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Heat, Charge and Spin Transport of Thin Film Nanostructures

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HEAT, CHARGE AND SPIN TRANSPORT OF THIN FILM NANOSTRUCTURES

A Dissertation
Presented to
the Faculty of Natural Sciences and Mathematics
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In Partial Fulfillment
of the Requirements for the Degree
Doctor of Philosophy

by
Devin Wesenberg
March 2018
Advisor: Professor Barry L. Zink
Abstract

Understanding of fundamental physics of transport properties in thin film nanostructures is crucial for application in spintronic, spin caloritronics and thermoelectric applications. Much of the difficulty in the understanding stems from the measurement itself. In this dissertation I present our thermal isolation platform that is primarily used for detection of thermally induced effects in a wide variety of materials. We can accurately and precisely produce in-plane thermal gradients in these membranes, allowing for thin film measurements on 2-D structures. First, we look at thermoelectric enhancements of doped semiconducting single-walled carbon nanotube thin films. We use the Wiedemann-Franz law to calculate contributions to thermal conductivity and find interesting underlying physics as we dope the films, thus changing the Fermi level. Adapting the tube diameter leads to structural differences, which greatly affects both phonon and electron contributions to thermal conductivity. These unique films can be designed as thermoelectric materials that are easy to manufacture and can be utilized in a variety of situations. Second, we look
at work measuring enhanced contributions to thermopower and thermal conductivity of unique ferromagnetic metals. We observe improved thermopower due to the ultra-low damping of the magnon system. For spintronic and spin caloritronic applications, having a low damping is important for device engineering and allows for long spin lifetimes. Third, we present on spin transport through disordered magnetic insulators. We observe spin Hall effect driven magnon transport through materials with no long-range order but with local antiferromagnetic exchange interactions. We are the first to observe this type of transport, which may lead spintronic investigations in a new and profound direction. Finally, we look at transverse effects in a thin ferromagnetic metal. Our observation of the planer Nernst effect and planar Hall effect across long length scales shows that effects in this range are dominated by traditional magneto-thermoelectric effects without any evidence of spin transport. A careful understanding of thermal and electric gradients is needed to aid in understanding of transport properties of thin films.
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Chapter 1

Transport and Effects

The physics of transport properties in materials is a key component in understanding the fundamental aspects of a material. It is also important in device development. This section will explore the main aspects of electrical, thermal and spin transport in a variety of thin film materials.

1.1 Electrical Transport

Charge carrier transport in a material is a highly studied phenomenon within solid state physics. Charge carriers can typically be described as electrons ($n$) or holes ($p$) and can be defined in terms of electrical conductivity, $\sigma$, or electrical resistivity, $\rho$. Carriers can move via an applied external voltage, or through diffusion.

One of the earliest methods to describe electrical conduction is the Drude model, proposed in 1900.[7] The model describes electrons as point sources with no local interaction. It is a simple model that obeys classical physics of kinetic theory that
is applicable to simple materials, namely metals. The Drude model defines a linear relationship between current density, $J$, and electric field, $E$,

$$ J = \sigma E, $$

(1.1.1)

with

$$ \sigma = \frac{nq^2\tau}{m} = \frac{nq^2l}{m_e v_F}, $$

(1.1.2)

where $n$ is the number density, $q$ is the electronic charge, $\tau$ is the mean free time between collisions, $m$ is the mass, and $v_F$ is the Fermi velocity. The mean free path, $l$, is defined as:

$$ l = v_F \tau. $$

(1.1.3)

The model does an acceptable job of estimating $\sigma$ of monovalent metals. The free electron gas, or Drude-Sommerfeld, model is an extension of the Drude model and takes into account Fermi-Dirac statistics and forms the basis of valence electron behavior.

Current understanding of electrical conductivity comes from band theory, which according to quantum mechanics claims that electrons occupy discrete energy levels. Electrons move to minimize the total energy by occupying the lowest available state, but are unable to sit in the same quantum state due to the Pauli exclusion principle. The level to which the electrons fill is called the Fermi level. The Fermi level forms the basis of electrical conduction as only the electrons near the Fermi level are available to move around. Electrical transport in thin films can vary widely compared to
bulk,[8] which is due to increased scattering events. Scattering is mainly due to impurities and phonon-electron scattering.

1.2 Thermal Conductivity

The thermal conductivity, $k$, of a material describes its ability to conduct heat. Thermal conductivity in steady state is defined as,

$$k = -\frac{\bar{Q}}{\nabla T},$$

(1.2.1)

where $\bar{Q}$ is the heat flux and $\nabla T$ is a temperature gradient. As we will see in the following chapters, thermal conductivity can vary widely depending on the material. Metals tend to have a high thermal conductivity compared to semiconductors or insulators. Total thermal conductivity can come from three contributions: phonons (lattice vibrations), electrons or magnons (spin waves). Thermal conductivity in insulators mainly stems from phonons, since electrons cannot flow. Semiconductors see contribution from all three. Metals are typically dominated by electrical mobility. Magnons typically contribute little but special materials see a significant contribution to thermal properties.

Kinetic theory of gases defines,

$$k = \frac{1}{3} C_v lv,$$

(1.2.2)
where $C_v$ is the heat capacity, $l$ is the mean free path and $v$ is the speed of sound in the material. To understand $k$’s expected temperature dependence we can look at each of its parts. Heat capacity is the materials ability to absorb energy. Individual atom vibrations are not independent, instead they can form quantized waves called phonons. Phonon information is defined by its frequency and wavelength. $C_v$ for phonons typically drops to zero at 0K with a $T^3$ dependence at low $T$ from the Debye model. $C_v$ is constant at $T$ above the Debye temperature, $\theta_D$, which is the temperature at which the crystal has its highest normal mode of vibration. The mean free path of a material defines the average distance traveled between collisions. Phonons can scatter off defects, boundaries and other phonons, which all decrease $l$. At low $T$ phonon mean free paths are long and mainly limited by the boundary. At high $T$ the phonon mean free path scales with a $1/T^x(x = 1 − 2)$ dependence due to phonon-phonon scattering. Combined, the phonon thermal conductivity typically takes the form of low at low $T$ and low at high $T$ with a peak in the middle. The electron contribution to $C_v$ has a $T$ dependence at low temperatures and constant at high $T$.

### 1.3 Thermopower

The thermoelectric effect, or the Seebeck effect, was discovered by Thomas Seebeck in 1821\cite{9} is the generation of an electric field in response to a temperature gradient, which is an intrinsic property in a material. The thermopower ($\alpha$), or Seebeck coefficient ($S$), gives the magnitude of the effect. Materials with high Seebeck coefficients
are useful when designing thermoelectric generators or coolers. The Seebeck coefficient can be given as

$$\Delta V = \int_{T_0}^{T} SdT.$$  \hspace{1cm} (1.3.1)

The thermopower in metals can be described as a logarithmic derivative of the electrical conductivity, $\sigma$, as

$$S = \frac{\pi^2 k_B^2 T}{3e} \left[ \frac{\partial \log \sigma(E)}{\partial E} \right]_{E=E_f},$$  \hspace{1cm} (1.3.2)

where $T$, $e$, $\sigma$, and $E_f$ are the temperature, electron charge, electrical conductivity and Fermi energy, respectively. The log portion of Eq. 1.3.2 is defined as

$$\frac{\partial \ln \sigma(E)}{\partial E} = \frac{\partial A}{\partial E} + \frac{\partial l}{\partial E},$$  \hspace{1cm} (1.3.3)

where $A$ is the area of the Fermi surface and $l$ is the mean free path of the charge carrier (electron or hole). The equations make up the Mott formula.[10, 11, 12]. Using this equation we can relate the changes in electrical conductivity with changes in energy at the Fermi level. This can be sensitive to the changes in the number of available scattering centers and also to the Fermi surface shape. For insulators and semiconductors, the charge carrier density of states is smaller than the density of thermally available states. This can lead to larger thermally driven charge carrier mobility above the Fermi surface increasing the thermopower. For metals those same energy states near the Fermi energy are filled, which leads to lower thermopowers.
The sign of $S$ can play a large role in detailing the dominate charge carrier of a material. Electrons diffuse from hot to cold, resulting in the cold side being negative. The sign of the thermopower is determined by the balancing of Eq. 1.3.3. For metals, electrons have higher energy so tend to scatter less, which results in a long average mean free path. This leads the second term in Eq. 1.3.3 to be positive. The first term will depend on the interaction between Fermi surface and the first Brillouin zone (FBZ). If the Fermi surface increases towards the FBZ, the area increases leading to a positive term. Once past the FBZ, the are decreases leading to a negative term. For semiconductors these terms can be greatly affected by doping concentration. In general, n-doping leads to overall negative thermopower and p-doping leads to positive thermopower.

1.4 Lorenz Number and Wiedemann-Franz Law

The Wiedemann-Franz law directly relates the electrons mobility to the thermal conductivity in a material using

$$\frac{k}{\sigma} = LT,$$

where $L$ is the Lorenz number. It was originally thought that the Lorenz number was a constant only applicable to metals.[13] To derive the law, we start with Eq. 1.2.2. If the electrons are treated as a Fermi gas, the heat capacity takes the form

$$C = \frac{1}{3} \pi^2 D(E_F) k_B^2 T.$$
We can then substitute $D(E_F) = 3N/(2k_BT_F)$, where $T_F$ is the Fermi temperature, for the density of states which gives

$$C = \frac{1}{2}\pi^2 nk_B T T_F.$$  \(1.4.3\)

Relating the Fermi energy to the Fermi temperature gives $k_BT_F = \frac{1}{2}mv^2_F$, which can be substituted into Eq. 1.4.3. The thermal conductivity then gives the form

$$k = \frac{1}{3}\pi^2 nk_B^2 T m^{-1} \tau.$$ \(1.4.4\)

We can then equation 1.1.2 and substitute that in our thermal conductivity equation 1.4.4 to yield

$$\frac{k}{\sigma} = \left[\frac{\pi^2}{3} \left(\frac{k_B}{e}\right)^2\right] T.$$ \(1.4.5\)

The quantity in the bracket of equation 1.4.5 is a constant known as the Sommerfeld theory for free electrons, $L_0 = 2.44 \times 10^{-8} \text{W}\Omega\text{K}^{-2}$. This value can be used to provide an estimate of the electron contribution to the thermal conductivity, such as $k_e = \sigma L_0 T$.

### 1.5 Spin Transport in Metals and Insulators

Spin transport, or spintronics, deals with the study of how the electron’s spin interacts in a material. The device applications are numerous, including tunnel magnetoresistance (TMR) and giant magnetoresistance (GMR) for magnetic read
heads, spin-transfer torque for magnetoresistive random-access memory, spin-wave logic devices, and many more.

Spin is the intrinsic angular momentum of an electron entirely separate from the orbital motion angular momentum. The magnitude can be defined as \( \frac{1}{2} \hbar \) along an arbitrary axis. Spin is a Fermion as described by Fermi-Dirac statistics and obeys the Pauli exclusion principle. For systems described here the spin in a material can act in sync with neighboring spins through both local and long range interactions and can greatly affect magnetic, thermal and electronic properties in the material.

1.5.1 Landau-Lifshitz-Gilbert Equation

Magnonics is the study of spin waves in a material and forms the bases for understanding spin interactions. Spin waves are able to propagate through magnetic materials but the frequency of precession can vary greatly in different materials. To understand the spin wave phenomenon we’ll first take a look at the Landau-Lifshitz-Gilbert (LLG) equation\(^{[14, 15]}\) of the precessional motion of magnetization, \( M \),

\[
(1 + \alpha^2_{\text{gil}}) \frac{dM}{dt} = -\gamma_0 M \times H_{\text{eff}} - \gamma_0 \frac{\alpha_{\text{gil}}}{M_s} M \times (M \times H_{\text{eff}}),
\]

(1.5.1)

where

\[
\gamma = \gamma_0 / (1 + \alpha^2_{\text{gil}}).
\]

(1.5.2)

The gyromagnetic ratio, \( \gamma = g|e|/2m_e \), where \( g \) is the g-factor (\( g = 2 \) for free electrons), \( e \) is the electron charge and \( m_e \) is the electron mass. \( \gamma \) is from the original LL equation and is smaller than \( \gamma_0 \) from the LLG equation by a small factor relating
to $\alpha_{\text{gil}}$. $H_{\text{eff}}$ is the effective field, a combination of the external magnetic field, the demagnetizing field, and other quantum mechanical effects, $\alpha_{\text{gil}}$ is a dimensionless constant called the damping factor,[16] and $M_s$ is the saturation magnetization. The first term, $\mathbf{M} \times \mathbf{H}_{\text{eff}}$, describes the magnetization precession around the effective magnetic field, $\mathbf{H}_{\text{eff}}$. The double vector product second term describes the damping processes relaxation to a minimum energy parallel to the effective field as well as state that the magnetization magnitude should remain constant.

Here we focus on $\alpha_{\text{gil}}$, the Gilbert damping constant, which describes the dissipation speed of magnetic procession in a material. This parameter is quite important as it directly impacts the magnetic energy loss of a system. The rate of remagnetization can lead some materials useful or useless for spintronic applications. In systems that require charge flow metals are often used, but these typically have high damping due to magnon-electron scattering in the conduction band, which increases the damping. Recent work by groups have found alloys that achieve low damping however,[2] which forms the bases of our work in Chapter 4. If a charge current is not required an often studied material is yttrium iron garnet (YIG), a ferrimagnetic insulator, due to it’s ultra low damping. The Gilbert damping constant can be experimentally determined using broadband ferromagnetic resonance (FMR) spectroscopy. FMR works by exciting a precession in the magnetization of the material by an applied external field, which exerts a torque on the sample. A transverse RF field is applied to the sample concurrently. Once the RF and magnetization precession coincide, the samples absorbs the energy and the dynamics can be measured.
The measured linewidth of the sample is directly related to the Gilbert damping parameter.

### 1.5.2 Spin Currents

Once a low magnetic damping material is achieved there are a variety of methods to create a spin current to transfer spin information. One of these is spin pumping,[17, 18, 19] which happens when a precessing magnetization in a ferromagnet is injected into an adjacent normal metal via a spin current. The inverse can also happen, where a spin polarized current in a normal metal exerts a torque on an adjacent ferromagnet, which is called spin-transfer torque.[20] Spin current can also be generated in ferromagnets by driving a charge current through the material, which creates a spin accumulation due to the difference in chemical potential of up and down spins.[21]

#### Spin Hall Effect

The spin Hall effect (SHE) is a popular method to generate a spin polarized current in a normal metal. The SHE consists of a spin accumulation at the lateral boundaries of a current carrying electrical conductor.[22] The direction of spin polarization obeys the right hand rule. This accumulation on the boundaries is due to spin orbit interactions, which couples spin and charge currents. Materials with high spin orbit coupling (SOC) make good SHE materials. On the contrary, if a polarized spin current is injected into a high SOC material a lateral voltage will be generated, which is called the inverse spin Hall effect (ISHE).[23]
1.6 Summary

In this chapter, we introduced transport properties of thin films. We discussed both electrical and thermal conductivity in metals. We showed how the Wiedemann-Franz law can be used to determine the electron, phonon, or magnon contributions to thermal conductivity and how the thermopower can be affected in different materials. We wrapped up with discussion of spin transport through metals and insulators. In the following chapter, our strategy for performing thermal and charge transport measurements are detailed.
Chapter 2

Thermal Isolation Platforms and Measurement Technique

This chapter discusses experimental techniques for measuring thermal conductivity, electrical conductivity, thermopower and spin transport. Our method of using a-Si-N thermal isolation platforms is detailed.

2.1 Introduction

Thermal and electrical transport properties of thin films can differ greatly from bulk values and are typically smaller. Compared to bulk, thin film structures tend to have more impurities, grain boundaries and disorder due to fabrication techniques that can all lower the thermal and electrical conductivity. Boundary scattering and phonon leakage are two additional factors that can contribute to lowered thermal conductivity in thin films. There are a variety of measurement techniques available
that each have advantages and disadvantages, which includes the $3\omega$ method,[24, 25] laser thermoreflectance,[26, 27, 28, 29] and Raman methods.[30, 31, 32]

$3\omega$ method can be both an optical or electrothermal technique, which can be performed in both cross-plane and in-plane. The $3\omega$ method utilizes a metal wire on top of a sample and substrate. An AC current of angular frequency $\omega = 2\pi f$ is applied to the wire, which in turn heats the sample at $2\omega$ frequency. The temperature amplitude generated will depend on the material properties. The temperature oscillations then follow the heater oscillations with a phase lag. The current driven at frequency $\omega$ leads to a resistance change of $2\omega$ which allows for detection of a RMS voltage at $3\omega$. Transport properties can then be extracted from the $3\omega$ signal. This technique has complications though, and is primarily used as a cross-plane detection technique. Issues then arise with the addition of a substrate background signal. The substrate background can be measured separately and subtracted off but this can lead to additional error due to the calculations being approximations. In-plane $3\omega$ measurements are less common and can have sensitivity issues.

The laser thermoreflectance method is another method for measuring thermal conductivity of thin films. This method relies on both the reflective properties of a surface and the induced thermal stress as it is heated. A pulse laser is used to generate acoustic heat waves and a probe laser can detect the reflecting waves through the piezo-optic effect. The data obtained is put through a thermal model, where the thermal conductivity and thermal conductance can be deduced. Laser thermoreflectance does allow for separation of thin film and background substrate, which is an advantage over the $3\omega$ method, but can also have sensitivity errors from
the phases as well as laser noise. Another technique is the Raman shift method, which is an indirect method that relies on a focused laser that thermally excites the sample and undergoes Raman scattering. The phonon energies of the sample can give the temperature profile. The thermal conductivity is then calculated from analytical or numerical models of heat diffusion. Its accuracy depends on the intensity of the Raman signal, which can depend on the material.

Here we present a direct method to measure thermal and electrical conductivity as well as thermopower of thin films. We make use of a 500 nm thick amorphous silicon nitride (a-Si-N) bridge suspended between two a-Si-N islands for accurate measurements of $k$, $\sigma$ and $\alpha$ all on the same film.[33, 34, 35, 36, 37]

### 2.2 Device Manufacture

All platforms are fabricated at the Center for Integrated Technology (CINT) in Albuquerque, New Mexico. Processing starts with a roughly 500 µm thick 100 mm $<100>$ single-side polished silicon wafer. A low stress (0-150 MPa) amorphous silicon nitride (a-Si-N) layer is deposited on the wafer via low pressure chemical vapor deposition (LPCVD).

Fig. 2.1 shows the layer processing of the material stack. A low RF plasma asher is used to help prep the surface and remove any organics before deposition. Patterns are constructed using photolithography. First, a 3-5 micron layer of negative photoresist (nLOF-2070) is spin coated on the Si-N coated wafer with a pre and post exposure bake. The coated side is covered in a 5” Soda Lime or Quartz mask that has been
Figure 2.1: Steps for fabrication process of $\alpha$-Si-N membranes thermal isolation platforms for thermal conductivity, electrical conductivity and thermopower measurements.
pre-patterned to construct our heaters and thermometers. Once the resist has been exposed to high intensity UV light and developed to expose the Si-N underneath, metal is deposited to form our structures. Electron beam evaporation (pressure \( \leq 1 \times 10^{-5} \) torr) is used to deposit a 10 nm chromium (Cr) sticking layer under 40 nm platinum (Pt). Metal liftoff in acetone is used to remove resist post deposition. After liftoff, another photoresist layer is used to help pattern our membrane structures. A positive photoresist (AZ-4330) is used with a pre-bake before exposure. A reactive ion etcher (RIE) with inductively coupled plasma (ICP) is to etch the a-Si-N to form the platforms. Once platforms are formed the entire wafer is inserted into a tetramethylammonium hydroxide (TMAH) wet chemical bath to etch away the exposed Si substrate and suspend the platforms. At 95°C <100> Si etches at approximately one micron per minute in the <111> plane. Standard samples were left in chemical bath for 360 minutes to fully suspend platforms. An image of platforms that have not been fully suspended is shown in Fig. 2.2. A complete platform device can be seen in Fig. 2.3. Length of bridge is 2050 µm, width is 88 µm and thickness is 500 nm. A close up of an island can be seen in Fig. 2.4. Here was can see the leads used as heaters, thermometers as well as two leads for \( \sigma \) and \( \alpha \) measurements.
Figure 2.2: Optical image of an $a$-Si-N test membrane that has not been fully released. Pink indicates fully released Si-N membrane. Green on the boarders and center of islands indicates Si-N with Si substrate underneath.
Figure 2.3: SEM image of $\alpha$-Si-N thermal isolation platform. White indicates Pt leads.
Figure 2.4: SEM image of right island of $\alpha$-Si-N thermal isolation with Pt leads. Two leads for $\sigma$ and $\alpha$ are present, one inner lead and one triangle lead.
2.3 Device Preparation and Measurement

To measure a film, a machined aluminum mask is fit over a thermal isolation platform leaving the bridge exposed. The mask is large enough for film to make contact on inner leads on each island for electrical conductivity and thermopower measurements. The devices are placed in a gold plated copper sample mount and wire bonded with Al-Si(1%) bond wire to make electrical connection with room temperature electronics. The sample mount is capped with a radiation shield to prevent radiative heating and mounted to the cold stage of a sample-in-vacuum cryostat. Measurements are made in a liquid nitrogen cryostat under vacuum at or below $1 \times 10^{-5}$ mbar. Temperatures range from 77 to 400 K.

