Resonant switching and vortex dynamics in spin-flop bi-layers

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Abstract

This thesis is a study of the static and dynamic behavior of the magnetization in spin-flop bi-layers, which consist of two soft ferromagnetic layers coupled by dipolar forces through a thin nonmagnetic spacer. The focus of the work is three fold: collective spin dynamics in the anti-parallel ground state; resonant switching in the presence of thermal agitation; and static and dynamic behavior of the system in the vortex-pair state, with a particular emphasis on the interlayer core-core interaction.

Two collective spin-flop resonance modes are observed and interpreted as acoustical and optical spin precessions, in which the moments of the two layers oscillate in phase and out of phase, respectively. An analytical macrospin model is developed to analyze the experimental results and is found to accurately predict the resonance frequencies and their field dependence in the low-field anti-parallel state and the high-field near saturated state. A micromagnetic model is developed and successfully explains the static and dynamic behavior of the system in the entire field range, including the C- and S-type spin-perturbed scissor state of the bi-layer at intermediate fields.

The optical spin-flop resonance at 3-4 GHz is used to demonstrate resonant switching in the system, in the range of the applied field where quasi-static switching is forbidden. An off-axis field of relatively small amplitude can excite large-angle scissor-like oscillations at the optical resonance frequency, which can result in a full 180-degree reversal, with the two moments switching past each other into the mirror anti-parallel state. It is found that the switching probability increases with increasing the duration of the microwave field pulse, which shows that the resonant switching process is affected by thermal agitation. Micromagnetic modeling incorporating the effect of temperature is performed and is in good agreement with the experimental results.

Vortex pair states in spin-flop bi-layers are produced using high amplitude field pulses near the optical spin resonance in the system. The stable vortex-pair states, 16 in total, of which 4 sub-classes are non-degenerate in energy, are identified and investigated using static and dynamic applied fields. For AP-chirality vortex-pair states, the system can be studied while the two vortex cores are coupled and decoupled in a single field sweep. It is found that the dynamics of the AP-chirality vortex pairs is critically determined by the polarizations of the two vortex cores and the resulting attractive or repulsive core-core interaction. The measured spin resonance modes in the system are interpreted as gyrational, rotational, and vibrational resonances with the help of the analytical and micromagnetic models developed herein.

A significant effort during this project was made to build two instruments for surface and transport characterization of magnetic nanostructures: a high-current Scanning Tunneling Microscope for studying transport in magnetic point contacts, and a Current In Plane Tunneling instrument for characterizing unpatterned magnetic tunnel junctions. The design and implementation of the instruments as well as the test data are presented.
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TO MY FAMILY
List of abbreviations

AC alternating current
ADC analog to digital converter
AF antiferromagnetic
AMR anisotropic magnetoresistance
AP antiparallel state
BL bit line
CIP current in plain
CIPT current-in-plane tunneling
CPP current perpendicular to the plain
DC direct current
DOS density of states
DSP digital signal processor
DW domain wall
EA easy axis
eq. equation
F, FM ferromagnet, ferromagnetic
FPGA field programmable gate array
GMR giant magnetoresistance
HA hard axis
HF high frequency
HV high voltage
I/V current-voltage characteristics
MR magnetoresistance
MRAM magnetic random access memory
MTJ magnetic tunnel junction
NM non-magnetic
P parallel state
PSD power spectral density
RF radio frequency
RL read line
RT room temperature
sec. section in the thesis
SD single domain
SF spin-flop
SS scissored state
STM scanning tunneling microscope
STT spin transfer torque
SW Stoner-Wohlfarth model, switching etc.
TMR tunnel magnetoresistance
TJ tunnel junction
WL word line
Chapter 1

Introduction

Magnetic recording of information has been around for a long time. Since 1894, when the Telegraphone was discovered by a Danish telephone technician Valdemar Poulsen, the technology of magnetic recording has been developed much further, with the recording density as well as the read/write speeds constantly improved. The evolution of this technology went from the invention of magnetic wire-based sound recorders to the latest hard drives with the storage capacity of over 1 TB and, recently, to non-volatile Magnetic Random Access Memory (MRAM). The discovery of the Giant Magneto-Resistance (GMR) in 1988 was a revolution, which started a great amount of basic and applied research worldwide. The majority of the research is geared towards the use of spin-based electronics in data storage and magnetic field sensing applications. The invention of GMR-based hard-drive read heads allowed to significantly increase the information density and set the stage for the development of MRAM. Historically the first type of MRAM had the Stoner-Wohlfarth (SW) [1] configuration, which didn’t become a successful commercial product due to issues with the magnetic bit stability. The work on MRAM continued and led to the invention of Toggle-MRAM in 2003 [2], which became a commercial product released by Freescale in 2006. Further advances in magnetic recording continued with the introduction of perpendicular-to-the-plane magnetic recording and Tunnel Magneto-Resistance (TMR) based read heads in 1 TB/in.sq. disc drives [3]. A great amount of research in the field is currently focused on Spin-Transfer-Torque [4] and Race-Track MRAM [5] invented at IBM in the last decade.

Today’s computer random access memory (RAM) utilizes a storage principle where the bits of information are stored as charges on small capacitors. This technology is known as CMOS memory and is highly scalable. However, it has a disadvantage as regards volatility – information loss after power down; frequent refreshing of the storage capacitors is necessary during operation, which leads to a relatively high power consumption. In magnetic types of memory, such as MRAM, information is stored in the form of the orientation of magnetization in the storage
element and the readout is performed using the TMR effect. MRAM is non-volatile and does not require refreshing. The recently developed MRAM has, in addition, nanosecond range write/read cycles, which makes it a candidate to replace not only flash-RAM but also some DRAM solutions in various applications.

Accompanying the market introduction of MRAM much attention is being paid to studying the high frequency properties of magnetic nanostructures used as storage elements in MRAM cells, since the operation frequency of today’s memory is well over 100 MHz. Designing memory cells with sub-micrometer dimensions and operating them at near-GHz frequencies is challenging. The magnetic storage element size usually exceeds the true single domain limit and, therefore, the behavior of its magnetization strongly depends on the material properties as well as the geometry. Thus, soft nano-particles of elongated in-plane shapes usually exhibit a single domain-like configuration at equilibrium, whereas nearly circular particles often exhibit non-uniform spin states, such as the vortex state. The error-free operation of MRAM at high speeds requires fine tuning the amplitude and timing of the applied field. This forces the magnetization to precess at high speeds around its equilibrium position and can lead to unwanted reversal in the situation where the precession is not well damped. On the other hand, the effect of the magnetization precession can be used to assist the magnetization reversal and decrease the amount of current needed for switching the memory cell. Since the computer memory usually operates at and slightly higher than 300 K, thermal agitation can influence the magnetization dynamics. Therefore, a thorough understanding of the dynamic behavior of various magnetization states in magnetic nanostructures under thermal agitation is necessary for improving and developing new high-speed magnetic memory devices.

This thesis is an in depth study of spin-flop bi-layers used in MRAM. The second focus of this work is novel measurement techniques and instrumentation for characterizing magnetic nanostructures in terms of surface morphology and magneto-transport. The thesis is organized as follows.

Chapter 2 describes the design and implementation of two instruments for surface and transport characterization of magnetic nanostructures developed within this thesis project. A Scanning Tunneling Microscope (STM) capable of high-current magneto-resistance (MR) measurements is described and the test data obtained using this instrument are presented. In addition, an instrument for micrometer scale electrical measurements on unpatterned tunnel junctions utilizing a multi-tip probe is described. The functional elements as well as the software developed for this instrument are discussed. The main operation mode is Current-In-Plane Tunneling (CIPT), which allows measurements of MR on unpatterned magnetic tunnel junctions (MTJ) using micrometer-sized four- and twelve-tip probes. This technique allows fast, simple and nondestructive characterization of MTJ’s without time-consuming and expensive lithography steps.

Chapter 3 is an introduction to spin dependent transport, micromagnetism, and magnetization dynamics in nanostructures.

Chapter 4 reviews the basics of the Stoner-Wohlfarth and Spin-flop (or Toggle)
MRAM, as well as the macrospin theory of the magnetization dynamics in spin-flop bi-layers. The effects of the strength of assisting microwave field and that of thermal agitation on resonant magnetization reversal are discussed.

Chapter 5 discusses vortex-pair states in spin-flop bi-layers, with an emphasis on their dynamic properties.

Chapter 6 describes the samples used in this study, the measurement and simulation techniques, and the data analysis employed.

The last Chapter summarizes the original results obtained in this work, which are detailed in the Papers appended.
Chapter 2

STM and CIPT instruments

Electrical characterization of the surface and interface properties in thin films and multilayers is an important step in developing applications based on such multilayers. The transport properties of magnetic tunnel junctions make them very attractive for applications in data storage [6]. The quality of a tunnel junction is determined by its uniformity, desired resistance-area (RA) product and MR ratio, which need to be controlled continuously during the fabrication and integration process. When a tunnel junction device is made using a multi-step lithographic process, the quality of the tunnel barrier can be discovered only at the last stage of the fabrication. Thus, valuable time and resources can be spent on process integration while the tunnel barrier is sub-optimal.

Part of this thesis was to develop instrumentation for characterizing thin film samples with little or no processing. The work focused on two interrelated projects: a scanning tunneling microscope having the capability to make high current MR measurements, and a current-in-plane tunneling instrument for measuring unpatterned MTJ multilayers. A variety of electrical measurements could be performed with these instruments; however, taking into account our specific interest in spin-dependent transport, the design was geared toward measuring GMR in spin-valve structures and TMR in MTJs.

2.1 High-current STM

STM is a well known technique for studying surface morphology and electronic structure [7]. The morphology can be measured with a very high spatial resolution using this technique. STM’s are also capable of extremely localized transport measurements on nanostructures [8–10]. The configuration of a typical STM is for surface topography and basic I/V characterization, limited to the current range of 100 nA. This range is typically insufficient for studying the transport properties in low-resistive devices, such as spin-valves and spin-torque-driven magnetic point contacts, currently of interest in the field of spintronics. We therefore designed and
produced in-house an STM with the capability to make magneto-resistive measurements up to 25 mA in current and in fields of up to ~1 kOe.

Studies of transport in MTJs or spin-valves are usually conducted on samples patterned by lithography into nanopillars. In order to measure transport, a current is sent through the nanopillar. A thick underlayer typically serves as the bottom electrode and there are two possibilities for the top contact - a lithographic contact produced using a multi-step process, or a surface probe capable of locating the pillar and performing the desired electrical measurements. Integrating the top contact requires a considerable effort, which makes the surface probe technique an attractive alternative.

The room-temperature STM design presented below allows characterization of magnetic multilayers in an efficient way, requiring only a relatively simple lithographic patterning of the multilayer film into an array of pillars. This avoids the demanding stage of top contact fabrication. The low-current STM mode is used to scan the surface and locate individual pillars, which is followed by contacting the selected pillar and performing an electrical measurement in the specially designed high current mode. For example, a magneto-resistance measurement can be made by applying a fixed current through the pillar and varying the magnetic field in the sample plane. The instrument is especially suitable where a combined optimization of morphology and transport is desirable; e.g. pillars can be lithographically produced using a dose gradient and STM-characterized to determine the area as well as the magneto-transport properties.

<table>
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<th>Technical Specifications</th>
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A schematic of the instrument is presented in Figure 2.1. The structure of the microscope does not differ much from those commonly used [11]. The microscope consists of 3 main blocks: a controller box; a scanning head incorporating the approach mechanism and current preamplifiers; and a power supply block providing power to all elements of the microscope, including the HV drive voltages for the piezos. The key element is the controller, which contains a Signal Ranger DSP [12] board running a real-time kernel. The main tasks of the TMS 320VX5402 100MHz DSP are controlling the eight built-in 16-bit analog outputs/inputs, generating the
2.1. HIGH-CURRENT STM

For the same reason, high voltage amplifiers were placed in the same box as the power supply. All parts of the scan head are made from nonmagnetic materials in order to provide magnetic field compatibility and avoid drifts during magnetic field sweeps. Manual positioning of the sample holder provides for rough positioning of the desired sample within a 8x8 mm area. The monolithic base, holding the positioning stage, serves as a support for the electromagnet capable of generating magnetic fields up to 800 Oe in the sample plane. The scanning head, which has a tripod design, rests on three titanium legs above the sample holder, which makes no mechanical contact with the magnet poles. The head-mounted two-range tunnel current preamplifier performs current measurements in two modes: a low-current mode for scanning the surface with the range of up to 100 nA, and a point contact mode with the current range of up to 25 mA. Low noise is achieved in the high-current mode by using DSP-based digital lock-ins operating at the first and second harmonics. The technical specifications of the instrument are shown in Table 2.1.

Our aim was to develop an instrument with a high level of design flexibility and the capability of easily modifying any part of the hardware or the software block. This being the case, for STM operations, we chose the Open Source GXSM software [13]. This package provides high quality software for scanning the surface, conducting transport measurements, and visualizing the data obtained. It contains an Open Source DSP kernel for real-time feedback during operation [14]. The ability to access low level DSP functions opens a wide range of possibilities for creation of special operation modes and further modernization of the instrument. Due to its flexibility and Open Source nature, the STM is very adaptive to any
particular task with only a basic knowledge of computer programming required. In
the present work, the software side was used without any significant changes except
for a small software add-on allowing control of the magnetic field.

The full experimental setup is depicted in Figure 2.2. In many respects, the
design of the microscope is not original, as many ideas were borrowed from instru-
ments designed in other groups. In particular, the slip-stick approach drive was
adopted from [15] and ref [16] was used as a concept for the design of the tun-
nel preamplifier. The low thermal drift of the microscope and the use of proper
vibro-isolation guarantee the quality of the measurements. The thermal drift of
the instrument was determined by taking a number of sequential scans of the same
area and measuring the shift of the area over time.

The operating principle of the STM requires a conductive tip (as well as sample),
so the tips are usually made of a highly conductive, stiff, chemically stable material
such as Pt/Pd alloy or tungsten wire. The easiest way to produce atomically
sharp probes is by simply cutting a conductive wire. This method is commonly
used in microscopy of thin films where there is no need for tips with high aspect
ratio. Imaging highly profiled surfaces, like those produced by e-beam lithography,
requires tips of high aspect ratio. We made such tips by wet etching a tungsten wire
[17], the method we found to be fast and sufficient to achieve a high tip curvature
of down to 10 nm, as shown in Figure 2.3(b). Wet etching uses a NaOH solution,
with the tip electrically biased as shown in Figure 2.3(a). The etching is continued
until the bottom portion of the wire tears off from the top. The tearing breaks
the electrical circuit and as a result the etching process stops. This method is
self-controlled and does not require any electronic monitoring for determining the
end point. It is simple, yet produces very sharp tips. Different tip curvatures
can be produced by adjusting the amount of NaOH in the solution, tuning the
applied current, and changing the length (and mass) of the tungsten wire beneath
the etching area. Example Scanning Electron Microscopy (SEM) images of the
2.1. **HIGH-CURRENT STM**

![Figure 2.3](image)

Figure 2.3: STM tip made from tungsten wire by electrochemical etching. (a) Schematic of the etching setup. (b, c) SEM images of the tip apex.

