Rich physics is involved with these domain walls and their propagation, and a lot of research is ongoing in relation to the racetrack memory mentioned in section 1.2.

Figure 2.2: A sketch of magnetization reversal in a magnetic thin film. A sample is saturated by a positive magnetic field, and then an increasingly large negative field is applied. Domains nucleate and expand through domain wall propagation.

The classic magnetic switching in the above sections occurs on time scales ranging from nanoseconds to seconds. Picking up the pace, we shall now have a look at ultrafast magnetization dynamics.

2.2 Ultrafast magnetization dynamics

The field of ultrafast magnetization dynamics started with Beaurepaire's observation of sub-ps demagnetization of a Ni-film upon excitation with a 60 fs laser pulse [5]. They modelled their observations with the phenomenological three-temperature model (3TM), which assigns a temperature and heat capacity to the three separate subsystem of interest, being the electrons, their spins and the lattice (see Figure 2.3). The laser pulse is absorbed by the electron system, which quickly thermalizes internally within 100 fs. This excess energy then flows to the lattice via electron-phonon scattering with a typical time scale of 1 ps. Part of this energy also finds its way to the spin system, causing a rise in spin temperature and a decrease in magnetization on a sub-ps time scale.

This phenomenological description does not give any insight in the microscopic mechanisms behind the ultrafast magnetic response. Aside from energy conservation, the total angular momentum should also be conserved in the system. During the demagnetization, angular momentum is rapidly transferred from the spin system to either the electrons or phonons. One of the main goals of this new research field has been to find channels through which this angular momentum transfer takes place.

The most obvious channel would be that of spin-lattice relaxation, but this was thought to occur on a time scale of more than 100 ps [13]; much too slow to explain the experimental results. Another explanation could be ultrafast interaction between the spins and...
2.2 Ultrafast magnetization dynamics

Figure 2.3: The flow of energy between the three subsystems in the 3TM

the electrons. But this was disproved by Stamm et al. [14], who used X-Ray Magnetic Circular Dichroism (XMCD) to probe both spin and orbital angular momentum. They showed that the electron orbital angular momentum does not increase during ultrafast demagnetization, and consequently that the electron system does not act as the reservoir for angular momentum. Another channel, that of direct transfer of the angular momentum of the photons to the spins, was also discarded as explanation due to a lack of photons to cause a significant change in magnetization [15].

Above explanations consider only local effects, that is, no spatial transport of spins/electrons takes place. Moving spins could possibly transfer angular momentum away from the area where demagnetization occurs, bypassing the need for local angular momentum conservation. Evidence for this superdiffusive spin transport was first observed by Malinowski et al. [16]. They observed a difference in demagnetization of two ferromagnetic layers with either a conducting and an insulating spacer layer in between. This was explained by spin transport through the spacer, over lengths in the order of 1 nm. Battiato et al. [17] modelled this spin transport using transport equations, and calculated that spin transport alone could completely account for the ultrafast demagnetization. These non-local transport effects were later also established to play a role in the AOS of GdFeCo [18]. But up to the present day, the dominant mechanism for ultrafast demagnetization remains a matter of debate.

One of the proposed local models is the Microscopic 3TM (M3TM) introduced by Koopmans et al. [19, 20], which will be used in this thesis to simulate ultrafast magnetization dynamics. In the spirit of the 3TM from Beaurepaire, the authors derived three coupled differential equations for the spin, electron and lattice temperatures. In this case the derivations start from microscopic Hamiltonians, giving a better insight in the underlying mechanisms.
Within this model, angular momentum conservation is explicitly taken into account. Angular momentum is transferred between the spin and lattice via an Elliot-Yafet spin-flip mechanism [21, 22]. This mechanism describes a spin flip induced by electron-phonon scattering, which is possible due to the spin orbit coupling (SOC). The electronic states in materials with SOC are not simply spin up and down states, but a mixture of the two. Due to this spin mixing there is a small probability $a_{sf}$ that an electron-phonon scattering event causes a spin flip, effectively transferring angular momentum from the spin system to the lattice.

2.3 M3TM

In this thesis we describe the ultrafast magnetization dynamics with the M3TM. We shall therefore now derive the M3TM as introduced by Koopmans et al. [20]. First a microscopic description of the electron, phonon and spin system will be given. Then the Hamiltonians describing the interactions between the systems will be presented, from which the scattering rates and thereby the energy transfer will be calculated using Fermi's Golden Rule. This finally gives three differential equations for the electron and phonon temperatures $T_e$ and $T_p$, as well as for the magnetization $m$.

A couple of approximations and assumptions are made along the way in the basic version of the model:

- All systems are in internal equilibrium, i.e. internal scattering happens extremely fast.
- The spin system can be described with $S=1/2$.
- The spin system does not influence the electron-phonon scattering.
- The ambient temperature lies above the Debye temperature, $T > T_D$.

2.3.1 Electron system

The electron system is modelled as a free electron gas with a constant density of states $g_e$ (DOS) around the Fermi level:

$$g_e(E_e) = D_F,$$  \hspace{1cm} (2.5)

where $E_e$ is the electron energy. The occupation of these states is given by Fermi-Dirac statistics:

$$f_e(E_e, T_e) = \frac{1}{1 + \exp \left( \frac{E_e}{k_B T_e} \right)},$$  \hspace{1cm} (2.6)
with $T_e$ the electron temperature and $k_B$ the Boltzmann constant. The heat capacity per atom is then

$$C_e = \frac{\partial}{\partial T_e}U_e(T_e) = \frac{\partial}{\partial T_e} \int_0^\infty E_e g_e(E_e) f_e(E_e, T_e) dE_e = \gamma T_e,$$  \hspace{1cm} (2.7)

with $\gamma = \frac{1}{3} \pi^2 D_F k_B^2$ a constant.

### 2.3.2 Phonon system

The phonon system is described with the Debye model\(^1\), assuming a linear dispersion relation up to a cut-off energy $E_D = k_B T_D$. The total number of modes is $N D_p$, with $N$ the amount of atoms and $D_p$ the amount of possible polarization states (one longitudinal and two transverse). The density of states is then given by

$$g_p(E_p) = \frac{3 D_p E_p^2}{k_B^3 T_D^3}.$$  \hspace{1cm} (2.8)

The occupation function is given by Bose-Einstein statistics:

$$f_p(E_p, T_p) = \frac{1}{\exp \left( \frac{E_p}{k_B T_p} \right) - 1}.$$  \hspace{1cm} (2.9)

The heat capacity per atom is thus given by

$$C_p = \frac{\partial}{\partial T_p} U_p(T_p) = \frac{\partial}{\partial T_p} \int_0^{E_D} E_p g_p(E_p) f_p(E_p, T_p) dE_p,$$  \hspace{1cm} (2.10)

which in general cannot be solved analytically. However, in the limit $T_p \gg T_D$ we find that $C_p = k_B D_p$. In practice, this approximation is already decent for $T_p = T_D$.

### 2.3.3 Spin system

We describe the spin system with a mean field Weiss model. We only consider the case of spin $S = \frac{1}{2}$, but the model can be extended to all possible values of $S$ (see section 2.3.5). The Weiss model describes $N D_s$ non-interacting spins that occupy two discrete energy levels according to Boltzmann statistics, with $N$ the number of atoms and $D_s$ the spin density. The occupation function of these levels is given by

$$f_S^\pm(\Delta_{ex}) = \frac{\exp \left( \frac{\pm \frac{1}{2} \Delta_{ex}}{k_B T_S} \right)}{\exp \left( - \frac{\Delta_{ex}}{k_B T_S} \right) + \exp \left( \frac{\Delta_{ex}}{k_B T_S} \right)}.$$  \hspace{1cm} (2.11)

\(^1\)Technically it is a modified Einstein model [20]
The splitting between energy levels due to the exchange interaction is
\[ \Delta_{ex} = 2k_B T_C m, \] (2.12)
with \( T_C \) the Curie temperature. The magnetization normalized to its saturation value
\[ m = \frac{M}{M_{sat}}, \]
is given by
\[ m = \exp \left( \frac{\Delta_{ex}}{2k_B T_S} \right) - \exp \left( -\frac{\Delta_{ex}}{2k_B T_S} \right). \] (2.13)
Combining Equation 2.12 and Equation 2.13, this simplifies (in our case of \( S = \frac{1}{2} \)) to the
well known relation
\[ m = \tanh \left( \frac{m T_C}{T_S} \right). \] (2.14)

### 2.3.4 Obtaining the differential equations

We assume that the individual subsystems remain in internal equilibrium, hence we neglect electron-electron and phonon-phonon scattering. The next assumption is that the presence of the spin system does not influence the energy flow between the electron and phonon system, since the spin specific heat is negligibly small. This leads to the following Hamiltonian for the electron-phonon interaction:
\[ H_{ep} = \frac{\lambda_{ep}}{N} \sum_k \sum_{k'} \sum_q N_D p c^+_k c_k (a^+_q + a_q), \] (2.15)
where an electron in state \( k \) is scattered into state \( k' \) by emitting \( (a^+_q + a_q) \) or absorbing \( (a_q) \) a phonon, and with \( \lambda_{ep} \) the electron-phonon coupling constant. This Hamiltonian can be evaluated using Fermi’s golden rule, giving the following expression for the energy flow between the electron and phonon system:
\[ \frac{dE}{dt} = \frac{2\pi \lambda_{ep}^2}{\hbar} \int_0^{E_D} E_p g_p(E_p) \left[ f_p(E_p) SI(-E_p) - \left( 1 + f_p(E_p) \right) SI(E_p) \right], \] (2.16)
where
\[ SI(\Delta E) = D_p^2 \int_0^{\infty} [1 - f_e(E)] f_e(\pm \Delta E) dE = D_p^2 \exp(\pm \Delta E/k_B T_e) - 1 \] (2.17)
is the integral over all electronic states in which the electron loses/gains an energy \( \Delta E \). Evaluating this integral gives:
\[ \frac{dE}{dt} = -3\pi \lambda_{ep}^2 D_p^2 D_e^2 D_p^2 E_p^4 \left[ \frac{1}{h k_B T_e^3} \right] \left[ \coth \left( \frac{E_p}{2k_B T_e} \right) - \coth \left( \frac{E_p}{2k_B T_p} \right) \right] \] (2.18)
\[ \rightarrow \frac{dE}{dt} = -\frac{3\pi \lambda_{ep}^2 D_p^2 D_e^2 k_B^2 T_e^3}{2h} (T_e - T_p) \] (2.19)
Defining the constant \( G_{ep} = \frac{3\pi^{2}k_{B}^{2}D_{p}k_{B}^{2}T_{D}}{2\hbar} \), we get the following two coupled differential equations for the electron and phonon temperature:

\[
\begin{align*}
C_{e}(T_{e}) \frac{dT_{e}(t)}{dt} &= G_{ep} [T_{p}(t) - T_{e}(t)] \\
C_{p}(T_{p}) \frac{dT_{p}(t)}{dt} &= G_{ep} [T_{e}(t) - T_{p}(t)]
\end{align*}
\]

(2.20)

To calculate the magnetization dynamics, the possibility of a spin flip is taken into account, giving the following Hamiltonian:

\[
H_{eps} = \sqrt{\frac{a_{sf}}{D_{s}}} \frac{\lambda_{ep}}{N^{3/2}} \sum_{k} \sum_{k'} \sum_{q} \sum_{j} c_{k}^{+} c_{k}(a_{q}^{+} + a_{q})(s_{j,+} + s_{j,-})
\]

(2.21)

where \( a_{sf} \) is the spin flip probability and \( s_{j,+} (s_{j,-}) \) is the raising (lowering) operator for the \( j \)th spin. There are four different scattering cases: phonon absorption or emission and spin flip upwards or downwards.

The total change in spin for phonon absorption, with a spin flip from up to down, is then

\[
\frac{dS}{dt} = D_{p}^{2}D_{p} \frac{a_{sf} \lambda_{ep}}{D_{s}} \left[ \sum_{k} \sum_{k'} \sum_{q} \sum_{j} c_{k}^{+} c_{k}(a_{q}^{+} + a_{q})(s_{j,+} + s_{j,-}) \right] \int_{0}^{E_{D}} E_{p} \left[ 1 + 2S \tanh \left( \frac{\Delta_{ex}}{2k_{B}T_{e}(t)} \right) \right] dE_{p}.
\]

(2.22)

A similar equation can be obtained for all four possible scattering cases. Summing them all up and simplifying with \( A = \frac{6\sigma_{sf} \Delta_{ex} \lambda_{ep}^{2}k_{B}D_{p}^{2}T_{D}}{\hbar k_{B}T_{D}D_{s}} \) gives

\[
\frac{dS}{dt} = -A \int_{0}^{E_{D}} E_{p} \left[ 1 + 2S \tanh \left( \frac{\Delta_{ex}}{2k_{B}T_{e}(t)} \right) \right] dE_{p}.
\]

(2.23)

Using \( S = -m/2 \) and solving the integral:

\[
\frac{dm}{dt} = AE_{D}^{2} \left[ 1 - m \tanh \left( \frac{\Delta_{ex}}{2k_{B}T_{e}(t)} \right) \right].
\]

(2.24)

This can be further simplified with \( R = \frac{8\sigma_{sf}^{2}T_{D}^{2}G_{ep}}{\pi k_{B}T_{D}D_{s}} \) and \( \Delta_{ex} = 2k_{B}T_{C}m \) to finally give

\[
\frac{dm}{dt} = RT_{p}(t)T_{C}m \left[ 1 - m \tanh \left( \frac{T_{C}}{T_{e}(t)}m(t) \right) \right].
\]

(2.25)

The magnetization dynamics is thus described by the following coupled differential equations:

\[
\begin{align*}
\gamma_{T_{e}}(t) \frac{dT_{e}(t)}{dt} &= G_{ep} [T_{p}(t) - T_{e}(t)] \\
C_{p} \frac{dT_{p}(t)}{dt} &= G_{ep} [T_{e}(t) - T_{p}(t)] \\
\frac{dm}{dt} &= RT_{p}(t)T_{C}m(t) \left[ 1 - m(t) \tanh \left( \frac{T_{C}}{T_{e}(t)}m(t) \right) \right].
\end{align*}
\]

(2.26)
2.3.5 Extended M3TM

It is possible to extend the M3TM as it was described above by dropping some of the restrictions. Roth et al. [23] used a real Debye model and included the spin specific heat. With this extension, demagnetization behaviours in Ni as a function of laser fluence and ambient temperature could be reproduced. A further extension was done by Schellekens and Koopmans [24]. They extended the model to $S = N/2$ instead of just $S = \frac{1}{2}$, and introduced more than one spin system. They were able to reproduce the AOS in ferri-magnets (which we shall discuss in section 2.4.3). The details of this extension will not be discussed here, they can be found in the respective publications.

2.4 Optical Switching

The results discussed in the previous sections raised the question to what degree it was possible to optically control magnetism on a fs time scale. More specifically it would be of huge technological interest if it were possible to all-optically switch magnetization. The first indication that this was possible was discovered by the Rasing group in Nijmegen in 2005 [25, 26]. They could coherently control the spin dynamics induced by an intense ultrashort laser pulse, based on the polarization of the light.