2.3.1 Thermal Conductivity Measurement

Thermal Conductivity of $a$-Si-N Bridge

The thermal isolation platforms are specifically designed to obey the steady state thermal model found in Fig. 2.5. Measurements begin by regulating the sample stage and device frame at $T_{ref}$. To calibrate the temperature, the resistance of each thermometer is measured using a four wire SIM921 AC Resistance Bridge while the sample stage is held at $T_{ref}$. The power dissipated in the thermometer is much lower than a nanowatt. The device frame thermometer, $T_0$, is very stable with a $\leq 3$ mK deviation throughout the temperature range.

In Fig. 2.6 we can see a calibration curve for one of the island thermometers. At each reference temperature after calibration one of the islands is heated by running
Figure 2.5: Thermal model of thermal isolation platforms for calculating thermal conductivity.

a current through the heater strip, which heats via joule heating allowing heat to flow down both the legs and the bridge to the other island. Once the island comes to equilibrium (<5 sec) the temperatures on the frame, hot island and cold island are measured via the resistances. The voltage of the applied current is also measured to determine power dissipated. This procedure is repeated for up to 11 heater currents, ranging from 0.2 µW to 60 µW. Fig. 2.7 shows a graph of temperature vs power applied for hot island, cold island and frame thermometer with reference temperature set to 78 K. As the heater power increases, the temperature of the hot island increase, the temperature of the cold island increases a small but measurable amount, and the frame thermometer remains at the reference temperature. The rate of heat flow can be written mathematically as
Figure 2.6: Example calibration of micromachined thermometer. Inset: SEM micrograph of Pt wires patterned as thermometer, heater as well as $\sigma$ and $\alpha$ leads.
\[ C_h \frac{\partial T_h}{\partial t} = -K_L(T_h - T_0) - K_B(T_h - T_s) + P_h, \]  
\[ C_s \frac{\partial T_h}{\partial t} = -K_L(T_s - T_0) - K_B(T_s - T_h) + P_s, \]

where \( T_0, T_s, \) and \( T_h \) are the temperatures on the frame, cold island and hot island, respectively. \( C_h, C_s, P_h, \) and \( P_s \) are the specific heats and power dissipated on hot and cold islands, respectively. \( K_L \) and \( K_B \) are the thermal conductance through the legs and through the bridge. Our measurements are performed in steady state, so the time dependent variable vanishes. We also do not apply power to cold island so that term also disappears. Above equations are then reduced to

\[ 0 = -K_L(T_h - T_0) - K_B(T_h - T_s) + P_h, \]  
\[ 0 = K_L(T_s - T_0) - K_B(T_s - T_h). \]

These can then be solved for \( T_h \) and \( T_s \)

\[ T_s = T_0 + \frac{(K_L + K_B)P_h}{(2K_B + K_L)K_L}, \]  
\[ T_s = T_0 + \frac{(K_B)P_h}{(2K_B + K_L)K_L}. \]

A straight line can be fitted to both \( T_h \) and \( T_s \) vs P, then \( K_B \) and \( K_L \) can be calculated from the slopes. A plot of the thermal conductance vs. temperature of a Si-N bridge can be seen in Fig. 2.8. Once we know \( K_B \), the thermal conductivity, \( k_{Si-N} \), of the Si-N can be determined via
Figure 2.7: Temperature vs. heater power at $T_{ref} = 78$ K for micromachined $a$-Si-N island. Red data ($T_H$) represents the thermometer temperature of the hot island. Blue data ($T_S$) represents the thermometer temperature of the cold island. Black data ($T_0$) represents the thermometer temperature of the frame. Hot and cold side slopes are used to determine $K_B$ and $K_L$. 
Figure 2.8: Thermal conductance vs. temperature of a Si-N membrane bridge.
\[ k_{Si-N} = \frac{K_B l}{w t}, \]  

(2.3.7)

where \( l, w, \) and \( t \) are the length, width, and thickness of the bridge, respectively. All of the geometry is known so the calculation is straightforward. Here we are measuring the total in-plane thermal conductance of our Si-N bridge. Fig. 2.8 shows thermal conductance and thermal conductivity vs temperature for a Si-N bridge. Here we see similar temperature dependence to other Si-N membranes grown via LPCVD.[33, 38] Our values are smaller here possibly due to a difference geometry which can affect the Si-N microstructure.

**Thermal Conductivity of Deposited Thin Films**

Once the thermal conductivity of the Si-N bridge is determined, a thin film can then be deposited on the bridge via e-beam evaporation or sputtering. The film will add a parallel thermal conductance path to the bridge which shows as an increased thermal conductance in our measurement. Here we measure the total thermal conductance, \( K'_B \), of both the Si-N bridge and film. Subtracting the background contribution from the Si-N yields the final total in-plane thermal conductance of our sample, \( K_{film} \). Formulas can be seen below

\[ K'_B = K_{Si-N} + K_{film}, \]  

(2.3.8)

\[ K_{film} = K'_B - K_{Si-N}, \]  

(2.3.9)
\[ k_{film} = \frac{K_{film} l}{w t}, \quad (2.3.10) \]

where \( l, w, \) and \( t \) are the length, width, and the thickness of the film, respectively.

Fig. 2.9 shows thermal conductance measurements vs temperature of both a blank Si-N bridge plus the total thermal conductance of the Si-N bridge plus a 50 nm AuPd thin film. Here we can see a clear contribution due to the addition of a film to the bridge. We can then simply subtract off the bridge thermal conductance to determine the thermal conductance of the AuPd film itself, which can be seen in Fig. 2.10.

### 2.3.2 Thermopower Measurement Technique

To measure the thermopower, or Seebeck coefficient, in a thin film we use the same setup as for thermal conductivity. We use a 500 nm thick \( \alpha \)-Si-N membrane to create in plane thermal gradients in thin films. To measure the thermopower of a thin film, we measure the voltage generated along the length of the film in present of a thermal gradient, \( \Delta T \), that is established in the thermal conductivity experiment. The thermopower can be determined from the slope of voltage in response to the \( \Delta T \) as seen in Fig. 2.12. Fig. 2.13 shows \( \alpha_{\text{rel}} \) measured at each temperature step to determine temperature dependence.

**Lead Contribution to Thermopower**

A majority of thermopower measurements include values of both the film and the lead contributions. In the case of our measurements, there is an additional
Figure 2.9: Thermal conductance vs. temperature for Si-N bridge and for Si-N bridge with 50 nm AuPd film. Measurements were taken on the same Si-N bridge.
Figure 2.10: Thermal conductance and thermal conductivity vs. temperature for a 50 nm AuPd film.
Figure 2.11: SEM image of Si-N membrane structure showing electrical connection to thin film for measuring thermopower and resistivity.
Figure 2.12: Longitudinal voltage vs. applied $\Delta T$ for 50 nm AuPd thin film at $T_{ref}=78$ K.
Figure 2.13: Measured $\alpha$ vs. temperature for 50 nm AuPd film.
contribution from our Cr/Pt leads on our platforms. A method has been devised to calculate the lead thermopower that can then be subtracted from the total value measured. This model is known as the effective Fuchs-Sondheimer (FS) model.[39] To determine the lead thermopower contribution, a series of varying thickness thin metallic films was deposited on the same membrane. The changes in $\sigma$, $\alpha$ and effective $l$ were measured as a function of film thickness. We expect leads fabricated via the same method and thickness should contribute to the lead thermopower equally. We can sum up the two expected thermopower contributions in the following way,

$$V_{\text{measured}} = (S_{\text{film}} - S_{\text{lead}})\Delta T.$$

(2.3.11)

Once the effective FS model is applied, the $S_{\text{lead}}$ contribution can be subtracted off of any measured $\alpha$ on the membranes. To apply the model a series of films are deposited on the membrane until the film hits the infinitely thick thin film (ITTF) limit. In typical thin metal films when the thickness is on order or smaller than the electron mean free path the transport properties will be dominated by electron-electron interactions. As the thickness is increased, the scattering will reduce. This is reflected by a change in the thermopower but also a reduction of the electrical resistivity. At the ITTF limit, the scattering of electrons is dominated by the grain boundaries and impurities in the film. The goal is to achieve a state where the scattering due to grain boundaries and impurity concentrations are constant with increased thickness, but the electron mean free path increases with the film thickness. This was achieved using a series of Au films deposited on the Si-N membrane.[40] Gold has a few advantages: it does not oxidize in air so the interface between films
Figure 2.14: Calculated $\alpha$ vs temperature for 10 nm Cr/40 nm Pt lead contribution to thermopower measurements made on Si-N membranes.

should be clean of oxide, it has a simple Fermi sphere so it is relatively straightforward to apply the effective FS model, and gold is also known to have fairly smooth grain boundaries which also aids in applying the effective FS model. The model allowed for a calculation of the absolute thin film thermopower, which can then be applied to equation 2.3.11.

Once the absolute thermopower was subtracted from the relative thermopower, the lead contribution could be determined, which can be seen in Fig. 2.14. Here
we can see that the lead contribution is quite small although similar to other metal films we have measured and varies from positive 3 $\mu$V/K at low temperatures to -5 $\mu$V/K around room temperature. This determination of the Cr/Pt lead contribution to thermopower will allow us to directly measure the absolute thermopower of any thin film.

2.3.3 Electrical Conductivity Measurement Technique

Measuring electrical conductivity is fairly straightforward with the thermal isolation platforms. Since the geometry is known, a 4-wire resistance measurement is all that is needed for calculating $\sigma$. A 4-wire measurement allows for a direct resistance measurement of our film, negating any lead resistance. An image of the setup can be seen in Fig. 2.15. Here, we drive a current through the triangle leads and measure a voltage at the inner leads using a Keithley 2400 sourceMeter. Typically, voltage is measured as current is swept from either $\pm 10 \mu$A or $\pm 5 \mu$A. The slope of this curve gives us our film resistance, as seen in Fig. 2.16. Similar to thermal conductivity measurement, our geometry is known so electrical conductivity or resistivity can be calculated using

$$\sigma = \frac{l}{Rwt},$$  \hspace{1cm} (2.3.12)

where $l$, $R$, $w$, and $t$ are length, resistance, width, and thickness of the film, respectively.

35
Figure 2.15: SEM image of Si-N membrane structure showing electrical connection to thin film for measuring electrical resistance.
Figure 2.16: Voltage vs current plot of 50 nm AuPd film at $T_{\text{ref}}=78$ K used for resistance measurement.
2.4 Lorenz Number Measurement Technique

As discussed above one of the major advantages of our membrane technique is our direct measurement of both the total thermal conductivity as well as the electrical resistance of the same film with the same geometry. This type of measurement allows for a direct calculation of the Lorenz number, ignoring any geometry affect. From Eq. 1.4.1 and since \( K = kA/L \) and \( R = \rho L/A \) we have

\[
L = \frac{k}{\sigma T} = \frac{k\rho}{T} = \frac{R_{\text{film}} K_{\text{film}}}{T} \tag{2.4.1}
\]

We can directly measure the Lorenz number, \( L \), of any deposited film, which give us insight into the various contributions to the total thermal conductivity.

2.5 Heat flow Modeling with MATLAB

To aid us in understanding the thermal model of our thermal isolation platforms, we have performed heat flow calculation of our structures. We created a thermal model of our Si-N membranes using the Partial Differential Equation (PDE) toolbox from MATLAB. The PDE toolbox uses a finite element analysis to define a 2D mesh geometry and formulate boundary conditions. The PDE solution is approximated using a piecewise linear function.

We import our geometry from Xic, our geometry layout and editor to define our structure. Next is to define a solution mesh, which can be seen in Fig. 2.19. After the mesh is established we select the heat transfer equation, in this case a steady state heat transfer elliptic PDE,
Figure 2.17: Thermal mesh for computing heat flow through a thermal isolation platform.
Figure 2.18: Boundary conditions for computing heat flow through a thermal isolation platform.
\[-\nabla \cdot (k \nabla T) = Q + h(T_{\text{ext}} - T),\]  

(2.5.1)

where \(k\), \(Q\), \(h\), and \(T_{\text{ext}}\) are the 2D thermal conductivity, heat source or power per unit area, the coefficient of convective heat transfer, and external temperature, respectively. The heat source \(Q = P/L(w)\) is defined as the applied power. Our experiment is performed in vacuum so \(h=0\). We calculate the 2D thermal conductivity, \(k_{2D}\), by multiplying \(k\) by the thickness of the material, \(t\). Subsequent stacks are added together. For example, we’ve measured the thermal conductivity of Si-N at \(\approx 3\) W/mK. Multiplying that by 500 nm gives a \(k_{2D}\) of \(1.5\) \(\mu\)W/K. We’ve also calculated the 2D Pt thermal conductivity as \(1.72\) \(\mu\)W/K. To set the thermal conductivity of the full stack we simply add them together, \(k_{2D} = k_{2D,\text{Si-N}} + k_{2D,\text{Pt}} = 1.5\) \(\mu\)W/K + \(1.72\) \(\mu\)W/K = \(3.22\) \(\mu\)W/K. The final step is to define the boundary conditions as seen in Fig. 2.18. Dirichlet boundary conditions are used to set the frame temperature of the system around the perimeter and are shown as red lines in Fig. 2.18. Neumann boundary conditions are used to define the edges of the platform with heat flux and heat transfer coefficient set to zero to represent the platform being in vacuum.

With our inputs set we can generate our heat flow model, as seen in Fig. 2.19, for creating thermal gradients used in our thermal conductivity and thermopower measurements. Here we can see our thermal gradient direction along the bridge, confirming our understanding of our thermal model. The model can also be used to estimate thermal gradients found in our spin transport measurements in Chapter 5.
Figure 2.19: Heat flow model for thermal conductivity and thermopower measurements using a thermal isolation platform. Heat is applied to heater on back of the right island with a 20K $\Delta T$ between right island and frame. The model was generated using the PDE Toolbox from MATLAB and assuming 2-D heat flow through the structure.
Figure 2.20: Heat flow model for spin transport measurements on thermal isolation platform. Heat is applied to lead on inner leg of the right island with a $\Delta T$ of 50 K between right island and frame.
2.6 Summary

In this chapter, we covered our use of thermal isolation platforms to measure electrical, thermal and spin transport in thin films. The membrane structures allow us to create and manipulate 2D thermal gradients in the thin films. We can calculate the Lorenz number directly from the film resistance and thermal conductance measured on the same film, eliminating any geometry effects. Heat flow modeling has been used to help us estimate and understand the thermal gradients in our membrane. The following chapters will cover transport measurements across a variety of thin film nanostructures.
Chapter 3

Thermal conductivity dependence of doped semiconducting single-walled carbon nanotube networks

In this chapter we present thermal conductivity, \( k \), electrical conductivity, \( \sigma \), and Seebeck voltage, \( \alpha \), of semiconducting single-walled carbon nanotube thin films. Three films will be presented, two p-type doped films from two manufacturing sources and one n-type doped film.
3.1 Introduction

The search for quality, reliable and efficient thermoelectric materials is always expanding. One of the latest materials of interest are arrays of semiconducting single-walled carbon nanotube (s-SWCNT) networks. Work in 2016 by our collaborators at the National Renewable Energy Lab (NREL, Golden, CO) have found thermoelectric power factors, higher than $340 \, \mu W m^{-1} K^{-2}$ at room temperature, comparable to conducting polymers and larger than other carbon nanotube films.[1] Even more recently the same group have found power factors upwards of $700 \, \mu W m^{-1} K^{-2}$ at room temperature for both n-type and p-type doped carbon nanotube films.[41] The same films show a peak material $zT \approx 0.12$, which is unprecedented in the carbon nanotube TE field.

3.2 Experimental Details

The experimental technique is very similar to the data taken on the CoFe alloy films presented in chapter 4 but the setup is quite different. The process of extraction and enriching SWCNT samples begins with raw SWCNT soot. A sonication tip is used along with a selectivity fluorene-based cleavable polymer to create polymer-rich exfoliated SWCNTs. A centrifuge is then used to separate semiconducting and metallic SWCNT. The remaining s-SWCNT solution is once again put in a centrifuge to separate the polymer with the s-SWCNT ink. The ink is then ultrasonically sprayed onto a our Si-N platform leaving a s-SWCNT network with excess polymer. The final step includes a TFA/Toluene treatment to fully remove the excess polymer.
This last step provides a dense network of s-SWCNT for characterization. The full removal of the sorting polymer allows for enhanced maximum electrical conductivity and TE power factor.

For $k$, $\sigma$ and $\alpha$ measurements performed on our Si-N platforms several steps are needed before the s-SWCNT spray deposition. A two-step 10 nm alumina ($\text{Al}_2\text{O}_3$) layer is deposited on both the bridge and the islands, as seen in fig. 3.1. The $\text{Al}_2\text{O}_3$ pre-passivation reasoning is two-fold. The layer on the bridge (Fig. 3.1 (1)) is to reduce the $k$ of the Si-N background.[37] As detailed in chapter 2, understanding the background $k$ of the Si-N bridge is very important. The $k$ of s-SWCNTs can be quite small, about 10% or less of the Si-N background. As found with previous films, a thin layer of material can drop the measured $k$ of the background by $5 - 10\%$. The background $k$ becomes ‘saturated’ by the addition of a several nm thick film due to a modification of surface scattering at the Si-N/film surface. The second layer of alumina (Fig. 3.1 (2)) is deposited to cover thermometer and heater leads. During the spraying of the s-SWCNT ink, the ink tends to undercut the mask and would electrically short the leads without a passivation layer. Once both alumina layers are deposited the platforms are soaked in a dopant, triethyloxonium hexachloroantimonate (OA) for p-type or a potassium-crown ether complex for n-type, to further saturate the platform. A background measurement is then performed on the pre-passivated, pre-doped platforms. Once complete the s-SWCNT ink can then be sprayed.
Figure 3.1: Location of 10 nm Al₂O₃ two step pre-passivation. Growth was performed in an ultra-low vacuum deposition chamber at starting pressures of $1 \times 10^{-9}$ torr and growth rate of 2 Å/s.
3.3 Results and Discussion

The initial state of the s-SWCNT films have a low electrical conductivity and a high thermal conductivity (15-20 W/m K).[1] As reference, individual SWCNT have extremely high thermal conductivities along the tube axis, with experimental results higher than 1000 W/m K at room temperature.[42, 43] Thermal conductivities of CNT networks vary widely based on CNT source and measurement technique.[44, 45] $k$ is expected to be smaller than metals due to electrons and phonon needing to ‘hop’ from tube to tube, with tubes being held lightly together via Van der Waals forces. Unique to our measurements, we look at the evolution of $k$, $\sigma$ and $\alpha$ with various doping levels, which can greatly affect carrier concentrations and transport. Fig. 3.2 shows a cartoon image of a phonon and electron traveling down a CNT network in addition to measured thermal conductivities of an undoped, doped and de-doped sample.

In Fig. 3.2 we can clearly see the effect that doping has on the transport properties. The initial undoped state has a low $\sigma$ and high $k$. Once doped, the sample $\sigma$ increased drastically, where the $k$ dropped. We understand that by adding doping molecules to the network the electron mobility increases causing an increase in $\sigma$. These same molecules add scattering sites for phonon with drops the overall $k$. The samples can be somewhat de-doped by sitting in vacuum for several days. This process causes the $\sigma$ to drop as the doping molecules are driven off but we don’t see a large change in thermal conductivity. We are unable to confirm what physical process happens while de-doping but it is theorized that the doping molecules aren’t full extracted. One possibility is as the sample de-dopes, the doping molecules are
Figure 3.2: (Top left) Cartoon image of undoped s-SWCNT network. Orange line shows phonon traveling along connected tubes freely. Green line shows charge carrier unable to cross tube to tube junction. (Bottom Left) Shows doped s-SWCNT network. Phonon (orange line) scatters off dopant. Charge carrier (green line) is able to traverse from tube to tube with addition of dopant molecule. (Right) Thermal conductivity vs electrical conductivity data for undoped, doped and de-doped PFO-BPy:LV thin film near 300 K. The dashed gray line indicates expected electronic thermal conductivity, $k_e$, contribution determined from the Wiedemann-Franz law.
cleaved in such a way to reduce electron mobility but still leave scattering sites for phonons.