![Figure 2.4](image)

Figure 2.4: STM scans of a Permalloy film. (a) 1x1µm and (b) 250x250 nm.

tungsten tips produced in this way are shown in Figure 2.3(b), 2.3(c).

Although there is a small amount of noise and image distortion, the resolution of the developed instrument is still sufficient for locating grains on a sample surface with lateral dimensions down to 20 nm, as shown in Figure 2.4. On the other hand, the maximal scan range of the STM (greater than 10x10 µm) is big enough to scan larger structures and locating nanoparticles or nanopillars over the whole area.

Figure 2.5 shows SEM (a) and STM (b) images of an array of 150x100 nm nanopillars fabricated using e-beam lithography. The scan range and resolution of the designed STM is sufficient for scanning the sample over a large area containing a number of nanopillars, locating a single pillar and contacting it. The sample in this experiment was prepared using a standard e-beam patterning technique [18], whereby the actual structure written by e-beam is transferred to the magnetic multilayer by reactive ion etching. During the etching process, some material from
CHAPTER 2. STM AND CIPT INSTRUMENTS

Figure 2.5: (a) SEM and (b) STM images of an array of e-beam patterned spin-valve pillars.

the etched areas was redeposited on top of the pillars, forming a conductive crown-like formation. This structure was resolved by both SEM and STM microscopes. An array of pillars with a crown formation at the edges and a plateau in the middle of each nanopillar was observed by STM. The sharpness of the STM tip is sufficient for contacting an individual pillar without contacting its neighbors.

Transport measurements of an individual pillar can be unstable due to the presence of adsorbed water or a thin oxide layer on the sample surface leading to an unstable electrical contact. Although it was found experimentally that gold capping is sufficient for contacting and measuring magnetic properties of unpatterned films, the deposition of a thin layer of gold on the top of magnetic pillars did not improve the stability of the electrical contact. The presence of the remaining baked resist, being an insulator, potentially impaired the electrical contact. As shown in ref [19], a Ru capping layer is optimal for providing a stable electrical contact between the conductive probe and the nanopillar being measured. As an example, Figure 2.6 shows the in-plane magnetoresistance of a Permalloy spin-valve film recorded in the high-current STM mode. In this measurement the magnetic film was covered by a thin gold layer which provided a good electrical contact and protected the sample surface from oxidation.

In summary, a scanning tunneling microscope was developed with a high-current point contact mode and variable magnetic field. The STM is based on software developed by the Open Source GXSM project and custom-made hardware. The spatial resolution of the microscope allows imaging and contacting lithographically produced nanopillars with high precision. Examples of morphology images and magneto-transport measurements are presented.
2.2 CIPT INSTRUMENT

Developing new magnetoresistive devices based on magnetic tunnel junctions involves optimizing new materials, processes, and device designs, and benefits greatly from fast turnaround measurements. Thus, the magnetoresistance and the resistance-area product of a junction are very informative and need to be measured frequently. These properties characterize the quality of the barrier and its suitability for a particular device. Using the full cycle of lithography for fabricating fully integrated junctions with top and bottom contacts is a large process overhead when the basic properties of the tunnel junction need to be determined in a large number of samples. The measurement procedure can be greatly simplified by patterning the magnetic multilayer film into an array of nanopillars for measurements by an STM instrument, as described in the previous section. However, even this simplified procedure requires a non-trivial step of patterning the tunnel junction material lithographically, which can lead to degradation of the sample. For example, the tunnel barrier can be shorted by redeposited material during the etching stage. The problems mentioned above can be solved by using the recently developed current-in-plane tunneling technique [20]. This method is based on measuring the resistance of unpatterned MTJ stacks and employs micrometer sized multi-tip probes [21] having different tip-to-tip distances. This probes was originally developed for electrical characterization of surfaces in the semiconductor industry [22–24]. Generally, macroscopically spaced tips probe the resistance of the bulk only. A current fed through a pair of surface contacts penetrates the sample to roughly the same depth as the contact spacing, making surface resistance measurements impossible when
CHAPTER 2. STM AND CIPT INSTRUMENTS

Figure 2.7: Electrical current fed through a pair of surface contacts penetrates the sample to roughly the same depth as the contact spacing; for this reason a small spacing is required to achieve surface sensitivity.

the tips are widely spaced. Thus, when the electrical properties of the surface are of interest, the spacing between the probing tips should be reduced significantly. Figure 2.7 illustrates the current flow in a sample for different spacings between surface contacts.

When electrical probes placed on top of a multilayer film are close to each other, the current flows predominantly through the top layer, making a separate measurement of the top layer resistance possible. If the spacing between the probes is relatively large, the current flows through the entire multilayer stack, and so the resistive properties of the top layer cannot be separated from the signal from the bottom layers. A 4-point probe measurement of the surface resistance was successfully implemented using a multi-tip STM [8, 25]

The CIPT technique can be used to measure the MR and RA values of un-patterned magnetic multilayers, which is accomplished by conducting a series of 4-point resistance measurements with varying spacing between the tips and subsequently fitting the data to a theoretical model [20]. An external magnetic field is used to pre-set the MTJ sample into the high- or low-resistive state, corresponding to the anti-parallel and parallel alignment of magnetization in the MTJ layers. For each alignment of magnetization the low and high resistance values and therefore the MR are measured. Thus, RA and MR of the tunnel barrier are obtained by fitting the measured $R(x, B)$ to the CIPT-model [20]. The resistance is measured using standard CIPT probes, consisting of a linear array of 4 or 12 cantilever-like tips separated by various (unequal) spacings, typically in the range of 1.5 - 20 µm. The use of a multiplexer allows any of the 12 micromachined tips to be addressed and set as a current source or voltage measurement contact in a 4-point resistance measurement. This scheme allows to choose any probe spacing ranging between 1.5 and 20 µm. As previously mentioned, in the case of relatively small separation between the tips, the current flows mostly through the top layer, since the effective tunnel barrier resistance of the area under the tips is significantly larger than the resistance of the top metal layer section of length equal to the tip spacing. When the spacing between the tips is large, then the effective tunnel barrier resistance
2.2. CIPT INSTRUMENT

Figure 2.8: (a) Resistor network model of CIPT. (b) Probe spacing dependence of the calculated resistance per square and MR. After D. C. Worledge [20]

is low and the current flows through both the top and the bottom metal layers in proportion to their respective resistances. In this case, essentially no magnetoresistance can be measured, due to a very small contribution from the Tunnel Junction (TJ) resistance to the total in-plane resistance of the MTJ stack. However, at some intermediate spacings between the tips the TJ resistance is comparable to the resistance of the metal layers and the TMR of the junction can then be measured.

The basics of the CIPT technique can be explained using a simplified resistor network [20], shown in Figure 2.8(a). The unpatterned MTJ film with two contacts of length \( L \), width \( W \), and spacing \( x \) between them (\( L>>x>>W \)) placed on top of the film can be electrically modeled by four resistors. Two horizontal resistors \( xR_T/L \) and \( xR_B/L \) represent the top and bottom metal layers, respectively. The two vertical resistors, each having area \( xL/2 \) and therefore having resistance \( 2RA/xL \), represent the tunnel junction. Analyzing the resistive network one can clearly see that in the case of closely spaced contacts, as \( x\to0 \), the resistance of the tunnel barrier diverges to infinity and the current flows only through the top layer, reducing the MR signal to zero. When the contacts are placed too far apart, the resistance of the barrier becomes negligible on the scale of the resistance of the metal layers causing the expected MR signal to vanish. From the dimensional analysis the optimal length scale at which the MR is significant can be estimated as:

\[ \lambda = \sqrt{RA/(R_T + R_B)} \]

where \( R_T \) and \( R_B \) are the resistance per square of the top and bottom metallic layers, respectively.

The solution of this model, shown in Figure 2.8(a), can be represented as follows:

\[
R = \frac{x}{L} \frac{R_TR_B}{R_T + R_B} \left( 1 + 4 \frac{R_T}{R_B} \frac{1}{4 + x^2\lambda^2} \right). \tag{2.1}
\]
The MR is then

\[
MR_{\text{clip}} = 100 \left( \frac{R_{\text{high}} - R_{\text{low}}}{R_{\text{low}}} \right),
\]  

(2.2)

where \(R_{\text{high}}\) and \(R_{\text{low}}\) can be determined using \(R_{\text{A high}}\) and \(R_{\text{A low}}\), respectively.

Using the typical MTJ parameters of \((R_T = R_B = 50 \ \Omega / \square, R_A = 1000 \ \Omega \ \mu m^2, \ MR=30\%\ \text{and} \ L=500 \ \mu m)\), the maximum measured magnetoresistance should be observable at the tip spacing on the order of micrometers. It follows from the model that particular attention should be paid to determining the correct tip spacing as it significantly affects the extracted MR values.

The resistivity measurement is performed using 4-point measurements. This technique reduces the influence of the contact resistance on the MR signal. For this case, the resistance \(R\) is given by [20]

\[
R = \frac{V}{I} = R_T \frac{R_B}{R_T + R_B} \frac{1}{2\pi R_B} \left[ R_t \times 
\right.
\]

\[
\left. \times \left[ K_0 \left( \frac{a}{\lambda} \right) + K_0 \left( \frac{c}{\lambda} \right) - K_0 \left( \frac{a + b}{\lambda} \right) - K_0 \left( \frac{b + c}{\lambda} \right) + \ln \left( \frac{(a + b)(b + c)}{ac} \right) \right] \right),
\]  

(2.3)

where \(a\) is the distance between the I+ and V+ contacts, \(b\) – the distance between V+ and V-, \(c\) – the distance between V- and I-, and \(K_0\) – the modified Bessel function of the second kind of zero order. The MR is then obtained using the equation 2.2. Figure 2.8(b) represents the calculated resistance per square for the low-resistance state and MR as a function of the tip spacing. The MR depends not only on \(\lambda\), but on the \(R_T/R_B\) ratio as well: red, blue and green curves represent the solutions of equation 2.3 for \(R_T/R_B = 10, 1, \) and \(0.1\), respectively [19].

Thus, the tunnel junction MR is strongly dependent on the contact spacing – the spacing different from the optimal length \(\lambda\) results in lower measured MR. Practically, the RA and MR is obtained by measuring the resistance for different tip combinations and fitting data to the above CIPT-model. The relative magnetoresistance of an MTJ sample (in percent, relative to RA) can be measured quickly and simply by using only one tip spacing. It is informative to note that the CIPT technique can also be used to measure the MR of samples with much smaller RA, such as spin-valves. In this case, \(R_T\) and \(R_B\) should be kept small in order to keep \(\lambda\) in the optimal range.

The CIPT instrument presented here uses standard multi-tip microprobes commercially available from Capres [26]. These probes, with the tip spacing of 1.5 to
2.2. CIPT INSTRUMENT

Figure 2.9: (a) SEM image of a CIPT microprobe. (b) Si-chip CIPT probe mounted onto a chip carrier. After Capres [26]

20 µm, are produced using a Si-based MEMS technology, similar to that used for making AFM probes. CIPT microprobes had 4 or 12 aligned silicon sharpened tips, extending from a silicon support chip, as shown in Figure 2.9(a). As a result of a well established processing technology, the silicon tips of 25 µm length and 3 µm width are positioned extremely accurately with respect to each other, which is very important for 4-point electrical measurements. The contact side of each silicon tip had an Au conductive coating. The high mechanical flexibility of the tips, whose spring constant is ~5 N/m, allows a reliable contact between the sample surface and the tips, even when the sample and the probe are slightly misaligned. The contacting force is around $10^{-8}$ - $10^{-7}$ N, which guarantees non-destructive measurements and allows further processing of the samples after testing. The probe is glued and electrically bonded to the ceramic probe base, as shown in Figure 2.9(b). The probe holder connects all of the tips on the silicon chip to the external electronics. The probe current was limited to 1 mA to avoid heating and potentially melting the Au conductive layers of the tips. Similarly, a safe threshold of 200 mV was selected for the applied voltage during probe engagement. Too high a voltage can result in sparks between the tip and the sample, which can damage the contact area.

The block diagram of the instrument is shown in Figure 2.10. The system was designed to have a fixed probe and a moving sample stage, as shown in Figure 2.11. This design allows for easy and fast changes of the sample without the danger of damaging the probe. A thin film sample is mounted on a 3D moving stage with manually adjustable X and Y axes, which is driven by a squiggle piezoelectric step motor on the Z axis. The step size in Z can also be accurately adjusted manually. In the case of the automatic approach, the system is preset for three discrete step sizes: 125, 250, and 500 nm. The instrument contains two separate blocks: the preamplifier block and the main unit, which includes the power supplies for the
electronic circuits and the electromagnet, the digital IC’s for signal conversion from 3.3 V to 5 V logic levels, the magnetic field and piezodrive controllers, as well as some additional auxiliary electronics. The signals from the probe holder are fed into the preamplifier block, consisting of a solid-state cross-point switch and digitally controlled amplifiers, and subsequently into the ADC unit on the FPGA board. The cross-point switch was chosen to have a low ON resistance to avoid signal loss, and as high as possible OFF resistance to reduce the crosstalk between the channels and the parasitic leakage. In the present design an AD75019 switch with 100 Ω ON and 10 MΩ OFF resistances and -96 dB crosstalk between the channels was used. The inputs of the switch were directly connected to the contacts on the probe holder whereas the outputs were connected to instrumentation amplifiers of type PGA204 with digitally controlled gain. The current source circuit was placed inside the same preamplifier block. In the instrument presented here, the magnetic field is generated by a horseshoe-type electromagnet embedded into the approach stage, capable of creating magnetic fields up to 350 Oe in strength, which is sufficient for magnetization switching in typical ferromagnetic materials used with CIPT. The setup was designed in such a way that the microprobe is situated between the poles, protruding slightly below the bottom edge of the magnet poles. During MR measurements, when the microprobe is in contact with the sample, the poles of the electromagnet are approximately 1 mm above the sample surface. The switching of the magnetization in the sample is forced by the stray field from the magnet gap, which was accurately calibrated. The close positioning of the magnet is such as to provide a strong magnetic field in the sample plane, without the sample being placed inside the magnet gap. This simplifies the magnet design and results in a very compact instrument layout.