The real breakthrough was achieved two years later by the same group, when they showed fully deterministic switching in a ferrimagnetic GdFeCo alloy [6]. The direction of the switch was determined by the helicity of the circularly polarized light, as can be seen in Figure 1.3. The time scale of this switching process was later shown to be as short as 30 ps [27], the fastest switch ever demonstrated. To understand the images from Stanciu \textit{et al.}, we take a look at a schematic overview of their experiment in Figure 2.4. In the center of the laser pulse, the sample is heated above the Curie temperature, causing a random multi-domain state to form when the sample cools down, a process called Thermal Demagnetization (TD). Deterministic AOS only occurs at a narrow range of fluences at the edges of the laser pulse. At the outermost edges of the laser pulse, the fluence is below the switching threshold. By sweeping the beam across the sample, a fully deterministically switched domain can be written\textsuperscript{2}.

The switching process was explained as the result of a two-fold effect of the laser pulse. First the sample is rapidly heated by the pulse, causing a sub-ps demagnetization as described in the previous sections, and bringing the spin system into a highly susceptible state. Then the symmetry is broken by an optically induced magnetic field, the direction of which is determined by the helicity of the light. We shall now discuss these two effects separately.

\textsuperscript{2}A video demonstrating this process can be found at \url{www.youtube.com/watch?v=Sa0kwR2dmpg}
2.4 Optical Switching

Figure 2.4: A schematic representation of an AOS switching experiment as in Figure 1.3. The laser beam is swept across a sample causing the magnetization to switch from $-M$ (black) to $+M$ (white). AOS only occurs in a narrow region on the edges of the pulse.

2.4.1 Heating

That the temperature greatly influences the magnetization of a material should be no surprise: magnetization is an ordering of spins and temperature fluctuations destroy this order. With the mean field model a good approximation of the magnetization as a function of temperature can be found in the form of the well-known Equation 2.13. This equation can be solved numerically to give an $M(T)$ curve as shown in Figure 2.5a. The magnetization decreases with increasing temperature up until the Curie temperature $T_C$, above which a ferromagnet becomes paramagnetic.

The coercivity $H_C$ is also affected by the temperature of the sample. The simple expression $H_C = \frac{2K}{\mu_o M_s}$ derived in section 2.1.2 would suggest that the coercivity increases with temperature because $M_s$ decreases. However, the anisotropy $K$ also strongly decreases upon increasing temperature [28]. Furthermore, we have seen in section 2.1.3 that magnetic switching is a thermally activated process. At higher temperatures, there is an increasing chance that the energy barrier of the anisotropy will be spontaneously overcome by the thermal fluctuations, effectively reducing the coercivity. The interplay of these temperature dependencies is not trivial and depends on the specific material parameters [12, 29, 30].
2.4 Optical Switching

Figure 2.5: The magnetization as a function of temperature for (a) a simple ferromagnetic materials, and (b) a ferrimagnet with two inequivalent sublattices. The compensation temperature $T_{\text{Mcomp}}$, where the two magnetizations of the sublattices are equal, is indicated as well.

But in general, the coercivity is found to decrease with increasing temperature, making it easier to switch a heated material. This is shown in Figure 2.6a, where both a normal and a heated hysteresis loop (with the reduced hysteresis and magnetization) are shown.

In ferrimagnets with multiple sub-lattices, such as GdFeCo, the effect of the temperature is more complicated. The saturation magnetization and Curie temperature of the different materials in the compound can often be tuned by changing the exact composition. This allows one to create ferrimagnets with one specific magnetic compensation temperature $T_{\text{Mcomp}}$ where the magnetization of the two sub-lattices are equal, as shown in Figure 2.5b. Since the magnetization of the sub-lattices are oppositely oriented, this means that the net magnetization of the material is 0 at this temperature. The role of this compensation temperature will be discussed at the end of section 2.4.3.

The reduction in the coercivity can be exploited for magnetic recording, a principle called Heat Assisted Magnetic Recording (HAMR, see for example [31]). For this technology high anisotropy (and thus high coercivity) storage media are used, which allow denser recording compared to normal media. Normally, one would need very high fields to switch the magnetization of these materials, making them unusable for hard disk recording. But if the material is locally heated (by a laser), the coercivity decreases, making it possible to switch the magnetization with a lower magnetic field. The material then cools down back to its highly stable high coercivity state. This process is shown in Figure 2.6. The technology is being heavily researched by the hard disk industry because of its high storage density, and working prototypes have already been demonstrated by for example Seagate\(^3\).

\(^3\)http://blog.seagate.com/business/seagate-demos-hamr-drives-at-intermag-conference/
2.4 Optical Switching

Figure 2.6: The effect of heating on the magnetic properties. (a) shows a normal hysteresis loop (blue), and a hysteresis loop after heating by an intense laser pulse (red). Both the coercivity and the saturation magnetization are reduced (by $\Delta H_C$ and $\Delta M_{demag}$ respectively). (b) shows the process of a HAMR write cycle which makes use of the coercivity reduction, reproduced from [31]. The medium is heated by a laser, allowing writing using the available field from the write head. The medium then cools down to the high coercivity state.

The principle of thermo-magnetic recording can also be taken to the regime of ultrafast dynamics. For example, Hohlfeld et al. [32] showed magnetization reversal in GdFeCo within several hundreds of ps by using fs laser pulses and an externally applied field lower than the coercivity. The temperature is quickly increased by the laser pulse, thereby quenching the magnetization and the coercivity so that a small applied field was enough to switch the magnetization. On the contrary, in the AOS experiments of Stanciu et al. [6] there was no applied field, and the ‘final push’ to switch the magnetization was provided by another mechanism, which we shall discuss in the next section.

2.4.2 Helicity Dependence

The helicity dependence of the AOS was initially explained by an optically induced magnetic field. The mechanism for this is called the inverse Faraday effect, as first described by Van der Ziel et al. [33]. The normal Faraday effect in which a magnetized medium causes a change in polarization of light upon propagation through this medium is well-known, and often used in experiments (see section 3.2). In contrast, the inverse effect in which magnetization is induced in a medium by polarized light, is relatively unknown. The static magnetization that is induced by high intensity laser radiation reads

$$M(0) = \frac{\chi}{16\pi} [E(\omega) \times E(\omega)^*],$$  \hfill (2.27)
where $\chi$ is the magneto-optical susceptibility, and $\mathbf{E}(\omega)$ the electric field of the light of frequency $\omega$. It follows from this relation that the induced magnetization is oriented along the wavevector ($\mathbf{k}$), and that left- and right-handed circularly polarized light induces opposite magnetizations. Effectively, the IFE was shown to act as a magnetic field in magnetic systems [26], following the same relation as Equation 2.27. The size of this magnetic field thus increases linearly with the laser power. In magnetic materials with typical laser fluences of some mJ/cm$^2$, estimates of the induced field strength range from 0.1 T to 20 T [26, 34].

The mechanism behind the IFE is similar to stimulated Raman scattering, combined with spin-orbit coupling. A detailed description can be found in the original publications [33, 35], and Kimel et al. [36] gave a description in the context of ultrafast magnetism. One thing to note is that photons are not absorbed during this process. Some photons lose a small amount of energy (the energy needed to create a magnon), but they do not lose any angular momentum.

Doubt was cast on this explanation with the IFE in later research. One thing that remained unclear was the lifetime of the magnetic field induced by the IFE. The laser pulse is Gaussian in time and the generated field pulse is expected to follow a similar temporal profile. It is thought to have a slower exponential decay, meaning that a magnetic field is still present after the laser pulse has disappeared, and when the sample starts cooling down from near the Curie temperature. This way, the magnetization grows from being quenched to the direction set by the IFE induced field. Estimates of this effective lifetime range from 0.2 ps [37, 34] to 3 ps [27] to several tens of ps [38], but the microscopic mechanism behind such a long lifetime remains unclear and heavily debated.

Furthermore, it was known from theory that the strength of the IFE in metals should have a strong frequency dependency, following a $1/\omega^3$ relation [39]. But this frequency dependency was not found in all-optical switching experiments by Steil et al. [40] and Khorsand et al. [9].

In this latter publication, Khorsand et al. therefore proposed a different explanation for the observed helicity dependence based on Magnetic Circular Dichroism (MCD). Right- and left-handed circularly polarized light experience different refractive indices$^4$ based on the magnetization of the material, and therefore a difference in absorption is possible. This difference in absorption is called the MCD. The HD-AOS was now explained by stating that for the one polarization enough light is absorbed to cross the switching threshold, whereas the other polarization is absorbed less and does not cause switching.

$^4$More specifically, the imaginary part of the dielectric tensor is different for different polarizations
2.4.3 Towards one theory on AOS

This theory of helicity dependent switching due to MCD, was fuelled by the discovery that heating alone was a sufficient stimulus to trigger AOS. Radu et al. [8] showed that it was possible to all-optically switch GdFeCo with just linearly polarized light, each subsequent pulse switching the magnetization back and forth (a toggle switch). They found an intricate switching behaviour by using X-ray MCD to separately probe the dynamics of the Gd and Fe sub-lattices, shown in Figure 2.7. Due to different demagnetization rates of the Fe and Gd, a transient ferromagnetic state arises even though the coupling between the two sub-lattices is anti-ferromagnetic. The Gd sub-lattice then continues to demagnetize and eventually switches direction due to the anti-ferromagnetic exchange interaction. These dynamics have been reproduced theoretically, using the atomic LLG [41] as well as the M3TM [24]. The transient ferromagnetic state can be explained by taking into account exchange scattering between the two sub-lattices.

![Figure 2.7: The separate magnetization dynamics of Gd and Fe in the GdFeCo alloy, reproduced from [8]](image)

Even though it was shown that the earlier experimental AOS results could also be explained by the MCD theory [9], the question remained if this explanation was complete. The results of Hohlfeld et al. [42] for example, that showed that AOS was more efficient at lower temperature (down to 10 K), remained unexplained. For this reason, Alebrand et al. [38] investigated the interplay of heating and helicity in a series of two-pulse experiments. They could discern between heating and helicity dependent effects by using one linearly and one circularly polarized pulse, and varying the fluence and delay between the two pulses. Surprisingly, they found that both a moderate amount of heating and helicity
were needed to achieve AOS, contradicting the earlier results. They also claimed that the helicity information from the light pulse was somehow stored (for example in the form of a magnetic field induced by the IFE) in the system for as long as 100 ps, much longer than previously assumed possible. We can conclude from these results that the theories on AOS are still incomplete.

Rather than relying on theory, Mangin et al. [7] therefore used a set of empirical rules to search for AOS in different types of materials. On top of the known RE-TM alloys, they found that also RE-TM multilayers and even RE-free synthetic ferrimagnets were able to switch all-optically. By varying the composition of their samples, they were able to determine what was and was not important to achieve AOS. The key to AOS turned out to have two antiferromagnetically coupled sub-lattices with the magnetic compensation temperature $T_{\text{Mcomp}}$ above room temperature. This $T_{\text{Mcomp}}$ is then crossed when the sample is heated by the laser pulse. Around $T_{\text{Mcomp}}$ the remanent magnetization is low and the susceptibility very high, facilitating an easy switch. This requirement was later reinterpreted by Hassdenteufel et al. [43], who stressed the importance of a low remanent magnetization for AOS (which is the case near $T_{\text{Mcomp}}$).

### 2.4.4 AOS in Ferromagnets

The understanding of AOS was challenged even further when Lambert et al. [10] demonstrated HD-AOS in ferromagnetic thin films such as [Co/Pt]$_N$ multilayers and granular recording media of FePt grains. For the thicker Co/Pt structures ($N > 3$) they only observed thermal demagnetization, but for thinner samples AOS was observed, as can be seen in Figure 2.8a. The authors speculate that dipolar fields play a role, something that was confirmed to be the case in ferrimagnets [44]. We speculate that another important factor could be the IFE. It is likely that the interfacial spin-orbit coupling is also involved, and maybe even direct transfer of the angular momentum of the photons occurs. But up to this day the dominant driving mechanism remains unknown.

An idea of the strength of the driving mechanism can be obtained from the switching experiments under an externally applied field, shown in Figure 2.8b. Here it can be seen that a field of around 10 Oe is sufficient to counteract the unknown mechanism. It should be noted, however, that this applied field is present during the entire switching process, whereas the helicity dependent process persists only for a few ps after laser excitation.

### 2.4.5 AOS in the M3TM

To get a more complete understanding of the mechanisms that could be responsible for AOS, we shall model AOS in ferromagnetic thin films using the M3TM. The results of simulations with this model will then be presented in Chapter 4. Several terms have to be added to the M3TM equations in section 2.3.4 to accurately describe AOS.
2.4 Optical Switching

Figure 2.8: AOS in a [Co/Pt]₃ multilayers, reproduced from [10]. (a) The helicity dependence is shown by using linear (L), left-handed (σ⁻) or right-handed (σ⁺) circularly polarized light. (b) The effect of an applied magnetic field opposite to the switching direction is shown.

Laser Pulse

First we introduce the effect of the laser pulse, which is assumed to only heat up the electron system. The pulse is approximately Gaussian in time with width Γ, total power $P₀$ and centred around $t = 0$:

$$P(t) = \frac{P₀}{\Gamma \sqrt{\pi}} \exp \left[ -\left( \frac{t}{\Gamma} \right)^2 \right]$$  \hspace{1cm} (2.28)

This term is added on the right-hand side of Equation 2.26.

Magnetic fields

To model the effect of magnetic fields, we can introduce an effective field by adding a Zeeman term to the exchange splitting:

$$\Delta_{ex} = 2k_BT_Cm + 2\mu_0\mu_BH_{eff}.$$  \hspace{1cm} (2.29)

We can break down this effective field into an externally applied field and a possible field pulse induced by the laser (caused by the IFE). The sign of this field pulse is dependent
on the helicity of the laser pulse, and it is directed along the wave vector of the light (which is nearly perpendicular to the sample in our case). Its temporal profile is given by the convolution of the Gaussian laser pulse with an exponential decay with lifetime $\tau$ (see [45, 34]):

$$H_{\text{pulse}}(t) = H_0 P_0 \frac{\Gamma}{\sqrt{\pi}} \exp \left( \frac{-t^2}{\Gamma} \right) \ast \exp \left( -\frac{t}{\tau} \right) = H_0 P_0 \frac{2}{\tau} \exp \left[ \frac{\Gamma^2}{4} - \frac{t}{\tau} \right] \left( 1 + \text{erf} \left[ \frac{t}{\Gamma} - \frac{\Gamma}{2\tau} \right] \right).$$

(2.30)

Note that we have defined $H_0$ as a peak power (power does not decrease with increasing lifetime) that scales with the laser power $P_0$.

With the effective field $H_{\text{eff}} = H_{\text{appl}} + H_{\text{pulse}}(t)$, we obtain for the exchange splitting:

$$\Delta_{\text{ex}} = 2k_B T_C m + 2\mu_0 \mu_B \left( H_{\text{appl}} + \frac{H_0 P_0}{2} \exp \left[ \frac{\Gamma^2}{4} - \frac{t}{\tau} \right] \left( 1 + \text{erf} \left[ \frac{t}{\Gamma} - \frac{\Gamma}{2\tau} \right] \right) \right).$$

(2.31)

To simplify the notation, we introduce a scaled field $b_{\text{eff}}$, such that

$$b_{\text{eff}}(t) = \frac{2\mu_B}{2k_B T_C} B_{\text{eff}} = \frac{2\mu_0 \mu_B}{2k_B T_C} H_{\text{eff}},$$

(2.32)

so $b_{\text{eff}} = 1$ corresponds to a Zeeman splitting equivalent to the exchange splitting at $T = 0$ K.

