Understanding of Pure Spin Transport in a Broad Range of Y$_3$Fe$_5$O$_{12}$-based Heterostructures

Dissertation

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Hailong Wang, M.S.

Graduate Program in Physics

The Ohio State University

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Dissertation Committee:

Professor Fengyuan Yang, Advisor
Professor P. Chris Hammel
Professor Ralf Bundschuh
Professor Andrew Heckler
Abstract

Spin currents carried by mobile charges in metallic and semiconducting ferromagnetic (FM) and nonmagnetic (NM) materials have been the central focus of spintronics for the past two decades, while spin transport in insulators is largely unexplored. Ferromagnetic resonance (FMR) driven spin pumping has awakened intense interest in magnon-mediated spin currents which can propagate in both conducting and insulating FMs and in antiferromagnets (AF). High quality $Y_3Fe_5O_{12}$ (YIG) epitaxial thin films have been grown by our off-axis sputtering technique and characterized by x-ray diffraction (XRD) atomic force microscopy (AFM), vibrating sample magnetometer (VSM) et al. Due to the exceptional low damping constants and insulating nature, YIG thin films have been regarded as one of the most promising candidates for microwave application and dynamic spin transport study, particularly for the detection of pure spin currents in YIG/NM bilayer structures.

FMR spin pumping is an emerging technique for dynamic injection of a pure spin current from a FM into a NM without an accompanying charge current, which offers the potential to enable low energy cost, high efficiency spintronics. The performance of these future spin-based applications relies on the efficiency of spin transfer across the FM/NM interfaces. In this thesis, I present our recent results on FMR spin pumping in YIG-based heterostructures, the large mV-level inverse spin Hall signal observed in our YIG/Pt bilayers provides a good platform to probe the underlying
spin transport and coupling mechanism across different materials, including metallic Cu layer and some insulators with different magnetic correlation strengths. We systematically study the spin transport in a series of six Pt/insulator/YIG trilayers where the insulators are diamagnetic (one), paramagnetic (one) and AF (four, having a wide range of ordering temperatures). We observe remarkably robust spin transport in the AF insulators and a distinct linear relationship between the spin decay length in the insulator and the damping enhancement in the YIG, suggesting the critical role of magnetic correlations in magnetic insulators for spin transport. In particular, we have observed highly efficient dynamic spin injection from YIG into NiO, and robust spin propagation in NiO as thick as 100-nm mediated by its AF spin correlations. Strikingly, the insertion of a thin NiO layer between YIG and Pt significantly enhances the spin currents injected into Pt, suggesting exceptionally high spin transfer efficiency at both YIG/NiO and NiO/Pt interfaces. This result opens a new venue for exploration of spin manipulation in tailored structures composed of metallic and insulating FM, AF and nonmagnetic materials and demonstrates that spin pumping in YIG-based heterostructure is a powerful and versatile tool for understanding spin Hall physics, spin-orbit coupling (SOC), and magnetization dynamics in a broad range of materials.
To my dear parents and loving wife, Chunhui Du, I love you forever and always.
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Vita

October 18, 1988 .......................... Born - Hefei, Anhui, China

2010 ................................. East China Normal University

2013 ................................. M.S. Physics

2015 ................................. Ph. D. Physics

2011-present ........................ Graduate Research Associate,
                                      The Ohio State University.

Publications

C. H. Du, H. L. Wang, P. C. Hammel, F. Y. Yang, “Y_{3}Fe_{5}O_{12} Spin Pumping for
Quantitative Understanding of Pure Spin Transport and Spin Hall Effect in a Broad

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R. Adur, C. H. Du, S. A. Manuilov, H. L. Wang, F. Y. Yang, D. V. Pelekhov,
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Fields of Study

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Chapter 1: Introduction

Spintronics study the interplay between spin and charge. Over the last two decades, spin currents have been extensively studied to control and manipulate the magnetization dynamics, such as giant magnetoresistance [1], magneto tunneling resistance [2], spin transfer torque induced magnetic resonance and switch[3, 4, 5]. Spin current has also been widely used in the industry, such as data storage, information processing and communications. Generation and manipulation of spin current is of central importance for modern spintronics applications. Typically, spin current can be generated by three different methods as shown in Fig. 1. First, electrical injection is the most widely used way to achieve angular momentum transfer from ferromagnetic materials (FM) to non-magnetic materials (NM). In this case, spin current is carried by mobile electrons. When electrical currents go through from FM to NM, spin currents can go across FM/NM interface and transmit within the spin diffusion length. For the electrical spin transfer, the injection efficiency is typically limited by spin polarization of FM and also the interface spin mismatch[6], thus a insulating magnetic tunneling layers like MgO, or Al$_2$O$_3$ will be inserted between FM and NM to enhance the transfer efficiency[6]. The electrical spin injection method has been extensively used to achieve angular momentum transfer from FM materials to semiconductors, like Si and recently graphene and other 2D materials. The widely
used non-local spin valve structure also bases on this technique. Typically, in order to obtain high spin injection efficiency, the electrical injection devices need to be in \( \mu m \) or even nanometer scale and appropriate interface treatment is important.

Thermal injection shown in Fig.1 (b), usually called spin Seebeck effect[7] is another way to achieve pure spin current transfer. It has been reported in ferromagnetic metal Py[7], ferrimagnetic insulator Yttrium Iron Garnet (YIG)[8], and
So far, the mechanism of spin Seebeck effect is still under debate, people from some groups claim that the thermal spin pumping signal can be contaminated in both longitudinal and transverse geometries[10, 11]. In transverse case, the single crystalline substrate could be treated as a good thermal conductor and the lateral temperature gradient will lead to a vertical temperature different between ferromagnetic conductor and spin detector, thus anomalous Nernst effect is essentially indistinguishable from the observed spin Seebeck effect[10]. On the other hand, in longitudinal geometry, the proposed proximity effect at ferromagnetic insulator/Pt interface could also contribute to the observed thermal spin pumping signal[11]. While the fundamental mechanism is still under debate, spin Seebeck effect has been proposed to be quite promising in energy renewal for future application, the current understanding believes that the thermal spin transfer efficiency in spin Seebeck effect is determined by the temperature difference of the magnon on FM side and the temperature of electrons on NM sides[12].

Recently, microwave-driven ferromagnetic resonance (FMR) spin pumping as shown in Fig.1 (c) has been demonstrated to be another effective way to achieve angular momentum transfer from FM to nearby nonmagnetic materials due to the dynamic exchange coupling between precessing magnetization of FM induced by magnetic resonance and the free electrons in NM at the interface. It injects pure spin current without accompanying charge current since no bias voltage needed. Although FMR spin pumping has been realized in various material systems,[13, 14, 15, 16], the underlying spin transport mechanism, spin transfer efficiency need further investigation. The general understanding believes that the FMR spin injection efficiency heavily depends on the interfaical condition, people from different groups reported
various surface treatment methods which could improve the spin pumping efficiency. Compared with electrical and thermal spin injection methods, the microwave could induce in-phase magnetization precession in which case pure spin current transfer can be achieved. Especially, FMR spin pumping technique provides a unique platform to study the spin transmission and conductance in insulators[17, 18, 19] which cannot be probed by the conventional electrical spin injection method. On the other hand, FMR driven spin pumping could be used to study the spin Hall physics, like the spin orbit coupling strength in NM materials[20, 21, 22, 23].

So far, these activities and research in spin Hall physics are realized only in material systems, often without precise atomic level control. Two dimensional material systems (2D materials), such as graphene, transition-metal dichalcogenides (TMDs), surface state of the topological insulators (TIs)[24] are an open research domain, providing a new platform for control of charge, magnetism, spin transport, and spin dynamic properties. Relatively low critical current density for switching was already reported in FM/topological insulator (TI) bilayers by Dr. Ralph and his collaborators, possibly due to spin-momentum locking at the surface state with large SOC[24]. Minimize the bulk conductivity or combination the TIs with ferrimagnetic insulator such as $Y_3Fe_5O_{12}$ (YIG) could potentially improve the switch efficiency using topological insulators. TMDs exhibit large spin-orbit coupling, combined with the 2D nature, they can also be promising candidates to generate large spin currents with significantly lower electrical current density. So far, the manipulation of FM magnetic dynamics using TMDs has not yet been explored and reported. Studying the spin current generation efficiency in various TMDs will broaden the general understanding in spin Hall physics in this new 2D material system. The theoretically proposed
strong spin orbit coupling in TMDs materials could also possibly lead to the manipulation of magnetization using DC charge current with significantly higher efficiency compared with the FM/NM heterostructures, which will be helpful for data storage and information processing.

Another hot topic in spintronics is optimization of the spin transfer efficiency using single crystalline structure, including the control of interfacial condition at FM/NM interface. People reported that an insulating antiferromagnetic (AF) spacer layer between Pt and FM materials could actually enhance spin transport capability\[19\]. The spin transfer torque efficiency which is actually a reversal process of FMR spin pumping is also reported by T. Moriyama et al. to be higher than the FM/NM bilayer structure when AF layer inserted. These interesting phenomena could potentially pave the way for the broad application of spin Hall physics in the future spintronics devices.

In the following chapters, chapter 2 will focus on the growth, characterization of high quality epitaxial YIG thin films and FMR spin pumping study in YIG/NM bilayer structures, chapter 3 probes the underlying spin pumping coupling mechanism and chapters 4 and 5 study the role played by magnetic insulators and metallic Cu spacer in spin transport and efficiency.
Chapter 2: Growth, characterization, and large spin pumping signal in YIG-based heterostructures

As modern electronics shrink towards the sub-10nm regime, new paradigms of information technology are urgently needed. Both semiconductor industry and the fundamental research community are actively seeking transformative technologies that will meet the demand of the fast growing digital economy. Spintronics is one of the leading candidates for future technologies, with the generation and manipulation of spins being of central importance to spin-based electronics. Emergent spin phenomena in magnetic insulators are promising for energy efficient approaches due to the extremely low magnetic damping in these materials, which also provide new understanding of spin/magnon transport and dynamic properties. Over the last couple of years, a great deal of research attention has been paid to the growth\[25, 26\], characterization, and (inverse) spin Hall effect\[27, 13, 21\]study in ferrimagnetic insulator YIG structures \[28, 29\] with an exceptionally low damping constant\[25, 26\].

2.1 Background

Fabrication of single crystalline YIG films were first reported by liquid phase epitaxial (LPE) method\[30\] with the thickness typically in the $\mu$m scale. With the emerging of spin dynamics study, since 2011, several pioneering groups have reported
the successful growth of nanometer thickness high quality YIG thin films with pulser laser deposition (PLD)[31, 26], laser molecular beam epitaxial (MBE), and also sputtering technique[25]. In 2012, Prof. Mingzhong Wu’s group first reported the deposition of 20nm YIG films with Gilbert damping constant about one order of magnitude smaller[26] of other metallic ferromagnetic materials, such as Py and CoFeB. Due to the insulating nature and low damping constant, YIG thin films have great advantage in minimizing the artifacts due to thermoelectric or magnetoelectric effects, such as anisotropic magnetoresistance (AMR) or anomalous Hall effect (AHE)[32, 33] in dynamic spin transport and spin pumping study.

Figure 2.1: **Schematic illustration of the device in Saitoh’s work[34]** Pt (i) acts the role as a spin injector though spin Hall effect and excites the magnetic precession of underneath YIG film. Pt (o) strip is used to detect the excited spin waves via inverse spin Hall Effect.
In 2010, Saitohs group in Japan reported the first, although kind of controversial result about induced magnetic precession by spin transfer torque at YIG/Pt interface[34]. The excited spin wave propagates in YIG and gets detected in a second Pt strip via inverse spin Hall effect (ISHE). The device structure is shown in Fig. 2.1. The thickness of YIG film is 1.3 μm with the damping constant about 6.7×10^{-5}. The separation between the two Pt strips is 1mm within the spin decay length of YIG which makes the spin wave transmission possible. The surface area of the YIG, Pt (i), and Pt (o) is 35, 27.5, and 0.5mm² respectively. Pure DC current was applied through the Pt (i) layer in y direction as shown in the Figure. The external H field is applied in x-y plane with certain angle θ relative to the current direction. When θ = 0° or 180°, the spin transfer torque is inactive, since the spin transfer torque cannot induce stable magnetization precession around the external field H direction. When θ = 90°, due to the small thermal vibration of the magnetization M at room temperature, M and spin polarization σ is not absolutely parallel with each other which can induce stable YIG magnetization precession around the H direction. In order to overcome the intrinsic damping in YIG layer, the spin transfer torque need to be large enough to achieve a critical point 6.0 × 10^8 A/m² to achieve magnetic precession. Above this value, the local M precession can be excited for the YIG layer underneath the Pt (i) strip. The excited magnetic precession will induce spin wave propagation in YIG due to the exceptional low damping constant with the spin wave decay length typically in several mm level scale. When the propagating spin wave gets to the Pt (o) strip, spin currents will inject into Pt (o) layer and converted to charge currents through the inverse spin Hall effect (ISHE) and electrically detected. The ISHE signal observed when DC current equals 1.0 × 10^9 A/m² is about 1nV. In order to eliminate
the possible contamination from the lateral spin Seebeck effect potentially induced by the Joule heating of the applied currents, authors also switch the external field $H$ direction to $-90^\circ$. The observed ISHE signal is in the opposite sign confirming that the electrical signal indeed depends on the spin polarization $\sigma$ direction. If the observed electrical signal comes from the lateral spin Seebeck effect, the temperature gradient should not depends on the DC current polarity. This paper reported a quite exciting and promising experimental result and pointed a very important direction to achieve spin precession by pure DC electrical current via SHE induced spin transfer torque effect, the spin wave information can also been transmitted via a ferrimagnetic insulator with mm-level spin decay length in macro-scale level. However, the results are not quite convincing and kind of controversial due to the almost negligibly small electrical observed ISHE signal which is down to 1nV. Second, other groups reported some issues when they tried to repeat the same experiment.

![Figure 2.2: Spin wave transmission in YIG-based waveguide reported by Prof. Mingzhong Wu’s group][18] Experiment setup to achieve the control of propagation spin wave in YIG with spin Hall effect induced spin transfer torque. External field $H$ is in $y$ direction and DC current is applied in $x$ direction in Pt layer.
After the work from Saitohs group, in 2012, Prof. Wu group in Colorado State University reported the tuning of spin wave propagation efficiency in YIG-based waveguide through SHE induced spin transfer torque as sample structure shown in Fig. 2.2[18]. Two Au microwave antennas with 50 μm wide and 2.2mm long were deposited on two sides of YIG films separated by 5.5 mm. Pt layer was deposited on top the YIG surface. The thickness of the YIG and Pt layer is 4.6 μm and 20nm respectively. Alternating (AC) currents in the first Au microwave antenna excite the spin wave precession, the propagating spin waves transmit through the YIG and gets to the second Au antenna and picked up as microwave signal. Pure DC current was applied though Pt layer in x direction. The induced spin polarization is in +y or -y direction depending on the DC current polarity. The key message of this experiment is that the SHE induced spin transfer torque at YIG/Pt interface can control the damping of the propagating spin waves by damping like spin transfer torque or anti-damping like torque depending on the relative H field orientation and DC current polarity as explained in details above. Thus, the magnitude of the microwave signal picked up in the second Au transducer can be controlled.

2.2 Fabrication of epitaxial YIG thin films by off-axis sputtering technique

2.2.1 Synthesis and characterization

Compared with LPE, PLD, and Laser MBE, off-axis sputtering technique [25, 35, 36] will provide a more accurate and flexible control of the oxygen flow which is critical for high quality complex oxide growth. Figure 2.3 shows the sputtering system we used for YIG growth. The sample holder is put about 3 inches away and 2 inches down from the sputtering gun, YIG target is 2 inches in diameter and made
from commercial powder. Single-crystalline YIG epitaxial thin films were grown on (111)-oriented Gd$_3$Ga$_5$O$_{12}$ (GGG) substrates in an off-axis ultrahigh vacuum (UHV) sputtering system with a base pressure below $5 \times 10^{-9}$ Torr. Horizontal sputtering sources and 90 off-axis geometry were used for film deposition. The optimal growth conditions include: a total Ar/O$_2$ pressure of 11.5 mTorr with an O$_2$ concentration of 0.15%, a substrate temperature 750°C, and a radio-frequency sputtering power of 50 W. The deposition rate for YIG is 0.33 nm/min and the film thickness ranges from 10 to 200 nm.

