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Spatially and Temporally Resolved Spin Seebeck Experiments

Orts- und Zeitaufgelöste Spin Seebeck Experimente

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Introduction

During the second part of the 20th century, microelectronics and computer technology have been an important research field for physicists and engineers. Even nowadays the constant need for faster and smaller devices pushes scientists to find new ways of processing information. Most technologies we use today are based on the transport and storage of electronic charge. Hence, further decreasing the device size while trying to increase the transistor speed results in heat dissipation due to the Joule effect, imposing a fundamental limit on electronic device performance. To avoid this, a new approach is to exploit the spin degree of freedom of electrons instead of their charge, in particular by using pure spin currents which are predicted to be dissipationless [1]. This would mean transition from electronics to so called spintronics.

Recently, Uchida *et al.* demonstrated that spin currents can be induced in ferromagnets by applying a temperature gradient [2]. This effect was called the Spin Seebeck effect (SSE) in analogy to the conventional Seebeck effect, a thermoelectric effect known since 1823 [3], where a charge current is induced by a temperature gradient.

Following the discovery of the SSE, many research groups have become interested in the emerging field of spin-caloritronics (calor (lat.) = heat) focussing on the interaction between spin- and heat-currents. They are attracted not only by the idea of dissipationless spin currents but also by the possibility of exploiting waste heat in devices to induce those currents. Scientifically, spin-caloritronics give access to magnetothermally induced phenomena involving the interaction of heat with spin and charge of electrons. This, for example, allows to establish a microscopic picture of the interactions between magnons and thermal phonons.

A spatially resolved technique to induce and detect spin currents in ferromagnetic thin films - using a laser beam to establish a local temperature gradient - has already been developed at the Walther-Meissner-Institut [4].

The objective of the thesis presented here was to follow up on this work and in particular to further characterize the Spin Seebeck effect in different ferromagnetic thin films. To this end, spatially resolved Spin Seebeck effect measurements were carried out in different samples. The setup was then extended to enable temporally resolved measurements of the SSE. With these experiments many parameters involved in the SSE were identified and it was found that the effect takes place on a time scale of less than 10 μs .

The basic physics behind the Spin Seebeck effect are briefly explained in Chapter 1, followed in Chapter 2 by a description of the setup used for the experiments. First, we discuss spatially resolved measurements in different ferromagnetic insulator thin films in order to determine the size of the SSE effect as a function of the layer composition. In a second series of measurements, we studied the dependence of the SSE on the incident laser power. Finally, since the physical origin of the SSE is not yet fully understood, we carried out time resolved measurements in order to better describe the mechanisms potentially involved. The results are presented and discussed in Chapter 3. In Chapter 4 we give a short summary of our findings and an outlook proposing further steps that can be taken based on this work.

Chapter 1

Theory

1.1 The Spin Seebeck effect

The Spin Seebeck effect (SSE) recently discovered by Uchida *et al.* [2] was given its name in analogy to the conventional Seebeck effect which describes the generation of a charge current when applying a temperature gradient to a conductor: the temperature difference induces a heat current, which, in metals, is dominantly carried by electrons, resulting in a charge redistribution. For example if we build an electrical circuit by connecting two different conductors and apply a temperature gradient between the contacts on both ends, a voltage $V = \int_{T_1}^{T_2} (S_1 - S_2)dT$ can be measured [see Fig 1.1 a)]. Here, S_i is the Seebeck coefficient (in V/K) of the given material, which depends on the density and scattering rate of the conducting electrons.

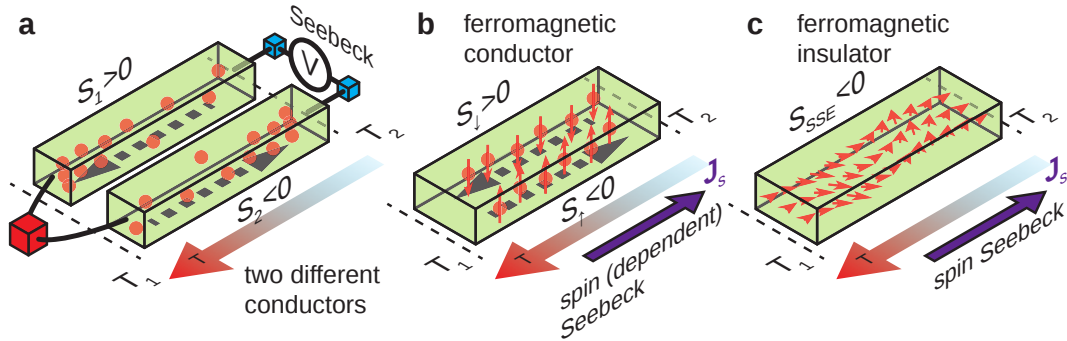


Figure 1.1: a) Seebeck effect in a thermocouple: an electrical voltage develops when applying a temperature gradient. b) Spin (dependent) Seebeck effect in a ferromagnetic conductor: a temperature gradient results in a spin polarized current transported by mobile charge carriers. c) In a ferromagnetic insulator heat and angular momentum are carried by magnons which also gives rise to a Spin Seebeck effect. The figure is taken from Ref. [4].

The Spin Seebeck effect was first discovered in a metallic ferromagnetic thin film [2], where spin up and down electrons have different densities and scattering

properties and therefore can be assigned different Seebeck coefficients. In analogy to the Seebeck effect this generates differing electron flows in the two spin channels which leads to a spin current, i.e. a net flow of angular momentum [see Fig. 1.1 b)]. In open circuit conditions, this results in an accumulation of opposite spin directions on the opposite ends of the ferromagnet.

Shortly after this, the Spin Seebeck effect was also observed in magnetic insulator thin films [5] in the absence of conduction electrons. In this case, the angular momentum cannot be carried by mobile charge carriers but is transported by spin-waves (magnons) which can carry heat as well, see Fig. 1.1 c). However, the actual mechanisms responsible for the observed effect are not yet completely understood. Furthermore, the exact characteristics of the temperature gradient within the thin film, as well as the influence of the interfaces between different layers is not well known yet.

1.2 Detection method: the inverse Spin Hall effect

Since we are studying pure spin currents, a suitable spin current detection method is necessary. To this end, the ferromagnetic thin films are covered with a Pt layer enabling us to exploit the inverse Spin Hall effect (ISHE) which converts a spin current into a charge current that can easily be detected [6] [7].

The conventional Spin Hall effect, a process that converts a charge current into a transverse spin current in a normal metal is shown in Fig. 1.2 a). A pure charge current (no net flow of angular momentum) J_c is applied along the x -axis. The up and down electrons are deflected in opposite directions ($J_c^\uparrow$ and $J_c^\downarrow$) normal to their group velocity by spin-orbit interactions, such as skew scattering, side jump scattering (both extrinsic due to impurities) and intrinsic interactions. These spin-orbit interactions therefore lead to a pure spin current $J_s = J_c^\uparrow - J_c^\downarrow$ perpendicular to J_c .

This mechanism also works the other way around as shown in Fig. 1.2 b): a spin current J_s induces a transverse charge current J_c resulting in the inverse Spin Hall effect.

The macroscopic ISHE can be described by the following equation [7]:

$$J_c = D_{\text{ISHE}} J_s \times \sigma, \quad (1.1)$$

where σ is the spin polarization of the electrons and D_{ISHE} a factor representing the ISHE efficiency in a given material. For Pt a very large ISHE effect is observed [7] due to pronounced spin-orbit interactions (proportional to Z^4 with Z the charge number of the atom [8]), we therefore used Pt as a spin current detector layer in our samples.

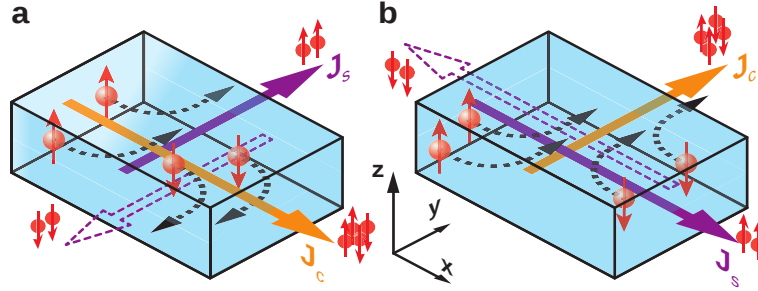


Figure 1.2: a) Spin Hall Effect: a pure charge current induces a transverse pure spin current as a consequence of spin-orbit interaction. b) The inverse Spin Hall effect converts a pure spin current into a pure charge current. The figure is taken from Ref. [4].

