Ultrafast Manipulation of Magnetic Order with Electrical Pulses

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Abstract

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During the last 30 years spintronics has been a very rapidly expanding field leading to lots of new interesting physics and applications. As with most technology-oriented fields, spintronics strives to control devices with very low energy consumption and high speed. The combination of spin and electronics inherent to spintronics directly tackles energy efficiency, due to the non-volatility of magnetism. However, speed of operation of spintronic devices is still rather limited (~nanoseconds), due to slow magnetization precessional frequencies.

Ultrafast magnetism (or opto-magnetism) is a relatively new field that has been very active in the last 20 years. The main idea is that intense femtosecond laser pulses can be used in order to manipulate the magnetization at very fast time-scales (~100 femtoseconds). However, the use of femtosecond lasers poses great application challenges such as diffraction limited optical spot sizes which hinders device density, and bulky and expensive integration of femtosecond lasers into devices.

In this thesis, our efforts to combine ultrafast magnetism and spintronics are presented. First, we show that the magnetization of ferrimagnetic GdFeCo films can be switched by picosecond electronic heat current pulses. This result shows that a non-thermal distribution of electrons directly excited by laser is not necessary for inducing ultrafast magnetic dynamics.

Then, we fabricate photoconductive switch devices on a LT-GaAs substrate, to generate picosecond electrical pulses. Intense electrical pulses with 10ps (FWHM) duration and peak current up to 3A can be generated and delivered into magnetic
films. Distinct magnetic dynamics in CoPt films are found between direct optical heating and electrical heating.

More importantly, by delivering picosecond electrical pulses into GdFeCo films, we are able to deterministically reverse the magnetization of GdFeCo within ~10ps. This is more than one order of magnitude faster than any other electrically controlled magnetic switching. Our results present a fundamentally new switching mechanism electrically, without requirement for any spin polarized current or spin transfer/orbit torques. Our discovery that ultrafast magnetization switching can be achieved with electrical pulses will launch a new frontier of spintronics science and herald a new generation of spintronic devices that operate at high speed with low energy consumption.

At last, to push ultrafast spintronics to practical use, ultrafast switching of a ferromagnetic film is desired. By exploiting the exchange interaction between GdFeCo and ferromagnetic Co/Pt layer, we achieved ultrafast (sub 10ps) switching of ferromagnetic film with a single laser pulse. This result will open up the possibility to control ferromagnetic materials at ultrafast time scale, critical for practical applications.
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Chapter 1

Introduction

1.1 Motivation: magnetism for devices
Magnetic order is a parameter that describes the arrangement of electron spins in solids (1). Depending on how electron spins are aligned, magnetic materials can be classified into ferromagnets, ferrimagnets, or antiferromagnets. In ferromagnets, spins are aligned in the same direction, while in ferrimagnets or antiferromagnets, spins are aligned in opposite directions, resulting in a reduced or cancelled magnetic moment. In magnetically anisotropic materials, the magnetic moment tends to align with an easy axis, which is an energy-favorable direction of spontaneous magnetization. Usually, magnetic anisotropy introduces a bi-stable magnetization state.

Magnetism is of great importance to information technology because it is widely used to represent binary data (e.g., 0 and 1) with the bi-stable directions of the magnetization vector \( \mathbf{M} \), such as in the ferromagnets used in hard drives to store data or magnetic random access memories (MRAM) (2). Additionally, spin logic devices have been proposed to replace charge-based logic devices due to better energy efficiency (3, 4).

Operation of all magnetic devices requires changes to magnetization states. Common methods are magnetic fields, currents, or light. Magnetization undergoes different dynamics depending on the manipulation method used. Ultimately, a fast yet practical and energy efficient way to manipulate the magnetic order is desired.

1.2 Magnetization dynamics in magnetic fields
The most common and straightforward way to manipulate magnetization of ferromagnetic material is to apply an external magnetic field (1). The electron spin \( \mathbf{S} \) with magnetic moment \( \mathbf{\mu} \) will experience torque \( \mathbf{\tau} \) in the external magnetic field \( \mathbf{B} \).

\[
\mathbf{\tau} = \mathbf{\mu} \times \mathbf{B} = -\gamma \mathbf{S} \times \mathbf{B} = \frac{d\mathbf{S}}{dt} \tag{1.1}
\]

Here, \( \gamma \) is the gyromagnetic ratio of the electrons. Equation 1.1 implies that electron spin precesses around the external magnetic field indefinitely. However, because of dissipation of energy and angular momentum, the system will eventually relax.
to its minimum energy state, in which the electron spin is aligned to the external magnetic field. This phenomenon is well described by the Landau-Lifshitz-Gilbert (LLG) equation (1),

$$\frac{dM}{dt} = -\gamma \mu_0 M \times H_{\text{eff}} + \frac{\alpha}{M} M \times \frac{dM}{dt}$$  \hspace{1cm} (1.2)

where $\mu_0$ is the permeability constant, and $\alpha$ is the phenomenological dimensionless damping constant describing how quickly $M$ relaxes to its equilibrium position, aligned with $H_{\text{eff}}$. Figure 1.1 shows a schematic of the magnetization precession and damping torque due to an external magnetic field ($H_{\text{eff}}$).

When using a magnetic field, the speed of magnetization reversal is limited by the magnetic precession frequency. The precession frequency and switching speed are inversely proportionate to the effective magnetic field amplitude. This sets a limit to the maximum attainable speed for magnetization manipulation, given by the maximum field amplitude that can be generated. The current record for reliable magnetic switching by an external magnetic field is approximately 200 ps using the Oersted field generated by a current pulse to switch the magnetization of an in-plane magnetized permalloy film (5). There are also efforts to use picosecond pulsed magnetic fields generated by high-intensity relativistic electron beams to switch the magnetization of CoCrPt films (6). With 28 GeV electron beams, a magnetic field of up to 10 T has been achieved. The magnetization of CoCrPt reverses after picosecond electron beam radiation; however, magnetization
reversal is nondeterministic and unreliable under such strong fields, making it impractical for device applications. Switching magnetization with external magnetic fields creates a significant obstacle to scaling magnetic devices into nanometer dimensions, due to the lack of a practical strategy to confine the magnetic field to nanometer scales.

1.3 Electrical control of magnetization

For the scalability of magnetic devices, electrical control of magnetization is desired. A well-known method to control magnetization electrically is to use spin-transfer-torque (STT) (7), for which a simple schematic is shown in Figure 1.2. Upon passing through the left ferromagnetic layer (i.e., red layer) the current becomes spin polarized due to the different mobilities of spin “up” and “down” carriers in the ferromagnet. This spin polarized current then flows into the right magnet (i.e., blue layer), and the angular momentum in the spin current is subsequently absorbed into the blue layer, resulting in torque in the localized magnetic moment.

\[
\frac{dM}{dt} = -\gamma \mu_0 M \times H_{eff} + \frac{\alpha}{M} M \times \frac{dM}{dt} + \gamma \mu_0 \frac{J_s}{M t} M \times (\sigma \times M) \tag{1.3}
\]

where \( J_s \) is the spin-current density, \( t \) is the thickness of the magnetic material absorbing the spin current, and \( \sigma \) is the unit vector along the polarization direction of the spin current. Equation 1.3 shows that spin currents tend to align \( M \) with \( \sigma \);
thus, STT can be used to electrically switch the orientation of the magnetization in a certain direction.

In addition to STT, spin-orbit torque (SOT) has been utilized to control magnetization (8). In heavy metals, such as Ta or Pt, large spin-orbit coupling causes a transverse spin current from a charge current, resulting in spin accumulation on the surface of the metal. The spins can be absorbed by the adjacent magnetic film and exert a torque, controlling the magnetization of the magnetic film (9, 10).

![Figure 1.3 Controlling magnetization of ferromagnets using SOT in a heavy metal Ta (10).](image)

Similar to STT devices, magnetization dynamics of SOT devices are also governed by the LLG equation, which means precessional switching is expected. Currently, the fastest STT/SOT devices switch magnetization around 100 ps (11–13).

1.4 Ultrafast laser-induced magnetization dynamics

Ultrafast pulsed lasers have provided access to magnetic dynamics on unprecedented timescales (14, 15). Measurements of the dynamic responses of magnetic material upon pulsed laser excitation were first conducted in 1996 by Beaurepaire et al. (14). After the excitation of a 60 femtosecond (fs), 2 eV laser pulse, the magnetization of Ni is quenched within several hundred femtoseconds, followed by a slow recovery. This behavior is explained phenomenologically by a three-temperature model; however, the microscopic origin is still under heated debate (16–20).
Ultrafast laser-induced demagnetization opens up a whole new femto-magnetism field in which femtosecond optical pulses are utilized to control magnetism on a sub-picosecond (ps) timescale. For example, with femtosecond laser excitation, an enormous pure spin current can be generated (21–25) that could potentially be used in spintronic devices. In addition, laser irradiation on magnetic materials can drive ultrafast magnetic phase transition (26). Recently, the ultrafast optical switching of magnetization in ferrimagnets has been demonstrated (27–30), opening up the possibility of writing a magnetic bit with a pure optical pulse.

Controlling magnetism with optical pulses allows for orders of magnitudes of faster operations than conventional methods, like using magnetic fields or an electrical current. However, the density of magnetic devices is limited by the diffraction limit of laser light, usually on the order of hundreds of nanometers. Also, the high cost and difficulty of integrating a femtosecond laser onto a chip limits the practical use of opto-magnetism phenomena.

In this thesis, we explore the possibility of replacing laser pulses with ultrafast electrical pulses to push the speed of spintronic devices to a higher level.
Chapter 2

Experimental techniques

2.1 Theoretical background of MOKE

To access the magnetic information of materials, we use optical methods that take advantage of the so-called magneto-optic Kerr effect (MOKE) \((31, 32)\). MOKE is very similar to the famous Faraday effect, in which the polarization of linearly polarized light is rotated when passing through a magnetic material. MOKE describes the polarization change of light reflected off a magnetic sample, which is proportional to the magnetization.

![Figure 2.1 A schematic of MOKE](image)

The magneto-optical property of magnetic materials is closely related to their dielectric tensor (Eq. 2.1). The diagonal terms do not depend on the magnetization, while the off-diagonal terms are proportional to the magnetization or the applied magnetic field. This can be understood with the conductivity tensor \(\sigma\). Electrons traveling in a magnetic medium or an external magnetic field will experience Lorentz force, which gives rise to off-diagonal terms in the conductivity tensor. Considering the linear relationship between dielectric and conductivity tensors, \(\epsilon = 1 + i \frac{4\pi}{\omega} \sigma\), the off-diagonal terms of \(\epsilon\) depend on magnetization as well:

\[
\epsilon = \begin{pmatrix}
\epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\
-\epsilon_{xy} & \epsilon_{yy} & \epsilon_{yz} \\
-\epsilon_{xz} & -\epsilon_{yz} & \epsilon_{zz}
\end{pmatrix}.
\] (2.1)

In an isotropic medium with \(M\) along the z direction, Equation 2.1 can be reduced to
\[ \epsilon = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\
-\epsilon_{xy} & \epsilon_{xx} & 0 \\
0 & 0 & \epsilon_{xx} \end{pmatrix}. \] (2.2)

The eigenmodes of Equation 2.2 can be calculated as follows:
\[ \begin{pmatrix} E_x \\ E_y \end{pmatrix} \pm = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm i \end{pmatrix}. \] (2.3)

where \( E_x \) and \( E_y \) are electric fields in the x and y directions.

These are left circularly polarized (LCP) and right circularly polarized (RCP) light with different eigenvalues \( \epsilon = \epsilon_{xx} \pm i \epsilon_{xy} \). This suggests that LCP and RCP light undergo a different phase change and/or absorption upon reflection off magnetic material. Thus, we can decompose linearly polarized (LP) light on an LCP and RCP basis and calculate the complex rotation \( \Phi \) to be
\[ \Phi = \theta + i \epsilon = \frac{\epsilon_{xy}}{(\epsilon_{xx} - 1)\sqrt{\epsilon_{xx}}} \] (2.4)

The variables \( \Phi, \theta, \) and \( \epsilon \) represent the complex Kerr angle, the Kerr rotation, and the Kerr ellipticity, respectively.

There are three different configurations of MOKE. They are polar MOKE, longitudinal MOKE, and transverse MOKE (33). Polar MOKE is used to measure the magnetization orthogonal to a sample surface. The light is usually close to normal incidence to maximize the signal. Most of the measurements in this thesis are conducted in polar MOKE geometry. Longitudinal MOKE is usually used to measure in-plane magnetization. It is sensitive to the in-plane magnetization component that lies in the plane of incidence. Transverse MOKE, in contrast, is used when the in-plane magnetization is perpendicular to the plane of incidence of the light. The magnetic contrast of transverse MOKE is much smaller than other two configurations, so it is not used in our experiments (33).

Figure 2.2 Different configurations of MOKE: a) polar MOKE, b) longitudinal MOKE, and c) transverse MOKE.
2.2 Measurement of MOKE

2.2.1 Balance detection

The Kerr rotation of magnetic materials is typically very small, on the order of milliradian for 10 nm GdFeCo film in polar MOKE geometry. In order to access both the static and dynamical magnetic properties, a high signal-to-noise ratio (SNR) MOKE is essential.

One way to measure such a small polarization rotation is to use balance detection. When linearly polarized probe light is reflected off the sample surface, it passes through a polarizing beam splitter. The two split beams will then be detected by the two reference ports of a balance photodetector. The balance detector then will output a signal that is amplified but proportional to the difference in the intensity of the two incoming beams. By rotating a half-wave plate in front of the polarizing beam splitter, the intensity of the two beams can be set to almost equal, such that the balanced signal is close to zero. Once the magnetization of the sample is changed (reversed to the opposite direction by an external field, for instance), the polarization of the reflected beam will rotate. Then the two beams are less balanced and the differential signal is proportional to the angle of the rotation, which results in the magnetization of the sample.

Overall, the balanced signal is small, so it is better to modulate the probe beam intensity and conduct AC measurement. To improve the SNR of the MOKE measurement, a high-frequency measurement is preferred to avoid laser noise at lower frequencies. The laser noise goes with $1/f$, so the higher the modulation frequency, the lower the laser noise. One simple way to modulate the signal is to use a mechanical chopper, which is typically on the order of several hundred hertz. To achieve a higher modulation frequency, we use an electro-optic modulator (EOM) (34). An EOM consists of a polarizer, an electro-optic crystal, and an analyzer. The electro-optic crystal is driven by a voltage signal up to the order of megahertz (MHz). Then the polarization of the beam is modulated at the frequency of the driving voltage. The analyzer will finally convert the polarization modulation back to intensity modulation. With MHz modulation, MOKE noise as low as $\mu$ rads can be obtained.

2.2.2 Polarization modulation

Aside from balance detection with intensity modulation, polarization modulation is another method of measuring the Kerr rotation and the Kerr ellipticity. And sometimes polarization modulation is more convenient than the other methods.
To modulate the polarization of the probe beam, we use a photo-elastic modulator (PEM). The PEM modulates the phase of one axis at a fixed frequency, which is around 50 kHz. The incoming probe beam is polarized at 45° to horizontal. The optical axis of the PEM is set parallel (0°) to horizontal, such that the phase of p-polarization is modulated periodically with respect to its s component. After the probe beam reflects off the sample, it passes through an analyzer at 0° and is then collected at a photodetector. The signal from the photodetector is then sent to a lock-in amplifier. By setting the reference frequency to either 1 f (50 kHz) or 2 f (100 kHz), the Kerr ellipticity or the Kerr rotation can be obtained.

Using the Jones matrix formalism, the incoming light can be expressed with a two-dimensional vector,
\[
\begin{pmatrix}
\frac{1}{\sqrt{2}} \\
n
\end{pmatrix},
\]
and the Jones matrices for the PEM, the Kerr rotation, the Kerr ellipticity, and the analyzer can be written as
\[
\begin{pmatrix}
1 & 0 \\
0 & e^{-i\beta\sin(\omega t)}
\end{pmatrix},
\begin{pmatrix}
\cos \theta & -\sin \theta \\
\sin \theta & \cos \theta
\end{pmatrix},
\begin{pmatrix}
1 & -i \varepsilon \\
i \varepsilon & 1
\end{pmatrix},
\begin{pmatrix}
0 & 0 \\
0 & 1
\end{pmatrix},
\]
where \(\beta, \omega, \theta,\) and \(\varepsilon\) are the modulation amplitudes of the PEM, the frequency of the PEM, the Kerr rotation, and the Kerr ellipticity, respectively.

By multiplying all the matrices, the intensity at the photodetector can be derived as follows:
\[
I(t) = I_0 \left(1 + 2\theta \cos(\beta \sin(\omega t)) - 2\varepsilon \sin(\beta \sin(\omega t))\right),
\]
given that
\[
\cos(\beta \sin(\omega t)) = J_0(\beta) + 2 \sum_{n=1}^{\infty} J_{2n}(\beta) \cos(2n\omega t),
\]
\[
sin(\beta \sin(\omega t)) = 2 \sum_{n=0}^{\infty} J_{2n+1}(\beta) \sin((2n + 1)\omega t) .
\]

(2.11 – 2.12)

\(J_n(x)\) is the Bessel function of the nth kind. We can expand Eq. 2.10 in the frequency domain:

\[
I(t) = I_0 (1 - 4\varepsilon * J_1(\beta) * \sin(\omega t) + 4\theta * J_2(\beta) * \cos(2\omega t) + \ldots) .
\]

(2.13)

If the right value is chosen for \(\beta\) to maximize \(J_1(\beta)\) and reference at \(\omega (2\omega)\), the Kerr rotation and the Kerr ellipticity can be measured.

Figure 2.3 A schematic for polarization modulation. The lock-in amplifier is referenced at the PEM frequency, which is typically 50 kHz.

2.3 Time-resolved MOKE

To assess the dynamical magnetization response, time-resolved MOKE (TR-MOKE) is used (35). To obtain temporal information, a pulsed ultrafast laser is exploited as the light source. In our experiments, two different pulsed laser sources are used. On some occasions, when low laser excitation is applied and a high SNR is needed, we typically use the oscillator. The Ti:Sapphire oscillator (Coherent Mantis) outputs laser pulses with a center wavelength of around 800 nm at 80 MHz with a pulse duration of around 100 fs for the full width at half maximum (FWHM). Due to the high repetition rate of the oscillator, we can modulate the laser beam at a relatively high frequency on the order of MHz. Considering the low laser noise of the oscillator, the MOKE noise can be largely suppressed. Figure 2.4 is a typical setup schematic with an oscillator as the light source. The laser pulse from the oscillator is split into two beams, one as a pump beam and the other as a probe beam. A
polarizing beam splitter is used to split the pump and probe beams because it is easy to allocate the power between the pump and probe beams by inserting a half-wave plate in front of the polarizing beam splitter.

The pump and probe beams will go through different optical paths until they overlap on the sample. The pump beam is sent onto a retroreflector on a linear delay stage (Thorlabs DDS 220). The reflecting beam is almost perfectly parallel to the incidental beam and is tuned to be parallel to the motion of the delay stage. In this way, the propagation direction remains static, while the delay stage moves back and forth. By changing the position of the delay stage, the relative time delay between the pump and probe beams arriving at the sample will be adjusted, which allows us to obtain temporal magnetization changes due to the pump beam excitation. The delay stage has a resolution of 0.1 um, which corresponds to a temporal resolution of 6.7 fs. The pump beam is eventually focused onto the sample by a plano-convex lens with various focal lengths depending on the spot size desired. The probe beam passes through an optical path with a similar length and is focused by a long working distance objective (Mitutoyo Corporation) onto the same spot on the sample. The overlap of the pump and probe spots is ensured by locating a beam profiler (WinCamD-XHR; DataRay, Inc.) at the sample position. Most of the time, polar MOKE is used. The reflected probe beam is collected by the same objective and directed to the detection path by a non-polarizing beam splitter. A half-wave plate and a Wollaston prism are used to achieve balance detection.