![Home-built off-axis sputtering system](image)

Figure 2.3: **Home-built off-axis sputtering system** The substrate holder is typically put 3 inches away and 2 inches down from the sputtering gun, the power applied and growth pressure depends on the specific materials and required stoichiometry.

YIG has a cubic crystal structure with space group *Ia-3d* as shown in Fig. 2.4. The cubic unit cell has a lattice constant of $a = 12.376$ angstrom and contains 8 formula units (f.u.) with 160 atoms, of which only the Fe$^{3+}$ ions carry magnetic
moment (5µB each). Of the 40 Fe$^{3+}$ ions in a unit cell, 16 are on octahedral sites and 24 are on tetrahedral sites. Each octahedral Fe is connected to 6 tetrahedral Fe and each tetrahedral Fe is connected to 4 octahedral Fe through corning sharing an oxygen, resulting in an intertwining octahedron-tetrahedron network. The magnetic moments of all the tetrahedral Fe are aligned anti-parallel to those of the octahedral Fe, resulting in ferrimagnetic order. The intertwining octahedron-tetrahedron Fe network leads to very low magnetic anisotropy as compared to FMs with hexagonal (e.g., Co), tetragonal (e.g., CrO$_2$), and simpler cubic (e.g., Ni) structures, which in
turn leads to exceptionally low damping in YIG. We will discuss later the critical importance of preserving the stoichiometry and ordering of YIG, both in the bulk of the films and at the interfaces, for achieving high-efficiency spin pumping.

2.2.2 X-ray diffraction and ferromagnetic resonance characterization of YIG thin films

The crystalline quality of the YIG films is determined by high-resolution x-ray diffraction (XRD). A representative $\theta$-2$\theta$ scan of shown in Fig. 2.5 indicates a phase-pure epitaxial YIG film. Figure 2.5 shows $\theta$-2$\theta$ scans near the YIG (444) peak for four films with thicknesses, $t = 10, 20, 50$, and 80 nm. Pronounced Laue oscillations are observed in all films, reflecting smooth surfaces, sharp YIG/GGG interfaces and high uniformity throughout the films. The XRD rocking curves exhibit a full width at half maximum (FWHM) of $0.027^\circ$, $0.0092^\circ$, $0.0072^\circ$, and $0.0053^\circ$ for the 10, 20, 50, and 80 nm thick films, respectively, which reach the resolution limit of conventional high-resolution XRD systems. These values of FWHM are among the narrowest of any epitaxial films, demonstrating excellent film crystalline quality.

To further investigate the surface condition of the YIG thin films deposited by off-axis sputtering technique, we conduct x-ray reflectometry (XRR) scan and Atomic force microscopy measurements as shown in Fig. 2.6. It shows an x-ray reflectometry (XRR) scan of a YIG/Pt bilayer with two periods of oscillations, corresponding to the 34-nm YIG and 4.1-nm Pt layers. A fit to the XRR scan gives a YIG/Pt interfacial roughness of 0.22 nm[22], indicating the sharpness of the interface. The smooth surface of our YIG films is confirmed by the AFM image from which we obtain a root-mean-square (rms) roughness of 0.10 nm over an area of 10$\mu$m×10$\mu$m.
Figure 2.5: XRD and rocking curve scan of YIG thin films with different thicknesses. Semilogarithmic $\theta$-2$\theta$ scans of 10-, 20-, 50-, and 80-nm-thick YIG films near the YIG (444) peak, all of which exhibit clear Laue oscillations corresponding to the film thickness[25]. The scans are offset from each other for clarity. Rocking curves of the four YIG films are taken for the first satellite peak to the left of the main peak at the 2$\theta$ angle marked by the up arrows.

Ferromagnetic resonance measurements [37] of the YIG films are carried out under room temperature in a cavity at a microwave frequency $f = 9.65$ GHz and power $P_{rf} = 0.2$ mW. Figure 2.8 shows a typical FMR derivative spectrum of a 20-nm YIG film with an in-plane magnetic field $H$ along the $x$-axis ($\Delta H = 90^\circ$ which gives a peak-to-peak linewidth ($\Delta H$) of 7.4 Oe. Figure 2.7 shows the specific FMR measurement geometry. The measured FMR linewidth contains two components, 1) Gilbert
Figure 2.6: AFM image and XRR scan of YIG/Pt bilayer. AFM image of a YIG film with a roughness of 0.10 nm. X-ray reflectometry scan (red) of a YIG(34 nm)/Pt(4.1 nm) bilayer shows the superposition of oscillations from both the Pt and YIG layers. The fit (blue) to the experimental data gives a YIG/Pt interfacial roughness of 0.22 nm.

damping component which can be obtained from the frequency dependence of the linewidth, another contribution comes from the inhomogeneity part which describes the linewidth under zero frequency. The angular dependence of the resonance field ($H_{res}$) of the YIG film is shown in Fig.2.9, where $H_{res}$ is defined as the field at which the derivative of the FMR absorption crosses zero. We obtain the effective magnetization, saturation magnetization = 1794 Oe, from a fit to $H_{res} (\Delta H)$ employing
Figure 2.7: FMR experimental geometry.

quantitative analysis. Saturation magnetization and $g$ factor of the YIG films were determined from FMR resonance field as a function of $H_{res}$. Resonant condition can be derived by minimizing the total free energy $F$. For a material with tetragonal symmetry, $F$ can be expressed by\cite{38}:

$$F = -\mathbf{H} \cdot \mathbf{M} + \frac{M}{2} \left[ 4\pi M_{\text{eff}} \cos^2 \theta - \frac{H_{4\perp}}{2} \cos^4 \theta ight.$$

$$- \frac{H_{4\parallel}}{8} \left( 3 + \cos 4\phi \right) \sin^4 \theta - H_{2\parallel} \sin^2 \theta \sin^2 \left( \phi - \frac{\pi}{4} \right) \left. \right]$$

where $\theta$ and $\phi$ are angles of magnetization $M$ in the equilibrium position with respect to the film normal and in-plane easy axes, respectively. The first term in equation is the Zeeman energy and the second term is the effective demagnetizing energy ($4\pi M_{\text{eff}}$) which includes both the shape anisotropy ($4\pi M_s$) and out of plane uniaxial
Figure 2.8: **FMR spectrum of 20-nm YIG thin film** Room-temperature FMR derivative spectrum $dI_{FMR}/dH$ vs $H$ of a 20-nm YIG film at $\theta_H = 90^\circ$ (field in-plane) gives a linewidth of 7.42 Oe.

The magnetocrystalline anisotropy $M_s - H_{2\perp}$, where $4\pi M_{eff} = 4\pi M_s - H_{2\perp}$)[38]. The remaining terms are out-of-plane cubic anisotropy ($H_{4\perp}$), in-plane cubic anisotropy ($H_{4||}$), and in-plane uniaxial anisotropy ($H_{2||}$). Since the the lattice constant mismatch between YIG and GGG substrate is essentially zero, the magnetocrystalline anisotropy induced from strain is negligible[39], so the obtained saturation magnetization is nearly identical to values reported for single crystal YIG which is typically around 1750Oe in the bulk.

To confirm the saturation magnetization value measured by FMR angular dependence, we also did the VSM measurement of the YIG thin film in Fig. 2.10 which shows an in-plane magnetic hysteresis loop taken at room temperature with a very small coercivity ($H_c$) of 0.35 Oe and exceptionally sharp reversal: the magnetic
switching is completed within 0.1 Oe. This indicates the essential absence of defects and high magnetic uniformity in the YIG film which provides a good platform for magnetic spin dynamics study.

### 2.3 Ferromagnetic resonance driven spin pumping study in YIG-based FM/NM bilayer structures

Ferromagnetic resonance (FMR) driven spin pumping\[20, 40, 41, 42, 43\] of pure spin currents has generated intense interest for its potential application in next-generation spintronics\[44, 41, 45\]. It is actually a reverse process of spin transfer...
torque in which process spins ejected from non-magnetic transition metals to manipulate the magnetization and spin dynamics in ferromagnetic materials\cite{3, 4, 5, 46, 47, 48}. Under the FMR resonance condition, the magnetization $M$ on the ferromagnetic materials will precess around equilibrium position with certain cone angle $\theta$ as shown in Fig. 2.11. The precession cone angle $\theta$ is expressed by\cite{20, 14, 49}:

$$\theta = \frac{\gamma h_{rf}}{2\alpha \omega} \hspace{1cm} (2.2)$$

Where $\gamma$ is gyromagnetic ratio, $h_{rf}$ is microwave field, $\alpha$ is Gilbert damping constant, and $\omega$ is the microwave frequency. The Gilbert damping constant describes the spin relaxation rate of the precession magnetization to the equilibrium position.
Figure 2.11: Schematic for the angular momentum transfer from precessing magnetization to the nearby nonmagnetic materials.

Due to the dynamic coupling, the free conduction electrons in the normal metals side could couple with the precessing magnetization $M$ of YIG and get spin polarized. Thus, there will be pure angular momentum transfer $J_S$ from YIG to nonmagnetic metal sides. The transferred spin currents can be phenomenologically expressed by the equation below:

$$J_S = g_{\uparrow\downarrow} f \theta^2 P e$$ \hspace{1cm}(2.3)$$

Where $g_{\uparrow\downarrow}$ is spin mixing conductance[50, 51, 52] which quantifies the interfacial spin transfer efficiency derived from scattering matrix, The factor $P$ arises from the ellipticity of the magnetization precession which is a constant in our experiment. Spin
mixing conductance \( g_{\uparrow\downarrow} \) can be experimentally determined from the Gilbert damping enhancement of ferromagnetic layer before and after the deposition of nonmagnetic layer. Several groups have reported high \( g_{\uparrow\downarrow} \) values at YIG/Pt interface confirming the angular momentum transfer. The spin current increases with increasing excitation of the FM magnetization which can be characterized by the opening angle of the cone described by the precessing magnetization. Generating a high spin current density with a modest radio-frequency (rf) field, \( h_{\text{rf}} \), requires a FM with low damping and YIG is highly attractive for this purpose.

### 2.3.1 Inverse spin Hall effect

The inverse spin Hall effect (ISHE)[53, 21, 20] as shown in Fig. 2.12 is an effective tool for studying spin pumping from FMs into nonmagnetic materials (NM). ISHE effect essentially is a inverse process of spin Hall effect [54, 55] in which pure spin currents are generated by electrical charge currents in non-magnetic materials with spin orbit coupling. The pure injected spin currents could be converted to charge currents due to the spin orbit coupling strength in NMs which is actually an inverse process of spin Hall effect which has been well established in metals and semiconductors. We first define the out-of-plane direction as up spin, thus down spin will be in the opposite direction, pure spin currents with down spin polarization move up equalize to the up spin moving down. Moving charges in the electric field \( E \) will experience effective \( B \) field due due to the realistic effect. The experienced \( B \) is proportional to the cross product of effective \( E \) and momentum \( P \). The Hamiltonian \( H \) is proportional to cross product of effective \( B \) field and spin polarization \( \sigma \). The net force experienced by spins carried by electrons is proportional to the gradient of the Hamiltonian. So for
the spin up and down and moving in the opposite directions, the experienced force is actually in the same direction. For the FMR driven spin pumping picture, pure spin currents injected from ferromagnetic materials side will be scattered to charge currents and electrically detected\[56, 57, 58, 59, 60\].

\[
B \sim E \times P \quad (2.4)
\]

\[
H \sim (E \times P) \bullet \sigma \quad (2.5)
\]

\[
H \sim \nabla[(E \times P) \bullet \sigma] \quad (2.6)
\]
Pt has been regarded as one of the most popular spin detectors due to the large atomic number and consequent large spin orbit coupling strength[20, 11]. In addition to Pt, \( \beta \)-phase W and Ta are expected to generate large ISHE voltages (though of the opposite sign), making them attractive in this role as well. In the following paragraphs, we report observation of ISHE voltages, \( V_{\text{ISHE}} \), of 2.10 mV (0.420 mV/mm) and 5.26 mV (1.05 mV/mm) for Pt(5nm)/YIG(20nm) and W(5nm)/YIG(20nm) bilayers, respectively, excited by a rf field of 0.3 Oe in a FMR cavity[25].

2.3.2 Large spin pumping signal in YIG/Pt and YIG/W bilayers

There are two common methods in generating magnetic resonance in FMs for spin pumping, cavity FMR and microstrip waveguide. FMR cavities produce modest-strength, uniform rf fields over a relatively large space (cm-scale); while microstrip waveguides produce rf fields typically in micron to sub-mm scale, and when made very close to the FMs, can generate fairly large \( h_{\text{rf}} \). The most relevant parameter for FMR spin pumping is the rf field, which is non trivial to obtain \( h_{\text{rf}} \) for microstrip spin pumping measurements. Our spin pumping measurements are conducted at room temperature on three bilayer samples: Pt(5nm)/YIG-1, Pt(5nm)/YIG-2 and \( \beta \)-W(5nm)/YIG-2, all made by off-axis sputtering technique. The samples with approximate dimensions of 1 mm \( \times \) 5 mm are placed in the center of the FMR cavity with \( H \) applied in the \( xz \)-plane while the ISHE voltage is measured across the 5 mm long Pt or W layer along the \( y \)-axis, as illustrated in Fig. 2.13. The transfer of angular momentum to the Pt or W conduction electrons resulting from FMR excitation of the YIG magnetization (\( M \)) can be described as a spin current \( J_s \) injected along the \( z \) axis with its polarization (\( \sigma \)) parallel to \( M \). This spin current is converted by spin-orbit
interactions to a charge current $J_s \sim \theta_{SH} \times J_s$, where $\theta_{SH}$ is the spin-Hall angle of Pt or W. Figure 2.14 shows the $V_{ISHE}$ vs. $H$ spectra for Pt/YIG-1 and W/YIG-2 at $\theta_{SH} = 90^\circ$ (field in-plane) and $P_{rf} = 200$ mW, which generates an estimated rf field $h_{rf} \sim 0.3$ Oe. At this moderate $h_{rf}$ excitation, $V_{ISHE}$ reaches a large value of 2.10 mV (0.35 mV/mm) in Pt/YIG-2 as shown in Fig. 2.14, significantly larger than previously reported spin pumping signals using cavity FMR. The W/YIG-2 bilayer exhibits an even larger $V_{ISHE}$ of -5.26 mV (-1.05 mV/mm), where the negative sign reflects the opposite spin Hall angles of W and Pt[25]. Figure 2.15 shows the rf-power dependence of $V_{ISHE}$ for Pt/YIG-2 and W/YIG-2 at $\theta_{SH} = 90^\circ$. The linear relationship between $V_{ISHE}$ and $P_{rf}$ indicates that the observed ISHE voltage is not near saturation and can potentially be further increased by larger $h_{rf}$ ($\sim 0.3$ Oe) in our measurements) since $V_{ISHE} \sim P_{rf} \sim h_{rf}^2$. Figure 2.16 shows a series of $V_{ISHE}$ normalized by the maximum value for varying $\theta_H$ at $P_{rf} = 200$ mW for the two samples. $V_{ISHE}$ is antisymmetric about $\theta_H=0$ as expected from FMR spin pumping.
since the reversal of $H$ switches $M$ (hence $\sigma$) and, consequently, changes the sign of $J_c$. When $H$ is rotated from in-plane ($\theta_H = 90^\circ$) to out-of-plane ($\theta_H = 0^\circ$ or $180^\circ$), $V_{\text{ISHE}}$ gradually vanishes. $M$ follows $H$ at all angles since $2500 \text{ Oe} \leq H_{\text{res}} \leq 5000 \text{ Oe}$, all larger than $4\pi M_{\text{eff}} = 1794 \text{ Oe}$ of our YIG film. The observed angular dependence of $V_{\text{ISHE}}$ for Pt/YIG-2 and W/YIG-2 normalized by the maximum magnitude of $V_{\text{ISHE}}$ at $\theta_H = 90^\circ$ demonstrates a clear sinusoidal shape which is characteristic of ISHE confirming that the observed ISHE voltage arises from FMR spin pumping. The spin pumping signals we observed in insulating YIG cannot be explained by artifacts due to thermoelectric or magneto-electric effects, such as anisotropic magnetoresistance (AMR) or anomalous Hall effect (AHE)[32, 11].