1.3 Spatially Resolved Spin Seebeck Effect: SRSSE

We have seen that the Spin Seebeck effect describes the generation of a pure spin current along an applied temperature gradient. The exchange coupled spins in the sample are oriented by applying an external magnetic field in order to create a macroscopic effect. We can then write for the spin polarization $\sigma = M/M_s$ with M the magnetization of the sample and M_s the saturation magnetization¹.

Many measurements (for example by Uchida *et al.* [2] [5]) have been conducted in the transverse configuration: the magnetization is parallel to a homogeneous temperature gradient applied in plane to the whole sample.

In the longitudinal configuration the magnetic field is applied perpendicular to the temperature gradient. Mathias Weiler developed a setup during his PhD thesis [4] at the Walther-Meißner-Institut using this configuration, where the magnetic field lies in the sample plane while a local temperature gradient is applied along the FM/Pt layer normal. This is achieved with a focussed scanning laser beam. Since the different layers absorb different amounts of heat, the laser beam creates a locally confined temperature gradient perpendicular to the sample plane. This method allows for spatially resolved SSE experiments.

The Spin Seebeck effect gives rise to a pure spin current J_s along the FM/Pt normal. The spin current is injected into the Pt layer, inducing a local electric field in the film plane due to the inverse Spin Hall effect

$$E_{\text{ISHE}}(x, y) = -S_{\text{SSE}}(x, y) \sigma \times \nabla T(x, y) \quad (1.2)$$

with the phenomenological Spin Seebeck coefficient S_{SSE} . This effect will, throughout this thesis, be called Spatially Resolved Spin Seebeck effect (SRSSE), in order to

¹The spin polarization can be parallel or antiparallel to the magnetization M , the exact sign is chosen depending on the convention used.

differentiate it from the more general term of Spin Seebeck effect. The S_{SSE} coefficient is in fact not the exact analogon to the conventional Seebeck coefficient S , which depends only on the material where the currents are induced. S_{SSE} rather depends on the interplay of many parameters, for example the thin film materials, in particular the spin current detecting normal metal, (D_{ISHE} , see Eq. 1.1), their layer thickness, the interface between different layers and others that have not been identified yet ². To gain further insight, we in particular analyzed the SRSSE in different ferromagnetic insulator and conducting thin films, but also in a few ferromagnetic conducting thin films. Spatially resolved SSE detection can be carried out in a time resolved fashion, to find out on what time scales the effects occur and thereby get a deeper understanding of the mechanisms involved.

In the following chapter we will describe the experimental setup used for both, spatially and time resolved measurements.

²In particular the sign of S_{SSE} is still unclear and also depends on the convention chosen for the spin polarization, see Footnote 1.

Chapter 2

Experimental Setup

2.1 Setup

For the magneto-thermo-galvanic (MTG) measurements the experimental setup shown in Fig. 2.1 was used. It was developed by Mathias Weiler during his PhD thesis [4] and later extended by Michael Schreier [9]. A laser diode with wavelength $\lambda = 660\text{ nm}$ and power $P_0 = 120\text{ mW}$ is coupled into a single mode optical fiber which terminates in a collimator with a lens with focal length $f = 11\text{ mm}$. The collimator is mounted on a xyz flexure stage (Thorlabs NanoMax 300) that can be remote-controlled by a computer. The stage is actuated by three stepper motors with a repetition accuracy of 500 nm and has a traveling range of 4 mm in all three directions. This allows to focus and scan the laser across the sample's surface.

For the measurements, an external magnetic field is applied, provided by a home-built 2D vector magnet featuring 4 identical coils that can each carry a maximum current of 4 A . The coils form two pairs, each of which is connected by an iron core, so that the sample can be positioned in a $2 \times 2\text{ cm}^2$ area within the magnetic field. The field with magnitude $\mu_0 H \lesssim 100\text{ mT}$ can be applied along any orientation α within the sample plane by an appropriate superposition of the magnetic fields induced by the two coils. Two Hall probes are used to determine the x and y components H_x and H_y of the magnetic field. The Hall probes are connected to two Lakeshore DSP 475 Gaussmeters that set the output currents of two Kepko BOP bipolar four quadrant power supplies using closed-loop PI control. In that way, the magnetic field can also be computer-controlled. The whole setup is controlled via a custom program written in National Instrument's LabVIEW.

2.2 Imaging: spatially resolved measurements

Acquiring the MTG-signal as a function of the laser spot position is the first step in the analysis of the Spatially Resolved Spin Seebeck effect. For this experiment, the laser beam at constant laser power P is focussed and scanned over the sample surface, shaped into a Hall bar geometry (see Fig. 2.1), following a scanning grid

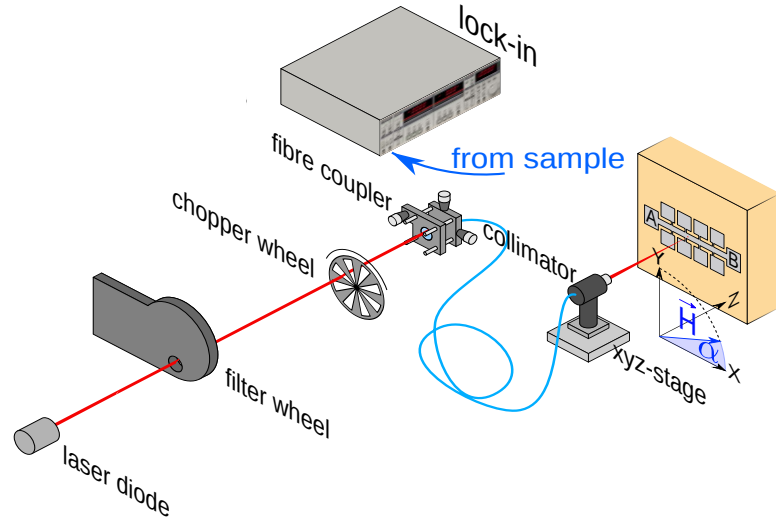


Figure 2.1: Schematic illustration of the experimental setup: a laser beam is focussed and scanned across the sample surface providing a temperature gradient. An external magnetic field $\mu_0 H$ can be applied at any orientation α within the sample plane. The filter wheel allows for measurements at different incident laser powers. The figure was taken from Ref. [9].

in the xy -plane. The sample is mounted on a chip carrier system with 20 electrical contacts directly attached to the center pins of the BNC connectors of a breakout box. The pads of the Hall bar are bonded to the contacts on the chip carrier. This system simplifies the exchange of different samples. Two Hall bar contacts can then be connected to the differential input of a Stanford Research SR830 lock-in detector by two coaxial cables to measure the magnetothermally induced voltage drop in the desired section of the hall bar. As a reference signal for the lock-in, a chopper wheel running at a constant effective frequency $\nu = 810 - 870$ Hz is placed within the beam path. Several lock-ins can then be used to measure the voltage drop between different sample contact pairs. The lock-in phase is the same for all detectors used, and chosen such that the entire signal is recorded in the x -channel. With this method, the voltage V_{ISHE} induced in the sample by the laser beam can be recorded for each point on the scanning grid. The control program transcribes these data into a 2-dimensional image showing the MTG-signal (color-coded) as a function of the laser position (x, y) , each pixel representing the voltage measured at one point of the scanning grid.

2.3 Laser power dependent measurements

In order to allow for laser power dependent measurements, a filter wheel with 6 slots containing 5 ND filters (Neutral Density filters) 01 to 05 and one open slot was put within the beam path (see Fig. 2.1). Each ND x filter reduces the laser power by a factor $10^{-x/10}$. By using an additional ND 06 filter after the filter wheel, overall 12 different values (from 100% to 8% of P_0) for the incoming laser power could be set. In this way, the MTG-signal as a function of the incident laser power can be studied.