**Heat diffusion**

In the experiments, samples cool down after excitation by heat diffusion to the environment. We neglect heat transport within the sample, i.e. the sample has a uniform temperature, since the films under investigation are extremely thin (a couple of nm). Heat transport does takes place between the sample and the substrate, which we assume to be an infinite reservoir with a high heat conductivity and ambient temperature $T_{\text{amb}}$. Effectively we thus model the sample and substrate as two uniform heat baths, between which heat diffusion takes place. This heat diffusion is then modelled as

$$C_e \frac{dT_e}{dt} = -\kappa(T_e - T_{\text{amb}}).$$

(2.33)

The heat diffusion constant $\kappa$ is chosen such that the heat diffusion has the same time scale as observed in experiments.

**Direct transfer of angular momentum**

Another effect that might play a role in AOS is the direct transfer of angular momentum from the photons to the spin system. This is incorporated into the M3TM by adding an
extra source term to the change in magnetization:

\[
\frac{dm}{dt} = \frac{dL_{\text{photon}}}{dt} = \frac{\mu_B}{g \mu_{\text{at}}} \frac{dN_{\text{foton}}}{dt} \frac{1}{N_{\text{atoms}}},
\]

(2.34)

with \( \mu_{\text{at}} = 1.72 \mu_B \) the atomic magnetic moment of Co and \( g \approx 2 \) the electron spin g-factor. The amount of photons per atom \( \frac{N_{\text{foton}}}{N_{\text{atoms}}} \) scales with the laser power \( P(t) \), and it is estimated that (see [15])

\[
\int_{-\infty}^{\infty} \frac{dN_{\text{foton}}}{dt} \frac{1}{N_{\text{atoms}}} dt \approx 0.01
\]

(2.35)

for a typical laser fluence of 1.5 mJ/cm\(^2\). Assuming that this fluence roughly corresponds to \( P_0 = 1 \) J/s, we get

\[
\frac{dL_{\text{photon}}}{dt} = \frac{0.01}{2 \times 1.72} P(t) \approx AP(t),
\]

(2.36)

where we have introduced the constant \( A \approx 0.03 \) J\(^{-1}\) that can be varied to change the strength of this effect.

**Final equations**

Adding the above terms to Equation 2.26 leads us to the following system of three coupled differential equations:

\[
\gamma T_e(t) \frac{dT_e(t)}{dt} = G_{ep} [T_p(t) - T_e(t)] + P(t) - \kappa [T_e(t) - T_{\text{amb}}]
\]

\[
C_p \frac{dT_p(t)}{dt} = G_{ep} [T_e(t) - T_p(t)]
\]

\[
\frac{dm}{dt} = \frac{RT_p(t) \left[ m(t) + b_{\text{eff}}(t) \right]}{T_C} \left[ 1 - m(t) \coth \left( \frac{T_C}{T_e(t)} \left[ m(t) + b_{\text{eff}}(t) \right] \right) \right] + AP(t)
\]

(2.37)

The above equations can be solved for \( T_p, T_e \) and \( m \) using a numerical differential equation solver in programs like Matlab or Mathematica.
Chapter 3

Methodology

In this chapter we describe the sample fabrication and characterization. We start by explaining the structure of our samples and how they are fabricated. Then the standard MOKE measurement technique, as well as the time resolved MOKE setup with all its components will be discussed. We end this chapter by explaining the experimental procedure of a switching experiment, one of the most used experiments in this thesis.

3.1 Sample growth and structure

In this thesis we study optical switching in ferromagnetic systems that could be used in future memory devices. In particular, we focus on Co/Pt layers that are already used as magnetic recording media in current devices. It was experimentally shown by Lambert et al. that AOS occurs in these materials, and the samples we study are therefore similar to the Co/Pt layers they investigated. A schematic overview of the sample composition is shown in Figure 3.1.

The Co/Pt layers can be repeated to create a multilayer structure, but we restrict ourselves to $N = 1$ and $N = 2$, which Lambert et al. showed to be optimal for AOS. The Co thickness is varied between 0.4 and 0.8 nm based on three considerations:

- The samples should still have PMA, setting an upper limit of about 1 nm on the Co thickness (see section 2.1.1)
- If the Co layer is thinner than 0.4 nm, we will no longer have a well-defined uniform Co layer, but a Co/Pt alloy.
- Lambert et al. showed that the power threshold for AOS decreases with decreasing Co thickness, making a thinner Co layer preferable for optical switching experiments.

The samples are grown layer by layer using a DC sputtering process in ultra high vacuum,
3.1 Sample growth and structure

Figure 3.1: A schematic overview of the samples that are used in this thesis, with the respective layer thickness in nm. Starting from a boron doped Si substrate, a Ta seed layer is sputtered to accommodate growth. The following Pt layer ensures proper growth of a PMA Co layer. The Co/Pt structure is then grown with a variable Co thickness $d_{\text{Co}}$. The samples are capped with a Ta layer to prevent oxidation.

Figure 3.2: A schematic overview of the sputtering process

see Figure 3.2. A high voltage is put between an anode ring and a cathode of the material that is to be deposited, called the target. An argon gas is inserted, which gets ionized and accelerated towards the target. There the high energy ions knock atoms off this target. Part of these atoms land on the substrate and condense, forming a thin layer. Polycrystalline layers can thus be grown with a near Ångström precision. In the case of Co, these layers are polycrystalline and consists of grains with slightly differently oriented crystal axes.
3.2 MOKE

The samples described in the previous section are all measured using the Magneto-Optical Kerr Effect (MOKE). This effect entails that when light is reflected off a magnetized sample, its polarization is changed dependent on the magnetization of the sample. First discovered by John Kerr in 1877 [46], this effect is similar to the earlier discovered Faraday effect where the polarization is changed upon transmission through a magnetized medium.

These effects find their microscopic origin in the spin-orbit coupling, which effectively couples the magnetic (determined by electron spin) with the optical (determined by electron orbital motion) properties of a material. We shall give only a phenomenological description of the effect. It is possible to describe it quantum mechanically, but for this the reader is referred to for example [45].

Linearly polarized light can be considered as a linear combination of left- and right-handed circularly polarized light. These two modes have both a different velocity and absorption/reflection in the magnetic material. This difference is determined by the dielectric tensor, whose off-diagonal components are dependent on the magnetization of the material. A linearly polarized beam will therefore experience rotation of the polarization and will also gain an ellipticity. We write this as the complex rotation

\[ \Theta = \theta + i \epsilon, \]  \hspace{1cm} (3.1)

where \( \theta \) is defined as the Kerr rotation and \( \epsilon \) as the Kerr ellipticity. This complex rotation is directly proportional to the magnetization of the sample, providing a way of measuring this magnetization by measuring the rotation.

Figure 3.3: A schematic overview of a standard (polar) MOKE setup, with the photo-elastic modulator (PEM) being optional. See text for a description of the components and their functions.

The Kerr rotation and ellipticity can be measured in a standard MOKE setup, as depicted in Figure 3.3. We will first consider the case without the photo-elastic modulator (PEM).
A laser beam is passed through the polarizer P1 to obtain a linearly polarized beam. When the light is reflected from the magnetic sample, it will pick up a Kerr rotation and ellipticity. The second polarizer P2, also called analyser, is rotated by 90° with respect to P1. If the polarization at the sample would not change, all light would thus be blocked by P2. But if there is a change in polarization, then a small amount would pass through P2 and reach the detector. It should be intuitively clear that in this crossed polarizer setup, the intensity on the detector is proportional to the change in polarization caused by the sample.

The signal-to-noise ratio can be increased by introducing the PEM, which introduces a time-dependent modulation of the polarization. Locking in to the modulation frequency will now remove polarization independent effects. It can be shown (see Appendix B for the derivation) that the intensity at the detector now consists of three parts: a DC component and a first and second harmonic in the PEM frequency. The first harmonic is proportional to the Kerr ellipticity and the second harmonic to the Kerr rotation, which both can be obtained by using a lock-in amplifier.

The two main configurations of a MOKE measurement, polar and longitudinal, are shown in Figure 3.4. With polar MOKE one is sensitive to the out of plane component of the magnetization, with longitudinal MOKE to the in plane component. Since all our samples have PMA, we will only use MOKE in the polar geometry, with an angle of incidence close to 0° with respect to the normal.

A standard MOKE setup uses a polarized laser beam to pick up a Kerr rotation. It is also possible to use a Kerr microscope, which uses MOKE in an optical microscope to generate a two-dimensional image of the magnetization in a material. The spatial resolution of this device is limited by the resolution of the optical microscope, which is around 200 nm in our case.
3.3 Time Resolved MOKE setup

3.3.1 Stroboscopic measurements

With the above mentioned MOKE and Kerr microscope techniques it is possible to make magnetic images with high spatial resolution. The temporal resolution, however, is limited by the signal acquisition time which is typically in the range of ms. A different tactic is thus required to study the ultrafast magnetization dynamics on a fs time scale. For this we use a stroboscopic pump-probe measurement. A high intensity pump pulse is used to excite the sample, after which its effect is measured with the low intensity probe pulse using MOKE. By varying the time delay between the two pulses, the magnetization can be measured as a function of time after the excitation with the pump.

3.3.2 Experimental setup

To do these stroboscopic measurements we redesigned and rebuilt a polar Time-Resolved MOKE setup (TRMOKE), which is shown schematically in Figure 3.5 and a picture is shown in Figure 3.6.

![Schematic diagram of the TR-MOKE setup](image)

Figure 3.5: A schematic overview of the TR-MOKE setup, see text for a description of the components and their functions. In reality the angle of incidence is much closer to 0° (with respect to the normal) than depicted here.

The beam from the pulsed laser is split into a pump and a probe beam using a beam splitter with an intensity ratio of 20:1. The pump is modulated using a mechanical chopper at
3.3 Time Resolved MOKE setup

Figure 3.6: A picture of the TR-MOKE setup, with the path of the probe beam in red and the pump beam in blue. The reflected and blocked pump beam is not shown. The QWP is not shown but can be inserted after the chopper.

A frequency of 60 Hz. It is then reflected by a retro-reflector mounted on a translation stage, which acts as a delay line to control the time delay between pump and probe pulses. A quarter wave plate (QWP) can be inserted into the pump beam path to change the polarization of the pump from linear to circular. The pump is then focused on the sample, after which the reflection from the sample is blocked.

The probe beam is used as a standard MOKE measurement beam, and it is passed through the linear polarizer P1 and the PEM, which modulates the polarization of the beam with a frequency of about 50 kHz. The probe is then focussed and reflected from the sample. It passes through the analysing polarizer P2, after which it hits the detector.

The signal from the detector is passed to the first lock-in amplifier. This lock-in is set to the frequency of the PEM (first or second harmonic), and gives the standard MOKE signal as explained in section 3.2. In principle it is possible to measure the time-resolved demagnetization with this signal, but the signal to noise ratio is rather poor. To improve this, the signal from the first lock-in is passed to the second lock-in which is set to the frequency of the chopper. Because the pump is modulated by this chopper, the second lock-in is sensitive only to pump-induced changes in the MOKE signal, which is the ultrafast demagnetization. The signal to noise ratio is greatly increased using this double modulation technique.

The sample is put inside an electromagnet coil that produces a uniform magnetic field,
which can be used to switch the magnetization direction of the sample. All experiments are performed at room temperature.

### 3.3.3 Laser parameters

Two different Ti:Sapphire lasers are used to generate the ultrashort laser pulses. The first laser is a Tsunami laser pumped by a Millennia Xs, the second is a Spirit-NOPA system. The Spirit-NOPA is an amplified system and has 200 times lower repetition rate but a 20 times higher pulse energy compared to the Tsunami. Furthermore, the Spirit-NOPA has customizable output parameters, whereas the Tsunami’s parameters are fixed. An overview of the different parameters of the laser beam is given in Table 3.1. The high repetition rate of the Tsunami gives a high average power but lower pulse energy compared to the Spirit-NOPA. Since the Spirit-NOPA system was installed only at the end of this project, most experiments in this thesis were performed with the Tsunami. The presented results are thus obtained with the Tsunami laser pulses, unless mentioned otherwise.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Spirit-NOPA</th>
<th>Tsunami</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pulse length</td>
<td>20-50</td>
<td>70</td>
</tr>
<tr>
<td>Wavelength</td>
<td>650-900</td>
<td>780</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>&lt;0.5</td>
<td>80</td>
</tr>
<tr>
<td>Output power</td>
<td>0.5</td>
<td>1.8</td>
</tr>
<tr>
<td>Max pump power at sample</td>
<td>50</td>
<td>800</td>
</tr>
<tr>
<td>Pump spot size at sample</td>
<td>14</td>
<td>7</td>
</tr>
<tr>
<td>Probe spot size at sample</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>Pump fluence at sample</td>
<td>8.2</td>
<td>3.3</td>
</tr>
</tbody>
</table>

Table 3.1: An overview of the laser beam parameters for the two different laser systems

The spot size at the sample is determined using the so-called knife-edge method. For a description of this method we refer to Appendix C. The quality and wavelength of the pulses are checked by measuring the spectrum using an OceanOptics spectrum analyser. The power of the laser beam is measured using a Spectra Physics thermopile power meter.

### 3.4 Experimental procedure for optical switching

In this thesis, we will often perform writing experiments, in which a region of a magnetic sample is switched by the laser. The procedure for such an experiment is schematically shown in Figure 3.7, and goes as follows:

1. The magnetization of the sample is saturated in one direction by applying a magnetic field higher than the coercivity. The pump beam is blocked.
3.4 Experimental procedure for optical switching

2. A bias field smaller than the coercivity is applied in the opposite direction. In the case of AOS the field is set to 0. The magnetization remains in the state set by the previous step.

3. The pump beam is unblocked and swept across the sample. The beam locally switches the magnetization, resulting in a line that is "written".

4. The pump is blocked again and the field is set to 0.

5. The probe is scanned across the area of interest to see the results of the switching experiment.

The size of the switched area can be determined as a function certain parameters such as bias field or pump power. This gives us an insight in the influence of these parameters on the optical switching process.

![Figure 3.7: The evolution of the magnetization during a typical writing experiment. The static hysteresis loop is shown in blue, the pump-induced loop in red.](image)

Unfortunately it is not possible to do time resolved switching experiments with this setup. The reason for this is that the sample is usually switched as soon as the first pump pulse hits the sample. All subsequent pump pulses do not induce any effect other than the standard demagnetization. To overcome this, the magnetic state of the sample would have to be reset by applying a field $H > H_C$ between every pulse pair. The laser we primarily used had a repetition rate of 80 MHz, meaning that one would have to apply timed field pulses that are shorter than a ns, with a typical strength of 20 mT. This was not possible with the available experimental tools.
Chapter 4

Modeling Optical Switching

The results of the simulations with the M3TM are presented in this chapter. We first present a standard M3TM demagnetization simulation to make the reader familiar with the output from the simulations. We then work our way towards AOS step by step, as will be done in the experiments. We start with predicting the results of optical switching experiments with linearly polarized pulses and an applied magnetic field, i.e. field assisted switching. The next step is then to simulate optical switching with circularly polarized pulses, by now including a magnetic field pulse induced by the IFE. Both the strength and length of this field pulse are a matter of great discussion, which is why the effect of these two parameters on the optical switching will be investigated. Finally we investigate the feasibility of all-optical switching, giving special attention to the experimental results of Lambert et al. [10].

All the results presented here are obtained with the standard M3TM, but several results were also qualitatively confirmed with the extended M3TM discussed in section 2.3.5 and [24].