While a spin current is generated by transfer of angular momentum from YIG to metal, simultaneously, the coupling between YIG and metal exerts an additional damping to the magnetization precession in YIG, resulting in increased linewidths for the three samples before ($\Delta H_0$) and after ($\Delta H_1$) the deposition of Pt or W. A
Figure 2.15: Linear rf-power dependence of $V_{\text{ISHE}}$ with a least-squares fit is shown for the YIG/Pt and YIG/W samples.

clear linewidth broadening is observed for all three samples: $\Delta H_1 - \Delta H_0 = 19.9$, 24.3 and 12.3 Oe for Pt/YIG-1, Pt/YIG-2 and W/YIG-2, which give $V_{\text{ISHE}}$ of 1.74, 2.10 and 5.26 mV, respectively. It is generally believed that the FMR linewidth of YIG largely determines the spin pumping efficiency. However, we note that the magnitude of $V_{\text{ISHE}}$ appears to be more correlated to the linewidth change than the original linewidths of the YIG films: Pt/YIG-2 has larger linewidth increase (24.3 Oe) and $V_{\text{ISHE}}$ (2.10 mV) than Pt/YIG-1 ($\Delta H_1 - \Delta H_0 = 19.9$ Oe, $V_{\text{ISHE}} = 1.74$ mV) although
YIG-2 ($\Delta H_0 = 11.7$ Oe) has a larger linewidth than YIG-1 ($\Delta H_0 = 7.4$ Oe). This can be understood from the perspective of interfacial spin mixing conductance which is determined by the linewidth broadening$^{[13, 17, 50]}$.

$$g_{\uparrow\downarrow} = \frac{e^2 \sqrt{3} \pi M_s \gamma t_F}{g \mu_B w} (\Delta H_f - \Delta H_0) \quad (2.7)$$

where $g_{\uparrow\downarrow}$, $\gamma$, $g$ and $\mu_B$ are real part of the spin mixing conductance, the gyromagnetic ratio, $g$ factor and Bohr magnetron, respectively. Using Eq. (2), we obtain $g_{\uparrow\downarrow} = 4.56 \times 10^{14} \Omega^{-1} m^{-2}$ and $2.30 \times 10^{14} \Omega^{-1} m^{-2}$ for Pt/YIG-2 and W/YIG-2, which agree with the theoretical calculations and are among the highest of reported experimental values.

Previously, spin pumping of Pt/YIG excited by similar cavity FMR as used here gave ISHE voltages in the $\mu$V range. The large spin pumping signals observed in our YIG thin films may be attributed to two possible reasons. First, the epitaxial
YIG films made by UHV off-axis sputtering are different in crystalline quality and magnetic resonance characteristics from the YIG films made by LPE and PLD. The second possibility is the small thickness (20 nm) of our films compared to LPE films (100 nm or larger) may play an important role, as suggested by a recent report that a 200-nm YIG film shows much higher spin pumping efficiency than 1-µm and 3-µm films excited by a microstrip waveguide. Compared to cavity FMR, microstrip waveguides can potentially provide much stronger rf fields, e.g. 16 Oe [15], which can significantly increase the magnitude of ISHE voltages ($V_{\text{ISHE}} \sim h_{\text{rf}}^2$ in the linear regime). Further investigation of spin pumping in these thin YIG films using microstrip waveguides will access larger dynamic range and reveal the determining factors for the observed large spin pumping signals. In addition, the mV-level ISHE voltages reported here using a moderate $h_{\text{rf}}$ will allow miniaturization of spin pumping structures while maintaining signals sufficiently large to explore opportunities such as magnon-based electronics and other next generation technologies. It also provides a material platform for probing the fundamental mechanisms in spin pumping for quantitative characterization of coupling mechanisms and interfacial phenomena.

2.3.3 Key factors contributing to large ISHE signals in our YIG-based structures

Lastly, we comment below on the important factors that lead to the exceptionally large ISHE signals observed in our YIG-based structures. (1) The spin current generated in YIG/NM bilayers depends on how far the YIG magnetization can be excited away from equilibrium, which can be described by precession cone angle [13, 17, 50]. This demands YIG films with low damping, a widely accepted criterion in the spin pumping field. (2) However, the measured values of $\alpha$ is a bulk property of the YIG
films, while spin pumping is determined predominantly by the YIG/NM interface because exchange interaction (effective distance ≈2 angstrom) is the dominant mechanism for spin pumping [50]. This requires pristine YIG surface that enables similarly large precession cone angle at the interface as inside the film. Thus, the YIG films need to maintain correct stoichiometry, high crystalline quality, and uniform magnetic ordering from inside to the top atomic layer of the YIG film. Post-deposition treatments, such as polishing, etching, or even annealing, could jeopardize the YIG surface. (3) Spin mixing conductance is a phenomenological parameter that describes the quality of the YIG/NM interface in conducting spin currents. The values of $g_{\uparrow\downarrow}$ vary significantly for the same YIG/Pt structure made by different techniques and research groups due to variation of the interface quality[26, 25]. Since characterizing the chemical, structural, and magnetic uniformity of the YIG surface is rather challenging, ISHE voltage is a good quantity for comparing the spin pumping efficiency and YIG/Pt interface quality. After all, $V_{\text{ISHE}}$ is a direct measure of the pure spin currents pumped into Pt. (4) We find that oxygen content during the YIG growth is critical for its spin pumping performance. YIG thin films with the most desirable spin pumping properties can only be grown within a narrow window of the oxygen partial pressure, outside which, the ISHE signals degrade dramatically. Some of these points are supported by experimental evidence such as the oxygen tuning of YIG quality and some are our speculations, such as the correlation between the interfacial condition and film quality. More detailed characterizations of the YIG/NM interfaces are needed to understand the nature of spin transfer from dynamically excited YIG to metals.
2.3.4 Spin pumping study of spin Hall angle values for a series of $3d$, $4d$, and $5d$ transition metals

Using FMR driven spin pumping technique, we can further determine the spin orbit coupling strength characterized by spin Hall angle values in a series of transition metals. Spin-orbit coupling (SOC) is the underlying mechanism for some of the most important phenomena in condensed matter physics such as magneto-crystalline anisotropy, spin Hall effect and recently topological insulators. SOC is determined by the product of spin and orbital moments. General understanding believes SOC should follow the $Z^4$ dependence on the atomic number ($Z$) and will only be significant in those heavy metals. In 2008, Tanaka[61, 62] predicted that the interaction is also sensitive to the orbital electron filling, in particular to those $d$ orbital electrons as

Figure 2.17: Tanaka’s prediction of spin Hall conductances in a series of $4d$ and $5d$ transition metals.

shown in Fig 2.17. The strength of SOC is reflected by the magnitude of spin Hall angles $\theta_{SH}$, which can be obtained in FMR spin pumping study. Due to the dynamic coupling, spin current is generated at the YIG/NM interface, $V_{\text{ISHE}}$ detected in NM reflects the magnitude of charge currents. Spin hall angle actually quantifies the spin-charge interconversion capability[20, 21].

$$\theta_{SH} = \frac{V_{\text{ISHE}}}{RW\lambda_{SD}\tanh(t_N/2\lambda_{SD})}$$  \hfill \text{(2.8)}
Where $\theta_{SH}$, $R$, $W$, $\lambda_{SD}$ are spin Hall angle, sample width, resistance, and spin diffusion length respectively. Spin diffusion length could be obtained from the nonmagnetic materials thickness dependence of the spin pumping signal. $R$, $W$, $V_{ISHE}$ could be measured experimentally. The spin Hall angle values determined from our spin pumping study are shown in Fig. 2.18.

This figure shows systematic variation of spin hall angles both in sign and magnitude obtained from our spin pumping measurements for 3$d$ and 5$d$ transition metals[23, 22]. X axes is the total number of electron in $d$ and $s$ orbitals. $\theta_{SH}$ changes sign when the number of $d$ orbital electrons go across half filling. Moreover, according to the $Z^4$ dependence, SOC in 3$d$ metals should be negligibly small, however, we observe surprisingly large spin hall angle in 3$d$ metals, especially Cr and Ni with a magnitude even comparable to the spin Hall angles of 5$d$ metals. The dramatical variation in magnitude and systematical change of the sign indicate difference in the SOC strength and multiple underlying mechanisms. For example, 5$d$ metal W gives the largest spin hall angle value, -0.14[22], while the value for 3$d$ Cu is about two order of magnitudes smaller, and of the opposite sign. Also, largest spin hall angle is observed when $d+s=6$ and 10. These results demonstrate important role played by $d$ orbital electron filling on the spin hall physics in transition metals[63], which agrees well with Tanaka’s prediction. Especially, the Cr and Ni give the two largest spin Hall angle values within the 3$d$ metals which are quite promising for future application in the spintronics based devices. To test the generally believed $Z^4$ dependence on atomic number, we purposely choose metals Cu, Ag, and Au with filled $d$ orbitals to minimize the contribution of $d$ orbital electron to SOC. The measured spin hall angles of these three metals roughly follow a nice $Z^4$ dependence[22] as shown in Fig.
Figure 2.19: The determined spin Hall angle values for the three transition metals Au, Ag, and Cu roughly follow the $Z^4$ dependence. 2.19, demonstrating the role of atomic number played in spin Hall physics. Combined with our previous results, we conclude that atomic number and $d$ orbital electrons can be comparably important in determining the spin orbit coupling in transition metals[22, 64].

2.3.5 Magnetoresistance observed in YIG/Pt bilayer structures

Recently, magnetoresistance was reported in YIG/Pt bilayer structures which are explained by either proximity effect[11] or spin Hall magnetoresistance(SMR)[65, 66, 67]. The proximity at YIG/Pt interface could induce some magnetization on Pt side which can give the anomalous Hall effect in Pt layer. Another explanation of the observed magnetoresistance comes from the integration of spin Hall effect and inverse
spin Hall effect which can change the resistance of the Pt layer due to the spin transfer torque [68, 69] depending on the relative orientation of the spin polarization $\sigma$ induced in Pt layer and the YIG magnetization $M$. For Pt thin layer exhibiting strong spin orbital coupling strength, due to spin Hall effect (SHE), the applied electrical current flow $J_e$ along the film plane will cause spin current flow $J_s$ traveling perpendicular to the film surface with spin polarization $\sigma$ parallel to the surface. Spin polarized electrons in Pt could exchange couple with YIG magnetization $M$ and get absorbed via spin transfer torque, thus spin current reflection at YIG/Pt interface will be suppressed. The spin current absorption is maximized when $\sigma$ is perpendicular to $M$ and minimum when parallel. Thus, SMR due to the combined effect of SHE and inverse spin Hall effect (ISHE) is maximum when $M$ is perpendicular to $\sigma$ and minimum when parallel. 5nm Pt was in-situ deposited on 20nm epitaxial YIG film and YIG/Pt bilayer has been patterned into standard Hall bar structure via photolithograph and ion milling process. We measure the angular dependence of $MR$ at two different angles $\alpha$ and $\gamma$ as shown in schematics Fig.2.20. Figure 2.21 shows the resistance change $\Delta R$ as a function of magnetic field $H$. In the experiment, $H$
was applied in the film plane and parallel with $J_e$ which gives $\alpha = 0^\circ$. As shown in the Fig. 2.20. YIG/Pt bilayer exhibits a clear resistance change in the field range $\Delta H \leq 3\text{Oe}$. Resistance increases when increasing magnetic field $H$ from 0Oe. When $H \geq 3\text{Oe}$, $R$ remains almost constant with a small linear background coming from the joule heating. The field range that resistance changes coincides with the re-magnetization process of YIG film. Figure 2.21 shows in plane magnetic hysteresis

Figure 2.21: Room temperature in-plane magnetic hysteresis loop of a YIG(20 nm)/Pt(5 nm) bilayer and field dependence of resistance and magnetoresistance of the YIG/Pt bilayer with an in-plane field which is applied parallel to the electric current.
loop measurement of a typical 20nm YIG film. The effective magnetization $4\pi M_{eff}$ is estimated to be around 1750Oe[22] indicating negligible strain induced in-plane anisotropy. The saturation of magnetization completes around $H = 3$Oe suggesting that the observed $MR$ comes from the different orientations of YIG magnetization. As shown in Fig. 2.21 When $\alpha = 0^\circ$, we observe 0.073% $MR$ ratio, larger than the value reported elsewhere, which most likely comes from optimal YIG/Pt interfacial condition and film quality. When $\alpha = 45^\circ$, $MR$ decreases to 0 approximately, while for $\alpha = 90^\circ$, the sign of $MR$ changes to negative. $\alpha$ angular dependence of $MR$ follows a $\sin^2\alpha$ trend as shown in Fig. 2.22. The in-plane angular dependence of $MR$ cannot exclude the possible contribution from anisotropic magnetoresistance (AMR)[32] if Pt gets magnetized at the YIG/Pt interface due to the proximity effect which also follows the similar angular dependence. When rotating $\gamma$, the angle between $J_c$ and $H$ changes

Figure 2.22: In-plane angular dependence of MR for the YIG(20 nm)/Pt(5 nm) bilayer from $\alpha = 0^\circ$ to $180^\circ$. 

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while the relative orientation between $\sigma$ and $M$ remains constant. If the observed resistance change $\Delta R$ mainly comes from SMR, $MR$ ratio should keep constant when changing $\gamma$, otherwise, $MR$ will still follow $\sin 2\gamma$ trend if AMR is the dominant effect. Figure 2.23 shows the $\gamma$ angular dependence of $MR$ ratio, $MR$ equals 0.081%, 0.084%, and 0.077% at 60°, 150°, and 240° as shown which roughly remains a constant value and does not change sign indicating that the observed $MR$ in our system mainly comes from $SMR$ induced from spin current absorption and reflection rather than AMR [70, 71]. Further study, such as the temperature dependence study could provide clear information about the relative contribution of SMR and proximity effect to the observed magneto-resistance in our YIG/Pt bilayer structures.

![Graph](image)

**Figure 2.23:** Out-of-plane angular dependence of MR from $\gamma = 0^\circ$ to $360^\circ$. 
Chapter 3: FMR spin pumping mechanism study

Generation and manipulation of spin currents is centrally important for spintronic applications. FMR driven spin pumping\[^{72, 73}\] has been demonstrated to inject a pure spin current through angular momentum transfer from an FM to an adjacent NM\[^{74, 75, 76, 77, 78}\]. It is generally believed that this dynamic coupling proceeds by means of the exchange interaction between the precessing magnetization ($M$) of the FM and the conduction electrons of the NM at the NM/FM interface. This mechanism will lead to a short-range coupling that decays exponentially on atomic length scales with separation between the FM and NM\[^{79, 80}\]. However, this mechanism has not been experimentally confirmed, partially due to the large dynamic range needed to measure such rapidly decaying spin pumping signal. Our demonstration of large spin pumping\[^{25}\] in Pt/YIG bilayers with mV-level inverse spin Hall effect (ISHE) voltage, $V_{\text{ISHE}}$, offers a material platform with signal-to-noise ratio sufficient to quantitatively characterize the spin pumping coupling range, enabling detailed insight into the spin pumping mechanism. We have done systematic measurements with four different barrier materials, including three oxide insulators and Si, to investigate the barrier thickness $t$ dependence of spin pumping in Pt/barrier($t$)/YIG heterostructures. We observe clear exponential decays of ISHE voltage with characteristic length scales of
0.2 nm for the oxide barriers; these data provide decisive evidence for the predicted exchange coupling model for spin pumping.