2.4 Time resolved measurements

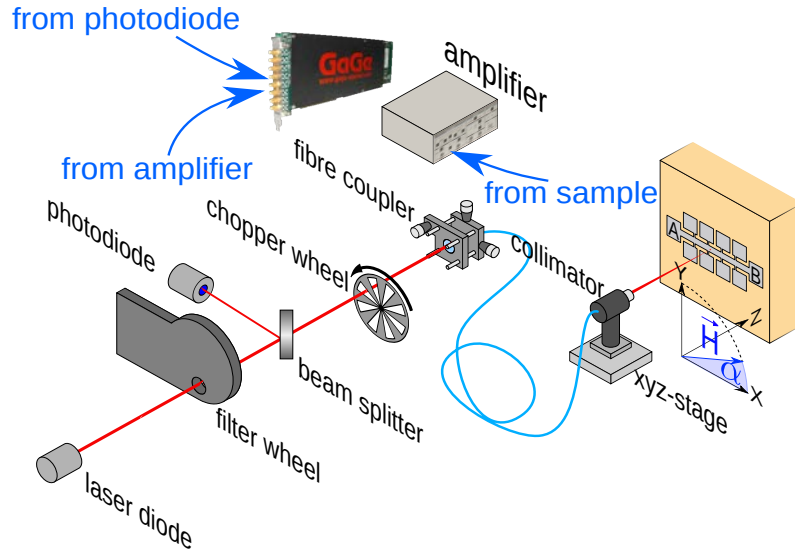


Figure 2.2: Schematic illustration of the setup used for the time resolved measurements: the same setup is used as in 2.1 but the MTG-voltage is preamplified and read out by a digitizer card with 200 Mega-samples per second. A beam splitter outcouples a part of the laser beam which is detected by a photodiode and gives the trigger signal for the read out program. The figure was taken from Ref. [9].

To determine the time scales on which the mechanisms responsible for the SRSSE work, the thermal voltage is measured as a function of the laser modulating frequency (switching the laser on and off periodically).

The setup described in section 2.1 was used with only a few modifications (see Fig. 2.2). First, the Hall bar position is determined by scanning over the sample surface as described in Section 2.2. The xyz stage is then set to a constant position so that the laser impinges on the desired area of the Hall bar. The thermal voltage

signal now is no longer detected by lock-ins, but fed into a Stanford Research SR560 voltage preamplifier and then read out by digitizer card with 200 Mega-samples per second. For a better signal to noise ratio, the signal is averaged in hard- and software.

As the chopper wheel can only run at frequencies up to 1 kHz, the laser diode is now connected to an Agilent Arbitrary Wavefunction Generator (AWG), to control the frequency (up to 80 MHz) and pulse width of the laserdiode current. A 40/60 beam splitter is inserted in the beam path, with the minor beam reaching the sample and the outcoupled beam being detected by a photodiode and used as the trigger signal for the read-out program. The chopper wheel is not used for the time resolved experiments discussed here.

2.5 Samples

Name	Layers
YIG19	GGG/YIG(20)/Pt(7)
YIG45	YIG(20)/Cu(9)/Pt(7)
YY14	YAG/YIG(70)/Pt(7)
YY21	YAG/YIG(45)/Pt(20)
YY26	YAG/YIG(45)/Pt(15)
NFO	MgAl ₂ O ₄ /NiFe ₂ O ₄ (620)/Pt (10)
SP1	Ni/Pt(7)
SP35	Fe ₃ O ₄ /Pt(7)

Table 2.1: Samples used throughout this thesis with composition and thickness (in nm) of the different layers. The substrate thickness is always 500 μm

Imaging and laser power dependent measurements were conducted for different samples. Due to the limited time allocated to this thesis, the frequency dependence was only examined on the YY14 sample. All samples are composed of a substrate with a thickness of 500 μm onto which different ferromagnetic thin film/Pt hybrid layers were deposited. The platinum is needed to exploit the inverse spin Hall effect for detection of the spin currents (see Chapter 1). The sample names used in this thesis are the lab names given after the preparation, they are listed in Table 2.1 with the corresponding layer composition and thickness in nm, the substrate thickness being always 500 μm . YIG stands for Yttrium Iron Garnet ($\text{Y}_3\text{Fe}_5\text{O}_{12}$), YAG for Yttrium Aluminum Garnet ($\text{Y}_3\text{Al}_5\text{O}_{12}$) and GGG for $\text{Gd}_3\text{Ga}_5\text{O}_{12}$. All samples, except for the NFO, were prepared at the Walther-Meissner-Institut by Matthias Althammer and Sibylle Meyer. The YIG thin films were obtained by pulsed laser deposition onto the substrate and the Pt was deposited in situ using electron beam

evaporation. For the NFO sample, the ferromagnetic layer on substrate was prepared by A. Gupta, University of Alabama, USA, the Pt was deposited at the University of Bielefeld, Germany. The Hall bars were patterned by photolithography and a subsequent etching process. All Hall bars except for the SP1 sample are identical, $80\text{ }\mu\text{m}$ wide and $1000\text{ }\mu\text{m}$ long as depicted in Fig. 2.3 The SP1 Hall bar has different dimensions ($200\text{ }\mu\text{m}$ wide and $1000\text{ }\mu\text{m}$ long) which was taken into account during data analysis.

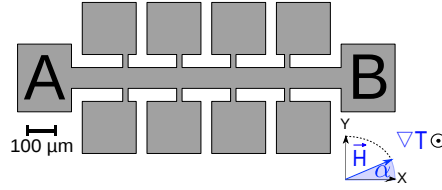


Figure 2.3: Schematic illustration of the Hall bar structure with dimensions $80 \times 1000\text{ }\mu\text{m}^2$ (true to scale) used for the samples. The figure was taken from Ref. [9].

The Hall bar structure (Fig. 2.3) and its position within the setup can also be seen in Fig. 2.1 and Fig. 2.2. The angle α indicates the orientation of the external magnetic field relative to the Hall bar. In all the following measurements, the voltage drop is measured between pads A and B, the temperature gradient goes into the sample plane and the magnetic field is applied perpendicular to the long side of the Hall bar. This configuration corresponds to a magnetic field orientation angle $\alpha = 90^\circ$.

Chapter 3

Experimental results

3.1 Imaging

For imaging experiments, the Spatially Resolved Spin Seebeck effect is measured for various laser spot positions on the Hall bar. Using the setup described in Section 2.1 and 2.2, the entire Hall bar is first scanned in magnetic saturation. Under these conditions, the magnetization is maximal and homogeneous throughout the Hall bar and we can actually use this as an imaging method for the Hall bar structure on the sample.

Figure 3.1 a) shows the voltage V_{ISHE} measured between pads A and B as a function of the laser spot position in the xy plane with an external magnetic field $\mu_0 H = 70 \text{ mT}$ applied at an angle $\alpha = 90^\circ$ relative to the long side of the bar. $\mu_0 H = 70 \text{ mT}$ was used because $\mu_0 H_c \ll 70 \text{ mT}$, with H_c the coercive field of the sample, and the measurement is therefore carried out in magnetic saturation. As we can see by comparison with the indicated Hall bar and with Fig. 2.3, a finite voltage is only observed when the laser hits the main bar. Since the sample is contacted on both ends of the bar, its full length can be represented. The sections on the left and right end where the signal is not very clear and even changes sign corresponds to the bond wires on the pads where the laser also induces a thermoelectric current due to the charge-Seebeck effect. But this is not relevant for the following discussion and therefore will not be evaluated further.

The incident laser power P impinging on the sample surface was measured before each experiment. In this case $P = 52 \text{ mW}$. The resolution of the scanning grid is $6 \mu\text{m}$, i.e., the distance between two consecutive data points in x and y -direction is $6 \mu\text{m}$. The Gaussian laser spot can be estimated to $5 \mu\text{m}$ in diameter. The magnetothermal mapping therefore is a good representation of the $80 \times 1000 \mu\text{m}^2$ Hall bar. We find an average value of $V_{\text{ISHE}} \approx 4.7 \mu\text{V}$ when the laser hits the main Hall bar.

