Magneto and spin transport in magnetically doped semiconductors and magnetic insulators

Dissertation

Presented in Partial Fulfillment of the Requirements for the Degree Doctor of Philosophy in the Graduate School of The Ohio State University

By

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Graduate Program in Electrical and Computer Engineering

The Ohio State University

2017

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Abstract

Over the last two decades, spin transistors that operate using both charge and spin properties of electrons have motivated extensive studies of injection, detection and manipulation of electronic spin current in various material systems. Dilute magnetic semiconductors, in which the spin polarized charge carriers are coupled to the magnetic moment, are of particular interest due to their compatible lattice structures and similar growth methods to current Si and GaAs technology. The first part of this thesis focuses on the structural, magnetic and magnetotransport properties of magnetically doped GaN and 2D MoS$_2$. The Gd doped AlN/GaN heterostructures are grown by plasma assisted molecular beam epitaxy. The Gd atoms are δ-doped at the AlN/GaN heterointerface where the two dimensional electron gas (2DEG) forms. These samples exhibit defect-induced room temperature ferromagnetism with an easy axis along the c-axis. However, the nonlinear Hall resistivity does not track the magnetization in these Gd doped samples indicating the lack of coupling between the conduction electrons in the 2DEG and the Gd-induced ferromagnetism. This makes Gd doped GaN not useful as a dilute magnetic semiconductor. Mn doped few-layer MoS$_2$ samples synthesized via sulfurization of Mn thin film on sapphire are fabricated in the aim of realizing a 2D dilute magnetic semiconductor. However, these samples mainly show paramagnetism implying the lack of ferromagnetic coupling between the Mn dopants. In addition to the electronic spin current, magnonic spin current has
recently received growing research interest since it serves as a new route for achieving novel thermoelectric generators and magnon transistors. The second part of the thesis focuses on the study of the transport properties of the thermally induced magnonic spin current via spin Seebeck effect. A nonlocal opto-thermal spin Seebeck configuration is proposed and implemented to measure the spin diffusion length in yttrium iron garnet (YIG). Finite-element modeling (FEM) based on heat and spin transport is employed to validate the detection of the pure diffusive magnonic spin current and to estimate an upper bound of the spin diffusion length by considering the Pt sinking effect from the unused Pt absorbers. Magnon dynamics are probed via the time-resolved longitudinal spin Seebeck effect. The time domain spin Seebeck waveforms consist of a fast rise (∼ ns) component and a slow rise (∼ ms) component at all temperatures. FEM modeling based on heat and spin transport suggests that the fast rise component is attributed to the interfacial electron and magnon temperature difference while the slow component is a result of bulk magnon diffusion. Different mechanisms (either spin or heat diffusion) are found to dominate the speed of the slow rise component at different temperatures. In the hope of putting the spin heat transport knowledge into practice, a novel Nernst thermoelectric generator with scalable power output using Galfenol wire in a coil geometry is demonstrated. The Nernst coefficient found in Galfenol is the highest among similar ferromagnetic materials. At last, the coupled thermal and condensed magnon transport are modeled to facilitate the design of the magnon Bose-Einstein experiment.
This work is dedicated to my beloved parents and my dear fiancée Shuo Zhang.
Acknowledgments

I would like start my acknowledgements by thanking the group members of Myers research group since most of my work is done with their help. I joined the group together with Dr. A.T.M Golam Sarwar in fall 2011. By then, there were three group members: Dr. Jing Yang, Dr. Santino Carnevale, and Dr. Thomas Kent. Dr. Jing Yang was a nice person and a good friend of mine. She helped me growing the samples and trained me on lots of material characterization/processing skills, like AFM, XRD, micro-polishing etc. Chatting with her was always stress-relieving during my early years in the group. Dr. Santino Carnevale was a MBE master and he is also the most logical and organized person that I have ever met. Although our research did not overlap, the way he plan, execute and finish research projects set up an example for me and guided me through the entire Ph.D. career. Dr. Thomas Kent was an optics genius and a brilliant MBE grower. He not only grew samples for my project, but more importantly, he gave me full package training on the clean room micro-processing techniques, SQUID and optics lab instruments which I had been benefited from through my entire time at OSU. Dr. Sarwar, my brother! I would like to express my heart full gratitude to him. Although we did not have a chance to work on the same project together, I have to say that he influenced my life and research from lots of aspects. He was always patient and insightful when we discussed research related problems and he was also a semiconductor sensai to me from whom
I almost re-learnt semiconductor device physics. We spent a lot of fun time together too. I can still recall that we frequently saw each other working at NTW at 1 am in the night and that we played 8-ball and FIFA etc. (of course he always defeated me) in RPAC lounge room together. These experiences are of my best memories at OSU.

The first person joined the group after me was Dr. Masihhur Laskar. I learnt lots of XRD tricks from him and I also had a chance to collaborate with him on the electrical transport on 2D materials. I spent most time in Myers group with Brandon Giles who joined the group in my second year. We started the spin caloritronics project together since 2013. Although there were lots of difficulties and setbacks, we were able to overcome most of them and accomplish research goals. In my third year, Brelon May and John Jamison joined the group. John was the new optics lab boss and a person very dedicated to science. We had a lot of fun working on some really complicated projects together. Brelon May was an excellent crystallographer and he also has endless interesting ideas on improving the MBE growth technique. I thank him for all the BBQ he organized. The latest student joined the group is Emilio Codecido. I am thankful for his trust in me when I tried to talk him over on working with me on the Galfenol Nernst coil project. I believe he has outstanding research instinct which will definitely help his future career.

I would like to thank my advisor Prof. Roberto Myers for giving me the opportunity to work in the group. He always courage me to try and pursue new and great ideas and always willing to discuss research related questions. With his advice and mentoring, I have grown from an undergraduate student who only knew how to solve homework problems to an independent researcher. Some of his perspective and mindset on research will definitely benefit me for my entire life.
During the route, I am very grateful that I had the chance to work with many talented people both inside and outside OSU. I am lucky enough to have the privileges to collaborate with Prof. Joseph Heremans group at ME department. Prof. Joseph Heremans is among one of the most knowledgeable and kind scholars that I have ever known. I cherish the moments when he would like to sit down and help me go through magneto-transport related problems. Dr. Hyungyu Jin carried me through the collaboration and the discussion with him had been always helpful for my research. Dr. Yibin Gao was a kind senior mentor to me and was collaborating with me on several side projects. Yuanhua Zheng provided lots of help on the Nernst coil project. I have also got a lot of help from Prof. Siddharth Rajan group at ECE department. Dr. Digbijoy Nath shared his growth recipe for the AlN/GaN HEMT on SiC which solved the leaking current issue. Zhichao Yang had lots of useful discussion with me on transport related problems. Besides, I appreciate the help from Jack Brangham and Prof. Fengyuan Yang for providing the Pt/YIG samples for the magnon transport study. In addition, I am really grateful for the help from Dr. Flebus Benedetta, Dr. Se Kwon Kim and Prof. Yaroslav Tserkovnyak on the thermal and condensed magnons transport simulations. Besides the academic facultys and Ph.D. students, I also would like to express my gratitude to the staff at NTW and ENSL. Lots of technical related problems cannot be solved without their assistance.

I would like to thank my roommate and my good buddy Yiping Sun. I felt really lucky to have him around during my start-up years and especially his company in the hospital when I had the car accident in Dec. 2012. My thanks also go with my friends Bin He, and Ge Tian. They were always around to courage me when I had difficulties in life and research. I would also like to thank all my friends from my undergrad
class in University of New South Wales, Australia. The endless discussions on the semiconductor physics and solar cells with my roommates Dr. Ruoyu Li, Dr. Yao Yao, Dr. Ziheng Liu and Dr. Jian Chen strongly inspired my interest in semiconductor devices. I worked with Dr. Lan Yu with the same advisor (Dr. Ivan Perez-Wurfl) for the undergrad thesis. I will never forget the hard working days when we had to prepare GRE, undergrad thesis and Ph.D. application at the same time. The experience of living and studying with them encouraged me to pursue a career in engineering and science.

I would like to thank my parents, Prof. Rongcun Yang and Yunnuan Jia. They have been always supportive through my entire Ph.D. journey and their concern on my life gave me the strength to overcome any difficulties in life. At last, I would like to thank the sweetheart of my life, Shuo Zhang. She sacrificed so much for me that I do not even know how to pay back. I am also really grateful for all the encouragement and prolonged phone calls between us. These helped me go through some of my toughest days in the last six years. I would not be able to finish my Ph.D. without her.
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Publications

Zihao Yang, John. S. Jamison, Brandon L. Giles, Jack Brangham, Fengyuan Yang and Roberto C. Myers, "Magnon dynamics in spin Seebeck effect from ns to ms range", In prep..

Brandon L. Giles, Zihao Yang, John. S. Jamison, and Roberto C. Myers, "Long range thermally drive spin current in YIG due to intrinsic spin Seebeck effect", In prep..

Zihao Yang, D. Scott Katzer, Neeraj Nepal, David J. Meyer and Roberto C. Myers, "Superconducting properties of epitaxial $\beta$-Nb$_2$N thin film on SiC substrate”, In prep..


**Fields of Study**

Major Field: Electrical and Computer Engineering
Magnetic Materials
Spin Caloritronics
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A.1 Etch rate of Kyma GaN and Gd doped GaN using Cl\textsubscript{2} based etching recipe.

A.2 2 probe IV measurements over Ti/Al/Ni/Au contacts on AlN/GaN heterostructure.

C.1 Etch rate of Pt using Cl\textsubscript{2} based etching recipe.

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Chapter 1: Introduction and Overview of the Thesis

The current logic circuit technology and its building blocks (such as transistors etc.) mainly utilize the charge properties of the charge bearing carriers (electrons and holes) in various metals, semiconductors and insulators. Besides their charge properties, electrons (holes) also exhibit one of two states of spin, i.e. spin up and spin down. It is envisioned that next generation electronics with lower power consumption will rely not only on the charge properties of the electrons (holes) but also on their spin properties [5, 6]. Datta and Das [7] proposed a field effect transistor that operates based on electronic spin. Research on the injection/detection of electronic spin from the source and drain, and manipulation of the electronic spin precession in the spin channel has attracted extensive research since 1990 [8, 9, 10, 11]. The family of dilute magnetic semiconductors with carriers coupled to their magnetism is an ideal type of material to facilitate the spin injection into other semiconductors. More importantly, they are suitable for integration into the current vertical transistor structures due to their similar crystal structure to Si and GaAs and thin film growth methods. However, the prototypical magnetic semiconductors GaMnAs and InMnAs suffer from a low Curie temperature ($T_c$) which hinders their capability for technological applications [12, 13]. In addition to the electronic spin current, there is a second
type of spin current, i.e. a spin wave spin current [14], where the spin current is carried by magnons. The transport of thermally induced magnon spin current has been widely studied since Uchida et al. [15] observed the spin Seebeck effect in permalloy. Understanding the transport properties of magnons and the interplay between electrons, phonons and magnons is crucial for building next generation thermoelectric generators [16, 3] and magnon based field effect transistors [17].

1.1 Dilute magnetic semiconductors

Magnetic semiconductors represent a semiconductor family that possess both ferromagnetic and semiconducting properties. These types of semiconductor include europium chalcogenides (such as EuO, EuS etc.) and semiconductor spinels (such as CoCr$_2$S$_4$ etc.) [1]. The latter usually contain repeated arrays of magnetic elements in their lattice as shown in Fig. 1.1c. Although the carriers are typically found to be strongly coupled to the magnetic ions in these magnetic semiconductors [18, 19], their complicated crystal growth methods and lattice-mismatched crystal structures [1] make them difficult to integrate into current logic electronic structures.

This problem can be overcome by considering a second type of semiconductor that exhibits similar ferromagnetic and semiconducting properties to magnetic semiconductors. These are dilute magnetic semiconductors. In this type of material, magnetic atoms are substitutionally doped into a non-magnetic host lattice as shown in Fig. 1.1a and b. The introduction of the magnetic impurity would lead to a n- or p-type semiconductor that exhibits ferromagnetism and contains spin polarized carriers. The ferromagnetism is either mediated by the carriers [10, 20] or by the defects
Figure 1.1: Dilute magnetic semiconductors. Schematic diagram of (a) non-magnetic semiconductor (b) dilute magnetic semiconductor and (c) magnetic semiconductor. This figure is adapted from [1].

[21, 22, 23]. A signature of the coupling between the carriers and the ferromagnetic phase in this type of material is the anomalous Hall effect.

1.2 Anomalous Hall effect

The anomalous Hall effect usually exists in magnetic metals and semiconductors. In these materials the time reversal symmetry is broken due to the spin orbit coupling [24]. The anomalous Hall resistivity usually tracks the magnetization of the material [10, 20, 18] and an example of the Hall resistivity $\rho_{xy}$ consisting of both ordinary and anomalous Hall effects is shown in Fig. 1.2a. When the sample temperature is lower than the Curie temperature $T < T_c$, the hysteretic open loop in $\rho_{xy}$ in the low field magnetic region is attributed to the anomalous Hall effect while the linear component
Throughout the entire magnetic field region is associated with the ordinary Hall effect. At an elevated temperature $T > T_c$, the anomalous Hall effect diminishes due to the ferromagnetic to paramagnetic phase transition and only the ordinary Hall component still is present (assuming the carrier concentration is independent of temperature). The contribution from the anomalous Hall effect alone can be extracted by subtracting the ordinary Hall component and is shown in Fig. 1.2b. The sharp saturation of the $\rho_{xy}$ and the open hysteretic loop resemble the magnetization behavior in ferromagnetic materials.

In Ref. [24], three mechanisms (i) deflection due to Berry phase (ii) side jump and (iii) skew scattering are proposed as possible origins for the anomalous Hall effect. Phenomenologically, these mechanisms can be interpreted as follows [24]. Deflection
due to Berry phase refers to spin up and spin down electrons acquiring different transverse velocities due to their Berry curvature in momentum space with the presence of a longitudinal electric field. The side jump means that electrons with different spins would undergo opposite deflection due to the reversed electric field direction they experience when bypassing an impurity from two different sides. Skew scattering refers to electrons with different spins experiencing asymmetric scattering owing to the different spin orbital interaction when scattering with the impurities from the same side. As the latter two mechanisms involve impurities, they are also called extrinsic mechanisms, while the first one is referred to as an intrinsic mechanism. Side jump and skew scattering result in different shapes of the anomalous Hall resistivity. For side jump $\rho_{xy} \propto \rho_{xx} \times m$ while for skew scattering $\rho_{xy} \propto \rho_{xx}^2 \times m$ where $\rho_{xx}$ is the longitudinal resistivity and $m$ is the magnetization [1, 24].

1.3 Spin waves and magnons

To discuss spin waves and magnons, a classic Heisenberg model is employed. A Heisenberg model describes the coupling interaction between two neighboring electrons with overlapping wave functions in a magnetically ordered structure. The energy possessed by the set of ordered spins is calculated by the sum over all bonds (assumed to be N) between the adjacent electrons and can be described as,

$$U_p = -2J \sum_{p=1}^{N} \vec{S}_p \cdot \vec{S}_{p+1}$$

where $J$ is the exchange energy between the adjacent spins and $\vec{S}_p$, $\vec{S}_{p+1}$ are the spin angular momentum at site $p$ and $p+1$. The spins would favor a parallel configuration (ferromagnet) if $J$ is positive or an antiparallel configuration (anti-ferromagnet) if $J$
is negative. By adopting this formalism, at absolute zero, the energy of the ground state of the 1D spin chain (all spins are static and aligned along one direction, not shown here) can be calculated as $U_0 = -2NJS^2$.

When the spin system that is originally in the ground state is subjected to an elevated temperature, the magnetic ordering leaves its ground state and tends to experience a distortion. This excited state is shown in Fig. 1.4 where all spins share the thermal energy and experience a small deviation from the ground state in the form of precession along a specific direction (z-direction in this case). The internal energy of the $p^{th}$ spin in the excited state can be calculated as [2],

$$U_p = -2J\vec{S}_p \cdot (\vec{S}_{p-1} + \vec{S}_{p+1})$$ \hspace{1cm} (1.2)

The exchange energy can be represented in the form of the production of its magnetic moment and the exerted effective magnetic field as,

$$U_p = -\vec{\mu}_p \cdot \vec{B}_p$$ \hspace{1cm} (1.3)
We can rewrite the magnetic moment of an electron as $-\vec{\mu}_p = -g\mu_B\vec{S}_p$ where $g$ is the g-factor and $\mu_B$ is the Bohr magneton. The effective exchange magnetic field $\vec{B}_p$ felt by the $p^{th}$ spin can be derived by comparing Eqn. 1.2 and 1.3. The resulting form is $\vec{B}_p = \left(\frac{2J}{g\mu_B}\right)(\vec{S}_{p-1} + \vec{S}_{p+1})$. All of the vectors used in the previous and following paragraphs are shown in Fig. 1.3.

The time-dependent magnetization $\vec{\mu}_p(t)$ under the effective exchange magnetic field $\vec{B}_p(t)$ can be solved by the Landau-Lifshitz-Gilbert (LLG) equation as follows,

$$\frac{\hbar}{g\mu_B} \frac{d\vec{\mu}_p(t)}{dt} = -\vec{\mu}_p(t) \times \vec{B}_p(t)$$ (1.4)

where $\vec{\mu}_p(t) \times \vec{B}_p(t)$ is the torque induced by $\vec{B}_p(t)$, and $\frac{\hbar}{g\mu_B} \frac{d\vec{\mu}_p(t)}{dt}$ is the change in magnetization induced by the torque. The dynamic magnetization $\vec{\mu}_p^{x,y}(ka, t)$ can then be solved and the solution form is analogous to a plane wave expressed as,

$$\vec{\mu}_p^{x,y}(ka, t) = \vec{\mu}_{p,0}^{x,y}(ka) \cdot e^{i(ka-\omega t)}$$ (1.5)
where the characteristic frequency and wave vector are \( \omega \) and \( k \). As a result, Eqn. 1.5 implies that the configuration of the 1D spin chain, as well as the individual local spins, exhibit an oscillatory motion (shown in Fig. 1.4). This coherent precession of the dynamic magnetization \( \vec{\mu}_{p,0}(ka) \) is defined as a spin wave. Analogous to the definition of a phonon (a collective excitation of the periodic and elastic arrangement of atoms), the quantized form of the spin wave is defined as a magnon.

1.4 Spin current carried by spin waves (magnons)

Ferromagnetic insulators on the other hand are electrically inactive, due to the absence of conduction carriers. However they are magnetically active due to the spins of localized electrons. The spin flux \( (j_s) \) is closely related to its magnetic precession. In the previous paragraphs, Eqn. 1.4 shows the simple form of the LLG equation. The complete description of the LLG equation, including the damping term reads \[ [14, 25], \]

\[
\frac{\partial \vec{M}(\vec{r},t)}{\partial t} = \gamma \vec{H}_{eff} \times \vec{M}(\vec{r},t) + \frac{\alpha}{M} \vec{M} \times \nabla^2 \vec{M}(\vec{r},t) \quad (1.6)
\]

where \( \gamma \) is the gyromagnetic ratio, \( \vec{H}_{eff} \) describes the total effective magnetic field, including external magnetic field and the exchange field, and \( \alpha \) is the Gilbert damping factor. For small distortions of the spin lattice, \( \vec{H}_{eff} = A \nabla^2 \vec{M}(\vec{r}) \) where \( A \) is the spin stiffness constant. Eqn. 1.6 can then be rewritten as,

\[
\frac{\partial \vec{M}(\vec{r},t)}{\partial t} = A\gamma \nabla^2 \vec{M}(\vec{r}) \times \vec{M}(\vec{r},t) + \frac{\alpha}{M} \vec{M} \times \nabla^2 \vec{M}(\vec{r},t) \quad (1.7)
\]

Applying the vector identity \( \vec{A} \times \nabla^2 \vec{A} = \nabla \cdot \left( \vec{A} \times \nabla \vec{A} \right) \), Eqn. 1.7 can be simplified as (neglecting the Gilbert damping term for simplicity),

\[8\]
\[
\frac{\partial \vec{M}(\vec{r}, t)}{\partial t} = \nabla \cdot \left[ A\gamma \nabla \vec{M}(\vec{r}) \times \vec{M}(\vec{r}, t) \right]
\]  \hspace{1cm} (1.8)

The above equation has the same form of a continuity equation and we can define a spin flux \( \vec{j}_s = A\gamma \nabla \vec{M}(\vec{r}) \times \vec{M}(\vec{r}, t) \). The LLG equation 1.4 can finally be expressed in terms of spin current \( \vec{j}_s \) as \cite{14},

\[
\frac{\partial \vec{M}}{\partial t} = -\nabla \cdot \vec{j}_s
\]  \hspace{1cm} (1.9)

This equation implies that time varying magnetic moments are equivalent to a flow of an spin flux \( \vec{j}_s \). Since we know the general solution form of the LLG equation is a plane wave, we can substitute \( \phi(\vec{r}, t) = M(\vec{r}, t) \) in Eqn. 1.9 and get \cite{14},

\[
\vec{j}_s = \hbar \gamma \sum_q \vec{v}_q n_q
\]  \hspace{1cm} (1.10)

where \( \vec{v}_q \) is the group velocity of the spin wave and \( n_q \) is the number of spin waves. A net spin current will be produced when \( n_q \) is different in k-space between \( q \) and \( q' \) points. This imbalance of the numbers of the spin waves can be induced by a temperature gradient.

### 1.5 Spin Hall effect and inverse spin Hall effect

The spin Hall effect (SHE), as shown in Fig. 1.5a, refers to the conversion of a charge current \( j_c \) into a transverse conduction electron spin current \( j_s \) via spin orbital interaction where spin accumulation of different signs can be found at the opposing boundaries. The induced spin current reads \cite{26},

\[
\vec{j}_s = \theta_{SH} \vec{\sigma}_s \times \vec{j}_x
\]  \hspace{1cm} (1.11)
where $\theta_{SH}$ is the spin Hall angle and $\sigma_s$ is the spin polarization vector.

The inverse spin Hall effect (SHE), as shown in Fig. 1.5b, refers to the conversion of a spin current $j_s$ into a transverse conduction electron current $j_e$ via spin orbital interaction. If the sample is measured under open circuit conditions, electron accumulation can be found at one of the boundaries.

### 1.6 Spin Seebeck effect

The spin Seebeck effect (SSE) is a type of spin caloritronic effect that consists of a two-step process where spin transfer takes place across the interface between a material with spin polarization (e.g. ferromagnet insulator FM) and a normal metal (NM). Specifically, in the first step, a temperature gradient is applied to the spin-polarized material, resulting in a heat flux $j_Q$. This temperature gradient also exerts a thermodynamic force on the spin carriers (e.g. magnons in ferromagnet insulator) that drives the spin carriers out of thermal equilibrium through phonon-magnon interactions. As a result, a pure spin flux/current $j_s$ (which can be viewed as...
as a nonequilibrium magnon flow or as a spin wave spin current flux as discussed in Sec. 1.4) is generated, subject to the applied temperature gradient. In the second step, due to the non-equilibrium nature of spin carriers between the FM and NM, the spin flux is injected into the NM across the FM and NM interface. When the NM possesses strong spin-orbit interactions (like Pt, W and Ta), the injected spin current $j_s$ generates a perpendicular charge current $j_c$ through the inverse spin Hall effect (discussed in Sec. 1.5). As a result, a transverse electrical field $E_{ISHE}$ develops across the NM. This electric field can be detected as a voltage $V_{ISHE}$ when measured in an open circuit condition. Phenomenologically, $V_{ISHE}$ can be expressed as $[4, 15]$,
\[ E_{ISHE} = (\theta_{SH}\rho_{NM})j_s \times \sigma_s \]  

where \( \theta_{SH} \) is the spin Hall angle, \( \rho_{NM} \) is the resistivity of NM, \( j_s \) is the spin current density and \( \sigma_s \) is the spin polarization vector.

The spin Seebeck effect was first observed by Uchida et al. in a ferromagnetic metal, permalloy [15]. Later, it was also observed in ferromagnetic semiconductors [27] and insulators [28]. In this thesis, we focus on the material systems consisting of yttrium iron garnet or YIG \((\text{Y}_3\text{Fe}_5\text{O}_{12})\) as the FM and platinum (Pt) as the NM. The spin Seebeck effect can be measured in two geometries, i.e. longitudinal [4, 29] and transverse [15, 27]. In longitudinal geometry as shown in Fig. 1.6, a temperature gradient in a ferromagnetic insulator (e.g. YIG) is applied along the z-direction, and a magnetic field is applied along the y-direction providing the spin-polarization \( S \) and net magnetization \( M_y \). The inverse spin Hall voltage \( V_{ISHE} \) (spin Seebeck signal) is measured in the x-direction. The transverse geometry is not covered in this thesis and is not discussed here.

1.7 Overview of the thesis

In the first part of this thesis (chapter 2 and 3), the structural, magnetic, and magnetotransport properties of magnetically doped GaN thin film and the 2D material MoS\(_2\) with Gd and Mn respectively, are studied. In chapter 2, a leakage free AlN/GaN 2 dimensional electron gas (2DEG) structure is grown on a SiC wafer using molecular beam epitaxy (MBE). The Gd dopants are introduced at the AlN/GaN interface where the 2DEG concentration peaks. Secondary ion mass spectroscopy (SIMS), high resolution X-ray diffraction (XRD) and atomic force microscopy (AFM)
confirm the successful growth of the target structure. Magnetization measurements on these Gd doped (and undoped) AlN/GaN heterostructure samples are performed using superconducting quantum interference device (SQUID) magnetometry. Room temperature defect-induced ferromagnetism with easy axis along the growth direction (c-axis) is found in the Gd doped AlN/GaN heterostructures. Magnetotransport measurements are performed using physical property measurement system (PPMS) at various temperatures. The comparison between the magnetization and non-linear Hall resistivity suggests the lack of coupling between the conduction electrons and the Gd-induced ferromagnetic phase. The non-linear Hall slope is likely a result of the mixing of the magnetoconductivity tensor into the Hall resistivity. In chapter 3, Mn doped few-layer MoS$_2$ is synthesized by sulfurizing the Mn:Mo thin film on sapphire substrate. Energy dispersive x ray spectroscopy (EDX), raman spectroscopy, XRD, and AFM characterization show the growth of single crystal few-layer Mn:MoS$_2$ along the (001) direction on a sapphire substrate with target Mn concentration of 12.5 % (atomic ratio to Mo). The magnetic properties of these Mn doped 2D materials are examined by SQUID. It is found that, contradictory to the theoretical prediction, few-layer Mn:MoS$_2$ is dominated by the paramagnetism and the Mn dopants do not experience long range ferromagnetic coupling.