### 3.1 Exchange coupling and dipole coupling

In the magnetism study, ferromagnetism comes from the long-range alignment of the atomic magnetic moments even without the external magnetic field. In classical mechanics, magnetic field could be created by moving charge carriers and an electric current in a closed loop producing a magnetic moment. In a microscopic scale, the circular movement of electrons around nucleus gives the orbital angular moment. In quantum mechanics, electrons also possess another intrinsic moment called spin which could take two states, up and down. The total magnetic moments of atoms come from both the spin moments and orbital angular momentum of the electrons. There are two main interactions between electrons, classical dipole interaction and quantum exchange interaction. In Heisenbergs picture, the exchange energy between spin moments can be expressed as:

\[ E_{ex} = -2\Sigma_i \Sigma_j^N J_{ij}^{ex} S_i S_j \]  

(3.1)

where \( J_{ij}^{ex} \) is exchange integral describing the coupling between \( S_i \) and \( S_j \). In ferromagnetic materials, \( J_{ij}^{ex} \) is positive, so when \( S_i \) and \( S_j \) point to the same direction, \( E_{ex} \) is the lowest which corresponds to the ground state. If \( J_{ij}^{ex} \) is negative, \( E_{ex} \) is the lowest when \( S_i \) and \( S_j \) are opposite, resulting in anti-ferromagnetic alignment. The coupling distance of exchange interaction is typically short, on the scale of atomic distance. On the other hand, two magnetic moments can couple over longer distances.
via dipolar interaction, which can be expressed as:

\[ E_{dip} = \frac{S_i S_j}{r_{ij}^3} - 3\frac{(S_i S_j)(S_j S_j)}{r_{ij}^5} \]  \hspace{1cm} (3.2)

where \( r \) is the relative distance between \( S_i \) and \( S_j \). Typically, the magnitude of dipolar interaction is much smaller than exchange interaction. In ferromagnetic materials, exchange interaction dominates and overcome the thermal fluctuation. This results in spontaneous parallel alignment of the magnetic moments, creating a macroscopic magnetic moment (magnetization \( M \)) even without external field.

### 3.2 Characterization of the insulating barrier materials

\( \text{Sr}_2\text{GaTaO}_6, \text{Sr}_2\text{CrNbO}_6 \) barriers are both insulators with the \( \text{A}_2\text{BB}'\text{O}_6 \) double Perovskite structure\[36, 81\], which can be viewed as combination of two different single Perovskites (similar to \( \text{SrTiO}_3 \)). Double Perovskites exhibit a broad range of interesting properties due to their complexity and tenability, and have been used in various applications. For example, \( \text{LSAT} \) is a commonly used single crystal substrate for epitaxial film growth with the composition \((\text{LaAlO}_3)_{0.3}(\text{Sr}_2\text{AlTaO}_6)_{0.7}\), of which the majority phase \((\text{Sr}_2\text{AlTaO}_6)\) is a double Perovskite very similar to the \( \text{Sr}_2\text{GaTaO}_6 \) used in this report. \( \text{Sr}_2\text{CrNbO}_6 \) has an almost identical lattice constant as \( \text{Sr}_2\text{GaTaO}_6 \) \((a = 0.788 \text{ nm})\) and have been used as an epitaxial buffer layer for subsequent growth of other double Perovskite epitaxial films. The in-plane bulk lattice constants of \( \text{Sr}_2\text{GaTaO}_6, \text{SrTiO}_3, \text{Sr}_2\text{CrNbO}_6 \) are 3.947, 3.905, and 3.945 angstrom respectively, which is actually pretty close with each other within 1% mismatch. We have successfully achieved epitaxial heterostructures based on combination of double Perovskites and reported the manipulation of magnetocrystalline anisotropy of double perovskite \( \text{Sr}_2\text{FeMoO}_6 \) on substrates or buffer layers with different lattice constants\[36, 82, 83\],
such high quality fabrication of those kinds of materials lay a solid foundation for our spin pumping study in YIG/barrier/Pt trilayer structures.

Thin Sr$_2$GaTaO$_6$ and Sr$_2$CrNbO$_6$ layers of various thicknesses were deposited on YIG films in the same sputtering system at room temperature by rf sputtering using power of 60 and 50 W respectively[36]. The Ar/O$_2$ sputtering pressure is 11.5 and 12.5 mTorr and the O$_2$ concentration is between 0.15% and 0.2%. The deposition rates are 1.3 and 0.50 nm/min for Sr$_2$CrNbO$_6$ and Sr$_2$GaTaO$_6$, respectively. The single crystalline nature of the double prevoskite Sr$_2$CrNbO$_6$ and Sr$_2$GaTaO$_6$ thin films can be verified by XRD scan as shown in Fig. 3.1. The Laue oscillations also demonstrate the smoothness, uniformity and sharp interface.

Figure 3.1: XRD scan of SGTO and SCNO thin films grown on different substrates. The single diffraction peak reveals the pure double Perovskite phase.
Diffuse reflectance ($R$) spectra of the $\text{Sr}_2\text{GaTaO}_6$ and $\text{Sr}_2\text{CrNbO}_6$ samples were collected on an Ocean Optics USB4000-UV-VIS miniature fiber optic spectrometer using a Spectralon standard. The percent reflectance was transformed using the Kubelka-Munk function:

$$F(R) = \frac{(1 - E)^2}{2R}$$

(3.3)

The spectra are shown in Fig.3.2. The intersection of the linear fit and photon energy axis gives band gaps of 4.91 eV ($\text{Sr}_2\text{GaTaO}_6$) and 2.36 eV ($\text{Sr}_2\text{CrNbO}_6$).

Figure 3.2: UV-visible absorption spectra for $\text{Sr}_2\text{GaTaO}_6$ (red) and $\text{Sr}_2\text{CrNbO}_6$ (blue) using the Kubelka-Munk function.
3.3 FMR spin pumping study in YIG/SrTiO$_3$/Pt trilayer structures

For YIG/Pt bilayer structure, the spin pumping measurements are conducted at room temperature using a 5-nm thick Pt layer on 20-nm YIG films as shown previously in Fig. 2.13. The samples (∼1mm wide and ∼5mm long) are placed in the center of an FMR cavity and in a DC magnetic field ($H$) applied in the $xz$-plane. At resonance, the precessing magnetization transfers angular momentum from the YIG to the conduction electrons in Pt by means of dynamical coupling, generating a pure spin current, $J_s$, in Pt directed along the $z$-axis with a polarization ($\sigma$) parallel to the YIG magnetization. In the Pt layer, $J_s$ is converted into a net charge current, $J_s \sim J_s \times \sigma$, via the inverse spin Hall effect, resulting in an ISHE voltage $V_{\text{ISHE}}$ along the $y$-axis. Figure 3.3 shows $V_{\text{ISHE}}$ vs. $H$ spectra for a Pt/YIG bilayer at $\theta_H = 90^\circ$ and $270^\circ$ (field in plane) at $P_{rf} = 200$ mW. The peak value of $V_{\text{ISHE}} = 1.0$ mV is generated at the FMR resonant condition as illustrated by the FMR derivative spectrum. As $H$ is reversed from $\theta_H = 90^\circ$ to $270^\circ$, $V_{\text{ISHE}}$ changes sign and maintains the same magnitude as expected since $\sigma$ changes sign with reversal of $M$ ($M$ is parallel to $H$ since $H$ exceeds $4\pi M_s$ at resonance field), resulting in a reversed sign of $V_{\text{ISHE}}$.

In order to probe the spin pumping coupling strength and length, we insert four different thin, insulating barriers, Sr$_2$GaTaO$_6$ (SGTO) with a band gap $E_g = 4.91$ eV, SrTiO$_3$, (STO) with $E_g = 3.40$ eV, Sr$_2$CrNbO$_6$ (SCNO) with $E_g = 2.36$ eV and amorphous Si, between Pt and YIG as illustrated in Fig. 3.4 and 3.5. Figures show the spin pumping spectra of the Pt/barrier/YIG structures with 0.5-nm Sr$_2$GaTaO$_6$, SrTiO$_3$, Sr$_2$CrNbO$_6$ and Si barriers; these barriers reduce the ISHE voltage to 20, 60, 100 and 440$\mu$V, respectively. We will discuss the variation of the decay rate of $V_{\text{ISHE}}$ in details.
Figure 3.3: $V_{\text{ISHE}}$ vs $H$ spectra and corresponding FMR derivative spectrum of a Pt (5nm)/YIG(20 nm) bilayer at $P_{rf} = 200$ mW.

below. The angular dependence of normalized $V_{\text{ISHE}}$ for Pt/barrier(0.5nm)/YIG with the four barriers all give the sinusoidal angular dependence which is characteristic of the ISHE, confirming that the observed signals are due to spin pumping, not artifacts due to thermoelectric or magnetoelectric effects such as anisotropic magnetoresistance (AMR). The power dependence of $V_{\text{ISHE}}$ from $P_{rf} = 0.2$ to 200 mW with an in-plane field ($\theta_H = 90^\circ$) for the four samples, all show a linear relationship between $P_{rf}$ and
$V_{\text{ISHE}}$ indicating that the measured inverse spin Hall signal still in the linear regime.

![Graphs showing $V_{\text{ISHE}}$ vs $H$ for Pt/barrier/YIG multilayers with different barriers.]

Figure 3.4: $V_{\text{ISHE}}$ vs $H$ of Pt/barrier/YIG multilayers with 0.5-nm thick (a) Sr$_2$GaTaO$_6$, (b) SrTiO$_3$.

To further probe the underlying spin pumping mechanism, we plot the barrier thickness dependence of the ISHE voltage as shown in Fig. 3.6. For SGTO spacer, the spin pumping signal decays more than two orders of magnitude when only 1nm SGTO spacer inserted between YIG and Pt. The systematic behavior of spin pumping across a thin insulating barrier becomes evident when the dependencies of $V_{\text{ISHE}}$ normalized by the Pt/YIG samples with direct contact on the barrier thickness are plotted as shown in Fig 3.7 for the Pt/barrier(t)/YIG heterostructures with Sr$_2$GaTaO$_6$, SrTiO$_3$, Sr$_2$CrNbO$_6$ and Si barriers. Four representative $V_{\text{ISHE}}$ vs. $t$ spectra for various barrier thicknesses are shown for each series. The mV-scale ISHE voltage of Pt/YIG allows us to observe dramatic, thousand-fold changes in $V_{\text{ISHE}}$.
Figure 3.5: $V_{\text{ISHE}}$ vs $H$ of Pt/barrier/YIG multilayers with 0.5-nm thick (a) Sr$_2$CrNbO$_6$, (b) Si.

As the barrier thickness increases, $V_{\text{ISHE}}$ exhibits a clear exponential decay for all three complex oxide barrier materials and eventually falls below the noise level at $t = 2$ nm for Sr$_2$GaTaO$_6$, SrTiO$_3$, Sr$_2$CrNbO$_6$ barriers and at $t = 5$ nm for Si barrier. From a least-squares linear fit shown in the semi-log plots in Figs. 3.7, we obtain an exponential decay length, $\lambda = 0.16$, 0.19, 0.23 and 0.74 nm for Sr$_2$GaTaO$_6$, SrTiO$_3$, Sr$_2$CrNbO$_6$ and Si barriers, respectively, following:

$$V_{\text{ISHE}} = V_{\text{ISHE}}(t = 0)e^{-\frac{t}{\lambda}}$$

(3.4)

Since $V_{\text{ISHE}} \sim J_s$, the exponential decay of $V_{\text{ISHE}}$ indicates that the pure spin current in Pt transferred from YIG layers also decreases exponentially with $t$, a signature behavior of short range exchange coupling between the FM and NM separated by an insulating barrier. This result is a direct quantitative evidence of the exchange coupling model for spin pumping.
3.4 Tunneling process for spin transport across nonmagnetic insulators

The exponential dependence of spin pumping on barrier thicknesses can be explained by a process in which the wave-function of the conduction electrons in Pt tunnels through the barrier, couples with the precessing magnetization of YIG through exchange interaction, and acquires spin polarization via spin-dependent scattering at the barrier/YIG interface. At a NM/FM interface, a spin current can be generated either by transmission of spin-polarized electrons from the FM into the NM, or by spin-dependent scattering of the conduction electrons in NM at the interface. Given
that YIG is an insulator, it is unlikely that spin-polarized electrons flow from YIG into Pt. This suggests that spin-dependent scattering of Pt conduction electrons at the Pt/YIG interface via the exchange interaction with the precessing magnetization of YIG is the dominant mechanism to achieve angular momentum transfer in YIG/Pt spin pumping process.
Figure 3.8: Schematic of band structures of Pt/barrier/YIG heterostructures with an estimated Schottky barrier height $\Phi_B$ at half of the band gap for each barrier material. The blue curve illustrates the quantum tunneling of the electron wave function from Pt into the barrier.

The ability of conduction electrons in Pt to tunnel through the barrier separating the Pt from the YIG will depend sensitively on the height of the energy barrier. At the interface between a metal and an insulator or a semiconductor, the relevant barrier is typically the Schottky barrier, $\Phi_B$, which depends on the work function of the metal and on the electron affinity, charge carrier type and concentration of the insulator/semiconductor. Here we estimate the values of $\Phi_B$ for Pt/Sr$_2$GaTaO$_6$, Pt/SrTiO$_3$, and Pt/Sr$_2$CrNbO$_6$ based on published results on metal/perovskite Schottky junctions and correlate them with our measured decay lengths. The Schottky barrier height in Au/SrTiO$_3$ (Nd-doped, carrier density $10^{17} \sim 10^{18}$ cm$^{-3}$) Schottky junctions, is reported to be in the range $1.4 \sim 1.7$ eV, about half of the SrTiO$_3$ band
gap. At lower carrier concentration, $\Phi_B$ is expected to remain in this range. Au and Pt have similar work functions (5.47 eV for Au and 5.64 eV for Pt), so we expect $\Phi_B$ in Pt/SrTiO$_3$ to be around 1.7 eV (about half of the band gap). Since both Sr$_2$GaTaO$_6$ and SrTiO$_3$ are also Sr-based perovskites, it is reasonable to expect $\Phi_B$ in Pt/Sr$_2$GaTaO$_6$ and Pt///SrTiO$_3$ interface to be half of their barrier band gaps as well. Using band gaps of 4.93 eV for Sr$_2$GaTaO$_6$ and 2.36 eV for Sr$_2$CrNbO$_6$ determined by optical absorption and the reported $E_g = 3.40$ eV for SrTiO$_3$, we estimate $\Phi_B = 2.5$, 1.7 and 1.2 eV for Pt/Sr$_2$GaTaO$_6$, Pt/SrTiO$_3$, and Pt/Sr$_2$CrNbO$_6$ interfaces, respectively. For a finite rectangular potential barrier, as illustrated in Fig. 3.8 for the three oxide barriers, the electron tunneling transmission coefficient $D$ is determined by the barrier height $\Phi_B$ and width $t$:

$$D = \exp\left[-\frac{2t}{\bar{h}} \sqrt{2m\Phi}\right] \quad (3.5)$$

where $m$ is the effective electronic mass and $\bar{h}$ is Planck's constant. Since $V_{\text{ISHE}} \sim J_s \sim D$, Eqs. 3.4 and 3.5 imply $\frac{1}{\lambda} \sim \sqrt{\Phi}$ for Pt/Sr$_2$GaTaO$_6$/YIG, Pt/SrTiO$_3$/YIG and Pt/Sr$_2$CrNbO$_6$/YIG. The three data points are quite consistent with a vanishing intercept, providing further evidence for the exchange coupling model in spin pumping and the role of oxide barrier characteristics in quantum tunneling.

For Si barriers, the decay length of 0.74 nm is much larger than the 0.16, 0.19 and 0.23 nm for samples with oxide barriers. Single crystalline Si has a smaller band gap (1.1 eV) than the three oxide barriers, thus we expect a larger decay length. If we use $\Phi_B = 0.55$ eV for Pt on amorphous Si, we estimate a decay length of 0.34 nm using the same rectangular potential barrier model, smaller than what we have observed, indicating a barrier height smaller than half of the Si band gap as we assumed for the oxide barriers. This is not unexpected, since the Schottky barrier heights of metal/Si
Figure 3.9: Inverse of decay length, $1/\lambda$, as a function of $\sqrt{\Phi_B}$ for Pt/Sr$_2$GaTaO$_6$/YIG, Pt/SrTiO$_3$/YIG and Pt/Sr$_2$CrNbO$_6$/YIG. The solid markers are experimental data and the solid line connecting the three points and origin is a guide to the eye.

junctions are sensitive to the doping type and carrier concentration. In addition to the tunneling mechanism already discussed, we should also consider the possibility that there may be carriers in the Si barrier allowing either an indirect exchange coupling or spin diffusion process through the semiconductor barrier[17]. Further investigation of the characteristics of the barriers in spin pumping is needed to obtain better insights into the spin pumping mechanisms.

