Spin Hall magnetoresistance in yttrium iron garnet | tungsten heterostructures

Bachelor’s Thesis
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1 Introduction

Humanity's desire to increase our knowledge of everything led us to constructing more and more powerful computers to assist us. While the growth in computing power followed an exponential increase over the past 50 years [1], this trend began to slow down since we are approaching the limit of increasing computing power by scaling down components [2]. Heating effects and the potential of quantum tunneling fundamentally limit our ability to improve processors [3], leading us to research alternative technologies to be paired with conventional computers or even replace them. Most prominently, this is attempted by introducing a new degree of freedom to information transport besides the electron charge that is in use today [4]. The field of spintronics uses the intrinsic electron spin as an additional degree of freedom and the transport of angular momentum in pure spin currents [5]. Spintronic devices are already in use today in data storage devices utilizing the giant magnetoresistance GMR [6] and the research field promises to realize devices that are much more energy efficient like magneto random access memory (MRAM) [7] and can transport information with much less heat dissipation [3]. In contrast to charge currents, spin currents can be present both in a normal metal (NM) without magnetic order as well as magnetically ordered insulators (MOI) where they propagate as spin waves [8].

The generation and detection of said currents in MOI|NM heterostructures is an important mechanism required to integrate spintronic devices into modern computers. The generation is possible via the spin Hall effect that arises in NMs with strong spin orbit coupling [9, 10, 11] and the detection by its counterpart, the inverse spin Hall effect [8, 12]. In MOI|NM heterostructures they generate spin Hall magnetoresistance (SMR), which we can use to directly measure the spin current transferred from the NM to the MOI. This transfer is dependent on the magnetization orientation of the MOI, allowing us to use the NM|MOI interface as a spin current switch just like in conventional electronics. An important property of generating spin currents this way is that the spin current can enter the MOI by a transfer torque of angular momentum, while the charge carriers that transport spin information in the NM can not enter the MOI. This opens the possibility of magnon based logic [13, 14]. The conversion efficiency of a charge current to a pure spin current via the SHE is material dependent, with the most commonly used NM being platinum with a conversion efficiency between 0.075 and 0.11 [15]. The search for a material with more efficient charge to spin current conversion is of upmost importance for advances in spintronic devices. Aside of this conversion efficiency, the investigation of cheaper materials is interesting as well from an economic standpoint.

In this thesis we investigated spin current generation in yttrium iron garnet (YIG)|tungsten (W) heterostructures to determine its conversion efficiency as well as finding an optimal W layer thickness for spin current generation. To accomplish this we developed a new magneton sputtering method that enables the characterization of a materials spin transport properties by fabricating only a single sample. After introducing the underlying theoretical concepts of spin currents and magnetoresistive effects in Sec. 2, the sample fabrication is explained and discussed in Sec. 3. In Sec. 4 we will discuss our results on the magnetoresistive properties of YIG|W for varying measurement conditions. From thickness dependent measurements we extract the spin transport properties of W and compare them to similar
measurements in YIG|Pt [15], YIG|Ta [16] and CoFeB|W [17]. A conclusion of the thesis and a short outlook are provided in Sec. 5.
2 Theoretical Concepts

In this chapter we cover the concept of pure spin currents and the direct and inverse spin Hall effect, which is commonly used to produce these currents. Additionally, we discuss the spin Hall magnetoresistance occurring in NM|MOI heterostructures. We will give a very short description of the Hanle magnetoresistance and discuss the ordinary MR occurring in materials with a two band structure.

2.1 Pure Charge and Spin Currents

The electric charge of an electron is used to carry information in every electronic device. But electrons also carry spin, which can be used to transport information as well. We describe moving electrons with an electric current density \( j_q \), which we can express by distinguishing the current contributions of the two spin directions spin-up \( |\uparrow\rangle \) and spin-down \( |\downarrow\rangle \). The original current is obtained by adding the two contributions \( j_\uparrow \) and \( j_\downarrow \) [4, 8]:

\[
j_q = j_\uparrow + j_\downarrow
\]

When considering a pure spin current, the two directions contribute with opposing sign. We can express a spin current by

\[
j_s = \frac{\hbar}{2e} (j_\uparrow - j_\downarrow)
\]

with the reduced Planck’s constant \( \hbar \) and the electron charge \( e \). An illustration of a pure charge current and a pure spin current is given in Fig. 2.1. We obtain a pure charge current when spin-up and spin-down electrons move in the same direction with the same current (Fig. 2.1(a)). This current transports charge but no angular momentum. On the other hand, if the spin orientations move in opposing directions with the same current, the overall charge current vanishes and we are left with a pure spin current (Fig. 2.1(c)) which transports angular momentum. If the current for both spin orientations are not equal, we obtain a spin polarized current (Fig. 2.1(c)). It is important to mention that while charge is a conserved transport quantity, angular momentum is not conserved since electrons can undergo spin-flip scattering. Thus the model of a pure spin current is only valid on the lengthscale of the spin diffusion length \( \lambda_{sf} \), i.e. the average length an electron can move through the conductor without a spin-flip.

2.2 Direct and Inverse Spin Hall Effect

In the last section we introduced the concept of a pure spin current. To study this quantity, we need to be able to generate and detect such currents. We can do so by using the spin Hall effect (SHE) and the inverse spin Hall effect (ISHE) that are depicted in Fig. 2.2. In conductors with finite spin orbit coupling (SOC), the charge carriers acquire a spin dependent transverse velocity, which results from both so-called extrinsic effects such as scattering with impurities and phonons like skew-scattering [19] or side-jump scattering [20] and so-
Figure 2.1: (a) Illustration of a pure charge current when spin-up. Both spin up and spin down electrons move in the same direction and produce a charge current $J_q$ while the spin current $J_s$ vanishes. (b) Illustration of a polarized spin current. The currents $J_\uparrow$ and $J_\downarrow$ are not equal, leading to a combination of charge and spin current. (c) Illustration of a pure spin current. Spin up and spin down electrons move in opposite directions, thus producing a pure spin current with no overall charge current. Taken from [18].

called intrinsic effects that are a result of the bandstructure [21]. The transverse velocities of the different spin orientations are in opposite direction, thus creating a pure spin current perpendicular to both the spin polarization and the direction of the charge current (SHE, see Fig. 2.2(a)). The current conversion via the SHE can be described by using the following equation [12]:

$$j^{\text{SHE}}_s = -\frac{\hbar}{2e}\theta_{\text{SH}}(j_q \times s)$$

The spin hall angle $\theta_{\text{SH}}$ is a material dependent quantity that represents the conversion efficiency between charge and spin current. The SHE was first described phenomenologically by Dyakonov and Perell [9] in 1971, a first theoretical description was published by Hirsch [10] in 1999 where the term spin Hall effect originated. It was experimentally verified in 2004 in a GaAs semiconductor by Kato et al. [22].

The ISHE is shown in Fig. 2.2(b). The moving charge carriers in the spin current acquire a transverse velocity again, but since they move in opposing directions, they will be deflected in the same direction. This converts the spin current back to a charge current perpendicular to $s$ and $J_s$. The ISHE can be described by [8]:

$$j^{\text{ISHE}}_q = -\frac{2e}{\hbar}\theta_{\text{SH}}(j_s \times s)$$

Note that charge and spin currents have different units, hence the factor $2e/\hbar$. The SHE and ISHE let us investigate spin currents while maintaining an all electrical input and output and thus utilizing conventional charge based instruments.

### 2.3 Spin Hall Magnetoresistance

In a conducting material, the SHE results in a spin accumulation at the boundary of the structure. This results in a gradient in the spin chemical potential $\mu_s(z) = \mu_\uparrow - \mu_\downarrow$ where $z$ is the direction of the spin current and $\mu_\uparrow, \mu_\downarrow$ are the chemical potentials for spin-up and
spin-down electrons. This in turn creates a spin diffusion current:

$$j_{s, \text{diff}} = \frac{\hbar \sigma}{2 e^2} \nabla \mu_s(z)$$  \hspace{1cm} (5)

It points in opposite direction to the current produced by the SHE and counteracts it, leading to a net zero spin current. This is the case if there is no other material attached to the NM to which the spin current can be transferred to. We call this an open circuit condition, i.e. the spin current can not leave the material. It is depicted in Fig. 2.3(a). However, if there is a NM|MOI interface, the spin current can flow into the MOI. The spin propagates in the MOI as spin waves whose excitation quanta are magnons. The transfer current can be described by [23]:

$$j_{s, \text{STT}} = \frac{\hbar}{4\pi e^2} \left( g_{1^+}^{\uparrow \downarrow} + g_{1^+}^{\uparrow \downarrow} \mathbf{m} \times \right) (\mu_s(0) \mathbf{s} \times \mathbf{m} - \hbar \mathbf{n}) + [g(\mu_{\text{mag}} + \mu_s(0) \mathbf{s} \cdot \mathbf{m}) + S(T_M - T_N)] \mathbf{m}$$  \hspace{1cm} (6)