![Figure 2.4 The optical layout for TR-MOKE with balance detection. PBS: polarizing beam splitter; EOM: electro-optic modulator; BS: beam splitter; Obj: objective; HWP: half-wave plate; WP: Wollaston prism; PD: photodetector.](image)
Although oscillator TR-MOKE provides a high SNR, it also has some limitations. The per-pulse energy for the oscillator is quite limited, on the order of nanojoules (nJ). However, in some experiments, high fluence excitation is preferred. In these cases, we use an amplified pulse laser source. The oscillator is used to seed a Ti:Sapphire regenerative amplifier (Coherent RegA 9050). The laser pulse from the RegA has a center wavelength of around 810 nm with an adjustable repetition rate of 10 kHz to 300 kHz and a pulse width ranging from 60 fs to 20 ps (FWHM). Instead of nJ per pulse energy from the oscillator, the RegA is able to output microjoule (µJ) energy per pulse. Because of the relatively low repetition rate, it is not suitable for high-frequency modulation with EOM. Instead of balance detection, we therefore use the polarization modulation technique with the PEM, as described previously.

![Diagram of optical layout for TR-MOKE](image)

**Figure 2.5** The optical layout for TR-MOKE with polarization modulation. PBS: polarizing beam splitter; PEM: photo-elastic modulator; BS: beam splitter; Obj: objective; HWP: half-wave plate; PD: photodetector.

One final point that needs to be made here is that sometimes the time domain reflectance signal is mixed with the magnetic signal. To obtain a purely magnetic signal, all the TR-MOKE measurements are carried out under opposite external magnetic fields. By subtracting the two measured TR-MOKE curves, the reflectance signal is minimized.
2.4 MOKE microscope

In addition to TR-MOKE, another frequently used technique is MOKE microscopy. It is very convenient to obtain static magnetization information with a MOKE microscope. Throughout this thesis, the magnetic hysteresis curves are often measured with a MOKE microscope.

The principle behind the MOKE microscope is the same as the other MOKE measurements, and the main difference is the light source and the detector used. Instead of using a laser as a light source, here we use LED to illuminate the sample. A CCD camera is also utilized to replace the photodetectors in a laser MOKE setup. The LED light is first polarized with a high-quality polarizer (Glan-Thompson model; Newport Corporation). Then the light is collected by a lens and focused to the back aperture of the objective. In this way, we can make sure that as much light as possible illuminates the sample uniformly. Reflected light passes through an analyzer, which is close to the cross position of the previous polarizer, before it reaches the CCD camera to form an image. For better contrast, differential images are usually taken to visualize the magnetic domains. To improve the SNR, we use a relatively long exposure time for the camera, usually around 500 ms, without saturating the camera. MOKE microscopy does not provide as high an SNR as laser MOKE due to the lack of high-frequency modulation. However, the optical alignment in MOKE microscopy is not as critical as in laser MOKE, so it is frequently used to quickly check magnetic contrast. Also, a big advantage of MOKE microscopy is that it offers spatial magnetic information. Figure 2.7 shows a typically differential MOKE image for ferrimagnetic GdFeCo domains. The MOKE microscope is integrated with a TR-MOKE setup so that it is convenient to switch between the static and time-resolved measurements.

![Figure 2.6 A schematic of the MOKE microscope.](image)
2.5 Depth-sensitive MOKE

In some of our experiments, we study multilayer magnetic film structures. To understand how individual layer interacts with the rest of the stack, access to the magnetic information for each separate layer is critical. To achieve this, a depth-sensitive MOKE technique is used.

The depth-sensitive MOKE technique is based on polarization modulation with the PEM. Instead of sending 45° linearly polarized light directly through the PEM, a quarter-wave plate (QWP) is placed behind the 45° polarizer. The rest of the MOKE setup remains the same. By tuning the angle of the quarter-wave plate, the linear combination of the Kerr rotation and the Kerr ellipticity can be measured. With the proper angle on the quarter-wave plate, the Kerr rotation and ellipticity can cancel each other, thus killing the magnetic signal of that specific layer. We can explain the cancelation of the magnetic signal with the following formalism.

The Jones matrix for the quarter-wave plate can be written as

\[
\begin{pmatrix}
\cos \alpha & \sin \alpha \\
-sin \alpha & \cos \alpha
\end{pmatrix}
\begin{pmatrix}
1 & 0 \\
0 & i
\end{pmatrix}
\begin{pmatrix}
\cos \alpha & -\sin \alpha \\
\sin \alpha & \cos \alpha
\end{pmatrix},
\]

where \( \alpha \) is the angle of the fast axis of the quarter-wave plate with respect to the horizontal plane. By adding Eq. 2.14 into the previous calculation, we eventually get a 2\( \omega \) signal from the lock-in amplifier that is proportional to
\[ \theta \ast (\sin 2\alpha)^2 + \varepsilon \ast \cos 2\alpha. \tag{2.15} \]

There is always a specific \( \alpha \) that makes Eq. 2.15 equal zero, thus killing the MOKE signal from the specific layer. Because different layers have different Kerr rotations (\( \theta \)) and Kerr ellipticities (\( \varepsilon \)), a different QWP angle \( \alpha \) is required to eliminate the magnetic contrast from that specific layer. Since the total MOKE signal is additive for the layers, the annihilation of the MOKE signal of one layer provides a magnetic signal for the remaining layers.

As an example, Figure 2.8 shows the magnetic hysteresis curves measured at several different quarter-wave plate settings on the same GdFeCo/Pt/Co/Pt sample. The GdFeCo layer and the Co/Pt layer are decoupled and have different coercivities. As shown in the figure, only the GdFeCo layer is probed with a quarter-wave plate angle of 102°, and only the Co/Pt layer is probed with a quarter-wave plate angle of 120°. At other quarter-wave plate angles, the hysteresis curves contain a magnetic signal from both layers.

Figure 2.8 Hysteresis curves for the GdFeCo/Pt/Co/Pt multilayer structure with different QWP angles. The GdFeCo and Co/Pt layers have different coercivities (~500 oe for GdFeCo and 150 oe for Co/Pt).
Chapter 3

Generation and characterization of picosecond electrical pulses

3.1 Low-temperature (LT) GaAs

Low-temperature GaAs is commonly used in ultrafast opto-electronics due to the unique properties of this semiconductor material. LT-GaAs is usually a molecular beam epitaxially (MBE) grown on a GaAs substrate at a relatively low temperature (between 200°C and 400°C) (36, 37). Because it is grown at low temperatures, LT-GaAs contains excess arsenic clusters. These arsenic clusters form mid-gap defect levels. Once electron-hole pairs are excited by laser pulses, these defect levels will lead to fast nonradiative recombinations within the order of one picosecond (ps). Due to good carrier mobility, high dark resistivity, and a short carrier lifetime, LT-GaAs is an ideal material for photoconductive switches (38). When not excited by a laser pulse, LT-GaAs is semi-insulating with a resistivity as high as 10^8 ohm*cm. With laser pulse excitation, electron-hole pairs become conductive for a short period of time, resulting in the generation of picosecond electrical pulses.

![Figure 3.1 The band structure of LT-GaAs (39).](image)

Figure 3.1 The band structure of LT-GaAs (39).
3.2 External THz pulse emitter

To generate picosecond electrical pulses, we first tried external LT-GaAs-based emitters. An external THz emitter is commercially available from Protemics GmbH. A coplanar stripline (CPS) is patterned on a flexible polyethylene terephthalate (PET) cantilever. In the middle of a narrow CPS section, a small (200 x 200 um²) LT-GaAs is transferred before the deposition of the CPS, serving as an active photoconductive switch (PCS) region (Figure 3.3). The CPS line, with a width of 50 um and a gap of 30 um, extends to the tip end of the PET cantilever. The other end of the CPS line on the emitter is connected to an SMP connector. A Keithley 2400 SourceMeter can then be used to bias the CPS line via the SMP connector. Once a laser pulse excites the active PCS region, the THz pulse can be generated and guided onto the sample via the capacitive coupling between the CPS lines on the emitter and the sample (Figure 3.4).

Figure 3.2 The time domain thermal reflectance measurement of LT-GaAs, which reveals a carrier lifetime of 2 ps.
Figure 3.3 An optical image of the external THz pulse emitter.

Figure 3.4 A schematic of guiding the THz onto the sample. The CPS on the sample guides the THz pulse to a metallic magnetic film, which is integrated into the CPS.
To characterize the THz pulse generated by the external emitter, we use a similar external LT-GaAs-based THz pulse detector. The detector is also an LT-GaAs photoconductive switch with a 2 µm activation gap to increase the spatial resolution. While the THz pulse is launched with a pump laser pulse, a probe laser pulse hits the tip of the external detector, generating transient photocarriers, at a certain time delay. Also at a certain time delay, the THz pulse arrives at the detector when the photocarriers are excited. The electric field in the THz pulse will accelerate the photocarriers and contribute to a net charge current. The THz pulses are generated at a high repetition rate (typically 250 kHz for the amplifier and 80 MHz for the oscillator), so an average charge current can be measured with a current amplifier that is proportional to the THz field strength. To improve the SNR, we modulate the pump laser beam with a mechanical chopper at 100 Hz and use a lock-in amplifier to measure the signal from the current amplifier. By scanning the time delay between the pump and probe laser pulses, the temporal profile of the THz pulse on the transmission line is obtained. As an example, a THz pulse with a 1.1 ps duration (FWHM) is measured on a 200 nm aluminum CPS line on the quartz substrate with an external THz emitter (Figure 3.7).

Figure 3.5 A schematic of measuring the temporal profile of the THz current pulse.
3.3 On-chip photoconductive switch
Although the external THz emitter is able to provide short enough THz electrical pulses, the strength of the pulses is not sufficient to induce any measurable ultrafast demagnetization on the magnetic device.
There are two main reasons for this. First, the capacitive coupling between the emitter and the CPS on the device is not effective enough. This is partially caused by the finite air gap between the THz emitter and the CPS on the device, which results in a relatively small capacitance. Also, the impedance of the CPS is different between the THz emitter and the device because of the different substrate materials used. The impedance mismatch causes a reflection of the THz at the emitter–device interface, thereby lowering the coupling efficiency. The second reason for the lack of pulse strength is the attenuation of the THz pulse during propagation. The active photoconductive area of the THz emitter is 1 mm from the end of the emitter. Also, the THz pulse needs to travel another 1 mm or so to reach the magnetic device for convenient pump or probe measurement. During this transmission process, part of the THz pulse energy is dissipated into the substrate and CPS line through joule heating.

In order to reach the desired THz current pulse strength, we finally fabricated our own photoconductive switch on an epitaxially grown LT-GaAs substrate and integrated the magnetic device into the transmission line. The LT-GaAs wafer, which was purchased from Pam-Xiamen, consists of a one-micron GaAs layer grown at a low temperature through molecular beam epitaxy on a GaAs substrate. Once the electrons are excited by a laser pulse from the valence band to the conduction band, the reflectivity will change due to the change in the conductivity. Measuring the transient reflectivity with a pump-probe scheme will provide information on how fast the electron-hole pairs recombine in LT-GaAs (40–42). The transient reflectivity measurements of the LT-GaAs are shown in Figure 3.2 and indicate a carrier lifetime of ~2 ps in the LT-GaAs layer.

A schematic is shown in Figure 3.8 of the device fabrication. The procedure involves three photolithography or lift-off steps. First, we cover the whole substrate, with the exception of a small area where the photoconductive switch will be located, with an insulating layer of MgO in order to increase the resistance between the signal and the ground lines of the CPW. The ~100 nm of MgO is deposited via RF sputtering. The dimensions of the patterned region from which the MgO is lifted are 100 um x 60 um. After completing a second round of lithography, we deposit the magnetic section of the CPW center line using DC magnetron sputtering. After a final photolithography step, we deposit Ti/Au layers that will form the transmission lines and the photoconductive switch. The 250 nm Au transmission lines are deposited via e-beam evaporation. To promote adhesion, a 20 nm Ti layer is evaporated onto the sample first. The Au center line of the CPW overlaps with a
small section of the magnetic wire on both sides. As a result, a final small section of the magnetic wire becomes uncoated with Ti and Au.

![Diagram](image)

**Figure 3.8** The fabrication procedure for coplanar waveguide devices.

The setup to measure the THz current pulse generated on-chip is similar to the external emitter case. The only difference is that the pump beam illuminates the photoconductive switch on-chip. With an on-chip photoconductive switch, we are able to generate 5 ps (FWHM) electrical pulses. Using the average photocurrent measured by the Keithley SourceMeter, we can measure the total charge carried by each electrical pulse. Taking into account the temporal profile of the current pulse and assuming that the entire charge is contained in the transient current pulse, the amplitude of the current pulse can be calculated. In the CPW devices, a peak current of up to 1 A is achieved at an elevated bias (70 V).
Figure 3.9 A schematic of the experimental setup for measuring the temporal profile of the electrical pulse.

Figure 3.10 The temporal profile of the current pulse generated by the CPW device, as measured with a Protemics probe positioned between the photoconductive switch and the magnetic wire.
Chapter 4

Ultrafast demagnetization of ferromagnets by picosecond electrical pulses

4.1 Introduction

The pioneering observation of ultrafast demagnetization in ferromagnetic nickel following optical irradiation has led to the discovery of a broad range of extraordinary magnetic phenomena \(^{(19, 22, 23, 26, 27, 29, 43, 44)}\). Despite this broad array of discoveries, the microscopic mechanisms that enable the sub-picosecond quenching of the magnetization in magnetic metals following ultrafast heating are unclear \(^{(16–20)}\). One aspect of optically induced ultrafast demagnetization that remains under debate is whether the initially nonthermal distribution of electrons is an important driver of ultrafast magnetic phenomena \(^{(16, 18, 45, 46)}\). In the first hundred femtoseconds following laser irradiation, electrons are nonthermal, i.e. Fermi-Dirac statistics provides a poor description of the excitation energies \(^{(47)}\). Several studies have predicted the initially nonthermal distribution impacts ultrafast demagnetization because electronic scattering rates depend on both the average energy and total number of electronic excitations \(^{(46, 48)}\). The average energy and total number of excitations can also impact transport phenomena, which may be important in the ultrafast demagnetization in metal multilayers. However, the lifetimes of eV-scale electronic excitations are typically tens of femtoseconds \(^{(49)}\). Demagnetization typically occurs over hundreds of femtoseconds \(^{(50)}\). Therefore, most models assume that highly excited electronic states can be disregarded when modelling magnetization dynamics \(^{(18)}\) and treat the electron distribution as thermal on all time-scales.

Here we demonstrate that the initially nonthermal distribution of electrons strongly impacts optically induced ultrafast magnetization dynamics. We deposit roughly equal amounts of energy into the electrons of a magnetic film with either a 2.6 picosecond optical pulse or 4 picosecond electrical pulse. Optical heating deposits energy by exciting a few electrons ~ 1.5 eV above the Fermi-level. In contrast, electrical heating simultaneously excites many electrons to only a few meV above the Fermi level. As a result of the differences in the initial electron distribution, we observe significant differences in the magnetization dynamics induced by optical vs. electrical heating of a ferromagnet. We attribute the distinct
dynamics that arise from electrical heating to enhanced electron-phonon scattering rates that result in rapid electron/phonon thermalization.

4.2 Methods

4.2.1 Sample and experimental details
We excite electrical pulses on a coplanar waveguide structure (CPW) using photoconductive Auston switches (Figure 4.1). Different from CPS as described in chapter 3, CPW has a signal line in the center and two ground plane on both sides. Upon optical irradiation of the biased photoconductive switch with an 830 nm laser, a transient electrical pulse with a FWHM of 5.5 ps is generated and propagates along the CPW (Fig 4.2). The current profile \( I(t) \) is measured with Protemics THz detector. The energy carried by the electrical pulses, \( \int I^2(t)Z_0 \, dt \), ranges from 1 to 200 pJ for DC biases across the photoswitch between 10 and 80 V. The impedance \( Z_0 \) of the waveguide is \( \sim 60 \) ohms. Upon passing through the ferromagnetic wire, the electrical pulse deposits part of its energy via Joule heating, thereby inducing ultrafast demagnetization. Since the power scales with \( I^2 \), the 5.5 picosecond current pulse corresponds to a \( \sim 4 \) ps heat pulse.

![Figure 4.1](image-url)

Figure 4.1 a) Schematic of the electrical demagnetization experiments. The Auston switch is illuminated with a 1.5 nJ laser pulse while biased between 10 and 80 V. The magnetization of
the magnetic wire is monitored via TR-MOKE. b) Optical image of the Auston switch. c) Optical image of the CoPt section of the waveguide.

![Diagram](image)

Figure 4.2 a) Temporal profile of the current pulse generated by the photoswitch, as measured with a Protemics probe positioned between the photoswitch and the CoPt wire. b) Average current across the device with 160 mW of laser power irradiating the photoswitch. The filled circles correspond to measurements while the photoswitch was irradiated with 1.5 nJ laser pulses at a rate of 80 MHz (photocurrent). Open circles are current measurements on the device without laser irradiation (dark current). A rapid increase in dark current occurs as the bias voltage across the photoswitch approaches the breakdown voltage of the device, ~90V. c) Dependence of the average current on average laser power with a bias voltage of 30 V.

Optical ultrafast demagnetization experiments are typically performed with laser fluences between 0-10 J m\(^{-2}\) (23, 51, 52), corresponding to irradiation of the film with 0.1 to 1 nJ of energy across a 100 µm\(^2\) region. Our CPW device delivers similar energy densities with an electrical pulse to a ferromagnetic wire. At a distance of 0.5 mm from the photoconductive switch, the center line width of the CPW and gap distance between the center line and ground are tapered down from ~30 um to ~4 um over 0.6 mm. The ratio between the centerline width and gap distance is constant in order to keep the waveguide impedance constant at 60 ohms. In the narrowed region of the CPW, a 6 um long section of the center line is made out of a thin film of a ferromagnetic metal. We are able to deliver electrical pulses with
energies as high as ~200 pJ to a 25 µm² ferromagnetic thin film, i.e. we can deliver up to ~8 J m⁻² of electrical fluence. Only a fraction of the incident energy is absorbed via Joule heating, e.g. 10 to 30%. The amount of electrical energy absorbed depends on the resistivity and dimensions of the ferromagnetic wire.

In addition to the photocurrent across the device, a constant but small dark current flows across the device in the absence of laser illumination of the photoswitch, see Figure 4.2b. In the experiments we describe below, the dark current is less than 20 µA because the bias voltage is kept below 60 V. The heat-current on the magnetic device due to the dark current is less than 20 mW cm⁻² and has no impact on the experiments.

We characterize the magnetization response of the CoPt samples to heating via time-resolved measurements of the polar magneto-optical Kerr effect (TR-MOKE). We modulate the pump beam with an electro-optic modulator at 1 MHz and use lock-in detection to monitor small changes to the magneto-optic response of the sample. The duration of the probe laser pulse is 0.3 ps, much shorter than the 2.6 ps pump pulse. The optical pump and probe beams possess different pulse durations because of dispersion from the electro-optic modulator that the pump beam passes through. Optical pulse durations are determined with an optical autocorrelator (APE corp.). The 1/e radius of the pump beam focused on the sample is ~20 µm. The 1/e radius of the probe beam is ~ 1.5 µm. The spot-size is determined in two ways. First, we use the knife-edge method. We place a knife edge at the focal point of the laser beam. By scanning the knife edge with a translational stage, a power meter records the integrated power of the laser beam that is not blocked by the knife edge. We can then fit the measured power vs. knife edge position curve to find out the radius of the laser beam. Second, we use a CCD camera image of the beam profile. Both agree to within 10%.