Next we take a look at the evolution of two samples, a 67.9 nm ± 15 nm high-pressure disproportionation of carbon monoxide (HiPCO, Los Alamos National Laboratory) and a 60 nm ± 15 nm plasma-torch (PT, NanoIntegris) sample. In each we look at three doping states to help us understand the contribution so \( k_{\text{total}} \). Fig. 3.3 shows temperature dependent \( K_{\text{total}} \) for the HiPCO sample. Data shows the sample minimally doped at 10,720 S/m, fully doped at 129,465 S/m, and then partially de-doped at 102,560 S/m. The thermal conductance follows the trend of low \( \sigma \), high \( k \) and vice versa. We can see here how small the thermal conductance of the SWCNT network is compared to the background thermal conductance. This small signal contributes to about half of the calculated error. After subtracting the background conductance, Fig. 3.4 shows just the SWCNT network thermal conductance and thermal conductivity. Here it is easier to observed a trend in \( k \). Interestingly when we look at the PT SWCNT thin film, we see the opposite effect as seen in Fig. 3.5 and 3.6. Here as \( \sigma \) decreases, \( k \) also decreases.

We can further analyze these two samples by calculating the Lorenz number, allowing us to separate and determine the \( k_{\text{phonon}} \) and \( k_{\text{electron}} \) contributions to the measured \( k_{\text{total}} \). The expected electron contribution to \( k_{\text{total}} \) comes from the Wiedemann-France law, \( L = K_{\text{film}} R_{\text{film}} / T \), with \( L \) set to \( L_o = \pi^2 k_b^2 / (3 e^2) \), the Sommerfeld theory for free electrons. It is important to note that \( R_{\text{film}} \) and \( K_{\text{film}} \) are measured on the same sample, resulting in a geometry free calculation. Tables 3.1 and 3.2 show thermal conductance, thermal conductivity and Lorenz number for each of the doping...
Figure 3.3: Thermal conductance vs. temperature for 67.9 nm ± 15 nm HiPCO SWCNT thin film including background. Three colors indicate three doping levels with empty squares indicating background thermal conductance from Si-N membrane. Measurements were all taken on the same film in vacuum. Error bars are dominated by uncertainty in film thickness but does not effect individual points, instead will simply shift all data up or down.
Figure 3.4: Thermal conductance (left axis) and calculated thermal conductivity (right axis) vs. temperature for 67.9 nm ± 15 nm HiPCO SWCNT thin film. Three colors indicate three doping levels.
Figure 3.5: Thermal conductance vs. temperature for 60.0 nm ± 15 nm PT SWCNT thin film including background. Three colors indicate three doping levels with empty squares indication background thermal conductance from Si-N membrane. Measurements were all taken on the same film in vacuum. Error bars are dominated by uncertainty in film thickness but does not effect individual points, instead will simply shift all data up or down.
Figure 3.6: Thermal conductance (left axis) and calculated thermal conductivity (right axis) vs. temperature for 60.0 nm ± 15 PT SWCNT thin film. Three colors indicate three doping levels.
Table 3.1: Thermal conductance, thermal conductivity and Lorenz number for 3 doping states of a HiPCO SWCNT thin film.

<table>
<thead>
<tr>
<th>(\sigma) (S/m)</th>
<th>(K) (nW/K)</th>
<th>(k) (W/m K)</th>
<th>(L \times 10^{-8}) (WΩ/K²)</th>
<th>(L/L_o)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10,720</td>
<td>6.09 ± 0.32</td>
<td>2.09 ± 0.72</td>
<td>64.96</td>
<td>26.62</td>
</tr>
<tr>
<td>129,465</td>
<td>4.05 ± 0.22</td>
<td>1.39 ± 0.43</td>
<td>3.58</td>
<td>1.47</td>
</tr>
<tr>
<td>102,560</td>
<td>5.65 ± 0.44</td>
<td>1.94 ± 0.70</td>
<td>6.30</td>
<td>2.58</td>
</tr>
</tbody>
</table>

Table 3.2: Thermal conductance, thermal conductivity and Lorenz number for 3 doping states of a PT SWCNT thin film.

<table>
<thead>
<tr>
<th>(\sigma) (S/m)</th>
<th>(K) (nW/K)</th>
<th>(k) (W/m K)</th>
<th>(L \times 10^{-8}) (WΩ/K²)</th>
<th>(L/L_o)</th>
</tr>
</thead>
<tbody>
<tr>
<td>191,071</td>
<td>11.93 ± 0.90</td>
<td>4.63 ± 1.49</td>
<td>9.15</td>
<td>3.75</td>
</tr>
<tr>
<td>131,612</td>
<td>7.29 ± 1.04</td>
<td>2.83 ± 1.12</td>
<td>7.17</td>
<td>2.94</td>
</tr>
<tr>
<td>103,535</td>
<td>6.13 ± 1.00</td>
<td>2.38 ± 0.98</td>
<td>7.66</td>
<td>3.14</td>
</tr>
</tbody>
</table>

states seen in figures above. To help reduce error, we assume no temperature dependence allowing us to average our data points around room temperature.

We can see that for the HiPCO film in table 3.1, the Lorenz number changes drastically with a change in doping. This indicates that this sample is largely dependent on phonon thermal conductivity. \(L/L_o\) indicates a phonon to electron contribution ratio with higher values leading to strong dependence on phonon thermal conductivity. Looking at the data for the PT sample in table 3.2, we see an almost constant Lorenz number. Even as the doping chances the respective phonon and electron contributions remain largely the same.

To help understand these differences in the two films we can look at both the intrinsic and extrinsic differences in the samples. As detailed in MacLeod et. al. [41], each of these source materials varies both intrinsically and extrinsically. The HiPCO SWCNTs have a smaller diameter, 1.0 nm ± 0.15 nm, compared to the diameter of the PT SWCNTs, 1.3 ± 0.10 nm. The HiPCO also have a smaller bundle size, 15
± 3nm, than the PT SWCNTs, 20 ± 4 nm. It appears that a smaller diameter and bundle size may lead to an overall lower thermal conductivity and a stronger reliance on phonons as a thermal transport mechanism. However, extrinsic properties may also play a role, such as film processing conditions and film morphology. It is difficult to determine at this time how each factor plays a role. What we can conclude at this time is that it appears that the intrinsic size of the SWCNT diameter and bundle affects how heat is transferred through the films.

Finally, we look at the Seebeck voltage, \( \alpha \), of both films as seen in Figs. 3.7 and 3.8. Both films follow expected \( \alpha \) shift with doping evolution. Both films were p-type doped, which shifts Fermi energy from the middle of the band gap towards the valence band resulting in a positive \( \alpha \) as seen in Fig. 3.9. HiPCO and PT film exhibit highest \( \alpha \) with lowest doping levels with \( \alpha \) decreasing with increasing doping.
Figure 3.7: Seebeck voltage, $\alpha$, vs. temperature for 3 doping states of 67.9 nm ± 15 nm HiPCO SWCNT thin film. Relative $\alpha$ includes Pt lead contribution but expected to be < 10% at room temperature and same for all doping.
Figure 3.8: Seebeck voltage, $\alpha$, vs. temperature for 3 doping states of 60.0 nm $\pm$ 15 nm PT SWCNT thin film. Relative $\alpha$ includes Pt lead contribution but expected to be $< 10\%$ at room temperature and same for all doping.
3.4 Conclusions

In this chapter, we have discussed thermal and electrical transport measurements performed on semiconducting single-walled carbon nanotube thin films. We took a close look at two separate but similarly thick films as we manipulated the charge doping levels. These films were manufactured in two different processes which lead each of them to exhibit separate dependence on both phonon and electron thermal conductivity as the doping levels changed.
Chapter 4

Magnon contributions to thermal conductivity and thermopower in a metallic thin film

In this chapter we present thermal conductivity and thermopower results of ultra-low magnetic damping ferromagnetic thin films. Main focus will be on two Co$_{25}$Fe$_{75}$ alloy films (ultra-low damping) and a Co$_{50}$Fe$_{50}$ alloy film as a baseline.

4.1 Introduction

Recent work has renewed interest in the role of magnons in the transport and thermoelectric properties of metallic ferromagnets. A multitude of groups have explored the role the magnon contribution plays to thermopower (cite work). This effect is called magnon drag, which is a consequence of the electron-magnon interaction
whereby the spin excitations in a magnetic material transfer momentum to the
electron system and increase the thermopower. The overall magnon contribution
is closely related to the magnetic damping parameter, which is a key component
in future device development. In order to utilize the spin degree of freedom the
damping must be understood as it greatly affects both the energy requirement and
the speed of operation. Colleagues at the National Institute of Science and Tech-
ology (NIST, Boulder, CO) have performed a thorough study to quantitatively
predict the magnetic damping of a binary alloy CoFe [2]. Finding a low damping
ferromagnet was considered difficult due to the scattering of magnons by a metal’s
conduction electrons. They theoretically predicted and experimentally demonstrated
a total damping of $\alpha_{tot} = 0.0021 \pm 0.00015$ (Fig. 4.1), where $\alpha_{tot} = \alpha_{int} + (\alpha_{rad}/2) + \alpha_{sp}$. $\alpha_{int}$, $\alpha_{rad}$ and $\alpha_{sp}$ are the intrinsic, radiative and spin pumping damping contributions
respectively. An intrinsic damping value of $\alpha_{int} = 0.0005$ was found for the Co$_{25}$Fe$_{75}$
alloy. Other metal and half metal systems have been theoretically predicted to have
ultra-low damping in the $10^{-4}$ regime[46] but only a few experimental results have
been realized as low as 0.001.[47, 48] The damping constant of Yttrium Iron Garnet
(YIG) has been measured as low as $(7.35 \pm 1.50) \times 10^{-5}$, but YIG is an insulator so
it is not available for some spintronics applications.

To account for the low $\alpha_{int}$ in presence of conduction electrons, electronic struc-
ture calculations were performed. These calculations showed that $\alpha_{int}$ is strongly
determined by Density of States (DOS) at the Fermi energy, $n(E_f)$, as seen in Fig.
4.2. A minimum in the DOS at $n(E_f)$ can be seen for the Co$_{25}$Fe$_{75}$ alloy com-
position. They concluded that the low intrinsic damping stems from the minimized
Figure 4.1: Total measured damping. The total damping, $\alpha_{\text{tot}}$ (red cross with lines), spin-pumping, $\alpha_{\text{sp}}$ (gray line) and radiative $\alpha_{\text{rad}}$ (green line) and intrinsic damping, $\alpha_{\text{int}}$ (black squares with lines) are plotted against the Co concentration. Reprinted by permission from Springer Nature License.[2]
Figure 4.2: Electronic structure of bulk Co$_x$Fe$_{1-x}$. DOS vs. Co concentration is show. $E_F$ is the Fermi energy. Reprinted by permission from Springer Nature License.[2]

$n(E_f)$ in the limit of intraband scattering, and that in order for a strong theoretical understanding all contributions to the damping are needed.

Work in this chapter explores the Co$_{25}$Fe$_{75}$ alloy further by looking at the thermal conductivity and thermopower temperature dependence to determine the effect of ultra-low damping in a ferromagnetic metal thin film. Specifically, we probed the balance between magnon and phonon contributions to thermal conductivity and the role of these excitations in the Seebeck effect in these films.
4.2 Experimental Details

Fabrication details of thermal isolation platforms is detailed in an earlier section (chapter 2). Platforms are masked with a micromachined shadow mask that allows for a continuous film to be grown on the bridge with electrical connection to voltage leads on the inner section of each island. The magnetic films are deposited via DC magnetron sputtering at an Ar pressure of approximately $5 \times 10^{-3}$ Torr and chamber base pressure of $4 \times 10^{-8}$ Torr. The alloy is co-sputtered from dual elemental targets (Co and Fe) with deposition rates calibrated by X-ray reflectometry (XRR). The deposition rate is kept to 0.25 nm/s. Samples are grown with a Ti(3 nm)/Cu(5 nm) seed layer to promote a BCC structure and an Al(5 nm) capping layer to prevent oxidation. Three films were grown in this manner. Two Co$_{25}$Fe$_{75}$ films were grown during the same deposition and one Co$_{50}$Fe$_{50}$ film was grown separately. Alloy thickness of 75 nm was used across all films. An example stack is shown in Fig. 4.3.
4.3 Results and Discussion

Four-wire resistance measurements were performed on both sets of films on thermal isolation platforms. A high density of states in the Co$_{50}$Fe$_{50}$ alloy leads to an increase in the electrical conductivity or a lowering in the electrical resistivity compared to the two Co$_{25}$Fe$_{75}$ films, which can be seen in Fig. 4.4a. The two Co$_{25}$Fe$_{75}$ films have a similar resistance indicating thickness, alloy composition, and impurities are roughly consistent. Fig. 4.4b compares the CoFe alloys to other films we’ve measured on thermal isolation platforms. Here we see a higher resistivity compared to elemental thin films as one expects from increased disorder scattering from the random alloy. Similar values for the CoFe alloys were reported by the group at NIST Boulder. The temperature dependence is as expected for a metal.

Next we take a look at the thermal conductivity, $k$, of our thin films. Fig. 4.5 shows the thermal conductivity of both Co$_{25}$Fe$_{75}$ and Co$_{50}$Fe$_{50}$ films. At first glance the data doesn’t appear to line up properly. We would expect the Co$_{25}$Fe$_{75}$ films to have similar values to each other. Instead, the Co$_{50}$Fe$_{50}$ film sits directly in between the two Co$_{25}$Fe$_{75}$ films. To help clear this up we first take a look at the expected electronic contribution to the thermal conductivity via the electrical resistance as determined by the Wiedemann-Franz law, $\kappa_e = L_0 T \sigma_e$, where $\kappa_e$, $L_0$, $T$ and $\sigma_e$ are the electronic thermal conductivity, Lorenz number, temperature and electrical conductivity respectively.

Fig. 4.6 shows the expected electrical thermal conductivity in comparison to the total thermal conductivity measured for all three films. Here we see that for the Co$_{50}$Fe$_{50}$ film in Fig. 4.6c, the entire contribution to the thermal conductivity is due
Figure 4.4: Temperature dependent resistivity of two Co$_{25}$Fe$_{75}$ and one Co$_{50}$Fe$_{50}$ film. 4-wire resistance data was taken and converted to resistivity with known sample geometry.
Figure 4.5: Temperature dependent total measured thermal conductivity, $k$, of both Co$_{25}$Fe$_{75}$ films and Co$_{50}$Fe$_{50}$ film.

to the electrons as heat carriers, with a depression at higher temperature. While the Co$_{50}$Fe$_{50}$ film is also at a lower damping than a typical metal alloy, this does not seem to affect the thermal conductivity compared to the ultra-low damping Co$_{25}$Fe$_{75}$ films. The ultra-low damping films show a greatly enhanced thermal conductivity in addition to the expected electron contribution, as seen in Fig. 4.6b and 4.6c. The additional contribution to thermal conductivity above the expected contribution from electrons could be due to either magnon or phonon effects. The Co$_{25}$Fe$_{75}$ films appear to have a peak due to a drag-like feature, which typically shows an increasing effect as the temperature raises, but are reduced at high temperatures due to additional phonon or magnon scattering.
We can then calculate the Lorenz number for each film,

\[ L(T) = \frac{k}{\sigma T} = \frac{K_{\text{film}} R_{\text{film}}}{T} \]  \hspace{1cm} (4.3.1)

where \( K_{\text{film}} \) and \( R_{\text{film}} \) are the sample thermal conductance and electrical resistance respectively. Notice here that these values are geometry independent, which can often be the largest source of error in transport measurements. In our samples we measure the \( K_{\text{film}} \) and \( R_{\text{film}} \) on the same sample canceling out the common geometry. Our use of the total \( k \) allows for determination of all sources of thermal conduction.

Fig. 4.7 shows the calculated \( L \) for all three films. Here we see the expected trend in our three films. The \( \text{Co}_{50}\text{Fe}_{50} \) sample sits directly at \( L_o \), the Sommerfeld value for free electrons, \( L_o = \frac{\pi^2 k_b^2}{3 e^2} \).

There is still a discrepancy between the two \( \text{Co}_{25}\text{Fe}_{75} \) films, which should have the same or similar value since they were grown at the same time on the same growth stage. Assuming the thickness is similar, there could be two possibilities for this difference. Either the phonon population/scattering is different between the two, or the magnon population/scattering is different. Initial thoughts lead to a different magnon population. A higher resistance should lead to a lower phonon thermal conductivity due to additional impurities that causes scattering. If we look at film in Fig. 4.6b), which has a higher resistance, it also has a higher \( k \), the opposite we’d expect if it was a phonon thermal conductivity. To probe the magnetic degree of freedom, the next step is to introduce both samples in a high magnetic field that could collapse the two films to a similar magnetic state. This next step is ongoing as of this writing. If the two films do not respond to a magnetic field then this discrepancy
Figure 4.6: Total thermal conductivity and calculated electrical thermal conductivity vs. temperature for 75nm thick CoFe films. a) Co$_{25}$Fe$_{75}$ alloy film. b) Co$_{25}$Fe$_{75}$ alloy film. c) Co$_{50}$Fe$_{50}$ alloy film.
Figure 4.7: Lorenz number vs. temperature for two Co$_{25}$Fe$_{75}$ films and a Co$_{50}$Fe$_{50}$ film. Green dashed line indicates Lorenz number for free electrons.

could be due to a dissimilar $k_{\text{phonon}}$ DOS, which may not be affected by an external field. This is an interesting possibility that other groups are currently exploring. Either the ultra-low damping causes an increase in $k_{\text{phonon}}$, or the minimum in the electronic density of states leads to both the ultra-low magnetic damping and an increase in $k_{\text{phonon}}$. This type of tuning may be useful for thermoelectrics, with the ability to manipulate both the resistance and thermal conductivity.

Fig. 4.8 shows the absolute Seebeck voltage, $\alpha_{\text{abs}}$, as a function of temperature for both Co$_{25}$Fe$_{75}$ films and Co$_{50}$Fe$_{50}$ film. Here we see nice agreement with the Co$_{25}$Fe$_{75}$ films. If we consider $\alpha$ purely from a DOS view, we’d expect the Co$_{25}$Fe$_{75}$ film to have a very small, near zero, Seebeck voltage due to the minimum in the DOS at the Fermi
Figure 4.8: Absolute Seebeck voltage vs. temperature for two Co$_{25}$Fe$_{75}$ films and a Co$_{50}$Fe$_{50}$ film.
Figure 4.9: Absolute Seebeck voltage vs. temperature for a variety of thin films grown on membranes.

level. The Co$_{50}$Fe$_{50}$ film would then have a small negative Seebeck voltage. Instead, we see a relatively large Seebeck voltage in the Co$_{25}$Fe$_{75}$ film, with a value larger than the Co$_{50}$Fe$_{50}$ film and a Co thin film as seen in Fig. 4.9. This enhancement could be due to either magnon-drag [49] or phonon-drag effects present in the ultra-low damping film. The Co$_{50}$Fe$_{50}$ alloy film is roughly half of what is seen in the Co$_{25}$Fe$_{75}$ alloy films due to a change in the DOS dropping both the diffusive and magnon-drag contributions.
Finally, we compare the thermopower of the CoFe alloys with Co and Fe elemental films. The Co$_{25}$Fe$_{75}$ films both have larger signal sizes than the elemental films, again indicating that something else is at work here. Interestingly we found almost identical temperature dependence and signal size between the Co$_{50}$Fe$_{50}$ film and a pure Co film of the same thickness. This indicates to us that any magnon-drag contribution exclusive to the low damping nature of the measured CoFe alloys is suppressed in the Co$_{50}$Fe$_{50}$ film. This isn’t to say that there is no magnon-drag component to the thermopower of the Co$_{50}$Fe$_{50}$ or Co film, just that any enhancement due to the low damping is suppressed in these two films. The Fe film is overall positive and exhibits a magnon-drag peak around 125 K, both in agreement with previous results.[50, 36]

4.4 Conclusions

In this chapter we presented thermal conductivity and thermopower data for three CoFe thin films. We observed enhanced thermal conductivity in the two ultra-low magnetic damping Co$_{25}$Fe$_{75}$ thin films and suppressed thermal conductivity in the Co$_{50}$Fe$_{50}$ thin film. Thermopower data gave us insight into a possible magnon-drag like contribution in addition to the expected metallic DOS driven thermopower.
Chapter 5

Long-distance spin transport in disordered magnetic insulators

In this chapter we present long distance spin transport measurements in amorphous yttrium iron garnet (a-YIG) and amorphous chromia (a-Cr$_2$O$_3$) thin films. We perform measurements on both membrane and substrate allowing for manipulation of thermal gradients.