Fast and precise control of the stage movement is required during approach. For this purpose we have used a commercially available nonmagnetic squiggle piezo motor with a motor controller [27]. The squiggle piezodrive allows for precise stepwise, continuous, high speed movement of the stage, which is convenient for fast disengagement of a sample from the probes. Standard precision machining typically introduces a small error in the dimensions of the parts, which can result in a small misalignment of the probe and the sample surface of typically less than 1 degree. If the probe approaches a strongly misaligned sample, the approach algorithm should estimate the angle of misalignment and stop the approach procedure when this angle exceeds a critical value. In the design presented here, this approach method was implemented using the following algorithm. After each step of the piezomotor, the resistance between the two rightmost and the two leftmost tips of the probe are monitored. Once the probe’s tips are in electrical contact with the sample, the drop in the resistance generates a stop event. If the probe and the sample are highly misaligned, the stop event is generated by only one pair of the outermost tips. In the case of ideal alignment, the stop event is generated by both pairs simultaneously. Once electrical contact is established, the movement is stopped and further action can be taken. In practice, the stop event is usually generated by only one tip pair, while the other side of the probe is not in contact. Then, in order to bring all of
At the core of the system is a National Instruments powered FPGA PCI card running a real time algorithm (developed in-house) that controls the instrument. The FPGA card performs the analog signal acquisition, current sourcing, digital communications with the amplifiers and the cross-point switch, ramping of the magnetic field, interfacing with the piezo motor controller, and communicating with the host PC. In addition, the FPGA card algorithm implements two independent digital lock-ins with the sample rate of up to 200 kS/s used for low noise resistance measurements. Low level software running on the FPGA can be divided virtually into two sequential main loops. The first of these controls the engagement of the sample and the establishment of stable electrical contact. The second loop, which runs during measurements, provides two independent digital lock-ins, a subroutine for ramping the magnetic field, and a routine controlling the communications between the FPGA card and the host computer. The noise and probe-position error reduction was implemented by a FPGA routine similar to the one, described in [28].

Figure 2.12 shows a screenshot of the control software, developed using LabView [29]. This software provides approach control, manual control of the instrument settings, and measurements of the sample in the automatic regime. The control software, having a user friendly GUI with full access to the real-time algorithm running on the FPGA card, allows changing the device parameters and saving the measured data on a continuous basis. It is built as 5 sequential subroutines: the first stage implements the approach mechanism and stops the engagement automatically when electrical contact is established. The second stage allows manual control of the contact quality. The specific tips of the probe to be used for the
resistance measurement can be chosen in the third stage. The current amplitude, time constant, and amplifier gain of the embedded lock-ins are configured in the fourth stage. The final stage governs magnetoresistive measurements and saving of data. In order to prolong the probe’s lifetime, each subroutine strictly follows the preceding one and cannot be executed out of order. For example, after establishing electrical contact between the probe and the sample, the software does not allow any further movement of the sample towards the probe, which can produce tip damage. A video capture subroutine, running simultaneously with the main program, as shown in Figure 2.12, allows visual inspection of the contacted area and the condition of the Si tips.

The data in Figure 2.13(b) represent experimentally measured resistance of a spin-valve (a) and that of an unpatterned MTJ provided by Capres [26] as a test sample (b). The MTJ sample had $R_T = 27.7 \, \Omega$, $R_B = 2.3 \, \Omega$, $R_A = 4169 \, \Omega/\mu m^2$ and $MR = 62 \%$. As previously discussed, for optimal MR measurements, the spacing of the tips should be comparable to $\lambda$. The data shown in Figure 2.13(b) was obtained using the tip spacing of 5 $\mu m$, which in this case is ~3 times shorter than $\lambda$ (estimated to be 15 $\mu m$). This explains the relatively low MR value obtained. It is important to mention that establishing stable, vibration-free electrical contact is very important for microprobe CIPT measurements. In the first implementation, the CIPT head was placed on a passively damped optical table, and was found to be quite sensitive to vibrations. Placing the instrument in a well isolated environment should improve its signal-to-noise ratio and reduce the drift caused by vibrations. This work is under way. During testing, it was also found that much of the resistance noise visible in the experimental data is caused by the drift of the electrical contact and not by the electronic circuitry or the mechanics. In order to establish a stable electrical contact, the top layer of the MTJ should be covered with a thin Ru or Au film. Although the use of a capping layer reduces the
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Figure 2.12: Screenshot of the software developed for controlling CIPT measurements.

Figure 2.13: (a) Example of spin-valve MR measured using the developed CIPT instrument. (b) TMR measured for the test sample provided by Capres Co. [26]

noise caused by contact drifts, the capping layer itself serves as a shortcut for the source current, reducing the amplitude of the useful signal in inverse proportion to the capping film thickness. Figure 2.13(a) shows an MR measurement of a spin-valve film produced in-house. The low MR value observed in this case is due to shorting of the MR signal by the 10 nm thick Au capping layer.

To summarize, a CIPT instrument was developed and successfully tested. The
software controlling the instrument allows for a fully automatic approach process, surface detection, and measurements of transport properties of thin films. The embedded optical system provides visual control of the condition of the probe as well as choosing a clean, dust-free area of the sample. Test measurements of the switching characteristics in MTJ and spin-valve blanket films are presented. The instrument can significantly simplify the process of developing new MTJ materials in virtue of fast, accurate, and nondestructive measurements of the relevant MR and RA values.
Chapter 3

Introduction to spintronics

The magnetoresistance effect is one of the most fascinating discoveries in thin film magnetism, engendering numerous spintronics devices, and becoming an essential driver of rapid progress in computer technology. GMR and TMR based spintronics devices, such as read heads of hard disk drives and magnetic random access memory, provide new opportunities in the high-density magnetic disk storage and high-speed non-volatile computer memory. This chapter will introduce basic principles of spin dependent transport and magnetism at the nanoscale.

3.1 Spin-dependent transport in nanostructures

The transient properties of electrons in conducting materials are determined by the electric field applied and electron scattering in the material. At elevated temperatures, such as room temperature, electrical conductance in non-magnetic metals is limited predominantly by scattering on thermal phonons [30]. Magnetic materials introduce addition, magnon scattering, which is due to interaction of the spin of the electron with localized atomic spins. Non-magnetic metals (NM) have a symmetrical density of states (DOS) and their electrical conductivity is due to the \(s\)-band electrons, as shown in Figure 3.1 (a). In the case of magnetic materials, there is a shift in the density of states between the spin-up \((s_\uparrow)\) and spin-down \((s_\downarrow)\) electrons, which is determined by the exchange energy \(E_{\text{ex}}\), as shown in Figure 3.1 (b). Electrical properties of magnetic metals are determined to a larger degree by the \(s\)-band “free” electrons, while the less numerous \(d\)-band electrons carry magnetic moment owing to the fact that the \(d\)-band is spin-polarized. In the transition metal ferromagnets, the \(4s\) and \(3d\) bands overlap and become hybridized, which results in a decreased mobility of the \(s\) conduction electrons through an increase in their effective mass. The hybridization also means that the \(4s\) electrons can scatter into the \(3d\) states. Since the \(d\)-band is partially filled (e.g., half-filled), \(s_\uparrow\) electrons of only one spin polarization can scatter into the \(d\)-band, which results in polarization of the charge current in the ferromagnet.
Figure 3.1: Simplified band structure of normal metal (left) and ferromagnet (right).

The electrical resistivity of a ferromagnet (FM) can be described using a simplified two channel model [31–33], based on different scattering rates for $s_\uparrow$ and $s_\downarrow$ electrons. The total resistivity can then be represented by two parallel resistivities $\rho_\uparrow$ and $\rho_\downarrow$, where $\rho_\uparrow$ is the lower of the two:

$$\rho = \frac{\rho_\uparrow \rho_\downarrow}{\rho_\uparrow + \rho_\downarrow}.$$  \hspace{1cm} (3.1)

In this two-channel model, the polarization $P$ of the magnetic metal is given by:

$$P = \frac{\sigma_\uparrow - \sigma_\downarrow}{\sigma_\uparrow + \sigma_\downarrow} = \frac{D_\uparrow - D_\downarrow}{D_\uparrow + D_\downarrow},$$  \hspace{1cm} (3.2)

where $\sigma_\uparrow, \sigma_\downarrow = 1/\rho_\uparrow, \rho_\downarrow$ is the conductivity and $D_\uparrow, D_\downarrow$ – the DOS for the majority and minority spin bands.

When the current, polarized inside the ferromagnet, is injected into a non-magnetic metal, the current's polarization is conserved and results in a spin accumulation in NM. This spin imbalance in NM decreases exponentially away from the injection point over a characteristic length known as the spin diffusion (or spin-flip) length:

$$l_{sf} = \sqrt{D\tau_{sf}},$$ \hspace{1cm} (3.3)

where $D = v_F l_f/3$ is the spin-independent diffusion coefficient, $v_F$ – the Fermi velocity, $l_f$ – the mean free path, and $\tau_{sf}$ – the spin-flip rate. In the case where another ferromagnet is added within the spin diffusion length to form an FM1/NM/FM2 structure, the interaction between the polarized electrons and the magnetization of the second ferromagnet leads to a change in the electrical resistance of the overall structure due to spin-dependent scattering in FM2. This is known as the GMR effect, which is described below.
AMR

Magnetoresistance was first discovered in 1856 by William Thompson [34] (more commonly known as Lord Kelvin), while he was conducting measurements of the electrical resistance of a ferromagnet in a magnetic field. In the course of these experiments, he observed that the resistance of Fe and Ni is a function of the angle between the direction of the magnetization and that of the electrical current. This effect is referred to in the literature as anisotropic magnetoresistance (AMR). Although the AMR effect in ferromagnets is relatively small, a few percent in magnitude in such materials as NiFe alloys, it could be used in AMR sensors as a replacement for the inductive read heads in magnetic storage discs in the late 1970s. Normally, maximum resistivity corresponds to the parallel alignment of the current and magnetic field, whereas the resistivity is minimal for the perpendicular orientation. Thus, the resistivity depends on the angle \( \theta \) between the current and the magnetic field:

\[
\rho = \frac{|E|}{|J|} = (\rho_\parallel - \rho_\perp) \cos^2(\theta) + \rho_\perp.
\]

The nature of this effect is in the spin-orbit coupling affecting the scattering rate of the conducting electrons in a ferromagnet [35, 36].

GMR

Giant magneto-resistance is found in FM/NM/FM multilayers also known as spin-valves. GMR can be much larger than AMR (hence the name), up to 20%. This allows successful use of the effect in spintronic devices.

GMR was independently discovered in 1988 by two groups, led by A. Fert [37] and P. Grunberg [38], when it was observed that applying a magnetic field significantly reduces the resistance of an anti-ferromagnetically coupled FM/NM multilayer: Fert’s group used a Fe/Cr multilayer, whereas Grunberg’s group used a Fe/Cr/Fe tri-layer. The discovery of the GMR effect triggered a wide range of research on spin-dependent transport due to the tremendous technological potential of the effect for applications in the fields of magnetic storage and sensor technology; this discovery has been rightly called the birth of spintronics, and received Nobel Prize in 2007.

The resistance of a spin-valve structure with the magnetic moments of the ferromagnetic layers aligned at an arbitrary angle \( \theta \) can be expressed as follows:

\[
R = R_P + \frac{\Delta R (1 - \cos(\theta))}{2} = R_P + \Delta R \sin^2\left(\frac{\theta}{2}\right),
\]

where \( \Delta R = R_{AP} - R_P \), \( R_{P,AP} \) is the resistance of the spin-valve for parallel and antiparallel magnetization alignment.
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Figure 3.2: Schematic of the two-channel model of GMR. The parallel alignment of magnetization is shown on the left with the anti-parallel configuration on the right. Bottom panels represent the respective equivalent electrical circuits.

Physically, the origin of GMR can be understood by considering a metal as having two independent conductive channels. The probability of spin-flip scattering in metals is normally small compared to the probability of scattering in which the spin of the electron is conserved. This means that $s_\uparrow$ and $s_\downarrow$ electrons do not mix over long distances and the electrical conduction occurs in two independent spin-polarized channels. The difference in scattering rates for $s_\uparrow$ and $s_\downarrow$ electrons can be quite large in magnetic metals, and results in different effective resistances for each spin channel. The schematic of the two-channel model and its effective electrical circuit is shown in Figure 3.2. Upon parallel alignment of the magnetization in the ferromagnets, the $s_\uparrow$ electrons experience lower total resistance than the $s_\downarrow$ electrons. In the anti-parallel state the two spin channels have the same effective resistance. Electrical analysis of the equivalent circuits yields the total resistance for each alignment:

$$R_P = \frac{R_\uparrow R_\downarrow}{R_\uparrow + R_\downarrow} \quad \text{and} \quad R_{AP} = \frac{R_\uparrow + R_\downarrow}{2}. \quad (3.6)$$

The GMR ratio is then determined by:

$$GMR = \frac{\Delta R}{R} = \frac{R_{AP} - R_P}{R_P} = \frac{(R_\downarrow - R_\uparrow)^2}{4R_\downarrow R_\uparrow}. \quad (3.7)$$

**TMR**

A magnetic tunnel junction is obtained by replacing the non-magnetic spacer in the spin-valve structure discussed above with an insulating (I) tunnel barrier. Its
magnetoresistance, the TMR, was first reported in 1975 by Julliere, who measured approximately 14% in an Fe/Ge/Co trilayer at 4K [39]. Julliere proposed a model which gives the TMR of a FM/I/FM structure as:

$$TMR = \frac{R_{AP} - R_P}{R_P} = \frac{2P_1P_2}{1 - P_1P_2},$$  \hspace{1cm} (3.8)$$

where $P_1, P_2$ represent the polarization of the two ferromagnetic electrodes.

The Julliere model provides a simple physical picture which explains the nature of the TMR effect. However, this model does not take into account the height and the thickness of the tunnel barrier and considers only DOS at the Fermi energy. As a result, the bias voltage dependence of the TJ resistance is not explained by this model.

In 1989 a more accurate theoretical model, which took into consideration the height and the thickness of the barrier, was proposed by Slonczewski [40]. His model is based on the Schrödinger equation with a single-electron Hamiltonian, and assumes a rectangular barrier separating two free-electron-like ferromagnets. Slonczewski derives the conductance as a function of angle $\theta$ between the directions of magnetization of the ferromagnets to be:

$$G(\theta) = G_0(1 + P^2 \cos(\theta)).$$  \hspace{1cm} (3.9)$$

The equivalent expression for the MTJ resistance can be written as:

$$R(\theta) = \frac{R_0}{1 + P^2 \cos(\theta)},$$  \hspace{1cm} (3.10)$$

where $P$ is the effective spin polarization of tunneling electrons:

$$P = \left(\frac{k_\uparrow - k_\downarrow}{k_\uparrow + k_\downarrow}\right) \left(\frac{k_\uparrow^2 - k_\uparrow k_\downarrow}{k_\uparrow^2 + k_\uparrow k_\downarrow}\right).$$  \hspace{1cm} (3.11)$$

Here $k_{\uparrow, \downarrow}$ are the Fermi wave vectors and $k$ – the wave number of the electron wave function inside the barrier, which depends on the barrier height $U$:

$$k = \sqrt{(2m/\hbar^2)(U - E_F)}.$$  \hspace{1cm} (3.12)$$

The first term in equation 3.11 represents the spin polarization, similar to that in Julliere's model. The second term is due to the interface, and depends on the barrier height $U$. The effective polarization decreases for small barrier heights. For a high potential barrier, $k$ tends to infinity and spin polarization reduces to that predicted by Julliere's model.
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\[ \lim_{k \rightarrow \infty} P_F = \frac{k^\uparrow - k^\downarrow}{k^\uparrow + k^\downarrow} \]  

(3.13)

Slonczewski’s model is more realistic and gives the spin-polarization of the MTJ conductance, which is not entirely an intrinsic property of the ferromagnetic electrodes. Features that this model does not take into account are voltage, temperature, and barrier thickness dependence of the TMR. However, Slonczewski’s model provides a very good approximation in the case of thick barriers and small barrier heights. A detailed comparison of Julliere’s and Slonczewski’s models with first principles numerical calculations was performed by MacLaren et al. [30].