The experimental setup includes an integrated microscope that uses bright field imaging to monitor the pump and probe beams on the sample surface. The vibration isolation provided by our optics table ensures sub-micron stability so that the spatial jitter experienced by the laser beams is much less than the micron scale features of the devices.

In addition to performing electrical demagnetization experiments, we perform optical demagnetization experiments by altering two things in our experimental setup. Instead of focusing the pump beam on the photoconductive Auston switch, we overlap the pump and probe beams on the sample. Additionally, the optical demagnetization experiments are not performed on the 5 µm x 5 µm section of the CoPt wire that we pass the electrical current through because the pump beam
radius of 20 µm is much larger than 5 µm. Instead, we move the pump and probe beam to a separate area of the sample where a large section of the Co/Pt multilayer film remains unpatterned.

We performed both optical and electrical ultrafast demagnetization experiments on two Co/Pt multilayers. The geometry of the first and second film are (3 nm Ta / 15 nm Pt / [0.7 nm Pt / 0.6 nm Co] x 8 / 5 nm Pt), and (1 nm Ta / 1 nm Pt / [0.7 nm Pt / 0.6 nm Co] x 8 / 1.7 nm Pt), respectively. Below, we refer to these as the Pt/CoPt and the CoPt sample, respectively.

4.2.2 Three temperature model
To describe how magnetization evolves upon laser/current pulse excitation, we adopt the phenomenological three temperature model. The three temperature model is based on the assumption of three thermal reservoirs, electrons at temperature $T_e$, phonons at temperature $T_p$, and spins at temperature $T_s$. We assume that rate of energy transfer between the spin degrees-of-freedom and phonons is negligible, which is consistent with theoretical calculations (53). Furthermore, we assume that because of the short life-times of spin-excitations in ferromagnetic metals that the diffusion of heat in the spin reservoir is negligible. Then, the three temperature model consists of three equations for each layer in the sample,

$$C_e \frac{dT_e}{dt} = g_{ep}(T_p - T_e) + g_{es}(T_s - T_e) + \Lambda_e \frac{d^2T_e}{dx^2} + q(t, z) \quad (4.1)$$

$$C_p \frac{dT_p}{dt} = g_{ep}(T_e - T_p) \quad (4.2)$$

$$C_s \frac{dT_s}{dt} = g_{es}(T_e - T_s) \quad (4.3)$$

Here, $C$ denotes specific heat, $g$ denotes energy transfer coefficient, $\Lambda$ denotes thermal conductivity, and the subscripts $e$, $p$, and $s$ refer to the thermodynamic reservoirs of electrons, phonons, and spins. The heat is transferred to the electron reservoir with a rate $q(t, z)$. We solve these equations numerically with a Crank-Nicholson method.

For the CoPt multilayer, we treat the entire stack as an effective medium. We set the electronic heat capacity to 0.15 MJ m$^{-3}$ K$^{-1}$ based on first principles calculations. We set the phonon heat capacity to 3.05 MJ m$^{-3}$ K$^{-1}$, a weighted average of the bulk values for Pt and Co. We set the spin heat capacity to 0.2 MJ m$^{-3}$ K$^{-1}$, the value of Nickel at room temperature which has a magnetization and Curie temperature that is comparable to the Co/Pt multilayers. The value of $g_{es}$ is set to $5 \times 10^{17}$ W m$^{-3}$ K$^{-1}$ based on measurements of FePt:Cu, which possess a similar Curie temperature.
The value of the electronic thermal conductivity is fixed using the Wiedemann Franz law and measurements of the electrical conductivity, $\rho \approx 5 \times 10^{-7} \Omega m$.

We treat the Pt/CoPt sample as a three-layer system: 5 nm Pt / 10.4 nm PtCo / 18 nm Pt. We set the thermal properties of the CoPt layer equal to those for the CoPt sample. We assumed a phonon and electrical interface conductance between layers of 300 MW m$^{-2}$ K$^{-1}$ and 10 GW m$^{-2}$ K$^{-1}$, both typical values for phonon-phonon and electron-electron interface conductances (54). We set the Pt electron/phonon coupling constant to $35 \times 10^{17}$ W m$^{-3}$ K$^{-1}$ based on Reference (23). Finally, we set the electronic thermal conductivity of Pt using the Wiedemann Franz law, and an estimate of the electrical resistivity of $\rho \approx 4 \times 10^{-7} \Omega m$ based on sheet resistance measurements.

The depth dependence of $q(t, z)$ for the optical experiments is calculated using a multilayer optical matrix method. The time-dependence of $q(t, z)$ is included in our three temperature model calculation. The depth dependence of the absorption for the optical demagnetization experiments is shown in Figure 4.3. For the electrical demagnetization experiments, we deposit the heat from the electrical pulse uniformly in the Co/Pt sample. Alternatively, in the Pt/CoPt sample, we assume the amplitude of the volumetric heating is $\sim 20\%$ higher in the surface and bottom Pt layer than the [Co/Pt] multilayer because the CoPt sample has an electrical resistivity $\sim 20\%$ higher than the Pt/CoPt sample.

![Figure 4.3](image)

Figure 4.3 Multilayer calculation of optical absorption through the depth of the two samples. For this calculation we used literature values for the optical constants: $n_{Co} = 2.5 + 4.8i$, $n_{Pt} = 2.85 + 4.96i$, $n_{Ta} = 3.43 + 3.66i$ (beta-phase), $n_{MgO} = 1.72$, and $n_{GaAs} = 3.68 + 0.09i$. 
4.2.3 Electrical absorption calculation

We determine the electrical energy absorbed in the magnetic wire by two steps. First, we calculate the attenuation of the electrical pulse when propagating on the CPW to the sample section. We take the Fourier transform of the electrical pulse (voltage) $V(t)$ in time domain to get the frequency domain spectrum $\tilde{V}(\omega)$. The energy spectral density is then proportional to $|\tilde{V}(\omega)|^2$. The voltage on the CPW for an individual frequency $\omega$ signal at a given position $x_0$ away from the photo-switch is given by

$$\tilde{V}(\omega, x_0) = \exp\left(-\int_0^{x_0} \gamma dx\right) \tilde{V}(\omega, 0)$$  \hspace{1cm} (4.4)

where $\gamma$ is the propagation constant defined as follows

$$\gamma = \sqrt{(R + i\omega L)(G + i\omega C)}$$  \hspace{1cm} (4.5)

$R$, $L$, $G$, $C$ are the resistance, inductance, conductance and capacitance per unit length for the CPW. $R$ is estimated to be $4 \times 10^4$ $\Omega/m$ and $3 \times 10^3$ $\Omega/m$ for 5 µm wide and 30 µm wide CPW regions. $G$ is estimated to be 0.14 S/m and 0.014 S/m for 5 µm wide and 30 µm wide CPW regions. $L$ and $C$ can be calculated with the equations given in Ref. (55). The energy attenuation for a single frequency $\omega$ is then given by

$$\alpha_1(\omega) = \exp\left(-2\text{Re}\left(\int_0^{x_0} \gamma dx\right)\right)$$  \hspace{1cm} (4.6)

As a second step, we calculate the absorption of the electrical pulse in the magnetic load. We use a multilayer absorption calculation, using the same method as in the optical absorption calculation shown in Figure 4.3. Here we assume that the electro-magnetic wave travels from the gold CPW into a film thickness given by the gap distance between Au section of the CPW (length of the magnetic wire), and then exits back into the gold CPW line. The length of the Pt/CoPt and CoPt wires are 5 and 7 µm, respectively. The effective complex refractive index is given by

$$n(\omega) = \left(\frac{\gamma c}{i\omega}\right)^*$$  \hspace{1cm} (4.7)

where $c$ is the speed of light. The difference of complex refractive indices of the magnetic CPW and gold CPW section comes from the difference in $R$. For the Pt/CoPt and CoPt CPW sections, $R$ is estimated to be $2.7 \times 10^7$ $\Omega/m$ instead of $4 \times 10^4$ $\Omega/m$ for the 5 µm wide gold section. The product of $\alpha_1(\omega)$ and $\alpha_2(\omega)$ for the Pt/CoPt and CoPt samples are shown in Figure 4.4.

Finally, the total absorption in the magnetic section of the CPW can be calculated as follows.
\[
\alpha = \frac{\int |\vec{V}(\omega)|^2 \alpha_1(\omega)\alpha_2(\omega) d\omega}{\int |\vec{V}(\omega)|^2 d\omega} \tag{4.8}
\]

We calculate the total absorption in the Pt/CoPt and CoPt sample to be ~11 and 29%.

Figure 4.4 Calculation of the percent energy absorbed by the Pt/CoPt and CoPt wires due to Joule heating as a function of frequency component of the electrical pulse, i.e. \( \alpha_1(\omega)\alpha_2(\omega) \).

4.3 Results: electrical versus optical demagnetization

Figures 4.5 and 4.6 show the response of the two samples to ultrafast heating of the electrons via optical (Figure 4.5) and electrical pulses (Figure 4.6). The absorbed optical fluence for the Pt/CoPt and CoPt samples is 0.2 J m\(^{-2}\) and 0.7 J m\(^{-2}\), respectively. We use a 50 V bias voltage in the electrical demagnetization experiments shown in Figure 4.6. All demagnetization curves are normalized by the demagnetization at 10 ps to facilitate comparisons.
Figure 4.5 Optically induced demagnetization of (a) Pt/CoPt and (b) CoPt samples. The markers are normalized TR-MOKE measurements of the demagnetization of the CoPt ferromagnetic layer following optical absorption of ~0.2 J m$^{-2}$. The shaded region represents the power vs. time of the irradiating laser. The solid lines are a three-temperature model prediction for the magnetization dynamics with an electron phonon energy transfer coefficient predicted by scattering theory, $g_{ep} = 15 \times 10^{17} W m^{-3} K^{-1}$. To explain the demagnetization data on picosecond scales, the net energy-transfer coefficient must be reduced from the theoretical value to $g_{ep} = 7 \times 10^{17} W m^{-3} K^{-1}$, see dashed lines.
Figure 4.6 Electrically induced demagnetization in (a) Pt/CoPt and (b) CoPt samples. Markers are normalized TR-MOKE measurements of the magnetization of the samples after heating by an electrical pulse with ~0.5 A peak amplitude. The shaded region represents the power profile of the electrical pulse, as measured by the Protemics probe. The heating time-scales are slightly different for the two samples due to differences in the Auston switch devices. The solid lines are three-temperature model predictions for the demagnetization with the theoretically calculated value of \( g_{ep} = 15 \times 10^{17} W m^{-3} K^{-1} \). For comparison with the optical experiments, the dashed lines show predictions with a reduced electron-phonon energy transfer coefficient of \( g_{ep} = 7 \times 10^{17} W m^{-3} K^{-1} \). A higher rate of energy transfer between electrons and phonons in the electrical heating experiments explains why there is no recovery of the magnetization in the picoseconds following electrical heating.

At large heating fluences, nonlinear effects are known to impact the magnetization dynamics. An example of such a nonlinear effect is the increase in the demagnetization time-scale that occurs if the fluence is sufficient for the per-pulse temperature rise to approach the Curie temperature \((18, 50, 51)\). Here, we intentionally use small fluences in order to exclude nonlinear effects from the dynamics and simplify analysis. The peak demagnetization in the optical and electrical experiments shown in Figures 4.5 and 4.6 is below 2%, and the peak per pulse temperature rise is less than 20 K. The peak demagnetization is calculated by normalizing the maximum transient Kerr rotation in our pump/probe experiments by the static Kerr rotation. The maximum temperature rise is calculated with a thermal model \((51)\), which is described in detail in the previous section. We experimentally verify that our experiments are in a linear regime by performing optical and electrical experiments across a range of absorbed fluences between 0
and 0.5 J m$^{-2}$. The shape of the dynamics does not depend on the bias voltage in the electrical experiments, or the laser energy in the optical experiments, as shown in Figure 4.7.

Figure 4.7 Peak current (a) and incident laser fluence (b) dependence of the demagnetization of the CoPt sample. The shape of the demagnetization is independent of the peak current (c) and incident laser fluence (d) in both the electrical and optical experiments.

For both electrical and optical heating, the magnetization of the Co/Pt multilayer decreases rapidly. However, clear differences exist for the two types of heating. We attribute the differences to differences in the initial distribution of excited electrons.

Optical irradiation excites electrons between 0 and 1.55 eV above the Fermi level and the initial distribution is nonthermal, i.e. can’t be described with Fermi-Dirac
statistics \((56, 57)\). In contrast to optical heating, when electrons are electrically heated their energies only increase a few meV. The largest longitudinal electric field that occurs in the ferromagnetic wire during our experiments is \(j_{\text{max}}/\sigma \approx 4\,MVm^{-1}\), where \(j_{\text{max}}\) is the maximum current density and \(\sigma\) is the electrical conductivity of the ferromagnet. Assuming a scattering time of \(\sim 30\,\text{fs}\), a value typical for transition metals \((58)\), the average increase in kinetic energy of an electron due to acceleration in the electric field prior to scattering is \(\Delta E \approx \frac{(eE\tau)^2}{2m_e} \approx 1\,\text{meV}\), where \(m_e\) is the mass of an electron. Because \(\Delta E \ll k_B T\), the distribution of excited electrons is thermal. Therefore, by comparing the response of CoPt and Pt/CoPt to optical vs. electrical heating, we are directly probing the impact of an initially nonthermal vs. thermal electron distribution on the magnetization dynamics.

Demagnetization of the CoPt following optical heating, as shown in Figure 4.5, displays “type I” dynamics \((18)\). The sample demagnetizes during laser irradiation, followed by a smaller increase in the magnetization as the electrons and phonons thermalize. Our “type I” categorization agrees with prior studies of Co/Pt \((59)\), whose large spin-orbit coupling is credited with abnormally strong coupling between electronic and spin degrees-of-freedom. Magnetization dynamics without a recovery in the magnetization in the picoseconds following irradiation are “type II” dynamics \((18)\). (The category of “type II” dynamics also includes observations of demagnetization on multiple time scales, as is observed for Gd.) In contrast to the “type I” dynamics displayed following optical irradiation, the magnetization of neither CoPt or Pt/CoPt display a significant recovery in the picoseconds following electrical heating.

### 4.4 Analysis

After energy is added to the electronic system via optical or electrical pulses, the electrons transfer energy to other degrees of freedom via electron-phonon scattering, and scattering between the electrons and spin degrees-of-freedom \((25)\). We model the redistribution of energy from optically excited electrons to phonons and spin degrees-of-freedom with a phenomenological three temperature model \((14, 51)\) (as described in previous section).

While the three temperature model as described in Equations 4.1-4.3 has many parameters, the model predictions for picosecond dynamics are only sensitive to \(q(t,z), g_{ep}, C_p\). The values of the other parameters are largely irrelevant. We
quantitatively demonstrate this in Figure 4.8 with sensitivity curves. We define the sensitivity of the model predictions to parameter $\alpha$ as

$$S_\alpha = \frac{\Delta(dM)/dM}{\Delta\alpha/\alpha}$$  \hfill (4.9)$$

As seen in Figure 4.8, the model predictions are dominated by the parameters $C_p$ and $g_{ep}$ at short time-delays, with $S_{g_{ep}} \approx -1.5$ and $S_{C_p} \approx -1.5$ at 1 ps; in other words, a 10\% increase in $C_p$ or $g_{ep}$ produces a 15\% change in $\Delta(dM)/dM$ at $t \sim 1$ ps.

The only parameters other than $C_p$, $g_{ep}$, or the pulse duration $\tau_0$ that the model has non-negligible sensitivity to are $C_s$, $g_{es}$, and the film thickness $h$. The model predictions are more than 4x as sensitive to $C_p/g_{ep}$ than to $C_s/g_{es}$. Furthermore, the sensitivity to $C_s$ and $g_{es}$ is zero at $t \approx 1$ ps when the demagnetization is a maximum because the primary impact of $C_s$ and $g_{es}$ is to shift the peak demagnetization by introducing a lag between the maximum in $q(t)$ and the maximum in $dM$. The film thickness is accurately determined with via X-ray reflectivity measurements, and is therefore well known and doesn’t add uncertainty to our analysis. Therefore, the sensitivity curves in Figure 4.8 provide quantitative support for our qualitative argument in the following analysis that the amplitude of the initial demagnetization is only determined by $C_p$, $g_{ep}$ and $\tau_0$. 
Figure 4.8 Thermal model sensitivities for Pt/CoPt when undergoing optical demagnetization. (a) Sensitivity of the demagnetization to the pulse duration $\tau_0$ and energy transfer coefficients. (b) Sensitivity of the model to the electron, spin, and phonon heat-capacities. (c) Sensitivity to the electronic thermal conductivity, electron interface conductance between the Pt and CoPt layers, and the film thickness $h$.

The heating profile $q(t)$ ($\tau_0$) for the optical experiments is accurately measured using an optical autocorrelator (APE corp.) that has fs resolution. The heating profile $q(t)$ ($\tau_0$) for the electrical experiments is accurately measured by measuring the electric field vs. time of the current pulse with a Protemics THz detector. The Protemics detector possess sub-ps resolution. The phonon heat-capacity is set to 2.85 and 3.1 MJ m$^{-3}$ K$^{-1}$ for the Pt and [Co/Pt] multilayers based on literature values, respectively. So $g_{ep}$ is the only parameter left to determine the shape of the demagnetization curve.
We fix $g_{ep}$ in our thermal model using the scattering theory original derived by Allen (60). According to scattering theory, $g_{ep} \approx \pi \hbar k_B D_f \lambda \langle \omega^2 \rangle$, where $\hbar$ is the reduced Planck’s constant, $k_B$ is Boltzmann’s constant, $D_f$ is the density of states at the Fermi level, $\lambda$ is the electron-phonon coupling constant in the Eliashberg generalization of BCS theory, and $\langle \omega^2 \rangle$ is the second frequency moment of the phonons. We approximate $\langle \omega^2 \rangle$ by assuming a Debye density of states, $\langle \omega^2 \rangle \approx 0.6 \cdot k_B^2 \Theta_D^2 / \hbar^2$, where $\Theta_D$ is the Debye temperature. For Pt, $D_f \sim 9 \times 10^{47} \text{ J}^{-1} \text{ m}^{-3}$ (61), $\lambda = 0.66$ (62), and $\Theta_D \approx 240 K$. Therefore, theory predicts $g_{ep} \approx 1.5 \times 10^{18} \text{ W m}^{-3} \text{ K}^{-1}$. We use the theory prediction for $g_{ep}$ of Pt only because no experimental measurement of $\lambda$ exists for Co. First-principles calculations suggest $g_{ep}$ is higher for Co than Pt (63, 64), therefore $1.5 \times 10^{18} \text{ W m}^{-3} \text{ K}^{-1}$ can be viewed as a theoretical estimate of the lower limit for $g_{ep}$ in our layers.