Figure 3.1 b) was recorded at the inverted magnetic field $\mu_0 H = -70 \text{ mT}$ and $P = 52 \text{ mW}$. The resolution was lower (about $18 \mu\text{m}$) but sufficient to determine $V_{\text{ISHE}} \approx -4.75 \mu\text{V}$. This is the expected value since at opposite magnetic field and thus opposite magnetization orientation, only the sign of the ISHE voltage should change, while the absolute amplitude stays the same [cf. Eq. (1.2)].

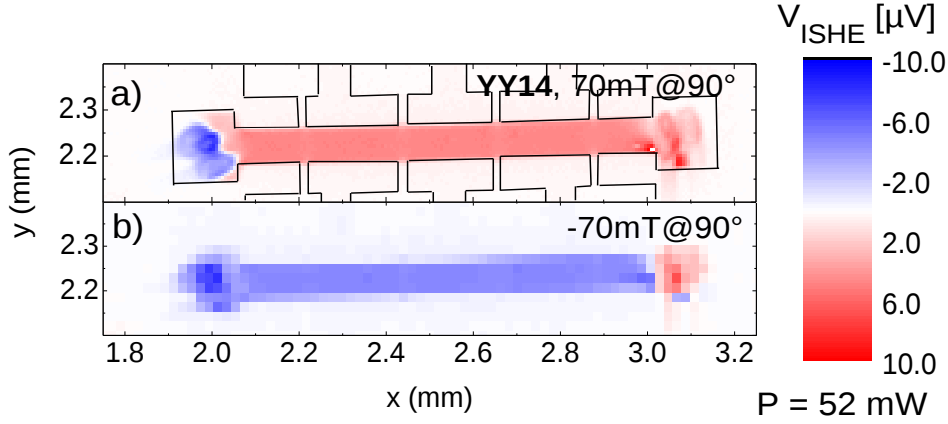


Figure 3.1: a) V_{ISHE} signal as a function of the laser position in the xy -plane at an applied magnetic field $\mu_0 H = 70 \text{ mT}$ perpendicular to the long side of the bar. The Hall bar structure including the pads was drawn onto the panel to show the accuracy of this imaging method. b) Same experiment as in a) but with $\mu_0 H = -70 \text{ mT}$.

To better characterize the field dependence of the Spin Seebeck effect which induces V_{ISHE} in the Pt layer, and also make sure that the magnetization is indeed at saturation at $\mu_0 H = 70 \text{ mT}$, a fieldsweep is carried out. The laser is set at a constant position hitting a single point within the main Hall bar. The magnetic field is then swept from $+70 \text{ mT}$ to -70 mT and to $+70 \text{ mT}$ again. V_{ISHE} is measured between pads A and B and shows a hysteretic behaviour (see Fig. 3.2). This is consistent with the theory outlined in Chapter 1: the measured voltage $V_{\text{ISHE}}(x, y) = -S_{\text{SSE}}(x, y) \sigma \times \nabla T(x, y)$ depends on the magnetization in the sample which exhibits hysteresis during a fieldsweep. All the other parameters were held constant and had no influence on the hysteresis. Also the values for $V_{\text{ISHE}} \approx \pm 4.7 \mu\text{V}$ at saturation at positive and negative field are consistent with the previous results (cf. Fig. 3.1).

The same measurements, both imaging and fieldsweep, were conducted for the other samples and the resulting V_{ISHE} is displayed in Fig. 3.3. The voltage was measured between pads A and B of the Hall bar for each sample.

In most cases the images shown in the bottom panels, taken at negative magnetic field, have lower resolution ($18 \mu\text{m}$) since there only the negative saturation value of V_{ISHE} is relevant, while the exact structure of the Hall bar can be seen in the top

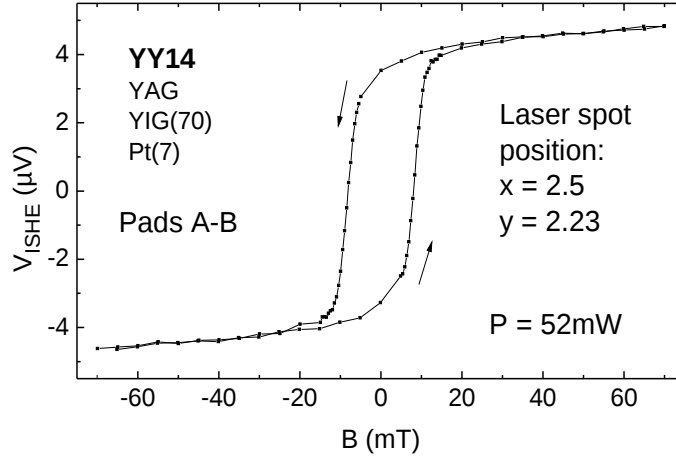


Figure 3.2: Fieldsweep YY14: the external magnetic field is swept down from $\mu_0 H = 70$ mT to $\mu_0 H = -70$ mT and up again. The result is a hysteresis loop for V_{ISHE} .

panel. Again, V_{ISHE} changes sign when the magnetic field is inverted, while the V_{ISHE} magnitude remains constant.

Furthermore, not all datasets were taken at same laser power due to a problem with the optical fiber: the transmission efficiency of the fiber decreased during the experiment such that a few measurements were taken at only 22 mW and the fiber had to be exchanged. For time reasons the measurements could not be repeated. This is one of the reasons why a power dependent measurement as carried out in the next section is interesting. Since a comparison of the results with different laser power would not make much sense at this point, a detailed analysis will be postponed to Section 3.2 where data on the power dependence of V_{ISHE} will allow a more meaningful discussion.

Nevertheless, it is evident from Fig. 3.3 that all the samples show a magnetothermal voltage signal V_{ISHE} of different amplitude for different sample compositions, as well as a hysteretic behaviour.

Since the voltage for some samples is very small compared to the thermal signal coming from the bonds on both ends, the colorscale was adjusted such that the V_{ISHE} signal from the bar is well discernible. This way the bonds can no longer be represented on the chosen scale (saturation) and some pixels appear in black and white.

There are no imaging pictures for YIG19 due to a technical problem during the measurement. But the fieldsweep (see Fig. 3.4) was conducted for one point within the Hall bar, and we can see a similar behaviour, namely a thermally induced voltage depending on the magnetic field.