With $g_{1^+}^{\uparrow \downarrow} = g_{1^+}^{\uparrow \downarrow} + g_{1^+}^{\uparrow \downarrow}$ the complex spin mixing interface conductance, $\mathbf{m}$ the direction of magnetization in the MOI, $\mu_{\text{mag}}$ the magnonic chemical potential, $g$ the spin conductance, $S$ the spin Seebeck coefficient, $T_N$ the temperature of the electronic system in the NM and $T_M$ the temperature of the magnonic system in the MOI. This equation describes a variety of different effects including the SMR. The first term describes the spin current that flows across the NM|MOI interface. It describes elastic spin-flip scattering at the boundary that exert a spin transfer torque (STT) on the magnetization of the MOI. The second term describes thermally activated inelastic spin-flip scattering that transfer energy from the electrons at the interface to magnons in the MOI [8]. We can simplify Eq. (6) as we are only interested in the SMR. We can set $\mu_{\text{mag}}$, $\mathbf{n}$ and $T_M - T_N$ equal to zero and acquire the simplified expression

$$j_{s, \text{int}} = \frac{\hbar}{4\pi e^2} \left( g_{1^+}^{\uparrow \downarrow} + g_{1^+}^{\uparrow \downarrow} \mathbf{m} \times \right) \mu_s(0) \mathbf{s} \times \mathbf{m} + g\mu_s(0)(\mathbf{s} \cdot \mathbf{m}) \mathbf{m}$$  \hspace{1cm} (7)

If the magnetization $\mathbf{M}$ of the MOI is parallel to the spin polarization $\mathbf{s}$, the first term in Eq. (7) vanishes. The second term however remains and would produce a finite spin transfer in parallel configuration. We can neglect the second term, as we know from theoretical calculations that $g << |g_{1^+}^{\uparrow \downarrow}|$, so $g \simeq 0$ is a good approximation [8]. Thus we get no spin transfer current, i.e. the spin current gets reflected just as in open circuit condition. However,
The theoretical concepts

Figure 2.3: (a) Open circuit condition for the spin Hall effect. The spin current leads to a spin accumulation at the boundary, which leads to a difference in the chemical potential $\mu_s$, creating a spin diffusion current that counteracts the spin current produced by the SHE. The depicted steady state shows the charge carriers traveling undisturbed by the SHE. (b) Closed circuit condition for the SHE. Attaching a MOI layer to the NM allows the spin current to flow into the insulator, reducing the diffusion current generated by $\mu_s$ and thus leading to a finite SMR. Figure taken from [16].

If $\mathbf{M} \perp \mathbf{s}$, the first term does not vanish and a finite spin current can flow across the interface. This transfer lowers the spin accumulation at the interface and thus the diffusion current $j_s^{\text{DIFF}}$. The spin current converted back to a charge current via the ISHE is now lower, resulting in a higher resistance in the NM layer. This is called a closed circuit condition and can be seen in fig. 2.3(b) [8, 12].

If we consider a NM|MOI structure, we can change the direction of the magnetization while simultaneously measuring the resistance in the NM. We should be able to see a difference between $R_{\parallel} = R_{|\mathbf{M}|\parallel\mathbf{s}}$ and $R_{\perp} = R_{|\mathbf{M}|\perp\mathbf{s}}$. According to the theoretical description by Chen et al. [24], the total resistance is given by

$$R_{\text{long}} = R_0 + R_1 m_t^2$$

with $R_0$ the intrinsic electric resistance of the NM layer, $R_1 = R_{\parallel} - R_{\perp} < 0$ and $m_t$ the projection of the magnetization onto the transverse $\mathbf{n} \times \mathbf{j}_q$ of the current direction and the surface normal $\mathbf{n}$. We can conclude from this expression that a continuous rotation of the magnetization will result in a $\cos^2$ behaviour, as $m_t^2 \propto \cos^2(\angle(\mathbf{M}, \mathbf{s}))$. The SMR with different magnetization orientations can be seen in Fig. 2.4.

The longitudinal SMR ratio is best expressed by using the resistivity instead of the resistance, since we want to get a result independent of the NM geometry. The ratio is then given by [24]

$$\frac{\rho_1}{\rho_0} = \frac{\theta_{\text{SH}}^2 (2 \lambda_{sf}^2 \rho_{NM}) (t_{NM})^{-1} g_t^{\downarrow\downarrow} \tanh^2 \left( \frac{t_{NM}}{2 \lambda_{sf}} \right)}{hc^{-2} + 2 \lambda_{sf} \rho_{NM} g_t^{\downarrow\downarrow} \coth \left( \frac{t_{NM}}{\lambda_{sf}} \right)}$$

with $t_{NM}$ the thickness of the structure in spin current direction, $\lambda_{sf}$ the spin diffusion length and $\rho_{NM}$ the resistivity of the NM. In normal cases, this resistivity is not structure dependent. However, for thin structures this is not the case as scattering processes at surfaces and boundaries increase the resistivity and we have to modify the resistivity with
Theoretical concepts

Figure 2.4: SMR in NM|MOI(FMI) heterostructures. (a) $\mathbf{M} \parallel \mathbf{t}$, no spin current is transferred to the MOI, so the spin diffusion current counteracts the SHE spin current. Equivalent to open circuit condition. (b) $\mathbf{M} \perp \mathbf{t}$, some spin current is transferred to the MOI, reducing the spin diffusion current. Subsequently, the ISHE current is reduced, leading to a finite SMR. Equivalent to closed circuit condition. Taken from [25].

layer thickness and temperature dependent terms. The thickness dependent resistivity can be described by the Fuchs-Sondheimer approximation which can be derived from the Boltzmann equation [26]:

$$\rho(t, T) = \rho_\infty(T) \left[ 1 + \frac{3}{8(t - h)} l_\infty(1 - p) \right]$$

with $\rho_\infty$ the resistivity for infinite $t$, $h$ the interface roughness, $l_\infty$ the mean free path for infinite $t$ and $p$ the mirroring parameter. By replacing $\rho_{NM}$ in Eq. (9) by the Fuchs-Sondheimer approximation, we can get an accurate formula that shows the thickness dependency of various spin transport parameters. By measuring the SMR ratio for different thicknesses, we can determine those parameters, i.e. the conversion efficiency $\theta_{SH}$, the spin mixing conductance $g_\uparrow \downarrow$ and the spin diffusion length $\lambda_{sf}$ for any NM that is suitable for generating pure spin currents. In the case of this Bachelor thesis, this was attempted for tungsten.

The theoretical framework above describes the resistive properties of the SMR in longitudinal direction which is parallel to the charge current $j_q$. There are also transverse effects being generated that are described by Chen et al. , but we only investigate the longitudinal portion of the effect, as including other directions would be beyond the scope of this thesis.

### 2.4 Hanle Magnetoresistance

The SMR is not the only magnetoresistive effect that has a $\cos^2$ behaviour. Another effect is the Hanle magnetoresistance (HMR) as described by Dyakonov [27] in 2007. An electric current induces spin accumulation at the sample edges via the SHE. Applying a magnetic field perpendicular to the spin polarization causes a spin precession. This causes spin dephasing, decreasing the spin accumulation at the edges and resulting in an increase in resistance via the ISHE. To speak loosely, the magnetization $\mathbf{M}$ is replaced by the magnetic
field $H$ that now modulates the resistance. It shows identical angular behaviour in angle dependent magnetoresistance (ADMR) measurements [28] and occurs without the need of an MOI Layer. The HMR ratio for the longitudinal resistivity can be described by [27]

$$\frac{\rho_1}{\rho_0} = \left(\frac{\rho_1}{\rho_0}\right)_{H \to \infty} \left[1 - \left(\frac{1 + \sqrt{1 + x^2}}{2(1 + x^2)^{1/2}}\right)^{1/2}\right]$$

whith $x = \Omega \tau$, $\Omega = g \mu_B B / \hbar$ the spin precession frequency ($g$ is the Lande factor and $B$ the magnetic field) and $\tau$ the effective life time of the spin polarization. By measuring the field dependence of the MR this effect can be distinguished from the SMR.

