MASTER

The nanostencil process
nanostructuring sub-micron spin-torque devices

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Nanostructuring sub-micron spin-torque devices

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Abstract

The prediction of a completely new physical effect, called Spin-Transfer-Torque (STT), whereby the magnetization can be manipulated using a current of electrons, attracted a lot of research attention and is now truly a hot topic within the field of spintronics.

In this thesis we are aiming at the creation of a device exploiting the STT effect. However, before the results on the creation of these devices are presented, this thesis will provide a formal derivation of spin-transfer-torque, deriving the critical features needed in the devices. For the actual fabrication of STT devices we adapted the so-called nanostencil process, where a predefined nanometer sized stencil is created and, hereafter, the device structure is deposited into.

We have been successful in the fabrication of the desired nanostencils and we succeeded in depositing, imaging and measuring structures grown inside the nanostencil. For the creation of the nanostencils, we have used both UV and E-beam Lithography, as well as Focussed Ion Beam milling to create the nanostencil opening. The lateral dimension of our nanostencils are typically on the order of 200 nm and the sputtered STT structures consist of a Co/Cu/Co trilayer, only a few nanometers thick. Moreover, a quantitative numerical model describing the growth inside the nanostencil has been developed. Finally, we were able to perform a first preliminary transport measurement on a fully integrated nanostencil STT device. Further optimization of the growth and shape of the device structure grown inside the nanostencil is currently in progress.
# Contents

1 General Introduction .................................................. 2  
1.1 The exploding digital universe ................................. 2  
1.2 From spins to spintronics ........................................ 3  
1.3 Motivation for this work ......................................... 5  
1.4 Guide to this thesis .............................................. 6  

2 Spin transfer torque: A theoretical description .............. 8  
2.1 What is magnetoresistance ...................................... 8  
2.2 Giant magnetoresistance ........................................ 9  
  2.2.1 Charge transport in magnetic heterostructures .......... 9  
  2.2.2 Calculating the giant magnetoresistance effect ........ 13  
  2.2.3 Geometries for the giant magnetoresistance effect .... 16  
2.3 Spin transfer torque ............................................... 19  
  2.3.1 Spin filters and spin polarizers ............................ 20  
  2.3.2 Spin accumulation in the non-magnetic layer .......... 21  
  2.3.3 Definition of spin current density and spin transfer torque 26  
  2.3.4 Spin torque as a result of spin filtering ................. 27  
  2.3.5 Spin torque near the interface ............................. 30  
  2.3.6 Current-induced switching .................................. 34  

3 Spin transfer torque devices ........................................ 37  
3.1 General device geometries ....................................... 37  
3.2 The nanostencil process ......................................... 39  
  3.2.1 What is a nanostencil ....................................... 39  
  3.2.2 Why nanostencils .......................................... 39  
  3.2.3 Recent literature results ................................... 41  
  3.2.4 Critical factors in the nanostencil process ............. 43  

4 An experimental toolbox ................................................ 46  
4.1 Fabrication techniques ........................................... 46  
  4.1.1 Material addition ........................................... 47  
  4.1.2 Pattern generation .......................................... 51  
  4.1.3 Material removal ............................................ 54  
4.2 Characterization and measurement techniques ............... 56  
  4.2.1 Scanning Electron Microscopy (SEM) ....................... 57
Chapter 1

General Introduction

One of the most remarkable advances in technology, in the last decades, is made in the area of digital data storage. Especially the field of magnetic data storage, due to several scientific breakthroughs, has undergone an incredible progress to store more and more information on an ever decreasing area. Recent development of spin-transfer-torque (STT) based devices has opened an active new area for the exploration of magnetic nanostructures. Although this research is strongly technologically driven, it is also of great scientific interest. In this thesis we will determine what spin-transfer-torque is and how can we design devices exploiting this new effect. This introductory chapter will first present the relevance for society for the research performed in this thesis in section 1.1. Then an overview of the birth of spintronics is given in section 1.2. Hereafter, section 1.3 elaborates on the motivation of this work and finally, a guide to this thesis is presented in section 1.4.

1.1 The exploding digital universe

Our modern day society seems to have an insatiable hunger for creating, manipulating and storing more and more (digital) information. Nowadays, digital information surrounds us everywhere. We see digital bits on our big-screen HDTVs, listen to them on our Ipods and create new ones every time we take a picture with our digital cameras. Advances in technology, over the last decades, made this increasing amount of digital information possible. Especially the field of magnetic data storage has undergone an incredible progress to store more and more information on an ever decreasing area.

It has been only half a century ago that IBM introduced the 305 RAMAC as the first commercial computer that used a magnetic hard disk drive to store its information. It existed of fifty 24 inch disks, each containing 5 MB of data and the whole system could be housed in a room of about 9 by 15 m. To put
things into perspective, Googles clusters nowadays process more than twenty petabytes of data per day \[1\]. To store this amount of data on RAMAC systems would require a surface of 540 000 km$^2$, roughly the size of a country like Spain.

In 2006, the amount of digital information created, captured, and replicated was estimated to be on the order of 160 billion gigabytes. This is about 3 million times the information in all the books ever written \[2\]. By 2011, the digital universe will be 10 times the size it was in 2006 \[3\].

To deal with this explosion of our digital universe, new technologies are needed to provide enough digital storage capacity. Also, these new technologies need to be faster, cheaper, more energy efficient and easy to make. In the next sections, we will introduce the scientific breakthroughs enabling the incredible increase in magnetic storage capacity and present new technologies, possibly enabling future increasing storage capacity as well as other future devices. Although both strongly technologically driven, they are also of great scientific interest.

### 1.2 From spins to spintronics

Until recently, the charge of an electron and its spin were considered separately and in traditional electronics, only the electrons charge is used to carry the information. However, this picture started to change in 1988, when Peter Grünberg and Albert Fert discovered the Giant Magneto Resistance (GMR) effect and opened the way to efficient control of charge transport through the spin of the electron.

An elaborate discussion of the theory behind the GMR will be presented in chapter 2, but now we will briefly summarize the effect. The GMR effect is exploited in a structure consisting out of two ferromagnetic layers, separated by a non-magnetic spacer layer. The device is constructed in such a way that the magnetization of the ferromagnetic layers are oriented either parallel or antiparallel. The electrical resistance of the device will depend on the magnetic configuration, with a low electrical resistance if the magnetizations are parallel aligned and a high resistance in the anti-parallel configuration. As will be described in section 2.2, it turns out that the orientation of the spin of the electron actually causes this effect.

Nowadays, we could say the GMR effect is, most successfully, exploited commercially by manufacturers of hard disk drives and magnetic sensors. By incorporating GMR sensors in the read heads of harddisks, the manufacturers were able to increase the exponential growth rate of the areal storage density, as shown in figure 1.1.

This figure shows the exponential increase in areal data density, starting from the introduction of the RAMAC in 1956. The areal density of the RAMAC
is approximately 2000 bits/in². Nowadays, in a laboratory environment, TDK researchers have achieved an areal density as high as 800 Gb/in². This is an increase of a factor 400 million times over the last 52 years. Again, to put things into perspective, 1 Tbit/in² is an areal density obtained when using an individual bit size of only $20 \times 30 \text{ nm}^2$.

Besides the great technological impact, the discovery of the GMR effect has led to a whole new research field. In this field, researchers try to incorporate the electron spin into (new) electronic devices and therefore this field is called spintronics. For the discovery of GMR and the birth of spintronics, Fert and Grünberg were rewarded with the Nobel prize in 2007.

Readers interested in a more detailed description of the discovery and impact of spintronics are referred to the following articles. An excellent review describing the journey to the Nobel Prize can be found in [6]. Another highly recommended paper is Fert’s Nobel lecture, entitled: Origin, Development, and Future of Spintronics [7]. For a review regarding spintronics and the emergence of spin electronics in data storage, please refer to [8, 9].
1.3 Motivation for this work

Until recently, external applied magnetic fields are used to manipulate the magnetization in spintronic devices. However, in 1996 Slonczewski [10] and Berger [11] independently predicted that it could be possible to alter the magnetization by applying a current through a nanometer sized device. The prediction of this completely new physical effect, called Spin-Transfer-Torque (STT), attracted a lot of research attention and is now truly a hot topic within the research field of spintronics. Since the STT effect covers completely new physics, thorough theoretical studies are being performed on this subject. However, the STT effect also creates new possibilities for future spintronic devices and it is therefore being studied from an experimental point of view as well.

One of the devices which can be created using the STT effect is a new type of Random Access Memory. STT-RAM is a next-generation, non-volatile memory solution that overcomes the limitations of conventional technologies. While existing memory technologies become more difficult to produce and operate with decreasing size, STT-RAM shows excellent scalability with shrinking design rules. This translates to greater density and, ultimately, lower cost. STT-RAM also consumes less power than existing mainstream memories, and

Typically STT devices are 50 – 100 nm in height and have lateral dimensions on the order of 100 – 300 nm.
provides unlimited endurance as well as fast read/write capability.

Another future spintronic device, relying on the STT effect, is the so-called Racetrack Memory, as proposed by Parkin [12]. This is a type of memory were the bits, characterized by different magnetic domains, are shifted using appropriate current pulses. Recently the controlled motion of a series of domains along magnetic nanowires is reported [13].

The motivation to devote a master thesis project on the research of STT is the fact that the STT effect is of both technological as well as scientific interest. Within the Physics of Nanostructures group (FNA), where the research presented in this thesis is performed, there is a treasure of knowledge and experience in the field of spintronics. New research is aimed towards the fabrication and understanding of nanostructured (magnetic) devices. Therefore, the aim of this project is not only understand the underlying physics, but also design a process to create STT devices.

Nowadays, researchers are assiduously attempting to create STT devices using different fabrication processes. In this thesis we will use one of those processes, namely the so-called nanostencil process. A schematic representation of a STT device created using this process is shown in figure 1.2. This method uses a predefined stencil to create the STT device structure into and the exact geometry of this structure, as shown in [12], as well as the reason for the different layers involved, will be explained further on in this thesis. The main reason for the choice of the nanostencil process are the fact that it is a versatile and scalable process, allowing easy alteration of the exact STT device structure. Besides these properties, does the nanostencil process provide us the opportunity to explore the usage of the recently acquired Focused Ion Beam setup as a nanostructuring tool.

1.4 Guide to this thesis

In the previous sections we saw an example of the impact of the discovery of GMR and the subsequent birth of the new research field of spintronics. Likewise we mentioned that one of the new, promising, discoveries succeeded from the spintronics research is the spin-transfer-torque effect. Accordingly, the question one would like to answer is:

What is spin-transfer-torque and how can we design a device exploiting and thereby analyzing this new effect?

To answer this question, this thesis is basically made up out of two parts. In the first part we will try to explain the STT effect. This is done both in a conceptional manner as using a formal approach. This formal derivation is deliberately presented in all its details, because this master thesis work was the
first research performed on STT within the Physics of Nanostructures group. The second part of this thesis deals with the design and fabrication of the STT device and this part will mainly present our obtained experimental results. We will now provide the reader with an overview of the different chapters presented in this thesis.

- **Chapter 2**: *Spin transfer torque: A theoretical description*. In this chapter we will first discuss the GMR effect, where we will introduce a number of essential parameters for explaining the STT effect. Then, the STT effect is first explained in a conceptional manner and thereafter using a formal approach.

- **Chapter 3**: *Spin transfer torque devices*. In this chapter we will discuss our choice for using the nanostencil process to create STT devices and summarize the critical factors in the nanostencil process.

- **Chapter 4**: *An experimental toolbox*. The techniques used to fabricate and characterize our nanostencils and STT devices are presented in this chapter.

- **Chapter 5**: *Creating a nanostencil*. This chapter concentrates on the actual creation of a nanostencil. Therefore material choices and their growth conditions are discussed first and thereafter the results leading to the fabrication of our nanostencils are presented.

- **Chapter 6**: *Growth inside a nanostencil*. The growth inside a nanostencil, after it has been created, will be discussed in this chapter. We will first show the results of structures deposited inside the nanostencil and then present a model describing these depositions.

- **Chapter 7**: *Transport measurements*. This chapter will present the first preliminary transport measurements on STT devices created inside the nanostencil. However, before growing devices inside a nanostencil, we investigate non-patterned devices. We will show interesting GMR measurements and discuss their implications for future STT devices.

- **Chapter 8**: *Conclusions and Outlook*. The final chapter of this thesis will summarize the obtained results, showing that our main research question is answered satisfiable. This chapter will also present a brief outlook for future research on nanostencil based STT devices.

The reader only interested in the experimental results, is suggested to continue reading at chapter 5. However, to fully appreciate how these results are presented, a brief look at section 3.2 may be useful.
Chapter 2

Spin transfer torque: A theoretical description

In this chapter we will first discuss the introduction of magnetoresistance in section 2.1. Thereafter, in section 2.2, the transport of electrons in magnetic heterostructures is described to explain the giant magnetoresistance effect. The concepts introduced in this section are needed to explain the spin-transfer-torque effect in section 2.3. First, the STT effect is explained in a conceptional manner and thereafter using a formal approach. For an extensive review regarding almost all aspects of STT, the reader is referred to [14].

2.1 What is magnetoresistance

Magnetoresistance (MR) is used to refer to all effects where the resistance of a material, or combination of materials, changes in response to an external applied magnetic field. The first observations of magnetoresistance date from more than 150 years ago by Lord Kelvin [15]. Kelvin demonstrated a dependence of the resistance on the angle between the direction of electrical current and the applied magnetic field for nickel and iron. This effect, now called Anisotropic Magnetoresistance (AMR), is attributed to a larger scattering probability in the direction of magnetic field and has, in most ferromagnetic materials, a maximum value when the direction of current is parallel to the magnetic field.

Despite the fact that AMR had several technological applications, the impact on (the scientific) society was not quite as big as the discovery of Giant Magnetoresistance (GMR) in 1988 by Grünberg [16] and Fert [17]. Where the AMR effect is on the order of a few percent, the GMR effect can be on the order
of 10 – 100% depending on the exact geometry and material system. Nowadays, we could say the GMR effect is, most successfully, exploited commercially by manufacturers of hard disk drives and magnetic sensors.

Another type of magnetoresistance is Tunneling MagnetoResistance (TMR), which received a lot of attention recently. The effect is based on spin dependent tunneling between two ferromagnetic layers as proposed in 1975, but only in the last decade been proved experimentally. The TMR effect can be up to 600% at room temperature, using special material combinations and fabrication processes.

Although the mechanisms responsible for AMR, GMR and TMR are different, they are all derived from the interaction of the current carrying electrons and magnetism. Because the GMR effect will be used in the devices described in this thesis and some concepts are needed for the understanding of spin transfer torque, a more detailed theoretical description will be presented in the next section. Therefore we will start the theoretical description of GMR by discussing the transport of electrons through magnetic and nonmagnetic materials.

2.2 Giant magnetoresistance

2.2.1 Charge transport in magnetic heterostructures

To derive relations describing the charge transport in magnetic and nonmagnetic materials we first need to know how the current carrying electrons can be described inside the material. In the case of a ferromagnetic material we are especially interested in the interaction between the electrons and the magnetism. To do this, we introduce the spin polarized density-of-states (DOS).

In a first approximation, we can split the density-of-states into two parts, one for the conduction s-electrons, as shown in figure 2.1(a), and one for the bound d-electrons, as shown in figure 2.1(b). We will use the Stoner model approach, which states that the DOS, for the d-electrons in a ferromagnetic material, is nearly identical for the majority and minority electrons, but the states are shifted in energy with respect to each other. This is due to exchange interactions between the bound d-electrons, by which their energies are shifted by the exchange energy $E_{ex}$. This implies that the DOS at the Fermi energy $E_F$ becomes different for the two spin sub-bands, and therefore is spin polarized.

In the rest of this thesis, majority electrons (denoted with the symbol $\uparrow$) are defined as those having their spin parallel to the magnetization of the magnetic material and minority electrons (denoted by $\downarrow$) as those having their spin anti-parallel.

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1 This will be explained in the next section.
2 For a complete review of spin dependent tunneling please refer to e.g. [18].
2.2 Giant magnetoresistance

(a) s-band (b) d-band

(c) fcc Co (d) fcc Cu

Figure 2.1: Spin dependent density of states (DOS). At the Fermi level, in a ferromagnetic material, the density of states for the d-electrons is different for the spin up and spin down electrons. In this simplified picture the conduction s-electrons (a) and the bound d-electrons (b) are treated as two independent bands. The calculated total DOS \[ \text{total DOS} \] for fcc Co (b) and fcc Cu (c) show a much more complicated structure.

The conducting electrons at the Fermi energy are those responsible for electric transport. The calculated total DOS for Co is shown in figure 2.1(c) and shows that the conduction electrons are spin polarized, and a charge current is accompanied by a current of spins. For a normal non-magnetic metal as Cu, the density of states at the Fermi level is equal for both spin directions, as shown in figure 2.1(d), and therefore the conduction electrons are, in principle, not spin polarized. However, later we will show that we are able to inject a current of spin polarized electrons into a normal non-magnetic metal like Cu.

We are now going to treat the conduction electrons, in a ferromagnetic material, as being made up out of two independent channels. This co-called two current model, as first introduced by Mott [24, 25], will be used to describe the spin dependent electrical properties of the ferromagnet. The total conductance \( \sigma \) of ferromagnetic materials can, in this model, be described as the sum of the spin up \( \sigma^\uparrow \) and spin down conductance \( \sigma^\downarrow \):

\[
\sigma = \sigma^\uparrow + \sigma^\downarrow.
\]  

The two current model is a fair approximation for ferromagnets based on Co, Fe or Ni at temperatures well below the Curie temperature. Using a free electron approximation, the spin dependent conductivities are given by the Drude formula [23]:

\[
\sigma^\uparrow(I) = \frac{e^2}{m^*v_F} n^\uparrow(I) \lambda^\uparrow(I) = \frac{e^2}{m^*} n^\uparrow(I) \tau^\uparrow(I),
\]

where \( e \) is the electron charge, \( m^* \) the effective electron mass, \( v_F \) the Fermi velocity, \( n \) the electron density, \( \lambda \) the electron mean free path, and \( \tau \) the electron...
momentum relaxation time which are related by:

$$\lambda_{\uparrow}(\downarrow) = v_F \tau_{\uparrow}(\downarrow). \quad (2.3)$$

Although the Drude model is usually expressed in terms of conductivity, in most cases it is more clear to think in terms of the resistivity. Therefore we introduce the spin dependent resistivity $\rho$, which can be written as:

$$\rho_{\uparrow}(\downarrow) = \frac{1}{\sigma_{\uparrow}(\downarrow)}. \quad (2.4)$$

The Drude formula explains the resistivity only in the case where solely s-bands are crossing the Fermi level, as shown in figure 2.1(a). However in ferromagnetic materials, there are also d-band states at the Fermi level. Due to this overlap of the s and d wavefunctions, the conduction s-electrons can scatter into the available bound d-electrons at the Fermi level. This process is called s-d hybridization [26]. The timescale which governs this process is the relaxation time and, using Fermi’s golden rule, the spin dependent relaxation time can be written as [23]:

$$\tau_{\uparrow}(\downarrow) = \frac{1}{|V_{\uparrow}(\downarrow)|^2 N_{\uparrow}(\downarrow)(E_F)}, \quad (2.5)$$

where $V_{\uparrow}(\downarrow)$ is the spin dependent scattering potential and $N_{\uparrow}(\downarrow)(E_F)$ the spin dependent total DOS at the Fermi level. If we now look at the calculated density of states for ferromagnetic Co and non magnetic Cu, figure 2.1(c) and (d), we can conclude that, because of the difference in DOS at the Fermi level, there will be different relaxation times for spin-up and spin-down in Co whereas there is no difference for Cu. In the case of Co, more scattering into d-bands will take place for the minority electrons, lowering their relaxation time and, using equation 2.2 and 2.4, increasing the resistivity. Besides the differences in the DOS at the Fermi level, a difference in the scattering potential also causes spin dependent resistivity.

We now have shown that the current through a ferromagnetic material is carried by the two independent spin channels, both having their own conductivities given by equation 2.2. But this approximation is only valid when the spin channels are truly independent. However, in a real material there is the possibility that a certain spin state scatters into the other. This is schematically shown in figure 2.2(a) where a spin-down electron flips its spin state after a certain amount of scattering events. The average distance the electron travels in the ferromagnetic material between two spin flip events is called the spin mean free path $\lambda_{sf}^F$.

Figure 2.2(a) also shows the electron momentum free paths for spin-up $\lambda_{F}^\uparrow$ and spin-down $\lambda_{F}^\downarrow$ electrons inside a ferromagnetic material. In a normal
2.2 Giant magnetoresistance

Figure 2.2: Spin dependent transport. (a) The electron mean free path inside a ferromagnet is different for spin-up $\lambda^F_\uparrow$ and spin-down $\lambda^F_\downarrow$ electrons. The average distance between two spin flip events is given by the spin mean free path length $\lambda^F_{sf}$. (b) The electron mean free path $\lambda^N$ for spin up and spin down electrons are the same in a normal metal. The interface between the two layers causes an interfacial spin dependent resistance $r$.

nonmagnetic material, there is no difference for spin-up or spin-down states and the electron momentum mean free path is given by $\lambda^N$, as shown in figure 2.2(b). Inside a normal metal, however, the spin can still flip and the spin mean free path is given by $\lambda^N_{sf}$. We now introduce another length scale, namely the spin diffusion length, which in a normal metal is given by [27, 28]:

$$l^N_{sf} = \sqrt{\frac{\lambda^N \lambda^N_{sf}}{6}}$$  \hspace{1cm} (2.6)

and in a ferromagnet reads:

$$l^F_{sf} = \sqrt{\frac{\lambda^F_\uparrow \lambda^F_\downarrow \lambda^F_{sf}}{3(\lambda^F_\uparrow + \lambda^F_\downarrow)}}.$$  \hspace{1cm} (2.7)

Finally, we want to introduce one more scattering event. Besides scattering in the bulk of the material, an electron can also scatter at the interface between two different materials as shown in figure 2.2(b). In the case of a perfect interface between two materials electrons still can scatter at the interface [29]. This so-called specular interface scattering occurs when the potentials inside the two materials are different. However, in reality there are no perfect interfaces, and interface scattering also results from defects at the interface, interfacial roughness or interdiffused atoms. This type of scattering is called diffuse scattering. The interface resistance $r$, as caused by interface scattering, can now be different for the majority and minority electrons and can be derived from the spin electronic structure matching [30].

We have derived that the resistivities inside a ferromagnetic material are different for the spin-up and spin-down electrons. Furthermore, the interface resistance between a ferromagnet and a normal metal is different for the two
2.2 Giant magnetoresistance

Figure 2.3: Spin dependent transport through a magnetic heterostructure. Two magnetic layers separated by a non-magnetic spacer where the magnetizations of the layers are (a) parallel or (b) antiparallel aligned. In the parallel alignment, the spin-down electrons experience a higher resistance $R_H$ in both layers whereas the spin-up electrons experience a lower resistance $R_L$. In the antiparallel alignment, both spin directions have a higher resistance in one of the two layers. The bottom panels show the equivalent resistor networks.

spin states. These two effects turn out to be the origin of the GMR effect, which will be explained further in the next section.

2.2.2 Calculating the giant magnetoresistance effect

Figure 2.2 shows that there is a differences in the electron mean free path, and thereby resistivity (through equation 2.2), for spin-up and spin-down electrons in a ferromagnetic material. However, there is no difference in mean free path in a non-magnetic metal. If we now construct a device consisting of two magnetic layers, whose magnetization direction we can manipulate independently and separate the two layers by a non-magnetic spacer, we will show in the following that the resistance depends on the relative orientation of the magnetization.

To describe the spin dependent resistivities, we now introduce the bulk spin asymmetry coefficient $\beta$ and the bulk ferromagnet resistivity $\rho_F$. The spin asymmetry coefficient, which magnitude is $0 \leq \beta \leq 1$, is a dimensionless parameter related to the polarization of the current. In the case of a short mean free path, we introduce the high resistivity $\rho_H$ as:

$$\rho_H = \frac{2\rho_F}{1 - \beta}. \quad (2.8)$$

In the original paper by Valet and Fert [27], the bulk ferromagnet resistivity $\rho_F^*$ is introduced. The two are related by $\beta$ as: $\rho_F = \rho_F^* (1 + \beta) (1 - \beta)$. 

2.2 Giant magnetoresistance

In the case of a long mean free path, the low resistivity $\rho_L$ can accordingly be described as:

$$\rho_L = \frac{2\rho_F}{1 + \beta}. \quad (2.9)$$

If the current is completely polarized, $\beta = 1$, one spin channel has an infinite high resistivity and the complete current will flow through the other channel. However, for a non-polarized current, as is the case in bulk non-magnetic metals, $\beta = 0$ and the current will be equally divided over the two spin channels. Until now we discussed resistivities, the resistance $R$ is related to the resistivity $\rho$ as:

$$R = \frac{\rho l}{A}, \quad (2.10)$$

where $l$ is the length of the layer and $A$ the cross-sectional area.

We now introduce a device consisting of two magnetic layers, whose magnetization direction we can manipulate independently and separate the two layers by a non-magnetic spacer, as shown in the top panels of figures 2.3(a) and (b). In the case of a parallel alignment of the two magnetic layers, the spin-down electrons experience a high resistance $R_H$ in both ferromagnetic layers whereas the spin up electrons experience a low resistance $R_L$, due to their spin dependent mean free paths. The bottom panels of 2.3(a) and (b) show the equivalent resistor networks\(^4\), where the parallel resistance $R_P$ can be described as a summation of the spin-up and spin-down channel:

$$\frac{1}{R_P} = \frac{1}{2R_L} + \frac{1}{2R_H} = \frac{R_L + R_H}{2R_LR_H}. \quad (2.11)$$

In the antiparallel alignment, both spin directions undergo a higher resistance in one of the two layers, and the antiparallel resistance $R_{AP}$ can be described as:

$$\frac{1}{R_{AP}} = \frac{1}{R_L + R_H} + \frac{1}{R_H + R_L} = \frac{2}{R_L + R_H}, \quad (2.12)$$

which always\(^5\) results in a higher total resistance for the whole device compared to the parallel case. We have now indeed shown that the resistance of the two ferromagnetic layers depends on their relative magnetic orientation and this exceptional effect is called the giant magnetoresistance effect. By a relatively simple resistance measurement we are now able to describe the magnetic state of our device. To determine the magnitude of the GMR effect, we calculate the relative change in resistance between the parallel and antiparallel alignment as:

$$GMR = \frac{\Delta R}{R_P} = \frac{(R_L - R_H)^2}{4R_LR_H} = \frac{\beta^2}{1 - \beta^2}. \quad (2.13)$$

\(^4\)Where, for now, the resistance of the non-magnetic spacer layer is neglected and there is no resistance associated with the interface between the layers. These two effects will taken into account for in the next part of this section.

\(^5\)As long as $R_H \neq R_L$. 
2.2 Giant magnetoresistance

Figure 2.4: Magnetic moment and resistance curves as a function of the applied magnetic field. Figure (a) shows the magnetization of a spin-valve, whereas figure (b) shows the resistance and the related MR ratio. The top and bottom horizontal arrows indicate the magnetization directions of the fixed and free layer, respectively.

where $\Delta R = R_{AP} - R_P$. For a typical ferromagnetic material as Co, $\beta$ is 0.46 [31] and this corresponds to a (theoretical) GMR ratio of 27%. From equation 2.13 we can conclude that a higher value of $\beta$ will, in theory, result in a higher GMR effect. Moreover, when using a material with a spin asymmetry coefficient $\beta \approx 1$, the GMR ratio would be almost infinite. These materials are called half-metals and the search for these materials is still a big issue in the field of modern magnetism [32].

So far, we deduced that a GMR structure has two different states corresponding to a high and low resistance, but we have not showed how this resistance changes as the magnetization of the layers changes. Therefore we will observe the resistance of a GMR device where the magnetization of one of the two layers is fixed. Now, by applying an external field, we will alter the magnetization of the other, free, magnetic layer.

We will now show the general shape of a magnetoresistance curve, as is shown in figure 2.4(b). This graph is collected by measuring the resistance of the GMR structure, while sweeping the applied field from positive to negative fields and backwards. The corresponding normalized magnetic moment of the structure is shown in figure 2.4(a). We will now go through this loop, explaining the exact shape.

The top and bottom horizontal arrows indicate the magnetization direction of the fixed and free magnetic layer respectively. If we start with a large positive applied magnetic field the total magnetization will be positive. Since the magnetization of both layers are parallel aligned, the resistance will be low. When decreasing the magnetic field, the bottom free layer will switch
its magnetization at a certain negative applied field. Now, the magnetization direction of the two magnetic layers are antiparallel aligned, resulting in zero magnetization and a higher resistance of the device, as shown in figure 2.4.

When increasing the applied field again, the magnetization of the free layer will switch back to its original state, while the magnetization of the fixed layer remained unaffected. Now, the magnetization of both layers are parallel aligned again and the resistance will be low.

### 2.2.3 Geometries for the giant magnetoresistance effect

Now we have shown that the origin of the GMR effect lies in spin dependent scattering and derived an expression describing the magnitude of the effect, let us discuss how to experimentally exploit the effect. Therefore we have to construct a device consisting of two magnetic layers separated by a non-magnetic metallic spacer. Such a device is often called a spin valve. Figure 2.5 shows two general spin valve geometries. In the left picture the net Current direction is In the Plane of the layers (CIP) and it is important that the mean free path of the electrons as given by equation 2.3, is longer than the sum of the layer thicknesses so that the electrons effectively sample the different layers. In figure 2.5(b), the net Current direction is Perpendicular to the Plane of the layers (CPP) and the critical length scale becomes the spin-diffusion length, as described by equation 2.6 and 2.7.

We will now continue this section by

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6 Generally, the GMR effect can be exploited in two different ways. In the first manner, as in the original experiments of Grünberg [16] and Fert [17], antiferromagnetically coupled layers are used. In the second manner, the giant magnetoresistance effect is used in the so called spin valve geometry. In this geometry, two ferromagnetic layers are separated by a non-magnetic metallic spacer layer, thick enough to decouple the magnetizations of the two layers. This will be the geometry used in this thesis. For more information about antiferromagnetically coupled GMR devices, please refer to [33].

7 For at least one spin direction.
2.2 Giant magnetoresistance

introducing two models to describe the two different geometries. For the CIP geometry we will discuss the Camley-Barnas (CB) model, and for the CPP geometry the Valet-Fert (VF) model will be presented.

Current In Plane (CIP) GMR

A theoretical description of the CIP GMR, using a semi-classical Boltzmann transport model, is proposed by Camley and Barnas [34]. A complete description of the model is out of the scope of this thesis, therefore we will only discuss the key concepts and briefly treat some results.

Within the CB model, electrons are essentially regarded as point like particles (hence the term classical), but certain consequences of quantum mechanics are taking into account (therefore the addition of the term semi). The normal metal and ferromagnetic layers are treated as a free electron gas and Boltzmann equations are used to describe the transport through the layers. The free parameters in the CB model are the spin dependent scattering in the bulk of the layers and the spin dependent scattering at the outer boundaries and interfaces between the layers. Scattering at the interfaces can be either reflection, transmission or diffusive scattering, as described in the last part of section 2.2.1.

From a general analysis of the CB model, we can conclude that, for achieving the highest GMR ratio possible, we need:

- highly spin dependent resistivities in the bulk of the ferromagnetic layers  
  or,
- highly spin dependent interface scattering probabilities,
- ideally thin non-magnet spacer thickness,
- optimal ferromagnetic layer thicknesses and
- optimal growth of the layers.

