Gallium Nitride and Aluminum Gallium Nitride Heterojunctions for Electronic Spin Injection and Magnetic Gadolinium Doping

THESIS

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By

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Abstract

As the power requirements of computation for large scale and mobile applications increase and the fundamental limits of current technology are being approached, the development of novel computational schema is required to continue the meteoric improvements in processing. One such schema, spintronics, relies on manipulation of the magnetic state of electrons in computation. Such approaches to computation would be facilitated by the development of high-quality ferromagnetic semiconductors capable of room temperature operation and by controlled injection of magnetically polarized electrons into conventional semiconductors. The first part of this work explores the utility of gadolinium doped GaN/AlGaN heterojunctions as a magnetic semiconductor system. The extremely large magnetization per magnetic dopant atom observed in gadolinium doped gallium nitride is also observed in some, but not all, of these heterojunctions. The second part of this work involves the fabrication of spin valve devices on undoped GaN/AlGaN heterojunctions. Such devices present one potential avenue for the control of magnetically polarized current within conventional semiconductor systems.
Dedication

This thesis is dedicated to my wife, Gail Clendenin, for her support and love.

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I want to acknowledge my advisors, Roberto Correa Myers and Ezekiel Johnston-Halperin for their passion for the pursuit of knowledge, for their patience in guiding students, and for their continual insistence on performing the best research.
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Fields of Study

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Chapter 1
Introduction

1.1 The miniaturization of charge-based transistors

For decades the number of transistors on computing chips has doubled approximately every two years, a remarkable trend popularly known as Moore's Law. The semiconductor industry has maintained this remarkable pace of advancement largely by shrinking transistor sizes. This trend may continue in the near future; Intel expects to continue increasing transistor density by shrinking transistors to 10 nm by 2015 [1]. However, the miniaturization of the charge-based transistors will soon become impractical because of fundamental limits imposed by quantum effects, which become increasingly important as the size of transistors drops. Take a field effect transistor (FET) as a concrete example. The off state of a FET corresponds to a state wherein carriers are confined to the source electrode by the energetic barrier of the gate voltage. As devices shrink, the barrier width decreases. Since tunneling rates exponentially increase with decreasing width, the probability of electrons tunneling through the barrier increases extremely rapidly as transistor size decreases. When the tunneling probability becomes
high enough, enough electrons tunnel under the barrier to provide a current to the drain which registers as the on state. This breakdown of the transistor functionality can be avoided by increasing the voltage to provide an arbitrarily high tunnel barrier, but increasing barrier heights introduces new problems. In order to switch from the off state to the on state or vice versa, the gate must be charged or uncharged. The electrical energy associated with switching the state of the barrier will be dissipated as heat. As gate lengths shrink and gate voltages rise, transistors start to generate heat sources strong enough to melt themselves. Thus the interrelated problems of increased gate leakage and heat dissipation as the size of transistors shrink impose a fundamental and rapidly approaching limit on the size of transistor fabricated using traditional complementary metal-oxide semiconductor (CMOS) technology.

The energy dissipated during computation is also increasingly important for mobile device applications and because of environmental concerns. Although the amount of energy used to power computer systems is difficult to calculate, estimates of the power used to power personal computers, server systems, and computing networks are approximately 2% of the global energy consumption and global carbon emissions[2, 3]. Since the amount of computation performed is projected to dramatically increase in the near future, even small increases in the energy efficiency of computing could dramatically decrease the carbon dioxide and other toxins emitted to generate power for computing systems. Lowering the power required to manipulate and store data would also have a profound influence on the mobile devices industry, enabling the development of mobile devices with longer battery lives and improved performance.
1.2 The potential of spin-based logic

To continue the dramatic increase in the performance of computer systems beyond the near term requires that the CMOS transistor be replaced by a fundamentally distinct computing method[4-7]. Among the many approaches proposed is a class of computational devices which manipulates intrinsic angular momentum, or spin, instead of charge. Such devices are termed spintronics. The energy theoretically required to perform spin-based computation is significantly lower than the energies required to move charge in transistors[8-10]. Thus the fundamental limit on the energy dissipation by spintronic computational devices is lower than that of current charge-based computers.

Spintronic computation also enjoys the advantage of being able to utilize materials deposition and fabrication techniques developed by the CMOS industry. Indeed spintronics already serve a vital role in data storage; spin valve memory devices are already standard technology in current hard drives. To extend the application of spintronics to logic, a number of spin-transistors[5, 11] and integrated logic-memory devices have been proposed[10]. Important milestones in spin-based logic have already been reached: all-electronic spin injection and detection at room temperature[12-16] in metals and semiconductors and the fabrication of prototype spin based logic devices[17, 18]. Because of the similarities between the existing and highly successful CMOS technologies and semiconductor spintronics, spin-based logic device prototypes could likely be commercialized with much less investment in research and development than other proposed computational schema such as biological or entanglement approaches.
1.3 Current challenges in spintronics