Three temperature model predictions are in excellent agreement with the electrically induced demagnetization data, see Figure 4.6. However, the three-temperature model predictions are in poor agreement with the optical experiments, see Figure 4.5. In order to achieve good agreement between the thermal model and the optical demagnetization data for both the CoPt and Pt/CoPt sample, we must reduce the value of the electron-phonon coupling constant by half to $7 \times 10^{17} \text{ W m}^{-3} \text{ K}^{-1}$. The factor of two difference in the peak electrical vs. electrical demagnetization cannot be explained by the differences in optical vs. electrical pulse duration. The thermal model predicts that a 35% change in pulse duration from 4 to 2.6 ps will only alter the peak demagnetization by ~10%. Instead, we posit that the disagreement between the optical demagnetization data and the three-temperature model is because the three-temperature model does not account for the initially nonthermal distribution of excited electrons in the optical experiments (57). We discuss this further in next section.
Figure 4.9 Demagnetization versus amplitude of the current pulse. Markers are TR-MOKE measurements of the demagnetization 10 ps after the electrical pulse heats the Pt/CoPt (red) and CoPt (blue). The dashed line are the predictions of our thermal model, using the value of \((1/M) dM/dT\) derived from optical demagnetization experiments and our calculation of the energy absorbed by the electrons via joule heating.

In the above analysis, we restricted our comparison between the demagnetization data and thermal model predictions to the shape of the demagnetization. Now, we compare the magnitude of the demagnetization at 10 ps delay time to the predictions of our thermal model. In Figure 4.9, we plot the demagnetization as a function of the peak current of the pulse. Uncertainty in our electrical absorption calculations is ~30% due to uncertainties in the film resistivity and dimensions. In order to make predictions with the thermal model for the demagnetization, we must have knowledge of the temperature dependence for the magnetization. We set \((1/M) dM/dT \approx 10^{-3} K^{-1}\) by comparing the optical demagnetization at 10 ps to the per pulse temperature rise, \(hC_{tot}/F\), where \(h\) is the metal film thickness, \(C_{tot}\) is the total volumetric heat capacity, and \(F\) is the absorbed fluence. The agreement between data and model predictions supports our conclusion that the observed ultrafast magnetic response of both samples is due to electrical heating. Finally, let us consider the influence on the ultrafast dynamics of the Oersted magnetic field that accompanies the electric field pulse. The magnetic field will point in opposite directions at the top and bottom of the magnetic film, causing the
magnetic field amplitude to vary across the metal layer. Using Ampere’s law, we estimate the variation of the field across the Co/Pt multilayer in both samples for a peak current of 0.5 A. In the Pt/CoPt sample, the amplitude varies from ~40 mT at the surface of the [Co/Pt] multilayer to -5 mT at the bottom, corresponding to an average field of 20 mT. In the CoPt sample, we estimate the field will vary from ~40 mT at the surface to -40 mT at the bottom of the Co/Pt multilayer, corresponding to an average field of ~0 mT. Calculating the ultrafast magnetic response of the Co/Pt to a non-uniform magnetic field would require atomistic spin simulations and is beyond the scope of the current work. However, we can rule out magnetic field induced dynamics as a significant contributor to the MOKE signal in our experiments by considering how the Co/Pt multilayer would respond to uniform magnetic fields of comparable magnitude. The Landau Lifshitz Gilbert equation predicts only a 0.1 and 0.025% reduction of the z-component of the Co/Pt magnetization as a result of a 5.5 picosecond duration external magnetic field with an amplitude of ~40 or 20 mT, respectively. The initial reduction occurs on the time-scale of the magnetic pulse, and then processes back to equilibrium on a time-scale of hundreds of picoseconds. In this LLG calculation, we assume an anisotropy of 0.26T, consistent with literature values for similarly prepared Co/Pt multilayers.

4.5 Discussion and conclusion
Photoemission experiments suggest the nonthermal electron distribution that is initially excited by an optical pulse persists for tens to hundreds of femtoseconds in transition metals such as Al (57), Au (56), Ni (47), and Fe (65). While an electron-electron equilibration time of 10 fs < \( \tau_{ee} \) < 100 fs is much shorter than the picosecond time-scales of our heat pulses, that does not imply a nonthermal distribution will have no effect. The relevant comparison is not between the time-scale for heating and \( \tau_{ee} \). Instead, the relevant comparison is between \( \tau_{ee} \) and \( \tau_{es} \) or between \( \tau_{ee} \) and \( \tau_{ep} \). Unless \( \tau_{ee}/\tau_{es} << 1 \) or \( \tau_{ee}/\tau_{ep} << 1 \), a significant fraction of the spin and vibrational degrees-of-freedom in the optical demagnetization experiments occurs while the electrons are nonthermal. An implication of the small fluences we use in our experiment is the heat induced dynamics are linear, i.e. superposition applies. Therefore, the picosecond heating in our experiment can be viewed as the summation of dynamics caused by a sequence shorter optical pulses (66). If the rate of energy exchange between electrons, spins, and phonons is sensitive to the initially excited distribution of electrons, this sensitivity remains regardless of the duration of the heat pulse.
The rate that energy is exchanged between electrons and phonons is strongly altered while the electrons are nonthermal because the scattering rate between the electrons and phonons is proportional to the number of electronic excitations \( (57) \). The total number of electrons excited above the Fermi level increases by orders of magnitude after the absorbed energy is redistributed from a nonthermal to a thermal distribution via electron-electron scattering. Therefore, a nonthermal distribution of excited electrons will take hundreds of femtoseconds longer to equilibrate with the lattice than if the initial distribution is thermal. This hypothesis has previously been supported indirectly by comparing the value of \( g_{ep} \) derived from pump/probe measurements to theory \( (57, 67) \), but has never been directly validated by comparing energy transfer rates for thermal vs. nonthermal electron distributions. The value of \( g_{ep} \) derived by fitting pump/probe measurements with a thermal model are often lower than theoretical predictions, presumably in order to compensate for the lower electron/phonon scattering rate while the electron distribution is nonthermal \( (57) \).

In addition to influencing the rate of energy transfer to phonons, there are a number of ways for a nonthermal distribution to influence the demagnetization dynamics. For example, Elliot-Yafet scattering is thought to play a central role in ultrafast demagnetization \( (18) \) and depends on the total electron-phonon scattering rate. The scattering rate between electrons and spin-excitations \( (25) \), e.g. magnons, may also depend on the number of excited electrons. The high average energy of excitations in a nonthermal distribution may allow the generation of nonthermal spin excitations, e.g. Stoner excitations with sub-eV energies \( (47) \). Finally, the rate that electrons thermalize with the lattice will indirectly impact the magnetization dynamics. The rate of energy transfer to the spin degrees-of-freedom depends on how long the electrons remain hot \( (51) \). A faster exchange of energy between electrons and phonons favors slower demagnetization \( (51) \).

A nonthermal distribution of excited electrons can also impact how energy and angular momentum are transported, e.g. allow for superdiffusive spin and heat currents \( (19, 21) \). Therefore, in addition to an altered electron-phonon interaction, it is also possible that the significant differences we observe between electrical and optical demagnetization are partially due to superdiffusive spin transport. We note that superdiffusion and changes to e-p scattering rates are related phenomena, as electron-phonon scattering rates are an important component of super diffusive transport theory \( (68) \). One motivator for the geometry of our two samples is to investigate the impact of superdiffusion on our results. The Co/Pt multilayer in the CoPt/Pt sample is sandwiched between Pt layers that are 5 and 15 nm thick,
comparable to the spin diffusion length in Pt of ~ 8 nm. In contrast, the Co/Pt multilayer in the CoPt sample is sandwiched between only 1 and 1.7 nm of Pt. Therefore, the CoPt/Pt sample possesses a significant Pt reservoir for superdiffusive spins to be transported into, while the CoPt sample does not. The differences between optical and electrical demagnetization are similar for both samples. Therefore, we cannot conclude from the current experiments that superdiffusion is an important contributor for the differences in optical and electrical demagnetization. One possible explanation for the similar data for both samples is that the hot electron velocity relaxation length in Pt is much shorter than 8 nm. Recent observations of THz emission are consistent with a hot electron length of only ~1 nm (69).

Several prior studies have demonstrated that indirect optical excitation can induce ultrafast demagnetization. For example, Eschenlohr et al. reported differences in dynamics that occur following optical excitation of a Au/Ni vs. Ni sample in order to examine how nonthermal electron transport impacts magnetization dynamics (46). Similar experiments have been performed by Vodungbo et al. with Al (70) and Bergeard et al. with Cu (71). In these types of experiments, the average energy, spin-polarization, and number of excited electrons is altered depending on whether the energy is directly absorbed by the ferromagnet, or indirectly delivered via hot electrons from an adjacent metal layer, e.g. Au (46), Al (70), or Cu (71). However, significant controversies remain concerning the interpretation of these experiments because they require modeling of how the initially excited distribution of electrons evolve in time and space after optical irradiation. Eschenlohr et al. use scattering rates from first principles calculations to estimate the distribution and transport of excitations into the ferromagnetic layer from an adjacent film, however their interpretation remains controversial (54, 72). Alternatively, Bergeard et al. suggest hot electron transport between metal layers is ballistic (71). Other studies report results that are consistent with thermal diffusion (22, 23, 54).

The electrical vs. optical heating approach we utilize here is a more direct method for testing the impact that the initially excited electron distribution has on the magnetization dynamics because no predictions or assumptions are necessary for how an excited electron distribution evolves in time and space.

In conclusion, we observe rapid demagnetization in Co/Pt wires due to picosecond electrical heating. We observe large differences in the demagnetization rates of Co/Pt for optical vs. electrical heating. We attribute the large differences to the initially nonthermal vs. thermal distributions of excited electrons. The rate of scattering processes responsible for transferring energy from the electrons to the
lattice degrees of freedom is strongly affected by the number and average energy of excited electrons. Our experimental results will require a reexamination of the belief that the physics of optically induced demagnetization is well described by assuming the electron system is thermalized (18).
Chapter 5

All optical switching of ferrimagnetic GdFeCo

5.1 Introduction

Ultrafast optical excitation of magnetic materials causes distinctive dynamics of great interest for applications (73–75) and fundamental science (14, 76, 77). In particular, short pulse laser irradiation of a magnetic thin film can reverse the direction of the magnetic moment, even in the absence of an external magnetic field, a phenomenon known as all optical switching (AOS) (27, 28, 76, 78, 79). Many AOS studies have only observed deterministic switching if the laser pulse irradiating the sample is circularly polarized (27, 78, 79). However, in ferrimagnetic GdFeCo films, AOS is observed with linear polarized light and has been described as an ultrafast thermal effect (28, 76).

Despite intense study, the mechanisms of AOS remain unclear due to the rich physics that are found after a sub-100 femtosecond pulsed laser excitation. In the first hundred femtoseconds, highly non-equilibrium phenomena such as non-thermal carrier excitation (80, 81) and super-diffusive spin-currents (19) may take place. In the next few hundred femtoseconds, electrons become thermalized with each other resulting in a high electronic temperature $T_e$, but remain out of thermal equilibrium with the lattice and spin degrees of freedom (14). In addition to these nonequilibrium phenomena, the strong dependence of equilibrium magnetic properties on temperature could also play a central role in AOS (73, 82), as it does in heat assisted magnetic recording (HAMR) technology (73). Finally, magneto-optical phenomena such as the inverse Faraday effect (83) or magnetic circular dichroism (MCD) (84) complete the wide range of coexisting mechanisms that may play a role in AOS, making it a fascinating but challenging problem to understand.

The energy absorbed by the metal film, and the resulting transient temperature response, are known to play a central role in ultrafast demagnetization of single element ferromagnets (14, 50, 51). However, due to the large number of mechanisms that may contribute to AOS, it has been difficult to determine the primary role of energy and temperature during AOS. Temperature rise can directly or indirectly facilitate magnetization switching in a number of ways. For example, in HAMR, the lattice temperature $T_p$ of the system is heated close to the critical Curie temperature $T_C$ to reduce the anisotropy before an applied field favors a
particular direction for the magnetization upon cooling (73). In contrast, AOS models for ferrimagnets (28, 64, 76, 85–89) do not require the lattice temperature of the film to approach the Curie temperature. Instead, these models rely on transient electron temperatures that are out of equilibrium with the lattice to induce the dynamics of the Gd and Fe magnetic sublattices (76).

Despite the centrality of temperature to prevailing theories for AOS, the energy required for switching, and the resulting temperature response of the electrons and phonons remains unclear. This is largely related to uncertainties in the minimum absorbed fluence required for switching (i.e. critical fluence $F_C$) and unknown thermal parameters. Peak temperatures and subsequent cooling are determined by $F_C$, the electron phonon coupling parameter $g_{ep}$, and the electronic heat capacity $C_e$. $C_e$ and $g_{ep}$ are generally set by assuming typical values for transition metals. However, reported values for $g_{ep}$ for transition metals vary by an order of magnitude (64, 90).

Indeed, reported $F_C$ values for GdFeCo vary from 0.75 mJ/cm$^2$ to 3.14 mJ/cm$^2$ (91). As an example, assuming the carefully determined threshold from Ref. (84) $F_C$=2.6±0.2 for a d = 20 nm thick film and a total heat capacity of $C = 3±0.2$ MJ/(m$^3$K) (92), the transient $T_p$ can be calculated through $T_p = T_0 + F_C/(d*C)$, where $T_0$ is the initial temperature. $T_p$ should rise to about 750K, well above $T_C$≈ 550 K (91). Crossing $T_C$ would imply a loss of memory of the magnetic order. The final magnetization would then be determined by the cooling conditions, analogous to HAMR, which is in contrast with what most AOS models assume (28, 64, 76, 85–89).

Here, we carefully measure $F_C$ for the helicity-independent AOS of GdFeCo films, through single shot switching and stroboscopic pump-probe experiments. $F_C$ values are then obtained as a function of the sample temperature $T_0$ and the laser pulse duration $\Delta t$. We observe AOS for pulse durations as long as $\Delta t = 15$ ps and identify two distinct mechanisms that prevent AOS at longer pulse durations. By using the three temperature model (51), we calculate that for $\Delta t = 55$ fs, $T_e$ reaches 1600 K, while for a $\Delta t = 12.5$ ps pulse $T_e$ reaches 530 K. We conclude that the electron peak temperature does not play a key role in the switching mechanism, and raise questions about the conclusions in various AOS models. Finally, we performed pump-probe experiments as a function of the pulse duration and showed that 10ps pulses result in switching times of $\sim$ 13 ps.
5.2 Experiment details
The experiments were carried out on two Gd$_x$(Fe$_{90}$Co$_{10}$)$_{100-x}$ films of concentrations $x = 24.5\%$ and $27.5\%$ grown by co-sputtering of the following stacks (in nm): Si/SiO$_2$(300)/Ta(2.5)/GdFeCo(14)/Ta(3.6)/Ta$_2$O$_5$(2.8). The layer thicknesses were confirmed by X-ray reflectivity. Both samples exhibited perpendicular magnetic anisotropy, which was determined via MOKE hysteresis measurements (as shown in the inset of Fig 5.1). A Curie temperature of about 540 K was obtained by fitting the normalized polar Kerr rotation (NPKR) via phenomenological formula (93)

\[ \text{NPKR} = \frac{(T_C-T)}{(T-300)^{0.39}} \]

(see Fig 5.1). This Curie temperature is close to previously reported values (77). The compensation temperature $T_M$ was measured by monitoring the coercivity and polarity of the magnetic hysteresis via MOKE as the sample was heated with an electric heater (77). We found $T_M \approx 430K$ for sample $x = 27.5\%$. Sample $x = 24.5\%$ presented a hysteresis with the opposite polarity to that of $x = 27.5\%$ at room temperature (see inset of Figure 5.1), meaning its compensation temperature was below room temperature. We did not have the capability to measure below ambient temperature. The two samples will respectively be addressed as Gd$_{24}$FeCo and Gd$_{27}$FeCo throughout the text.

![Graph showing temperature dependence of normalized Kerr rotation and magnetic hysteresis.](image)

**Figure 5.1** Temperature dependence of the normalized Kerr rotation of Gd$_x$(Fe$_{90}$Co$_{10}$)$_{100-x}$ with $x = 27.5\%$. Inset shows the magnetic hysteresis as a function of the out-of-plane external field $H$, at room temperature, for samples with $x = 27.5\%$ and $x = 24.5\%$.

Time resolved MOKE measurements were carried with amplified laser pulses (RegA) at 250 KHz. The laser pulse duration full-width at half maximum (FWHM) was
tunable from $\Delta t = 55$ fs to $\Delta t = 25$ ps by adjusting the final pulse compressor in the chirped pulse amplifier (94). Individual single-shot laser pulses could be obtained from our laser system. A MOKE microscope was used for imaging the sample magnetization after each single laser pulse shot.

In single shot experiments, the laser beam was incident with an angle of $40^\circ$ with respect to the normal. The spatial beam profile was obtained by the knife-edge technique (95) and the energy of each pulse was monitored with a calibrated photodiode connected to a 6 GHz oscilloscope. To accurately determine the fluence absorbed in the GdFeCo film, a multilayer absorption calculation was performed (96) using an effective index of refraction of $n = 3.7 + 4.2i$ for Ta/GdFeCo/Ta measured by ellipsometry. An absorption of 29% was found (absorption profile in Figure 5.2). The magnetization of the film was saturated with an external magnetic field $H \approx 100$ Oe. Following removal of the external field, the film was then exposed to a single linearly polarized laser pulse.

![Figure 5.2 Optical absorption vs depth for GdFeCo films.](image)

**5.3 Results**

As shown in Figure 5.3, after each laser pulse of the same energy, the magnetization in a small region reliably toggles between white (‘up’) and black (‘down’) back and forth. Our observation of helicity independent toggling of the GdFeCo magnetization is consistent with the helicity independent AOS reported in Ref. (28).
Figure 5.3 (a) MOKE micrographs of the magnetization of the Gd$_{27}$FeCo film exposed to successive linearly polarized laser pulses of $\Delta t = 55$ fs on an initially “down” ($M_-$) magnetized sample. Reliable all-optical switching of the magnetization independently of the helicity of light is demonstrated. (b) Evolution of the normalized polar Kerr rotation of GdFeCo samples induced by a linearly polarized $\Delta t = 55$ fs pump, under a constant perpendicular external field of 55 Oe. The blue lines correspond to the evolutions under no external field, which show no difference during the first nanosecond.

In the absence of domain wall motion, the reversed domain size is determined by the area within the Gaussian laser profile with a fluence above $F_C$ (84). However, in our films, we noticed that domain wall motion reduces the size of the reversed domain in the seconds following laser irradiation. We observe a critical domain size (10 µm) below which optically switched domains shrink and collapse after several seconds. Instability of small magnetic domains is a well-understood phenomenon that occurs whenever the domain wall energy is larger than the domain stabilizing pinning and dipolar energy terms (97). In order to minimize the effect of such relaxation mechanisms on our measurement of the critical fluence, the pump diameter (FWHM) was chosen to be relatively large (0.16 mm). The absorbed critical fluences $F_C$ were then obtained by decreasing the pump fluence until no switching was observed. For $T = 300$ K and $\Delta t = 55$ fs, we found $F_C = 0.82 \pm 0.16$ mJ/cm$^2$.

We performed time-resolved pump-probe MOKE measurements on both samples. For these experiments, a constant, perpendicular external field of 55 Oe was
applied to reset the magnetization between pump pulses. The pump beam, incident at 40°, had a spot diameter (FWHM) of 100 µm, whereas the probe, at normal incidence, was kept much smaller with a spot diameter of 6 µm. As shown in Figure 5.3 for a fluence of 0.86 mJ/cm² the reversal occurs, against the external magnetic field, for both samples within a few picoseconds. The opposite sign of the signal at negative time delay for samples with Tₘ above and below room temperature is due to the sensitivity of our 810 nm probe to the FeCo sublattice magnetization (98). When T < Tₘ the external field drives the dominant Gd sublattice whereas at T > Tₘ the field drives the FeCo (98).