### 2.5 Two Band Model Magnetoresistance

Materials with two different types of charge carrier have a different Hall effect (HE) than materials where we only need to consider one charge carrier. Magnetoresistive effects become a relevant contribution to ADMR measurements for some materials. Although the band structure is much more complicated for tungsten, we can use a two band model MR to roughly approximate the effect. We start with the linearized Boltzmann transport equation:

$$\left(\frac{\partial f_0}{\partial E}\right) v(k) \cdot (eE + v(k) \cdot \nabla g(k)) + \frac{e}{\hbar} (v \times B) \cdot \nabla \kappa g(k) = \left(\frac{\partial f}{\partial t}\right)_{\text{coll}}$$

with $f_0$ the equilibrium distribution, $E$ the energy, $E$ the electrical field, $B$ the magnetic field, $v$ the speed of the charge carrier, $k$ their wave vector, $g(k) = f(k) - f_0(k)$ and $(\partial f / \partial t)_{\text{coll}}$ the collision integral. If we consider an isotropic and parabolic conducting band and a homogenous material ($\nabla r = 0$), we can simplify Eq. (12) to

$$eE \cdot v(k) \left(- \frac{\partial f_0}{\partial E}\right) = \frac{g(k)}{\tau} + \frac{e}{\hbar} (v(k) \times B) \cdot \nabla \kappa g(k)$$

With the average collision time $\tau$. A proper derivation of this and the following expression can be found in [26]. If we consider only free charge carriers, we eventually arrive at a expression for the electrical field produced by the transverse current:

$$E = \frac{1}{\sigma_i} j_{q_i} + \frac{e \tau_i}{m_i} \left( B \times \frac{1}{\sigma_i} j_{q_i} \right)$$

with $\sigma_i$ the conductivity, $m_i$ the mass and $j_{q_i}$ the current for the charge carriers with charge $q_i$. From this we can derive the equations for the resistivity by using Ampere’s law:

$$\rho(B) = \frac{\rho_1 \rho_2 (\rho_1 + \rho_2) + (\rho_1 R_1^2 + \rho_2 R_2^2)B^2}{(\rho_1 + \rho_2)^2 + (R_1 + R_2)^2 B^2}$$

With $R_i$ the Hall constant and $\rho_i$ the resistivity of the two charge carriers. The magnetoresistance is then given by

$$MR = \frac{\rho(B) - \rho(B = 0)}{\rho(B = 0)}$$
and provides a theoretical description of the field dependency. We will use these equations later to describe additional magnetoresistive effects occurring at low temperatures.

2.6 Kohlers Law

A qualitative connection between the magnetic field dependence and the temperature dependence of the magnetoresistance in a metal was formulated by Max Kohler [29] and is known as Kohlers Law. The basic assumption is that all charge carriers have the same characteristic collision time \( \tau(T) \) that is temperature and purity dependent. Additionally an arbitrary eigenvalue distribution of the electrons is demanded. Solving the corresponding statistical function one arrives at an expression for the longitudinal magnetoresistance:

\[
\frac{\rho_1}{\rho_0}(x) = \psi(x)
\]

with \( x = \frac{B}{\rho_0} \) and \( \psi(x) \) a material dependent function. \( \psi(x) \) can be approximated with a parabola (i.e. the magnetoresistance as described by a two band model) for small arguments \( x < x_k \) where \( x_k \) is some critical value. At values \( x > x_k \) the behaviour of \( \psi \) is hard to describe theoretically. For the following argumentation we assume some differentiable function. We obtain the \( T \) dependence by differentiation of Eq. (17)

\[
\frac{d\rho_1}{dT} = \frac{d\rho_0}{dT} \left[ \psi(x) - x \frac{d\psi}{dx}(x) \right]
\]

The sign of this term is determined by the sign of \( d\rho_0/dT \) and the sign of the second factor. Assuming \( d\rho_0/dT < 0 \) (the reasoning behind this will become clear when we discuss resistive data measurements in Section 4.1), whether the magnetoresistance either increases or decreases with increasing temperature is determined by comparing \( \psi(x)/x \) and \( \psi'(x) \). The MR will decrease with temperature when \( \psi(x)/x > \psi'(x) \) and increase if it is the other way around. We will provide a intuitive graphical explanation in Sec 4.1. Eq. (18) gives us a relation between the field dependence and temperature dependence of an ordinary MR that can be used to distinguish ordinary MR-contributions from others.
3 Experimental Setup

In this section we discuss the structure of the samples used as well as the fabrication process. We go over how to realize different NM layer thicknesses on a single sample by using a new sputtering technique and we present the lift-off process used to pattern the NM layer. Additionally, we will explain the angle dependent magnetoresistance measurement technique that was used to measure the SMR and distinguish it from other magnetoresistive effects.

3.1 Sample Structure and Patterning Process

To achieve a NM | MOI interface, substrates of gadolinium gallium garnet (GGG; Gd₃Ga₅O₁₂) were prepared with a 1 µm layer of yttrium iron garnet (YIG; Y₃Fe₅O₁₂) that was grown using liquid phase epitaxy (LPE). The YIG provides the MOI layer on which a tungsten | tantalum bilayer is subsequently patterned. The addition of a Ta film is necessary to prevent the oxidation of the W layer. The sample structure is sketched in Fig. 3.1(a). As structure on the YIG layer we used a stripped down version of a Hall bar (Fig. 3.1(b)). Since we are only interested in longitudinal measurements, the transverse resistivity measurement points were removed from the design to optimize usage of the available surface area. The remaining bar has a length of 940 µm and a width of 20 µm with aluminium bond pads of 200 µm × 200 µm at the end. These dimensions were chosen to again optimize surface area usage while maintaining a high length to width ratio (l:w=47:1).

![Figure 3.1: Illustration of the sample setup. (a) A YIG film is grown on a GGG substrate and a W | Ta bilayer is applied by magneton sputtering. (b) Structure of the W | Ta bilayer, including Al bond pads. The strip is 940 µm long and 20 µm wide with 20 µm by 20 µm area being in contact with the bond pads. (c) Complete sample structure. Half of the strips are shifted to increase efficiency in using available space.](image)

The structural setup of a sample is shown in Fig. 3.1(c). A single sample holds a total of 48 bar structures. Every second structure is shifted with respect to its neighbours so the bond pads are using up available space better. The sample has a size of 5 mm by 6 mm, although only a section of 3.5 mm by 4.5 mm was used for the structures because of the nature of the
Experimental setup

lift-off process used to create them. The reasoning for this will be explained later in this section.

To apply the structures onto the YIG film, we used a multi step lift-off process that is illustrated in Fig. 3.2. As a first step (Fig. 3.2(b)), the resist AR-600K is spin coated on top of the YIG at 4000 rounds per minute and then baked out for 5 min at 170°C. Subsequently a second resist (AR-950K) is applied using the same procedure. Since the sample is insulating, we need an additional conductive resist t (SX AR-PC 5000/91.1) on the surface. It is coated at 4000 rpm and baked out for 2 min at 90°C. In the next step (Fig. 3.2(c)) the patterns are written using a NanoBeam nb5 EBL system and the conductive resist is subsequently removed by washing the sample with deionized water for 30 s and the resist bilayer is developed using an AR 600-56 developer for two minutes. To stop the developing process, the sample is washed twice in isopropanol for 5 s and 25 s respectively. We avoided writing the patterns too close to the sample edges because although the spin coating process produces a layer of constant thickness, the resist bulges up at the edges due to surface tension of the resist. Writing structures in this thicker part could risk removing parts of the pattern during lift-off. After writing and development, we are left with a resist layer where we do not want any metal to be present later. We can now apply a material layer by means of magneton sputtering (Fig. 3.2(d)). The final step is the resist lift-off from the substrate. The sputtered sample is placed in acetone for 10 min at 70°C. After that a pipette is used to create a fluid flow around the sample, removing most of the resist. The sample is then put into room temperature acetone in an ultrasonic bath for 1 min at the lowest power setting. What remains is the sample with the sputtered structure (Fig. 3.2(e)). This process is done three times for every sample. In the first run, alignment markers are applied to ensure the correct placement of the following runs. In the second run, the W | Ta bilayer bars are applied while in the final run the Al bond pads are patterned.

![Figure 3.2: Sketch of the patterning process. (a) The sample before the process. (b) AR-600K, AR-950K and the conductive resist are spin coated on the substrate. (c) The sample after EBL and development of the resins. (d) The sample after the sputtering process. (e) The sample after the lift-off process. The undercut structure of the resin ensures a clean lift-off. Taken from [16].](image)

3.2 Material Gradient Magneton Sputtering

Magneton sputtering is typically used to create material layers of a fixed thickness. A metal target is bombarded by charged ions (argon in our case). An impacting ion creates a collision
Experimental setup

A chain that usually ends with a particle of the target material getting emitted. It can travel a few centimeters before colliding with the sample and condensing onto it. The sample gets exposed to the target for a fixed amount of time at a predetermined sputtering rate. This way one can get a very thin film with a very precise thickness. In this thesis we tested a different sputtering method that used a moving wedge-shutter between the sample and the magneton to create a material gradient on the sample.

3.2.1 Sputtering Setup

The goal of the new technique is to do thickness dependent ADMR measurements on a single sample by creating a linear gradient along the long side of the sample (compare Fig. 3.1(c)). The bar structures are perpendicular to the gradient to ensure a constant thickness over a single structure. With this setup, a single sample holds $2 \times 29$ structures with different thicknesses. Note that the thickness difference over the bar width is neglected since the surface roughness is greater than this difference.