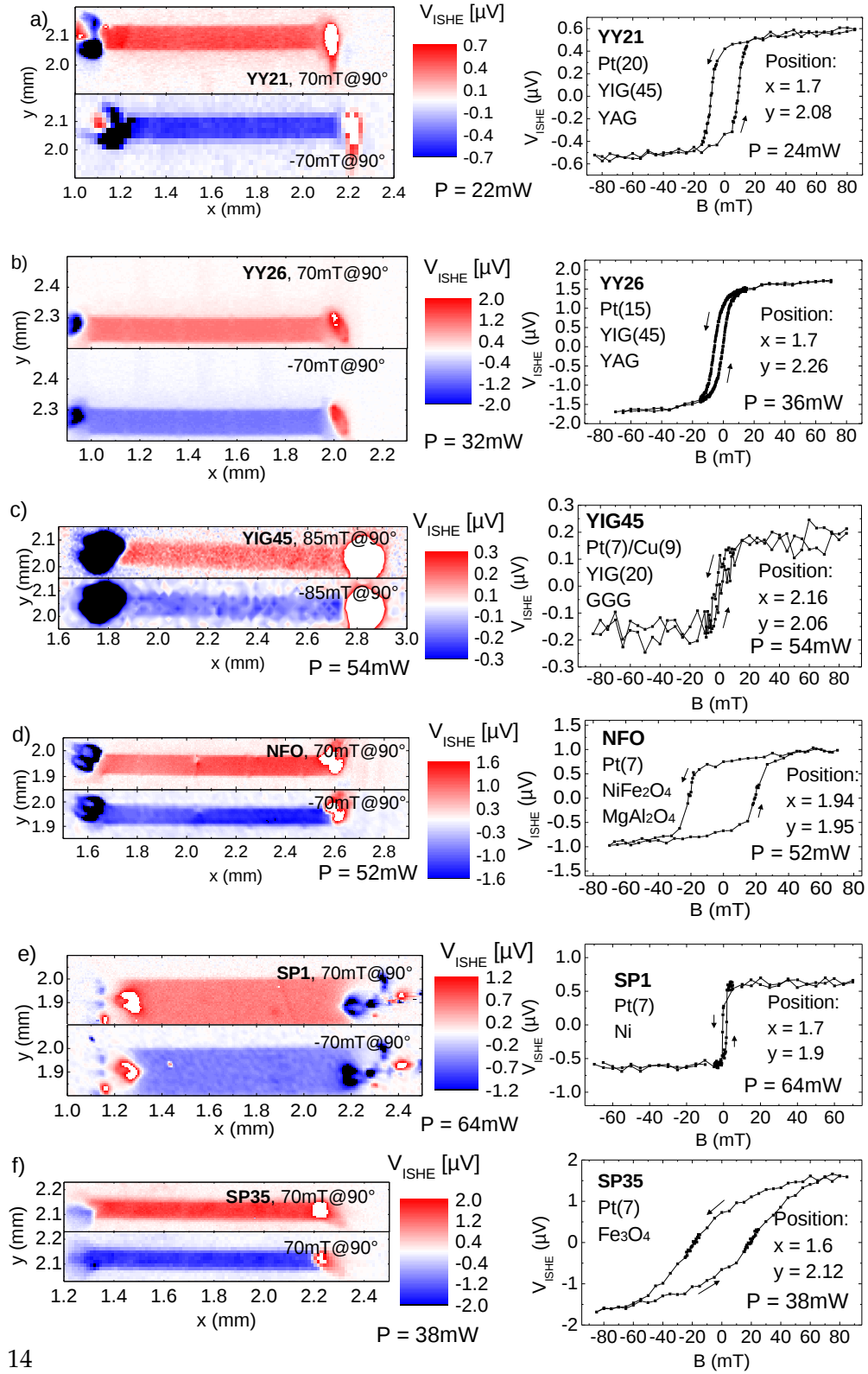


Figure 3.3: Panels on the left: same experiment as in Fig. 3.1 a) and b) for the other samples, V_{ISHE} as a function of the laser spot position. Panels on the right: fieldswep as in Fig. 3.2, the constant laser spot position as well as the sample composition are included in the graph.

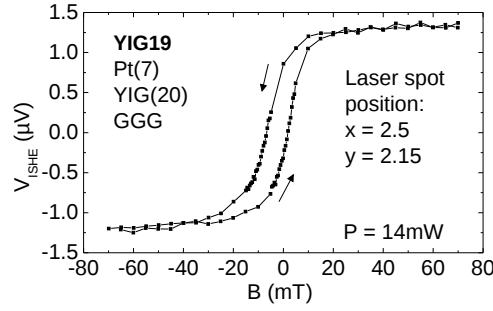


Figure 3.4: Fieldsweep on the YIG19 sample, analogous to Fig. 3.2. The imaging could not be carried out due to technical problems.

3.2 Laser power-dependent measurement

The next important step in describing the Spatially Resolved Spin Seebeck effect is to experimentally determine the dependence of V_{ISHE} on the incident laser power. As mentioned before, the magnitude $V_{\text{ISHE}}(x, y) = -S_{\text{SSE}}(x, y)\sigma \times \nabla T(x, y)$ depends linearly on the temperature gradient ∇T perpendicular to the sample plane. It is therefore interesting to be able to calculate this gradient depending on different parameters. It was shown by Reichling and Grönbeck [10]³ that the temperature gradient induced by a laser beam with Gaussian profile impinging on a multilayered sample can be calculated analytically and that ∇T is proportional to the incident laser power P . We would consequently expect this to show in the experimental data.

Using the setup described in Section 2.3 with different filters, a corresponding experiment was conducted for all 8 samples. The results are shown in 3.5.

The black squares represent the experimental data which are in all cases well described by a linear fit (red line) going through the origin (0,0). We find that the power dependence is linear for all the samples but the slopes are different, as summarized in Table 3.1. To simplify the analysis, this slope will from now on be called m . For the SP1 sample the differing dimensions of the Hall bar were taken into account by multiplying the voltage, and therefore the slope, by a factor 5/2. This is justified because it was shown [4] that the magnitude V_{ISHE} in a given thermal landscape is inversely proportional to w , the width of the Hall bar. The width of the SP1 Hall bar is 200 μm whereas for all the other samples $w = 80 \mu\text{m}$.

³Reichling and Grönbeck extended the work of Jackson *et al.* [11] to a two layer system.

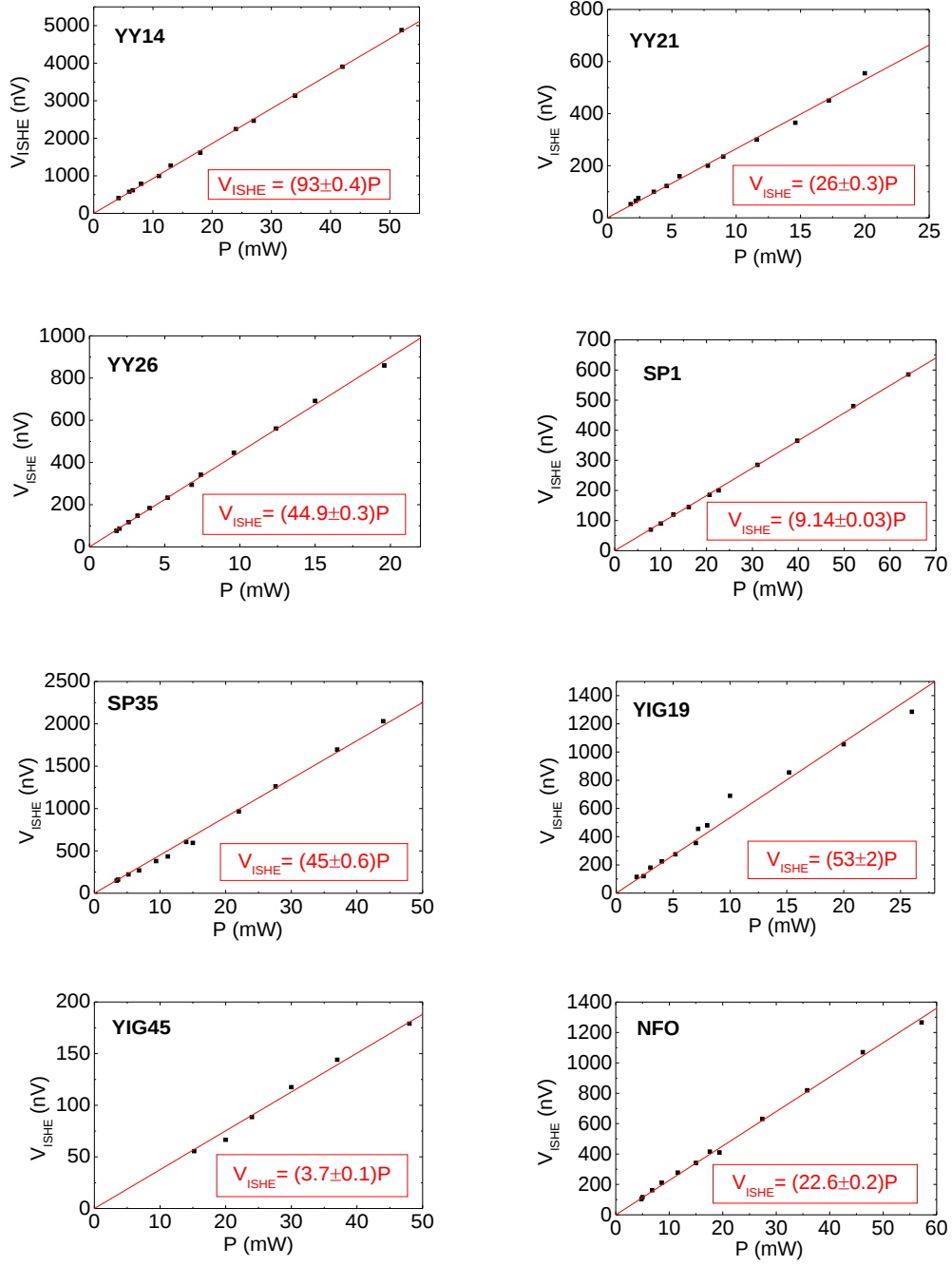


Figure 3.5: V_{ISHE} as a function of the incident laser power P measured for all 8 samples. The black squares represent the experimental data which are in good agreement with the linear fit (red line). The slopes (nV/mW) with their standard error are included in each graph.