In the second part of this thesis chapters 4, 5, 6 and 7, magnon transport properties and dynamics are probed via the spin Seebeck effect and compared to finite element method (FEM) simulations. In chapter 4, we employ a new measurement scheme, the nonlocal opto-thermal spin Seebeck configuration, to detect the diffusion of thermally generated magnons. In this experiment, a Ti:sapphire laser is used to remotely excite magnons and a lock-in amplifier is used to detect these diffused
magnons as a function of distance to the laser. The spatial dependence of the diffused spin signal shows an exponential decay with a characteristic decay length of $\sim 47 \, \mu\text{m}$ at 23 K. FEM simulations based on the thermal and spin transport equations are employed to confirm that the detected signal is purely from magnon diffusion. An upper bound of the spin diffusion length ($\sim 63 \, \mu\text{m}$) is also determined through the spin transport modeling. In chapter 5, the time domain spin Seebeck waveforms are measured through the time resolved longitudinal spin Seebeck effect. The temporal spin Seebeck signal exhibits a fast and slow rise component. Based on the theory of the spin Seebeck effect and the simulation results, the fast rise is attributed to the spin current due to the interfacial electron magnon temperature difference while the slow rise arises from the bulk magnon diffusion. Coupled thermal and spin transport modeling points out that spin diffusion governs the speed of the slow rise component at lower temperature and thermal diffusion is the dominate mechanism at higher temperatures. The fast rise time is found to be temperature independent and on par within the laser power rise time, while the slow rise time continuously grows as temperature increases. In chapter 6, a thermoelectric generator based on the spin Seebeck effect consisting of Pt and Fe$_2$O$_3$ is proposed and attempted first. However, such a device performs poorly due to the low quality Pt/Fe$_2$O$_3$ interface. A modified thermoelectric generator based on the ordinary (anomalous) Nernst effect is realized using a magnetostrictive material, Galfenol. Scalable Nernst voltage/power generation is demonstrated by varying the Galfenol wire length. The Nernst coefficient of Galfenol is found to be $\sim 2.6 \, \mu\text{V/KT}$ which is the highest among different types of ferromagnetic materials. In chapter 7, the thermal and condensed magnon
transport equations are carefully re-derived from Ref. [30] so that they can be implemented in the finite element method software COMSOL. The magnon chemical potential, together with thermal and condensed magnon current profiles, is solved and discussed under different conditions. An experiment design on probing the magnon Bose-Einstein condensation is proposed last.
Chapter 2: Anisotropic Defect-induced Ferromagnetism and Magneto-transport in Gd-doped GaN Two-dimensional Electron Gasses

This chapter is adapted from the previously published work in Ref. [31] by the authors Zihao Yang, Thomas F. Kent, Jing Yang, Hyungyu Jin, Joseph P. Heremans and Roberto C. Myers.

There have been plenty of studies on material systems that contain tunability of both charge transport in semiconductors and spin transport in ferromagnets, since these types of material systems open up the possibility of integrating new functionality into current electronic devices. There are multiple ways of realizing these types of material systems. Among them, the growth and characterizations of semiconductors exhibiting ferromagnetic behavior has been a major focus since Ohno et al. [32] discovered MBE grown Mn doped GaAs possessed ferromagnetic magnetization characteristics. In this type of material, although Mn dopants (usually to $10^{20} \text{ cm}^{-3}$) are randomly distributed throughout the film, the spins of Mn are coupled and exhibit collective ferromagnetic ordering, and therefore they are also called dilute magnetic semiconductors (DMS). Later on multiple devices like Hall-bar based transistors and LED structures have been made, demonstrating that GaMnAs exhibits the anomalous Hall effect and spin polarized electroluminescence emission. These studies prove
that the carriers associated with Mn (mainly holes) are coupled to the ferromagnetic phase and thus are spin polarized. However, the Curie temperature ($T_c$) of GaMnAs is well below room temperature, limiting its practical applications.

Besides Mn-doped III-arsenides, III-nitrides are also widely studied as host materials for magnetic transition metal doping. They are particularly good host materials for the magnetic rare earth element gadolinium (Gd). The Gd ion contains 7 unpaired f-shell electron spins and it has relatively high solubility in GaN, which opens the possibility of engineering magnetism into nitride semiconductors [21, 33]. In the dilute doping regime, Dhar et al. showed Gd:GaN possessed a colossal ferromagnetic moment (per Gd ion) with a $T_c$ persistent up to room temperature [21]. This magnetism is considered to originate from the crystallographic point or line defects, due to its colossal nature [21, 22, 34, 35]. This room temperature ferromagnetic phase may point to a new magnetic semiconductor material system that overcomes the disadvantage of the $T_c$ being too low in the prototypic DMS GaMnAs [36]. However, there are no reports on the electrical transport properties together with the magnetic properties of the Gd-doped GaN or its related structures. Lo et al. observed the anomalous Hall effect in Gd ion-implanted GaN/AlGaN 2DEGs using a Van der Pauw geometry, however the magnetic properties of these samples were not presented [37]. Recently, Buβ et al. suggested the absence of exchange coupling between Gd ions or Gd associated polarized lattice ions and conduction band electrons in Gd ion-implanted GaN thin films through time-resolved magneto-optical spectroscopy [38]. Given the controversies in the previous literature, the use of Gd doped GaN as a ferromagnetic semiconductor is still unclear.
To elucidate this problem, we performed magnetization and magnetotransport measurements in Gd delta-doped polarization-induced AlN/GaN two-dimensional electron gas (2DEG). The direct probing of the coupling between the anomalous ferromagnetic phase and the conduction band electrons in Gd-doped GaN is made possible in these samples, since the conduction electrons that are confined in quantum wells at the AlN/GaN interface are spatially overlapped with the magnetically doped region. However, the leakage current through the GaN buffer layer and the regrowth interface in these GaN HEMT devices (Sec. 2.1) introduces contaminations to the transport signal from the 2DEG which prevents accurate magnetotransport measurements through the 2DEG. In Sec. 2.2, the method of eliminating such leakage current and the growth of leakage-free Gd doped GaN samples on SiC is discussed. In Sections 2.4 and 2.5, the measurements of magnetic properties and magnetotransport properties and their comparison to Gd doped 2DEG are discussed. The possible explanation of the nonlinear Hall slope and the lack of coupling between ferromagnetic phase and conduction electrons is given in Sec. 2.5 and 2.6.

2.1 Leakage current in AlN/GaN heterostructures grown on GaN templates

The presence of a leakage current through the GaN buffer and the regrowth interface is a known issue in GaN HEMTs. A HEMT structure with an AlN back barrier is initially grown with the aim of eliminating the leakage current. The samples (without any Gd doping) are grown on a Kyma GaN template (i.e. GaN on sapphire) and consist of the following layers (from top to bottom) 4 nm AlN capping layer/ 150 nm smooth GaN / 2 nm AlN back barrier / 100 nm GaN buffer / GaN template / Sapphire. The band diagram of this structure is shown in Fig. 2.1. It can be
found that the quantum well and 2DEG is formed at the AlN/GaN interface close to the surface. In addition, due to the difference in piezoelectric charge density between the smooth GaN layer and the strained AlN back barrier, an electric field is built up in the smooth GaN region bending the GaN conduction band up as it approaches the regrowth interface. This electric field repels electrons from flowing towards the regrowth interface and thus reduces the leakage current.

Figure 2.1: Band diagram of AlN/GaN heterostructure grown on Kyma GaN template with AlN back barrier to minimize the leakage current.

A buffer leakage test is performed on this device to examine the presence or absence of the leakage current. Two devices were fabricated on the same sample where the first device kept the conducting 2DEG channel between the two contacts while the second one had the 2DEG channel etched away (meaning the only possible conduction path is through the buffer and the regrowth interface). The IV characteristics are shown in Fig. 2.2 for both devices. The magnitude of the current conducted through the GaN buffer is only $\sim 10$ times smaller than the 2DEG in this device which
implies that AlN back barrier does not completely eliminate the leakage current. Since
the magnetotransport signals are expected to be very sensitive to the contamination
current, a better sample design with complete elimination of the leakage current is
required. The growth and characterizations of the leakage-free AlN/GaN 2DEG on
6H-SiC are discussed in the following sections.

![Figure 2.2: Comparison between the current conducted through 2DEG and through
buffer/regrowth interface in the AlN/GaN heterostructure grown on Kyma GaN tem-
plate with AlN back barrier before and after removing of the 2DEG conduction chan-
nel.](image)

2.2 Growth of leakage-free Gd doped AlN/GaN heterostructures on SiC wafers

All samples discussed in this chapter are grown by plasma-assisted molecular beam
epitaxy (PAMBE) on a Veeco GEN930 PAMBE system. The source materials Al,
Ga, Gd in the effusion cells have a purity of 6N5, 7N and 4N7 respectively, while the
purity of the nitrogen plasma source is 6N [33, 39]. The base pressure of the PAMBE system is typically in the mid 10^{-11} torr range.

The Gd doping rate is first calibrated to achieve precise control of the Gd doping concentration. This is done by performing SIMS measurement on a calibration sample consisting of different layers of Gd doped GaN thin films grown at various Gd effusion cell temperatures. To be more specific, the first Gd doped GaN layer is grown at a Gd effusion cell temperature of 900 °C on an AlN on sapphire (Kyma) template for 30 minutes. The subsequent Gd doped GaN layers are grown on top of the first Gd doped GaN layer with an increasing Gd effusion cell temperatures from 950 °C to 1150 °C with 50 °C step for 15 minutes.

A PHI6600 Quadrupole secondary ion mass spectrometry (SIMS) instrument with 5 keV O\textsubscript{2} primary ion bombardment is used to measure the Gd concentration in these layers with different growth conditions. Prior to the SIMS measurement, Rutherford backscattering spectroscopy (RBS) on a Gd ion implanted standard sample with an implant dose of 1.01 \times 10^{15} cm\textsuperscript{-2} was performed for calibration purposes. The Gd doping concentration for five major Gd isotopes of the calibration sample are measured separately and plotted as thin colored lines while the averaged final Gd doping profile is plotted as a thick black line in Fig. 2.3a. The final averaged Gd doping concentration (black dots on a linear scale) is then plotted as a function of the reciprocal of the Gd cell temperature as shown in Fig. 2.3b. This temperature dependency can be described and fit (red dotted line) using the Clapeyron equation \( n_{Gd} \propto p_{Gd} = A \times e^{-b/T} \) where \( n_{Gd} \) is the Gd doping concentration and \( p_{Gd} \) is the equilibrium partial pressure of the Gd beam [40]. The Gd doping rate (in units of effective Gd monolayer (ML) coverage per second) is then calculated by assuming
GdN remains in the wurtzite structure. The doping rate is shown as a function of the reciprocal of the Gd cell temperature on a semilog scale in Fig. 2.3c. The Gd growth rate for all samples used in this chapter is 0.007 ML/s (blue open circle) which is extrapolated from the Clapeyron fitting in Fig. 2.3c. This corresponds to a Gd effusion cell temperature of 1169.4 °C.

Fig. 2.4 shows the structures of the two sets of samples that will be discussed in this chapter. All of them are grown on the Si face of 6H-SiC wafers with different Gd doping concentrations. Their structures consist of the following layers (from top to bottom): 5 nm AlN (Al-face) capping layer / 0.4 nm GaN spacer / 0 ML or 0.2 ML or 0.4 ML Gd-δ-doping / 300 nm smooth unintentionally doped (UID) GaN (Ga-face) / 140 nm rough UID GaN / 70 nm UID AlN nucleation layer / 6H-SiC wafer. The sample without any Gd doping served as a control sample in this study. The denotations of the samples are listed in Table 2.1. It should be noted here that Kent et al. demonstrated the forming of rocksalt GdN nano-islands in a GaN matrix with a Gd delta dosage greater than 1.2 ML [33]. The Gd dosage used in this study is well below this critical value and therefore it is expected that only single phase wurtzite Gd doped GaN would form.

<table>
<thead>
<tr>
<th>Gd doping</th>
<th>Denotation</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 ML</td>
<td>Control Sample 1</td>
</tr>
<tr>
<td>First Set</td>
<td>0.2 ML $N_{2D}^{Gd}=0.2$ML</td>
</tr>
<tr>
<td></td>
<td>0 ML Control Sample 2</td>
</tr>
<tr>
<td>Second Set</td>
<td>0.4 ML $N_{2D}^{Gd}=0.4$ML</td>
</tr>
</tbody>
</table>
Figure 2.3: Gd doping calibration. (a) Gd concentration depth profile of the five major Gd isotopes (colored line) and the final averaged concentration (black line) of the Gd doped GaN calibration sample grown with different Gd effusion cell temperatures characterized by SIMS. (b) Averaged Gd doping concentration of the Gd doped GaN calibration sample as a function of the reciprocal of the Gd cell temperature with fitted curve using Clapeyron equation. (c) Calculated Gd doping rate as a function of Gd cell temperature with fitted curve using Clapeyron equation.

During the growth, the chamber pressure is held at $2 \times 10^{-5}$ torr. The nitrogen limited growth rates of GaN for the first and second set of samples are 250 and 290 nm/hour respectively. The growth rate of AlN is slightly slower (by $\sim 0.8\%$) than
Figure 2.4: Schematic of the sample structures used in this study.

The GaN growth rate. The detailed growth conditions of each layer are discussed as follows. The first AlN nucleation layer is grown under a nitrogen rich regime (Al/N flux ratio of 0.6) at 780 °C. The subsequent rough GaN layer is grown under a Ga rich intermediate regime (Ga/N flux ratio of 1.7). The smooth GaN layer on top is grown under a droplet regime (Ga/N flux ratio of 2.6) at 710 °C. After the growth of the smooth GaN, the shutter for the Ga effusion cell is closed and the shutter for the Gd effusion cell is opened for 30 seconds (0.2 ML Gd-doping) or 1 minute (0.4 ML Gd-doping). An additional 0.4 nm GaN spacer is grown upon finishing the Gd dopants deposition to ensure a good AlN/GaN heterointerface. The top most AlN capping layer is grown under the Al-rich intermediate regime (Al/N flux ratio ∼ 1) at 710 °C.

A similar leakage current test on these AlN/GaN heterostructures grown on SiC wafers with a AlN nucleation layer is taken to examine the removal of the current path through the GaN buffer and regrowth interface. The result is shown in Fig. 2.5. Compared to the samples grown on Kyma templates, the samples grown on SiC exhibit much lower leakage current, where the ratio between the leakage and 2DEG
current is around $10^6$. The AlN nucleation layer used in the samples grown on SiC wafers provides a physical barrier for the electrons that can diffuse to the regrowth interface. The successful removal of the leakage current ensures the magnetotransport measurement in Sec. 2.5 is solely from the 2DEG.

![Figure 2.5: Comparison between the current conducted through 2DEG and buffer/regrowth interface in the AlN/GaN heterostructure grown on Kyma GaN template with AlN back barrier in log scale (main panel) and linear scale (right inset) before and after removing of the 2DEG conduction channel of the Control Sample 1. Left inset: the sample structures used in the leakage test.](image)

**2.3 Band diagram and structural properties of Gd doped AlN/GaN heterostructures**

In order to understand the electrical transport in these samples, we simulated the band diagram of the AlN/GaN structure without Gd doping (control samples) using the one dimensional Poisson Schrodinger solver, BandEng [41]. The calculated band alignments and carrier concentration profiles are shown in Fig. 2.6a. The fermi level in the AlN capping layer is pinned at 3.1 eV below the conduction band minimum due to the presence of surface states. The conduction band offset between AlN and GaN...
$(\Delta E_c^0)$ is set to be 2.1 eV. The background doping concentration in the smooth GaN region is set to be n-type with a concentration of $10^{16} \text{cm}^{-3}$ due to the incorporation of oxygen impurities [42].

Here we compared two cases where the AlN layer is either (i) fully strained (dotted lines) or (ii) partially relaxed (solid lines). For case (i), fully strained AlN exhibits stronger piezoelectric charge density and results in a sharper band bending in GaN and a deeper quantum well at the AlN/GaN interface. The carrier concentration is calculated to be around $4.2 \times 10^{13} \text{cm}^{-3}$ for case (i). This is different than the carrier concentration measured in Sec. 2.5.

This discrepancy can be explained by carefully revising the input parameters in the simulation. First, the AlN capping layer is partially strain relaxed rather than fully strained. In this case, the piezoelectric charge density would decrease, leading to a shallower quantum well at the AlN and GaN interface. Second, the conduction band offset of 2.1 eV between AlN and GaN is overestimated and needs to be revised since the AlN band gap tends to shrink under the biaxial tensile strain. Assuming all the band gap shrinkage in AlN contributes to the reduction of conduction band offset, the amount of reduction can be calculated as [43],

$$\Delta E_c^0 - \Delta E_c = -[(a_{cz} - D_1)e_\perp + 2(a_{ct} - D_2)e_\parallel]$$

(2.1)

where $e_\perp$ and $e_\parallel$ are uniaxial and biaxial strain, $d_1$ and $d_2$ are deformation energy, $\Delta E_c$ is the new conduction band offset when AlN is strained [43, 44]. Third, due to the non-parabolic energy dispersion in the triangular quantum well, the resulting electron effective mass in GaN will increase and can be calculated by the Ando formula as [45],
\[ \frac{\Delta m^*}{m^*} = \sqrt{1 + 4 \times \frac{< K >_i + E_f}{E_g}} - 1 \]  

(2.2)

where \(< K >_i \) is the kinetic energy of the motion perpendicular to the interface and \(E_F\) is the Fermi energy measured from the lowest subband. Here, \(< K >_i \) can be calculated as \(< K >_i = E_i/3\) with \(i = 1\) and \(E_i\) being the subband energy measured from the conduction band minimum. This is due to the fact that most of the electrons reside in the first subband. A simulation using these revised parameters on a fixed structure of 5 nm AlN / 300 nm GaN is then conducted. It is found that with \(R = 68\%, \Delta E_c = 1.93 \text{eV}, m_0 = 0.24 m_0 \) and \(R = 74\%, \Delta E_c = 1.96 \text{eV}, m^* = 0.24 m_0\), the simulated carrier concentration matches the measured one for Control Sample 1 and 2 respectively. The AlN in Control Sample 1 is slightly less relaxed compared to Control Sample 2. This agrees with the slightly higher concentration in Control Sample 1. The simulated band diagram and carrier concentration profile of the case (ii) partially relaxed AlN layer with revised physical parameters of Control Samples 2 is shown as the solid line in Fig. 2.6a. In this case, the reduced piezoelectric charge and band offset leads to a shallower quantum well and lower carrier concentration. For Gd doped samples, due to the lack of information on how the band offset and electron effective mass depend on the Gd doping, the band diagrams and carrier concentration of the Gd doped samples are not attempted here. However, the observation of a partially relaxed AlN layer in the control samples suggests a similar situation exists in Gd doped samples.

The calibrated Gd concentration depth profile is measured using SIMS with 2 keV \(CsM^+\) primary ion as shown in Fig. 2.6b. It can been seen that the Gd doping concentration peaks at 0.4 nm away from the AlN/GaN interface with a surface
concentration of $2.1 \times 10^{14} \text{ cm}^{-2}$. This doping concentration corresponds to 0.2 ML equivalent of Gd dopant atoms. Comparing Fig. 2.6a and 2.6b, it can be concluded that the Gd dopants are spatially overlapped with the 2DEG region. It also should be noted that although there is a long tail of Gd concentration that extends into the underlying GaN region which is indicative of the Gd dopants of thermal diffusion, the majority of Gd dopants are still present in the quantum well near the interface. In addition, the Ga, Al, and N concentrations are not calibrated. Although they are just raw signals, they can be used to identify the AlN and GaN regions and mark out their interface. It has to be noted that the decrease in Al concentration near the AlN/air interface is an artifact in the SIMS measurement.
The structural properties of the AlN/GaN heterostructure are characterized by high resolution X-ray diffraction (XRD) and atomic force microscopy (AFM). A representative $\omega - 2\theta$ XRD scan on the Control Sample 1 is shown in Fig. 2.7a. Wurtzite (002) GaN and AlN peaks can be clearly observed, demonstrating the epitaxial growth and single crystal nature of such layers. The rocking curve for the (002) GaN peak is shown as an inset in Fig. 2.7a. The full width half maximum (FWHM) of the (002) and (102) GaN peaks are 792 and 1116 arcsec respectively [46, 47]. These values are higher than the MBE grown GaN in the previous reports. The density of screw type threading dislocations can be estimated from the FWHM of the rocking curve based on the Hirsch model [48, 49, 50] which reads,

$$D_s = \frac{\beta_{002}^2}{4.35 \times |b_s|^2}$$  \hspace{1cm} (2.3)

where $|b_s|$ ($|b_s|=0.52$ nm) is the Burgers vector of the screw type dislocations and $\beta_{002}$ is the FWHM of the (002) GaN rocking curve. The screw type dislocation density is then calculated to be around $6.3 \times 10^8$ cm$^{-2}$. In terms of the other type of threading dislocation, the edge threading dislocation, it has been shown in Ref. [49] that similar FWHM of the (002) and (102) peak results in similar screw and edge type threading dislocations. Therefore, the edge threading dislocation density is estimated to be $\sim 10^9$ cm$^{-2}$ which is similar to the screw type one.

The surface morphology of the 0.2 ML and 0.4 ML Gd doped samples are characterized using AFM and are shown in Fig. 2.7b and c, where the surface roughness (RMS) are 0.56 and 0.43 nm respectively. The step flow growth mode of the GaN and AlN layers can be identified from the terrace features and smooth RMS of both samples.
Figure 2.7: Structural properties of Gd doped AlN/GaN heterostructures. (a) High resolution X-ray diffraction $\omega - 2\theta$ scan of Control Sample 1. Inset: rocking curves of the (002) and (102) diffraction peaks. AFM characterization of the (b) 0.2 ML and (c) 0.4 ML Gd doped sample.

2.4 Magnetic properties of the undoped and Gd doped AlN/GaN heterostructures

The key to reveal the existence or absence of the coupling between the Gd induced magnetic phase and conduction electrons in GaN is to directly compare the magnetization and Hall resistivity in the Gd doped samples. The magnetic properties of the control samples and Gd doped samples are measured using the superconducting quantum interference device (SQUID) in a Quantum Design MPMS XL. The magnetization of these samples are shown in Fig. 2.8 after subtracting the diamagnetic background from SiC. The volume diamagnetic susceptibility (in c.g.s unit) of the SiC is calculated to be around $1.17 \times 10^{-5} \text{ cm}^3/\text{mol}$. This value is close to the previous reported value of $1.06 \times 10^{-5} \text{ cm}^3/\text{mol}$ [51]. Also, all the magnetization data
presented in this chapter are assumed to be free of transition metal contamination since all samples were cleaned with HCl acid and solvents and prepared in the clean room environment with plastic tweezers.

The out-of-plane magnetization data of the control samples and Gd doped samples at 300 K are shown in Fig. 2.8a. The magnetization data have been normalized by the sample area due to the 2D nature of the Gd doping. The 0.2 ML Gd doped sample exhibits a sharp change in the magnetization below 5 kOe and a full saturation up to a value of $6.5 \times 10^{-7}$ emu/mm$^2$ at 20 kOe. This behavior implies the presence of the room temperature ferromagnetism in these samples. Similar magnetization is observed in the 0.4 ML Gd doped sample with a smaller saturation magnetization.

This large variation in the magnetization in samples with different Gd doping concentrations was also observed in Ref. [23]. Same measurement was carried on the 6H-SiC wafer and control samples to examine the role of Gd doping on this ferromagnetic phase. The magnetization measured on the 6H-SiC wafer shown in the top left inset in Fig. 2.8a only contains a very weak ferromagnetic response with a magnetization less than $10^{-7}$ emu/mm$^2$. The origin of this weak marginal ferromagnetism is attributed to the spin polarized defects in SiC[52]. Remarkably, the Control Sample 1, which does not have any Gd doping, exhibits a stronger ferromagnetic behavior compared to the SiC wafer with a saturation magnetization of $1.9 \times 10^{-7}$ emu/mm$^2$. This ferromagnetism in wide bandgap semiconductors is suggested to be originated from the spin polarized point defects or cation vacancies, that exhibit long range coupling [53]. As for the Control Sample 2, it exhibits a stronger paramagnetic response comparing to the SiC wafer as shown by the 5 K magnetization data in the bottom right inset in Fig. 2.8a. Based on these points, we concluded that the control samples
Figure 2.8: (a) Magnetization hysteresis loops of the SiC wafer, control samples and Gd doped samples with magnetic field applied along [0001] (out of plane) at 300 K after diamagnetic background subtraction (same background subtraction applies to the rest of the figures). Inset: comparison of the magnetization from control samples and SiC wafer at 5 K (bottom right) and 300 K (top left). (b) Magnetization hysteresis loops of the 0.2 ML and 0.4 ML Gd doped samples at 5 K and 300 K. (c) Magnetization hysteresis loops of the 0.2 ML Gd doped sample with applied magnetic field parallel to [0001] (out of plane) and perpendicular to [0001] (in plane). (d) Magnetization of the Gd doped samples after zero field cooling with applied magnetic field parallel to [0001]. Inset: raw temperature dependent magnetization of the 0.4 ML Gd doped sample and SiC wafer.
without Gd doping contain free spins that can be either ferromagnetically coupled thus showing ferromagnetism at room temperature (Control Sample 1) or uncoupled thus exhibiting paramagnetism (Control Sample 2). The difference in magnetic response in these control samples is attributed to the variation in the density of spin polarized defects which is very sensitive to the growth condition. Although some of these control samples are not totally free of ferromagnetic response, we did observe a boost in magnetism upon Gd doping which agrees with previous report on Gd doped GaN [21, 54].

Detailed comparison of the out-of-plane magnetization of the Gd doped samples at 5 K and 300 K are shown in Fig. 2.8b. The magnetization data are normalized by the total amount of Gd dopants. The saturation magnetization of the sample doped with 0.2 ML Gd is 30 and 38$\mu_B$/Gd$^{3+}$ taken at 300 K and 5 K respectively. These values are taken at a magnetic field of 20 kOe; more detailed data are shown in Table 2.2. The values of the saturation magnetization are higher than the atomic moment of a single Gd atom (7$\mu_B$/Gd$^{3+}$). Therefore, the magnetization in these Gd doped samples does not mainly come from Gd ions alone. This is not totally surprising since the control samples even without any Gd doping, i.e. Control Sample 1, contain ferromagnetic defects (see discussion above). Multiple origins explaining the Gd induced ferromagnetism in Gd doped GaN have been proposed. Bedoya-Pinto et al. suggested that doping with Gd in GaN is likely to catalytically generate spin polarized accepter-like defects and a defect band will form at a Gd doping level of $2 \times 10^{16}$ cm$^{-3}$, $6 \times 10^{18}$ cm$^{-3}$ [54]. Besides acceptor-like defects, interstitial nitrogen, oxygen in octahedral sites and Ga vacancies have been suggested as the possible origins for the defect induced ferromagnetism in Gd doped GaN in Ref. [34, 35].
Rover et al. later ruled out gallium vacancies and gallium vacancy clusters and suggested that extended defects are more likely to play an import role to contribute to this defect induced ferromagnetism [23]. As for the samples doped with higher Gd concentration, 0.4 ML, the saturation magnetization decreases to 7 and 8.4 $\mu_B/Gd^{3+}$ at 300 K and 5 K respectively. These values are taken at a magnetic field of 20 kOe; more detailed data are shown in Table 2.2. The decrease in the saturation magnetization with the increasing Gd doping concentration is in accordance with previous reports [21, 23]. The saturation magnetization and the coercive field with an in-plane applied magnetic field listed in Table 2.3 only experience slight difference at 5 K and room temperature. This implies that the Curie temperature of these Gd doped samples is well about room temperature.

<table>
<thead>
<tr>
<th>Sample $N_{2D}^{Gd}$</th>
<th>T (K)</th>
<th>$H_c$ (Oe)</th>
<th>$M_s$(Scan Direction) ($\mu_B/Gd^{3+}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2 ML</td>
<td>5</td>
<td>41.1±0.87</td>
<td>-72±134.6(+→−) 113.4±107(−→+)</td>
</tr>
<tr>
<td>300</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.4 ML</td>
<td>5</td>
<td>9.2±1</td>
<td>64.3±255.4(+→−) 203.2±275.1(−→+)</td>
</tr>
<tr>
<td>300</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The magnetic anisotropy of the Gd doped samples is shown in Fig. 2.8c. Two surprising features is observed by comparing the in-plane and out-of-plane magnetic hysteresis loops measured on the 0.2 ML Gd doped samples. First, the magnetic
Table 2.3: In-plane Magnetic Properties

<table>
<thead>
<tr>
<th>Sample</th>
<th>T (K)</th>
<th>H_c (Oe)</th>
<th>M_s(Scan Direction) (µ_B/Gd^{3+})</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>33.2±3</td>
<td>-310.8±83.3(+→−) 365.4±135.4(−→+)</td>
</tr>
<tr>
<td>N_{Gd}^{2D}=0.2ML</td>
<td>300</td>
<td>18.7±2.5</td>
<td>-195.2±94.4(+→−) 186.3±283.5(−→+)</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>7.4±0.1</td>
<td>-325.3±75.3(+→−) 289.3±44.4(−→+)</td>
</tr>
<tr>
<td>N_{Gd}^{2D}=0.4ML</td>
<td>300</td>
<td>2.2±0.4</td>
<td>-282.6±56.8(+→−) 156.4±103.4(−→+)</td>
</tr>
</tbody>
</table>

easy axis is in the out-of-plane direction. This implies that the crystalline anisotropy dominates over the shape anisotropy in these samples, although the latter one is usually much stronger in ferromagnetic thin films. The dominance of the crystalline anisotropy also rules out the possible ferromagnetic dust contamination whose magnetization is usually expected to be isotropic. This out-of-plane easy axis is also in contrast to the previous observation of an in-plane easy-axis in Gd doped GaN thin films report by Perez et al. in Ref.[8]. Our result hints the possible role of the treading dislocations being the origin of the anomalous ferromagnetism. Second the in-plane magnetization does not converge to the out-of-plane one even up to the maximum applied field 50 kOe. This behavior is also consistent with exchange coupled spins locked along the crystallographic c-axis that cannot be rotated in-plane.