The first two points have been already addressed in the previous sections, the last three point will be discussed in section 7.1.

Current Perpendicular to Plane (CPP) GMR

The semiclassical theory for the CPP geometry has been developed by Valet and Fert [27, 28]. Using Boltzmann equations, the transport properties of a structure consisting of magnetic and non-magnetic layers is calculated for currents perpendicular to the layers. In their approach, the interfaces are sufficiently separated, whereby quantum interference effects due to reflection from the in-

---

8Such as the quantum mechanic relation between energy and momentum, Fermi-Dirac statistics and quantum mechanic scattering probabilities.
2.2 Giant magnetoresistance

Figure 2.6: Current Perpendicular to the Plane (CPP) GMR. Two magnetic layers separated by a non-magnetic spacer where the magnetizations of the layers are (a) parallel or (b) antiparallel aligned. The bottom panels show the equivalent resistor networks, where there are resistances associated with both the bulk of the layers $R$ as well as the interfaces $r$.

Table 2.1: Material parameters for a Co/Cu system. The table shows the spin dependent resistivity $\rho$, interface resistance $r$, spin diffusion length $l$, the bulk spin asymmetry coefficient $\beta$ and the interfacial spin asymmetry coefficient $\gamma$ for a sputtered Co/Cu system measured at 4.2 K. [35, 31]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Material</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho$</td>
<td>Cu</td>
<td>5</td>
<td>n$\Omega$·m</td>
</tr>
<tr>
<td>$\rho_{\uparrow}$</td>
<td>Co</td>
<td>41</td>
<td>n$\Omega$·m</td>
</tr>
<tr>
<td>$\rho_{\downarrow}$</td>
<td>Co</td>
<td>110</td>
<td>n$\Omega$·m</td>
</tr>
<tr>
<td>$r_{\uparrow}$</td>
<td>Co/Cu</td>
<td>0.12</td>
<td>f$\Omega$·m$^2$</td>
</tr>
<tr>
<td>$r_{\downarrow}$</td>
<td>Co/Cu</td>
<td>0.90</td>
<td>f$\Omega$·m$^2$</td>
</tr>
<tr>
<td>$l_{sf}$</td>
<td>Cu</td>
<td>450</td>
<td>nm</td>
</tr>
<tr>
<td>$l_{sf}$</td>
<td>Co</td>
<td>59</td>
<td>nm</td>
</tr>
<tr>
<td>$\beta$</td>
<td>Co</td>
<td>0.46</td>
<td>-</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>Co/Cu</td>
<td>0.75</td>
<td>-</td>
</tr>
</tbody>
</table>

Interfaces can be neglected. The theory shows that the total resistance can be broken into contributions from the bulk regions and to resistances associated with each interface, as is shown in figure 2.6.

We already introduced the bulk spin asymmetry coefficient $\beta$ in section 2.2.2. This parameter describes the spin dependence of the bulk resistance of the layers. Accordingly, the interfacial spin asymmetry coefficient $\gamma$ is introduced. This parameter describes the spin dependent interface resistance as:

$$r_{\uparrow(\downarrow)} = \frac{2r_b}{1 + (-)^{\gamma}}.$$  \hfill (2.14)

\[\text{In the original paper by Valet and Fert [27] the interface resistance } r_b \text{ is introduced. The two are related by } \gamma \text{ as: } r_b = r_b^* (1 + \gamma) (1 - \gamma)\]
Using the two spin asymmetry coefficients $\beta$ and $\gamma$, the CPP GMR can be described by the equivalent resistor network shown in the bottom panel of figure 2.6. The exact equations describing the bulk and interface resistances can be found in the original article of Valet and Fert [27], here we only want to briefly mention some typical values. For a Co/Cu system, a material combination also used in this thesis, the typical parameters are given in table 2.1. We can now calculate the bulk $R_b$ and interface $R_i$ resistances of a device consisting of a Co and Cu layer. If the layers have a cross-sectional area of $100 \times 100$ nm$^2$ and a thickness of 10 nm, then, using equations 2.8–2.10 and table 2.1, the bulk resistance for majority electrons in the Co layer is $R^\uparrow_b = 41$ m$\Omega$ and the interface resistance $R^\uparrow_i = 12$ m$\Omega$. For minority electrons, the bulk resistance is $R^\downarrow_b = 110$ m$\Omega$ and the interface resistance $R^\downarrow_i = 90$ m$\Omega$, which are both, as expected, higher than for the majority electrons. For the Cu layer, the bulk resistance is only $R^\uparrow_b = 5$ m$\Omega$. This analysis clearly shows that the interface resistance is of the same order as the bulk resistance in these devices.

2.3 Spin transfer torque

Until now we have used external applied magnetic fields to manipulate the magnetization of the ferromagnetic layers. However, in 1996 Slonczewski [10] and Berger [11] independently predicted that the current flowing through a ferromagnetic heterostructure could influence the magnetization by transferring angular momentum from the electrons to the magnetization. In this section we will derive expressions describing this so called Spin-Transfer-Torque (STT) effect. The STT effect is a purely quantum mechanical effect, which only occurs in nanometer sized magnetic heterostructures. Before we will give a more formal derivation of the spin-transfer-torque, we will start by drawing a qualitative picture for the mechanisms behind it, using some of the spin dependent properties derived in section 2.2.1. This qualitative picture, which will be presented in the next sections, is based on three concepts:

- Interfacial spin-dependent scattering: section 2.3.1
- Ferromagnets as spin polarizers: section 2.3.1
- Spin accumulation: section 2.3.2

Finally, before the concept of spin-transfer-torque is unraveled, we want to clarify two different currents, namely the electron current $\vec{j}$ and the accompanying spin current $\vec{Q}$. The electron current $\vec{j}$ is a vector related to the motion of the charge of the electron, as is shown in figure 2.7(a). Conversely, the spin current $\vec{Q}$ is related to the moving spin. Because both the motion $\vec{v}$ and the spin $\vec{s}$ are vectors, the spin current is described by a tensor. For this tensor, the different spin currents are described by a tensor. For this tensor, the different...
2.3 Spin transfer torque

\begin{center}
(a) \hspace{1cm} \text{(electron) current} \hspace{2cm} (b) \hspace{1cm} \text{spin current}
\end{center}

\begin{align*}
\vec{j} &= j_y \quad & \vec{v} \quad & \vec{s} \\
\vec{Q} &= Q_{yz} \\
\vec{j} &= j_x \quad & \vec{v} \quad & \vec{s} \\
\vec{Q} &= -Q_{xz}
\end{align*}

Figure 2.7: Electron and spin current. Using the \(xyz\)-axis as defined in the picture, the vector (electron) current \(\vec{j}\) is shown in (a) and the tensor spin current \(\vec{Q}\) in (b). In our notation, an electron with its spin in the \(z\)-direction and moving in the \(x\)-direction, causes a current \(j_x\) and an accompanying spin current \(Q_{xz}\). Note that an electron with its spin in the negative \(z\)-direction (spin down) but moving again in the \(x\)-direction, causes a current \(j_x\), however now with an accompanying spin current \(-Q_{xz}\).

columns are related to the direction of motion of the electron, whereas the rows are related the the spin directions. Therefore, in our notation, an electron with its spin in the \(z\)-direction and moving in the \(x\)-direction, causes a current \(j_x\) and an accompanying spin current \(Q_{xz}\). Note that an electron with its spin in the negative \(z\)-direction (spin down) but moving again in the \(x\)-direction, causes a current \(j_x\), however now with an accompanying spin current \(-Q_{xz}\), as can be shown in figure 2.7(b).

2.3.1 Spin filters and spin polarizers

The first two concepts, interfacial spin-dependent scattering and ferromagnets as spin polarizers, are shown in figure 2.8. As a current of electrons scatters at the interface between a normal metal and a ferromagnet, due to interfacial spin-dependent scattering, as described in section 2.2.1, the majority electrons are preferentially transmitted and the minority electrons are preferentially reflected. This causes a spin polarized current to enter the ferromagnet, as is shown in figure 2.8(a). Accordingly, when a spin polarized current is injected from the ferromagnet into the normal metal, as shown in figure 2.8(b), the current maintains its polarization over distance given by the spin diffusion length \(L_{sf}^{N}\), given by equation 2.6. When combining these two concepts, we are now able to introduce the concept of spin accumulation.
2.3 Spin transfer torque

![Figure 2.8: Spin dependent scattering and spin polarization.](image)

(a) As a current of electrons scatter at the interface between a normal metal and a ferromagnet, the majority electrons are preferentially transmitted and the minority electrons are preferentially reflected, which causes a spin polarized current to enter the ferromagnet. (b) Accordingly, when a spin polarized current is injected from the ferromagnet into the normal metal, the current maintains its polarization over distance given by the spin diffusion length $l_{sf}^N$.

2.3.2 Spin accumulation in the non-magnetic layer

A single ferromagnetic layer

When the two structures of figure 2.8 are put together, the structure shown in the top panels of figure 2.9 arises. Here an electron current is passed through a structure consisting of a single ferromagnetic layer sandwiched between two normal metals. Far from the interface, the spin current $Q_s$ as described in section 2.3 and figure 2.7 depends on the spin dependence of the resistivity. In the non-magnetic normal metal there is no spin dependent resistivity and therefore the current is unpolarized, leading to a zero spin current. However, in the ferromagnet the resistivities are spin dependent and there is a finite spin current. This is the case far away from the interface. Now let us look at what happens near the interface between the two materials.

Due to the spin-dependent scattering at the first metal–ferromagnet interface, minority electrons will accumulate in the region close to the interface, while the majority electrons travel freely through the ferromagnet. The spin accumulation $\delta \vec{m}(x)$ is the offset in the local equilibrium spin density, which is a function of the position from the interface. Because of spin flip scattering, the system will reach a steady state and the length scale over which the spin accumulation will fade away is given by the spin diffusion length $l_{sf}^N$:

$$\delta \vec{m}(x) \propto \exp\left(-\frac{x}{l_{sf}^N}\right),$$  \hspace{1cm} (2.15)

where $l_{sf} = l_{sf}^N$ in the normal metal and $l_{sf} = l_{sf}^F$ in the ferromagnet. Since the spin accumulation is a vector in spin-space, a minority accumulation leads, in our notation, to a negative spin accumulation $\delta m_z$ in the $z$-direction as shown

\footnote{At least the spin diffusion length $l_{sf}^N$.}
2.3 Spin transfer torque

Figure 2.9: Spin accumulation resulting from a single ferromagnetic layer. (a) A current is passed through a structure consisting of a single ferromagnetic layer (FM1) sandwiched between two normal metals. In figure (a) the current flows in the positive $x$-direction, whereas in (b) it flows in the negative $x$ direction. Due to the spin-dependent scattering at the first metal–ferromagnet (NM1–FM1) interface, minority electrons will accumulate in the region close to the interface, while more majority electrons travel through the ferromagnet. Therefore, at the second (FM1–NM2) interface, a majority spin accumulation results. The middle panels show the spin accumulation $\delta m_z$ for both current directions. The bottom panels show the resulting spin currents $Q_{xz}$.

Figure 2.10: Spin accumulation resulting from two ferromagnetic layers. A current is passed through a structure consisting of two ferromagnetic layers sandwiched between two normal metals and separated by a thin normal metal layer, with the magnetization of the two layers parallel (a) or antiparallel (b). The middle panel shows that there is almost no spin accumulation in the parallel case, whereas a large majority accumulation arises in the antiparallel case. The resulting spin currents are shown in the bottom panels.
2.3 Spin transfer torque

in figure 2.7. Furthermore, we will assume that the ferromagnetic and spacer layers are much smaller than the spin diffusion lengths. The leads, however, may be longer and therefore a fading spin accumulation, as given by equation 2.15, is visible in the middle panels of figure 2.9.

We showed that the spin accumulation near the first interface is due to accumulation of minority electrons and more majority electrons will travel through the ferromagnetic layer. Therefore, at the second interface, a majority spin accumulation results in the normal metal lead, causing a positive $\delta m_z$. We deduced that there is no spin current far away from the interface. However, because there is a fading spin accumulation in the normal metal near the interface, there must be a resulting spin current. Indeed, it can be shown [36] that the spin current $Q$ in the normal metal can be described by:

$$Q_{\alpha\beta} \propto -\vec{\nabla}_\alpha \cdot \vec{m}_\beta,$$

(2.16)

where, in this case, $\alpha$ is in the $x$ spatial direction and $\beta$ is in the $z$ spin direction. Equation 2.16 states that a spin current in the normal metal is due to a spatially inhomogeneous spin accumulation. When the current is flowing in the positive $x$-direction, this results in a negative spin current as is shown in figure 2.9(a). If the current is in the opposite direction, 2.9(b), a negative spin current arises.

The detailed forms of the spin accumulation and spin currents can be found in appendix A of [36]. Here we only want to mention that the origin of the discontinuity in the spin accumulation lies in the spin dependent interface resistance, whereas the gradient in the spin accumulation is due to the difference in bulk resistivity. On the other hand, the spin current is continuous over the interface because we neglect spin flip scattering resulting from the interface.

**Two collinear ferromagnetic layers**

We now can extend the situation to a structure consisting of two ferromagnetic layer sandwiched between two normal metals and separated by a thin metal layer, as shown in figure 2.10. This structure is similar to the spin valve structure introduced in section 2.2.3. If the magnetizations of the two layers are collinear, the spin accumulation and spin current for this structure can be seen as a superposition of the values of the single layer. When the magnetizations are parallel aligned, figure 2.10(a), there is a large minority accumulation near the first interface, almost no accumulation in the spacer layer and a large majority accumulation near the last interface. Also a large spin current flows through the whole device. However, for an antiparallel alignment, figure 2.10(b), there is almost no spin accumulation in the leads and a large majority spin accumulation in the spacer layer. The spin current through the device is also much smaller then in the case of parallel alignment. This difference in spin current is the origin of the giant magnetoresistance effect, where more current can flow through the lower-resistance majority channel in the case of parallel alignment.
2.3 Spin transfer torque

Figure 2.11: Spin accumulation resulting from two ferromagnetic layers with a non-collinear magnetization. A current is passed through a structure consisting of two ferromagnetic layers sandwiched between two normal metals and separated by a thin metal layer. (a) The current is passed from the left to the right or (b) from the right to the left. The second panel shows the spin accumulation $\delta m_z$ in the $z$-spin direction, whereas there is a spin accumulation $\delta m_x$ in the $x$-spin direction is shown in the fourth panel. The accompanying spin currents are shown in the third ($Q_{xz}$) and fifth ($Q_{xx}$) panel, where the absorption of the transverse spin current is clearly visible. This absorption of spin current leads to the spin-transfer-torque effect, as shown in the bottom panels, where a torque acting on the magnetization rotates it in the direction of the incoming spin.
Two non-collinear ferromagnetic layers

In the subsequent part of this section we are going to use the same structure consisting of two ferromagnetic layers sandwiched between two normal metals and separated by a thin metal layer, but now the magnetizations are non-collinear as is shown in figure 2.11. In this case we can still describe the spin accumulation and spin current in the same way as for the collinear magnetizations, but with an additional assumption, which will be justified in the next sections of this chapter, that the transverse component of the spin current will be absorbed at the interface.

Considering spins moving from left to right, as shown in figure 2.11(a), there will be an accumulation of minority spins near the first interface and a large majority spin current passing through the first ferromagnetic layer into the spacer layer. When this spin current reaches the interface with the second ferromagnetic layer, whose magnetization is transverse, it will be absorbed at the interface. This absorbed spin current leads to a discontinuity of the spin current at the interface, so there must be another mechanism counteracting this effect. This counteraction is taken into account by a torque acting on the magnetization of the ferromagnet, trying to rotate it towards a parallel alignment. This torque, which effectively transfers angular momentum from the current carrying electrons to the magnetization of the ferromagnet, is called the spin-transfer-torque and is schematically shown in the bottom panels of figure 2.11.

The case that the current is flowing in the positive $x$-direction is shown in the bottom panel of figure 2.11(a). Here, majority electrons from the first ferromagnetic layer will effectively be absorbed at the interface with the second ferromagnetic layer, thereby applying a torque in the $z$-direction. In the case of electrons flowing in the negative $x$-direction, there is a minority accumulation $\delta m_z$ in the spacer layer. The absorption of this spin current will result in an opposite torque acting on the second ferromagnetic layer.

Using the concepts of interfacial spin dependent scattering and ferromagnets as polarizers, we have introduced the concept of spin accumulation. By assuming absorption of the transverse spin current at the normal metal–ferromagnet interface, we have successfully explained the concept of spin-transfer-torque in a conceptual manner. Our next objective is presenting a more formal derivation of the STT effect and especially focussing on the physics near the interface.

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12 The electrons are majority with respect to the first ferromagnetic layer and are illustratively shown darker, whereas the minority electrons are shown as lighter circles.
13 Again with respect to the first layer.
2.3 Spin transfer torque

2.3.3 Definition of spin current density and spin transfer torque

In the following sections we will present a mathematical derivation of the spin transfer torque using two different models. A more formal elaborate derivation of the equations in these sections can be found in appendix A. Here we will deliberately omit the intermediate steps and focus on the physical consequences of the equations derived. In the following derivation, we will use the currents and coordinate system as showed in figure 2.7.

In this section we want to describe the equations of spin transport in a quantum mechanical way. Therefore we want to relate the electron and spin currents, as already conceptual introduced in figure 2.7, to the wavefunction of the electron. For a more formal derivation of these quantum mechanic equations, please refer to e.g. [37] or [38].

Considering an electron with wavefunction $\psi$, electron density $n$ is given by:

$$n = \psi^* \psi. \quad (2.17)$$

We can now describe the electron current density $\vec{j}$ as:

$$\vec{j} = \text{Re} \left( \psi^* \hat{v} \psi \right), \quad (2.18)$$

where $\hat{v} = -\frac{\hbar}{m} \vec{\nabla}$ is the velocity operator, which shows an analogy to the classical electron moving with a velocity $\vec{v}$ as shown in figure 2.7(a). Using conservation of electron current, we can write the continuity equation as:

$$\frac{\partial n}{\partial t} + \vec{\nabla} \cdot \vec{j} = 0. \quad (2.19)$$

We are interested in the steady state solution where there are no sources or sinks, $\frac{\partial n}{\partial t} = 0$ and therefore $\vec{\nabla} \cdot \vec{j} = 0$.

Now looking at the spin of the electron, the analog for the electron density, equation 2.18, is the spin density $\vec{m}$:

$$\vec{m} = \psi^* \vec{s} \psi, \quad (2.20)$$

which is a vector in spin space and where $\vec{s} = \frac{\hbar}{2} \vec{\sigma}$ with $\vec{\sigma} = (\sigma_x \sigma_y \sigma_z)$ the Pauli matrices in Cartesian coordinates. Instead of a electron current density, equation 2.18, we now have the spin current density, which has both a direction in spin-space and real space and is therefore given by a tensor $\vec{Q}$:

$$\vec{Q} = \frac{\hbar^2}{2m} \text{Im} \left( \psi^* \vec{\sigma} \otimes \vec{\nabla} \psi \right), \quad (2.21)$$

where $\vec{\sigma} \otimes \vec{\nabla} \psi$ stands for the dyadic product\(^{14}\) of the Pauli matrices\(^{15}\) and the velocity operator\(^{16}\) working on the wavefunction. The analog for the continuity

---

\(^{14}\)Since $\vec{\sigma}$ and $\vec{\nabla} \psi$ are both vectors of the same length, the tensor product in this case called a dyadic product.

\(^{15}\)Connected to the spin part of the tensor.

\(^{16}\)Connected to the spatial part of the tensor.
2.3 Spin transfer torque

Equation 2.19 in the case of the spin current is given by [39]:

\[ \frac{\partial \vec{m}}{\partial t} + \vec{\nabla} \cdot \vec{Q} = -\frac{\delta \vec{m}}{\tau_{sf}} - \vec{n}_{ext}, \]  \hspace{1cm} (2.22)

where \( \delta \vec{m} \) is the spin accumulation, the offset in the local equilibrium spin density as discussed in the previous section, \( \tau_{sf} \) is the spin relaxation time, as described in section 2.2.1, and \( \vec{n}_{ext} \) is the external torque density. This torque density is made up out of the Zeeman interaction, the magnetocrystalline anisotropy and other terms in the Hamiltonian which do not commute with the spin density [39]. In the steady state case, \( \frac{\partial \vec{m}}{\partial t} = 0 \) and where there is no spin flip, \( \tau_{sf} = \infty \), the continuity equation 2.22 states that the net flow of spins into a region plus the torques acting on that region must be zero:

\[ \vec{n}_{ext} = -\vec{\nabla} \cdot \vec{Q}. \] \hspace{1cm} (2.23)

If we only take into account current induced torques, \( \vec{n}_{ext} = \vec{n}_{st} \), and we integrate the continuity equation over a certain volume of ferromagnetic material, using Gauss’ theorem, we find that we can write the spin torque term \( \vec{\tau}_{st} \) as:

\[ \vec{\tau}_{st} = -\int \hat{n} \cdot \vec{Q} d^2 r, \] \hspace{1cm} (2.24)

where \( \hat{n} \) is the unit normal vector to the surface of the volume as shown in figure 2.13(a). Equation 2.24 tells us that if there is a difference in the spin current flowing into and out of a volume, there must be a counteracting torque acting on this volume. Now let us determine this net spin current using two different models.

2.3.4 Spin torque as a result of spin filtering

We conceptually derived the STT effect in section 2.3.2 using the concept of spin accumulation by observing the layered structure as shown in figure 2.12(a). In this section, we will derive the STT in a formal manner, focusing on the second, spin analyzing layer, as shown in figure 2.12(b).

In this section we will derive an expression for the spin transfer torque term \( \vec{\tau}_{st} \) by first considering the net change in the spin current after passing through a layer of ferromagnetic material. This non-equilibrium spin current through the volume of the ferromagnetic layer can be calculated by integrating the spin current density normal to the surface of this volume. By conservation of angular momentum, this non-equilibrium spin current is then compensated by a spin transfer torque of the electrons acting on the magnetization of the ferromagnetic layer. We will use the geometry shown in figure 2.13(b). A spin-polarized current flows from a metallic non-magnet into a metallic, single-domain ferromagnet through an ideal, flat interface. The particle current density vector...
2.3 Spin transfer torque

Figure 2.12: Deriving the spin-transfer-torque using different approaches. (a) We conceptually derived the STT effect in section 2.3.2 using the concept of spin accumulation by observing the layered structure. (b) Focussing on the second, spin analyzing layer, the STT is derived in a formal manner in section 2.3.4 (c) In section 2.3.5, the STT is derived by determining the effect near the normal metal–ferromagnet interface.

$j\parallel$ is along the positive $x$–axis and we point the ferromagnetic magnetization vector $\vec{M}$ along the positive $z$–axis, furthermore, we fix the interface at $x = 0$. Summarizing, the derivation will have the following sequence:

- first determine the spin current density tensor,
- then determine the spinor wavefunctions,
- calculate the accompanying spin current densities and
- finally calculate the spin torque from the net spin current

We will start the derivation by determining the wavefunction $\psi$ of an electron with an arbitrary spin state. This wavefunction can be described by a coherent superposition of the spin-up $|\uparrow\rangle$ and spin-down $|\downarrow\rangle$ states, as shown in figure 2.13(c), and this is called a spinor wavefunction. For a plane wave, with wavevector $k$ and traveling in the $x$ direction, the spinor wavefunction is given by:

$$\psi = \frac{e^{ikx}}{\sqrt{\Omega}} (a |\uparrow\rangle + b |\downarrow\rangle) = \frac{e^{ikx}}{\sqrt{\Omega}} \begin{pmatrix} a \\ b \end{pmatrix},$$

where $|a|^2$ and $|b|^2$ are the spin up and spin down probabilities, respectively, and $\Omega$ is a normalization volume. The two notations in equation (2.25) are identical, we will choose the second notation and represent a spinor as a vector. The spin current density tensor, equation (2.21), can be written as:

$$\frac{\mathbf{Q}}{2m} = \frac{\hbar^2}{2m} \text{Im} \begin{pmatrix} \psi^* \sigma_x \frac{\partial \psi}{\partial x} & \psi^* \sigma_y \frac{\partial \psi}{\partial y} & \psi^* \sigma_z \frac{\partial \psi}{\partial z} \\ \psi^* \sigma_y \frac{\partial \psi}{\partial x} & \psi^* \sigma_y \frac{\partial \psi}{\partial y} & \psi^* \sigma_y \frac{\partial \psi}{\partial z} \\ \psi^* \sigma_z \frac{\partial \psi}{\partial x} & \psi^* \sigma_z \frac{\partial \psi}{\partial y} & \psi^* \sigma_z \frac{\partial \psi}{\partial z} \end{pmatrix},$$

where the columns of this tensor are related to the motion of the electron, whereas the rows are related the the spin directions. Because we are looking at an electron described with a plane wave wavefunction in the $x$ direction, $\frac{\partial \psi}{\partial y} = \frac{\partial \psi}{\partial z} = 0$ and $\frac{\partial \psi}{\partial x} \neq 0$, therefore we can conclude that the spin current
density tensor, in this case, only has non-zero values for the $x$ spatial direction. After calculating the terms $\psi^* \sigma_i \frac{\partial \psi}{\partial x}$ for $i = x, y, z$ we can write the spin current density in the $x$ direction as:

$$\vec{Q}_x = \frac{h^2 k}{2m \Omega} \begin{pmatrix} 2 \text{Re} (ab^*) \\ 2 \text{Im} (ab^*) \\ |a|^2 - |b|^2 \end{pmatrix}$$

(2.27)

To describe an electron with a non-collinear spin state, we have to apply a matrix rotation. This matrix can be calculated using the Pauli matrices. Starting in the spin up state and rotation the spin over an angle $\theta$ in the $x-z$ plane, and using equation (2.25) the wavefunction of the incoming electron with wavevector $k$ traveling in the $x$ direction can be described by the spinor:

$$\psi_{in} = \frac{e^{ikx}}{\sqrt{\Omega}} \begin{pmatrix} \cos \theta \\ \sin \frac{\theta}{2} \end{pmatrix}.$$  

(2.28)

The transmitted $\psi_{tr}$ and reflected $\psi_{re}$ wavefunctions are calculated using the spin dependent transmission $t$ and reflection $r$ coefficients [40]:

$$\psi_{tr} = \frac{e^{ikx}}{\sqrt{\Omega}} \begin{pmatrix} t_1 \cos \frac{\theta}{2} \\ t_1 \sin \frac{\theta}{2} \end{pmatrix}$$  

(2.29)

$$\psi_{re} = \frac{e^{-ikx}}{\sqrt{\Omega}} \begin{pmatrix} r_1 \cos \frac{\theta}{2} \\ r_1 \sin \frac{\theta}{2} \end{pmatrix}.$$  

(2.30)

To derive the spin current density $\vec{Q}_x$ in the $x$ spatial direction we apply equation (2.27) which gives for the incoming spin current density:

$$\vec{Q}_{x,\text{in}} = \frac{h^2 k}{2m \Omega} \begin{pmatrix} \sin \theta \\ 0 \\ \cos \theta \end{pmatrix},$$

(2.31)
2.3 Spin transfer torque

for the transmitted spin current density:

\[
\vec{Q}_{x,\text{tr}} = \frac{\hbar^2 k}{2m\Omega} \begin{pmatrix}
\sin \theta \text{Re}(t_1 t_1^*) \\
\sin \theta \text{Im}(t_1 t_1^*) \\
|t_1|^2 \cos^2 \frac{\theta}{2} - |t_1|^2 \sin^2 \frac{\theta}{2}
\end{pmatrix}
\]  
(2.32)

and for the reflected spin current density:

\[
\vec{Q}_{x,\text{re}} = -\frac{\hbar^2 k}{2m\Omega} \begin{pmatrix}
\sin \theta \text{Re}(r_1 r_1^*) \\
\sin \theta \text{Im}(r_1 r_1^*) \\
|r_1|^2 \cos^2 \frac{\theta}{2} - |r_1|^2 \sin^2 \frac{\theta}{2}
\end{pmatrix}.
\]  
(2.33)

The spin transfer torque \(\vec{\tau}_{st}\) on an area \(A\) of the ferromagnetic layer is equal to the net spin current, \(\vec{Q}_{x,\text{in}} + \vec{Q}_{x,\text{re}} - \vec{Q}_{x,\text{tr}},\) transferred from the electrons to the ferromagnet. Using equation 2.24, \(\vec{\tau}_{st}\) can be written as:

\[
\vec{\tau}_{st} = -\int \hat{n} \cdot \vec{Q} \, d^2 r = A \hat{x} \cdot \left( \vec{Q}_{\text{in}} + \vec{Q}_{\text{re}} - \vec{Q}_{\text{tr}} \right).
\]  
(2.34)

Substituting equations 2.31 - 2.33 and using \(|r_1|^2 + |t_1|^2 = 1\) and \(|r_1|^2 + |t_1|^2 = 1\) and using the fact that there is no spin flip, the spin torque term simplifies to:

\[
\vec{\tau}_{st} = A \frac{\hbar^2 k}{2m\Omega} \begin{pmatrix}
\sin \theta \left[1 - \text{Re}(r_1 r_1^* + t_1 t_1^*)\right] \\
-\sin \theta \left[\text{Im}(r_1 r_1^* + t_1 t_1^*)\right]
\end{pmatrix}.
\]  
(2.35)

From this simple model we can conclude that there is only a spin torque in the directions transverse to the magnetization (i.e. \(\tau_{st,z} = 0\)). Furthermore, there is only a spin torque when the incoming spin current is non-collinear to the magnetization (i.e. \(0 < \theta < \pi\)). And finally the spin torque result from spin filtering of the ferromagnetic layer (i.e. \(t_1 \neq t_1^*\) and \(r_1 \neq r_1^*\)). In the case of perfect spin filtering, where only spin up electrons are transmitted \((t_1 = 1, t_1 = 0)\) and all spin down electrons are reflected \((r_1 = 0, r_1 = 1)\), equation 2.35 turns into:

\[
\vec{\tau}_{st} = A \frac{\hbar^2 k}{2m\Omega} \sin \theta \hat{x}
\]  
(2.36)

and we can conclude that, in this model, the spin torque is equal to the complete component of the spin current transverse to the magnetization.

2.3.5 Spin torque near the interface

In the previous section, we derived the STT in a formal manner, focussing on the second, spin analyzing layer, as shown in figure 2.12(b). In this section we
In the final part of 2.3.2 we made the assumption that the complete transverse spin current is absorbed at the normal metal–ferromagnet interface. Therefore we will look at the spin torque term near the interface in this section. Assuming an incoming spin polarized electron, with a wavefunction described by equation 2.28, we now model the ferromagnetic layer using a Stoner model as shown in figure 2.14. Due to the exchange interaction, the energies of the spin-up and spin-down electrons are different inside the ferromagnetic layer. The spin down (minority) electrons are higher in energy than the spin up (majority) electrons and the difference in energy is the exchange energy $E_{ex}$. The system is modeled by a potential energy step, at a position $x = 0$. The derivation is quite similar to that in the previous section:

\[ E_{ex} \]

\[ (2.37) \]

The incoming electron has a free-electron dispersion relation and its energy is therefore given by:

\[ E = \frac{\hbar^2 k^2}{2m}. \]

(2.37)

The physics near the metal–ferromagnet interface can now be described as a quantum mechanical scattering problem, where the incoming, transmitted and reflected spinor wavefunctions are given by:

\[ \psi_{in} = e^{ikx} \begin{pmatrix} a_1 \cos \frac{\theta}{2} \\ a_1 \sin \frac{\theta}{2} \end{pmatrix} \]  

(2.38)

\[ \psi_{tr} = e^{ik_{t_1}x} \begin{pmatrix} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{pmatrix} \]

(2.39)
\[ \psi_{re} = e^{-ikx} \begin{pmatrix} r_\uparrow \cos \frac{\theta}{2} \\ r_\downarrow \sin \frac{\theta}{2} \end{pmatrix}, \] 

where \( k \) is the wavevectors inside the normal metal and \( k_\uparrow \) and \( k_\downarrow \) the wavevec-
tors for spin-up and spin-down electrons in the ferromagnet\(^{17}\).