5.1 Introduction

Motivated by new paradigms for information processing, spintronics research has recently focused on the transport of spin information via spin-wave, or magnon, excitation in magnetic insulators. Much of this work uses yttrium iron garnet, Y$_3$Fe$_5$O$_{12}$ (YIG), as the spin transport medium due mostly to its very low damping of magnetization dynamics and the resulting long spin-wave propagation lifetime.[51, 52]
In its bulk crystalline form YIG is a ferrimagnet with an electronic bandgap of \( \approx 2.8 \text{ eV} \), which is also achieved in thin films,[53] so that electronic excitations can not contribute to transport. The ferrimagnetism arises due to the location of \( \text{Fe}^{3+} \) ions in two inequivalent sites in the relatively complicated unit cell, leading to antiferromagnetic exchange interactions between octahedrally- and tetrahedrally-coordinated \( \text{Fe}^{3+} \) ions but with somewhat different moments, leaving a net imbalance of magnetization and macroscopic properties often described using the typical language of ferromagnets.

5.2 Experimental Details

5.2.1 Deposition

Devices used in these experiment are the same as seen in earlier chapters (2, 3, and 4). 200 and 100 nm thick \( a \)-YIG films were sputtered from a stoichiometric YIG target in argon gas on 1 cm \( \times \) 1 cm blank Si-N coated silicon substrates and \( a \)-Si-N platforms. The substrates were held near room temperature with growth rates at \( \sim 0.5 \text{ nm/min} \). 100 nm thick \( a \)-\( \text{Cr}_2\text{O}_3 \) was evaporated using an electron beam. In both these cases the disordered magnetic material was grown on top of Pt leads. A secondary set of devices were fabricated with leads grown on top of \( a \)-YIG. Resist was patterned via electron beam lithography to construct a set of parallel strips. 20 nm of Pt and Cu were deposited using DC sputtering. A low energy ion mill was performed before metal deposition to clean the surface.
5.2.2 X-ray Diffraction

X-ray diffraction data were collected in the Bragg-Brentano symmetrical $\theta - 2\theta$ reflection geometry by using CuK$_\alpha$ characteristic energy (8 keV), excited at 30 kV and 30 mA. Reflected intensity was scanned by a proportional detector every 0.05° in the angle $2\theta$ for 10 s per step. XRD was measured on a 200 nm thick $a$-YIG layer deposited on a 500 nm thick Si-N coated Si substrate, where the $a$-YIG was grown in the same deposition as films on thermal isolation platforms tested for spin transport and magnetization. For polycrystalline YIG the spectrum was normalized to the (420) peak, which had a raw value of 13,000 counts.

5.2.3 Non-local Transport Measurements

Three iterations of the non-local measurements were made on $a$-YIG and $a$-Cr$_2$O$_3$. The series performed on the Si-N isolation platforms on both membrane and substrate used Pt injection and detection leads underneath the $a$-YIG and $a$-Cr$_2$O$_3$ films. Measurements were performed by sourcing a current $I$ down the length of a Pt lead and measuring a voltage on a parallel but completely separate Pt strip. On the membrane, the Pt leads transverse both legs of one Si-N island with a total length $\sim$ 2 mm and separation of $\sim$ 9 microns and are entirely on the suspended membrane. On the substrate portion of the thermal isolation platforms, the Pt leads sit on the outer edge of the device. Here the total length was $\sim$ 10 - 15 mm had a and separation of 10 - 210 microns. The final device iteration we measured on Si-N blanks. Measurements performed on the Si-N blanks had Pt leads on top of $a$-YIG.
and crystalline YIG films with lengths of $\sim 3 - 9$ mm and separation of 10 - 150 microns.

We measured the non-local signal as a function of temperature from cryostat temperatures of $\sim 80 - 390$ K. Driving large $I$ through the Pt wire joule heating occurs. Currents used on the substrate measurements saw substrate temperatures rise by several Kelvin increasing the measured voltage by a few microvolts. This effect was evident but easily accounted for. Any $\Delta T$ was out of plane due to the substrate acting as a heat sink. Measurements performed on the Si-N membranes resulted in significant heating, upwards of a $\Delta T$ of 200 K from island to frame. This thermal gradient of almost entirely confined to the plane due to the 2D structure.

All zero-field measurement were performed under vacuum, or exchange gas, at $10^{-6}$ Torr or better. Field-dependent measurements were performed in ambient conditions, with sample placed between the 10 cm diameter pole pieces of an electromagnet with a gap of $< 1$ cm. To reduced background temperature variance along with background thermoelectric voltages, voltage measurements recorded while cycling a set bias current on and off were averaged over several cycles. The remaining background drift is linear over small time periods and was removed with a simple linear fit.

5.3 Results and Discussion for $a$-YIG

Among the most exciting spin transport studies in YIG are experiments demonstrating electrical excitation of spin waves in the YIG via the spin Hall effect,\[54, 55, \]
56, 22] and subsequent detection of the spin information some distance away from
the injection site via the reciprocal inverse spin Hall effect (ISHE). This non-local
generation and detection of spin information transport in YIG, shown schematically
in Fig. 5.1a, was first described by Kajiwara, et al. [3] where very long length scale
propagation was claimed. Only recently have other reports of similar experiments
also on YIG emerged, showing shorter propagation length scales.[57, 58, 59, 60, 61]
All of these experiments focus on crystalline or epitaxial YIG, though depending
on the process steps used in fabrication some level of disorder could arise. The
study by Kajiwara, et al. also reported the excitation of magnetization dynamics
in crystalline YIG by SHE-driven torques, a phenomenon also recently reported
using different device structures,[62, 63, 64, 65, 66] including some that deliberately
enhance the role of thermal gradients.[64] The SHE excitation of YIG magnetization
has also been theoretically described.[67, 68, 69] The characteristic feature of this
SHE spin-wave excitation is an onset of the dynamics at a critical current where
the applied spin torque balances the damping of the magnetization dynamics in the
YIG. Furthermore, thermal gradients and spin-wave excitations have been shown
to have other dramatic interactions in YIG,[70, 71] opening the possibility that the
application of thermal gradients in these experiments, whether intentional or unin-
tentional, could play a strong role in measured effects. This has been observed by
some groups,[57, 59] though the applied thermal gradients tested to date are over-
whelmingly perpendicular to the plane of the YIG/Pt interfaces. Nevertheless, the
thermal generation of a population of magnons that subsequently diffuses through
the YIG is one possible mechanism for the long-distance spin flow.[72]
Figure 5.1: Schematic views of experiments in long-distance spin transport. a) Non-local spin transport in crystalline YIG, a ferrimagnetic insulator. b) Spin transport through a disordered magnetic insulator, a-YIG, relying only on magnetic correlations.
Other recent reports have shown that spin transport is possible through a much wider range of materials than previously thought. These include studies of spin transport, and possible enhancement of spin flow, through very thin nickel oxide\cite{73, 74, 75} and other nominally antiferromagnetic insulating\cite{76} layers inserted between YIG and Pt layers, and through thin native oxides of nickel and Permalloy between transition metal ferromagnets and heavy metal films.\cite{77} These initially unexpected experimental results have stimulated theoretical consideration of spin transport by magnons in antiferromagnetic insulators.\cite{78, 79, 80} In addition to these studies, where spin transport was shown via electrically-detected measurements of the ISHE in response to spin pumping, the longitudinal spin Seebeck effect has been demonstrated in antiferromagnets, \cite{81, 82, 83} paramagnets,\cite{84} and ferromagnets above the Curie temperature.\cite{85} These results clearly demonstrate that long-range magnetic order is not a requirement for spin transport in an insulator, which is also implicit in any spin transport experiment using a very thin film of a material that is antiferromagnetic in bulk, but with a blocking temperature well below the temperature of the experiments.\cite{73, 74, 76, 77} New experiments to test a broader range of disordered magnetic insulators, where magnetic correlations persist due to strong local exchange interactions despite the lack of a low symmetry state, are therefore critical for spintronics.

Here we show that a disordered magnetic insulator allows long-distance spin transport. Fig. 5.1a) shows non-local spin transport in crystalline YIG, a ferrimagnetic insulator. Charge current driven through a platinum strip causes a spin current in the Pt thickness direction flow via the spin Hall effect (SHE), generating spin torque
and/or spin accumulation at the Pt/YIG interface, exciting magnons that carry spin information to a Pt detector where spin current injected generates a charge voltage via the inverse spin Hall effect (ISHE). The X-ray diffraction (XRD) pattern shows Bragg reflections consistent with randomly oriented polycrystalline YIG. A simplified 2D schematic spin structure of YIG shows two sublattices of Fe spins, where nearest neighbor interactions are antiferromagnetic. Fig. 5.1b) shows spin transport through a disordered magnetic insulator, $a$-YIG, relying only on magnetic correlations. XRD on a 200 nm thick YIG layer sputtered on a Si substrate coated with 500 nm of $a$-Si-N shows no YIG diffraction peaks, indicating the lack of any medium- or long-range order in the YIG layer. Strong peak at $\sim 35^\circ$ is due to the Si substrate (200) Bragg reflection. A simplified 2D schematic random spin structure of $a$-YIG, illustrates the high degree of frustration, and lack of long-range order despite strong AF interactions between neighboring spins. We demonstrate non-local spin transport (see Fig. 5.1b), with large signal voltages indicating propagation over dozens of microns, through amorphous YIG ($a$-YIG), a magnetic insulator with strong local antiferromagnetic exchange interactions but neither magnetic nor structural long-range order. We describe non-local spin transport in $a$-YIG films sputtered both on suspended amorphous Si-N sample platforms and on bulk Si substrates. Comparing these allows us to identify a strong effect on in-plane thermal gradients. We show two separate contributions to the non-local spin transport, with one showing a clear onset at well-defined critical current density in the Pt across a fairly broad range of samples and measurement conditions, while the other is linear with applied current through the strip. Finally, when the non-equilibrium spin carriers are injected into $a$-YIG the
temperature profile suggests efficient heat transport by this spin population, which
echoes the strong magnon-phonon coupling often observed in crystalline YIG. These
results open a new frontier in insulating spintronics, proving that magnetic order is
not required, and may not be desirable, for an efficient spin-transport medium.

Amorphous YIG was originally studied, though far from exhaustively, decades
ago. Results indicated that disordered YIG (rarely grown in thin-film form) showed
a broad peak in $M$ vs. $T$ between 50 and 100 K,[4, 5] with a splitting between
curves measured in zero field cooled and field cooled conditions.[5] Above this split-
tting, some groups reported reasonable agreement of $M$ vs. $T$ with a Curie-Weiss
law, $M \propto 1/T - \theta$ with a large negative $\theta$ on the order of 100 K indicating the
presence of strong antiferromagnetic (AF) exchange interactions. Since the expecta-
tion for $a$-YIG is that the nearest-neighbor environment is largely unchanged from
the crystalline state, local AF interactions are reasonable, though existing reports
disagree on this issue. [4, 5] The lack of long-range order gives rise to frustration,
pushing $a$-YIG toward spin glass or more complex non-equilibrium behavior. Here
one expects strong AF correlations between neighboring spins up to a temperature
scale comparable to the bulk transition temperature, with lower temperature freezing
phenomena that depend on the balance of the competing interactions in a particular
structure.

Using techniques previously shown to produce high-quality epitaxial YIG films
when the proper crystalline substrate was used and the proper post-annealing was
conducted,[86] we sputtered 100 nm and 200 nm thick films of $a$-YIG on amor-
phous silicon-nitride ($a$-Si-N) coated Si substrates and also on $a$-Si-N thermal isola-
tation platforms[87] developed for thermal and thermoelectric characterization of thin films and nanostructures.[88, 89] Figure 5.1 shows x-ray diffraction (XRD) patterns comparing a polycrystalline bulk YIG sample (a)) to an a-YIG film on the Si-N coated Si substrate (b)). The former indicates randomly oriented polycrystalline YIG, whereas the latter exhibits no medium- or long-range order in the YIG layer. We also performed magnetization measurements of a similar a-YIG sample on a Si substrate via SQUID magnetometry. $M$ vs. $T$ (after subtraction of backgrounds from the substrate and sample mount as described in Supplemental Materials) shows a broad peak near 50 K described in literature[4, 5] and a second, not previously observed feature near 230 K discussed further below.

**5.3.1 Membrane**

Results from the membrane experiments appear in Fig. 5.2. Fig. 5.2a) shows Schematic cross-section of the Si-N platform with 200 nm of a-YIG on 500 nm thick Si-N membrane, with locations of injection (purple) and detection (green) Pt strips indicated. Fig. 5.2b) shows optical micrograph of the thermal isolation platform. Fig. 5.2c) shows false color scanning-electron micrograph depicts the suspended non-local spin transport measurement. Bottom panels of Fig. 5.2d-g) display $V_{nl}$ vs. $I$ for four different base temperatures, $T_o$. In each, an abrupt onset of non-local voltage occurs above 500 $\mu$A ($10 \times 10^8$ A/m$^2$), with positive $V_{nl}$ developed for positive $I$ and negative $V_{nl}$ for negative $I$. As shown in the inset to Fig. 5.2d), a similar pattern was reported for crystalline YIG,[3] though in that case the field must be reversed (red and blue lines) to achieve the opposite polarization of the spin current in the YIG.
Figure 5.2: Non-local spin transport through suspended $\alpha$-YIG. a) Schematic cross-section of the Si-N platform. b) Optical micrograph of the thermal isolation platform. c) False color scanning-electron micrograph depicts the suspended non-local spin transport measurement. Bottom panels d-g) display $V_{nl}$ vs. $I$ for four different base temperatures, $T_o$. 
In the disordered YIG there is no special direction set by the film magnetization, allowing transport in both channels with no external field. The open circles in panel Fig. 5.2f) for $T_o = 300$ K show the result of the same non-local measurement performed on a Si-N structure with no $a$-YIG layer, and is essentially zero for all $I$, as expected. Each top panel shows the concurrent measurement of the temperature of the Si-N island made via an entirely separate thin film thermometer. For large $I$ this $T$ first slows its rise with increasing $I$ then for higher $T_o$ actually cools due to increased heat transport by spin excitations. Note that the open circles in panel Fig. 5.2f) for $T_o = 300$ K result from the non-local measurement performed on a Si-N structure with no $a$-YIG layer, and is essentially zero for all $I$, as expected. Note also that there is a finite but very small amount of charge current leakage through the YIG (resistance from the injector to the detector is always $> 100$ kΩ at room temperatures and much larger at low temperatures) that is always too small to account for the measured non-local voltages (for additional details see Supplemental Materials). Across all four measured base $T$, a non-linear component to $V_{nl}$ with a sign change that rules out simple heating effects, is reminiscent of the pattern seen in the original experiments on magnon spin currents in YIG,[3] keeping in mind that the disordered material has no net magnetization and no preferred direction so that either sense of spin current can propagate. Despite this similarity, which suggests that a non-equilibrium spin population could become self-oscillatory when enough spin-torque is provided by the SHE, the voltages we measure are many orders of magnitude larger. We also reiterate that this large voltage was measured across a distance of nearly 10 microns.
Another extremely unusual feature of the data is seen most clearly in Fig. 5.2f) and g), where the measured temperature of the Si-N island coated with a-YIG actually drops dramatically with increasing $I$. The thermometer is measured using an AC technique, which is very unlikely to suffer interference from the large DC current applied to the Pt strip. This also cannot be due to the Peltier effect,[90] which would be linear with applied $I$, causing heating with one polarity and cooling with the other. We believe this large drop in the temperature of the island, which we have observed on multiple platforms and with different a-YIG thickness, is driven by the addition of a new channel for heat conduction created in the a-YIG in response to the SHE injection of the non-equilibrium spin population. In other words, a non-equilibrium conductance, $K_{\text{spin}}$, is added to the thermal conductance of the leg (as defined in the thermal model of Fig. 5.2g). We estimate this $K_{\text{spin}}$ could exceed the thermal conductance of the a-YIG by more than 2 orders of magnitude (for further details see supplemental materials).

As shown in the inset to Fig. 5.2f) and in supplementary materials, we use 2D finite-element analysis software to estimate the size of in-plane thermal gradients generated in the suspended thermal platform during non-local spin transport. The image depicts $T$ calculated for the condition where $\Delta T = 50$ K between the Si frame held at 300 K and the island thermometer. Since heat is dissipated in the Pt lead that runs along the entire length of the legs of the structure, the peak $T = 360$ K is actually on the leg. In general, the in-plane thermal gradient along the leg has components along both the $\hat{x}$ and $\hat{y}$ directions, with $\nabla T_x$ reaching absolute values near 6 K/mm in the region between the two Pt leads at the peak $T$ location, and with
Figure 5.3: Intentional manipulation of direction of thermal gradient.  

a) Compares $T$ and $V_{nl}$ measured on the island thermometer vs. $I$ applied to the Pt spin injector for the a-YIG coated Si-N membrane structure with and without He exchange gas surrounding the membrane.  

(b) and (d) compare the total non-local voltage measured in the membrane with exchange gas to the a-YIG on the bulk Si substrate. Subtracting the linear term in these plots, as shown in (c) and (e), reveals a similar non-linear signal as seen in the membrane, though with reduced signal size. 

a maximum value of 18 K/mm achieved near the connection to the bulk Si frame. 

Peak values of $\nabla T_y \approx 70$ K/mm along the legs occur in a similar region. These much larger gradient areas could dominate the additional heat-sinking via spin excitations that drives the overall cooling of the suspended island. Finally, we note that $\nabla T_x$, which we hypothesize plays a role in increasing the non-local voltage signal, actually varies in magnitude and sign across the structure, suggesting that similar devices optimized to produce large and uniform $\nabla T_x$ could lead to even more dramatic spin transport effects in suspended a-YIG.
5.3.2 Membrane vs. Substrate: Manipulating Thermal Gradient Direction

Figure 6.3 describes two different approaches to manipulate the direction of the applied thermal gradient in the non-local spin transport experiment. First, we compare $V_{nl}$ measured in vacuum as in Fig. 5.2 with the signal measured on the same sample but with helium gas added to the cryostat to thermally short the Si-N structures to the sample environment. As is clear from the measured $T$ as a function of $I$ shown in Fig. 6.3a), in-plane gradients are nearly entirely eliminated, and the dominant gradient is normal to the interface of the heated Pt strip and the gas and therefore very similar to the situation when the Pt/a-YIG is supported on a bulk substrate. As shown in Fig. 6.3b), this reduces the size of the non-local signal. Despite the reduced size, both a linear and a non-linear term remain easily measurable when only an out-of-plane gradient exists as shown in Figs. 6.3c-f). Figs. 6.3c) and d) show the total measured $V_{nl}$, while e) and f) show the signal after subtraction of the linear term (determined via least-squares fit to the small $I$ region) in order to examine non-linear contributions. Note that in both experiments, where the exciting Pt strips have very different width, the non-linear $V_{nl}$ turns on at similar current density, $10 \times 10^8 \text{ A/m}^2 < j < 20 \times 10^8 \text{ A/m}^2$. This large difference in magnitude of $V_{nl}$ between in-plane gradient and out-of-plane gradient cases could relate to the presumed large difference in the absolute magnitude of thermal gradients produced in the two experiments. However these comparisons are complicated by the difficulty in estimating out-of-plane gradients when the constituent materials’ thermal properties and nature of the interfaces between them are poorly known.
Here we can use FEM to roughly estimate a value near 0.8 K/mm for the out-of-plane gradient, with negligible in-plane gradients on distances greater than even one micron away from the Pt current strip. In our view the most reasonable assumption is that our experiments on the substrate do not involve significant thermal gradients, and instead probe purely electrical spin generation, transport, and detection though further experiments are required to confirm this.