In conclusion, experimental observation of a clear exponential decay of dynamic spin pumping provides decisive evidence for the short range exchange coupling and quantitative understanding of a fundamental spin pumping mechanism. This result
also points to the important ability to tune characteristics of spin functional devices and reveal new phenomena.
Chapter 4: FMR spin pumping study of spin transport mechanism in antiferromagnetic insulators

Spin transport in ferromagnetic (FM) and nonmagnetic materials (NM) has been extensively studied. Pure spin currents driven from FMs to metals or semiconductors by ferromagnetic resonance (FMR) or thermal spin pumping have attracted especially intense interest. Though antiferromagnets (AF) should in principle be able to transport spin currents via AF spin waves, they have been largely neglected in this role. Here, we report observation of spin transport from YIG to Pt across an AF insulator, NiO, with thickness ($t_{NiO}$) up to 100 nm by FMR spin pumping[19]. Remarkably, the insertion of a thin NiO layer enhances the already large spin currents driven into Pt by as much as a factor of 7, suggesting higher spin mixing conductances in YIG/NiO/Pt trilayers than for YIG in direct contact with Pt. For $1 \text{ nm} \leq t_{NiO} \leq 50 \text{ nm}$, the spin pumping signals decay exponentially with a length scale of $\sim 10 \text{ nm}$. Together with spin pumping measurements on other magnetic insulators, this result demonstrates robust spin transport through an AF insulator carried by AF magnons, thus opening exciting new opportunities for utilizing pure spin currents in potential applications.
4.1 Spin wave and magnon study in antiferromagnetic insulators

Spin currents in insulators propagate via precessional spin wave modes, e.g., magnons in ordered FMs and AFs. However, it is challenging to excite AF magnons which, for example, requires THz frequency in NiO[84]. Furthermore, the AF ordering temperatures in thin films decrease at lower thicknesses and conventional magnons cannot be sustained above the ordering temperatures. Here, we leverage the established technique of FMR spin pumping in YIG-based structures to excite the AF insulators via exchange coupling to the precessing YIG magnetization and to probe spin transfer in these insulators.

4.2 Spin transport study across NiO

FMR spin pumping in FM/NM bilayers relies on transfer of angular momentum from the precessing FM magnetization to the conduction electrons in the NM to generate spin currents. Pure spin currents in FM insulators by means of magnons have been reported. Theoretical work predicted simultaneous spin and magnon accumulation at a NM/FM-insulator interface and the interconversion between spin current $J_s$ and magnon current $J_m$. Another important class of magnetic materials, antiferromagnets, both metallic and insulating, can also sustain spin wave excitation and propagation. We have demonstrated growth of high-quality YIG epitaxial thin films which enable mV-level inverse spin Hall effect (ISHE) spin pumping signals in YIG/Pt bilayers and confirmed the exchange coupling mechanism for spin pumping[22]. The effectiveness of this platform encouraged our search for spin transport in antiferromagnets.
Recently, inverse spin Hall effect has been reported in antiferromagnetic metal layers \([58, 85, 86]\), spin transport and dynamics in antiferromagnet is a hot topic and attracts a lot of research attention, in this chapter our FMR spin pumping study focuses on three series of YIG/Pt bilayers and YIG/NiO/Pt trilayers prepared from three 20-nm YIG films labeled YIG-1, YIG-2 and YIG-3 with FMR linewidths (bare YIG) of 8.5, 18 and 24Oe, respectively. We find that the YIG film quality is sensitive to the oxygen partial pressure during growth. For the three different series of YIG films studied, we carefully tune the oxygen partial pressure during the film deposition which gives different FMR linewidth. Meanwhile, all three YIG films show equally high crystalline quality as indicated by XRD. We also performed atomic force microscopy (AFM) measurements on the three YIG films as shown in Fig. 4.1. The rms roughnesses of the three films are comparable, ranging from 0.17 to 0.20 nm. These excellent surface conditions play an important role to study the spin transport across AF insulators which will be discussed in details later.

![AFM images of YIG-1, YIG-2, and YIG-3 films over an area of 10 \(10\mu m^2\) give rms roughness of 0.20, 0.17, and 0.19 nm, respectively.](image)

Next, we characterize the spin pumping signal of the three YIG/Pt samples. Fig 4.2 show \(V_{\text{ISHE}}\) vs. \(H\) spectra for the three YIG/Pt bilayers at \(P_{rf} = 200\) mW,
which give $V_{\text{ISHE}} = 3.04\, \text{mV}, 604\, \mu\text{V}, \text{and} 146\, \mu\text{V}$, respectively. The YIG-1/Pt sample exhibits the highest $V_{\text{ISHE}}$ (3.04 mV) we have obtained to date and the three YIG/Pt bilayers are selected to have a wide range of ISHE voltages due to the difference in YIG film/interface quality. The linewidth enhancements after Pt deposition for the three YIG thin films vary from 4.1 to 11.7 Oe, indicating clear difference in exchange coupling strength between YIG and Pt, which is primarily responsible for the difference in observed spin pumping signals. The larger linewidth enhancement directly correlates with larger spin pumping signals, reinforcing the importance of interfacial quality in spin transport.

Figure 4.2: $V_{\text{ISHE}}$ vs $H - H_{\text{res}}$ spectra of at $P_{rf}=200$ mW using (a) YIG-1, (b) YIG-2, and (c) YIG-3.

To explore potential spin transport in AF insulators, we insert a layer of NiO, an AF with a Neel temperature over room temperature, between YIG(20 nm) and Pt(5 nm) in all three YIG series. The insulating nature of the NiO films is confirmed by electrical measurements. We add 1 nm NiO spacer layer and measure the spin pumping signal. Figure 4.3 (a) to (c) shows the $V_{\text{ISHE}}$ vs $H$ spectra for YIG/NiO(1 nm)/Pt trilayers of the three series. Strikingly, we observe a significant enhancement

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of spin pumping signals for all three samples: $V_{\text{ISHE}} = 4.71 \text{ mV}$ (from 3.04 mV), 1.20 mV (from 604 µV), and 1.03 mV (from 146 µV), which is a factor of 1.55, 1.99, and 7.05 increase with the insertion of 1-nm NiO for the YIG-1, YIG-2, and YIG-3 samples, respectively. This is in notable contrast to our previous study of the exponential decay of ISHE voltages by more than two orders of magnitude when a 1-nm oxide insulating spacer inserted between YIG and Pt. Thus, the root of this enhancement of spin pumping efficiency in YIG/NiO(1 nm)/Pt trilayers must lie in the magnetic character of NiO.

Figure 4.3: $V_{\text{ISHE}}$ vs $H - H_{\text{res}}$ spectra of at $P_{rf}=200$ mW using (a) YIG-1/NiO(1 nm)/Pt, (b) YIG-2/NiO(1 nm)/Pt, and (c) YIG-3/NiO(1 nm)/Pt.

The dependence of the spin current injected into Pt on the thickness of the NiO interlayer provides clues as to length scale, and hence the mechanism underlying spin pumping observed here. Figure 4.5 (a) to (c) shows semi-log plots of the dependencies of $V_{\text{ISHE}}$ on $t_{\text{NiO}}$ for the three series of trilayers from 1 to 100 nm. In order to probe the variation of spin pumping at low NiO spacer thickness regime, figure 4.4 (a) to (c) plot the $V_{\text{ISHE}}$ as a function of NiO thickness from 0 to 10 nm for the three series of samples, where the horizontal dashed lines mark the values of $V_{\text{ISHE}}$ for
the YIG/Pt bilayers. We observe three important features. First, at $t_{NiO} \leq 1$ or 2 nm, the ISHE voltages of YIG/NiO($t_{NiO}$)/Pt trilayers increase with increasing $t_{NiO}$. After peaking, the spin pumping signals of the trilayers remain higher than the values of corresponding YIG/Pt bilayers up to $t_{NiO} \geq 5$ nm for the YIG-1 and YIG-2 series and $t_{NiO} \geq 10$ nm for the YIG-3 series. The enhanced ISHE voltages suggest that the overall spin mixing conductance of the entire YIG/NiO/Pt trilayer is higher than the YIG/Pt bilayer with direct contact, indicating the YIG/NiO and NiO/Pt interfaces must be exceptionally efficient in transporting spins. This is quite unusual considering that the trilayers have two interfaces sandwiching NiO, as compared to a single interface in YIG/Pt, and the spin mixing conductance of YIG/Pt is already among the highest in YIG/metal systems. We also note that the YIG-3 sample with the lowest quality of the three series gives the largest enhancement (7 times) of $V_{ISHE}$, suggesting that the combined YIG/NiO and NiO/Pt interfaces are much more transparent in conducting spins than YIG/Pt interface. These observations suggest that NiO has a healing effect for interfacial spin transport.

Figure 4.4: $V_{ISHE}$ as a function of NiO thickness from 0 to 10 nm for the three series of samples, where the horizontal dashed lines mark the values of $V_{ISHE}$ for the YIG/Pt bilayers.
Second, all three series of YIG/NiO($t_{\text{NiO}}$)/Pt trilayers exhibit a clear exponential decay of $V_{\text{ISHE}}$ between $t_{\text{NiO}}$=5 and 50 nm, implying diffusive spin transport in the AF insulator NiO. From least-squares fits to the semi-log plots in Fig. 4.5 in the range $5 \text{ nm} \leq t_{\text{NiO}} \leq 50 \text{ nm}$, we obtain magnon diffusion lengths $\lambda=8.8$, 9.4, and 10.8 nm for the YIG-1, YIG-2, and YIG-3 series, respectively, using $V_{\text{ISHE}} = V_{\text{ISHE}}(t_{\text{NiO}}=1 \text{ nm})e^{-t_{\text{NiO}}/\lambda}$. Here, data for $t_{\text{NiO}} \leq 5 \text{ nm}$ are excluded from the exponential fits.

![Figure 4.5: Semi-log plots of the NiO thickness dependencies of the ISHE voltages for YIG(20 nm)/NiO($t_{\text{NiO}}$)/Pt(5nm) trilayers using (a) YIG-1, (b) YIG-2, and (c) YIG-3.](image)

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Lastly, at $t_{NiO} \geq 50$ nm, the decay of $V_{ISHE}$ becomes slower, as seen from the data points for $V_{ISHE} = 100$ nm (well above the fitting lines) which give $V_{ISHE} = 1.85, 0.61, \text{ and } 0.51 \mu V$, respectively. Clearly, the observation of spin pumping signals in Pt across a 100-nm insulating NiO spacer from YIG excludes tunneling as a mechanism as we discussed previously. The insulating nature of YIG and NiO also rules out anisotropic magnetoresistance (AMR) or anomalous Hall effect (AHE)[32]. In addition, magnetic proximity effect in Pt is not expected given that Pt is on top of antiferromagnetic NiO[11, 87].

For all the three 20-nm YIG series studied, FMR linewidth first decreases when a thin NiO spacer is inserted, then increases and reaches saturation at large NiO thicknesses. We believe that in the low NiO thickness regime, the AF ordering has not been fully established in NiO[88] (AF blocking temperature below room temperature) and AF fluctuation is the dominant mechanism for spin (magnon) transport. At larger NiO thicknesses, the AF ordering is stabilized and the exchange coupling at YIG/NiO interface enhances the damping in YIG, resulting in the increase in FMR linewidth. Eventually, at very large NiO thicknesses (above 50 nm), the AF ordering remains constant and the FMR linewidth reaches saturation. In another control experiment of YIG(100 nm)/NiO($t_{NiO}$)/Pt series, the FMR linewidth does not decrease in the low spacer thickness regime, while we still observe the enhancement of spin pumping. This indicates that the enhancement of spin pumping does not have an obvious correlation with linewidth, demonstrating the YIG/NiO interfacial coupling plays a major role here.

The long-range spin pumping phenomenon most likely arises from magnon-mediated angular momentum transfer in the antiferromagnetic NiO spacer. At the YIG/NiO
interface, a strong, short-range exchange interaction couples the FM magnetization in YIG with the AF moments in NiO. At YIG resonance, the precessing YIG magnetization excites AF spin waves (magnons) in NiO through exchange coupling. The AF magnons carry the angular momentum across the NiO thickness to the NiO/Pt interface, where the angular momentum is transferred across the NiO/Pt interface, generating a spin current in Pt via AF spin pumping. Since the AF magnon transport
in NiO is diffusive, the spin currents show exponential decay as a function of the NiO thickness. The fact that YIG is a ferrimagnet insulator with five Fe$^{3+}$ ions (3 up and 2 down) per formula unit, thus possesses a significant AF sub-lattice magnetization, may play a role in enabling strong coupling between the YIG uniform magnetization and the AF magnetization of NiO for efficient spin transport.

To verify that the observed spin transport across NiO in YIG/NiO/Pt trilayers is mediated by AF magnons, and does not arise from other spurious effects, we grow four different heterostructures on YIG-1 and measure their spin pumping signals as control experiments, as shown in Fig. 4.6. The first sample, YIG/NiO(5 nm)/Cu(10 nm)/NiO(5 nm)/Pt(5 nm), in which we insert a 10-nm Cu (NM metal) spacer in between two 5-nm NiO layers, exhibits $V_{\text{ISHE}} = 1.25\mu V$. This value is about three orders of magnitude smaller than the value of 1.42 mV for YIG-1/NiO(10 nm)/Pt as shown in Fig. 4.4, indicating that spin current can still propagate from YIG to Pt across the three spacers, but the combined spin mixing conductances of the three-layer/four-interface system is much smaller than for the two interfaces involved with the NiO trilayer system. Replacing the 10-nm Cu with a 5-nm SiO$_x$ (NM insulator) layer in control sample (2) eliminates any detectable spin pumping signal, demonstrating that Cu can conduct spin current while nonmagnetic insulator SiO$_x$ blocks spin flow. The third control sample, YIG/NiO(5 nm)/Cu(10 nm), shows no ISHE signal, confirming that the observed spin pumping signal for YIG/NiO/Cu/NiO/Pt indeed comes from the ISHE in the Pt top layer. Lastly, the YIG/Cu(10 nm)/NiO(10 nm)/Pt structure shows a small but still clear ISHE signal of 0.20$\mu V$.

Altogether, this suggests the following multiple-stage spin conversion[89, 90] at the four interfaces in the YIG/NiO/Cu/NiO/Pt multiplayer: 1) at the YIG/NiO
interface, the precessing YIG magnetization injects angular momentum into the first NiO, producing a magnon current $J_m$; 2) the $J_m$ in the first NiO layer carries the angular momentum to the first NiO/Cu interface, where it is converted to a spin current $J_s$ carried by the conduction electrons in Cu; 3) the $J_s$ in Cu propagates to the second Cu/NiO interface where it is converted back to $J_m$ in the second NiO layer; and 4) the $J_m$ in the second NiO layer transfers the angular momentum to the interface with Pt, where it is converted to a spin current in Pt, resulting in an ISHE voltage.

Figure 4.7: (a) FMR derivative absorption spectra taken at $f = 9.65$ GHz and frequency dependencies of FMR linewidth of a bare YIG-1 film, a YIG-1/NiO(20 nm) and a YIG-1/SiO$_x$(20 nm) bilayer. To further study the coupling between YIG and NiO, we measure the FMR linewidth of YIG, YIG/NiO and YIG/SiO$_x$ samples. Figure 4.7a shows the FMR derivative absorption spectra of a bare YIG-1, a YIG-1/NiO(20 nm) and a YIG-1/SiO$_x$(20 nm) bilayer measured at $f = 9.65$ GHz, which demonstrates that a 20-nm NiO significantly broadens the linewidth while SiO$_x$ has essentially no effect on the
YIG linewidth. This suggests that the AF ordering in NiO plays an important role in the damping of YIG. To confirm this, figure 4.7b gives the frequency dependencies of $\delta H$ for the three samples shown in Fig. 4.7a, all of which exhibit a linear relationship with frequency. From the slopes of least-squares fits to the data in Fig. 4.7b, we obtain the Gilbert damping constant $\alpha = 5.9 \times 10^{-4}$, $5.9 \times 10^{-4}$, and $2.5 \times 10^{-3}$ for YIG-1, YIG-1/SiO$_x$, and YIG-1/NiO, respectively. The 20-nm NiO clearly enhances the damping in YIG while the damping in YIG/SiO$_x$ is almost the same as in bare YIG. This indicates that the AF moments in NiO exchange couple to the YIG magnetization in a way similar to the exchange bias in FM/AF bilayers, which causes additional damping and magnetic relaxation in the FM.