Just as in the previous section, the spin torque term can be calculated by determining the net spin current density in the \( x \) spatial direction. For this we first need to derive expressions for the incoming, transmitted and reflected wavefunctions. These can be derived by matching the wavefunctions and their derivatives at the interface between the metal and ferromagnetic layer and after solving these equations we can write the coefficients as:

\[ a_\uparrow = a_\downarrow = \frac{1}{\sqrt{\Omega}} \] 

\[ r_\uparrow = \frac{1}{\sqrt{\Omega}} \frac{k - k_\uparrow}{k + k_\uparrow} \] 

\[ r_\downarrow = \frac{1}{\sqrt{\Omega}} \frac{k - k_\downarrow}{k + k_\downarrow} \] 

\[ t_\uparrow = \frac{1}{\sqrt{\Omega}} \frac{2k}{k + k_\uparrow} \] 

\[ t_\downarrow = \frac{1}{\sqrt{\Omega}} \frac{2k}{k + k_\downarrow} \] 

To calculate the spin current density (equation \( 2.21 \)), the gradient and complex conjugates of the wavefunctions have to be determined and then it follows that, just as in the previous section, the spin current density only has an \( x \) spatial direction. After some derivation, we can write the incident spin current as:

\[ \vec{Q}_{x,in} = \frac{\hbar^2}{2m\Omega} \begin{pmatrix} \sin \theta \\ 0 \end{pmatrix} , \] 

the transmitted spin current as:

\[ \vec{Q}_{x,tr} = \frac{\hbar^2}{2m\Omega} \begin{pmatrix} \text{Re} \left( t_\uparrow t_\uparrow^* \right) \sin \theta \left( k_\uparrow + k_\downarrow \right) \cos \left( (k_\downarrow - k_\uparrow) x \right) \\ \text{Im} \left( t_\uparrow t_\uparrow^* \right) \sin \theta \left( k_\uparrow + k_\downarrow \right) \sin \left( (k_\downarrow - k_\uparrow) x \right) \\ |t_\uparrow|^2 k_\uparrow \cos^2 \frac{\theta}{2} - |t_\downarrow|^2 k_\downarrow \sin^2 \frac{\theta}{2} \end{pmatrix} \] 

and the reflected spin current as:

\[ \vec{Q}_{x,re} = -\frac{\hbar^2}{2m\Omega} \begin{pmatrix} \text{Re} \left( r_\uparrow r_\uparrow^* \right) \sin \theta \\ \text{Im} \left( r_\uparrow r_\uparrow^* \right) \sin \theta \\ -|r_\uparrow|^2 \cos^2 \frac{\theta}{2} + |r_\downarrow|^2 \sin^2 \frac{\theta}{2} \end{pmatrix} . \] 

\(^{17}\)In this model we have assumed \( k_\downarrow < k < k_\uparrow \).
Again, the spin torque $\vec{\tau}_{st}$ is calculated from the net spin current and using equations 2.46 – 2.48 yields:

$$\vec{\tau}_{st} = \frac{A}{\Omega} \frac{h^2}{2m} \begin{pmatrix} \tau_{st,x} \\ \tau_{st,y} \\ \tau_{st,z} \end{pmatrix},$$

(2.49)

where:

$$\tau_{st,x} = \sin \theta \left[ k \left( 1 - \text{Re} \left( r_{1} r_{1}^* \right) \right) - (k_{\uparrow} + k_{\downarrow}) \text{Re} \left( t_{1} t_{1}^* \right) \cos \left( (k_{\uparrow} - k_{\downarrow}) x \right) \right]$$

(2.50)

$$\tau_{st,y} = \sin \theta \left[ -k \text{Im} \left( r_{1} r_{1}^* \right) - (k_{\uparrow} + k_{\downarrow}) \text{Im} \left( t_{1} t_{1}^* \right) \sin \left( (k_{\uparrow} - k_{\downarrow}) x \right) \right]$$

(2.51)

$$\tau_{st,z} = 0$$

(2.52)

After this derivation we can deduce a couple of insightful results. From this model we can conclude that there is, just as in the previous model, only a spin torque in the directions transverse to the magnetization, and there is only a spin torque when the incoming spin current is non-collinear to the magnetization. We now like to distinguish three sources for spin-transfer-torque.

First there is the process of spin filtering. Spin filtering occurs when the transmission and reflection coefficients are spin dependent, as discussed in section 2.3.1. This leads to different transmitted and reflected spin currents in the ferromagnet and normal metal, respectively, and thereby a discontinuity in the spin current at the interface. This discontinuity is then counteracted by the STT acting on the magnetization.

A second source of STT is the so called spin precession. Because in our model the spin-up and spin-down electrons have different wavevectors in the ferromagnet, spatially varying phase factors turns up in the transmitted spin currents:

$$Q_{xx, tr} \propto \cos \left( (k_{\uparrow} - k_{\downarrow}) x \right)$$

(2.53)

$$Q_{xy, tr} \propto \sin \left( (k_{\uparrow} - k_{\downarrow}) x \right).$$

(2.54)

These two terms represent a precession of the spin about the z-axis as the spin penetrates the ferromagnetic layer. The period of this precession $\frac{2\pi}{k_{\uparrow} - k_{\downarrow}}$ is on the order of a few atomic layers in typical ferromagnets like Co. Now consider electrons entering the ferromagnetic layer from different directions, corresponding to different states at the Fermi surface. As they move through the ferromagnet, they all travel different paths and therefore precess with a different angle about the z-axis. This results in a classical dephasing and when summing up over all relevant parts of the Fermi surface, the transverse component of the spins vanishes.

Finally, the third source is, although not visible from our model, spin rotation. In this model, we start with a spinor in the xz-plane. However, starting
with a spinor which also has a \( y \)-component, adds a phasefactor \( e^{i\phi} \), where \( \phi \) is the azimuth angle, to the incident wavefunction (equation 2.38). Then the third source of STT, \textit{spin rotation}, occurs if the product \( r^\uparrow r^\downarrow \) is not real. The reflected spin will be rotated with respect to the incident spin direction and a difference in phasefactors \( \Delta \phi \) arises \[41\]. Thereby there is a discontinuity in the spin current and there will be an associated SST. This is a purely quantum mechanical concept which has no classical analog.

Besides showing the different sources of STT, we also justified the assumption, made in the last part of section 2.3.2, that the transverse spin current will be absorbed near the normal metal–ferromagnet interface. Partially it is absorbed at the interface due to spin filtering and the complete (remaining) transverse component is absorbed in the region close to the interface due to the spin precession.

One final point we want to show is that, although there is a net spin current density \( (Q_{x,\text{in}} + Q_{x,\text{re}} - Q_{x,\text{tr}} \neq 0) \), resulting in the STT, there is no net current flowing through the interface. Therefore we need to calculate the current densities \( \mathbf{j} \), given by equation 2.18, which, in this case, only have a component in the \( x \) spatial direction. The incoming, transmitted and reflected currents in the \( x \) spatial direction are given by:

\[
\begin{align*}
  j_{x,\text{in}} &= \frac{\hbar k}{m\Omega} \\
  j_{x,\text{tr}} &= \frac{h}{m\Omega} \left( |t^\uparrow|^2 \cos^2 \frac{\theta}{2} + |t^\downarrow|^2 \sin^2 \frac{\theta}{2} \right) \\
  j_{x,\text{re}} &= -\frac{\hbar k}{m\Omega} \left( |r^\uparrow|^2 \cos^2 \frac{\theta}{2} + |r^\downarrow|^2 \sin^2 \frac{\theta}{2} \right).
\end{align*}
\]

The continuity equation (2.19) tells us that there is, in this case, indeed no net current flowing:

\[
  j_{x,\text{in}} + j_{x,\text{re}} - j_{x,\text{tr}} = 0. \tag{2.58}
\]

### 2.3.6 Current-induced switching

In the previous sections we showed both a qualitative as well as a formal derivation the spin-transfer-torque and derived that by transfer of angular momentum from the electrons to the magnetization, we are able to switch the magnetization. In this section we want to present a equation relating the current needed to switch the magnetization to the properties of the ferromagnetic layer.

In general, when we apply a external magnetic field to ferromagnetic layer, the magnetization of this layer will respond. Spin dynamics is the field of research describing the response of the magnetization to external influences like the applied magnetic field. It is beyond the scope of this thesis to present a comprehensive description of the magnetization dynamics and the reader is therefore referred to \[42\].
The response of the magnetization in a ferromagnet after applying an external magnetic field, is governed by the well-known Landau–Lifshitz–Gilbert (LLG) equation. The effect of spin angular momentum transfer can now be incorporated into the LLG equation through the addition of an extra spin torque term.

For simple geometries and under a macro-spin approximation, this adapted LLG equation can be linearized and solved for its stability boundary. For a thin ferromagnetic layer in a collinear geometry the current $I_c$ at which the magnetization switches, can be calculated [42]:

$$I_c \propto \left( \frac{2e}{\hbar} \right) \left( \frac{\alpha}{\eta} \right) m.$$  \hfill (2.59)

In this equation $e$ is the elementary electron charge, $\hbar$ Planck’s constant, $\alpha$ the Gilbert damping, $\eta$ the spin-torque efficiency and $m = VM_s$ is the total magnetic moment of the ferromagnetic layer, where $M_s$ the saturation magnetization and $V$ the volume of the layer. The spin-torque efficiency $\eta$ is given by:

$$\eta = \frac{I_\uparrow - I_\downarrow}{I_\uparrow + I_\downarrow},$$

where $I_\uparrow$ and $I_\downarrow$ are the majority and minority spin-polarized currents, respectively.

In an experimental structure, the Gilbert damping $\alpha$ and the saturation magnetization $M_s$ are material parameters. Therefore, we can derive from equation (2.59) that in order to lower the critical switching current, we have to lower the ferromagnetic layer volume. The critical switching current as

![Figure 2.15: Switching current.](image-url)
a function of the lateral structure size, using experimental switching current densities on the order of $10^6 - 10^7$ A/cm$^2$ [43], is shown in figure 2.15. From this figure we can derive that structures created using conventional techniques like shadow masks, exhibit a critical switching current on the order of kA, which is totally unfeasible in an experiment. However, using nanostructuring techniques, as will be discussed in section 4.1.2, the switching current is on the order of mA, a value totally feasible.

We have now formally derived the spin-transfer-torque and discussed the different characteristics needed to exploit this effect. Furthermore, we showed an equation describing the current needed to switch the magnetization and concluded that the STT effect is only feasible in nanometer sized structures.
Chapter 3

Spin transfer torque devices

In the previous chapter we drew a qualitative picture of the spin-transfer-torque effect using the concepts of interfacial spin dependent scattering and spin accumulation. Also a more formal derivation of STT was given by considering the net change in the spin current. In this chapter we will discuss how we can design devices to investigate the STT effect experimentally. Therefore we will first present an overview of the STT device geometries presently known in literature in section 3.1. Then, the different steps in the nanostencil process are briefly described in section 3.2.1 and the choice for the nanostencil process is discussed section 3.2.2. Hereafter, some recent results obtained using the nanostencil process are presented in section 3.2.3 and finally, the critical factors in the nanostencil process are given in section 3.2.4.

3.1 General device geometries

In the previous chapter, when explaining the spin-transfer-torque effect using the concept of spin accumulation, we concluded that, in general, to exploit the STT effect we need a device consisting of:

- a spin polarizing layer,
- a spin analyzing layer,
- separated by a spacer layer
- and contacted with electrodes.

First the polarizing layer, polarizes the incoming current\(^1\). After passing through the spacer layer, this spin current can reorient the magnetization of the second magnetic layer by transfer of angular momentum. If we now measure the resistance of the device, by contacting it through the electrodes, we can determine

\(^1\)By spin dependent reflection and transmission
3.1 General device geometries

Two general experimental approaches are used to create spin torque devices. (a) Mechanical or (b) lithographic defined point contacts or nanopillar devices which can be made using subtractive techniques (c) and (d) or using a nanostencil approach (e).

![Figure 3.1: Schematic representation of different spin torque devices geometries.](image)

In all STT devices, there are common critical factors. First, the layers must be created in a well defined way, with the thickness of the layers on the order of nanometers. Furthermore, interfacial spin dependent scattering requires layers with low interfacial roughness. In section 2.3.6, we also deduced that, to obtain a considerable STT effect, a large current density, in the order of $10^6 - 10^7$ A/cm$^2$ \[43\], is needed. Therefore, a region of small cross-sectional area is needed in order to create these high current densities. We will now discuss how to create a STT device, addressing these critical factors.

Generally, two devices geometries have been used experimentally to create the small cross-sectional area needed, namely point contact devices and nanopillar devices. In a point contact device the polarizing, analyzing and spacer layer, together with the bottom electrode are created first. Then, to realize the small cross-sectional area, a sharp point\[4] contacts the functional layers and acts as the top electrode. This device geometry is schematically shown in 3.1(a) and was first realized by Tsoi et al.\[44\]. Instead of using a mechanical point contact, a lithographically defined point contact can be created. This

\[2\] Usually a sharpened Ag or Au tip is used and the contact area is on the order of 100 nm$^2$.\n
geometry is shown in figure 3.1(b) and the first results were reported by Myers et al. [45].

Beside point contact devices, another method to create the small cross-sectional area is to create so-called nanopillars. Nanopillar devices can be constructed by patterning at least one of the magnetic layers to a desired area using nanofabrication techniques. The nanopillar devices are schematically shown in figures 3.1(c) and (d). Kantine et al. were the first to report on current-driven magnetization reversal in Co/Cu/Co nanopillars with a diameter of 130 nm [46].

One other method to produce nanopillar STT devices is using the so-called nanostencil method, as first presented by Sun et al. [47]. This method uses a predefined stencil, located on top of the bottom electrode, to deposit the nanopillar into and the geometry is schematically shown in figure 3.1(e). In this thesis we use this nanostencil process to fabricate STT devices. Hence, we will now first briefly go through the process steps needed to create a nanostencil based STT device.

3.2 The nanostencil process

3.2.1 What is a nanostencil

Although we already got a glimpse of a STT device created using the nanostencil process, to fully understand the process, the consecutive process steps are now discussed. The first step in the nanostencil process is defining the bottom and top electrodes, later on used to contact the device. The electrodes are separated by an insulating layer and the resulting structure is schematically represented in figure 3.2(a). Then an opening in the top electrode is created by locally removing material as shown in figure 3.2(b). Hereafter an undercut is created in the insulating layer to clear the bottom contact under the opening. This step is illustrated in figure 3.2(c). After this undercut is created, the desired STT device, consisting of the polarizing, spacer and analyzing layer, can be deposited inside the nanostencil as shown in figure 3.2(d). The opening created in the second step now determines the size of the nanopillar. Finally, as shown in figure 3.2(e), the nanopillar is contacted to the top electrode.

3.2.2 Why nanostencils

As discussed in the first section of this chapter, there are a number of possible methods to create STT devices. Therefore we would like to treat the main advantages as well as the disadvantages of the nanostencil process compared to other methods. The main advantages of the nanostencil process are:
3.2 The nanostencil process

![Diagram of nanostencil process]

Figure 3.2: Schematic representation of the critical steps in the nanostencil process. (a) The first step in the process is defining the bottom and top electrodes, separated by an insulating layer. (b) Then, the opening of the nanostencil is created by locally removing the top electrode. (c) Hereafter an undercut is created in the insulating layer to clear the bottom contact under the opening. (d) After this, the desired STT device is grown inside the nanostencil and (e) finally the nanopillar is contacted to the top electrode.

- **Additive:**
  The nanostencil process is an additive process, whereas the nanopillar methods are subtractive. This basically denotes that, in the nanostencil process, the STT device is not affected after it is created. While, in the nanopillar method, the patterning happens after the STT device is created. This difference will be discussed in more detail in section 4.1.

- **Versatile:**
  The size and shape of the nanostencil opening, created in the second process step in figure 3.2, determine the size and shape of the STT device deposited inside the nanostencil. Because these can be altered easily, the nanostencil process is therefore a versatile process. This in contrast to the point contact devices, where there is only a limited control over the size and shape of the actual contact.

- **Scalable:**
  We can fabricate multiple nanostencils on a single sample. If desired, it is even possible to produce multiple nanostencils on the same electrodes.

- **Multiple purposes:**
  Although the nanostencil process is designed to create STT devices, other devices or materials can be deposited inside the stencil as well.
Obviously, the nanostencil process has also disadvantages:

- **Multiple process steps:** Although figure 3.2 only shows five different process steps, each of those steps again consists out of multiple steps, causing the nanostencil process to be time consuming serial process. However, it is possible to fabricate a large number of different nanostencils on one single sample.

- **Exact control:** Another disadvantage is the fact that it is difficult to completely control the shape of the STT device inside the nanostencil, because we are depositing the functional layers through a mask instead of patterning them using nanofabrication techniques.

### 3.2.3 Recent literature results

Sun et al. were the first to report on current induced magnetic switching measurements of STT devices using the a nanostencil method [47]. In their original paper they proposed a nanostencil process as described in the section 3.2.1 and showed their results for nanostencils of $50 \times 100\,\text{nm}^2$ in size. However, Sun et al. used a combination of electron beam lithography (described in section 4.1.2) and ion milling (described in section 4.1.3) to create the nanostencil opening. A cross-sectional SEM view of an array of nanostencils, created using their approach, is shown in figure 3.3(a). Figures 3.3(b) and (c) show a cross-sectional TEM view of a STT device created inside the nanostencil using a sputtering and evaporation technique, respectively.

Özyilmaz et al. proposed an alternative method to create the nanostencil opening using a focused ion beam (described in section 4.1.3) milling step [49]. In his thesis, this alternative method, however slightly adapted, is also used to create the nanostencil opening.

Both the method proposed by Sun as well as Özyilmaz, require additional nanostructuring steps for contacting their STT devices. However, we adapted the process so that these additional steps are not necessary. This is possible because our electrodes are completely pre-defined. For more details, please read the last part of this chapter.

Recent results for current induced magnetoresistance measurements on nanostencil based STT devices, are shown in figure 3.3 [50]. The resistance measurements are performed on a nanostencil with a cross-sectional area of $50 \times 100\,\text{nm}^2$ and clearly show the two resistance states of the device.
3.2 The nanostencil process

Figure 3.3: Recent literature results on nanostencils. (a) Cross-sectional SEM view of an array of nanostencils. Picture adapted from [47]. (b) Cross-sectional TEM view of a STT device inside a nanostencil created by a sputtering and (c) evaporation technique. Pictures adapted from [48]. (d) Field-induced and (e) current-induced switching of a 50 × 100 nm² nanostencil based STT device.
3.2.4 Critical factors in the nanostencil process

The critical factors in the nanostencil process can, as will be discussed in more detail below, be summarized by:

◦ the growth of the electrodes,
◦ the growth of the insulating layer,
◦ the growth of the functional layers and contacts,
◦ controlling the dimensions of the nanostencil and
◦ controlling the growth inside the nanostencil.

The first three items are addressed in chapter 5, whereas the last two items will be discussed in chapter 6. We will end this section by discussing the critical factors in the separate process steps, as represented in figure 3.2, in some more detail. Although this section is a must-read for the reader intended to reproduce the nanostencil process, the reader only interested in the actual results is suggested to omit this final part.

Growing the electrodes and insulating layer (chapter 5)

The first step in the nanostencil process is to create bottom and top electrodes, separated by an insulating layer, as shown in figure 3.2(a). This involves a number of critical steps. First, the actual creation of the layers. We want to create smooth, well defined layers having the desired electrical properties, namely good conducting electrodes with a decent insulating layer in between. For that we need to find candidates for these materials and subsequently determine the optimal growth conditions.

Next, we want to design our electrodes in such a way that they have relatively large contact pads with adjoining small leads. If the bottom and top electrodes are constructed perpendicular to each other, this results in four contact pads enabling resistance measurement of the device. The contact pads need to be large enough to be able to contact them using mechanical probes or a wirebonding technique. The leads connecting the contact pads need to have an area with a small lateral dimension in order to minimize the possibility of shunting the current through the insulating layer. To achieve the desired small lateral dimension, we will define the electrodes using a lithography process.

Finally, we want to create the insulating layer only in a small area around the nanostencil. This corresponds to the area around the overlapping electrodes. The insulating layer only serves to separate the two electrodes. In order to be able to contact the bottom electrode, the insulating layer should not be on top of this bottom contact. Therefore, we will also define the insu-

\footnote{The area where the two electrodes overlap and where the nanostencil thus will be made.}
3.2 The nanostencil process

Creating the nanostencil opening (chapter 5)

The nanostencil opening will determine the lateral dimensions of the nanopillar grown inside the nanostencil. Therefore an opening, with the desired dimensions, must be created in the top electrode. In order to successfully create an undercut in the insulating layer, the opening needs to be deeper than the thickness of the top electrode. Conversely, the hole must not be deeper than the insulating layer in order to prevent any damage to the bottom electrode, as is schematically shown in figure 3.2(b). Therefore the critical points in this step are the control over the desired dimensions of the opening.

Creating an undercut in the insulating layer (chapter 5)

The purpose of the undercut in the insulating layer, depicted in figure 3.2(c), is to create an opening between the bottom and top electrodes which, hereafter, is used to deposit the desired STT nanopillar into. For this reason, the undercut needs to be big enough to locally clear the electrodes, but small enough to still provide the necessary structural integrity in order that the electrodes do not deform. We will produce this undercut using a selective wet etch procedure, where only the insulating material is being removed. Therefore, one other material property for the electrodes is that they must withstand this wet etch. The critical point in this step is to find the wet etch parameters producing a desirable undercut.

Growth of the nanopillar inside the nanostencil (chapter 6)

Once the actual nanostencil, consisting of the locally opened top electrode, the insulating layer with undercut, and bottom electrode, is prepared, the nanopillar STT device can be deposited inside this stencil. This process step is schematically shown in figure 3.2(d). The critical point in this step is the control over the exact structure grown inside the stencil. For that reason, the variables governing the growth of the different layers needs to be optimized in order to produce smooth and well defined layers.

Moreover, the nanostencil itself influences the growth inside the nanostencil by effectively masking a part of the incident flux of deposited material. It is therefore crucial to know how this masking is related to the dimensions of the nanostencil.
3.2 The nanostencil process

Contacting the nanopillar device (*chapter 6*)

Finally, after the desired STT nanopillar is grown inside the nanostencil, the nanopillar needs to be contacted to the top electrodes in order to perform resistance measurements on the device. The critical point in this process step, shown in figure 3.2(e), is to completely contact the nanopillar with the top electrode. Nevertheless, we need to make sure that the only contact from the bottom to the top electrode is through the nanopillar, thus the two electrodes may not be shunted elsewhere.

After the nanopillar has been deposited inside the nanostencil and the STT device is contacted to the electrodes, we want to remove all layers deposited on top of the electrodes, remaining only the bare top electrode. Nevertheless, the STT device must remain unaffected thereby. To accomplish this, a sacrificial layer on top of the sample is introduced. Although not shown in the figure, this layer is already introduced in the first step and will not be removed until the last step.
Chapter 4

An experimental toolbox

In chapter 2 we already deduced that the dimensions of STT devices have to be in the sub-micron range. We also showed an overview of the nanostencil process and summarized its critical fabrication points in chapter 3. In this chapter we will first discuss the techniques needed to produce nanostencil based STT devices in section 4.1. Then, in section 4.2, we will discuss the techniques used for the characterization of the different process steps and finally we will briefly discuss the techniques used to perform measurements on STT devices. Readers completely familiar with fabrication and characterization techniques can continue with the next chapter, where the actual fabrication of a nanostencil is addressed in more detail.

4.1 Fabrication techniques

In this section we will discuss the techniques used to produce nanostencil based STT devices. Generally, the fabrication process can be divided into three parts, the addition of material, structuring and the removal of material. Additionally, we can distinguish the fabrication process between subtractive and additive process steps, as shown in figure 4.1.

In a subtractive process, figure 4.1(a), the desired material is first deposited onto the substrate and hereafter a masking layer is introduced. This masking layer can either be in physical contact with the structure, or float above the surface. Then, the unmasked material is removed, the masking layer is removed subsequently and the desired patterned structure is left over. In an additive process, however, the pattern is first defined and then the desired material is deposited, as shown in figure 4.1(b). Consequently, the integral nanostencil

\footnote{Typically our STT devices are 50 – 100 nm in height and have lateral dimensions on the order of 100 – 300 nm.}

\footnote{Completely or merely a part of the unmasked material.}
4.1 Fabrication techniques

**Figure 4.1: Subtractive and additive nanostructuring process steps.**

Generally, nanostructuring can be divided into (a) subtractive and (b) additive processes. Basically, both structuring processes consist out of the addition of material, patterning of the desired structure and the removal of material. In a subtractive process, the desired material is first deposited and patterned hereafter. In an additive process, however, the pattern is first defined and the desired material is deposited subsequently.

The process can be seen as an additive process, since the functional layers are deposited in a pre-defined stencil (see figure 3.2(d)). Nevertheless, separate steps in the process may still be subtractive.

Table 4.1 provides an overview of the different fabrication techniques used in this thesis, by categorizing the techniques into the addition of material, the generation of patterns and the removal of material. In the next sections we will briefly discuss the techniques, focusing on the aspects relevant for the nanostencil process. The reader interested in an introduction to a variety of (other) nanostructuring techniques, is referred to [51].

### 4.1.1 Material addition

STT devices are characterized by their small cross sectional areas, on the order of 50 – 300 nm, and their even smaller thickness, which is on the order of only a few nanometers. Also the patterning of structures often requires thin films, having high thickness homogeneity, covered over the whole substrate.
4.1 Fabrication techniques

Table 4.1: An overview of fabrication techniques. Generally, the device fabrication process can be divided into three parts, namely the addition of material, the generation of patterns and the removal of material. The table provides an overview of the different fabrication techniques used in this thesis. The numbers between the brackets denote the page number describing the specific technique.

<table>
<thead>
<tr>
<th>Material addition (section 4.1.1)</th>
<th>Pattern generation (section 4.1.2)</th>
<th>Material removal (section 4.1.3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>◦ Sputtering (48)</td>
<td>◦ Optical lithography (51)</td>
<td>◦ Wet etching (55)</td>
</tr>
<tr>
<td>◦ Spin coating (50)</td>
<td>◦ Electron beam lithography (51)</td>
<td>◦ Dry etching (55)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>◦ Focused ion beam milling (56)</td>
</tr>
</tbody>
</table>

Surface. Therefore, well defined growth or deposition of these thin layers is crucial. A considerable advance in thin film deposition has been made over the last decades. Methods such as, sputtering, thermal evaporation, molecular beam epitaxy (MBE) and atomic layer deposition (ALD) are nowadays common practice. In this section we will focus on the two techniques used in this thesis, namely sputtering for the fabrication of thin film layers and spin coating for the layers needed in the structuring of the sample.

Sputter deposition

Nowadays, sputter deposition is one of the most commonly used and well known methods for the deposition of thin\(^3\) films. Accordingly, numerous comprehensive reviews regarding sputter deposition technology can be found, see e.g. [52]. For sputter deposition, a target material is exposed to a bombardment of particles, whereby the target atoms (or clusters of atoms) are ejected and deposited onto a substrate surface. We will now discuss the sputter deposition is somewhat more detail.

Figure 4.2(a) gives a schematic overview of the sputtering process. A flow of an inert gas\(^4\) is injected into a vacuum system, close to the target material. By applying a large potential difference between the target, acting as anode, and a shielding ring, acting as cathode, a plasma is created. The high energetic plasma ions will bombard the target surface, ejecting the target atoms (or clusters of atoms) into the vacuum chamber and thereby depositing the target material onto the substrate. Using a shutter, placed between the target and substrate, the flux of material onto the target can be completely shielded,

---

\(^3\)Typically, sputtered layers are on the order of 0.5 – 500 nm thick.

\(^4\)Argon in our case.
Figure 4.2: Techniques used for the addition of material. For the addition of material, in this thesis, two different techniques are used. (a) In a sputtering process an Ar plasma is used to eject atoms (or clusters of atoms) from a target material. Due to the low background pressure inside the vacuum chamber, the atoms then can be deposited onto the substrate. A shutter can be opened or closed to control the sputtering onto the substrate. (b) An other method to produce thin layers is spin coating. Here a solution of the desired material is placed onto a centrally rotating plate. Rotation of the substrate results in a centrifugal force, whereby the floating of the material and the evaporation of the solvent lead to a decrease in layer thickness.

effectively stopping the deposition.

Because a current must be able to flow from the cathode to the anode, only conducting targets can be sputtered with the process described above. To sputter insulating materials, like SiO$_2$, a radio frequency (RF) sputtering technique is used. Here, an RF alternating voltage is applied, which can be coupled through any kind of impedance and thereby removing the restriction that the target should be conducting.

To confine the plasma in a smaller region close to the target, magnetron sputtering can be used in both of the techniques described above. In that case, magnets are placed behind the target, confining the plasma by the created magnetic field. Using magnetron sputtering, greater plasma densities are achieved, resulting in a higher sputter yield.

The sputter yield differs for various target materials. Some materials, like Cu, are sputtered relatively easy, whereas others, like SiO$_2$ have a significantly lower sputter rate. In general, the sputter rate is determined by the power put into the plasma, controlled by the applied voltage, and also the pressure,

---

5 In our case using a RF frequency of 13.56 MHz.
6 Defined as amount of material sputtered per unit of time and in the case of thin film sputtering denoted as nm/s.
regulated by the flow of argon gas. A higher pressure means more ions, resulting in a higher bombardment of the target material. Also the distance from the target to the substrate can influence the sputter rate, since more material will be deposited onto the substrate compared to the surroundings. In our setup, the Ar flow is typically on the order of 1.4 – 10 SCCM, resulting in a pressure of \(10^{-3} – 10^{-2}\) mbar. We use a plasma power in the range of 10 – 120 W and we can vary the target to substrate distance from 50 to 95 mm. Typically, this results in sputter rates of 0.2 – 2 Å/s.

Once a target atom is ejected into the vacuum chamber, we can describe the transport from target to substrate using kinetic theory of gases. One key parameter is the mean free path, which is the average distance the particle travels between collisions with other moving particles. Using a Maxwell-Boltzmann distribution of velocities, the mean free path \(\lambda\) can be described by the following relation:

\[
\lambda = \frac{k_B T}{\sqrt{2 \pi p \sigma}}
\]  

(4.1)

where \(k_B\) is the Boltzmann constant, \(T\) the temperature, \(p\) the pressure and \(\sigma\) is the cross section for scattering of the target atom onto the background gas atoms. Although a lower gas pressure means less material is ejected, we can deduce from equation (4.1) that the mean free path is increased accordingly. Likewise, at low pressures, the kinetic energy of the target atoms is higher, due to less scattering resulting from their longer mean free path. This can influence the quality of the deposited layers as will be shown in section 4.2.3.