5.3.3 Temperature and Distance Dependence

Though the largest effects come on the membrane, there the exact temperature of the a-YIG transporting spin is difficult to discuss. In light of this we explore the $T$- and $L$-dependence of the effect in detail using the substrate-supported case, as shown in Figs. 6.5 and 6.4. Fig. 6.5a) shows the component of $V_{nl}$ purely linear in $I$ (determined from fits to the slope of $V_{nl}$ vs $I$ at each $T$) for both 100 nm and 200 nm thick a-YIG films. Fig. 6.5b) shows the maximum recorded value of the non-linear component (here taken at $I = 8$ mA), $V_{nl,max}$ vs. $T$. Both components become measurable only above $\sim 230$ K. Fig. 6.5c) indicates that this temperature correlates with the disappearance of spin freezing in the a-YIG. Here we plot $\Delta m$ vs $T$, the component of magnetization due to the a-YIG film deposited on a Si-N coated Si substrate (isolation of this component from total measured SQUID magnetization is described in supplementary materials) for both zero-field-cooled (red symbols) and field-cooled (black symbols) states using a magnetic field of 5000 Oe. In contrast to existing literature on a-YIG, we see splitting of these curves at two temperatures, near the expected 50 K peak in the ZFC curve, and at a temperature nearly equal to
Figure 5.4: $V_{nl}$ vs. $T$ from 5 to 300 K indicating spin transport through 100 and 200 nm thick $a$-YIG on the substrate. a) Linear component (slope) of $V_{nl}$. Inset: Schematic view of the non-local experiment. b) Maximum non-linear $V_{nl}$ provides an estimate of the component potentially related to ST-driven spin excitations. Inset: Optical micrograph of isolation platform frame showing the location of the substrate-supported non-local measurement. c) Magnetization of the $a$-YIG vs. $T$ from 5 K to 300 K cooled in zero field (ZFC, red symbols) and in the 5000 Oe measuring field (FC, black symbols).
the observed onset of spin transport effects. This suggests that spin transport occurs in the presence of disorder and strong spin correlations but only when sufficient thermal energy is available to overcome spin freezing.

As seen in non-local spin transport in crystalline YIG, Figure 6.4 indicates a sharp drop in spin signal with increased separation between Pt strips, $L$, for both the linear and non-linear components of $V_{nl}$. These data do not fit a simple exponential dependence. We require more data to effectively probe existence of diffusive and relaxation regimes\cite{59}, and more detailed examination of separation dependence is ongoing. We are able to clarify that any thermal component to $V_{nl}$ here is small, and has a different dependence on $L$, further evidence that electrical effects dominate spin transport in the experiment on the substrate.

5.3.4 Applied Magnetic Field Dependence

In disordered spin systems, even above any freezing temperature, strong AF spin correlations typically lead to small magnetic susceptibility and very large saturation fields. This is the case for $\alpha$-YIG, where $M$ is a very small fraction of either the saturation magnetization of crystalline YIG or of the even larger estimated $M$ of free Fe atoms at the same density. Despite achieving a magnetization less than 10% of the YIG value (described further in supplemental materials), as shown in Fig. 6.6 there is an observable effect of applied field on $V_{nl}$. Figure 6.6a) shows $V_{nl}$ as a function of applied $I$ for the substrate-supported $\alpha$-YIG film, here measured in air at room temperature. Fig. 6.6b) isolates the nonlinear component, which is near zero for $I \leq 2$ mA. We applied fields up to 14 kOe perpendicular to the film, large enough
Figure 5.5: Distance-dependence of $V_{nl}$ on the substrate. a) Voltage components related to spin drop off sharply with distance. Here blue symbols indicate slope (right axis) and black symbols maximum non-linear component (left axis). Stars, boxes, and circles indicate three different samples (Insets Nonlinear spin signals after linear subtraction show clear effects even for $L > 100 \mu m$.) b) Estimation of the (small) heating effects drops off much more slowly, reinforcing that the spin signals are not simply temperature driven but require SHE excitation.
Figure 5.6: Dependence of $V_{\text{nl}}$ on applied field. Here $H$ up to 14,000 Oe was applied perpendicular to the substrate as shown inset in a), which shows $V_{\text{nl}}$ vs. $I$, here measured in ambient conditions, and displaying the same linear and non-linear contributions as earlier Figs. b) The non-linear component isolated by subtraction of the linear term. These clarify that when biased at $I = 2$ mA the signal is dominated by the linear term, where at 8 mA the non-linear term contributes. Panels c) and d) show that at both bias points, clear field dependence is observable, with similar relative magnitude and trends.
to have saturated $M$ and completely eliminated spin transport in crystalline YIG.[60] Figs. 6.6c) and d) show $\Delta V_{nl}/V_{nl}$ vs. $H$ for $I = 8$ mA and $I = 2$ mA, respectively, and show that both the linear and non-linear regimes react to $H$ in a similar manner as expected if the field dependence arises from magnetic-field dependent properties of the medium. Here $\Delta V_{nl}/V_{nl} = (V_{nl}(H) - V_{nl}(H = 14 \text{ kOe}))/V_{nl}(H = 14 \text{ kOe})$.

Reduction from maximum $H$ does increase the signal, with the zero field values slightly reduced from a peak that occurs at intermediate fields. The slight asymmetry in the peak value when starting from either value of maximum field is likely due to error on the subtraction procedure. The small shifts in $V_{nl}$ are consistent with the small shift in total magnetization achieved here. Despite the small size, this field dependence is strong evidence that $V_{nl}$ for $a$-YIG relies on spin transport.

5.3.5 Pt leads grown on top of $a$-YIG

A second set of devices were fabricated using Si-N ‘blanks’ where Pt leads were deposited on top of the $a$-YIG film. Growth on top of film lead to a different Pt/$a$-YIG interface. $V_{nl}$ vs. $I$ can be seen in Fig. 5.7 for 20 nm thick Pt leads grown on top of 200 nm thick $a$-YIG with a 20 $\mu$m separation. Here we see a non-linear contribution on the same order of magnitude as our previous results with Pt leads under the $a$-YIG. The linear contribution is greatly reduced however, most likely due to the different interface.

We also present temperature dependent $V_{nl}$ from 200 to 320 K. In this case, we observe a severe drop in spin transport signal with signals sizes becoming unmeasurable at 250 K. This is slightly higher than the previously presented data with
Figure 5.7: Non-local spin transport through 200 nm thick $a$-YIG on a substrate with leads grown on top of film. $T_0$ of 295 K with a 20 $\mu$m separation between leads. Total strip length of $\sim$ 3 mm.
Figure 5.8: $V_{nl}$ vs. $T$ from 200 to 330 K indicating spin transport through 200 nm thick $a$-YIG on a substrate with leads grown on top of film. $V_{nl}$ includes both linear and non-linear component at $I=8$ mA ($1 \times 10^{-10}$ A m$^{-2}$).

leads underneath ($\sim 230$ K). We believe this to be due to the slightly increased lead separation (20 $\mu$m vs. 10 $\mu$m). As with data presented earlier in this chapter, signal sizes decrease dramatically with increased separation between Pt strips for both linear and non-linear components of $V_{nl}$.
5.3.6 Lead Material: Pt vs. Cu

To help rule out charge leakage a device with Cu leads was measured using the same method as detailed above. A current is driven down one lead and a non-local voltage is measured at a second electrically isolated lead. The lead width is 40 µm, the lead separation is 40 µm and the overall length of the leads are ≈3 mm. The data for both 300 K and 380 K can be seen in Fig. 5.9 and Fig. 5.10, respectively. For the 300 K data, the Pt shows a very slight non-linear signal with a linear term indistinguishable from the background. The measured voltage for Cu at 300 K shows mainly background drift from the instrumentation. Once the temperature is raised to 380 K, the Pt leads show a clear spin transport signal due to a SHE driven spin affect in the $a$-YIG. Both a non-linear and linear component are present at 380 K using Pt leads. Cu leads show zero spin transport signal at this temperature.

If we compare Figures 5.9 and 5.10 for Pt leads grown on top with previous data presented for Pt leads underneath the $a$-YIG layer a pattern emerges. The non-linear component appears to be similar in size for both sets of devices but the linear component is much smaller in the case of the lead material grown on top, which we believe is due to an interface effect. Further analysis of the interface between the Pt lead and $a$-YIG layer is needed.
Figure 5.9: Total non-local voltage vs. $I$ for both Pt and Cu leads at a base temperature of 300 K. Total strip length of $\sim 3$ mm for both materials.
Figure 5.10: Total non-local voltage vs. $I$ for both Pt and Cu leads at a base temperature of 380 K.
5.4 Results and Discussion for \( \alpha\text{-Cr}_2\text{O}_3 \)

With successful spin transport through \( \alpha\text{-YIG} \) we sought a second disordered magnetic material. \( \text{Cr}_2\text{O}_3 \), or chromia, is an antiferromagnet up to its Néel temperature of 307 K in its bulk crystalline form\[91\] and has a relatively complicated unit cell.\[92\] Recent work on spin transport in chromia films have focused on its crystalline form.\[93, 94\] Our goal was to perform similar measurement on \( \alpha\text{-Cr}_2\text{O}_3 \) as in the \( \alpha\text{-YIG} \) films.

5.4.1 Membrane

Results for membrane data on \( \alpha\text{-Cr}_2\text{O}_3 \) can be seen in Fig. 5.11. Here we present data with a base temperature, \( T_0 \), of 300 K. A non-linear component to \( V_{nl} \) is similar to data seen for \( \alpha\text{-YIG} \) in Fig. 5.2, with a similar sign change that rules out any heating affect. Due to the disorder there is no net magnetization and no preferred direction so that spin transport can propagate in either direction. As with the \( \alpha\text{-YIG} \), \( V_{nl} \) values we measure are orders of magnitude larger previously seen across distances up to 10 microns.

We also observe a shift away from the expected parabolic term in temperature as \( I \) reaches its max value. The expected behavior for heating of the Si-N membranes can be seen in Fig. 5.2f, where a clear parabolic contribution is do to joule heating as \( I \) is increase in both directions. For the case of \( \alpha\text{-Cr}_2\text{O}_3 \), a slight reduction from a parabolic dependence can be seen. We believe the slight deviation in the temperature is due to the addition of a new channel for heat conduction created in response to the
SHE injection and subsequent non-equilibrium spin population. This deviation is not as aggressive for $a$-Cr$_2$O$_3$ as it is for $a$-YIG possibly due to it’s different structural ordering in this temperature range. Work is currently being performed to understand the magnetic ordering of the material but is unavailable at the time of this writing.

5.4.2 Substrate

Fig. 5.12 plots total non-local voltage vs. current for $a$-Cr$_2$O$_3$ on bulk Si substrate at room temperature. The voltage signal here shows a parabolic heating term and no spin transport signal. At room temperature we believe we are below the spin “freezing” temperature of $a$-Cr$_2$O$_3$. To achieve a spin current, we increased the base temperature of to 380K, which can be seen in Fig. 5.13. Fig. 5.13 clearly shows a linear and non-linear component to the total voltage. At this elevated temperature the non-linear component is on order the same signal size as seen in $a$-YIG. The linear component is greatly reduced compare to the $a$-YIG however, which could give us insight into the spin propagation mechanism in the two disordered systems. To better understand this information, further work is required on understanding the $a$-Cr$_2$O$_3$ magnetization which is currently ongoing as of this writing.
Figure 5.11: Non-local spin transport through suspended $\alpha$-Cr$_2$O$_3$. Bottom panel shows $V_{nl}$ vs. $I$ for base temperature, $T_0$, of 300 K. Top panel shows concurrent measurement of the temperature of the Si-N island made using an entirely separate thin film thermometer.
Figure 5.12: Total non-local voltage vs. current for $a$-Cr$_2$O$_3$ on bulk Si substrate at base temperature of 300 K. Thermal gradient direction is perpendicular to the film normal. A small negative linear contribution from instrument drift was subtracted from this data.
Figure 5.13: Total non-local voltage vs. current for $a$-Cr$_2$O$_3$ on bulk Si substrate at base temperature 380 K. Thermal gradient direction is perpendicular to the film normal. To achieve this plot, the 300 K data from Fig. 5.12 was treated as a background signal and subtracted from the raw data.
5.5 Additional Background Measurements: SiO$_2$, CNT

In addition to the “blank” Si-N membrane measurement shown in Fig. 5.2c), we verified the absence of a spin signal in two other films, an insulating (but non-magnetic) 100 nm sputtered SiO$_2$ film and a 90 nm doped carbon nanotube (CNT) network film that we previously characterized in detail for thermal, electrical, and thermoelectric properties.[6] Results from non-local transport experiments on these samples are shown in Fig. 5.14. As in Fig. 5.2 the top panel shows temperature measured by the separate island thermometer, while the lower panel shows the measured non-local voltage, $V_{nl}$, both as a function of applied current, $I$, to the injector strip. The resulting heating (free from non-monotonic behavior associated with spin excitations, as was the bare Si-N platform) is nearly identical between the two platforms.

In both platforms, small non-local voltages are measurable, though orders of magnitude smaller than the signals seen in $a$-YIG and with very different dependence on $I$. $V_{nl}$ for the SiO$_2$ film shows a roughly linear behavior, possibly due to a drift of the base temperature for this measurement. $V_{nl}$ for the CNT film is predominantly proportional to $I^2$ with a constant offset voltage, obvious from the $I = 0$ value. Both these terms have a simple thermoelectric origin, with the constant offset due to the small temperature difference between the sample region and the room temperature electronics, and the $\propto I^2$ term likely due to a thermal gradient that develops in the platform as current is driven down the Pt injector strip. We previously measured
Figure 5.14: a) Temperature and b) Non-local voltage, $V_{nl}$, vs. current applied to the Pt injector strip as described in the main text. Boxes show results for a thermal isolation platform coated with a 100 nm-thick film of sputtered SiO$_2$, and circles for a platform coated with a 90 nm-thick doped carbon nanotube network thin film (described in more detail elsewhere[6]). The fill color indicates the temperature in both panels, and the same color scale is used for all data sets.
Figure 5.15: The same two data sets shown in Fig. 5.14 compared on a larger scale to the $\alpha$-YIG non-local data for the membrane with He exchange gas, and in vacuum. This clarifies the small scale of the background voltages in Fig. 5.14, even compared to the smaller, largely linear spin signal generated when the in-plane thermal gradient is cancelled. This plot also clarifies the small $V_{nl}$ term $\propto I$ in the $\alpha$-YIG with an in-plane thermal gradient, which is not visible on the full scale plots shown in Fig. 5.2.
the Seebeck coefficient of this CNT film to be \( S \approx 70 \, \mu\text{V/K} \) in this temperature range,[6] suggesting that a total temperature difference across the sample region of approximately 0.2 K develops at the maximum applied \( I \). This is \( \approx 0.3 \% \) of the maximum temperature of the island, likely due to small asymmetries that exist in the thermal platform or the sample itself, and is in line with background signals seen in measurements of other transverse thermovoltages.[89, 95] Note that this CNT film also has an easily measurable *electrical* conductivity, such that the charge resistance between the two Pt strips is \( \sim 100–200 \, \Omega \). This much more efficient charge transport channel than exists in the \( a \)-YIG does NOT lead to the sign-reversing signal we identify with spin transport. To clarify this, we plot \( V_{\text{nl}} \) for these two backgrounds with the data for suspended \( a \)-YIG both in vacuum and with exchange gas in Fig. 5.15. Here even the comparably smaller linear term that dominates in \( a \)-YIG nonlocal transport when the in-plane thermal gradient is cancelled is much larger than either background measurement. When a significant in-plane thermal gradient is applied, the non-linear part of \( V_{\text{nl}} \) dominates and is many orders of magnitude larger (Fig. 5.15 shows only a small portion of this data).

## 5.6 Consideration of Leakage Current in \( a \)-YIG

Though the CNT data is strong evidence that a conducting path between Pt strips does not lead to the signals we associate with spin transport, we also checked carefully for any significant charge conductance through the \( a \)-YIG film. Figure 5.16 details 2-point resistance measurements between metal features on the thermal isolation
Figure 5.16: a) Two-point resistance measurements through the 200 nm thick $\alpha$-YIG layer deposited on the thermal isolation platform between various metal features all show very large values of resistance, and all show the same roughly exponential behavior as a function of $T$. At 200 K and below $R$ became too large for the standard digital voltmeter used here to measure. These results are all consistent with charge flow through a large band gap semiconducting layer, taking the geometry into consideration. b) Two-point resistance as a function of separation for the strips used in Fig. 6.4 shows the simple, roughly linear increase with separation expected from the geometry.

platform for various temperature. All of these values are orders of magnitude larger than the “leakage” path present in the CNT film discussed above. To compare to measured non-local voltages generated by spin transport, one can calculate $R_{\text{spin}} = V_{\text{nl}}/I$ for the linear and non-linear components to $V_{\text{nl}}$. These values fall in the range of $10^{-1} - 100 \, \Omega$, many orders of magnitude less than measured charge resistance.

To further eliminate the possibility of leakage currents or Schottky barrier effects contributing to non-local voltages we measure here, we have directly measured current flowing between the Pt strips under large bias voltages. As the application of large currents to the Pt injector, with $R \sim 1000 \, \Omega$ in the case of the substrate-supported experiment, causes a large voltage drop across the entire length of the injector wire,
Figure 5.17: Comparison of a) voltage-biased, measured current and b) standard current-biased non-local voltage (right) measurements. Both a) and b) measurements are on the same Pt strips and are on the substrate. Dashed lines in both main plots indicate a linear fit to the data near zero current, extrapolated across the range of data. The top plots show the deviation from this linear curve by subtracting it from $V$. A very small non-linearity under large voltage bias changes the $\approx 650 \, \text{k}\Omega$ effective resistance between the Pt strips by $< 1\%$. This small non-linear effect, likely the result of Schottky barriers formed at the Pt/a-YIG interface, cannot explain any of the non-local signals. c) The voltage-biased measurement for the Si-N membrane shows similarly tiny non-linearity compared to the very large effects under current bias.
some portion of the $a$-YIG between the Pt strips experiences an effective bias voltage up to potentially 8 V. Here we perform a more rigorous check, by applying $V_{\text{bias}}$ up to 8 V between the strips across their entire length. In Fig. 5.17a we plot measured current, $I_{\text{meas}}$ against $V_{\text{bias}}$ (with measured current on the $x$-axis) to simplify comparison to the non-local IV curves for the substrate-supported Pt/$a$-YIG. We do see a very small non-linearity under voltage bias, likely indicating the presence of Schottky barriers at the Pt/$a$-YIG interface. However, as the overall effect here is to shift the effective resistance (the slope of this curve) by $< 1\%$ at large voltage biases, this cannot explain the much larger non-linear voltage components seen when large bias current drives spin transport in the $a$-YIG.

We note that though simplistic network models or finite-element calculations do allow that linear voltage components on the order of mV or less are possible in the non-local measurement due to leakage of charge through the $a$-YIG, there are two pieces of strong evidence against a charge-leakage origin for the linear term. The first is that the linear component drops off dramatically with distance, while as shown in Fig. 5.16b, the measured resistance through the $a$-YIG due to charge effects goes as expected from a simple increased length of the current path approximately linearly with distance. Specifically, the slope of the IV curve drops by a factor of more than 20 between the 10 $\mu$m and 110 $\mu$m separations, while the $a$-YIG resistance increases by less than a factor of 3. The second piece of evidence that argues against a charge leakage explanation, as described in more detail in the main text is the field-dependence of both the linear and non-linear terms.
We have also tested the case of voltage bias as a function of temperature for the experiments on \(a\)-Si-N membranes. Fig. 5.17c) shows two voltage-bias experiments for two different membrane temperatures (where heat is added to the island heater for the 380 K data). Here current-biased experiments are not shown, but result in the very large non-linear contributions apparent in Fig. 5.2. Again under voltage bias we see very nearly linear responses, with non-linearly much less than 1%. The leakage resistance is temperature dependent, but no large non-linear terms appear even at elevated temperatures. We can furthermore again rule out any contribution of voltage leakage as a complete explanation of the linear term in non-local experiments with large current biases. Consider that the leakage resistance through \(a\)-YIG drops by \(\sim 60\%\) between 300 and 380 K, where the linear term in non-local measurements with large current bias increases by more than a factor of ten. The non-linear voltage under current bias at a membrane temperature of 380 K is 350× larger than the linear term, while the charge leakage under voltage bias changes by < 1%. This clarifies that neither the linear nor non-linear components of the non local voltages arises from charge transport through the \(a\)-YIG.