The temperature dependency of the magnetization in the Gd doped samples is shown in Fig. 2.8d. The zero field cooled magnetization reveals a decreasing but non-vanishing magnetization up to room temperature which supports the observation
of the room temperature ferromagnetism discussed above. A local peak around 50
K is observed in both Gd doped samples. This temperature is close to the Curie
temperature of cubic GdN whose $T_C$ is reported to be between 40 to 70 K depending
on the growth conditions [33, 55, 56]. However, GdN is not likely the cause of this
peak since the presence of a second ferromagnetic phase would lead to a step-like
feature rather than a peak in the magnetization around $T_C$. The oxygen contamination
due to the air leak could potentially results a peak at 40 to 50 K due to the para-
antiferromagnetic phase transition of solid oxygen [57]. We can also rule out this
possibility by examining the magnetization of the SiC wafer as shown in Fig. 2.8d
inset where its raw magnetization is very small ($1 \times 10^{-7}$ emu which approaches the
SQUID detection limit) and does not show any peak around 50 K. The physical origin
of this peak thus requires further studies. Besides the local peak at around 50 K, a
sharp increase in the magnetization due to paramagnetic spins is also observed at
extremely low temperatures in both Gd doped samples.

2.5 Magnetotransport measurements in undoped and Gd doped
AlN/GaN heterostructures

In order to compare to the magnetization measurement discussed in the last sec-
tion, magnetotransport measurements are performed on these samples using a Quan-
tum Design PPMS 7T Model 6000. The 1-3-3-1 Hall-bar geometry is used based on
the design criteria in ASTM F76 [58]. The dimensions and the measurement config-
uration are shown in Fig. 2.9a where $I_{xx}$ is the injection current, $V_{xy}$ and $V_{xx}$ are
the measured Hall and longitudinal voltage respectively. The image of the processed
device based on this Hall bar design and its 3D schematic are shown in Fig. 2.9b and
2.9c. Due to the small cross section of the 2DEG conduction channel, Joule heating
could be significant. Therefore, a $I_{xx}$ of 10 µA is used for all magnetotransport measurements. As discussed in Sec. 2.1, a leakage current through the regrowth interface severely contaminates the conduction current from the 2DEG in AlN/GaN 2DEG samples grown on GaN/sapphire templates. Thus, a leakage test similar to the one discussed in Sec. 2.2 is performed on the AlN/GaN 2DEG samples grown on SiC. The results are shown in Fig. 2.5 where the leakage current is five orders of magnitude smaller than the 2DEG current. This ensures that the Hall resistivity and magnetoresistance measured from the magnetotransport measurements only come from the 2DEG.

An example of the raw Hall resistivity $\rho_{xy}$ measured on the 0.4 ML Gd doped sample at 20 K is shown in the top left inset of Fig. 2.11a. The $\rho_{xy}$ measured at high magnetic fields shows a linear relationship with respect to the magnetic field and is attributed to the ordinary Hall effect (OHE). This linear component is observed in all samples. The carrier concentration and mobility shown in Fig. 2.10 are extracted from the slope of this linear relationship and the zero field resistivity. The carrier concentrations from all samples are almost constant throughout the entire temperature range due to the temperature independent spontaneous and piezoelectric charge density. The mobilities from all samples increase and saturate with decreasing temperature due to the switching of the scattering mechanism in the 2DEG from the polar phonon scattering to impurity scattering [59]. Both of these behaviors are in good agreement with previous studies on similar structures [60, 59, 61, 62]. It should be noted that the carrier concentration increases with the presence of Gd dopants. This could be a result of the distortion of the crystal potential field and conduction band offset between AlN and GaN at the heterointerface induced by the Gd dopants.
Figure 2.9: Design and schematic of the Hall bar mesa. (a) Dimensions of the 1-3-3-1 Hall bar with labels of input current and measured voltages. (b) Real image of the fabricated Hall bar devices (c) 3D schematic showing the AlN/GaN heterostructure and the fabricated Hall bar mesa.
The change in the local band structures would strongly alert the band diagram and carrier concentration in these samples. Therefore, the boost in carrier concentration in Gd doped samples does not necessary contradict to the presence of acceptor like defects in Gd doped GaN observed by Bedoya-Pinto et al. [54] as discussed in the previous section.

Figure 2.10: Carrier concentration and mobility. (a) Carrier concentration and (b) mobility as a function of temperature of the control samples and Gd doped samples calculated from the Hall measurement (without taking MR mixing effect).

An offset between the linear fit of $\rho_{xy}$ in the high positive and negative magnetic field region in the 0.4 ML Gd doped sample can also be observed in the top left inset of Fig. 2.11a. This indicates a non-linear component exists in the raw $\rho_{xy}$ trace. This
non-linear component in the Hall resistivity $\rho_{xy}^{NL}$ can be extracted by subtracting the OHE component from the raw $\rho_{xy}$. The resulting $\rho_{xy}^{NL}$ as a function of magnetic field for all samples is shown in the main panel of Fig. 2.11a. It can be seen that $\rho_{xy}^{NL}$ from control samples and Gd doped ferromagnetic samples shows similar magnetic field dependence where $\rho_{xy}^{NL}$ first increases to half of the maximum value around 5 kOe and 10 kOe (depending on the sample) and finally saturate around the highest magnetic field of 50 kOe. Due to this similarity in $\rho_{xy}^{NL}$ observed in all samples, it is evident that the Gd-induced defect ferromagnetism does not significantly couple to the conduction electrons in the 2DEG although they are physically overlapped with each other. This is further confirmed by directly comparing the $\rho_{xy}^{NL}/\rho_{xx}^{\gamma}$ and magnetization $m$ in Gd doped samples. In ferromagnetic metals and semiconductors, $\rho_{xy}^{NL}$ follows [1]

$$\rho_{xy}^{NL} = \rho_{xy}^{AHE} = c \times \rho_{xx}^{\gamma} \times m$$  \hspace{1cm} (2.4)

where $c$ is a temperature independent proportionality constant, $\rho_{xx}$ is the magnetoresistance, $\gamma$ is a power constant and $m$ is the magnetization of the sample. Here, $\gamma$ value is determined based on the type of the specific magnetic scattering mechanism, i.e. $\gamma = 1$ (skew-scattering) or $\gamma = 2$ (side jump). By rearranging the above equation, it is expected that the ratio of $\rho_{xy}^{NL}/\rho_{xx}^{\gamma}$ should be proportional to the magnetization $m$ in a real ferromagnetic semiconductor. Therefore, the direct comparisons of $\rho_{xy}^{NL}/\rho_{xx}^{\gamma}$ and the magnetization $m$ for all samples are shown in Fig. 2.11b-e. It is obvious that the $\rho_{xy}^{NL}/\rho_{xx}^{\gamma}$ does not fit the magnetization in Gd doped samples. Here we take the 0.2 ML Gd doped sample as an example (Fig. 2.11d). It can be seen that the
Figure 2.11: Comparison between the magnetotransport and magnetization measurements. (a) Ordinary Hall background corrected $\rho_{xy}^{NL}$ from the control samples and Gd doped samples. Top right inset: raw $\rho_{xy}$ (blue dot) with linear fitting of the high field data (pink dashed line) of the 0.4 ML Gd doped sample at 20 K. Comparison between $-\rho_{xy}^{NL}/\rho_{xx}^{\gamma}$ at 20 K and the magnetization (after diamagnetic background subtraction) at 5 K for the (b) Control Sample 1 (c) Control Sample 2 and (e) 0.4 ML Gd doped sample. Same comparison for (d) 0.2 ML Gd doped sample at 20 K. Here, $\gamma$ could be 1 or 2 depending on the origin of the scattering.
magnetization experiences a very fast saturation within 5 kOe while $\rho_{xy}^{NL}/\rho_{xx}^\gamma$ does not fully saturation until 50 kOe.

Since $\rho_{xy}^{NL}$ in all samples exhibits a Brillouin function like characteristic. Therefore, it might be supposed that the nonlinearity in the Hall resistance originates from the paramagnetic 2DEGs. However, this possibility is ruled out due to the distinct difference between the temperature dependence of the magnitude of the nonlinear Hall resistivity $\Delta \rho_{xy}^{NL}$ and paramagnetism as shown in Fig. 2.12. The paramagnetic response exhibits a $1/T$ dependence while $\Delta \rho_{xy}^{NL}$ shows a much slower change as a function of temperature. In Ref. [63], the authors reported a similar nonlinear Hall resistance in a regular AlGaAs/GaAs two dimensional hole gas and suggested that it is a genuine anomalous Hall effect that originates from the skew scattering of spin polarized charge carriers. The spin polarization between the spin up and spin down holes is induced by the Zeeman splitting due to the presence of the external magnetic field. However, this cannot explain our observation of the nonlinearity in the control samples since the Zeeman splitting (1.3 $K/T$ for a g factor of 2) is trivial compared to the thermal energy $k_B T$ at the magnetic field where the nonlinearity is observed. An alternative explanation of the shape of $\rho_{xy}$ is discussed in Sec. 2.6.

The absence of the ferromagnetic like response in the nonlinear component in $\rho_{xy}$ indicates that the electrons in the 2DEGs do not interact with the defect induced ferromagnetism despite the intentional physical overlapping between them. This implies that the electrons are locally depleted in the regions where the ferromagnetic spins present. Among the possible defects in our system, acceptor like point defects are not likely to fully deplete the electrons since the electron density in the 2DEG is as high as mid $10^{13}$ cm$^{-2}$. In Sec. 2.3, the threading dislocation density in these samples is
Figure 2.12: Comparison of the temperature dependent $\Delta \rho_{xy}^{NL}$ and Brillouin function with parameters of $S=3/2$, $g=4.5$ for the control samples and Gd doped samples.

in the order of $10^9$ cm$^{-2}$ and they are known as deep acceptors in GaN with large electron depletion width [64, 65]. We therefore hypothesized that the defect-induced ferromagnetism is present in large scale defect clusters, such as threading dislocations, that can locally repel the conduction electrons in the 2DEG.

The magnetoresistance $\rho_{xx}$ is also measured using the same Hall bar on all samples. The raw magnetoresistance is then converted to sheet magnetoresistance using $\rho_{xx} = \frac{V_{xx}}{I_{xx} \cdot w}$ (in the unit of $\Omega/\square$). The comparison of the sheet magnetoresistance among all samples is plotted in Fig. 2.13a. A negative magnetoresistance consisting of a fast decrease in the low magnetic field region and a linear decrease in the high magnetic field region is observed in all samples. This characteristic suggests that there are more than one magnetic field dependent scattering mechanisms dominating the lateral transport. The 0.4 ML Gd doped sample possesses the highest zero field sheet resistance of $\rho_{xx} = 1158 \ \Omega/\square$ due to its low electron mobility. This is expected since the heaviest Gd doping generates the highest defect density comparing to other samples. $\rho_{xx} = 412$ and 488 $\ \Omega/\square$ is found for Control Sample 1 and 2 respectively.
while the 0.2 ML Gd doped sample has a $\rho_{xx}$ of 427 $\Omega/\square$. $\rho_{xx}$ is slightly lower in the 0.2 ML Gd doped sample comparing to the Control Sample 2, which agrees with the higher carrier concentration measured in the 0.2 ML Gd doped sample. The small difference in $\rho_{xx}$ in two control samples is due to the slight difference in their mobilities as shown above.

The percentage change in the magnetoresistance in calculated based on $MR = \frac{\rho_{xx}(H) - \rho_{xx}(0)}{\rho_{xx}(0)}$ (in the unit of %) and is shown in Fig. 2.13b for all samples. The magnitudes of the MR are $-4.5\%$ and $-3\%$ for the Control Sample 1 and 2 and $-3.7\%$ and $-3.3\%$ for the 0.2 ML and 0.4 ML Gd doped samples. The shapes of the MR for all samples show a similar behavior comparing to the sheet magnetoresistance, which consists of a fast decrease in the low magnetic field region and a linear decrease in the high field region. The linear fits of the high field MR are shown as dotted lines in Fig. 2.13b. The shape of the MR implies the existence of different magnetic scattering processes in these samples. A detailed discussion of each specific scattering possess is proved in Appendix B.

2.6 Origin of the nonlinear Hall slope

The focus of this section is to discuss and explain the origin of the nonlinear component in the Hall resistivity observed in all samples. Here, we proposed the mixing of the magnetoconductivity tensor $\sigma_{xx}$ into the Hall resistivity $\rho_{xy}$ as an explanation for the observed nonlinear $\rho_{xy}^{NL}$. In two dimensional transport, this effect can be significant when there is a large enough change in the magnetoresistance. With the presence of an out-of-plane magnetic field along z-axis $B \approx \mu_0 H$, the Hall resistivity (transverse resistivity) $\rho_{xy}$ and magnetoresistance (longitudinal resistivity) $\rho_{xx}$ can
be expressed in terms of the off-diagonal and diagonal elements of the conductivity tensor, $\sigma_{xy}$ and $\sigma_{xx}$ as follows [66]

\begin{align}
\rho_{xx} &= \frac{\sigma_{xx}}{\sigma_{xx}^2 + \sigma_{xy}^2} \quad \text{(2.5a)} \\
\rho_{xy} &= \frac{\sigma_{xy}}{\sigma_{xx}^2 + \sigma_{xy}^2} \quad \text{(2.5b)}
\end{align}

where the conductivity tensors are functions of electron concentration $n$ and mobility $\mu$ as follows

Figure 2.13: Non-linear magnetoresistance. (a) Magnetoresistance $\rho_{xx}$ (plotted in $\Omega/\square$) and (b) MR (plotted in %) the control samples and Gd-doped samples at 20 K. The linear fit of data in the high magnetic field region is shown as dotted lines.
\[ \sigma_{xx} = \frac{-ne\mu}{1 + \mu^2 B^2} \]  \hspace{1cm} (2.6a)
\[ \sigma_{xy} = \frac{-ne\mu B^2}{1 + \mu^2 B^2} \]  \hspace{1cm} (2.6b)

By substituting Eqn. 2.6 into Eqn. 2.5, the measured physical quantities \( \rho_{xx} \) and \( \rho_{xy} \) can be simplified to \( \rho_{xx} = 1/ne\mu \) and \( \rho_{xy} = B/ne \) where \( \rho_{xx} \) is a constant with respect to the magnetic field while \( \rho_{xy} \) is proportional to the magnetic field. However, \( \rho_{xx} \) shows a negative magnetoresistance with multiple distinct regions rather than a constant as a function of magnetic field as shown in Fig. 2.13. This implies a magnetic field dependent correction term \( \delta \sigma_{xx} \) is needed to be taken into account into the conductivity tensor \( \sigma_{xx} \) where the corrected one should be \( \sigma_{xx} = ne\mu/(1 + \mu^2 B^2) + \delta \sigma_{xx} \). Once this correction term is included, \( \rho_{xy} \) will be affected by \( \sigma_{xx} \) and thus no longer proportional to the magnetic field.

The functional form of \( \delta \sigma_{xx} \) is formulated based on the weak localization and electron electron scattering theory in Appendix B. The corrected conductivity tensor \( \sigma_{xx} \) together with \( \sigma_{xy} \) is then calculated based on the fitting parameters for different scattering mechanisms and then transformed back to \( \rho_{xx} \) and \( \rho_{xy} \). These calculated \( \rho_{xx} \) and \( \rho_{xy} \) are compared to the measured values to determine the fitting parameters. Fig. 2.14 is an example of the best fit of the Control Sample 2. A good agreement between the measured and calculated \( \rho_{xx}, \rho_{xy} \) and \( \rho_{xy}^{NL} \) at 20 K can be observed. This validates the mixing of the \( \sigma_{xx} \) into \( \rho_{xy} \) as the origin of the nonlinearity in the Hall slope. The carrier concentration \( n \) and mobility \( \mu \) for all samples used in the calculation and fitting at 20 K are listed in Table 2.4. These values are very close to the ones that are calculated based on the Hall slope and zero field resistivity. No
Figure 2.14: Comparison of calculated and measured (a) $\rho_{xx}$, (b) $\rho_{xy}$ and (c) $\rho_{xy}^{NL}$ measured from the Control Sample 2 at 20 K.
nonlinearity in $\rho_{xy}$ is observed at 300 K as shown in Fig. 2.15a due to the perfect overlapping of the linear fit in the high positive and negative magnetic field regions. This is due to the negligible magnetoresistance at 300 K as shown in Fig. 2.15b. This similar temperature dependence between the magnetoresistance and $\rho_{xy}^{NL}$ further confirms the mixing of $\sigma_{xx}$ into $\rho_{xy}$ as the reason for the observed $\rho_{xy}^{NL}$. The carrier concentration $n$ and mobility $\mu$ for all samples at 300 K are calculated from the Hall slope and zero field resistivity and are listed in Table 2.5.

Figure 2.15: Comparison of measured $\rho_{xx}$, $\rho_{xy}$ at 20 K and 300 K in Gd doped samples. (a) Hall resistivity $\rho_{xx}$ (brown circles) measured from the 0.2 ML Gd doped sample with linear fit of the high positive (red dotted line) and negative (blue dotted line) magnetic field data at 300 K . (b) Comparison of the magnetoresistance from the 0.2 ML Gd doped sample at 20 K and 300 K.
Table 2.4: Carrier concentration and mobility at 20K

<table>
<thead>
<tr>
<th>Sample</th>
<th>( n_{2D}/(\text{cm}^2) )</th>
<th>( \mu/(\text{cm}^2/\text{Vs}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control Sample 1</td>
<td>( 3.3 \times 10^{13} )</td>
<td>564</td>
</tr>
<tr>
<td>( N_{Gd}^{2D} = 0.2 \text{ ML} )</td>
<td>( 2.9 \times 10^{13} )</td>
<td>513</td>
</tr>
<tr>
<td>Control Sample 2</td>
<td>( 4.4 \times 10^{13} )</td>
<td>407</td>
</tr>
<tr>
<td>( N_{Gd}^{2D} = 0.4 \text{ ML} )</td>
<td>( 3.1 \times 10^{13} )</td>
<td>195</td>
</tr>
</tbody>
</table>

Table 2.5: Carrier concentration and mobility at 300K

<table>
<thead>
<tr>
<th>Sample</th>
<th>( n_{2D}/(\text{cm}^2) )</th>
<th>( \mu/(\text{cm}^2/\text{Vs}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control Sample 1</td>
<td>( 2.4 \times 10^{13} )</td>
<td>554</td>
</tr>
<tr>
<td>( N_{Gd}^{2D} = 0.2 \text{ ML} )</td>
<td>( 2.3 \times 10^{13} )</td>
<td>445</td>
</tr>
<tr>
<td>Control Sample 2</td>
<td>( 3.1 \times 10^{13} )</td>
<td>335</td>
</tr>
<tr>
<td>( N_{Gd}^{2D} = 0.4 \text{ ML} )</td>
<td>( 2.7 \times 10^{13} )</td>
<td>198</td>
</tr>
</tbody>
</table>

2.7 Conclusions

The Gd δ-doped AlN/GaN 2DEGs grown on the 6H-SiC wafers exhibit a ferromagnetism with a Curie temperature above room temperature. The saturation magnetization is found to be larger than the \( 7\mu_B \) (Gd ion) indicating the defect-induced ferromagnetism nature in these Gd doped samples. This ferromagnetism exhibits an easy axis parallel to the crystalline c-axis rather than the basal plane. More importantly, these defect induced ferromagnetic spins are locked along the polar c-axis direction. Weak ferromagnetism and paramagnetism are observed in the control samples. This difference in magnetic behavior in the control samples is attributed to the difference in the spin polarized defect density in control samples.
Hall measurements on ferromagnetic Gd doped 2DEGs show a nonlinear Hall resistivity as well as the control samples without any Gd doping. Furthermore, this nonlinear Hall resistivity does not track the magnetization in Gd doped samples. These observations indicate the lack of coupling between the Gd induced ferromagnetic phase and conduction electrons in the 2DEGs although they spatially overlap with each other. We speculated that the ferromagnetic spins reside within threading dislocations or defect clusters at the interface. This defect clusters are acceptor type which locally deplete the electron and induce strong charge scattering. In this case, electrons in 2DEG are repelled from these ferromagnetic defect clusters which hinders the effective coupling between them. The nonlinearity in the Hall resistivity is consistent with the mixing of the conductivity tensor $\sigma_{xx}$ into the Hall resistivity $\rho_{xy}$. 
Chapter 3: Growth and Magnetic Properties of Mn Doped 2D MoS$_2$ on Sapphire Substrates

The content in this chapter is done in collaboration with Lee Edwin, Lu Ma, Yijing Wu, and Siddharth Rajan and is served as a supplemental material for the ongoing project on the Mn doped 2D MoSe$_2$.

The 2D material family attracts extensive studies for their potential of being used in low cost and flexible electronic and optoelectronic applications. The prototypical 2D material graphene [67, 68] is gapless and thus limits its usage in logical circuits [69]. On the other hand, 2D transition-metal dichalcogenides (TMD), such as MoS$_2$, are ideal for integration into current transistor structures for novel applications due to their semiconducting properties and atomically-thin nature. In terms of their magnetic properties, previous experimental work mainly focused on the magnetism in MoS$_2$ nano-sheets [70] and single crystals [71] while much less work is done on layered TMDs with magnetic transition-metal doping. Recently, a theoretical study based on the density functional theory predicts that the Mn-doped monolayer MoS$_2$ is a room temperature ferromagnetic semiconductor which makes it a promising candidate for realizing a room-temperature 2D dilute magnetic semiconductor for practical spintronic applications [72].
In this chapter, we tested this prediction by growing and characterizing the few-layer Mn:MoS$_2$. In Sec. 3.1, the synthesis method of few-layer Mn:MoS$_2$ samples are by sulfurizing a metallic layer of Mn doped Mo thin film sputtered on a (0001) sapphire substrate is discussed. The structural properties of these few-layer Mn:MoS$_2$ samples are characterized by a variety of methods: atomic force microscopy (AFM), high resolution X-ray diffraction (XRD) and Raman spectroscopy in Sec. 3.2. The magnetic properties are characterized using the superconducting quantum interference device (SQUID) magnetometry and the corresponding results are discussed in Sec. 3.3.

3.1 CVD growth of Mn doped MoS$_2$ on sapphire substrates

The growth of Mn doped MoS$_2$ is done via a two-step process (i) deposition of Mn doped Mo thin films on (0001) sapphire substrates (ii) sulfurization of Mn doped Mo thin films using MoS$_2$ precursor at a high temperature. The metal deposition in the step (i) is done using the AJA Orion RF/DC Sputter Deposition Tool. The base pressure is $< 3 \times 10^{-7}$ torr. A RF power of 100 W and Ar pressure of 3 mTorr are used to achieve a growth rate of 1 nm / 25 sec. 5 nm Mn doped Mo is deposited on the sapphire substrate using this growth rate. A control sample consisting of 5 nm Mo without any Mn dopants is also prepared using the similar sputtering method. In step (ii), the samples are placed in a sealed quartz tube along with the MoS$_2$ precursor. Sulfurization of the sputtered Mn:Mo (or Mo) thin films occurs at 1100 °C at which the precursor starts to vaporize and react with the metal thin films. The duration of the growth is 4.5 hours. This growth method is discussed in more details in Ref. [73].
After the sulfurization of the Mn:Mo thin films, energy-dispersive X-ray spectroscopy (EDX) in Sirion SEM is performed to characterize the chemical composition in these films. 2D EDX maps of Mo, Mn and S elements are shown in Fig. 3.1a, b and c. All elements are uniformly distributed in the film indicating the absence of large chucks agglomerations or clusters of any given elements. However, we can not rule out the presence of small Mn clusters until an EDX scan with a finer resolution is done. The detailed line scans of the atomic concentration of these elements are shown in Fig. 3.1d. The target concentration between Mn and Mo is $\sim 12.5 : 87.5$ which corresponds to a Mn:Mo:S ratio of $\sim 4 : 30 : 67$. This is very close to the measured atomic
concentration shown in Fig. 3.1d. This implies the targeted stoichiometry of Mn:Mo and Mo:S are well maintained during the sputtering and sulfurization processes.

3.2 Structure properties of Mn doped MoS$_2$ on sapphire substrates

![XRD and Raman scans](image)

Figure 3.2: Structural properties of the few-layer MoS$_2$ and Mn:MoS$_2$ samples. (a) XRD scans of the MoS$_2$ and Mn:MoS$_2$ samples reveal the single crystal nature of these few-layer samples. (b) Raman spectroscopy confirms the presence of similar vibration modes as in the bulk MoS$_2$ samples.

High resolution X-ray diffraction scans on sulfurized Mo and Mn:Mo films are shown in Fig. 3.2a. Single crystal MoS$_2$ peaks along the (0001) direction can be observed in both samples. The (0002) MoS$_2$ peak in the Mn:MoS$_2$ sample is more broadened and distorted possibly due to the more deformed lattice structure due to
the higher density of small crystallites. Multiple thickness fringes can be observed in both samples indicating the good interface and surface quality. The calculated thickness for the MoS$_2$ and Mn:MoS$_2$ samples are $\sim$ 9 and $\sim$ 6 nm suggesting their few-layer nature. In Fig. 3.2b, Raman spectroscopy reveals the $E_{2g}^{1}$ (in-plane vibration mode) and $A_{1g}$ (out-of-plane vibration mode) [74] peaks at 382 cm$^{-1}$ and 408 cm$^{-1}$ with a peak to peak ratio of 1.5. These characteristics are very similar to the ones measured on the bulk MoS$_2$ [75, 76, 77, 78]. The higher intensity in MoS$_2$ samples could be a result of the better crystalline quality or the larger thickness in the undoped samples. The full-width-half-maximums of these two peaks are comparable possibly due to the relatively low resolution of the Raman scan, and therefore, cannot be directly related to the crystalline quality of these few-layer samples.

The AFM images of the few-layer MoS$_2$ and Mn: MoS$_2$ are shown in Fig. 3.3a and b. Uniform thin sheets of MoS$_2$ and Mn:MoS$_2$ are found to cover the entire sapphire substrate. Small hexagon crystallites are observed on the surface arising from the basal plane symmetry of MoS$_2$. Line cuts across the green and blue line in Fig. 3.3a and b are shown in Fig. 3.3c. The step height is 0.6 nm corresponding to one mono-layer of MoS$_2$. This reveals the nature of the layered structure of these few-layer MoS$_2$ and Mn:MoS$_2$ samples.