As a final remark, we would like to note that since, in our case, the mean free path of the target atoms is typically less than the target to substrate distance, the flux arriving at the substrate does not all arrive at a normal incidence angle. Rather, the amount of flux at a certain angle, compared to the sample surface, is proportional to the sine of that angle [53]. This is in contrast to other deposition methods, like Molecular Beam Epitaxy (MBE), where the mean free path is much longer than the target to substrate distance and the deposition is effectively normal to the substrate plane.

**Spin coating**

Spin-coating has been used for several decades as a convenient method to quickly produce thin films. A typical spin coating process, shown in figure 4.2(b), involves the application of a solution of the desired material onto a substrate which is placed on a centrally rotating plate. The centrifugal force acting on the fluid, created by the rotation, causes the fluid to spread out over the substrate and eventually off the edges, leaving a thin film on the surface of the substrate. Simultaneously, the solvent component of the solution will evaporate, effectively decreasing the layer thickness, increasing the viscosity of the
4.1 Fabrication techniques

fluid and thereby eventually stopping the flow. Moreover, this will, after the complete solvent is evaporated only leave the desired material on the surface. The thickness of spin coated films depends on the properties of the desired material solution (viscosity, evaporation rate, concentration of the desired material) as well as the parameters chosen for the actual spin process (final rotational speed, acceleration, spin time). Accurate control of these parameters, enables the preparation of layers with highly reproducible thicknesses ranging from 10 nm – 10 µm. In our experiments we have used a Convac 1001S spinner, which allows us to change the rotational speed, the acceleration and the spinning time.

4.1.2 Pattern generation

In the previous section we discussed the fabrication techniques used for the deposition of material. In this section we will discuss how to pattern the acquired layers. There are numerous of methods to pattern structures to sub-micron sizes such as lithography, mechanical surface modifications using a scanning force microscopy (STM) tip or even using self-structuring masks. Essentially, the integral nanostencil process is also a method to pattern sub-micron sized structures. In this section, however, we will only discuss lithography steps used in the nanostencil process to create the electrodes and define the insulating layer. For those new to the field of lithography, please refer to e.g. for an introduction to the science of microlithography. For a more elaborate review of all aspects regarding microlithography and microfabrication, as well as detailed process recipes and information, please refer to . In general, a patterning process combines several steps in sequence. An overview of a typical patterning process using lithography steps is shown in figure 4.3.

The first step in the process, is the preparation of the sample by subsequent cleaning steps. Organic and inorganic contaminations present on the surface are removed by using different chemicals treatments. In fact, this is actually an example of a technique to remove material as will be discussed in section . After the sample is cleaned, often a so-called adhesion promotion step is included to ensure proper adhesion of the resist to the substrate.

Then, depending on the final desired structure and the actual used lithography method, a certain resist is applied to the surface as shown in the top of figure 4.3. This is usually done by spin coating the resist on the surface, as discussed in section . Hereafter, the resist is usually soft baked to evaporate remaining solvent.

Often, nanostructured devices are prepared using multiple process steps. Therefore, subsequent process steps have to be aligned to each other and an

\(^7\)Therefor often an additional drying step is included, where the substrate is heated for a certain period.
After the substrate has been carefully prepared, a layer of photoresist is applied. This can either be a so-called positive or negative resist. After the desired pattern is exposed, the exposed positive resist will be removed during the development step, whereas the unexposed resist is removed in the case of a negative resist. The subsequent step is either to use the acquired photoresist as a mask for deposition or etching. Then, the resist layer is removed. This step is known as a lift-off in the case there is material deposited on top of the resist. After the resist removal (or lift-off step), we end up with one of the four structures shown above.
extra *alignment* step is necessary before the actual *exposure* step can take place. There are a couple of possible sources for the exposure of the resist, leading to different lithography techniques. In this thesis we use optical lithography, using UV-light as a source, and Electron Beam Lithography (EBL), using, as the name implies, electrons as an illumination source. The key characteristics of both techniques will be briefly described in the end of this section.

The resist material undergoes a chemical reaction during exposure. Adopting this chemical reaction, we can divide the resist into two categories, namely positive and negative resist. A positive resist is normally insoluble to the chemicals referred to as resist developers. After the desired pattern is exposed, the resist is made more soluble to the developer and the exposed part will therefore be removed during the *development* step. In the case of a negative resist, the exact opposite occurs and the unexposed resist will be removed by the developer. By selectively exposing certain areas of the resist, a desired pattern can be created after developing, as is schematically shown in the second and third panel of figure 4.3. Before and after the exposure, additional baking steps can be included to further optimize the development step and resist quality respectively.

Once the lithography steps are performed, either deposition or removal of material can take place using for instance one of the techniques described in section 4.1.1. Then, the resist layer is removed. In the case that there is material deposited on top of the resist, this step is known as a lift-off process. After the resist removal (or lift-off), we end up with one of the four structures shown in the bottom panel of figure 4.3.

Before we proceed to discuss the optical and electron beam lithography, we will first summarize the steps in a typical lithography process:

- Substrate cleaning
- Adhesion promotion
- Application of resist
- Softbake resist
- Alignment of substrate
- Exposure of resist
- Post-exposure bake
- Development of resist
- Hardbake of resist

From this overview it is immediately clear that there is an even more extensive compilation of parameters influencing the final result. Each step has particular variables like, which resist to use, how long to expose and which dose to use, what temperature used to bake and how long, and so on. One can immediately understand that finding the optimal values for each desired pattern can be
4.1 Fabrication techniques

(a) Isotropic (b) Anisotropic

Masking layer

Substrate

Etched Layer

Figure 4.4: Two approaches for the removal of material. In general, we can distinguish between (a) isotropic and (b) anisotropic removal of material, where the resulting etched layer is shown in the figure.

a prolonged task. It would be beyond the scope of this chapter to present a detailed overview of all the parameters used during the fabrication of our nanostencil based STT device. However, this information is of critical value for those wanting to repeat our process. Therefore, this information can be found in appendix B.

We will end this section by summarizing the main advantages and disadvantages of both the UV lithography (UVL) as well as the electron beam lithography (EBL). The primary advantage of EBL is the fact that electrons are used for exposure, enabling patterned structure resolutions in the sub-100 nanometer regime. Another advantage of EBL over UVL is the fact that it is a form of maskless lithography, ensuring a large degree of flexibility. The primary advantage of UVL is the fact that this technique can be used to pattern a large area in one exposure, whereas in a EBL process, each pattern needs to be exposed independently. Our UVL exposures are performed using the Karl Suss MA6 Mask Aligner and the EBL in our FEI Nova Nanolab 600i dualbeam system using a Raith EBL module.

4.1.3 Material removal

Next to the deposition of material and the patterning of structures, the removal of material is one other key component in the fabrication of nanostencil STT devices. We can characterize this step by two features, namely isotropy and selectivity, which will be explained using figure 4.4. In this figure a part of the substrate is protected by a masking layer. If the material is removed isotropic, as is shown in figure 4.4(a), the local removal rate is equal for all spatial directions, whereas there is a preferential direction in the case of anisotropic removal.

8Using our UV lithography process we are able to pattern an area of $15 \times 15 \text{ mm}^2$. In industry, however, wafers up to a diameter of 30 cm can be patterned in one process step.
as shown in figure 4.4(b). The selectivity of the material removal determines the amount of desired material removed, compared to the other materials. In the case of a high selectivity, only the desired material is removed, as shown in figure 4.4(a).

To remove material, there are several methods possible. In this section we will discuss wet etching, dry etching and focused ion beam milling techniques as methods to, in some cases locally or selectively, remove material.

Wet etching

The two fundamental types of etching are liquid-phase (wet etching) and plasma-phase (dry etching). Each of these exists in several varieties. In a wet etch process, the substrate is usually immersed in a bath of the etchant, or the etchant is directly applied to the surface using for instance a rinsing bottle.

Different specialized etchants are used for the removal of specific materials. We already mentioned the cleaning of samples as a wet etch process. Here, organic and inorganic contaminations, present on the surface, are removed by using different chemicals. Often used chemicals for cleaning are isopropyl alcohol, acetone and ammonia.

Besides cleaning, the wet etch is used during one of the critical steps in the nanostencil process, namely for the creation of an undercut in the insulating layer. Therefore we will use an isotropic wet etch with a high selectivity for the insulating material. This can be accomplished by using SiO$_2$ as the insulating layer and hydrofluoric acid (HF) as etchant. In this case, the dissolution process of the solid SiO$_2$ into the aqueous HF solution can be described as follows:

$$\text{SiO}_2(s) + 6 \text{HF}(aq) \rightarrow \text{H}_2[\text{SiF}_6](aq) + 2 \text{H}_2\text{O}(l).$$

The etch rate$^9$ depends on the etching time, the concentration of the etchant in the solution, the temperature and also the replenishment of the etchant.

Dry etching

In a dry etching process, material is removed by exposing the substrate to a plasma, causing a bombardment of ions onto the substrate and thereby ejecting portions of the material from the exposed surface. This process is quite similar to the sputtering process, we now only remove material from our substrate instead from our target. Unlike most wet etching processes, the dry etching process typically etches anisotropically, as shown in figure 4.4(b). Usually, dry etching has a low selectivity. However, in the case of reactive ion etching (RIE),

$^9$Defined as the amount of material removed per unit of time and in this case usually denoted as nm/min.
the source gas for the plasma usually contains small molecules rich in chlorine or fluorine enhancing the etching of certain materials. A plasma containing oxygen is used to remove photoresist or other organic materials and is therefore often used as a cleaning step.

**Focused Ion Beam (FIB) milling**

One other way to locally remove material is using a Focused Ion Beam (FIB). Here a Liquid Metal Ion (LMI) source is used to generated ions, which are then accelerated and subsequently focused onto the sample by a set of electrostatic lenses. The FIB therefore very much resembles a scanning electron microscope (SEM), which will be discussed in section 4.2.1. However, the SEM uses a focused beam of electrons instead of ions.

Just as in the case of dry etching, the ions will bombard the sample surface and thereby removing (milling) material. However, unlike the dry etching which will remove material from the whole sample surface, we can now use a well defined focused beam of ions to locally remove material. By defining a pattern which the ion beam has to pass over, we can define the areas which need to be removed. If the beam is focussed longer onto a certain position, or if that position is patterned more often, more material can be removed. Also changing the flux of ions, the ion beam current, will cause more material to be removed. Just like dry etching, FIB typically removes material anisotropically, as shown in figure 4.4(b).

In our experiments the FIB we have used is incorporated in a FEI Nova Nanolab 600i dual beam system. This system consists of both an ion beam column as well as an electron beam column, hence the term dual beam. For an elaborate description of this particular setup, please refer to chapter 4 of [56]. Here we only want to mention that, besides the ion and electron column, the Nanolab 600i is equipped with a series of accessories and detectors, such as the EDAX EDX detector which enables the imaging of X-rays emanating from the substrate as will be described in section 4.2.2. The Nanolab 600i uses Ga ions, which can be accelerated from 1 to 30 keV, the ion beam current can be varied from 1.5 pA up to 20 nA and the minimum spot size can be on the order of 5 – 10 nm. In our experiments, we have used the lowest beam current of 1. pA at an acceleration voltage of 30 keV.

### 4.2 Characterization and measurement techniques

The previous sections of this chapter presented an overview of fabrication techniques divided into three parts, namely the addition of material, the generation of patterns and the removal of material. The final section of this chapter deals with the different characterization and measurements techniques used in this
4.2 Characterization and measurement techniques

Table 4.2: An overview of structural and surface characterization techniques. Using electrons, ions or photons as incident excitation species, the accordingly emitted electrons, ions or photons can be analyzed. Diverse techniques use different exited and emitted probes. The techniques described in this thesis can be found in the section between the brackets. The abbreviations used in this table are: Scanning Electron Microscopy (SEM), Energy Dispersive X-ray Spectroscopy (EDS), Focussed Ion Beam Microscopy (FIBM), Secondary Ion Mass Spectroscopy (SIMS), X-ray Photoelectron Spectroscopy (XPS) and X-Ray Reflectometry (XRR).

<table>
<thead>
<tr>
<th>Incident excitation probe</th>
<th>Emitted and analyzed response</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrons</td>
<td>SEM (4.2.1) EDS (4.2.2)</td>
</tr>
<tr>
<td>Ions</td>
<td>FIBM SIMS</td>
</tr>
<tr>
<td>Photons</td>
<td>XPS XRR (4.2.3)</td>
</tr>
</tbody>
</table>

thesis. Table 4.2 provides an overview of several structural and surface characterization techniques. Using electrons, ions or photons as incident excitation species, the accordingly emitted electrons, ions or photons can be measured and analyzed. In this section we will discuss the Scanning Electron Microscopy (SEM), Energy Dispersive X-ray Spectroscopy (EDS) and X-Ray Reflectometry (XRR). It is beyond the scope of this thesis to explain these techniques in all their details, therefore the reader interested in a more complete overview of the possibilities of scanning electron microscopy and X-ray analysis is referred to [57]. For a more elaborate description of X-ray reflectometry, please refer to e.g. [58].

4.2.1 Scanning Electron Microscopy (SEM)

Since our nanostencil structures are typically on the order of hundreds of nanometers and smaller, to examine these structures we will need a microscope capable of achieving high enough magnifications. A Scanning Electron Microscope (SEM) is capable of creating images with a magnification of up to 1500 000 times. To obtain this high magnification, a focused beam of electrons is created and scanned across the surface of the sample. The electrons will interact with the atoms that make up the sample producing signals which can be detected. In this section we will give a brief description of the origin of the detected signals and how we will use the SEM in this thesis.

As was already mentioned in section 4.1.3, our SEM is incorporated in the FEI Nova Nanolab 600i dual beam. Here, the electrons are generated by a Field Emission Gun (FEG) and subsequently accelerated to 1–30 keV. To collect an image, the sample is locally bombarded with the high energetic electrons.
4.2 Characterization and measurement techniques

Figure 4.5: Schematic overview of the processes occurring in a sample, caused by a bombardment of electrons. A focused beam of primary electrons hits the sample and penetrates into it. Electrons then will undergo both elastic and inelastic scattering events, whereby electrons will be backscattered, transmitted or create secondary electrons or X-rays.

These primary electrons (PE) generate a cascade of secondary electrons (SE), backscattered electrons (BSE) and forward scattered electrons (FSE) as shown in figure 4.5. It is beyond the scope of this thesis to discuss all events which can take place in the sample into detail. We would like to focus on the implications of the events on our measurements.

An SEM image is collected by simultaneously scanning the electron beam and subsequently detecting the generated particles. In a standard SEM image, the secondary electrons are collected. It is important to note that contrast differences in such an image are not only directly related to the depth information. This is caused by the fact that the detected electrons originate not only from the sample surface, but also from the sample within. Therefore the contrast in an SEM image can be caused by a couple of effects. If there are sharp edges present at the sample, much more secondary electrons are emitted from this edge compared to a smooth sample. One other source of contrast is caused by the differences in materials. This contrast is especially important in an SEM image where the backscattered electrons are collected, because the fraction of backscattering depends on the atomic mass. One final remark is that, although the electron beam can be focused to a spot size smaller than 1nm, the detected electrons generally originate from a larger area of the sample, ultimately limiting the maximum achievable resolution.
4.2 Characterization and measurement techniques

4.2.2 Energy Dispersive X-ray Spectroscopy (EDS)

In the previous section it was already mentioned that when the electrons of the focused electron beam hit the sample, a number of processes will occur inside the sample. One of such processes is the creation of X-rays. The Nova Nanolab 600i is equipped with a special detector, the EDAX EDX detector, which enables the imaging of X-rays emitted from the sample. Energy Dispersive X-ray Spectroscopy (EDS or EDX) is the analytical technique used for the elemental characterization of a sample using the characteristic X-rays.

Although there are numerous processes occurring inside the sample throughout the bombardment with electrons and during the creation and detection of the generated X-rays, this section only provides a brief overview of the EDS technique. Readers interested in a more detailed description are referred to [56, 57].

The origin of the characteristic X-rays is schematically shown in figure 4.6, where an incoming primary electron (PE) can excite an electron in an inner shell of the atom, ejecting it as a secondary electron (SE) and thereby creating an electron hole. The primary electron then continues as a backscattered (BSE) or forward scattered (FSE) electron. After a certain amount of time, the atom relaxes by an electron from an outer, higher-energy shell, filling the hole, as shown in figure 4.6(b). The difference in energy between the higher-energy shell and the lower energy shell may be released in the form of an X-ray. The number of X-rays and their energies are then collected into a so-called EDX spectrum. As the energies of the X-rays are characteristic of the difference in energy between the two shells, and of the atomic structure of the element from...
4.2 Characterization and measurement techniques

Figure 4.7: Typical EDX spectra of a sample consisting of 50nm Co$_{60}$Fe$_{20}$B$_{20}$ on top of 20nm Pt on a Si substrate. The spectra are collected using (a) 7 and (b) 15 keV electrons and the different characteristic X-ray peaks are visible.

which they were emitted, this allows the elemental composition of the specimen to be measured.

An example of a EDX spectrum collected from a sample consisting of 50nm Co$_{60}$Fe$_{20}$B$_{20}$ on top of 20nm Pt on a Si substrate, is shown in figure 4.7, where the spectra are collected using (a) 7 and (b) 15 keV electrons. The appearance of peaks at definite positions in the spectrum makes it possible to identify the presence of certain elements. These peaks are generally referred to as X-ray lines and appear in series, labeled as K, L, M etc. corresponding to the transitions between different shells. Figure 4.7(a) shows the K-lines of boron (BK), carbon (CK) and silicon (SiK), the L-lines of iron (FeL) and cobalt (CoL) and finally the M-line of platinum (PtM), collected using 7 keV primary electrons. The overlap of the Co and Fe peaks, caused by the small difference in X-ray energy, is clearly shown. However, when using a higher electron energy of 15 keV, as shown in figure 4.7(b), we are also able to collect the Co and Fe K-lines which do not overlap. Although the same sample is used in both cases, the intensity of the peaks can be totally different. One of the effects causing the difference in intensities is the penetration depth of the incoming electrons. The higher the energy of the electrons, the greater the penetration depth and thereby more X-rays can be generated in the Si substrate, resulting in a higher Si peak.

We already have discussed that, although the electron beam can be focused to a spot size smaller than 1 nm, the detected electrons generally originate from a larger area of the sample, as shown in figure 4.5. This is also true for the generated X-rays. Therefore besides the difference in penetration depth, the electron energy also influences the radial distance were the X-rays are gener-
ated. This results in a lower spatial resolution of EDS than is achievable with the SEM.

In most cases, however, one is not only interested in the possible presence of an element (qualification), but also in its relative abundance. The process of determining relative abundances is called quantification. In contrast to the other qualitative imaging techniques, EDS can provide quantitative information about the composition of a sample. This is done by measuring the peak intensity of certain elements and comparing them to a reference spectrum of that same element.

4.2.3 X-Ray Reflectometry (XRR)

After a thin film is created, for example using sputter deposition, we would like to determine its thickness to calibrate the growth rate. A technique which provides information about the thickness of thin films is X-ray reflectometry (XRR). Besides thickness information, XRR also can provide information on the crystalline properties of the material and the roughness of the film. The principle of XRR is based on the constructive and destructive interference of X-rays that are reflected from a crystal lattice or a layer interface.

When we want to determine the thickness of the deposited layer, we use XRR in a glancing incidence angle configuration, which means that the source and the detector are almost in the same plane as the sample. In this case, constructive interference will occur between reflections from the top interface and the interface between the film and the substrate. The condition for this constructive interference is than given by Braggs law:

\[ 2d \sin \theta = n \lambda, \]  

where \( d \) is the film thickness, \( \theta \) the angle between the incoming X-rays and the sample surface, \( n \) an integral number and \( \lambda \) the wavelength of the X-rays.

A typical XRR measurement is shown in figure 4.8, where the XRR measurements are performed on a sample consisting of a Cu layer sandwiched between two 2nm thick Pt layers. These experimental results can than be compared with simulated data using GIXA (Glancing Incidence X-ray Analysis) software which is used to estimate the layer thickness and gives an indication of the roughness. From (a) we can derive that the period of the oscillations is 0.22° corresponding to a Cu layer thickness of 360nm. Figure (b) also shows a Cu layer, only now deposited using different sputter conditions. Because in this measurement the period of the oscillations is smaller, the layer has to be thicker. In this case the Cu layer is has a thickness of 430nm. Besides the fact that the Cu layer is thicker, the roughness of the layer is higher. This can be deduced from the amount of oscillations still visible at higher angles. In the case of roughness, the local thickness varies, and destructive interference
between reflections from different sample regions occurs, making the high order (larger angle) reflections harder to see. As a final note, we would like to mention that Braggs law is a first order approximation based on interference in a single layer. Interference patterns can also be simulated using dedicated software to obtain more accurate information about layer thickness, density and especially roughness [59].
Chapter 5

Creating a nanostencil

The previous chapter showed the fabrication and characterization techniques used in the nanostencil process. In this chapter we present the results for the fabrication of a nanostencil, focusing on the first three critical steps discussed in section 3.2.4. The first three sections address the first critical step, namely the electrodes and insulating layer. We will therefore briefly treat the material choices in section 5.1. Then the optimal growth conditions are investigated in section 5.2 and the patterning is described in section 5.3. The results to accomplish the second critical step, the creation the nanostencil opening, are presented in section 5.4. Finally, the results for the third critical step, the creation of an undercut in the insulating layer, are shown in section 5.5.

5.1 Material choices

The first step in the nanostencil process is the creation of the electrodes and the insulating layer in between. As already discussed in section 3.2.4 we want to create smooth, well-defined layers having the desired electrical properties. For well-conducting electrodes, there are numerous materials possible. Often used electrode materials are Au, Cu, Pd, Pt, Ru and Ta. However, our electrode material must withstand the wet etching step needed to create an undercut in the insulating layer later on. Therefore we have chosen Pt as electrode material\(^1\), because it is a decent conductor and because of its property to withstand most wet etchants. For the insulating layer material there are also a lot of materials possible. We will use SiO\(_2\), because, when grown properly, it is an excellent insulator and we can use an HF etch, as discussed in section 4.1.3 to create the desired undercut.

The electrodes and insulating layer will be created using a lithography process. But before continuing to discuss the patterning, we first need to determine

\(^1\)At least for the parts of the electrodes which can come into contact with the etchant.
5.2 Optimal growth conditions

Both the Pt and SiO$_2$ thin films will be grown by sputter deposition as discussed in section 4.1.1. In the sputter process there are a lot of parameters influencing the deposition rate as well as the roughness of the sputtered thin films. In this section we will investigate the effect of the sputter pressure, by changing the argon flow, the sputtering power, the sample to target distance
and the influence of different seeding and capping materials on the deposition rate and roughness of the sputtered films. Our goal is to find the optimal growth conditions, denoted by a low as possible film roughness together with a high as possible deposition rate.

The first parameter we would like to investigate is the sputter pressure. The pressure in the sputter chamber depends on the sputter gas flow rate and the pump capacity during sputtering. Because in our system the pump capacity remains constant during sputtering, we will use the argon flow rate as the optimization parameter. Figure 5.1(a) shows the XRR intensity as a function of the angle $2\theta$ for different argon flow rates $\phi$, whereas the sputter power is 80W, the sample to target distance is 95mm, and the seed and cap layers are Pt. For reasons of clarity, the different graphs are shifted in intensity in such a manner that they do not overlap. As discussed in section 4.2.3, the amount of oscillations per angle $2\theta$ is a measure of the film thickness and the angle $2\theta$ to where the oscillations are visible is a measure of the roughness. Thereby figure 5.1(a) clearly shows that a smaller Ar flow results in a lower film roughness. Using dedicated analysis software [59], we determined the roughness of the layers. The roughness to thickness ratio of the sample sputtered using 7.0 sccm is 5.5% $\pm$ 0.5%, whereas the sample sputtered using 1.4 sccms has a ratio $< 1.0% \pm 0.5%$. The inset shows the dependence of the deposition rate on the Ar flow, indicating a maximum around 3sccm. Since our mail goal is to produce low roughness films, we opt for the lowest possible Ar flow which is, in our system, 1.4 sccm.

Another important parameter in the sputter deposition is the used sputter power. At low power ($< 40W$) the Ar flow needed to keep the deposition plasma stable starts rising significantly and it is no longer possible to use the desired flow of 1.4sccm. At higher power ($> 120W$) the power source is not able to couple all the power into the plasma. Therefore the sputter power was varied between 40 and 120W and the results are shown in figure 5.1(b), where the Ar flow is 1.4sccm, the sample to target distance is 95mm and the seed and cap layers are Pt. We did not see any significant change in film roughness. All three samples have a roughness to thickness ratio of $< 1.0% \pm 0.5%$. However, the deposition rate did change significant from 0.06 Å/s at 40W to almost 0.24 Å/s at 120W. Since the roughness of the films did not change significant, we will use the highest sputter power possible.

Yet another parameter directly related to the growth rate and roughness of the films is the distance between sample surface and the sputter target. We investigated this effect by varying the distance from 65mm to 95mm, the minimum and maximum value we can use in our system. When using a Ar flow of 1.4sccm, a sputter power of 80W and a seed and cap layers of Pt, the results can be found in figure 5.1(c). We can conclude that the maximum sample to target distance of 95mm results in the film with the lowest roughness. The
5.3 Patterning the electrodes and insulating layer

Table 5.1: Optimal growth conditions for SiO$_2$, Pt, Co, Co$_{60}$Fe$_{20}$B$_{20}$ and Cu. The table shows for each element the Ar flow, sputter power $P$ and sample to target distance $d$.

<table>
<thead>
<tr>
<th>Element</th>
<th>$\phi$ (sccm)</th>
<th>$P$ (W)</th>
<th>$d$ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO$_2$</td>
<td>1.4</td>
<td>120 RF</td>
<td>95</td>
</tr>
<tr>
<td>Pt</td>
<td>2.1</td>
<td>60 DC</td>
<td>95</td>
</tr>
<tr>
<td>Co</td>
<td>7.0</td>
<td>20 DC</td>
<td>95</td>
</tr>
<tr>
<td>Co$<em>{60}$Fe$</em>{20}$B$_{20}$</td>
<td>7.0</td>
<td>20 DC</td>
<td>95</td>
</tr>
<tr>
<td>Cu</td>
<td>3.0</td>
<td>20 DC</td>
<td>95</td>
</tr>
</tbody>
</table>

The deposition rate will decrease with increasing sample to target distance.

One final parameter we investigated is the seeding and capping material. Although it is not expected that a different material choice will influence the deposition rate, the roughness of the film, however, might be different. The results for Pt, Ta and Pd are shown in figure 5.1(d) and we can conclude that the roughness and the deposition rate of SiO$_2$ is more or less equal for all material choices we investigated, because all three samples have a roughness to thickness ratio of $<1.0\% \pm 0.5\%$.

To conclude this section, we have investigated that the optimal growth conditions for SiO$_2$ in our case are an Ar flow of 1.4sccm, a sputter power of 120W and a sample to target distance of 95mm. We also have shown that we are able to grow SiO$_2$ onto Pt with a low film roughness. Using dedicated analysis software, we determined the roughness of this SiO$_2$ layer using a 4-point resistance measurement technique and showed that the resistance is $>10\text{M}\Omega$, ensuring a well-insulating layer.

A similar investigation has been carried out for the other materials used in this thesis, namely Pt, Co, Co$_{60}$Fe$_{20}$B$_{20}$ and Cu. This analysis results in the optimal growth conditions as given in table 5.1.

5.3 Patterning the electrodes and insulating layer

Now the optimal growth conditions are determined, we want to pattern our electrodes in such a way that they have relatively large contact pads with adjoining small leads. The contact pads are needed to provide a way to create an electrical contact between the STT device and our measurement equipment. Therefore, the size of the pads needs to be large enough to ensure contacting using sharp needles and micromanipulators is possible. Conversely, smaller contact pads we are able to create more nanostencils onto one sample. We will use an optical lithography technique as described in section 4.1.2 to pattern the electrodes.
5.3 Patterning the electrodes and insulating layer

Figure 5.2: Patterning of the electrodes and insulating layer. (a) A top view showing the patterned Pt bottom electrode after deposition and subsequent lift-off. (b) A close-up of the 2 μm wide junction area, showing the patterned 30 × 30 μm² insulating SiO₂ layer after deposition and lift-off.

First, a UV mask has been fabricated. The mask includes 225 electrodes with contact pads of 200 × 200 μm² and leads down to 2 μm wide. The exact layout of the created mask can be found in appendix C. Using this mask, we are able to create the electrodes on our sample using the process steps as described in section 4.1.2. The exact process steps are given in appendix B here we will only present the results.

A top view of the patterned Pt bottom electrode after deposition and subsequent lift-off is shown in figure 5.2(a). This SEM top view shows a part of the sample surface, where the bright areas are covered with Pt and the dark area is the Si substrate. The electrodes are separated 1 mm apart. The image clearly shows the 200 × 200 μm² contact pads and the adjoining small leads. The small crosses, surrounding to the contact pads are alignment marks needed in the next process steps. For convenience, numbers have been patterned next to the electrodes to provide a way to quickly retrieve certain devices on the sample.

Next, the insulating SiO₂ layer is created. Since we only want the insulating layer in a small area around the nanostencil, we use an electron beam lithography process to pattern this layer. In this manner, we are able to exactly determine the size and position of the insulating area. An area of 30 × 30 μm² centered on the junction area is patterned and the results, after deposition and lift-off, are shown in figure 5.2(b). Here, a SEM close-up of one of the electrodes is presented. The patterned square SiO₂ layer is visible on top of the 2 μm wide leads. The small crosses on the image are used as alignment marks during the EBL process.
The last step in the patterning process is to repeat the first UV procedure, however, in this case with the mask rotated 90°. The alignment marks, depicted by the small crosses in figure 5.2, ensure the top electrode is created on the desired position on the sample with respect to the bottom electrode. After this second UV lithography process, we created the our desired structure consisting of the two electrodes separated by the insulating area. Hereafter, we are ready to create a nanostencil in the $2 \times 2 \mu m^2$ wide junction area.

After the electrodes have been made, the sacrificial layer on top of the sample is deposited. After the nanostencil has been created and the desired STT device has been deposited inside, this layer enables the removal of all deposited material on top of the electrode, while the deposition remains unaffected. The sacrificial layer is a spin coated 180 nm thick PMMA layer and the exact process steps to create this layer are given in appendix B.

5.4 Creating the nanostencil opening

The next step in the nanostencil process is creating the nanostencil opening which will determine the lateral dimensions of the nanopillar grown inside the nanostencil. We will therefore use the focused ion beam milling technique, as discussed in section 4.1.3, to create an opening in the PMMA layer and top Pt electrode. For the proceeding steps, the opening needs to be deeper than the PMMA layer and top electrode, but not deeper than the insulating SiO$_2$ layer. To determine the optimal milling time, we have developed a measuring sequence where we deliberately mill through the complete structure. Subsequently, the optimal milling time is determined using the EDS technique as described in section 4.2.2. We will now explain the measuring sequence in some more detail and thereafter show the results.