5.7 SQUID Magnetometry of \(a\)-YIG films

We measured magnetization as a function of temperature of \(a\)-YIG using a Quantum Design MPMS SQUID Magnetometer. As stated in the main text, 200 nm thick \(a\)-YIG was simultaneously deposited on Si-N thermal isolation structures and 1 cm × 1 cm Si-N coated Si substrates. One of these substrates was cut into
Figure 5.18: Moment vs. $T$ in 5000 Oe applied field for both FC (black box) and ZFC (red circle) states. The blue line includes three terms, with two background contributions as described in associated supplemental text. Here $B = 1.73 \times 10^{-8}$ emu/K and $C = 80.3$ $\mu$emu, these two terms are subtracted from both FC and ZFC data for the plot in the main text.
smaller pieces, placed in a gelatin capsule, held in place using cotton batting, and mounted in a drinking straw that was mounted in the magnetometer. $M$ vs $T$ for first zero-field-cooled and then field-cooled conditions was measured from $5 - 300$ K in 5000 Oe, an applied field much smaller than the typical exchange energy of the AF coupling in a-YIG. The raw voltage was converted to moment using typical fitting procedures, and the resulting raw moment is shown in Fig. 5.18. Despite presence of both a significant temperature-independent paramagnetic background and a smaller, linear in $T$ paramagnetic background, all essential features of the a-YIG magnetization are obvious. The large $T$-independent offset is most likely due to the background from the cotton batting,[96] though more quantitative background determination is required to rule out an origin in iron clusters in the a-YIG itself that are too small to be observed in XRD as nanocrystallites. The linear background term we associate with the temperature-dependent Van Vleck paramagnetism of the semiconducting Si substrate.[97, 98] The plots in the main text subtract these two background terms, which gives the expected $M \propto 1/(T - \theta)$ dependence above the 50 K freezing temperature for the FC curve with $\theta$ on order of $-100$ K.

Figure 5.19 shows estimated magnetization vs. $H$ for the a-YIG film. The main plot compares the upper bound of $4\pi M$ for the a-YIG to the expected value for high-quality bulk and thin films of ordered YIG. This upper bound is given by converting the entire measured moment of the sample, substrate, and mounting materials using the volume of the film. $4\pi M$ determined this way is at least an order of magnitude below the YIG value up to at least 5000 Oe (well past the expected in-plane saturation), and remains linear with $H$ with large susceptibility, indicating no
Figure 5.19: Estimated $4\pi M$ vs. $H$ at 300 K. Even taking the entire measured moment as the contribution of $\alpha$-YIG (which is very unlikely but provides an upper limit) results in overall magnetization a small fraction of that commonly seen in ordered YIG.
approach to saturation, which is a common feature of disordered magnetic systems even far above the freezing temperature.\[99, 100, 101\] In such systems, very strong AF interactions are common, and here we expect the field required to approach saturation to be well out of the scale of typical laboratory superconducting magnets.

5.8 Finite Element Thermal Modeling

We performed finite element modeling in 2d using a common commercially available software package \[102\]. This package allows solution of the 2d heat flow equation (for our purposes limited to steady-state):

\[
\frac{\partial}{\partial x} \left( k_{2D} (x,y) \frac{\partial T(x,y)}{\partial x} \right) + \frac{\partial}{\partial y} \left( k_{2D} (x,y) \frac{\partial T(x,y)}{\partial y} \right) = P_{2D} (x,y), \tag{5.8.1}
\]

where \( k_{2D} = k \cdot t \) with \( k \) the thermal conductivity (in W/mK) of the constituent materials. In the case of models of the essentially 2d suspended structures, \( t \) is taken to be the known thickness of each film. Where two films overlap, \( k_{2D} \) is the sum of both contributions. We also estimate out-of-plane gradients for the sample-on-substrate case by taking \( t \) to be a uniform thickness (here 1 \( \mu \)m) of the hypothetical cross-section. As long as the heat flow is dominated by the bulk substrate so that in-plane thermal transport is negligible on long length scales, such a model gives a reasonable estimate of the out-of-plane thermal gradient at the Pt/a-YIG interface. To match our experimental conditions for the cross-sectional simulations (sample in vacuum, with substrate clamped at the bottom to a thermal bath), we choose the Dirichlet boundary condition at the base of the Si substrate (fixing \( T = 300 \) K),
Figure 5.20: Calculated thermal profile in the suspended platform. **Upper Right:** Plot of $T(x, y)$ resulting from the 2d FEM simulation. Heating is in response to the large current density passed through the Pt injector strip as described in the main text. Dashed, colored lines indicate the locations of the 1d plots. **Lower Right:** $T$ vs. $y$ and $dT/dy$ vs. $y$ along the leg between the Pt injector and detector strips. **Upper Left:** $T$ vs. $x$ and $dT/dx$ vs. $x$ along the center of the bridge connecting the two islands. **Lower Left:** Zoomed view of the $T$ vs. $x$ and $dT/dx$ at the point of maximum $T$ along the leg. Shaded boxes indicate the locations of Pt strips and Si-N/a-YIG areas, where the thermal gradient is largest as expected.
and Neumann boundary conditions elsewhere with no radiative or convective heat flow. The 2d models of the membrane assume the Si frame is clamped to the base temperature, and Dirichlet boundary conditions are used at all edges of the Si-N structure.

Values of the thermal conductivity of the Pt lead are estimated from the Wiedemann-Franz law, and a-YIG by using (low) values taken from the thermal platform measurements. For the Si-N underlayer, which is critical for realistic modeling, we take the value $\sim 3 \text{ W/m K}$ that we measure frequently for this Si-N using the suspended Si-N platforms[87], and use literature values for Si thermal conductivity ($\sim 2000 \text{ W/m K}$) [103]. For simplicity we use temperature-independent thermal conductivity (which is most likely a good assumption for the sample-on-substrate models, but could introduce inaccuracy for the thermal platform measurements where large temperature differences occur), and also make the simplifying assumption that all Joule heat is dissipated evenly in the injecting Pt wire.

For the membrane measurements we set $P_{2d}$ dissipated in the injector strip by matching the temperature to that measured by the island thermometer. For substrate measurements, the known current applied is converted to the appropriate volumetric power dissipation. The FEM problem is then solved using an adaptive mesh with $> 5000$ nodes. The resulting solution for $T(x, y)$ for the membrane is shown in Fig. 5.2f) and in greater detail in Fig. 5.20. Values from this solution are exported and a numerical derivative of this curve as a function of the appropriate dimension gives the thermal gradient.
5.9 Estimation of Spin Thermal Conductance in \( a\)-YIG

We can estimate the size of this new heat pathway by comparison to the bare Si-N thermal isolation platform. There the effective thermal conductance of the legs connecting the central Si-N island to the thermal bath is typically \( K_{L,\text{eff}} = P/\Delta T \sim 2.4 \mu W/K \) near 300 K, where \( P \) is the power dissipated and \( \Delta T \) the resulting temperature difference across the leg. Achieving \( \Delta T \simeq 150 \) K as shown for the bare Si-N platform in Fig. 5.2f) then requires an average power dissipation of \( P \simeq 360 \mu W \). The reasonable assumption that the same applied \( I \) in the \( a\)-YIG coated platform causes the same average applied \( P \) allows the estimation for the much smaller temperature difference caused by the addition of the spin excitation thermal conductance channel once dynamics are excited in \( a\)-YIG such that \( K_{L,\text{eff}} = (360 \mu W)/(70 \text{ K}) \simeq 5 \mu W/K \), suggesting the spin excitations contribute a roughly equal heat conduction to the existing Si-N leg with its Pt leads. Simple estimates based on the \( a\)-YIG film geometry indicate a spin thermal conductance \( k_{\text{spin}} > 100 \) W/m K, orders of magnitude larger than the \( \sim 1 \) W/m K total thermal conductivity of the \( a\)-YIG film seen in our measurements with no applied charge current in the Pt strip, and on the order of electronic thermal conductivities seen in polycrystalline metal films.[88]
5.10 Conclusion

The recent theories that explain spin transport in antiferromagnetic insulators[78, 79, 80] invoke a well-defined antiferromagnetic magnon spectrum that is either absent or substantially modified in the case of a truly disordered systems as we use here. The magnon spectrum of disordered magnets has been rarely explored in the past, though existing work suggests an analogy to phonon spectra in glassy systems.[104] Vibrational modes of amorphous systems certainly exist, and a long history of study shows that whether called a phonon or given a more specific name (such as propagon), heat transport via a broad spectrum of vibrational excitations is possible in amorphous systems [105]. Recent work[106, 107, 108, 87] shows that this transport is often surprisingly efficient, with long phonon mean free paths despite the disorder. Our work is the first indication of similar effects for spin transport via magnetic correlations in a disordered system.

In fact, use of a disordered system has potential advantages for magnonics. Two traditional challenges for magnonic materials are the presence of a gap in the magnon spectrum and the highly anisotropic nature of the magnon transport introduced in a crystal [109]. Neither should occur in a disordered system. A central question is if spin transport effects in disordered systems persist over long enough length scales to be useful technologically. The data shown here proves emphatically that they do. The easy compatibility of the $a$-YIG and $a$-Cr$_2$O$_3$ materials in any device process is also compelling, suggesting a potential paradigm shift in materials science for magnon transport.
Chapter 6

Relation of Planar Hall and Planar Nernst effects in Thin Film Permalloy

6.1 Introduction

Recent years have seen an intense effort to understand the interplay of thermal, electronic, and spin degrees of freedom in a wide range of nanoscale magnetic systems and devices. This new field of spincaloritronics continues to expand, driven by the promise of new potential routes to energy harvesting, information storage, and logic devices enabled by spin.[110, 111, 112] Though significant effort in the field now focuses on interactions of magnons and electrons at an interface between a magnetic insulator and a non-magnetic metal with strong spin-orbit coupling [113, 114, 115,
interest in thermal generation of spin currents in purely metallic systems remains high [117, 118, 119, 120]. Thermal gradients applied to metallic ferromagnets have by now been confirmed to generate spin accumulation and pure spin currents only when heating is applied on a very short length scale comparable to the spin diffusion length in the metallic ferromagnet [121, 122, 123, 124, 125, 126, 127, 21, 128], or in experimental configurations that rely on magnon spin transport over somewhat longer distances [129]. Experiments that probe thermal gradients on much longer length scales, especially when a thin film ferromagnet is heated on a bulk substrate, have proven to produce signals dominated by traditional magnetothermoelectric effects. Depending on the exact orientation of the thermal gradient on the film at the location of the voltage probes, these effects can involve the planar Nernst effect, the anomalous Nernst effect, or a combination [130, 89, 131, 132, 133, 134, 135, 136, 137, 138, 139, 140, 141, 142]. Here the emphasis on the transverse effects (the Nernst effect being the thermal analog to the Hall effect) comes since the inverse spin Hall effect (ISHE) [54, 55, 56, 22] is typically used to probe the presence of spin currents, such that the signal of interest should be a voltage transverse to the applied thermal gradient.

Since control of the direction of the thermal gradient is so critical in identifying the physical processes that produce transverse voltages when metallic FM thin films are heated, we have pioneered thermal isolation platforms where a 500 nm thick free-standing silicon-nitride membrane replaces the bulk substrate beneath the FM.[87, 143] This effectively confines the thermal gradient to the plane of the thin film FM sample deposited on the membrane. Our first experiments designed to probe thermal
generation of spin currents when an exclusively planar thermal gradient is applied to permalloy (Py, the nickel-iron alloy with 80% nickel content) and nickel thin films showed no sign of spin currents.[89] All magnetic-field dependent effects instead showed the symmetry of the planar Nernst effect (PNE),[144, 145] as confirmed by other groups using similar suspended structures.[131, 137, 142] Further work also showed the expected tight link between the magnetic field dependence of the standard Seebeck effect and the PNE,[146] though these studies left several open questions, including the physical origin of a scaling factor needed to explain the total signal size and the cause of a magnetic field-independent background transverse voltage.

Metallic ferromagnets show several important responses to currents and thermal gradients, which often share a common origin and are related by simple expressions. In the Seebeck effect, a longitudinal thermal gradient, \( \nabla T \) applied to a conducting sample along the \( x \)-direction excites phonons and electrons that transport energy through the film. When no steady-state current can flow through the sample, charge flows only until the electric field balances the heat flow through the film such that \( E_x = -\alpha_{xx} \partial T / \partial x \). If the thermal gradient is uniform between the voltage measurement leads separated by \( \ell \), then \( \Delta V = E_x \ell, \partial T / \partial x = \Delta T / \ell \), and the longitudinal thermopower or Seebeck coefficient is given by \( \alpha = -\Delta V / \Delta T \) with \( \Delta T \) the temperature difference across the sample. Furthermore, the Seebeck coefficient is related to the electrical resistivity of the sample, \( \rho \), via the Mott equation:

\[
\alpha = -\frac{\pi^2 k_B T}{3e} \frac{1}{\rho} \left[ \frac{\partial \rho}{\partial E} \right]_{E=E_F}.
\]  

(6.1.1)
The interaction between conduction electrons and sample magnetization adds additional electrical and thermoelectric effects in ferromagnetic metals. One example of this interaction is the anisotropic magnetoresistance (AMR), where spin-dependent spin-orbit scattering generates a change in $\rho(H)$ that depends on the angle of the magnetization with respect to current flow that is even in applied field $H$. Examination of Eq. 6.1.1 indicates that the field-dependence of $\rho$ will be reflected in $\alpha$.

In addition to longitudinal thermopower, ferromagnetic conductors exhibit transverse thermopowers, where a voltage develops in the direction perpendicular to the applied thermal gradient. The anomalous Nernst effect (ANE) and planar Nernst effect (PNE) are thermal analogs to the well-known anomalous Hall effect (AHE) and planar Hall effect (PHE) in FM metals. In these effects, spin-dependent scattering of electrons in the presence of the internal magnetic field of the ferromagnet adds transverse momentum, which leads to voltages in the $\hat{y}$-direction when either current or thermal gradient is applied in the $\hat{x}$ direction. In the ANE a magnetic field applied perpendicular to the plane of a sample and a $\vec{\nabla}T$ in the plane of a sample generates an electric field transverse to the applied $\vec{\nabla}T$. In contrast to the ANE, the PNE depends on the angle between the in-plane sample magnetization and $\vec{\nabla}T$. The PNE coefficient is defined by [147]

$$\alpha_{\text{PNE}}(H) = \frac{1}{2}[\alpha(H_{\parallel}) - \alpha(H_{\perp})]\sin 2\theta. \qquad (6.1.2)$$

In this equation, $\alpha(H_{\parallel})$ and $\alpha(H_{\perp})$ are longitudinal thermopower coefficients measured in external fields oriented parallel and perpendicular to the applied $\vec{\nabla}T$. $\theta$ is the angle between the film magnetization, $\vec{M}$, and $\vec{\nabla}T$. The resulting angular depen-
dence of the PNE is proportional to \( \sin 2\theta \). The transverse electric field generated is then \( E_{y,\text{PNE}} = \alpha_{\text{PNE}(H)} \partial T/\partial x \) and again if the thermal gradient is uniform the transverse PNE voltage is

\[
V_{T,\text{PNE}} = \alpha_{\text{PNE}(H)} \frac{\Delta T}{\ell} w, \tag{6.1.3}
\]

where \( w \) is the width of the sample in the transverse direction. Thus the PNE is the thermal analog to the planar Hall effect, where transverse voltage is generated depending on the angle between applied current and in-plane magnetization with coefficient:

\[
\rho_{\text{PHE}}(H) = \frac{1}{2} [\rho(H_{\parallel}) - \rho(H_{\perp})] \sin 2\theta. \tag{6.1.4}
\]

Here, assuming uniform current density, the transverse electric field is \( E_{y,\text{PHE}} = \rho_{\text{PHE}}(H) I/(t \cdot w) \), with the sample thickness \( t \) and width in the transverse direction \( w \) defining the cross-sectional area. The transverse PHE voltage is then

\[
V_{T,\text{PHE}} = \rho_{\text{PHE}(H)} \frac{\Delta T}{\ell} w. \tag{6.1.5}
\]

This shows that just as measurements of longitudinal \( \rho(H) \) allow prediction of the planar Hall voltage, measurements of longitudinal \( \alpha(H) \) allow prediction of the planar Nernst voltage. One powerful feature of our thermal isolation platforms is that all these quantities can be measured on the same sample. If the measured \( V_T \) do not match the expectations from Eqs. 6.1.3 and 6.1.5, this indicates that the simple
assumptions regarding uniformity of current density and/or thermal gradient must be examined.

The ability to measure $\rho_{\text{PHE}}$ and $\alpha_{\text{PNE}}$ on the same sample also allows a unique exploration of the existence of a Mott-like relation between the planar Hall and planar Nernst effects. A transverse Mott relation between the ANE and the AHE has been described theoretically and proven for dilute magnetic semiconductors[148, 149], but the relation for planar transverse effects has not been explored or demonstrated to our knowledge. Our group and others have previously shown that the Mott relation can be demonstrated in metallic FM samples by changing the values of $\rho$ and $\alpha$ via
applied magnetic fields at a fixed $T[146, 150, 151]$. These experiments have been interpreted by some as a demonstration that the energy derivative of $\rho$ in Eq. 6.1.1 is independent of field, and can be used to determine a numerical value for this derivative. However, as we discuss further below, such demonstrations of the Mott relation as a function of applied field cannot rule out a field dependence of $\partial \rho / \partial E$ that has the same angular dependence as shown by the PNE and PHE themselves. Thus existing work cannot rule out an angular dependence of this derivative, leaving open the question of anisotropy of the scattering of electrons with applied field relative to the direction of applied gradients.

In this paper we present results from an optimized thermal isolation platform designed to more comprehensively probe thermal effects in thin film permalloy excited by well-controlled and quantified planar thermal gradients. These platforms employ wider samples than used in our earlier studies with both platinum strips and point contacts (as described in more detail below), in order to clarify the source of transverse voltages. We also produced platforms with no additional transverse electrical conduction path to examine closely any reduction in signal that these paths could produce. These platforms also allow voltage measurements on the same sample when either thermally biased or biased with an applied electrical current. This allows measurement of the planar Hall effect and the planar Nernst effect on exactly the same sample, and a close examination of the expected link between these various manifestations of spin-orbit scattering in metallic FM films. Since this comparison suggested disagreement when we used the simplest estimation of thermal gradient in the thermal isolation platform, we also performed 2d finite element modeling.
thermal analysis to calculate expected thermal gradients in the suspended structure as a function of position. Using the resulting thermal gradients gives excellent agreement between expected PNE and PHE signals and the corresponding Seebeck and anisotropic magnetoresistance values, comprehensively ruling out any signal corresponding to spin current generation in this nm-length scale experiment. Finally, we consider the form of a Mott relation between the planar Nernst coefficient and the planar Hall resistivity and compare this expectation to the Mott relation between longitudinal thermopower and electrical resistivity. The results show the same apparent field-independence of the scattering that was previously reported, though we add consideration of the field-dependence of the estimated absolute Seebeck effect that suggests a possibly anisotropy in the scattering with field direction.

6.2 Experimental Details

We originally measured $\alpha(H)$, AMR and PNE in previous thermal isolation platforms of sizes much smaller than current platforms. Here we designed new platforms (Fig. 6.1) to further probe the potential long-range tSSE along with the PNE. We fabricated these using 500-nm-thick low-stress Si-N, with each platform micromachined from the same 100-mm Si wafer. We patterned 40-nm-thick Pt leads with a 10-nm-thick Cr adhesion layer via photolithography to serve as thermometers, heaters and voltage leads. This extremely low thermal mass membrane yields effectively 2D heat flow and a unidirectional thermal gradient across the majority of
bridge. We are able to measure four-wire electrical resistivity, longitudinal thermopower, and transverse voltages at three locations along Ni-Fe films. The sample studied here was grown on the Si-N structures before release from the Si substrate via e-beam evaporation from alloy source material under high vacuum \((7 \times 10^{-7} \text{ Torr})\) at 20 nm/s. The Si wafer was rotated during film deposition. The Si-N structures were subsequently released via deep-trench Si etching from the backside of the wafer.