For comparison of Fₗ obtained through pump-probe experiments with Fₗ from single shot experiments, we first checked that the switching behavior was not affected by the constant external field. For this purpose, pump-probe demagnetization experiments at low fluences with no external field were performed on both samples (blue lines in Figure 5.3). No difference in the Kerr signal was observed with respect to experiments performed with the 55 Oe external field. In addition, we performed experiments on sample Gd₂₄FeCo (black down triangles in Figure 5.3) at T > Tₘ where no transition through Tₘ was possible due to laser heating. This means that field induced switching scenarios due to crossing of Tₘ can be discarded (98). Figure 5.4 shows the fluence dependence of the magnetization evolution in Gd₂₄FeCo at T = 300 K and for ∆t = 55 fs. The curve at 0.79 mJ/cm² presents relatively higher noise at long time delays, which we interpret as the result from laser intensity fluctuations when the fluence approaches Fₗ. We thus find Fₗ ≈ 0.8 mJ/cm², which is consistent with our single shot technique for measuring critical fluences (see Figure 5.5).
Figure 5.4 In solid lines, the evolution of the magnetization of Gd$_2$FeCo after a $\Delta t = 55$ fs linearly polarized pump pulse, at room temperature. The switching threshold is close to 0.8 mJ/cm$^2$, agreeing with the single-shot experiments.

In order to study the importance of the lattice temperature in AOS, the critical fluence was recorded as a function of the initial temperature $T_0$, which was varied by mounting the sample on a resistive heater. A threshold at which a multidomain pattern was observed was also recorded (see pic #3 in Figure 5.5). The measurement of this threshold has a large uncertainty due to the stochastic nature of domain nucleation and the instability of the small multidomain patterns. Within experimental accuracy we found the multidomain thresholds for both samples to be equal.
In the case where the whole system reaches $T_C$, a multidomain magnetization pattern is expected to arise as the sample cools down from the paramagnetic state and is re-magnetized randomly. Indeed, the transition from pure AOS to multidomain is observed (pics #2 and #3 in Fig 5.5) at a particular threshold fluence (blue crosses in Fig 5.5). Transient temperatures for electrons and the lattice were calculated with the three temperature model (51), and the threshold at which the overall temperature (back in equilibrium) exceeds $T_C$ is plotted as a blue dashed line in Figure 5.5. The model will be discussed later in the text. As both samples have very similar compositions, resulting in similar total heat capacities and Curie temperatures, the transient equilibrium temperature and thus the multidomain fluence threshold are expected to be similar. Therefore, the demagnetization threshold sets a limit above which no AOS can be observed.

The critical fluence for Gd$_{24}$FeCo is independent of ambient temperature, while the critical fluence of Gd$_{27}$FeCo decreases by a factor of two upon a change in ambient temperature from 300 to 450 K. We believe the different temperature dependence is related to the difference in energy transfer rates between sublattices in both samples, as has been predicted (86). A discussion on the energy transfer rates will follow later. However, both samples display a weaker temperature dependence than we would expect if AOS was an equilibrium phenomena analogous to HAMR.
If changes to equilibrium magnetic properties were the primary driver of AOS in a manner analogous to HAMR, the peak lattice temperature reached following laser irradiation at $F_C$ would be insensitive to ambient temperature. At ambient temperatures of 300 K and 470 K, the calculated transient temperature rise in the lattice following irradiation is $\sim 150$ K and $\sim 70$ K, respectively. Therefore, the peak lattice temperature during AOS varies from 450 K to 540 K ($\sim T_C$) for ambient temperatures from 300 to 470 K. Therefore, as expected, we confirm that unlike in HAMR, heat induced changes to equilibrium magnetic properties are not the primary driver of AOS.

5.4 Analysis

As discussed above, numerous models predict that the transient temperature response of the electrons $T_e$ following laser irradiation is responsible for AOS (28, 64, 76, 85–87). In atomistic calculations typically $T_e$ is coupled to a Langevin random field term which is then entered into a Landau-Lifshitz-Gilbert equation. The equations are,

$$\frac{\delta S_i}{\delta t} = -\frac{\gamma_i}{(1+\alpha_i^2)\mu_i}(S_iH_i + \alpha_i S_i[S_iH_i])$$  \hspace{1cm} (5.1)

Where $S_i$ is the reduced atomic localized spin, $\gamma_i$ is the gyromagnetic ratio of the sublattice, $\alpha_i$ is an effective damping parameter (a channel to dissipate angular momentum to the lattice), and $H_i$ is an effective field given by

$$H_i = \eta_i + \frac{\delta E_i}{\delta S_i}$$  \hspace{1cm} (5.2)

Where $\eta_i \propto \alpha_i T_e$ is the Langevin noise of the sublattice, proportional to $T_e$, and $E_i$ is the energy of the sublattice, including exchange, anisotropy and Zeeman terms. These models can successfully reproduce the switching behavior through a three-step process. In the first step $T_e$ has to quickly overcome $T_C$ in order for the Langevin field (thermal excitations) to overcome the exchange field. This induces the independent demagnetization of the sublattices. Due to their different damping (rate of dissipation of angular momentum) and magnetic moments, demagnetization for different lattices will occur at different rates, the Fe demagnetizing faster (76). The second step involves the cooling of $T_e$ which allows the re-magnetization of the completely demagnetized Fe sublattice. At this stage the exchange fields become dominant again, and as the Gd demagnetizes towards its equilibrium magnetization (at $T_e$) conservation of angular momentum induces the switching of the Fe sublattice. The third step consists in the antiparallel alignment of the Gd spins relative to the Fe spins due to the exchange interaction.
However, in these models it is often clearly claimed (28, 76, 85) that initially $T_e$ needs to quickly overcome $T_C$ in order to decouple the sublattices and allow a faster demagnetization of the Fe sublattice. Other microscopic models (88, 89) that treat the energy and angular momentum exchange through scattering processes, also reach similar conclusions, and state the necessity of short and intense pulses for the initial demagnetization of both sublattices to happen at different rates.

To test the importance of the peak electron temperature, single shot AOS experiments as a function of the pulse duration (FWHM) $\Delta t$ were performed. As $\Delta t$ increases, the laser peak intensity drops as $1/\Delta t$ resulting in a lower peak $T_e$. However, since energy transfer rates depend on temperature differences between heat baths, electrons actually lose less energy when they are cool. The result is a drop of the peak $T_e$ by a factor of 3 when going from $\Delta t = 50$ fs to 10 ps (51). If the peak $T_e$ is a key parameter for AOS, as $\Delta t$ is increased the critical fluence should increase proportionally. We observe a relatively weak dependence of the critical fluence on the pulse duration (see Figure 5.5.b). The energy needed for AOS increases 50% as the pulse duration increases by over two orders of magnitude. Similar trends have been reported in the context of helicity-dependent AOS (91, 99). However the analysis in Refs.(91, 99) was made in terms of helicity-induced opto-magnetic fields. Furthermore, high critical fluences were reported that would easily heat the lattice above $T_C$. As we have shown, such high lattice transient temperatures should result in a random multidomain state instead of a helicity-independent AOS. In our experiments, as shown in Figure 5.5.b, we observe single shot helicity-independent AOS for pulses as long as $\Delta t = 15$ ps pulse in the Gd$_{27}$FeCo sample. For $\Delta t > 15$ ps $F_C$ exceeds the multidomain critical fluence. The result is then a fully demagnetized pattern (pic #4 in Figure 5.5.b) and no AOS is ever observed for these pulse durations. This is in sharp contrast with the $\Delta t = 55$ fs multidomain state (pic #3 in Figure 5.5.a) where the outer part of the Gaussian laser beam does induce AOS.

We performed time resolved pump probe experiments on Gd$_{27}$FeCo at various pulse durations in order to see how the switching dynamics are affected by the electron’s heating rate. This is shown in Figure 5.6. The probe pulse duration was kept equal to the pump, which results in a loss of time-resolution and smoothing of the data for longer pulse durations. Due to a decrease of the AOS fluence window at long pulse durations, the probe was tightly focused through a 50x objective onto 2 µm spot at the center of the pump spot. A constant 200 Oe magnetic field was applied to reset the magnetic state of the film between pulses. As the pump duration increases, the switching time (crossing of 0 on the y axis) increases from 2
ps up to 13 ps for \( \Delta t = 55 \) fs and 10 ps respectively. The switching happens in all cases after all of the energy of the optical pulse is deposited in the film. This result shows that using 10 ps optical pulses, we can still perform a rather fast switching of the magnetization, which releases the constraint on using femtosecond lasers for the study of AOS and for applications.

![Figure 5.6](image)

Figure 5.6 In solid lines, the evolution of the magnetization of Gd\(_2\)FeCo after a linearly polarized pump pulse, at room temperature, for \( \Delta t = 60 \) fs, 1 ps, 6.4 ps, 10 ps and \( F_C \sim 0.8 \text{mJ/cm}^2, 0.9 \text{mJ/cm}^2, 1.0 \text{mJ/cm}^2, 1.6 \text{mJ/cm}^2 \) respectively. A 10 ps pump intensity profile is depicted in light grey. The probe duration was kept equal to the pump duration, which results in a loss of resolution and a smoothing of the long pulse duration curves. The switching time (crossing of 0) increases with the pump duration, and always happens after all the energy has been deposited on the film. Note that zero time delay was readjusted since tuning the compressor introduces small changes (2 mm) in the pump and probe paths. Zero time delay was set by assuming that the maximum slope of the demagnetization corresponds to the peak of the pump pulse.

The transient temperature response of the electrons and phonons during AOS with \( \Delta t = 55 \) fs, \( \Delta t = 1 \) ps, and \( \Delta t = 12.5 \) ps pulses at fluences equal to \( F_C \) are shown in Figure 5.7. We calculated the temperature responses using the three temperature model (51). We fixed the electron heat capacity \( C_e = \gamma T_e \), with \( \gamma = 300 \text{J/(m}^3\text{K}^2) \) based on first principle band structure calculations of amorphous GdFe\(_2\) (100). The lattice heat capacity is set to 2.3 J/(m\(^3\)K), a weighted average of the lattice heat capacity of Ta and GdFe\(_2\) (92). The spin heat capacity in our model as a function of
temperature was fixed by subtracting the electronic and lattice heat capacities from the total heat capacity of GdFe$_2$ (92). The electron-spin coupling constant was fixed to $10^{17}$ W/(m$^3$K) and the electron-phonon coupling constant was set to $6 \times 10^{17}$ W/(m$^3$K). These two values were set based on separate thermal transport measurements of Au/GdFeCo metallic bilayers that we have made (101).

We do not consider the spin temperature in our three temperature model calculation to be a valid descriptor of the thermodynamic state of the spin system. The transient magnetic states that occur following laser irradiation do not occur in the equilibrium phase diagram of GdFeCo, and therefore cannot be described with an effective temperature. Therefore, the sole purpose of the spin temperature in our model is to account for the impact of energy transfer between the electrons and magnetic sublattices on the transient temperature response of the electrons. This channel for energy exchange needs to be considered, especially when close to $T_C$ where the magnetic heat capacity is as large as 40% of the total heat capacity.

![Graph showing electronic (solid lines) and lattice (dotted lines) temperatures after different delays](image)

**Figure 5.7** Calculation of electronic (solid lines) and lattice (dotted lines) temperatures after a $\Delta t = 55$ fs $F_C = 0.8$ mL/cm$^2$ pulse (red), a $\Delta t = 1$ ps $F_C = 0.9$ mL/cm$^2$ (blue) and a $\Delta t = 12.5$ ps $F_C = 1.35$ mL/cm$^2$ pulse (black) according to the three temperature model. The dashed line indicates $T_C$. For $\Delta t = 12.5$ pulses, $T_e$ gets very close to $T_C$. Whether $T_e$ needs to reach $T_C$ or not is unclear due to the uncertainties ($\sim 20\%$) of the critical fluences $F_C$. 

55
The small increase in $F_C$ as $\Delta t$ increases implies that the peak electron temperature of the system is not particularly important for the helicity-independent AOS. In fact, as shown in Figure 5.7, for $\Delta t = 12.5$ ps pulses, $T_e$ will only be heated to 530 K. We are not able to exactly determine whether $T_e$ needs to reach $T_C$ or not, due to the uncertainty (20%) in the critical fluence. Despite this open question, our result raises questions on the proposed scenario where very high electron temperatures (1000–2000 K) are necessary for AOS (28, 64, 76, 85–89).

We posit that helicity independent switching is a three-step process, where there is no need for high electron temperatures. First, after optical absorption, the energy per Fe spin degree of freedom becomes slightly higher than the energy per Gd spin degree of freedom (i.e. the Fe is hotter), as proposed by Wienholdt et al (87). Second, the Fe and Gd spins exchange energy and angular momentum on a timescale faster than the time scale of angular momentum dissipation into the lattice. This corresponds to a semi-adiabatic process and the dynamics of the system are thus governed by the principle of maximization of entropy as described by,

$$ (2J_{FF}S_F - 2J_{GG}S_G + J_{FG}S_G - J_{FG}S_F)dS_F > 0 \tag{5.3} $$

where the left side of the equation corresponds to the change in internal energy of the system. $J$ is the exchange constant and $S$ is the total spin angular momentum of sublattices Fe (F) or Gd (G). In GdFeCo, $J_{FF}$ and $J_{GG}$ are negative, $J_{FG}$ is positive, so that $S_F$ and $S_G$ have initially opposite signs. Conservation of angular momentum (d$S_F$ = d$S_G$) is implied.

To fulfill Equation 5.3 we find that $S_F$ and $S_G$ must decrease, meaning demagnetization of the sublattices will occur. If the Fe sublattice is initially hotter, the Fe will reach full demagnetization first. With the Fe fully demagnetized ($S_F = 0$) Equation 5.3 implies the switch and growth of the Fe sublattice parallel to the Gd spins, leading to a transient equilibrium ferromagnetic state (87). In other words, on timescales over which angular momentum is conserved, the temporary equilibrium state will be ferromagnetic because entropy is maximized with ferromagnetically aligned Gd and Fe spins.

In the third and last step, the Gd switches in order to be antiferromagnetically aligned with the now hot and dominating Fe lattice (77) and both sublattices re-magnetize as they cool down. Re-magnetization occurs on much longer time-scales than demagnetization, so spin angular momentum is not conserved anymore.

In the proposed three step scenario, the magnetization can switch sign without ever reaching the Curie temperature. There are two requirements. First, the Fe spin
system must be preferentially heated with respect to the Gd spins (87). Second, exchange of energy between sublattices should happen faster than the timescales of dissipation of angular momentum into the lattice (102). Moreover, the lattice temperature should remain below $T_c$ at all times, otherwise resulting in a multidomain final state. However, as shown in Figure 5.5.b the total energy necessary for AOS is not strongly dependent on the pulse duration, indicating that a minimum amount of energy (the critical fluence) needs to be deposited on the sample in order to drive sufficient energy and angular momentum transfer between sublattices.

The difference in maximum pulse duration between samples Gd$_{24}$FeCo and Gd$_{27}$FeCo shown in Figure 5.5.b resides most probably in the differences in energy transfer rates between the Fe and Gd spin sublattices, which depend on the composition (86). Experimentally examining the relationship between alloy composition and energy transfer rates will be the subject of a future work.

5.5 Conclusion

In summary, we carefully extracted critical fluences for AOS in two GdFeCo samples as a function of the initial temperature of the sample and pulse duration by single-shot and stroboscopic measurements. We confirm that lattice heating is not the main mechanism for AOS. We then showed that AOS is possible for pump laser pulse duration up to 15 ps. We attribute the maximum pulse duration for AOS in the Gd$_{27}$FeCo sample to overheating of the lattice. We performed pump-probe experiments as a function of the pulse duration and showed that the switching time increases as the pump duration increases, with 10 ps pulses resulting in switching times of 13 ps. We estimated the temperature rise for electrons and the lattice via the three temperature model and showed that the peak electron temperature is not a major parameter for AOS as it varies from 1600 K for $\Delta t = 55$ fs to $T_c$ for $\Delta t = 12.5$ ps. AOS with 15 ps pulses challenges previous models for helicity-independent AOS where high electron temperatures are assumed responsible. Finally, we suggest a three step thermodynamic model of the switching based on the preferential heating of Fe spins compared with Gd spins, and on the fast energy exchange between the sublattices. In the case of the Gd$_{24}$FeCo sample, we attribute the pulse duration limit to a slowing down of the energy transfer rates between sublattices.
Chapter 6

Ultrafast magnetic switching of GdFeCo with electronic heat currents

6.1 Introduction

In previous chapter, we show that AOS of GdFeCo can be achieved with optical pulses as long as 12 ps. In all prior AOS experiments, GdFeCo electrons are excited with an ultrafast laser pulse to eV energies above the Fermi level (77). Subsequently, the FeCo sublattice demagnetizes within a few hundred femtoseconds (76). The Gd sublattice also loses magnetic order, but at a slower rate. The differing rates of demagnetization, together with the transfer of angular momentum from the Gd to FeCo sublattice, enables reversal of the magnetic moment on ps time-scales (28, 77, 87). While initial studies credited the ultrafast reversal of the magnetization to a helicity-dependent light-matter interaction (27), subsequent investigations with linearly polarized lasers show the reversal is driven solely by energy absorption (28, 84).

While the phenomenology of AOS is well documented (15, 52, 77, 91), many of the mechanisms underpinning ultrafast demagnetization and AOS remain poorly understood. The initial response of the GdFeCo to optical irradiation is an ultrafast quenching of the magnetization of the FeCo sublattice that causes an abrupt change in the net magnetization of the system. However, the role of thermal vs. nonthermal electrons in ultrafast demagnetization and subsequent magnetization reversal remains controversial (16, 19, 46, 72, 103–106).

Here, we demonstrate direct laser irradiation of GdFeCo is not necessary for deterministic reversal of the magnetization. We show the magnetic response of h nm Au/ 10 nm GdFeCo bilayers to optical irradiation at the Au surface. By varying the Au thickness from 5 to 91 nm, we control the ratio of laser energy absorbed by the GdFeCo to that is absorbed by the Au. The total absorbed fluence required for switching increases by only a factor of three when the Au thickness is increased from 5 to 91 nm, despite a factor of twenty decrease in the amount of energy directly absorbed by the GdFeCo. Our results suggest thermal currents can reverse the magnetization as is possible by direct optical heating.
6.2 Experiment details
Six Au/GdFeCo bilayer samples were prepared via magnetron sputter deposition on sapphire substrates. The Au film thicknesses for the six samples are 5, 10, 37, 58, 73 and 91 nm. The GdFeCo film thickness is measured by XRR to be ~11.5 nm in all six samples.

We use an amplified Ti:sapphire laser with 810 nm center wavelength in our experiments (Coherent RegA 9050). We run the laser amplifier at a repetition rate of 250 kHz for time-resolved pump/probe measurements, or instead eject single laser pulses. In the majority of experiments, the pump laser is incident on the Au surface of the bilayer, while the probe laser is incident on the GdFeCo film through the sapphire substrate (Figure 6.1).

![Figure 6.1 A schematic of the front-pump-back-probe experiment on the Au/GdFeCo bilayer structure.](image)

We use a MOKE microscope for monitoring the GdFeCo magnetization after laser irradiation. The MOKE microscope focuses on the GdFeCo film through the sapphire substrate. In these experiments, an external magnetic field $H \approx \pm 100$ Oe saturates the magnetization of the sample out-of-plane. Following removal of the external field, a single linearly polarized laser pulse irradiates the Au surface. As shown in Figure 6.2 for the Au (91 nm)/GdFeCo bilayer, if a laser pulse of sufficient energy irradiates the Au film surface, the magnetization in the irradiated region reliably toggles between white ('up') and black ('down').
Figure 6.2 The MOKE images from GdFeCo side of Au(91nm)/GdFeCo sample, after consecutive laser pulse irradiation on Au side.