4.1 Simulating demagnetization

In section 3.1 we have introduced the samples for which we will study the optical switching properties. The magnetic material in these structures are thin Co layers, wedged in between two Pt layers. In the simulations we will therefore use the material parameters of a Co thin film as input. One exception is the spin-flip rate $a_{sf}$, which is only about 0.03 for normal Co. However, it was found by Kuiper et al. [47] that it is greatly enhanced to $a_{sf} \approx 0.12$ in Co/Pt structures due to enhanced spin-orbit coupling, which is the value we have used in our simulations. An overview of all the material parameters can be found in Table 4.1.

We derived in section 2.3.1 that the electron heat capacity per atom is given by $C_e = \gamma T_e = \frac{1}{3} \pi^2 D_F k_B^2 T_e$, where substitution gives $\gamma = 7.33 \times 10^{-8}$ eV/K$^2$. For the phonon heat
4.2 Field assisted switching

Having obtained all the necessary tools, we can simulate the switching experiments described in section 3.4. In these experiments, a high power laser pulse is applied together
4.2 Field assisted switching

Figure 4.1: A typical solution for $T_e$ (black), $T_p$ (blue) and $m$ (red) of the final set of differential equations (with no external field or IFE in this case). Both the electron and phonon temperature are scaled to the Curie temperature as explained in the text.

with a bias field. The pulse brings the electron, phonon and spin temperature above $T_C$, causing a rapid demagnetization. Remagnetization occurs when the temperature drops below $T_C$. In absence of any other symmetry breaking effects (such as the IFE or direct transfer of angular momentum), the direction of this remagnetization is determined by the bias field, as shown Figure 4.2. It is thus possible to have an optically induced switch of the magnetization, as was shown experimentally by Hohlfeld et al. [32].

The time scale of this reversal is mainly determined by the speed of the heat diffusion, i.e. how fast the system cools down from $T_C$. In this case, after about 10 ps the magnetization is deterministically reversed. The subsequent growth back to equilibrium is much slower and takes place on a scale of some hundreds of ps. More details on the time scale of this reversal and how to decrease it can be found in the thesis of Schellekens [45].

Another thing that should be noted is that a rather high (0.1 T) magnetic field is needed to switch the magnetization even when it is almost fully quenched. If no laser pulse would be applied, the ‘coercive field’ is even around 1 kT. This is in stark contrast with the experimental coercive fields of the samples, which are in the 10 mT range. The reason for this is that the M3TM is a mean field model, which ignores the effect of thermal fluctuations and local correlations such as domain formation. In the model we technically have an infinite perpendicular anisotropy, as the magnetization can only change in size but not in orientation. The coercivity is therefore the field at which the exchange field is overcome, which is indeed around a kT. To improve on this, an atomistic spin model such
4.2 Field assisted switching

Figure 4.2: The results of a simulation with $P_0 = 0.8$. In (a) the electron (red) and phonon (blue) temperature are shown. The magnetization as a function of time is shown in (b) for no externally applied field (red) and for $B_{\text{ext}} = \pm 0.1 \, \text{T}$ (black and blue respectively)

as that of Kazantseva et al. [48] could be used.

Since we cannot do time-resolved switching measurements with our setup (see section 3.4), we shall now investigate the influence of the bias field on something we can measure: the size of the switched domain. At a certain bias field, there is a threshold power $P_{\text{threshold}}$ for which switching occurs. The size of the domain is directly related to this threshold power via the Gaussian laser spot profile

$$P(x) = P_0 \exp \left( -\frac{x^2}{\sigma^2} \right),$$

where $P_0$ is the peak power and $\sigma$ the width of the laser pulse. If $P_0$ is above $P_{\text{threshold}}$, ...
then a domain at the center of the laser spot will switch. At the edge of this domain, the power of the laser pulse equals the threshold power \( P(x) = P_{\text{threshold}} \). This gives for the domain size \( \Delta x \):

\[
\Delta x = 2\sigma \sqrt{-\ln \left( \frac{P_{\text{threshold}}}{P_0} \right)},
\]

(4.2)

where the factor two comes from the domain being switched on both sides of the center of the pulse.

We have determined the threshold power for increasing bias fields, which is shown as the black curve in Figure 4.3. The switching power threshold decreases approximately linearly with the external bias field. When the bias field gets very close to zero, the threshold diverges, as even a full demagnetization will not lead to switching without the symmetry breaking bias field. As the bias field reaches the ‘coercive field’ of 1 kT, the threshold reduces to 0, since spontaneous switching will now occur. Note that in experiments we shall never reach these very high fields.

From \( P_{\text{threshold}} \) we can calculate the domain size using Equation 4.2, which gives the red curve in Figure 4.3. At both ends of the curve we see an increase in slope, caused by the flattening of the Gaussian profile at the center and edges of the pulse. In the intermediate range the curve is approximately linear. When the power of the pulse is decreased to 0.5, it can be seen that the minimum bias field for a switch is increased, as to be expected.

To conclude, we have shown in this section that an externally applied field is necessary to cause optical switching with linearly polarized pulses. We have predicted the written domain size as a function of applied field, which will be compared to the experimental results in section 5.3.2.

### 4.3 AOS using circularly polarized pulses

Having looked at optical switching with just linearly polarized pulses, we shall now look at the effect of circularly polarized pulses. This means that we shall now include a magnetic field pulse that is induced by the IFE, in the form as it was introduced in section 2.4.5. Because the IFE in ferromagnets lacks a full theoretical description or a direct experimental observation, we can make rough estimates for realistic values of the strength \( B_{\text{pulse}} \) and lifetime \( \tau \) of this field pulse. By investigating the effect of these two parameters, we can discuss the possibility of AOS by the IFE in ferromagnets. Another effect that could play a role is the direct transfer of angular momentum from the photons to the spin system, which will be briefly investigated at the end of this section.
4.3 AOS using circularly polarized pulses

4.3.1 Strength of the IFE

The easiest way to determine the effect of the strength of the field pulse on the switching process is to compare it with an externally applied field $B_{\text{ext}}$. Experimentally this was done by Lambert et al. [10] in their experiments on Co/Pt. The static applied field is constant throughout the entire switching process, while the field pulse only persists for a lifetime $\tau$, which is typically 0.3 ps (see the next section). We have compared these two by determining the threshold power needed for switching as a function of both $B_{\text{ext}}$ and $B_{\text{pulse}}$, resulting in the phase diagram shown in Figure 4.4. The range of $B_{\text{pulse}}$ is chosen based on estimates of the IFE strength in literature [25, 26, 34], which range from 0.1 T to 20 T. Since Figure 4.4 is a rather complex phase diagram, we shall go through it step by step.

First of all, it should be noted that the cross section at $B_{\text{pulse}} = 0$, indicated with the dotted line, gives the same result as (the very left-most part of) Figure 4.3. We see again that the threshold power quickly diverges as the applied field approaches zero. The behaviour in the range $-2 \text{T} < B_{\text{pulse}} < 6 \text{T}$ is rather predictable: both increasing $B_{\text{pulse}}$ or $B_{\text{ext}}$ lowers...
4.3 AOS using circularly polarized pulses

Figure 4.4: The threshold power for switching as a function of bias field $B_{\text{ext}}$ and field pulse strength $B_{\text{pulse}}$, for a field pulse with lifetime $\tau = 0.3$ ps. We start the simulation from $m = -1$, so that a positive field assists in switching. No switching takes place in the black area.

the threshold power. In this range, we can state that the effect of the field pulse is roughly a factor 100 smaller than that of the static applied field.

If we increase the pulse field above 6 T, we see that AOS with $B_{\text{ext}} = 0$ becomes possible. For even higher pulse fields, the switching becomes more and more dependent on the pulse field. The externally applied field becomes less important, and switching even occurs for negative applied fields.

It should furthermore be clear that there is no straightforward relation between the effect of the static field and that of the field pulse. The contour lines are near horizontal in the bottom left of Figure 4.4, indicating that the static applied field is dominating the switching behaviour. But they become more slanted and eventually vertical as we move to the top right, which means that the pulse field starts to dominate over the static field.

In the switching experiments, we change the helicity of the light by rotating a QWP, thereby effectively varying $B_{\text{pulse}}$. We follow a similar procedure as in section 4.2 to predict the results of these experiments. We calculate the size of a switched domain, this time as a function of $B_{\text{pulse}}$, the result of which is shown in Figure 4.5a. The result shown is for an
applied field of +2 mT, but the curves are similar (but shifted) for different applied fields in a range of a couple mT.

We see that if the pulse power is above 0.83, switching will occur for all field pulse strengths. But as one would expect, the domain size is significantly (up to three times) larger if the field pulse is in the correct direction, i.e. if it is positive in this case of a negative starting magnetization. For pulse powers below 0.83, we see that switching only occurs if the field pulse is large enough and in the correct direction. This is schematically shown in Figure 4.5b.

4.3.2 Lifetime of the IFE

Having looked at the strength of the field pulse, we now turn our attention towards its lifetime. This lifetime has been a matter of great debate since the IFE was suggested as mechanism for HDAOS (see for example [38, 34] and section 2.4.2), and estimates vary between 0.2 and several ps. One thing that is clear, is that the lifetime is crucial in explaining AOS with the IFE, which can be understood by looking at Figure 4.6. To bring the system in a highly susceptible state, it is necessary that it is heated above \( T_C \), as shown in Figure 4.6a. This means that the magnetization remains quenched for some time after the laser pulse. The field pulse can now only have an effect if it is still present when the temperature drops below \( T_C \). This is illustrated by the difference between the two curves in Figure 4.6b. The field is not sufficiently high when the sample is cooled below \( T_C \) to cause a switch in case of the shorter field pulse, in contrast to the longer pulse which does cause a switch. Both magnetic field pulses as well as the laser pulse are shown in Figure 4.6c.

The influence of the lifetime of the IFE was first investigated by Vahaplar et al. [27], who refined their results in a later publication [34]. The authors use a multiscale approach as developed by Kazantseva et al., which uses first principle calculations as well as the Landau-Lifshitz-Gilbert (LLG) and the Landau-Lifshitz-Bloch (LLB) equation. Combining this with a two-temperature model for the electrons and the lattice, they investigated the magnetization reversal by an IFE induced field pulse. With this model, Vahaplar et al. constructed a phase diagram of the magnetization as a function of the field pulse lifetime (which they dubbed the optomagnetic pulse \( H_{OM} \)) and the laser pulse power, which is shown in Figure 4.7a.

We have performed a similar simulation using the M3TM with equivalent parameters and the result is shown in Figure 4.7b. The results of both models are very similar, showing that both models can be used to describe optical switching. We shall now have a look at the three distinct phases and their boundaries.

For low laser pulse powers we do not see any switch because \( T_C \) is not yet reached. If the power is increased, reversal becomes possible for long lifetimes, as was the case in Figure 4.6. Further increasing the power, we see that from a certain point only thermal demagnetization is observed. The sample is now heated to such a degree that it cannot cool
down in the set simulation time, resulting in zero final magnetization (unless a strong field is present). If the simulation is extended to a larger time scale, non-deterministic switching is observed in which numerical fluctuations determine the direction of the remagnetization. This effect is seen as noise at the triple point in the phase diagram of Figure 4.7b.

We see that a lifetime of at least 0.15 ps is necessary to achieve AOS in this case. The actual lifetime of the IFE is still heavily debated, but it is most often assumed to be around 3 ps, well above the required value. Alebrand et al. [38] even suggested that the helicity information is stored for up to 100 ps, but did not link this to the IFE. Lifetimes as long as these would result in rather wide reversal windows. Therefore despite the fact that the exact parameters of the induced magnetic field pulse are still under discussion, one can conclude that the lifetime will not be the limiting factor for all-optical switching.

The threshold for the lifetime is of course dependent on the specific simulation parameters. If the demagnetization rate is increased, the magnetization dynamics will speed up, and one would expect that an even shorter lifetime is sufficient for switching. Changing the demagnetization constant $R$, we see in Figure 4.7c that the reversal window indeed becomes larger. A lifetime as short as 0.05 ps is now sufficient for switching, which is even below the laser pulse duration of 0.1 ps. This shows that enhancing the demagnetization rate increases the feasibility of AOS, which is of crucial importance for applications. Such an enhancement was experimentally realized by Kuiper et al. [47] in Co/Pt multilayers compared to pristine Co due to an enhanced spin-flip scattering at the Co/Pt interfaces.
4.3 AOS using circularly polarized pulses

Figure 4.5: The written domain size as a function of the pulse field strength. This field pulse strength corresponds to the polarization of the light, which changes from RCP to LP to LCP as indicated by the colors. In (a) the domain size is given for pulses of different power $P_0$, with a lifetime $\tau = 3$ ps, under applying an external field of $+2$ mT. We start the simulation from $m = -1$, so that a positive field assists in switching. A sketch of the written domain is given in (b) for the six indicated situations.
Figure 4.6: The results of a simulation with $P_0 = 0.8, B_{\text{pulse}} = -5T$. In (a) the electron (red) and phonon (blue) temperature are shown. The magnetization as a function of time is shown for (b) $\tau = 0.1\,\text{ps}$ (black) and $\tau = 0.3\,\text{ps}$ (red). Plotted on shorter time scale for clarity, (c) shows the normalized profile of the laser (red) and field (black) pulse for $\tau = 0.3\,\text{ps}$ (solid) and for $\tau = 0.1\,\text{ps}$ (dashed).
4.3 AOS using circularly polarized pulses

Figure 4.7: The final magnetization as a function the lifetime $\tau_{(om)}$ of the field pulse and the pulse power, starting with $m = 1$. The results of Vahaplar et al. [34] with an atomistic LLG model are shown in (a). Our results with the M3TM with the standard parameters is shown in (b), and the result with a twice as fast demagnetization in (c). All simulations use a laser pulse duration of 0.1 ps, and a field pulse strength of 10 T–30 T, scaling with the laser power.
4.3 AOS using circularly polarized pulses

4.3.3 The combined influence of the lifetime and strength of the IFE

Having determined the separate influence of both the lifetime and the strength of the IFE, we shall now vary both these parameters. This results in a phase diagram for the threshold power as shown in Figure 4.8. We see that AOS is possible for a wide range of parameters. In this case, there is a lower limit for $\tau$ of about 0.15 ps, but this can lowered if the demagnetization rate is increased, as discussed in the previous section. Following the threshold between switching and no switching, we see that the field pulse should be longer as the strength becomes weaker. As $B_{\text{pulse}}$ approaches zero, the field pulse will need to become infinitely long to still achieve switching, in which case the field pulse becomes a static field like in section 4.2.

It should be noted that the parameters for which we predict AOS switching here are well within the range of estimates found in literature. We can therefore conclude that AOS should be achievable in ferromagnets. The switching is determined by the circular polarization of the laser pulse, which induces a magnetic field pulse due to the IFE. The range of fluences for which this switch occurs can be increased if the demagnetization rate is enhanced.

![Figure 4.8: The threshold power for switching as a function both the strength and the lifetime of the field pulse. We start the simulation from $m = -1$, so that a positive field assists in switching. The jagged profile at the border between no and deterministic switching is due to discretization.](image-url)
4.3.4 Direct angular momentum transfer

Another helicity dependent contribution could come from the direct transfer of the angular momentum of the photons to the spin system. This mechanism was found too weak to explain ultrafast demagnetization [15], but it might be sufficient to break the symmetry in the demagnetized state and cause AOS. To test this we have compared the strength of this effect (characterized by the constant $A \approx 0.03$ in Equation 2.36) to the effect of an external field, as was done for the IFE in Figure 4.4. The result is another phase diagram that is shown in Figure 4.9.