**3.3 Magnetic properties of Mn doped MoS$_2$ on sapphire substrates**

To test the existence/absence of the long-range ferromagnetic coupling between Mn dopants in the few-layer Mn:MoS$_2$, the magnetization of the Mn:MoS$_2$ together with the undoped MoS$_2$ samples are performed using the SQUID magnetometry with the magnetic field applied along the in-plane direction. As shown in Fig. 3.4a, the
magnetization of Mn:MoS$_2$ samples shows a ferromagnetic behavior with a sharp switching of the saturation magnetization within 6 kOe at 300 K. The undoped MoS$_2$ samples also exhibit ferromagnetism but with a much smaller saturation magnetization compared to the Mn:MoS$_2$ samples. The origin of this marginal ferromagnetism might be the spin polarized defects as discussed in Sec. 2.4 [34, 35]. However, we did observe a boost in the ferromagnetic response upon Mn doping in the few-layer Mn:MoS$_2$.

One might argue that the Mn dopants in these samples experience long-range spontaneous coupling based on the room temperature ferromagnetism in Mn:MoS$_2$. However, the majority of Mn dopants are not ferromagnetically coupled. This is clearly shown in Fig. 3.4b where the magnetization of the same Mn:MoS$_2$ sample
Figure 3.4: Magnetic properties of Mn doped few-layer MoS$_2$. (a) Diamagnetic background corrected magnetization of updoped MoS$_2$ and Mn doped MoS$_2$ samples at 300 K. (b) Diamagnetic background corrected magnetization of uMn doped MoS$_2$ samples at 5 K with Brillouin function fit.

is measured at 5 K. At low temperature, paramagnetism is observed to dominate the magnetic response. This paramagnetic behavior can be fitted with the Brillouin function $M = N_g \mu_B J \left[ \frac{2J+1}{2J} \coth \left( \frac{2J+1}{2J} \frac{g \mu_B J B}{k_B T} \right) - \frac{1}{2J} \coth \left( \frac{1}{2J} \frac{g \mu_B J B}{k_B T} \right) \right]$ with the amount of Mn dopants in the sample ($N = 4.1 \times 10^{14} \text{ cm}^{-2}$). Here, $g = 2$ is the g-factor, $\mu_B$ is the Bohr magneton and $J$ is the total angular momentum quantum number. If the majority of the Mn dopants exhibits ferromagnetic coupling, a much larger ferromagnetic response (goes with $1 - T^\frac{3}{2}$) rather than a paramagnetic one should
be observed at 5 K. Therefore, we concluded that the majority of Mn spins in these few-layer Mn:MoS$_2$ samples either (i) just freely resides on the Mo site (not coupled to other Mn spins) or (ii) do not incorporate into the correct lattice site. There might be a very small amount of ferromagnetically coupled Mn spins present in Mn:MoS$_2$ contributing to the slightly larger ferromagnetism at room temperature compared to the undoped sample. The reason for the lack of ferromagnetic coupling could be that the Mn dopants are not incorporated the Mo site (a requirement in for ferromagnetism in Ref. [72]). The Mn dopants could reside in the interstitial sites in the lattice or form small clusters. A more detailed structural measurement to study the how the Mn atoms are incorporated in the MoS$_2$ lattice is needed.

3.4 Conclusions

Single crystal few-layer MoS$_2$ and Mn:MoS$_2$ samples are successfully synthesized by sulfurizing the sputtered Mo and Mn:Mo metallic thin films on sapphire substrates. Single crystals MoS$_2$ peaks along the (001) direction are observed in both samples. The thickness estimated based on the thickness fringes indicates the few-layer nature of these samples. Both XRD and Raman characterizations imply the undoped MoS$_2$ samples exhibit a better crystalline quality. According to Ref. [72], doping with a Mn concentration of 12.5% (ratio to Mo) in 2D MoS$_2$ would result in a collective ferromagnetic coupling between the Mn dopants. However, few-layer Mn:MoS$_2$ samples with the correct Mn doping concentration contain mostly paramagnetic spins from the Mn dopants. Only a very small amount of Mn dopants experience ferromagnetic coupling and contribute to the ferromagnetism evidenced by the slightly larger ferromagnetic response than the undoped samples at 300 K. Currently, we are working
on the magnetically (Mn) doped 2D material MoSe$_2$ grown by the molecular beam epitaxy on Si wafers. The magnetic properties of such 2D films as a function of Mn doping concentration and film thickness will be investigated.
Chapter 4: Nonlocal Opto-thermal Spin Seebeck Effect and Magnon Spin Diffusion Length in YIG

The spin Seebeck effect (SSE) consists of the generation of a spin current in a ferromagnetic material (for example yttrium iron garnet, $Y_3Fe_5O_{12}$ or YIG) due to an external temperature gradient and the subsequent detection of such spin current using a spin transducer (usually heavy metals with large spin orbital coupling such as Pt) via the inverse spin Hall effect [15, 27, 4]. This thermally induced spin current is carried by magnons that typically possess much higher energies (500-6000 GHz) than the coherent microwave frequency magnons (1-10 GHz) [79]. Although these thermally induced magnons usually couple more strongly to phonons and thus leading to a shorter lifetime and a smaller diffusion length than the coherent microwave magnons, the investigation of the transport properties of these thermally induced magnons are of great importance for discovery of new physics in the field of spin caloritronics. For example, Flebus et al. recently proposed the existence of the Bose-Einstein condensation of superfluid magnons in YIG induced by the constant thermal pumping [30]. However, the realization of such new experiments is hindered due to the lack of information on the length scale over which these thermally generated magnons could exist.
In this chapter, we developed a nonlocal opto-thermal spin Seebeck measurement on Pt/YIG that allows us to measure the magnon spin diffusion length in YIG at 23 K. In this nonlocal configuration, multiple Pt spin injectors along with a spin detector are fabricated on YIG. A scanning laser is applied on the spin injector to excite the nonequilibrium thermal magnons. The decay of the diffused magnon spins are measured by a lock-in amplifier and compared to the results from the thermal and spin modeling to determine the spin diffusion length. Since all measurements are performed on a single Pt/YIG sample, the YIG crystal and Pt/YIG interface qualities are identical between all measurements. Later, Cornelissen et al. employed a similar detector/injector geometry to measure the spin diffusion length for the thermally driven magnons at different temperatures [80]. The magnon spins are injected via the Joule heating from the AC current in the Pt injector while the diffused spins are detected on a separate Pt detector using a lock-in amplifier. The samples used in this measurement typically contain only one or a few detector/injector devices. Therefore, a large number of samples with different detector/injector spacing is needed to map out the decay of the diffused spins. In this case, the consistency between different samples and measurements is hard to control due to the variations in the YIG crystal and Pt/YIG interface qualities.

Prior to the detailed discussions of the nonlocal opto-thermal SSE measurement, the growth of single crystal YIG thin films on gadolinium gallium garnet (Gd$_3$Ga$_5$O$_{12}$ or GGG) substrates is first discussed in Sec. 4.1. The local opto-thermal spin Seebeck measurement is performed and discussed on the Pt/YIG (single crystal thin film)/GGG sample and the Pt/YIG (bulk single crystal) sample in Sec. 4.2. It is found that the opto-thermal SSE signal is greater in the later one, and therefore, this
sample is used in the actual magnon spin diffusion length measurements. In Sec. 4.3 and 4.4, we modified the local opto-thermal SSE measurement into a nonlocal configuration which allows us to probe the decay of the diffused magnonic spin current using only one Pt/YIG sample. Finite element method simulations on heat and spin transports in this non-local configuration are discussed in Sec. 4.5 and 4.6 to verify the absence of the possible contamination signals induced by the stray heat beneath the spin detector and to estimate an upper bound of the spin diffusion length.

4.1 Growth of YIG single crystal thin films

The YIG thin film samples discussed in this chapter are grown by the radio-frequency (RF) magnetron sputtering on a Kurt Lesker LAB 18 tool. The base pressure of the LAB 18 chamber is around $1 \times 10^{-7}$ torr. The 3-inch YIG sputtering target is commercially obtained from the Kurt Lesker Company. The YIG thin films are grown on double side polished (111) GGG substrates purchased from the Saint-Gobain Company.

To obtain YIG single crystal thin films, two methods (i) in-situ heating during sputtering [81] (ii) post-annealing after sputtering [82] are attempted. The growth of single crystal YIG thin film using the method (i) is first discussed here. Prior to the growth, the GGG substrate is cleaned using solvent and baked for dehydration in the sputtering chamber. The RF power and Ar pressure used for the growth are 80 W and 3 mTorr. The growth rate of YIG is extremely slow ($\sim 8$ nm/hour). This low growth rate is limited by the maximum available RF power that the YIG target can tolerate. Using a higher RF power than 80 W would potentially damage the sputter target through (1) melting the indium bond between the YIG target and Cu
back protecting plate (2) creating a large temperature gradient between the target front and back surface which increases the risk of cracking the target. The in-situ substrate heater is set to 550 °C with a ramp rate of 10 °C/min. An example of the XRD scan on the film grown using the method (i) is shown in Fig. 4.1 along with the GGG substrate reference. The nominal thickness of this YIG thin film is 37 nm. Two shoulders on each side of the GGG substrate peak can be observed indicating the formation of bcc-YIG along the (444) crystal direction. However the absence of any thickness fringes indicates the low crystallinity and surface quality of this thin film. This preliminary result shows the possibility of achieving a better YIG single crystal thin films using an optimized in-situ heating temperature [81]. However, the further optimization process is hindered due to the poor design of the LAB 18 tool. In LAB 18 tool, the substrate heater and substrate holder are in close vicinity. During normal operations, the deposited materials penetrate through the substrate holder through the pilot holes that are left open and attach themselves onto the substrate heater. These unwanted materials on the heating filament reemit onto the pristine sample introducing contamination materials when the substrate heater is set to a high temperature. The YIG thin film samples with the in-situ heating method discussed here were grown right after the LAB 18 is installed. YIG thin films grown using this method afterwards show no single crystal features.

Due to the technical difficulty of using the in-situ heating method discussed above, the method (ii) that uses post-annealing after sputtering was adopted to achieve a better crystallinity of the YIG thin film. The sample preparation and growth conditions are similar to the ones used in method (i). The only difference is that the in-situ substrate heater is disabled during the thin film sputtering. The post-annealing
Figure 4.1: XRD scan on the sputtered YIG thin films with in-site heating at 550 °C. XRD on the GGG substrate is served as a reference. Single crustal YIG peak along (111) direction can be identified on the shoulders of GGG substrate peak.

is done in a furnace. The comparison of the XRD data measured on the as-deposited sample and samples with different posit-annealing temperatures is shown in Fig. 4.2. The as-deposited YIG thin films only show (444) GGG substrate peak implying their amorphous nature. The YIG thin films undergone a post-annealing 800 °C and 900 °C for 2 hours both show a (444) YIG single crystal peak. Multiple thickness fringes can be observed in these YIG thin films indicating the a good YIG/GGG interface quality. The thickness calculated based on the thickness fringes yields a value of ~ 40 nm.

The AFM images of the as-grown YIG thin film and thin films with 800 °C and 900 °C post-annealing are shown in Fig. 4.3a, b and c. The as-grown YIG thin film exhibits a very smooth surface morphology similar to the GGG substrate. The post annealing process roughens the surface and the surface roughness increases at a higher post-annealing temperature. Small hillock-like features can be observed in the
Figure 4.2: XRD scan on the as-deposited sputtered YIG thin films (w/o in-situ heating) and YIG thin films that undergo different post-annealing conditions. Single crystal YIG peaks along (111) direction are observed with multiple thickness fringes indicating good crystal and interface quality.

Sample with 800 °C post-annealing while much larger YIG crystallites start to form at a post-annealing temperature of 900 °C. Based on these points, the post-annealing temperature of 800 °C is chosen since it yields a relative smooth surface morphology.

Figure 4.3: AFM images on the (a) as-deposited (b) 800 °C post-annealed and (c) 900 °C post-annealed YIG thin films.
Ar ion milling (beam voltage 400 V and emission current 21 mA) can be used to reduce the surface roughness in these annealed samples. The surface morphologies of the YIG thin films before and after the ion milling are shown in Fig. 4.4a, b and c for 0, 30 sec and 1 min 30 sec ion milling duration. The surface roughness gradually decreases with increasing ion milling duration. However, the improvement of the surface roughness does not lead to a greater SSE signal as shown in Fig. 4.4d. This is possibly a result of the increased damping of magnons due to the YIG lattice damage from the high energy Ar particles. Here, the SSE signals are induced by a laser chopped at 2 kHz and are measured with a lock-in amplifier. The measurement method will be discussed in more detail in the following sections.

The magnetization of the annealed YIG thin film is measured by SQUID at 300 K and is shown in Fig. 4.5. The paramagnetic background from the GGG substrate is subtracted to reveal the magnetic response from the YIG thin film. After the subtraction, a ferromagnetic response from the YIG thin film is observed with a coercive field of 50 Oe and a saturation magnetization of 70 emu/cm$^3$. The open hysteresis loop in the low magnetic field region is clearly shown in the top left inset. These magnetic characteristics of the YIG thin film are similar to the ones in the previous reports [83, 84].

4.2 Opto-thermal spin Seebeck measurement on the Pt/YIG(thin film)/GGG and Pt/YIG(bulk) samples

The Opto-thermal spin Seebeck measurement was first demonstrated in Ref. [85]. In this section, we employed a similar measurement configuration to measure the opto-thermal SSE signal. To be more specific, the thermal gradient in the sample is generated using a Chameleon Ultra II Ti:sapphire laser. This laser outputs a 140
fs pulse with a frequency of 80 MHz. This pulse train is further modulated using a mechanical chopper at around 2 kHz and then focused on the sample using a 100 mm achromatic lens. The wavelength of the laser is 715 nm. The opto-thermal spin Seebeck signals are picked up by a Zurich Instrument lock-in amplifier in the current mode. In this mode, one of the two wires in Pt is grounded while the other one is connected to a current preamp and then to a lock-in amplifier using a SMA cable.

The opto-thermal spin Seebeck measurement using the measurement setup discussed above is carried out on the Pt/YIG (thin film)/GGG and Pt/YIG (bulk)
Figure 4.5: Magnetization of the single crystal YIG thin film measured at 300 K shows the ferromagnetic nature of the YIG thin films. The open hysteresis loop in the low magnetic field region is shown in the inset.

Figure 4.6: Comparison of the optothermal spin Seebeck signal between the Pt/YIG(thin film)/GGG and Pt/YIG(bulk) samples. Due to the lower crystal and interface quality, the Pt/YIG(thin film)/GGG sample yields a lower spin Seebeck signal.
samples. The (111) YIG thin film is grown using the method discussed in the previous section while the (100) YIG bulk single crystal is commercially obtained from the Princeton Scientific Corp. A 10 nm Pt thin film is deposited on both YIG samples via e-beam evaporation. The comparison of the opto-thermal spin Seebeck signal on these samples is shown in Fig. 4.6. The average laser power used in this experiment is 64 mW. It can be seen that the opto-thermal spin Seebeck signal is much larger in the Pt/YIG (bulk) sample. This observation is different than the results in Ref. [29] where the YIG thin film is 4 μm thick and is grown using a different method. It has been suggested by Ref. [86, 87] that the spin transfer across the Pt/YIG interface and the resulting SSE signal strongly depends on the interface quality. In Ref. [81], it is also reported that the quality of the YIG thin film strongly affects the efficiency of the spin pumping from YIG to Pt. Therefore the smaller signal measured in the YIG thin film sample is attributed to the lower Pt/YIG interface and YIG film quality.

4.3 Design of the nonlocal opto-thermal spin Seebeck effect experiment

The content from Sec. 4.3 to Sec. 4.7 is adapted from the previously published work in Ref. [88] by the authors Brandon L. Giles, Zihao Yang, John S. Jamison, and Roberto C. Myers. In this work, my responsibilities are sample design and fabrication and thermal and spin transport modeling.

As discussed in the previous sections, since we observed a much larger SSE signal in the Pt/YIG (bulk) sample comparing to the Pt/YIG (thin film) sample. Therefore, all measurements carried out in the following sections are performed on the bulk YIG single crystal samples. The conventional and the opto-thermal spin Seebeck effect is discussed in Sec. 1.6 and 4.2. In these measurements, the spin injection and detection
are achieved in a single Pt thin film. In this section, we patterned the Pt film into a Pt spin detector and an array of Pt spin injectors separating the spin injection and detection processes. These remote Pt injectors are both spatially and electrically isolated from the Pt spin detector. The schematic diagram of the sample design is shown in Fig. 4.7a. The same Ti:sapphire laser (as used in the previous section) is focused on a Pt spin injection pad producing a local temperature gradient $\nabla T$ in the YIG beneath. This temperature gradient $\nabla T$ excites non-equilibrium magnons that can diffuse in the YIG. The lateral diffusion of these non-equilibrium magnon spins contribute to a spin current $J^s_z$ and can be injected into the Pt spin detector via spin pumping. The injected spin current into Pt is then converted into a transverse voltage along the $y$ direction ($V_y$) via the inverse spin Hall effect. This allows us to probe the magnon spin diffusion of the thermally excited magnons in YIG. Here, we denoted the measurement configuration in which spins are injected and detected only using the Pt detector as the ”local opto-thermal SSE measurement” and denoted the one in which the spins are injected and detected using the spatially separated Pt spin injector and detector as the ”nonlocal opto-thermal SSE measurement”.

The microscope image of the actual device used for the non-local opto-thermal SSE measurement is shown in Fig. 4.7b. The single side polished (100) bulk YIG single crystal samples are purchased from the Princeton Scientific Corp with dimensions of $5 \times 5 \times 0.4$ mm. The Pt thin film is deposited using e-beam evaporation in a Kurt Lesker LAB 18 tool and the patterning of the Pt thin film is achieved through photolithography and Cl$_2$/CF$_4$ based dry etch in a Plasma Therm SLR770 tool. The Pt spin detection pad is $265 \times 265$ $\mu$m while the Pt spin injection pads are $10 \times 10$
Figure 4.7: Schematic diagram of the nonlocal configuration of the optothermal spin Seebeck measurement. The spin current injection and detection geometry are achieved on spatially and electrically isolated Pt pads. (b) Microscope image of the sample used in the measurement.

µm with a 3 µm spacing. Both the Pt spin detector and injector have a thickness of 10 nm. The detailed process recipe can be found in Appendix C.

4.4 Measurements of the magnon spin diffusion length in YIG

The schematic diagrams of the local and nonlocal opto-thermal SSE measurements discussed in the last section are shown in Fig. 4.8a and c respectively. In both configurations, the voltage measurements are taken on the Pt spin detection pad using a lock-in amplifier in the voltage mode. The laser is placed either on the spin detector or on the remote spin injector to facilitate the local or the nonlocal configuration measurement. The measured raw transverse voltage $V_y$ hysteresis loops are shown in Fig 4.8b and d for these two configurations at 23 K. The input laser
power is 29.9 mW at 23 K. Here, Δx is the distance between the edge of the spin detector and the center of the spin injector (i.e. Δx = 0 means the local configuration). In the local configuration, the magnetic field dependent $V_y$ tracks the magnetization of YIG and saturates to a value of $746 \pm 3 \text{nV}$. In the nonlocal configuration ($\Delta x = 54 \text{ µm}$), $V_y$ also tracks the magnetization but saturates to a much smaller value comparing to the local measurement. We believe this nonlocal signal is generated by the diffusion of the thermally induced magnon spins from the spin injector. To confirm this statement, finite element method simulations based on the heat transport equations are performed and discussed in Sec. 4.5 and 4.6. The half of the total magnitude of the measured voltage $V_y$ is defined as $V_L$ and $V_{NL}$ for the local and nonlocal measurement configurations.

Figure 4.8: Schematic diagram of the (a) local and (c) nonlocal configuration of the optothermal spin Seebeck measurement. The measured optothermal spin Seebeck signals for these two configurations are shown in (b) and (d) respectively. The magnetic field decadent $V_y$ track the magnetization of YIG in both cases while the magnitude of $V_y$ is much smaller in the non-local configuration.
To map out the spatial dependence of $V_{NL}$, the laser is moved onto multiple spin injection pads with different $\Delta x$. The spatial dependencies of the measured $V_{NL}$ with different laser powers ranging from 10 to 30 mW at 23 K are shown in Fig. 4.9. It can be seen that $V_{NL}$ decays as $\Delta x$ increases (equivalent to moving the laser away from the detector). These decay profiles are fitted with a single exponential function $V_{NL} = A \cdot e^{-\Delta x/\lambda_s}$ where $A$ is the pre-factor and $\lambda_s$ is the effective magnon spin diffusion length. The extracted $\lambda_s$ from the measured data shown in Fig. 4.9 is $\sim 47 \, \mu m$ and is found to be independent of laser power. This indicates that the magnon spin scattering mechanism is not sensitive to the applied temperature gradient. This spin diffusion length is further discussed and compared to the modeling results in Sec. 4.6.

![Figure 4.9: The decay of measured $V_{NL}$ as a function of injector to detector distance $\Delta x$. The decay is fitted with an exponential function $A \cdot e^{-\Delta x/\lambda_s}$. The characteristic decay length $\lambda_s$ is found to be around 47 $\mu m$ and is independent of input laser power.](image)
4.5 Simulations of the heat transport in the nonlocal optothermal SSE measurement

The thermal modeling based on diffusive heat transport equations is first performed using the finite element method to rule out the possible contamination signals induced by the stray heat beneath the detector. This is done by using the three-dimensional finite element method solver COMSOL with the same sample structure and laser power as used in the actual measurements.

![Figure 4.10: Temporal response of the temperature rise in YIG along the optical axis after the laser is turned on. The laser power profile is shown as red dotted line. The corresponding temperature rise at Pt/YIG interface and 10 µm below the interface are shown as dark blue circles and light blue rectangles. The fast saturation of the temperature rise in YIG to the steady state value implies that the experiment can be approximately treated as in quasi-steady-state.](image)

Since the laser used in the experiment is chopped at 2 kHz, the time domain temperature profiles in the sample are first examined to determine the time scale for the sample to reach thermal steady-state after the laser is turned on. The 715 nm laser is modeled as a Gaussian beam with a radius or 3.75 µm. The temporal
profile of the input laser power is modeled as a square wave modulated at 2 kHz. The input parameters used in the model are listed in Table 4.1. In the local configuration, the laser is absorbed in the center of the detector and is turned on between 0.614 and 0.886 ms as shown in Fig. 4.10. The resulting temperature rise at the Pt/YIG interface and in YIG 10 µm beneath the interface are shown as dark blue circles and light blue rectangles. These time domain temperature profiles show a rapid saturation with a 10% - 90% rise time of ∼ 12.6 and 23.5 µs, respectively. This indicates that the sample rapidly reaches thermal steady-state after the laser is turned on at 23 K. Therefore all simulations discussed below are solved under the steady-state condition.

Table 4.1: Material parameters used to solve temperature profiles in the local and nonlocal opto-thermal SSE measurements.

<table>
<thead>
<tr>
<th>Material</th>
<th>T (K)</th>
<th>n</th>
<th>α (1/m)</th>
<th>ρ (kg/m³)</th>
<th>κ (W/mK)</th>
<th>C (J/°K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pt</td>
<td>23</td>
<td>1.89+4.54i</td>
<td>78.3 × 10⁶ [89]</td>
<td>21450 [90]</td>
<td>350 [91]</td>
<td>11.7 [92]</td>
</tr>
<tr>
<td>YIG</td>
<td>23</td>
<td>2.07 [90]</td>
<td>-</td>
<td>5245 [90]</td>
<td>109.5 [93]</td>
<td>5.6 [93]</td>
</tr>
</tbody>
</table>

Since the presence of any finite temperature gradients at the detector will result in a conventional SSE signal, the temperature profiles at the spin detector induced by the remote laser heating need to be examined to ensure the absence of this type of the possible contamination signals. The vertical (∇T_z) and lateral (∇T_x) temperature gradients with a 29.9 mW laser being focused on the Pt spin injector at an ambient temperature of 23 K are shown are Fig. 4.11. Although the majority of the temperature gradients are localized beneath the Pt injector, there could be a finite ∇T_z and ∇T_x beneath the Pt detector when the distance between the spin injector and detector is close enough. FEM modeling shows that both ∇T_z and ∇T_x
beneath the detector decay to below 1% of their peak values when probing 23 µm away from the injector. This implies that any $V_{NL}$ measured beyond $\Delta x > 23 \mu m$ should be attributed to the pure diffusion of the thermally induced magnon spins. In addition, there is no evidence of the stray heat induced contamination in the measured signal within 23 µm. This is derived from the analysis of the fitting qualities of $V_{NL}$ in regions with different $\Delta x$. The details of this analysis are discussed in the supplementary material in Ref. [88].

Figure 4.11: 2D contour map of the simulated $\nabla T_z$ and $\nabla T_x$ in YIG ($z=0$ represents the Pt/YIG interface) with the laser focused on the Pt injection pad at $\Delta x = 60 \mu m$. The edges of the injector are labeled by the black dashed lines. The majority of $\nabla T_z$ and $\nabla T_x$ are localized beneath the injector and are isolated from the edge of the spin detector ($\Delta x = 0$). Here, the color scale is in logarithmic.

The exclusion of the possible contamination signals induced by the stray heat ($\nabla T_z$ or $\nabla T_x$) is more clearly shown in Fig. 4.12 where the spatial dependence of the measured signal $V_{NL}$ and the maximum temperature gradient at the Pt/YIG interface beneath the detector are directly compared. Both $\nabla T_z$ and $\nabla T_x$ decay much faster than the measured $V_{NL}$ indicating that neither $\nabla T_z$ nor $\nabla T_x$ is the dominant driving
force for the measured $V_{NL}$. Therefore the effective magnon spin decay length of 47 µm truly reflects the diffusion characteristic of the thermally induced magnonic spins. This length scale is much longer than the magnon thermal mean-free path of 4 µm at 20 K measured in the bulk YIG in Ref. [93]. This magnon thermal mean-free path refers to the length scale over which a thermally driven magnon can propagate without experiencing any inelastic scattering event. Two possible reasons for this discrepancy are offered here. First, the thermally induced magnons can preserve their spin even after multiple inelastic scattering events. This scenario is similar to electrons whose charge scattering rate is typically much faster than the spin-flip rate. Therefore, like electrons, magnonic spins could remain unaffected even after several linear momentum scattering events. Second, the measured $V_{NL}$ is generated by a subset of thermally driven magnons called subthermal magnons that possess a lower energy and experience a weaker magnon phonon coupling comparing to higher energy thermal magnons. In this case, the higher energy thermal magnons that carry the majority of the heat experience significant inelastic scattering to phonons thus dominating the measured magnon thermal mean-free path. On the contrary, the low energy subthermal magnons that contribute to the measured $V_{NL}$ have a much longer thermal mean-free path. This long magnon thermal mean-free path just for the subthermal magnons could be close to the measured magnon spin diffusion length from the nonlocal opto-thermal SSE measurement.

In addition, multiple heat loss mechanisms such as the convective cooling loss, radiation loss and heat loss due to metal wires need to be evaluated and determined whether they should be included in the thermal modeling. The convection loss is determined by $Q_{\text{conv}} = hA(T_{\text{surface}} - T_{\text{ambient}})$ [94] where $h$ is the heat
Figure 4.12: Comparison of the spatial dependence of $V_{NL}$, $\nabla T_z$ and $\nabla T_x$ as a function of $\Delta x$. The different characteristic decay profiles suggest the possible contamination signals due to the spurious temperature gradients can be ruled out in the non-local configuration.

