NOVEL TECHNIQUES FOR DETECTION AND IMAGING OF SPIN RELATED PHENOMENA: TOWARDS SUB-DIFFRACTION LIMITED RESOLUTION

DISSERTATION

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By

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Abstract

The idea that the spin degree of freedom of particles can be used to store and transport information has revolutionized the data storage industry and inspired a huge amount of research activity. Spin electronics, or spintronics, provides a plethora of potential improvements to conventional charge electronics that include increased functionality and energy efficiency.

Scientists studying spintronics will need a multitude of characterization tools to sensitively detect spins in new materials and devices. There are already a handful of powerful techniques to image spin-related phenomena, but each has limitations. Magnetic resonance force microscopy, for example, offers sensitive detection of spin moments that are localized or nearly so but requires a high vacuum, cryogenic environment. Magnetometry based on nitrogen vacancy centers in diamond is a powerful approach, but requires the nitrogen vacancy center to be in very close contact to the spin system being studied to be able to measure the field generated by the system. Spin-polarized scanning tunneling microscopy provides perhaps the best demonstrated spatial resolution, but typically requires ultrahigh vacuum conditions and is limited to studying the surface of a sample. Traditional optical techniques such as Faraday or Kerr microscopy are limited in spatial resolution by the optical diffraction limit.

In this dissertation I will present three new techniques we have developed to address some of these issues and to provide the community with new tools to help push forward spintronics and magnetism related research. I will start by presenting the first experimental demonstration of scanned spin-precession microscopy. This technique has the potential to turn any spin-sensitive detection technique into an imaging platform by providing the
groundwork for incorporating a magnetic field gradient with that technique, akin to magnetic resonance imaging, and the mathematical tools to analyze the data and extract the local spin information. This use of magnetic field gradients also allows for imaging below the diffraction limit, even with optical techniques, and for subsurface imaging.

Second, the technique of solid-state Hanle magnetometry will be presented. This is a novel technique for mapping the vector components of a magnetic field. We use the same measurement protocol as for a traditional Hanle type measurement, but we provide the framework for extracting the local magnetic field experienced by the spins being measured. The magnitude of the three vector components of the local field can be extracted by analyzing the lineshape of the Hanle type measurement, and the direction of one of the components can be extracted. We demonstrate this technique by measuring optically pumped spins in GaAs in the presence of a magnetic field from macroscopic permanent magnets and a micromagnetic particle.

Finally, I will show how we have discovered a new effect using nitrogen vacancy (NV) centers in diamond which can be used to locally detect magnetization dynamics in adjacent magnetic materials. This effect occurs far from the NV centers’ own magnetic resonance, but provides us with sensitive detection of magnetization dynamics, such as ferromagnetic resonance, spin wave modes or domain dynamics, even hundreds of nanometers away from the NV centers. The technique works at room temperature in ambient conditions and has the potential to allow for the imaging of ferromagnetic phenomena and spin transport at the nanoscale.
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3.1 The simulations shown illustrate the key physics underlying scanned spin-precession microscopy. The inhomogeneous magnetic field of a micromagnetic particle (gray sphere with black arrow indicating the magnetization direction) generates well understood spatially varying spin dephasing. Spins being injected into the semiconductor reach a steady-state density which results from a combination of the injection rate, the local spin properties and the local magnetic field, provided primarily by the particle. The colored arrows in the images represent the steady state spin density vector. The color scale represents the \( \hat{z} \) component of the spin density (parallel to the orientation of injection). (a) Demonstrates the case of a spatially uniform injection rate and highlights the extent of the influence of the micromagnetic particle’s field. The bottom left hand corner shows the coordinate system used throughout this chapter as well as various components of the particle’s field. (b) and (c) show simulations similar to (a) but with a Gaussian profile of injection and for two different position of the particle.

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3.6 The spin signal, $\Sigma(r_p)$, for various applied magnetic fields, $B_t = B_{\text{external}}$, plotted as a function of the magnetic particle’s position along the $\hat{y}$ direction (left plot) and $\hat{x}$ direction (right plot). The data at different fields are offset vertically for clarity. The open circles are experimental data while the solid lines are fits obtained using equations 3.6b and 3.7c. We used the independently obtained values of the magnetic moment, $m = 2 \times 10^{-9}$ J/T, the dipole’s height (using the diameter of the particle), $z_p = 8$ µm, and the spin lifetime, $\tau_s = 2.33$ ns (obtained using the Hanle measurement from figure 3.7). The peaks marked by the green dashed lines result when the laser spot, and thus the peak in spin density, $\rho$, lies directly under the magnetic particle. The net field in this position is mostly due to the particle’s field in the $z$-direction, $B_{pz}$. This dominates the total field even when the applied field is at the maximum 0.145 T. This results in a large signal. The second set of peaks seen only in the line scans in the $x$-direction and marked by the blue dashed line occur when the transverse field from the magnetic particle cancels the applied magnetic field, $B_{px} = -B_{\text{external}}$. The resolution of our technique can be estimated by the noise and slope of the data seen in the line scans. For the bottom right black curve we obtain a resolution of 1.2 µm (see section 3.4.5).

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4.1 An example of how an extra magnetic field can change the lineshape of the data in a Hanle type measurement.

4.2 Figure taken from [27] showing Hanle magnetometry performed using \(^{87}\)Rb gas. This shows the change in signal from a square pules of magnitude \( 3 \times 10^{-14} \) T. This extreme sensitivity is due to a narrow Hanle half-width and a large signal to noise ratio.

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4.11 Optical image of the GaAs membrane used in the experiment to measure the magnetic field from a micromagnet. It is shown with the photoluminescence spot as well as the NdFeB micromagnetic particle which is glued to the back of the membrane. Also shown is a green arrow representing the externally applied magnetic field in the x-direction for Hanle measurements.

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5.1 The optical setup used for experiments with NV centers. Details of the individual components are presented in table 5.1.

5.2 The signal processing setup used for experiments with NV centers. Details of the components are in the text and presented in table 5.1.

5.3 Experimental schematic: The sample is a 20-nm-thick epitaxial YIG film with nanodiamonds dispersed on top with a thickness of about 500 nm. To apply microwaves to the sample a silver microwire is lithographically patterned on the YIG. Green laser light is focused onto the nanodiamonds near the wire, and the intensity of the resulting photoluminescence from the NV centers is measured. The inset is an SEM image of dispersed nanodiamonds showing the size range from about 50-200 nm.

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preamplifier.

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directly on top of YIG, for comparison. (c) Line cuts of the data in (a) and
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5.11 Experimental schematic: Experiments were performed on a 5 m thick YIG film with a lithographically defined microwave antenna. Nanodiamonds were dispersed on top and were in contact with the YIG. Changes in the luminescence of the NV centers in the nanodiamonds were recorded as a function of the static magnetic field, $H_0$ (large red arrow) and the frequency of the microwave field, $H_1$ (elliptical green arrow), at various locations on the sample. Positions 1, 2, and 3 indicate the locations of the focal spot where the NV signal was measured and correspond to panels (c), (d) and (e) respectively of figure 5.12. Magnetization dynamics in the ferromagnet were measured simultaneously with the luminescence by monitoring the reflected microwave power from the microwire on a diode. This reflection signal is spatially averaged over the entire sample in contrast with the NV optical signal which measures local dynamics.

5.12 Broadband and localized spectroscopy of YIG using an ensemble of NV centers in nanodiamonds: (a) Spatially localized optical NV signal at three positions (blue lines, left axis) and global reflected microwave power (solid black line, right axis) as a function of field $H_0$ at a microwave frequency of 1.8 GHz. (b, c, d) Field-frequency 2D maps of the broadband dependence of NV PL as a function of field $H_0$ and frequency of $H_1$ for positions 1, 2 and 3 (see Fig. 1), respectively. The green lines indicate the extrema of the powder spectra of the NV ground (upper two lines) and excited state (bottom two lines) magnetic resonances. Note the low field features, below about 4 mT, which indicate sensitivity to dynamics associated with domains in unsaturated YIG. (e) 2D map of the reflected microwave power as a function of field $H_0$ and frequency of $H_1$. The superposed red lines show the calculated dispersions for several spinwave branches of this YIG film.

5.13 Measurement of ferromagnetic dynamics using NV centers in magnetic fields exceeding those typically used for conventional ODMR magnetometry: NV optical signal as a function of magnetic field at various microwave frequencies, showing the evolution of the ferromagnet spectrum.

A.1 (a) The MOCVD growth structure for the GaAs samples. (b) A GaAs membrane glued to a sapphire substrate.

A.2 (a) Optical image of NdFeB micromagnetic particles glued to a GaAs membrane. Lines are scratched into the GaAs to aid in locating the particles. (b) An SEM image of the same NdFeB particles for size calibration.

B.1 (a) Needle with epoxy, attached to a micro-manipulator, and near a bare cantilever. (b) Cantilever after a small amount of glue has been applied to the tip with the needle. (c) and (d) The cantilever after a NdFeB particle has been placed on the tip, focused on the cantilever and particle, respectively. (e) A sideview of the cantilever after the epoxy has been cured. (f) The cantilever mounted in the microscope with optical fiber to perform interferometry.

B.2 Cantilever magnetometry performed by monitoring the frequency of the cantilever oscillation as a function of applied magnetic field. By fitting to the equation shown we obtain a magnetic moment of 4.576e-9 J/T.
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Utilization of the spin degree of freedom in materials systems for information storage and processing has revolutionized information technology and inspired a huge amount of research activity. This field of research combining electronics with spin, known as spintronics, aims to make useful devices by understanding how spins behave in solid-state systems. This is accomplished by studying spin relaxation, dynamics and transport in many different materials including metals, semiconductors, and insulators as well as across interfaces and in heterostructures [5, 15, 16, 34, 42, 69, 79, 89, 91, 101, 107, 109, 113]. This includes the study of many effects for the generation, manipulation, storage, and detection of spin polarization. Adding spin-based functionality to memory and logic devices has the potential for (and in some cases already realized) nonvolatile memory, increased processing speed, less power consumption, and higher device density [6, 35, 89, 108, 113]. There are many challenges still facing this field, which include but are not limited to optimizing spin lifetime in device structures, detection of spin at the nanoscale, and transport and manipulation at relevant length and time scales [6, 89].

Traditional spintronic devices rely on the alignment of spins and the ability to detect that alignment with a measurable quantity, such as a current. This is highlighted in what some consider to be the prototypical spintronic device: the spin valve based on giant magneto-resistance (GMR). The discovery of this effect in the late 1980’s, which earned Albert Fert and Peter Grünberg the 2007 Nobel Prize, can be regarded as the beginning of the field of spintronics [6, 7, 17, 64, 89, 102, 108, 113]. A spin valve is typically made of two
ferromagnetic layers with a non-magnetic metallic layer sandwiched between. The resistance of the device relies on the relative orientation of the magnetization of the two ferromagnetic layers, namely parallel or anti-parallel. Devices which rely on tunneling magnetoresistance (TMR), or magnetic tunnel junction (MTJs), also consist of two ferromagnetic layers, but with an insulating layer between instead of metallic, and can exhibit higher relative changes in the resistance as compared to GMR devices [6, 71, 72, 108].

Like most discoveries, these were preceded and supported by a lot of other work. For example, the description of spin polarized transport introduced by Mott and expanded by others [20, 74, 108, 113] was invaluable in understanding GMR. This description, called the two current model, considers two parallel and independent channels of conduction: one for each orientation of the spin of a carrier. TMR was first measured and modeled by Jullière in ferromagnet/insulator/ferromagnet junctions [48, 113]. Today devices based on GMR and TMR have applications including field sensors, spin valve based read heads, and magnetic random access memory (MRAM) and are already influencing multi-billion dollar industries [6, 35, 108].

These devices, based on decades of research in magnetic field control of the magnetization of ferromagnetic bits for binary encoding, are probably just the beginning of the influence of spintronics on information technology [6, 35]. The merging of magnetic with electronics and even photonics could lead to many new multifunctional devices. One such possible device, introduced by Datta and Das in 1990 [22, 113], is the spin field effect transistor (spin-FET). This device relies on the injection of spin into a semiconducting channel by a ferromagnetic contact, which is then manipulated by an electric field through spin-orbit coupling, and read out by a second ferromagnetic contact on the other side of the channel. While such electrical injection of spin into a semiconductor has been achieved [31, 113], a working spin-FET has not yet been achieved. Other means of spin injection into semiconductors are also promising, such as optical orientation [58, 69, 113].

The future of spintronics is promising, but relies on a deeper fundamental understanding of spin interactions in novel materials and structures, particularly on the nanoscale and with regard to the role of low dimensionality, defects, and band structure [35, 108]. There have
been many recent discoveries that are encouraging. This includes, to name just a few, spin transfer torques which could lead to direct current control of the magnetic orientation of a bit [6, 81], multiferroics which exhibit coupling of the magnetic state to other properties of the material [6, 29], the spin Hall effect and inverse spin Hall effect which create spin currents from charge currents and vice versa in spin-orbit coupled materials [6, 44, 54, 73, 87].

The field of spintronics has progressed remarkably fast and has been extremely forward looking in utilizing effects whose fundamentals are still being extensively studied by scientists [6, 35].

1.1 Motivation

To move the field of spintronics forward, a better understanding of materials and devices at the nanoscale is required. There are already a multitude of tools available for spin sensitive imaging, but they all have limitations. Magnetic resonance force microscopy has pushed magnetic resonance imaging into the nanoscale. It has been used to performed 3D imaging well into the nanoscale [23] and has achieved single spin sensitivity [84]. Ferromagnetic resonance can quantitatively measure ferromagnetic material parameters, and ferromagnetic resonance force microscopy has combined that ability with sensitive nanoscale imaging. This technique has the capability to map ferromagnetic properties in materials and at length scales relevant to spintronic devices [3, 62, 111]. These techniques, however, generally are only capable of detecting spin moments that are localized or nearly so, and require a high vacuum, cryogenic environment.

Magnetometry based on nitrogen-vacancy (NV) centers in diamond is a powerful approach. It has been shown to be capable of magnetometry with a sensitivity on the order of a few $nT/\sqrt{Hz}$ [9]. It has also been shown to be able to image even a single nuclear spin [94]. This technique generally requires the NV center to be in very close contact with the spin system being studied to enable it to measure the field generated by the spins. There has been progress in creating usable NV centers very close to the surface of a diamond and even in scanned cantilevers [51, 65], but the technique is still limited to measuring spins
near the surface of a sample, which is not ideal for studying a device with buried interfaces.

There are several well-established optical techniques for making spin-sensitive measurements. These include Faraday and Kerr microscopy, Brillouin light scattering, and spin-resolved photoluminescence. The magneto-optic Faraday and Kerr effects are used to study magnetism and perform microscopy on a wide variety of materials [12, 30, 80]. Spin-resolved photoluminescence measurements offer a way to study the spin polarization of carriers in solids [69]. It is a technique which has been invaluable in studies of electronic structure, however it is limited to optically active materials with an appreciable spin-orbit coupling. These optical techniques are all limited in spatial resolution by the optical diffraction limit.

These are just a few of the many spin-sensitive techniques, of which there are too many to properly cover here. Each of them has its own experimental requirements and limitations. To address some of these limitations, we wanted to create new techniques that can be used in addition to or in conjunction with other techniques. We wanted to create tools with imaging resolution into the nanoscale and which can be used to study spins in working devices. To this end it is also important to be able to measure buried interfaces, which are ubiquitous in current spintronics research and technologies.

The first technique we developed was scanned-spin precession microscopy [15, 16]. This technique aims to combine the sub-diffraction limit spatial resolution and ability to measure buried interfaces of magnetic resonance force microscopy with the detection advantages of other spin-sensitive techniques. By utilizing the Hanle effect, which describes the response of spins to a transverse magnetic field, and a magnetic field gradient, such as used in magnetic resonance imaging, any spin sensitive detection scheme can be transformed into an imaging platform. This potentially includes electrical detection schemes such as magneto-resistive effects in spin valves [32, 110]. This versatility in detection method as well as the intrinsic long range of dipolar magnetic fields could allow for the imaging of spins in their intended device environments, including in buried interfaces.

In the same vein of measuring spins in their intended environments, we developed the technique of solid-state Hanle magnetometry. This again utilized the Hanle effect, but this time to quantitatively measure the vector magnetic field experienced by the spins which
are being measured. The Hanle effect has been used to measure magnetic fields since the 1960’s [27]. This was done in a large volume of rubidium gas. We have extended this technique to solid state devices by measuring in GaAs with a spatial resolution limited by our focal spot. We have also expanded the technique to be able to measure all three vector components of the magnetic field. This technique could be applied in a straightforward manner to electrical devices, which could increase the sensitivity as well as push the spatial resolution down below the diffraction limit. This could also be a powerful technique for measuring the local magnetic fields experienced by spins in magnetic heterostructures.

Traditional NV magnetometry is a very powerful tool for measuring static magnetic fields. NV centers are also very sensitive to fluctuating magnetic fields, even away from the NV centers resonance [41, 59, 109]. We show that NV centers’ spin states are affected by coupling to magnetization dynamics in adjacent ferromagnets. These results give us insight into how spins interact across interfaces, even in insulator/insulator systems. They also give us a tool with which we can potentially image ferromagnetic dynamics at the nanoscale. The NV center is intrinsically nanoscale, and single NV center studies are routinely performed [33, 39, 47, 50, 59, 65, 82, 99]. We have also found that this effect can extend even hundreds of nanometers and across interfaces. All of this means that this technique has great potential for the imaging of ferromagnetic dynamics on the nanoscale.

These techniques provide three new ways that spintronics devices and materials can be studied and imaged. They have the potential to be very powerful tools, and in conjunction with other techniques, to push forward the field of spintronics and magnetics research in general.

1.2 Organization of this dissertation

In chapter 2 the basic physical concepts will be discussed which govern the experiments outlined in this dissertation. First, the Hanle effect will be outlined. This effect describes the dephasing of a continually pumped ensemble of spins in a transverse magnetic field. This will be important for understanding the techniques of scanned spin-precession microscopy
and Hanle magnetometry. Second, the optical pumping and readout of spin polarization
in GaAs will be explained. This will be important for understanding our experimental
demonstrations of scanned spin-precession microscopy and Hanle magnetometry. Third, the
basics behind the atomic and electronic structure of the nitrogen-vacancy center in diamond
will be discussed. And finally, optically detected magnetic resonance and the associated spin
dynamics in nitrogen-vacancy centers will be covered. These last two will be important for
understanding the experiments showing the off-resonant detection of magnetic dynamics
using nitrogen-vacancy centers.

Chapter 3 will show the theoretical background and first experimental demonstration
of scanned spin-precession microscopy. The extention of the Hanle effect to situations
with inhomogeneous magnetic fields will be explained. Next, using this understanding, it
will be shown how a spin density profile can be extracted from a globally averaged spin
signal with the use of what we have dubbed the "precessional response function.” This
function describes the response of a spin to the magnetic field profile used for imaging.
Next, the setup and protocol that was used to perform these experiments will be explained.
Following this, the results will be shown where spin-resolved photoluminescence from a
GaAs membrane is measured as a function of the position of the focal spot relative to
the inhomogeneous magnetic field created by a micromagnetic NdFeB particle. Then, the
processing of these data to determine the precessional response function and finally the
extraction of an unknown spin density profile will be demonstrated. To conclude, progress
towards extending this work to cantilever based scanned magnetic probes will be shown as
well as opportunities for future studies.

In chapter 4 the technique of solid-state Hanle magnetometry will be shown. The
understanding of the Hanle effect in inhomogeneous magnetic fields from chapter 3 will be
used to show how the vector components of the local magnetic field can be extracted from
a Hanle type measurement. Then results will be shown demonstrating this technique in the
presence of two SmCo permanent magnets in an anti-Helmholtz orientation. Also shown
will be the mapping of the vector field from a micromagnetic particle. This technique will
be compared to other magnetometry techniques.
Chapter 5 will present the off-resonant detection of magnetic dynamics using nitrogen-vacancy centers in diamond. First, the experimental setup will be shown along with the data processing used in these experiments. Next, the off-resonant nitrogen-vacancy detection of uniform mode ferromagnetic resonance in a thin yttrium iron garnet film will be presented. Then the detection of other ferromagnetic excitations in a thicker yttrium iron garnet film will be shown. These include spinwaves and domain dynamics in the unsaturated state. Following this the off-resonant nitrogen-vacancy detection of the electron paramagnetic resonance of P1 centers in diamond will be shown. This chapter will conclude with a discussion of possible explanations of this effect.
Chapter 2
BACKGROUND

In this chapter I will provide the background that is necessary to understand the three experiments presented in this dissertation. First, I will discuss the precession of spins in a magnetic field and the effect it has on the steady state spin polarization balanced by constant injection and relaxation of spins, known as the Hanle effect. Second, I will discuss the more specific example, relevant to our experiments, of optically pumped gallium arsenide (GaAs). I will discuss the band structure and how it plays a role in the creation of a non-equilibrium spin polarization and the measurement of said polarization.