Recent experiments report TMR of more than 70 % for AL\textsubscript{2}O\textsubscript{3} based tunnel junctions [41] and over 1000 % for MgO-based junctions [42, 43]. In this work the TMR effect was used for detecting various magnetization states in free layers of spin-flop magnetic tunnel junctions based on AL\textsubscript{2}O\textsubscript{3}.

3.2 Theory of micromagnetism

The dynamic behavior of magnetization of a uniform ferromagnet is well understood, and is governed by the Landau-Lifshitz-Gilbert equation [44]. However, when the dimensions of the ferromagnet exceed the single domain limit, the system must be considered using the micromagnetic approach. In micromagnetic simulations, a ferromagnetic particle is divided into a number of smaller cells with the assumption that the magnetization within each cell is uniform. In this way, a uniform magnetic moment (super-spin) of strength \( M_s \) times the cell’s volume can be associated with each cell. There are two main approaches to meshing a ferromagnetic body into small cells: the finite element (FE) approach where the meshing is into small tetrahedral elements, and the finite difference approach (FD) where the meshing is into small cuboids. Both of these approaches have their advantages and disadvantages: FE provides an efficient way to compute the demagnetizing field by using FFT, whereas the FD method better resolves shapes with a high degree of curvature.

The evolution of a micro-magnetic system in time follows from solving a system of coupled differential equations: the equation of motion should be solved for each cell taking into account the interactions with all the other cells. For accurate modeling, the mesh size should be smaller than the characteristic exchange length in the material [45]. When this requirement is fulfilled, the results of the micromagnetic modeling are independent of the cell size chosen.

Evolution of magnetization in magnetic material is governed by several competing energy terms. In the micromagnetic approach, four main energy contributions are used. These include magnetostatic energy originating from interactions between the magnetic poles in the system, the exchange energy forcing neighboring magnetic moments to align parallel or anti-parallel to one another depending on the magnetic nature of the material, magnetocrystalline anisotropy energy which tends
to align magnetic moments along certain preferred crystalline directions, and the Zeeman energy term responsible for alignment of the spins in the direction of the applied magnetic field [46]. At equilibrium, all energy terms compete in aligning the magnetic moment of each cell along the resulting effective field \( H_{\text{eff}} \).

### 3.2.1 Effective field calculation

The total energy of the system is obtained by summing all energy terms. The Zeeman energy is the energy of interaction between a magnetic moment and an external magnetic field. The total Zeeman energy is minimal when the moment is aligned with the external magnetic field:

\[
E_Z = -\mu_0 \int_V \vec{M} \cdot \vec{H}_{\text{ext}},
\]

(3.14)

where \( \mu_0 \) is the permeability of vacuum, \( \vec{H}_{\text{ext}} \) – the applied field, and \( \vec{M} \) – the magnetization of the sample.

The magnetostatic energy also known as the demagnetizing energy represents the interaction between a demagnetizing field \( \vec{H}_{\text{demag}} \) and the magnetization of the material. The demagnetizing field tends to minimize the total magnetization of the system, and consists of the internal demagnetizing field and the external stray field. Like the Zeeman term, it can be written as:

\[
E_{\text{demag}} = -\frac{\mu_0}{2} \int_V \vec{H}_{\text{demag}} \cdot \vec{M} dV.
\]

(3.15)

Maxwell’s equations give the demagnetizing field as

\[
\nabla \times \vec{H}_{\text{demag}} = 0 \quad \text{and} \quad -\nabla \cdot \vec{M} = \nabla \cdot \vec{H}_{\text{demag}}.
\]

(3.16)

Since the curl of \( \vec{H}_{\text{demag}} \) is zero, the demagnetizing field can be given by its scalar magnetic potential:

\[
\vec{H}_{\text{demag}} = -\nabla \phi,
\]

(3.17)

where \( \phi(\vec{r}) = q \frac{1}{4\pi r^3} \) is the scalar magnetic potential at distance \( r \) from a magnetic point charge \( q \), which needs to be integrated over the entire volume \( V \) in order to obtain the total demagnetizing field of the modeled particle. \( \vec{M} \) is continuous inside the magnetic material, and the magnetic charge density \( \rho \) existing inside the material is defined as the divergence of the magnetization:

\[
\rho = -\nabla \cdot \vec{M} = \nabla^2 \phi.
\]

(3.18)
In calculating the demagnetizing field, magnetic charges appearing on the boundaries of the material, should be taken into account: $\phi^{\text{out}} = \phi^{\text{in}}$. When computing the charge distributions, the solution of the Poisson equations can be presented as follows:

$$\phi(\vec{r}) = \int_V \frac{\rho(\vec{r}')}{|\vec{r} - \vec{r}'|} dV' + \int_\delta \frac{\sigma(\vec{r}')}{|\vec{r} - \vec{r}'|} dA. \quad (3.19)$$

Here, expression 3.17 can be used to obtain:

$$\vec{H}_{\text{demag}}(r) = -\nabla \phi_{\text{demag}}. \quad (3.20)$$

Because of the integration over the entire volume, the calculation of the demagnetizing field is the most time consuming stage, which severely limits the computational performance of micromagnetic simulations.

The exchange interactions between neighboring atoms align their magnetic moments. Its origin lies in a quantum mechanical effect resulting from the Pauli exclusion principle and the Coulomb repulsion. The micromagnetic exchange energy is given by:

$$E = A |\nabla \vec{m}|^2, \quad (3.21)$$

where $A$ is the exchange constant – a property of the material that reflects the strength of the exchange interaction and

$$|\nabla \vec{m}|^2 = (\nabla \vec{m}_x)^2 + (\nabla \vec{m}_y)^2 + (\nabla \vec{m}_z)^2. \quad (3.22)$$

The characteristic length scale associated with the exchange interaction is called the exchange length $l_{\text{ex}}$, and is given by:

$$l_{\text{ex}} = \sqrt{\frac{2A}{\mu_0 M_s^2}}. \quad (3.23)$$

Here, $M_s$ is the saturation magnetization. Within the exchange length, the magnetization in the material is uniform. Magnetization non-uniformities, such as domain walls and vortices have dimensions exceeding the exchange length. Therefore, in micromagnetic simulations, the exchange length is typically the maximal discretization size.

The numerical engine assumes that the magnetization changes little from cell to cell. Therefore, the mesh size should be chosen small enough, such that the direction of neighboring moments varies little from one cell to another. The effective exchange field can be found by differentiating expression 3.21, which yields:
3.2. THEORY OF MICROMAGNETISM

\[ H_{\text{ex}} = 2A(\nabla \vec{m})^2. \]  (3.24)

Due to spin-orbit coupling, the total energy of a ferromagnet can depend on the relative angle between the magnetization and the crystal axes. This property of magnetic materials is known as magnetocrystalline anisotropy and is responsible for magnetic hysteresis and coercivity. The form of magnetic anisotropy depends on the symmetry of the crystal, e.g., for hcp crystals, the easy axis can be along the c axis of the hexagonal cell. The simplest phenomenological anisotropy model is that for materials with uniaxial anisotropy, which is given by:

\[ E_{\text{ani}} = \int_V K_1 \sin^2(\theta) dV, \]  (3.25)

where \( K_1 \) is the first-order anisotropy constant of the magnetic material (J/m³), \( V \) is the volume of the magnet, and \( \theta \) is the angle between the magnetization of the sample and the preferred anisotropy direction. Although the first-order anisotropy term is the major contribution in most materials, there are times when higher order and/or non-uniaxial contributions should be taken into account. Higher-order anisotropy terms can be included as:

\[ E_{\text{ani}} = K_1 \sin^2(\theta) + K_2 \sin^4(\theta) + K_3 \sin^6(\theta). \]  (3.26)

The material studied in this work is Permalloy, which has a very low uniaxial anisotropy. For this reason, the contribution of the higher order anisotropy terms was not considered. In practice, it is often convenient to express anisotropy in terms of an effective anisotropy field:

\[ H_{\text{ani}} = \frac{2K_1}{\mu_0 M_s}, \]  (3.27)

where factor 2 allows direct comparison of \( H_{\text{ani}} \) with \( H_c \), the latter representing the coercivity field. It was determined experimentally that the samples used in this study had an anisotropy field of approximately 5-10 Oe, which corresponds to the anisotropy constant of 420 J/m³.

The relation between any of the energy density terms described above and the corresponding field terms can be expressed as follows:

\[ H_i = \frac{1}{\mu_0 M_s} \frac{\delta e_i}{\delta \vec{m}}. \]  (3.28)

The total, effective field can be written as:

\[ H_{\text{eff}} = (\mu_0 M_s \vec{H}_{\text{ext}} - \nabla \phi_{\text{demag}} + 2A\nabla^2 \vec{m} - 2\vec{K} \times (\vec{m} \times \vec{k})), \]  (3.29)
where $\vec{m}$ is the magnetic moment vector, and $K$ – the anisotropy constant having direction $\vec{k}$.

### 3.2.2 Energy minimization

When it is necessary to determine the equilibrium configuration of magnetization in an external magnetic field, there may be no need to calculate intermediate magnetization states – we are interested in the final, equilibrium state only. In this case, the ground state corresponds to the state of lowest energy when the magnetic moment of the ferromagnet under consideration is aligned with the effective field. The preceding section describes how to obtain all the energy terms of interest, so the total energy of the system is:

$$E_{\text{tot}} = E_{\text{ex}} + E_{\text{ani}} + E_Z + E_{\text{demag}}. \quad (3.30)$$

At equilibrium, the torque on the magnetic moment becomes zero as the magnetization becomes parallel to the effective field, which can be written in the simplified form of Brown's static equation:

$$\frac{\delta E}{\delta \vec{m}} = \vec{m} \times H_{\text{eff}} = 0. \quad (3.31)$$

This equation allows to find the equilibrium configuration of the magnetization within the magnetic body. It is important to note that it is nonlinear and is usually solved numerically.

In the present work, the Conjugate Gradient (CG) method implemented in the energy minimization module of the Object Oriented MicroMagnetic Framework OOMMF [47] was used to numerically calculate the energy minimum from the initial configuration. The CG algorithm takes the gradient of a function, minimizes the function in the gradient direction, and from this point calculates the gradient and a new (conjugate) direction in which to minimize. The gradient of the energy is $\vec{m} \times \vec{H}_{\text{eff}}$. Details of the CG algorithm can be found in [48]. This method is the most efficient for calculating, for example, quasistatic magnetization versus magnetic field, i.e., hysteresis loops.

### 3.2.3 Landau-Lifshitz-Gilbert equation of motion

The dynamic equation for the magnetization vector precessing about the effective field $H_{\text{eff}}$ was proposed by Landau and Lifshitz in 1935 [44]. This equation is phenomenological and takes into account all the quantum mechanical effects and anisotropy by means of the effective field. In the absence of dissipation, the equation is...
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\[
\frac{d\vec{M}}{dt} = -\gamma \vec{M} \times \vec{H}_{\text{eff}}.
\] (3.32)

Here \( \gamma \) is the gyromagnetic ratio given by \( \gamma = \frac{\mu_0 q_e}{m_e} = 2.21 \times 10^5 \text{A}^{-1} \text{ms}^{-1} \), where \( g \) is the Landé factor (~2), and \( q_e \) and \( m_e \) are electron’s charge and mass. This equation conserves the energy of the system and does not take into account magnetic dissipation. In order to describe the magnetization dynamics of a realistic system, dissipative processes must be taken into account. Landau and Lifshitz modified the equation of motion by adding a damping term proportional to \( \vec{M} \times (\vec{M} \times \vec{H}_{\text{eff}}) \):

\[
\frac{d\vec{M}}{dt} = -|\gamma| \vec{M} \times \vec{H}_{\text{eff}} - |\gamma| \alpha M_s \vec{M} \times \left( \vec{M} \times \vec{H}_{\text{eff}} \right).
\] (3.33)

The nature of dissipation is usually associated with eddy currents and magnon-phonon coupling. In 1955 Gilbert modified the dissipation term in the LL equation [49], making it proportional to the time derivative of the magnetization. The Landau-Lifshitz-Gilbert (LLG) equation has the form:

\[
\frac{d\vec{M}}{dt} = -|\gamma| \vec{M} \times \vec{H}_{\text{eff}} - \frac{\alpha}{M_s} \left( \vec{M} \times \frac{d\vec{M}}{dt} \right),
\] (3.34)

where \( \gamma \) is given by \( \gamma = (1 + \alpha^2)|\gamma| \), \( \alpha \) – a dimensionless phenomenological damping parameter. Mathematically, this equation is identical to the Landau-Lifshitz equation.

The Gilbert form of the equation allows an easy interpretation of the two contributions to the time evolution of magnetization. The first term is responsible for the precession of magnetization around the effective field direction, as shown in Figure 3.3. It keeps the angle between \( \vec{M} \) and \( \vec{H}_{\text{eff}} \) constant and, therefore, conserves the energy of the system. The precession frequency is given by: \( f = \frac{\mu_0 |\vec{H}_{\text{eff}}|}{2\pi} \).

The second term of the LLG equation represents the energy dissipation in the system, with the damping proportional to the rate of precession \( \delta \vec{M}/\delta t \). The dimensionless damping parameter determines the speed of magnetization relaxation in the direction of \( \vec{H}_{\text{eff}} \) and is usually much smaller than 1.