The heat transfer coefficient $h$ is zero since the sample is mounted in a cryostat with a pressure below $10^{-5}$ Torr. Therefore the convective heat loss is zero. The radiation loss is determined by

$$Q_{\text{rad}}^{\text{loss}} = \epsilon\sigma A(T_{\text{surface}}^4 - T_{\text{ambient}}^4)$$

where $\epsilon$ is the emissivity and $\sigma$ is the Stefan-Boltzmann constant. Using $\epsilon_{Pt} = 7.3 \times 10^{-4}$ [95], $A = 10 \times 10 \ \mu m^2$, $\sigma = 5.67 \times 10^{-8} \ W/m^2K^4$ and $T_{\text{surface}} = 30 \ K$ (this temperature is an overestimation and is higher than the actual surface temperature rise), we calculated $Q_{\text{rad}}^{\text{loss}} \sim 10^{-12} \ mW$ which is negligible comparing to the input laser power of 30 mW. The heat loss due to the heat conduction through the Au voltage wires is determined by

$$Q_{\text{wire}}^{\text{loss}} = \kappa A \nabla T$$

where $\kappa$ is the thermal conductivity of the wire. Using $\kappa_{Au} = 11.4 \ W/cmK$ [91], $A = \pi(25.4\mu m/2)^2 = 506.7 \ \mu m^2$ and $\nabla T = (25.015-23 \ K)/(1 \ cm) = 0.015 \ K/cm$, we calculated $Q_{\text{wire}}^{\text{loss}} = 1.7 \times 10^{-3} \ mW$ which is also insignificant comparing to the
input laser power of 30 mW. Based on the discussions above, the heat loss due to convection, radiation and conduction through the metal wires are not considered in the thermal modeling.

4.6 Simulations of the spin transport in the nonlocal optothermal SSE measurement

The finite element method modeling based on the spin diffusion theory is performed in this section for two reasons. First, there are multiple unused Pt pads in between the injector and detector as shown in Fig. 4.7. Since Pt represents a strong spin sink, the extracted $\lambda_s$ discussed in Sec. 4.4 based on the exponential fit serves as a lower bound for the spin diffusion length. The upper bound for $\lambda_s$ can be estimated through spin diffusion modeling that accounts for the loss of diffused magnons via spin sinking through the unused Pt pads. Second, the use of an exponential fit to extract the effective spin diffusion length in Sec. 4.4 still remains to be validated.

To answer these questions, nonequilibrium magnon diffusion equation is solved using the finite element method. The nonequilibrium magnon diffusion equation in YIG given by Ref. [96] reads

$$D \nabla^2 \delta m_m - \frac{\delta m_m}{\tau_{th}} = 0 \quad (4.1)$$

where $D$ is the magnon diffusivity, $\delta m_m$ is the nonequilibrium magnetic moment density (equals to $2\mu_B n_m$, $n_m$ is the nonequilibrium magnon density) and $\tau_{th}$ is the magnon life time. Here $\tau_{th} = 1 \times 10^{-6}$ s from Ref. [97] is used. The boundary condition is given by the magnon flux transferred across the Pt/YIG interface as follows [96]
\[- \hat{n}( - D \nabla \delta m_m ) = - G_{me} \delta m_m \] (4.2)

where $\hat{n}$ is the unit normal normal to the interface and $G_{me}$ is the spin convertance which can be estimated using [96]

$$G_{me} \sim \frac{\pi S a_0^5 J_{sd}^2 g_e(E_F)T}{h a_{0M} T_F}$$ (4.3)

where $a_{0I}$ and $a_{0M}$ are the lattice constant of YIG and Pt, $J_{sd}$ is the exchange energy, $g_e(E_F)$ is the density of states of electrons at the Fermi level in Pt and $T_F$ is the Fermi temperature of Pt. Using $a_{0I} = 12.376 \text{ Å}$, $a_{0M} = 3.90 \text{ Å}$, $J_{sd} = 1 \text{ meV}$, $g_e(E_F) = 1.164 \times 10^{23} \text{ eV}^{-1} \text{cm}^3$ [98], and $T_F = 9.812 \times 10^4 \text{ K}$ [98], the $G_{me}$ is calculated to be 31.6 m/s. These parameters are taken at 300 K and are considered to be temperature independent.

The partial differential equation 4.1 is solved with the boundary condition Eqn. 4.2 in a geometry that is identical to the one used in the actual experiment. The 2D map of the solved $\delta m_m$ with the laser focused on the Pt spin injector with $\Delta x = 47 \mu m$ is shown in Fig. 4.13a. Due to the presence of the Pt detector in the $\Delta x \leq 0 \mu m$ region, $\delta m_m$ at the detector side (left) near the Pt/YIG interface ($z=0 \mu m$) is much lower than to the other side of the Pt injector. The lowered $\delta m_m$ is attributed to the loss of nonequilibrium magnons due to the strong spin sinking of the Pt detector. The detailed interfacial $\delta m_m$ profile is shown in Fig. 4.13b by taking a line cut along $z=0$ in Fig. 4.13a. Three different scenarios with different detector/injector configurations on YIG are compared here: (i) only one Pt injector (ii) one Pt injector + one Pt detector (iii) one Pt injector + one Pt detector + unused Pt pads in between. It can be seen that the overall nonequilibrium magnons density decreases by adding more
Figure 4.13: Simulation on spin diffusion in YIG with laser heating. (a) 2D profile of $\delta m_m$ in YIG with laser at $\Delta x = 47 \, \mu m$ (same Pt detector and injector geometry as used in the experiment above.) (b) Interfacial $\delta m_m$ profile along x-direction under different scenarios (i) one Pt injector (ii) one Pt injector + one Pt detector (iii) one Pt injector + one Pt detector + unused Pt pads in between. (c) Spatial dependence of the interfacial magnon gradient along z-direction $\nabla_z \delta m_m$ under different scenarios. (d) Spatial dependence of the integrated spin current injected from YIG into Pt as a function of laser position with different parameters showing the characteristic exponential decay.

Pt pads on top of YIG due to the stronger spin sinking effect. The derivative of $\delta m_m$ along the z-direction evaluated at the Pt/YIG interface ($z=0$), $\nabla_z \delta m_m$, for the three different scenarios discussed above are also shown in Fig. 4.13c. This nonequilibrium
magnon density gradient represents the spin current that is vertically diffused from YIG into Pt across the interface. The presence of unused Pt pads in between the Pt injector and detector draws additional spin current from YIG as expected.

The total magnon spin current, $J_z^s$, at the detector is determined by

$$J_z^s = -\frac{D}{L} \int_{-L}^{0} \nabla_z \delta m dx$$

where $L$ is the length of the Pt detector. This value is calculated and shown in Fig. 4.13d as a function of laser position. The characteristic decay profile of $J_z^s$ follows an exponential function. To determine the upper bond of $\lambda_s$, the input parameter $D$ is varied to fit the simulated $J_z^s$ to the measured $V_{NL}$. A good fitting is achieved using a combination of $D = 0.0053$ m$^2$/s and $\tau = 10^{-6}$ s. The effective $\lambda_s$ can then be determined using $\lambda_s = \sqrt{D \tau_{th}}$ and is found to be $\lambda_s = 73$ $\mu$m for the above input parameters. Since the maximum value of $G_{me}$ and $\tau_{th}$ are used in the simulation, 73 $\mu$m servers as the upper bound of $\lambda_s$. To test the sensitivity of the model to the input parameters, a simulation with a much smaller value of $\tau_{th} = 1 \times 10^{-8}$ s is shown in Fig. 4.13d. In this case, $D = 0.41$ m$^2$/s is used to achieve the best fitting and the resulting $\lambda_s$ is calculated to be 64 $\mu$m. It can be seen that the shape of the $J_z^s$ decay profile and the value of $\lambda_s$ remains relative constant even by varying $\tau_{th}$ by two orders of magnitude. Based on the discussions above, we concluded that the spin diffusion length in YIG is $47 < \lambda_s < 73$ $\mu$m at 23 K.

### 4.7 Conclusions

In this chapter, single crystal YIG thin films are successfully grown on GGG substrates using RF magnetron sputtering with a post-annealing treatment. However comparing to the Pt/YIG(bulk) sample, the Pt/YIG(thin film)/GGG sample yields a much lower opto-thermal SSE signal, and therefore the former sample structure is used
to study the magnon spin diffusion length in YIG. To do so, a nonlocal opto-thermal spin Seebeck measurement configuration is proposed and employed in which the spin injection and detection are achieved on separate Pt pads. The measured nonlocal signal $V_{NL}$ is attributed to the pure diffusion of the thermally induced magnons. This is verified using the finite element modeling based on the heat and spin transport equations. The effective spin diffusion length is measured to be $47 < \lambda_s < 73 \, \mu m$ at 23 K in YIG which is much longer than the magnon thermal mean-free path in Ref. [93]. This indicates that the magnonic spins retain themselves even after several inelastic scattering events.

Currently, the temperature dependent magnon spin diffusion length is measured in a newly installed cryostat. The improved background noise allows us to probe the decay of $V_{NL}$ more accurately. It is observed that the decay of $V_{NL}$ fits better with a bi-exponential function with two decay length $\lambda_1$ and $\lambda_2$ at lower temperatures (below 150 K) while a single exponential function fits the measured $V_{NL}$ better at higher temperatures. Remeasuring the decay of $V_{NL}$ at 20 K gives $\lambda_1 \sim 10 \, \mu m$ and $\lambda_2 \sim 70 \, \mu m$. A revised spin transport model based on a new spin diffusion formalism from Ref. [30] suggests that the short decay length $\lambda_1$ is associated with the spin diffusion while the long decay length $\lambda_2$ is attributed to the intrinsic spin Seebeck effect signal due to the non-vanishing $\nabla T_z$ at the detector. The manuscript on the temperature dependent spin diffusion length in YIG and the interpretation on the two characteristic decay constant is being prepared [99].
Chapter 5: Magnon Dynamics from the ns to ms Range Probed by the Time Resolved Longitudinal SSE

This chapter is adapted from the manuscript (in preparation) by the authors Zihao Yang, John S. Jamison, Brandon L. Giles and Roberto C. Myers.

The steady-state diffusive magnon transport in YIG measured via the nonlocal opto-thermal spin Seebeck effect is discussed in chapter 4. Although there is plenty of reports on the steady-state thermally-induced magnon transport [88, 80, 100, 101], not much work has been done towards the dynamics of these magnon involved in the spin Seebeck effect. On the ns to ms time scale, Roschewsky et al. [102] studied the laser-induced spin Seebeck effect as a function of laser modulation frequency at room temperature in a Pt/YIG sample. The authors found that the bandwidth of the spin Seebeck effect is as high as 30 MHz and suggested a lower bond of the time constant of ~ 5 ns for the spin Seebeck effect. However, probing the accurate time constant is hindered by the RC low pass filter nature of the sample and experimental setup. Agrawal et al. [103] studied the spin Seebeck signal waveforms using laser heating in time domain at room temperature in a Pt/YIG sample. By comparing the temperature profiles from the thermal modeling with the measured spin Seebeck waveforms, the authors were able to differentiate the interfacial and bulk (due to magnon diffusion) contributions to the spin Seebeck effect. However, there are two
shortcomings in this study: (i) The 2D geometry used in the thermal modeling is not appropriate. Under a laser heating condition, the constant heat power surface is a concentric half circle in 2D, while in 3D it is a homocentric hemisphere. Therefore considering the 3D nature of the sample, the temperature profiles solved using the 2D geometry is not accurate. (ii) The model only contains thermal transport (with no magnon spin transport included). On the ultrafast time scales, Kimling et al. [104] observed the time scale for the angular momentum transfer at Pt/YIG interface is picosecond via the time-resolved magneto-optic Kerr effect measurement at 300 K. In this experiment, the measured signal is primarily originated from the interfacial spin current and thus contains no information of the bulk magnon spin current. Based on the discussion above, a complete quantitative study on the dynamics of the interface and bulk magnon spin currents involved in the spin Seebeck effect has not been performed between ns to ms at different temperatures.

In this chapter, a transistor-transistor-logic (TTL) modulated laser is applied on a Pt/YIG sample and the time domain waveforms of the laser-induced spin Seebeck voltage are probed using an oscilloscope at different temperatures. In Sec. 5.1, the spin Seebeck effect is separately probed using the lock-in amplifier and oscilloscope. A good agreement in the SSE signal magnitude between the two measurements is observed. The temporal spin Seebeck voltage waveforms measured using the oscilloscope exhibit a fast rise in the ns time scale (comparable to the rise time of the input laser power) and a slow rise in the ms time scale at all temperatures. In Sec. 5.2, time domain finite element method simulations based on the heat and spin transport equations are performed to understand the underlying physics of the different components in the measured signal. It is found that the spin current due to the interfacial
electron magnon difference $\Delta T_{me}$ is responsible for the fast rise component while the spin current due to the bulk magnon diffusion is associated with the slow rise component. More importantly, the magnon diffusion lags behind the thermal diffusion (the driving force for magnon diffusion) and dominates the speed of the slow rise component at low temperatures. On the contrary, the thermal diffusion is the limiting factor for the slow rise at higher temperatures. In Sec. 5.3, the magnon spin diffusivity as a function of temperature is extracted from by fitting the simulated time domain spin current to the measured spin Seebeck voltage waveforms.

5.1 The time domain spin Seebeck waveforms measured using the oscilloscope

In this experiment, a continuous wave laser at 980 nm from the Newport Company is modulated at 1 kHz using a function generator. The laser is focused on a Pt/YIG sample using a 40X objective. The (100) orientated single side polished single crystal YIG samples with dimensions of $5 \times 5 \times 0.5$ mm are commercially obtained from the Princeton Scientific. The 6 nm Pt is deposited via the off-axis sputtering. The Pt thin film is then pattered into a $50 \times 50 \mu$m square using photolithography and the Cl$_2$ and CF$_4$ based dry etching. The sample is mounted in a Montana cyrostat and the spin Seebeck voltage is measured with a Keysight oscilloscope using co-axis cables. The schematic diagram of the sample and measurement setup is shown in Fig. 5.1a. The radius of the laser spot is $\sim 3 \mu$m and the laser incident power is $\sim 14.7$ mW. The focused laser on the Pt thin film creates a temperature gradient $\nabla T_z$ in the z-direction and the magnetic field $H_x$ is applied along the x-direction. The generated spin Seebeck voltage $V_y$ is in the y-direction. An example of $V_y$ measured as a function of external magnetic field using a lock-in amplifier is shown in 5.1a.
The magnetic field dependent $V_y$ tracks the YIG magnetization. Here $V_{SSE}$ is defined as the magnitude difference between the saturation values at the high positive and negative magnetic field $V_{SSE} = \frac{V_s(+H) - V_s(-H)}{2}$.

The time resolved spin Seebeck waveforms are measured with a high speed digital storage oscilloscope with a sampling rate of 1.25 GSa/s. The bandwidth of the measurement setup $\sim$ 350 MHz is limited by the Stanford current preamplifier. This corresponds to a rise time of $\sim$ 1 ns which sets the time resolution of our time domain measurements. An example of the measured time domain spin Seebeck voltage traces at magnetic fields of $\pm$ 1.25 kOe at 30 K is shown in 5.1c main panel. The spin Seebeck voltage traces show opposite signs when the magnetic field direction is reversed. The background voltage is found to be stable during the course of the measurement and is possibly induced either by the regular Seebeck effect between the metal wires on the sample or by the small DC noise from the electronic circuits in the setup. The value of the saturated $V_{SSE}$ after subtracting the background voltage is $\sim$ 8.5 $\mu$V. This is different from the value $\sim$ 3.8 $\mu$V measured by the lock-in amplifier. This is due to the fact that the lock-in measures the RMS value for the 1st harmonic of the modulation frequency. Therefore, a factor (for the square wave) of $\frac{\sqrt{2}}{\pi} \sim 0.45$ should be used to convert the absolute value of 8.5 $\mu$V measured by the oscilloscope to the RMS value of $\sim$ 3.8 $\mu$V measured by the lock-in. Note, this conversion factor might be different at higher temperatures since the shape of the spin Seebeck voltage waveforms start deviating from the square wave assumption at higher temperatures.

The temperature dependence of the time domain spin Seebeck voltage waveforms are measured and shown in Fig. 5.2 at different temperatures between 20-300 K. These raw traces are plotted on the linear time scale in 5.2a. The magnitude of the
Figure 5.1: (a) Schematic diagram of the experimental setup. (b) $V_y$ as a function of magnetic field measured using the lock-in amplifier at 30K. (c) Time domain $V_y$ waveforms as a function magnetic field measured using the oscilloscope at 30 K. Inset: Photodiode response of the laser power measured using the oscilloscope. The rise time (10-90%) is $\sim 2.2$ ns.
Figure 5.2: Time domain spin Seebeck voltage waveform at different temperatures in (a) linear scale and (b) semilog scale.

$V_{SSE}$ increases from 30 K and peaks at 80 K and followed by a decrease to a minimum value at 300 K. This trend is similar to the spin Seebeck coefficient reported in Ref. [29]. In terms of the shape of the waveform, the spin Seebeck voltage rises and saturates rapidly at low temperatures while the rise rate in the spin Seebeck voltage slows down at higher temperatures. A minimum rise speed at 180K is observed.
The shape of these time domain spin Seebeck voltage waveform is better illustrated in Fig. 5.2b where the normalized signal (using $\frac{V_{SSE}}{V_{SSE}(t=500\mu s)}$) are plotted on the semilog time scale. After the laser is turned on at $\sim 10^{-8}$ s, the spin Seebeck voltage exhibits a fast rise within the first $\sim 3$ ns. This time scale is comparable to the rise time of the laser power. After the laser power saturates, a continuous slow rise upto ms is observed at all temperatures. The time constant for the slow rise component strongly depends on the measured temperature. Generally, it takes less time to saturate at low temperatures but much longer at high temperatures.

5.2 Time domain FEM simulations using heat and spin transport equations

Finite-element method (FEM) modeling is used here to understand the underlying physics behind the time domain spin Seebeck traces due to the complicated nature of the spin and heat transport in our sample structure. The commercially available FEM software COMSOL is used to solve the heat and spin transport equations [105]. Agrawal et al. [103] proposed a model that only contains thermal transport to explain the dynamics of the spin Seebeck effect. We first adopted the similar method as suggested in [103] to test the validity of this model at different temperatures. In this model, the heat transport equation solved in the heat transport model reads

$$\rho C \frac{\partial T}{\partial t} - \nabla \cdot (\kappa \nabla T) = Q$$  \hspace{1cm} (5.1)

where $C$ is the specific heat, $\rho$ is the density, $\kappa$ is the thermal conductivity and $Q$ is the volume heat source. The temperature dependent input parameters are taken from Ref. [93, 90, 91] and are not listed here. The time domain temperature profile is
solved in a 3D geometry similar to the sample structure used in the measurement. In
this geometry, the heat is only absorbed in Pt due to the wavelength of the laser 980
nm used in the experiment. As suggested by Agrawal et al. [103], the time domain
spin Seebeck voltage can be described as the combination of the surface and volume
integral of the thermal driving force \( \int \int \nabla T_z 2\pi r dr + \int \int \int \nabla T_z e^{-z/l_s} 2\pi r dr dz \) where \( l_s \) is the effective spin diffusion length. In this equation, the first term represents the
contribution from the interfacial spin current while the second term stands for the
spin current due to the bulk magnon diffusion. \( l_s \) governs the length scale where \( \nabla T_z \) is taken into consideration. The comparison between the normalized spin Seebeck
voltage waveforms and the surface and volume integrals of \( \nabla T_z \) is shown in Fig. 5.3
at different temperatures.

In Fig. 5.3, \( \int \int \nabla T_z 2\pi r dr + \int \int \int \nabla T_z e^{-z/l_s} 2\pi r dr dz \) (green dots) calculated from
the thermal modeling agree well with the measured time domain spin Seebeck wave-
forms at higher temperatures (200 K and 300 K). The values of the fitting parameter,
spin diffusion length \( l_s \), are comparable to the reported values in Ref. [80, 106].
The \( \int \int \nabla T_z 2\pi r dr \) term (red dots) corresponds to the fast rise in the measured time
domain SSE data due to the immediate interfacial temperate response after the
laser is turned on. The \( \int \int \int \nabla T_z e^{-z/l_s} 2\pi r dr dz \) term (blue dots) exhibits a much
slower rise after the introduction of the laser due to the extended temperature re-
response time in the bulk of YIG. However, there is a large discrepancy between
\( \int \int \nabla T_z 2\pi r dr + \int \int \int \nabla T_z e^{-z/l_s} 2\pi r dr dz \) calculated from the thermal modeling and the
measured time domain spin Seebeck waveforms at lower temperatures (30 K as an
example). The simulation result at low temperatures rises much faster than the mea-
sured time domain data. Since the interfacial component \( \int \int \nabla T_z 2\pi r dr \) still tracks
Figure 5.3: Comparison of the time domain spin Seebeck voltage waveform and the simulated temperature profile using a temperature model at 300 K, 200 K and 30 K.

The fast rise in the measured spin Seebeck waveform, the discrepancy is attributed to the calculated bulk magnon diffusion term. At low temperatures, $\nabla T_z$ in the bulk of YIG rise much faster compared to high temperatures due to the extremely large thermal diffusivity $\kappa/\rho C$. Therefore, the resulting bulk magnon diffusion term $\int\int\int \nabla T_z e^{-z/l_s} 2\pi r dr dz$ exhibits a much faster saturation at low temperatures. It can be seen that the calculated bulk magnon diffusion term saturates much earlier than the measured spin Seebeck trace even with $l_s = 50 \mu m$ (a much larger value than the reported values [80, 106]). Based on these observations, we concluded that it is
not sufficient to analyze the time domain SSE data with thermal modeling alone. A model consisting of both thermal and spin transports is required and discussed in the following paragraphs.

According to [107, 30], the spin current at Pt/YIG interface that contributes to the spin Seebeck voltage consists of a component proportional to the interfacial electron and magnon temperature difference $\Delta T_{me}$ and a component proportional to the interfacial spin chemical potential difference $\Delta \mu$. Therefore, we bulkd the model based on a three temperature model including electrons ($T_e$), phonons ($T_p$) and magnons ($T_m$) temperature plus a spin diffusion model. The thermal transport equations involved in the three temperature model in Pt reads [108]

$$\rho C_e \frac{\partial T_e}{\partial t} - \kappa_e \nabla^2 T_e = Q_{\text{laser}}(t, x, y, z) - G_{ep}(T_e - T_p) \quad (5.2a)$$

$$\rho C_p \frac{\partial T_p}{\partial t} - \kappa_p \nabla^2 T_p = G_{ep}(T_e - T_p) \quad (5.2b)$$

where $C_e$ and $C_p$ are the electron and phonon specific heat, $\kappa_e$ and $\kappa_p$ are the electron phonon thermal conductivity, $G_{ep}$ is the electron phonon coupling constant. The thermal transport equations involved in the three temperature model in YIG reads [108]

$$\rho C_p \frac{\partial T_p}{\partial t} - \kappa_p \nabla^2 T_p = -G_{pm}(T_p - T_m) \quad (5.3a)$$

$$\rho C_m \frac{\partial T_m}{\partial t} - \kappa_m \nabla^2 T_m = G_{pm}(T_p - T_m) \quad (5.3b)$$

where $C_m$ is the magnon specific heat, $\kappa_m$ is the magnon thermal conductivity, $G_{pm}$ is the phonon magnon coupling constant. The interface boundary conditions are given by the (i) $-\kappa_p \frac{dT_p}{dx} = \frac{1}{R_{th}} \Delta T_p$ (for phonons) (ii) $-\kappa_m \frac{dT_m}{dx} = \frac{1}{R_{th}} \Delta T_m$ (for electrons and
magnons) where $R_{th}^P$ and $R_{th}^m$ are the Kapitza resistance and effective thermal electron magon resistance.

![Graph showing time domain spin Seebeck voltage waveform and simulated spin current profile using a three temperature with spin diffusion model at 300 K, 200 K and 100 K.]

The time domain spin diffusion equation reads as [30]

$$\frac{\partial \delta n_x}{\partial t} - \sigma \nabla^2 \mu - \varsigma \nabla^2 T_m = -g_{n\mu} \mu$$

(5.4)
where $\delta n_x$ is the nonequilibrium magnon density that can be expanded to the first order as $\frac{n_x^{(0)}}{T} \mu$ ($n_x^{(0)}$ is the equilibrium magnon density at the equilibrium temperature), $\sigma$ is the spin conductivity, $\mu$ is the magnon chemical potential, $\varsigma$ is the intrinsic spin Seebeck coefficient, $g_{nm}$ represents the relaxation of magnons to the phonons. The interface boundary condition is given by the spin current continuity equation

$$\dot{j}_s = -\sigma \nabla \mu - \varsigma \nabla T_m = G(\mu_{Pt} - \mu) + S(T_e - T_m)$$

where $G$ and $S$ are the interfacial spin conductance and spin Seebeck coefficient. The spin current $j_s$ has a unit of $1/m^2s$ representing the magnon flux across the Pt/YIG interface. There are four unknowns $T_e, T_p, T_m$ and $\mu$ in this model. The temperature unknowns $T_e, T_p, T_m$ are first solved and the results are used as input to calculate $\mu$. The total interfacial spin current that is picked up by the detector can be calculated using 

$$j_s = \int \int S \Delta T_{me} 2\pi r dr + \int \int G \mu 2\pi r dr.$$ 

This value is proportional to the measured spin Seebeck signal and therefore it is used to compare to the time domain spin Seebeck traces at different temperatures. The parameters used in the simulations are listed in Appendix D.1.

The comparison between the simulated $j_s$ and measured spin Seebeck traces is shown in Fig. 5.4. It can be seen that the fast rise in the measured signal is associated with the $j_s^{int} = \int \int S \Delta T_{me} 2\pi r dr j_s|_{int}$. This term represents the spin pumping due to the interfacial spin torque transfer between magnons and electrons. Since the Pt film on top of YIG is ultrathin, the temperatures of electrons in Pt and magnons in YIG at the interface can instantaneously respond to the rise of the laser power. On the other hand, the second term $j_s^{diff} = \int \int G \mu 2\pi r dr$ is responsible for the slow rise in the measured SSE signal. The magnon chemical potential $\mu$ at the interface is governed by the magnon diffusion that is induced by $\nabla T$ in the bulk of YIG. Therefore $j_s^{diff}$ is sensitive to the combination of both thermal and spin diffusions. In Fig. 5.3, the
simulation result from the pure thermal diffusion is found to be much faster than
the actual slow rise of the measured time domain signal at low temperatures. After
taking the spin diffusion into consideration, a good agreement between the simulated
\( j^{\text{int}}_s \) and time domain data is achieved as shown in Fig. 5.4. This suggests that the
spin diffusion dynamics is much slower than the thermal diffusion in YIG at lower
temperatures. The comparison between the thermal and spin diffusion involved in
the SSE is discussed in details in Sec. 5.3.

5.3 Discussions on the fitting parameters used in the time
domain FEM simulations

The fitting parameters used in the simulations in the last section are \( S/G \) ratio
(interfacial and bulk spin current ratio), \( l_s \) (spin diffusion length) and \( D_s \) (spin diffusivity). Among these fitting parameters, the first fitting parameter, \( S/G \) ratio, governs
the relative ratio between the two simulated spin current components \( j^{\text{int}}_s \). The
\( S/G \) ratio is independent from the other two fitting parameters since it does not affect
the shape of the fast or slow rise. The temperature dependence of the \( S/G \) ratio de-
creases as temperature increases as shown in Fig. 5.5b. It also should be noted that,
from theory [30], \( S/G \) ratio should be close to \( k_B \) (this is the magnon thermopower),
while the simulation suggests a ratio more than 200 \( k_B \) at high temperatures. This
could be a result of the underestimation of \( \Delta T_{\text{me}} \) due to the calculated interfacial
magnon thermal resistance \( R_m \). The second fitting parameter, spin diffusion length
\( l_s = \sqrt{\sigma/g_{\mu}} \), is plotted as a function of temperature in Fig. 5.5b. It mainly af-
facts the curvature of the bulk magnon diffusion current \( j^{\text{diff}}_s = \int G\mu 2\pi r dr \). The
magnitudes of \( l_s \) are generally in good agreement with the reported values in Ref.
while their temperature dependencies are slightly different possibly due to the uncertainties in the input parameters used in the simulation.

Figure 5.5: The fitting parameters (a) spin diffusivity $D$ and (b) spin diffusion length $l_s$ and the ratio between interfacial spin Seebeck coefficient $S$ and interfacial spin conductance $G$ at different temperatures.