In conclusion, we have observed strong spin transport in AF insulator NiO and significant enhancement of spin pumping signals with insertion of a thin NiO spacer between YIG and Pt. The enhanced spin pumping indicates excellent spin mixing conductance at the YIG/NiO and NiO/Pt interfaces as well as robust magnon-mediated spin transport in NiO. The magnitude of spin currents in NiO decreases exponentially with decay lengths of $\sim 10$ nm within $5 \text{ nm} \leq t_{\text{NiO}} \leq 50$ nm. This result suggests a new path toward high-efficiency spin transport by engineering heterostructures involving antiferromagnets in addition to FM and NM materials.

### 4.3 Spin transport study across insulators with different magnetic correlation strengths

Using the diamagnetic insulator SrTiO$_3$, the spin decay length is only 0.19 nm dominated by exchange coupling for FMR spin pumping, for strong AF insulator NiO, we observe the long range spin transport phenomena with spin decay length around 10 nm, strong and robust spin pumping signal can still be observed even at
100nm NiO spacer thickness. In the following section, we report a systematic study and comparison of spin transport in six series of Pt/Insulator/YIG trilayers, where the insulators include diamagnetic SrTiO$_3$, paramagnetic (PM) Gd$_3$Ga$_5$O$_{12}$, and antiferromagnetic Cr$_2$O$_3$, amorphous YIG, amorphous NiFe$_2$O$_4$, and NiO. We observe surprisingly robust spin transport in the AF insulators, even in AF layers with low ordering temperatures. The spin transfer efficiency and spin current decay lengths appear related to the strength of magnetic correlation in these insulators. This result provides insights into the role of magnetic correlation for spin transport in magnetic insulators and opens a new venue for exploration of spin manipulation in tailored structures comprising metallic and insulating FM, AF and NM materials.

4.3.1 Characterization of insulators with different magnetic correlation strengths

In order to probe spin transport in insulators of various magnetic structures, we select six materials, including: 1) amorphous SrTiO$_3$, a diamagnet, 2) epitaxial Gd$_3$Ga$_5$O$_{12}$, a paramagnet with a large magnetic susceptibility $\chi$, and four antiferromagnets, 3) Cr$_2$O$_3$, 4) amorphous YIG (a-YIG)[91], 5) amorphous NiFe$_2$O$_4$ (a-NFO)[92], and 6) NiO. All insulator layers are deposited by off-axis sputtering. Lattice matched, strain-free Gd$_3$Ga$_5$O$_{12}$ films are epitaxially grown on YIG at high temperature confirmed by the XRD scan; the remaining five insulators are grown at room temperature to avoid straining the epi-YIG films which can significantly alter the magnetic properties in YIG. Electrical transport measurements confirm the highly insulating nature of all these films. Figures 4.8 (a) and (b) show the magnetic hysteresis loop and FMR measurement spectrum which indicate that the a-YIG film
has negligible magnetization and FMR absorption (a-NFO films exhibit similar behavior). The six insulators include a diamagnet, a paramagnet, and four AFs with a wide range of ordering temperatures, allowing us to probe magnetic excitations and spin propagation in insulators both above and below the AF ordering temperatures, hence illuminating the roles of both static and dynamic magnetic correlations.

Figure 4.8: (a) Original room temperature in-plane magnetic hysteresis loops of a 20-nm epitaxial YIG film (blue) and a 20-nm amorphous YIG film (red) grown on GGG, where the paramagnetic background comes from the GGG substrate. (b) FMR derivative absorption spectra of an epitaxial (blue) and an amorphous (red) 20-nm YIG film on GGG taken at $P_{rf} = 200$ mW.

Bulk Cr$_2$O$_3$ and NiO have Neel temperatures $T_N = 318$ and 525 K, respectively. Both YIG and NiFe$_2$O$_4$ are ferrimagnets when in crystalline form; however, amorphous YIG and NiFe$_2$O$_4$ become AFs due to the lack of crystalline ordering required for ferrimagnetism. The temperature $T$ dependence of exchange bias in FM/AF bilayers allows direct measurement of the blocking temperature, $T_b$, of the AFs. To
determine $T_b$ for each AF studied here, we use Ni$_{81}$Fe$_{19}$ (Py) as the FM and measure exchange bias in Py(5 nm)/AF(20 nm) bilayers grown on Si. To determine the antiferromagnetic ordering temperature, figure 4.9 shows the hysteresis loops of four Py/AF bilayers at $T = 5$ K after field cooling from temperature above $T_b$. All four samples exhibit substantial exchange bias: $H_E = 646, 1403, 568$, and $97$ Oe for Py/NiO, Py/$\alpha$-NFO, Py/$\alpha$-YIG, and Py/Cr$_2$O$_3$, respectively. The blocking temperature could be obtained from the temperature dependence measurement of bias field[93]. Figure 4.10 show the temperature dependencies of $H_E$ for the four bilayers, from which we determine $T_b = 20, 45, 70$, and $330$ K for $20$nm thickness Cr$_2$O$_3$, $\alpha$-YIG, $\alpha$-NFO, and NiO, respectively.

Figure 4.9: Magnetic hysteresis loops of four Py(5 nm)/AF(20 nm) bilayers at 5 K after field cooling, all demonstrating clear exchange bias, confirming the AF nature of those insulators.
4.3.2 Spin transport mechanism studied by FMR spin pumping technique

For each of the six insulators, we grow a series of Pt(5 nm)/insulator(t)/epi-YIG(20 nm) trilayers with various insulator thicknesses \( t \) on YIG films cut from the same YIG/GGG wafer to ensure consistency of the YIG quality. Since Pt is the only conductor in the trilayers, the voltage signals detected are exclusively from the ISHE \( (V_{\text{ISHE}}) \), which proportionally reflects the spin currents pumped into Pt across the insulators.

Room-temperature spin pumping measurements are conducted on all trilayers (\( \sim 1 \) mm wide and \( \sim 5 \) mm long) in an FMR cavity at \( f = 9.65 \) GHz and \( P_{rf} = 200 \) mW in an in-plane \( DC \) field \( (H) \). The mV-level ISHE voltages provide a dynamic range of more than three orders of magnitude for detecting the decay of spin current across the insulators. The rates at which \( V_{\text{ISHE}} \) decays with increasing insulator thickness
(t) differ dramatically among the six spacers. A 0.5-nm SrTiO$_3$ already suppresses $V_{\text{ISHE}}$ by a factor of 17 from the corresponding Pt/YIG bilayer. As we change the insulator from SrTiO$_3 \rightarrow$ Gd$_3$Ga$_5$O$_{12} \rightarrow$ Cr$_2$O$_3 \rightarrow$ $\alpha$-YIG $\rightarrow$$\alpha$-NFO $\rightarrow$ NiO as shown in Fig. 4.11, the spin currents exhibit substantially increasing propagation lengths.

Figure 4.11: Representative $V_{\text{ISHE}}$ vs $H-H_{\text{res}}$ spectra of Pt(5 nm)/insulator(t)/YIG(20 nm) trilayers taken at $P_{\text{rf}} = 200$ mW at varying spacer thicknesses for SrTiO$_3$, Gd$_3$Ga$_5$O$_{12}$, Cr$_2$O$_3$, amorphous YIG, amorphous NFO, and NiO.

Figure 4.12 summarizes the t-dependencies of the normalized peak $V_{\text{ISHE}}$ at YIG resonance, $H_{\text{res}}$, for all six series. From the linear relationship in the semi-log plots, we extract the spin decay lengths $\lambda$ in the insulators by fitting to $V_{\text{ISHE}} = V_{\text{ISHE}}(t=0)e^{-t/\lambda}$, which gives $\lambda = 0.18, 0.69, 1.6, 3.9, 6.3,$ and 9.8 nm for SrTiO$_3$, Gd$_3$Ga$_5$O$_{12}$, Cr$_2$O$_3$, $\alpha$-YIG, $\alpha$-NFO, and NiO, respectively. More surprisingly, $V_{\text{ISHE}}$ initially increases by a factor of 2.1 and 1.6 when a 1- or 2-nm NiO and $\lambda$-NFO, respectively, is inserted.
between YIG and Pt (the point for \( t = 0 \) is excluded from the exponential fit for NiO and NFO). This dramatic variation in the spin current propagation length-scale and the enhancement of spin pumping signal most likely arise from different magnetic characteristics of the six insulators.

Figure 4.12: Semi-log plots of \( V_{\text{ISHE}}(H_{\text{res}}) \) as a function of the insulator thickness for the six series normalized to the values for the corresponding Pt/YIG bilayers, where the straight lines are least-squares fits to each series, from which the spin decay lengths \( \lambda \) are determined.
For dynamically generated spin current to transmit across insulating spacers beyond the tunneling range (∼1 nm), magnetic excitations in the insulators should play a major role. Except SrTiO$_3$, all other five insulators have strong magnetic moments, including PM Gd$_3$Ga$_5$O$_{12}$ and four AFs with ordering temperatures either above or below room temperature. For the same AF material, the blocking temperature, $T_b$, below which the AF ordering is stable, can vary significantly depending on the film thickness. Among the four AFs, NiO is the most robust AF with a bulk $T_N = 525$ K, while for very thin NiO layers ($\leq 5$ nm), $T_b$ is well below 300 K. For α-NFO, α-YIG and Cr$_2$O$_3$, the AF ordering temperatures are below room temperature at all thicknesses. It is interesting to note that Gd$_3$Ga$_5$O$_{12}$ also exhibits magnetic order at very low temperatures. Thus, magnetic correlation of AF moments in thermal fluctuation is critically important for the observed robust spin transport.

At resonance, the precessing YIG magnetization generates magnetic excitations in the adjacent insulator (either with AF ordering or fluctuation) via interfacial exchange coupling, which in turn exerts an extra damping on YIG. We measure the Gilbert damping constant $\alpha$ from the frequency dependencies of FMR linewidth $\Delta H$ for six Insulator(20 nm)/YIG(20 nm) bilayers and a single epi-YIG film using a microstrip transmission line, as shown in Fig. 4.13. All the data show linear dependence following:

$$\Delta H = \Delta H_0 + 4\pi \alpha \frac{f}{(\sqrt{3}\gamma)} \quad (4.1)$$

where $\Delta H$ is the zero-frequency linewidth (y-intercept) and $\gamma$ is the gyromagnetic ratio. From the slopes of least-squares fits, we obtain $\alpha = (8.1 \pm 0.6) \times 10^{-4}$, $(8.6 \pm 1.0) \times 10^{-4}$, $(11 \pm 1) \times 10^{-4}$, $(12 \pm 1) \times 10^{-4}$, $(14 \pm 1) \times 10^{-4}$, $(17 \pm 2) \times 10^{-4}$, and $(26 \pm 3) \times 10^{-4}$.
$10^{-4}$ for the bare YIG, SrTiO$_3$/YIG, Gd$_3$Ga$_5$O$_{12}$/YIG, Cr$_2$O$_3$/ YIG, $\alpha$-YIG/ YIG, $\alpha$-NFO/ YIG, and NiO/ YIG.

Figure 4.13: Frequency dependencies of FMR linewidths of a bare epitaxial YIG film, SrTiO$_3$(20 nm)/YIG, Gd$_3$Ga$_5$O$_{12}$(20 nm)/YIG, Cr$_2$O$_3$(20 nm)/YIG, $\alpha$-YIG/(20 nm)/YIG, $\alpha$-NFO(20 nm)/YIG, and NiO(20 nm)/YIG bilayers.

The diamagnetic SrTiO$_3$ with essentially no magnetic moment does not enhance the damping of YIG within experimental uncertainty while its decay length is of only atomic distance ($\lambda = 0.19$ nm), which can be explained by quantum tunneling. Gd$_3$Ga$_5$O$_{12}$ has one of the highest PM susceptibility with a measured value of $\chi =$
6.45×10^{-2} \text{ emu/(Oe·mol)}, which gives a net magnetic moment of 0.030 \( \mu \text{B} \) per formula unit (f.u.) at in-plane YIG resonance field of 2630 Oe. The magnetic moment in \( \text{Gd}_3\text{Ga}_5\text{O}_{12} \) likely absorbs angular momentum via exchange coupling to YIG and conducts spin current through (weak) magnetic correlation, resulting in a longer \( \lambda = 0.69 \text{ nm} \) compared to \( \lambda = 0.19 \text{ nm} \) for \( \text{SrTiO}_3 \). The four AF insulators show much longer spin decay lengths together with larger damping enhancement on the underlying YIG due to stronger magnetic correlations. NiO is the most robust AF of the four and more than triples the damping of YIG while the spin decay length in NiO is almost 10 nm and clear spin current is detected over a NiO thickness of 100 nm as discussed in previous section.

The strong easy-plane anisotropy in NiO results in AF resonance frequency near 1 THz which is very different from the 9.65 GHz used in our FMR excitation of YIG. However, the dispersion relation for very thin NiO films is likely to be different from that for bulk NiO due to exchange coupling to YIG and possible existence of surface/interface anisotropy. Considering that strong AF spin correlation has been observed well above \( T_N \) for NiO, we believe the excitation responsible for spin transport in AFs must be magnonic in ordered AFs or AF fluctuations in insulators with low blocking temperatures. In either case, the strongly correlated AF spins are excited via exchange coupling to the precessing YIG magnetization at the AF/YIG interface and transfer the spin current across the spacer to the interface with Pt, where it is converted to spin-polarized current in Pt. This is analogous to the magnon accumulation at magnetic-insulator/metal interfaces and magnon current inside the magnetic insulators. It is well known that exchange coupling in an AF/FM bilayer can lead to two possible effects in the hysteresis loop: exchange bias at temperatures below
$T_b$ of the AF and the increase in coercivity which exists even at temperatures well above $T_b$. To verify the interfacial exchange coupling, we show in Fig. 4.14 the in-plane hysteresis loops of a single epi-YIG film and $\alpha$-YIG/YIG, $\alpha$-NFO/YIG, and NiO/YIG bilayers, which exhibit $H_c = 0.40, 0.79, 1.27, \text{ and } 2.06 \text{ Oe, respectively.}$ The variation of $H_c$ agrees well with the pattern observed in spin decay length and damping enhancement. The SrTiO$_3$/YIG, Gd$_3$Ga$_5$O$_{12}$/YIG and Cr$_2$O$_3$/YIG show negligible enhancement in $H_c$ within experimental uncertainty. No clear exchange bias is detected in the six bilayers at room temperature. However, we note that no exchange bias has been reported in the literature for YIG-based structures.

![Figure 4.14](image-url)  

Figure 4.14: Room temperature in-plane magnetic hysteresis loops of a single epitaxial YIG film, an $\alpha$-YIG(20 nm)/YIG, an $\alpha$-NFO(20 nm)/YIG, and a NiO(20 nm)/YIG bilayer, with a coercivity of 0.40, 0.79, 1.27, and 2.06 Oe, respectively.
From the three independent experimental measurements in spin decay length $\lambda$, magnetic damping enhancement $\Delta \alpha$ and coercivity $H_c$, we observe that following the sequence from SrTiO$_3$→Gd$_3$Ga$_5$O$_{12}$→Cr$_2$O$_3$→α-YIG→α-NFO→NiO, $\lambda$, $\Delta \alpha$ and $H_c$ all increase monotonically. Figures 4.15 shows the correlation between the spin decay length and the damping enhancement which gives a almost linear relationship except the SrTiO$_3$ point. This implies that a common mechanism is responsible for this systematic behavior. We hypothesize that strong magnetic correlation between ordered or fluctuating AF spins play a dominant role in the observed spin transport in insulators. The strength of magnetic correlation depends on the ordering temperature which in turn is determined by the material and film thickness. From our results, it appears that the correlation strength increases in the order SrTiO$_3$ (diamagnet)→Gd$_3$Ga$_5$O$_{12}$ (PM)→Cr$_2$O$_3$ (AF, $H_N$ 20 K)→α-YIG (AF, $H_N$ ～ 45 K)→α-NFO (AF, $H_N$ ～ 70 K)→NiO (AF, $H_N$ = 330 K). As the magnetic correlation increases, exchange interaction becomes stronger, which, 1) strengthens the exchange coupling at the insulator/YIG interfaces, 2) facilitates the propagation of spin current carried by magnetic excitations in the insulators, and 3) enhances the magnetic damping in the underlying YIG films. The surprising enhancement of spin pumping signals for the trilayers with 1- or 2-nm NiO and α-NFO indicates that the NiO/YIG and α-NFO/YIG interfaces with strong exchange coupling are more efficient in spin transfer than Pt/YIG with direct contact.