When using the FIB to create the nanostencil opening, the top PMMA layer is first removed. Subsequently the first Pt layer, SiO$_2$ layer and second Pt layer are milled away. When continuing milling, also the substrate material will be locally removed. We can now use the element specificity of the EDS technique to determine which materials are still present after a certain milling time. Before addressing this, we will first discuss the EDX spectra of an unmilled sample consisting of PMMA(180nm) / Pt(20nm) / SiO$_2$(50nm) / Pt(20nm) on a Si substrate, as shown in figure 5.3. Here, the EDAX detector signal is measured as a function of the x-ray energy for an electron beam voltage of 7 or 15 keV, figure 5.3(a) or (b), respectively. The figures clearly show peaks for C, O, Pt and Si corresponding to the elements present in the sample.

If we collect an EDX spectrum at one specific location and determine the

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4PMMA (polymethylmethacrylate) consists out of carbon, hydrogen and oxygen atoms and has the chemical formula $(C_5H_8O_2)_n$
5.4 Creating the nanostencil opening

Figure 5.3: EDX spectra of a sample consisting of PMMA(180nm)/Pt(20nm)/SiO$_2$(50nm)/Pt(20nm) on a Si substrate. The spectra are collected using (a) 7 and (b) 15 keV electrons.

area of the occurring peaks, we can assign a value, corresponding to the integrated area, for each element to that specific location. This procedure will be done automatically in the EDAX Genisis software. We are now able to determine if a certain element is present as a function of the position on our sample, if we scan the electron beam over the sample and collect and process a spectrum at every position. This procedure allows us to make so-called linescans.

We will exploit this by making a series of 300 nm diameter holes in our stacked sample, using a FIB milling current of 1.5 pA and an acceleration voltage of 30 keV. Then, we will increase the milling time from 0 to 27 s with 1 s steps for each hole. Using the process described above, EDX element specific linescans are collected across the holes and the results are shown in figure 5.4, where the linescans are collected using 15 keV electrons.

Figure 5.4 shows both the collected linescans as well as SEM images of the holes. Because the contrast in the SEM top view images is low, images under an angle of 52° are also incorporated. As the milling time increases, the SEM images of the holes become brighter, indicating the presence of the Pt layer. Although we want to mill away the material layer for layer, the SEM images indicate that this is not occurring. During the first 13 s of milling, the 52° SEM images show a convex shape arising in the milled holes. Then, during the next 3 s, a small left over dot of material in the middle of the hole is visible. This undesirable artefact is caused by redeposition of milled material inside the hole. If the milling time is further increased, this left over material is eventually removed.

The EDX linescans clearly show an periodic behavior with a width of approximately 3 nm.

Note that although the EDX measurements and the SEM images are from the same array of holes, they can be shifted in position because they are not taken simultaneously.
Figure 5.4: EDX linescans of a sample consisting of PMMA(180nm) / Pt(20nm) / SiO2(50nm) / Pt(20nm) on a Si substrate, for different milling times. The Si, Pt and C linescans are collected using 15 keV electrons and the EDX signal is plotted as a function of the position. This position corresponds to the array of 300nm holes shown in the accompanying SEM pictures (showing a top view and a view under an angle of 52°). For each hole, the milling time is increased by 1s.

proximately 300nm, corresponding to the diameter of the holes. To explain the origin of this periodic behavior, we have to think in terms of the elements still present inside the milled holes. In the case of an unmilled area, we expect to see a EDX signal from the Si, Pt and C present in our sample as shown in figure 5.3. Indeed, the EDX signal in the area in between the holes is constant in figure 5.4. If we now locally remove material from our sample, we expect to see a decrease in the EDX signal of the elements of the material being removed. Using the FIB we will first start to remove the top PMMA layer and hereafter the first Pt layer. This is apparent in figure 5.4 where the periodic behavior is first visible in the C signal and thereafter in the Pt signal. A decrease in the Pt signal, however, is accompanied by an increase in the Si signal. This is due
to the fact that the Pt layer effectively stops a part of the incoming electrons before they can reach the SiO$_2$ or Si layer, as discussed in section 4.2.2. If we would have milled away our complete structure, we would only expect a Si signal coming from our substrate. However, this will not be the case in our sample due to the non-local detection property of the EDS measuring technique as described in section 4.2.2.

We now explained the periodic behavior of the EDX linescans, but how can we use this effect to determine the optimal milling time. Therefore, we will determine the average EDX signal inside the hole and plot this value as a function of the milling time as is shown in figure 5.5(a) and (b), respectively.

Figure 5.5(a) shows the EDX linescans of Si, Pt and C for 12, 15, 18 and 21s milling time, where the dotted line represents the signal for the unmilled sample. The signal inside the hole is determined by averaging the signal of the hatched area. This procedure is repeated for all the holes and the results are shown in figure 5.5(b), where the dotted line represents the signal for the unmilled sample. We can distinguish five different areas in this figure, which represent the five different stages of the milling process as shown in figure 5.6(a).

◦ (1) During the first 12s, the top PMMA layer is milled away. This can be seen as the decline in the C signal. The Pt and Si signal however, remain unchanged. This is due to the fact that a light element as C, contrary to a heavy element like Pt, does not stop a large part of the incoming electrons.

◦ (2) The next 3s show a decrease of the Pt signal accompanied by an increase in the Si signal, caused by the removal of the top Pt layer.

◦ (3) Then, the Pt and Si signal are approximately constant for 4s. Although we actually are milling the SiO$_2$ layer, the signal coming from our Si substrate is so much greater that we do not see the effect of the milling in our measured Si EDX signal.

◦ (4) After the SiO$_2$ layer has been milled away, during the next 3s the second Pt layer is removed. This is clearly visible in another decrease in Pt signal and correlated increase in Si signal.

◦ (5) Continuing milling will effectively only remove material from our Si substrate. However, due to the already discussed non-local property, we will still measure a small Pt and C signal, although they are not present in the hole anymore.

We now derived the result we were aiming for in this section. Using the developed EDX linescans measuring sequence we have measured the milling depth as a function of the milling time, and thereby we can determine the optimal milling time. We want to mill away the complete PMMA layer and top Pt electrode, but not the complete SiO$_2$ layer, therefore the optimal milling time is marked by area 3 in figure 5.5(b) and is in the range of 15 to 19s for 300nm holes using a FIB milling current of 1.5pA. The ensure that inhomogeneities
5.4 Creating the nanostencil opening

Figure 5.5: EDX linescans of a sample consisting of PMMA(180nm) / Pt(20nm) / SiO2(50nm) / Pt(20nm) on a Si substrate, as a function of the milling time. (a) The EDX linescans of Si, Pt and C for 12, 15, 18 and 21s milling time, where the dotted line represents the signal for the unmilled sample. The average signal inside the hole, marked by the hatched area, is plotted as a function of the milling time in figure (b).

Figure 5.6: Comparison between the EDX measurement and the CASINO simulation. (a) The five different stages of the milling process. (b) The open symbols represent the normalized EDX signal of figure 5.5(b), whereas the closed symbols represent the CASINO simulation. The five different stages of figure (a) are also shown in the figure.
5.5 Creating the undercut

After the nanostencil opening is created, the undercut in the insulating layer must be made. This is done using a wet etch technique as described in section 4.1.3. To selectively etch away the SiO\textsubscript{2} layer we use HF as etchant. To determine the optimal etching parameters we have investigated a series of different HF solutions and etching times. From this analysis we conclude that a 5 minute etch using a 1\% HF solution in H\textsubscript{2}O results in the desired, while minimizing damage to our sample.

To determine the size of the undercut, we will use the SEM to image the nanostencil from a top view and a cross-sectional view. This cross-section is made using the FIB and the imaging with the SEM is performed under an angle of 52°. Figure 5.7 shows the results for a 200 nm nanostencil after 5 minutes HF (1\%) etch. Note that figure 5.7 shows the SEM images of a 200 nm nanostencil after the deposition of 50 nm Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} and subsequent lift-off. This is done because the PMMA layer on top of our sample complicates the imaging and creation of the cross-section. The undercut, marked by the dotted

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\footnote{Our samples are 25 × 25 mm\textsuperscript{2} and the detailed analysis, as discussed above, is performed in the center of the sample whereas the other analysis has been performed approximately 5 mm from the edge of our sample. The patterned area of our samples if 15 × 15 mm\textsuperscript{2}.}
Figure 5.7: The undercut present in a 200 nm nanostencil. (a) A top view showing the undercut, marked by the dotted line, and reflected by the bright area surrounding the nanostencil opening. (b) The cross-sectional SEM image clearly shows the undercut in the SiO$_2$ layer. The undercut in the 200 nm nanostencils is 450 nm in diameter. Both images are taken after sputtering of 50 nm Co$_{60}$Fe$_{20}$B$_{20}$ and subsequent lift-off.

We have now determined the optimal growth conditions for the materials and showed that we can pattern the electrodes and insulating layer. Furthermore we have showed an actual fabricated nanostencil and, moreover, using our developed EDX measuring sequence, we are able to determine the optimal milling time for an arbitrary nanostencil geometry. We will now continue by describing the growth inside our nanostencils in the next chapter.
Chapter 6

Growth inside a nanostencil

The previous chapter showed the actual fabrication of a nanostencil. The next step is to deposit material into this nanostencil. This chapter presents the results for the growth inside a nanostencil, where we are mainly interested in how the material is deposited inside the nanostencil. Therefore, the sputtering inside the nanostencil is investigated in section 6.1. Subsequently, a model describing the deposition inside a nanostencil is presented in section 6.2, where we will compare our measured results with the simulations. Finally, we end by presenting some interesting simulated depositions.

6.1 Sputtering inside a nanostencil

The results of the previous chapter enable us to create nanostencils with the desired dimensions. Next, we will investigate the growth inside the nanostencils by sputtering. Therefore a series of nanostencils with a diameter of 200 and 300 nm are prepared in a non-structured sample consisting of two 20 nm thick Pt electrodes separated by a 50 nm SiO$_2$ layer and covered with a spin coated 180 nm thick PMMA layer. The milling time for the 200 nm nanostencils is estimated to be 7.6 s.

After the nanostencils are created, a thin film of varying Co$_{60}$Fe$_{20}$B$_{20}$ thickness and 2 nm of Pt is sputtered on the sample surface and thereby also inside the nanostencil. We sputter a calibrated Co$_{60}$Fe$_{20}$B$_{20}$ thickness, varied from 10 to 70 nm, both on the 200 as well as the 300 nm nanostencils, producing a total set of 12 samples each containing 8 nanostencils. The sputter rate for the

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1 Non-structured in this case means that the sample is a homogenous thin film sample, without any electrodes patterned.

2 The optimal milling time for a 300 nm nanostencil, using a current of 1.5 pA, is 17 s. Based on the difference in area and assuming a cylinder of material is removed, thereby volume, a 200 nm diameter hole will take $(\frac{200}{300})^2 \times 17s = 7.6s$ to mill.
thin films is calibrated using the technique described in section 4.2.3. After sputtering, the samples undergo a lift-off process step, as described in section 4.1.2 whereby the PMMA layer, the Co$_{60}$Fe$_{20}$B$_{20}$ and Pt sputtered on top of the PMMA layer are removed. However the material deposited inside the nanostencil remains unaffected during the lift-off process.

To investigate the depositions, SEM images are collected of the nanostencils after sputtering and lift-off and the results are shown in figure 6.1 and 6.2. The granularity of the depositions inside the nanostencil is attributed to the growth of the Co$_{60}$Fe$_{20}$B$_{20}$ onto the Pt bottom electrode. The 20, 50 and 70 nm depositions inside the 300 nm nanostencil, show a larger brighter grain, in the middle of the deposition. We attribute this to left-over redepositions after the wet-etch step to create the undercut. These effects might be suppressed by an additional layer deposited before the rest of the structure is grown (introducing a so-called seed layer) and additional cleaning of the sample before sputtering. The result thereof, on the device grown inside the nanostencil, will be discussed in section 7.1.2.

Subsequently, EDX element specific linescans, for Si, Pt and Co are performed across the nanostencils. The results for the 200 nm nanostencils are shown in figure 6.1 and for the 300 nm nanostencils in figure 6.2, where all linescans are collected using 7 keV electrons.

From the EDX Co signal present in the linescans we can conclude that there is indeed Co$_{60}$Fe$_{20}$B$_{20}$ present inside the nanostencils. Although the linescans of Fe and B are not integrated in figures 6.1 and 6.2, the presence of the elements is confirmed by separate EDX spectra. We can conclude from the linescans that an increase in sputtered Co$_{60}$Fe$_{20}$B$_{20}$, results in an increase in Co EDX signal. This increase is accompanied by a decrease in the Pt signal, although the Pt thickness is constant, less X-rays are being generated inside the Pt due to the fact that more electrons are stopped inside the thicker Co$_{60}$Fe$_{20}$B$_{20}$ layer. A similar trend is shown in the Si EDX linescans, where an increase in Co$_{60}$Fe$_{20}$B$_{20}$ layer thickness is associated with a decrease of the Si signal inside the nanostencil. However, even if there is no Co$_{60}$Fe$_{20}$B$_{20}$ deposited inside the nanostencil, the Si signal shows a higher plateau inside the nanostencil area.

To investigate the origin of the plateau in the Si signal, we need to determine where the Si EDX signal is generated. From an analysis using CASINO simulations, it has been confirmed that most of the Si signal originates from the Si substrate and not from the 50 nm thick SiO$_2$ layer even at an electron energy of 7 keV. Furthermore, the undercut in the SiO$_2$ layer is approximately laterally 125 nm, further increasing the contribution of the Si signal from the substrate. In the area outside the nanostencil, the substrate is effectively covered with 40 nm of Pt, whereas the area inside the nanostencil only has 22 nm
Figure 6.1: EDX line scans of a nanostencil with a diameter of 200nm after sputtering and lift-off. The amount of Co$_{60}$Fe$_{20}$B$_{20}$ sputtered has been varied from 10 to 70nm, where the sputter rate is calibrated for a thin film sample. The Si, Pt and Co line scans are collected using 7 keV electrons and the EDX signal is plotted as a function of the position. This position corresponds to the 200nm holes shown in the accompanying SEM images.
6.1 Sputtering inside a nanostencil

Figure 6.2: EDX linescans of a nanostencil with a diameter of 300nm after sputtering and lift-off. The amount of Co$_{60}$Fe$_{20}$B$_{20}$ sputtered has been varied from 10 to 70nm, where the sputter rate is calibrated for a thin film sample. The Si, Pt and Co linescans are collected using 7 keV electrons and the EDX signal is plotted as a function of the position. This position corresponds to the 300nm holes shown in the accompanying SEM images.
6.1 Sputtering inside a nanostencil

of Pt on top of the Si. Thereby, inside the nanostencil area, more electrons will penetrate into the substrate generating a larger Si signal then in the area outside the nanostencil.

Besides the conclusion that the Co EDX signal increases with increasing Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} thickness, figures 6.1 and 6.2 also show that the Co signal, for the same sputtered Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} thickness, is higher inside the 300 nm nanostencils than in the 200 nm nanostencil. This indicates that more material is deposited inside the larger nanostencil. To investigate the amount of material deposited inside the nanostencil, EDX spectra are collected for all nanostencils using an electron energy of 7 and 15 keV. As an example, the EDX spectrum of the 300 nm nanostencils collected using 15 keV electrons is presented in figure 6.3(a). The correlation between the increase in sputtered Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} and the Co EDX peak is indeed shown. Also the increase in the Fe peak is visible, whereas the Pt peak, at an electron energy of 15 keV, remains almost constant.

Using a specialized software package called Stratagem [61], a quantification of the thickness of the Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} depositions is performed. In literature, almost all EDS measurements are performed on bulk or homogenous samples. EDS is then almost exclusively used to identify certain elements in a composition and often to determine their abundance. Stratagem, however, can account for interactions between different layers and thereby enables EDS quantification of layered samples. Provided the area of the different peaks in a EDX spectrum are calculated, Stratagem is capable to determine the thickness of layers. Stratagem is also able to use spectra collected at multiple electron energies, exploiting the availability of multiple X-ray peaks as discussed in section 4.2.2.

For all EDX spectra, Stratagem is used to calculate the Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} thickness inside the nanostencils and the results of this analysis are shown in figure 6.3(b). We already indicated that for the same sputtered Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} thickness, more material is deposited inside the larger nanostencil. This statement is confirmed by the results of figure 6.3(b). Furthermore, we now derived the amount of material deposited inside the nanostencil as a function of the sputtered thickness.

\textsuperscript{3}The bottom Pt electrode is 20 nm thick and the protective Pt on top of the Co\textsubscript{60}Fe\textsubscript{20}B\textsubscript{20} deposition is 2 nm thick.

\textsuperscript{4}In practice, to so-called k-ratio, is determined. This k-ratio is the ratio between the area of the peak in the layered sample, compared to a reference sample of that specific element. For more information about k-ratios, please refer to [57].

\textsuperscript{5}The EDX spectra for both the 200 and 300 nm nanostencils are collected using 7 and 15 keV electron energy.
6.2 Modeling the growth inside a nanostencil

The main goal of this section is to model the amount of material actually deposited inside the nanostencil and predict the shape of the deposition inside the nanostencil. To determine the deposition rate, we will introduce the so-called tooling factor (TF), which is calculated from the ratio between the height of the sputtered structure inside the nanostencil and the amount of sputtered material. When normally sputtering thin films onto a substrate surface, this tooling factor is equal to one, meaning that the sputtered layer thickness is equal to the calibrated thickness. The presence of the nanostencil will influence the TF and we expect less material being deposited inside the nanostencil, effectively decreasing the TF.

Although we have derived the amount of material deposited in the previous section, the shape of the deposition inside the nanostencil remains yet unclear, because the measured EDX linescans cannot be translated to height profiles directly. Despite a cross-section of the deposition, shown in figure 5.7, gave a glimpse of its profile, the spatial resolution of the SEM, even in the optimal conditions, is not quite high enough. Therefore an alternative approach is used to gain insight in the deposition profile and a numerical model is developed to describe the growth inside a nanostencil.

Figure 6.3: Analysis of the deposited Co$_{60}$Fe$_{20}$B$_{20}$ thickness inside the nanostencils. (a) EDX spectra, of the 300 nm nanostencils collected at 15 keV, showing the increase of the Co, Fe and Pt peaks for increasing sputtered Co$_{60}$Fe$_{20}$B$_{20}$ thickness. (b) The calculated thickness (using Stratagem) as a function of the sputtered Co$_{60}$Fe$_{20}$B$_{20}$ thickness, for both the 200 and 300 nm nanostencils.
6.2 Modeling the growth inside a nanostencil

The model is based on the fact that the nanostencil will mask a portion of the incident flux of material and thereby less material is deposited inside the nanostencil than on top of it, as shown in figure 6.3(b). We will now explain the geometry used for the model using figure 6.4 and 6.5. If we first look at a clear flat surface, the amount of material \( d \) deposited per unit of time \( t \) at a position \( (x, y) \) on the surface can be described by:

\[
d(x, y) = \int_{-\pi/2}^{\pi/2} \int_{0}^{\pi} f(\Theta, \varphi) \, d\Theta \, d\varphi,
\]

where \( f(\Theta, \varphi) \) is the incident flux of material as a function of the azimuth \( \varphi \) and polar angle \( \Theta \), as shown in figure 6.4(a). If the incident flux is created using a sputtering technique, this function will be axisymmetric along the azimuth angle and will roughly follow a sine law for the polar angle [53] (see section 4.1.1) and consequently can be described by:

\[
f(\Theta, \varphi) \propto \sin(\Theta).
\]

If, however, another technique is used to deposit the material, e.g. molecular beam epitaxy (MBE) or evaporation, the incident flux \( f \) can be described by another function of \( \varphi \) and \( \Theta \).

If we now look at the case of a nanostencil mask being placed above the surface, as shown in figure 6.4(b), the incident flux is now (partly) obstructed at certain positions on the surface. Therefore the integration boundaries for the polar angle in equation 6.1 are now functions of the position and azimuth angle.

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The incident flux in this case can be described as a Gaussian peak centered around a polar angle \( \Theta_0 \), where \( \Theta_0 \) is the angle between the normal of the plane and the source.
6.2 Modeling the growth inside a nanostencil

We will now model the nanostencil as the intersect of an ellipse based cone with a plane of thickness $t$ at a height $h$ above the surface. Figure 6.5(a) shows this geometry for a cross-section along the $xz$-plane, where the angle of the nanostencil edge is given by $\Theta_A$. The integration boundaries for the polar angle can now be deduced from this representation. For a given position $x$ along the semimajor axis, and azimuth angle $\varphi = 0$, the minimum and maximum polar angles are thereby shown in figure 6.4(a).

Using goniometric relations, it is possible to calculate the polar angle as a function of the position inside the nanostencil and the azimuth angle $\varphi$. Then, using the incident flux distribution given by equation 6.2, we are able to calculate the deposition rate $d$ as described in equation 6.1. The formal derivation of the deposition rate inside a nanostencil, for a cross-section along the $x$-axis, will be given in appendix D.

Once the deposition rate $d$ is known, the deposition profile $z$ of the deposited material inside the nanostencil can be derived by integration of $d$ over time:

$$z(x, y, t) = \int_0^t d(x, y) \, dt. \quad (6.3)$$

However, since $d$ is also a function of $z$, this problem cannot be solved analytically. For that reason a numerical model has been developed, which will now be described.

This numerical model will calculate the deposition rate $d$ for a cross-section along the $x$-axis for a given nanostencil geometry. The model will take into account that the deposition profile $z$ changes and thereby influences the deposition rate $d$. Also the fact that during the deposition not only material gets...
6.2 Modeling the growth inside a nanostencil

Figure 6.6: SEM image of a nanostencil used for modeling the growth inside the nanostencil. A layer of 10 nm Co$_{60}$Fe$_{20}$B$_{20}$ has been sputtered on top (and partially) inside (a) a 200 and (b) 300 nm nanostencil, where the PMMA layer has not yet been lifted off. The image shows the inner and outer radii of the FIB hole inside the PMMA and Pt and partially covered with the Co$_{60}$Fe$_{20}$B$_{20}$.

deposited inside the nanostencil, but also on top and on the edges is reckoned with. Both effects are schematically shown in figure 6.4b).

The input of the model is given by the geometry of the nanostencil and the desired number of deposition iterations. The parameters related to the nanostencil geometry are the semimajor and semiminor axis $a$ and $b$, the nanostencil height $h$, the top layer thickness $h$ and the angle of the nanostencil edge $\Theta_A$. The model can then be described by the following sequence of events:

- Create nanostencil geometry.
- Determine the minimum and maximum polar angles for all angles $\varphi$.
- Calculate the deposition rate at this position.
- Repeat this for all positions along the $x$-axis.
- Recalculate nanostencil geometry.
- Repeat the process.

A detailed flowchart for the model is presented in appendix E.

6.2.2 Results for the deposition profiles

Before presenting the results of this model, we first have to mention how certain parameters have been determined. The model incorporates the fact that the

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7In the case of a circular nanostencil $a = b = r$, with $r$ the radius of the nanostencil.
Figure 6.7: Imaged, measured and calculated deposition profiles. A cross-sectional SEM image of the deposition after sputtering of 50 nm Co$_{60}$Fe$_{20}$B$_{20}$ and subsequent lift-off for (a) a 200 nm and (b) 300 nm nanostencil. The measured EDX linescans are shown in the middle panels and the calculated profiles are presented in the bottom panels of the figure. The amount of sputtered Co$_{60}$Fe$_{20}$B$_{20}$ is varied from 10 to 70 nm.
holes created in the PMMA and top Pt layer, as described in section 5.4, are not perfectly cylindrical. Also the fact that the deposition takes place inside and on top of the nanostencil as well as on the edges of the nanostencil opening is taken into account for in this model. To determine the parameters governing this effect, we will use figure 6.6. Here, a layer of 10 nm Co$_{60}$Fe$_{20}$B$_{20}$ has been sputtered on top (and partially) inside a 200 and 300 nm nanostencil, where the PMMA layer has not yet been lifted off. From this picture we are able to determine the inner and outer semimajor and semiminor axis $a$ and $b$, as well as the angle $\Theta_A$ of the nanostencil edge. In this case we determined an angle $\Theta_A = 61 \pm 4^\circ$ for the 200 nm nanostencils and $\Theta_A = 58 \pm 3^\circ$ for the 300 nm nanostencils.

The sputter profiles are calculated for both the 200 and 300 nm nanostencils, where the amount of sputtered material is varied from 10 to 70 nm. The results of the calculated profiles are presented in the bottom panels of figure 6.7, whereas the measured EDX linescans are shown in the middle panels and the top panel of figure 6.7 shows cross-sectional SEM images of the deposition after sputtering of 50 nm Co$_{60}$Fe$_{20}$B$_{20}$ and subsequent lift-off in a 200 and 300 nm nanostencil.

From the simulations we can conclude that the nanopillar grown inside the 200 nm nanostencil has a $150 \pm 10$ nm wide plateau in the center adjacent by $100 \pm 10$ nm wide steep edges, whereas the 300 nm nanostencil has a $240 \pm 10$ nm wide plateau adjacent by $100 \pm 10$ nm wide steep edges. During the growth inside the nanostencils, the shape of this plateau becomes more convex, while the edges become steeper. Determining the angle of the nanopillar edge for the last deposition (70 nm Co$_{60}$Fe$_{20}$B$_{20}$ sputtered), yields an angle of $52 \pm 3^\circ$ for the 200 nm nanostencil and $54 \pm 3^\circ$ for the 300 nm nanostencil. The only known literature value for the angle of a sputtered nanopillar inside a nanostencil is $66^\circ$[48]. However the latter nanostencil has a diameter of only 80 nm and the angle is determined using a cross-section TEM view, as shown in figure 6.10(e). From our cross-sectional SEM image, as shown in the top panels of figure 6.7, we are not able to determine the angle of our grown nanostencils.

When comparing the measured EDX linescans in figure 6.7 with the calculated deposition profiles using our model, we can conclude that the EDX linescans are more rounded. This is due to the fact that an EDX measurement at a certain position inside the nanostencil, is not corresponding to the exact Co$_{60}$Fe$_{20}$B$_{20}$ thickness at that position but is an average over a larger area. This effect would cause the calculated (using Stratagem) height to be lower than expected. If we now compare this calculated height with the height determined from our simulations, shown in figure 6.8(a), we indeed notice that the modeled height is always higher than the measured height, although not significantly higher.

Now that the deposition profile is investigated, we can determine the tooling
6.2 Modeling the growth inside a nanostencil

Figure 6.8: Calculated and modeled sputter rates inside a nanostencil. (a) The height of the nanopillar as a function of the sputtered Co$_{60}$Fe$_{20}$B$_{20}$ thickness calculated using Stratagem and determined from our model. The 200 nm nanostencil represented using circles and the 300 nm nanostencils are represented by squares. The filled symbols represent the model, whereas the open symbols show the Stratagem results. (b) The tooling factor (TF) is calculated from the ratio between the height of the nanopillar and the amount of sputtered Co$_{60}$Fe$_{20}$B$_{20}$.

The tooling factor (TF), which, in this case, is calculated from the ratio between the height of the modeled deposition profile and the amount of sputtered Co$_{60}$Fe$_{20}$B$_{20}$. The tooling factor has been determined for all measured and calculated samples and the results are shown in figure 6.8(b), where the 200 nm nanostencil results are represented using circles and the 300 nm nanostencils are represented by squares. The filled symbols represent the model, whereas the open symbols show the Stratagem results. From this figure we conclude that the TF for the 200 nm nanostencils is 0.56 ± 0.03 and for the 300 nm nanostencils is 0.68 ± 0.02. Even though we determined a constant TF, a slight decreasing trend is observed in the figure 6.8(b). This decrease in TF with increasing amount of sputtered material can be attributed to the shrinking nanostencil opening, as shown in figure 6.5, resulting in a smaller incident flux in the nanostencil.

6.2.3 Model predictions

In a schematic representation of a STT device, the structure grown inside the nanostencil often has a pillar shape with perfectly flat layers, as is shown in figure 6.9(a). In reality however, this is not the case, as we have already shown in cross-sectional SEM pictures, figure 6.7, and concluded from our simulation analysis, and shown in figure 6.9(b).

From the modeled structure, as show in figure 6.9(b), we can conclude
that the layer thickness near the edges of the structure is particularly non-homogenous. For our sputtered structures this implies that the layers will intermix near the edges. In the case of an all-metal GMR device, such as the Co/Cu structure shown, this region will not contribute to the GMR effect, thereby effectively decreasing the GMR ratio of the whole device. The same argumentation holds for the STT effect in an all-metal device. If, however, we want to deposit insulating spacer layers to exploit the TMR effect, the intermixing of the layers near the edges will cause a shunting of the device, diminishing the TMR effect.

Although these imperfections near the edges of the structure will decrease the potential STT effect, the effect will still occur. Nevertheless, the steepest possible edges and the flat as possible layers are desired. One of the main advantages of describing the growth inside a nanostencil using a numerical model, is that we are now able to investigate how the geometry of the nanostencil and the deposition parameters will influence the deposition profile inside the nanostencil. In this manner we are able to simulate different geometries and select the geometry, resulting in the most optimal deposition profiles, for future nanostencil STT devices. To investigate how the geometry of the nanostencil influences the deposition profile, we model three different scenarios.

First we calculate the deposition profile for different heights of the nanostencil, where the height $h$ is given by the etched insulating SiO$_2$ layer, as shown in 6.4(a). Then, we investigate the influence of thickness $t$ of the top PMMA and Pt layer, as shown in 6.4(a). Thereafter, we determine the deposition profile for an ellipse shaped nanostencil, while changing the ellipse area. Finally, we want to determine how the deposition method influences the deposition profile inside the nanostencil. Therefore we model an evaporated deposition, where we will use a Gaussian distribution for the incoming flux.

In the previous section we calculated the deposition profile for circular nanostencils with a nanostencil height of 50 nm, a top layer thickness of 200
nm and using a sputter deposition. For all simulations, a deposition of 50 nm of sputtered material is modeled. We will now briefly discuss the results for the different geometries and deposition method.

**Nanostencil height**

The calculated profiles for the sputter deposition into a 300 nm circular nanostencil are presented in figure 6.10(a), where the nanostencil height is varied for 5, 25, 50 and 100 nm. From the deposition profiles we can conclude that the nanostencil height needs to be as low as possible to obtain steep edges and a flat plateau. Also we can conclude that the tooling factor decreases with an increasing nanostencil height. Caused by the fact that an increase in nanostencil height, effectively decreases the boundary angles of the incoming flux of material.

**Top layer thickness**

The influence of the top layer thickness is shown in figure 6.10(b), where the thickness is varied for 20, 100, 200 and 400 nm. From this we can conclude that a thicker top layer results in a flatter plateau area, whereas there is no significant change in the steepness of the edges. An increase in top layer thickness, decreases the TF. Also this effect can be attributed to the effective decrease of the boundary angles of the incoming flux of material.

**Ellipse area**

All experiments presented in this thesis are performed on circular nanostencils. However, in future STT devices, an ellipse shape is preferred. Until now, we are not yet able to fabricate appropriate ellipse shaped nanostencils. However, in our simulation we are able to adjust the shape of the nanostencil. Figure 6.10(c) shows the deposition profiles along the semiminor axis for three different ellipse areas, namely $50 \times 100$ nm$^2$, $100 \times 200$ nm$^2$ and $150 \times 300$ nm$^2$. Although the TF increases with increasing nanostencil area, the shape of the deposition profile does not change significantly.