The third fitting parameter, spin diffusivity $D_s$, is the most critical fitting parameter that determines the speed and the shape of the bulk magnon diffusion current $j_s^{diff}$. Here the spin diffusivity for magnons is defined as $D_s = \frac{\sigma}{n_s^2(k_B T)^{-1}}$ which is similar to the definition of thermal diffusivity for phonons $D_t = \frac{\kappa}{\rho C}$. The dynamics of $j_s^{diff}$
include two processes (i) diffusion of the thermal driving force (i.e. how fast to excite the nonequilibrium magnon) (ii) diffusion of the magnon spins (i.e. how long it takes for the excited magnons to diffuse from the bulk to interface). At high temperatures, the best fit is achieved when the spin diffusivity is larger than the thermal diffusivity as shown in Fig. 5.5a. Tuning the spin diffusivity does not significantly change the shape of simulated $j_s^{\text{diff}}$ as long as it is larger than the thermal diffusivity, therefore the thermal diffusivity serves as a lower bond for the spin diffusivity at temperatures above 200 K. This also implies that the entire slow rise of the measured signal is limited by the thermal transport in YIG. On the other hand, at low temperatures, the thermal driving force rises much faster than the slow rise in the measured signal due to the rapid increase in the thermal diffusivity. This means the other process, spin diffusion, is the limiting factor for the slow rise in $j_s^{\text{diff}}$. The extracted fitting parameter at 100 K is the exact value rather than an upper or lower bound estimation. The spin diffusivity at other temperatures and the underlying physics behind the opposite temperature dependencies between the spin and thermal diffusivity are still under investigation.

In Fig. 5.6, the measured rise time and magnitude for the fast and slow components are directly extracted from the time domain spin Seebeck waveform. In the inset of Fig. 5.6a, the fast rise times $\tau_l$ is almost temperature independent and is comparable to the rise time of the laser power at different temperatures. This indicates the intrinsic response time of the interfacial electron and magnon temperature is possibly faster than the ns time scale and therefore is limited by the rise time of the laser power. In contrast, the rise time of the slow component shows strong temperature dependency. These slow rise time is a combination result from the thermal
and spin diffusion. The detailed comparison between the extracted spin diffusivity and measured slow rise time will be discussed after the simulations over the entire temperature range are finished. In Fig. 5.6b, the magnitude of the slow component peaks at around 80 K and dominates the total signal magnitude at most temperatures while the magnitude of the fast component is relatively constant as a function of temperature up to around 200 K. The correlation between the measured magnitude and the S/G ratio is still under investigation.
5.4 Conclusions

In this chapter, the time domain spin Seebeck waveforms are measured on a Pt/YIG sample using an oscilloscope at different temperatures. Two distinct regions are found in the time domain spin Seebeck traces including a fast rise $\sim$ ns and a slow rise region $\sim$ ms. Comparing to the simulation results by solving the heat and spin transport equations, the fast and slow rise is attributed to be originated from the interfacial electron magnon temperature difference and bulk magnon diffusion. Thermal (or spin) diffusive transport is found to dominate the speed of the slow rise component in the measured SSE waveforms at high (or low) temperatures. The rise time of the fast component is found to be independent of temperature while the rise time of the slow component gradually rises as temperature increases. Simulations throughout the entire temperature range is still under investigation in order to extract the spin diffusivity and $S/G$ ratio. These fitting parameters will be compared to the measured rise time and the relative magnitude of the fast and slow rise components in the measured time domain spin Seebeck waveforms.
Chapter 6: Scalable Nernst Thermoelectric Power Using a Coiled Galfenol Wire

The content in this chapter is adapted from a submitted manuscript to Applied Physics Letters in Ref. [109] by the authors Zihao Yang, Emilio A. Codecido, Jason Marquez, Yuanhua Zheng, Joseph P. Heremans, Roberto C. Myers.

The magnon diffusion transport and dynamics involved in the spin Seebeck effect have been discussed in the previous chapters. Although the physics of spin Seebeck effect have been extensively studied, there has not been much studies on utilizing this spin thermal effect for real applications. It has been reported that the giant spin Seebeck coefficient could be as high as $\sim 2000 \, \mu V/K$ at 4.3 K in n-type InSb. As for YIG, it possesses a spin Seebeck coefficient of 30 nV/K (and $\sim 300 \, nV/K$ if a homoepitaxial YIG thin film is grown on the bulk YIG prior to the Pt deposition) at room temperature. Here we attempted to employ a cylindrical geometry consisting of a Pt thin film coil and a Fe$_2$O$_3$ bulk cylinder material to harvest the spin Seebeck voltage as a proof of concept for the spin caloritronic generator. In this geometry, the temperature gradient and magnetic field are applied along the radial and axial direction respectively. Comparing to the conventional planar structure, the electric field in Pt coil is generated along the azimuthal direction in the cylindrical geometry. Therefore, the output voltage can be easily scaled up by increasing the Pt coil length.
Unfortunately, the output voltage from the device discussed above is small due to the porous surface nature of the Fe$_2$O$_3$ sample. This results in a low-quality Pt/Fe$_2$O$_3$ interface which greatly reduces the efficiency of the spin injection from Fe$_2$O$_3$ into Pt. Later, we proposed a novel thermoelectric generator based on the Nernst and anomalous Nernst effect using the similar cylindrical geometry. The voltage generation from the spin Seebeck effect and the ordinary (or anomalous) Nernst effect share the same requirement of the orthogonally orientated applied temperature gradient and magnetic field to produce the output voltage. In addition, the thermopower generation using the ordinary (or anomalous) Nernst effect can be realized by using only one material thus eliminating the interface quality effect on the output signal in the spin caloritronic generator.

Although the Nernst thermopower is generally thought to be too weak in most metals for converting waste heat into useful electricity, its transverse orientation makes it more robust and advantageous over the conventional Seebeck effect on non-flat heater surfaces. In this geometry, a metal wire (Galfenol) is coiled around a hot cylinder where the temperature gradient and the magnetic field are applied along the radial and axial directions. The voltage generated from the azimuthal electric field along the wire is measured as a function of temperature gradient, magnetic field and wire length. Galfenol (FeGa), a magnetostrictive metal, is used as the thermoelectric generator material since it is envisioned that this type of material exhibits a larger anomalous Nernst coefficient due to the enhanced magnon-phonon coupling.

In Sec. 6.1, we first discussed the design of a spin caloritronic generator utilizing the spin Seebeck effect on a cylindrical ferrimagnetic oxide Fe$_2$O$_3$ coated with a coiled Pt thin film. Due to the low performance of the spin caloritronic generator, a better
Nernst thermopower generator using a coiled FeGa wire is introduced in Sec. 6.2. The output thermoelectric voltages as a function of magnetic field, temperature gradient and wire length are discussed in Sec. 6.3. Finite element method simulations are performed in Sec. 6.4 to solve the temperature profiles in the thermoelectric generator. In Sec. 6.5, the Nernst coefficient for Galfenol is calculated based on the results from the simulation and the measured thermovoltage. This Nernst coefficient is compared with the measurements on the bulk FeGa sample and other magnetic materials.

6.1 Attempt of building a spin caloritronic generator using the spin Seebeck effect

As discussed in the previous chapters, an inverse spin Hall voltage can be generated via the thermal spin pumping from the ferromagnetic material into the spin transducer through the spin Seebeck effect. Therefore, a power generator based off the spin Seebeck effect could potentially be realized to convert the waste heat energy into electricity. First, we take the conventional Pt/YIG system as an example. The output voltage follows $V_{ISHE} = L \cdot S \cdot (\vec{j}_s \times \vec{\sigma})$ where $L$ is the length of the Pt, $S$ is the spin Seebeck coefficient, $j_s$ is the spin current and $\sigma$ is the spin polarization vector in YIG. At a fixed temperature gradient (fixed $j_s$) and a fixed magnetic field (fixed $\sigma$), the output voltage scales with the length of the spin transducer Pt. In this case, a coiled Pt thin film on a cylindrical YIG sample is suitable to harvest large output voltage and power since it can easily accommodates a long Pt spin transducer. This geometry is illustrated in Fig. 6.1 where a heater is used to create the radial temperature gradient (radial $j_s$) and an external magnetic field is applied along the axial direction. The spin current injected from Fe$_2$O$_3$ into Pt induces an azimuthal
electric field at every point in Pt and generates an output voltage through the entire
length. The readily available Fe$_2$O$_3$ material is used instead of YIG in this experiment.

![Figure 6.1: Schematic of the spin caloritronics generator using spin Seebeck effect. The temperature gradient is along radial direction and the applied magnetic filed is along axial direction. The generated output electric field is along azimuthal direction.](image)

The Fe$_2$O$_3$ sample with a diameter of 0.5 cm and a length of 2.5 cm is commercially
obtained from the Fair-Rite Products Corp. The Fe$_2$O$_3$ sample is first hand polished
to achieve the mirror finish surface and then coated with 10 nm Pt thin film using e-beam evaporation. This cylindrical coating of Pt is achieved by rotating the sample
along its own axial axis during the e-beam deposition. The rotation is driven by an external motor outside the e-beam evaporation chamber. A lathe tool is used to patter the Pt into the coil geometry after the Pt deposition. The cut on the Pt coil can be observed in the inset in Fig. 6.2.

The inverse spin Hall voltages as a function of magnetic field with different input heater powers are shown in Fig. 6.3a. All the data are recorded with respect to the lab time. No significant voltage due to the spin Seebeck effect is observed at a stable magnetic field of $\pm$ 0.2 T. The peak in the output voltage observed during
the magnetic field ramping is due to the time-varying magnetic flux from Fe$_2$O$_3$. The nonzero voltage measured close to the end of the lab time with a heater voltage of 12 V is likely to be induced by the background drift in the measurement setup. The absence of the thermally induced voltage in this device is attributed to the porous surface morphology of Fe$_2$O$_3$ as shown by the AFM images in Fig. 6.3b. The surface roughness of Fe$_2$O$_3$ is around 300 nm resulting a low quality Pt/Fe$_2$O$_3$ interface and a non-continuous Pt film. Both of them tend to decrease the spin transfer efficiency between the Fe$_2$O$_3$ and Pt thus yielding an almost zero output voltage. In the following sections, we demonstrated a novel Nernst thermoelectric generator that is built on the similar coiled geometry.
Figure 6.3: Output voltage and the surface morphology of the spin Seebeck generator. (a) The output spin Seebeck voltage as a function of lab time. The voltages are measured at different applied magnetic field with various heater powers. (b) AFM image of the porous surface morphology of Fe$_2$O$_3$ with a RMS of $\sim$ 300 nm.

6.2 Design of the novel Nernst thermoelectric generator using a coiled Galfenol wire

Converting the waste heat energy into electricity through thermoelectric means is considered as an environmentally friendly approach. Among all thermoelectric phenomena, the Seebeck effect is most widely used to achieve this purpose. In this case, a longitudinal electric field is generated parallel to the applied thermal gradient. Alternatively, there is another transverse thermoelectric effect, called the Nernst effect.
In this case, a transverse electric field is produced perpendicular to the orthogonally orientated temperature gradient and magnetic field. Although the Nernst effect is usually considered as a weaker effect compared to the Seebeck effect, the Nernst effect are more flexible and favorable for the thermopower generation in certain cases [110]. To be more specific, unlike the Seebeck effect, the Nernst electric field is not in the same direction as the applied thermal gradient. Therefore the Nernst voltage can be scaled up linearly by increasing the length of the thermoelectric generator material. In addition, since only one material is needed in the Nernst thermoelectric generator, the conventional thermopile structure with series connected n- and p-type semiconductor used in the Seebeck thermoelectric generator can be avoided thus simplifying the generator fabrication process. This feature is particularly useful for a non-flat heater surface, i.e. a cylindrical heater. In this case, a thermovoltage generator metal wire is wrapped around a hot cylinder producing the Nernst voltage that can be scaled up linearly with the number of turns of the wire. This device was first prosed by Norwood in Ref. [111]. This thermoelectric generator based on the Nernst effects offers an attractive alternative way for the waste heat recovery on non-flat surfaces, such as hot exhaust pipes.

The Nernst effect usually includes an ordinary ($E_{ONE}$) and an anomalous ($E_{ANE}$) components in magnetic materials. They can be expressed as a function of temperature gradient and magnetic field as follows [110]

\[
E_{ONE} = N_{ONE} \cdot \mu_0 \cdot \left( \vec{H} \times \vec{\nabla}T \right)
\]

(6.1a)

\[
E_{ANE} = N_{ANE} \cdot \mu_0 \cdot \left( \vec{M} \times \vec{\nabla}T \right)
\]

(6.1b)
where $N_{\text{ONE}}$ and $N_{\text{ANE}}$ are the ordinary and anomalous Nernst coefficients and $\vec{H}$ and $\vec{M}$ are the applied magnetic field and magnetization, respectively. Recently, Sakuraba [110] estimated the performance of a coiled cylindrical thermoelectric generator and concluded that a larger Nernst coefficient than those typically found in metals is needed to achieve the power density for any practice usage. It has been shown that the phonon drag could enhance the Nernst coefficient at low temperatures in bismuth [112, 113]. More recently, it is reported that the anomalous Nernst coefficient in single crystal Fe is benefited from the magnon drag up to 200 K [114]. In this study, we propose to use Galfenol ($\text{Fe}_{100-x}\text{Ga}_x$), a magnetostrictive metallic metal, as the thermoelectric generator material since it potentially has a large Nernst coefficient due to the presence of the strong magnon phonon coupling in this material.

Galfenol stands for a type of magnetostrictive alloys that are low cost but with high strength and great flexibility. The magnetostriction in the Tb doped $\text{Fe}_{83}\text{Ga}_{17}$ is shown to be as high as $\sim 886$ ppm [115]. The magnetostriction could be further enhanced in Galfenol by increasing the Ga content up to 20 at%. However, at 15 at% Ga, Galfenol metal exhibits the ductile-to-brittle transition [116, 117, 118]. Therefore, to achieve both high magnetostriction and flexibility, ductile Galfenol wires with 15 at% Ga and 85 at% Fe obtained from the Etrema Products INC are used in the experiment. The diameter of the wire is $\sim 508 \mu\text{m}$. The fabrication process of the wire is similar to the process in Ref. [118].

The Galfenol wire is first encased in a polytetrafluoroethylene (PTFE) shrink tube with a wall thickness of 254 $\mu\text{m}$. The PTFE shrink tube provides the electrical insulation to the heater surface and wire itself. The Galfenol wire together with the PTFE shrink tube are wrapped around a cylindrical cartridge heater and placed
between the magnetic poles. The entire setup is shown in Fig. 6.4. In cylindrical coordinates, the temperature gradient generated by the cartridge heater is along the radial direction, $\vec{\nabla} T = T_r \cdot \vec{r}$, and the applied magnetic field is along the axial direction, $\vec{H} = H_z \cdot \vec{z}$. According to Eqn. 6.1, the Nernst electric field is generated along the azimuthal direction, $\vec{E}_{NE} = N_{ONE} \mu_0 H_z \nabla r \hat{\phi} + N_{ANE} \mu_0 M_z \nabla T_r \hat{\phi}$, at every point along the coiled wire, and a resulted voltage drop, $V$, can be measured between the two leads of the wire. The measurement setup consists of a Keithley 2700 multimeter, a cartridge heater from the Omega Engineering and an electromagnet from the SES Instruments.

Figure 6.4: Schematic of the Nernst generator. The temperature gradient is along radial direction and the applied magnetic field is along axial direction. The generated output electric field is along azimuthal direction.

### 6.3 Scalable thermoelectric voltage from the coiled Nernst thermoelectric generator

In this section, the output thermoelectric voltage from a coiled Galfenol wire wrapped around a hot cylinder is characterized as a function of heater power, magnetic field and wire length. This serves as the first experimental proof-of-concept for a novel
thermoelectric generator based on the Nernst effect on a non-flat heater surface which shows scalable output thermovoltage generation.

An example of the raw Nernst voltage generated from the Galfenol coil with a wire length of $\sim 59$ cm and a radial temperature gradient of $\sim 652$ K/m is plotted in blue open circles as a function of applied magnetic field in Fig. 6.5a. Here, the magnetic field is swept in a hysteretic manner. The magnetic field dependent raw Nernst voltage is generally proportional to the magnetic field reaching a maximum voltage of $\sim 166 \mu V$ at a magnetic field of 0.18 T and an input heating power of 4.1 W. A good agreement between the raw Nernst voltage and the linear fit (red dotted line) is observed where the slope of the fitted curve is 0.98 mV/T. The background subtracted Nernst voltage measured in the low magnetic field region is shown in the bottom right inset in Fig. 6.5a. An open hysteresis loop is observed indicating the anomalous contribution to the total Nernst voltage. This characteristic is observed in multiple samples with different wire lengths. The background voltage is around 6 $\mu V$ and is much smaller than the total magnitude of the measured raw Nernst voltage. This small background voltage could be a result of the Seebeck effect that is induced by the small temperature difference between the electrical contacts at the two ends of the wire.

To study this anomalous contribution, a qualitative magnetization measurement is performed on this Galfenol wire. Due to the large sample size and its coil geometry, an accurate magnetization measurement using the standard magnetometry is not straightforward. Alternatively, a digital tesla-meter is placed right next to the coil to measure the total local magnetic field $B_l$ as a function of applied field $H_l$. The applied magnetic field $H_l$ is obtained by measuring the electromagnet alone with the
Figure 6.5: Output voltage from the Nernst generator. (a) Nernst voltage as a function of applied magnetic field with a radial temperature gradient of $\nabla T_r = 652 \text{ K/m}$. Top right inset: local magnetization of the Galfenol wire at room temperature. Bottom left inset: Nernst voltage measured in the low magnetic field region. (b) 2D map of Nernst voltage as a function of applied magnetic field and radial temperature gradient. The dashed black line cut represents the data in Fig. 6.5a.

coil removed. The local magnetization of the Galfenol coil can then be determined from the difference between the two based on $\mu_0 M_l = (B_l - \mu_0 H_l)$. The resulting local magnetization $M_l$ is plotted in the top left inset in Fig. 6.5a. A finite remnant magnetization at zero magnetic field can be observed in the $M_l - H_l$ characteristic indicating the ferromagnetic nature of the Galfenol coil. In addition, the magnetization does not saturate at the maximum applied field of 0.18 T. This hard axis behavior is attributed to the shape anisotropy from the coil geometry. Although the anomalous
Nernst contribution is expected to dominate the total Nernst voltage in magnetic materials, the difference in the shape between the local magnetization and total Nernst voltage suggests the presence of both ordinary and anomalous Nernst components. The background corrected Nernst voltage as a function of applied magnetic field and temperature gradient is shown in Fig. 6.5b. At a fixed radial temperature gradient, the Nernst voltage shows a linear relationship with respect to the magnetic field where the slope increases from 67 V/T to 1.42 mV/T with an increasing temperature gradient from $\sim 30$ to 1000 K/m. The black dashed line represents the data shown in Fig. 6.5a. At a fixed applied magnetic field, the Nernst thermopower can be calculated based on $\alpha_{r,\phi z} = \frac{E_{\phi}}{\nabla T_r} |_{\mu_0 H_z}$ where a magnitude of 0.4 $\mu$V/K is found at an applied magnetic field of 0.18 T.

The key advantage of the Nernst voltage generation from this coil geometry is its scalability with wire length (L). This is demonstrated by measuring the output voltage at a fixed temperature gradient on various devices with L = 12, 35, and 59 cm as shown in Fig. 6.6a. At a fixed magnetic field, the Nernst voltage shows a boost by increasing the wire length. This length dependence if the Nernst voltage is directly plotted in Fig. 6.6b. A clear proportionality between the Nernst voltage and the wire length L is observed at different thermal gradients.

### 6.4 Simulations on temperature profiles in the coiled Nernst thermoelectric generator

The temperature drop across the Galfenol wire needs to be evaluated to extract the thermopower and Nernst coefficient for Galfenol. However, a direct measurement of the temperature drop is not feasible due to its coiled geometry. Even if it is possible to mount thermocouples on the generator, the measured temperature difference

\[ 112 \]
Figure 6.6: Scalable Nernst voltage generation. (a) Wire length dependent Nernst voltage as a function of magnetic field at a fixed radial temperature gradient of $\nabla T_r = 1000 \text{ K/m}$. (b) Wire length dependent Nernst voltage as a function of radial temperature gradient.

will be dominated by the PTFE shrink tube due to its much lower thermal conductivity compare to the Galfenol wire. In addition, an active air cooling is employed in the measurement setup to stabilize the temperature. The flowing air creates an azimuthally dependent temperature profile in Galfenol that is also not straightforward to measure. Therefore, finite element method simulations based on the thermal transport equations are performed using COMSOL to estimate the temperature profile in our device [105].
Figure 6.7: Temperate profile simulation in Galfenol Nernst generator. (a) Surface temperature profile of the Galfenol and cartridge heater of the Nernst generator and wind speed profile. (b) Comparison of measured and simulated surface temperature on grey pentagon in Fig. 6.7a. (c) Average temperature and temperature gradient of the Galfenol wire as a function of input cartridge heating power. (d) Absolute temperature and temperature gradient profile along the black dotted line in Fig. 6.7a at cartridge heater power of 6.4 W.
The surface temperature profile of a device with a wire length of \( L = 12 \text{ cm} \) is shown in Fig. 6.7a together with the wind speed profile along the \( r - z \) plane. The speed of the inlet air flow is measured to be \( \sim 7 \text{ m/s} \) which corresponds to a Reynolds number in the turbulent air flow regime. In this case, a low Reynolds \( k - \epsilon \) model is used to solve for the air flow profile. The resulted air flow profile is then coupled to the heat transfer model in which the temperature profiles are solved based on heat conduction and convention equations. The input heating power is calculated based on Joules law. A thermocouple is attached to the surface of the coil (grey pentagon in Fig. 6.7a) serving as a temperature reference point to cross check the simulation results. The thermal conductivity \( (\kappa) \), specific heat \( (C_p) \), density \( (\rho) \), ratio of specific heat \( (\gamma) \), and dynamic viscosity \( (\mu) \) used in simulations are shown in Table 6.1. The thermal properties of Galfenol are obtained from a separate measurement on a bulk sample with slightly different composition \( (\text{Fe}_{81.6}\text{Ga}_{18.4}) \) from the same vendor [66].

The density of Galfenol is estimated based on the relative composition of Fe and Ga. In addition, the parameters used in the simulations are assumed to be temperature independent due to the relative small temperature rise in the actual measurements. In Fig. 6.7b, the simulated temperature at the reference point (grey pentagon in Fig. 6.7a) shows a good agreement compared to the measured one at different heating powers. The radial temperature profile and temperature gradient across the entire device along the black dotted line in Fig. 6.7a are shown in Fig. 6.7d. A temperature drop of \( \sim 0.3 \text{ K} \) is found across the Galfenol wire at an input heater power of 6.4 W. Most of the temperature drop happens in the PTFE tube and adjacent air layers due to their low thermal conductivities. The simulated average temperature \( T_{\text{average}} \) and radial temperate gradient \( \nabla T_r \) in the Galfenol wire is shown in Fig. 6.7c at different
input heating powers. These values are used to estimate the Nernst coefficient of the Galfenol wire in the next section.

Table 6.1: Material parameters used in temperature simulation in Nernst generator

<table>
<thead>
<tr>
<th>Material</th>
<th>$\kappa$ (W/Km)</th>
<th>$C_p$ (J/kgK)</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$\gamma$</th>
<th>$\mu$ (Pa·s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>0.024 [119]</td>
<td>1005 [119]</td>
<td>1.2 [120]</td>
<td>1.4 [120]</td>
<td>$2 \times 10^{-5}$ [121]</td>
</tr>
<tr>
<td>Heater (copper)</td>
<td>387 [122]</td>
<td>385 [122]</td>
<td>8930 [122]</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Shrink tube (PTFE)</td>
<td>0.25 [123]</td>
<td>1300 [124]</td>
<td>2200 [125]</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Galfenol</td>
<td>17</td>
<td>437</td>
<td>7329</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

6.5 Large Nernst coefficient in Galfenol

In this section, the Nernst coefficient defined as $N = \frac{\partial \alpha_{\phi_2}(\mu_0 H_z)}{\partial \mu_0 H_z}$ in Ref. [114] is computed based on the measured Nernst voltage (Fig. 6.5b) and the simulated temperature gradient (Fig. 6.7) for Galfenol. The calculated Nernst coefficient is shown as black triangles in Fig. 6.8. The Galfenol wire exhibits a Nernst coefficient of $\sim -2.6 \, \mu V/KT$ near room temperature and shows a weak temperature dependency up to 315 K. This value is compared to the results measured from another independent measurement on a bulk polycrystalline Galfenol (Fe$_{81.6}$Ga$_{18.4}$) sample which is shown as red stars in Fig. 6.8. The bulk sample has a size of $6 \times 2 \times 1$ mm and is obtained from the same vendor. This measurement setup is also used to determine the thermal conductivity and specific heat of Galfenol discussed in the previous section. The Nernst coefficients obtained from two independent measurements agree with each other with a difference less than 20%. To the best of our knowledge, this is the first study on the Nernst coefficient in Galfenol and this value is found to be higher than the previous reported values of 0.189 $\mu V/KT$ in Ref. [126], 0.4 $\mu V/KT$ in Ref. [114],
and 0.56 \( \mu \text{V/KT} \) in Ref. [127] in single crystal \( \text{Fe}_3\text{O}_4 \) bulk, single crystal Fe bulk, and epitaxial FePt thin film measured at room temperature. The Nernst coefficients from Ref. [126] and Ref. [127] are calculated based on the saturation magnetization (\( \mu_0 M_s \)) of the sample and are converted to the saturation magnetic field (\( \mu_0 H_s \)) to compare to our result. The Nernst coefficient measured on the bulk Galfenol sample exhibits a continuous increase from 80 to 300 K. The physical origin of this temperature dependence needs to be further studied. In addition, the Nernst thermopower does not exhibits a saturation up to 1.4 T on the bulk Galfenol sample. This is consistent with unsaturated Nernst voltage measured on the coiled wire generator in Fig. 6.5a.

![Figure 6.8: Nernst coefficient measured on bulk Fe\text{81.6}Ga\text{18.4} and wire Fe\text{85}Ga\text{15} samples. Inset: Magnetic field dependent Nernst thermal power measured in bulk samples at 82 K and 300 K.](image)

### 6.6 Conclusions

In this chapter, we first attempted to realize a spin caloritronics generator operating based on the spin Seebeck effect using a coiled Pt thin film on a \( \text{Fe}_2\text{O}_3 \) cylinder.
However, the device performance is low due to the porous surface morphology of Fe$_2$O$_3$. Later, this cylindrical geometry is used for the generation of the Nernst voltage from a coiled Galfenol wire wrapped on a hot cartridge heater. This device serves as the first proof-of-concept for a novel Nernst thermoelectric generator on a non-flat heat source with a scalable output thermovoltage. The scalability can be easily achieved by increasing the coiled wire length. A giant Nernst coefficient of $\sim -2.6$ $\mu$V/KT is found in the Galfenol coiled generator at room temperature. This result is in good agreement with the measurement performed on the bulk Galfenol sample with a slight composition difference. In addition, Galfenol represents a category of magnetostrictive materials that exhibits a stronger magnon phonon coupling. The enhanced interaction between the magnons and phonons could potentially lead to a larger Nernst coefficient. The current magnon drag Nernst theory predicts that the Nernst coefficient follows $N \approx 0.05 \frac{C_m}{n e^2}$ where $C_m$ is the magnon specific heat and $n$ is the conduction electron density. This formalism does not take the magnon phonon coupling into consideration. Therefore, a model involves strong hybridized magnon phonon excitations (or magnon polarons) is needed to reveal the physics behind the large Nernst coefficient and its temperature dependence in magnetostrictive materials. [105]
Chapter 7: FEM Simulations on the Thermal and Condensed Magnon Transport

This chapter is adapted from a manuscript in preparation by the authors Zihao Yang, Se Kwon Kim, Yaroslav Tserkovnyak and Roberto C. Myers.