The LLG equation is widely used in numerical simulations of the magnetization time evolution. This can be done by performing numerical integration of the LLG equation with recalculation of the effective field \( \vec{H}_{\text{eff}} \) at every time step, since it changes with changes in the magnetization distribution. In this work, we have used the Euler and 4th-order Runge-Kutta methods of integration, implemented in OOMMF.
3.2.4 Temperature in micromagnetic simulations

The usual way to simulate time dependent evolution of a magnetic system is by numerical integration of the LLG equation [44] over the entire volume of the magnetic particle. When the effect of thermal fluctuations is to be taken into account, it is treated numerically as a random fluctuating field acting on the magnetic moment $\vec{h}_{\text{fluct}}$ in addition to $\vec{H}_{\text{eff}}$ discussed above. Thus, the LLG equation becomes a stochastic differential equation (Langevin equation) of the form:

$$\frac{d\vec{M}}{dt} = -|\gamma| \vec{M} \times (\vec{H}_{\text{eff}} + \vec{h}_{\text{fluct}}) - |\gamma| \alpha \vec{M} \times (\vec{M} \times [\vec{H}_{\text{eff}} + \vec{h}_{\text{fluct}}]).$$  \hspace{1cm} (3.35)

Integration of the stochastic LLG equation can be done numerically. For this, two parameters (mean value and variance) of the fluctuating field $\vec{h}_{\text{fluct}}$ should be set. At the start of the simulation, $<\vec{h}_{\text{fluct}}>$ should be set to zero in order to avoid driving the system in a specific direction. The strength of the thermal field follows from the fluctuation-dissipation theorem:

$$h_{\text{th}} = \sqrt{\frac{\alpha k_B T}{\gamma M s V}}.$$ \hspace{1cm} (3.36)

Usually, $\vec{h}_{\text{fluct}}$ is taken in the form of uncorrelated white noise in time, space, and/or direction. The time scale of fluctuations is assumed to be much shorter than the intrinsic time scale of the LLG dynamics, given by the Larmor frequency. Micromagnetic simulations of temperature effects can fail at very low temperatures or at extremely high $\vec{H}_{\text{eff}}$ due to the limited precision of numerical integration.

In this work, thermal fluctuations were modeled using the additional Evolver class in OOMMF developed by Oliver Lemcke [50].

Figure 3.3: Precession (left) and dissipation (right) terms of the Landau-Lifshitz-Gilbert equation.
Chapter 4

Spin-flop dynamics

4.1 Introduction to MRAM

In conventional charge-based random access memory, information is stored on small capacitors and needs to be refreshed frequently in order to avoid data loss due to leakage currents. This results in a relatively high power consumption. MRAM has the potential to be a universal memory due to its non-volatility, high speed, low power requirements, and therefore can in principle serve as a replacement for flash-RAM and DRAM. In MRAM, information is stored as relative orientations of magnetization and does not need refreshing or power for maintaining the memory state. Once the power is removed, the magnetic bits stay magnetized, providing nonvolatility, and can be read immediately following next startup of the computer.

The evolution of the field-write MRAM begins in the early 1950s when the ferrite core memory was introduced [51] as the operating memory of the computers at that time. The principle of operation is based on the reversal of magnetization of a small ferrite toroid caused by current pulses through write coils wound onto the toroid, with the direction of the magnetization (clockwise, CW, or counterclockwise, CCW) representing a digital bit. The discovery of the GMR effect [37, 38] and the subsequent discovery of TMR [52] provided the foundation for developing new types of MRAM.

A magnetically soft thin film elliptical nanomagnet is always aligned with the effective anisotropy field, defining the easy axis (EA), typically along the long axis of the ellipse. In magnetic memory applications, the storage cell is usually fabricated from material of low intrinsic anisotropy, such as Permalloy, in order to control the EA using shape anisotropy only. In order to distinguish memory states using the direction of magnetization, a reference direction must be defined. Therefore, a magnetic memory cell consists of at least two magnetic parts: an easily switchable (free or storage) layer and a reference layer rigidly pinned in one direction. The two layers are separated by a non-magnetic spacer. The magnetization orientation in the free layer relative to that in the reference layer in a spin-valve or MTJ yields two
stable states – the low-resistive parallel state, and the high-resistive anti-parallel state. In digital memory, these resistive states are associated with the binary “0” and “1”, as illustrated in Figure 4.1. By reversing the magnetization in the free layer, new information is written to the memory cell. Reading of the binary state of the cell is done by measuring the voltage across a current-biased storage element. Depending on the material of the spacer between the reference and free layers, the readout uses either the GMR or the TMR effect. From a technological point of view, the TMR readout is more efficient on account of the high signal to noise ratio and low bias current requirements. Memory cells based on GMR require an additional amplifier to match the low readout voltage to the operating voltage of CMOS electronics, which complicates the design and leads to additional power consumption.

While the data storage mechanism in all types of magnetic memory is more or less the same and is based on relative orientation of the magnetization in a memory cell, MRAM can be divided into classes according to how data is written: field write, current write, and domain wall movement. The most obvious way to switch magnetization in a magnetic particle is to apply a magnetic field opposite to the direction of magnetization, of sufficient strength to switch the particle into the opposite magnetic state, as shown in Figure 4.2(a). Current pulses through embedded write lines generate the necessary fields. Magnetization can also be reversed by applying a sufficiently high current across a spin-valve or an MTJ – here, the mechanism is based on the spin transfer torque effect [4, 53] (see Figure 4.2(b)), which is the foundation of the newly developed STT-MRAM. The recent proposal for a “racetrack” type memory [5] is based on current-induced domain wall motion inside a long ferromagnetic strip, as illustrated in Figure 4.2(c). Here, the magnetic domains encoding the bits are created by a locally applied magnetic field, and the readout is implemented using a TMR sensor.
The subject of this study is spin-flop bi-layers, found at the core of the newly developed Spin-flop or Toggle-MRAM, which is of field-write type. We, therefore, next consider the basics of this memory type.

4.2 Stoner-Wohlfarth and Toggle-MRAM

Recent advances in the MTJ technology, with the TMR of over 70 % for AlO$_2$ [54] and over 200 % for MgO barriers [55] at room temperature, have provided the MTJ based MRAM designs with a high signal-to-noise ratio at ns-range access times [56]. The additional benefit of the MTJ’s is their high resistance compared to GMR devices, which allows using CMOS compatible electronics for readout without any intermediate amplification of the signal.

In a practical memory device the cells are arranged as a two-dimensional array, placed at the row-column intersections (bit-word line cross points). Writing or reading out a memory bit requires addressing the respective memory cell. The readout is usually performed by comparing the measured voltage across a current-biased MTJ to a reference voltage provided on chip. The cell must be addressed before the actual readout can take place. Implementing a transistor in series with the MTJ provides the necessary cell selectivity. The writing is performed by sending current pulses through the word line (WL) and bit line (BL) adjacent to the cell.

The structure of a typical storage element is shown in Figure 4.3(a) (top panel). It consists of three key blocks: a free layer is interfaced through a tunnel barrier with a flux-closed reference layer. Generally, the layer structure is more complicated and can consist of more than ten layers. However, the structure presented reflects the main functionality. The magnetic element typically has an elliptical shape, which provides shape anisotropy, with the easy axis along the length of the elliptical particle. The behavior of the free layer in the form of a single ferromagnetic layer of elliptical shape is usually described by the Stoner-Wohlfarth model for coherent rotation of magnetization in a single domain particle [1]. The magnetization in the reference layer does not change under applied write fields due to its very high
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Figure 4.3: (a) Layout of single free layer Stoner-Wohlfarth (top) and spin-flop bi-layer (bottom) MRAM storage elements. (b) Respective cell alignment versus the word and bit lines.

anisotropy. Therefore, only the orientation of the free layer relative to the EA direction of the elliptical particle needs to be considered.

The writing is performed by applying two magnetic fields simultaneously, along the EA and hard axis (HA) of the particle, as illustrated in Figure 4.3(b) (upper Figure). Switching occurs when the resulting vector field is outside the so-called astroid boundary, given by

\[ H_{HA}^2 + H_{EA}^2 = \left( \frac{2K}{M_s} \right)^2. \]  

(4.1)

The SW switching principle is illustrated in Figure 4.4: digital “1” or “0” are written into the selected memory bit by choosing a suitable combination of the EA and HA fields. This approach works well for individual bits but produces errors due to the so-called half-select problem in all bits situated on the same word and bit lines as the bit being written. Thermal excitations of the half-selected bits, subject to one-half of the writing field, result in a relatively high rate of memory loss due to thermally induced reversal. As shown in Figure 4.4, the activation energy of the half-selected SW bits decreases dramatically with increasing the writing field, which leads to potential data loss [57].

The stability as well as reproducibility of the magnetization reversal are significantly improved in the Toggle MRAM design, originally proposed by L. Savchenko et al. [2]. The bottom panel of Figure 4.3(b) illustrates the layer structure of the storage element in this design: the free layer consists of two dipole coupled soft magnetic layers [57] – the spin-slop bi-layer. The two magnetic moments in such
a bi-layer align antiparallel in the ground state. Similar to the SW structure, the spin-flop bi-layer is interfaced with a reference layer through a tunnel junction, which dominates the resistance of the stack. The memory cell is aligned at 45° to the write field lines, as shown in Figure 4.3(b). Switching is accomplished by a sequential application of the word and bit lines – the so-called toggle sequence. The switching boundary changes significantly from the SW astroid to a rectangular field boundary shown in Figure 4.4. The bottom panel of the Figure 4.4 shows the activation energy plotted as a function of the write field strength, \( H_{BL} \), which increases at higher fields, which in fact means a higher stability against thermal agitation for half-selected bits.

The magnetization reversal in Toggle MRAM is performed differently from that in SW MRAM. Figure 4.5 shows the magnetic phase diagram of the spin-flop bi-layer. Due to the inter-layer dipolar interaction, the magnetic moments in the two free layers are aligned antiparallel to each other at zero applied magnetic field. Applying an EA magnetic field of strength higher than the critical spin-flop field \( H_{SF} \) results in a spin-flop transition, where the magnetic moments discontinuously jump from the AP to the scissor state. In an ideal bi-layer, there is no way to control the rotation direction of the individual moments during the spin-flop – one moment flips clockwise and the other counterclockwise. A similar transition occurs when the EA field is reduced from above \( H_{SF} \) to zero. In order to perform a controlled rotation of the magnetization, the toggle field excursion is applied, such as shown by the dashed line in Figure 4.5. Applying this field sequence forces the magnetic moments to rotate into a scissor state and continuously follow the vector field, eventually deterministically rotating by 180 degrees. It is important that the applied word and bit line fields exceed the critical spin-flop field \( H_{SF} \) (e.g., those at point 2) in order to achieve the magnetization reversal. For sub-critical fields (white region), the magnetic moments rotate into a scissor-like configuration, and never reverse (e.g., points 1 and 3 in the phase diagram).

Figure 4.6 shows the time sequence of the applied magnetic field pulses during a toggle excursion. Here, the green and red arrows in the top panel represent the magnetic moments of the spin-flop pair. In equilibrium, no magnetic field is applied and the magnetic moments are aligned antiparallel to each other, as depicted in time frame \( t_0 \). We can define this state as “0” and the state where the bi-layer is rotated by 180° as “1”. Sequentially applying magnetic field pulses, as shown in Figure 4.6, leads to a clockwise rotation of the scissored magnetic moments into the opposite AP state (“0” to “1”). Repeating this sequence one more time results in switching of the magnetization state from “1” back to “0”. This sequence is known as toggling. Naturally, toggling can be performed counterclockwise, by starting the sequence with a bit-line pulse.

The magnetic toggling described above can be affected by various imperfections that can be present in an MTJ. The ideal spin-flop MTJ consists of two identical free layers and a fully flux-closed reference layer. However, even with most advanced fabrication processes it is difficult to produce spin-flop MTJ’s and avoid material or geometrical imperfections. We illustrate three possible sources of such imperfections
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Figure 4.4: Illustration of the Stoner-Wohlfarth and Toggle switching criteria. (a) For a Stoner-Wohlfarth particle switching occurs for fields outside the critical field curve known as the “astroid”. (b) For a spin-flop bi-layer crossing the L-shaped critical field curve is required for 180 degree reversal. The half-select field significantly decreases the activation energy of a SW particle (right panel in a), while the stability of the spin-flop bi-layer in fact improves (right panel in b). After D. C. Worledge [57].

Figure 4.5: Schematic of the magnetic phase diagram of the spin-flop bi-layer. $H_{SF}$ corresponds to the spin-flop critical field and $H_{XSAT}$ represents the saturation field along the EA of the elliptical particle. The gray area represents the 2D region in applied magnetic fields where magnetization can be toggled to the opposite AP state. The unshaded area is the region of zero switching probability. Magnetization can be reversed by a toggle box excursion through any point within the gray area (e.g., point 2).
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Figure 4.6: Time diagram of the toggle sequence. The green and red arrows represent the magnetic moments of the top and bottom magnetic layers. The wide blue and red arrows represent the magnetic fields acting on the bi-layer.

Figure 4.7: Illustration of possible mechanisms for magnetic asymmetry in an MTJ: (a) Neel coupling due to surface roughness, (b) thickness imbalance, and (c) fringing field from a not fully compensated reference layer.

in Figure 4.7. A rough magnetic surface produces microscopic dipoles, as shown in Figure 4.7(a), which can couple across the spacer and affect the switching properties of the system. Such coupling can reach as high as 100 Oe in extreme cases [58, 59]. Magnetic asymmetry can also be created by a small difference in thickness of the two layers forming the spin-flop pair (Figure 4.7(b)), or by a non-ideally compensated reference layer illustrated in Figure 4.7(c). Most of the resulting magnetic asymmetry can be compensated by suitable external field biasing. In our samples, the combined magnetic asymmetry was in the range of 5 to 10 Oe.

4.3 Macrospin theory

The first approximation in describing the magnetization behavior of a magnetic nanoparticle is the macrospin or single-domain (SD) model, which has been used
very successfully used for modeling particles with lateral dimensions of 100 nm and smaller. In this limit the model is essentially exact and the switching occurs through coherent rotation of magnetization rather than domain wall nucleation and propagation. As regards the spin-flop system, it has been found that even larger particles, 100-500 nm in size, can be well approximated by the macrospin model when it comes to their quasistatic properties, such as the phase diagram [57, 60–63] and the critical switching curve [64, 65]. In this section we overview the results of this model for the dynamics of the spin-flop system, including such key properties as the collective spin-resonance modes and their variation with applied magnetic fields. We find that, in extended regions of the magnetic phase space of the system, the model provides a good description of the experimentally observed behavior, and that other regions of the phase space are best analyzed using micromagnetic simulations (Papers 1, 2, and 5).

The spin-flop bi-layer is typically taken to consist of two ellipsoids of revolution, in order to have the demagnetizing coefficients expressed analytically. The second common approximation is that the magnetic moments are confined to the film plane – this is true to a good accuracy for thin films where the large shape anisotropy forces the spins into the plane. The intrinsic anisotropy is taken to be uniaxial, and coaxial with the shape anisotropy. The dipole field outside each layer was taken to equal the demagnetizing field inside the respective layer – a very good approximation when the magnetic layers are separated by a thin spacer. A more detailed case, including a thickness imbalance, arbitrarily directed anisotropy, and a Néel coupling between the layers is found in Ref. [63].