![Figure 4.9: The threshold power for switching as a function of bias field $B_{\text{ext}}$ and angular momentum transfer constant $A$. We start the simulation from $m = -1$, so that a positive field assists in switching.](image)

We see that changing the strength of the direct transfer has very little effect on the switching threshold. AOS can not be observed even if we exaggerate the strength of the effect by a factor 50. Several combinations of simulation parameters were tried (varying for example the laser pulse length and demagnetization rate as in the previous sections), but this did not give any different results.

We can thus conclude that the direct transfer of angular momentum of the photons does not play a role in AOS. One important reason for this is that the transfer takes place only during the laser pulse. This means that most of the angular momentum transfer takes place before the magnetization is fully quenched, making it a very inefficient mechanism, as explained in section 4.3.2. This could be overcome by designing a two-pulse experiment,
in which the first pulse heats the sample and the second pulse uses the direct transfer once the sample starts cooling down. In a scenario like that it should be possible to switch the magnetization with the angular momentum of the photons.

4.4 Simulating AOS experiments in ferromagnets

In the previous sections we have shown that is possible to achieve AOS with a circularly polarized laser pulse, if the IFE is present to generate a field pulse. We shall now use this to simulate an AOS experiment as it was schematically shown in Figure 2.4. The results from these simulations will be compared to the experimental results of Lambert et al. [10], as they currently have the only experimental proof of AOS in ferromagnets.

4.4.1 Including spatial effects in the M3TM

We have to extend our model beyond the simple macrospin approach to be able to reproduce images like Figure 2.4, where the spatial profile of the laser pulse plays a major role. To do so, we now consider an array (1D case) or a grid (2D case) of $N$ magnetic domains and solve the M3TM for each of these domains. Several aspects are included:

- The Gaussian spatial profile of the laser pulse is included by varying the laser power across the grid.
- Multiple pulses are swept across the sample in the writing experiments. This is simulated by using the calculated final magnetization profile as input for the next simulation. For each subsequent simulation, the center of the laser pulse is displaced by one domain. This scan rate is fixed for all simulations and roughly corresponds to the scan rate in experiments.
- In real experiments, there will be inhomogeneities in the sample and the magnetic field. In the simulation, we include this as a Gaussian distributed random magnetic field noise with an amplitude of around 0.15 mT.
- In section 4.4.3 we shall further extend the model by also including spatial correlations in the form of dipolar coupling between neighbouring domains.

With this extension, we shall be able to reproduce the magnetization profiles from Lambert et al., most specifically the one shown in Figure 4.10. This specific result shows the clearest example of AOS, and the scans with a counteracting applied field give us an idea of the strength of the switching mechanism.
4.4 Simulating AOS experiments in ferromagnets

Figure 4.10: The results of the AOS experiment from Lambert et al. for a [Co(0.4 nm)/Pt(0.7 nm)]₃ multilayer, as in Figure 2.8b. We shall reproduce the magnetization profiles of the highlighted areas.

4.4.2 Simulating magnetization profiles

Before we discuss the results of a sweep of multiple pulses, we shall first consider the simple case of a one pulse experiment without an applied field. The result of such an experiment is shown in Figure 4.11, where all dimensions are scaled to the pulse width $\sigma$, since this is the only length scale in the model. In the center of the pulse we see thermal demagnetization; an area of randomly oriented domains. Here the sample gets heated well above $T_C$, and the direction of remagnetization is determined by the random noise in the simulation. At the edges of the pulse we see a small (blue) area where the magnetization is deterministically switched. In this ring, the power is just right to enable a switch by the magnetic field pulse that is induced by the circularly polarized light.

Having explained the effect of a single pulse, we now discuss the simulations where a train of pulses is swept across the sample, which are shown in Figure 4.12. Starting with the case of no applied field in Figure 4.12a, we see that the magnetization profile of area a. in Figure 4.10 can be perfectly reproduced. The sweep across the sample leaves behind a single switched domain. At the end of the sweep, at the center of the last pulse, we see the thermal demagnetization as in Figure 4.11.

When a small positive field of 0.5 mT is applied (so that it opposes switching), this thermal demagnetization disappears (see Figure 4.12b). In the center of the last pulse we now see an area with a uniform positive magnetization. This is understandable since the applied field is
4.4 Simulating AOS experiments in ferromagnets

Figure 4.11: The magnetization profile after a single pulse experiment, with the spatial dimensions scaled to the pulse width $\sigma$. We have $P_0(0,0) = 1$, $B_{\text{pulse}} = -3\, \text{T}$, $\tau = 0.3\, \text{ps}$, $B_{\text{noise}} = 0.15\, \text{mT}$, no applied field, and a grid of 100x100 domains.

several times higher than the noise that is responsible for the random domain orientation. Comparing this to the experimental results, we see that the thermal demagnetization persists even for the highest applied field of 1.15 mT (11.5 Oe, area d in Figure 4.10). The discrepancy could be solved by increasing the noise, but the noise would have to be unrealistically high to be able to reproduce the experimental data. This suggests that the model is too simplified to fully explain AOS. We shall address this issue at the end of this section by including dipolar fields.

If the applied field is increased further to 1.8 mT in Figure 4.12c, the switching becomes non-deterministic. The applied field is now exactly in balance with the IFE field, so that the noise once again determines the direction of remagnetization. In the experiments, this situation corresponds to the image at 0.7 mT (7 Oe, area c), where we also see that the switching is no longer deterministic. Larger connected domains are formed, in contrast to the simulation in which just random domains are present. This again shows that the model could be improved. Further increasing the applied field to 2.1 mT in Figure 4.12d, we see
that the applied field now dominates over the field pulse and that no switching takes place, as in the experiment at 1.15 mT (area d in Figure 4.10).

Figure 4.12: The magnetization profiles after a train of pulses is swept across the sample. The experiments are performed for an applied field of (a) 0 mT, (b) 0.5 mT, (c) 1.8 mT and (d) 2.1 mT. The four figures can be compared to the four regions of interest in Figure 4.10. The simulation parameters are equivalent to those in Figure 4.11.

We have thus seen that the general optical switching behaviour in ferromagnets can be reproduced, with realistic values for the magnetic fields involved. However, what could not be reproduced was the presence of thermal demagnetization at high applied fields and the formation of large connected domains. This is because we have ignored any spatial correlation effects. We shall therefore now improve the model by including such a spatial coupling between domains through dipolar fields.

4.4.3 Dipolar coupling

In magnetic films with PMA, there are strong dipolar fields that support the formation of a multi-domain state. The dipolar field one magnetic domain experiences is caused by the
surrounding domains (see Figure 4.13), and can be calculated using standard electromagnetism calculations. We add these fields to the effective field term in Equation 2.37. In its simplest form, taking the 1D case and only nearest neighbour dipole interaction, the effective field term now reads

$$b_{\text{eff}}(n, t) = b_{\text{appl}} + b_{\text{noise}}(n) + b_{\text{pulse}}(n, t) + b_{\text{dip}}[m(n + 1, t) + m(n - 1, t)], \quad (4.3)$$

where $n$ is the (discrete) position coordinate. The dipole term can be refined by also taking $m(n \pm 2, t)$ and further terms into account (with the appropriate scaling factor).

We calculate the dipole fields in the 2D case by calculating the magnetic field of an uniformly magnetized cuboid domain of 0.2 by 0.2 pm and a thickness of 1 nm (as shown in Figure 4.13). This field is integrated and averaged over the volume of a neighbouring domain. Using the saturation magnetization of Co ($M_s \approx 1400 \text{kA/m}$), we find that the average dipolar field of a domain is around 0.6 mT at the position of the nearest neighbour. This field rapidly decays to below 0.1 mT for further neighbours. Because of this rapid decay, only the 12 nearest neighbours are used in the calculation of the dipolar fields.

Figure 4.13: The dipolar fields in a thin film with PMA, that favour an opposite orientation of neighbouring domains.

We now have a system of $N$ coupled differential equations for $m$, instead of just $N$ separate equations (ignoring the equations for $T_e$ and $T_p$ that are uncoupled anyway). This greatly increases the computation time needed to solve these equations, typically by a factor 1000. The following simulations therefore use a much coarser grid and less steps (so a shorter sweep of the laser beam) then the simulations without dipole coupling.

We run a similar set of simulations as in Figure 4.12, but now include the dipole interaction, the result of which is shown in Figure 4.14. We see a similar general behaviour as in Figure 4.12; at low applied fields there is AOS which disappears at higher fields. But now we do see thermal demagnetization at the center of the last pulse when an external field of 0.5 mT is present (see Figure 4.14b), as in the experiment. This thermal demagnetization disappears again at higher fields, in contrast to the experiment (compare Figure 4.14c with area c in Figure 4.10). We can conclude that the dipole fields do indeed support the formation of a multi domain state, but the fields are not strong enough to fully explain the thermal demagnetization observed in the experiment of Figure 4.10.
4.4 Simulating AOS experiments in ferromagnets

This seems counter-intuitive if one compares the total strength of the dipole fields, which is around 2 mT, to the applied field which is several times smaller. However, the area in the center of the laser spot gets fully demagnetized and therefore no dipole fields are present at that time. Remagnetization occurs only after several picoseconds, and only from this point in time the dipole fields start playing a role. This is contrast to the externally applied field, which is present the entire time and has therefore a much stronger effect on the direction of the remagnetization.

Figure 4.14: The magnetization profiles after a couple of pulses are swept across the sample, now including the dipole interaction. The experiments are performed for an applied field of (a) 0 mT, (b) 0.5 mT, (c) 1.8 mT and (d) 2.1 mT. The four figures can again be compared to the four regions of interest in Figure 4.10. The simulation parameters are equivalent to those in Figure 4.11, but we now have a grid of 35x50 cells with 0.2 µm size. The film thickness, which is used to calculate the dipole fields, is assumed to be 1 nm.

The lifetime and strength of the magnetic field pulse (see section 4.3) that are used in the above simulations are well within the range of estimates in literature. We can therefore conclude that AOS in ferromagnets, and more specifically the experiments of Lambert et al. on Co/Pt, can be explained by taking the IFE as the driving mechanism. We furthermore
conclude that the (modified) M3TM is a valuable tool in describing AOS.

4.5 Summary

In this chapter, we have simulated (all)-optical switching with the M3TM. We started with simulating field assisted switching, and could make a prediction for the written domain size as a function of applied field. In section 5.3.2, we shall compare this prediction with the experimental results.

We then examined the effect of the helicity of the light pulse on the switching process. We first investigated the influence of both the strength and lifetime of a magnetic field pulse induced by the IFE. The written domain size as a function of field pulse strength was determined, which will be used in section 5.3.3. For the influence of the lifetime, a good agreement with literature results from an atomistic model was found. It was furthermore shown that the parameters for which we predict AOS lie well within the estimated actual values. We therefore state that AOS can be explained with the IFE. A different helicity dependent mechanism, that of direct angular momentum transfer, was found to be too weak to explain AOS.

We can summarize the requirements to achieve AOS with the IFE as follows:

- The magnetization should be (almost) fully quenched by the laser pulse.
- The IFE should have a strength of at least 3 T to 5 T (dependent on the lifetime).
- The lifetime of the IFE should be at least 0.2 ps to 0.4 ps (dependent on the strength).
- A faster demagnetization rate will lead to less strict requirements on the lifetime and strength of the IFE.

A real AOS experiment was simulated using an extended version of the M3TM, with the IFE as the driving mechanism. With a first simple extension, the general switching behaviour as it was reported by Lambert et al. could be reproduced for realistic values of all simulation parameters. Adding dipolar fields to the model resulted in an even better correspondence between experiments and simulations. We can thus for the first time explain AOS in ferromagnets, by invoking the IFE as the driving mechanism.
Chapter 5

Optical switching experiments

The previous chapter has provided us with a better theoretical understanding of the different mechanisms behind optical switching. We shall now present the results from the optical switching experiments in this chapter. As in the previous chapter, we shall work our way towards AOS step by step. We start with characterizing our Co/Pt samples by measuring the hysteresis loops using MOKE, and special attention will be paid to the coercivity. After this static characterization, we shall do time-resolved measurements of the demagnetization using the TR-MOKE setup.

Having characterized the samples, we shall perform optical switching experiments. First, the reduction of the coercivity will be investigated. The coercivity is reduced by heating with the laser pulse, and this reduction is crucial in facilitating optical switching. Then, we shall perform field assisted switching experiments for both linearly and circularly polarized light. We compare the results from these experiments with the predictions made in the previous chapter. Finally, we comment on the experimental feasibility of AOS in Co/Pt systems.

5.1 Static characterization

The static characterization of the samples is done by measuring the hysteresis loops (see section 2.1.2) with the standard polar MOKE technique (see section 3.2). All samples show square loops with sharp switches as shown in Figure 5.1a, which indicates that they all have PMA.

The observed linear background is caused by Faraday rotation in the lens (which is close to the magnet) and can be subtracted. If the signal is then also normalized to the signal at saturation, the graph of Figure 5.1b is obtained. We have measured the hysteresis loops of ten different samples, and fitted them with two error functions to obtain the coercivity.
5.1 Static characterization

Figure 5.1: A typical hysteresis loop, in this case from a single Co layer of 0.6 nm thickness. The raw data is presented in (a), the corrected loop with in (b), which also includes the fit with two error functions in red. The small jump at zero field is a measurement artefact due to the current reversal in the electromagnet.

The results are shown in Figure 5.2, in which a clear trend can be observed. The coercivity increases approximately linearly with the Co thickness. The double layer samples have a higher coercivity than the monolayers, which is to be expected if they are considered as one thicker Co layer. The increase of the coercivity with thickness is not trivial. From Equation 2.2 and 2.4, one would expect that the coercivity decreases with Co thickness because the effective anisotropy increases (considering $M_s$ is roughly constant). This trend can be observed at low temperature [49], but as already discussed in section 2.1.3, magnetic switching is a thermally activated process.

At room temperature the coercivity is determined by nucleation and domain wall motion. A similar trend in the coercivity as in Figure 5.2 was also found in previous research by Lavrijsen and Franken [50, 51]. They explained this trend by a difference in domain wall velocity. Thinner films have less pinning of the domain walls and hence a higher domain wall velocity. A higher domain wall velocity in turn means a lower coercivity, as any nucleated domain will quickly propagate and switch the entire sample.

Furthermore, we note that samples with the same composition (in theory) do not always have the same coercivity. There are two factors that cause this inconsistency. The first is that the conditions during the sputtering of the samples (especially the base pressure of the sputter chamber) might differ from sample to sample, causing the actual Co thickness to vary from the set value. The second factor is the inhomogeneity of the samples, caused by growth defects, impurities or dust particles on the surface of the samples. This causes the coercivity to fluctuate throughout the sample, so measuring at different positions will give a slightly different coercivity. For example, the coercivity will be lower when measured
5.2 Time-resolved characterization

Figure 5.2: The coercivity of our mono and double layer Co samples, as a function of the Co thickness. The lines are a guide to the eye. The error bars are based on the step size in the magnetic field sweep.

near a defect, because a domain can easily nucleate at the defect (see section 2.1.3). So although the trend in the coercivity is clear, one should take care when comparing the coercivity of different samples.