The finite element method (FEM) simulations is particularly useful tool to understand the physical phenomena that can be described using partial differential equations with proper initial and boundary conditions. For example, this method has been used to simulate the electron, phonon and magnon temperatures in ultrafast demagnetization physics [128, 129]. Recently, this method has been widely used to study the magnon transport related physics due to the complexity nature of physics involved in these measurements. Scherier et al. [130] simulated the electron magnon temperature difference with an effective electron magnon heat conductance at the Pt/YIG interface. The interfacial magnon electron heat current associated with this temperature difference strongly affects the magnon temperature profile in YIG near the interface region. The authors found out that the calculated spin Seebeck voltages can match to their measurements only when this electron magnon heat conductance is included in their thermal modeling. Furthermore, Cornelissen et al. reported the temperature dependent magnon spin conductivity from the nonlocal magnon transport measurements in Ref. [80, 100, 106]. Specifically, this is done by fitting the measured spin
signal decay to the simulated spin current solved from the spin transport modeling. Later, Shan et al. [101] determined an intrinsic Spin Seebeck coefficient in YIG by comparing the simulated magnon spin current from the coupled spin/heat transport model to the measured spin decay of the thermally induced magnons [80, 100].

Flebus et al. proposed a two fluid theory with a set of transport equations for thermal and condensed magnons based on the Boltzmann transport theory and linear response theory in Ref. [30]. In this chapter, we adopted these transport equations and solved them using the FEM for different scenarios to better understand the transport properties of thermally induced magnons involved in the experiments in chapters 4 and 5. Moreover, in the light of the two fluid thoery, we proposed an experiment design to probe the magnon Bose-Einstein condensation. The commercially available FEM software COMSOL is used to solve the heat and spin transport equations [105]. In Sec. 7.1, we started from the simplest scenario in which these conditions (i) only thermal magnon (ii) 1D geometry and (iii) linear temperature profile are satisfied. The magnon chemical potential together with magnon spin current is solved in the Pt/YIG/Pt and Cu/YIG/Cu structures. In Sec. 7.2, the condition (iii) is dropped off, and the magnon chemical potential and magnon spin current with an arbitrary temperature profile are examined. The magnon chemical potential and spin current are solved in a more realistic scenario with laser heating in a Pt/YIG/Cu structure in Sec. 7.3. In Sec. 7.4, both conditions (i) and (ii) are dropped off, and the transport equations are solved for the coupled thermal and condensed magnons. The corresponding spin current due to thermal and condensed magnons are discussed along with other magnon condensation parameters. Finally, an experiment to probe the magnon BEC is proposed in Sec. 7.5 based on the solutions discussed in Sec. 7.4.
7.1 1D steady-state thermal magnon transport under a linear temperature profile

As discussed above, Flebus et al. proposed a set of transport equations for thermal and condensed magnons in Ref. [30]. We first rearranged the partial differential equation and boundary conditions for the thermal magnon transport so that they can be properly implemented in the FEM software COMSOL [105]. Note the Boltzman constant $k_B$ is assumed to be 1 J/K and the spin Peltier effect are neglected in this chapter.

![1D Domain](image)

Figure 7.1: 1D domain (green) with the definitions of axis direction (black) and boundary normal vector (red).

Considering a 1D geometry (YIG) $(0 < x < L)$ as shown in Fig. 7.1, the Boltzmann transport theory for thermal and condensed magnon reads [30]

$$\partial \hat{n}_x + \nabla \cdot \hat{j}_x = \frac{n_c \hbar \omega - \mu}{\tau_{ex} T_m} - g_{np} \mu - g_{nT} (T_m - T_p) \quad (7.1)$$

where $n_x$ and $n_c$ are the equilibrium thermal and condensed magnon density, $j_x$ is the thermal magnon spin current, $\mu$ is the thermal magnon chemical potential, $\tau_{ex}$ is the
condensate-thermal magnon scattering time, \( g_{n\mu} \) and \( g_{nT} \) describe the relaxation of \( \mu \) to the phononic environment and \( T_m \) and \( T_p \) are the magnon and phonon temperature respectively.

Since the condensed magnons are not considered in this section, the term associated with the condensed magnons \( \frac{n_c \hbar \omega - \mu}{\tau_{c\mu} T_m} \) is omitted. In addition, since the equation is solved under the steady-state condition, the time derivative term \( \partial \dot{n}_x \) can also be neglected. The simplified Eqn. (7.1) reads

\[
\nabla \cdot \mathbf{j}_x = -g_{n\mu}\mu - g_{nT}(T_m - T_p) \tag{7.2}
\]

At relatively high temperatures, the inelastic magnon-phonon scattering length (\( \sim \text{nm} \)) is shorter than other relevant length scales (magnon diffusion length \( \sim \mu\text{m} \) in YIG). Therefore magnons and phonons are in local thermal equilibrium \( (T_m \approx T_p) \) and \( T \) is used to represent \( T_m \) and \( T_p \) in this section. In this case Eqn. (7.2) can be rewritten as

\[
\nabla \cdot \mathbf{j}_x = -g_{n\mu}\mu \tag{7.3}
\]

The thermal magnon spin current, \( j_x \), can be expanded as \( j_x = -\sigma \nabla \mu - \varsigma \nabla T \). In this section, the magnon chemical potential and magnon spin current under a linear temperature profile are firstly examined. Under the linear temperature profile, \( \nabla^2 T = 0 \). After substituting the expansion of \( j_x \) into Eqn. (7.3) with \( \nabla^2 T = 0 \), we get

\[
\nabla^2 \mu - \frac{g_{n\mu}}{\sigma} \mu = 0 \tag{7.4}
\]
The effect of the applied temperature gradient $\nabla T$ and relaxation rate to phononic environment $g_{n\mu}$ on the magnon chemical potential and magnon spin current are shown in Fig. E.1 and E.2 in Appendix 7.

As for the boundary conditions, the thermal spin current flowing across the interface ($x = 0$) are given by [30]

$$\left. j_x \right|_{x=0} = \left[ G [\mu_{Pt} - \mu(0)] + S(T_e - T) \right]_{x=0} \hat{x} \quad (7.5)$$

where $\mu_{Pt}$ is the spin accumulation in Pt, $T_e$ is the electron temperature in Pt, $G$ is the interfacial spin conductance and $S$ is the interfacial spin Seebeck coefficient. Since there is no phonon Kapitza resistance or electron magnon thermal conductance is considered in this model, the interfacial electron temperature and magnon temperature are equal $T \approx T_e$ at the interface. Therefore the above boundary conditions Equation (7.5) can be simplified as

$$\left. j_x \right|_{x=0} = \left[ G [\mu_l - \mu(0)] \right]_{x=0} \hat{x} \quad (7.6)$$

The spin current at the boundary must satisfy the continuity condition. Therefore Eqn. (7.6) equals to the spin current in YIG in the close vicinity of the boundary $j_x(0) = -\sigma \nabla \mu - \varsigma \nabla T$. Therefore, we can rearrange the boundary condition in terms of the unknown, magnon chemical potential $\mu$, which leads to [30]

$$G(\mu_l - \mu(0))\hat{x} = -\sigma \nabla \mu - \varsigma \nabla T$$

$$-\nabla \mu = \frac{G}{\sigma} (\mu_l - \mu(0)) \hat{x} + \frac{\varsigma}{\sigma} \frac{T(L) - T(0)}{L} \hat{x} \quad (7.7)$$

Now since the boundary unit vector $\hat{n}$ always points outwards with respect to the domain of interest, the vector form of the boundary condition can be rewritten in the
scalar form at boundary $x = 0$ as follows

\[-\hat{n} \cdot \nabla \mu |_{x=0} = \left[ \frac{G}{\sigma} (\mu_l - \mu(0)) + \frac{\zeta}{\sigma} \frac{T(L) - T(0)}{L} \right] \hat{x} \cdot \hat{n} \]

\[-\hat{n} \cdot \nabla \mu |_{x=0} = -\frac{G}{\sigma} (\mu_l - \mu(0)) - \frac{\zeta}{\sigma} \frac{T(L) - T(0)}{L} \]  

(7.8)

The same process is used to derive the boundary condition at at $x = L$ which leads to

\[-\hat{n} \cdot \nabla \mu |_{x=L} = \frac{G}{\sigma} (\mu(L) - \mu_r) + \frac{\zeta}{\sigma} \frac{T(L) - T(0)}{L} \]  

(7.9)

Now, the partial differential equation (7.4) along with boundary condition equations (7.8) and (7.9) forms a complete equation set that describes the 1D steady-state thermal magnon chemical potential profile $\mu$ with the linear temperature profile as input. With appropriate input parameters, the steady-state magnon chemical potential $\mu$ and magnon spin current $j_x$ can be solved. The Cu/YIG/Cu structure is first considered here. Cu represents a poor spin sink and is assumed to have zero spin current flowing across the interface. Therefore the boundary conditions Eqn. (7.8) and (7.9) for the Cu/YIG interface at $x = 0$ and $x = L$ can be then simplified as

\[-\hat{n} \cdot \nabla \mu |_{x=0} = -\frac{\zeta}{\sigma} \frac{T(L) - T(0)}{L} \]  

(7.10a)

\[-\hat{n} \cdot \nabla \mu |_{x=L} = +\frac{\zeta}{\sigma} \frac{T(L) - T(0)}{L} \]  

(7.10b)

With Eqn. 7.4 and Eqn. 7.10, the magnon chemical potential $\mu$ and the different components of magnon spin current ($j_x = \sigma \nabla \mu - \zeta \nabla T$) in the 1D Cu(left boundary)/YIG($0 < x < 200\mu m$)/Cu(right boundary) structure are plotted in Fig. 7.2. The input parameters are calculated and discussed in Appendix E. Here, the
Figure 7.2: Magnon chemical potential $\mu$ and different components in the spin current, $j_x \propto \nabla \mu, j_x \propto \nabla T$ and total $j_x$ in a 1D Cu/YIG/Cu structure with linear temperature gradient of $\nabla T = 1 \text{ K/\mu m}$. YIG (0 to 200 $\mu$m) is sandwiched between the cold Cu side at $x = 0$ and the hot Cu side at $x = 200$. The applied temperature gradient is $\nabla T = 1 \text{ K/\mu m}$. With this temperature gradient, the magnon is driven out of thermal equilibrium. These nonequilibrium thermal magnons produce a magnon diffusive flow from the hot to cold end. At the hot end, a negative magnon chemical potential $\mu$ is established due to the depletion of the nonequilibrium magnons. On the contrary, the accumulation
of the nonequilibrium magnons results in a positive magnon chemical potential at the cold end. The magnon chemical potential is zero in the center region of YIG due to the absence of nonequilibrium magnons. The length scale of the deviation of the magnon chemical potential from zero are governed by the magnon spin diffusion length $\sqrt{\sigma/g_{\mu}}$ which is $\sim 10 \, \mu m$ in this case.

Figure 7.3: Comparison of the magnon chemical potential $\mu$ and different components in the spin current, $j_x \propto \nabla \mu$, $j_x \propto \nabla T$ and total $j_x$ in a 1D Cu/YIG/Cu and Pt/YIG/Pt structure with linear temperature gradient of $\nabla T = 1 \, K/\mu m$ at 300 K.
The total spin current consists of two components, \( j_x \propto \nabla \mu \) and \( j_x \propto \nabla T \). As for the spin current component \( j_x \propto \nabla \mu \), a positive nonzero \( j_x \propto \nabla \mu \) is observed in the region near the two boundaries within the spin diffusion length due to the depletion (accumulation) of magnons at the hot (cold) end. As for the spin current \( j_x \propto \nabla T \), a constant negative \( j_x \propto \nabla T \) is observed throughout the entire YIG due to the constant applied \( \nabla T \). Combining these two parts, the total spin current \( j_x \) gradually increases from zero at the Cu/YIG interface (due to the boundary condition) and saturates outside of the spin diffusion length.

Now, the Cu is switched into Pt and the magnon chemical potential and magnon spin current are solved in the Pt/YIG/Pt structure. Pt represents a good spin sink and is assumed to have aero spin accumulation at the boundary \((\mu_l = \mu_r = 0)\). The boundary conditions can be simplified as

\[
-\hat{n} \cdot \nabla \mu |_{x=0} = \frac{G}{\sigma} \mu(0) - \frac{\xi}{\sigma} \frac{T(L) - T(0)}{L}
\]

\[
-\hat{n} \cdot \nabla \mu |_{x=L} = \frac{G}{\sigma} \mu(L) + \frac{\xi}{\sigma} \frac{T(L) - T(0)}{L}
\]  

(7.11a)  
(7.11b)

Using the new boundary conditions for the Pt/YIG boundary, the magnon chemical potential and the different components of magnon spin current in the 1D Pt/YIG/Pt structure are solved and compared to the 1D Cu/YIG/Cu structure in Fig. 7.3. In order to clearly demonstrate the difference, only the data in the region close to the cold end are illustrated. Due to the spin sinking ability of Pt, less magnon accumulation is observed near \( x = 0 \) compared to the Cu/YIG boundary. This also applies to the hot end where less magnon depletion is observed at the Pt/YIG boundary. This results in a reduction in the spin current component \( j_x \propto \nabla \mu \) in the Pt/YIG/Pt structure. In addition, the Pt/YIG boundary condition allows a finite spin current flowing across the interface due to the difference in the magnon chemical potential in
YIG and Pt at the interface. This is directly contradictory to the zero spin current at the interface in the Cu/YIG/Cu structure. The net spin current flowing out of/into YIG at the cold/hot side at Pt/YIG boundaries can be converted into an electrical potential via the inverse spin Hall effect which can be measured as the spin Seebeck signal. This injection and detection of thermally induced magnons in the Pt/YIG structure are the physics behind the spin Seebeck effect.

### 7.2 1D steady-state thermal magnon transport under a arbitrary temperature profile

The sample structure and linear temperature profile discussed in the last section apply to the measurements where the YIG is heated with a conventional resistive heater on one side and is attached to a copper heat sink on the other side. However, the Pt/YIG used in the previous chapters is heated by a laser. Therefore, a more robust temperature profile is used as input to solve for the magnon chemical \( \mu \) potential and magnon spin current \( j_x \) in this section.

For the non-linear temperature profile, the \( \nabla^2 T \) term in the magnon spin current equation \( \nabla^2 j_x = -\sigma \nabla^2 \mu - \varsigma \nabla^2 T \) is not zero as discussed in the last section. Therefore, Eqn. 7.4 is rewritten as [30]

\[
\nabla^2 \mu - \frac{g_{\text{mu}}}{\sigma} \mu + \frac{\varsigma}{\sigma} \nabla^2 T = 0 \tag{7.12}
\]

The boundary conditions (7.8) and (7.9) are still valid with non-linear input temperature profile. Therefore the partial differential equation (7.12) along with boundary condition Eqn. (7.8) and (7.9) forms a complete equation set that describes the
1D steady-state magnon chemical potential profile $\mu$ with a non-linear temperature profile.

Figure 7.4: Magnon chemical potential profile $\mu$ (top panel) and the different components in the spin current, $j_x \propto \nabla \mu$, $j_x \propto \nabla T$ and total $j_x$ (bottom panel) induced by the exponential temperature profile in the 1D Cu/YIG/Pt.

In Fig. 7.4, the temperature profile $T$, magnon chemical potential $\mu$ and the different components of the magnon spin current in a 1D Cu(left boundary)/YIG(0 < $x < 200\mu m$)/Pt(right boundary) structure are plotted. The non-linear temperature

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profile shown as the green line in the top panel in Fig. 7.4 follows an exponential decay function $T \sim T(x = 200)e^{-(x-200)/200}$ with a characteristic decay length $\sim 20 \mu m$. The temperature drop is only significant near the hot Pt end on the right side while the majority of the sample remains close to the ambient temperature. The corresponding magnon chemical potential $\mu$ is shown as the black line in the top panel in Fig. 7.4. A similar magnon depletion as discussed in the last section can be observed in the close vicinity of the hot Pt/YIG boundary. An abnormal positive magnon chemical potential is found next to the magnon depletion region indicating the presence of magnon accumulation. This is due to the following reasons (i) $\nabla^2 T$ term in Eqn. 7.12 acts as a bulk magnon generation term (ii) the temperature decay length of (20 $\mu m$) is longer than the magnon spin diffusion length (10 $\mu m$). In the close vicinity of the hot Pt/YIG boundary, the depletion of magnons due to the relaxation to phonons dominates over the magnon generation leading to a negative magnon chemical potential. As moving away from the interface, the generation of magnons starts to take over from one spin diffusion length into YIG to the region where the sample temperature equilibrates with the ambient temperature.

The presence of the additional magnon generation leads to a sign switching in the magnon spin current component $j_x \propto \nabla \mu$ near one spin diffusion length away from the Pt/YIG boundary ($x = 200 \mu m$) as shown in the bottom panel in Fig. 7.4. The other spin current component $j_x \propto \nabla T$ simply follows the decay of the temperature profile. The combination effect of these two components on the total spin current $j_x$ shows an interesting feature where the total spin current peaks neither deep in the bulk nor at the boundary, but rather near one spin diffusion length away from boundary. The
magnon chemical potential and magnon spin current in a 2D Pt/YIG/Cu structure with laser heating are shown in Fig. E.3 in Appendix 7.

### 7.3 1D steady-state thermal magnon transport with the three temperature model under the laser heating condition

The assumptions of $T_m \approx T_p$ is not applicable at low temperatures when the magnon thermal mean free path becomes comparable with the magnon spin diffusion length. In this section, we separated the one temperature bath into three different temperature baths. These temperature baths describe the temperatures of the particles (electrons) and quasi-particles (phonons and magnons) in Pt and YIG. In this case, Eqn. 7.4 can be rewritten as [30]

$$\nabla^2 \mu - \frac{g_n \mu}{\sigma} \mu - \frac{g_n T}{\sigma} (T_m - T_p) + \frac{\varsigma}{\sigma} \nabla^2 T = 0 \quad (7.13)$$

The structure solved in this section consists of 10 $\mu$m Cu (left boundary) / 100 $\mu$m YIG/ 10 nm Pt (right boundary). The heat transport equations are firstly solved to determine the electron ($T_e$), phonon ($T_p$) and magnon ($T_m$) temperature profiles. The solved temperature profiles are shown in Fig. 7.5a. At the Pt/YIG interface, the phonon Kapitza resistance is given by $-\kappa_p \frac{dT_p}{dx} = \frac{1}{R_{th}^p} \Delta T_p$ and the interfacial electron magnon heat current is given by $-\kappa_m \frac{dT_m}{dx} = \frac{1}{R_{th}^m} \Delta T_m$. Here, $\kappa_p$ and $\kappa_m$ are the phonon and magnon thermal conductivity in YIG and $R_{th}^p$ and $R_{th}^m$ are the Kapitza resistance for phonons and the effective interfacial heat resistance for electrons and magnons. The values of these parameters are taken from Ref. [130, 80, 30, 93] and are listed in Appendix E. The phonon Kapitza resistance results in a step in $T_p$ at the interface while the magnon electron interfacial heat current brings $T_m$ close to $T_e$ at
the interface. The temperature exhibits mostly linear decrease away from the surface and finally reaches the ambient temperature at the Cu/YIG interface ($x = 0$).

The spin transport equations are evaluated to determine the magnon chemical potential and magnon spin current after solving the temperature profiles for electrons, phonons and magnons. In Fig. 7.5b, the magnon chemical potential $\mu$ in YIG is similar to the one shown in Fig. 7.2 where magnon depletion/accumulation is observed close to the hot/cold end. Due to the presence of the additional decay term $\frac{\sigma_{\text{mT}}}{\sigma}(T_m - T_p)$, the magnon chemical potential exhibits a more drastic change close to the interface. The corresponding fast decay in the magnon spin current close to the right Pt/YIG interface is shown in Fig. 7.5c (entire YIG) and d (zoom in on the Pt/YIG interfacial region). This fast decay in the magnon chemical potential and magnon spin current happens over $\sim$ nm near the interface much shorter than the decay induced by $\frac{\sigma_{\text{mT}}}{\sigma}\mu \sim \mu m$ in both $j_x \propto \nabla \mu$ and $j_x \propto \nabla T$ components. The fast decay in $j_x \propto \nabla \mu$ is a result of the additional strong relaxation mechanism due to the non-equilibrated phonons while the fast decay in $j_x \propto \nabla T$ is a direct result of the sharper $T_m$ profile near the interface induced by the interfacial magnon electron heat current. The length scale of this decay is governed by the magnon thermal mean free path which is $\sim 40$ nm at room temperature in YIG. Note the absolute temperature rise of 100 K is to exaggerate the influence of $\frac{\sigma_{\text{mT}}}{\sigma}(T_m - T_p)$. All parameters used in the simulation are taken at 300 K and are assumed to be temperature independent.

7.4 1D steady-state thermal and condensed magnon transport under a linear temperature profile

In Ref. [30], it is predicted that a magnon bath that possesses coherent precessions (magnon condensation) could exist under a constant thermal magnon pumping via the
Figure 7.5: Solutions to the three temperature based heat transport model and spin transport model in Cu/YIG/Pt structure at 300 K. (a) Electron $T_e$, phonon $T_p$ and magnon $T_m$ temperature close to the Pt/YIG interface. (b) Magnon chemical potential $\mu$ and the different components of spin current $j_x$ in the YIG. (d) Zoom in of the spin current components near the Pt/YIG interface.
spin Seebeck effect. This mechanism is explained in detail in Ref. [131, 30, 132]. In this section, we mainly focus on the transport properties of these condensed magnons and their interplay to the thermal magnons. The transport equations are solved under the steady-state condition with the assumption that \( T_m \approx T_p \) in YIG. With the presence of condensed magnons, the thermal magnon transport equation Eqn. 7.1 can be written as [30]

\[
-\sigma \nabla^2 \mu = \frac{n_c}{\tau_{cx}} \frac{\hbar \omega - \mu}{T} - \frac{g_{\text{ax}}}{\sigma} \mu + \varsigma \nabla^2 T \tag{7.14}
\]

where \( n_c \) is the equilibrium condensed magnon density, \( \tau_{cx} \) is the condensate-thermal magnon scattering time, \( \omega = \dot{\phi} \) is the condensate frequency (\( \phi \) being the precessional angle of the magnetization density in the x-y plane with an applied magnetic field in the z-direction) and \( \hbar \omega \) is the condensate energy. The equilibrium condensed magnon density as a function of magnetic field and temperate is calculated and shown in Appendix E. It can be seen that the magnon condensation can be destroyed by tuning the system to greater magnetic fields or higher temperatures.

For the condensed magnons, the transport equation reads [30]

\[
\nabla \cdot j_c = -\frac{n_c}{\tau_{cx}} \frac{\hbar \omega - \mu}{T} - 2\alpha \omega n_c \tag{7.15}
\]

where \( j_c \) is the condensate spin current which equals to \( j_c = n_c \nabla \cdot v_c \) (\( v_c = \frac{\hbar}{m} \frac{d\phi}{dx} \) is the condensate velocity), \( m \) is the effective magnon mass and \( \alpha \) is the Gilbert damping.

At thermal equilibrium, all physical quantities are uniform in space. Under this condition, the condensate magnon current velocity \( v_c = 0 \), condensate frequency \( \omega = 0 \), and normal magnon chemical potential \( \mu = 0 \). Under a small perturbation, the change in \( v_c \), \( \omega \) and \( \mu \) is assumed to be small. This allows us to implement the
linear response theory to linearize the above equation. The resulting equation reads

\[ n_c \nabla \cdot \mathbf{v}_c = -n_c \frac{\hbar \omega - \mu}{\tau_{cx} T} - 2\alpha \omega n_c \]  

(7.16)

Since the small perturbation assumption is used, the value of \( n_c \) with the presence of the external thermal bias is same to the equilibrium value. Therefore, the only unknown in the partial differential equation 7.16 is the condensate velocity \( \mathbf{v}_c \). This condensate velocity governs the condensed magnon spin current profile.

The boundary conditions for the condensate velocity are given by [30]. For the Pt/YIG/Pt structure, the zero spin accumulation boundary condition \( \mu_l = \mu_r = 0 \) still applies. Therefore the boundary conditions for the condensed magnons can be simplified as [30]

\[ \mathbf{v}_c(0) = -g^{\uparrow \downarrow} \omega / 2\pi s \]  

(7.17a)

\[ \mathbf{v}_c(L) = g^{\uparrow \downarrow} \omega / 2\pi s \]  

(7.17b)

where \( g^{\uparrow \downarrow} \) is the spin mixing conductance and \( s \) is the saturation magnetization of YIG,

For the Cu/YIG/Cu structure, the zero spin current boundary condition \( n_c \mathbf{v}_c + j_x = 0 \) still applies. Therefore the boundary conditions for the condensed magnons can be simplified as [30]

\[ \mathbf{v}_c(0) = \frac{1}{n_c} (\sigma \nabla \mu + \varsigma \nabla T) \]  

(7.18a)

\[ \mathbf{v}_c(L) = \frac{1}{n_c} (\sigma \nabla \mu + \varsigma \nabla T) \]  

(7.18b)
Figure 7.6: Coupled condensed and thermal magnon properties in 1D Cu/YIG/Cu structure with linear temperature profile. (a) Input temperature (b) thermal magnon chemical potential (c) condensate velocity (d) condensate velocity in YIG at 240 K with applied $\nabla T = 0.01 \text{ K/\mu m}$.
In order to solve the coupled condensed and thermal magnon transport equations in COMSOL, a third partial differential equation besides Eqn. 7.14 ($\mu$ is the unknown) and Eqn. 7.16 ($v_c$ is the unknown) is needed since the condensate frequency in the boundary conditions is also an unknown parameter. Considering the small perturbation assumption, the condensate frequency is a constant through YIG, the third equation to solve $\omega$ reads

$$\nabla \omega = 0 \quad (7.19)$$

Since Eqn. 7.16 for the condensate velocity is a first order partial differential equation, one of the two boundary conditions are redundant. Therefore the boundary condition for $\omega$ is rearranged using one of the two boundary conditions for $v_c$. For the Pt/YIG/Pt structure, this boundary condition can be rewritten as

$$\omega(L) = \frac{2\pi s v_c(L)}{g^{\uparrow \downarrow}} \quad (7.20)$$

while for the Cu/YIG/Cu structure, this boundary condition can be rewritten as

$$\omega(L) = \frac{1}{\hbar} \left[ \frac{v_c(L) 2\pi \hbar s}{g^{\uparrow \downarrow}} + \mu(L) + \frac{n_c v_c(L)}{G} \right] \quad (7.21)$$

Combing the partial differential equations for thermal magnons Eqn. 7.12 and for condensed magnons Eqn. 7.16 and 7.19 with the boundaries conditions Eqn. 7.8 for thermal magnons and Eqn. 7.21 and 7.18 for condensed magnons, the thermal magnon chemical potential, condensate velocity and condensate frequency can be solved in the 1D Cu/YIG/Cu structure with $\nabla T = 0.01 \, \mu m$. These results are shown in Fig. 7.6a-d. The key input parameters for the condensed magnon transport are $n_c = 1.4 \times 10^{26} \, m^{-3}$ (extracted from Fig. E.4 in the Appendix E) at 240 K with zero magnetic field and $\tau_{cx} = 1 \times 10^{-10} \, s$. The other input parameters for thermal magnon transport equation are listed in Appendix E. The thermal magnon chemical potential
Figure 7.7: Condensed and thermal magnon current. (a) Condensed, thermal magnon and total spin current in 1D Cu/YIG/Cu structure with linear temperature profile. (b) Comparison of the condensation current in 1D Cu/YIG/Cu and Pt/YIG/Pt structure under the same thermal bias.
\( \mu \) is similar to the one in Fig. 7.2 where the thermal magnon depletion/accumulation can be found at the hot/cold end. For the condensed magnons, the most obvious feature is their zero condensate frequency. This implies that the magnon spins are static canted along the magnetic field direction with a phase angle \( \phi \) [30]. At the cold end, the condensed magnon bath experiences positive damping (anti-damping) from the thermal magnon bath [131, 30]. In this case, the thermal magnon bath feeds the condensed magnon bath inducing a spatially changed condensate phase angle \( \phi \). This generates a positive condensate velocity gradient \( \nabla v_c \) and induces a finite condensate velocity in the bulk of YIG. At the hot end, the condensed magnon bath exhibits negative damping from the thermal magnon bath [131, 30]. In this case, the condensed magnon bath feeds the thermal magnon bath inducing an opposite spatial change of \( \phi \) compared to the cold end. This induces a negative condensate velocity gradient \( \nabla v_c \) which decreases the condensate velocity to \( \sim 0 \) at the cold end. The mechanism behind the damping and anti-damping of condensed magnons to thermal magnons is discussed in details in Ref. [131].