Sample	Layers	Slope m [nV/mW]
YY14	YAG/YIG(70)/Pt(7)	93 ± 0.4
YY21	YAG/YIG(45)/Pt(20)	26 ± 0.3
YY26	YAG/YIG(45)/Pt(15)	44.9 ± 0.3
SP1	Ni/Pt(7)	23.85 ± 0.03
SP35	Fe ₃ O ₄ /Pt(7)	45 ± 0.6
YIG19	GGG/YIG(20)/Pt(7)	53 ± 2
YIG45	GGG/YIG(20)/Cu(9)/Pt(20)	3.7 ± 0.1
NFO	MgAl ₂ O ₄ /NiFe ₂ O ₄ (620)/Pt (10)	22.6 ± 0.2

Table 3.1: List of the analysed samples with their power dependence m (nV/mW) and standard error extracted from the linear fits in Fig. 3.5.

The coefficient m is now independent of the laser power and the Hall bar dimensions. Therefore only the dependence of m on the layer composition and thickness remains to be analysed. Since the samples are very different from each other, a comparison will not allow exact predictions but at least give an idea of the parameters that will increase or decrease the magnitude of m .

We will first discuss the dependence of m on the Pt layer thickness. The YY21 and YY26 samples have the same structure except for their Pt layer. From these two the following equation can be determined, assuming a linear dependence on the Pt layer thickness d_{Pt} :

$$m = -3.8 \cdot d_{\text{Pt}} + \text{const} \quad (3.1)$$

Next, we can compare two samples that differ only in their YIG layer thickness and have the same d_{Pt} : YIG19 and YY14. For now, we take the assumption that the substrate has no major influence on the MTG-signal and that in this case GGG and YAG have similar characteristics. Again, the following linear dependence can be found:

$$m = 0.8 \cdot d_{\text{YIG}} + \text{const}, \quad (3.2)$$

where d_{YIG} is the YIG layer thickness.

Using Eq. (3.1), we can further analyze the dependence of m on the YIG layer thickness for YY14, YY21, YY26, YIG19, by eliminating the d_{Pt} dependence and calculating $m_{\text{Pt}(7)}$ for $d_{\text{Pt}} = 7$ nm:

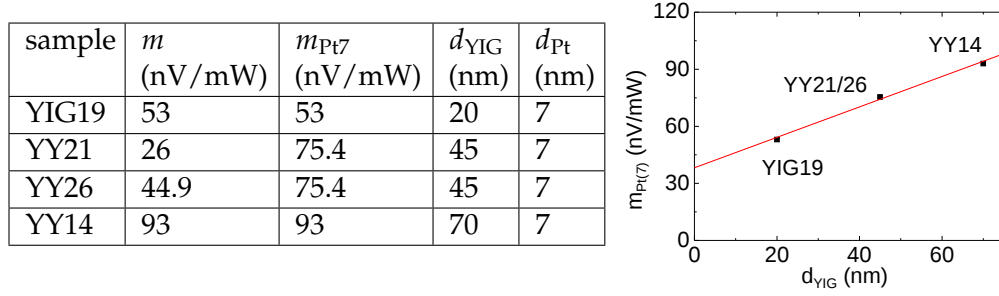


Figure 3.6: $m_{Pt(7)}$ was calculated for the samples listed here, eliminating the dependence on the Pt layer thickness with Eq. 3.1. The data was plotted (black squares) and fitted (red line) showing a linear dependence on d_{YIG} .

The results are shown in Fig. 3.6 and we find:

$$m = (0.8 \pm 0.09) \cdot d_{YIG} + \text{const}, \quad (3.3)$$

with the standard error of the linear fit (red line). The results for YY21 and YY26 fit in well with the other two samples and the slope of the linear fit is consistent with Eq. (3.2). Since only 4 data points were analysed this only gives a trend, but it appears that a thinner YIG film leads to a smaller m and thus a smaller Spin Seebeck effect.

This analysis can now be repeated for the dependence of m on d_{Pt} . To this end, $m_{YIG(20)}$ was calculated for a YIG layer thickness of 20 nm using Eq. (3.3).

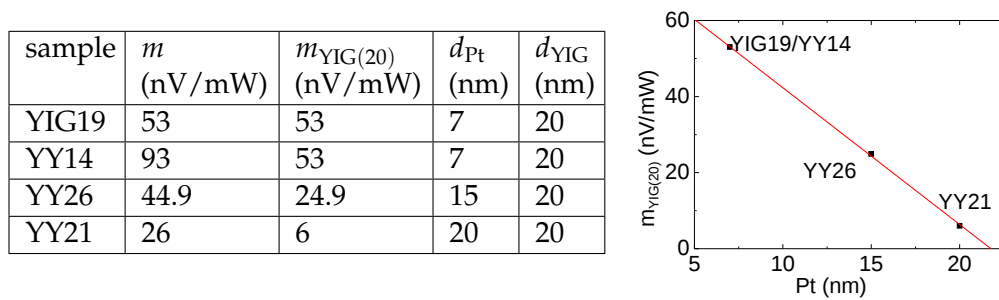


Figure 3.7: $m_{YIG(20)}$ was calculated for the samples listed here, eliminating the dependence on the YIG layer thickness with Eq. 3.3. The data was plotted (black squares) and fitted (red line) showing a linear dependence on d_{Pt} .

We find again a linear dependence as shown in Fig. 3.7:

$$m = (-3.6 \pm 0.1) \cdot d_{\text{Pt}} + \text{const} \quad (3.4)$$

which is in good accordance with Eq. (3.1). This analysis shows that the magnitude of the MTG-signal decreases with increasing d_{Pt} by a factor $\approx -3.6 \text{ nV/mW nm}$, whereas a thinner YIG layer gives rise to an effect larger by a factor $\approx 0.8 \text{ nV/mW nm}$. The thickness of the Pt layer has a great influence on the MTG-signal at a high laser power, but changing d_{YIG} has only a weak impact. The factor m is one of the sample parameters that plays an important role in the Spin Seebeck coefficient introduced in Eq. (1.2) and allows for predictions concerning further samples with similar composition (substrate/YIG/Pt). This has to be verified by analysing further samples. Moreover, it would be interesting to find out the limits of these predictions, especially if there is a minimal d_{Pt} or maximal d_{YIG} beyond which the MGT-signal decreases again. It is clear that using no Pt at all destroys the signal since there is no detection medium left for the spin currents. Therefore, there has to be a d_{Pt} for which V_{ISHE} reaches its maximum.

The results also show that the previous assumption of the substrate nature (GGG or YAG) being irrelevant seems legitimate, since no considerable deviation from the linear behaviour in Eq. (3.3) and (3.4) has been found.

The YIG45 sample was not included in this analysis since its MTG-signal is extremely small, $m = 3.9 \text{ nV/mW}$. This weak signal is due to the 9 nm Cu layer between YIG and Pt. There are different reasons why this sample gives rise to a very low thermal voltage: Cu has a weaker spin-orbit interaction (SOI) than Pt because the strength of the SOI is proportional to Z^4 with Z the charge number of the atom [8]. Since Cu is much lighter than Pt, the inverse Spin Hall effect depending on the SOI is negligible in Cu. The additional layer interface also leads to losses during the spin current injection from the YIG layer, the Cu layer absorbs most of the spin currents before they reach the Pt layer. This is all consistent with the assumption that the Spin Seebeck coefficient S_{SSE} depends on a complex interplay between different material parameters [see Eq. (1.2)].