The samples were processed in the SUPERBOWLS which is composed of two deposition chambers, one for ferromagnetic and one for non ferromagnetic materials. The sample is placed on a substrate manipulator that positions it over the correct magneton. In Fig. 3.3 the setup of the magneton and the manipulator is illustrated. The manipulator contains the wedge-shutter which is used in normal cases to prevent accidental material deposition. To create the material gradient, we used the shutter to block a part of the particle stream. Before the magneton is activated, the shutter is positioned in such a way that it just barely covers the sample. It then gets retracted at a constant speed, exposing more and more of the sample to the magneton until it is fully visible. When a constant sputtering rate is

Figure 3.3: Superbowl sputtering apparatus. (a) face-to-face magneton sputtering setup. The speed of material deposition is controlled by the distance of the sample to the target and through power regulation of the magneton. (b) closeup of the wedge shutter protecting the sample. There is a gap of approx. 4 mm from the upper edge of the shutter to the sample (varies for different sample holders). (c) the Superbowl substrate manipulator. A sample is transferred from the loadlock with an arm and secured in place by the mask holder to secure a steady sputtering process. Adapted from [30].
maintained throughout the whole process we are left with a linear material gradient. The thickness as a function of position and sputter rate can be calculated by

\[ t_{\text{NM}}(x) = \left(1 - \frac{x}{l}\right) \frac{f_{\text{sputter}}}{v_{\text{shutter}}} \]  

(19)

with \( x \) the position along the long side of the sample, \( l \) the difference of start and end position of the shutter, \( f_{\text{sputter}} \) [nm/s] the sputter rate and \( v_{\text{shutter}} \) the shutter speed. Eq. (19) describes an ideal situation where the shutter perfectly blocks all deposition beyond its current position.

In practice the deposition process is more complicated. As displayed in Fig. 3.3(b), there is a gap between the sample and the wedge shutter. This changes the conditions of the material deposition. Fig. 3.4(a) and (b) show the sputtering conditions for different distances of the wedge-shutter to the sample. When travelling from the magneton to the sample, the material particles can collide with the Ar atoms (the sputtering process is done in an Ar atmosphere at \( 5 \cdot 10^{-3} \) mbar) and other sputtered particles and gain a velocity component in transverse direction to the particle stream. This allows them to pass through the gap between shutter and sample where they get deposited in an area that should be blocked. At best this creates a vertical shift of the gradient, i.e. a layer of constant thickness is added to the gradient while leaving its form intact (this means adding a fixed thickness \( t_a \) to Eq. (19)). However, the amount of additional material deposited is very hard to approximate.

![Figure 3.4: Illustration of gradient magneton sputtering. (a) A snapshot mid-process in an ideal situation. The shutter blocks material deposition in area II, while area I receives the full deposition rate. (b) The particles acquire a transverse velocity. Deposition in area II is still blocked, however there is additional deposition with a lower rate beyond the shutter position. (c) Final result of the sputtering process. A W layer with a thickness gradient is deposited on top of the YIG and a thin Ta layer is added to prevent oxidation of W.](image)

3.2.2 Thickness Measurements

The area where additional material gets deposited was tested at different distances between the target and the sample holder. The wedge-shutter was locked at the center of the sample during the sputtering process to make the area visible. For every test sputtering, the area was visible to the naked eye at a width of approx. 1 mm or larger (up to approx. 2 mm). Moving the mask holder further away from the target increases the width quite significantly. A longer distance from target to sample means there is more time for scattering events to take place so we expect an increase in transverse velocity and subsequently a bigger deposition area in the blocked region. This limits the sputtering rate that can be used with gradient sputtering since an increased distance reduces the sputter rate but makes the
Experimental setup

The layer thickness of the deposited material was measured using atomic force microscopy (AFM) and x-ray diffraction (XRD). Exemplary measurements are shown in Fig. 3.5. The AFM uses the force a sample exerts on a small probe tip. The deflection is measured by a laser and translates to an accurate image of the sample's surface (Fig. 3.5(a)). The AFM was used to measure the W | Ta bilayer thickness of multiple bar structures. Since we cannot distinguish between the bilayers different composites with an AFM measurement, we need to measure the Ta layer thickness by means of XRD. Monochromatic x-rays get diffracted under low angles of incident resulting in a diffraction pattern. Fitting the reflectometry (Fig. 3.5(b)) gives the layer thickness as a parameter. This method does not work for the gradient, since the x-rays get diffracted differently by different parts of the sample, resulting in a washed out reflectometry. However, XRD can give us the Ta layer thickness as it is constant. For every sputtering process we installed an additional silicon oxide sample on the mask holder to determine the Ta thickness. The sample was placed in a spot where a gradient would be sputtered as well, which makes it impossible to measure the Ta layer. For future iterations of gradient sputtering placing the Si sample in a spot where it is completely exposed during the deposition is advisable. For this thesis, a sample from a different sputtering process with the same Ta layer settings was used to approximate the layer thickness via XRD measurement.

The XRD reflectometry curve is fitted by the program leptos to extract layer thickness and roughness parameters. For the Ta | TaO layer we get 1.56 nm | 2.37 nm which allows us to estimate that the layer of the structured sample has a combined height of approx. 4 nm. By adding this layer to our expected value of the gradient we can compare the expected thickness with the AFM measurements. This is shown in Fig. 3.6. The deposition did not produce a linear behaviour as expected. Ignoring the thickest structures allows a linear

![Graph](image-url)
Figure 3.6: structure thickness measured by AFM versus expected thickness of the YIG M1-E2 sample. The dashed line indicates a perfect match. The thinner structures can be linearly fitted, however with a different slope than expected.

fit, but not with the same slope as expected. For this thesis the linear fit will be used since the thicker structures do not produce a MR for the particular set of samples that were used. There are a number of factors that can cause non linear behaviour. In our previous explanation the material deposited in the blocked region was constant throughout the whole deposition process. This is a simplification and the actual deposition in this area may be much more complicated. The area were the material is deposited is much bigger than initially expected and it widens as the sample is moved further away from the magneton. Another potentially crucial factor is the sputter target itself. The W forms an oxide layer that changes the sputter rate and makes the deposition rate inconsistent. To avoid this, the target was sputtered for at least half an hour before using it for deposition after the SUPERBOWL chamber had been opened. This should remove the oxide layer, but since the sputter rate can not be measured while simultaneously depositing, a variation of the rate could have caused the non linear behaviour as well. The shutter opening speed may also have varied, although that is not a very likely explanation since the shutter position turned out to be very precise as determined in various test depositions.

In conclusion, gradient sputtering can produce structures with different thicknesses on a single sample, but it is advisable to measure the thickness of each structure individually to avoid using the wrong thickness values for subsequent calculations. An auxiliary substrate is needed if an additional layer e.g. an oxidation protection is applied and has to be positioned on the mask holder in a position where it is either completely covered by the shutter or completely exposed to the magneton during gradient deposition. The deposited layer will be thicker than can be calculated from the shutter speed and deposition rate unless the distance between sample and shutter can be reduced significantly, since the plasma can
get into the space between them. Maintaining a sample position further away from the magneton will make this effect worse. It is not advisable to use this method to create steep gradients over short distances.

### 3.3 Measurement Technique

As described in section 2.3, the occurrence of an SMR is dependent on the angle between the transverse to the current density $j_C$ and the magnetization $M$. We can measure the SMR amplitude by performing a full rotation of $M$. This technique is called angle dependent magnetoresistance (ADMR) which was mentioned already in section 2.3. The term is misleading since the SMR is technically not angle dependent but dependent on the orientation of $M$, but we will use the term loosely since it simplifies the discussion. We can control the direction of $M$ by applying an external magnetic field $h = H/|H|$ to the MOI that is bigger than its saturation magnetization $M_s$. There are three axes that define three rotation planes for an ADMR measurement. First, the in plane (IP) orientation is defined by the normal $n$ and the angle $\alpha$ (Fig. 3.7(a)) where $h$ is rotated in the interface plane. Additionally, there are two out of plane orientations. The OOPJ plane where $h$ is rotated around the current vector $j_C$ (Fig. 3.7(b)) with the angle $\beta$ and the OOPT plane that is defined by the transverse vector $t = n \times j_C$ and the angle $\gamma$ (Fig. 3.7(c)). As we recall from section 2.3, the resistivity of the NM layer is proportional to $m^2$. From this behaviour we can distinguish the SMR from other magnetoresistive effects. We expect to see a $\cos^2$ behaviour of the SMR in the IP and OOPJ rotation planes as we rotate $h$. In the OOPT plane however, $h \perp t$ is true for all angles $\gamma$ so $m_t$ is always zero. According to Eq. (8) we will not see a change in resistance of the NM layer in this case.

To conduct ADMR measurements, the prepared samples are glued on a chip carrier that also provides 20 contact points to connect measurement devices to. The sample structures are connected to them by bonding wires to the bond pads and the contacts on the carrier. Pictures of a sample on the chip carrier and before bonding are in Fig. 3.8. The sample is then mounted on a dipstick and inserted into the MORIA superconducting magnet cryostat (Magneto-Optical-Resistance-Investigation-Apparatus) with a 1D field and a maximum field strength of $\mu_0 H = 7$ T. The dipstick is rotated in the static magnetic field by a motor,
allowing us to rotate the sample in one plane. Upon switching the rotational plane, the sample needs to be remounted within the dipstick. In addition, the cryostat is able to cover a temperature range of $3.5 \, \text{K} < T_{\text{cryo}} < 300 \, \text{K}$ with a precision of $\pm 10 \, \text{mK}$, although uncertainties of $\pm 1 \, \text{mK}$ can be reached in certain cases. The temperature is measured at the dipstick close to the sample position.