The model geometry is shown in Figure 4.8. The WL and BL field directions are aligned at 45° to the EA of the particles. The blue and red arrows in Figure 4.8 represent the magnetic moments in the top and bottom magnetic layers directed at angles \( \theta_1 \) and \( \theta_2 \), respectively.
The total energy of the system, $E$, is the sum of the energy terms for each magnetic particle and a term representing the interaction between the particles. These include the Zeeman energy, magnetostatic energy, intrinsic anisotropy of the material, and dipolar inter-particle interaction. Specifically:

$$E(\theta_1, \theta_2)/VM_S = -H_x(\cos \theta_1 + \cos \theta_2) - H_y(\sin \theta_1 + \sin \theta_2) + (N_x M_S - j) \cos \theta_1 \cos \theta_2$$

$$+ (N_y M_S - j) \sin \theta_1 \sin \theta_2 + \frac{1}{2} [(N_y - N_x) M_s + H_i] \times (\sin^2 \theta_1 + \sin^2 \theta_2),$$

(4.2)

where $V = \frac{1}{4} \pi abt$ is the particle volume, $H_{x,y}$ – the $x$ and $y$ components of the applied field, $N_{x,y} = 4\pi n_{x,y} \frac{t}{b}$ – the demagnetizing factors, $H_i$ – the intrinsic anisotropy field, and $j = \frac{J t}{M_s}$, with $J$ being the interlayer exchange coupling constant and $t$ – the thickness of the magnetic layer. The $j$-term can be neglected because of negligible interlayer exchange coupling in the samples used in this work (true for the TaN spacers used). The demagnetizing factors $n_{x,y}$ are given in Refs. [57, 60, 61]. The samples used in this study have the aspect ratio of $a/b = 1.2$, which corresponds to $n_x \approx 0.62$ and $n_y \approx 0.82$. In Eq. 4.2 it is assumed that the dipole field outside the nanoparticle, acting on the other layer is equal to the internal demagnetizing field of the particle. This is true in the limit of small separation between the layers, and can be adjusted by a constant of order 1 for thicker spacers.

The minimum of the energy described by Eq. 4.2 represents a stable state of the bi-layer system. The global energy minimum at zero field corresponds to the AP alignment of the magnetic moments. In an external magnetic field, the energy minimum shifts its position in terms of $\theta_1$ and $\theta_2$, and the two magnetic moments rotate. When a certain critical field applied along the EA is reached, switching into a scissors state takes place. By calculating the positions of the energy minima at different magnetic fields, the phase diagram of the system can be obtained [60, 61].

The key field scales are:

$$H_{sf} = \sqrt{H_i \left(8\pi M_s n_y \frac{t}{b} - \frac{2J}{M_s t} + H_i\right)},$$

(4.3)

$$H_x = \left(8\pi M_s n_y \frac{t}{b} - \frac{2J}{M_s t} + H_i\right) \times \sqrt{H_i \left(8\pi M_s n_y \frac{t}{b} - \frac{2J}{M_s t} + H_i\right)},$$

(4.4)

$$H_{xxsat} = 8\pi M_s n_x \frac{t}{b} - \frac{2J}{M_s t} + H_i,$$

(4.5)

$$H_{ysat} = 8\pi M_s n_y \frac{t}{b} - \frac{2J}{M_s t} + H_i.$$
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The magnetization dynamics of the spin-flop system can be analytically modeled by solving the system of two coupled LLG equations:

\[
\frac{d\vec{M}_{1,2}}{dt} = -|\gamma|\vec{M}_{1,2} \times \vec{H}_{\text{eff}}^{1,2} + \alpha \vec{M}_{1,2} \times \frac{d\vec{M}_{1,2}}{dt},
\]

where \(\vec{H}_{\text{eff}}^{1,2}\) are the effective magnetic fields acting on each of the two ferromagnets.

The result is that the system has two main resonance modes: the acoustical mode, where the two magnetic moments oscillate in phase with a constant angle between them; and the optical mode, where the moments exhibit scissor-like out-of-phase oscillations with varying angle between them. Figure 4.9 illustrates these acoustical (a) and optical (b) resonance modes, as well as a simple mechanical analogy based on two pendula connected by a spring (c, d). Depending on the specifics of the applied magnetic field, the acoustical, the optical, or both modes can be excited. For example, an alternating in-plane magnetic field applied off the EA forces optical oscillations and suppresses the acoustical mode.

Paper 1 details the dynamics of the spin-flop system and shows that, indeed, collective spin resonances replace the dynamics of the individual layers. The dynamics in the AP ground state differs significantly from that in the scissor state at intermediate fields. The analytically calculated dependence of the resonance frequencies on the applied EA magnetic field is presented in Figure 4.10. The approximate expressions for the optical and acoustical resonance frequencies in the AP ground state are:
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Figure 4.10: Analytically calculated optical and acoustical resonance frequencies for different values of the EA magnetic field [66].

\[ f_{\text{AP}}(H_{EA} = 0) \approx \gamma \sqrt{(4\pi M_s + H_i)H_i}, \]  
\[ (4.8) \]

\[ f_{\text{o}}(H_{EA} = 0) \approx \gamma \sqrt{(4\pi M_s + H_i)H_{\text{ysat}}}, \]  
\[ (4.9) \]

where \( \gamma = 27.8 \text{ GHz/T} \). These eigen-frequencies quantitatively are given in Figure 4.10 for a 400x490 nm spin-flop bi-layer.

Taking into account the fact that the anisotropy field is substantially smaller than the saturation field, \( H_i \ll 4\pi M_s \), equation 4.9 can be simplified by substituting equation 4.6 into 4.9 to obtain the dependence of the optical spin-resonance frequency on the width of the particle, successfully verified on the experiment in Paper 1:

\[ f_{\text{o}}(b) \approx 4\pi \gamma M_s \sqrt{2n_y b} \sim b^{\frac{1}{2}}. \]  
\[ (4.10) \]

The high speed memory operation requires write and read times of the order of ns. It was shown experimentally that MRAM is capable to perform fast writing operations on sub-ns time scale [67]. Fast precessional switching, resulting in sub-ns magnetization reversal, was considered in [68]. A single-domain numerical study of the spin-flop dynamic reversal was performed in [69] and showed that particular attention should be paid to the form and duration of the field pulses as well as to temperature fluctuations during the high-speed writing. High-frequency
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magnetization "ringing" should be avoided. This can be done by fine tuning the magnetic field pulses and/or the material of the free layer. On the other hand, the pronounced dynamic properties of the spin-flop system described above can be used for resonantly enhancing the switching process – the topic of the next section.

4.4 Microwave and thermally assisted switching

There is a term, magnetic recording trilemma, used in the field of magnetic hard disc drives [70–73], but perhaps can be extended to the MRAM field as well. Generally, this term is used regarding the tradeoffs between the signal-to noise ratio (SNR), thermal stability, and writability of the magnetic medium. For MRAM, the magnetic fields necessary to reverse magnetization in a memory cell and thereby write data should be as small as possible. Thermal fluctuations of magnetization should not lead to data loss, and the magnetoresistance ratio should be high enough to provide a high signal-to-noise ratio for readout. These issues are especially important for further miniaturization of MRAM. The activation energy of a magnetic particle decreases with shrinking dimensions, which leads to data loss due to thermal agitation. Thermal stability and writability require fine tuning of the magnetic material parameters [74] or changes in the shape of the magnetic storage elements [75]. Thermal stability can be increased by using magnetically stiffer materials or by increasing the shape anisotropy. This, however, results in higher write fields (currents) and therefore higher power consumption.

Finding methods for low-field switching while keeping thermal stability in the safe range is a challenging task for memory developers. Different approaches have been taken to address this issue [75, 76], and microwave assisted switching of magnetization is considered to be one of the promising alternatives. It consists of using a relatively weak alternating magnetic field, typically at the resonance frequency of the free layer, applied simultaneously with a quasistatic reversing magnetic field. The resulting resonant oscillation of magnetization can significantly decrease the reversing field requirements, while the static thermal stability of the memory cell is kept unchanged.

Recently, attention has been paid to microwave assisted switching using magnetic fields [77–81] as well as spin-polarized currents [82]. A substantial decrease in coercivity was reported [80], which potentially can be used in novel magnetic memory devices.

Resonantly enhanced switching of a single magnetic particle has been of great interest for the past few years, yet resonant switching of the spin-flop system remains unexplored. In this section, we overview the physics – the details, including our experimental results and micromagnetic analysis are presented in Paper 5.

The ground state of the spin-flop bi-layer is a uniform AP state. Continuous driving the bi-layer with an off-axis microwave field results in a scissor-like motion of the two magnetic moments, described in the previous section. Predominantly the optical resonance mode is excited, while the acoustical mode is suppressed.
4.4. MICROWAVE AND THERMALLY ASSISTED SWITCHING

High-amplitude optical-mode oscillations can lead to a dynamic magnetization reversal. In an ideal bi-layer, once the critical amplitude of the microwave pumping is reached the system should continue to switch between the two AP states as long the microwave field is on. In the presence of magnetic asymmetry (e.g., a small DC field bias) unidirectional resonant switching of magnetization can be obtained.

The resonant switching process can be modeled in detail using the macrospin model with two coupled LLG equations for the two moments. This approximation is particularly accurate for not too large driving amplitudes. For very high amplitudes the motion of the magnetization becomes partially un-harmonic and can be more accurately modeled using the micromagnetic approach. (See Paper 5 for details on both.)

Here, in order to better visualize the physical process involved we discuss a mechanical analogy. The system consists of two coupled harmonic oscillators of mass $m_1$ and $m_2$, connected by a spring, placed in a double well potential, as illustrated in Figure 4.11. The two potential wells schematically represent the two stable AP states of the spin-flop bi-layer. The spring represents the dipolar coupling between the layers, forcing the particles to occupy opposite wells at equilibrium (AP alignment of magnetization). Magnetic asymmetry can be represented by different particle masses: the heavier particle $1$ would tend to occupy the lower well, forcing the lighter particle $2$ via the spring (dipole repulsion) to occupy the opposite well.

The externally applied alternating magnetic field is represented here by tilting of the potential left and right every half-period of the field, thereby forcing the particles to oscillate in the wells and impinge onto the respective barriers. At low microwave amplitudes, both particles oscillate in a harmonic fashion at the bottom of the potential wells. By increasing the excitation amplitude (amount of tilting), the particles oscillate with a larger amplitude and, at some critical amplitude, can escape to the opposite well (switching of magnetization or spin-flop). In this process particles 1 and 2 exchange their positions. At sufficiently large amplitudes of the excitation, the particles switch between the two states continuously. The described process has a resonant nature - when the frequency of the microwave field matches the resonant frequency of the harmonic oscillators, the oscillation amplitude is greatly enhanced, which leads to a much high probability of switching. For a certain range in the resonant field amplitude, no switching will take place outside the resonance region.

The quasistatic magnetic field applied to the bi-layer can be pictured as a static tilting of the potential. At some critical tilting (DC magnetic field bias) the repulsive force of the spring connecting the particles can no longer hold them apart in the opposite wells, and both particles roll to the lower potential well thus forming a scissor-like magnetization state.

At zero temperature, the spin-flop switching threshold should be very sharp. A continuous-wave excitation with an amplitude slightly lower than the critical amplitude necessary for dynamic spin-flop switching will result in steady state magnetization oscillations without reversal. Field amplitudes slightly higher than the threshold would then result in immediate magnetization reversal, so that further
CHAPTER 4. SPIN-FLOP DYNAMICS

Figure 4.11: Simplified model of microwave assisted and thermally assisted switching, picturing two harmonic oscillators in a double well potential. The spring connecting the particles represents the dipolar coupling between the spin-flop layers, which forces the particles to occupy opposite wells (AP alignment of magnetization). Magnetic asymmetry is represented by a tilt of the potential or a difference in mass between the particles, $(m_1 \neq m_2)$. Microwave magnetic field with amplitude $h$ and frequency $\omega$ causes the tilt to oscillate left and right (red and blue dashed lines). If the amplitude of the excitation is insufficient for magnetization reversal, thermal fluctuations can provide additional energy resulting in the escape of the particles, bringing the system to the opposite AP state.

Increasing the excitation amplitude would not have any significant effect. Thermal fluctuations, however, can alter this resonant switching behavior. Oscillations in the system with amplitude slightly lower than the dynamic switching threshold will, in the presence of thermal noise, result in the particles escaping from well to well (spin-flop), with the probability proportional to the strength of the thermal noise. The switching mechanism becomes statistical in nature: the probability that the particle has escaped increases with increasing the waiting time in the subcritical "tilted" state. The presence of thermal noise can be modeled analytically as well as micromagnetically, with the thermal fluctuation term included as was described in section 3.2.4.

Thermal stability is a critical issue for magnetic memory applications. Once information is written, it should be stable (non-volatile) over a certain period of time. In the memory technology, non-volatility is usually defined by a maximum error rate of $10^{-9}$ over a period of 10 years [6]. In regards to the above double well potential, the energy barrier between the two wells should be about $60 k_BT$ in order to provide the desired stability. Thus, the energy barrier to thermal agitation, also known as the activation energy should be sufficiently large to provide stability, and at the same time not too high for power consumption reasons related to the
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Writability.

Thermally activated switching has mostly been studied on single ferromagnetic particles [69, 81, 83]. Only a few studies report the effect of temperature on the magnetization reversal in exchange or dipole coupled ferromagnets [84, 85]. Nevertheless, it is now established that thermal fluctuation can play a significant role in dynamic switching processes [69, 86], with the thermal agitation effectively lowering the potential barrier to reversal.

The switching probability, \( p(t) \), is an exponential function of the energy barrier relative to the thermal energy [6, 84]:

\[
p(t) = f_\gamma \exp \left( -\frac{E_b}{k_B T} \right) < 1, \tag{4.11}
\]

where \( f_\gamma \) is the Larmor spin precession frequency of the order of GHz, and \( E_b/k_B T \) – the energy barrier to activation (in terms of \( k_B T \)). Fitting experimental data on the switching probability and extrapolating using the SD model [60, 61], allows to determine the activation energy of the bi-layer in the ground state, which corresponds to the state of long term data storage in MRAM.

As mentioned earlier, an analytical description of resonant activation in submicrometer spin-flop bi-layers can be rather complex and/or insufficiently accurate, and one then resolves to micromagnetic simulations. For other systems, relatively simple analytical solutions for the resonant activation behavior can be derived, e.g., for one-dimensional systems. One example is a current-biased Josephson junctions (JJ) analyzed by Devoret et. al. [87]. In this work the JJ was biased with a subcritical current and excited with a weak microwave current, at the plasma resonance frequency of the system. The measured resonant thermal activation was explained using a model of the particle (quasi-charge in the JJ case) escaping from a tilted potential well obtained by proper biasing of the JJ [88]. The spin-flop system contains two moments and therefore at least two dimensions (neglecting the out-of-plane precession), and should require a more sophisticated theory, including the Fokker-Planck type treatment of the switching probability.
Chapter 5

Vortex pair dynamics

Ferromagnetic nanoparticles usually exhibit uniform magnetization distribution. However, when the size exceeds the single domain limit and the shape is circular or nearly circular, the magnetization distribution in equilibrium can become highly non-uniform. One example is the vortex state [89], which can be the ground state of a magnetic nanoparticle, stable enough to survive moderate (~100 Oe) external magnetic fields. In a vortex, most of the spins curl around, forming a flux-closed structure. The spins in the middle of the particle, forced by the exchange interaction, point out of plane forming a vertically polarized vortex core. The size of the core is of the order of the exchange length [90], and its polarization (or polarity) can be up or down. Recent studies of spin vortices [91–93] indicate their potential for magnetic data storage.