As shown in Fig. 6.1, the updated thermal platforms consist of two 800 x 800 \(\mu\text{m}\) Si-N islands each connected to a supporting Si frame by 4 Si-N legs. The islands are connected with a bridge of length 2050-\(\mu\text{m}\) and width of 380-\(\mu\text{m}\) all suspended over a cavity. A 75-nm-thick NiFe film with width 353 \(\mu\text{m}\) was patterned on the bridge, which makes electrical contact with large Pt triangular leads for longitudinal thermopower and Pt voltage contacts for transverse voltage measurements. The platforms allow for “zero substrate” heating of our Ni-Fe thin films, which eliminates unintended thermal gradients and pushes our system to the 2D limit. Two varieties of voltage contacts are used for making transverse voltage measurements: strips (Fig. 6.1c and d)) and point contacts (Fig. 6.1b and e)) . These contacts are placed at either end of the film as well as the center. We also tested platforms with either point voltage contacts (Fig. 6.1b) or Pt strips (Fig. 6.1c) and no other metallic connections to the film, produced by removing the large triangular longitudinal thermopower measurement pads. These “no shorts” lead patterns eliminate any current shunting effects when measuring a transverse voltage on an island.

All measurements are taken using a cryostat under vacuum of \(10^{-6}\) Torr or better to prevent convective heating. We mount the platforms to a radiation-shielded gold-
plated copper mount to prevent radiative heating. Wire bonds are used to make connection with room-temperature electronics. Base temperatures of 276 K are used for all thermal measurements, so that raising the temperature of the platform island to 50 K above this base brings the average temperature of the sample itself near to room temperature, and 300 K for all electrical measurements. A small 20 µA current is used for resistance measurements to prevent film heating. The small mass of the membranes allows the islands to come to thermal equilibrium extremely rapidly (≪ 1.5 s). We measure longitudinal thermopower by applying a series of heating powers to one island’s heater. We then measure not only the voltage generated at either longitudinal or transverse contacts, but also the temperature of each island’s separate sample thermometer. We also monitor a similar micromachined resistive Pt thermometer on the supporting Si frame to ensure thermal stability during thermal measurements. Further details of thermopower measurements with the thermal isolation platforms are published elsewhere [152, 153, 146]. Transverse thermopower measurements are made by cycling a constant heating power on and off to remove any possible contribution from thermoelectric effects in cryostat wiring. The platform allows easy reversal of the direction of thermal gradient by heating either island, but also allows measurements with near zero thermal gradient by heating both islands simultaneously to the same temperature. Further details on this quasi-ac technique for transverse voltage measurements were published previously [89]. Heat flow modeling was performed on the updated Si-N membranes using the Partial Differential Equation (PDE) toolbox from MATLAB[102]. The geometry of the membrane is imported directly from the photolithography layout files to
define our model. The PDE toolbox uses a finite element analysis to define a 2D mesh geometry and formulate boundary conditions. Here we use a $k_{2D}$ calculated by multiplying measured $k$ values by thickness, $t$. Each layer has an additive contribution to total $k_{2D}$. Power was numerically applied uniformly to the geometry of the island heater, and chosen to achieve average island temperatures needed to model the experimental situations as required.

6.3 Results

Fig. 6.2 displays longitudinal thermopower, $\alpha_{rel}(H)$, and four-wire $R(H)$ at 300 K in perpendicular and parallel field orientations. The $R(H)$ measurements are typical of AMR in permalloy and show a film coercivity of $\sim 3$ Oe in perpendicular and parallel configurations, about the same as seen on the narrower and thinner films previously measured [146, 89]. As expected, at zero field and $\vec{M}$, both parallel
and perpendicular orientations have similar values (the small difference in $\alpha_\parallel$ and $\alpha_\perp$ at $H = 0$ is most likely due to a small misalignment in the field angle for this measurement). $R(H)$ and $\alpha(H)$ exhibit similar, even field dependent patterns, indicating both are a result of spin-dependent scattering. $\Delta R/R$ for the various devices measured (not all are shown here) are in the 0.8 – 1% range. We will use the quantities $\alpha_\parallel$, $\alpha_\perp$, $\rho_\parallel$, and $\rho_\perp$ in Eqs. 6.1.2 and 6.1.4 to determine the expected PNE and PHE coefficients.

Fig. 6.3 details transverse thermopower and PHE measurements made on Ni-Fe using a thermal isolation platform with point contacts (as shown in Fig. 6.1a) and e)). Panel a) shows transverse voltage $V_T$ at the center of the platform bridge as a function of field for four different orientations of $H$ with respect to $\nabla T$. For example in the $\theta = 0^\circ$ orientation, $\nabla T$ is parallel to applied field. Also shown in each sub-panel are measurements for $\nabla T = 0$ (magenta symbols), $\nabla T = 14.9$ K/mm (black symbols) and $\nabla T = -14.9$ K/mm (blue symbols). As discussed in detail below, these values of thermal gradient are the result of 2d heat flow simulations and are significantly lower than the simple expectation based on the measured temperature difference between the islands. Note that heating both islands such that $\nabla T = 0$ gives a totally field-independent background voltage. Panel b) shows transverse voltage at the same center point contacts in response to applied charge current for $I = \pm 30$ $\mu$A and 15 $\mu$A. These show qualitatively similar patterns, though no background voltage appears in the PHE case since no significant temperature differences arise in the platform for these measurements.
Figure 6.3: Magnetic field dependence of $V_T$ using point contacts at the center of the platform bridge as a function of angle for both applied $T$ and $I$. a) $V_T$ at center in response to applied thermal gradient, with four different field orientations. b) $V_T$ at center in response to applied current, with four different field orientations. c) Saturated $V_T$ from a) vs. field orientation angle. Green line indicates predicted $V_T$ calculated from Eq. 6.1.3. d) Saturated $V_T$ from b) vs. field orientation angle. Green line indicates predicted $V_T$ calculated from Eq. 6.1.5.
Figs. 6.3c) and d) summarize the results of these experiments by plotting the saturated values of $V_T$ as a function of the angle $\theta$. In the case of thermal measurement, the $\nabla T = 0$ voltage was first subtracted though of course this does not alter the field dependence of the signal in any way. Here the $\sin 2\theta$ dependence of the PNE is clear, and the maximum value of these signals indicates a PNE component with voltage near 150 nV. As expected, the purely electric measurement also shows the $\sin 2\theta$ dependence resulting from the PHE with $V_T = 780$ nV. In each plot the green line shows $V_T$ predicted by Eq. 6.1.3 or 6.1.5, with the appropriate coefficient determined from the data in Fig. 6.2 using Eq. 6.1.2 or 6.1.4. The values of $\nabla T$ used here will be discussed in detail below.

Fig. 6.4 depicts the same series of experiments on the same Ni-Fe film, but with $V_T$ measured at point contacts on the right end of the film near the triangular lead visible in Fig. 6.1e). Here for PNE experiments, the $\nabla T = 0$ background must only be removed from the orientation of $\nabla T$ that results in heating of the right island, since there is little temperature rise compared to the base temperature on the non-heated island. The proximity of the measurement location to the triangle lead and to the island itself has a large effect on the signals measured, leading to an apparent field-dependence of the $\nabla T = 0$ background and adding large offset voltages to the PHE data. As shown in Figs. 6.4c) and d), the overall size of the PNE and PHE signals also drops, by a factor of three in the case of PNE and more than a factor of 2 for PHE, though the field dependence remains entirely $\sin 2\theta$.

As shown in Fig. 6.5, comparison of these results with platforms using Pt strips in both the standard and “no shorts” configurations clarifies the physics and puts
Figure 6.4: Magnetic field dependence of $V_T$ using point contacts on right side of the platform bridge as a function of angle for both applied $T$ and $I$. a) $V_T$ at right in response to applied thermal gradients, with four different field orientations. b) $V_T$ at right in response to applied current, with four different field orientations. c) Saturated $V_T$ from a) vs. field orientation angle. d) Saturated $V_T$ from b) vs. field orientation angle.
Figure 6.5: $V_T$ vs. angle comparing center and right side strip contacts as well as "no shorts" contacts. a) $V_T$ vs. angle for center strip contacts. b) $V_T$ vs. angle for right side strip contacts. c) $V_T$ vs. angle for center strip contacts with "no shorts". d) $V_T$ vs. angle for right strip contacts with "no shorts".
an additional limit on any spin current generation in these mm-scale thermal experiments. Figs. 6.5a) and b) show saturated $V_T$ as a function of $\theta$ measured on Pt strips at the center of the bridge and right end, respectively. Both show exclusively $\sin 2\theta$ field dependence, with no sign of the $\cos \theta$ symmetry that would indicate presence of the transverse spin Seebeck effect (tSSE)[154]. Comparison of Fig. 6.5a) to Fig. 6.3c), where the same experiment was performed with point contacts, shows that the overwhelming effect of the Pt strip is to partially shunt the transverse voltage, such that the maximum measured PNE signal component is reduced from 150 nV to 135 nV. These shunting effects become greater near the triangular leads, though a significant reduction in the signal size to $\sim 70$ nV occurs even with no transverse shorts. This indicates reduced and non-uniform thermal gradients, as the thermal simulations below bear out. Fig. 6.5c) shows that the center strip location is not meaningfully affected by removal of the triangular leads at the ends, as expected. The removal of the triangular leads also causes field-independent background voltages of opposite sign for the opposite orientations of $\nabla T$ as shown in Fig. 6.5d), a phenomenon also seen in the original thermal isolation platform experiments we used to demonstrate the PNE and search for the tSSE in metallic ferromagnets[89]. These results clearly show that such sign changes can easily be generated solely from non-uniform thermal gradients. None of the saturated $V_T$ measurements for Pt strips have ever shown larger voltages than the corresponding measurement with point contacts, which puts a firm limit on the presence of thermal spin current effects when truly in-plane thermal gradients are applied on mm-length scales in metallic FMs.
Figure 6.6: 2D finite element analysis modeling of PNE structures using PDE toolbox from MATLAB. Red line indicates left side heating with $\Delta T = 50$ K between left island and frame. Blue line indicates right side heating with $\Delta T = 50$ K between right island and frame. Black line indicates both left and right island heating with $\Delta T = 0$ K between islands and 50 K between islands and frame.

### 6.4 Discussion

According to Eqs. 6.1.2 and 6.1.4 and using the data from Fig. 6.2, we calculate the maximum value of the PNE coefficient that occurs when $\sin 2\theta = 1$, $\alpha_{PNE,max} = 30$ nV/K, and the maximum value of the PHE coefficient, $\rho_{PHE,max} = 2$ nΩ m.

Using the geometry of the Ni-Fe film, the expected PHE transverse voltage signal from Eq. 6.1.5 is shown in Fig. 6.3d) as the green solid curve and has a maximum value of 800 nV. This is in excellent agreement with $V_{T,max} = 780$ nV measured for the PHE on the center point contacts, as is obvious in Fig. 6.3d). However, if we use
the simple assumptions regarding thermal gradients that result in the expression in Eq. 6.1.3, we expect a thermal gradient near 23 K/mm and \( V_{T,\text{max}} \sim 240 \text{ nV} \). The actual measured values of this PNE voltage contribution even for the ideal case of the center point contacts are far lower (150 nV), indicating that the real thermal gradient at the center of the bridge of the thermal isolation platform is lower.

To explore this possibility in greater detail we performed 2D finite element analysis heat flow simulations in steady state using the actual geometry of our platforms imported directly from lithography layout files. As a first approximation, we use temperature independent thermal conductivities but take these values from our extensive experience using similar platforms to measure thermal conductivity of metallic thin films and the Si-N supporting structure[87, 88]. Figure 6.6 reports results of these simulations, with panel a) showing the color-mapped solution of the thermal Laplace equation overlaid on the representation of the FEM mesh used in the calculation, and panel b) showing the resulting thermal gradient along the center of the sample bridge as a function of position \( x \), for the three different heating conditions used in the PNE measurements. These simulations clearly show that when the desired 50 K temperature difference between heated island and frame is achieved, the thermal gradient at the center of the bridge is very uniform for a large range of the structure, but indeed is much lower than the simple estimation. Using the simulated values of \( \nabla T = \pm 14.9 \text{ K/mm} \) to predict the PNE \( V_T \) gives the green curve in Fig. 6.3c, which nearly exactly matches the measured \( V_T \).

With this understanding of the thermal gradient and transverse shorting issues, we can also examine limits on the existence of the long-length scale thermal spin
Table 6.1: Slope $M_{\text{Mott}}$ and derivatives $\frac{\partial \rho}{\partial E}$ comparing three different membrane-supported FM metal films.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$M_{\text{Mott}} \left( \frac{\Omega \cdot \text{m}}{K} \right)$</th>
<th>$\frac{\partial \rho}{\partial E} \left( \frac{\text{V} \cdot \text{m}}{\text{eV}} \right)$</th>
<th>$\frac{\partial \rho}{\partial E_{\text{planar}}} \left( \frac{\text{V} \cdot \text{m}}{\text{eV}} \right)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni-Fe (75 nm)</td>
<td>$-3.42 \times 10^{-12}$</td>
<td>$4.7 \times 10^{-7}$</td>
<td>$4.7 \times 10^{-7}$</td>
</tr>
<tr>
<td>Ni-Fe (20 nm)[146]</td>
<td>$-2.6 \times 10^{-12}$</td>
<td>$3.5 \times 10^{-7}$</td>
<td>-</td>
</tr>
<tr>
<td>Ni (20 nm)[146]</td>
<td>$-2.8 \times 10^{-12}$</td>
<td>$3.8 \times 10^{-7}$</td>
<td>-</td>
</tr>
</tbody>
</table>

current generation, or tSSE, in metallic ferromagnets. The original reports of the tSSE suggested spin Seebeck coefficients near $6 \times 10^{-11}$ V/K for Ni-Fe. Using the geometry of the thermal isolation platforms discussed here, the resulting signal would be a $\cos \theta$ contribution with amplitude near 300 nV. Here we have conclusively shown no $\cos \theta$ signal within the $\sim 10$ nV error bar of our transverse voltage measurements. This puts a stringent limit on the existence of the tSSE, which must be at least $30 \times$ lower than original reports.[155]

Finally, we can examine the question of a Mott-like relation between the PNE and PHE coefficients. Eq. 6.1.1 shows that at a fixed temperature, and if the energy derivative of $\rho$ is independent of magnetic field, a plot of $\alpha(H)$ vs. $1/\rho(H)$ will be linear with a slope given by

$$M_{\text{Mott}} = -\frac{\pi^2 k_B^2 T}{3e} \left[ \frac{\partial \rho}{\partial E} \right]_{E=E_F}.$$  

When examining the Mott relation involving the longitudinal Seebeck coefficient, one must measure multiple $\alpha$ and $1/\rho$ and determine this slope, since any measurement of longitudinal thermopower includes the contribution from the voltage lead itself. In other words, all measured longitudinal thermopower values are relative rather than
absolute, such that $\alpha_{\text{rel}} = \alpha_{\text{film}} - \alpha_{\text{lead}}$. Examining only the field dependence is one way to correct for this lead contribution, though this technique also throws away any portion of the sample thermopower that is field-independent. Since at fixed $T$ the first fraction in Eq. 6.4.1 is entirely constant, determining this slope from the saturated values of $\alpha$ and $\rho$ as shown in Fig. 6.2, allows calculation of the energy derivative of the electrical resistivity with respect to energy. This is a quantity that is difficult to measure directly, so these measurements have fundamental value for exploring the electron-energy dependence of the scattering events that contribute to $\rho$. The first two columns of Table 6.1 compare $M_{\text{Mott}}$ and $\partial \rho / \partial E$ for the Ni-Fe thin film measured here, as well as Ni-Fe and Ni films previously measured by our group[146]. Despite differences in thickness and growth technique (75 nm films were e-beam evaporated and 20 nm films were sputtered), the values for $\partial \rho / \partial E$ at $E = E_F$ are remarkably similar, varying by less than 25%.

As a first attempt at writing an expression that relates $\alpha_{\text{PNE}}$ and $\rho_{\text{PHE}}$, one might simply replace the corresponding longitudinal coefficients in Eq. 6.1.1, as is effectively the case for the ANE and PHE[148, 149]. However, this results in the unphysical situation where smaller PHE causes larger PNE. Instead we first assume the Mott relation holds separately for $\alpha_\perp$ and $\alpha_\parallel$, and use the definition of the PNE coefficient to determine the relationship:

$$
\alpha_{\text{PNE}} = -\frac{1}{2} \left( \frac{\pi^2 k_{\text{B}} T}{3e} \right) \left( \frac{1}{\rho_\parallel} \left[ \frac{\partial \rho_\parallel}{\partial E} \right]_{E=E_F} - \frac{1}{\rho_\perp} \left[ \frac{\partial \rho_\perp}{\partial E} \right]_{E=E_F} \right) \sin 2\theta.
$$

(6.4.2)
With the further assumption that
\[
\begin{bmatrix}
\frac{\partial \rho}{\partial E} \\
\end{bmatrix}
\bigg|_{E=E_F} = \begin{bmatrix}
\frac{\partial \rho}{\partial E} \\
\end{bmatrix}
\bigg|_{E=E_F} \equiv \begin{bmatrix}
\frac{\partial \rho}{\partial E} \\
\end{bmatrix}
\bigg|_{E=E_F},
\tag{6.4.3}
\]
and using Eq. 6.1.4, then
\[
\alpha_{\text{PNE}} = \frac{\pi^2 k_B T \rho_{\text{PHE}}}{3e} \frac{\partial \rho}{\partial E} \bigg|_{E=E_F}. \tag{6.4.4}
\]

As we have experimentally determined all coefficients in this equation apart from the energy derivative of \( \rho \), we can determine \( (\partial \rho/\partial E)_{\text{planar}} \) directly as shown in Table 6.1. The result exactly matches the quantity determined from the standard Mott relation, as expected based on the assumptions made in this calculation.

However, we note that any angular dependence of this derivative is likely to have the same functional dependence of the AMR and magnetothermopower, and the apparent agreement of the various values in Table 6.1 cannot reveal an anisotropy in the scattering because of the underlying assumption that the Mott relation holds for the separate field directions. However, we can examine this assumption more closely by comparing the AMR ratio and its thermal analog. By the traditional definition, the AMR ratio for our Ni-Fe sample is
\[
\frac{\Delta \rho}{\rho_o} = \frac{\rho_{\|} - \rho_{\perp}}{\frac{1}{3} \rho_{\|} + \frac{2}{3} \rho_{\perp}} = 8.40 \times 10^{-3}. \tag{6.4.5}
\]

The thermal analog is simple to write, but we clarify that this requires determination of the \textit{absolute} Seebeck coefficient, which is challenging for thin film structures since
thin films even of nominally pure materials cannot be expected to have bulk values of the Seebeck coefficient. We have explored a number of techniques to approximate the lead contribution to longitudinal thermopower [95, 156] and estimate a room-temperature contribution from the Pt leads used in these thermal isolation platforms to be \( \alpha_{\text{lead}} = -5 \, \mu V/K \). If we keep the definition of \( \alpha_{\parallel} \) and \( \alpha_{\perp} \) as relative thermopower used above, then the corrected magnetothermopower ratio is

\[
\frac{\Delta \alpha}{\alpha_o} = \frac{\alpha_{\parallel} - \alpha_{\perp}}{\frac{1}{3}\alpha_{\parallel} + \frac{2}{3}\alpha_{\perp} + \alpha_{\text{lead}}} = 4.20 \times 10^{-3}, \tag{6.4.6}
\]

which is a factor of two lower than for the AMR. This is the first indication of a break with the strict relationship between \( \alpha \) and \( \rho \) prescribed by the Mott relation and may be the first evidence of a field-induced anisotropy between the thermally-driven and electric-field driven scattering of electrons in ferromagnetic metals.

### 6.5 Conclusion

In summary, we have used unique thermal isolation platforms to explore the relation between the planar Nernst effect and planar Hall effect in thin films of ferromagnetic metallic nickel-iron alloys. To confirm the uniformity of thermal and current gradients, we measured transverse voltages at various locations on the film, and explicitly tested the effect of transverse current shorting paths. The comparison between the current-driven planar Hall effect and the thermally-driven planar Nernst effect is extremely tight after the correct value of thermal gradient was determined for this structure using 2D finite element analysis. As all measured signals show
field dependence of $\sin 2\theta$, these results put a stringent limit on the long-distance transverse spin Seebeck effect in ferromagnetic metals. Comparison of the AMR and magnetothermopower ratios, after estimation of the absolute Seebeck coefficients, is the first evidence of a possible deviation in field dependence of the thermal and current-based effects.
Chapter 7

Conclusion

In this work we have utilized a-Si-N thermal isolation platforms to measure transport properties across a variety of thin film nanostructures. The Si-N platforms allow for a 2D system that allows us to achieve precise thermal gradients in thin films. We used these thermal gradients to measure thermal conductivity, thermopower and spin transport through thin films. The platforms also allow us to make electrical measurements of thin films with and without the thermal gradients.