We measured both thermal and resistive effects by applying a constant charge current to the bar structures. While thermal effects in the NM are generated by Joule heating due to the structure’s finite resistance, the resistive response arises from magnetoresistive effects. Both responses can be obtained by measuring the voltage between both ends of the bar. We can distinguish between the contributions by using a current reversal measurement. The current direction is repeatedly switched from positive to negative and the respective voltages $V_+$ and $V_-$ are measured using a nanovoltmeter. The resistive contribution is dependent on the current direction, so we can define the resistive answer as

$$V_{\text{res}} = \frac{V_+ - V_-}{2}$$

(20)

The thermal contribution however is independent of current direction and is eliminated in this expression. To calculate the thermal answer, we use the symmetric function

$$V_{\text{therm}} = \frac{V_+ + V_-}{2}$$

(21)

Analogously, resistive effects are eliminated in this expression aswell. Note that $V_-$ has a negative sign, so we obtain a thermal answer only if $|V_+| \neq |V_-|$. Since the SMR is a resistive contribution, we expect the thermal answer to be $\approx 0$ at lower currents. We conducted the current reversal method anyways to ensure our contributions were purely resistive.

Up to six bar structures could be measured simultaneously. The wiring is shown in Fig. 3.9. A current $I = 10 \, \mu\text{m}$ is applied with a Keithley 2400 Sourcemeter. The bar structures are all supplied with the same source signal and are wired in series so the current $I$ is equal for all structures. For each bar the voltage $V_n$ between the two ends is measured with a Keithley 2182 Nanovoltmeter as shown.
In all measurements we use a repeating filter setting which averages the voltage 30 times intrinsically for each voltage readout. The output is a single value, so the exact values cannot be retrieved in hindsight. The current reversal is performed three times for each angular configuration to increase signal to noise ratio. The timing and procedure is taken out and recorded by a Labview program. For a set temperature $T_{\text{cryo}}$ and field $\mu_0 H$, the sample is rotated from $-20^\circ$ to $380^\circ$ and back in steps of $10^\circ$. The backwards rotation is necessary to exclude any hysteretic behaviour of the measurement. At each angle, a current reversal measurement is conducted.
4 Magnetoresistance in YIG | W

The investigation of the SMR in tungsten is motivated both from an economic standpoint as well as a scientific one. Platinum which is the most common material when producing spin currents is very expensive, while W is roughly a tenth of the material cost. Additionally W has been observed to have a spin hall angle of up to 0.49 [17], depending on the oxidation and phase of the layer [31]. While standard α-phase W has a spin hall angle of 0.07, a pure but metastable β-phase W phase can have a spin hall angle of up to 0.4 [32] without using oxygen during film growth. In this section we want to investigate the SMR in W | YIG heterostructures by ADMR measurements for varying magnetic field, temperature and thickness to determine the optimal conditions for the SMR in W. The samples were prepared as described in Sec. 3.3. The measurements were carried out with a fixed external magnetic field in the interval $0.05 \mu_0 H \leq 7 \text{T}$. The samples are rotated in the three planes (IP, OOPJ, OOPT) for each static field. This procedure is repeated for different temperatures with $5 \text{K} \leq T \leq 300 \text{K}$. This allows us to observe dependence of orientation, field, temperature and thickness for the MR in W.

![Figure 4.1](image)

**Figure 4.1:** Normalized ADMR measurement results for $t = 6.7 \text{nm}$. The panels (a), (b) and (c) were measured at $T = 300 \text{K}$ while panels (d), (e) and (f) were measured at $T = 5 \text{K}$. Panels (a) and (d) show the IP orientation, (b) and (e) show the OOPJ orientation and panels (c) and (f) show OOPT measurements for said temperatures.

Since we conduct DC measurements resistance and voltage are proportional to each other as described by Ohm’s law. Thus we can obtain the magnetoresistance by normalizing the
angle dependent voltage to its minimum by using the following formula:

$$MR = \frac{V - V_{\text{min}}}{V_{\text{min}}}$$  \hspace{1cm} (22)$$

An exemplary graph of the MR in the three rotation planes at different external fields between 1 T and 7 T for $T = 300$ K and $T = 5$ K is depicted in Fig. 4.1 for a 6.7 nm structure and in Fig. 4.2 for a 10 nm structure. The data is shifted by a few degrees to account for a present offset. In IP orientation plane, we observe minima at $\alpha = 0^\circ$ and $\alpha = 180^\circ$ where the magnetization $M$ is collinear to the spin polarization $s$. In this position, no spin transfer torque (STT) is applied and the resistance of W behaves as if no YIG layer is attached. Transitioning to $90^\circ$ and $270^\circ$, spin transfer into the YIG is maximized when $M \parallel s$ as described by Eq. (7). The MR curve shows a $\cos^2(\alpha)$ behaviour as expected from Eq. (8).

In OOPJ orientation (panels (b) and (e) of Fig. 4.1 and Fig. 4.2) the same argument can be applied. We observe minima at $90^\circ$ and $270^\circ$ where $M \parallel s$ and maxima at $0^\circ$, $180^\circ$ and $360^\circ$. At $300$ K, no MR is observed in the OOPT orientation just as expected for a pure SMR response. At $5$ K we observe an additional positive MR in OOPJ and OOPT orientation of higher amplitude than the MR in IP rotation plane. For the OOPJ rotation plane this additional MR exhibits the same angle dependence as the SMR (panel (e) and (f) of Fig. 4.1 and Fig. 4.2). The MR is of the same periodicity in the angle $\beta$ and $\gamma$ as a $\cos^2$ function but does not resemle a $\cos^2$ behaviour. We attribute this additional MR to an ordinary

Figure 4.2: Normalized ADMR measurement results for $t = 10.1$ nm. The upper row was measured at $T = 300$ K while the lower row was measured at $T = 5$ K. Panels (a) and (d) show the IP orientation, (b) and (e) show the OOPJ orientation and panels (c) and (f) show OOPT measurements for said temperatures.
magnetoresistance in the W structure that is not spin sensitive. The strong field dependence suggests a field induced magnetoresistance with a dependence of the angle between the field orientation $\mathbf{h}$ and the surface normal $\mathbf{n}$. We will study this additional MR in more detail in section 4.1 when we discuss the field dependency of all magnetoresistive effects. To further investigate this effect, the sample structure could be replicated on a substrate without an MOI layer to negate any SMR contributions (with the exception of the Hanle magnetoresistance). This was not realized during this thesis due to time constraints.

When comparing the in plane measurements, we observe an increase of the signal amplitude for lower temperatures and higher field strengths. The temperature dependence will be discussed in section 4.2. Comparing Fig. 4.1 with Fig. 4.2 shows a thickness dependence of the signal for all orientations. The amplitude is reduced as the thickness increases. The thickness dependence will eventually lead to acquiring the spin current parameters $\theta_{\text{SH}}$ and $\lambda_{\text{sd}}$ and will be dealt with in section 4.3.

4.1 Magnetic Field Dependence

To better analyze the magnetic field dependence, we fitted the measured voltage for each ADMR measurement with a $\cos^2$ function. The MR ratio is obtained by dividing the amplitude $A$ by the y-intercept $y_0$ which is equivalent to the base voltage $V_0$. Since we can apply Ohm’s law here, this ratio is equivalent to the resistance ratio.

We already saw in Fig. 4.1 and Fig. 4.2 that the MR in tungsten has a strong field dependence. This is in contrast to comparable measurements for YIG|Pt [15]. This section will focus on the field dependency of both the SMR and the ordinary MR at low temperatures. Fig. 4.3 shows the field dependence of the MR at different temperatures for two different thicknesses (6.7 nm and 10.1 nm). In IP orientation the MR follows a linear behaviour that increases with field strength at all temperatures. The same is true for OOPJ orientation at temperatures of 75 K and above. Strong magnetic field dependence of the SMR was observed before in YIG|Ta [16], where it was accounted for by pinning effects. Since W/Ta bilayers were used during this thesis a contribution of Ta to the SMR can not be excluded, especially since XRD measurements (cmp. Sec. 3.2) show a thin amorphous Ta layer that is not oxidized. This layer can contribute to the creation of a spin current via SHE but since it does not have an MOI interface we expect the SMR behaviour to be determined by the W layer. This topic will be further discussed in Sec. 4.2. For $\mu_0H = 7$ T, we find comparable SMR amplitudes at low temperatures to those in YIG|Pt that Meyer et al. found [15]. For higher temperatures the SMR in YIG|Pt is a lot higher and makes Pt more promising when conducting non local magnetotransport phenomena. Similarly the strong field dependence limits the usage of W for magnon transport since it is constrained by the high fields that are required to get a good SMR signal from W. This indicates that W is not a good alternative for magnetotransport experiments.