In an elliptical magnetic element, the vortex state of magnetization can be more energetically favorable than the uniform state, depending on the element’s aspect ratio (length to width) [89, 94]. The demagnetizing field is reduced in the vortex state due to its flux-closed structure, except for the small region of the vortex core, where magnetization is aligned normal to the film plane. The vortex state is then characterized by the polarity of the vortex core (either up or down) and the chirality of the spins at the periphery of the vortex (CW or CCW). For a single vortex, the two polarity and two chirality states are energetically degenerate. This is no longer true for two interacting vortices where, for example, the core-core interaction energetically distinguishes the states of parallel and anti-parallel core polarizations.

Figure 5.1 shows a typical spin vortex in a 100 nm Permalloy disk. According to [95, 96], a vortex can be created during a magnetization reversal process in relatively thick (~10 nm) ferromagnetic particles. Thin ferromagnetic particles can also exhibit stable vortex states, created for example by nucleation from a spin-disordered state produced by a strong magnetic field excitation.

The dynamics of a vortex in a thin film ferromagnet can depend on the vortex core polarization. Displaced from its equilibrium position, the core moves to the
equilibrium position in a circular trajectory, the direction of which (clockwise or counterclockwise) depends on the polarization of the vortex core. This was theoretically predicted by Thiele for the motion of a domain wall in a two-dimensional ferromagnet [97] and modified for the case of a magnetic vortex by Huber [98]:

\[ \vec{G} \times \frac{d\vec{X}}{dt} + \vec{F} = 0 , \]  

(5.1)

where \( \vec{G} \times \frac{d\vec{X}}{dt} \) or \( \vec{G} \times \vec{v} \) is the so called \textit{gyroforce}, directed orthogonal to the velocity and forcing the motion of the core along a circular trajectory.

The next approximation was to attribute a mass \( M \) to the vortex core [99, 100] and, thereby, add an inertial term to the Thiele equation:

\[ \vec{G} \times \frac{d\vec{X}}{dt} + \vec{F} = M \frac{d^2\vec{X}}{dt^2} , \]  

(5.2)

where \( \vec{v} \) and \( \vec{a} \) are respectively the velocity and acceleration of the vortex core, \( \vec{F} \) represents the restoring force, \( \vec{G} = G\vec{e}_z \) is the gyrovector – the product of the gyroscopic constant \( G \) and a unit vector orthogonal to the film plane, \( \vec{e}_z \). Figure 5.2 illustrates this classical model of vortex dynamics, which is in essence similar to the movement of a particle in a harmonic potential.

Thiele’s equation explains the movement of a vortex core displaced from the equilibrium position and reaching its energetic minimum in a stationary circular orbit under a continuous-wave excitation. This \textit{gyrational} mode of the core motion has the largest amplitude and is most easily detectible. Therefore, in the literature mainly this mode of the vortex core motion was considered [101, 102]. Recently, high-frequency modes of vortex core dynamics have been observed [103] and interpreted numerically and analytically [104] as a low-frequency gyration with superposed high frequency oscillations. There are two high-frequency eigen-modes
Figure 5.2: Illustration of the Thiele equation describing the forces acting on a magnetic vortex core displaced from its equilibrium position.

having opposite precessional directions. A phenomenological equation of third order in time for the core coordinate $\vec{X}$ was proposed by Mertens et.al. [105] to better describe the high frequency core dynamics, and has the form:

$$G_3 \left( \vec{e}_z \times \frac{d^3 \vec{X}}{dt^3} \right) + M \frac{d^2 \vec{X}}{dt^2} + G \left( \vec{e}_z \times \frac{d \vec{X}}{dt} \right) = \vec{F}. \quad (5.3)$$

This equation describes non-Newtonian dynamics of the complex core motion. In addition to the standard gyrotropic term, equation 5.3 contains an inertial term proportional to the core mass $M$, and a gyrotropic term of higher order describing the high frequency oscillations. A general solution of this equation can not be obtained. However, the eigen-frequencies of the core oscillations $\omega_n$ can be expressed in terms of $G_3$, $M$, $G$ and $\kappa$ for small displacements of the core from the equilibrium position [106, 107]. Using the inequalities $\omega_0 \sim \Delta \omega \ll \bar{\omega}$, $2\bar{\omega} = \omega_2 + \omega_1$ and $\Delta \omega = \omega_2 - \omega_1$, the following simple relations can be derived [106]:

$$\omega_0 = \frac{\kappa}{G}, \quad G_3 \omega^2 = G, \quad G_3(\Delta \omega + \omega_0) = M,$$

where

$$G_3 = 0.625 \frac{R}{4\pi \gamma^3 M_s}, \quad M = 0.58 \frac{L}{\gamma^2}, \quad G = \frac{2\pi LM_s}{\gamma}, \quad (5.5)$$

$R$ is the radius of the nanoparticle, $L$ - its thickness. The force acting on the vortex in a circular particle is $\vec{F} = -\delta U/\delta \vec{X} = -\kappa(\vec{X})\vec{X}$, where the dependence
of the restoring force coefficient $\kappa \left( |\vec{X}| \right)$ on $|\vec{X}|$ determines the nonlinearity of the system.

The resonance frequencies of the core motion are

$$\omega_0 = 20\gamma M_S L/9 R ,$$

$$\bar{\omega} \approx 0.9 \cdot 4\pi\gamma M_s \sqrt{L/R} , \quad \Delta \omega \approx 3.5\omega_0 .$$

These analytically obtained resonant frequencies for a single vortex core agree well with the numerically simulated ones, as was shown in [104].

Magnetization states in spin-flop bi-layer particles are rather different from those observed in single ferromagnetic particles. The ground state usually has two near-single-domain particles aligned anti-parallel due to the dipolar interaction across the nonmagnetic spacer. For certain aspect ratios (thickness/diameter), however, stable vortex pair states in bi-layers have been predicted [108]. The samples used in this work have elliptical in-plane geometry favoring antiparallel alignment of the two magnetic moments in the ground state at zero field, due to a relatively strong dipolar coupling. Paper 3 shows that vortex pairs can nevertheless be very stable states of the spin-flop system, even though they are not the ground state of the system. Due to the small thickness of our Permalloy layers, a vortex cannot be nucleated during the magnetization reversal process. Instead, a strong alternating magnetic field must be applied to create a vortex pair.

Laterally separated magnetic nanoparticles each in the vortex state [95, 109–111] as well as vortex pairs in a single particle [112, 113] were studied recently and found to exhibit a variety of gyrational modes. Furthermore, the interaction of two vertically stacked vortices has recently been studied theoretically [114] and experimentally [115]. In all these studies, however, the interaction between the vortex cores was negligible. Indeed, in order to study the core-core interaction, the cores should be in the immediate vicinity to each other. Since the magnetic field decays away from the vortex core as $1/r^3$, the core-core separation should be of the order of 1 nm in order for the core-core coupling to be significant. This limit can be realized in spin-flop bi-layers, with both layers in the vortex state, separated by a thin non magnetic spacer. A model of such a vertical vortex-pair system has recently been reported by Guslienko et. al. [108], again however neglecting the core-core interaction considered insignificant due to the small size of the cores. Numerically, some aspects of the system have been modeled by S-H. Jun et. al. [116], however the relatively thick spacer and large mesh size made the core-core effects negligible. Thus, the limit of strongly coupled vortex cores and their dynamics remains unexplored, and is discussed at length in Paper 4 appended.

Since the magnetic vortex in a single nanomagnet has 4 degenerate in energy configurations (chirality clockwise/counterclockwise and vortex core polarity up/down), a sandwich of two magnetic layers has the total of 16 vortex states.
Basic symmetry operations reduce these 16 states to 4 non-degenerate classes with 4 vortex-pair states in each. The 4 non-degenerate classes can be represented by the vortex pairs shown in Figure 5.3: Parallel vortex cores and Parallel chirality (P-P); Antiparallel cores, Parallel chirality (AP-P); P-cores, AP-chirality (P-AP); and AP-cores, AP-chirality (AP-AP). Parallel alignment of the vortex cores results in a bound vertical core-core pair at zero field, while AP cores repel each other when on-axis. These properties of the core-core interaction can be used to experimentally identify the individual vortex polarity/chirality configurations, as shown in Paper 4.

The response of a vortex pair to an external magnetic field is expected to be strongly dependent on the vortex chirality in each magnetic layer, even in the absence of core-core coupling. The two vortex cores, forced by the field, move in the same direction in the case of like chiralities ($C_1=C_2=1$) or in the opposite directions and perpendicular to the field in the case of opposite chiralities ($C_1=0$; $C_2=1$) [117]. When separated by field or non-interacting directly due to a thick spacer the cores gyrate about their equilibrium positions at sub-GHz frequencies, similar to the single vortex case discussed earlier [102]. For strongly coupled cores, e.g., in the P-AP state, the single-core gyrational motion is effectively suppressed and a new rotational resonance appears, in which the two cores rotate about their magnetic center of mass. Similar to the single vortex case, the high-frequency modes (due to the non-linear dynamics) are present, although the strong coupling between the cores suppresses their amplitude.

The vortex-pair dynamics can be described using the basic non-linear model developed for single vortices [104]. The trajectory of the vortex cores is described by a phenomenological equation of third order in time based on the Thiele equation:

$$G_3 \left( \vec{e}_z \times \frac{d^3 \vec{X}}{dt^3} \right) + M \frac{d^2 \vec{X}}{dt^2} + G \left( \vec{e}_z \times \frac{d\vec{X}}{dt} \right) = \vec{F}.$$  (5.8)
The main difference between the equation for a single vortex and for a pair of interacting vortices is that the potential energy of the vortex core has an additional term \( \kappa(\vec{r}_1 + \vec{r}_2)^2 \), responsible for the magnetostatic interaction between the cores:

\[
U = \frac{1}{2} \left[ k_1(x_1^2 + x_2^2) + k_2(y_1^2 + y_2^2) + \kappa(\vec{r}_1 + \vec{r}_2)^2 \right],
\]  

(5.9)

where \( k_{1,2} \) are the coefficients of the returning force, which are different for the two in-plane axes of the elliptical particle, and \( \kappa \) – the coefficient determining the interaction between the vortex cores, with \( \vec{r}_{1,2} \) being the vortex cores’ coordinates in layers 1 and 2, respectively. Taking into account that \( \vec{F}_{1,2} = -\delta U/\delta \vec{r}_{1,2} \), equation 5.8 transforms into a system of two independent equations:

\[
G_3 \frac{d^3 x}{dt^3} + M \frac{d^2 y}{dt^2} + G \frac{dx}{dt} + (k_1 + 2\kappa)y = 0,
\]

(5.10)

\[
-G_3 \frac{d^3 y}{dt^3} - M \frac{d^2 x}{dt^2} + G \frac{dy}{dt} + (k_2 + 2\kappa)x = 0.
\]

(5.11)

The solutions are of the form

\[
x_\alpha = \sum_\alpha A_\alpha \cos(\omega_\alpha t + \phi_{0,\alpha}), \quad y_\alpha = \sum_\alpha B_\alpha \cos(\omega_\alpha t + \phi_{0,\alpha}),
\]

(5.12)

where \( \alpha = 0, 1, 2 \) are three normal modes of the core oscillations, \( \phi_{0,\alpha} \) – the initial phases and \( A_\alpha \) and \( B_\alpha \) – the amplitudes of each mode, related as

\[
\frac{A}{B} = \sqrt{\frac{k_1 + 2\kappa - M\omega^2}{k_2 + 2\kappa - M\omega^2}}.
\]

(5.13)

The frequencies of the eigen-modes are determined by the dispersion relation:

\[
\omega^2(G_3\omega^2 - G)^2 - [M\omega^2 - (k_1 + 2\kappa)] [M\omega^2 - (k_2 + 2\kappa)] = 0.
\]

(5.14)

Equation 5.14 can be simplified assuming that the difference in coefficients \( k_1 \) and \( k_2 \) is negligible for elliptical particles with small aspect ratio (our case):

\[
G_3\omega^3 - M\omega^2 - G\omega + \bar{k} + 2\kappa = 0.
\]

(5.15)

Solving the cubic equation 5.15 gives explicit expressions for the three resonant modes:
\[ m = \pm 1, \quad \tilde{k} = \sqrt{k_1 k_2} = \frac{40\pi M_s^2 L^2}{9R}, \]
\[ \omega_0 = \frac{(\tilde{k} + 2\kappa)}{G}, \quad \omega^2 = \frac{G}{G_3}, \quad \Delta \omega = \frac{M}{G_3 - \omega_0}. \quad (5.16) \]

In the case of coupled vortex cores (P-AP/P states), coefficient \( \kappa \) in equation 5.15 is much more significant than \( \tilde{k} \), which can therefore be neglected. In the case of decoupled vortex cores, \( \tilde{k} \) is much stronger than \( \kappa \) and equation 5.15 yields solutions, which are essentially those for single non-interacting vortices. These predictions are in good agreement with our experimental data and micromagnetic simulations, discussed in detail in Papers 3 and 4.

We will briefly mention another interesting aspect of vortex core dynamics discussed in the literature. It has been reported that a vortex core can be an extremely stable formation, requiring a perpendicular to the plane field of over 1 kOe for switching its polarity [118]. However, the polarity can be dynamically reversed relatively easily by applying short ns-range magnetic field pulses [92, 119]. Once a magnetic field pulse is applied, the vortex core starts to move perpendicular to the direction of the applied field. When the speed of the core movement exceeds some critical value, a vortex-antivortex pair can be formed right behind the moving original core. At above-critical amplitudes of the excitation (the critical speed of the vortex core movement in Permalloy was reported to range from 250 m/s [91] to 320 m/s [120]), the original core annihilates with the newly formed antivortex. The resulting core is the newly created vortex, which turns out to be of opposite polarity compared to the original one [121].