Next, I will show the basics of nitrogen vacancy centers in diamond. I will introduce the atomic and electronic structure of the system, and highlight the electronic states most relevant to our experiments. I will then describe the dynamics associated with this structure that allow for optical readout of the spin state. This makes possible the powerful technique of optically detected magnetic resonance as well as our experiments with off-resonant detection of magnetic dynamics.

2.1 Precession of Spins in a Magnetic Field and the Hanle Effect

The dependence of steady state spin polarization on a transverse magnetic field is known as the Hanle effect. This effect is very often used as a way to distinguish spin-related phenomena from other effects [69]. More precisely, a measurement of the steady state spin polarization in a system as a function of magnetic field transverse to the orientation of
generated spin polarization will have a Lorentzian lineshape with a width proportional to the relaxation rate. This effect is due to the competition between the generation of spins, the precession of the spins in a magnetic field, and the relaxation of the spins.

Let us first consider simply a spin, such as that of an electron, in a magnetic field. It will experience a torque given by \( \tau = \mu \times B \), where \( \mu = g \mu_B S \). \( \mu \) is the magnetic moment of the spin, \( g \) the g-factor, \( \mu_B \) the Bohr magneton, and \( S \) the spin. This torque will cause the spin to begin to precess about the field as shown in Fig. 2.1. The frequency of the precession is the so-called Larmor frequency and is given by \( \omega = \gamma B \), where \( \gamma \) is the gyromagnetic ratio (see the caption of Fig. 2.1). If we define the direction perpendicular to the magnetic field as the z-direction, then we can write the z-component of the spin as sinusoidally oscillating at the Larmor frequency: \( S_z(t) \propto \cos(\gamma B t) \).

![Figure 2.1: An illustration of a particle with a magnetic moment, \( \mu \) (in red), precessing about a magnetic field, \( B \) (in blue). The particle will experience a torque, \( \tau \) (in green), that will cause precession about the field with a frequency \( \omega \) (in gray). The magnetic moment, \( \mu \), and the gyromagnetic ratio, \( \gamma \), are related by \( \mu = \gamma S \), where \( S \) is the spin of the particle. The gyromagnetic ratio is related to several fundamental properties of the particle such as the charge, \( q \), mass, \( m \), and g-factor, \( g \), by \( \gamma = \frac{q g}{2m} \).]

We can represent spin relaxation as an exponential decay of spin density in time, and so we can also write: \( S_z(t) \propto e^{-t/\tau_s} \), where \( \tau_s \) is called the spin relaxation time. This can be thought of as an ensemble of spins with a distribution of spin lifetimes where the majority of spins will relax before the time \( \tau_s \) and a smaller and smaller fraction will survive for longer and longer times. If we combine this with the sinusoidal precession about the magnetic...
field, we end up with an equation for the z-component of the spin density given by

\[ S_z(t) = S_0 \cos (\gamma B t) e^{-t/\tau_s} \]  

(2.1)

where \( S_0 \) is the spin density at \( t = 0 \), when the spin is created/injected. This behavior can be seen pictorially in Fig 2.2. When an ensemble of spins is injected (at \( t = 0 \)) with an initial polarization along the z-direction and they experience a magnetic field in the x-direction, then the net spin density will precess about the x-axis with a frequency \( \gamma B \) and its amplitude will exponentially decay with time \( \tau_s \). As the magnetic field amplitude in increased, the spins will precess faster and thus rotate more before relaxing. This causes the steady-state, or time-averaged, polarization, \( \overline{S_z} \), to reduce with field. Mathematically we can write this time average as an integral of \( S_z(t) \) from \( t = 0 \), when the spins are created, to \( t = \infty \), thus sampling all possible lifetimes:

\[ \overline{S_z} = \int_0^\infty S_z(t) dt = \int_0^\infty S_0 \cos (\gamma B t) e^{-t/\tau_s} \]

(2.2a)

\[ = \frac{S_0 \tau}{\gamma^2 B^2 \tau^2 + 1} \]  

(2.2b)

We have now arrived at the well known lineshape of the Hanle effect where monitoring the steady state \( \overline{S_z} \) as a function of transverse magnetic field will yield this Lorentzian shape with a half-width proportional to \( 1/\gamma \tau_s \).

To understand the scanned spin-precession microscopy and Hanle magnetometry experiments we performed it is useful to approach the problem in a way which will make it more straightforward to later incorporate a spatially varying magnetic field with components in all three spatial directions. We consider the motion of an ensemble of spins in two dimensions where we can use the 2D drift-diffusion equation and consider the following as the equation of motion[34]:

\[ \frac{\partial S}{\partial t} = \mathbf{G} + D \nabla^2 \mathbf{S} + \zeta (\mathbf{E} \cdot \nabla) \mathbf{S} + \gamma \mathbf{B} \times \mathbf{S} - \frac{\mathbf{S}}{\tau_s}, \]  

(2.3)
Figure 2.2: The behavior of a spin distribution created by a constant excitation (in the z-direction) for several different magnetic fields (in units of one over the relaxation time multiplied by the gyromagnetic ratio). The images show what happens to spin density as a function of time in a constant magnetic field in the x-direction (out of the page). The spins precess about the x-axis (in the y-z plane) and simultaneously the amplitude is exponentially decaying with time. What we measure is the steady state spin density in the z-direction, $S_z$, and it can be seen that the sum of the projections along the z-axis decays as the magnetic field is increased from (a) to (b) to (c). (Figure adapted from [69])

where $S$ is the spin density, and its components, $S_x$, $S_y$, and $S_z$, give the difference between the density of spin up and spin down in that direction. $G$ represents the rate at which spin density is injected into the system (with, in general, a dependence on space and time). The second and third terms dictate diffusion and drift, respectively, where $D$ is the diffusion constant, $\zeta$ is the carrier mobility, and $E$ is the electric field. The fourth term represents precession of the spins around a magnetic field. $B$ is the net magnetic field experienced by the spins and can depend on space and time as well. $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio, with, again, $g$ being the g-factor, $\mu_B$ the Bohr magneton, and $\hbar$ Planck’s constant. The final term represents spin relaxation where $\tau_s$ is the spin relaxation time.

In all of our experiments $E = 0$, $\tau_s$ is spatially uniform, and $B$ and $G$ vary in space but not time. Also, in our experiments the spin injection is constrained to being along the z-axis (see Sec. 2.2); so $G = G_z \hat{z}$. In our samples we have seen that diffusion is negligible and so we take $D \to 0$, and in our measurements we are sensitive to the steady state spin
density where $S = S(x, y, t \to \infty)$ and $\frac{\partial S}{\partial t} \to 0$. After we take all of this into account we are left with

$$\frac{\partial S}{\partial t} = G_z \hat{z} + \gamma B \times S - \frac{S}{\tau_s} = 0 \quad (2.4)$$

Let us look at the situation that is typical for a Hanle effect measurement. Say we apply a magnetic field that is transverse to the injected spin direction ($\hat{z}$), so for instance $B = B_{\text{applied}} \hat{x}$. In this case $\gamma B \times S = \gamma B_{\text{applied}} (S_y \hat{z} - S_z \hat{y})$, and we can write Eq 2.4 as a system of three equations:

$$-\frac{S_x}{\tau_s} = 0 \quad (2.5a)$$
$$-\gamma B_{\text{applied}} S_z - \frac{S_y}{\tau_s} = 0 \quad (2.5b)$$
$$G_z + \gamma B_{\text{applied}} S_y - \frac{S_z}{\tau_s} = 0, \quad (2.5c)$$

The solutions for the vector components of $S$ from this system of equations can be solved for algebraically and are:

$$S_x = 0 \quad (2.6a)$$
$$S_y = \frac{G_z \gamma B_{\text{applied}} \tau_s^2}{1 + (\gamma B_{\text{applied}} \tau_s)^2} \quad (2.6b)$$
$$S_z = \frac{G_z \tau_s}{1 + (\gamma B_{\text{applied}} \tau_s)^2} \quad (2.6c)$$

We typically measure $S_z$, the spin density along the same direction as injection (see Sec. 2.2). In equation 2.6(c) we again recover the Lorentzian lineshape of $S_z$ as a function of applied magnetic field, equivalent to Eq. 2.2(b). This method of solving for the spin density will be useful when the situation gets more complicated by adding in other spatial components of the magnetic field, instead of just having the x-component, and also when
the injection, $G$, and the magnetic field, $B$, become spatially dependent. We tackle this case in Sec. 3.1.

2.2 Optical Pumping and Detection of Spin Polarization in GaAs

Optical pumping (sometimes referred to as optical orientation) of carriers in semiconductors is the effect of achieving different populations of nearly degenerate levels through an assymetry in optical absorption. The effect is a direct consequence of the conservation of the angular momentum of the absorbed light, and the effect can be much enhanced by the use of circularly polarized light. The simplest case would be if one transition, say, from the valence band to the spin +1/2 levels is allowed and from the valence band to the spin -1/2 is not. In most systems this is not the case and most or all transitions are allowed, but with different intensities [69]. We will discuss the case of GaAs, which is directly relevant to our experiments, in more detail below.

Photo-excited carriers in semiconductors live in the conduction band for a time $\tau_p$, and then recombine and relax back to the valence band. If the carrier is polarized and its spin lifetime is greater than the recombination time ($\tau_s > \tau_p$), then the emitted light can also be polarized [113]. By measuring the polarization of the luminescence we can monitor the spin polarization in the material. GaAs is one of the canonical materials for optical pumping and spin-resolved photoluminescence, and because it has been so extensively studied it makes a perfect testbed for our new techniques of scanned spin-precession microscopy and solid-state Hanle magnetometry.

2.2.1 Optical Pumping In GaAs

GaAs is a III-V semiconductor with a zinc-blende crystal structure. The s and p orbitals in the gallium and arsinic hybridize in the crystal but not completely. The valence band is more p-like, and the conduction band is more s-like. Its band structure near $k=0$ is shown in Fig 2.3. It has a band-gap of 1.52 eV and the valence band is split by a spin-orbit coupling of
0.34 eV. We are interested in calculating the transition rates between the different states in the valence band and the conduction band when under illumination by circularly polarized light and will follow the same logic as [113] and [14].

Figure 2.3: GaAs band structure near \( k = 0 \). The conduction band consists of s-like orbitals with orbital angular momentum \( l = 0 \) and total angular momentum \( j = 1/2 \). The valence band is more complicated, having p-like orbitals with orbital angular momentum \( l = 1 \) and total angular momentum \( j = 1/2 \) or \( j = 3/2 \). Because of spin orbit coupling the \( j = 1/2 \) and \( j = 3/2 \) bands are split as shown.

First, the s-like conduction band, with orbital angular momentum \( L=0 \), can be represented as:

\[
\Psi_c^\pm \propto Y_0^0 \left| \frac{1}{2}, \pm \frac{1}{2} \right>,
\]  

(2.7)

where \( Y_0^0 = 1/4\pi \) is the orbital part of the angular momentum and the first spherical harmonic, and \( |S, m_s\rangle = |1/2, \pm 1/2\rangle \) is the spin angular momentum part of the wave function. Second, the valence band is more complicated because the angular momentum, \( L \neq 0 \), and the crystal field of the nuclei combine to create a sizable spin-orbit coupling. This interaction is of the form \( \mathbf{L} \cdot \mathbf{S} \), and it follows that \( m_l \) and \( m_s \) are no longer good quan-
tum numbers because \( L_z \) and \( S_z \) no longer commute with the Hamiltonian. Introducing the total angular momentum, \( J = L + S \), and its \( z \)-component, \( J_z \) gives us quantities that do commute with the Hamiltonian, and thus have good quantum numbers for us to define our eigenstates \([37]\). At the zone center, \( k = 0 \), there is a four-fold degenerate band with \( |j, m_j\rangle = |3/2, \pm 3/2\rangle, |3/2, \pm 1/2\rangle \), and lowered by the spin-orbit coupling is the split-off band with \( |j, j_z\rangle = |1/2, \pm 1/2\rangle \). We can write the eigenstates of this system in terms of \( l, m_l, s \), and \( m_s \) by using the Clebsch-Gordon coefficients, and then in terms of the spherical harmonics, \( |l, m_l\rangle = Y^m_l \). We will also write \( m_s = \pm 1/2 \) as spin up or down, |↑⟩ or |↓⟩:

\[
|j, j_z\rangle = |l, m_l, s, m_s\rangle \quad (2.8a)
|3/2, 3/2\rangle = |1, 1, 1/2, 1/2\rangle = Y^1_1 |↑\rangle \quad (2.8b)
|3/2, 1/2\rangle = \frac{1}{\sqrt{3}}|1, 1, 1/2, -1/2\rangle + \sqrt{\frac{2}{3}}|1, 0, 1/2, 1/2\rangle = \frac{1}{\sqrt{3}}Y^{-1}_1 |↓\rangle + \sqrt{\frac{2}{3}}Y^0_1 |↑\rangle \quad (2.8c)
|3/2, -1/2\rangle = \frac{1}{\sqrt{3}}|1, 1, 1/2, -1/2\rangle + \sqrt{\frac{2}{3}}|1, 0, 1/2, -1/2\rangle = \frac{1}{\sqrt{3}}Y^{-1}_1 |↑\rangle + \sqrt{\frac{2}{3}}Y^0_1 |↓\rangle \quad (2.8d)
|3/2, -3/2\rangle = |1, -1, 1/2, -1/2\rangle = Y^{-1}_1 |↓\rangle \quad (2.8e)
|1/2, 1/2\rangle = \sqrt{\frac{2}{3}}|1, 1, 1/2, -1/2\rangle - \frac{1}{\sqrt{3}}|1, 0, 1/2, 1/2\rangle = \sqrt{\frac{2}{3}}Y^1_1 |↓\rangle - \frac{1}{\sqrt{3}}Y^0_1 |↑\rangle \quad (2.8f)
|1/2, -1/2\rangle = -\sqrt{\frac{2}{3}}|1, -1, 1/2, 1/2\rangle + \frac{1}{\sqrt{3}}|1, 0, 1/2, -1/2\rangle = -\sqrt{\frac{2}{3}}Y^{-1}_1 |↑\rangle + \frac{1}{\sqrt{3}}Y^0_1 |↓\rangle \quad (2.8g)
\]

To calculate the transition rates between the valence states and the conduction states we will use Fermi’s golden rule:

\[
R_{\text{valence} \rightarrow \text{conduction}} = \frac{2\pi}{\hbar} |\langle \Psi_{\text{conduction}} | H_{\text{dipole}} | \Psi_{\text{valence}} \rangle|^2 \delta(E_g - \hbar\omega), \quad (2.9)
\]

where \( H_{\text{dipole}} \) is the electric dipole interaction between the light and the electronic states, \( E_g \) is the band gap, and \( \hbar\omega \) is the energy of the light. For right and left circularly polarized light, \( \sigma^\pm \), the dipole interaction is proportional to the spherical harmonics \( Y_1^\pm \). For these transitions there is a quantum mechanical selection rule only allowing transitions where
We want to know the relative amplitude of the rates of allowed transitions. Let us consider the $|\langle \Psi_{\text{conduction}} | H_{\text{dipole}} | \Psi_{\text{valence}} \rangle|^2$ part of the equation. To calculate the rates we will use the selection rule and the fact that the spin up and spin down states are orthogonal, such that $\langle \uparrow | \uparrow \rangle = \langle \downarrow | \downarrow \rangle = 1$ and $\langle \uparrow | \downarrow \rangle = \langle \downarrow | \uparrow \rangle = 0$. For example the rate of a transition from $|j, j_z\rangle = |\frac{3}{2}, -\frac{3}{2}\rangle$ to the conduction band state $\Psi_{-}^{c} = Y_{0}^{0}|\frac{1}{2}, -\frac{1}{2}\rangle$ with right circularly polarized light, $\sigma^{+}$, relative to the transition from $|j, j_z\rangle = |\frac{3}{2}, -\frac{1}{2}\rangle$ to the conduction band state $\Psi_{+}^{c} = Y_{0}^{0}|\frac{1}{2}, \frac{1}{2}\rangle$ also with right circularly polarized light, can be calculated:

$$\frac{|\langle \Psi_{-}^{c} | Y_{1}^{0} | \frac{3}{2}, -\frac{3}{2}\rangle|^2}{|\langle \Psi_{+}^{c} | Y_{1}^{0} | \frac{3}{2}, -\frac{1}{2}\rangle|^2} = \frac{(Y_{0}^{0}Y_{1}^{1}Y_{1}^{-1})^2|\langle \downarrow | \downarrow \rangle|^2}{\frac{1}{2}(Y_{0}^{0}Y_{1}^{1}Y_{1}^{-1})^2|\langle \uparrow | \uparrow \rangle|^2 + \frac{2}{3}(Y_{0}^{0}Y_{1}^{1}Y_{1}^{0})^2|\langle \uparrow | \downarrow \rangle|^2} = 3 \quad (2.10)$$

We can repeat this for all possible transitions, and the results are shown schematically in Fig. 2.4. The number of arrows in the figure represent the relative intensity of the transition, and the solid red/dashed blue lines represent excitation with right/left circularly polarized light, respectively. As can be seen, light with enough energy to excite from the split-off band to the conduction band can excite all the transitions, and there will be equal populations of spin-up and spin-down electrons in the conduction band. However, if the energy of the light is enough to excite from the heavy hole/light hole bands but not enough to excite from the split-off band then the occupation in the conduction band is polarized. For instance if the excitation is with right circularly polarized light then the polarization will be $P \equiv (n_{\uparrow} - n_{\downarrow})/(n_{\uparrow} + n_{\downarrow}) = (1 - 3)/(1 + 3) = -1/2$, where $n_{\uparrow}$ and $n_{\downarrow}$ are the number of up and down spins, respectively.

### 2.2.2 Spin Resolved Photoluminescence In GaAs

To determine the spin polarization of the electrons in the conduction band we can measure the circular polarization of the photoluminescence. This works in a way similar to the
Figure 2.4: Schematic of the selection rules and relative transition rates for GaAs. The solid red lines represent excitation by positive helicity (right circularly polarized) light, and the dashed blue lines represent excitation by negative helicity (left circularly polarized) light. The number of arrows represents the relative intensity of the transition rate, as described in the text. Polarization of the conduction band is achieved by illumination with circularly polarized light that has energy greater than the band gap, but less than the band gap plus the spin-orbit splitting. This excites electrons to either the spin-up or spin-down band with a 3:1 ratio, which can create a polarization of 50% [14, 113].

Optical pumping. The circular polarization of the photoluminescence is defined as:

$$P_{PL} \equiv \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)},$$

(2.11)

where $I(\sigma^\pm)$ is the intensity of light with right/left circular polarization. The polarization of the light emitted when an electron relaxes follows the same relative rates as the excitation process. Light that is emitted with energy greater than the bandgap plus the spin-orbit splitting will thus be unpolarized. However, light with energy greater than the bandgap but less than the bandgap plus the spin-orbit splitting will be polarized. For example, light from the spin-up state will relax with a ratio of 3:1 right circularly polarized relative to left. Using this and equation 2.11 we can write the polarization of the photolu-
minescence when the GaAs is under illumination with right circularly polarized light:

\[
P_{PL} = \frac{(n_+ + 3n_-) - (3n_+ + n_-)}{(n_+ + 3n_-) + (3n_+ + n_-)} = \frac{2(n_- - n_+)}{4(n_- + n_+)} = -\frac{P}{2} = \frac{1}{4},
\]

where \( n_\pm \) is the number of photons from the spin-up/spin-down state respectively, and \( P \) is the polarization of the electrons in the conduction band (as discussed in the previous section). The polarization of the photoluminescence thus gives us a measure of the polarization of the carriers in the conduction band. The spin polarization relaxes back to equilibrium by either carrier recombination and photon emission or by spin relaxation, as discussed in section 2.1.

### 2.3 Nitrogen-Vacancy Centers in Diamond

Research pertaining to nitrogen-vacancy (NV) center defects in diamond has exploded in that last decade or so. A powerful combination of properties makes it a highly stable source of photons and perhaps most interestingly allows its spin state to be optically initialized and read. This has made it an important system in a variety of new technologies such as quantum information, nanoscale magnetic resonance imaging, bio-imaging, sub-diffraction limit imaging, high sensitivity magnetometry, thermometry, and other application in metrology [1, 2, 8, 25, 39, 47, 96]. The ability to measure single atomic-scale centers has made all of these more exciting and important. Although there has been five decades of research on NV centers and many of its properties are well established, there are still unknown features of its electronic structure and dynamics [25]. The extensive amount of reasearch leading to the current understanding of the NV center as well as the considerable depth of this understanding are beyond the scope of this overview. As such, this section will only cover the basics of the atomic and electronic structure as well as the electronic dynamics that lead to the optical sensitivity of the center to its spin state, which is relevant to the research presented in chapter 5.