At lower temperatures we see a deviation from the expected SMR behaviour due to the ordinary MR in OOPJ and OOPT orientation, which follows a steeper curve and seems to saturate for larger magnetic fields. A simplified explanation can be given by using the two band model MR introduced in 2.5. This description only produces a transversal MR
Figure 4.3: Field dependence of the MR at different temperatures between 5 K and 300 K. The upper row is for $t = 6.7$ nm, the lower row for $t = 12.2$ nm. (a) and (d) are in plane orientation, (b) and (e) are OOPJ orientation and (c) and (f) are OOPT orientation.

when $h \perp j_c$ which produces the same angular dependency we observe. This is due to the assumption of a spherical fermi surface of the material.

In Fig. 4.4 the MR in OOPJ orientation is plotted for a single structure thickness, showing a saturating behaviour at high magnetic fields. The two band model produces a parabolic behaviour of the MR at small field strengths which is contrary to what we observe. Fitting a normalized form of Eq. 15 can reproduce the saturating behaviour at high fields with the two charge carriers having only a slight difference in their Hall constants. Including additional types of charge carriers or using a high field approximation for open fermi surfaces like described in [25] may result in better descriptions of the MR but this would require additional measurements to better the fit parameters. The fermi surface of tungsten is rather complicated [33], so a two band model can not describe the occuring effect sufficiently. Similar to the SMR, this MR is thickness dependent and is stronger the thinner the structure. We can further confirm that the MR is caused by the electronic structure of W by applying Kohlers law to the field dependency. As we discussed in Sec. 2.6 field and temperature dependence are dependent on eachother. Since the two band model does not reproduce the field dependence accurately, for the following arguments we use a polynomial fit instead. Using Eq. (18) we can see that we can deduce the behaviour in temperature from the function $\psi_{long}$ that describes field dependence. Specifically, we compare the sign of the two terms $\rho_0 \psi/B$ and $\rho_0 \cdot d\psi/dB$. Fig. 4.5, the two terms and $\psi$ are shown. We can identify the parts of Eq. (18) as $\rho_0 \cdot d\psi/dB = \tan \phi$ and $\rho_0 \cdot \psi/B = \tan \xi$, where $\phi$ is the angle between the $B$-axis and the connection line from the origin to a value of $\psi$ and $\xi$ is the angle between the $B$-axis and the tangent of $\psi$ at the same point. If $\alpha > \beta$, the MR decreases with temperature.
Figure 4.4: Field dependence of the magnetoresistance in OOPJ orientation at $t = 6.7$ nm and $T = 5$ K. The data is fitted with Eq. (15) that was derived in the two band model.

Figure 4.5: Field dependence of the magnetoresistance in OOPJ orientation at $t = 6.7$ nm and $T = 5$ K. The data is fitted with a polynomial of second order. The black tangential line to the function has an angle $\beta$ to the x-axis, the blue connection line between the origin and the tangential intersect has an angle $\alpha$ to the x-axis.
and vice versa. In case of our measurements, $\alpha > \beta$ is true for all structures, so we expect a decrease of the MR with increasing temperature. This is true for thin structures where we observe $d\rho_0/dT<0$.

### 4.2 Temperature Dependence

In Fig. 4.6 the temperature dependence of the MR is plotted in all orientations for $t = 6.7 \text{ nm}$ and $t = 10.1 \text{ nm}$ and different fields. The temperature dependence follows the same behaviour for thin and thick W layers. In IP orientation, the MR grows in linear fashion as temperature decreases until around 75 K and then drops again for lower temperatures. In OOPJ orientation we observe the same dependence until 75 K, where the ordinary MR arises and overlays the SMR as can be confirmed by the temperature dependence in OOPT orientation. The temperature dependence is different than in YIG|Pt heterostructures\cite{34,15} where the MR decreases with temperature for similar thickness and production method of the YIG film (LPE, sputtering). The signal in YIG|Pt is approximately a factor of 5 bigger than in YIG|W. At the maximum field strength of $\mu_0 H = 7 \text{ T}$, the MR signal is of comparable size to YIG|Pt at low temperatures (smaller by a factor of 2). The temperature dependence of the SMR is strongly dependent of the quality of the NM|MOI interface, so additional samples may be required in the future to verify this dependence as well. Whether this dependency still holds for $\beta$-phase W might be a subject worth investigating as well. Initially, this was planned to be determined in the scope of this thesis, however, complications and difficulties in the gradient sputtering process limited the amount of measurements that

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.6}
\caption{Temperature dependence of the magnetoresistance for the three orientations IP ((a) and (d)), OOPJ ((b) and (e)) and OOPT ((c) and (f)). The upper row is for $t = 6.7 \text{ nm}$, the lower row for $t = 10.1 \text{ nm}$}
\end{figure}
could be carried out. We can exclude a significant contribution of amorphous Ta in the SMR signal from the temperature dependence. As discussed earlier, a thin Ta layer that is not oxidized remains on our samples. Comparing the temperature dependence of YIG|W to YIG|Ta we see a different behaviour for both elements. The SMR amplitude drops for W at very low temperature where that of Ta increases [16] which confirms that the Ta contribution is insignificant.

The ordinary MR has a strong dropoff with increasing temperature. To verify that the MR is indeed ordinary, this behaviour needs to be in agreement with the description from Sec. 2.6. From our results for the field dependence we expect a decrease with temperature, which is precisely what we observe. This confirms that the observed MR arises due to the electronic structure in our samples.

4.3 Thickness Dependence and Resistivity

The thickness dependence of the SMR as described by Eq. (9) is dependent on the spin mixing conductance $g_{↑↓}$, the Spin Hall angle $\theta_{SH}$ and the spin diffusion length $\lambda_{sf}$. We can extract these from thickness dependent SMR measurements. Since we are dealing with very thin structures, the thickness dependence of the resistivity must be included as well. Fig. 4.7 shows the resistivity as a function of temperature and thickness. We can see that for thin structures (Fig. 4.7(a)), the resistivity decreases with temperature while it increases for thicker structures (Fig. 4.7(b)). The transition occurs between 12.1 nm and 13.5 nm. We assume that these different behaviours come from the two different crystalline phases from W. We assume that thinner structures contain mostly amorphous W while thicker structures are in a combination of crystalline $\alpha$- and $\beta$-phase. The phase is very sensitive to the sputtering conditions, like for example chamber pressure, sputter time and deposition rate. A similar behaviour was found in YIG|Ta structures that has two phases similar to W [16].

The thickness dependence of $\rho$ is shown in Fig. 4.7(b). We can see a decrease of $\rho$ with increasing thickness as expected. From theory we expect a $1/t$ behaviour as described by Eq. (10) which is not the case for our samples. Again, the most likely explanation is the contribution of two phases that have a different resistivity $\rho_{∞}$ and different mean free electron paths $l_{∞}$. Fitting two separate functions will result in an overparametrization as we would have three parameters for each crystalline phase. Although our data does not
align up well with theory, for further calculations of the spin Hall angle we need a function that describes $\rho(t, T)$. For each temperature a second order polynomial was used to fit the resistivity. These functions are needed to express $\rho_{NM}$ in Eq. (7).

The thickness dependence of the SMR is shown in Fig. 4.8 for all orientations at 5 K and 300 K. Before we discuss the SMR thickness dependence, we return to the ordinary MR that is visible in OOPJ and OOPT orientation at 5 K (Fig. 4.8(b) and(c), respectively). We can see a strong thickness dependence of the MR in both orientations where the MR decreases with increasing thickness. We observe this behaviour for all magnetic field strengths that are sufficiently high to tilt the magnetization into the out-of-plane direction. For studying the SMR in W, we focus on IP measurements since no other MR contributions are present in this orientation. We extract a decreasing SMR signal with increasing thickness for both 5 K and 300 K. This still holds for all temperatures investigated. At 300 K we can additionally compare the IP signal and the OOPJ signal. The SMR ratio matches well between both orientations inside of measurement uncertainties, which is in perfect agreement with our SMR theory.