Magnetic vortices have the potential to be used in non-volatile magnetic memory, where a bit of information can be represented by the two core polarizations, spin up or down. Furthermore, it is conceivable that a spin vortex can carry two bits of information stored in the vortex core polarization and chirality (sense of rotation) [122]. Possible technical implementations were proposed recently [123–125], where the writing of information can be realized by magnetic field [126] or spin-polarized current pulses [127, 128].
Chapter 6

Experimental methods

6.1 MRAM samples

The spin-flop magnetic tunnel junctions studied in this thesis were produced using the methods described in [129]. The samples were elliptical Permalloy bi-layers of width 350-500 nm and aspect ratio ~1.2. The layer structure of the samples was similar to the one described in [74], consisting of two dipole-coupled Permalloy nanoparticles of 5 nm thickness, separated by a TaN spacer with negligible exchange coupling through it. An AlO$_2$ tunnel barrier serves as an interface between the soft magnetic bi-layer and the synthetic antiferromagnet (SAF) flux-closed reference layer, as shown in 6.1. The resistance of the MTJ stack is determined predominantly by the resistance of the tunnel barrier and is in the range of 1-1.5 kΩ. The TMR is approximately 20 % and is determined by the relative magnetization orientation of the bottom free and the top reference layers: their anti-parallel alignment results in a high-resistance state whereas parallel alignment results in a low-resistance state of the MTJ. For this reason, only the magnetization direction of the bottom free layer can be determined from experimentally measured resistance. The magnetic state of the top free layer contributes to the MR signal through its effect on the bottom free layer. The average magnetization direction in the bottom free layer can be obtained using equation 3.10, assuming that the magnetization in the CoFeB reference layers is fixed along the EA. The magnetic moments of the two free layers align anti-parallel in the ground state due to the strong dipole interaction through the spacer. The moments can be rotated by intermediate magnetic fields whereas the magnetization of the reference layers is aligned along the long axis (EA) of the elliptical particles and is not affected by applied magnetic fields. This guarantees that the measured magnetoresistive signal comes predominantly from the magnetization rotation in the free layers.

The high frequency magnetic field was applied to the junctions using integrated write lines embedded near the MTJ stack and angled at 45 degrees to the EA of the Py free layers. The write lines had 50 Ω resistance for impedance matching.
CHAPTER 6. EXPERIMENTAL METHODS

Figure 6.1: Layer structure of a typical MTJ sample studied in this work. The MTJ is connected at the top and bottom to integrated read out lines. A separate write line (not shown) is electrically decoupled from the junction.

The write line was electrically decoupled from the read out lines connected to the top and bottom of the nanopillar junction. The write and read lines had pads for surface probe access. In order to minimize the high-frequency signal distortions, the surface probes had the bandwidth of 40 GHz, significantly above the highest-speed signals used.

6.2 Measurement techniques

6.2.1 Quasistatic measurements

The electrical measurements were performed using a room temperature probe station. A toroidal electromagnet was used for magnetoresistance measurements and setting the spin-flop bi-layer magnetization into a desired ground state [74]. The magnetoresistance was measured quasistatically by stepwise changing the external magnetic field and simultaneously measuring the voltage drop across a current biased tunnel junction. The samples were placed in the centre of the toroidal magnet designed to produce two-directional in-plane magnetic fields of strength up to 350 Oe. Figure 6.2.1 shows a sample placed into the measurement setup. The magnetic field perpendicular to the sample plane was produced by bringing a suitably shaped and positioned permanent magnet closer or farther away from the sample, which gave up to 1500 Oe of a vertically directed field.

6.2.2 Dynamic measurements

The high speed measurements were conducted by sending a high frequency current through the write lines while simultaneously measuring the MTJ resistance. The schematic of the measurement setup used for high-speed measurements is shown in Figure 6.2.2. In order to decrease the impedance mismatch between the TJ and the high-frequency measurement circuits, the junctions for this study were made with substantially smaller resistance (much thinner tunnel barrier) than
6.2. MEASUREMENT TECHNIQUES

Figure 6.2: Image of the probe station. Sample, placed inside a toroidal magnet, is contacted by two high bandwidth probes: one for applying alternating magnetic fields and the other for reading out the resistance response of the tunnel junction.

Figure 6.3: Schematic of the experimental setup used for high frequency measurements. Microwave fields were applied to the MTJ by sending pulsed or continuous wave current through the write line. The static and dynamic resistance components were measured while current biasing the MTJ.

the ones described in [129]. The DC bias and the radiofrequency resistive response across the junctions were separated at the surface probe using a bias tee. The dynamic measurements were performed using two methods: real-time resistance measurements of the free spin-oscillations following an short (~100 ps) field pulse and microwave spectroscopy. These methods are illustrated in Figure 6.2.2.
Figure 6.4: Schematic of the real-time and spectroscopic techniques. The frequency spectra were obtained by recording the MTJ resistance response to a 200 ps field pulse and performing FFT of the recorded signal. The MTJ's resonance properties were measured also using the microwave spectroscopy technique, consisting of measuring the quasistatic resistance under a continuous-wave field excitation of a given frequency, and repeating the measurement while stepping the frequency.

The real-time technique utilizes time-domain measurements of the MTJ’s resistance as a response to a short in-plane magnetic field pulse. The magnetization in the free layers, excited by the magnetic field pulse, starts to oscillate, causing a change in the resistance proportional to the amplitude of the oscillation. The FFT of the time evolution of the recorded resistance gives information about the resonant properties of the spin oscillations in the free layer after the field pulse is removed. We used 200 ps full-width-at-half-maximum pulses for magnetically exciting the junctions. The subsequent resistance response was amplified by +64 dB and recorded using a 40 GS/s Agilent DSO80604B real-time oscilloscope with 6 GHz real-time bandwidth.

10 ns time traces containing the resistance response to a field pulse were averaged up to $10^6$ times in order to improve the signal to noise ratio. It was determined experimentally that the optimal amplitude for the pulse excitations is 20 Oe, which was used to obtain the real-time experimental and micromagnetically simulated data detailed below.

The microwave spectroscopy measurement [66, 130] consists of excitation the sample with a swept-frequency microwave field and measuring the sample resistance quasistatically. An alternating magnetic field forces the magnetic moments in the
ferromagnetic bi-layer to oscillate towards and away each other in a scissor-like manner, which in turn causes the layer magnetization averaged over time to deviate from its equilibrium state along the EA. This is reflected in the deviation of the resistance from its ground state. The deviation is proportional to the oscillation angle of magnetization and is greatly enhanced at spin-resonance frequencies of the bi-layer, especially at the optical spin-resonance where the precession is scissor-like. This makes the optical resonance mode easily detectable by microwave spectroscopy, as opposed to the acoustical mode, which is suppressed by the continuous-wave field excitation. In general, magnetization oscillations in both the free layers and the reference layers can contribute to the resistance response. However, the response from the strongly pinned CoFeB reference layer is much weaker than that from the free layer. Our micromagnetic simulation of the reference layer magnetization in response to moderate magnetic field excitations confirmed that the deviation of the magnetization from its equilibrium position is negligible. Therefore, the magnetoresistive response is determined mostly by the magnetization dynamics in the free layer. Microwave spectroscopy is more sensitive than the real-time technique; however only the resistance response to continuous-wave excitations can be measured.

Both the real-time and the microwave spectroscopy techniques require calibration of the applied microwave field. The data presented in this work were collected after proper calibrations were applied to the field amplitude in the full frequency range.

6.3 Micromagnetic modeling

Both quasistatic and dynamic responses of the magnetization to external magnetic fields were modeled micromagnetically, using the finite difference software package OOMMF [47] . OOMMF allows modeling of the magnetization behavior in magnetic elements of arbitrary shape by solving the dynamic LLG equation or by using energy minimization. The micromagnetically modeled structures had the same geometry as the experimental samples: two elliptically-shaped Py layers separated by a nonmagnetic spacer, through which the exchange coupling was negligible. Only the Py layers was modeled micromagnetically; the reference layer was omitted from consideration due to its negligible influence on the magnetization dynamics studied. The modeling was conducted using the configurations employed in the experiment, with quasistatic and/or high-frequency magnetic fields, continuous-wave or pulsed.

The typical material parameters for Permalloy were used: saturation magnetization $M_s=840 \times 10^3$ A/m, exchange constant $A=1.3 \times 10^{-11}$ J/m, damping constant $\alpha=0.013$. The intrinsic anisotropy field was equivalent to what was measured experimentally, approximately 5-10 Oe. The exchange between top and bottom Py layers was neglected. The simulated volume was meshed into cells of size 1 to 5 nm. Fine meshing with the cell size of 1x1x1 nm was necessary [131] in order to
reliably model highly non-uniform states of magnetization, such as vortices. The small mesh size was also desirable for modeling the 1 nm thin spacer, which was very computer intensive. In order to have reasonable simulation times, for non-critical tasks the spacer was taken to be slightly thicker, which was verified to not affect the results in any significant way.

Most of the simulations were performed without taking into account the influence of temperature on the magnetization behavior. However, under certain conditions thermal agitation can play a significant role in the magnetization reversal process (Paper 5). In such cases, the micromagnetic simulations were performed with thermal fluctuations included. The time step for the numerical solver is an important parameter and determines the accuracy of the simulation such that a smaller time step gives a more accurate result but requires more time to complete the simulation, while increasing the time step introduces errors into the solution. The time step used was kept as small as possible in order to maintain the angle between the neighboring spins in the range of 30 degrees. This gives reasonable results [45] and keeps the simulations time manageable. Both room and zero temperatures were modeled using the same numerical solver and the same time step, in order to have comparable results. As was shown in Ref [132], the size of the elementary cell can play a significant role specifically when thermal agitation is included. Therefore, in our modeling of the effect of temperature the mesh size was kept as small as possible, 2.5 nm being a reasonable compromise.

6.4 Data processing

The experimental and the micromagnetic data were processed in a similar way in order to compare the results. In order to avoid confusion between the two, the micromagnetic data is presented in the normalized units of $\langle M_x/M_s \rangle$ and the experimental data as measured in $\Omega$.

The data measured quasistatically did not require any additional processing, as opposed to the data obtained during dynamic measurements. The data acquired in real-time dynamic measurements reflect the resistance response of the junction versus time. The power spectrum density spectrograms were computed by using Hann filtering of the recorded signal followed by FFT. A similar procedure was applied to the data obtained by micromagnetic modeling the dynamic, time-dependent magnetization response. The details of the post-processing used during the real-time measurements are given in [133].

The experimental data obtained using microwave spectroscopy did not require any processing. On the other hand, the spectroscopic micromagnetic data required a careful smoothing procedure. Good results were obtained by averaging one full period of the oscillations. The micromagnetic data obtained were in some cases scaled to the experimental data to better visualize the comparison.

In order to trace the motion of small magnetic irregularities such as spin vortex cores, the time dependence of their position had to be calculated with special pre-
cision. A weighted average of the $M_z$ component scaled against a chosen threshold was used to define the position of the vortex core. Thereafter the vortex core path was traced based on the magnetization vectors stored with a 5 ps time interval.
Chapter 7

Results and Conclusions

This thesis presents a study of the static and dynamic properties of dipole-coupled ferromagnetic bi-layers, which are at the core of the recently developed MRAM. A macrospin model provides a good description of the magnetization dynamics in the AP ground state and in the near saturated state. The resonance frequencies obtained using this model agree well with the experimental and micromagnetic data. A micromagnetic study of the intermediate in field, spin-flop region confirms highly non-uniform states of magnetization, with the magnetization dynamics governed not by scissor-like precession of the two macropins but by non-uniform spin oscillations in C- and S-states of magnetization (Paper 1).

Resonant properties of spin-flop bi-layers in the AP magnetization ground state were experimentally studied by means of real time measurements of the resistance oscillations following a short magnetic field pulse and by means of microwave spectroscopy. It was found that strong in-plane magnetic field excitations can lead to magnetization reversal in the region where no quasistatic switching can occur. The reversal process has a pronounced resonant character – for moderated magnetic field amplitudes switching occurs only near the optical spin-resonance frequency and never outside the optical resonance region. A further increase of the microwave field amplitude leads to a broadening of the switching region due to excitation of strong micromagnetic modes (spin waves). The experimental results and the results of the micromagnetic simulations confirm the fact that thermal agitation plays a significant role in the magnetization reversal (Papers 2 and 5).

The AP ground state of the magnetization is very stable in dipole coupled bi-layers. However, it was found that a strong alternating magnetic field applied at the frequency close to the optical spin resonance leads to a creation of vortex states of magnetization in both top and bottom magnetic layers, having intermediate resistance, between the high- and low-resistance states of the MTJ. These vortex states are stable states of magnetization and withstand ~100 Oe in-plane magnetic fields. The quasistatic and dynamic properties of the observed vortex-pair states are quite different from those of the uniform magnetization states and are governed
by the relative orientation of the vortex core polarization and the vortex chirality. These results are presented in Paper 3.

A detailed experimental and micromagnetic investigation of the vortex-pair dynamics shows that the coupling between the vortex cores in a spin-flop bi-layer significantly changes the magnetic response of the system to external fields (Paper 4). The core-core coupling-decoupling process significantly alters the resonance frequencies of the vortex cores and changes the trajectory of the core motion. The presented micromagnetic analysis explains this dynamic behavior and allows to identify three main modes of the core motion: a low-frequency gyrational mode corresponding to decoupled vortex cores, moving independently about their equilibrium positions in circular trajectories; a rotational mode corresponding to strongly coupled vortex cores rotating about their magnetic center of mass; and a high frequency vibrational double, corresponding to oscillations of the vortex cores with very high frequency about the lower-frequency gyrotrropic or rotational trajectories.

Two measurement instruments have been built: a scanning tunneling microscope, specially designed for high-current MR measurements and a current-in-plane tunneling instrument, for characterizing unpatterned MTJ multilayers. Both instruments were designed to characterize unprocessed (CIPT) or minimally processed (STM) magnetic multilayer films in terms of the quality of the TJ barrier, the magnetoresistance, and the resistance area product. The CIPT instrument works in a fully automatic regime, and the STM allows to perform scanning the surface of lithographically patterned samples and manually choose the place for performing I/V or MR measurements.

In conclusion, the thesis has a two-fold focus. Two novel instruments are designed and built for characterizing nanostructures in terms of surface morphology and magneto-transport. These STM and CIPT instruments provide efficient tools in developing new magnetic multilayered materials. The second focus is the magnetization dynamics in spin-flop bi-layers. A comprehensive study, including measurements, analytics, and micromagnetics, of both the spin-uniform anti-parallel state and the vortex-pair state of the system is presented. New effects of resonant activation and vortex core-core dynamics are described. The results can prove useful for developing new and improved magnetic memory devices.
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Chapter 8

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Chapter 9

Appended papers

9.1 List of papers

   **My contribution:** Conducted microwave spectroscopy measurements, took part in the preparation of the manuscript.

   **My contribution:** Conducted measurements and micromagnetic modelling, wrote the manuscript.

   **My contribution:** Conducted measurements and the micromagnetic modelling, wrote the manuscript.

4. S. S. Cherepov, V. Korenivski, D. C. Worledge, A. Yu. Galkin, B. A. Ivanov, Core-core dynamics in spin vortex pairs, manuscript
   **My contribution:** Conducted the measurements, analysed the data and prepared the manuscript.

5. S. S. Cherepov, V. Korenivski and D. C. Worledge, Resonant activation of a synthetic antiferromagnet, manuscript
   **My contribution:** Conducted the measurements and micromagnetic modelling, analysed the data, took part in preparing the manuscript.
9.2 List of papers not included in this thesis

  
  **My contribution:** Performed a subset of the measurements, participated in the discussions on the manuscript.