6.3 Results and analysis
The goal of our study is to examine systematically how heating GdFeCo electrons via thermal currents instead of optical absorption influences energy requirements for magnetization reversal. For the Au/GdFeCo bilayers with Au layers thicker than 50 nm, electronic heat currents flowing from the adjacent Au layer deposit the majority of heat in the GdFeCo layer. In Figure 6.3, we report the total fluence, $F_T$, the bilayer must absorb to induce magnetization reversal of the GdFeCo. Irradiation of the Au surface with sufficient fluence results in reversal of the GdFeCo magnetization in all samples, despite negligible GdFeCo absorption in the bilayers with thick Au films. As a control experiment, we also perform measurements on a Pt (5 nm)/Au(75 nm)/MgO(3 nm)/Au(5 nm)/GdFeCo(10 nm) sample. For this sample, the Pt absorbs most of the optical pulse, and the insulating MgO layer prevents electronic heat currents into the GdFeCo. No deterministic magnetization switching is observed in this sample at any fluence.
To interpret our experimental data, we use a thermal model to predict the temperature responses of the electrons and phonons in the bilayers, as shown in Figure 6.4. Details of the thermal model are in Ref. (101). Following optical heating of the Au electrons, the Au electrons reach a temperature in excess of 2000 K in bilayers where the majority of energy is absorbed in the Au. The high diffusivity of the Au electrons allows rapid heat diffusion (54), resulting in TW m\(^{-2}\) picosecond heat currents into the GdFeCo.
Figure 6.4 Temperature response of a Au (91 nm) / GdFeCo bilayer after the Au electrons absorb 17 J m\(^{-2}\) from a 55 fs laser. Each curve represents an average temperature across the layer. The large temperature difference between the Au electrons and GdFeCo electrons for the first few picoseconds following irradiation generates electronic heat currents in excess of 2 TW m\(^{-2}\). The large electronic heat currents are sufficient to reverse the GdFeCo magnetization.

The Au/GdFeCo bilayers with thicker Au layers require more absorbed fluence to induce magnetization reversal. The increase in \(F_T\) with increasing Au thickness is due to only a fraction of the energy absorbed by the Au diffusing to the GdFeCo electrons. In parallel to energy transfer from the hot Au electrons to the GdFeCo electrons, significant energy is transferred to the Au phonons via electron-phonon scattering (54). The characteristic length-scale over which the electronic heat can diffuse before the hot Au electrons transfer most of their energy to the phonons is 

\[
d_{ep} \approx \sqrt{\Lambda_{e,Au} / g_{ep,Au}} \approx 100 \text{ nm},
\]

where \(\Lambda_{e,Au} \approx 250 \text{ W m}^{-1} \text{ K}^{-1}\) is the thermal conductivity of the Au electrons and \(g_{ep,Au} \approx 2.8 \times 10^{16} \text{ W m}^{-3} \text{ K}^{-1}\) is the electron-phonon coupling constant of Au (107).

Figure 6.5 shows our thermal model’s estimate for the total fluence transferred between the Au and the GdFeCo for each experiment. Figure 6.5.a shows the optical and electronic heat currents into the GdFeCo for the Au (38 nm)/GdFeCo and Au (91 nm) / GdFeCo bilayers. Integrating both the optical and electronic heat-currents over the time interval of the experiment yields the total fluence absorbed by the GdFeCo electrons, \(F_{GFC}\), as a function of Au thickness (Figure 6.5.b). We observe in all samples that a total fluence between 5 and 6 J m\(^{-2}\) must be absorbed
by the GdFeCo from electronic and/or optical heat currents for magnetization reversal to occur. Especially, in the sample with 91 nm Au, as shown in Figure 6.5.b, almost the energy comes from electronic heat current in gold layer, which means electronic heating alone could induce magnetic switching of GdFeCo.

Figure 6.5 (a) Heat currents into the GdFeCo electrons from both direct optical absorption and from the adjacent Au film. Solid and dashed lines are calculations for the Au (38 nm) and Au (91 nm) bilayers with an absorbed fluence of 9.4 and 17 J m\(^{-2}\), respectively. Red curves represent electronic heat-currents from the adjacent Au layer, while blue curves are heat-currents from direct optical absorption of the laser energy by the GdFeCo electrons. (b) Fluence absorbed by GdFeCo electrons vs. Au film thickness. The red open circles demark the fluence from heat currents via the Au electrons, the blue open circles represent the fluence from direct optical absorption, and the filled black circles represent the total fluence absorbed by the GdFeCo electrons from all sources. Lines are to guide the eye.

In our analysis above, we assume exciting the GdFeCo electrons via heat-currents across the Au/GdFeCo interface will generate a thermal distribution. Prior to heating the GdFeCo electrons, energy must travel across the Au film. In the time it takes the heat to travel from the Au, the electron-electron and electron-phonon scattering in the Au will move the initially nonequilibrium Au electron distribution towards a thermal distribution. The time-scale for energy to diffuse across a 73 nm thick Au film via hot electrons is \(\tau \approx 0.6\) ps. Alternatively, the time-scale for energy to ballistically travel across the film is given by \(\tau \approx h_{Au}/v_F \approx 50\) fs, where \(v_F \approx 1.4 \times 10^6\) \(m/s\) is the Fermi velocity of Au.
To test the time-scale for energy transport across the Au, we prepared four new samples for time-resolved pump/probe measurements. Pump/probe measurements of the Au/GdFeCo bilayers were not possible because the low optical absorption of Au requires a high incident laser fluence to induce magnetization dynamics. A high fluence causes sample damage when we operate the laser at a repetition rate of 250 kHz instead of the single shot mode we use in the experiments described above. The geometries of the four new samples are Au (10nm) /MgO (170 nm) /Ta (2nm)/ GdFeCo (10 nm) / Au ( hAu )/ Pt (5 nm), with hAu = 0, 30, 74 and 120 nm. The addition of Pt to the stack lowers the incident fluence necessary to induce magnetization dynamics by 5x, and eliminates the sample damage problem. The addition of the Au/MgO bilayer adjacent to the GdFeCo enhances the MOKE contrast (108). Figure 6.7 shows pump/probe measurements on these four samples. The insertion of a 30, 74 and 120 nm thick Au layer between the Pt and GdFeCo causes a delay of ~0.1, ~0.7 and ~1 ps in the demagnetization of the GdFeCo layer. These sub-picosecond time-scales are longer than the 20, 50 and 80 fs time-scales we expect for ballistic travel. The consistency of zero delay time between samples is better than 30 fs, and is primarily determined by the 1.6 µm depth of focus of the objective lens. Therefore, our pump/probe measurements provide strong support for our thermal modelling.
Figure 6.7 Pump probe data of Pt / Au / GdFeCo samples. The pump laser is incident on the Pt. The 0.1 and 0.7 ps delay in demagnetization of the samples with 30 nm Au and 75 nm Au between the Pt absorber and GdFeCo layer is consistent with diffusive heat transfer by hot Au electrons.

6.4 Conclusion

In conclusion, by adding a gold layer adjacent to GdFeCo to serve as an optical absorber, we excite GdFeCo with electronic thermal currents. With front-pump-back-probe technique, we confirm that in the samples with thick (>70nm) gold spacer, the hot electrons are already thermalized before they reach GdFeCo layer. The pure electronic thermal currents are sufficient to cause ultrafast magnetic switching in GdFeCo. The discovery that purely thermal currents are effective in magnetization reversal of GdFeCo signals new opportunities for potential device applications of ultrafast magnetization switching.
Chapter 7

Ultrafast magnetization reversal of ferrimagnets by picosecond electrical pulses

7.1 Introduction

Spintronic devices are promising candidates for future low-energy electronics that take advantage of the non-volatility of nanoscale magnets (4). For example, magnetic random access memory (MRAM) is emerging as a universal integrated on-chip memory (2). Investigations into spintronic logic devices are also underway due to their potential for low-power computing (3, 109–111). A significant obstacle that impedes wide spread adoption of spintronic devices is their speed. Spintronic mechanisms such as spin-transfer torque (STT) or spin-orbit torque (SOT) can theoretically transfer angular momentum in the low picosecond timescale and induce fast magnetization dynamics (112, 113). However, so far, the record switching time of spintronic devices is still on the order of hundreds of ps (5, 11–13). For comparison, silicon field-effect transistors possess switching delays less than 5 ps (114). In order for spintronic technologies to challenge charge-based devices in information technologies, increases in the speed of operation are necessary.

Research over the past two decades in the field of ultrafast magnetism demonstrates that precessional speed limits for manipulating magnetic order can be broken if the electrons are excited on time-scales faster than the electron-phonon relaxation time, i.e. excited on ps or sub-ps time-scales (15). For example, the magnetization of a ferromagnetic thin film can be quenched within 300 fs upon 60 fs laser irradiation (14). Furthermore, multiple studies have demonstrated the ability of single 100 fs laser pulses to deterministically and repetitively switch the magnetization of the ferrimagnetic metal GdFeCo on sub-picosecond time-scales (27, 28, 52, 76), a phenomenon known as all optical switching (AOS), as discussed in previous chapters.

Here, we take advantage of the physics responsible for AOS in GdFeCo to demonstrate a new regime of purely electrical ultrafast spintronics. Instead of optically exciting electrons, we use picosecond charge current pulses to excite the mobile conduction electrons of a GdFeCo metal film. We observe repeatable
ultrafast magnetization reversal in the GdFeCo film with single sub-10 ps electrical pulse excitation.

7.2 Methods
To generate picosecond electrical pulses, we fabricated picosecond photoconductive switches (38), in a gold coplanar stripline (CPS) geometry, on a “low temperature (LT) grown” GaAs substrate (37), as discussed in chapter 3. Figure 7.1.a shows a schematic of the CPS device. The CPS is tapered from 50 µm to 5 µm, contacting on top of both sides of a patterned GdFeCo film, leaving a 4 µm by 5 µm uncovered GdFeCo section. By illuminating the DC biased photoconductive switch with amplified 60 fs laser pulses at 810 nm wavelength, we are able to generate current pulses that have a duration of ~9 ps at full width half maximum (FWHM) and a current density up to ~10^9 A/cm^2 through the GdFeCo section (Figure 7.1.b).

Figure 7.1 Schematic of the CPS device and characterization of electrical pulse. a) Schematic of electrical switching experiment. The photo-switch is illuminated with laser pulses while biased with a DC source. Magnetization dynamics of GdFeCo is monitored with TR-MOKE. Left Inset: Optical image of the photo-switch. Right Inset: Optical image of GdFeCo section of CPS. Scale bar: 20 µm. b) Calculated temporal current density profile through the GdFeCo section, based on the temporal current profile measured with Protemics Spike probe positioned 1 mm before the GdFeCo section.
Figure 7.2 Temporal current density profiles in CPS. Temporal current density profile generated by the photo-switch in 50 µm wide CPS Au line with different biases, as measured with Protemics Spike probe positioned 1mm before the GdFeCo section. The smaller electrical pulse following the main peak is attributed to electrical reflection from the GdFeCo section in the CPS.

In our experiments, we studied the magnetic response of a Ta(5 nm) / Gd$_{30}$Fe$_{63}$Co$_{7}$(20 nm) / Pt(5 nm) multilayer. The film presented perpendicular magnetic anisotropy with a coercivity of 80 Oe at room temperature. The compensation temperature for this sample, i.e. the temperature where the net moment is minimized in ferrimagnets, is ~270 K (Figure 7.3). Consistent with prior studies (28, 52, 76), as shown in the differential MOKE images, upon irradiation by a sequence of single laser pulses, the magnetization of the GdFeCo film toggles after each pulse. We checked the AOS ability at different laser pulse durations and found the GdFeCo film switches with laser pulse durations between 60fs (FWHM) and 10 ps, consistent with the results reported in Ref.(52).
Figure 7.3 Magnetic Properties of the GdFeCo stack. a) Magnetic moment of the GdFeCo film measured with a superconducting quantum interference device (SQUID) at various temperatures. An out-of-plane external magnetic field of 500 Oe is applied during all the measurements. b) Magnetic hysteresis curve of the GdFeCo film at room temperature, measured with the MOKE microscope.

To calculate the energy deposited by picosecond electrical pulses, we use the same methods as described in chapter 4. We take the Fourier transform of the electrical pulse (voltage) $V(t)$ in time domain to get the frequency domain spectrum $\tilde{V}(\omega)$. The energy spectral density is then proportional to $|\tilde{V}(\omega)|^2$, as plotted in Figure 7.4.
Figure 7.4 Energy spectral density of the electrical pulse, calculated as the square of the Fourier transform of the electrical pulse in the time-domain.

The attenuation before the GdFeCo load $\alpha_1(\omega)$, as well as absorption $\alpha_2(\omega)$ are calculated similarly as chapter 4 and plotted in Figure 7.5.

We calculate the total absorption in the GdFeCo to be 13%. The total energy carried by the initial electrical pulse is estimated by $\int I^2 \times Z \, dt = 4.3 \, \text{nJ}$, which means that we deliver about 570 pJ of electrical energy into the GdFeCo load.

To calculate the optical energy absorbed in the GdFeCo stack, we use a multilayer absorption calculation (52). The electric field inside the stack is obtained through the transfer matrix method. The absorption was then obtained by calculating the divergence of the Poynting vector inside the stack. The resulting absorption per layer is reported in Table 7.1. The absorption profile is shown in Figure 7.6. The whole GdFeCo stack absorbs 35% of the incident energy.
<table>
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<th>Thickness (nm)</th>
<th>Refractive Index</th>
<th>Absorption (%)</th>
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<td>-</td>
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<tr>
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<td>10.4</td>
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Table 7.1 Optical multilayer calculation parameters and results

Figure 7.6 Calculated optical absorption profile for the Pt/GdFeCo/Ta stack at 810 nm wavelength. Incident angle is 40 degrees to normal.

7.3 Results

We examined the response of the GdFeCo film to electrical pulses. Figure 7.7.b shows the differential MOKE images of the device after a sequence of individual 9 ps electrical pulses with current density $\sim7\times10^8$ A/cm² through the GdFeCo section. The magnetization of the GdFeCo section toggles after each electrical pulse, just as in the optical experiments. The switching behavior is driven by transient heating of the electrons, as described in prior studies of AOS in GdFeCo.
Figure 7.7 Single-shot optical and electrical switching of GdFeCo. a) Differential MOKE images of bare GdFeCo film after sequential 6.4 ps optical pulse irradiation. Absorbed fluence is 1.8 mJ/cm². After each optical pulse, the magnetization of GdFeCo toggles to the opposite direction. The contrast indicates change in magnetization. b) Differential MOKE images of GdFeCo CPS section after sequential 9 ps electrical pulse excitation. After each electrical pulse, the magnetization of GdFeCo toggles. Yellow and blue dash lines indicate gold CPS and GdFeCo section. Scale bar: 5 µm.

We performed time resolved MOKE measurements in order to temporally resolve the switching dynamics following the arrival of an electrical pulse. Figure 7.8.a shows the magnetic dynamics that result from electrical pulses of different amplitude. For electrical pulses with an absorbed energy density in the GdFeCo section less than 0.8 mJ/cm² (with reference to the surface area), the MOKE signal (primarily indicative of the FeCo sublattice magnetization) shows demagnetization within 20 ps, followed by a recovery to the initial magnetization state on longer time-scales. With increased current pulse amplitude, the FeCo demagnetization is larger. For the electrical pulses with an absorbed energy density greater than 1.3 mJ/cm², the magnetic moment of the FeCo sublattice reverses within ~10 ps of the electrical pulse arrival at the GdFeCo film. Following reversal, the FeCo magnetization recovers rapidly towards the opposite direction. It reaches 70% of saturation within just 30 ps. We attribute the non-monotonic evolution of the magnetization, e.g. the decreasing of magnetization at ~40 ps, to the arrival of several low amplitude electrical pulses that arise from reflections of the initial pulse from various electrical discontinuities in the CPS structure.
Figure 7.8 Magnetization dynamics of GdFeCo after electrical and optical excitations. a) Electrical demagnetizing and switching of GdFeCo. All fluences are calculated absorbed fluences. With increasing electrical pulse amplitude, the GdFeCo demagnetization amplitude increases, and eventually switches around 10 ps. b) Comparison of electrical and optical switching of GdFeCo. Both electrical and optical fluences are calculated absorbed fluences.

For comparison, we performed time resolved AOS experiments on the same material, deposited in parallel on a SiO$_2$ (100 nm)/Si substrate (Figure 7.9). We used optical pulses with a FWHM of either 1 ps or 6.4 ps. The latter pulse duration is equivalent to the duration of electrical heating from a 9 picosecond electrical pulse because Joule heating is proportional to square of the current. The temporal evolution of the FeCo magnetization following optical irradiation is shown in Figure 7.8.b. We also include in Figure 7.8.b the magnetization dynamics that results from a 9 picosecond electrical pulse. The 1 ps optical pulses switched the magnetization in ~4 ps. Both the 6.4 ps optical pulse and the 9 ps electrical pulse switch the magnetization in ~10 ps. While the time-scale for switching is comparable, significant differences exist between optical and electrical switching. After the magnetization crosses through zero, the recovery of the magnetization in the opposite direction takes hundreds of ps longer for optical vs. electrical switching. This indicates the system reaches a higher equilibrium temperature after optical pulse excitation, given that electrons, spins and lattice are almost certainly in thermal equilibrium after 50 ps. We determined the absorbed critical fluences required for optical switching to be 1.2 mJ/cm$^2$ and 1.6 mJ/cm$^2$ for 1 ps and 6.4 ps durations, respectively. These fluence values for AOS are on the same order as our observations in previous work. Shorter optical pulses require somewhat less energy to reverse the GdFeCo magnetization, consistent with prior studies. We
determined the critical energy deposited in the GdFeCo during the electrical switching experiment to be 1.3 mJ/cm². Therefore, both our time-domain measurements of the magnetization and our estimates of the absorbed fluence indicate that electrical switching of GdFeCo requires less energy than optical switching.

Figure 7.9 TR-MOKE for 6.4 ps optical pulses with different fluences. The critical fluence for switching is around 1.6 mJ/cm². The GdFeCo film becomes completely demagnetized above 1.73 mJ/cm² because the lattice temperature exceeds Curie temperature.

7.4 Discussion and conclusion

One possible explanation for the different energy requirements of electrical vs. optical switching is the thermal vs. nonthermal nature of electrical vs. optical heating. In chapter 4, we have demonstrated that thermal vs. non-thermal heating of ferromagnetic metals results in distinct magnetization dynamics. The transfer of energy from electronic to spin degrees of freedom relies on scattering events, such as Elliot-Yafet scattering and electron-magnon scattering (18). Total scattering rates will depend strongly on both the number of excited electrons, and the average energy of excited electrons (57). Electrical heating results in a large population of excited electrons with average energies less than 10 meV, while optical heating initially excites a much smaller number of electrons with eV scale energies (115). The ability to induce magnetization reversal in GdFeCo by
picosecond electrical heating demonstrates that exciting a nonthermal electron distribution is not necessary for magnetization reversal. The ability to switch a magnetic metal such as GdFeCo with a single short electrical pulse has significant potential technological impact. For experimental convenience, we used an optoelectronic switch to generate picosecond electrical pulses, however this is not necessary. It is currently possible to generate and deliver sub-10 ps current pulses on-chip in conventional CMOS electronics. For example, a 5 ps gate delay has been demonstrated with 45 nm CMOS technology (114). Therefore, it may be possible to implement GdFeCo based ultrafast on-chip memory and logic devices. A memory device would also require electrical read out. The addition of an oxide tunnel junction to the GdFeCo stack would enable electrical read out of the magnetic state (116).