NV centers in diamond consist of a substitutional nitrogen atom in the diamond lattice adjacent to a vacancy [2, 67, 96]. This can be seen in figure 2.5(a). The center is optically
active and exists in two charge states. The neutral NV$^0$ state has five electrons: one each from the three carbon atoms adjacent to the vacancy and two more from the nitrogen. This form of the NV center has a zero phonon line (ZPL) luminescence of 575 nm. The NV$^0$, however, does not exhibit optical sensitivity to the spin state that is crucial to many of the applications mentioned above. The negatively charged NV$^-$ center has an extra electron trapped from the lattice, most likely from single substitutional nitrogen defects known as P1 centers. The NV$^-$ center has a ZPL luminescence of 637 nm as well as a sizeable phonon side band [1, 25], as can be seen in figure 2.5(b). We are primarily concerned with the negatively charged NV$^-$ center which exhibits all of the interesting optical and spin properties which we will use and study. From here on a reference to NV centers will refer to the negative charged variety.

![Figure 2.5](image1.png)

**Figure 2.5:** (a) An illustration of a nitrogen-vacancy center in diamond. It consists of a substitutional nitrogen atom (yellow) adjacent to a vacancy (blue) surrounded by the carbon atoms (black) that make up the rest of the lattice. (b) Typical photoluminescence spectrum for NV centers. Indicated are the zero phonon lines (ZPLs) of the NV$^0$ and NV$^-$ centers at 575 and 637 nm, respectively. For clarity the data shown are taken at 45 K, where the ZPL is larger relative to the phonon side band than at room temperature.
2.3.1 Electronic Structure of Nitrogen-Vacancy Centers

Much of the NV center’s electronic configuration can be qualitatively determined by considering that it has $C_{3v}$ symmetry and consists of six electrons. While a complete group theoretical discussion is beyond the scope of this dissertation, we will discuss just what is needed to understand the basic physics of the defect. We can use the method of linear combination of atomic orbitals (LCAO) first used in [63] and again in [1, 24, 60, 67, 96]. We start with four $sp^3$ orbitals: one along the axis between the vacancy and nitrogen atom, we will call $n$, and three along the axes between the vacancy and the three nearest neighbor carbon atoms, we will call $c_1$, $c_2$, and $c_3$, as can be seen in figure 2.6(a).

![Figure 2.6](image)

Figure 2.6: (a) Many NV center properties can be understood by considering a linear combination of atomic orbitals. We will consider the four $sp^3$ orbitals around the vacancy in the NV center, labeled $n$, $c_1$, $c_2$, and $c_3$. (b) The linear combination of orbitals creates the states $a'_1$, $a_1$, $e_x$, and $e_y$. They are arranged as shown and fill according to Hund’s rules.

Using these orbitals we can construct four orbitals which transform with the $C_{3v}$ symmetry group:

\begin{align}
    a_n & \propto n \\
    a_c & \propto c_1 + c_2 + c_3 \\
    e_x & \propto 2c_1 - c_2 - c_3 \\
    e_y & \propto c_2 - c_3.
\end{align}
The Coulomb interaction mixes $a_n$ and $a_c$ to create the states $a_1$ and $a'_1$. The orbitals are arranged in energy as depicted in figure 2.6(b). They fill according to Hund’s rules. Note that all of $a'_1$ is predicted to lie below the diamond Fermi level and the rest of the states have been shown to be in the diamond band gap. The lowest energy states consist of $a_1$ and $a'_1$ fully filled and $e_x$ and $e_y$ partially filled. The lowest of these is referred to as $^3A_2$, from the group theory representation, and is an orbital singlet and has a spin of 1. This is further broken up into three states with $m_s = 0, +1,$ or $-1$. Their wavefunctions can be written as $|^3A_2, 0) = |e_x \uparrow, e_y \downarrow\rangle + |e_x \downarrow, e_y \uparrow\rangle$, $|^3A_2, +1) = |e_x \uparrow, e_y \uparrow\rangle$, and $|^3A_2, -1) = |e_x \downarrow, e_y \downarrow\rangle$, respectively. Also with this configuration of electrons is the $^1A_1$ state, which is an orbital singlet and has a spin of 0. Its wavefunction can be written $|^1A_1) = |e_x \uparrow, e_x \downarrow\rangle + |e_y \uparrow, e_y \downarrow\rangle$.

Lastly, for this configuration of electrons we can have the $^1E$ states which are spin 0 but have orbital angular momentum. These states can be written $|^1E_X\rangle = |e_x \uparrow, e_x \downarrow\rangle - |e_y \uparrow, e_y \downarrow\rangle$ and $|^1E_Y\rangle = |e_x \downarrow, e_y \uparrow\rangle - |e_x \uparrow, e_y \downarrow\rangle$.

The first excited configuration of the NV center occurs when one electron is promoted to from $a_1$ to an $e$ orbital. This also has states with $^1E$ symmetry, with spin 0 and having orbital angular momentum. These states can be written $|^1E_{X, excited}\rangle = |a_1 \uparrow, e_x \downarrow\rangle - |a_1 \downarrow, e_x \uparrow\rangle$ and $|^1E_{Y, excited}\rangle = |a_1 \uparrow, e_y \downarrow\rangle - |a_1 \downarrow, e_y \uparrow\rangle$. Also with this excited configuration of electrons there are states with $^3E$ symmetry, which have a spin of 1 and also have orbital angular momentum. These wavefunctions can be written: $|^3E_{X, 0}\rangle = |a_1 \uparrow, e_x \downarrow\rangle + |a_1 \downarrow, e_x \uparrow\rangle$; $|^3E_{X, +1}\rangle = |a_1 \uparrow, e_x \uparrow\rangle$; $|^3E_{X, -1}\rangle = |a_1 \downarrow, e_x \downarrow\rangle$; $|^3E_{Y, 0}\rangle = |a_1 \uparrow, e_y \downarrow\rangle + |a_1 \downarrow, e_y \uparrow\rangle$; $|^3E_{Y, +1}\rangle = |a_1 \uparrow, e_y \uparrow\rangle$; $|^3E_{Y, -1}\rangle = |a_1 \downarrow, e_y \downarrow\rangle$. The $^3E$ states with $m_s = |1|$ mix at room temperature leaving a single spin triplet manifold [1, 83].

The Hamiltonian of the NV center is dominated by spin-spin interaction. Other interaction such as spin-orbit or hyperfine play a role, but are beyond the scope of this dissertation. Much more can be seen in [1, 25, 96] among other texts. Here will just consider spin-spin interactions as well as the Zeeman interaction with a magnetic field. These give us the
following Hamiltonian:

\[ H = DS_z^2 + E(S_x^2 + S_y^2) + g\mu_B \vec{S} \cdot \vec{B}, \]  

(2.14)

where \( D \approx 2.87 \text{ GHz} \) is the axial zero field splitting, \( E \) is the transverse zero field splitting (which in strain free diamond is 0), \( \vec{S} = (S_x, S_y, S_z) \) are the Pauli spin matrices for \( S = 1 \), \( g \approx 2 \) is the electron g-factor, \( \mu_B \) is the Bohr magneton, and \( \vec{B} \) is the magnetic field.

Using these considerations and many experiments and calculations [24, 36, 47, 60, 67, 83] it has been determined that at room temperature the NV center can be considered an eight level system, as shown in figure 2.7 [1, 25]. \( ^3A_2 \) is the ground state and the \( m_s = 0 \) and \( m_s = \pm 1 \) sublevels are separated by the zero field splitting \( D = 2.87 \text{ GHz} \). The \( m_s = \pm 1 \) sublevels will split in magnetic field due to the Zeeman interaction. The \( ^3E \) states can be reached via optical transitions with a ZPL of 637 nm and is refered to as the excited state. The \( m_s = 0 \) and \( m_s = \pm 1 \) sublevels of the excited state are separated by a zero field splitting of 1.43 GHz. The \( ^1E \) and \( ^1A_1 \) states are not accessible from the ground state through optical transitions, but do lie intermediate in energy to the ground and excited states. The \( ^1E \) and \( ^1A_1 \) states are separated from each other by a ZPL of 1042 nm. This eight level system is the basis for all the coupling of the optical properties with the spin degree of freedom of the NV center. The dynamics within this system and how that leads to the ability to perform optically detected magnetic resonance are presented in the next section.

### 2.4 Optically Detected Magnetic Resonance of NV Centers

The sensitivity of an NV center’s photoluminescence to its spin state is the reason it is such an exciting and useful system with applications in metrology and quantum information [2, 39, 47, 96]. Tied in with this is the ability to easily polarize an NV center using unpolarized light. In this section the dynamics that take place within the NV center will be discussed. First, the different transitions in the NV center that produce spin polariza-
Figure 2.7: The NV center as an eight level system. The ground \( ^3A_2 \) and excited \( ^3E \) states have zero field splittings of 2.87 GHz and 1.43 GHz, respectively. The ground and excited states are separated in energy by a ZPL of 637 nm. The singlet states \( ^1E \) and \( ^1A_1 \) are separated by a ZPL of 1042 nm. While they are intermediate between the two, where exactly these lie with respect to the ground and excited states is still not clear.

2.4.1 Optical Pumping of NV centers

Figure 2.8 shows the eight level system described in the previous section and the transitions that lead to optical pumping of the spin. First, the NV center is excited, typically with a green laser, from the \( ^3A_2 \) states to the \( ^3E \) states, which is depicted by the green arrow. This is a spin conserving (\( \Delta m_s = 0 \)) transition. The NV center can then decay back to the ground state in a couple different ways. Most common is by emitting a photon, which can be measured as red photoluminescence, and is depicted by red arrows. The \( m_s = 0 \) state will almost always decay in this way. The \( m_s = \pm 1 \) states, however, have an alternate decay path. Through an intersystem crossing mechanism these states can non-radiatively decay to the \( ^1A_1 \) state, then through an infrared radiative decay or another non-radiative process will decay to the long lived \( ^1E \) states. Finally, the NV center will decay from the
$^1E$ states back to the $^3A_2$ states via a second non-radiative intersystem crossing process.

Figure 2.8: Schematic of the eight level NV center system with transition rates responsible for the optical pumping process. The NV center can be excited from the $^3A_2$ ground state into the $^3E$ excited state by spin conserving optical transitions. The NV center then decays by either emitting a photon or through a spin-selective intersystem crossing into the short lived $^1A_1$ state and then down to the long lived $^1E$ state. This then decays back to the ground state with a second, slightly spin-selective, intersystem crossing. Under continuous optical illumination a population of NV centers will become highly polarized in the $m_s = 0$ spin state.

The relative rates of all of these different processes is important for the physics of the NV center optical pumping and ODMR. The rate of optical excitation and photoluminescent decay between $^3A_2$ and $^3E$ is typically about 77 MHz [67], but does depend on the laser power. The first intersystem crossing, from the $^3E$ states to the $^1E$ states, is highly spin selective. The rate from $^3E$, $m_s = \pm 1$ to $^1A_1$ is about 30 MHz, but the rate from $^3E$, $m_s = 0$ to $^1A_1$, while not forbidden, is negligible [36]. The decay down to the $^1E$ state from $^1A_1$ is very fast at about 1 GHz, but then the $^1E$ states are long lived, with a rate back to $^3A_2$ being about 3.3 MHz [67]. This second intersystem crossing, from $^1E$ to $^3A_2$, is also spin selective but not nearly as dramatically. The ratio of decaying from $^1E$ to $^3A_2$, $m_s = 0$ is about 1.2 time greater than decaying from $^1E$ to $^3A_2$, $m_s = \pm 1$ [25, 82].

These transitions and rates lead to a depopulation of the $^3A_2$, $m_s = \pm 1$ states and a
polarization of the $^3A_2$, $m_s = 0$ state. This is because over multiple optical cycles a center in the $m_s = 0$ state will stay in $m_s = 0$, but a center with $m_s = \pm 1$ will be selectively depopulated by the first intersystem crossing. This all happens on timescales much faster than NV center spin lifetimes where $T_2$ is typically on the order of microseconds and $T_1$ is several milliseconds (both can be drastically longer at low temperature) [25]. This can all lead to a polarization of the $m_s = 0$ state of up to 96% [25], though it varies widely in the literature.

### 2.4.2 ODMR and Magnetometry with NV Centers

The process of optical pumping in NV centers is the same process which enables ODMR. When a microwave frequency magnetic field is applied to an NV center with energy equal to the difference between the $m_s = 0$ and $m_s = \pm 1$ levels it will cause transitions between them. When the NV center transitions to the $m_s = \pm 1$ states it will again be able to decay through the non-radiative intersystem crossing. This will reduce the average photoluminescence from the NV center because when it is polarized in the $m_s = 0$ state it decays radiatively most every time, giving a maximum photoluminescence. This can be seen in figure 2.9(a) where the photoluminescence from an ensemble of NV centers is measured as a function of applied microwave frequency. In zero magnetic field when 2.87 GHz microwaves are applied, transitions from $m_s = 0$ to $m_s = \pm 1$ are seen as a dip in the photoluminescence.

When an NV center is in a magnetic field the $m_s = \pm 1$ states will experience a Zeeman shift equal to $m_s g \mu_B B$, as shown in figure 2.9(b). When photoluminescence is monitored as a function of applied microwave frequency, dips will be seen when the frequency is equal to the sum of the zero field splitting with the Zeeman shift, as shown by the red arrows in figure 2.9(b). Magnetometry can be performed by measuring the positions of the peaks in these continuous wave measurements, where the laser and microwaves are both on continuously.

Shown in figure 2.9(c) is a broadband measurement of NV center photoluminescence as a function of applied microwave frequency and magnetic field. We can see the pronounced features at the zero field splitting of 2.87 GHz and 1.43 GHz. We can also see the Zeeman shift with magnetic field (highlighted by the blue arrows). The data do not simply exhibit
Figure 2.9: Optically detected magnetic resonance and magnetometry with NV centers. (a) An ODMR measurement of an ensemble of NV centers at zero magnetic field. Microwaves at the zero field splitting of 2.87 GHz cause transitions from $m_s = 0$ to $m_s = \pm 1$ and a subsequent reduction in average photoluminescence. (b) A level diagram showing the zero field splitting of the ground and excited NV center states (green) along with the Zeeman splitting of the $m_s = \pm 1$ states (blue). The transitions measured in an ODMR spectrum will be the zero field splitting combined with the Zeeman shift of the $m_s = \pm 1$ states (red). Magnetometry can be performed by measuring the Zeeman shift. (c) Broadband measurements of the NV center photoluminescence (represented in the color map, red being maximal and blue being minimal). The zero field splittings of the ground and excited states can be seen, along with the Zeeman splitting of the levels (blue arrows).

two peaks because it is not a single NV center being measured but an ensemble of nanodiamonds implanted with NV centers. Due to the zero field splitting, if the magnetic field is not along the NV center axis then the Zeeman splitting will not be exactly equal to $m_s g \mu_B B$. With an ensemble of nanodiamonds like this we have NV centers whose axes are pointing in all different directions, and this leads to what is known as a powder pattern. With a bulk diamond this measurement would typically show eight distinct peaks because the four different NV orientations in the diamond lattice will all, in general, have a different angle.
to the magnetic field, and thus a different splitting. In fact, these four different splittings and knowledge of the diamond orientation can enable the calculation of all three vector components of the magnetic field. The features in figure 2.9(c) below 1 GHz are artifacts of harmonics produced in our microwave generator, which is discussed in more detail in section 5.2.4.

For the experiments in chapter 5 it will be most important to understand that a change in NV center photoluminescence corresponds to a change of the spin state of the center. Much more information on performing magnetometry with NV centers can be found in [1, 2, 8, 41, 59, 61, 95, 96].
Chapter 3
SCANNED SPIN-PRECESSION MICROSCOPY

In this chapter I will present the first demonstration of the technique we have dubbed scanned spin-precession microscopy. This technique was inspired in part by magnetic resonance imaging, which converts a magnetic field response into spatial information through the use of a magnetic field gradient, and by scanned gate imaging [13, 97], which uses a local gate to manipulate a global electrical signal making it sensitive to local effects. Here we rely on the ubiquitous phenomenon of spins precessing in a magnetic field and introduce a spatially inhomogeneous magnetic field. This can transform a spin-sensitive measurement into an imaging tool, analogous to magnetic resonance imaging. As our experimental source of spatially varying magnetic field we use a micron scale magnetic particle, which is essentially a magnetic analog of the scanned electrical gate.

Scientists studying spin phenomena will need a multitude of characterization tools to address sensitive spin detection in new materials and device structures [21, 43, 56, 75, 88]. Magnetic resonance force microscopy [23, 62], for example, offers sensitive detection of spin moments that are localized or nearly so, but requires a high vacuum, cryogenic environment. Spin-polarized scanning tunneling microscopy [52] perhaps provides the best spatial resolution among spin detection techniques, but typically requires ultrahigh vacuum conditions and is limited to studying surfaces. Magnetometry based on nitrogen-vacancy (NV) centers in diamond [40, 66, 68, 85, 92] is a powerful approach that will be discussed in more detail later, but it typically requires bringing the spin system being studied into
intimate contact with a nitrogen-vacancy-containing diamond surface or a scanned diamond microstructure, allowing for measurements of the fields generated by the spin system. There has been progress in generating NV centers near the surface of diamond, but measuring buried interfaces would still be difficult. These and other tools, many of which are based on optical techniques such as Kerr microscopy [49, 53], are limited to certain materials or by other constraints, which was a motivating factor in the development of scanned spin-precession microscopy. We wanted to develop a tool to this list that could study devices in technologically relevant geometries with nanoscale resolution and across a wide range of materials. The potential to use any spin sensitive signal in combination with a scanned magnetic probe, whose dipole field can extend below surfaces and through interfaces, could create new opportunities to further the field of spintronics.

This technique is complimentary to other techniques of measuring and imaging spin related properties because it offers versatility in the detection mechanism, which means that whatever spin-sensitive measurement best fits the particular system or device that is being studied can be used and then integrated with our protocol for imaging. Regardless of the chosen detection mechanism we provide the framework for understanding and analyzing spin-sensitive data taken in the presence of inhomogeneous magnetic fields. Scanned spin-precession microscopy can thus enable the imaging of spin properties in devices with buried interfaces and in materials that are not optically active, enabling imaging using techniques that previously were globally averaged, such as electrical spin detection. It can also enhance the resolution of optical techniques, such as Faraday or Kerr microscopy, beyond the optical diffraction limit.

The effect on spin polarization from the field of a micromagnetic particle is illustrated in the simulations shown in figure 3.1. The simulations highlight the key physics governing scanned spin-precession imaging. The inhomogeneous magnetic field of the magnetic particle (shown in the figure as a gray sphere with a black arrow representing the direction of magnetization) causes spins to dephase with a well understood spatial variation. Spins which are being injected into the semiconducting sample reach a steady-state density which results from a combination of the spin injection rate, the local spin properties (such as spin
lifetime), and the local magnetic field, which is provided entirely by the particle in these simulations. We can also add an in-plane spatially uniform magnetic field in our experiments. The colored arrows in the images represent the steady-state spin density vector at that spatial location, and the color scale represents the \( \hat{z} \) component of the spin density, which is parallel to the orientation of the spin polarization upon injection. In the bottom left of panel (a) the coordinate system used throughout this chapter is shown. The spins are injected and measured along the z-direction, and the sample is in the x-y plane. Also, as shown, it will be important to distinguish between the components of the magnetic field from the particle that are parallel (\( \hat{z} \)) or perpendicular (\( \hat{x} \) or \( \hat{y} \)) to the injected spin direction.

Figure 3.1: The simulations shown illustrate the key physics underlying scanned spin-precession microscopy. The inhomogeneous magnetic field of a micromagnetic particle (gray sphere with black arrow indicating the magnetization direction) generates well understood spatially varying spin dephasing. Spins being injected into the semiconductor reach a steady-state density which results from a combination of the injection rate, the local spin properties and the local magnetic field, provided primarily by the particle. The colored arrows in the images represent the steady state spin density vector. The color scale represents the \( \hat{z} \) component of the spin density (parallel to the orientation of injection). (a) Demonstrates the case of a spatially uniform injection rate and highlights the extent of the influence of the micromagnetic particle’s field. The bottom left hand corner shows the coordinate system used throughout this chapter as well as various components of the particle’s field. (b) and (c) show simulations similar to (a) but with a Gaussian profile of injection and for two different position of the particle.
The main image in panel (a) of figure 3.1 highlights the extent of the influence of the micromagnetic particle’s magnetic field by considering the case where spins are injected uniformly everywhere in the sample. In an annular disk centered around the particle there are strong fields in the plane, perpendicular to the injected spins. This causes those spins to precess and dephase. Directly under the magnetic particle, however, is a region of reduced dephasing. This is caused by the strong magnetic field parallel to the injected spins which reduces their dephasing. The details of this physics will be discussed later in this chapter. Panels (b) and (c) show a similar situation, but now instead of a spatially uniform injection profile we have a Gaussian shape. In (b) the particle is directly centered on the Gaussian profile and most of the spins are kept from dephasing by the strong parallel magnetic fields, but in (c) the magnetic particle is offset from the Gaussian profile and the majority of the spins are sitting in the region where there are strong perpendicular fields. This causes the majority of the spins to become dephased. This is essentially where the imaging capability of scanned spin-precession microscopy comes from. The micromagnet will cause some spins to dephase and other not, depending on their position relative to the particle. By understanding exactly when and to what extent this will happen we can measure the average spin density in the sample as a function of the particle’s position and then decode the spin information using standard deconvolution techniques.