While the spin decay length and damping enhancement show an excellent linear relationship for the five magnetic insulators, neither the spin decay length nor damping enhancement depends linearly on the blocking temperature. As reported previous work, strong magnetic correlation was observed in NiO even at 850 K[94], well above
Figure 4.15: Excellent linear correlation between spin decay length $\lambda$ and Gilbert damping enhancement $\Delta \alpha = \alpha_{\text{Insulator/YIG}} - \alpha_{\text{YIG}}$ for the six insulators. The line is a least-squares linear fit to all data points excluding SrTiO$_3$.

the ordering temperature, and the AF to PM phase transition is continuous. The nature of the dependence of spin decay length and damping enhancement on the AF ordering temperature is not fully understood. More experiments on other important parameters of the heterostructures and theoretical investigations are needed to provide insights into these intriguing phenomena.

In summary, we observe clear spin current transfer in a series of PM and AF insulators mediated by magnetic correlations due to AF ordering or AF fluctuations. This result brings a large family of insulators, in particular, AF insulators, into the exploration of spintronic applications utilizing pure spin current.
Chapter 5: Spin transport across metallic Cu spacer probed in YIG/Cu/Pt trilayer structures

Generation of pure spin currents from ferromagnets (FM) to normal metals (NM) has been widely studied by thermal \cite{7, 9, 8} and ferromagnetic resonance (FMR) spin pumping\cite{95, 96}, where the NM is typically a spin-sink metal such as Pt, W and Ta\cite{15, 16}, while lighter metals such as Cu is rarely used. The efficiency of spin pumping is largely determined by the spin mixing conductance $g_{\uparrow\downarrow}$ at the FM/NM interface\cite{97, 98}. Here, we report a comparative study of spin pumping in YIG/Cu/Pt and YIG/Cu/W trilayers with varying Cu thicknesses. We observe a very large intrinsic spin mixing conductance of the YIG/Cu interface and obtain the spin mixing conductances of the Cu/Pt and Cu/W interfaces\cite{99}. While the generated spin current in YIG/Cu/Pt series decreases with the insertion of a Cu spacer, the spin current in YIG/Cu/W is enhanced by more than 4 times as compared to YIG/W due to improved spin impedance matching\cite{100}. This result points towards a route to design heterostructures of FMs and NMs with optimal spin mixing conductances and high spin pumping efficiency.
5.1 Spin diffusion in metallic systems

As indicated in previous chapter, the spin current injected from YIG into metal layer along the normal direction marked as $Z$ direction is shown in Fig. 5.1 could be written as[101]:

$$J_S(Z) = \frac{\sinh[(t_N - Z)/\lambda_{SD}]}{\sinh(t_N/\lambda_{SD})} J_S(0)$$  \hspace{1cm} (5.1)

Figure 5.1: Schematic for the spin diffusion in the transition metal in YIG/NM bilayer structures.

where $t_N$, $\lambda_{SD}$ are the thickness and spin diffusion length of the metal layers, respectively. $J_s(0)$ is the spin current at the YIG/NM interface ($Z = 0$), which can be
expressed as [101]:

\[ J_S(Z) = \frac{g_{t\uparrow} \gamma^2 h_{rf}^2}{8\pi\alpha^2 [(\gamma 4\pi M_s)^2 + 4\omega^2]} \]  

(5.2)

where \( \gamma \) is the gyromagnetic ratio, \( g_{t\uparrow} \) is the spin mixing conductance, \( \alpha \) is the Gilbert damping constant, \( h_{rf} \) is the radio frequency (rf) field, \( \omega = 2\pi f \) is the FMR frequency.

The injected spin current is converted to charge current \( j_c \) due to inverse spin Hall effect (ISHE) in the normal metal layer. The average charge current could be obtained by [101]:

\[ J_c = \theta_{SH} \left( \frac{1}{t_N} \right) \left( \frac{2e}{\hbar} \right) \int_0^{t_N} J_S(z) \, dz \]  

(5.3)

which gives

\[ J_c = \theta_{SH} \frac{\lambda_{SD}}{t_N} \frac{2e}{\hbar} \tanh \left( \frac{t_N}{2\lambda_{SD}} \right) J_S(0) \]  

(5.4)

where \( \theta_{SH} \) is the spin Hall angle of the normal metal. From the charge current, we can find the electromotive force due to ISHE effect. In the following section of this chapter, we will study and discuss the variation of Cu spacer thickness of both the magnitude of ISHE voltage [16] and spin current in YIG/Cu/NM trilayer structures. The analysis of the spin pumping results combines two independent and complementary experimental methods: (1) determination of spin-pumping enhanced Gilbert damping from the frequency dependence of the FMR linewidth (as opposed to the more common measurement of linewidth broadening at a single frequency), from which we obtain the (effective) spin mixing conductance; and (2) determination of spin currents using the measured (large) ISHE voltages. These two methods are powerful techniques for studying spin pumping, but have rarely been used together, especially for YIG systems. The combination of the two complementary methods eliminates
some assumptions used in analysis of spin pumping when only one technique is applied thus improving confidence in the analysis of the result.

5.2 FMR spin pumping study of YIG/Cu/Pt and YIG/Cu/W trilayer structures

FMR spin pumping measurement is performed at room temperature and radio-frequency ($rf$) $f = 9.65$ GHz on YIG (20 nm)/Cu(t)/Pt (5 nm) and YIG (20 nm)/Cu(t)/W (5 nm) trilayers with varying Cu thickness $t$ from 0 to 20 nm. All metal layers are deposited by off-axis ultrahigh vacuum sputtering to minimize damage to the YIG/NM interfaces. Samples of $\sim$1 mm wide and $\sim$5 mm long are placed in the center of an FMR cavity and in a DC magnetic field ($H$) applied in the $xz$-plane, as the same schematic experimental set-up used previously. At resonance, the precessing YIG magnetization transfers angular momentum to the conduction electrons in Cu. Since the Cu thickness is much shorter than the spin diffusion length ($\lambda_{SD}$) [102, 103], the spin accumulation in Cu spacer will drive a spin current $J_s$ with a polarization ($\sigma$) parallel to YIG magnetization into the Pt or W layer. Once inside Pt or W, $J_s$ is converted into a net charge current, $J_c \sim J_s \times \sigma$, via inverse spin Hall effect, resulting in a voltage signal ($V_{ISHE}$) along the $y$-axis. Figure 5.2 (a) and (b) show the $V_{ISHE}$ vs. $H$ spectra for YIG/Pt and YIG/W bilayers at $\theta_H = 90^\circ$ and $270^\circ$ (both in-plane fields) at $rf$ input power $P_{rf} = 200$ mW, which gives $V_{ISHE} = 552\mu$V and -2.04 mV, respectively. The negative sign for YIG/W reflects the opposite spin Hall angles of W and Pt which has been reported previously in chapter 2. As $H$ is reversed from $\theta_H = 90^\circ$ to $270^\circ$, $V_{ISHE}$ changes sign and maintains the same magnitude as expected since $\sigma$ changes sign with reversal of $M$. 

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Figure 5.2: ISHE voltage measurements and $V_{\text{ISHE}}$ vs. $H-H_{\text{res}}$ spectra of (a) YIG/Pt bilayer, (b) YIG/W bilayer. The thicknesses of all YIG, Pt and W layers are 20, 5 and 5 nm, respectively.

In order to study the effect of Cu spacer in FMR driven spin pumping, figure 5.3 (a) and (b) show the $V_{\text{ISHE}}$ vs. $H$ spectra for YIG/Cu (20 nm)/Pt and YIG/Cu (20 nm)/W trilayers. The peak values decrease to $V_{\text{ISHE}} = 1.21 \mu V$ and $-16.3 \mu V$ as compared to YIG/Pt and YIG/W with direct contact, respectively. However, the decrease of spin pumping signal does not necessarily indicate the decrease of spin current due to the shunting effect of Cu spacer with the much smaller electrical resistance.

The magnitude of spin current is another important quantity to describe the spin pumping efficiency. To compare the effect of Cu spacer on spin current $J_s$ generated in the two trilayer systems, we show in Fig 5.3 (a) and (b) the Cu thickness dependence of $J_s$ normalized by $J_s(0)$ of the YIG/Pt and YIG/W bilayers. Spin current $J_s$ can be calculated from:

$$J_s = \frac{V_{\text{ISHE}}}{\theta_{SH} \lambda_{SD} \tanh\left(\frac{t_N}{2\lambda_{SD}}\right) \omega R}$$  \hspace{1cm} (5.5)
where $t_N$ and $\theta_{SH}$ are the thickness and spin Hall angle of Pt or W, $R$ and $w$ the total resistance and width of the trilayers, respectively. Both Figs. 5.4 (a) and (b) show that with the insertion of Cu, the spin current initially decreases dramatically when the Cu thickness $t_{Cu} \leq 5 \text{ nm}$ and reaches a plateau at $t_{Cu} \leq 10 \text{ nm}$. However, YIG/Cu/Pt and YIG/Cu/W structures show opposite plateau values of the normalized spin current: for YIG/Cu/Pt, $J_s$ is only about 20-25% of $J_s(0)$, while for YIG/Cu/W, $J_s$ is 4 to 4.5 times of $J_s(0)$. The initial decrease of $J_s$ at $t_{Cu} \leq 5 \text{ nm}$ may be related to the much higher resistivity of thin Cu layers due to finite size effect which could induce significant spin flipping when the Cu spacer is below critical thickness. At $t_{Cu} \geq 10 \text{ nm}$, the resistivity of Cu layers is greatly lowered, the accumulated spins in Cu are partially reflected back into YIG and partially transmitted into Pt or W to produce ISHE signal. The ratio between these two fractions depends on the values of interfacial spin mixing conductance $g_{Cu/Pt}^{\uparrow\downarrow}$, $g_{Cu/W}^{\uparrow\downarrow}$, and $g_{YIG/Cu}^{\uparrow\downarrow}$.
Figure 5.4: Spin current $J_s$ in (a) YIG/Cu/Pt and (b) YIG/Cu/W trilayers normalized to $J_s(0)$ in YIG/Pt and YIG/W bilayers, respectively, as a function of the Cu thickness.

To uncover the mechanism behind the different spin pumping behavior of the two trilayers systems, we need to determine the spin mixing conductances $g_{\uparrow \downarrow}$, which can be obtained from the enhancement of Gilbert damping due to spin pumping. We first measure the frequency dependence of the FMR linewidth $\Delta H$ by placing samples on a microstrip transmission line. In all cases the linewidth increases linearly with frequency from 10 GHz to 20 GHz. Gilbert damping constant $a$ can be obtained using:

$$\Delta H = \Delta H_{inh} + \frac{4\pi a f}{\sqrt{3} \gamma}$$  \hspace{1cm} (5.6)$$

where $\Delta H_{inh}$ is the inhomogeneous broadening and $\gamma$ is the gyromagnetic ratio. The enhanced Gilbert damping due to spin pumping is defined as\\(104, 95\\):

$$\alpha_{SP} = \alpha_{YIG/Cu/NM} - \alpha_{YIG/Cu/NM}$$  \hspace{1cm} (5.7)$$
Figure 5.5 shows that the $\alpha_{SP} = (2.1 \pm 0.1) \times 10^{-3}$ for YIG/Pt is about twice as large as the value of $(1.1 \pm 0.1) \times 10^{-3}$ for YIG/Cu(20 nm)/Pt, which agrees with the previous damping study on Py/Cu/Pt and YIG/Cu/Pt multilayers. However, the order is reversed for YIG/Cu/W trilayers: the enhancement of Gilbert damping for YIG/Cu(20 nm)/W, $\alpha_{SP} = (2.1 \pm 0.2) \times 10^{-3}$, is more than 3 times larger than the value of $(6.3 \pm 0.6) \times 10^{-4}$ for YIG/W.

Figure 5.5: Frequency dependencies of FMR linewidths of (a) YIG/Cu/Pt, (b) YIG/Cu/W.

Room-temperature cavity FMR measurements also confirm this trend as shown in Fig 5.6 (a) and (b). The linewidth change for YIG/Cu(20 nm)/Pt compared to the bare YIG is 6.0 Oe which is smaller than the value of 12.6 Oe for YIG/Pt. However, for YIG/Cu(20 nm)/W trilayer, the linewidth change is 13.1 Oe which is larger than the value of 6.4 Oe for YIG/W. This implies a significant difference at the interfacial spin transport in YIG/Cu/Pt and YIG/Cu/W compared to YIG/Pt and YIG/W. In
order to understand this behavior, we need to determine the interfacial spin mixing conductance of $g_{YIG/Cu}^{↑↓}, g_{Cu/Pt}^{↑↓}$, and $g_{Cu/W}^{↑↓}$ as well as $g_{YIG/Pt}^{↑↓}$ and $g_{YIG/W}^{↑↓}$.

Figure 5.6: FMR derivative absorption spectra of YIG/Cu/Pt, YIG/Cu/W series at $f = 9.65$ GHz.

5.3 The mechanism for the enhancement of spin current

Tserkovnyak et al.[52] provides a theory for quantitative analysis of interfacial spin mixing conductance. This theory is developed for metallic FM/NM bilayers or
trilayers where the real part of $g_{↑↓}$ is dominant. Since the imaginary part of $g_{↑↓}$ of YIG/NM interface has been recently reported to be negligibly small, this theory is also applicable for YIG/NM systems. The spin pumping induced Gilbert damping enhancement can be expressed as [52]:

$$\alpha_{SP} = \frac{g_\mu B}{4\pi M_S t_F} g_{e\text{ff}}^{↑↓}$$

where $g_{e\text{ff}}^{↑↓}$ is the effective interfacial mixing conductance which includes the spin current back flow driven by spin accumulation, $g$, $\mu_B$, $M_S$ and $t_F$ are the $g$ factor, Bohr magnetron, saturation magnetization of YIG, and the thickness of the YIG layer, respectively. For ideal spin sink materials, such as Pt and W, the pumped spin current relax almost immediately near the FM/NM interface by fast spin-flip process which prevents spin accumulation build-up and backflow of spin current, thus $g_{e\text{ff}}^{↑↓} \approx g_{↑↓}^{↑}$. In spin diffuse systems, such as FM/Cu bilayer structure:

$$g_{e\text{ff}}^{↑↓} = g_{FM/NM}^{↑↓}[1 + g_{FM/NM} \frac{1}{4\sqrt{\epsilon/3} \tanh(t_N/\lambda)g_{N}^{Sh}}]^{-1}$$

where $g_{FM/NM}^{↑↓}$ is intrinsic spin mixing conductance of the FM/NM interface normalized by Sharvin conductance $g_{N}^{Sh}$ and $\epsilon$ is the spin flip probability [52, 105, 106]. Jia et al. computed the spin mixing conductance for YIG/Ag interface and gives $g_{YIG/Ag}^{↑↓} \approx g_{Ad}^{Sh}$. It is reasonable to assume $g_{YIG/Cu}^{↑↓} \approx g_{Cu}^{Sh}$ which reduces Eq.(4) to the following expression,

$$g_{e\text{ff}}^{↑↓} = g_{YIG/Cu}^{↑↓}[1 + \frac{1}{4\sqrt{\epsilon/3} \tanh(t_N/\lambda)}]^{-1}$$

For lighter normal metals, such as Cu, the spin backflow current into the FM can be significant due to spin accumulation and consequently, $g_{e\text{ff}}^{↑↓}$ is much smaller than $g_{YIG/Cu}^{↑↓}$ depending on $t_N$ and $\epsilon$. For YIG/Cu/NM trilayer structures where the NM
is taken to be a perfect spin sink, in the limit of vanishing spin flip in Cu spacer layer, the total effective spin mixing conductance $g_{\text{eff}}^{\uparrow\downarrow}$ is simply given by the sum of two interfacial contributions and spin resistance in Cu:

$$\frac{1}{g_{\text{eff}}^{\uparrow\downarrow}} = \frac{1}{g_{\text{YIG/Cu}}^{\uparrow\downarrow}} + R_{\text{Cu}}^{\uparrow\downarrow} + \frac{1}{g_{\text{Cu/NM}}^{\uparrow\downarrow}}$$  \hspace{1cm} (5.11)

where $R_{\text{Cu}}^{\uparrow\downarrow} = \frac{2e^2}{h\sigma}$ ($\sigma$ is the electrical conductivity) is the spin resistance of the Cu spacer. In order to quantitatively determine the intrinsic $g_{\text{YIG/Cu}}^{\uparrow\downarrow}$, we grow a 2-µm (\(\gg \mu_{SD}\)) Cu layer on YIG to minimize the backflow of spin current. Figure 5.7 (a) shows the Gilbert damping enhancement of two YIG/Cu bilayers compared to a bare YIG film. The values of $\alpha_{sp}$ for YIG/Cu(10 nm) and YIG/Cu(2µm) are \((1.1\pm0.1)\times10^{-4}\) and \((1.8\pm0.1)\times10^{-3}\) (16 times difference), respectively, which clearly indicates significant backflow of spin current driven by spin accumulation when $t_{\text{Cu}}\ll\lambda_{SD}$. This is also confirmed by cavity FMR measurement as shown in figure 5.7 (b). The in-plane linewidth enhancement for YIG/Cu(2µm) is 8.7 Oe, much larger than the value of 1.2 Oe for YIG/Cu(10 nm). From Eqs. (9) and (10) and using the parameters $\epsilon = 1/700$ and $\lambda_{SD} = 250$ nm [52], we calculate the intrinsic YIG/Cu interfacial spin mixing conductance $g_{\text{YIG/Cu}}^{\uparrow\downarrow} = (4.3\pm0.4)\times10^{19}\text{m}^{-2}$ and $(6.1\pm0.6)\times10^{19}\text{m}^{-2}$ for the YIG/Cu(2µm) and YIG/Cu(10 nm) bilayers, respectively. The slight difference between the two may come from the spin-dependent scattering of the top copper oxide layer which brings some additional contributions to the Gilbert damping especially when the Cu layer is $\ll \lambda_{SD}$[52].