A non-volatile ultrafast embedded MRAM technology based on ultrafast electrical excitation of the GdFeCo electrons would not only allow low static power dissipation due to the non-volatility of GdFeCo, but also require low dynamic energy consumption. The energy density required to switch the magnetization in our device is 13 aJ/nm². For a cell-size of (20 nm)³, which is typical for memory devices (117), switching should be possible with a current pulse with a peak current of 3 mA that delivers ~4 fJ of energy. The energy required for switching remains low despite the high current density required because the electrical pulse duration is short. We conclude that picosecond electrical switching of GdFeCo can be as energy efficient as STT and SOT schemes (11–13, 118, 119), yet more than one order of magnitude faster (Figure 7.10).
Figure 7.10 Switching energy per unit area vs switching time for various magnetic devices. For STT and SOT devices, switching energy per area is derived from data in Ref. (120) and switching time is assumed equal to the switching pulse duration. Only the energy dissipated in the magnetic area is calculated. Resistivity of Pt wire in SOT device is assumed to be 50 $\mu\Omega \cdot cm$ (Ref. (121)). Red star represents our 30nm thick electrically switched GdFeCo. Switching energy is set equal to the absorbed energy in GdFeCo film.

An important merit of MRAM over other memory devices is the nearly unlimited cycling endurance (2). Electrical heating of GdFeCo shows strong potential for high endurance in our experiments. We observe no degradation of electrical or magnetic properties in our devices after more than 10 hours of pump-probe experiments, which were performed with a laser repetition rate of 252 kHz. Ten hours of experiments corresponds to more than 1010 switching cycles. Although the peak current density during switching is high ($\sim 7 \times 10^8$ A cm$^{-2}$), the average current density is only $\sim 2 \times 10^3$ A cm$^{-2}$, therefore we do not expect electromigration to be an issue in device durability. A lifetime in excess of $10^{10}$ cycles is many orders of magnitude higher than most resistive RAM, phase-change memory or conductive bridging RAM (120).

So far, the peak current requirements for switching that we observe represent significant technological challenges for practical implementation. Further device
and structure optimization may allow for significant reductions in the peak current and energy required for electrical switching. For example, switching energy per unit area could be significantly lowered by reducing the thickness of the GdFeCo stack from the 30 nm used here. Further optimization of the magnetic stack to reduce the peak switching current requirement is a subject of ongoing research. New memory circuit architectures may also be needed that support such short electrical pulses.

In summary, we demonstrate that picosecond heating by electrical current pulses can reverse magnetic order efficiently. The observed switching is an order of magnitude faster than prior methods for electrical switching based on spin polarized currents or spin torques. Our discovery bridges the gap between the fields of spintronics and ultrafast magnetism, which will open a new frontier for ultrafast spintronic science and related devices.
Chapter 8

Model for all optical switching of ferromagnets with multiple pulses

8.1 Introduction
The all optical switching was first observed in ferrimagnetic GdFeCo alloys (27), later in a variety of ferrimagnet alloys and synthetic ferrimagnets (78), and eventually in metallic ferromagnets (79). It has been recently shown (122) that two different types of switching need to be considered: First, a single-shot helicity-independent AOS found in ferrimagnets such as GdFeCo (28), and second, a multishot helicity-dependent AOS found in TbCo and ferromagnets (122). Although some models have been proposed that explain the switching in two-sublattice systems such as ferrimagnets (28, 64, 76, 86), modeling for AOS in ferromagnets was lacking until very recently. To our knowledge, the only mechanism suggested so far for AOS in ferromagnets is a combination of heating and the inverse Faraday effect (IFE) (123, 124). The IFE corresponds to the generation of an effective magnetic field on the sample induced by the helicity of the light. However, IFE is still very difficult to characterize experimentally and in the cited works the amplitude of such fields is treated as a free parameter.

Here we present an alternative model that describes multishot helicity-dependent AOS in ferromagnetic materials based on a purely heat-driven mechanism. We first present the switching mechanism which is based on a combination of MCD and stochastic switching close to the Curie temperature $T_C$. This is followed by an in-depth description of the problem and of the way physical parameters are chosen. AOS is shown to be possible within a range of temperatures (i.e., laser fluences), for a large range of MCD values, but only after a certain number of laser shots. Finally, we reproduce previous AOS results by simulating the sweeping of the laser beam.

8.2 Methods
In our model, the mechanism driving the switching is a very simple and intuitive one: Whenever a laser heats a state will be dramatically lowered as the anisotropy drops. For a circularly polarized beam, regions of the magnet with opposite
magnetization will absorb different amounts of light due to MCD, resulting in hotter $T_{\text{hot}}$ and cooler regions $T_{\text{cold}}$. The difference in temperatures will lead to a difference in magnetic stability. If $T_{\text{hot}} \approx T_C$ and the MCD is large enough, cool regions will remain stable, whereas hot regions will be prone to stochastic switching. Repeating the process (laser heating and cooling) multiple times will statistically lead the magnet to full switching.

In order to numerically test this idea, we represent the magnetic material by an array of $N_x$ by $N_y$ cells, grains, or macrospins (as in Figure 8.1.a), presenting a strong out of plane magnetic anisotropy of energy density $K$. For the sake of simplicity, we ignore exchange and dipolar couplings between cells. These hypotheses will be discussed later in the text. This means that the magnetic state of the cell can be represented by a symmetric double well potential, as shown in Fig 8.1.c, where the magnetization can be only in the states up or down. The characteristic hopping time for the magnetization from state a (up) to state b (down) is given by the Néel-Brown formula (125):

$$\tau_{ab}(T) = \tau_0 \exp\left(\frac{E_{ab}(T)}{k_B T}\right)$$  \hspace{1cm} (8.1)

where $\tau_0$ is an attempt time typically estimated to be on the order of 0.1 ns (126), $k_B$ is the Boltzmann constant, $T$ is the temperature of the cell, and $E_{ab}$ is the energy barrier that prevents the magnetization from switching. The barrier will have the temperature dependence $E_{ab}(T)=K(T)\times V$, where $V$ is the volume of the cell. A high anisotropy at room temperature $T_0$ leads to long-term stability of the magnetization. However, when heating the cells with a laser pulse (assuming a step-like heating profile of amplitude $\Delta T$ proportional to the laser fluence and duration $t_{\text{hot}}$), the probabilities that determine the final state (a or b) of the magnetization when starting in state a are given by (127)

$$P_{ab} = \frac{1}{2} \left(1 - \exp\left(-\frac{t_{\text{hot}}}{\tau_{ab}}\right)\right)$$  \hspace{1cm} (8.2)

$$P_{aa} = 1 - P_{ab}$$  \hspace{1cm} (8.3)

The probabilities are defined the same way when starting in state b, but the hopping time will be given by $\tau_{ba}$. The probability function spans from 0 to $1/2$ as the energy barrier decreases from infinite to 0. Intuitively, it means that when the cell is heated close to $T_C$ and the barriers disappear, the magnetization has no preferential direction.

If the laser pulse is right $\sigma^+$ (left $\sigma^-$) circularly polarized, b (a) states will absorb more heat due to MCD. This difference will result in hot and cold cells where, assuming a temperature-independent phonon heat capacity the temperatures are
given by $T_{\text{hot/cold}} = T_0 + (1 \pm \text{MCD}/2)\Delta T$ (Figure 8.1.e). Because of this difference in temperatures, hot and cold cells will have different switching probabilities (Figure 8.1.f) leading to a higher number of reversals of the hot cells (Figure 8.1.d). If we now heat the cells and allow them to cool back to $T_0$ N times, the cumulative probability for the magnetization to end in state b is given by

$$P_B = \left( P_{tb} - \frac{P_{ab}}{P_{ab} + P_{ba}} \right) \left( 1 - P_{ab} - P_{ba} \right)^{N-1} + \frac{P_{ab}}{P_{ab} + P_{ba}}$$

(8.4)

where the subscript i refers to the initial state. As N increases, the probability is given by the last term in Eq. 8.4 and when $P_{ab} \gg P_{ba}$ we find $P_B \approx 1$. Consequently, as long as enough heating cycles (i.e., laser pulses) are used and as long as there is a significant difference in the energy barriers for a and b cells, deterministic switching is expected. The barrier height difference originates in the difference in absorption due to MCD which leads to a different temperature rise for cells in states a and b. Because of this temperature difference and the strong $dK/dT$ close to $T_C$, $E_{ab}$ will be very different for the two initial magnetization states.

![Figure 8.1 The AOS model supposes an a) $N_x \times N_y$ grid of cells, which present b) temperature-dependent magnetic anisotropy energy barriers $E_{ab}$, resulting in c) two stable possible states a (up) and b (down). As a d) circularly polarized laser pulse arrives onto the grid, e) cells in a and b states will absorb different amounts of energy due to MCD inducing a temperature distribution in the grid of hot and cold cells. This will lead to f) different energy landscapes for the hot and cold cells, resulting mostly in stochastic switching of the hotter cells.](image_url)
Numerical simulations were conducted by considering an FePt-C-L10 granular film, for which AOS has been reported (79). The typical size of grains is around 5 nm wide and 7 nm thick (128), where grains are separated by a 1-nm-thick carbon matrix (129). This matrix ensures thermal isolation, as well as magnetic exchange isolation. We can therefore safely neglect the exchange interaction and assume that a nonhomogeneous temperature distribution can exist in the sample.

The temperature dependence of the magnetization $M_S$ and $K$ was extracted from Ref. (73), and corresponds to an Fe$_{50}$Pt$_{50}$ film. First, $M_S$ was fitted with phenomenological equation $M_{s0}[(T_\text{C}-T)/(T_\text{C}-T_0)]^\gamma$ where $M_{s0}=1.15\times10^6$ A/m is the magnetization at $T_0=300$ K, $T_\text{C}=775$ K, and $\gamma=0.34$ is the phenomenological fitting exponent used for Fe (93). Then $K$ was fitted with (73, 130) $K_0(M_S/M_{s0})^2$ where $K_0=4.5\times10^6$ J/m$^3$. The fits are shown in Figure 8.2.

The MCD was calculated, for a wavelength $\lambda = 810$ nm, by using the nonmagnetic complex index of refraction $n=3+4i$ and the complex nondiagonal term $\sigma_{xy}=-(1.4+1.7i)\times10^{14}$ s$^{-1}$ (cgs) of the optical conductivity tensor. These values were extracted from ellipsometry, Kerr rotation, and Kerr ellipticity measurements in Refs.(131, 132) through the relations reported in Refs.(131, 133). Through Maxwell equations, the complex index of refraction for left and right circular polarized light $n\pm$ are found to be $n\pm=(n^2 \pm 4\pi\sigma_{xy}/\omega)^{1/2}$, where $\omega = 2\pi c/\lambda$ and $c$ is the speed of light.
Fresnel equations were then used to obtain the reflectance and absorption $A_+$ and $A_-$ for both helicities in the case of an infinitely thick film and normal incidence. Finally, the MCD was calculated as $2(A_+-A_-)/(A_++A_-)$. An MCD of 5.8% is obtained for Fe$_{50}$Pt$_{50}$.

8.3 Results

We first compute Equation 8.4 as a function of the temperature, where the starting state a is heated more than b due to MCD. We begin with the calculated MCD = 5.8%, a cell volume of $5\times5\times7=175$ nm$^3$ (128), and $t_{hot}=1$ ns. As shown in Figure 8.3 for a single pulse ($N=1$, black line) the probability of switching is 0 below a certain temperature threshold and 0.5 above. No full switching is thus possible with a single shot. As we increase the number of shots, a narrow range of temperatures around $T_C$ results in a probability of switching that increases up to 1 eventually ensuring full switching. In this case the absorbed critical fluences for AOS will be around $F=\Delta T h C \approx 1.2$ mJ/cm$^2$, where $\Delta T=T_C-T_0$, $h=7$ nm is the thickness, and $C=3.5\times10^6$ J/(m$^3$K) is the heat capacity of FePt (51).

![Figure 8.3](image)

Figure 8.3 All-optical switching probability as a function of the laser temperature increase $\Delta T$ for different numbers of shots $N$. Three different temperature regimes are observed. At low temperature no switching is possible due to strong anisotropy barriers. Close to $T_C$ a certain amount of AOS occurs as the number of shots increases. With enough pulses full switching becomes possible. At higher temperatures the sample gets randomly demagnetized into a multidomain structure.
Since this model assumes a sudden step-like temperature increase in the sample, cooling dynamics are not taken into account. Cooling in the presence of strong dipolar fields would make a full switching process less probable. However, this argument is consistent with the observation (79) that only thin films, with a small magnetization volume and thus smaller dipolar fields, exhibit nearly full AOS. Thicker films always show some degree of demagnetization (multidomains), and full switching is not observed.

In this calculation, the ratio $t_{\text{hot}}/\tau_0$ that acts as a prefactor in the exponential of Equation 8.2 was set equal to 10. This parameter varies for different heat dissipation in the sample, but mostly offsets the temperature range at which AOS is observed, as shown in Figure 8.4.

![Figure 8.4 AOS probability after N = 100 shots, as a function of the temperature rise $\Delta T$ for $t_{\text{hot}}$ ranging from 1 ns to 10 ns. $\tau_0$ is kept constant at 0.1 ns.](image)

As shown in Figure 8.5, the temperature window for AOS becomes larger as the MCD increases. However, even for a small MCD value of 0.5%, some AOS is still possible in a narrow range of temperatures.
Next, the lateral spatial heating profile was assumed to be Gaussian, according to the laser intensity profile. The $1/e^2$ radius was set to 115 cells and the temperature rise to $\Delta T=600$ K (Figure 8.6.a) and $\Delta T=500$ K (Figure 8.6.b). As shown in Figure 8.6.a, with white and black corresponding to opposite magnetizations, one shot on an initially saturated grid results in a circular demagnetized pattern. As the number of shots increases, an outer ring, within the temperature window for AOS, fully switches. The fact that the first laser shots only result in a demagnetized area agrees with the observations on Co/Pt from Ref. (122). The final state, a ring domain with an inner demagnetized area, resembles strongly the AOS results in FePtC reported by Lambert et al (79).
Figure 8.6 Simulations of the magnetization state after assuming a Gaussian temperature increase induced by the laser intensity profile. a) Simulation of successive shots with left circularly polarized ($\sigma^-$) light on an initially saturated black (“down”) grid. After ten laser shots a fully switched white ring develops. b) Simulations scanning the beam with left circular ($\sigma^-$), right circular ($\sigma^+$), and linearly polarized ($\pi$) laser shots at a ten shots/cell sweeping speed. Initially, the grid consisted of a left-half up (white) magnetization and right-half down (black) magnetization. Each helicity favors one magnetization direction, which is determined by the edge of the Gaussian profile, whereas the linearly polarized laser beam only demagnetizes the sample.

We next scan the Gaussian beam (temperature profile) at a speed of one cell/$t_{rep}$ where $t_{rep}$ is the laser repetition period, and as shown in Figure 8.6.b, we are able to write a magnetic domain by swiping the outer edge across the grid. Initially the grid was set so that the left half of the grid was up (white) and the right one down (black). Left circular ($\sigma^-$) and right circular ($\sigma^+$) polarized light causes up and down magnetizations, respectively, whereas linearly polarized ($\pi$) light only demagnetizes the sample and results in multidomain states. This is an expected behavior when considering the Gaussian profile of the laser intensity and the existence of a helicity-dependent AOS threshold (27). In fact, this laser sweeping
technique was used to reveal the AOS in ferromagnets such as FePtC by Lambert et al. (79) and lead to qualitatively similar magnetic domain patterns. The switching time via this mechanism is obviously not ultrafast, in contrast to that found in ferrimagnets (76). Since the film needs to be heated and cooled multiple times before observation of a full switching, the number of pulses needed for switching, cooling rates, and the laser repetition rate will define the switching time. However, such minimum switching time should also be fundamentally limited by the time scales of the thermal excitations (given by $\tau_0$).

8.4 Discussion and conclusion
The suggested AOS mechanism should thus operate in materials with large MCD, small dipolar fields, limited in-plane heat diffusion, and a strong temperature dependence of the anisotropy close to $T_C$. We note that this does not restrict the mechanism to ferromagnets; thin ferrimagnetic films (such as TbCo, GdFeCo) or even antiferromagnetic materials are also strong candidates for this switching mechanism due to their small dipolar fields and significant MCD.

Finally, we would like to discuss the validity of the model for ultrathin ferromagnetic films such as Pt/Co multilayers. These films do have significant MCD and exhibit low dipolar fields, however, they lack the granular structure that allows for magnetic and thermal isolation. Nevertheless, exchange interaction is not necessarily detrimental for the AOS. In such materials, we provide the following qualitative description: The first laser shot demagnetizes the sample. The magnetization spontaneously breaks into domains of various sizes. Under negligible dipolar fields, domain relaxation is dominated by the wall energy and pinning. The smallest domains will thus disappear because shrinking forces induced by the wall energy increase as the bubble diameter decreases (97), whereas larger ones (100–1000 nm wide) will remain pinned. These larger domains will then remain cool on the next laser shot, while the rest of the film will restart the same process over. Repetition of this mechanism will eventually finish when various large domains merge together, resulting in full AOS.

In summary, we have proposed a multi-shot all-thermal mechanism for helicity-dependent AOS in magnetic materials, which is based on temperature distributions induced by the MCD when heating the films close to $T_C$. This mechanism could possibly coexist with other mechanisms such as the IFE (123, 124). We calculated the cumulative probability for AOS after a certain number of pulses, and numerically estimated it for the case of an FePtC granular film. The AOS window as
a function of MCD, temperature, and the number of pulses was presented showing that even with as little as 0.5% of MCD, multi-shot switching should still be possible. Finally, we simulated the resulting domain patterns after N laser shots, qualitatively reproducing various reported experimental results (79, 122).
Chapter 9

Single-shot ultrafast all optical switching of ferromagnets

9.1 Introduction

AOS and a plethora of other ultrafast phenomena has been associated to rapid heating of electrons following the absorption of an optical pulse (14, 22, 28, 134). Moreover, in previous chapters we demonstrated that picosecond electrical pulses could also serve the purpose of rapid electronic heating, extending the physics of ultrafast magnetism to the realm of spin electronics. This supposes a great opportunity for applications exploiting ultrafast magnetism physics.

However, so far, only GdFeCo ferri-magnetic films have reproducibly shown single-shot and ultrafast (~ ps) AOS. AOS in GdFeCo has been attributed to the ability of the sublattices to demagnetize at different rates and to exchange angular momentum with each other (76). In order to extend ultrafast magnetism into the field of spintronics, more materials with perpendicular magnetic anisotropy (PMA) presenting AOS are required.

Ferro-magnetic PMA materials such as Co/Pt, FePt, Co/Ni or CoFeB have all been widely studied. They have well known and reliable growth properties and present various advantages for applications such as very high thermal stabilities, high spin polarizations and/or high tunneling magneto resistance ratios. Some of these systems have also been widely studied for the possibility of controlling magnetic topological states (domain walls and skyrmions) via electrical currents and magnetic fields, which could also lead to ultrahigh density memories (135, 136). Unfortunately, ferromagnets that present AOS require up to hundreds of optical pulses for a complete reversal, making the overall mechanism rather slow and energy demanding (79, 122).