In this first demonstration of scanned spin-precession microscopy we performed spin-dependent photoluminescence measurements in GaAs because it is a very well understood system. We create a spatially varying spin density and image it using the inhomogeneous magnetic field from a micro-magnetic particle. We achieve an imaging resolution of 1.2 \( \mu m \), which is much less than the \( \sim 10 \ \mu m \) optical feature size. However, this resolution is most likely limited by the feature size and not the intrinsic limit of the technique, which is set by the magnetic field gradient of our magnetic particle.
3.1 Hanle Effect in Inhomogeneous Magnetic Fields

As mentioned above and previously in section 2.1, scanned spin-precession microscopy relies on spin precession in a magnetic field. This effect is commonly used to distinguish spin related phenomena, such as in a Hanle effect measurement [69]. We use a spatially varying magnetic field to encode local information into a globally averaged spin signal through the selective precession of spins localized near the inhomogeneous magnetic field produced by a micromagnetic particle. This effect has been discussed by Bhallamudi et al. [15, 16] and is illustrated in figure 3.1. To decode this signal and produce spatial maps of spin dependent properties we need to understand how spins behave in magnetic fields whose amplitude and direction vary in space.

First, let us look at the drift diffusion equation which was introduced in equation 2.4, but with the spatial dependence explicitly added:

$$\frac{\partial S(r, t)}{\partial t} = G(r) + D \nabla^2 S(r, t) + \zeta (E(r) \cdot \nabla) S(r, t) + \gamma B(r) \times S(r, t) - \frac{S(r, t)}{\tau_s}, \tag{3.1}$$

where \(r = (x, y, 0)\) and is the position in the sample, which is a GaAs membrane detailed in appendix A, \(S(r)\) is again the spin density, whose components represent the difference between spin-up and spin-down densities in the \(x, y\) or \(z\)-direction. \(G(r)\) represents the (spatially dependent) rate at which spin density is injected into the system, and in our experiment it is related to the intensity of the pumping laser light. The third term on the right hand side of the equation, \(D \nabla^2 S(r)\), represents diffusions of spins, where \(D\) is the diffusion constant. The fourth term, \(\zeta (E(r) \cdot \nabla) S(r)\), represents drift of spin in an electric field, \(E(r)\), where \(\zeta\) is the electron (or hole) mobility. In GaAs the spin diffusion is coupled to charge diffusion [107], and in our samples we have seen that the approximation of zero diffusion \((D = 0)\) works well. We apply no electric field, so \(E(r) = 0\), and we can simplify the equation to:

$$\frac{\partial S(r, t)}{\partial t} = G(r) + \gamma B(r) \times S(r, t) - \frac{S(r, t)}{\tau_s}, \tag{3.2}$$

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In this limit, equation 3.2 has an algebraic solution. We can write this more conveniently by introducing the following scaled field matrix:

$$B = \begin{pmatrix} 0 & B^*_z & -B^*_y \\ -B^*_z & 0 & B^*_x \\ B^*_y & -B^*_x & 0 \end{pmatrix},$$

where we have applied a scaling to $B$ (and its vector components $B_x, B_y, B_z$), $B^* = \gamma \tau_s B$. We can solve for the steady state solution by again (see section 2.1) setting $\partial S(r, t)/\partial t = 0$, and we get the compact equation:

$$S(r, t) = [I - B(r)]^{-1} \rho(r),$$

(3.3)

where $\rho(r) = (0, 0, \rho(r))$, and $\rho(r) \equiv G_z(r) \tau_s$, which represents generation of spin polarization in the z-direction. It should be noted that $\rho(r)$ is proportional to the steady state spin density in the absence of any magnetic field and is the quantity that we are interested in measuring in this experiment, extracting it from a signal that is averaged over all $r$.

Seen in the above equation is the fact that the absence of both drift and diffusion eliminates all coupling between neighboring positions. Thus, in this particular limit, the steady-state solution can be determined algebraically by the magnetic field values at a given position. The algebraically determined solutions for the three components of $S$ are determined only by $B$ and $\rho$ at a particular $r$, and are given by:

$$S_x = \frac{B^*_yB^*_z - B^*_y}{1 + B^*_x^2 + B^*_y^2 + B^*_z^2 \rho},$$

(3.4a)

$$S_y = \frac{B^*_yB^*_z + B^*_x}{1 + B^*_x^2 + B^*_y^2 + B^*_z^2 \rho},$$

(3.4b)

$$S_z = \frac{1 + B^*_z^2}{1 + B^*_x^2 + B^*_y^2 + B^*_z^2 \rho},$$

(3.4c)

where $S_x, S_y, S_z, B^*_x, B^*_y, B^*_z$, and $\rho$ are all functions of $r$. Remember this is with spin injected in the z-direction, and we measure the z-component of the spin density through the
polarization of the photoluminescence (see section 2.2.2). We can see here that if $B_y^* = 0$ and $B_z^* = 0$, then we recover the form of the Hanle effect discussed in section 2.1. In the following section I will describe how we use this form of $S_z$, generalized to vector magnetic fields that can vary in space, to describe the spatially averaged signal we measure when we perform an optical experiment near a magnetic particle’s inhomogeneous magnetic field.

3.2 Global Spin Signal and the Precessional Response Function

In this section I will describe how we theoretically describe our measured signal, which is averaged over the laser spot on our sample and measured as a function of a magnetic particle’s position relative to the laser spot. Then I will show how we can take that signal and calculate the spin density in the sample, unperturbed by the magnetic particle. We use the idea of a response function, or point spread function, similar to that sometimes used in optical imaging [77].

3.2.1 Globally Averaged Spin Signal

In our experiment we measure photoluminescence which is averaged spatially over the sample. Practically this means we measure from an area that includes our entire laser spot. We can describe this signal as:

$$\Sigma \propto \int_A S_z(r) dA,$$

where $A$ represents the area of detection and can be assumed to extend to infinity. This is because the laser spot, and thus the spin density, dies off with a Gaussian shape. We will use the expression for $S_z$ from equation 3.4 to gain further understanding of this signal. The magnetic field that determines $S_z$ derives from an externally applied, spatially uniform magnetic field in the x-direction as well as the field form the micromagnetic particle near the detection area. Assuming the magnetic particle to be a point dipole, which is a good approximation with the spherical NdFeB particles we used, the field experienced by a spin
at position $r$ in the sample is given by

$$B(r, r_p) = B_{\text{dipole}} + B_{\text{external}} \hat{x}$$ (3.6a)

$$= \frac{\mu_0}{4\pi} \frac{3R(m \cdot R) - mR^2}{R^5} + B_{\text{external}} \hat{x},$$ (3.6b)

where $R \equiv r - r_p$ is the position in the sample relative to the position of the magnetic particle, $r_p$. $B_{\text{external}}$ is the uniform applied magnetic field, $m$ is the magnetic moment of the magnetic particle, and $\mu_0$ is the permeability of free space. Substituting equations 3.4 and 3.6b into equation 3.5, we can write the measured signal as a function of the magnetic particle’s position:

$$\Sigma(r_p) \propto \int_A S_z(r, r_p) dA$$ (3.7a)

$$\propto \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left( 1 + \frac{\gamma^2 \tau_s^2 B_z^2(R)}{1 + \gamma^2 \tau_s^2 (B_x^2(R) + B_y^2(R) + B_z^2(R))} \right) \rho(r) dx dy$$ (3.7b)

$$\propto (H_B * \rho)(r_p),$$ (3.7c)

where $B(R)$ is the spatially varying magnetic field from equation 3.6b, $\gamma$ is the gyromagnetic ratio, and $\tau_s$ is the spin lifetime. The * in the third line refers to a two dimensional convolution. $H_B$ is what we refer to as the precessional response function, which is defined as

$$H_B(R) \equiv \frac{1 + \gamma^2 \tau_s^2 B_z^2(R)}{1 + \gamma^2 \tau_s^2 (B_x^2(R) + B_y^2(R) + B_z^2(R))}$$ (3.8a)

$$= \frac{1}{1 + \theta_B^2(R)}$$ (3.8b)

$$= \frac{1}{1 + \theta_B^2(R)},$$ (3.8c)

and is equivalent to the signal measured from a point source spin density (i.e. if $\rho(r) = \delta(r)$, a two dimensional delta function). We refer to $\theta_B$ as the effective dephasing factor,
which is introduced to emphasize and help understand the physics at play. It is given by

$$\theta_B(R) = \frac{\gamma^2 \tau_s B_B^2(R)}{1 + \gamma^2 \tau_s B_B^2(R)}$$

(3.9)

where $B_B = \sqrt{B_x^2 + B_y^2}$ and $B_B = B_z$, and they refer to the magnetic field component perpendicular or parallel to the injected spin direction, $\hat{z}$. What we learn from this is that if $B_B$ is large (compared to $\gamma \tau_s$ and $B_B$) then the dephasing factor is large and the response function is small, making the spin signal small. Physically, this is because magnetic fields transverse to spins will cause them to precess and dephase; this is the Hanle effect described in section 2.1. However, when $B_B$ is large (compared to $\gamma \tau_s$ and $B_B$) then the dephasing factor is small, making the response function close to 1, and the spin signal maximum. This is because, while magnetic field parallel to the spins cannot create spin density, the spins precess about the total field, and the dephasing effect of a magnetic field perpendicular to the spins is lessened by this parallel field. It should also be noted that the precessional response function also depends on the applied uniform magnetic field, $B_{external}\hat{x}$, and thus can be tuned as is seen in section 3.4.3.

### 3.2.2 Spin Density Profile as a Deconvolution

The fact that our signal can be represented by the convolution shown in equation 3.7c allows us to use well developed mathematical techniques, specifically of deconvolution, to analyze our data. In the end we want to extract the unperturbed spatially varying spin density in the sample, that is the spin density without any magnetic field applied and no micromagnetic particle nearby. Mathematically we do this by performing a deconvolution:

$$\Sigma(r_p) = (H_B * \rho)(r_p)$$

(3.10a)

$$\Rightarrow \rho(r) = \Sigma(r_p) \star H_B(R),$$

(3.10b)

where $\star$ represents a deconvolution, $r$ is the position in the sample, $r_p$ is the position
of the micromagnetic probe, and $R = r - r_p$ is their relative position. Using this we can extract the spin density, $\rho$, from our measured global signal, $\Sigma$, by performing this deconvolution, provided we know the precessional response function, $H_B$.

We can obtain the precessional response function in two different ways. First, we can calculate it theoretically by modeling our micromagnetic particle as a dipole and plugging that into the equations from the previous section. We can also obtain it experimentally if we can measure a signal for a known spin density profile by performing another deconvolution:

$$H_B(R, B_{\text{external}}) = \Sigma(r, B_{\text{external}}) \ast \rho(r), \quad (3.11)$$

where the dependence on the external field, $B_{\text{external}}$, is added explicitly because the precessional response function depends strongly on this field, and we later perform this operation for several different fields. We show the precessional response function, both theoretical and experimental in section 3.4.3, and the spin density profile in section 3.4.4.

### 3.3 Experimental Setup and Protocol

To perform scanned spin-precession microscopy measurements we built a microscope that is capable of spin resolved photoluminescence measurements, as described in section 2.2, on GaAs membrane samples, described in more detail in Appendix A, at cryogenic temperatures, typically about 17 K, and with the capability to scan the objective lens relative to the sample. Here the experimental setup will be described, and more detail can be found in [14]. The measurement protocol used to obtain the data presented later in this chapter will then be presented.

#### 3.3.1 The Optical Setup

Optical pumping of spin polarization in GaAs is based on the energy and circular polarization of the excitation laser light, and the detection is based on measuring the circular polarization of the PL, which in turn is proportional to the spin polarization within the sample. The set-up used for optical pumping and spin resolved photoluminescence mea-
measurements is shown in Fig. 3.2. Also, provided in Table S1 is a detailed list of the various optical components used and their purpose.

Figure 3.2: The optical set-up for the optical pumping and spin resolved photoluminescence detection. See Table 3.1 and Sec. 3.3.1 for more details of the various components.

The pump laser is chosen to be 780 nm to excite photoluminescence in GaAs and allow for the build up of spin polarization, as detailed in section 2.2. The laser light is linearly polarized in the vertical direction and directed through the Electro-Optic Modulator (EOM), which allows us to modulate the polarization of light, and thus modulate the spin polarization within the sample, between $\pm \hat{z}$, and enable lock-in measurements. The EOM modulates the pump laser between two linear polarization states. A dichroic mirror made for use with 780 nm light is used to reflect the laser light towards the sample. The two linearly polarized states of the light are converted into the two circular states by a quarter-wave plate. The circularly polarized laser light is focused on the sample (which sits inside an optical cryostat at a temperature of 17 K) using an objective. A wire mesh can be placed in the path of the pump beam to produce the spin density profile used in section 3.4.4.
<table>
<thead>
<tr>
<th>Description</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>A GaAs sample</td>
<td>see Appendix A for more details</td>
</tr>
<tr>
<td>B Electromagnet</td>
<td>Power supply controlled via DAQ card</td>
</tr>
<tr>
<td>C Optical crystat</td>
<td>Operated at 17K</td>
</tr>
<tr>
<td>D Objective lens</td>
<td>Nikon Plan Fluor 10X</td>
</tr>
<tr>
<td>E Quarter-wave plate</td>
<td>Thor Labs WPQ10M-780</td>
</tr>
<tr>
<td>F Dichroic mirror</td>
<td>Reflects pump towards the sample, but attenuates it during transmission</td>
</tr>
<tr>
<td>G Notch Filter</td>
<td>For 780 nm</td>
</tr>
<tr>
<td>H Cube beam splitter</td>
<td>Non-polarizing</td>
</tr>
<tr>
<td>I Focusing lens</td>
<td>$f = 100$ mm</td>
</tr>
<tr>
<td>J Wollaston prism</td>
<td>Separates the two linear polarization components</td>
</tr>
<tr>
<td>K Bandpass filter</td>
<td>Centered at 820 nm, 10 nm width</td>
</tr>
<tr>
<td>L Photodiode bridge</td>
<td>Thor Labs PDB210A, the difference channel is amplified by 100000</td>
</tr>
<tr>
<td>M focusing lens</td>
<td>$f = 200$ mm</td>
</tr>
<tr>
<td>N Cube beam splitter</td>
<td></td>
</tr>
<tr>
<td>O Long pass filter</td>
<td>Thor Labs FEL0850, cut off at 850 nm</td>
</tr>
<tr>
<td>P CMOS camera</td>
<td>For spin-insensitive imaging and tracking</td>
</tr>
<tr>
<td>Q Photodiode</td>
<td>Thor Labs DET100A, measures the DC PL</td>
</tr>
<tr>
<td>R Wire mesh</td>
<td>To create a diffraction pattern</td>
</tr>
<tr>
<td>S Electro-optic modulator</td>
<td>Thor Labs EO-AM-NR-C1</td>
</tr>
<tr>
<td>T Polarizer</td>
<td>Polarisizes vertically, as needed for the EOM operation</td>
</tr>
<tr>
<td>U Polarizer</td>
<td>Allows for fine control of pump power</td>
</tr>
<tr>
<td>V ND filters</td>
<td>For coarse adjustment of pump power</td>
</tr>
<tr>
<td>W Pump laser</td>
<td>Melles Griot 56RCS012/HS, 780 nm, stable laser source, intensity modulated</td>
</tr>
<tr>
<td>X Function generator</td>
<td>SRS DS360, for modulating EOM, $f_{mod} \sim 73$ Hz</td>
</tr>
<tr>
<td>Y Voltage preamplifier</td>
<td>SRS SR560, Amplifies the A-B channel of the diode bridge, Gain of 200</td>
</tr>
<tr>
<td>Z Voltage preamplifier</td>
<td>SRS SR560, Amplifies the B channel of the diode bridge, Gain of -1000</td>
</tr>
<tr>
<td>AA Voltage preamplifier</td>
<td>SRS SR560, Amplifies the A channel of the diode bridge, Gain of 1000</td>
</tr>
<tr>
<td>AB Lock-in amplifier</td>
<td>Signal Recovery 7265, demodulates the pump power modulation, typical time const. 200-500ms</td>
</tr>
<tr>
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</tr>
<tr>
<td>AD Function generator</td>
<td>SRS DS360, for modulating pump intensity, $f_{mod} \sim 1100$ Hz</td>
</tr>
<tr>
<td>AE Computer</td>
<td>Controls the instruments and performs data acq., thru GPIB and DAQ card</td>
</tr>
</tbody>
</table>

Table 3.1: A list of the various optical components and electronic instruments used in the experiment. The letters in the first column correspond to the letters in Fig. 3.2 and Fig. 3.4.
Photoluminescence from the sample (near the band edge of GaAs, 819 nm) is then collimated by the objective, and circular polarization is converted back into the linear states by the quarter-wave plate. The Wollaston prism splits the two orthogonal linear states; each of which is then collected by a separate photodiode that is part of a diode-bridge circuit. More details of how the photodiode signal is processed are presented in the next section.

The end of a multi-mode fiber is placed ∼1 mm away from the sample (on the side opposite to laser injection, i.e. on the same side as the micro-magnetic probe’s side) as seen in Fig. 3.3(a). The fiber is used for illuminating the sample with unpolarized broadband light (from a halogen lamp) to assist with camera imaging and tracking of the micromagnetic probe’s position during the measurements. The back illumination gives negligible spin signal for our lock-in measurements.

Also, a variety of optical filters are used in various parts of the optical set-up to isolate the photoluminescence in the detection. Please see Table 3.1 for more details.

### 3.3.2 Scanning and Data Collection

The data from section 3.4 are obtained by scanning the objective lens, and thus the pump laser beam, relative to the sample and the micromagnetic particle, using motorized stages. Note that with a homogenous sample, as in our experiments, scanning the laser spot around the membrane and particle is equivalent to scanning the particle around an unmoving laser spot and membrane. The position of the micromagnetic probe relative to the pump laser beam was tracked using a home-built Labview-based software solution. The program uses an image from a camera (labeled P in Fig. 3.2 and Table 3.1) like the one shown in Fig. 3.3(b) in conjunction with pattern recognition algorithms from National Instruments. This tells us the measured position, in pixels, of the micromagnetic probe, which we convert to microns using an SEM image, shown in Fig. 3.3(c), of the same micromagnet as a calibration. Tracking of the particle was necessary because the step size of the translation stages is somewhat variable. This leaves us with spatial maps without evenly distributed data, as shown in figure 3.8. To allow for further data analysis, we interpolate the two-dimensional data using Igor Pro software making uniform spatial grids. The data shown in figure 3.9 is
Figure 3.3: (a) Schematic of the sample showing, on the left, polarization dependent optics being performed through the sapphire substrate along with, on the right, backlight illumination with broadband unpolarized light to enable tracking. (b) Optical image from the camera (P in Fig. 3.2 and Table 3.1) used for tracking of the micromagnetic particle relative to the laser spot. (c) SEM image of the NdFeB micro-magnetic particle used to calculate the size of the particle, the optical spot size, and the pixel/μm ratio used to calibrate the position tracking.

After the Wollaston prism splits the light into the two orthogonal linear polarization states, each beam is incident on a photodiode in a photodiode bridge (L in Fig. 3.2, Fig. 3.4, and Table 3.1). The photodiode bridge allows us to compute the average steady state spin polarization in the sample given by the relation,

\[ \Sigma \propto \frac{V_A - V_B}{V_A + V_B}, \]  

(3.12)
where $V_A$ and $V_B$ are proportional to the light incident on each of the two photodiodes in the bridge. The photodiode bridge has three outputs, proportional to $V_A$, $V_B$, and $V_A - V_B$. Figure 3.4 shows a schematic of the instrumentation and hardware signal processing used to process and measure these signals.

The difference signal, $V_A - V_B$, is amplified and then measured in a lock-in amplifier referenced to the modulation frequency of the light polarization, the same as the one controlling the EOM (typically at about 73 Hz). This signal is related to the spin polarization in the GaAs, but it may be contaminated by spurious changes in the reflectivity from the sample as we scan the laser spot’s position. These changes can come from changes in the surface condition of the GaAs or from the relative position of the micromagnetic particle behind the GaAs membrane. An example of changes in photoluminescent intensity as a function of position can be seen in Fig. 3.5 at $y_p=20\mu$m where we see a dip in the sum $V_A + V_B$ (green dashed line), in the DC photoluminescence measurement (black dot-dash line), and (less obviously) in the difference $V_A - V_B$ (blue dotted line). To normalize for
Figure 3.5: Background removal and normalization of the spin signal, $\Sigma$. The normalized signal (red) corresponds to the top left curve in figure 3.6. The two lock-in signals $V_A - V_B$ (blue dotted line) and $V_A + V_B$ (green dashed line) correspond to the difference and sum of the signals from the two photodiodes in the diode bridge. The spin density in the GaAs is proportional to the normalized signal. The normalization is necessary because of spurious changes in $V_A - V_B$ due to changes in reflectivity as the laser spot is scanned around the sample. These changes in reflectivity are apparent in the DC photoluminescence signal (black dot-dash line) and in the sum signal. For more information please see 3.3.2.