**Evaporated depositions**

The final parameter we investigated is the influence of the deposition method. Until now, all experiments and simulations are performed on sputtered depositions. Although using other deposition methods in experiments can be

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8In an ellipse shaped STT device, the magnetization of the ferromagnetic layers can be oriented along the semimajor axis due to the induced shape anisotropy.
Figure 6.10: Calculated deposition profiles for different nanostencil geometries and deposition parameters. (a) Increasing nanostencil height from 5, 25, 50 to 100 nm. (b) Increasing nanostencil top layer thickness from 20, 100, 200 to 400 nm. (c) Increasing area for an ellipse shaped nanostencil. The area is increased from $50 \times 100 \text{ nm}^2$, $100 \times 200 \text{ nm}^2$ to $150 \times 300 \text{ nm}^2$. (d) Increasing Gaussian width, typical for evaporated depositions. We varied the FWHM of the incoming flux distribution from 1, 5, 10, 20, 30 to 40°. (e) A cross-section TEM view of a sputtered STT device inside a nanostencil. Picture adapted from [48]. (f) A cross-section TEM view of an evaporated STT device inside a nanostencil. Picture adapted from [48].
impractical or time consuming, it can easily be implemented in our model. For the evaporated depositions, equation 6.2 is altered and the incident flux is described as a Gaussian peak centered around 90°. Figure 6.10(d) shows the deposition profiles for different Gaussian peak widths. We varied the FWHM of the incoming flux distribution from 1, 5, 10, 20, 30 to 40°, where 1 – 4° would be a typical value for evaporation techniques. From the calculated deposition profiles we can conclude that, as expected, the nanostencils become more rounded and the TF decreases as the FWHM of the incoming flux distribution increases.

Determining the angle of the nanopillar edge for the evaporated deposition, yields an angle of 70 ± 2° for the 1° FWHM Gaussian distribution and 52 ± 2° for the 40° FWHM Gaussian distribution. The only known literature value for the angle of a evaporated nanopillar inside a nanostencil is 82° [48]. However, we do have to note that this measurement, as shown in figure 6.10(f), is performed on a different nanostencil geometry as has been calculated here.

From this analysis we can conclude that nanostencils created using an evaporation technique, with a narrow incoming flux distribution, exhibit steeper edges and have a flatter plateau than sputtered nanostencils. Therefore, for an optimal deposition profile, an evaporation method is preferred for future STT device fabrication.

The main goal of this chapter was to investigate the deposition profile inside a nanostencil. We achieved this goal by imaging, measuring and modeling the growth inside our nanostencils. Furthermore, we simulated the deposition profiles for different nanostencil geometries and deposition methods. The next step in the process would be to create an actual STT device inside our nanostencil and perform electrical measurements on this device.
Chapter 7

Transport measurements

In chapter 5 we showed that we are able to create nanostencils with a desired shape and size. Then, the growth inside this nanostencil was discussed in chapter 6, where we are able to predict the shape and size of the deposition by altering the geometry of the nanostencils. The next step would be to fabricate an actual STT device. However, before growing structures inside a nanostencil, we first perform measurements on non-patterned GMR devices in section 7.1. Here, we will investigate the influence of the deposition conditions focusing on the implications for future STT devices. Subsequently, we present the first preliminary transport measurement on a fully integrated STT device in section 7.2.

7.1 Results on CIP-GMR devices

The STT devices we are aiming to create inside our nanostencils essentially consist of a GMR stack where the current is flowing perpendicular to the plane of the layers. Therefore, before attempting to grow, analyze and optimize this structure inside a nanostencil, it is sensible to investigate homogenous non-patterned samples first. Using non-patterned homogenous samples, we are able to investigate the GMR effect, without affecting the growth by the presence of the nanostencil. Using this approach we are able to investigate the deposition related parameters and determine their implications for future STT devices.

Because we require homogenous non-patterned samples, we will fabricate Current-In-Plane (CIP) GMR devices, which are discussed in section 2.2.3. In contrast to Current Perpendicular to Plane (CPP) GMR devices, discussed in section 2.2.3, CIP-GMR devices do not require additional nanostructuring to pattern structures of small cross-sectional area. Although the exact geometry is different, growth related parameters influencing the GMR effect in both device geometries can be compared.
Despite the tremendous experience in the fabrication of magnetic thin film structures, no CIP-GMR devices have been fabricated in our research group before. Therefore, the following discussion may appear rather technological, however, we do want to emphasize its relevance for future STT devices.

7.1.1 CIP-GMR device fabrication

We will now present the analysis for the CIP-GMR of so-called Exchange-Biased Spin-Valves (EB-SV’s). These GMR devices, discussed in section 2.2.3, consist essentially of two ferromagnetic (FM) layers separated by a non-magnetic (NM) spacer and an antiferromagnet (AF) in contact with one of the ferromagnetic layers. The purpose of this AF layer is to fix the magnetization of the contacting FM layer by an exchange interaction between the layers. The effect of this layer will be explained later on, when the magnetoresistance curves will be discussed in more detail.

For the fabrication of our EB-SV’s we use a sputter deposition process, as described in section 4.1.1. Using a so-called shadow mask during sputtering, we can produce devices of 1 cm in length and 200 µm wide. During the sputtering process we apply a magnetic field of 25 mT. This field is directed in the sample plane, aligning the magnetization of the ferromagnetic layers along the applied field and thereby creating a preferential magnetization direction.

Our EB-SV’s are sputtered onto a Si substrate and typically consist of the following layered structure: Pt / Co / Cu / Co / IrMn / Pt, where IrMn is the AF and the Pt layers act as seeding and capping layers, which we will discuss in some more detail later on. The number of parameters influencing the GMR ratio of these sputtered EB-SV’s are immense. Not only the materials themselves can be altered, also the thicknesses of the layers and the growth conditions are of great influence on the GMR. It would be beyond the scope of this thesis to present an all-embracing analysis, readers interested in an extensive review are referred to [62, 33]. However, the effect of the substrate cleaning, sputter pressure, seeding and capping layers and the spacer layer thickness are investigated and the results will be briefly discussed in the next section, focusing on the implication for future STT devices. However, to fully understand our measured results, we will first discuss the general shape of the magnetoresistance curve of an EB-SV. In the following, all measurements are performed at room temperature.

7.1.2 Magnetoresistance curves

The general shape of a magnetoresistance curve is shown in figure 7.1(b). This graph is collected by measuring the resistance of the device, while we sweep the applied field from positive to negative fields and backwards. The corresponding
normalized magnetic moment of the device is shown in figure 7.1(a). We will now go through this loop, explaining the exact shape.

We start with a large positive applied magnetic field, denoted by region (1) in figure 7.1. The top and bottom horizontal arrows indicate the magnetization direction of the fixed and free magnetic layer respectively. In this region, the total magnetization will be positive. Since the magnetization of both layers are parallel aligned, the resistance will be low, as described in section 2.2.2.

When decreasing the magnetic field, the bottom free layer will switch its magnetization at a certain negative applied field. Now, the magnetization direction of the two magnetic layers are antiparallel aligned, resulting in zero magnetization and a higher resistance of the device, as shown in region (2) of figure 7.1.

Eventually, when a large enough negative external field is applied, the fixed magnetic layer will also switch. In this region (3), both magnetizations will be pointing to the left and therefore in our notation negative. Since the magnetization of both layers are parallel aligned, the resistance will be low.

When increasing the applied field again, the magnetization of the fixed layer will switch back to its original state, while the magnetization of the free layer remains unaffected. Here, we enter a new region (4) of antiparallel alignment, where there is no net magnetization and a high resistance and related MR. Further increasing the applied field will bring us back in region (1) again.

Next, we start our analysis of the different growth conditions and materials. Therefore we introduce a reference magnetoresistance curve in figure 7.2(a).
Figure 7.2: Magnetoresistance curves as a function of the applied magnetic field for different growth and material parameters. (a) The reference magnetoresistance curve for a sample consisting of Pt(20) / Co(50) / Cu(23) / Co(50) / IrMn(100) / Pt(20), where the thickness of the layers in Angstroms is denoted between the brackets. (b) The MR curve for the same stack using different substrate cleaning methods. (1) (△) Substrate only cleaned using chemicals. (2) (○) Substrate cleaned using chemicals and vacuum annealed before sputtering. (3) (□) Substrate cleaned using chemicals, vacuum annealing and plasma cleaning before sputtering. (c) MR curves for varying Co sputter conditions. The Argon flow and thereby the sputter pressure is varied from 3 sccm (△), 5 sccm (○) to 7 sccm (□). (d) The MR curves for different seeding and capping materials, where Al is denoted with open triangles (△), Pd is represented by open circles (○) and Pt by open squares (□).
This reference EB-SV structure consists of Pt(20) / Co(50) / Cu(23) / Co(50) / IrMn(100) / Pt(20), where the thickness of the layers (in Ångstroms) is denoted in the brackets. For this sample we used the growth conditions given in table 5.1. The resulting parallel resistance of this structure is 67 Ω and the GMR ratio, as defined in equation 2.13 is 7.4%. We will investigate the effect of the substrate cleaning, sputter pressure, seeding and capping layers and the spacer layer thickness by comparing the results with our reference sample.

### Substrate cleaning

How thorough the substrate is cleaned, determines to a large degree the substrate growth conditions. Although a substrate may optically look clean, small organic and inorganic contaminations may still be present on the surface. Before sputtering the GMR device, we clean our substrates using one of the following sequences:

- Method 1: chemical cleaning, were we put the sample 10 minutes in an ultra-sonic bath using a sequence of ammonia, ethanol and iso-propanol.
- Method 2: chemical cleaning, as described before, followed by vacuum annealing, where we heat the sample to 150 °C for 1 minute at a pressure of $10^{-5}$ mbar.
- Method 3: chemical cleaning, vacuum annealing and followed by plasma cleaning, where the sample is exposed to a 15 W oxygen plasma for 5 minutes.

The MR curves for the reference sample, now using different substrate cleaning methods are shown in figure 7.2(b), where method 1 is denoted by the open triangles (△), method 2 denoted by the open circles (○) and method 3 denoted by the open squares (□).

From figure 7.2(b) we can conclude that the substrate cleaning method has a drastic impact on the GMR ratio, decreasing the MR to only 4.1% when only chemicals are used to clean the substrate. This decrease in MR ratio is caused by a higher roughness of the sputtered films due to the contaminations present on the substrate surface before sputtering. The implication for our STT devices is that the STT effect will decrease significantly if the bottom electrode is not properly cleaned before sputtering into the nanostencil. Therefore, an additional cleaning step, after the undercut is made, and before the deposition takes place, should be included.

### Sputter pressure

Next, we investigated the influence of the Co sputter pressure on the GMR effect. Therefore, the argon flow and thereby the sputter pressure is varied and
the results are shown in figure 7.2(c), where an Ar flow of 3 sccm is denoted by
the open triangles (△), 5 sccm by the open circles (○) and 7 sccm by the open
squares (□). From this figure we can conclude that the GMR ratio increases
with increasing argon flow and thereby sputter pressure. We determined a
maximum GMR ratio when using an Ar flow of 7 sccm, corresponding to a
sputter pressure of $10 \cdot 10^{-3}$ mbar.

This increase in GMR with increasing Co sputter pressure can be explained
in terms of the kinetic energy distribution of the Co atoms arriving at the
sample surface. For low sputter pressure, the atoms arriving at the surface
have a larger kinetic energy compared to using a high pressure. This high ki-
netic energy results in dense, but strongly intermixed layers. At higher sputter
pressures the kinetic energy of the Co atoms is lower, accompanied by lower
intermixing of the layers, therefore a higher interface quality and resulting in a
higher GMR ratio.

The observation of a higher GMR ratio when using a higher sputter pres-
sure, is also confirmed for Py/Cu/Py EB-SV’s, where the optimal Py sputter
pressure was determined in the range of $4 \sim 7 \cdot 10^{-3}$ mbar [62].

To end this section we briefly want to mention a theoretical study on the
deposition process suggesting the use of a two step sputtering process [63] to
obtain deposited layers, resulting in high GMR ratios. In this process, the first
part of a layer is sputtered using a high pressure, resulting in a low intermixing.
Subsequently, the remainder of the layer is sputtered using a higher pressure,
resulting in a denser layer. Although we have not yet fabricated layers using
this approach, it might be useful for further optimizing future STT devices.

### Seeding and capping layers

Furthermore, we want to investigate the influence of the seeding and capping
material on the GMR effect. The purpose of these layers is two-fold. First the
bottom seeding layer serves to protect the device from interdiffusion with the
substrate, whereas the top capping layer serves to prevent oxidation. But the
seed layer also serves to influence the microstructure of the layers deposited
on top, creating, for example, preferential crystalline directions or affecting the
grain size and roughness of the film.

We investigated the influence of these layers by using Pt, Pd and Al as
different materials and the resulting MR curves are shown in figure 7.2(d), where Al
is denoted with open triangles (△), Pd is represented by open circles (○) and
Pt by open squares (□).

From this figure we can conclude that substituting Pt for Pd decreases the
GMR ratio to 6.6%. When using Al as seeding and capping material, the GMR
ratio decreases to only 1.4 %. In the case of Pt, a distinct switching of the free
7.1 Results on CIP-GMR devices

Figure 7.3: GMR as a function of the Cu spacer layer thickness. GMR as a function of the Cu spacer layer thickness. The solid line is a guide to the eye, whereas the dotted line is a fit using equation 7.1 for the Cu thickness ≥ 23 Å. The corresponding GMR versus field curves for a Cu spacer layer of (i) 15, (ii) 23 and (iii) 50 Å are shown in the top panels.

and fixed layer can be seen. The MR curve for Al, however, shows these two regimes merging and the characteristic GMR plateau vanishes. This can be attributed to magnetic coupling between the two ferromagnetic layers. Since for all three graphs the different layer thicknesses are the same, this means that, effectively, the Cu spacer layer must be thinner. This can be caused by a high layer roughness or island growth of Cu on Al.

In our STT devices, we use Pt as the bottom electrode material and thereupon we deposit our STT device. If, however, another material is used as a bottom electrode, the previous analysis shows that this can greatly influence the growth of the device. Therefore, we can conclude that, the effect of first depositing a seeding layer before depositing the rest of the STT device, should be optimized and different seeding materials could be attempted.
Spacer layer thickness

Finally, we investigate the influence of the Cu spacer layer on the CIP-GMR. The dependence of the GMR ratio on both the ferromagnetic and normal metal spacer layer thickness has been studied extensively [64, 65]. Therefore, a series of EB-SV’s with varying Cu layer thickness $t_{Cu}$ are fabricated, with a structure similar to our reference sample: Pt(20) / Co(50) / Cu($t_{Cu}$) / Co(50) / IrMn(100) / Pt(20). For all samples the MR curves are measured and subsequently the GMR ratio is calculated. The results for a Cu thickness varying from 10 to 100 Å are shown in figure 7.3 where the top panels show the corresponding GMR versus field curves for a Cu spacer layer of (i) 15, (ii) 23 and (iii) 50 Å.

From figure 7.3 we can conclude that the GMR ratio increases with increasing Cu layer thickness reaching a maximum value of 7.4% at 23 Å and a further increase in Cu layer thickness results in a decrease in GMR. We will now discuss the general shape of this curve in some more detail.

The dependence of the GMR ratio on both the ferromagnetic and normal metal spacer layer thickness has been studied extensively [66, 64, 65]. For low Cu spacer layer thickness, the GMR increases with Cu thickness. This increase in GMR is caused by magnetic decoupling of the two magnetic layers. If the spacer layer thickness is very small (< 20 Å), the two ferromagnetic layers can no longer be described as two separate magnetic layers, but their magnetizations will act as one magnetic layer, diminishing the GMR effect. To describe the decreasing part of the curve, we will now only introduce a phenomenological equation describing the GMR ratio as a function of the thickness $t_{NM}$ of the non-magnetic spacer [67]:

$$\frac{\Delta R}{R} = A \exp\left(\frac{-t_{NM}}{l_{NM}}\right) \frac{1}{1 + \frac{t_{NM}}{a}}$$ (7.1)

where $A$, and $a$ are phenomenological parameters and $l_{NM}$ is related to the bulk electron mean free paths of the normal metal, as introduced in section 2.2. In this equation the coupling of the magnetic layers is not taken into account for. To obtain the largest GMR ratio possible, the Cu spacer layer should be ideally thin compared to this electron mean free path. As the thickness of the Cu spacer layer increases, the orientation of the magnetization in one of the Co layers only influences the current density in the other Co layer weakly. This term is expressed by the exponential part in equation (7.1) As the spacer thickness increases, the shunting of the current through this layer also increases and thereby decreasing the GMR effect. This is expressed in denominator of equation (7.1).

The dotted line in figure 7.3 shows a fit, for the Cu thickness $\geq$ 23 Å, using equation (7.1). From this fit we determine $A = 14 \pm 1\%$, the theoretical maxi-
7.2 Results on STT devices

We fabricated a series of both 200 and 300 nm nanostencils and we deposited a STT device inside the nanostencil, consisting of the following structure: Cu / Co(t_{Co,1}) / Cu(t_{Cu}) / Co(t_{Co,2}) / Cu. The resulting modeled structure is schematically exhibited in figure 7.4(a).

The bottom Cu layer serves as a seed layer and its thickness is 10 nm. Hereafter, two Co layers separated by a Cu spacer layer are deposited. By choosing \( t_{Co,2} \gg t_{Co,1} \), the first Co layer will, due to a difference in the switching fields, act as the analyzing layer, whereas the second layer is the spin polarizing layer. In our STT devices, we used \( t_{Co,1} = 3 \) nm, \( t_{Cu} = 10 \) nm and \( t_{Co,2} = 12 \) nm, values also often used in literature. Then, finally, the top Cu layer is deposited. This layer is needed to ensure an electrical contact between the device and the top electrode. This Cu layer therefore needs to be thick enough to contact the device, but it must not be so thick that it will interfere with the lift-off process.

Figure 7.4: STT device deposited inside a nanostencil. Figure (a) shows the modeled layered structure of a STT device inside a nanostencil (not to scale), whereas (b) shows an SEM image of an actual device where the poor electrical contact is clearly exposed.
7.2 Results on STT devices

Our STT devices are sputter deposited inside the nanostencils while a 25 mT external applied field is present during sputtering. This field is directed in the sample plane, aligning the magnetization of the Co layers along the applied field and thereby creating a preferential magnetization direction.

After sputtering and subsequent lift-off, to free the bottom electrodes, we are able to perform electrical measurements on the STT device. We can contact our electrodes using small needles, positioned on the sample using micromanipulators while observing the sample through a microscope. For a more detailed description of this technique, please refer to section 2.1.4 of [69]. Then, field-induced magnetoresistance measurements can be performed by measuring the device resistance while changing the applied magnetic field.

However, we were not able to measure the resistance of the majority of our STT devices. The devices we were able to contact, showed a resistance on the order of 20 – 25 Ω. This device resistance is one order of magnitude higher than the 1 – 5 Ω resistance expected from literature values. This might point to a poor electrical contact between the device and the electrodes, which is confirmed by SEM analysis as shown in figure 7.4(b).

Although we have performed magnetoresistance measurements on the STT devices that we were able to contact, no MR is observed when a magnetic field is used to switch the magnetization alignments. The lack of MR and thereby STT can be caused by a number of factors, which we will now briefly discuss.

First, the poor electrical contact might diminish the MR. Since the measured resistance is one order of magnitude higher than expected, the MR ratio, if present, would be one order of magnitude smaller and thereby maybe unmeasurably small for the setup we used. Nevertheless, the issue of poor electrical contact could be easily addressed by performing an analysis, whereby the top Cu layer thickness is increased and the device resistance is measured. Once the device resistance is on the order of 1 – 5 Ω, a decent electrical contact is assured.

Although the higher than expected resistance might be caused by a poor electrical contact, growth related non-uniformities within the deposited STT structure can also result in a higher resistance. The measurements are performed on nanostencils created without the additional cleaning step as suggested in the previous section. Incorporating this cleaning step before the deposition of the STT device and further optimization of the growth parameters is therefore suggested for future devices.

Another, more likely, cause of absence of MR could be the fact that, until now, we have produced circular nanostencils. However, in literature it is shown that an ellipse shaped STT device is preferred to ensure alignment of the magnetization with the semimajor axis through shape anisotropy. Therefore, we have started the fabrication of ellipse shaped nanostencils. Currently, the criti-
cal point in this process is the creation of the ellipse shaped opening in the top electrode. At this point, we are not yet able to produce the desired opening. Further optimization of the milled pattern is therefore needed. Although at the time of writing this thesis, no ellipse shaped nanostencils have been fabricated, there are no additional difficulties expected for the growth inside these nanostencils as already briefly discussed in §6.2.3.

We will end this chapter with the conclusion that we are now able to create nanostencils, subsequently deposit structures inside and thereafter perform resistance measurements on the created device. Albeit the devices deposited so far did not show any MR or STT, further optimization of the growth, combined with the transition to ellipse shaped nanostencils will provide a promising outlook for future research.

\footnote{The software of our FIB (FEI Nova Nanolab 600i) is not capable of milling other patterns than lines, circles and rectangles. However, additional patterns are possible with the newly installed Raith EBL setup. Since patterning using the Raith software produces slightly different structures than the FEI software, the optimization, as described in section §5.4 has to be repeated.}
Chapter 8

Conclusions and Outlook

8.1 Conclusions

The goal of this master thesis work, as stated in section 1.4, was to answer the question:

*What is spin-transfer-torque and how can we design a device exploiting and thereby analyzing this new effect?*

This question is addressed by first deriving what spin-transfer-torque is, subsequently determining the features needed to exploit this effect experimentally and thereupon designing a process to produce STT devices. We will now summarize the conclusions made in the main chapters of this thesis.

*Chapter 2 Spin transfer torque: A theoretical description.*

- We showed how to explain the spin-transfer-torque effect by using the concept of spin accumulation.
- We presented a formal derivation of the spin transfer torque term, proving that a net change in the spin current after passing through a normal metal–ferromagnet interface is the cause of the STT effect.
- We derived that a STT device should consist of a spin polarizing layer and a spin analyzing layer separated by a spacer layer and contacted with electrodes to the outer world.
- We derived that the critical currents needed to exert a STT and switching the magnetic layer require devices of small cross-sectional area.

*Chapter 3 Spin transfer torque devices.*

- In this chapter we presented a number of different STT device geometries and showed the nanostencil process in somewhat more detail.
8.1 Conclusions

- We discussed that the main advantages of the nanostencil process are its versatility and scalability, allowing the creation of a variety of nanostencil shapes, sizes and configurations.
- We derived the critical factors in the nanostencil process which can be summarized by the parameters influencing the growth of the nanostencil itself and the control over the depositions inside the nanostencil.

Chapter 5: Creating a nanostencil.

- In this chapter we discussed our choice for Pt as the electrode material and SiO$_2$ as insulating spacer layer and we determined the optimal growth conditions for the materials.
- We showed that we can pattern the electrodes and insulating layer into the desired large contact pads and small connecting leads.
- Using a newly developed EDX linescan measuring sequence, we determined the milling time needed to open the top electrode.
- We showed that we are able to produce nanostencils with the desired opening size and undercut.

Chapter 6: Growth inside a nanostencil.

- In this chapter we showed that we are able, using our adapted nanostencil process, to deposit material only inside the nanostencil by using sputtering and subsequent lift-off techniques.
- We used a similar EDX linescan measuring sequence as in the previous chapter to determine the amount of material deposited inside the nanostencil.
- A model, quantitatively describing the growth inside a nanostencil, has been developed and the modeled results are in agreement with the measured thickness.
- Using the model, we are able to simulate the growth inside nanostencils of different geometry and even using different deposition methods.

Chapter 7: Transport measurements.

- In this chapter we showed that, even within the limited timeframe of this master project, we were able to perform a first preliminary transport measurement on a fully integrated nanostencil STT device.
- We attributed the lack of measured MR mainly on the fact that we need to fabricate ellipse shaped nanostencils.
- Furthermore, we showed the influence of substrate cleaning, sputter pressure and seeding layers for the GMR effect. From this we concluded that an additional cleaning step and further optimization of the growth inside a nanostencil may be necessary.
To summarize, we have been successful in the fabrication of the desired nanostencils and we succeeded in depositing, imaging and modeling structures grown inside the nanostencil. Finally, we were able to perform a first preliminary transport measurement on a fully integrated nanostencil STT device. Although, further optimization is needed, the nanostencil process is most definitely a promising method to fabricate future STT devices. In the final section of this thesis we will provide a brief outlook for future research as well as describing currently addressed issues.

8.2 Outlook

During the time of the writing of this thesis, a new master student, Paul Soto Rodriguez, continued the research on STT devices using the nanostencil process. The issues, as described in this thesis, we are currently addressing are:

Currently addressed issues

- Optimizing the fabrication of ellipse shaped nanostencils, by creating a method to control the milling of the ellipse shaped nanostencil opening. We are aiming for minimal influence of redeposition during milling and better defined nanostencil edges.
- Enhancing the electrical contact of the device with the top electrode, by optimizing the top Cu layer thickness.
- Incorporating the additional cleaning step before the deposition of the STT structure and thereby improving the growth inside the nanostencil.

Proposed further optimization

- Optimizing the thickness of the ferromagnetic and spacer layers, to increase the MR ratio.
- Investigating the effect of the use of different ferromagnetic materials on the MR ratio and the current-induced switching.
- Investigating the effect, both on the STT structure as well as on the STT effect, when using an evaporation technique to deposit the ferromagnetic and spacer layers.

This is only a brief summary of the seemingly endless list of possible research opportunities. Although we used the nanostencils to create STT devices within, nanostencils can be used in a broader perspective than the research presented in this thesis. Therefore, we would like to end this thesis by providing the reader two promising future research opportunities using nanostencils.
8.2 Outlook

Figure 8.1: Outlook for future research using nanostencils. (a) Exploiting the nanostencil process for the research on phase-locking in nano oscillators, where two STT devices are created in close proximity. (b) The nanostencil process can be implemented for the fabrication of lateral devices, by depositing the different layers under multiple angles.

Future research using nanostencils

○ Besides switching the magnetization of a device, using a current, through the STT effect, it is also possible to force the magnetization into a (stable) processional state. This effect, exceptionally interesting from a fundamental point of view, also enables the creation of new future spintronic devices. In these devices, so-called nano-oscillators, an DC current will result in a AC voltage, where the frequency is in the GHz regime[70]. Nanostencils could be used to perform research on the so-called phase-locking phenomena, where STT devices grown inside the nanostencils are able to "magnetically communicate" with each other [71]. Therefore it is necessary to create two, or even multiple, devices in close proximity, or even in the same nanostencil, possibly sharing mutual layers, as is schematically shown in figure 8.1(a).

○ Another promising opportunity for the exploitation of the nanostencil process is in the creation of lateral structures. Lateral structures are often preferred in devices where it is crucial to avoid intermixing of the different layers by the deposition process. An example is the creation of a organic spin-valve structure, where an organic material is used as the spacer layer in a spin-valve device [72]. Creating lateral devices is possible using the nanostencil process, while deposition under multiple angles as shown in figure 8.1(b). Using nanostencils, it is possible to create structures with materials, like most organic materials, normally not allowed to be patterned using subtractive processes.
Appendix A

Formal derivation of spin transfer torque

This chapter is made up out of three parts. In the first part, the formal definition of the spin current density and the spin torque term will be derived. Using these definitions, the second part will give a derivation of the spin torque by considering the net change in the spin current before and after passing through a layer of ferromagnetic material. The last part will use a Stoner model to describe the physics that take place near the metal–ferromagnet interface.

A.1 Definition of spin current density and spin transfer torque

Before we describe the equations of spin transport, which will lead to the spin torque term, it is instructive to first show the quantum mechanic equations of particle transport. For a more formal derivation of these equations, please refer to e.g. [37] or [38].

Looking at an electron with wavefunction $\psi$, the particle equations we want to introduce are the particle density $n$:

$$n = \psi^* \psi$$

(A.1)

and the particle current density $\vec{j}$:

$$\vec{j} = \text{Re} (\psi^* \hat{v} \psi),$$

(A.2)

where $\hat{v}$ is the velocity operator given by:

$$\hat{v} = -\frac{i\hbar}{m} \vec{\nabla}.$$  

(A.3)

106
A.1 Definition of spin current density and spin transfer torque

Using conservation of particle number, we can write the continuity equation as:

\[ \frac{\partial n}{\partial t} + \vec{\nabla} \cdot \vec{j} = 0. \] (A.4)

Because we are interested in the steady state solution where there are no sources or sinks, \( \frac{\partial n}{\partial t} = 0 \) and therefore \( \vec{\nabla} \cdot \vec{j} = 0 \).

Now looking at the spin of the electron, the analog for the number density, equation A.1 is the spin density \( \vec{m} \):

\[ \vec{m} = \psi^* \vec{s} \psi, \] (A.5)

which is a vector in spin space and where \( \vec{s} \) is:

\[ \vec{s} = \frac{\hbar}{2} \vec{\sigma}, \] (A.6)

where \( \vec{\sigma} = (\sigma_x \ \sigma_y \ \sigma_z) \) are the Pauli matrices, in Cartesian coordinates, given by:

\[ \sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \] (A.7)

\[ \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \] (A.8)

\[ \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \] (A.9)

Instead of a number current density, equation A.2 we now have the spin current density, which has both a direction in the spin-space and real space and is therefore given by the tensor \( Q \):

\[ \vec{Q} = \text{Re} \left( \psi^* \vec{s} \otimes \hat{v} \psi \right), \] (A.10)

where \( \vec{s} \otimes \hat{v} \) stands for the dyadic product\(^1\) of the Pauli matrices and the velocity operator. Using equations A.3 and A.6 we can write the spin current density as:

\[ \begin{align*}
\vec{Q} &= \text{Re} \left( \psi^* \frac{\hbar}{2} \vec{\sigma} \otimes \left( -\frac{i\hbar}{m} \hat{\nabla} \psi \right) \right) \\
&= \frac{\hbar^2}{2m} \text{Im} \left( \psi^* \vec{\sigma} \otimes \hat{\nabla} \psi \right) 
\end{align*} \] (A.11)

The analog for the continuity equation A.4 in the case of the spin current is given by [39]:

\[ \frac{\partial \vec{m}}{\partial t} + \vec{\nabla} \cdot \vec{Q} = -\frac{\delta \vec{m}}{\tau_s} - \vec{n}_{ext}, \] (A.12)

\(^1\)Since \( \vec{s} \) and \( \hat{v} \) are both vectors of the same length, the tensor product in this case is a dyadic product.
A.2 Spin torque as a result of spin filtering

Figure A.1: Deriving the spin torque term. (a) The spin torque is calculated from integration of the net spin current density normal to the surface. (b) The spin filter properties of a ferromagnetic layer are described by spin dependant transmission and reflection coefficients. The spin torque term is calculated using the difference in incoming and outgoing spin currents. (c) An electron with an arbitrary spin state can be described by a coherent superposition of the spin-up $|\uparrow\rangle$ and spin-down $|\downarrow\rangle$ states.

where $\delta\vec{m}$ is the spin accumulation, as described in section 2.3.2, $\tau_{sf}$ is the spin relaxation time, described in equation 2.5, and $\vec{n}_{ext}$ is the external torque density [36]. In the steady state case, $\frac{\partial}{\partial t} = 0$, where there is no spin flip, $\tau_{sf} = \infty$, the continuity equation 2.22 states that the net flow of spins into a region plus the torques acting on that region must be zero:

$$\vec{n}_{ext} = - \nabla \cdot \vec{Q}. \tag{A.13}$$

If we only take into account for the current induced torques, $\vec{n}_{ext} = \vec{n}_{st}$, and we integrate the continuity equation over a certain volume of ferromagnetic material, we find that we can write the spin torque term $\vec{\tau}$ as:

$$\iiint \vec{n}_{st} d^3r = - \iiint \nabla \cdot \vec{Q} d^3r$$

$$\vec{\tau}_{st} = - \int \vec{n} \cdot \vec{Q} d^2r, \tag{A.14}$$

where $\vec{n}$ is the unit normal vector to the surface of the volume.