The NFO sample was not included either, since its composition is too different from the YIG samples to make a meaningful comparison. Nevertheless, its MTG-signal is in the same order of magnitude as for example in YY21, making it a promising thin film combination for further experiments. The SP1 and SP35 samples cannot be analysed in detail at this point because Ni and Magnetite (Fe_3O_4) are not ferromagnetic insulators. The thermal voltage measured here is not due exclusively

to the Spin Seebeck effect, but also to the Anomalous Nernst effect observed in ferromagnetic conductors [4]. It is therefore not clear to what extent the measured voltage can be attributed to the SSE.

3.3 Laser frequency-dependent measurement

To go one step further in the investigation of the Spin Seebeck effect, the last series of measurements was conducted to determine the effect's dependence on the laser modulating frequency. The measurements were carried out in the YY14 sample since it gave the largest V_{ISHE} . As described in Section 2.4, the laser was again set to a constant position heating one spot on the Hall bar. A square wave signal was used acting as an "on-off" switch for the incident laser power. In all measurements it was set to 50 % duty cycle, which means that the "on" and "off" time were equally long. We expect the same voltage levels when the laser is on as in the previous experiments (see Sections 3.1 and 3.2).

About 30 different frequencies in regular intervals ranging from 860 Hz to 200.060 kHz were tested. A few representative measurements are shown in Fig. 3.8.

At 860 Hz [Fig. 3.8 a)] the levels are clearly visible and the difference between "on" and "off" equals $\approx 4.5 \mu\text{V}$ which is consistent with the previous data ($P = 52\text{mW}$). The absolute values on the y -axis are offset such that $V_{\text{ISHE}}(\text{laseroff}) = 0$ and $V_{\text{ISHE}}(\text{laseron})$ corresponds to the voltage induced by the laser beam. Two horizontal lines were included in each panel to enable a straightforward identification.

When increasing the frequency to 20.060 kHz, sharp peaks in V_{ISHE} of high amplitude appear at every switching. At higher frequencies a "ringing" appears on both laser on and off levels, starting with a sharp peak and dampening exponentially. The ringing frequency was determined in Origin by FFT and found to be $\approx 1.6 \text{ MHz}$. At laser modulating frequencies of 200.060 kHz and above, this phenomenon makes data analysis impossible since levels are no longer discernible. We tentatively attribute this ringing to electromagnetic crosstalk, possibly interfering with the laser driver. A better shielding from interfering signals might be necessary but would implicate a modification of the experimental setup. This will be discussed in more detail in Chapter 4.

Nevertheless, for frequencies ν up to 160 kHz the level amplitude could be analyzed and the results as a function of ν are summarized in Fig. 3.9. A line was drawn in the graph at the average value $V_{\text{ISHE}} = 4.4 \mu\text{V}$. Within experimental uncertainties V_{ISHE} is independent of ν in the range $860 \text{ Hz} \leq \nu \leq 160 \text{ kHz}$. The values at higher frequency seem to decrease slightly, but this might be due to the fact that a precise

3.3 Laser frequency-dependent measurement

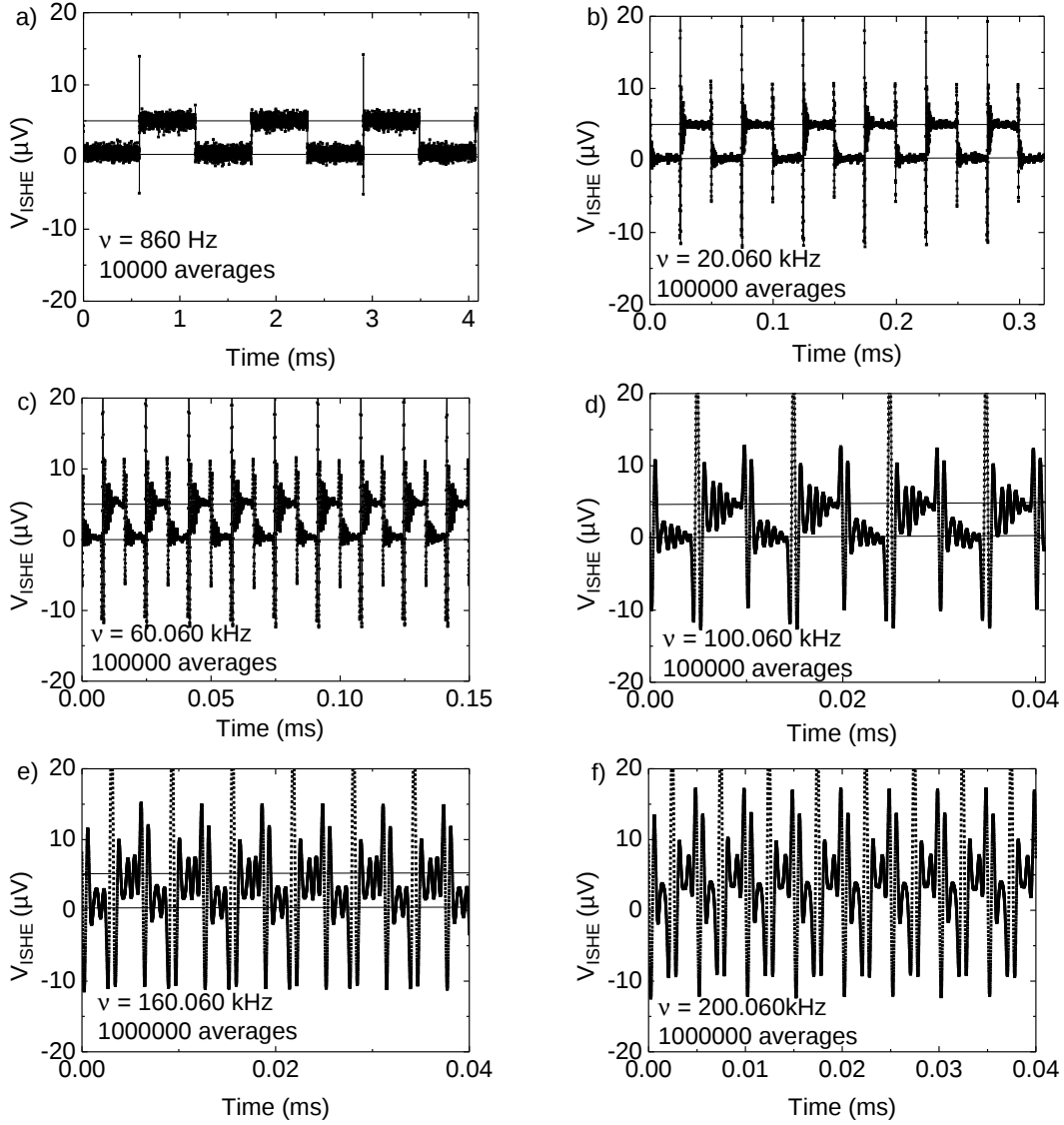


Figure 3.8: MTG-voltage measured for 6 different laser modulating frequencies. The higher level appears when the laser is on, the lower one when it is off. Two lines were drawn in each graph to make the reading easier.

analysis of the data becomes impossible due to the already discussed ringing of the signal.

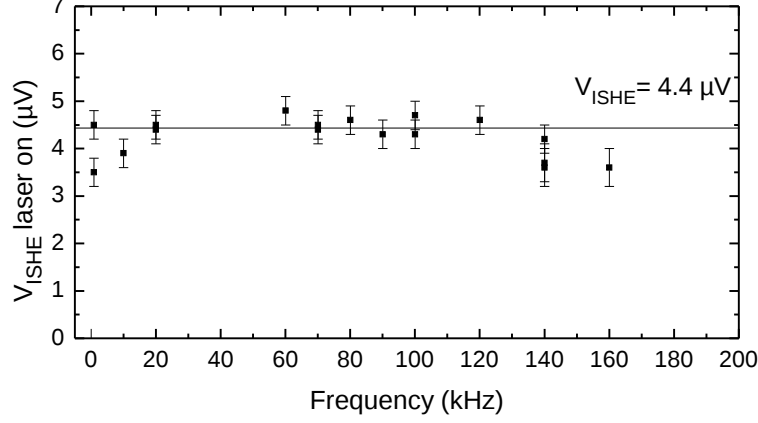


Figure 3.9: Voltage difference between laser "on" and "off" switching for different modulating frequencies. A line indicating the average value and error bars corresponding to a reading error are included.