However, this effect is negligibly for the YIG/Cu(2µm) sample since the Cu thickness is $\gg \lambda_{SD}$. Here, we take the value of $g_{\text{YIG/Cu}}^{\uparrow\downarrow} = (4.3\pm0.4)\times10^{19} \text{m}^{-2}$ for the following calculation.
We also did damping measurement on a YIG/Cu/Py trilayer structure\[23\] as another independent way to determine the spin mixing conductance of the YIG/Cu interface. The figure 5.8 shows the frequency dependencies of the FMR linewidth of a YIG(20 nm)/Cu(20 nm)/Py(3 nm) trilayer and a YIG(20 nm) single layer. Gilbert damping is clearly enhanced in the YIG/Cu/Py trilayer $\alpha_{YIG/Cu/Py} = 4.16 \times 10^{-3}$ as compared to the bare YIG film $\alpha_{YIG} = 8.7 \times 10^{-4}$, from which we obtain the total effective spin mixing conductance of the YIG/Cu/Py trilayer using $g_{\text{eff, trilayer}}^{\uparrow \downarrow} = 6.34 \times 10^{18} \text{ m}^{-2}$. If we assume that both of the 20-nm Cu layer and the Cu/Py interface are 100% spin transparent (i.e., infinite $g^{\uparrow \downarrow}$), this value ($6.34 \times 10^{18} \text{ m}^{-2}$) is the intrinsic spin mixing conductance of the YIG/Cu interface. However, as given by Tserkovnyak et al. in RMP 77, 1375 (2005) [page 1398, Eq. (81)], the YIG/Cu and Cu/Py interfaces as well as the Cu layer must all be included in calculating spin conduction, especially when the spin mixing conductances are large, which is the case here. Using Eq. 5.11, we can estimate the intrinsic spin mixing conductance of the YIG/Cu
interface, \( g_{YIG/Cu}^{\uparrow\downarrow} \). The spin resistance of the Cu layer, \( R_{Cu} = (2e^2t_{Cu})/\hbar\sigma = 6.2 \times 10^{-20} \) m\(^2\), which corresponds to a spin conductance \( g_{Cu} = 1.6 \times 10^{19} \) m\(^{-2}\) for the 20-nm Cu. Thus, the calculation of \( g_{YIG/Cu}^{\uparrow\downarrow} \) is sensitive to the value of \( g_{Cu/Py}^{\uparrow\downarrow} \). We first take \( g_{Cu/Py}^{\uparrow\downarrow} \approx 2g_{Cu}^{Sh} = 3.0 \times 10^{19} \) m\(^{-2}\) as suggested, which results in \( g_{YIG/Cu}^{\uparrow\downarrow} = (1.6 \pm 0.2) \times 10^{19} \) m\(^{-2}\). In practice, the reported experimental values of spin mixing conductance at Py/NM interfaces are slightly smaller: \( \approx 2.0 \times 10^{19} \) m\(^{-2}\). If we use this value, we obtain \( g_{YIG/Cu}^{\uparrow\downarrow} = (2.2 \pm 0.2) \times 10^{19} \) m\(^{-2}\). These two values of \( g_{YIG/Cu}^{\uparrow\downarrow} \) are comparable to our calculation of \( g_{YIG/Cu}^{\uparrow\downarrow} = 1.8 \times 10^{19} \) m\(^{-2}\) obtained from the first method for the YIG/Cu(2\( \mu \)m) bilayer. In particular, these two independent measurements of \( g_{YIG/Cu}^{\uparrow\downarrow} \) in our YIG/Cu(2\( \mu \)m) bilayer and YIG/Cu/Py trilayer indicate that the spin mixing conductance at the YIG/Cu interface is higher than the predicted values of \( 7 \times 10^{18} \) to \( 1 \times 10^{19} \) m\(^{-2}\) for YIG/Ag\[105\] given in Europhys. Lett. 96, 17005 (2011).

Using the \( g_{eff}^{\uparrow\downarrow} \) calculated from the damping enhancement of YIG/Cu(20 nm)/Pt and YIG/Cu(20 nm)/W trilayers by Eq. (9), we obtain the spin mixing conductance \( g_{Cu/Pt}^{\uparrow\downarrow} = (2.6 \pm 0.4) \times 10^{18} \) m\(^{-2}\) which is smaller than \( g_{YIG/Pt}^{\uparrow\downarrow} = (3.9 \pm 0.3) \times 10^{18} \) m\(^{-2}\) and \( g_{Cu/W}^{\uparrow\downarrow} = (6.1 \pm 0.7) \times 10^{18} \) m\(^{-2}\) which is much larger than \( g_{YIG/W}^{\uparrow\downarrow} = (1.2 \pm 0.1) \times 10^{18} \) m\(^{-2}\).

Using the interfacial spin mixing conductances calculated above for each interface in the YIG/Cu/Pt and YIG/Cu/W trilayers, we can understand the opposite behavior of spin currents shown in Figs. 5.3 (a) and (b) for the two systems. To further understand the underlying mechanism, figure 5.9 schematically shows the series circuits of spin current flow from YIG→Pt (or W) and from YIG→Cu→Pt (or W). Here, we use spin mixing resistance \( R_{Cu}^{\uparrow\downarrow} = 1/g_{Cu}^{\uparrow\downarrow} \) for each interface due to the convenience for series circuits. For the YIG/Cu(20 nm)/Pt trilayer, the total spin
resistance \[ R_{(YIG/Cu/Pt)}^{↑↓} = R_{(YIG/Cu)}^{↑↓} + R_{(Cu/Pt)}^{↑↓} = 2.3 \times 10^{-20} + 6.2 \times 10^{-20} + 3.9 \times 10^{-19} \text{ m}^2 = 4.8 \times 10^{-19} \text{ m}^2, \]
which is larger than the \[ R_{YIG/Pt}^{↑↓} = 2.6 \times 10^{-19} \text{ m}^2 \] for YIG/Pt.

As a result, the spin current at \( t_{Cu} \gg 10 \text{ nm} \) is smaller (20-25\%) than \( J_s(0) \) for YIG/Pt. One notes that the Cu/Pt interface is the dominating barrier for spin current flow from YIG to Pt across Cu, much larger than the YIG/Cu interface and the Cu spacer. In the case of YIG/Cu(20 nm)/W, the total spin resistance \[ R_{YIG/Cu/W}^{↑↓} = R_{YIG/Cu}^{↑↓} + R_{Cu}^{↑↓} + R_{Cu/W}^{↑↓} = 2.3 \times 10^{-20} + 6.2 \times 10^{-20} + 1.6 \times 10^{-19} \text{ m}^2 = 2.5 \times 10^{-19} \text{ m}^2, \] which is smaller than the \[ R_{YIG/W}^{↑↓} = 8.3 \times 10^{-19} \text{ m}^2 \] for YIG/W. This is why the spin current plateau for YIG/Cu/W is ~4.5 times larger than \( J_s(0) \) for YIG/W as shown in Fig. 5.4b.
Figure 5.9: Schematic comparison of spin mixing resistance ($R = 1/g$) of (a) YIG/Pt (red), YIG/Cu (yellow) and Cu/Pt (green) interfaces, and (b) YIG/W (blue), YIG/Cu (yellow) and Cu/W (purple) interfaces. The calculated values of spin mixing conductances explain the opposite behavior of Cu spacer thickness dependence of spin currents: the insertion of a Cu layer suppresses the magnitude of spin current from YIG to Pt, but enhances the magnitude of spin current generation from YIG to W.

Currently, proximity effect at YIG/Pt interface[107, 108] is a hot topic in the spin pumping field, however, we believe proximity is unlikely to affect the observed enhancement of spin current in our YIG/Cu/W trilayer structures. The AFM measurements of our YIG film show with rms roughness below 0.2nm over a large scan area[100], showing the smooth surfaces of our films. We expect that any proximity effect should be eliminated by much thinner Cu like 1 to 2 nm. In figure 5.4 (a), the
initial decrease of $J_S$ happens until $t_{Cu} = 5$ nm, suggesting that proximity effect is not the reason for the observed behavior.

![Graph showing the normalized spin current $J_S(t_{Cu})/J_S(0)$ as a function of $t_{Cu}$ for YIG/Cu(Py)(3 nm)](image)

Figure 5.10: Spin current $J_S$ in YIG/Cu/Py trilayers normalized to $J_S(0)$ in YIG/Pt and YIG/W bilayers, respectively, as a function of the Cu thickness.

Besides the YIG/Cu/NM trilayer structures, we also probe the variation of the magnitude of spin current in YIG/Cu/Py heterostructures. Using the obtained spin Hall angle for Py in chapter 2, we can estimate the spin currents pumped into Py from YIG across a Cu spacer of various thicknesses $t_{Cu}$ with $J_0 = 1.4 \times 10^6$ A/m$^2$ for the YIG/Pt(3 nm) bilayer. Figure 5.10 shows the $t_{Cu}$ dependence of $J_{S(t_{Cu})}$ normalized by $J(0)$, where the initial decrease of spin current is very similar to what we have observed in YIG/Cu/Pt and YIG/Cu/W trilayer structures which may be related to the high resistivity of the Cu spacer at $t_{Cu} \leq 10$ nm. At $t_{Cu} \geq 10$ nm, the spin current reaches a plateau; interestingly, $J_s$ is more than 4 times larger than $J(0)$ for YIG/Py with direct contact (similar to the YIG/Cu/W system), indicating a larger effective...
spin mixing conductance in YIG/Cu/Py trilayers than that in the YIG/Py bilayer. This results also points to the potential optimization of spin transfer efficiency in FM metallic Py based heterostructures.

In summary, we have systematically studied FMR spin pumping in YIG/Cu/Pt and YIG/Cu/W trilayers as well as YIG/Cu bilayers. From the spin pumping enhancement of Gilbert damping, we quantitatively determined the interfacial spin (mixing) conductances at YIG/Pt, YIG/W, YIG/Cu, Cu/Pt and Cu/W interfaces. The values of these spin mixing conductances explain the suppression of spin currents pumped into Pt in YIG/Cu/Pt trilayers and the enhancement of spin currents in YIG/Cu/W trilayers. This discovery potentially paves a path toward significant improvement of spin pumping efficiency by engineering multilayer with optimized spin impedance matching of the interfaces, a powerful capability for future spin-functional devices.
Chapter 6: Conclusion

We have demonstrated that off-axis sputter technique could be used as a unique technique to achieve single crystalline YIG thin film growth [25]. The as-deposited 20-nm films show high quality crystalline structure demonstrated by the clear Laue oscillations in the XRD scan. The FMR spectrum gives the peak to peak linewidth as narrow as 7.4Oe with Gilbert damping about $9.1 \times 10^{-4}$ [22]. The YIG thin film with insulating nature provides a previously unattainable platform to study the spin transport and dynamics in heterostructures. We observed mV-level large spin pumping in YIG/Pt bilayers [25] structures which is order of magnitudes larger than the spin pumping signal reported previously using the similar cavity FMR technique. This ISHE signal partially comes from the optimal surface condition with roughness below 0.2nm indicated by both AFM and XRR characterization.

Based on these excellent YIG thin films, we probe the spin Hall physics, especially the spin orbit coupling strength represented by spin Hall angles values in a series of 3$d$, 4$d$, and 5$d$ transition metals [22, 63]. Our results confirm the central role of atomic number ($Z$) played in the spin Hall physics in non-magnetic transition metals. Furthermore, the systematic variation of spin Hall angle values both in sign and magnitude demonstrate the importance of $d$-orbital filling as predicted by Tanaka.
et al.,[61]. The large spin Hall angle values observed in 3d Ni and Cr [22, 63] are quite promising for future spintronics based functional devices.

In YIG/spacer/Pt trilayer structures, the observed exponential decay of spin pumping signal over the insulating non-magnetic SrTiO$_3$, Sr$_2$GaTaO$_6$, Sr$_2$CrNbO$_6$ oxide barriers with decay length $\approx 0.2$nm clearly demonstrates that the FMR-driven spin pumping is dominated by the short range exchange coupling. Further investigation show that the spin decay length over non-magnetic oxide barriers is determined by the barrier height at Pt/barrier interface. To understand the role of magnetic ordering in spin transport in insulators, we expand the insulators to paramagnetic Gd$_3$Ga$_5$O$_{12}$, antiferromagnetic Cr$_2$O$_3$, amorphous YIG, NiFe$_2$O$_4$, and NiO. The spin decay lengths $\lambda$ obtained for those insulators highly depend on the magnetic correlation strength of the insulating spacers. $\lambda$ in antiferromagnetic NiO, amorphous YIG, NiFe$_2$O$_4$, and Cr$_2$O$_3$ are longer than the spin decay length in paramagnetic Gd$_3$Ga$_5$O$_{12}$ and diamagnetic SrTiO$_3$. Within the four AF spacer layers, for NiO and amorphous NiFe$_2$O$_4$ with strong AF ordering, we observed surprising enhancement of spin pumping signal when the spacer thickness is below 5nm[19]. In this regime, the blocking temperature is below the room temperature, thus we believe that not only the AF ordering but also the AF fluctuation play the role in interfacial spin transport in insulators. The measured damping enhancements in the series of YIG/insulators bilayers also correlate to the $\lambda$, confirming the role of magnetic ordering in spin transfer in YIG-based heterostructures.

In the end, we also insert metallic Cu layer between YIG and transition metals with large spin orbit coupling strength, like Pt and W. Surprisingly, the insertion of a metallic layer with long spin diffusion length and weak spin orbit coupling enhance the
magnitude of spin current in YIG/Cu/W trilayers by a factor of 4.5 and suppress the
magnitude of spin current in YIG/Cu/Pt structures [100]. By measuring the intrinsic
spin mixing conductance at YIG/Cu interface and analyzed by the series circuits of
spin current flow, we determine the spin conductance at Cu/Pt and Cu/W interface
and demonstrate that the optimal interfacial condition at Cu/W is the underlying
mechanism for the observed spin current enhancement.

In conclusion, the insulating YIG thin films with exceptional low damping constant
fabricated by off-axis sputtering technique provide a good material platform to study
dynamic spin transport. Our collaborators also observed the coupling between the
Nitrogen vacancy (NV) centers and YIG [74] and the intralayer spin pumping [78]
mechanism within the YIG thin films from the localized spin wave modes to the
surrounding area. All those discoveries potentially pave a path toward high efficiency
spin transport by engineering YIG-based heterostructures involving both metals or
insulators in addition to FM and NM materials.
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