5.2 Time-resolved characterization

Having done the static characterization, we can move on to the time-resolved characterization of our samples. To do so, we saturate the sample and irradiate it with the pump beam which induces the ultrafast demagnetization. The magnetization as a function of time is obtained by varying the delay between the pump and the probe beam. We measure at both positive and negative saturation and subtract these two signals to exclude any non-
magnetic effects. As mentioned in section 3.3.2, we use a double modulation scheme. The signal from the second lock-in is almost free of noise and also proportional to the magnetization. Absolute calibration of this signal can be done with the use of the (much noisier) signal from the first lock-in or the saturation magnetization measured in the hysteresis loops (see section 5.3.1).

An example of a demagnetization trace is given in Figure 5.3a. We see an ultrafast demagnetization within 0.5 ps and a subsequent remagnetization. Eventually the sample cools down due to heat diffusion to the substrate. We see that there still is a demagnetization at negative time delay, meaning that the sample has not enough time to completely cool down between two pulses. This is not unexpected considering the very high repetition rate of the laser pulses (80 MHz). A similar behaviour was found for all different samples, as shown in Figure 5.3b.

![Image](image)

Figure 5.3: Demagnetization trace of (a) a sample with 0.8 nm Co, fitted with Equation 5.1. In (b) the demagnetization traces of several samples are shown. For clarity they are all scaled to the maximum demagnetization and subsequent curves are offset.

We can fit the demagnetization using an analytical solution for the 3TM derived by Dalla Longa [52]. Neglecting the spin specific heat and assuming a low fluence it was shown that

\[
-\frac{\Delta M(t)}{M_0} = \left[ \frac{A_1}{\sqrt{t/\tau_0} + 1} - \frac{(A_2\tau_E - A_1\tau_M)e^{-t/\tau_M}}{\tau_E - \tau_M} - \frac{\tau_E(A_1 - A_2)e^{-t/\tau_E}}{\tau_E - \tau_M} \right] \Theta(t) + A_3 \delta(t) * P(t),
\]

where \( P(t) \) is the laser pulse, \( \Theta(t) \) the step function and \( \delta(t) \) the Dirac delta function. The constant \( A_1 \) is the value of \( -\frac{\Delta M(t)}{M_0} \) in equilibrium, \( A_2 \) is proportional to the initial rise in electron temperature, and \( A_3 \) represents the state filling effects that occur due to temporal overlap of the pump and probe pulse. The de- and re-magnetization are parametrized by time scales \( \tau_E \) and \( \tau_M \), that describe the electron-phonon interaction and the magnetization
loss respectively. Heat diffusion is described by the inverse square root term, with time scale $\tau_0 >> \tau_E, \tau_M$.

The demagnetization traces of different samples were measured and fitted with Equation 5.1. The results for the three most important parameters, being the maximum demagnetization, $\tau_E$ and $\tau_M$, are plotted in Figure 5.4.

![Figure 5.4: The three most important parameters obtained from the demagnetization fits: (a) the maximum demagnetization and (b) $\tau_E$ and $\tau_M$ as a function of the Co thickness. The lines are provided as a guide to the eye. The error bars are based on the accuracy of the fits of the pumped hysteresis loops (for (a)) and the demagnetization traces (for (b)).](image)

The maximum demagnetization is found to increase with decreasing thickness. This is because the Curie temperature decreases when the thickness of the Co layer is decreased. A similar increase in temperature (all experiments use the same laser fluence) will then result in a higher demagnetization for the samples with the lower Curie temperature (as can be seen from the curves in Figure 2.5).

With the highest demagnetization being around 40%, we are still far from completely quenching the magnetization. It seems therefore highly questionable if we shall be able to achieve AOS, since we have seen in the previous chapter that a full demagnetization is necessary for AOS. However, there is one thing we have overlooked, which is the spatial profile of both the pump and probe laser spot. Because of the Gaussian profile of the pump spot, the demagnetization in the center of the spot will be much higher than at the edges. Because the probe spot is of a similar size as the pump, we will measure an average demagnetization and not the maximum demagnetization in the center of the spot. In Appendix D we show that when the pump and probe spot are the same size, the maximum demagnetization is roughly twice the measured demagnetization. In this case that would mean that there is a maximum demagnetization of 80% at the center of the laser spot. This puts AOS within reach.
We find that $\tau_M$ is mostly constant with a value around 0.15 ps, which corresponds with both theoretical and experimental values in literature [20, 47]. It is not trivial that $\tau_M$ remains constant with the cobalt thickness. It is mainly dependent on the demagnetization rate $\tau_M \propto R^{-1} \propto \frac{\alpha_{\text{sf}}^2}{T_C^2}$. As discussed above, a decrease in film thickness will lower $T_C$ and thus increase $\tau_M$. But $\alpha_{\text{sf}}$ will also increase because the spin flip is mostly an interface effect, which will decrease $\tau_M$. It seems that in our range of thicknesses, these two effects cancel each other.

The electron-phonon relaxation time $\tau_E$ increases with decreasing thickness. The value of $\tau_E = 0.4$ ps again corresponds with theoretical and experimental values in literature [20, 45]. The increase in $\tau_E$ for the thin samples might be explained by the higher demagnetization, as it was shown by Roth et al. [23] that $\tau_E$ increases with increasing maximum demagnetization. Furthermore, it could play a role that $\tau_E$ is not only determined by the Co, but also by the Pt layers. Changing the thickness will change the relative amount of Co and might thereby influence $\tau_E$. A systematic study of the thickness dependence of both $\tau_M$ and $\tau_E$ would give more insight into which effects play a role, but this is beyond the scope of this thesis.

We can compare the demagnetization traces of Figure 5.3b to the simulated demagnetization in Figure 4.1. The most striking difference is the amplitude of the remagnetization. In the simulation, an almost full remagnetization is obtained within a ps, whereas in the experiments we see only a slight ultrafast remagnetization and a much slower full remagnetization due to heat diffusion. Our simulations do correspond to results from literature on Co/Pt multilayers or just single Co layers [47]. The low remagnetization in our experiments is most likely caused by the sample composition. Our samples consist of a single Co layer sandwiched between Pt, and capped by a relatively thick Ta layer. Heat transport on the nano-scale between these layers might play a role. This is not accounted for in the model and might therefore explain the difference between experiment and simulation.

A limited amount of TR-MOKE experiments were also performed with the higher fluence Spirit-NOPA laser. Unfortunately the lower average power of this laser causes the signal to be much noisier. Added to the fact that a lower resolution delay line had to be used because the high resolution delay line malfunctioned, this lead to very noisy demagnetization traces that could not be fitted (an example is given in Appendix E). We did observe that there was less demagnetization at negative time delay compared to the Tsunami experiments. This was to be expected due the lower repetition rate of this laser, giving the sample more time to cool down between pulses.

It was furthermore found that a 30% to 40% demagnetization could be obtained with about a quarter of the full laser power, which is indeed the power for which the fluences of both laser systems are more or less equal (3 mJ/cm$^2$, see section 3.3.3). By increasing the power a demagnetization of 50% could be achieved. However, damaging of the sample occurred at about half of the full laser power, indicated by deformation of the hysteresis loops. No full quenching of the magnetization was thus observed, but considering the
earlier argument on the probe spot size it is likely that a full demagnetization in the center of the pulse can be achieved without damaging the sample.

5.3 Optical switching

5.3.1 Coercivity reduction

In the previous section we have seen that the pump pulse induces an ultrafast demagnetization. But as was discussed in section 2.4.1, the heating by the pump pulse also causes a decrease in the coercivity. We can measure this decrease by comparing a standard (no pump irradiation) hysteresis loop with a loop when the pump beam does hit the sample. An example of this is shown in Figure 5.5. We see a similar figure as in Figure 2.6a. There are three main differences between the normal and pumped loops that we shall discuss: we see a plateau in the switching process, and the saturation magnetization is decreased, as well as the coercivity.

![Hysteresis loops of a sample with 0.8 nm Co.](image)

Figure 5.5: Hysteresis loops of a sample with 0.8 nm Co. The loops are shown for four cases: before any pump irradiation was on the sample, during pump irradiation for both a negative and positive time delay, and after pump irradiation.

The plateau in the switch can be explained as a consequence of the spatial profile of both the pump and probe spot. Because the center of the probe spot is the hottest, this area will switch first. The probe spot is slightly larger than this switched domain, and therefore ‘sees’ both up and down magnetization, resulting in an average signal somewhere in between up and down (the exact signal depends on the size of the spot and the switched domain).
5.3 Optical switching

While the field is being increased, the switched domain will expand until the entire sample is switched. This is seen as the slanted switch towards the saturation magnetization.

This plateau is only observed for the thick films with a Co thickness of 0.8 nm. For these samples, the dipolar fields already discussed in section 4.4.3 are rather significant and support the formation of small domains. In the thinner films these fields are suppressed and larger domains are favoured, so that a large area under the probe spot is switched at once and consequently no plateau will be visible. Furthermore, the domain wall velocity is higher for thinner samples (see section 5.1), so that any nucleated domain will quickly expand beyond the spot size.

The decrease in saturation magnetization is simply the demagnetization. We see that also for negative time delay there is a decrease in the step size of the switch. This means that there still is a slight demagnetization, as discussed in the previous section. For a positive time delay of 0.5 ps, which corresponds to the peak in Figure 5.3, there is a large demagnetization.

The coercivity is greatly reduced by a factor of 3. This reduction is independent of the time delay. One would expect that because it is a thermally activated process, the coercivity would be reduced more when the sample is hotter, which is for positive time delay. But as Roth et al. [53] pointed out, one cannot accurately measure the dynamics of the coercivity this way, since switching by an external field is an irreversible process. One would need to reset the magnetic state of the sample before every pulse, as was mentioned in section 3.4. As it is now, we always measure the maximum reduction in coercivity, and we cannot say at which time delay this maximum reduction occurs. Liu et al. [54] indeed showed that the coercivity at negative time delay is barely reduced if the sample magnetization is reset before every pump pulse.

There are two other noteworthy features in Figure 5.5. The first is that there is a high amount of noise in the loop measured at short positive time delay, which is caused by the temporal (and spatial) overlap of the pump and probe pulses. This can cause interference between the two which results in noise. To solve this, one could employ a so-called two-color measurement scheme, where the pump and probe pulses have a different frequency.

The second feature is that there is a slight difference between the loops before and after the sample is irradiated by the pump beam. This is due to irreversible modification of the sample by the high intensity pump pulses. In this case, it is likely that interdiffusion between the different layers of the sample takes place, causing a slight change in the magneto-optical properties. With the higher pulse energy of the Spirit-NOPA, we see actual damaging of the sample, resulting in a modified coercivity and non-square loops. At these high intensities it is possible that the materials from the sample get evaporated\(^1\). By measuring the hysteresis loop before and after pump irradiation, we can check if such damaging has occurred and if the laser power should thus be reduced.

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\(^1\)A process that can actually be used to grow thin films, called Pulsed Laser Deposition
By fitting the hysteresis loops, the reduced coercivity of the different samples is determined. The reduction in the coercivity as a function of Co thickness is plotted in Figure 5.6. The error bars are rather large because the fitting is complicated by the switch plateau and noise in the pumped loops. In the 0.4 nm Co case, the coercivity is so small that a change is hardly measurable. For the other samples, the relative reduction in coercivity is more or less constant around 70%.

![Figure 5.6](image)

Figure 5.6: The reduction of the coercivity by the pump pulse, as function of the Co thickness in samples with a monolayer Co. The normal and reduced coercivity are shown in (a), the percentual reduction in (b). The lines are a guide to the eye.

We can explain this if we consider that the laser power was the same for all measurements, so the samples are all heated by the same amount. We can (naively) regard a magnetic switch as thermal activation over an energy barrier described by a Boltzmann factor $H_C \propto \exp\left(-\frac{E_{\text{barrier}}}{k_B T}\right)$. The relative reduction in coercivity will then always be the same for the same temperature difference, regardless of the absolute value of the coercivity. It should be noted that this a very simplified view, and many more effects are present that can influence the coercivity reduction (such as the temperature dependence of the anisotropy).

In conclusion, we have seen in this section that the coercivity of our magnetic samples is reduced by about 70% when heated with the pump pulse. The coercivity does not completely disappear for the used laser power, indicating that we still need an externally applied field to switch the sample. We shall now discuss this field assisted switching.

### 5.3.2 Field assisted switching

The drastic reduction in the coercivity upon heating by the pump pulse allows us to perform the writing experiments described in section 3.4. The result of typical writing experiment
can be seen in the Kerr microscope image in Figure 5.7. A well-defined line of opposite magnetization is written\(^2\).

We see that there is a difference in the width of the line at the beginning and end of the writing process. This is because there still is a bias field present after the magnetization has switched. The domains that are switched will therefore slowly expand (see section 2.1.3). In the beginning of the line the bias field is present for quite some time after switching, whereas at the end this field is turned off shortly after switching. This gives a different time for which the domains walls can propagate, and thereby a difference in width.

Figure 5.7: A Kerr microscope image after a switching experiment (at 12 mT bias on a sample with 0.8 nm Co), with the length scale shown at the top of the figure. An L-shape with a different contrast is visible, where the magnetization is opposite to the rest of the film. The writing process started in the top left (1), moved to the right (2) and then down (3). The width of the line is shown at the beginning and the end of the scan.

The Kerr microscope provides a way to directly measure the size of a written domain, which we usually take as the width of a written line. But to make these Kerr images, the sample needs to be removed from the TR-MOKE setup, making this a cumbersome measuring method. We therefore use the probe beam of the TR-MOKE to scan across a line to determine the width. We want to compare these widths for different bias fields.

\(^2\)This is also how the cover image of this thesis was obtained
Considering the discussion in the previous paragraph, it is crucial that each experiment uses the same laser scan speed and that the size of the domain is measured at the same position along the line.

Another complication can come from the fact that the domain wall velocity is dependent on the applied field. This might on itself cause a difference in observed domain size for different bias fields (as opposed to a difference in size of the nucleated domain). We have determined that this effect is rather small (in the order of 1 µm), by measuring the line width at several places along the line for several bias fields. We can therefore conclude that we really are measuring the size of the switched domain as a function of bias field.

Figure 5.8: A scan across a written line domain for different bias fields, for a sample with Co thickness of 0.8 nm, and \( H_c = 23 \text{ mT} \). The solid black lines are fit to the data using Equation 5.2.

A typical measurement in which the probe beam is scanned across the line is shown in Figure 5.8. For low bias fields, it looks like no full switch is observed. This is a consequence of the finite size of the probe spot. Because the domains we are measuring are of roughly the same size as the Gaussian probe spot, we cannot completely resolve these domains. Instead, we get a convolution of the Gaussian probe and the measured domain. Taking this domain as a rectangular function with width \( d \) (assuming that the edge, which is the domain wall, is much thinner than the spot size) we get for the intensity profile:

\[
P(x) = \frac{\pi}{2} \sigma^2 \left[ \text{erf} \left( \frac{d - x}{\sigma} \right) + \text{erf} \left( \frac{x + d}{\sigma} \right) \right],
\]  

(5.2)
where \( x = 0 \) is taken as the center of the domain, and \( \sigma \) is the probe spot size. With this equation we can fit the data from Figure 5.8 and obtain the domain size \( d \).

These measurements were done for several different samples and the results are shown in Figure 5.9a. For comparison, the prediction from the simulations in section 4.2 is shown in Figure 5.9b. The overall form of the curves is similar for the experimental and simulation results. We see a linear increase in domain size in the middle field region, and the slope increases at both low and high fields. However, in the experiment the domain size diverges well before the coercive field is reached, in contrast to the prediction. We attribute this to the thermal activation of the switching, which is not incorporated in the model.