Here, we demonstrate single-shot ultrafast AOS of the magnetization in various ferromagnetic Co/Pt multilayer structures, which are coupled to a ferrimagnetic GdFeCo layer via a metallic spacer. By irradiating a single 70 femtosecond (FWHM) linearly polarized optical pulse on the stacks, we achieve the reversal of both ferrimagnetic and ferromagnetic layers. The ferrimagnetic layer drives the switching of the Co/Pt layer via the exchange interaction. We show that the Co/Pt multilayers can be either antiferromagnetically (AFM) or ferromagnetically (FM) exchanged coupled to the GdFeCo layer, and present AOS in all cases. Depth-
sensitive time-resolved magneto-optical experiments reveal that the reversal of the ferromagnetic layer happens within 7 picoseconds. Our approach of coupling GdFeCo with other magnets will extend greatly the possibilities for applications of ultrafast magnetism, as well as bringing new insights in the physics of AOS itself.

9.2 Methods
We grew different series of Ta/GdFeCo/Co/Pt(d)/Co/Pt stacks by magnetron sputter deposition on Si/SiO$_2$ (100 nm) substrates. The spacer thickness d was varied between 0 and 5 nm. The layer thicknesses were determined from the deposition rate of each material. All of the samples presented perpendicular magnetic anisotropy (PMA). The compensation temperature $T_m$, temperature at which the magnetic moment is minimized, determines the dominant magnetic moment, FeCo for $T_m<300$ K, and Gd for $T_m>300$ K.

The static magnetic properties of the stacks were characterized by performing hysteresis loops with a magneto optical Kerr effect (MOKE) microscope (Figure 9.1) and an out-of-plane magnetic field $H_\perp$. By tuning the angle of half-wave plate, depth-sensitive magnetization can be measured.

![Figure 9.1 The depth-sensitive MOKE microscope setup.](image)

To access time-resolved magnetization dynamics, TR-MOKE similar to what’s described in chapter 2 was used. Due to the stroboscopic nature of pump-probe experiments, the magnetization of both layers needs to be reset after each laser pulse. For this purpose, we patterned small Au coils on top of the films in order to
deliver short intense magnetic field pulses in between pump pulses, and avoid any constant external field that could affect the magnetization dynamics. We performed the experiment at 54 kHz and used 2 µs wide electrical pulses of 1 A in amplitude generated by an amplified waveform generator and synced via a delay generator triggered by a photodiode within the laser cavity. The 54 kHz repetition rate allows for ~16 µs of cooling time for the sample after the optical and electrical pulses, so that the initial state is always in equilibrium and at room temperature. All time-resolved curves in Figure 9.4 where obtained by performing the experiment for positive and negative magnetizations and taking the difference of the two signals in order to eliminate non-magnetic contributions to the signal.

![Figure 9.2 The depth-sensitive time-resolved MOKE setup.](image)

We use the static MOKE hysteresis loops to analyze the directions of each sublattice. First, a hysteresis of the bare film Gd$_{28}$FeCo(20 nm)/Ta(3 nm) is obtained (top hysteresis in Figure 9.3.c), which indicates that the compensation temperature is below room temperature. This conclusion is achieved by comparing the polarity of the hysteresis to previously characterized samples in Refs. (52). The dominant magnetic moment in the ferrimagnet is thus the FeCo moment at room temperature. We conclude that the highest coercivity (~100 Oe) in the weakly coupled stacks is given by the GdFeCo layer, by comparing the direction of the
coercive jumps (in other words, we compare the polarity of the hysteresis). Moreover, this is confirmed by the large Kerr signal (bigger jump) which we attribute to the thicker (20 nm) FeCo lattice.

Depth-sensitive MOKE was used to slightly increase the sensitivity to the thin Co/Pt layer. Single layer sensitivity is demonstrated in the top hysteresis loops of Figure 9.3.d, where distinct coercivities for each layer are distinguishable. We then fix the quarter wave plate at an angle, maximizing sensitivity to either layer, and perform hysteresis loops on all the samples. We repeat the experiment with a second angle of the quarter wave plate that provides high sensitivity to the other layer. These hysteresis loops are reported in Figure 9.3.d. We assume that the sensitivity to a single layer remains valid as the thickness of the spacer changes, as a non-magnetic spacer should not influence the complex Kerr signal. From the polarity of the cycles we extract the relative magnetization direction of the probed lattice.

To calculate the optical energy absorbed in the stack, we use a multilayer absorption calculation. The electric field inside the stack is obtained through the transfer matrix method (52). The absorption was then obtained by calculating the divergence of the Poynting vector inside the stack. The resulting absorption per layer is reported in Table 9.1. The absorption profile is shown in Figure 9.3. The whole stack absorbs 33% of the incident energy.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Thickness (nm)</th>
<th>Complex index of refraction</th>
<th>Absorption by layer (%)</th>
</tr>
</thead>
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<tr>
<td>air</td>
<td>-</td>
<td>1</td>
<td>-</td>
</tr>
<tr>
<td>Pt</td>
<td>3</td>
<td>2.85 + 4.96i</td>
<td>6.4</td>
</tr>
<tr>
<td>Co</td>
<td>0.6</td>
<td>2.50 + 4.84i</td>
<td>1.0</td>
</tr>
<tr>
<td>Pt</td>
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<td>2.85 + 4.96i</td>
<td>8.6</td>
</tr>
<tr>
<td>Co</td>
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<td>2.50 + 4.84i</td>
<td>0.5</td>
</tr>
<tr>
<td>GdFeCo</td>
<td>23</td>
<td>2.66 + 3.56i</td>
<td>16</td>
</tr>
<tr>
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</tr>
<tr>
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<td>0</td>
</tr>
<tr>
<td>Si</td>
<td>-</td>
<td>3.70 + 0.005i</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 9.1 Multilayer calculation parameters and absorption per layer.
9.3 Results

We first study a series of PMA samples where a single Co/Pt layer is grown on a GdFeCo/Co structure, separated by a Pt spacer of thickness $d$, as depicted in Figure 9.4.a. The thin Co layer adjacent to the GdFeCo is strongly coupled to the FeCo lattice, effectively modifying the composition of the alloy. In the following, we will refer to the FeCo/Co system as simply FeCo. Hysteresis loops are shown in Figure 9.4.c. Four remnant states are present in samples $d=4$ and 5 nm, and only two in the strongly coupled $d=1.5-3$ nm. We add a quarter wave plate in the optic path to enable depth-sensitive MOKE (137), which allows us to obtain layer sensitivity, as demonstrated in the loops of Figure 9.4.d. The polarity of the loops allows us to infer the directions of the magnetic moments in the stack. We indicate the direction of the Gd, FeCo and Co/Pt sublattice magnetizations by orange, green and blue arrows respectively. We note that the net moment of the GdFeCo/Co structures is dominated by the FeCo lattice.
Figure 9.4 a) GdFeCo/Co/Pt(d nm)/Co stack series with Pt spacer of thickness d. b) Schematic of exchange interactions between magnetic sublattices. c) Magnetic hysteresis loops for (top) bare GdFeCo(20 nm)/Ta(3 nm) and the GdFeCo/Co/Pt/Co stacks with different Pt spacer. Green, orange and blue arrows represent Gd, FeCo and Co/Pt magnetizations, respectively. Minor loops switching only the Co/Pt magnetization, presenting a positive exchange bias $H_b$, on samples d=4 and 5 nm. d) Depth sensitive MOKE hysteresis loops of the stacks, with maximum sensitivity to FeCo (green) and Co/Pt (blue) magnetic lattices.
Figure 9.5 Minor hysteresis loop of the Co/Pt layer in a Gd rich sample Si/SiO₂(100nm)/Ta(5nm)/(Gd₃₆Fe₅₇.₆Co₆.₄)(20 nm)/Pt(4.1nm)/Co(1.2nm)/Pt(5nm). Blue, green and orange arrows represent Co/Pt, FeCo and Gd magnetizations. Note the opposite sign of the exchange bias $H_b$ shift with respect to the one observed in FeCo rich samples (Fig 9.4.c). The Co/Pt layer is thus coupled with the sublattices instead of the net moment, just as expected for interlayer exchange coupling.

Next, we characterize the type of the interlayer coupling of the stacks. We obtained the coupling for samples d=4 and 5 nm by performing minor hysteresis loops, where only the Co/Pt magnetization is switched (blue circles in Figure 9.4.c. The interlayer bias field $H_b$ corresponds to the shift of the minor loop. The GdFeCo and Co/Pt net moments present an AFM coupling for thick spacers, as was found in Refs. (138–140). An RKKY-type of exchange or a dipolar orange peel coupling (138, 141) could explain such AFM coupling. However, in Gd dominated samples, $H_b$ has an opposite sign (Figure 9.5), demonstrating that the coupling does not follow the direction of the net moment, but rather the orientation of the sublattices. In addition, as shown by the magnetic moments depicted in Figures 9.4.c and d the coupling changes sign and becomes FM as the spacer is made thinner. We thus attribute the coupling to an RKKY-type of exchange as reported in Refs. (138–140), and assume any dipolar coupling to be negligible. The exchange between the different sublattice spin baths is depicted in Figure 9.4.b. The Gd and FeCo
sublattices in GdFeCo are AFM coupled via two direct exchange interactions $J_{5d-4f}$ and $J_{5d-3d}$ (142). Since most of the conduction in GdFeCo is carried by the FeCo sp electrons and conduction electrons are responsible for the exchange interaction, in the following we will refer to the interlayer coupling ($J_{3d-3d}$) as the one between the FeCo sublattice and Co/Pt. This is consistent with previous reports on TbFeCo/[Pt/Co] (143), TbFe/[Pt/Co] (144) bilayer and GdFe/Ta/DyCo (145) trilayer systems.

![Figure 9.6](image)

Figure 9.6  a) The schematic of experiment with GdFeCo/Co/Pt(d nm)/Co stack series presented in Fig 9.4. b) MOKE images of a sequence of AOS events on films with different Pt spacers. Green, orange and blue arrows represent Gd, FeCo and Co/Pt magnetizations, respectively. c) Absorbed critical fluence $F_c$ for AOS (in black), $F_r$ for relaxation of parallel states (in red), corresponding to the dotted lines in Fig 9.6.b, and estimated peak lattice temperature rises $\Delta T$ (in blue) as a function of the Pt spacer thickness.

We then check the all-optical switching capabilities of all the samples by irradiating the Co/Pt side (depicted in Figure 9.6.a). MOKE micrographs of the AOS are shown in Figure 9.6.b. The samples are initialized via a positive external magnetic field into a state where the FeCo and Co/Pt magnetizations are parallel. The magnetic field is then turned off. After the first single laser shot on sample $d=5$nm we observe two
new regions of different contrast, a centered white circle and the surrounding grey ring (delimited by the white dashed line). The new regions present opposite anti-parallel alignments of FeCo and Co/Pt. In the center area, the laser intensity is above the critical fluence $F_C$ for AOS, which results in the reversal of the GdFeCo magnetization. The Co/Pt magnetization remains in its initial orientation in order to relax the structure into a more stable AFM state. In the surrounding ring, the fluence is below threshold for AOS and GdFeCo does not switch. However, the hot Co/Pt layer does switch in order to relax into the AFM state. There is thus a second threshold $F_C$ for the relaxation of the parallel state to antiparallel. For even lower fluences, the heating is insufficient to generate any observable changes. When the sample is shot with a second laser pulse, the magnetization in the center region switches fully, reversing both GdFeCo and Co/Pt moments. The ring region remains stable after the first shot, as it is already in a stable AFM state. We then repeat the experiment for different thicknesses $d$ of the Pt spacer. Sample $d=4$ nm presents the same type of switching as sample $d=5$ nm. For samples $d=1.5$, $2$ and $3$ nm, where the coupling is FM, we observe only the switching of the center area, as expected, as no more remnant states are available.

We illuminate the samples repeatedly and obtain reliable toggle switching of both ferromagnetic and ferrimagnetic layers for up to more than 100 pulses. The switching is reproducible in all FM and AFM coupled samples. This therefore presents the first demonstration of a single-shot AOS of a ferromagnetic film.

The absorbed critical fluences $F_C$ for AOS, for relaxation as well as the estimated equilibrium lattice temperature rises $\Delta T$ are reported in Figure 9.6.c by black, red and blue points, respectively. The transient equilibrium temperature rise for the whole stack is calculated by the following formula, $\Delta T = \frac{F_C}{C \cdot t'}$, where $F_C$ is the critical fluence, $C \sim 2.9$ MJ/(Km$^3$) is the weighted heat capacity of the full stack (using the heat capacity of GdFeCo of 3 MJ/(Km$^3$) from Ref. (52)), and $t'$ is the total thickness of the stack. We obtain a temperature rise of $\sim 150$ K for all films, irrespective of the type of coupling. The Co/Pt is thus heated to $\sim 450$ K, very close to its expected Curie temperature of $\sim 470$ K. This indicates that for AOS to be possible the Co/Pt needs to be nearly fully demagnetized. Another possible mechanism to consider for switching are hot electron spin currents between the layers during demagnetization. If present, spin currents should have a different sign for parallel and antiparallel alignments, resulting in different critical fluences. Because $F_C$ for AOS is equal for AFM and FM coupled films, we believe that spin currents are probably not relevant to the switching mechanism.
The MOKE micrographs in Figure 9.6.b provide clear evidence of the AOS of Co/Pt, but cannot provide information on whether we are achieving ultrafast control over the exchange interaction, which is the ultimate goal of our study. In order to access the fast magnetization dynamics, we perform depth-sensitive time-resolved MOKE measurements with no external magnetic field. For this purpose, we grow a second series of two PMA samples with an extra Co/Pt repeat (Figure 9.7.a), in order to increase the sensitivity to the Co/Pt layer. A patterned Au coil grown on top of the magnet was used to deliver short intense magnetic field pulses at the laser repetition rate of 54 kHz to repetitively reset the magnetization during pump-probe experiments. The Pt spacer thickness $d$ of the samples was chosen equal to 1.5 and 4 nm, to obtain a strong FM and a weaker AFM coupling, respectively. We confirmed the AOS of the full stack in both samples.

Figure 9.7 a) GdFeCo/Co/Pt($d$ nm)/[Co/Pt]$_2$ stack series with Pt spacer of thickness $d$. b) Depth-sensitive MOKE magnetic hysteresis loops on sample $d=4$ nm. c) Depth-sensitive time-resolved demagnetization curves for antiparallel (AP) or parallel (P) initial states of the stack $d=4$ nm. Green, orange and blue arrows represent Gd, FeCo and Co/Pt magnetizations, respectively. d) Depth-sensitive demagnetization and AOS experiments at various fluences on sample $d=1.5$ nm.
We then perform low fluence demagnetization experiments in order to check the differences in demagnetization for a sample in an antiparallel (AP) vs a parallel (P) state. For this purpose, we work on sample d=4 nm which has 4 remnant states. We first select the quarterwave plate angles in order to maximize layer sensitivity to the Co/Pt and FeCo moments, as shown in Figure 9.7.b. By working at low fluence we avoid the full reversal of the Co/Pt layer, which has too high of a coercivity (~500 Oe) for the field delivered by the patterned coil. The results of the demagnetization are shown in Figure 9.7.c. The Co/Pt and FeCo peak demagnetizations are similar in both parallel (P) and antiparallel (AP) cases, but the long timescale (~2-20ps) dynamics of the Co/Pt magnetization are very different. We attribute the slower recovery of the parallel case to the intrinsic AFM coupling field of the stack that pulls the Co/Pt against the anisotropy field. If spin currents were relevant during demagnetization, they should be maximized during the fast demagnetization and have opposite signs for P and AP cases. The similar peak-demagnetization amplitudes for both P and AP cases indicate that spin currents do not play a major role.

Finally, we perform time-resolved switching experiments. Unfortunately, these experiments could not be carried on sample d=4 nm because the coercivity of the Co/Pt layer is larger than the amplitude of the magnetic field we can generate with the patterned coils. We thus perform the AOS experiments on a thinner d=1.5 nm sample, where both layers are strongly FM coupled and present a single and smaller coercivity (H~150 Oe). We vary the incident average power from 40 to 59 mW. As shown in Figure 9.7.d, the dynamics of the FeCo and Co/Pt layers are quite different. At the lowest power, 40 mW, we only obtain a demagnetization of both FeCo and Co/Pt magnetizations. Increasing the power to 53 mW results in the switching of FeCo within ~3 ps, during which the Co/Pt demagnetizes nearly completely. The magnetization of Co/Pt recovers during the next 4 ps as the system cools down. Eventually, due to the exchange field induced by the FeCo sublattice, the Co/Pt magnetization switches after ~30 ps. At even high power, 59 mW, the FeCo demagnetizes first and then grows in the opposite direction, switching in only ~7 ps.

The two-steps of the 7 ps switching event of Co/Pt shown in Fig 9.7.d, - an initial full demagnetization and a subsequent switching - strongly supports the idea that the exchange interaction is responsible for the reversal of the softened (hot) Co/Pt magnetization. The curves at 53 and 59 mW indicate that the fastest switching
occurs when the fluence is such that the temperature of the Co/Pt reaches exactly (or very close to) $T_C$.

9.4 Conclusion
We have shown that, despite using thick (up to 5 nm) metallic spacers with strong spin-orbit coupling, such as Pt, to separate a Co/Pt multilayer from GdFeCo, we can still achieve a single-shot 7 ps AOS of the ferromagnetic layers by exploiting the exchange interaction. This general method can be extended to other ferromagnets, ferrimagnets and antiferromagnets. As shown in Refs.(116, 146), adding a Co ferromagnetic layer to GdFeCo can improve the TMR from 0.5% to about 40%. The stacks we studied are particularly promising, since they offer a way to largely increase the GMR and TMR ratios of GdFeCo based junctions, opening a new avenue for ultrafast opto-spintronic applications. In addition, the possibility of rapidly switching ferromagnets with lasers and without magnetic fields might be highly appealing for the hard-drive industry, in particular for their laser-based heat-assisted magnetic recording technology. Furthermore, our approach opens up the possibility of probing various layers independently during AOS which could lead to a better understanding of the physics of AOS in films with multiple magnetic lattices.
Chapter 10
Conclusions and future work

We have achieved the ultrafast control of magnetic order with picosecond electrical pulses. Like femtosecond laser pulses, picosecond electrical current pulses are also capable of introducing ultrafast demagnetization in ferromagnets. The ultrafast demagnetization dynamics induced by electrical pulses are a distinct from caused by laser pulses, featuring a different electron distribution upon the ultrafast excitation. Despite the different dynamics, ultrafast electrical pulses are also capable of reversing the magnetization of GdFeCo on a picosecond timescale due to the rapid heating of the GdFeCo electrons. The ultrafast electrical switching of GdFeCo is thus the fastest magnetic switching through electrical methods and corresponds to a totally new electrical switching mechanism without involving any spin transfer or orbit torques. These results will hopefully open up a new regime of ultrafast spintronics.

In parallel, we have achieved the ultrafast, single-shot, all-optical switching of ferromagnetic films by exploiting the exchange between ferromagnetic films and GdFeCo films.

Looking forward, we would like to demonstrate the ultrafast electrical switching of GdFeCo–ferromagnet hybrid films, which will serve as a general method to control ferromagnets with ultrafast electrical pulses. Building functional spintronic devices, such as MTJs, with GdFeCo–ferromagnet hybrid films is another crucial step in making the ultrafast electrical control of practical use. Ultimately, the integration of such spintronic devices with CMOS transistors will demonstrate a truly practical ultrafast spintronic system.
Reference


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