A.2 Spin torque as a result of spin filtering

In this section we will derive an expression for the spin transfer torque term $\vec{\tau}_{st}$ by first considering the net change in the spin current before and after passing through a layer of ferromagnetic material, using the geometry shown in figure A.1(a). This non-equilibrium spin current through the volume of the ferromagnetic layer can be calculated by integrating the spin current density normal to the surface of this volume. By conservation of angular momentum, this non-equilibrium spin current is then compensated by a spin transfer torque of the
electrons acting on the magnetization of the ferromagnetic layer.

We will start of the derivation by determining the wavefunction $\psi$ of an electron with an arbitrary spin state. This wavefunction can be described by a coherent superposition of the spin up $|\uparrow\rangle$ and spin down $|\downarrow\rangle$ states and for this spinor, the wavefunction of a plane wave, with wavevector $k$ and traveling in the $x$ direction, is given by:

$$\psi = \frac{e^{ikx}}{\sqrt{\Omega}} (a |\uparrow\rangle + b |\downarrow\rangle) = \frac{e^{ikx}}{\sqrt{\Omega}} \begin{pmatrix} a \\ b \end{pmatrix},$$  \hspace{1cm} (A.15)

where $|a|^2$ and $|b|^2$ are the spin up and spin down probabilities respectively and $\Omega$ is a normalization volume. Using equation (A.15) the gradient $\nabla \psi$ and the complex conjugate $\psi^*$ of the electron wavefunction are given by:

$$\nabla \psi = ik \frac{e^{ikx}}{\sqrt{\Omega}} \begin{pmatrix} a \\ b \end{pmatrix} \hat{x}$$ \hspace{1cm} (A.16)

and

$$\psi^* = \frac{e^{-ikx}}{\sqrt{\Omega}} \begin{pmatrix} a^* \\ b^* \end{pmatrix}.$$ \hspace{1cm} (A.17)

To calculate the spin current density, first the dyadic product $\vec{\sigma} \otimes \nabla \psi$ in equation (A.11) must be determined:

$$\vec{\sigma} \otimes \nabla \psi = \begin{pmatrix} \sigma_x \frac{\partial \psi}{\partial x} & \sigma_x \frac{\partial \psi}{\partial y} & \sigma_x \frac{\partial \psi}{\partial z} \\ \sigma_y \frac{\partial \psi}{\partial x} & \sigma_y \frac{\partial \psi}{\partial y} & \sigma_y \frac{\partial \psi}{\partial z} \\ \sigma_z \frac{\partial \psi}{\partial x} & \sigma_z \frac{\partial \psi}{\partial y} & \sigma_z \frac{\partial \psi}{\partial z} \end{pmatrix}.$$ \hspace{1cm} (A.18)

Multiplying this with the complex conjugated wavefunction $\psi^*$ yields:

$$\psi^* \vec{\sigma} \otimes \nabla \psi = \begin{pmatrix} \psi^* \sigma_x \frac{\partial \psi}{\partial x} & \psi^* \sigma_x \frac{\partial \psi}{\partial y} & \psi^* \sigma_x \frac{\partial \psi}{\partial z} \\ \psi^* \sigma_y \frac{\partial \psi}{\partial x} & \psi^* \sigma_y \frac{\partial \psi}{\partial y} & \psi^* \sigma_y \frac{\partial \psi}{\partial z} \\ \psi^* \sigma_z \frac{\partial \psi}{\partial x} & \psi^* \sigma_z \frac{\partial \psi}{\partial y} & \psi^* \sigma_z \frac{\partial \psi}{\partial z} \end{pmatrix}.$$ \hspace{1cm} (A.19)

and the spin current density tensor can be written as:

$$\bar{Q} = \frac{\hbar^2}{2m} \text{Im} \begin{pmatrix} \psi^* \sigma_x \frac{\partial \psi}{\partial x} & \psi^* \sigma_x \frac{\partial \psi}{\partial y} & \psi^* \sigma_x \frac{\partial \psi}{\partial z} \\ \psi^* \sigma_y \frac{\partial \psi}{\partial x} & \psi^* \sigma_y \frac{\partial \psi}{\partial y} & \psi^* \sigma_y \frac{\partial \psi}{\partial z} \\ \psi^* \sigma_z \frac{\partial \psi}{\partial x} & \psi^* \sigma_z \frac{\partial \psi}{\partial y} & \psi^* \sigma_z \frac{\partial \psi}{\partial z} \end{pmatrix}.$$ \hspace{1cm} (A.20)

Using equation (A.16) we can conclude that the spin current density tensor, in this case, only has non-zero values for the $x$ spatial direction and therefore we can write it as:

$$\bar{Q} = \bar{Q}_x.$$ \hspace{1cm} (A.21)

\footnote{\frac{\partial \psi}{\partial y} = \frac{\partial \psi}{\partial z} = 0 \text{ and } \frac{\partial \psi}{\partial x} \neq 0}$
Substituting equations A.7 - A.9, A.16 and A.17, the spin current density in the $x$ direction is given by:

$$\vec{Q}_x = \frac{\hbar^2}{2m} \text{Im} \left( \begin{array}{c} Q_{xx} \\ Q_{xy} \\ Q_{xz} \end{array} \right),$$

where we can write $Q_{xx}$, $Q_{xy}$ and $Q_{xz}$ as:

$$Q_{xx} = \psi^* \sigma_x \frac{\partial \psi}{\partial x} = \frac{e^{-ikx}}{\sqrt{\Omega}} \begin{pmatrix} a^* & b^* \end{pmatrix} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} ik \frac{e^{ikx}}{\sqrt{\Omega}} \begin{pmatrix} a \\ b \end{pmatrix} = ik \frac{1}{\Omega} (a^*b + ab^*)$$

$$Q_{xy} = \psi^* \sigma_y \frac{\partial \psi}{\partial x} = \frac{e^{-ikx}}{\sqrt{\Omega}} \begin{pmatrix} a^* & b^* \end{pmatrix} \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} ik \frac{e^{ikx}}{\sqrt{\Omega}} \begin{pmatrix} a \\ b \end{pmatrix} = ik \frac{1}{\Omega} (-ia^*b + iab^*)$$

$$Q_{xz} = \psi^* \sigma_z \frac{\partial \psi}{\partial x} = \frac{e^{-ikx}}{\sqrt{\Omega}} \begin{pmatrix} a^* & b^* \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} ik \frac{e^{ikx}}{\sqrt{\Omega}} \begin{pmatrix} a \\ b \end{pmatrix} = ik \frac{1}{\Omega} (a^*a - b^*b)$$

and $\vec{Q}_x$ becomes:

$$\vec{Q}_x = \frac{\hbar^2}{2m} \frac{k}{\Omega} \begin{pmatrix} a^*b + ab^* \\ -ia^*b + iab^* \\ a^*a - b^*b \end{pmatrix}$$

This can be simplified\(^3\) to:

$$\vec{Q}_x = \frac{\hbar^2}{2m} \frac{k}{\Omega} \begin{pmatrix} 2 \text{Re}(ab^*) \\ 2 \text{Im}(ab^*) \\ |a|^2 - |b|^2 \end{pmatrix}$$

\(^3\)Using the following complex relations:

$$a = k + li$$
$$a^* = k - li$$
$$b = u + vi$$
$$b^* = u - vi$$

$$a^*b + ab^* = (k - li) (u + vi) + (k + li) (u - vi) = 2 (ku + lv)$$
$$-ia^*b + iab^* = -i (k - li) (u + vi) + i (k + li) (u - vi) = 2 (kv - lu)$$
$$ab^* = (k + li) (u - vi) = (ku + lv) - (kv - lu) i$$

$$\text{Re}(ab^*) = (ku + lv) = \frac{1}{2} (a^*b + ab^*),$$
$$\text{Im}(ab^*) = (kv - lu) = \frac{1}{2} (-ia^*b + iab^*)$$
To describe an electron with a non-collinear spin state, we will take the general wavefunction of equation [A.15] and by applying an Euler rotation we will find the desired spinor wavefunction. This rotation can be described by the rotation matrix $R$:

$$R(\alpha\beta\gamma) = e^{-\frac{i}{2}i\alpha\sigma_z}e^{-\frac{i}{2}i\beta\sigma_y}e^{-\frac{i}{2}i\gamma\sigma_z},$$  \hspace{1cm} (A.28)

where $\alpha$, $\beta$ and $\gamma$ are Euler angles and $\sigma_y$ and $\sigma_z$ the Pauli matrices given by equations [A.8] and [A.9] respectively. We now start with a spin up state and rotate the spin over an angle $\theta$ in the $x$–$z$ plane. This rotation is governed by $\beta$ thus we will apply the following rotation matrix:

$$R(\beta) = e^{-\frac{1}{2}i\beta\sigma_y}. \hspace{1cm} (A.29)$$

Recalling that:

$$\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \hspace{1cm} (A.30)$$

and considering a rotation over an angle $\theta$ we obtain the following expression:

$$R = e \begin{pmatrix} 0 & -\frac{\theta}{2} \\ \frac{\theta}{2} & 0 \end{pmatrix}. \hspace{1cm} (A.31)$$

In general, we can write a matrix exponential as:

$$e^A = I + A + \frac{1}{2}A^2 + \frac{1}{3!}A^3 + \ldots, \hspace{1cm} (A.32)$$

which in our case results in:

$$R = \begin{pmatrix} \cos \frac{\theta}{2} & -\sin \frac{\theta}{2} \\ \sin \frac{\theta}{2} & \cos \frac{\theta}{2} \end{pmatrix} \hspace{1cm} (A.34)$$

The expansion of:

$$e^{\begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix}} = I + \begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix} + \frac{1}{2} \begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix} \left( \begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix} \right) + \ldots \hspace{1cm} (A.33)$$

where the identity matrix is:

$$I = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$$

Calculating the matrix terms individually:

$$\begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix} \begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix} = \begin{pmatrix} -a^2 & 0 \\ 0 & -a^2 \end{pmatrix}$$

$$\begin{pmatrix} -a^2 & 0 \\ 0 & -a^2 \end{pmatrix} \begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix} = \begin{pmatrix} 0 & a^3 \\ -a^3 & 0 \end{pmatrix}$$
A.2 Spin torque as a result of spin filtering

If we now start with a spin up state and rotate the spin over an angle $\theta$ in the $x$–$z$ plane, this results in the spinor:

$$\begin{pmatrix} a \\ b \end{pmatrix} = R \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \begin{pmatrix} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{pmatrix}$$  \hspace{1cm} (A.35)

Using equation A.15, the wavefunction of the incoming electron with wavevector $k$ traveling in the $x$ direction can therefore be described by:

$$\psi_{\text{in}} = e^{ikx} \begin{pmatrix} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{pmatrix}.$$  \hspace{1cm} (A.36)

The transmitted $\psi_{\text{tr}}$ and reflected $\psi_{\text{re}}$ wavefunctions are calculated using the spin dependent transmission $\vec{t}$ and reflection $\vec{r}$ coefficients [40]:

$$\vec{t} = \begin{pmatrix} t_1 \\ t_1 \end{pmatrix}$$  \hspace{1cm} (A.37)

$$\vec{r} = \begin{pmatrix} r_1 \\ r_1 \end{pmatrix}$$  \hspace{1cm} (A.38)

and are given by:

$$\psi_{\text{tr}} = e^{ikx} \begin{pmatrix} t_1 \cos \frac{\theta}{2} \\ t_1 \sin \frac{\theta}{2} \end{pmatrix}$$  \hspace{1cm} (A.39)

$$\psi_{\text{re}} = e^{-ikx} \begin{pmatrix} r_1 \cos \frac{\theta}{2} \\ r_1 \sin \frac{\theta}{2} \end{pmatrix},$$  \hspace{1cm} (A.40)

where the minus sign in front of the wavevector is due to the fact that the reflected wave propagates in the opposite direction. To derive the spin current density $\vec{Q}_x$ in the $x$ spatial direction we make use of equation A.27 which results

$$\begin{pmatrix} 0 & a^3 \\ -a^3 & 0 \end{pmatrix} \begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix} = \begin{pmatrix} a^4 & 0 \\ 0 & a^4 \end{pmatrix}$$

we get that:

$$e^{\begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix}} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix} + \frac{1}{2} \begin{pmatrix} -a^2 & 0 \\ 0 & -a^2 \end{pmatrix} + \frac{1}{3!} \begin{pmatrix} 0 & a^3 \\ -a^3 & 0 \end{pmatrix} + \frac{1}{4!} \begin{pmatrix} a^4 & 0 \\ 0 & a^4 \end{pmatrix} + \ldots$$

in this way:

$$e^{\begin{pmatrix} 0 & -a \\ a & 0 \end{pmatrix}} = \begin{pmatrix} 1 - \frac{1}{2}a^2 + \frac{1}{4!}a^4 + \ldots & -a + \frac{1}{2}a^3 - \ldots \\ -a + \frac{1}{2}a^3 - \ldots & 1 - \frac{1}{2}a^2 + \frac{1}{4!}a^4 + \ldots \end{pmatrix} = \begin{pmatrix} \cos(a) & -\sin(a) \\ \sin(a) & \cos(a) \end{pmatrix}$$

and in our case we have $a = \frac{\theta}{2}$.
A.2 Spin torque as a result of spin filtering

\[ \vec{Q}_{x,\text{in}} = \frac{\hbar^2 k}{2m\Omega} \begin{pmatrix} 2 \text{Re} (ab^*) \\ 2 \text{Im} (ab^*) \end{pmatrix} = \frac{\hbar^2 k}{2m\Omega} \begin{pmatrix} 2 \text{Re} \left( \cos \frac{\theta}{2} \sin \frac{\theta}{2} \right) \\ 2 \text{Im} \left( \cos \frac{\theta}{2} \sin \frac{\theta}{2} \right) \end{pmatrix} \]

\[ = \frac{\hbar^2 k}{2m\Omega} \begin{pmatrix} \sin \theta \\ 0 \end{pmatrix}, \quad (A.41) \]

for the incoming spin current density,

\[ \vec{Q}_{x,\text{tr}} = \frac{\hbar^2 k}{2m\Omega} \begin{pmatrix} 2 \text{Re} \left( t_\uparrow \cos \frac{\theta}{2} t_\downarrow^* \sin \frac{\theta}{2} \right) \\ 2 \text{Im} \left( t_\uparrow \cos \frac{\theta}{2} t_\downarrow^* \sin \frac{\theta}{2} \right) \end{pmatrix} \]

\[ = \frac{\hbar^2 k}{2m\Omega} \begin{pmatrix} \sin \theta \text{Re} \left( t_\uparrow t_\downarrow^* \right) \\ \sin \theta \text{Im} \left( t_\uparrow t_\downarrow^* \right) \end{pmatrix}, \quad (A.42) \]

for the transmitted spin current density and

\[ \vec{Q}_{x,\text{re}} = -\frac{\hbar^2 k}{2m\Omega} \begin{pmatrix} 2 \text{Re} \left( r_\uparrow \cos \frac{\theta}{2} r_\downarrow^* \sin \frac{\theta}{2} \right) \\ 2 \text{Im} \left( r_\uparrow \cos \frac{\theta}{2} r_\downarrow^* \sin \frac{\theta}{2} \right) \end{pmatrix} \]

\[ = -\frac{\hbar^2 k}{2m\Omega} \begin{pmatrix} \sin \theta \text{Re} \left( r_\uparrow r_\downarrow^* \right) \\ \sin \theta \text{Im} \left( r_\uparrow r_\downarrow^* \right) \end{pmatrix}, \quad (A.43) \]

for the reflected spin current density. The spin transfer torque \( \vec{\tau}_{st} \) on an area \( A \) of the ferromagnetic layer is equal to the net spin current, \( \vec{Q}_{x,\text{in}} + \vec{Q}_{x,\text{re}} - \vec{Q}_{x,\text{tr}} \), transferred from the electron to the ferromagnet. Using equation \( A.14 \), \( \vec{\tau}_{st} \) can be written as:

\[ \vec{\tau}_{st} = -\int \hat{n} \cdot \vec{Q} \, d^2r = A \hat{x} \cdot \left( \vec{Q}_{x,\text{in}} + \vec{Q}_{x,\text{re}} - \vec{Q}_{x,\text{tr}} \right) \]

\[ = A \left( \vec{Q}_{x,\text{in}} + \vec{Q}_{x,\text{re}} - \vec{Q}_{x,\text{tr}} \right). \quad (A.44) \]

---

Using the trigonometric relations:

\[ 2 \sin x \cos x = \sin 2x \]
\[ \cos^2 x = \frac{1}{2} \left( 1 + \cos 2x \right) \]
\[ \sin^2 x = \frac{1}{2} \left( 1 - \cos 2x \right) \]
Substituting equations A.41 – A.43 yields:

\[
\vec{\tau}_{st} = A \frac{\hbar^2 k}{2 m \Omega} \begin{pmatrix}
\sin \theta & 0 \\
0 & \cos \theta
\end{pmatrix} - A \frac{\hbar^2 k}{2 m \Omega} \begin{pmatrix}
\sin \theta \text{Re} \left( r_\uparrow r_\uparrow^* \right) \\
\sin \theta \text{Im} \left( r_\uparrow r_\uparrow^* \right)
\end{pmatrix} |r_\uparrow| \left[ \cos^2 \theta \frac{\theta}{2} - |r_\uparrow| \sin^2 \frac{\theta}{2} \right]^{-1}
\]

\[
\vec{\tau}_{st} = A \frac{\hbar^2 k}{2 m \Omega} \begin{pmatrix}
\sin \theta \text{Re} \left( t_\uparrow t_\uparrow^* \right) \\
\sin \theta \text{Im} \left( t_\uparrow t_\uparrow^* \right)
\end{pmatrix} |t_\uparrow| \left[ \cos^2 \theta \frac{\theta}{2} - |t_\uparrow| \sin^2 \frac{\theta}{2} \right]^{-1},
\]

which turns into:

\[
\vec{\tau}_{st} = A \frac{\hbar^2 k}{\Omega 2 m} \begin{pmatrix}
\sin \theta \left[ 1 - \text{Re} \left( r_\uparrow r_\uparrow^* + t_\uparrow t_\uparrow^* \right) \right] \\
\sin \theta \left[ \text{Im} \left( r_\uparrow r_\uparrow^* + t_\uparrow t_\uparrow^* \right) \right]
\end{pmatrix} \cos \theta - \left( |r_\uparrow|^2 + |t_\uparrow|^2 \right) \cos^2 \frac{\theta}{2} + \left( |r_\uparrow|^2 + |t_\uparrow|^2 \right) \sin^2 \frac{\theta}{2}.
\]

Using \(|r_\uparrow|^2 + |t_\uparrow|^2 = 1\) and \(|r_\downarrow|^2 + |t_\downarrow|^2 = 1\), this simplifies to:

\[
\vec{\tau}_{st} = A \frac{\hbar^2 k}{\Omega 2 m} \begin{pmatrix}
\sin \theta \left[ 1 - \text{Re} \left( r_\uparrow r_\uparrow^* + t_\uparrow t_\uparrow^* \right) \right] \\
- \sin \theta \left[ \text{Im} \left( r_\uparrow r_\uparrow^* + t_\uparrow t_\uparrow^* \right) \right]
\end{pmatrix}.
\]

In the case of perfect spin filtering, where only spin up electrons are transmitted \((t_\uparrow = 1, t_\downarrow = 0)\) and all spin down electrons are reflected \((r_\uparrow = 0, r_\downarrow = 1)\), equation \(A.45\) turns into:

\[
\vec{\tau}_{st} = A \frac{\hbar^2 k}{\Omega 2 m} \sin \theta \hat{x}
\]

### A.3 Spin torque near the metal–ferromagnet interface

In this section we will look at the spin torque term near the interface between the normal metal and ferromagnetic layer. Assuming an incoming spin polarized electron, with a wavefunction described by equation \(A.36\), we now model the ferromagnetic layer using a Stoner model as shown in figure \(A.2\). Due to the exchange interaction, the energies of the spin-up and spin-down electrons are different inside the ferromagnetic layer. The spin down (minority) electrons are higher in energy than the spin up (majority) electrons and the difference in energy is the exchange energy \(E_{ex}\). The system is modeled by a potential energy step, at a position \(x = 0\). The incoming electron has a free-electron dispersion relation and its energy is therefore given by:

\[
E = \frac{\hbar^2 k^2}{2m} = E_F.
\]
A.3 Spin torque near the metal–ferromagnet interface

Figure A.2: Model used for deriving the spin torque near the normal metal–ferromagnet interface. Using a Stoner model approach, the normal metal – ferromagnet interface is modeled and the spin torque is calculated from the absorption of the transverse spin current near the interface.

The physics near the metal–ferromagnet interface can now be described as a quantum mechanical scattering problem, where the incoming, transmitted and reflected spinor wavefunctions are given by:

\[
\psi_{\text{in}} = e^{ikx} \left( \frac{a_\uparrow \cos \theta}{a_\uparrow} \right) \left( \frac{a_\downarrow \sin \theta}{a_\downarrow} \right) \quad (A.48)
\]

\[
\psi_{\text{tr}} = \left( e^{ik_\uparrow x} \frac{t_\uparrow}{t_\uparrow} \cos \frac{\theta}{2} \right) \left( e^{ik_\downarrow x} \frac{t_\downarrow}{t_\downarrow} \sin \frac{\theta}{2} \right) \quad (A.49)
\]

\[
\psi_{\text{re}} = e^{-ikx} \left( \frac{r_\uparrow \cos \theta}{r_\uparrow} \right) \left( \frac{r_\downarrow \sin \theta}{r_\downarrow} \right) \quad (A.50)
\]

where the wavevectors inside the normal metal \( k \) and for the spin up \( k_\uparrow \) and spin down \( k_\downarrow \) state in the ferromagnet are given by:

\[
k = k_F \quad (A.51)
\]

\[
k_\uparrow = \frac{\sqrt{2mE}}{\hbar} \quad (A.52)
\]

\[
k_\downarrow = \frac{\sqrt{2m(E - E_{\text{ex}})}}{\hbar} \quad (A.53)
\]

Just as in the previous section, the spin torque term can be calculated by determining the net spin current density in the \( x \) spatial direction. For this we first need to derive expressions for the incoming, transmitted and reflected wavefunctions. These can be derived by matching the wavefunctions and their derivatives at the interface between the metal and ferromagnetic layer:

\[
\psi_{\text{in}}|_{x=0} + \psi_{\text{re}}|_{x=0} = \psi_{\text{tr}}|_{x=0} \quad (A.54)
\]

\[
\frac{\partial \psi_{\text{in}}}{\partial x} \bigg|_{x=0} + \frac{\partial \psi_{\text{re}}}{\partial x} \bigg|_{x=0} = \frac{\partial \psi_{\text{tr}}}{\partial x} \bigg|_{x=0}. \quad (A.55)
\]
A.3 Spin torque near the metal–ferromagnet interface

Solving these equations gives:

\[ e^{ikx} \begin{pmatrix} a_↑ \cos \frac{\theta}{2} \\ a_↓ \sin \frac{\theta}{2} \end{pmatrix} \bigg|_{x=0} + e^{-ikx} \begin{pmatrix} r_↑ \cos \frac{\theta}{2} \\ r_↓ \sin \frac{\theta}{2} \end{pmatrix} \bigg|_{x=0} = \begin{pmatrix} t_↑ e^{ik_1 x} \cos \frac{\theta}{2} \\ t_↓ e^{ik_1 x} \sin \frac{\theta}{2} \end{pmatrix} \bigg|_{x=0} \]

and

\[ ik e^{ikx} \begin{pmatrix} a_↑ \cos \frac{\theta}{2} \\ a_↓ \sin \frac{\theta}{2} \end{pmatrix} \bigg|_{x=0} - ik e^{-ikx} \begin{pmatrix} r_↑ \cos \frac{\theta}{2} \\ r_↓ \sin \frac{\theta}{2} \end{pmatrix} \bigg|_{x=0} = \begin{pmatrix} ik_1 t_↑ e^{ik_1 x} \cos \frac{\theta}{2} \\ ik_1 t_↓ e^{ik_1 x} \sin \frac{\theta}{2} \end{pmatrix} \bigg|_{x=0} \]

At the interface, \( x = 0 \), we can derive the following relations:

\[ a_↑ + r_↑ = t_↑ \] \hspace{1cm} (A.56)
\[ a_↓ + r_↓ = t_↓ \] \hspace{1cm} (A.57)

and

\[ ika_↑ - ikr_↓ = ik_1 t_↑ \] \hspace{1cm} (A.58)
\[ ika_↓ - ikr_↑ = ik_1 t_↓ \] \hspace{1cm} (A.59)

Solving these equations and using the same normalization constant as in the previous section yields:

\[ a_↑ = a_↓ = \frac{1}{\sqrt{\Omega}} \] \hspace{1cm} (A.60)
\[ r_↑ = \frac{1}{\sqrt{\Omega}} \frac{k - k_1}{k + k_1} \] \hspace{1cm} (A.61)
\[ r_↓ = \frac{1}{\sqrt{\Omega}} \frac{k - k_1}{k + k_1} \] \hspace{1cm} (A.62)
\[ t_↑ = \frac{1}{\sqrt{\Omega}} \frac{2k}{k + k_1} \] \hspace{1cm} (A.63)
\[ t_↓ = \frac{1}{\sqrt{\Omega}} \frac{2k}{k + k_1} \] \hspace{1cm} (A.64)

and we can write the wavefunctions as:

\[ \psi_{in} = \frac{1}{\sqrt{\Omega}} e^{ikx} \begin{pmatrix} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{pmatrix} \] \hspace{1cm} (A.65)
\[ \psi_{tr} = \frac{1}{\sqrt{\Omega}} \begin{pmatrix} \frac{2k}{k + k_1} e^{ik_1 x} \cos \frac{\theta}{2} \\ \frac{2k}{k + k_1} e^{ik_1 x} \sin \frac{\theta}{2} \end{pmatrix} \] \hspace{1cm} (A.66)
\[ \psi_{re} = \frac{1}{\sqrt{\Omega}} \begin{pmatrix} \frac{k - k_1}{k + k_1} e^{-ikx} \cos \frac{\theta}{2} \\ \frac{k - k_1}{k + k_1} e^{-ikx} \sin \frac{\theta}{2} \end{pmatrix} \] \hspace{1cm} (A.67)
To calculate the spin current density (equation [A.11]), the gradient and complex conjugates of the wavefunctions have to be determined:

\[
\nabla \psi_{\text{in}} = \frac{1}{\sqrt{\Omega}} i k e^{i k x} \left( \begin{array}{c} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{array} \right) \hat{x} \quad (A.68)
\]

\[
\nabla \psi_{\text{tr}} = \frac{1}{\sqrt{\Omega}} \left( \begin{array}{c} i k t^*_1 e^{-i k x} \cos \frac{\theta}{2} \\ i k t^*_1 e^{-i k x} \sin \frac{\theta}{2} \end{array} \right) \hat{x} \quad (A.69)
\]

\[
\nabla \psi_{\text{re}} = \frac{1}{\sqrt{\Omega}} \left( \begin{array}{c} -ik r^*_1 e^{-i k x} \cos \frac{\theta}{2} \\ -ik r^*_1 e^{-i k x} \sin \frac{\theta}{2} \end{array} \right) \hat{x} \quad (A.70)
\]

\[
\psi_{\text{in}}^* = \frac{1}{\sqrt{\Omega}} e^{-i k x} \left( \begin{array}{c} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{array} \right) \quad (A.71)
\]

\[
\psi_{\text{tr}}^* = \frac{1}{\sqrt{\Omega}} \left( \begin{array}{c} t^*_1 e^{-i k x} \cos \frac{\theta}{2} \\ t^*_1 e^{-i k x} \sin \frac{\theta}{2} \end{array} \right) \quad (A.72)
\]

\[
\psi_{\text{re}}^* = \frac{1}{\sqrt{\Omega}} \left( \begin{array}{c} r^*_1 e^{i k x} \cos \frac{\theta}{2} \\ r^*_1 e^{i k x} \sin \frac{\theta}{2} \end{array} \right) \quad (A.73)
\]

Just as in the previous section, the spin current density only has an \( x \) spatial direction and therefore can be written as:

\[
\vec{Q}_x = \frac{\hbar}{2m} \text{Im} \left( \begin{array}{c} Q_{xx} \\ Q_{xy} \\ Q_{xz} \end{array} \right), \quad (A.74)
\]

where again:

\[
Q_{xx} = \psi^* \sigma_x \frac{\partial \psi}{\partial x} \quad (A.75)
\]

\[
Q_{xy} = \psi^* \sigma_y \frac{\partial \psi}{\partial y} \quad (A.76)
\]

\[
Q_{xz} = \psi^* \sigma_z \frac{\partial \psi}{\partial z} \quad (A.77)
\]

Calculating this for the incident, transmitted and reflected wavefunctions gives for the incident wavefunction the same result as in the previous section:

\[
Q_{xx,\text{in}} = \frac{1}{\sqrt{\Omega}} e^{-i k x} \left( \begin{array}{c} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{array} \right) \left( \begin{array}{cc} 0 & 1 \\ 1 & 0 \end{array} \right) \frac{1}{\sqrt{\Omega}} i k e^{i k x} \left( \begin{array}{c} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{array} \right) = \frac{i k}{\Omega} \cos \frac{\theta}{2} \sin \frac{\theta}{2} \sin \theta \quad (A.78)
\]

\[
Q_{xy,\text{in}} = \frac{1}{\sqrt{\Omega}} e^{-i k x} \left( \begin{array}{c} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{array} \right) \left( \begin{array}{cc} 0 & -i \\ i & 0 \end{array} \right) \frac{1}{\sqrt{\Omega}} i k e^{i k x} \left( \begin{array}{c} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{array} \right) = \frac{i k}{\Omega} \left( -i \cos \frac{\theta}{2} \sin \frac{\theta}{2} + i \cos \frac{\theta}{2} \sin \frac{\theta}{2} \right) = 0 \quad (A.79)
\]
A.3 Spin torque near the metal–ferromagnet interface

\[ Q_{xx, in} = \frac{1}{\sqrt{2}} e^{-ikx} \left( \begin{array}{cc} \cos \frac{\theta}{2} & \sin \frac{\theta}{2} \\ -\sin \frac{\theta}{2} & \cos \frac{\theta}{2} \end{array} \right) \left( \begin{array}{c} 1 \\ 0 \\ 0 \\ -1 \end{array} \right) \frac{1}{\sqrt{2}} e^{ikx} \left( \begin{array}{c} \cos \frac{\theta}{2} \\ \sin \frac{\theta}{2} \end{array} \right) \]

\[ = \frac{i k}{\Omega} \left( \cos^2 \frac{\theta}{2} - \sin^2 \frac{\theta}{2} \right) \frac{1}{\cos \theta} \]