Figure 5.9: The written domain size as a function of the applied bias field (scaled to the coercivity). Experimental data are shown in (a) for two different samples with 0.8 nm Co, where the black line corresponds to the fits in Figure 5.8, and one sample with 0.6 nm Co measured with both laser systems. The lines are a guide to the eye. The error bars are hidden for clarity, the error is for all measurements below 5 \( \mu \)m. The prediction from the simulations is shown in (b), which is a reproduction of Figure 4.3.

Comparing the different samples, we see that the results of the two samples with 0.8 nm Co are consistent with each other. The sample with 0.6 nm Co shows slightly larger domain sizes for the same laser power. This can be explained by the lower Curie temperature, so that less power (heat) is needed to switch these samples. For this sample, one series of experiments was also performed with the higher power Spirit-NOPA system. Not surprisingly, these higher pulse energies result in larger domain sizes. Unfortunately, no accurate measurements could be performed on samples with a Co thickness below 0.6 nm, because either the domains were too unstable or the samples got damaged by the laser.

In summary, we have investigated the field assisted optical switching experiments with linearly polarized laser pulses in this section. The switched domain size as a function of applied field follows the trend that was predicted with the simulations using the M3TM.
5.3.3 Helicity dependence

Having performed field-assisted switching, we take the next step towards HDAOS by investigating the helicity dependence of the switching. Therefore we insert the QWP in the pump beam as was shown in Figure 3.6. By rotating the QWP the polarization changes from left-handed circular (LCP) to linear (LP) to right-handed circular (RCP).

It would be instructive to compare the time-dependent demagnetization curves for different helicities, as was done for Ni by Dalla Longa et al. [55]. Unfortunately this was experimentally not possible due to imperfections in the QWP, such as contaminations and intrinsic deformations in the sheet of birefringent material. Because of these imperfections, when the QWP gets rotated to change the polarization, the path of the pump beam changes slightly. This changes the spatial overlap between pump an probe beam, which has a significant effect on the signal (see Appendix F). The apparent change in demagnetization is thus only a measurement artefact, any true changes in the demagnetization due to the polarization are obscured by this change in overlap.

To overcome this change in overlap, we shall therefore again measure the size of a switched domain, now as a function the angle of the QWP (and thus of the helicity of the light). Here the change in overlap does not matter because the information is obtained by the probe after the pump beam has caused a switch. The results of these experiments are shown in Figure 5.10. More experiments were performed on different samples and for different bias fields than shown in Figure 5.10, but the results were often noisy and irreproducible and are thus not shown.

We see a distinctive effect of the polarization of the light on the switched domain size. One would expect the curve to be periodic with a 180° period because the QWP is symmetric. Any deviations from this periodicity (such as the peak in the red curve at 320°) are again due to imperfections in the QWP. These imperfections might also cause a different transmission trough the QWP for different angles, leading to a difference in incident power, which would in turn influence the domain size. This was not the case, as the incident power was measured to be constant for all angles of the QWP (save a 1% noise).

The difference in domain size is thus purely caused by the difference in polarization of the beam. We see that for RCP light the domain sizes are significantly smaller than for the LCP light. The sizes for LP light are in between these two values. At first glance, this seems to roughly correspond with the predictions from Figure 4.5: the size of the switched domain is larger if the polarization of the pulse has the ‘correct’ orientation, causing a IFE field pulse in the right direction.

But when the experiment is performed for the two opposite bias fields (so performing a switch ‘up’ and a switch ‘down’), the results are unexpectedly exactly the same. RCP corresponds to a negative magnetic field pulse, and should therefore facilitate a switch

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3We call it right-handed, but we did not unambiguously determine the handedness.
5.3 Optical switching

Figure 5.10: The written domain size as function of the angle of the QWP, for (a) two different samples of 0.8 nm Co measured with the Tsunami and (b) 0.6 nm Co measured with the Spirit-NOPA. A domain size of 0 means that no switch in magnetization took place. The background color indicates the angle of the QWP for which the beam is circularly polarized (right- or left-handed). The lines are a guide to the eye. The error bars are again hidden for clarity, the error is for all measurements below 5 µm.

‘down’, and oppose the switch ‘up’. The situation is reversed for LCP, and the two curves should therefore be shifted by 90° with respect to each other. The fact that this is not the case excludes the IFE as the cause for the observed helicity dependence.

Aside from the IFE, another explanation for the helicity dependence could be MCD (see section 2.4.2). The different polarizations would have a different absorption in the sample which could also explain the change in domain size (more absorption leads to more heating and thus a larger domain). But the same argument as for the IFE holds: the two curves for the two opposite bias fields should have a phase difference of 90°. After all, if we start the switching experiment with a positive magnetization, LCP will be absorbed more than RCP, and vice versa for starting with a negative magnetization.

We have thus observed a polarization dependence that cannot be attributed to either the IFE or MCD. We shall now explain this polarization dependence as a measurement artefact. Since we do not have a perfect perpendicular incidence, we can define a polarization direction parallel (p) and perpendicular (s) to the plane of incidence. We know from literature that the absorption for these two polarizations differs slightly. This would have no consequence if our light would be perfectly circularly polarized, as both LCP and RCP would have the same ratio of s and p polarization. But due to the imperfect QWP, our light is not perfectly circularly polarized, but rather elliptically. This causes LCP and RCP light to have a different ratio of s and p polarization, and therefore a different absorp-
tion, which in turn causes a difference in written domain size. The absorption difference is irrespective of the magnetization, as was seen in the experiment. Measurements of the reflectivity (and thus the absorption) as a function of the QWP angle confirmed that the absorption is indeed different for RCP and LCP.

We can conclude from this section that although a helicity dependence was found, this can not be ascribed to the IFE. The helicity dependence can be explained as a measurement artefact. We can thus neither confirm nor disprove the presence of the IFE in our experiments. If the IFE is indeed present, a rough estimate estimate based on the simulations learns that the effect is smaller than 5 T.

5.3.4 AOS

The experiments in the previous section were attempted for several different samples, but in none of these AOS was observed. Switching could only be achieved when an external bias field was applied. This is not surprising considering the maximum demagnetization of 80% that was found in section 5.2. We know from our simulations as well as from literature that an almost full quenching of the magnetization is necessary to achieve AOS, something that could not be obtained with the low power Tsunami laser.

With the Spirit-NOPA laser, a higher demagnetization was possible. But before a full demagnetization could be observed, the high laser power caused damage to the samples. AOS could also not be found in the limited time that was available with this laser, despite trying several different samples and laser fluences.

The main parameter that determines if optical switching will occur is this laser fluence. Unfortunately, the fluence reported by Lambert et al. \(1 \times 10^{-3} \text{mJ/cm}^2\) is several orders of magnitude too low to induce any significant magnetization dynamics, and can therefore not be correct. However, it is reasonable to assume that the fluence threshold for AOS in Co/Pt is similar to that in ferrimagnets, which is around 5 mJ/cm\(^2\) [6, 34]. In the experiments with the Tsunami laser the fluence is a bit lower than this, but with the Spirit-NOPA we have achieved these fluences. AOS was not observed for either of these cases.

There are several possible reasons that could explain why we were not able to achieve AOS. The first reason is that the fluence we used in our experiments is simply not high enough. Very few experiments were done with the higher power Spirit-NOPA. This system should be able to reach high enough fluences, but our samples got damaged before these fluences were reached. This might be prevented by optimizing the samples, especially in terms of heat conductivity. Furthermore it should be noted that AOS often occurs only in a very narrow range of fluences, so a very careful adjustment of the fluence is necessary to observe AOS.

Another reason could be the relatively high repetition rate of both our lasers. This, in
combination with a too low heat conductivity of the substrate, can lead to heat accumulation in the sample which can cause damage. Furthermore, Lambert [56] found that a higher repetition rate tends to favour thermal demagnetization instead of AOS. Considering that our lowest repetition rate is still 100 times higher then in their experiments, it is plausible that even if the fluence would be high enough, we would only observe thermal demagnetization.

A final reason for the different results could simply be a slight difference in sample structure. The exact crystallographic structure of the thin films is highly dependent on the growth conditions during sputtering, as already mentioned in section 5.1. Therefore our samples are unavoidably slightly different than in the experiments by Lambert et al. But since the microscopic mechanism behind AOS remains unknown, it is difficult to predict if this will have a significant influence.

5.4 Summary

In this chapter, the results of the optical switching experiments on the Co/Pt samples were presented. We started with both a static and a time-resolved characterization of the samples. With the static measurements it was found that the coercivity increased linearly with the Co thickness in the range of 0.4 nm to 0.8 nm.

The time resolved measurements showed an ultrafast demagnetization followed by a slower remagnetization, with time scales comparable to those found in literature. The maximum demagnetization was found to increase with decreasing Co thickness because of the change in Curie temperature. For the thinnest sample, a demagnetization of 80% was achieved.

The next step towards optical switching was taken by investigating the reduction of the coercivity caused by the pump pulse. The coercivity was reduced by around 70%, independent of Co thickness. This reduction allowed us to perform field assisted switching experiments with linearly polarized pulses. The measured dependence of the switched domain size on the bias field corresponded well with the prediction based on the simulations.

The following step towards AOS was to perform switching experiments with CP pulses. A clear dependence of the switched domain size on the polarization of the light was found. However, this dependence could not be ascribed to the IFE or MCD, due to the fact that it was independent of the direction of the starting magnetization. The helicity dependence was explained as a measurement artefact caused by an imperfect QWP.

Even though the switching experiments were performed on several different samples, AOS was not observed. One reason for this is that the laser power that was used, was too low to cause a full demagnetization. Experiments with higher powers resulted in sample damage. Further optimization of the samples and experimental setup is therefore necessary to achieve AOS.
Chapter 6

Conclusion and Outlook

In this thesis, we have investigated the optical switching of ferromagnetic Co/Pt thin films. It was recently shown by Lambert et al. [10] that it is possible to all-optically switch these ferromagnetic structures. Their results challenge the current understanding of AOS, and the true mechanism behind it remains a matter of debate. With this work we try to gain a better understanding of AOS in ferromagnets. In this chapter we shall summarize the most important conclusions of our work towards achieving AOS. We shall finally provide an outlook for further research and applications.

6.1 Conclusions

The first step towards AOS was taken by simulating and measuring the ultrafast demagnetization after excitation with an ultrashort laser pulse. The ultrafast magnetization dynamics was modelled with the M3TM. This macrospin model is based on microscopic Hamiltonians and consists of three coupled differential equations for the electron, phonon and spin temperature, the latter of which determines the magnetization. A demagnetization curve was then be obtained by solving these equations, using a heat pulse to model the effect of the laser.

Experimentally the ultrafast demagnetization was measured using the TR-MOKE setup. A sub-ps demagnetization and subsequent slower remagnetization could be observed in correspondence with literature. The maximum demagnetization was found to increase with decreasing Co layer thickness, because of the decrease in Curie temperature. A demagnetization of 50% was found for the thinnest sample. However, due to the Gaussian spatial profile of the laser spot, it is likely that an (almost) full quenching of the magnetization is achieved at the center of the spot.

The laser pulse did not only cause a demagnetization, but the heating also decreases the coercivity. The reduction in coercivity was found to be around 70% for our specific laser
parameters, and independent of the Co thickness. This reduction was used to perform field assisted switching, where an applied field (in between the normal and the reduced coercivity) together with the linearly polarized laser pulse causes a switch. To quantify the effect of the applied field, a line was optically written and its width was measured. The switched domain size was thus measured as a function of the applied field. This could be compared to the predictions from the simulations, where the threshold power as a function of the applied field was determined, from which the experimental switched domain size could be derived. A good agreement between simulations and experiment was found.

The final step towards AOS was taken by investigating the effect of the helicity of the light. In the simulations this was done by including a magnetic field pulse. This field pulse is induced by the IFE and is dependent on the polarization of the light. Simulations with this field pulse gave similar results as those from literature where an atomistic model was used, showing the validity of this model to describe AOS. Little is known about the IFE in metals, and there is only a rough range of estimates for the strength and lifetime of the induced field pulse. But it was found that the parameters necessary for AOS are well within these estimates, showing that the IFE can be used to explain AOS. On a side note, direct transfer of angular momentum of the photons was shown to be too weak of an effect to play any role.

Having shown the possibility of (helicity dependent) AOS, a real AOS experiment was then simulated using an extended 2D version of the M3TM including the IFE. This way, we were able to reproduce the magnetization profiles reported by Lambert et al. for realistic values of all simulation parameters. At first try, only the general switching behaviour could be reproduced, but the thermal demagnetization that occurs in the center of the laser pulse could not. This was solved by including dipolar fields in the model, which supports the formation of a multi domain state. Hence, we can explain AOS in ferromagnets by invoking the IFE as the driving mechanism. To the best of our knowledge, such an explanation has not yet been provided.

Experimentally, the effect of the helicity of the light on the switched domain size was measured. A significant dependence between the helicity and the domain size was found. However, this dependence was not caused by the IFE, or even MCD, as it was independent of the direction of the starting magnetization. The dependence was found to be a measurement artefact caused by an imperfect QWP. We can thus neither confirm nor disprove that the IFE is present in the experiments. We can only conclude that if the IFE was present, the effect was rather weak.

AOS was not observed in any of these experiments; switching always required an externally applied field (although this field was much lower than the coercivity). This is not surprising for the experiments with the low power laser system, since the maximum demagnetization was only 80% and not fully quenched as required for AOS. With the new high power laser, a full demagnetization at the very center of the laser pulse might be possible. But no AOS was observed in the few experiments performed with this laser either, and at the highest laser powers sample damage occurred. The difference between this result and that
of Lambert et al. is most likely due to differences in the experimental parameters and samples.

In summary, we have shown with simulations that AOS in ferromagnets can be explained by the IFE. Experimentally, we were not able to achieve AOS. This demonstrates that it is a complex process that is not trivial to achieve, especially since little is known on the underlying mechanisms. More research is necessary to be able to fully understand AOS and use it for technological applications, which will be discussed in the following section.

6.2 Outlook

6.2.1 Modelling

With respect to the simulations, several improvements of the model are possible. A first extension would be to include domain wall motion in the 2D simulations. In the single pulse case with a time scale of several ps this will not make a difference, but for the multi-pulse sweeps nucleation and motion of domains can become rather important. Assuming a domain wall velocity of 1 m/s [57] and a repetition rate of 500 kHz, we obtain a displacement of 2 µm in the time between two pulses, which is indeed significant. We have already seen this effect of domain wall motion in the experiments in section 5.3.1 and 5.3.2. Since the motion is very much dependent on temperature, the domain wall velocity will be extremely high in the area heated by the laser pulse.

Other temperature dependencies could be added to the model as well. It has for example been suggested by Alebrand et al. [38] that the transfer of the helicity of the light to the spin system is more efficient at lower temperatures. Simulating their two-pulse experiments with such a temperature dependent helicity transfer could provide an extra insight in the interplay of heating and helicity.

The most rigorous extension of the M3TM would be to go from a scalar to a vectorial magnetization. So far the magnetization was considered to lie along the axis perpendicular to the thin film. In reality it can be slightly canted from this axis, giving rise to precessional dynamics described with the LLG equation. We have already discussed an atomistic model based on the LLG in section 4.3.2 [34]. Elements from this model could be incorporated into the M3TM to give a more complete description of AOS. The thickness dependent domain formation could then for example be simulated.