We now discuss that V_{ISHE} is indeed expected to be independent of ν in this frequency range.

In the samples the temperature gradient arises by propagation of thermal phonons. Phonons propagate at the speed of sound in solids $v_{\text{ph}} \approx 3000 \text{ m/s}$ (for Pt at room temperature). The time needed to establish a temperature gradient in the Pt layer can therefore be estimated to:

$$\tau = \frac{d_{\text{Pt}}}{v_{\text{ph}}} = \frac{10 \text{ nm}}{3000 \text{ m/s}} = 3.3 \text{ ps}, \quad (3.5)$$

where d_{Pt} is the thickness of the Pt layer.

This corresponds to a frequency $\nu \approx 300 \text{ GHz}$, which is far beyond the frequencies that were used in this experiment. Furthermore, measuring a voltage of the magnitude of a few μV on a time scale of ps is impossible with the technology at hand. Hence, the phononic temperature gradient can be considered to build up instantaneously.

An explanation for the Spin Seebeck effect in ferromagnetic insulators was given by Xiao *et al.* [12]. This theory introduces an effective magnon temperature T_{F}^* in the ferromagnetic insulator F and an electron temperature T_{N} in the normal metal N (Pt). The spin current J_{s} is proportional to the temperature difference $T_{\text{F}}^* - T_{\text{N}}$. Moreover it is assumed that the electron temperature T_{N} equals the lattice temperature of N. The magnons in F are colder than the lattice and electrons in N due to the absorption of the laser power which takes place dominantly in N as F is optically close to

transparent at the laser wavelength. Hence, a Spin Seebeck effect is expected to build up on the time scale of magnon-phonon relaxation which is $\tau_{\text{mp}} \approx 1 \mu\text{s}$ [12]. Using laser modulating frequencies of 1 MHz and beyond should enable us to see a change in the observed voltage. This is a frequency that can easily be attained with an improved setup and modifications in this direction are currently undertaken.

Nevertheless, for now, the results from Fig. 3.9 are consistent with expectations, showing no discernible change in V_{ISHE} when the laser is on at frequencies up to 200 kHz. In future experiments, the laser modulating frequency will be extended beyond 1 MHz as discussed in Chapter 4. This will allow to investigate the SSE on the timescale of magnon-phonon interaction.

Chapter 4

Summary and Outlook

In this thesis we examined the Spatially Resolved Spin Seebeck effect (SRSSE), a newly discovered magneto-thermo-galvanic (MTG) effect, in various ferromagnetic thin film/Pt hybrids, the majority of which were based on the ferromagnetic insulator Yttrium Iron Garnet (YIG). When applying a local temperature gradient to the thin film hybrid normal using a laser beam, spin currents are induced along the temperature gradient in the ferromagnetic layer. These can be detected in the Pt layer using the Inverse Spin Hall effect (ISHE) which transforms a pure spin current into a transverse charge current that can be measured. To this end, all samples were patterned into a Hall bar geometry.

In a first series of measurements, the laser beam was scanned over the surface of the magnetically saturated sample and we showed that a finite MTG-voltage V_{ISHE} can be observed in all samples whenever the laser impinges on the Hall bar region enclosed by the used electrical contacts. With this spatially resolved method, the V_{ISHE} was mapped as a function of the laser spot position (see Section 3.1). This enables recording spatially resolved maps of the Spin Seebeck effect in ferromagnetic thin films. Furthermore, it was found that the magnitude of V_{ISHE} depends on the layer composition of the samples.

In a second set of experiments, the dependence of the MTG-signal on the incident laser power was determined for each sample (Section 3.2). We found that V_{ISHE} scaled linear with laser power and thus temperature gradient for all samples and extracted from these data a material dependent coefficient m , proportional to the SSE coefficient. The influence of the layer composition and thickness on the SSE coefficient was also analyzed. It was found that the signal increases with decreasing thickness of the Pt-layer and that increasing the thickness of the YIG layer also gives rise to a slightly larger V_{ISHE} . The choice of the substrate used for the thin films - Yttrium Aluminum Garnet (YAG) or Gadolinium Gallium Garnet (GGG) - did not have any discernible influence on the measured voltage.

In a final set of experiments, time resolved traces of V_{ISHE} were recorded for laser modulation frequencies in the range from 860 Hz to 200 kHz (Sec. 3.3). This way, the temperature gradient induced by the laser was switched on and off periodically. The voltage difference ΔV_{ISHE} between the laser on and off states was analyzed for

frequencies up to $\nu = 200$ kHz, and found to be independent of ν . This finding is in accordance to the present understanding of the Spin Seebeck effect that suggests a spin current generation on the timescale of the magnon-phonon interaction (1 μ s).

The results of the spatially resolved measurements enable us to estimate the ISHE voltage magnitude for further samples. More YIG/Pt thin films need to be prepared with different layer thicknesses to systematically verify these predictions and see if there is indeed a linear dependence between V_{ISHE} and the layer thickness. It might also be interesting to find out if there is a maximal YIG-layer and/or minimal Pt-layer thickness beyond which the signal again decreases. As Pt acts as spin current detector via the ISHE, the optimal Pt thickness is expected to correspond to the spin diffusion length of Pt. Our results suggest a spin diffusion length of less than 7 nm in Pt and that there is an optimal Pt layer thickness $d_{\text{Pt}} \leq 7$ nm for which V_{ISHE} is maximal.

This optimization aspect is important especially for future technological applications of the Spatially Resolved Spin Seebeck effect, for example in microelectronics, where a voltage of only a few μ V is not sufficient. It might therefore also be useful to find new promising materials for thin film hybrids (as in the NFO sample). But this makes a more detailed understanding of the fundamental physics involved in the SSE necessary. Especially the temperature gradient induced by the laser beam in the thin film hybrid needs to be characterized to allow a quantitative determination of the SSE coefficient. Furthermore, the laser beam also induces in-plane temperature gradients, the influence of which has not been determined in detail yet.

The setup used during this thesis offers many advantages: it allows for spatially resolved measurements and the imaging of microscopical structures, in particular of magnetic domains (see Ref. [4]). Moreover, as demonstrated in this work, it allows temporally resolved measurements by using a laser beam to induce a temperature gradient that can be switched on and off on very short time scales (about 10 μ s). For a more detailed understanding of the Spin Seebeck effect, these time scales need to be further reduced, at least by one order of magnitude. Therefore, improving the experimental setup for the temporally resolved measurements is an important next step in our investigation. For this purpose a new laser system was already ordered based on the results of this thesis. Moreover a better electromagnetic shielding of the experimental is needed in order to be able to resolve V_{ISHE} traces with high temporal resolution. This can be achieved by reducing the cable length to a minimum. Also, the sample carrier, where the thin wires from the sample are connected to the break-out box, needs better shielding to ensure a better signal to noise ratio. These improvements will enable us to carry out measurements on the time scale of magnon-phonon interaction and beyond.

Another interesting approach for time resolved measurements might be to use not only simple on and off switching of the laser beam (square wave signal), but more advanced profiles, for example a triangle or a sinus wave. Finding a way to reverse the temperature gradient, as proposed by Walter *et al.* [13], on the time scales where the SSE takes place might also give a more detailed picture of the mechanisms involved.

This discussion shows that the investigation of the Spatially and Temporally Resolved Spin Seebeck effect has by far not reached its end. Many material dependent parameters remain to be determined and more measurements need to be carried out in order to extend the microscopical model of the complex mechanisms involved in the Spin Seebeck effect. In this thesis some of these material parameters were extracted and we demonstrated that spatially resolved SSE experiments can be carried out with temporal resolution. The results of these time resolved measurements were consistent with the present understanding of the SSE and suggest promising findings in further experiments.

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