Using the IP orientation data we can extract values for spin mixing conductance, Spin Hall angle and Spin diffusion length. Those parameters are highly dependent on each other, which makes an independent extraction of all three parameters from our measurements impossible. Fig. 4.9(a) shows exemplary data simulations of Eq. (9) (red line and blue
Magnetoresistance in YIG|W (line) for 300 K IP measurements (purple diamonds). For this simulation, $g_{\uparrow \downarrow}^r$ was fixed at 0.81 nm$^{-2}$. The spin mixing conductance was taken from spin pumping experiments in CoFeB|W heterostructures conducted by Jhajhria et al. [35]. Although $g_{\uparrow \downarrow}^r$ is material dependent, this value is comparable to that of YIG|Pt at $g_{\uparrow \downarrow}^r = 1$ nm$^{-2}$ which is most commonly used as a reference for SMR experiments with other materials. We further fixed $\theta_{SH} = 0.08$ and $\theta_{SH} = 0.04$. Earlier research regarding the SHA of tungsten found $\theta_{SH}^{\alpha-W} = 0.07$ [17], so these values were chosen to approximate the upper and lower bounds. The corresponding curves match almost perfectly, since we can find pairs of $\theta_{SH}$ and $\lambda_{sf}$, where we obtain the same function. The measurement data does not cover the SMR at very thin structures, which are essential to find a distinct tuple ($\theta_{SH}$, $\lambda_{sf}$). Especially, since the SMR signal exhibits a maximum at $t \approx 2\lambda_{sf}$. At thinner structures we obtain a weak spin accumulation and the effect declines, while we reduce the signal in thicker structures due to diffusion losses. Due to difficulties in the sputtering process we produced higher thicknesses on the samples as expected and subsequently did not measure structures near $2\lambda_{sf}$. This is confirmed by the fit parameters shown which estimate $0.78 \text{ nm} < \lambda_{sf} < 1.66 \text{ nm}$ at 300 K. Nevertheless, with this approach we can reasonably estimate upper and lower bounds of the spin diffusion length for the temperature range we investigated to gain information on the feasibility of W for magnon transport experiments.

Fig. 4.9(b) shows the temperature dependence of $\lambda_{sf}$ simulated with constant spin mixing conductance $g_{\uparrow \downarrow}^r = 0.81$ nm$^{-2}$ and two different SHA values. For both SHA values, $\lambda_{sf}$ shows an increase with decreasing temperature up until $T = 75$ K from where it remains approximately constant. Since the simulation gives more reasonable values of $\lambda_{sf}$ for $\theta_{SH} = 0.08$, it is likely that the true SHA is near this value. The simulation with $\theta_{SH}$ produces spin diffusion lengths of up to 3.66 nm at 75 K which would result in the maximum of Eq. (9) being placed at around 7 nm. We do not observe this in our data, suggesting that the SHA is greater than 0.04. Additionally, this suggests that there is mostly $\alpha$-phase W present in our samples, as mixed phase tungsten has a higher SHA of $\theta_{SH}^{\beta-W} = 0.18$ where $\alpha$-phase W

![Figure 4.9](image-url)
Magnetoresistance in YIG/W

is around 0.07 \cite{17}. This confirms our assumption made earlier that a high sputter rate produces mostly $\alpha$-W. The temperature dependence strongly resembles that of the SMR ratio shown in Fig. \ref{fig:4.6}(a),(d). Fixing two parameters carries over the temperature dependence of the SMR onto the remaining free parameter. Similar results were obtained for YIG/Pt by Meyer \textit{et al.} \cite{15}, although their data suggests that not the spin diffusion length but the SHA is dependent on temperature. The spin diffusion length increases with decreasing temperature due to reduction in spin-flip scattering events. The observed temperature dependence does decrease for very low temperatures, however, which is most likely to be caused by impurities in W also contributing to electron scattering. This causes the spin diffusion length to no longer increase at low temperatures. While additional research is necessary to confirm this assumption, it would be beyond the scope of this thesis to consider both cases of temperature dependent SHA and $\lambda_{sf}$. The spin mixing conductance is mostly constant in temperature in most materials, which is why we fixed it in our simulations as well.

For low temperatures $\theta_{SH}$ and $\lambda_{sf}$ are similar to that of YIG/Pt and YIG/$\alpha$-Ta. For Pt, spin diffusion lengths between 1.5 nm and 1.6 nm were reported \cite{15} which is comparable to our lower bounds spin diffusion length simulation. In YIG/Ta two different thickness regimes were found \cite{16} similar to our observations in W where thin structures resistivity increases as temperature decreases and the opposite behaviour for larger thickness structures. For Ta spin diffusion lengths were reported in an interval of approx. $0.6 \text{ nm} \leq \lambda_{sf} \leq 1.2 \text{ nm}$ for thin structures and approx. $2 \text{ nm} \leq \lambda_{sf} \leq 5 \text{ nm}$ which is again similar to the upper and lower bounds for W we established. This leads us to $\alpha$-W to be a reasonable alternative for magnon transport at lower temperatures, although one has to consider the impact of the ordinary MR on said experiments. Additionally, the resistivity of our samples is very high when compared to Pt, which causes more Joule heating of the NM. This increases unwanted energy dissipation.

\section{Sample Aging}

Since W is an easily oxidizing material, we conducted ADMR measurements about two months apart with samples from the same sputtering runs. If we find a significant oxidation of W, we expect the resistivity as well as the SMR to increase from previous studies of SMR in W, which suggest a much bigger SHA for oxidized W thin films \cite{17}. The comparison is depicted in Fig. \ref{fig:4.10}. We observe a similar curve for the resistivity (Fig. \ref{fig:4.10}(a)), but the data is negatively shifted by about 10\% with respect to the $\rho$-axis. This suggests that the W structures did not oxidize over the time period, which is also confirmed by XRD measurements of equally aged single thickness samples with a fixed thickness W/Ta bilayer. Here no sign of oxidized W showed up. A more likely explanation is that a part of the $\beta$-phase W transformed into $\alpha$-W since it is metastable. This results in less $\beta$ to $\alpha$ phase boundaries in the structure and therefore reduces the resistivity as boundaries between crystalline phases add to electron scattering and subsequently to resistivity. However, this is in contrast to our previous measurements suggesting that little $\beta$-W was present in the first place. It is also in contrast to the SMR thickness dependence shown in Fig. \ref{fig:4.10}(b) where we can not observe a noticeable difference between the fresh and aged samples. One possible
Figure 4.10: Comparison of a fresh sample (YIG M1-E2) and a sample that is two months old (YIG M1-E4). (a) Resistivity as a function of thickness. The aged sample has an overall lower resistivity while the curve is very similar. (b) SMR as a function of thickness. Both samples show no significant difference in their SMR amplitude.

explanation is that the decrease in resistivity due to phase conversion gets counteracted by a decrease in magnetoresistance. In the limit of almost no $\beta$-W this is a reasonable behaviour. The fact that the SMR does not change significantly is a good thing however, since this was a concern with regards to the long time usability of W as a NM for spin current generation. Whether this is still the case for pure $\beta$-W remains to be determined however, mostly because we can deduce from our experiments that $\alpha$-W does not have significant advantages over YIG|Pt both in ease of use as well as in spin current generation efficiency.

4.5 Summary

In this chapter we investigated the magnetoresistive effects in YIG|W heterostructures dependent on external magnetic field strength, temperature and layer thickness. We found that the effect peaks at around 75 K. in IP and OOPJ orientation. Below this temperature, an additional magnetoresistive effect arises in OOPJ and OOPT orientation that could be identified as a positive ordinary magnetoresistance caused by the electronic band structure of W. This ordinary MR obeys Kohler’s law. Similar to the SMR in YIG|Ta \cite{16} we found a strong field dependence of the SMR that is not observed in YIG|Pt and is not described in SMR theory by Chen et al. \cite{24}. The field dependence is most likely caused by pinning effects at the interface of YIG|W. The resistivity of W showed two different behaviours with temperature that may be caused by a change in crystalline structure. We expect the thin layers to be amorphous while the thicker layers may be crystalline, mostly $\alpha$-phase W. Thickness dependent SMR results show the SHA to be in agreement with earlier research results for $\alpha$-W. The temperature dependence of the spin diffusion length $\lambda_{sf}$ was simulated using the experimental results. Here we find an increase in $\lambda_{sf}$ with decreasing temperature which saturates at 75 K.
5 Summary and Outlook

We investigated the generation of spin currents in YIG|W hererostructures within this thesis. We first developed a new sputtering technique that allowed us to conduct angle dependent magnetoresistance (ADMR) measurements for different layer thicknesses of W on a single sample, thereby enabling classification of the spin Hall angle (SHA), the spin diffusion length and the spin mixing conductance in one measurement run. The new technique creates a material gradient over the sample that can be tuned in thickness and slope by changing sputter parameters and sample structure. We used a moving wedge shutter during the deposition process to block parts of the sample to create the gradient. As it turned out, a requirement for this to work properly is a small distance between shutter and sample. This reduces material deposition in regions blocked by the shutter that arises due to transverse particle velocity. We found that creating a linear gradient requires a constant sputter rate and close positioning of the sample to the magnetron to reduce unintentional deposition, should there be a gap between shutter and sample. The patterning was accomplished using a lift-off process now commonly used at the Walther-Meißner-Institute.