The spin current components due to the thermal magnon \( j_x \) and condensed magnon \( j_c \) and the total spin current \( j_{\text{total}} \) are shown in Fig. 7.7a. It can be seen that the thermal magnon spin current \( j_x \) profile resembles the one in Fig. 7.2 and 7.5 where no condensation is included. This is due to the similar magnon chemical potential profile in both cases. The condensed magnons form at the cold end due to the anti-damping to the thermal magnons while they evaporate at the hot end due to the damping to the thermal magnons. Therefore, a conveyor belt flow consisting of the condensed and thermal magnon spin current \( j_c \) and \( j_x \) is observed in Fig. 7.7a. This agrees well with the results in Ref. [30]. The overall spin current in YIG reduces
upon the formation of the magnon condensation due to the counter flow nature of the thermal and condensed magnon spin current. Note there is no spin current flowing out of the Cu/YIG boundary due to the zero spin current boundary condition. The comparison of the condensed magnon spin current $j_c$ in two different structures is shown in Fig. 7.7b. $j_c$ in the Cu/YIG/Cu structure is higher than the one in the Pt/YIG/Pt structure due to the larger thermal magnon accumulation at the cold end. The higher number of the non-equilibrium magnons enhances the anti-damping for the condensed magnons thus resulting in a higher condensate current density.

7.5 Design of the magnon Bose-Einstein experiment

In this section, the experimental design to probe the magnon Bose-Einstein condensation is proposed. Two measurable physical quantities associated with condensed magnons, i.e. total spin current in the z-direction $j_z$ and the condensate frequency $\omega$, are examined in Fig. 7.8 with different temperature gradients in different structures. The comparison of the total spin current in the 1D Pt/YIG/Cu structure with and without magnon condensation (this can be tuned by changing the magnetic field) at 240 K are shown in Fig. 7.8a. Upon forming the magnon condensation, the total spin current $j_x + j_c$ in the bulk exhibits a smaller value compared to the case without the magnon condensation. However the interfacial spin current that can be detected as an electrical voltage via the inverse spin Hall effect only shows slight difference at the interface for these two scenarios as shown in the inset in Fig. 7.8a. The absolute and percentage change of the interfacial spin current in the z-direction $j_z$ that can be collected by the the spin transducer and converted to a measurable electrical signal are shown in 7.8b as a function of applied thermal bias. Although the absolute change
in the total spin current $j_z$ can be scaled up by increasing the temperature gradient, its percentage change is found to be independent to the thermal driving force and remains at a low value $\sim 5\%$. Hyungyu et al. [29] and Kikkawa et al. [133] reported the suppression of the spin current in the spin Seebeck signal at high magnetic fields due to the change in the Zeeman energy. Since this effect could overwhelm the spin current reduction due to the formation of the magnon condensation, the detection of the condensed magnon by measuring the reduction of the total spin current is not feasible.

The other measurable physical quantity to probe the magnon condensation is the condensate frequency $\omega$. The change in the condensate frequency as a function of thermal gradient is shown in 7.8c. The condensate frequency can be tuned by the external thermal bias and is found to be in between the MHz to GHz range at around 240 K with a temperature gradient smaller than 0.1 K/µm. This coherent precession frequency can be picked up by putting an antenna on top of YIG. The coherent precession frequency and its dependence on external thermal gradient, magnetic field and sample temperature can be examined to study the presence/absence of the magnon condensation. A schematic diagram of a realistic sample structure consisting of different layers: Pt(pick up antenna)/YIG/Cu that can be used for this experiment is shown in Fig. E.5 in Appendix E.

7.6 Conclusions

In this chapter, the thermal magnon transport in YIG with a linear temperature is first examined in the Pt/YIG/Pt and Cu/YIG/Cu structures based on the finite-element method simulation results. The magnon chemical potential profile
Figure 7.8: (a) Comparison of the total spin current in YIG with and without magnon BEC by tuning the magnetic field in Pt/YIG/Cu structure. Inset: zoom in of the spin current at the interface. (b) The absolute and percentage change of spin current at Pt/YIG interface as a function of applied temperature gradient in Pt/YIG/Cu structure. (c) The condensate frequency as function of applied temperature gradient.
exhibits a negative/positive value at the hot/cold end indicating a magnon depl-
iton/accumulation in these regions. The characteristic decay length for these nonequi-
librium magnon to exist is the spin diffusion length. The thermal magnon spin cur-
rents due to $\nabla \mu$ and $\nabla T_m$ possess an opposite sign and the combined total thermal
magnon current can be detected using Pt in the Pt/YIG/Pt structure. When a non-
linear temperature profile is applied, the bulk magnon generation term due to $\nabla^2 T_m$
strongly affects the magnon chemical potential and magnon spin current profile. In
addition, when the temperatures of electrons, phonons and magnons are treated sep-
arately in a Pt/YIG/Cu structure with laser heating, the magnon chemical potential
and magnon spin current show multiple distinct decay regions in which their decay
length scale can be associated with the spin diffusion length and magnon thermal
mean-free path.

Besides the thermal magnon transport, Flebus et al. [30] also predicted that
the magnon condensation can be realized through constant thermal pumping via the
spin Seebeck effect in YIG. The coupled condensed and thermal magnon spin current
shows a conveyor belt flow pattern similar to the Ref. [30]. The condensate current
in the bulk of YIG is found to be higher in the Cu/YIG/Cu structure compared to
the Pt/YIG/Pt structure. In terms of designing an experiment to probe the magnon
condensation, measuring the total spin current at the Pt/YIG interface as a function of
magnetic field is not feasible. The measurement of the condensate frequency that can
be picked up by a Pt antenna in the Pt/YIG/Cu structure as a function of thermal
bias and sample temperature is more promising.
Chapter 8: Conclusions and Outlook

In this dissertation, multiple research topics ranging from the magnetically doped semiconductors to the transport of thermally induced magnons are covered. In terms of the magnetically doped semiconductors, ferromagnetic Gd doped AlN/GaN heterostructures grown by the molecular beam epitaxy are fabricated. Gd δ-doping at the AlN/GaN interface where the two dimensional electron gas forms is achieved to facilitate the study of the coupling between the conduction electrons and the Gd-induced ferromagnetic phase. The comparison between the magnetotransport and magnetization measurements suggests the lack of effective coupling between them. We speculated the Gd-associated defect-induced magnetism presents in large defect clusters which could locally deplete the electrons in the 2DEG as the reason for the absence of the coupling. This hypothesis is supported by the relatively high threading dislocation density and the spin locking along the c-axis observed in the Gd doped samples. In order to confirm this hypothesis, a structural study on the current samples to identify the location of the Gd dopants is needed. Furthermore, a study on a series Gd doped AlN/GaN samples with different threading dislocation densities near the interface should be performed. The correlation between the threading dislocation density and the magnetotransport measurement would verify or disprove the hypothesis.
In addition, we also synthesized Mn:Mo₂ on sapphire substrates using a CVD-like method like method. Structural studies suggest the few-layer and single crystal nature of these 2D Mn:MoS₂ samples. The magnetization measurements reveal that, unlike the theoretical prediction, such Mn doped few-layer MoS₂ samples exhibit mainly paramagnetism, which is indicative of the absence of the ferromagnetic coupling between Mn dopants. In the future, a detailed structural study on the location of the Mn dopants in these samples needs to be done since the theoretical prediction of the ferromagnetism in Mn:MoS₂ originates from the Mn dopants that substitute the Mo site. If the Mn dopants are found to be in large clusters or resides in interstitial sites in the MoS₂ lattice, the current growth method should be improved. Meanwhile, other magnetically doped 2D materials such as Mn:MoSe₂ are being studied in the aim of realizing a 2D dilute magnetic semiconductor.

In terms of the studies on the transport properties of thermally induced magnons in YIG, we first proposed a non-local optothermal spin Seebeck measurement configuration from which the magnon spin diffusion length is probed. Thermal simulation is used to verify the detection of the pure diffusion of the thermally induced magnons. By comparing the the decay of the nonlocal opto-thermal spin signal with the spin transport simulation, the spin diffusion length in YIG is found to be $47 < \lambda_s < 63 \mu\text{m}$ at 23 K. Currently, the new nonlocal spin signal decay profiles taken in a better measurement setup reveal a bi-exponential characteristic at lower temperatures and a single-exponential characteristic at higher temperatures. In the future, a detailed simulation study based on the heat and spin diffusion equations should be carried out to study the physical origin for these decay lengths.
The magnon dynamics are measured using the time-resolved longitudinal spin Seebeck effect. The time domain spin Seebeck waveforms contain a fast component corresponding to the interfacial electron magnon temperature difference and a slow component associating with the bulk magnon diffusion. The rise time for the fast component is found to be temperature independent while the rise time for the slow component gradually increases from low to high temperatures. The simulations based on the heat and spin transport equations suggest that the limiting factor for the speed of the slow rise should be attributed to the thermal (or spin) diffusion at higher (or lower) temperatures. Currently, more simulations at different temperatures need to be finished. The temperature dependence of the extracted simulation parameter, spin diffusivity, and its correlation to the slow rise component in the measured signal should be investigated. In the future, the time-domain longitudinal SSE should be performed on the Pt/YIG thin film samples at different temperatures. The current results predict that there will be no slow rise due to the bulk magnon diffusion in this samples due to their thin film nature.

In light of the experiments discussed above, a novel Nernst thermoelectric generator with a scalable power output is build using a Galfenol wire in a coiled wire geometry. The measured Nernst coefficient of Galfenol is the highest among the similar ferromagnetic materials. In the future, a thinner insulation material should be used to coat the Galfenol wire, which will create a larger temperature drop in the Galfenol wire. The output thermosvoltage can be boosted up by orders of magnetite if a few um insulation material is used. In addition, new materials with the large magnetostriction and remnant magnetization should be explored in order to make the Nernst coil generator feasible for practical applications.
At last, a complete simulation study on the thermal and condensed magnon transport in different structures with different conditions are discussed. An experimental design for realizing the magnon Bose-Einstein condensation is proposed at last.
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Appendix A: Processing Recipes for AlN/GaN High Electron Mobility Transistor

Ga droplet removal,

1. Dip the as-grown AlN/GaN HEMT in solution with HCl and DI H₂O (volume ratio 5:5) for 5 minutes. Dip the sample DI water for 5 mins.

Pre-cleaning step before processing,

3. Clean the sample in acetone bath with sonication for 5 minutes. Spray with methanol and IPA.

4. Dehydration bake in N₂ ambient at 150 °C for 5 minutes. Hall bar mesa definition,

5. Apply S1813 photoresist and soft bake at 115 °C for 1 minute (500 rpm/100 rpm (ramp) for 5 sec and 3000 rpm/5000 rpm (ramp) for 1 min.)

6. Expose using ALN2 (EVG 620) at NTW with exposure time of 2.4 secs. (Clean the mask with IPA after usage.)

7. Developed in MF319 for 1 minute. Clean off the developer leftover with water.

8. Hard bake on hot plate at 95 °C for 3 mins.

Hall bar mesa etch,
9. Etch using Plasma-Thermo ICP-RIE with either manual or batch mode. 
(Cl₂/Bcl₃/Ar : 10/1.2/1.2 sccm, RIE: 90 V (DC), ICP: 650 W, He: 10 mTorr, Chamber pressure: 5 mTorr.) The etch depth as a function of etch time of Kyma GaN and Gd doped AlN/GaN heterostructure are shown in Fig. A.1. The etch rates are \( \sim 21 \) nm and 18.8 nm for Kyma GaN and Gd doped AlN/GaN heterostructure respectively.

![Figure A.1: Etch rate of Kyma GaN and Gd doped GaN using Cl₂ based etching recipe.](image)

Ohmic contact fabrication:

10. Define the metal contact region using step 6-8.

11. Deposit metal stack of Ti/Al/Ni/Au or Ti/Al/Ti/Au with thickness of 20/150/30/50 nm or 20/170/5/50 nm.

12. Rapid thermal annealing of the metal stack at 800 °C (Ti/Al/Ni/Au) and 750C (for Ti/Al/Ti/Au) for 30 sec. (Delay: 600 sec. Ramp: 100 sec to 350 °C. Stay at 350 °C for 10 sec. Ramp: 100 sec to 850 °C (750 °C ). Stay at 850 °C (750 °C) for 30 sec.)
The effect of RTA of the Ti/Al/Ni/Au on AlN/GaN 2DEG device is shown in Fig. A.2. The IV characteristic is taken with two probe measurement and therefore the contact resistance is included. It can be seen that the overall resistance, i.e. contact resistance plus the 2DEG resistance, decreases as increasing the RTA annealing time at 800 °C. The inset is the zoom in of the data without any RTA annealing. The annealing recipe for Ti/Al/Ti/Au metal stack on AlN/GaN 2DEG device is determined using same methodology.

![Figure A.2: 2 probe IV measurements over Ti/Al/Ni/Au contacts on AlN/GaN heterostructure.](image-url)
Appendix B: The Corrected Functional Form of the Magneto-conductivity Tensor

In this appendix, we calculate a function form for $\delta \sigma_{xx}$. The fast decrease in magnetoresistance in the low field region $< 5$ kOe is a result of the weak-localization effect induced by the defects. The correction to $\sigma_{xx}$ due to this weak-localization effect is given by Ref. [134] and reads,

$$
\delta \sigma_{xx}^{WL} = \sigma_0 \left[ \psi\left(\frac{1}{2} + \frac{\hbar}{4eDB\tau_i}\right) - \psi\left(\frac{1}{2} + \frac{\hbar}{4eDB\tau_e}\right) + \ln\frac{\tau_i}{\tau_e} \right] \tag{B.1}
$$

where $\sigma_0 = e^2/2\pi^2\hbar$, $\psi$ is the digamma function, $D$ is the electron diffusion constant, $\tau_e$ and $\tau_i$ are the elastic and inelastic scattering rates, respectively. $D$ is calculated by $D = v_F^2\tau_e/2$ where $v_F$ is the Fermi velocity. We can then rewrite the conductivity tensor as $\sigma_{xx} = \frac{n_c\mu}{1 + \mu^2 B^2} + \delta \sigma_{xx}^{WL}$. In this case, after the matrix inversion, we find that the calculated $\rho_{xx}$ could fit the measured data in the low magnetic field region. However, the calculated $\rho_{xx}$ in high magnetic field region shows saturation rather than the linear decrease in the measured data. This implies that additional correction terms in $\sigma_{xx}$ need to be included.

As suggested in Ref. [134, 135], electron-electron scattering would result in a continuous reduction of magnetoresistance. Here, the detailed expression of $\delta \sigma_{xx}^{EEI}$ is
not evaluated here and a single value of $\delta\sigma_{xx}^{EEI}$ is used to account for the electron-electron scattering since it is predicted that the $\delta\sigma_{xx}^{EEI}$ is a magnetic field independent term when $k_B T \tau_e < 1$. Therefore, the conductivity tensor could be further rewritten as $\sigma_{xx} = \frac{-ne\mu}{1 + \mu^2 B^2} + \delta\sigma_{xx}^W + \delta\sigma_{xx}^{EEI}$. In this case, after the matrix inversion, we find that the calculated $\rho_{xx}$ exhibits a $B^2$ dependent parabolic decrease in the high magnetic field region. This is still different than the linearly decreasing $\rho_{xx}$ in the measurement. To achieve a better fitting between the calculated and measured $\rho_{xx}$, a second weak-localization correction term $\sigma_{xx}^{WL2}$ is added which indicates a second type of defects presented in the sample. The elastic scattering time $\tau_{e2}$ is much faster in $\sigma_{xx}^{WL2}$ than in $\sigma_{xx}^W$. This will modifies the curvature in the calculated $\rho_{xx}$ from a parabolic to a linear decrease in the high magnetic field region. The final form of $\sigma_{xx}$ and $\sigma_{xy}$ are given as follows

$$\sigma_{xx} = \frac{-ne\mu}{1 + \mu^2 B^2} + \sigma_{xx}^W + \sigma_{xx}^{EEI} + \sigma_{xx}^{WL2}, \quad (B.2a)$$
$$\sigma_{xy} = \frac{-ne\mu B^2}{1 + \mu^2 B^2}. \quad (B.2b)$$

The weak-localization effect and electron-electron scattering have no effect on $\sigma_{xy}$. Eqn. B.2 is converted to $\sigma_{xx}$ and $\sigma_{xy}$ through matrix inversion and then fitted to the measured $\sigma_{xx}$ and $\sigma_{xy}$. The variables $n$, $\mu$, $\sigma_{xx}^{EEI}$, $4eDB\tau_e$, $4eDB\tau_i$, $4eDB\tau_{e2}$ and $4eDB\tau_{i2}$ can be extracted from the least-squares fitting and $n$, $\mu$ is listed in tables in the main text. The discussion of $\sigma_{xx}^{EEI}$, $4eDB\tau_e$, $4eDB\tau_i$, $4eDB\tau_{e2}$ and $4eDB\tau_{i2}$ is beyond the scope of this paper and is not discussed here.
Appendix C: Processing Recipes for the Devices Used in the Nonlocal Opto-thermal SSE measurements

Pre-cleaning step before processing,

1. Clean the sample in acetone bath with weak sonication for 1 minute. Spray with methanol, IPA and DI water.

2. Dehydration bake on hot plate at 150 °C for 5 minute.

Spin detector fabrication, (this step is done with etching not lift off to keep a pristine Pt/YIG interface)

3. In-situ baking at 150 °C for 1 hour in LAB18.

4. Deposit Pt with 0.18 A/s (actual power may vary from 17% - 21% depending on the e-beam location and the amount of Pt material in the crucible) for 10 nm (or 6 nm).

5. Apply S1813 or S1805 photoresist and soft bake at 115 °C for 1 minute. (S1813: 500 rpm/100 rpm(ramp) for 5 sec and 3000 rpm/5000 rpm(ramp) for 1 minute.) (S1805: 6000 rpm/5000 rpm(ramp) for 30 sec and soft bake at 115 °C for 1 minute.)


7. Developed in MF319 for 1 minute. Clean off the developer leftover with water.
8. Etching Pt (10 nm) using (Cl₂/CF₄/Ar : 20/20/60 sccm, RIE: 100 V (DC),
IPC: 200 W He: 5 mTorr, Chamber pressure: 10 mTorr.) The etch rate is shown
in Fig. C.1. Etch time step (× 5 times): 20 sec(etch) plus 3 min(dwell for cooling
sample) with the last 6th step of 25 sec. For Pt (6nm) using the same recipe.

![Etch rate of Pt using Cl₂ based etching recipe.](image)

Figure C.1: Etch rate of Pt using Cl₂ based etching recipe.

9. Clean the sample in acetone bath with sonication for 1 min 45 sec. Spin injector
fabrication, (this step is done with life off)

10. Same as step 5, 6, 7 to define features.

11. Clean the PR left over using low power oxygen plasma in ASH01 at NTW.
(Power: 90%, oxygen: 25 sccm, duration: 30 sec.)

12. Same as step 4. (This also applies for different metals other than Pt.)

13. Lift off metal by sonicate the sample in acetone bath for 1-2 minutes (time
may vary depending on the thickness of the metal.)
Appendix D: Parameters Used in the Time Domain Heat and Spin Transport Simulations

Table D.1: Parameters used in the time domain heat and spin transport simulation

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value at 300 K</th>
<th>Temperature dependence</th>
</tr>
</thead>
<tbody>
<tr>
<td>pulse width</td>
<td>50 µs</td>
<td>-</td>
</tr>
<tr>
<td>laser power</td>
<td>14.7 mW</td>
<td>-</td>
</tr>
<tr>
<td>Pt reflectivity</td>
<td>97 % [136]</td>
<td>-</td>
</tr>
<tr>
<td>Pt absorption coefficient</td>
<td>2 × 10^7 1/m [89]</td>
<td>-</td>
</tr>
<tr>
<td>$R_{th}$</td>
<td>1/2.79 × 10^8 W/m²K [130]</td>
<td>[137, 138]</td>
</tr>
<tr>
<td>$R_m$</td>
<td>1/2.34 × 10^7 W/m²K [130]</td>
<td>-</td>
</tr>
<tr>
<td>$G_{ep}$</td>
<td>1.1 × 10^18 W/m³K [139]</td>
<td>[139]</td>
</tr>
<tr>
<td>$G_{pm}$</td>
<td>1 × 10^13 W/m³K [130, 93]</td>
<td>[130, 93]</td>
</tr>
<tr>
<td>$C_{Pt}$</td>
<td>130 J/kgK</td>
<td>-</td>
</tr>
<tr>
<td>$\rho_p$</td>
<td>5170 kg/m³ [93]</td>
<td>[93]</td>
</tr>
<tr>
<td>$\rho_p^{Pt}$</td>
<td>21450 kg/m³ [90]</td>
<td>[90]</td>
</tr>
<tr>
<td>$\kappa_p$</td>
<td>8 W/mK [90]</td>
<td>[90]</td>
</tr>
<tr>
<td>$\kappa_p^{Pt}$</td>
<td>64 W/mK [90]</td>
<td>[90]</td>
</tr>
<tr>
<td>$\kappa_p^{YIG}$</td>
<td>6 W/mK [93]</td>
<td>[93]</td>
</tr>
<tr>
<td>$\kappa_m^{YIG}$</td>
<td>0.3 W/mK [93]</td>
<td>[93]</td>
</tr>
<tr>
<td>$C_{p}$</td>
<td>570 J/kgK [93]</td>
<td>[93]</td>
</tr>
<tr>
<td>$C_{p}^{YIG}$</td>
<td>3.2 J/kgK [93]</td>
<td>[93]</td>
</tr>
<tr>
<td>$C_{p}^{YIG}$</td>
<td>130 J/kgK [91]</td>
<td>[91]</td>
</tr>
<tr>
<td>$C_{e}$</td>
<td>10 J/kgK [91]</td>
<td>[91]</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>2.3 × 10^{43} 1/Jsm [30, 80]</td>
<td>[30, 80]</td>
</tr>
<tr>
<td>$\zeta$</td>
<td>$k_B \sigma$</td>
<td>-</td>
</tr>
<tr>
<td>$G$</td>
<td>2 × 10^{48} 1/Jsm² [30, 130]</td>
<td>[30, 130]</td>
</tr>
<tr>
<td>$n_{e0}$</td>
<td>6.3 × 10^{26} 1/m³ [30]</td>
<td>[30]</td>
</tr>
</tbody>
</table>
Appendix E: Equilibrium Condensed Magnon Density and Parameters Used in the Coupled Thermal and Condensed Magnon Transport Equations

The effect of temperature gradient and relaxation rate to phononic environment are shown in Fig. E.1 and E.2.

Figure E.1: Effect of the applied temperature gradient $\nabla T$ on magnon chemical potential $\mu$ (top panel) and total spin current (bottom panel).
Figure E.2: Effect of the relaxation to phonon parameter $g_{nn \mu}$ on magnon chemical potential $\mu$ (top panel) and total spin current (bottom panel).

The 2D steady-state thermal magnon transport with arbitrary temperature profile is shown in Fig. E.3.

The equilibrium condensed magnon density can be calculated as based on the equations in Ref. [30] and the resulting equilibrium condensed magnon density as a function of magnetic field and temperature is shown in Fig. E.5.

The parameters used to solve the thermal and condensed magnon transport is calculated based on the equation given in Ref. [30]

$$\varsigma = \frac{T}{T_c} \frac{s^{2/3}l}{\hbar}$$  \hspace{1cm} (E.1)

$$\sigma = \frac{T}{T_c} \frac{s^{2/3}l}{\hbar}$$  \hspace{1cm} (E.2)
\[ g_{n\mu} = \frac{\alpha s (T/T_c)^{3/2}}{\hbar} \]  
(E.3)

\[ G = \frac{g_{\uparrow\downarrow}^{14}}{\pi \alpha s} g_{n\mu} \]  
(E.4)

Assuming \( T = 300 \, K \), these parameters can be calculated based on the material parameters for YIG [30]

\[ \zeta = \frac{300K \, (10nm^{-3})^{2/3} \times 1\mu m}{560K \, 1.054 \times 10^{-34}Js} \]
\[ = 5.1 \times 10^{46} \, J^{-1} \, s^{-1} \, m^{-1} \]

\[ \sigma = \frac{300K \, (10nm^{-3})^{2/3} \times 1\mu m}{560K \, 1.054 \times 10^{-34}Js} \]
\[ = 5.1 \times 10^{46} \, J^{-1} \, s^{-1} \, m^{-1} \]

\[ g_{n\mu} = \frac{10^{-4} \times 10nm^{-3} \times (300K/560K)^{3/2}}{1.054 \times 10^{-34}Js} \]
\[ = 3.72 \times 10^{57} \, J^{-1} \, s^{-1} \, m^{-3} \]

\[ G = \frac{1.3 \times 10^{18}m^{-2}}{\pi \times 10^{-4} \times 10nm^{-3}} \times 3.72 \times 10^{57} \, J^{-1} \, s^{-1} \, m^{-3} \]
\[ = 1.54 \times 10^{51} \, J^{-1} \, s^{-1} \, m^{-2} \]

The variables in the partial differential equation and boundary conditions are defined as follows
\( n_x \) normal magnon density
\( j_x \) spin current density
\( n_c \) condensed magnon density
\( \tau_{cx} \) collision time between condensed and thermal magnon
\( \mu \) chemical potential of the thermal cloud
\( T \) magnon temperature
\( T_p \) phonon temperature
\( g \) parameterize relaxation of magnon by phononic environment
\( \sigma \) bulk spin conductivity
\( \varsigma \) intrinsic spin Seebeck coefficient
\( T_l \) electron temperature
\( G \) interfacial magnon spin conductance
\( S \) spin Seebeck coefficient
\( L \) length of YIG

The constants used to calculate the variables are as follows
\( T_c \) Curie temperature, 560 K for YIG
\( s \) saturation spin density, 10 \( nm^{-3} \) for YIG
\( \alpha \) Gilbert damping, \( 10^{-4} \) for YIG
\( l \) energy independent scattering length, \( \sim \mu m \) for YIG
\( g^{\uparrow\downarrow} \) spin mixing conductance, \( 1.3 \times 10^{18} m^{-2} \) for YIG
Figure E.3: Joule heating induced (a) temperature $T$ (b) magnon chemical potential $\mu$ and different spin current components (c) $j_y \propto \nabla T$ (d) $j_y \propto \nabla \mu$ (e) total $j_y$ profile in the x-y plane. Note, x and y direction represents the length and depth respectively.
Figure E.4: Equilibrium condensed magnon density as a function of applied magnetic filed and temperature.
Figure E.5: (a) Schematic of the sample geometry to pick up magnon condensation frequency. (b) Temperature $T$ (c) magnon chemical potential $\mu$ and (d) condensate frequency $\omega$ in the x-z plane in Pt/YIG/Cu structure.