I have presented data on semiconducting single wall carbon nanotube thin films as the electronic doping level are manipulated. By observing the change in electrical and thermal conductivity, we were able to utilize the Wiedemann-Franz law to determine the electron and phonon thermal conductivity contributions. We found an interesting pattern that was in part dependent on the nanotube diameter and bundle size, with smaller size leading to a greater change in phonon thermal conductivity as the electronic doping changed.
I also presented on ultra-low magnetic damping alloys. We compared ultra-low damping Co$_{25}$Fe$_{75}$ films to Co$_{50}$Fe$_{50}$ and elemental films and found both enhanced thermal conductivities and thermopower. The increase in thermal conductivity was most likely due to a magnon contribution to the total thermal conductivity, although phonons may have also played a role. Co$_{25}$Fe$_{75}$ also showed a greatly increased thermopower, most likely due to magnon-drag.

The most interesting results of this thesis include spin transport through $a$-YIG and $a$-Cr$_2$O$_3$, which are both disordered magnetic insulators. By using the spin Hall effect in Pt leads, we detected a spin signal via the inverse spin Hall effect at a secondary lead some distance away. We found that by manipulating the thermal gradient between injection and detection lead we were able to achieve large signal sizes orders of magnitude larger than previous studies on crystalline YIG. This is quite a surprising result and may help pave future endeavors in spintronics.

Finally, results of planar Nernst effect and planar Hall effect on a permalloy thin film were discussed. The thermal isolation platforms allows for direct control of in-plane thermal and electrical gradients. Two transverse effects (PNE and PHE) were measured in response to these gradient.
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preparation).
Appendix A

Error Analysis

A.1 Thermal Conductance Error

Much of the error in our thermal conductance measurement comes from the linear fit of the $T$ vs. $P$ plots as well as the conversion from $K$ to $k$ due to uncertainty in thickness of the film.

To account for the error in our thermal conductance measurement we must first look how we perform our measurement. We look at the increase in $T$ with increasing applied $P$, which can be seen in Fig. A.1. Theses equations can be written as

\begin{align}
T_h &= m_{Th} P + T_0 \quad \text{(A.1.1)} \\
T_s &= m_{Ts} P + T_0 \quad \text{(A.1.2)}
\end{align}
Figure A.1: Temperature vs. heating power for the hot and cold island of a 500 nm thick $\alpha$-Si-N membrane structure. The linear fit of the slope is used in calculating $K_B$ and $K_L$. 

$T_H = (1,945,670)P + 78$

$T_S = (230,970)P + 78$
The steady state heat flow equations for our model can then be solved for $K_B$ and $K_L$ as

$$K_L = \frac{P}{(T_h - T_0) + (T_S - T_0)},$$  \hspace{1cm} (A.1.3)

$$K_B = \frac{1}{2} \left[ \frac{P}{T_h - T_s} - K_L \right].$$  \hspace{1cm} (A.1.4)

We can then substitute $(T_h - T_0) = mT_h P$, $(T_S - T_0) = mT_s P$, and $(T_h - T_s) = mT_h P - mT_s P$ into Eqns. A.1.3 and A.1.4. This simplifies into $K_B$ and $K_L$ as a function of slopes, our directly measured values

$$K_L = \frac{1}{mT_h + mT_s},$$  \hspace{1cm} (A.1.5)

$$K_B = \frac{mT_s}{(mT_h + mT_s)(mT_h - mT_s)}.$$  \hspace{1cm} (A.1.6)

The uncertainty of the slope of a line takes the form of a least square fit

$$\delta m = \delta T \sqrt{\frac{N}{N \sum P^2 - (\sum P)^2}},$$  \hspace{1cm} (A.1.7)

where $N$ is the number of data points.

Next is the uncertainty in temperature. To determine $T$, a 4th or 5th order polynomial is fit to the $T(R)$ data from each thermometer. We can multiply the derivative of this function for each thermometer by the SRS AC resistance bridge resolution
\((\delta R)\) which gives the error in \(T\) for each thermometer \(\delta T = \delta R \frac{dT}{dR}\). The slope uncertainty is calculated using error propagation \(\delta(m_{Th} \pm m_{Ts}) = \sqrt{(\delta m_{Th})^2 + (\delta m_{Ts})^2}\).

Once \(\delta T, \delta m_{Th}, \delta m_{Ts}\), and \(\delta(m_{Th} \pm m_{Ts})\) we can propagate the error out for the fractional uncertainty of \(K_B\) and \(K_L\),

\[
\frac{\delta K_L}{K_L} = \sqrt{\left(\frac{\delta(m_{Th} \pm m_{Ts})}{m_{Th} + m_{Ts}}\right)^2}, \tag{A.1.8}
\]

\[
\frac{\delta K_B}{K_B} = \sqrt{\left(\frac{\delta m_{Ts}}{m_{Ts}}\right)^2 + \left(\frac{\delta(m_{Th} \pm m_{Ts})}{m_{Th} + m_{Ts}}\right)^2 + \left(\frac{\delta(m_{Th} \pm m_{Ts})}{m_{Th} - m_{Ts}}\right)^2}. \tag{A.1.9}
\]

### A.2 Thermopower Error

To calculate the thermopower, or Seebeck coefficient, we again take the slope but this time of the longitudinal \(\Delta V\) as a function of \(\Delta T\). In this case the uncertainty of the slope becomes

\[
\delta\alpha = \delta V \sqrt{\frac{N}{N\Sigma \Delta T^2 - (\Sigma \Delta T)^2}} \tag{A.2.1}
\]

where \(V\) is the voltage resolution from a Keithley 2400 Sourcemeter, a Keithley 2182A Nanovoltmeter, or a Keithley 2000 Multimeter.

### A.3 SWCNT Error

We found that the thermal conductance, \(K\), of the SWCNT films is quite small compared to the background \(K\) of the Si-N bridge. This was unfamiliar territory where normally the insulators or metals we measure are on order the same size \(K\) as
the background, leading to a uncertainty in the 2-5 % range. Because of the small measured $K$, when we went to subtract the background Si-N signal we were getting upwards of 20% error in our calculations. To remedy this we considered our data further and noticed that across the typical temperature range we measured (280 - 320 K) the $K$ was constant. This allowed us to treat each data point as a separate sample. We calculated the mean of our set of data and used the standard error of the mean as our error,

$$\sigma_m = \frac{\sigma}{\sqrt{N}}$$  \hspace{1cm} (A.3.1)

where $\sigma_m$ is the standard error, $\sigma$ is the standard deviation, and $N$ is the number of samples.
Appendix B

Spin Transport Measurement

Challenges

Several challenges were realized during the measurements on spin transport on the substrate. Since the \( a \)-YIG, \( \text{Cr}_2\text{O}_3 \) and \( \text{SiO}_2 \) was grown on top of the leads, a diamond scribe was used to remove the film to expose the Pt leads. Once exposed, wire bonds were used to make electrical connection. This method was tricky and the scribe etching wasn’t always accurate. The scribe could easily cut through both the film, Pt leads, Si-N and into the underlying Si substrate. While it didn’t occur on every device, exposing the Si sometimes had consequences on the observed voltage. Here the working theory is that the Pt lead made some small electrical connecting with the Si, allowing for charge to flow. This type of signal can be seen in Fig. B.1, which we call a disconnected signal.
Figure B.1: Non-local voltage vs. current for disconnected spin transport device on substrate.
The disconnected signal was observed across all types of films measured including on blank devices with no deposited film but with etches marked on the device. Fig. B.2 shows data for three different devices. We see almost no change in data before and after an SiO$_2$ film deposited indicating this effect is not determined by the deposited film. Comparing devices, in each case the signal would ‘turn on’ all at the same 5 mA applied current. The overall signal size also changed dramatically from device to device with no apparent pattern.

Finally, we look at one final device measured on 200 nm of a-YIG in Fig. B.3. Here we see both our typical non-local voltage signal indicating spin transport, as well as our disconnected signal. By observing both our spin signal as well as the disconnected signal, we reason that each of these signals are different effects, with the disconnected signal only appearing under certain circumstances.
Figure B.2: Non-local voltage vs. current for disconnected spin transport device on substrate. Cyan data is a non-local voltage measurement of a blank device. Black data is a background device with SiO$_2$ deposited on top of Pt leads. Gray data is data from a 100 nm Cr$_2$O$_3$ device.
Figure B.3: Non-local voltage vs. current for 200 nm α-YIG device. Graph shows both the disconnected signal and spin transport signal.
Appendix C

LabVIEW Vi’s for Experiment and Analysis

C.1 Thermal conductance and spin transport

7FirstThermalKSRS-DU-TEPandSkip-K2000-V8.vi

Main vi for thermal conductance and thermopower measurements with skip. Will take R calibration at defined delta T and full data point every third temperature step. Takes start, end, temp step size and max current. Typical runs go from 78 to 326 K with delta T of 2 K. Typical max current used ranges from 160 to 250 $\mu$A (can start with 200 $\mu$A). This experiment takes up to three and a half days.

Temp-regulation Slope2DWithRangeScalingv4.vi

Main temperature regulation vi to set and stabilize at a given temperature. Resistance slope of 0.00510 typically used for thermal conductivity measurements, which
gives a nice stable data point (about 30 minutes to come to equilibrium at 30 mK stabilization). Resistance slope of 0.0150 used for resistance measurements, takes about 15 minutes to come to equilibrium (not stable enough for thermal conductance). Input start, end and delta T and max current.

**Temp-regulation Slope2DUWithRangeScalingv3.vi**
Secondary temp regulation vi to set and stabilize at a given temperature.

**Find V-Icurveof Filmv3withSRSTemp.vi**
V-I curve for single temp. Used in spin transport experiments to check for any charge leakage between injector and detector strips. Biases low to high input voltage and measures current and resistor for each step. Input start, max and voltage step size.

**FilmVI4WK2400withTempRegv2.vi**
V-I curve vs. temperature. Uses ‘Find V-Icurveof Filmv3withSRSTemp.vi’ at each temp step. Plots R-film vs. temp. Input start T, max T, delta T, SRS channel, start V, max V, voltage step.

**Find Rof Filmv3withSRSTemp.vi**
I-V curve for single temp. Used for spin transport experiment when you have a resistor hooked up. Sweeps from low to high input current and measures voltage and resistor for each step. Input resistance channel, start, max and current step size.

**SetMeasP-2400-V2.vi**
Sets current for Keithley 2400. Outputs heater power and thermometer resistance.

**SetMeasV-2400-V2.vi**
Sets voltage for Keithley 2400. Outputs heater power and thermometer resistance.

*8FirstThermalKSRS-DU-TEP-K2000-V8.vi*

Main vi for thermal conductance measurements without skip. Does not take intermediate calibration points. Recommended only for quick runs at delta T of at least 6K. Do not use at 2K step size, will take a long time. Input start, end and delta T and max current.

*Find Rof-Filmv2.vi*

I-V curve for single step. Used for spin transport experiment when you don’t have a resistor. Sweeps from low to high input current and measures voltage for each step. Input start, max and current step size. Will not look at temp, only use to quick check.

*Film4WK2400withTempRegv2.vi*

I-V curve vs. temp. Uses ‘Find Rof-Filmv2.vi’ so isn’t able to measure hot resistor.

*Film4WK2400withTempRegandThermov3.vi*

I-V curve vs. temp with resistor check each current. Use this for most comprehensive look. Uses ‘Find Rof Filmv3withSRSTemp.vi’.

*Film4W2400withWaitbetweenTempStepsV3.vi*

I-V curve vs. temp. Use this when you don’t have a resistor for temp regulation. Should wait for given time between each temp step instead of stabilizing off a resistor.

*ThermalPropertiesAnalysisV9.vi*
Use to calculate thermal conductance. Input main file from ‘7FirstThermal
KSRS-DU-TEPandSkip-K2000-V8.vi’ or ‘8FirstThermalKSRS-DU-TEP-K2000-V8.vi.’
Outputs are T calibration files for each thermometer, KallPs2Is file, KbKIAll file,
TEP file, polynomial fits for each thermometer, and thermometer sensitivity files.
Saving output files are optional.

**ScatterCorrectionAnalysis.vi**

Allows correction of single data point from ‘KallPs2Is’ output file. Can remove
low or high current step.

### C.2 PNE and PHE

*Vi’s below need updating to most recent field set, ‘FieldSetBZv3.vi’.*

**FieldSetBZv3.vi**

Most current field set vi. Uses NI box to set Kepco current for magnet.

**MasterSSEViWithRealFieldbzv3-Hysteresis.vi**

Main vi for SSE and PNE measurements. Heats left, right, both islands and
measures voltage. Sweeps from pos to neg field and back.

**IslandHeating-Zero-Averaging.vi**

Cycles heater power: off, on, off while measuring both nanovoltmeters. Can set
delay of voltage measurement after heater power on (useful for larger PNE devices).

**Master-AMR-Film-WithField-DeltaV3b.vi**

Uses delta mode to pole resistance as field is swept.

**FieldSetBZv2.vi**
Sets desired field using 6220.

**Master-AMR-Delta-Hysteresis.vi**

Sweeps AMR vi up and down.
Appendix D

List of devices measured
<table>
<thead>
<tr>
<th>PNE</th>
<th>Name</th>
<th>Measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>CINT 14</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PNE1</td>
<td>K (No film)</td>
<td></td>
</tr>
<tr>
<td>PNE2</td>
<td>K (No film)</td>
<td></td>
</tr>
<tr>
<td>SSE1</td>
<td>AMR</td>
<td></td>
</tr>
<tr>
<td>CINT 15</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PNE1</td>
<td>PNE, AMR, PHE</td>
<td>K (No film)</td>
</tr>
<tr>
<td>PNE1 NoShorts</td>
<td>PNE (100K), AMR</td>
<td>K (No film)</td>
</tr>
<tr>
<td>PNE1 Oxide NoShorts</td>
<td></td>
<td>K (No film)</td>
</tr>
<tr>
<td>PNE1 TwoLead NoShorts</td>
<td></td>
<td>K (No film)</td>
</tr>
<tr>
<td>PNE2</td>
<td>PNE</td>
<td>K (No film)</td>
</tr>
<tr>
<td>SSE1</td>
<td>PNE (100K), AMR, PHE</td>
<td>K (film)</td>
</tr>
<tr>
<td>SSE2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SSE1 Oxide</td>
<td></td>
<td>K (No film)</td>
</tr>
<tr>
<td>SSE2 Oxide</td>
<td></td>
<td>K (No film)</td>
</tr>
<tr>
<td>SSE1 NoShorts</td>
<td></td>
<td>PNE</td>
</tr>
</tbody>
</table>

Figure D.1: List of PNE devices that were measured. K indicates that thermal conductance, thermopower and film resistance (not for all samples) were measured.
<table>
<thead>
<tr>
<th>Spin Transport</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Film on top</strong></td>
</tr>
<tr>
<td>Yig 200nm (06/2016 CSU)</td>
</tr>
<tr>
<td>A3W2</td>
</tr>
<tr>
<td>A5W2</td>
</tr>
<tr>
<td>Yig 200nm (09/2016 CSU)</td>
</tr>
<tr>
<td>A10W2</td>
</tr>
<tr>
<td>A11W2</td>
</tr>
<tr>
<td>Yig 100nm (09/2016 CSU)</td>
</tr>
<tr>
<td>A7W2</td>
</tr>
<tr>
<td>A8W2</td>
</tr>
<tr>
<td>Cr2O3 (03-05/2017 DU)</td>
</tr>
<tr>
<td>A15W2</td>
</tr>
<tr>
<td>A16W2</td>
</tr>
<tr>
<td>Cr2O3 (06/2017 DU)</td>
</tr>
<tr>
<td>A17W16</td>
</tr>
<tr>
<td>SiO2 (08/2016 DU Xin)</td>
</tr>
<tr>
<td>A4W2</td>
</tr>
<tr>
<td>A6W2</td>
</tr>
<tr>
<td><strong>Leads on top</strong></td>
</tr>
<tr>
<td>Cu on A7A8A9 100nm YIG</td>
</tr>
<tr>
<td>Cu on A7A8A9 100nm YIG</td>
</tr>
<tr>
<td>Pt on A10A11 200nm YIG</td>
</tr>
<tr>
<td>Pt on CryYIG</td>
</tr>
<tr>
<td>Niobium</td>
</tr>
<tr>
<td>EBL Blank</td>
</tr>
</tbody>
</table>

Figure D.2: List of spin transport devices measured.
<table>
<thead>
<tr>
<th><strong>Thermal</strong></th>
<th><strong>K</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td>CoFe (11/2016)</td>
<td></td>
</tr>
<tr>
<td>B1W2 (3nm Cu 3nm Ta 75nm Co25Fe75 3nm Ta)</td>
<td>K</td>
</tr>
<tr>
<td>B2W2 (3nm Cu 3nm Ta 75nm Co25Fe75 3nm Ta)</td>
<td>K (Field Dependence)</td>
</tr>
<tr>
<td>CoFe (08/2017)</td>
<td></td>
</tr>
<tr>
<td>C1W16 (3nm Ti 3nm Cu 75nm Co25Fe75 5nm Al)</td>
<td>K</td>
</tr>
<tr>
<td>C2W16 (3nm Ti 3nm Cu 75nm Co25Fe75 5nm Al)</td>
<td>K (Field Dependence)</td>
</tr>
<tr>
<td>C3W16 (3nm Ti 3nm Cu 75nm Co50Fe50 5nm Al)</td>
<td>K</td>
</tr>
<tr>
<td>s-SWCNT</td>
<td></td>
</tr>
<tr>
<td>2015</td>
<td></td>
</tr>
<tr>
<td>AA01</td>
<td>Pre</td>
</tr>
<tr>
<td>AA02</td>
<td>K (Doped)</td>
</tr>
<tr>
<td>AA03</td>
<td>K (Doped multiple, Spin transport)</td>
</tr>
<tr>
<td>AA04</td>
<td>K (Doped multiple)</td>
</tr>
<tr>
<td>2017</td>
<td></td>
</tr>
<tr>
<td>AB01</td>
<td>K (Doped)</td>
</tr>
<tr>
<td>AB03</td>
<td>K (Doped)</td>
</tr>
<tr>
<td>AC01</td>
<td>Pre</td>
</tr>
<tr>
<td>AC02</td>
<td>Pre</td>
</tr>
<tr>
<td>AC03</td>
<td>K (HiPCO SMP OA Doped)</td>
</tr>
<tr>
<td>AC04</td>
<td>K (PT SMP OA Doped)</td>
</tr>
<tr>
<td>AC05</td>
<td>K (CE Doped)</td>
</tr>
<tr>
<td>AC06</td>
<td>K (Blank with ALD Alumina)</td>
</tr>
<tr>
<td><strong>Bulk 2015</strong></td>
<td></td>
</tr>
<tr>
<td>Si chunk</td>
<td>K</td>
</tr>
<tr>
<td>100 Ohm Resistor</td>
<td>K</td>
</tr>
<tr>
<td>AlSi Bond Wire</td>
<td>K</td>
</tr>
<tr>
<td>Apiezon N Grease</td>
<td>C</td>
</tr>
<tr>
<td>CoS2</td>
<td>K and C</td>
</tr>
<tr>
<td>Co1-.05Fe.05S2</td>
<td>K and C</td>
</tr>
<tr>
<td>Co1-.1Fe.1S2</td>
<td>C</td>
</tr>
<tr>
<td><strong>Miscellaneous</strong></td>
<td></td>
</tr>
<tr>
<td>E1W16 (50nm Fe - Bad)</td>
<td>K</td>
</tr>
<tr>
<td>D2W3 (10nm Cr 300nm Au)</td>
<td>K</td>
</tr>
<tr>
<td>P1W3 (50nm AuPd)</td>
<td>K</td>
</tr>
<tr>
<td>P2W3 (AuPd multiple deps)</td>
<td>K</td>
</tr>
<tr>
<td>P3W3 (AuPd multiple deps)</td>
<td>K</td>
</tr>
<tr>
<td>A1Wh1 (Half-size)</td>
<td>K</td>
</tr>
</tbody>
</table>

Figure D.3: List of devices used for thermal conductance measurements. K indicates that the thermal conductance, thermopower and film resistance were measured.