In this section I will be showing the spin-resolved photoluminescence data we have measured in the sample described in appendix A. I will show how we can use the theoretical framework described in section 3.2 not only to fit the data we measure very well, but also to extract the spin density profile in the GaAs. We first use a Gaussian injection laser spot to determine the precessional response function from a known spin density (namely the laser spot imaged with a camera). We then put a metal mesh in front of our laser beam to create a diffraction pattern of three spots on the sample and show how we can use the previously determined...
precessional response function to extract the spin density in the sample from the measured global signal.

3.4.1 Spatial Linescans of Spin-Resolved PL Around a Micromagnet

The magnetic fields parallel and perpendicular to the injected spins, $B_{\parallel}$ and $B_{\perp}$, have distinct spatial variation in our experiments due to the micromagnetic particle and our ability to apply an external uniform field in the x-direction. The consequences of these effects are evident in figure 3.6, where we show line scans along the x and y-directions for several values of the externally applied magnetic field, $B_{\text{external}}$. Also shown are fits to the data obtained using equations 3.6b and 3.7c, in which the probe is modeled as a dipole with a moment $m = m\hat{z}$ located at a height $z_p$ above the sample (see figure 3.6 caption for more details).

These scans were taken with the Gaussian shaped laser spot (see figure 3.9). The peaks at the center of the scans, marked by green dashed lines at $x_p = 0$ or $y_p = 0$, occur when the probe is located directly above the point of maximum spin density. At this position there is a maximum in the magnetic field parallel to the spins, $B_{\parallel}$, due to $B_{pz}$. We estimate this to be about 0.8 T. This field preserves the spin density $S_z$ by reducing the dephasing due to the transverse magnetic field.

When the magnetic particle is far from the spins, say at $y_p = 40 \mu m$, the signal decreases with increasing external magnetic field (moving from the red curve down to the black curve in figure 3.6). This decrease, from curve to curve, will have a Lorentzian lineshape because it is essentially a Hanle measurement, and it exhibits a half-width, $B_{1/2} = 1/\gamma \tau_s$, of 0.0111 T for this experiment. This is larger than expected for the doping level of our GaAs sample because the measured spin lifetime is limited by carrier recombination dynamics in our experiments (see section 2.2).

A second peak is seen, marked by the blue dashed line, in the line scans along the x-direction. These occur when the x-component of the magnetic field from the micromagnetic particle cancels the applied magnetic field. As the applied magnetic field is increased, the position where this cancellation occurs moves closer to the particle, where the field is larger.
Figure 3.6: The spin signal, $\Sigma(r_p)$, for various applied magnetic fields, $B_t = B_{\text{external}}$, plotted as a function of the magnetic particle’s position along the $\hat{y}$ direction (left plot) and $\hat{x}$ direction (right plot). The data at different fields are offset vertically for clarity. The open circles are experimental data while the solid lines are fits obtained using equations 3.6b and 3.7c. We used the independently obtained values of the magnetic moment, $m = 2 \times 10^{-9}$ J/T, the dipole’s height (using the diameter of the particle), $z_p = 8 \, \mu$m, and the spin lifetime, $\tau_s = 2.33$ ns (obtained using the Hanle measurement from figure 3.7). The peaks marked by the green dashed lines result when the laser spot, and thus the peak in spin density, $\rho$, lies directly under the magnetic particle. The net field in this position is mostly due to the particle’s field in the $z$-direction, $B_{pz}$. This dominates the total field even when the applied field is at the maximum 0.145 T. This results in a large signal. The second set of peaks seen only in the line scans in the $x$-direction and marked by the blue dashed line occur when the transverse field from the magnetic particle cancels the applied magnetic field, $B_{px} = -B_{\text{external}}$. The resolution of our technique can be estimated by the noise and slope of the data seen in the line scans. For the bottom right black curve we obtain a resolution of 1.2 $\mu$m (see section 3.4.5).
The fits indicate the effectiveness of equation 3.7c in describing our data. The various parameters for the fit were either measured independently or constrained by measured quantities. The half-width, as mentioned above, was measured at a spot on the GaAs membrane far from the magnetic particle and found to be 0.0111 T, which corresponds to a lifetime of 2.33 ns, assuming a g-factor of -0.44 for GaAs. This Hanle measurement with Lorentzian fit can be seen in figure 3.7.

![Figure 3.7: Hanle curves measured far away from the micromagnetic probe for two spin density profiles: \( \rho_c \) (red circles), which corresponds to the Gaussian beam shape and is used to calibrate the precessional response function, and \( \rho_u \) (blue plus signs), which is a diffraction pattern of three spots and can be assumed to be unknown until a deconvolution of the signal spatial map with the precessional response function is performed. The lifetime, obtained from the Lorentzian fit (green line) determined half-width, \( B_{1/2} = 1/\gamma \tau_s \), is limited by carrier recombination dynamics in our experiment. This lifetime depends on the laser power because of this, and so we adjust the power of the laser until we obtain the same lifetime for both spin density profiles so that the precessional response function is the same for both.](image-url)
A magnetic moment of $2 \times 10^{-9} \text{ J/T}$ and a moment height of $z_p = 8 \mu m$ were also used. These parameters provided better fits to the data (in terms of mean squared error) than did the values of $3 \times 10^{-9} \text{ J/T}$ and $z_p = 9 \mu m$, which is what would nominally be expected from a NdFeB particle (with $10^6 \text{ J/Tm}^3$) that has a radius of $9 \mu m$, as determined from the SEM image shown in figure 3.3. This discrepancy may be due to limitations of a simple single dipole model that we used for the magnetic particle.

We also achieve the best fits by assuming zero diffusion ($D = 0$). The negligible diffusion seen in our data is most likely a result of the fact that the luminescent region (used for spin-resolved photoluminescence detection) is limited by the availability of holes for recombination. Holes have very short diffusion lengths [113]. The low doping level of our sample, which is below the metal-insulator transition (and consequently resulting in insulating samples at the low temperatures in which our measurements are performed) might also contribute to a small diffusion constant. Lastly, for the fitting, the injection spot is assumed to be a Gaussian spot with a half-width of $5 \mu m$, which is close to the values obtained from fitting the camera image in figure 3.3 to a two dimensional Gaussian.

### 3.4.2 Measuring the Global Spin Signal

We then measured two dimensional maps of the spin-resolved photoluminescence around the micromagnetic particle. This data is shown in figure 3.8. The spatial positions of the data are not in a well-defined grid because of experimental constraints with scanning (see section 3.3). To allow further processing we interpolated the raw data to create data on a well-defined spatial grid. These data are shown in figure 3.9 (d) and (e). In (d) we can see clearly the ring around the magnetic particle where most of the injected spins are dephased by the transverse magnetic fields of the micromagnetic particle. The blue and red dots indicate positions of the laser spot relative to the micromagnet that are similar to the simulations from figure 3.1 (b) and (c), respectively. Figure 3.9 (e) shows the map of the global spin signal when we apply an external field in the x-direction, $B_t = 0.145 \text{ T}$. Here we can see that when the laser spot is away from the micromagnetic particle the external field causes all of the spins to rapidly dephase and thus show a small signal. However, when the
particle is right over the laser spot the field parallel to the spin, in the z-direction, is much stronger than the applied field and thus reduces the dephasing.

Figure 3.8: An example of raw, uninterpolated data measured with position tracking and plotted as a function of pixel position. The data will later be interpolated to make further processing possible and the positions will be scaled, using the calibration from figure 3.3(b) and (c).

Next, these data will be used to help determine the precessional response function, which can then be used to determine the spin density profile from another set of global spin signal maps.
Figure 3.9: (a) Schematic of the sample with pump laser beam and detected photoluminescence on one side of the sample, and a micromagnetic particle glued to the other. (b) Experimental camera image showing the photoluminescent spot and the NdFeB micromagnetic particle. (c) Camera image of the photoluminescence that is proportional to the spin density in the sample in the absence of the particle and any magnetic fields. (d) The interpolated global spin signal corresponding to the injection profile from (c) and with no external magnetic field, \( B_{\text{external}} = 0 \). A blue dot and a red dot indicate the position of the magnetic particle that is equivalent to the panels (b) and (c) in figure 3.1. (e) The interpolated global spin signal also corresponding to the injection profile from (c) but with an external magnetic field \( B_{\text{external}} = 0.145 \) T.
3.4.3 Theoretically Calculating and Experimentally Determining the Precessional Response Function

Presented in this section will be the procedure for both theoretically calculating and experimentally determining the precessional response function, $H_B$. We need to determine this function to be able to extract an unknown spin density profile from a spatial map of the global spin signal.

We can first theoretically calculate what the precessional response function should look like by using equations 3.6b and 3.8 and by using the same values from the fitting of the line scans in section 3.4.1. We perform this calculation for two different values of the externally applied field, $B_{\text{external}}$, and the results can be seen in figure 3.10 (c) and (d).

![Figure 3.10: (a) Precessional response function, $H_B$, obtained through Wiener deconvolution of the spin density profile (using the camera image of the photoluminescence from figure 3.9(c)) from the experimental data in figure 3.9(d), when the externally applied field is 0. (b) Experimental precessional response function for an externally applied magnetic field $B_{\text{external}} = 0.145$ T; also obtained through Wiener deconvolution, this time with the same camera image but the experimental data from figure 3.9(e). (c) and (d) Theoretically calculated precessional response functions obtained using equations 3.6b and 3.8 and the dipole moment and dipole position from the fits in figure 3.6 for external fields $B_{\text{external}} = 0$ T or $B_{\text{external}} = 0.145$ T, respectively. The experimental data have been normalized to highlight the close match between the theory and experiment.](image-url)
The theoretical calculation can be used in conjunction with experimental data maps of the global spin signal and equation 3.10 to obtain a spin density profile, however we can also experimentally determine the precessional response function. We do this by using a known spin density profile and deconvolving with the global spin signal associated with that known spin density. Our spin density profile should be proportional to that of our photoluminescence, and so we use the camera image of our photoluminescence shown in figure 3.9(c). We use equation 3.11 with the camera image and the data from figure 3.9(d) and (e) to obtain the precessional response functions shown in figure 3.10(a) and (b) for external magnetic fields of $B_{\text{external}} = 0$ T and $B_{\text{external}} = 0.145$ T, respectively. We use the Wiener algorithm [86] to implement this deconvolution. The deconvolution was implemented in Mathematica with a regularization parameter of 20. In figure 3.10(b) and (d) we can see the lobe to the left of center where $B_x$ cancels $B_{\text{external}}$.

### 3.4.4 Spin Density Profile Imaged by Scanned Spin-Precession Microscopy

To prove the fidelity of our imaging process, we used the experimentally determined precessional response functions from figure 3.10(a) and (b) with equation 3.10 to determine a nominally unknown spin density profile. We generated this new spin density profile by placing a wire mesh, with a wire spacing of about 0.5 mm, in front of our laser beam (see figure 3.2). The pump laser power was adjusted to keep the Hanle half-width the same as with the Gaussian beam profile, as shown in figure 3.7. This was to keep the lifetime and the precessional response function the same. The measured spin-resolved photoluminescence maps with external magnetic fields of $B_{\text{external}} = 0$ and $B_{\text{external}} = 0.145$ T are shown in figure 3.11 (a) and (c). We extract the spin density profile by again using the Wiener deconvolution algorithm performed in Mathematica with a regularization parameter of 10. These extracted spin density profiles are shown in figure 3.11 (b) and (d). Also shown in the figure, as an independent verification, is a camera image of the photoluminescence, which in our experiments is proportional to the spin density profile. The line cuts in figure 3.11 (e) present a more quantitative comparison of the extracted and measured data.
Figure 3.11: Obtaining the spatial variation of an unknown spin density from the measured signal, $\Sigma$: (a) Spin signal, $\Sigma_u$, measured for the nominally unknown spin density profile, $\rho_u$, with external field $B_{\text{external}} = 0$. (b) An image of $\rho_u$ extracted from the data in (a) by using the precessional response function from figure 3.10 (a) and performing a Wiener deconvolution. (c) and (d) Similarly measured spin signal, $\Sigma_u$, and extracted spin density profile, $\rho_u$, for an external field of $B_{\text{external}} = 0.145$ T. The precessional response function used for this deconvolution was from figure 3.10 (b). (e) Line cuts of the spin density profile taken along the lines in panels (b) (dashed blue line) and (d) (dashed green line). The red line in (e) was taken from (f) along the red dashed line, which is from a camera image of the photoluminescence. The spin density in our experiments is proportional to the photoluminescence, so this represents an independent measurement of the spin density profile to which we can compare our results.
We can also extract the spin density by deconvolving the signal with the theoretically calculated precessional response functions. This is shown in figure 3.12. The deconvolved images of the spin density at high field (figure 3.11 (d) and the right panel of figure 3.12) may match the true spin density profile more closely. This might be because the precessional response function at zero external magnetic field is finite even when extending the spatial coordinates out to infinity. This can result in numerical errors due to the unavoidable truncation of the precessional response function in our deconvolution process. Also, the camera image, while being very close, may not be an exact map of the spin density. This is because surface topology can impact the measured light intensity.

![Figure 3.12: Spin density profiles obtained by the deconvolution of the global spin signal from figure 3.11 (a) and (c) with the theoretically calculated precessional response functions from figure 3.10 (c) and (d), respectively.](image)

The ability to extract the spin density with both high and low external magnetic field provides evidence for excluding spurious effects, such as reflectivity changes as a function of the magnetic particle's position (which is also accounted for as described in section
3.3). Also, the external magnetic field gives us a knob that could be used to optimize the precessional response function to suit particular imaging needs. For instance, high field imaging might provide a more intuitive precessional response function in that it is nearer to a delta function and produces a measured global signal map which closely resembles the spin density profile, as can be seen in figure 3.11 (c) and (d). Low field imaging, on the other hand, might be more useful for non-local electrical devices, where a large transverse field would dephase spins before they could reach the detector.

3.4.5 Spatial Resolution of Scanned Spin-Precession Microscopy

We can estimate the spatial resolution of our technique by considering the ratio of the noise in our measured signal to the slope of the signal vs. position, as shown in figure 3.13. Using the numbers shown in the figure, we calculate that our resolution is about $1.2 \mu m$. We chose the particular linescan shown for this calculations because it showed the maximum slope of the data shown. This, however, was just the largest slope we have in our data and does not indicate the limit of the technique. Instead, it is probably limited by the feature size we are imaging. We could also increase this measure of resolution by increasing averaging by virtue of a higher signal-to-noise ratio.

Possibly the most conservative way to calculate an upper bound for the imaging resolution would be to look at the size of the features being imaged. To do this we can use the data from figure 3.11 (e). A Gaussian fitting of the narrowest lobe gives us a half width of $5.5 \mu m$. However, as in magnetic resonance imaging, the ultimate resolution of the technique, in the absence of diffusion, is set by the magnetic field gradient[15]. We can calculate the theoretical resolution by multiplying the magnetic field gradient by the half-width of our Hanle curve, which defines the field dependence of our signal. Magnetic field gradients of up to $4 \times 10^6 \ T/m$ have been achieved [23] by nanoscale fabrication of magnetic particles to submicron sizes, much smaller than what was used in these experiments. The gradients in these experiments are probably an order of magnitude smaller, and so an improvement in this aspect could greatly improve the imaging resolution. Even with the relatively large magnetic particle we used, if we assume a relatively modest gradient of $1 \times 10^5 \ T/m$, we can
Figure 3.13: The upper limit of the spatial resolution of our measurement can be estimated by taking the ratio of the noise in our signal to the slope of the signal vs. position. From this we calculate a resolution of about 1.2 µm. The data shown here are the linescan data in the x-direction, parallel to the external field, and taken at high external field, $B_{\text{external}} = 0.145$ T. This is the same data as in figure 3.6.

calculate the predicted resolution as $(0.0111 \text{ T})/(1 \times 10^5 \text{ T/m}) = 111$ nm. This is already ten times smaller than what we estimate above for this experiment, which again may be limited simply by the size of the features being imaged.

Resolution will be limited by diffusion as well, and by the unavoidable reduction of the spin signal as the region of affected spins shrinks. While diffusion can degrade the resolution, the spatial precessional response can be numerically analyzed to obtain valid and useful spatially resolved data. For large enough magnetic field gradients, sub-diffusion length and sub-diffraction limit resolution should be achievable. Images are obtained in the presence of spin diffusion in magnetic resonance imaging [11, 46, 98]; this should be feasible for spin-precession imaging as well.
3.5 Cantilever Based Scanned Spin-Precession Microscopy

The results shown previously in this chapter of the first successful demonstration of scanned spin-precession microscopy were all performed by gluing a micromagnetic particle to the back of a GaAs membrane. The laser spot pumping spins into the GaAs was scanned around the membrane near the particle. In our experiment this is equivalent to a stationary spin density profile with a scanned magnetic particle because the GaAs is uniform. However, to create a tool that is capable of studying more versatile samples and devices we want to be able to scan the magnetic particle. Towards this goal, we have taken a magnetic particle and glued it onto a modified AFM cantilever capable of scanning around the back side of our GaAs membranes. In this section spin-resolved photoluminescence data will be presented which is taken near one of these cantilevers and with the magnetic particle at several different distances from the GaAs membrane.

To create the cantilevers we used in these experiments we used spherical NdFeB particles. These created magnetic field profiles that were very close to an ideal dipole. Details of the creation of these cantilevers are shown in appendix B.

The geometry of the experiment is shown in figure 3.14. As in the previous experiments in this chapter, we have a GaAs membrane which is glued to a sapphire substrate. Spin polarization is created in the GaAs by a circularly polarized pump beam and spin-resolved photoluminescence measurements are performed on the resulting photoluminescence. The difference is that instead of gluing a particle to the GaAs membrane, we now have a cantilever behind the membrane with a particle glued to the tip. The cantilever is mounted at small angle relative to the membrane to allow for the approach, but the particle is magnetized such that its magnetization is out of plane.

We measured the spin-resolved photoluminescence as a function of positions relative to the micromagnetic particle. We again scanned the laser spot around and not the magnetic particle because of the constraints of our system, but again this should be equivalent to scanning the particle relative to the pump beam in our homogenious samples. These measurements are performed in zero applied transverse magnetic field and with different laser
Figure 3.14: Geometry of the scanned spin-precession microscopy experiments performed with a magnetic particle on a cantilever. Spin-resolved photoluminescence measurements are performed the same as in the rest of this chapter, but now instead of having a micromagnetic particle glued to the back of the membrane, we instead have one glued to the tip of a cantilever which can be brought close to the back of the membrane.

These data are shown in figure 3.15.

We measured with the cantilever either 2 µm away, 1 µm away, or touching the sample, and we performed the measurements with either and ND14.5, 08, or 03 filter in front of the pump laser beam. The different distances alter the data by changing the field profile in the sample from the micromagnet. When it is close there are stronger fields and higher gradients. The different pump powers alter the data by creating higher polarization in the center of the beam, and also measurable polarization further from the center of the beam.

These data show that SSPM can be performed with a particle on a cantilever, and that the technique is sensitive to the details of the spins being measured and the micromagnetic particle.
Figure 3.15: Spin-resolved photoluminescence data maps around a magnetic particle mounted on a cantilever. The data are taken with different pump laser powers, which is reduced from its maximal value of 53 mW by different ND filters, as shown. Data are also taken with the cantilever different distances away from the GaAs membrane.
3.6 Opportunities for Future Studies

In this chapter the first ever demonstration of scanned spin-precession microscopy\cite{16} was presented. This is a new technique for imaging spin properties using the well-understood precessional response of spins to the magnetic field of a micromagnetic particle. While we have imaged variations in the spin density, the technique is potentially more general than this because the response of the spins is sensitive to a variety of spin characteristics including spin lifetime\cite{15}. Scanned spin-precession should also make it possible to image spin species with different gyromagnetic ratios, such as electrons and holes in spin devices with p-n junctions \cite{45, 112} with a resolution better than the depletion width.

This technique should also be capable of subsurface imaging. This is because the nature of the magnetic dipole interaction between the magnetic particle and the spins being studied means the interaction can extended to the study of heterostructures where the spins might even be a few microns deep. The technique should also be applicable to a wide variety of materials and systems because it relies on proven spin polarization techniques, and can, in principle, be integrated with and of them. It should be able to enhance the imaging resolution of optical detection schemes, as was shown in this chapter, and transform any spin-sensitive measurement technique into a spin-sensitive imaging tool.
Chapter 4

Hanle Magnetometry

This chapter will show how the Hanle effect can be used to measure all three vector components of a magnetic field. The Hanle effect describes the reduction of a steady state spin density as a transverse magnetic field is applied. The measured spin density will have a Lorentzian lineshape, as shown in section 2.1, but when the ensemble of spins is subject to an extra magnetic field in addition to the applied transverse field the lineshape is affected in very specific ways that will allow us to calculate the vector components of this extra field. An example of how the lineshape can be changed is shown in figure 4.1.

Figure 4.1: An example of how an extra magnetic field can change the lineshape of the data in a Hanle type measurement.