After realizing samples with different layer thickness structures, we performed ADMR measurements of the longitudinal resistivity in YIG|W, where the magnetization of YIG is rotated using an external magnetic field. This was done in three planes. One in plane of the sample and two out of plane where the magnetization was rotated around the current direction and its transverse. These measurements were conducted using different field strengths in the interval \(0.05 \leq \mu_0 H \leq 7\) T and different temperatures in the interval \(5 \leq T \leq 300\) K for each orientation. The samples were installed in the MORIA cryostate and measurements at a DC current of \(I = 10\) µA were conducted. We observed a resistive \(\cos^2\) dependence in IP and OOPJ orientation as described by SMR theory \([24]\). We extracted the relative ratio of change in resistivity (SMR amplitude) from these measurements. Using these amplitudes we investigated temperature, field and thickness dependence of the magnetoresistance. We observed an increase of the SMR amplitude with decreasing temperature, with a saturation around 75 K from where the amplitude decreases again. Additionally we observed a magnetoresistive effect besides the SMR at low temperatures in out of plane orientations. Since it showed a saturating field dependence which matched its temperature dependence, described by Kohlers law \([29]\), we identified it as an ordinary MR caused by the electronic band structure of W.

In contrast to SMR theory, we observed a strong field dependence of the SMR. Although such field dependent effects are usually associated with Hanle magnetoresistance (HMR), we excluded this as a possible explanation since the HMR has a quadratic field dependence that we did not observe in our samples. We attribute the field dependence to pinning effects at the YIG|W interface.

We also investigated the thickness dependence of resistivity and magnetoresistance in YIG|W heterostructures. The resistance revealed two thickness domains. In thin structures (<13 nm) the resistivity decreases with increasing temperature, while thicker structures (>13 nm) show an increase in resistivity with temperature. This is caused by the crystalline structure of the W layer, where a transition between amorphous and crystalline W takes place in this thickness regime.
The SMR showed a strong thickness dependence as described by Chen et al. [24]. Using the thickness dependent measurements in IP orientation we determined the SHA to be between 0.04 and 0.08 which confirms that our samples mostly consist of α-phase tungsten. We determined the spin diffusion length to be dependent of temperature and increasing in analogous fashion to the SMR amplitude. Upper and lower bounds of the spin diffusion length were estimated and are comparable to YIG|Pt [15]. Previous research only studied the spin current properties of W in combination with conducting magnetic materials like CoFeB, while we investigated its interface properties with an insulating magnetic material for the first time to better compare the results obtained for Pt and Ta. Because of the lower SHA in combination with a higher resistivity, we determine that α-W is not a good alternative to Pt for spin current generation. Finally, we investigated the long term durability of YIG|W by measuring samples in a time interval of two months. Although we observe a decrease in resistivity, the SMR does not change significantly. Whether this observation holds for β-W is a topic worth investigating since it could be a good alternative for Pt, but one should be aware of the likely increase in resistivity and thus Joule heating.

We want to give a brief outlook over future development of gradient magneton sputtering as well as new research regarding efficient spin current generating materials. As worked out in this thesis, the current sputtering method is imprecise due to transverse travelling sputter atoms. A possible solution to this is using an array of collimation tubes [36]. A schematic illustration is shown in Fig. 5.1. If a sputtered atom has a transverse velocity component, it will be deposited on the side of a collimation tube instead of reaching the sample. Atoms that pass the collimator tubes have little to no transverse velocity. Using this we can block the particle stream with a wedge shutter that has a gap between the shutter edge and the sample. The downside is a reduction in sputter rate depending on the aspect ratio of the collimator. We can control the collimation by varying the aspect ratio of the collimator. Longer tubes result in a better collimation, while strongly reducing sputter rate, so one needs to find balanced collimator dimensions that produce a collimated enough...
stream while keeping sputter rates at an acceptable value.

Recent investigation of different materials show a new material to have very promising properties for SMR measurements. A mixture of gold and platinum with a mixing ratio of 0.25:0.75 of Au:Pt showed a giant SHA of 0.58 determined by direct spin orbit torque measurements with cobalt as a magnetic layer [37], bypassing even oxidized β-W [17]. A giant spin Hall angle allows for much more efficient switching of the magnetization in a magnet via the spin current, making this material very attractive in spin orbit torque memory devices for example. Another advantage of this material composition is its much lower resistivity of 0.83 μΩm compared to our samples where we observed up to 3 μΩm, greatly reducing losses due to Joule heating. In comparison this material has much better properties than the tungsten we investigated in this thesis but was yet again measured in combination with a conducting magnet while we measured SMR with an insulating magnet, which might lead to changes in the SMR properties, as spin transfer is an interface effect. One may argue that materials like these make investigating others not that promising, but one has to consider the significant cost difference between a material like W and AuPt. Pairing W with other materials to significantly reduce its resistivity while keeping its spin transport properties could offer a viable low cost alternative for future spintronic devices, especially since the gold price is exceptionally dependent on the economic status.

The search for the perfect material for spintronics, i.e. a material with large spin Hall angle, low resistivity and low cost is only limited by the amount of different materials we can realize, reaching from simple elemental conductors like Pt or W to much more complicated materials. We are only at the beginning of testing materials we deem worthy as potential candidates. Thus, far more results in this direction are to be expected in the future.
A Appendix

A.1 Additional Information to Section 4

Figure A.1: ADMR measurement of a 6.7 nm structure at $\mu_0H = 7 \text{T}$ and $T = 125 \text{K}$ in oopj orientation that has not been normalized. The Amplitude $A$ is the difference in voltage between the lowest and highest point, $y_0$ is the voltage at the lowest point and the SMR ratio is $A/y_0$.

A.2 List of Samples
<table>
<thead>
<tr>
<th>Name</th>
<th>Substrate</th>
<th>Material (thickness)</th>
<th>Sputter rate, mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wolfram-1</td>
<td>SiOx</td>
<td>W (10 nm) Ta (2 nm)</td>
<td>5.33 Å/s (RF), tilt-out 0.5 Å/s, tilt-in</td>
</tr>
<tr>
<td>Wolfram-2</td>
<td>SiOx</td>
<td>W (10 nm) Ta (2 nm)</td>
<td>1.46 Å/s (RF), tilt-out 0.5 Å/s, tilt-in</td>
</tr>
<tr>
<td>W-3</td>
<td>SiOx</td>
<td>W (10 nm) Ta (2 nm)</td>
<td>5.85 Å/s (DC), tilt-out 0.5 Å/s, tilt-in</td>
</tr>
<tr>
<td>W-4</td>
<td>SiOx</td>
<td>W (10 nm) Ta (2 nm)</td>
<td>1.86 Å/s (DC), tilt-out 0.5 Å/s, tilt-in</td>
</tr>
<tr>
<td>YIG M1-E2</td>
<td>GGG/YIG (LPE, 1 μm)</td>
<td>W (6.7 nm-47 nm) Ta (2 nm) Al (100 nm)</td>
<td>5.85 Å/s (DC), tilt-out 0.5 Å/s (DC), tilt-in 0.94 Å/s (DC), tilt-out</td>
</tr>
<tr>
<td>YIG M1-E4</td>
<td>GGG/YIG (LPE, 1 μm)</td>
<td>W (6.7 nm-47 nm) Ta (2 nm) Al (100 nm)</td>
<td>5.85 Å/s (DC), tilt-out 0.5 Å/s (DC), tilt-in 0.94 Å/s (DC), tilt-out</td>
</tr>
<tr>
<td>W6x10ramp-1</td>
<td>SiOx</td>
<td>W (6.7 nm-47 nm) Ta (2 nm) Al (100 nm)</td>
<td>5.85 Å/s (DC), tilt-out 0.5 Å/s (DC), tilt-in 0.94 Å/s (DC), tilt-out</td>
</tr>
<tr>
<td>Wedge_Center 1</td>
<td>SiOx</td>
<td>W (ca. 30 nm) Ta (2 nm) Al (100 nm)</td>
<td>5.85 Å/s (DC), tilt-out</td>
</tr>
<tr>
<td>Wedge_Center 3</td>
<td>SiOx</td>
<td>W (ca. 20 nm) Ta (2 nm) Al (100 nm)</td>
<td>1.86 Å/s (DC), tilt-out</td>
</tr>
<tr>
<td>Wedge-Center 2</td>
<td>SiOx</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>YIG M1-K1</td>
<td>GGG/YIG (LPE, 1 μm)</td>
<td>W (0 nm-15 nm) Ta (2 nm) Al (100 nm)</td>
<td>0.31 Å/s (DC), tilt-out 0.5 Å/s (DC), tilt-in 0.94 Å/s (DC), tilt-in</td>
</tr>
<tr>
<td>YIG M1-K1 Clone</td>
<td>GGG/YIG (LPE, 1 μm)</td>
<td>W (0 nm-15 nm) Ta (2 nm) Al (100 nm)</td>
<td>0.31 Å/s (DC), tilt-out 0.5 Å/s (DC), tilt-in 0.94 Å/s (DC), tilt-in</td>
</tr>
</tbody>
</table>

**Figure A.2:** List of samples used during this Thesis. The mode references to the sputtering setting. Tilt-out means the sample is in static position over the magneton, Tilt-in means the magneton gets tilted at an angle and the sample is rotated continuously at 20 rpm.
References


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