Repeating the procedure for the transmitted wavefunctions gives:

\[ Q_{xx, tr} = \frac{1}{\Omega} \left( \begin{array}{cc} t_1^* e^{-ik_1 x} \cos \frac{\theta}{2} & t_1^* e^{-ik_1 x} \sin \frac{\theta}{2} \end{array} \right) \left( \begin{array}{cc} 0 & i \\ i & 0 \end{array} \right) \left( \begin{array}{c} k_1 t_1 e^{ik_1 x} \cos \frac{\theta}{2} \\ k_1 t_1 e^{ik_1 x} \sin \frac{\theta}{2} \end{array} \right) \]

\[ = \frac{1}{\Omega} \left( t_1^* e^{-ik_1 x} \cos \frac{\theta}{2} i k_1 t_1 e^{ik_1 x} \sin \frac{\theta}{2} + t_1^* e^{-ik_1 x} \sin \frac{\theta}{2} i k_1 t_1 e^{ik_1 x} \cos \frac{\theta}{2} \right) \]

\[ = \frac{i}{\Omega} 2 \Re \left( t_1 t_1^* \right) \frac{1}{2} \sin \theta \left( k_1 e^{-i(k_1-k_1)x} + k_1 e^{i(k_1-k_1)x} \right) \] (A.80)

\[ \Im (Q_{xx, tr}) = \frac{1}{\Omega} \Re \left( t_1 t_1^* \right) \sin \theta \Re \left( k_1 e^{-i(k_1-k_1)x} + k_1 e^{i(k_1-k_1)x} \right) \]

\[ = \frac{1}{\Omega} \Re \left( t_1 t_1^* \right) \sin \theta \left[ k_1 \cos \left( (k_1 - k_1) x \right) + k_1 \cos \left( (k_1 - k_1) x \right) \right] \]

\[ = \frac{1}{\Omega} \Re \left( t_1 t_1^* \right) \sin \theta \left( k_1 + k_1 \right) \cos \left( (k_1 - k_1) x \right) \] (A.81)

\[ Q_{xy, tr} = \frac{1}{\Omega} \left( \begin{array}{cc} t_1^* e^{-ik_1 x} \cos \frac{\theta}{2} & t_1^* e^{-ik_1 x} \sin \frac{\theta}{2} \end{array} \right) \left( \begin{array}{cc} 0 & -i \\ i & 0 \end{array} \right) \left( \begin{array}{c} i k_1 t_1 e^{ik_1 x} \cos \frac{\theta}{2} \\ i k_1 t_1 e^{ik_1 x} \sin \frac{\theta}{2} \end{array} \right) \]

\[ = \frac{i}{\Omega} \left( -t_1^* e^{-ik_1 x} \cos \frac{\theta}{2} i k_1 t_1 e^{ik_1 x} \sin \frac{\theta}{2} + t_1^* e^{-ik_1 x} \sin \frac{\theta}{2} i k_1 t_1 e^{ik_1 x} \cos \frac{\theta}{2} \right) \]

\[ = \frac{i}{\Omega} 2 \Im \left( t_1 t_1^* \right) \frac{1}{2} \sin \theta \left( -i k_1 e^{-i(k_1-k_1)x} + i k_1 e^{i(k_1-k_1)x} \right) \] (A.82)

\[ \Im (Q_{xy, tr}) = \frac{1}{\Omega} \Im \left( t_1 t_1^* \right) \sin \theta \Im \left( -i k_1 e^{-i(k_1-k_1)x} + i k_1 e^{i(k_1-k_1)x} \right) \]

\[ = \frac{1}{\Omega} \Im \left( t_1 t_1^* \right) \sin \theta \left[ k_1 \sin \left( (k_1 - k_1) x \right) + k_1 \sin \left( (k_1 - k_1) x \right) \right] \]

\[ = \frac{1}{\Omega} \Im \left( t_1 t_1^* \right) \sin \theta \left( k_1 + k_1 \right) \sin \left( (k_1 - k_1) x \right) \] (A.83)

\[ Q_{xz, tr} = \frac{1}{\Omega} \left( \begin{array}{cc} t_1^* e^{-ik_1 x} \cos \frac{\theta}{2} & t_1^* e^{-ik_1 x} \sin \frac{\theta}{2} \end{array} \right) \left( \begin{array}{cc} 0 & 0 \\ 0 & -1 \end{array} \right) \left( \begin{array}{c} i k_1 t_1 e^{ik_1 x} \cos \frac{\theta}{2} \\ i k_1 t_1 e^{ik_1 x} \sin \frac{\theta}{2} \end{array} \right) \]

\[ = \frac{1}{\Omega} \left( t_1^* e^{-ik_1 x} \cos \frac{\theta}{2} i k_1 t_1 e^{ik_1 x} \cos \frac{\theta}{2} - t_1^* e^{-ik_1 x} \sin \frac{\theta}{2} i k_1 t_1 e^{ik_1 x} \sin \frac{\theta}{2} \right) \]

\[ = \frac{i}{\Omega} \left| t_1 \right|^2 k_1 \cos^2 \frac{\theta}{2} - \left| t_1 \right|^2 k_1 \sin^2 \frac{\theta}{2} \] (A.84)

\[ \Im (Q_{xz, tr}) = \frac{1}{\Omega} \left( \left| t_1 \right|^2 k_1 \cos^2 \frac{\theta}{2} - \left| t_1 \right|^2 k_1 \sin^2 \frac{\theta}{2} \right) \] (A.85)
Finally, for the reflected wavefunctions the spin current densities are:

\[
Q_{xx, re} = \frac{1}{\Omega} \left( r_1^* e^{ikx} \cos \frac{\theta}{2} \right) \left( 0 \ 1 \right) \left( \begin{array}{cc} 0 & 1 \\ -ikr_1 e^{-ikx} \cos \frac{\theta}{2} \\ -ikr_1 e^{-ikx} \sin \frac{\theta}{2} \end{array} \right)
\]

\[
= -\frac{1}{\Omega} \left( r_1^* e^{ikx} \cos \frac{\theta}{2} + r_1^* e^{ikx} \sin \frac{\theta}{2} ikr_1 e^{-ikx} \cos \frac{\theta}{2} \right)
\]

\[
= -ik \frac{1}{2} \sin \theta 2 \mbox{Re} (r_1^*) \quad \text{(A.87)}
\]

\[
\text{Im} (Q_{xx, re}) = -k \frac{1}{\Omega} \sin \theta \mbox{Re} (r_1^*) \quad \text{(A.88)}
\]

\[
Q_{xy, re} = \frac{1}{\Omega} \left( r_1^* e^{ikx} \cos \frac{\theta}{2} \right) \left( 0 \ -i \right) \left( \begin{array}{cc} 0 & -i \\ -ikr_1 e^{-ikx} \cos \frac{\theta}{2} \\ -ikr_1 e^{-ikx} \sin \frac{\theta}{2} \end{array} \right)
\]

\[
= -i \frac{1}{\Omega} \left( r_1^* e^{ikx} \cos \frac{\theta}{2} + r_1^* e^{ikx} \sin \frac{\theta}{2} i kr_1 e^{-ikx} \cos \frac{\theta}{2} \right)
\]

\[
= -ik \frac{1}{2} \sin \theta 2 \mbox{Im} (r_1^*) \quad \text{(A.89)}
\]

\[
\text{Im} (Q_{xy, re}) = -k \frac{1}{\Omega} \sin \theta \mbox{Im} (r_1^*) \quad \text{(A.90)}
\]

\[
Q_{xz, re} = \frac{1}{\Omega} \left( r_1^* e^{ikx} \cos \frac{\theta}{2} \right) \left( 1 \ 0 \right) \left( \begin{array}{cc} 1 & 0 \\ -ikr_1 e^{-ikx} \cos \frac{\theta}{2} \\ -ikr_1 e^{-ikx} \sin \frac{\theta}{2} \end{array} \right)
\]

\[
= \frac{1}{\Omega} \left( -r_1^* e^{ikx} \cos \frac{\theta}{2} + r_1^* e^{ikx} \sin \frac{\theta}{2} i kr_1 e^{-ikx} \sin \frac{\theta}{2} \right)
\]

\[
= \frac{1}{\Omega} ik \left( -|t_1|^2 \cos^2 \frac{\theta}{2} + |r_1|^2 \sin^2 \frac{\theta}{2} \right) \quad \text{(A.91)}
\]

\[
\text{Im} (Q_{xz, re}) = k \frac{1}{\Omega} \left( -|t_1|^2 \cos^2 \frac{\theta}{2} + |r_1|^2 \sin^2 \frac{\theta}{2} \right) \quad \text{(A.92)}
\]

Concluding, we can write the incident spin current as:

\[
\vec{Q}_{x, in} = \frac{\hbar^2 k}{2m \Omega} \left( \begin{array}{c} \sin \theta \\ 0 \\ \cos \theta \end{array} \right), \quad \text{(A.93)}
\]

the transmitted spin current as:

\[
\vec{Q}_{x, tr} = \frac{\hbar^2 k}{2m \Omega} \left( \begin{array}{c} \text{Re} \left( t_1^* \right) \sin \theta \left( k_1 + k_1 \right) \cos \left( \left( k_1 - k_1 \right) x \right) \\ \text{Im} \left( t_1^* \right) \sin \theta \left( k_1 + k_1 \right) \sin \left( \left( k_1 - k_1 \right) x \right) \\ \left| t_1 \right|^2 k_1 \cos^2 \frac{\theta}{2} - \left| t_1 \right|^2 k_1 \sin^2 \frac{\theta}{2} \end{array} \right) \quad \text{(A.94)}
\]
and the reflected spin current as:

\[
\vec{Q}_{x, re} = -\frac{\hbar^2 k}{2m\Omega} \begin{pmatrix}
\text{Re} \left( r^* \right) \sin \theta \\
\text{Im} \left( r^* \right) \sin \theta \\
-|r|^2 \cos^2 \frac{\theta}{2} + |r|^2 \sin^2 \frac{\theta}{2}
\end{pmatrix}.
\] (A.95)

Again, the spin torque \( \vec{\tau}_{st} \) is calculated from the net spin current as:

\[
\vec{\tau}_{st} = -\hat{n} \cdot \vec{Q}_{d} = A \hat{x} \cdot \left( \vec{Q}_{in} + \vec{Q}_{re} - \vec{Q}_{tr} \right)
\] (A.96)

Substituting equations A.93 – A.95 yields:

\[
\vec{\tau}_{st} = A \frac{\hbar^2}{2m} \begin{pmatrix}
\tau_{st,x} \\
\tau_{st,y} \\
\tau_{st,z}
\end{pmatrix},
\] (A.97)

where, using:

\[
\tau_{st,z} = k \cos \theta - \left( k |r|^2 + k |t|^2 \right) \cos^2 \frac{\theta}{2} + \left( k |r|^2 + k |t|^2 \right) \sin^2 \frac{\theta}{2}
\]

\[
= k \cos \theta - \left( k \frac{k - k^*}{k + k^* + k} + k \frac{2k}{k + k^*} \right) \cos^2 \frac{\theta}{2}
\]

\[
+ \left( k \frac{k - k^*}{k + k^* + k} + k \frac{2k}{k + k^*} \right) \sin^2 \frac{\theta}{2}
\]

\[
= k \cos \theta - k \cos^2 \frac{\theta}{2} + k \sin^2 \frac{\theta}{2} = 0
\] (A.100)

this turns into:

\[
\tau_{st,x} = \sin \theta \left[ k \left( 1 - \text{Re} (r^* \right) - (k_1 + k) \text{Re} \left( t^* t^* \right) \cos \left( (k_1 - k_1) x \right) \right]
\] (A.98)

\[
\tau_{st,y} = \sin \theta \left[ -k \text{Im} \left( r^* \right) - (k_1 + k) \text{Im} \left( t^* t^* \right) \sin \left( (k_1 - k_1) x \right) \right]
\] (A.99)

\[
\tau_{st,z} = 0
\] (A.100)

Although there is a net spin current density \( (Q_{x, in} + Q_{x, re} - Q_{x, tr} \neq 0) \), we no want to show that there is no net current flowing through the interface. Therefore we need to calculate the number current densities \( \vec{j} \):

\[
\vec{j} = \frac{\hbar}{m} \text{Im} \left( \psi^* \vec{\nabla} \psi \right),
\] (A.101)

which, in this case, only have a component in the \( x \) spatial direction. We first determine \( \psi^* \vec{\nabla} \psi \) and using equations A.68 [A.70] and A.71 [A.73] we can write:

\[
\psi^*_{in} \vec{\nabla} \psi_{in} = \frac{1}{\sqrt{\Omega}} e^{-ikx} \left( \cos \frac{\theta}{2} \sin \frac{\theta}{2} \right) ik \frac{1}{\sqrt{\Omega}} e^{-ikx} \left( \cos \frac{\theta}{2} \sin \frac{\theta}{2} \right)
\]

\[
= ik \frac{1}{\Omega}
\] (A.102)
The incoming, transmitted and reflected currents in the $x$ spatial direction are given by:

$$j_{x,\text{in}} = \frac{\hbar k}{m\Omega} $$

(A.105)

$$j_{x,\text{tr}} = \frac{h}{m\Omega} \left( k|t|e^{-ikx} \cos \frac{\theta}{2} + k|t|e^{ikx} \sin \frac{\theta}{2} \right) $$

(A.106)

$$j_{x,\text{re}} = -\frac{\hbar k}{m\Omega} \left( |r|e^{ikx} \cos \frac{\theta}{2} + |r|e^{-ikx} \sin \frac{\theta}{2} \right). $$

(A.107)

The continuity equation (A.4) tells us that there is, in this case, no net current flowing:

$$\vec{\nabla} \cdot \vec{j} = 0 $$

(A.108)

$$j_{x,\text{in}} + j_{x,\text{re}} - j_{x,\text{tr}} = 0. $$

(A.109)
Appendix B

Process Steps

B.1 Bottom electrode

Cleaning the sample:
- Ammonia in ultrasonic bath for 10 min
- Rinse with iso-propanol
- Iso-propanol in ultrasonic bath for 10 min
- Blow dry using N\textsubscript{2}

Cleaning the mask:
- Acetone in ultrasonic bath for 15 min
- Iso-propanol in ultrasonic bath for 15 min
- Rinse in H\textsubscript{2}O until resistance > 12 M\text{Ω}cm
- Blow dry using N\textsubscript{2}

Preparing sample for UV lithography:
- Plasma clean in O\textsubscript{2} plasma at 300 W for 10 min\textsuperscript{1}
- Rinse in H\textsubscript{2}O for 10 min
- Blow dry using N\textsubscript{2}
- Put in HMDS\textsuperscript{2} primer oven\textsuperscript{3}
- Spin coat\textsuperscript{4} ma-N 415 resist\textsuperscript{5} at 4000 RMP for 35 s
- Softbake at 90\textdegree{}C for 2 min on hotplate

\textsuperscript{1}Stripper name
\textsuperscript{2}Hexamethyldisilazane (HDMS)
\textsuperscript{3}HDMS oven name
\textsuperscript{4}Convac 1001S Spinner
\textsuperscript{5}ma-N 415 negative resist. From Micro Resist Technology GmbH
B.2 Insulating layer

Exposure:
- Load mask into mask aligner\(^6\)
- Load sample and align with mask
- Expose for 20 s
- Post exposure bake at 90°C for 2 min on hotplate

Develop:
- Develop in ma-D 332S\(^7\) pure for 2 min
- Rinse in H\(_2\)O until resistance > 10 MΩcm
- Blow dry using N\(_2\)

Metallization:
- Load sample into sputterclock\(^8\)
- Sputter 50 nm of Pt at 60 W 2.1 SCCM argon flow

Lift-off:
- Acetone vapor for 30 min
- Rinse with acetone
- Acetone in ultrasonic bath for 5 min
- Rinse with iso-propanol
- Iso-propanol in ultrasonic bath for 5 min
- Blow dry using N\(_2\)

B.2 Insulating layer

Preparing sample for E-Beam Lithography:
- Bake out sample at 200 °C for 2 min on hotplate and cool down afterwards for 5 min
- Spin coat 495PMMA A6\(^9\) at 2000 RMP for 50 s
- Hardbake at 200°C for 2 min on hotplate and cool down afterwards for 5 min
- Spin coat 950PMMA A2\(^10\) at 5000 RMP for 50 s

---
\(^6\)Karl Suss MA6 Mask Aligner
\(^7\)ma-D 332S developer for ma-N 415 resist. From Micro Resist Technology GmbH
\(^8\)Kurt J. Lesker UHV sputter facility
\(^9\)6% polymethylmethacrylate (PMMA) positive resist with a molecular weight of 495 000 solved in anisole. From MicroChem Corp.
\(^10\)2% polymethylmethacrylate (PMMA) positive resist with a molecular weight of 950 000 solved in anisole. From MicroChem Corp.
B.3 Top electrode

- Hardbake at 200°C for 2 min on hotplate and cool down afterwards for 5 min

**Exposure:**

- Load sample into EBL system
- Acceleration voltage 30 keV and set the beam current to 0.15 nA
- Measure current in Faraday Cup
- Use an Area Dose of 280 µC/cm²

**Develop:**

- Develop in MIBK:IPA 1:3 for 30 s
- Rinse in iso-propanol for 30 s
- Blow dry using N₂

**Insulating layer:**

- Load sample into sputterclock
- Sputter 50 nm of SiO₂ at 120 W 1.4 SCCM argon flow

**Lift-off:**

- Acetone vapor for 30 min
- Rinse with acetone
- Acetone in ultrasonic bath for 5 min
- Rinse with ethanol
- Iso-propanol in ultrasonic bath for 5 min
- Blow dry using N₂

B.3 Top electrode

**Cleaning the mask:**

- Acetone in ultrasonic bath for 15 min
- Iso-propanol in ultrasonic bath for 15 min
- Rinse in H₂O until resistance > 12 MΩcm
- Blow dry using N₂

**Preparing sample for UV lithography:**

---

11 FEI Nova Nanolab 600i using Raith module
12 Methyl Iso-Butyl Ketone (MIBK)
13 Iso-Propyl Alcohol (IPA) also known as iso-propanol
B.4 Making the undercut

- Plasma clean in O₂ plasma at 300 W for 5 min
- Rinse in H₂O for 10 min
- Blow dry using N₂
- Put in HMDS primer oven
- Spin coat maN-415 resist at 4000 RMP for 35 s
- Soft bake at 90°C for 2 min on hotplate

**Exposure:**

- Load mask into mask aligner
- Load sample and align with mask
- Expose for 20 s
- Post exposure bake at 90°C for 2 min on hotplate

**Develop:**

- Develop in ma D332 pure for 2 min
- Rinse in H₂O until resistance > 10 MΩcm
- Blow dry using N₂

**Metallization:**

- Load sample into sputter clock
- Sputter 20 nm of Pt at 60 W 2.1 SCCM argon flow

**Lift-off:**

- Acetone vapor for 30 min
- Rinse with acetone
- Acetone in ultrasonic bath for 5 min
- Rinse with iso-propanol
- Iso-propanol in ultrasonic bath for 5 min
- Blow dry using N₂

B.4 Making the undercut

**Preparing sample for the Focused Ion Beam:**

- Spin coat 950PMMA A14 resist at 5000 RMP for 45 s
- Hard bake at 200°C for 2 min on hotplate

---

14% polymethylmethacrylate (PMMA) positive resist with a molecular weight of 950 000 solved in anisole. From MicroChem Corp.
B.5 Making GMR stack

Making nanostencil:
  - Load sample into DualBeam system
  - Acceleration voltage 30 keV and set the beam current to 1.5 pA

Opening undercut:
  - Put sample in HF(1%) for 5 min
  - Rinse in H₂O until resistance > 12 MΩcm
  - Blow dry using N₂

B.5 Making GMR stack

Metallization:
  - Load sample into sputterclock
  - Sputter desired GMR stack using appropriate tooling factors

Lift-off:
  - Acetone vapor for 60 min
  - Rinse with acetone
  - Acetone in ultrasonic bath for 5 min
  - Rinse with iso-propanol
  - Iso-propanol in ultrasonic bath for 5 min
  - Blow dry using N₂

---

15 FEI Nova Nanolab 600i
16 1% Hydrofluoric acid solution in H₂O
Appendix C

UV Lithography

Figure C.1: Drawing of one section of the FNA UV Mask. The actual mask is made up of 9 (3 × 3) sections and therefore contains 225 electrodes.
Figure C.2: Drawing of one electrode of the FNA UV Mask. (a) Overview of one electrode showing the $200 \times 200 \, \mu m^2$ contact pads and (b) a close-up of the $2 \, \mu m$ wide junction area.
Appendix D

Formal derivation of the growth inside a nanostencil

In this appendix we will give a derivation of the expressions for the deposition rate $d$ inside a nanostencil using the geometry shown in figures 6.4 and D.1.

If we first look at a clear flat surface, the amount of material $d$ deposited per unit of time $t$ at a position $(x, y)$ on the surface can be described by:

$$d(x, y) = \int_{-\pi/2}^{\pi/2} \int_{0}^{\pi} f(\Theta, \varphi) \, d\Theta \, d\varphi,$$

where $f(\Theta, \varphi)$ is the incident flux of material as a function of the azimuth $\varphi$ and polar angle $\Theta$, as shown in figure 6.4(a). If the incident flux is created using a sputtering technique, this function will be axisymmetric along the azimuth angle and will roughly follow a sine law for the polar angle [53] (see section 4.1.1) and consequently can be described by:

$$f(\Theta, \varphi) \propto \sin(\Theta).$$

If, however, another technique is used to deposit the material, e.g. molecular beam epitaxy (MBE) or evaporation, the incident flux $f$ can be described by another function of $\varphi$ and $\Theta$.

If we now look at the case of a nanostencil mask being placed above the surface, as shown in figure 6.4(b), the incident flux is now (partly) obstructed at certain positions on the surface. Therefore the integration boundaries for the polar angle in equation (D.1) are now functions of the position and azimuth.

1 The incident flux in this case can be described as a Gaussian peak centered around a polar angle $\Theta_0$, where $\Theta_0$ is the angle between the normal of the plane and the source.
Figure D.1: Geometry used for modeling the growth inside a nanostencil. Figures (a) and (b) show a three dimensional representation of the nanostencil placed above the surface. Figures (c) and (d) represent a top view (xy-plane) of the nanostencil, showing the semimajor inner $a_r$ and outer $a_{r+c}$ axis, the semiminor inner $b_r$ and outer $b_{r+c}$ axis and the azimuth angle $\phi$. Figures (e) and (f) represent a side view (xz-plane) showing the height of the nanostencil and the polar angle $\Theta$. For this position $x$ and angle $\phi$ the minimum and maximum incident angle are given by $\Theta_{r+c}^-$ and $\Theta_{r+c}^+$ respectively.
angle and making use of relation \[D.2\] equation \[D.1\] becomes:

\[
d(x, y) = \frac{1}{2\pi} \int_{-\frac{\pi}{2}}^{+\frac{\pi}{2}} \int_{\Theta_{\text{min}}}^{\Theta_{\text{max}}} \sin(\Theta) \, d\Theta \, d\varphi
\]

\[
= \frac{1}{2\pi} \int_{-\frac{\pi}{2}}^{+\frac{\pi}{2}} \left[ \cos(\Theta_{\text{min}}(\varphi, x, y)) - \cos(\Theta_{\text{max}}(\varphi, x, y)) \right] \, d\varphi,
\]

(D.3)

where \(\Theta_{\text{min}}\) and \(\Theta_{\text{max}}\) are the minimum and maximum polar angles. To calculate these angles for a given position \(x\) along the x-axis and azimuth angle \(\varphi\), we first need to calculate the corresponding coordinates \([x^\pm, y^\pm]\) on the inner and \([x_{r+c}^\pm, y_{r+c}^\pm]\) on the outer ellipse, as shown in figure [D.1(c) and (d)]. These coordinates are described by the following set of equations:

\[
\tan(\varphi) = \frac{y^\pm_r}{x - x^\pm_r}
\]

(D.4)

\[
\left(\frac{x^\pm_r}{a_r}\right)^2 + \left(\frac{y^\pm_r}{b_r}\right)^2 = 1
\]

(D.5)

for the inner ellipse and

\[
\tan(\varphi) = \frac{y_{r+c}^\pm}{x - x_{r+c}^\pm}
\]

(D.6)

\[
\left(\frac{x_{r+c}^\pm}{a_{r+c}}\right)^2 + \left(\frac{y_{r+c}^\pm}{b_{r+c}}\right)^2 = 1
\]

(D.7)

for the outer ellipse. During the deposition, not only material gets deposited inside the nanostencil, but also on top and on the edges. To take into account for this effect the inner and outer radii of the nanostencil are described by:

\[
a_r = a - s_{xx}^y
\]

(D.8)

\[
b_r = b - s_{xx}^y
\]

(D.9)

\[
a_{r+c} = a + c - s_{xx}^{r+c}
\]

(D.10)

\[
b_{r+c} = b + c - s_{xx}^{r+c}
\]

(D.11)

\[
c = \frac{t + s_{r+c}^z - s_r^z}{\tan(\theta_A)} + s_{r+c}^{xy} - s_r^{xy},
\]

(D.12)

where \(s_{xx}^y\) and \(s_r^z\) represent the growth on the edges of the nanostencil, \(s_{xx}^{r+c}\) and \(s_{r+c}^z\) on top of it and \(\theta_A\) the angle of the nanostencil edge, as shown in figure [D.2(b)]. Solving the set of equations \[D.4\] - \[D.7\] for \(x^\pm_r, y^\pm_r, x_{r+c}^\pm,\) and \(y_{r+c}^\pm)\)
Figure D.2: Geometry used for modeling the growth inside a nanostencil. (a) Represents a three dimensional representation showing the semi-major inner $a_r$ and outer $a_{r+c}$ axis, the semiminor inner $b_r$ and outer $b_{r+c}$ axis, the azimuth angle $\varphi$ and polar angle $\Theta$. (b) Represents a side view (xz-plane) showing the height of the deposition inside the nanostencil $z(x)$, the different growth parameters $s$, the distance between the outer and inner ellipse $c$ and the angle of the nanostencil edges $\Theta_A$.

gives:

\[ x^\pm_r (\varphi, x) = \frac{a_r^2 x \cos^2 (\varphi) \pm a_r b_r |\cos (\varphi)|}{\sqrt{(...)}} \quad (D.13) \]

\[ \sqrt{(...)} = \sqrt{(b_r^2 - a_r^2 + x^2) \cos^2 (\varphi) + a_r^2 - x^2} \]

\[ y_r^\pm (\varphi, x) = \tan (\varphi) (x - x^\pm_r) \quad (D.14) \]

\[ x^\pm_{r+c} (\varphi, x) = \frac{a_{r+c}^2 x \cos^2 (\varphi) a_{r+c} b_{r+c} |\cos (\varphi)|}{\sqrt{(...)}} \quad (D.15) \]

\[ \sqrt{(...)} = \sqrt{(b_{r+c}^2 - a_{r+c}^2 + x^2) \cos^2 (\varphi) + a_{r+c}^2 - x^2} \]

\[ y_{r+c}^\pm (\varphi, x) = \tan (\varphi) (x - x^\pm_{r+c}) \quad (D.16) \]

Using these equations we can derive expressions for the minimum and maximum incident angles for both the inner and outer ellipse, where we have to differentiate between $|x| \leq a_r$ and $a_r < |x| < x_{\text{max}}$. In the first case, the minimum and maximum angles are given by:

\[ |x| \leq a_r \]
\[ \Theta_r^{-}(\varphi, x, z(x)) = \arctan \left( \frac{h - z(x)}{\sqrt{(x - x_r^{-})^2 + (y_r^{-})^2}} \right) \quad (D.17) \]
\[ \Theta_r^{+}(\varphi, x, z(x)) = \pi - \arctan \left( \frac{h - z(x)}{\sqrt{(x - x_r^{+})^2 + (y_r^{+})^2}} \right) \quad (D.18) \]
\[ \Theta_{r+c}^{-}(\varphi, x, z(x)) = \arctan \left( \frac{h_{r+c} - z(x)}{\sqrt{(x - x_{r+c}^{-})^2 + (y_{r+c}^{-})^2}} \right) \quad (D.19) \]
\[ \Theta_{r+c}^{+}(\varphi, x, z(x)) = \pi - \arctan \left( \frac{h_{r+c} - z(x)}{\sqrt{(x - x_{r+c}^{+})^2 + (y_{r+c}^{+})^2}} \right) \quad (D.20) \]

where \( z(x) \) is the deposition profile inside the nanostencil and \( h_{r+c} \), as shown in figure D.2(b), is given by:
\[ h_{r+c} = h + t + s_{r+c}^z \quad (D.21) \]

The exact geometry and position inside the nanostencil now determine \( \Theta_{\min} \) and \( \Theta_{\max}^{\varphi} \). The deposition rate inside the nanostencil, equation D.3, can now be written as:
\[
\begin{align*}
  d(x, z(x)) &= \frac{1}{2\pi} \int_{-\pi/2}^{\pi/2} \left[ \cos \left( \max \left( \Theta_r^{-}, \Theta_{r+c}^{-} \right) \right) \\
  &\quad - \cos \left( \min \left( \Theta_r^{+}, \Theta_{r+c}^{+} \right) \right) \right] d\varphi \quad (D.22)
\end{align*}
\]

The second case, where \( a_r < |x| < x_{\max} \), is shown in figure D.3(a) and (b). In this case, there is a maximum value to the azimuth angle \( \varphi_{\max} \), which is given by:
\[ \varphi_{\max} = \arctan \left( \frac{b_r}{\sqrt{x^2 - a_r^2}} \right) \quad (D.23) \]

The maximum \( x \) value \( x_{\max} \) is obtained as \( \Theta_{r+c}^{-} \) is equal to \( \Theta_{r+c}^{+} \) and, in the case \( z(x) = 0 \) is given by:
\[ x_{\max} = \frac{a_{r+c}h + a_r h_{r+c}}{h_{r+c} - h} \quad (D.24) \]

\(^2\)In the case of figure D.1(e) and (f), the edges of the outer ellipse limit the incident flux and thus form the integration boundaries for the polar angle. Therefore \( \Theta_{\min} = \Theta_{r+c}^{-} \) and \( \Theta_{\max} = \Theta_{r+c}^{+} \).

\(^3\)Once deposition in the nanostencil takes place, \( z(x) \) will increase and therefore \( x_{\max} \) will decrease.
Using equations [D.13] – [D.16] we can again derive expressions for the minimum and maximum incident angles, which are in this case given by:

\[ a_r < |x| < x_{\text{max}} \]

\[
\Theta^{-}_r (\varphi, x, z(x)) = \arctan \left( \frac{h - z(x)}{\sqrt{(x - x_r)^2 + (y_r)^2}} \right) \quad (D.25)
\]

\[
\Theta^{+}_r (\varphi, x, z(x)) = \arctan \left( \frac{h - z(x)}{\sqrt{(x - x_r)^2 + (y_r)^2}} \right) \quad (D.26)
\]

\[
\Theta^{-}_{r+c} (\varphi, x, z(x)) = \arctan \left( \frac{h_{r+c} - z(x)}{\sqrt{(x - x_{r+c})^2 + (y_{r+c})^2}} \right) \quad (D.27)
\]

The deposition rate inside the nanostencil, equation [D.3] can in this case be written as:

\[
d(x, z(x)) = \frac{1}{2\pi} \int_{-\varphi_{\text{max}}}^{+\varphi_{\text{max}}} \left[ \cos \left( \max \left( \Theta^{-}_r, \Theta^{-}_{r+c} \right) \right) - \cos \left( \Theta^{+}_r \right) \right] d\varphi \quad (D.28)
\]
Appendix E

Numerical model for the growth inside a nanostencil
Figure E.1: Flowchart for the model describing the growth inside a nanostencil.
Bibliography