The Hanle effect was first used to measure magnetic fields in 1969 using a gas of rubidium
atoms [27]. In this work they measure the response of polarized $^{87}Rb$ to very weak magnetic fields. They show a sensitivity to magnetic fields as low as 30 fT. This sensitivity comes from a narrow linewidth of 500 pT with a signal to noise ratio of $2.5 \times 10^3$. This measurement can be seen in figure 4.2.

![Figure 4.2: Figure taken from [27] showing Hanle magnetometry performed using $^{87}Rb$ gas. This shows the change in signal from a square pulse of magnitude $3 \times 10^{-14}$ T. This extreme sensitivity is due to a narrow Hanle half-width and a large signal to noise ratio.](image)

In the following sections it will be shown that this technique can be extended to the solid-state by using a GaAs device. By focusing a laser to optically pump spins in the GaAs, as discussed in section 2.2, we map vector magnetic fields with a spatial resolution limited only by the spot size. We have used our knowledge of spin precession in vector magnetic fields, outlined in sections 2.1 and 3.1, to show that by fitting Hanle curves we can extract all three vector components of the magnetic field. In this work we have shown a sensitivity down to 13 $\mu$T and a spatial resolution of about 10 $\mu$m, but these are not the limits of the technique, which will be discussed in sections 4.2.1.
4.1 Calculating the Local Field by Fitting a Hanle Curve

In this section I will outline how we extract the three vector components of the local magnetic field from a Hanle type measurement. We can do this because each component of the local magnetic field has a distinct effect on the Hanle lineshape. The geometry in this experiment is as shown in figure 4.3. Spins are optically pumped such that there is a net polarization in the z-direction. An external field, $B_{Hx}$, is applied to perform the Hanle type measurement. If there is a local magnetic field then the lineshape of the Hanle curve will be changed.

![Figure 4.3: Schematic showing the geometry of the experiment. Spins (shown in green) are injected such that there is a net polarization in the z-direction. A Hanle type measurement is performed by applying a field, $B_{Hx}$, in the x-direction (shown in blue). If there is a local magnetic field (shown in red) the lineshape of the measured Hanle curve will be altered.](image)

To see how to calculate the local field we refer back to equation 3.4 and take the solution for the steady state solution of the spin density $S_z$ in a vector magnetic field and rewrite it as

$$S_z = \frac{1 + (\gamma \tau B_z)^2}{1 + (\gamma \tau B_x)^2 + (\gamma \tau B_y)^2 + (\gamma \tau B_z)^2} \rho = \frac{1}{1 + \frac{B_z^2 + B_y^2}{1/\gamma^2 + B_x^2}} \rho,$$

(4.1)
where once again $\gamma$ is the gyromagnetic ratio of the spins, $\tau$ is the the spin relaxation time, $(B_x, B_y, B_z)$ is the magnetic field experienced by the spins, and $\rho$ is proportional to the spin density in the absence of magnetic fields. We see here that fields transverse to the spins will reduce the spin density, as in a Hanle measurement, and fields parallel to the spins will reduce the dephasing of these transverse fields. We can write this in terms of a local magnetic field, $\vec{B}_L$, and the applied field for the Hanle measurement in the x-direction, $B_{Hx}$.

$$
S_z = \frac{\rho}{(1 + \frac{B_L^2}{\gamma^2 \tau^2}) + \frac{B_{Hx}^2 + B_L^2}{\gamma^2 \tau^2}}
$$

(4.2)

When we perform a Hanle type measurement we will see that the data has a Lorentzian form and fit it to the following equation:

$$
S_z = \frac{A}{1 + \frac{(B_{Hx} - B_{shift})^2}{B_{1/2}^2}}
$$

(4.3)

The effect of the three free parameters in this fit (the amplitude $A$, the peak position $B_{shift}$, and the half width $B_{1/2}$) on the lineshape of our measurements is shown in figure 4.4.

Figure 4.4: An example of how a local field can change the amplitude $A$, the peak position $B_{shift}$, and the half width $B_{1/2}$ of the Hanle curve.
We can use equations 4.2 and 4.3 to see that $A$, $B_{shift}$, and $B_{1/2}$ are a function of the local field and several material parameters. When we solve for these we get:

\begin{align*}
B_{shift} &= -B_{Lx} \\
B_{1/2}^2 &= \frac{1}{\gamma^2 \tau^2} + B_{Ly}^2 + B_{Lz}^2 \\
A &= \frac{\rho}{1 + \frac{B_{Ly}^2}{1/\gamma^2 \tau^2 + B_{Lz}^2}}
\end{align*}

From these equations we can now calculate the three components of the local field:

\begin{align*}
B_{Lx} &= -B_{shift} \\
B_{Ly}^2 &= B_{1/2}^2 (1 - \frac{A}{\rho}) \\
B_{Lz}^2 &= \frac{A}{\rho} B_{1/2}^2 - \frac{1}{\gamma^2 \tau^2}
\end{align*}

Notice that we can get the magnitude and direction of the component of the field in the x-direction, but only the magnitude of the components in the y- and z-directions. To calculate these fields we also need to know the intrinsic amplitude of the signal, $\rho$, and the intrinsic half width, $1/\gamma \tau$. These are obtained by performing a measurement where there is no local field and fitting the measured curve.

### 4.2 Mapping Vector Magnetic Fields

In this section measurements will be outlined where we have mapped the vector magnetic field in a GaAs membrane from two SmCo permanent magnets in an anti-Helmholtz geometry. The sample used was a 1 $\mu$m thick GaAs membrane, similar to that used in scanned spin-precession microscopy experiments. The preparation of these membranes is outlined in appendix A. We pump and measure spin polarization in the GaAs as described in section 2.2.
The setup for this experiment is shown in figure 4.5. The GaAs membrane is shown, and the photoluminescence from the focal spot of the pump laser is near the center of the image. A schematic of the SmCo magnets is shown in blue around the GaAs, and the magnetization direction is shown with arrows. Green arrows show the orientation of the local field created by these SmCo magnets. The red arrow shows the orientation of the externally applied magnetic field used to perform the Hanle type measurements.

Figure 4.5: The experimental geometry for mapping the vector magnetic field from two SmCo permanent magnets in an anti-Helmoholtz orientation. In the middle is a camera image of the photoluminescence from the GaAs membrane with a focal spot of about 10 \( \mu \text{m} \) which can be scanned around the sample. The SmCo magnets are shown in blue with their magnetic orientation shown with blue arrows. In green is shown the magnet field lines created by the SmCo magnets. In red is shown the externally applied magnetic field in the x-direction to perform Hanle measurements.

We perform measurements at different positions by moving the objective lens, which is on stages as discussed in section 3.3. Shown in figure 4.6 are measurements taken at seven different positions on the GaAs membrane relative to the SmCo magnets. The schematic on the left shows approximately the orientation of these positions. They are all in a line where the y-position is 0 and the x-position is from \(-1.27 \text{ mm} \) to \(+1.27 \text{ mm} \). The data are shown by dots, and the Lorentzian fits are shown with solid lines.

The most obvious change in the lineshape between scans is the peak position, which equation 4.5 tells us is equal and opposite to the component of the local field in the x-
direction. This can be understood because when the applied field cancels the local field then the spins will see a net field in the x-direction of zero, which is where the peak in the spin signal will occur. The calculated x-component of the local field is plotted vs. position in figure 4.7. We see that the extracted field values fit very well to our expectation of a linear field profile due to the anti-Helmholtz SmCo magnets. We also see that the error bars are much smaller than the field values measured. In addition, the amplitude and width of the curves change as we change position on the sample; these effects are due to local fields in the y and z-directions.

We measured Hanle curves over the 2D plane of the GaAs membrane over approximately a $1 \times 2$ mm area. We extracted the magnetic fields, $B_x$, $|B_y|$, and $|B_z|$, and these data are plotted in figure 4.8. The x-component of the field, $B_x$, is shown in figure 4.8(a). We can see the slope of the field along the x-direction, as we did in figure 4.7, which was a linecut of this larger 2D map. We see again the magnitude and direction of $B_x$. There appears to be a slight tilt to the field gradient such that the field also changes in the y-direction. This could be because the permanent magnets are slightly tilted about the z-axis.

The magnitude of the y-component of the field, $|B_y|$, is shown in figure 4.8(b). Notice that the field scale for these data is smaller than in (a). For the anti-Helmholtz geometry we expect the y-component of the magnetic field to vary along the y-axis in such a way that $B_y$
\[ B_x = -B_{shift} \]

Figure 4.7: Values of the x-components of the local field extracted from the data in figure 4.6. The values fit our expectation of a linear field gradient due to the anti-Helmholtz configuration of the SmCo permanent magnets around the GaAs membrane. The extracted values are plotted as points with error bars showing the Lorentzian fitting error (too small to be seen on this scale). A linear fit is shown by the solid line.

is zero along \( y = 0 \), positive above \( y = 0 \), and negative below \( y = 0 \). We see in the data that there is a minimum near \( y = 0 \) and an increase in magnitude above and below, and while we cannot tell the sign from the measurement, the sign can be determined by using our knowledge of \( B_x \) and the anti-Helmholtz geometry of the permanent magnets. The fact that the minimum in \( |B_y| \) is not zero indicates that our membrane, and thus the (x,y) plane of our measurement, is not exactly lined up with the center of the permanent magnets. Based on the gradient observed over the mm scale of the map of about 100 \( \mu m/mT \), we predict a misalignment of about 200 \( \mu m \). This could be in the positive or negative z-direction. We also see a slight tilt to the minimum of \( |B_y| \) in the map. This is probably due to the same slight tilt that was seen in the map of \( B_x \).

The magnitude of the z-component of the field, \( |B_z| \), is shown in figure 4.8(c). Again, notice that the field scale is small on this plot, relative to the other two. Nominally we would expect \( |B_z| \) to be zero everywhere for perfect alignment, but we see a small field as well as a small gradient in the field. The small field is further evidence for a misalignment of the membrane’s plane to the center of the magnets. The gradient in \( |B_z| \) that seems to be in the x-direction is most likely due to a slight tilt in the membrane about the y-axis.
Figure 4.8: 2D maps of the vector components of the magnetic field in the GaAs membrane. (a) $B_x$ as a function of $x$ and $y$. We can see that it has a linear slope along the $x$-direction, as expected. We can measure the magnitude and the direction of this field over more than 2 mm of the sample. (b) $|B_y|$ as a function of $x$ and $y$. Notice the smaller field scale on this plot. We expect the $y$-component of the magnetic field to change along the $y$-axis in such a way that it is positive in below $y=0$ and negative above $y=0$. While we cannot tell the sign, we do see it a minimum in the center near the line along $y=0$. The fact that this does not go all the way to zero indicates that our membrane is not exactly lined up with the center of the permanent magnets. (c) $|B_z|$ as a function of $x$ and $y$. Again, notice the smaller scale on these data. We would expect this to be zero everywhere for a perfect alignment, but we see a very field and a small gradient in the field. The small field could again be due to the misalignment with the membrane plane to the center of the magnets. The gradient could be due to a slight tilt in the membrane with respect to the magnets.
4.2.1 Field Sensitivity

Here I will talk about the field sensitivity we achieve in these measurements. We performed Hanle measurements at three positions relatively close to each other, at \( y = 0 \) and \( x = 0, 13, \) and \( 18 \ \mu m \). These data are shown in figure 4.9. We fit the curves and measure the x-component of the local magnetic field to be \( 1753 \pm 16 \ \mu T \), \( 1843 \pm 13 \ \mu T \), and \( 1911 \pm 14 \ \mu T \). These values are plotted vs. the position in the inset. From these data we get a minimum sensitivity, given by the fitting error for the scan at \( x = 13 \ \mu m \), of \( 13 \ \mu T \). This value could be smaller if the y and z-components of the local field were smaller, thus making the amplitude greater and the half-width smaller.

![Figure 4.9: Three Hanle measurements taken to show the sensitivity of the technique.](image)

While \( 13 \ \mu T \) is the best sensitivity we get from fitting, the intrinsic sensitivity, \( \zeta \), of the technique is given by a ratio of the intrinsic half-width and noise to the signal amplitude:

\[
\zeta = \frac{2B_{1/2}N}{A},
\]  

(4.6)
where $B_{1/2} = 1/\gamma \tau$ is the intrinsic half-width of the GaAs, $N$ is the noise, and $A$ is the amplitude of the signal. Figure 4.10 shows a Hanle measurement of the GaAs with no local field. From this measurement we see that the intrinsic linewidth is 0.423 mT. The data is normalized to 1 and the corresponding noise figure is $0.0045 \sqrt{Hz}^{-1}$. This number is based on the standard deviation of the noise in the data along with the time constant used in the lock-in amplifier. Using equation 4.6, this gives us a sensitivity of $3.807 \frac{\mu T}{\sqrt{Hz}}$.

![Signal Amplitude](image)

**Signal Amplitude**

$A = 1$

**Half-width**

$B_{1/2} = 0.423 \text{mT}$

**Noise**

$N = 0.0045 \sqrt{Hz}^{-1}$

![Graph](image)

Figure 4.10: Shown is a Hanle measurement of the GaAs with no local magnetic field. From fitting this curve we see that the intrinsic half-width is 0.423 mT. We have normalized the amplitude of the curve to 1, and the corresponding noise is $0.0045 \sqrt{Hz}^{-1}$. Using equation 4.6, this gives us a sensitivity of $3.807 \frac{\mu T}{\sqrt{Hz}}$.

### 4.3 Mapping the Vector Field From a Micromagnet

In this section measurements will be shown where we have mapped the vector magnetic field in a GaAs membrane from a 20 $\mu m$ diameter spherical NdFeB micromagnetic particle. The sample was the same exact sample as used in the first scanned spin-precession microscopy experiments, which is described in appendix A and section 3.3. The measurements protocol is the same as used in the previous section.

The sample for this experiment can be seen in figure 4.11. Shown is an optical image of the 1 $\mu m$ thick GaAs membrane with the photoluminescent focal spot as well as the NdFeB
micromagnetic particle, which is glued to the back of the membrane. The magnetic particle is magnetized out of plane (the z-direction) in a 2 T field. Also shown is a green arrow representing the externally applied magnetic field in the x-direction for Hanle measurements.

Figure 4.11: Optical image of the GaAs membrane used in the experiment to measure the magnetic field from a micromagnet. It is shown with the photoluminescence spot as well as the NdFeB micromagnetic particle which is glued to the back of the membrane. Also shown is a green arrow representing the externally applied magnetic field in the x-direction for Hanle measurements.

We performed measurements at different positions by scanning the objective lens so that the focal spot was at specific locations relative to the micromagnetic particle. Shown in figure 4.12 are Hanle measurements performed at five different positions. The circles are the data, and the solid lines are Lorentzian fits. Shown in red is the data taken at a position far from the micromagnetic particle. This measurement gives us that intrinsic linewidth and amplitude of a Hanle measurement performed on the GaAs membrane.

Shown in blue are the data taken at a position directly over the micromagnetic particle. At this position we expect the z-component of the field, out of the plane of the membrane, to be much larger than the x- or y-components and even the externally applied magnetic field. This is supported by the fact that the width of this curve is much larger than any of the others as well as larger than the maximum applied field of 40 mT. The x- and y-components of the field are expected to be zero, but a shift of the peak and a reduction in amplitude...
Figure 4.12: Hanle measurements at different positions around the micromagnetic particle. The blue curve shows the data and fit for \((x,y)=(0,0)\) where the focal spot is directly over the particle. Also shown are data for positions where the focal spot is 10 \(\mu m\) away in the \(x\) and \(y\)-directions and data for a position several hundred microns away from the micromagnetic particle.

The black and yellow data are taken at positions 10 microns in the positive or negative \(x\)-direction relative to the micromagnetic particle, respectively. Their peaks are shifted in field, as would be expected for positions to the side of a dipole. Their widths are larger than the red curve, indicating fields in the \(y\)- or \(z\)-directions. We expect a field in the \(z\)-direction because we are displaced in the \(z\)-direction relative to the micromagnet. Any field in the \(y\)-direction would be due to a misalignment with the line at \(y=0\) or again because of the finite size of our laser spot. At these positions spins at the top and bottom of the laser spot would experience fields in the \(y\)-direction, while spins directly at the center should experience no field in the \(y\)-direction. The green data are at a position 10 microns in the \(y\)-direction. The curve is the same as that taken 10 microns in the negative \(y\)-
direction, because we are insensitive to the sign of $B_y$. This curve has a much reduced amplitude and larger width than the red curve, as expected for a large component of the field in the y-direction.

![Image](image1.png)

**Figure 4.13:** Data taken along the $y=0$ line (shown by the red line). We can see in the data that the maximum in the Hanle curves follows the familiar shape of the transverse component of the field from a dipole.

Data was taken at many more positions along the $y=0$ and $x=0$ lines. The data taken along the $y=0$ line is plotted in figure 4.13. Similar data was taken along the $x=0$ line. These data were fit and the extracted parameters of the half-width, amplitude, and peak shift are plotted in figure 4.14(a). Qualitatively we see some features we expect. When $B_z$ is expected to be very large, directly over the particle, we see a dramatic increase in the half-width in the data in both the x- and y-directions. When $B_y$ is expected to be to be large, in the positive and negative y-directions, we see a dramatic decrease in amplitude. And where $B_x$ is expected to be large, in the positive and negative x-directions, we see the large shift of the peak position. The shift in the peak in the scans along the y-direction are probably due to a slight misalignment of the laser spot to the center of the micromagnet.

However, there are a few features that were not initially expected in these measurements.
Most dramatic is the decrease in amplitude, indicating a component of the field in the y-direction, that shows up in the scans along the x-direction. Also indicating that something more complicated is going on are the data shown in figure 4.14(b). Determining \( B_x \) is straightforward, but when the valued from (a) are used to calculate \(|B_y|\) and \(|B_z|\) there is a problem. We see that a few of the values for \(|B_y|\) are not real, as well as all of the values for \(|B_z|\) at positions of x and y greater than about 10 microns. We have determined that this is most likely due to the fact that there are such large gradients in the micromagnet’s field that the local magnetic field in different positions within the laser spot is drastically different.

This can explain why the amplitude decreases in the scans along the x-directions because even if the center of the spot is directly on \(y=0\), spins in the top and bottom of the laser spot will experience fields in the y-direction, thus reducing the amplitude of the spin polarization at those positions. The Hanle curves measured to get this data can be thought of as a sum of Hanle curves from each position within the laser spot. This will result in data which is not strictly Lorentzian in shape, which can be seen most dramatically in the blue data from figure 4.12. The curve is assymetric and has a peak towards the positive field, which is real and is due to the spot being slightly to the left of center such that a majority of spins in the laser spot experience a negative \( B_x \), while a minority experience positive \( B_x \).
Figure 4.14: Extracted data from Hanle curves measured at several positions along the $y=0$ and $x=0$ lines. (a) The extracted fitting parameters, half-width, amplitude, and peak shift, from Lorentzian fits to the data. (b) Calculated local fields from the data in (a). The calculation of $|B_y|$ and $|B_z|$ produces values that are not real for much of the data. This is because the Lorentzian fits are not good when there is a magnetic field gradient across the laser spot that is large compared to intrinsic half-width divided by the spot size.
Chapter 5
Off-resonant Detection of Magnetic Dynamics Using Nitrogen Vacancy Centers in Diamond

Understanding the transport of spin and energy between dissimilar materials is a topic of intense current interest, reflecting both the scientific richness of the topic as well as its technological potential [5, 42, 69, 79, 91, 101, 113]. Metal/metal interfaces have been extensively studied and, to a lesser degree, so have metal/semiconductor and metal/insulator systems. However, the transfer of angular momentum between two insulating materials has been more challenging to study due to a lack of suitable detection methods.

Nitrogen vacancy (NV) centers in insulating diamond provides an excellent platform for performing spin-sensitive measurements. The paramagnetic NV center is optically active and its photoluminescence is dependent on the relative occupation of the lowest-lying electronic spin state [39, 103], the physics of which was presented in sections 2.3 and 2.4. This enables sensitive optical measurement of the NV center’s spin state, making optically detected magnetic resonance of NV centers an area of intense research activity [8, 10, 28, 33, 38, 50, 61, 65, 66, 68, 93, 95, 99]. However, previous work relies on direct manipulation of NV center spins using magnetic resonance.

In this chapter experimental evidence will be presented that shows the NV center spin state can be modified nonresonantly (i.e. by irradiation with microwave magnetic fields at...
frequencies far from NV center Larmor frequencies) by coupling to the dynamics of spins in close proximity to the NV centers. We show that NV centers will couple to dynamics in a proximal ferromagnetic insulator including uniform mode ferromagnetic resonance, as well as spinwave excitations and domain dynamics. We also show that NV centers will couple to paramagnetic spins resonating at much different frequencies than the NV center. This coupling is measured as a change in the NV center photoluminescence, as is done for conventional microwave-driven resonant spin manipulations. This effect may provide valuable insight into interactions between spins in adjacent, dissimilar materials. The cases presented here all are between insulating materials, and the interaction can be effective over long distances (greater than 300 nm). More generally, this provides a method for manipulating NV center spins which could enable sensitive, nanoscale imaging of ferromagnetic phenomena by means of nanodiamonds or even single atomic scale NV centers [8, 38, 39, 65, 95].