6.2.2 Experiments

It should be noted that the Spirit-NOPA laser system only became available at the end of this research project, only allowing us to do so pilot studies. More experiments should
therefore be performed with this high power laser system. The samples can be grown on other substrates with a higher heat conductivity (for example sapphire) to reduce the chance of damaging them. By careful tuning of the fluence it should then be possible to achieve AOS. The experiments of Lambert et al. with AOS under an opposing applied field, can be reproduced to check the strength of the driving mechanism. Once AOS is achieved, there are several possible experiments that will give an insight on this mechanism.

First of all it would be interesting to do time-resolved measurements of the switching process, instead of just static images of the result, and compare these to the results from the simulation. As discussed in section 3.4, these measurements require the magnetic state of the sample to be reset after every pulse. This can be done by using a laser with a lower repetition rate and a very ‘fast’ (low inductance) coil.

Secondly, two-pulse experiments could be performed, as was done by Alebrand et al. [38] for GdFeCo. These experiments will provide an insight in the interplay between helicity and heating. The lifetime of the helicity storage can also be determined this way.

It can furthermore be investigated specifically if the IFE plays a role in AOS in ferromagnets. The best way to prove the role of the IFE would be to do determine the switching threshold as a function of the laser wavelength. In section 2.4.2 this was already mentioned in relation to AOS in GdFeCo. A $P_{\text{threshold}} \propto \lambda^3$ dependence should be found if the IFE is indeed the responsible mechanism [39].

Another factor that could play a role and should therefore be explored is the magnetic moment in the Pt layers that is induced by the magnetic proximity effect. In general this induced moment is coupled to the magnetization of the Co layer, but at a fs time scale this coupling can diminish so that the two layers have different dynamics. This can be measured by doing element specific TR-MOKE measurements, for example by using XMCD or the QWP measurement technique introduced by Schellekens et al. [58].

Finally, the possibility of AOS in other ferromagnetic materials should be investigated. Up until now AOS has only been demonstrated in FePtC grains and the Co/Pt layers studied in this thesis. Experiments could also be carried out on for example Ni, which has a lower Curie temperature and should therefore in theory require less power to be switched. Determining which material parameters play a crucial role in the AOS process will provide a further insight into the mechanism behind it.

In this search for other materials it should be taken into account that a high demagnetization rate will increase the feasibility of AOS. In section 4.3.2 we have already discussed that for Co a faster demagnetization can be achieved by making Co/Pt multilayers. Such a strategy might also benefit other ferromagnetic materials, and other high spin-orbit coupling materials such as Tb and Dy could be used instead of Pt. Another way to enhance the demagnetization rate is to use the spin currents between two different layers (see [16]). Using these two strategies should make it easier to achieve AOS in other materials, which is especially important for future applications.
6.2 Outlook

6.2.3 Technological application

Even if the underlying mechanism behind AOS is unveiled, a lot of additional steps need to be taken before it can be implemented in memory devices. We shall discuss a few of the challenges that still need to be overcome.

One of the major challenges is the generation of the high power ultrashort laser pulses. Currently these pulses are generated by laser systems that are bulky, power-consuming, expensive, complex and not very robust against external disturbances. On the research side, some of the restrictions on the laser pulses might be loosened once the mechanism behind AOS is unveiled and other materials are used. For example, it has been shown by Steil et al. [40] that AOS is also possible with longer laser pulses up to 3 ps. Nevertheless, great improvements have to be made on the technological side to be able to integrate these lasers into a device.

To compete with current technologies, the size of the domain that is all-optically switched should be reduced to the current bit size in memory devices, which is around 20 nm. This is below the diffraction limit of the laser light which typically has a wavelength of 800 nm, so special techniques have to be used to achieve such a small size. First of all, the threshold behaviour of AOS can be exploited by using low laser fluences so that only the center of the pulse will cause a switch, effectively reducing the switched domain size. Secondly, samples can be microstructured so that the incoming pulses create complex interference patterns that effectively reduce the spot size [59, 60]. This has the added benefit of reducing the energy required for a switch. Finally, plasmonic antennas can be used to focus the laser beam beyond the diffraction limit [61]. This technology, which has received a lot of attention from industry in relation to HAMR, has been used by Liu et al. [4] in GdFeCo, and they achieved domain sizes down to 50 nm.

The final challenge is to integrate AOS into the hybrid magneto-photonic device introduced in section 1.2. Especially finding a suitable material of the racetrack is challenging, as it is has to both facilitate efficient AOS and have high domain wall velocities so that data can be quickly transported through the memory. This is the reason why it is crucial to understand AOS in Co/Pt, as it has been shown to allow very high domain wall velocities [62]. Another good candidate is the synthetic antiferromagnet as investigated in relation to AOS by Mangin et al. [7] (a Co/Ir multilayer). Structures like these have been shown to allow even higher domain wall velocities [63]. A lot of progress has been made in making a racetrack memory out of these materials, but here there are also still many technological challenges to overcome (see [64]).

We can conclude that although AOS is a promising new way of writing data, there are still many steps ahead of us. The mechanism behind it is not yet fully understood, especially in ferromagnets, which hinders technological application. Several technological improvements are necessary before AOS can be integrated into memory devices. But the fact remains that AOS has a very high potential, and time will learn if it can live up to this potential.
Bibliography


Acknowledgements

Here I would like to thank all the people who have helped and supported me throughout this project. I was especially blessed with the luxury of three supervisors!

First of all I want to thank Rosa, my daily supervisor. In the beginning you taught me the ins and outs of time resolved MOKE, and later on you gave me the freedom to find my own way, but always there to guide me in the right direction. Your enthusiasm for results of which I didn’t realize were any good helped me to stay motivated.

I also want to thank Mark, whom I have dubbed experimental supervisor as we spent quite some time together in the lab, trying to master that tricky setup. Especially your help with redesigning and rebuilding the setup with the new laser is much appreciated.

Furthermore I want to thank Bert, my supervising professor. You helped me stay on track with your guidance and suggestions, and your scientific enthusiasm was always a great motivator. Your tremendous amount of knowledge on both magnetism and optics helped me overcome many of the issues I encountered.

My thanks also goes out to the rest of the FNA group for all the (scientific) discussions at the coffee table but certainly also for all the fun activities such as futsal and the group outing. Special thanks goes out to the entire ‘fs team’ for the fruitful biweekly discussions, and for sharing the frustrations of the TR-MOKE setups and its components that kept breaking down. My office mates also deserve a special word of thanks for creating a nice work environment, and for keeping up with my rants against the computer whenever I did something stupid.

Finally I want to thank my friends and family for all their support and helping me to get my mind of things with Borrels, BBQ’s, All Terrain trainings, outdoor weekends and many more.
## Appendix A

### Acronyms

An overview of frequently used acronyms in this thesis is given below.

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>AOS</td>
<td>All-Optical Switching</td>
</tr>
<tr>
<td>AOHDS</td>
<td>All-Optical Helicity Dependent Switching</td>
</tr>
<tr>
<td>HAMR</td>
<td>Heat Assisted Magnetic Recording</td>
</tr>
<tr>
<td>HDD</td>
<td>Hard Disk Drive</td>
</tr>
<tr>
<td>IFE</td>
<td>Inverse Faraday Effect</td>
</tr>
<tr>
<td>LLG/LLB</td>
<td>Landau-Lifshitz-Gilbert (Bloch) equation</td>
</tr>
<tr>
<td>LP</td>
<td>Linearly Polarized</td>
</tr>
<tr>
<td>(M)3TM</td>
<td>(Microscopic) Three-Temperature Model</td>
</tr>
<tr>
<td>MCD</td>
<td>Magnetic Circular Dichroism</td>
</tr>
<tr>
<td>PEM</td>
<td>Photo-Elastic Modulator</td>
</tr>
<tr>
<td>PMA</td>
<td>Perpendicular Magnetic Anisotropy</td>
</tr>
<tr>
<td>QWP</td>
<td>Quarter Wave Plate</td>
</tr>
<tr>
<td>RCP/LCP</td>
<td>Right/Left handed Circularly Polarized</td>
</tr>
<tr>
<td>RE-TM</td>
<td>Rare-Earth Transition-Metal</td>
</tr>
<tr>
<td>SOC</td>
<td>Spin-Orbit Coupling</td>
</tr>
<tr>
<td>TD</td>
<td>Thermal Demagnetization</td>
</tr>
<tr>
<td>(TR)-MOKE</td>
<td>(Time-Resolved) Magneto Optical Kerr Effect</td>
</tr>
</tbody>
</table>
Appendix B

MOKE

In this section we shall derive the signal at the detector of a standard MOKE setup with PEM, as in Figure 3.3. We do this by describing (the polarization of) the laser beam using the Jones formalism \[65\].

After the first polarizer, which is oriented at 45° with respect to the horizontal the Jones vector reads

\[
\begin{pmatrix}
E_x \\
E_y
\end{pmatrix}
= \frac{1}{\sqrt{2}} \begin{pmatrix}
1 \\
1
\end{pmatrix}.
\]

(B.1)

The beam subsequently passes through the PEM, reflects of the sample and is passed through the analyser, which is oriented at 0°. The Jones matrices of these components are given by

\[
M = \begin{pmatrix}
1 & 0 \\
e^{-iA\cos(\omega t)} & 0
\end{pmatrix},
S = \sqrt{R} \begin{pmatrix}
1 & -\Theta \\
\Theta & 1
\end{pmatrix},
P = \begin{pmatrix}
1 & 0 \\
0 & 0
\end{pmatrix},
\]

(B.2)

where \(A\) is the amplitude and \(\omega\) the frequency of the polarization modulation by the PEM, \(R\) the reflection coefficient and \(\Theta = \theta + i\epsilon\) the complex Kerr rotation of the sample. The beam after the analyser is then described by

\[
\begin{pmatrix}
E_x \\
E_y
\end{pmatrix}
= \frac{1}{\sqrt{2}} PSM \begin{pmatrix}
1 \\
1
\end{pmatrix}.
\]

(B.3)

Using this expression we find for the intensity \(I = EE^*\) on the detector:

\[
I = R \left( \frac{1}{2} + \theta \cos[A\cos(\omega t)] + \epsilon \sin[A\cos(\omega t)] \right),
\]

(B.4)

where the terms in \(\theta^2\) and \(\epsilon^2\) are neglected since the Kerr rotation is very small. This can
be expanded in terms of the harmonics $\cos(n\omega t)$, of which the first three terms are:

$$I_{DC} = R \left[ \frac{1}{2} + \theta J_0(A) \right], \quad \text{(B.5)}$$

$$I_{1f} = \epsilon R J_1(A) \cos(\omega t), \quad \text{(B.6)}$$

$$I_{2f} = \theta R J_2(A) \cos(2\omega t), \quad \text{(B.7)}$$

where $J_n$ is the $n$-th order Bessel function of the first kind.

Either the Kerr rotation $\theta$ or the ellipticity $\epsilon$ can now be measured by using a lock-in set on the first and second harmonic of the PEM frequency (with $A$ chosen accordingly).
Appendix C

Laser spot size determination

The size of the probe (and also the pump) laser spot is determined using a knife-edge method. By moving towards the edge of the sample, less power is reflected as the laser spot partly hits the non reflective background. The spatial laser beam profile (in one dimension) can then be obtained by measuring the reflected power as a function of the distance from the edge. This process is shown in Figure C.1.

![Figure C.1: The reflected laser power of the sample as a function of the distance to the edge of the sample. The position of the laser spot on the sample is schematically shown for three points along the scan.](image)

The laser beam is expected to have a 2D Gaussian profile:

\[
I(x, y) = I_0 \exp \left( -\frac{(x^2 + y^2)}{2\sigma^2} \right), \quad (C.1)
\]
where $\sigma$ is the width of the beam and $I_0$ the peak intensity. To obtain the expected measured power as a function of the distance from the edge $x_0$, we integrate:

$$P(x_0) = \int_{-\infty}^{+\infty} \int_{-\infty}^{x_0} I(x,y) \, dx \, dy = \frac{I_0}{2\pi\sigma^2} \left[ \text{erf}\left(\frac{x_0}{\sigma}\right) + 1 \right] = P_0 \left[ 1 + \text{erf}\left(\frac{x_0}{\sigma}\right) \right]$$

(C.2)

The width of the beam $\sigma$ can now be obtained by fitting the measurements with the above equation, and generally ranges from 5 \(\mu\)m to 20 \(\mu\)m.
Appendix D

Measuring demagnetization

The finite size of the pump and probe spot causes the measured demagnetization to not be equal to the actual maximum demagnetization. We assume that the demagnetization depends linearly on the laser fluence, which is justified in the low fluence limit. We then obtain a Gaussian (de)magnetization profile after the pump pulse has heated the sample:

\[ M(x, y) = M_s \left( 1 - \Delta M_{\text{max}} \exp \left( -\frac{(x^2 + y^2)}{2\sigma_{\text{pump}}^2} \right) \right), \]  

(D.1)

so that there is a maximum demagnetization \( 0 < \Delta M_{\text{max}} < 1 \) in the center of the spot. If we assume perfect overlap between the pump and probe pulse, the observed demagnetization is now given by

\[ \Delta M_{\text{observed}} = \frac{\int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} M_s \left( 1 - \Delta M_{\text{max}} \exp \left( -\frac{(x^2 + y^2)}{2\sigma_{\text{pump}}^2} \right) \right) \exp \left( -\frac{(x^2 + y^2)}{2\sigma_{\text{probe}}^2} \right) dxdy}{\int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} M_s \exp \left( -\frac{(x^2 + y^2)}{2\sigma_{\text{probe}}^2} \right) dxdy}, \]  

(D.2)

Evaluation of this integral gives:

\[ \frac{\Delta M_{\text{observed}}}{\Delta M_{\text{max}}} = \frac{(\sigma_{\text{pump}})}{\sigma_{\text{probe}}}^2 \frac{1}{1 + (\sigma_{\text{pump}}/\sigma_{\text{probe}})^2} \]  

(D.3)

This function is plotted in Figure D.1. Usually the pump and probe spot are roughly the same size, for which we get that only half the maximum magnetization value is measured.
Figure D.1: The observed demagnetization as a function of the ratio between the pump and the probe spot size
Appendix E

Demagnetization traces with the Spirit-NOPA

A limited amount of experiments were performed with the high fluence Spirit-NOPA laser system. An example demagnetization trace is shown in Figure E.1. It can be seen that the signal is much noisier compared to that of the Tsunami laser, as explained in the main text.

Figure E.1: Demagnetization trace obtained with the Spirit-NOPA of a sample with a Co thickness of 0.6 nm.
Appendix F

Demagnetization traces with the QWP

It was attempted to measure demagnetization traces for different polarizations of the pump light, by rotating a QWP. The unprocessed data of these measurements are shown in Figure F.1. It can be seen that there are large difference in demagnetization amplitude (up to a factor two). These differences are too large to be attributed to the change in polarization of the light. The figure also shows that the QWP is not symmetric, since the the 0° and 180° traces are completely different. This all indicates that there are imperfections in the QWP, such as slight bends in the sheet of dichroic material. Due to these imperfections, the overlap between the pump and probe pulse slightly changes upon rotation of the QWP. This was confirmed by measuring the position of the pump spot with a diaphragm. The change in overlap causes significant changes in the demagnetization signal, that are unrelated to the actual demagnetization. Any change in the demagnetization due to the polarization are obscured by this overlap-related change in signal.
Figure F.1: Demagnetization traces obtained for different angles of the QWP.