5.1 Experimental Setup

In this section the experimental setup will be described which we use to do sensitive measurements of photoluminescence, which allows us to perform optically detected magnetic resonance and our off-resonant detection of magnetic dynamics using NV centers. First the optical setup which is used to excite the NV centers and isolate and measure the photoluminescence will be described. Next, the various microwave circuits will be shown that allow us to perform the broadband measurements as a function of the microwave frequency or power. Finally, I will describe the processing we perform after data collection.

5.1.1 Optical Setup

To detect the spin state of the NV centers in our experiments we need to be able to perform sensitive measurements of the intensity of the photoluminescence from the NV centers, the physics of which is described in sections 2.3 and 2.4. The optical setup used is shown in figure 5.1. Details of the components are shown in table 5.1.
Figure 5.1: The optical setup used for experiments with NV centers. Details of the individual components are presented in table 5.1.

Shown in figure 5.1 the NV excitation starts with a green laser (light brown box), either a 532 nm SNOC 250 mW laser or a 520 nm Coherent OBIS 520 LX 40 mW laser. Green light is chosen because it will efficiently excite the NV centers and can then be filtered out of the detection. This 1 mm diameter beam (shown in green) is then expanded to about 5 mm to fill the back of the objective lens. The light is then reflected by a dichroic mirror and into the objective lens. We typically use either a 20× objective lens with a working distance of 20 mm and an NA of 0.42 or a 40× lens with a working distance of 0.72 mm and an NA of 0.75. The objective is also mounted on three stages to allow for the focusing and position of the laser spot where we want on the sample. These stages are made by Newport, are controlled using an ESP300 unit, and have a closed loop positioning system allowing us to have accurate repositioning as we scan around a sample.

The laser then excites photoluminescence in the NV centers on the sample which is collected again by the objective lens. Around the sample is an electromagnet used to apply an in-plane static magnetic field. The field was from either two small electromagnets from Magnetech Corporation, model OP-1212, with a maximum field of about 35 mT, or larger.
electromagnet, from GMW, with a maximum field of about 180 mT. The photoluminescence is then transmitted through the dichroic mirror and passes through several color filters to remove any green light remaining. The photoluminescence is then split by a 50/50 mirror. Half of the light is deflected and focused onto a camera which is used to help focus the laser and to move the focal spot to the position of our choosing on the sample. The second half of the light sometimes passes through a second set of ND filters to adjust the intensity of the photoluminescence on the photodiode to optimize detection (usually no filter is used here).

The photoluminescence then passes to the detection. We will either focus the light directly onto a photodiode, as shown in Detection 1 in the bottom left of figure 5.1, or the light will be coupled into a multi-mode optical fiber and then into a photodiode, as shown in Detection 2 in the bottom right of figure 5.1. The fiber coupling is preferred because it acts to give us a confocal microscope as well as allow for easy swapping between different photodiodes, but the coupling is not perfect so some light is lost in the process which can reduce the signal. We use a variety of photodiodes in our different experiments. We choose based primarily on the sensitivity and bandwidth we need for a particular experiment. We have used photodiodes from Thorlabs: both the DET110 and the DET100A. We also have used an avalanche photodiode from Laser Components: the A-CUBE-S1500-10. The processing of the current (or voltage from the APD) from the photodiodes will be described in section 5.1.3.

5.1.2 Microwave Setup

To excite magnetic resonance in our samples we have to apply a microwave magnetic field. We have employed several different tactics and designs for this purpose. The simplest is the use of a small diameter wire, usually 50 \( \mu m \) diameter gold wire, held above the sample. Usually, however, we pattern microwires on our samples using photolithography and depositing 100-400 nm of gold or silver.

We modulate the microwaves using the amplitude modulation of the Wiltron microwave generator, which allows for lock-in detection of the reflected microwave power, and thus
5.1.3 Signal Processing

A lock-in measurement is performed on the photodiode signal by modulating the amplitude of the applied microwave field. To do this we use the scheme shown in figure 5.2. A signal generator, an SRS DS360, creates a square wave at about 1000 Hz and controls the amplitude modulation in the microwave generator. The photoluminescence from the NV centers is incident on the photodiode. If the photodiode used is one of the Thorlabs photodiodes, then the current is fed into a current preamplifier. The ACUBE APD has a built in preamplifier.

The voltage is fed into a lock-in amplifier with a reference from the signal generator modulating the microwave amplitude. This allows us to very sensitively measure changes in the photoluminescence that are only due to the application of microwaves. The voltage is also fed into a data acquisition card which is used to measure the DC intensity of the photoluminescence. This allows us to normalize the lock-in signal when we move the laser spot to different positions on the sample which may have different reflectivity, NV center density, or nanodiamond quantity.

We also measure the reflected microwave power from the sample using a microwave diode. The voltage from the diode is fed into a second lock-in amplifier also referenced to the frequency of the microwave amplitude modulation. This allows us to measure changes in the reflection, and thus absorption of the microwaves on the sample and to simultaneously measure the ferromagnetic resonance of the sample while performing optical experiments. The DAQ and lock-in signals are read by LabVIEW virtual instruments on the computer.
Figure 5.2: The signal processing setup used for experiments with NV centers. Details of the components are in the text and presented in table 5.1.
<table>
<thead>
<tr>
<th>Description</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pump Laser</td>
<td>Either a 532 nm 250 mW laser from SNOC or a 520 nm laser from Coherent, model OBIS 520 LX</td>
</tr>
<tr>
<td>ND filters</td>
<td>Filters to modify the intensity of the pump laser</td>
</tr>
<tr>
<td>Beam Expander</td>
<td>To expand the laser beam to fill the back of the objective lens</td>
</tr>
<tr>
<td>Dichroic Mirror</td>
<td>To reflect the green pump laser towards the sample and transmit the photoluminescence towards the detection</td>
</tr>
<tr>
<td>Objective Lens</td>
<td>To focus the pump laser onto the sample and collect photoluminescence, mounted on stages for 3D motion</td>
</tr>
<tr>
<td>Magnet</td>
<td>Iron core electromagnet to apply an in plane static magnetic field</td>
</tr>
<tr>
<td>Sample</td>
<td>Sample with NV centers; specifics will be explained in relevant sections</td>
</tr>
<tr>
<td>Color Filters</td>
<td>550 nm and/or 570 nm longpass filters to block whatever pump laser light gets through the dichroic mirror</td>
</tr>
<tr>
<td>50/50 Mirror</td>
<td>50/50 mirror to divert some of the photoluminescence into the camera</td>
</tr>
<tr>
<td>Lens</td>
<td>200 mm lens to focus light into the camera</td>
</tr>
<tr>
<td>Camera</td>
<td>To help position the focus spot on sample</td>
</tr>
<tr>
<td>ND Filters</td>
<td>To reduce the photoluminescence to optimize the detection, specifically when using the single photon counter</td>
</tr>
<tr>
<td>Light Detection</td>
<td>One of two detection schemes: focusing directly on a photodiode or fiber coupling before incidence on photodiode</td>
</tr>
<tr>
<td>Lens</td>
<td>Lens to focus onto the photodiode or optical fiber</td>
</tr>
<tr>
<td>Photodiode</td>
<td>To detect the photoluminescence</td>
</tr>
<tr>
<td>Preamplifier</td>
<td>To amplify the current from the photodiode and convert it to a voltage</td>
</tr>
<tr>
<td>DAQ</td>
<td>Data acquisition card</td>
</tr>
<tr>
<td>Lock-in 1</td>
<td>Lock-in amplifier measuring the photoluminescence, referenced to the modulation of the microwave amplitude</td>
</tr>
<tr>
<td>MW Source</td>
<td>Microwave generator with built in amplitude modulation</td>
</tr>
<tr>
<td>Signal Generator</td>
<td>To apply a square wave modulation to the microwave generator and reference to the lock-in amplifiers</td>
</tr>
<tr>
<td>MW diode</td>
<td>Microwave diode to measure the reflected power from the sample</td>
</tr>
<tr>
<td>Lock-in 2</td>
<td>Lock-in amplifier measuring the reflected microwave power</td>
</tr>
<tr>
<td>Computer</td>
<td>To run LabVIEW VI's controlling the experiments</td>
</tr>
</tbody>
</table>

Table 5.1: Components used in optical setup for NV center experiments. Corresponds to figure 5.1.
5.2 Detection of Uniform Mode Ferromagnetic Resonance

For the experiments in this section we chose the well-known ferrimagnetic insulator with exceptionally low damping, yttrium iron garnet (YIG), $Y_3Fe_5O_{12}$ [19, 90]. Here an epitaxial 20 nm thick YIG film was grown on a gadolinium gallium garnet (GGG) (111) substrate by off-axis sputtering [26, 105, 106]. Continuous-wave microwave fields are applied to the sample as described in section 5.1.2 by means of a 300 nm thick and 30 µm wide silver microstrip line patterned on top of the YIG. Nanodiamonds, 50-200 nm in size (as shown in the scanning electron microscopy (SEM) image in figure 5.3) and containing up to a few thousand NV centers each, were dispersed on top of the lithographically defined microstrip line, as shown in figures 5.3 and 5.4. Atomic force microscope (AFM) measurements indicate that the nanodiamonds form about a 500 nm thick film on top of the YIG, as shown in figure 5.4.

Figure 5.3: Experimental schematic: The sample is a 20-nm-thick epitaxial YIG film with nanodiamonds dispersed on top with a thickness of about 500 nm. To apply microwaves to the sample a silver microwire is lithographically patterned on the YIG. Green laser light is focused onto the nanodiamonds near the wire, and the intensity of the resulting photoluminescence from the NV centers is measured. The inset is an SEM image of dispersed nanodiamonds showing the size range from about 50-200 nm.
5.2.1 Modification of Photoluminescence by Ferromagnetic Resonance

NV center photoluminescence was measured, using a lock-in amplifier as described in section 5.1, as a function of an applied in-plane magnetic field and the applied microwave frequency. These data are shown in figure 5.5. Data for a control sample with nanodiamonds on a GGG substrate without YIG is shown in figure 5.5(a). We observe the intrinsic and well-known magnetic resonances of the NV center ground and excited states, which originate from 2.87 and 1.43 GHz, respectively, at zero field. Shown in figure 5.5(b) are the same data overlaid with the theoretically expected resonance conditions for the NV centers. The features seen in the PL below 1.25 GHz in figure 5.5(a) and 5.5(b) occur when the harmonics from the microwave synthesizer match the ground-state resonances (see section 5.2.4 for more information).

The data obtained from the nanodiamonds on top of the YIG film are shown in figure 5.5(c). The key difference between figures 5.5(a) and 5.5(c) is the feature in (c) that extends up from the lower left-hand corner. This is highlighted in figure 5.5(d) where the data from
5.5(c) is overlaid with blue dots and a solid blue curve, showing the YIG ferromagnetic resonance condition. The blue dots show the YIG resonance conditions as measured by reflected microwave power. These data are fit using the equation for the uniform mode ferromagnetic resonance mode in a thin film [55] (see section 5.2.2), and the fit is shown as a blue solid line. From the fit we obtain a magnetization $\mu_0 M_s$ of 183 mT. As can be seen, the intensity of the photoluminescence from the NV centers strongly changes precisely when the YIG ferromagnetic resonance is excited.
Figure 5.5: NV photoluminescence data while YIG undergoes ferromagnetic resonance: (a) Raw data showing the change in the intensity of the photoluminescence from the NV centers in the nanodiamonds dispersed on top of GGG, without YIG, as a function of microwave frequency and magnetic field. (b) The same data overlaid with theoretical resonance conditions for the NV centers. The black dashed lines show the resonance conditions for an NV center with the magnetic field parallel to the NV axis for the ground and excited states. The gray crosses show the resonance condition for an NV center with the magnetic field perpendicular to the NV axis. (c) Similar data from nanodiamonds dispersed on the YIG sample with the distinct feature corresponding to the YIG ferromagnetic resonance condition. (d) The data from (c) with the NV center resonance conditions overlaid as well as data corresponding to the ferromagnetic resonance measured by using reflected microwave power (blue dots). Also shown is a fit (blue line) to the calculated dispersion relation for YIG-film ferromagnetic resonance with the magnetic field in plane.
There are two key points to note about the ferromagnetic resonance-induced feature in the NV signal. First, it is seen at frequencies and fields well separated from the NV centers’ own resonance conditions. This is in clear contrast to the quantum computing and magnetometry techniques being developed, where the spin state of NV centers is coherently manipulated by microwave fields meeting the magnetic resonance condition \[8, 10, 28, 33, 38, 50, 61, 65, 66, 68, 93, 95\]. Instead, we see a change in the photoluminescence that correlates to the excitation of the YIG magnetization into precession by means of ferromagnetic resonance. It is remarkable that excitations at energies as much as three to six times smaller than any NV center resonance have such a large effect on the NV center spin state.

Second, the ferromagnetic resonance-induced feature is comparable in amplitude to the intrinsic NV resonances. The large amplitude of the signal implies that a significant number of NV centers in our laser spot must be contributing to the signal. This suggests that since the nanodiamond film is 500 nm thick, the coupling must be either long range (extending hundreds of nanometers) or that spin transport by means of spin diffusion plays a role\[18\].

5.2.2 Simultaneous Inductive Detection of Ferromagnetic Resonance

The ferromagnetic resonance of the YIG was measured by monitoring the reflected microwave power from the microwire on the sample which was grounded at one end. We measured the reflected microwave power as we swept the magnetic field at a given microwave frequency and these data are plotted in figure 5.6(a). A typical linecut of this data is shown in figure 5.6(b), and we see a dip in the reflected microwave power as the YIG undergoes resonance and absorbs microwave power. By fitting the peak position vs. microwave frequency, shown in figure 5.6(c), to the standard Kittel equation for the uniform mode ferromagnetic resonance in the plane of a film and neglecting cubic anisotropy\[55\]:

\[
\omega_0 = \gamma \sqrt{B_0(B_0 + \mu_0 M)}
\]

We use these data to confirm that the signal seen in the NV photoluminescence correlates
Figure 5.6: FMR data: (a) Reflected microwave power measured as a function of magnetic field and microwave frequency. The microwaves were applied at approximately 35 mW. Overlaid is the fit from (c). (b) A linecut from (a) at a microwave frequency of 1.6 GHz. We see a dip when the YIG undergoes ferromagnetic resonance indicating absorption of microwave power. (c) The positions of the peaks, in field, from the reflected microwave power data are plotted against the microwave frequency and then fit to the equation for uniform mode ferromagnetic resonance. From this we obtain $M_s = 183$ mT.

with the ferromagnetic resonance in the YIG. The data and fit shown in figure 5.6(c) are the same as shown in figure 5.5(d).

### 5.2.3 Microwave and Laser Power Dependence

To test the robustness of this phenomenon, we have measured the microwave and laser power dependence of the FMR-related feature, and the data are shown in figure 5.7. The magnitude of this feature, as measured by the area under the peak, is presented in figure 5.7(b). This area was calculated for frequency sweeps (figure 5.7(a)) performed at a field of about 2.5 mT, where the ferromagnetic resonance feature is well-separated from an NV resonances. The data show that the feature can be measured in a broad window of experimental parameters and that the magnitude is almost linear in both laser and microwave
power, within our measurement range.

Figure 5.7: Power dependence of the FMR-induced feature: (a) Frequency dependence of the NV signal as a magnetic field of 2.36 mT for various microwave (top panel) and laser (bottom panel) powers. (b) The area under the ferromagnetic resonance peak (identified in (a), between the dashed lines) is shown as a function of the microwave (red dots, bottom axis) and laser (blue dots, top axis) powers. A linear guide to the eye is provided which indicates that the ferromagnetic resonance-induced feature has a fairly linear response to both laser and microwave power in our measurement range.

Also, while we have studied one spot on one sample in greater detail and are presenting those results in this section, we have seen this feature in all of the YIG samples that we have measured. These include samples grown both by sputtering and liquid phase epitaxy of several different thicknesses. Some of these results will be presented in section 5.3. We have also consistently found it over multiple spots within various samples, with varying nanodiamond density. The amplitude of the effect varies between samples and spots within
a sample, but is consistent with the variability of microwave field strength expected in these measurements. We also note that the increased amplitude of the NV excited-state peak in figure 5.5(c) as compared to figure 5.5(a) most likely results from differing microwave fields experienced in the two samples as a consequence of variability in the microwave antenna fabrication (e.g. lithography or wire contacts) and is probably not linked to the YIG ferromagnetic resonance.

5.2.4 Control Experiments

We have ruled out numerous possible explanations for the observed behavior. First, we verified that the harmonics of the microwave source, which cause low-frequency features in figure 5.5, are not causing the ferromagnetic resonance-related peak. We did this by measuring the signal with microwave low-pass filters to block higher harmonics coming from the microwave generator and show these measurements in figure 5.8. The data in figure 5.8(a) show the NV photoluminescence signal from nanodiamonds on GGG at zero magnetic field. Peaks occur, as seen in the red solid line, when the microwave synthesizer frequency is set to $2.87/n$ GHz, $n=(3, 4, 5, 6)$. These peaks are present because the synthesizer produces harmonics at 2.87 GHz, the resonance frequency of the NV ground state magnetic resonance transition. To verify this, we repeated the measurement with and without microwave low pass filters at the output of the synthesizer. As can be seen in the green and blue dashed lines, the peaks vanish when either an 800 or 1800 MHz low pass filter is in place. This confirms that the peaks seen are indeed caused by harmonics from our synthesizer.

Shown in figure 5.8(b) is NV photoluminescence data from nanodiamonds on YIG with and without the 800 MHz microwave low pass filter. Seen in the red solid line is the ferromagnetic resonance-induced feature along with several harmonic peaks. The green dashed line shows that when an 800 MHz low pass filter is in place the harmonic peaks disappear, but the ferromagnetic resonance peak is still present, thus confirming that the ferromagnetic resonance-induced feature is not related to the harmonics from the synthesizer.

We have also considered the effect of heating, caused by the absorption of microwaves by
the YIG on resonance, on the NV center photoluminescence. Several control experiments and estimates of the possible effect render this potential explanation for the ferromagnetic resonance-induced feature highly unlikely. The temperature dependence of the NV center photoluminescence intensity in nanodiamonds have been reported to be 0.2% per K [78]. Here we will estimate the change in the photoluminescence that can result from the heating due to microwave absorption on ferromagnetic resonance. It is difficult to theoretically estimate the temperature change of the nanodiamonds, even with a known power absorption on ferromagnetic resonance, due to the uncertainties in estimating the thermal resistances at the various material interfaces. However, temperature change on ferromagnetic resonance in YIG has been measured by An et. al [4], and we will use their measurements to estimate the temperature change in our experiments. We will compute the upper limit of the effect by assuming the nanodiamond temperature tracks exactly the instantaneous YIG temperature.

Since we are measuring a lock-in signal, we are interested in determining the temperature change on ferromagnetic resonance during a half-cycle of our modulation period when the microwaves are on. If heating was the cause of the signal we measure, then the photoluminescence change due to this temperature change would approximately correspond to
the lock-in signal we measure when the YIG is on ferromagnetic resonance. We will assume
that the change in temperature is related to the experimental parameters in the following
manner:

\[ \Delta T_{FMR} \propto E_{abs} \propto H_1^2 t \tau \propto \frac{P_{in}}{f} t \tau, \quad (5.2) \]

where \( \Delta T_{FMR} \) is the change of temperature due to microwave abosorption on ferromag-
netic resonance, \( E_{abs} \) is the energy absorbed per unit area, \( H_1 \) is the microwave magnetic
field, \( t \) is the thickness of the YIG, \( \tau \) is the time the microwaves are on, \( P_{in} \) is the microwave
power into the sample, and \( f \) is a factor that is related to the decrease in the strength of
\( H_1 \) due to being a certain distance away from the microwave antenna.

Table 5.2 gives a comparison of our experimental parameters with those in An et. al [4].
Based on these values and their measured change of 10 K/s on ferromagnetic resonance, we
estimate temperature changes on ferromagnetic resonance smaller than 100 \( \mu K \) and changes
in photoluminescence too small to be measured in our experiment.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Our Experiment</th>
<th>An et. al</th>
</tr>
</thead>
<tbody>
<tr>
<td>( P_{in} ) (mW)</td>
<td>35</td>
<td>230</td>
</tr>
<tr>
<td>( f )</td>
<td>1</td>
<td>10</td>
</tr>
<tr>
<td>( \nu ) (nm)</td>
<td>20</td>
<td>30000</td>
</tr>
<tr>
<td>( t ) (ms)</td>
<td>3.5</td>
<td>1000</td>
</tr>
</tbody>
</table>

Table 5.2: The parameters needed to estimate the temperature change in the YIG when
absorbing microwaves on ferromagnetic resonance for a time \( t \) using equation 5.2. The
center column shows the parameters relevent for our experiments. The right column shows
the parameters used by An et. al [4].

The result of the analysis above is consistent with our control experiments. First, we
used Joule heating of the microwire to induce temperature changes in the nanodiamonds
which were on GGG. The heating power was modulated between 0 and 3 mW at 150
Hz, the same frequency as was used for the data in figure 5.5. Based on our reflected
microwave power data, we expect the power absorbed on ferromagnetic resonance to be
more than 2 orders of magnitude smaller than 3 mW. The measured change in the NV photoluminescence signal due to Joule heating, even at 3 mW, is 2 orders of magnitude smaller than the ferromagnetic resonance-induced peak seen in our data in figure 5.5(c).

We performed a second control experiment to investigate the role heating may play in our signal. We measured the ferromagnetic resonance-induced peak as a function of modulation frequency of the microwaves. We varied the modulation frequency from 15 Hz up to 5 kHz, the cutoff frequency of our preamplifier. Over this range the ferromagnetic resonance-induced feature behaves exactly the same as the NV ground state peak and does not have a strong dependence on modulation frequency, as shown in figure 5.9. The slight reduction in amplitude with frequency is consistent with the specifications of the preamplifier. These data show that if the effect is thermally driven then it has a time constant much faster than 5 kHz and would be inconsistent with the data from An et. al [4]. Our analysis and control experiments together show that heating does not play a significant part in our measured signal.

![Graph](image_url)

Figure 5.9: Thermal Effect Measurements: Peaks in the optical signal, at the YIG ferromagnetic resonance condition and the NV center resonance condition, are plotted vs. the frequency of the microwave amplitude modulation. These peaks are normalized to their values at 15 Hz. The red circles represent the amplitude of the peaks induced by the YIG ferromagnetic resonance, and the green triangles represent the amplitude of the NV center resonance peaks. We attribute the drop in the signal with frequency to the response of our preamplifier.
The linear response to microwave power rules out nonlinear effects such as second harmonic generation in the YIG, resulting in microwaves at the NV resonances, as a likely cause. It is unlikely that the YIG resonance emits microwaves strong enough to cause NV spin excitations at the far tails of the NV resonance Lorentzian absorption spectrum. This would require microwave fields that are 20 times stronger than those applied with the stripline in order to obtain the observed signal magnitude at 0.5 GHz. Such enhancement is unlikely and should result in a much larger signal at the intersection of the NV excited state with the ferromagnetic resonance condition. Furthermore, the NV signal away from resonance is below the noise at all microwave powers, even in the GGG sample, indicating the difficulty in exciting these Lorentzian tails. The ferromagnetic resonance-induced feature seems to be resulting from an as-yet unrecognized mechanism for transferring angular momentum between ferromagnets and NV centers. More discussion on the mechanism causing this effect can be found in section 5.4.

5.2.5 Long Range Coupling

To probe the spatial extent of the coupling, we repeated the measurement with our laser spot focused on the nanodiamonds on top of the microstrip line, where the nanodiamonds are separated from the YIG by more than 300 nm. This data can be seen in figure 5.10(a) and compared to the signal when the nanodiamonds are in direct contact with the YIG in figure 5.10(b). The NV excited state resonance is more pronounced in 5.10(a) probably because the microwave field is stronger at the position on top of the wire. Linecuts showing the frequency dependence of the NV signal at 16.5 mT are shown in figure 5.10(c). These were normalized to make the amplitude of the NV resonance the same in the two. These show that the ferromagnetic resonance-induced feature is reduced relative to the NV peak, but clearly persists even when the nanodiamonds are not in direct contact with, and in fact 300 nm away from the YIG.
Figure 5.10: NV center photoluminescence data on YIG and on the wire above YIG: (a) Data taken with the laser spot focused on top of the 300 nm thick patterned microwire. The data show that the signal corresponding to the YIG ferromagnetic resonance is still present, though reduced. The coupling must extend at least 300 nm through the wire. (b) Data from nanodiamonds directly on top of YIG, for comparison. (c) Line cuts of the data in (a) and (b) at 16.5 mT. The ferromagnetic resonance-induced peak is reduced by three times relative to the NV excited state resonance peak for the case of nanodiamonds on top of the microstrip, compared to the case directly on YIG.
5.3 Detection of Ferromagnetic Dynamics: Spinwaves and Domain Dynamics

Many spintronic [6, 35, 108, 113] and magnonic [57, 90] devices rely on the dynamics of ferromagnetic elements and understanding these dynamics at the nanoscale is crucial for their development. It will be important to understand angular momentum transfer across interfaces, spin wave propagation in low dimensional and nanoscale systems, domain wall motion, microwave driven switching, relaxation, and damping in small structures. There is also much interest in materials and systems which extend beyond the ferromagnetic phase and into magnetic textures such as the conical or helical phases and the so called skyrmion phase. However there is a need for new techniques which can study the dynamics in these systems spectroscopically and to do this at the nanoscale. Electrical detection schemes have been used for studying some of these phenomena, for example domain wall motion, but these schemes do not have imaging capabilities. Optical techniques, such as Brillouin light scattering, while very powerful, are restricted in their imaging resolution by the optical diffraction limit.

Our technique of using NV centers in diamond to detect ferromagnetic dynamics provides us with a very promising tool for nanoscale, spectroscopically sensitive detection of magnetic excitations. In this section I will show how we use the same technique, the first demonstration of which was shown in section 5.2, to detect the rich spectrum of excitations in a 5 µm thick YIG film. We again see uniform mode ferromagnetic resonance, but now we also measure a band of high-k dipolar-exchange spin waves as well as dynamics associated with the multi-domain state of the YIG. We also see that this technique is sensitive to the local density of magnetic excitations highlighting the potential for this technique to be extended to nanoscale imaging of magnetic dynamics.

5.3.1 Experimental Setup

For the experiments in this section we used a 5 µm thick yttrium iron garnet (YIG) film grown on a gadolinium gallium garnet (GGG) substrate by liquid phase epitaxy (LPE). A
microwave field is applied to the sample by means of a 400 µm wide, 300 nm thick gold microwire which was lithographically patterned on top of the YIG. Nanodiamonds (the same as shown in figure 5.3) and containing up to a few thousand NV centers each, were dispersed on top of the sample. In the center of the microwire was left a small gap 25 µm wide and 200 µm long, which allows for nanodiamonds to be in contact with the YIG in a position where the microwave field, $H_1$, was parallel to the static magnetic field, $H_0$, shown as position 1 in figure 5.11. This sample also allowed for the measurement of NV centers in positions where $H_1$ was perpendicular to $H_0$, as in positions 2 and 3 in figure 5.11.

Figure 5.11: Experimental schematic: Experiments were performed on a 5 m thick YIG film with a lithographically defined microwave antenna. Nanodiamonds were dispersed on top and were in contact with the YIG. Changes in the luminescence of the NV centers in the nanodiamonds were recorded as a function of the static magnetic field, $H_0$ (large red arrow) and the frequency of the microwave field, $H_1$ (elliptical green arrow), at various locations on the sample. Positions 1, 2, and 3 indicate the locations of the focal spot where the NV signal was measured and correspond to panels (c), (d) and (e) respectively of figure 5.12. Magnetization dynamics in the ferromagnet were measured simultaneously with the luminescence by monitoring the reflected microwave power from the microwire on a diode. This reflection signal is spatially averaged over the entire sample in contrast with the NV optical signal which measures local dynamics.
Photoluminescence is excited in the NV centers using either a 520 or 532 nm laser focused down to a $< 2 \mu m$ spot, and is collected by a photodiode. A lock-in measurement is performed by modulating the amplitude of $H_1$ in the way described in section 5.1. The signal is normalized by the overall intensity of the photoluminescence to allow for the comparison of data from different positions on the sample. We also measure a lock-in signal of the microwave power reflected from the shorted microwire. This gives us a measure of the absorption of power by the YIG, indicating excitation of ferromagnetic dynamics. The reflected microwave power is a measure of the dynamics over the entire sample, while the NV signal is sensitive to magnetic excitation only in the region near the laser spot.

5.3.2 Broadband Spectroscopy

The measurement is performed as a function of the magnitude of the static magnetic field, $H_0$, and the frequency, $\omega$, of the microwave field, and the data is presented in figure 5.12. In these data we see that the photoluminescence signal exhibits strong features associated with excitations in the YIG across a broad range of fields and frequencies. Figure 5.12(a) shows the magnetic field dependence of the NV photoluminescence signal as well as the reflected microwave power with a microwave frequency of 1.8 GHz. These data highlight the different excitations we have been able to measure. At around 20 mT we see the uniform mode ferromagnetic resonance, and at lower field, around 12 mT, we see a feature we associate with band of high-k dipolar-exchange spinwaves. Around zero field we see the largest of the features, which occurs when the YIG is in a multi-domain state.

Also evident in figure 5.12(a) is the high signal-to-noise ratio we are able to achieve in these measurements using a modest lock-in time constant of 500 ms and no other averaging. Shown in figure 5.12(b), (c) and (d) is the NV signal measured over frequencies from 0.1-3.7 GHz and fields from -25 to 25 mT. This shows that we can measure these signals over a broad range of fields and frequencies. This is especially of note because there does not need to be any overlap with the NV centers own resonance conditions, which in this case is a powder spectrum the extrema of which are shown by green lines.
Figure 5.12: Broadband and localized spectroscopy of YIG using an ensemble of NV centers in nanodiamonds: (a) Spatially localized optical NV signal at three positions (blue lines, left axis) and global reflected microwave power (solid black line, right axis) as a function of field $H_0$ at a microwave frequency of 1.8 GHz. (b, c, d) Field-frequency 2D maps of the broadband dependence of NV PL as a function of field $H_0$ and frequency of $H_1$ for positions 1, 2 and 3 (see Fig. 1), respectively. The green lines indicate the extrema of the powder spectra of the NV ground (upper two lines) and excited state (bottom two lines) magnetic resonances. Note the low field features, below about 4 mT, which indicate sensitivity to dynamics associated with domains in unsaturated YIG. (e) 2D map of the reflected microwave power as a function of field $H_0$ and frequency of $H_1$. The superposed red lines show the calculated dispersions for several spinwave branches of this YIG film.
5.3.3 Local Spectroscopy

Another important feature of these data is that the NV signal is sensitive to the position of the laser spot on the YIG sample. Particularly, the NV signal is sensitive to the local microwave environment in the $< 10\mu m^2$ laser spot and the magnetic dynamics excited most efficiently in that area. This can be seen clearly in figure 5.12(a) when comparing the NV signal at position 1, 2, and 3 to each other and with the global reflected microwave signal. Position 1 (blue dashed line), in the center of the wire, shows the strongest signal from the multi-domain state and also seems to match mostly closely to the global reflected microwave signal. This is probably because the reflected microwave signal will be dominated by the region under the wire where most of the microwave power is absorbed. Position 2 (dotted light blue line), at the edge of the wire, shows a smaller signal from the multi-domain state but a larger signal from the high-k spinwaves. This is due to the fact that the spinwaves are most efficiently excited in this region of the sample where $H_1$ has the largest amplitude and is also inhomogeneous.

Position 3 (solid purple line), 500 $\mu m$ from the edge of the wire, still exhibits all three features (uniform mode FMR, spinwave band, and domain dynamics), but interestingly the spinwave band seems to be shifted to higher field. This is most likely because the spinwaves are not excited at position 3, but at the edge of the wire. They then propagate across the sample to position 3. The field is shifted because the spinwaves at the center of the band have small group velocity, while the ones at higher field have a larger group velocity and can thus travel the 500 $\mu m$ before decaying.

The signal arising from dynamics in the multi-domain state of the YIG may be of special significance. Domain wall motion is currently a field of intense interest and technological promise. The fact that we see such strong signals from dynamics of this state presents a promising potential avenue for optically reading out domain wall motion.
5.3.4 High Field Spectroscopy

We were also able to measure this NV signal at higher field and frequency than is shown in figure 5.12. Figure 5.13 shows the evolution of the ferromagnetic spectrum up to 6.55 GHz and 180 mT. At 6.55 GHz (blue line) we see the uniform mode at 165 mT. This data is of note because the field is much higher than is typically used in magnetometry experiments with nitrogen vacancy centers. We are not sure if there is a limit to the field or frequency at which this effect will be present. 180 mT is near the limit of our current experimental setup. These data highlight the potential versatility of this effect to be used as a tool to study magnetic dynamics over a broad range of fields and frequencies.

Figure 5.13: Measurement of ferromagnetic dynamics using NV centers in magnetic fields exceeding those typically used for conventional ODMR magnetometry: NV optical signal as a function of magnetic field at various microwave frequencies, showing the evolution of the ferromagnet spectrum.
5.4 Mechanism of NV-Ferromagnetic Coupling

While the exact mechanism behind the signals we see in the NV photoluminescence is still unknown, there is a lot of work currently underway to learn what it is. Meanwhile, we can point out a few possibilities and what makes them more or less likely.

First, there are several effects which are highly unlikely to be the cause of the effect. Temperature has been shown to affect NV center photoluminescence and the energy levels of the spin sublevels [78, 99], and the heating of the YIG on resonance was considered as a cause for the signal we measure. This is unlikely for several reasons, which were discussed in section 5.2.4. Also, the insulating nature of both materials rules out long-range carrier-mediated transport of spins.

One explanation could be that the YIG acts as a microwave field amplifier. This idea has several problems, however. The signal persists over a wide range of field and frequency, and this explanation would only cause transitions in the NV centers when the YIG resonance was near the NV resonance. While the YIG could enhance the microwave field, it is unlikely that it would enhance it enough to cause the signal at 500 MHz, for example. Also, if this were the case, when the YIG resonance conditions matched that of an NV center transition, there should be a much larger effect than we observe.

An interesting feature of these experiments is that both the YIG magnetization and the NV center spins are out of equilibrium. The YIG is being driven by the microwaves and the NV centers are being optically pumped by the green laser. This is in contrast to other systems where angular momentum is transferred between spin subsystems, e.g. dynamic nuclear polarization or FMR spin pumping [69, 91, 100], where one spin subsystem relaxes by transferring polarization to another spin subsystem. The transfer of angular momentum between the YIG and the NV centers could result in relaxation towards equilibrium for both spin systems, and thus be highly desirable for the total system.

In this vein we have considered effects which can couple spin systems. The spins seem to be too far away (several hundred nanometers) for exchange coupling to play a large role, but dipole coupling could. Dipolar induced transitions in NV centers have been considered
in the literature [70, 104]. Work is being done to calculate transition rates for an NV center coupled to the YIG. However, this possible mechanism does not seem to explain everything about our signal. For example, it does not explain the seemingly small frequency/field dependence of the size of the effect. The amplitude of the measured signal stays relatively constant over our measurement range, whereas this effect should be enhanced at specific places in the spectrum, namely level crossings.

Regardless of the mechanism, this phenomena offers an opportunity to probe spin dynamics in ferromagnetic materials and heterostructures, and with the possibility of ultra-high-resolution imaging.
Chapter 6

Conclusion

In this dissertation I discussed three novel techniques for spin-sensitive imaging in condensed matter systems. The technique of scanned spin-precession microscopy combines the power of using magnetic field gradients for imaging with, in principle, any spin-sensitive detection technique. This is achieved by combining sensitive spatially-averaged measurement techniques with high magnetic field gradients provided by micromagnetic particles. By using a detailed understanding of the behavior of spins in inhomogeneous magnetic fields we have shown that local spin information can be extracted. Scanned spin-precession microscopy allows for the imaging of spins in buried interfaces and with spatial resolution only limited by the magnetic field gradient.

The second technique shown was solid-state Hanle magnetometry. This was shown to be capable of imaging the vector components of a magnetic field in a solid-state system. While the magnetic field sensitivity is good, it is not near the limits of other magnetometers. It could, however, find a niche in studying the local magnetic fields in spintronic devices. This would be achieved by using the spins in the devices themselves as the magnetometer, where other magnetometers would not have have access.

The third technique shown was the off-resonant detection of magnetic dynamics with nitrogen vacancy centers in diamond. We showed that nitrogen vacancy centers can be used as broadband and local sensors of magnetic dynamics in adjacent materials. We specifically showed sensitivity to uniform mode ferromagnetic resonance, spin waves, and domain dynamics in yttrium iron garnet. This technique is very promising because of its
versatility and because of the proven ability to measure single NV centers, which could push this technique into the nanoscale.

These three techniques represent several small steps forward in the understanding of the behavior of spin in condensed matter systems as well as providing new ways in which magnetism can be studied. They are all well suited to studying systems relevant to the field of spintronics and have potential for spatial resolution into the nanoscale.
REFERENCES


pages 1 and 10.


92(7). Cited on pages 5, 18, and 22.


51.


Appendix A

GaAs Membrane Samples

This appendix provides details of the structure and preparation of samples used for scanned spin-precession microscopy and solid state Hanle magnetometry. These were 1 µm thick membranes of n-GaAs (100) which were attached to a piece of sapphire. We started with metal organic chemical vapor deposition (MOCVD) grown samples with a structure as pictured in figure A.1(a), where the top layer becomes the membrane. An approximately 3×3 mm piece of this multi-layer structure is first glued to a 5×5 mm single crystal (0001)-sapphire substrate using Epotek 301-2 optical grade epoxy to provide support for the GaAs membrane and allow optical studies to be performed on the back of the membrane through the sapphire. The c-axis orientation of the sapphire was chosen to minimize birefringence during polarization-dependent optical measurements. The Epotek 301-2 epoxy was chosen to reduce any background photoluminescence. The GaAs substrate is then etched away with a citric acid-hydrogen peroxide-water solution as detailed by Otsubo et al [76]. The nominally lattice matched InGaP layer is grown between the top GaAs layer and the substrate to act as a stop-etch layer due to the high selectivity of the aforementioned etching procedure. The InGaP is then etched in HCl for approximately one minute. We are left with the 1 µm GaAs membrane on sapphire as pictured in figure A.1(b).

For the first demonstration of scanned spin-precession microscopy and for solid state Hanle magnetometry of a micro-magnetic particle, we then glued a NdFeB particle with diameters from 1-30 µm directly to the GaAs membranes using the same Epotek 301-2 optical epoxy. This can be seen in figure A.2. We first made several scratches in the GaAs
membrane using a needle attached to a micro-manipulator. We then use the needle to place small drops of glue in the locations between the scratches. NdFeB micromagnetic particles were then be picked up, using static forces, with another needle and placed on the drops of glue. This is all performed using micro-manipulators under an optical microscope with long working distance objective lenses. The glue is then cured in an oven, and the particles are magnetized out of the plane in a 2 T magnetic field.

Figure A.2: (a) Optical image of NdFeB micromagnetic particles glued to a GaAs membrane. Lines are scratched into the GaAs to aid in locating the particles. (b) An SEM image of the same NdFeB particles for size calibration.
Appendix B

Cantilevers with Magnetic Particles

To create the cantilevers we used in these experiments we used spherical NdFeB particles. These created magnetic field profiles that were very close to an ideal dipole. We glued the particles, typically with Epotek 301-2 epoxy, to cantilevers which were either bought without tips (AppNano SICONG-TL tipless cantilevers) or with tips which were removed using a focused ion beam.

This process is shown in figure B.1. We take a glass slide and put a small amount of epoxy on one side and a small number of NdFeB particles on the other. We then attach a cantilever to the end of one micromanipulator and a needle to another micromanipulator. The needle is dipped into the epoxy, so that just a small amount is on the tip, as see in figure B.1(a). The needle is then carefully touched to the tip of the cantilever so that a small amount of glue come off, as in figure B.1(b). A particle is chosen from the slide and is then picked up with the needle and carefully attached to the cantilever. Figure B.1(c) and (d) shows the cantilever after the particle is placed with the focus on the cantilever or particle, respectively. The cantilever is then carefully placed in an oven for the epoxy to cure. A sideview of the cantilever with particle is shown in figure B.1(e). And finally, figure B.1(f) shows the cantilever mounted in our microscope with an optical fiber brought to within approximately 50 μm. This is to perform interferometry for monitoring the cantilever position.

When peforming scanned spin-precession microscopy it is useful to know the properties
of the magnetic particle. To this end, we perform magnetometry on the cantilever by placing it in a magnet with the moment of the particle aligned with the field. By monitoring the oscillation frequency of the cantilever as we sweep the magnetic field we can determine the moment of the micromagnetic particle. A typical measurement is shown in figure B.2. By fitting to the equation shown we determine that the magnetic moment of the particle in this measurement is $4.576 \times 10^{-9}$ J/T. We know that NdFeB has a moment density of $1 \times 10^{-12}$ J/T/$\mu m^2$, and using this we confirm the size of the particle, in this case a radius of 10.299 $\mu m$. 

Figure B.1: (a) Needle with epoxy, attached to a micro-manipulator, and near a bare cantilever. (b) Cantilever after a small amount of glue has been applied to the tip with the needle. (c) and (d) The cantilever after a NdFeB particle has been placed on the tip, focused on the cantilever and particle, respectively. (e) A sideview of the cantilever after the epoxy has been cured. (f) The cantilever mounted in the microscope with optical fiber to perform interferometry.
Figure B.2: Cantilever magnetometry performed by monitoring the frequency of the cantilever oscillation as a function of applied magnetic field. By fitting to the equation shown we obtain a magnetic moment of $4.576 \times 10^{-9} \text{ J/T}$. 

$$\frac{\Delta f}{f_0} = \frac{\Delta k}{2k_0} = \frac{m}{2k_0 L_e^2} \Delta B$$