The commercial adaptation of spin logic devices may proceed only after basic technical hurdles are overcome. One challenge is preserving the information during computation and data storage, requiring the use of spin lifetimes longer than the operation and read-out time of the device. In order to integrate new spintronic devices within the existing CMOS framework, current or voltage inputs, controls, and outputs like those used in CMOS architectures should also be used in spintronic devices to induce a non-equilibrium spin-polarization within the device, manipulate that spin polarization, and detect the final spin state[4]. All-electronic manipulation of spin ensembles is challenging, and much of the foundational work on spin lifetimes and spin manipulation in semiconductors was done using optical techniques, such as magneto-optical Kerr-effect (MOKE) measurements[19] and spin-LEDs[6, 20-22]. One of the chief challenges of all-electronic spin logic is the low spin polarization of electron ensembles injected directly from ferromagnetic metals into semiconductors. The injected spin polarization can be increased by using tunnel barrier contacts between the ferromagnetic metal and semiconductor, but such tunnel barriers are prone to leakage and surface states which degrade the injected spin polarization. This problem could be entirely avoided by the use of high-quality room temperature ferromagnetic semiconductors, an area of intensive research today.
1.4 Nitride heterostructures as a materials system

In conventional electronic devices, III-N heterostructures are becoming increasingly important for high power and high frequency applications through high electron mobility transistors (HEMTs)[23]. Nitride HEMTs are based on the GaN/AlGaN heterojunction. When AlGaN is deposited on top of GaN during gallium-polar growth, a sheet of electrons builds up at the interface due to the difference in the unit cell polarizations and the interfacial strain. The structure is referred to as a two-dimensional electron gas (2DEG). A similar heterostructures can be created by depositing GaN on top of AlGaN during gallium-polar growth. By inverting the sign of the polarization difference across the interface, the charge of the carrier sheet at the interface is also inverted and a hole gas is formed at the interface in this two-dimensional hole gas (2DHG) structure.

Nitride-based heterojunctions provide materials improved carrier mobilities, breakdown fields, and drift velocities compared to impurity-doped bulk semiconductors. The carrier mobilities are higher because doping due to the difference in polarization of the unit cells results in a conduction charge which is spatially isolated from its source, whereas in impurity-doped semiconductors the impurity ions serve as scattering centers for carriers. The lower scattering rates of 2DEG and 2DHG heterojunctions often result in much higher carrier mobilities than typical for impurity-doped bulk semiconductors. The breakdown fields are higher because the band gaps of the gallium nitride and aluminum nitride are significantly larger than the band gaps of commonly used impurity-doped semiconductors. For example, the band gap of gallium nitride is 3.2 eV and the
band gap of aluminum nitride is 6.4 eV and both are much larger than silicon's band gap of 1.1 eV. This allows for larger electric fields to be applied to GaN and AlN before electronic breakdown occurs. The combination of high mobilities and large fields permit very large electron drift velocities. Large drift velocities enable high power and radio frequency operation of HEMTs. Although the growth and processing of high-quality nitride films remains a significant technological challenge, the materials system offers superior intrinsic materials properties as compared with silicon. For this reason, the demonstration of all-electronic spin injection and detection in this material could foster spintronic logic development.

1.5 Gd-doped III-nitride heterostructures

In the first research project discussed in this thesis, the effects of gadolinium doping on GaN/AlGaN two-dimensional electron gas (2DEG) structures on the magnetic properties of the quantum wells is explored. Previous work has shown that gadolinium doped GaN becomes ferromagnetic, and that the magnetism is much stronger than one would expect from complete magnetization of the gadolinium dopants. Since a robust ferromagnetic semiconductor system could be used in spin-gain devices for spintronic logic and to bypass the resistance mismatch problem in electrical spin injection, the magnetic properties of the gadolinium doped GaN/AlGaN heterojunction are of technological interest. In this project, 2DEG structures were grown with varied distribution of gadolinium dopants along the growth direction and the morphology, electronic transport, and magnetic properties of these heterjunctions were studied.
1.6 All-electronic spin injection and detection in GaN/AlGaN heterojunctions

The ability to induce, manipulate, and detect a non-equilibrium net magnetization into a materials system is foundational to spintronic logic devices. A number of methods, including optical, magnetic resonance, and electronic techniques have been used for this process of spin injection, manipulation, and detection. Of these methods, all-electrical techniques offer the best compatibility with existing CMOS technologies. With this motivation, the spin injection and detection project seeks to demonstrate all-electronic spin injection and detection in GaN/AlGaN heterojunctions. In this project, 2DEG structures are modeled and grown specifically for spin injection and detection. The morphology and electronic properties of the 2DEGs characterized, and spin injection/detection devices are fabricated from the nitride thin films.
2.1 Gadolinium doped gallium nitride

The operation of some spintronics logic devices requires a room temperature magnetic semiconductor with controllable carrier concentrations, so the development of a class of materials known as dilute magnetic semiconductors (DMS) is pivotal for the development of spintronic logic. These semiconductors become ferromagnetic after low concentration doping; their ferromagnetic and semiconducting properties arise from distinct mechanisms. Among dilute magnetic semiconductors, gallium manganese arsenide has been extensively studied. In GaMnAs the hole concentration controls the magnetization of the DMS[24]. Thus the magnetism of the sample can be adjusted by varying a gate voltage applied to the material. Unfortunately, its Currie temperature is well below room temperature, making it an impractical material for technological applications.

The dependence of the saturation magnetization of GaMnAs on the hole concentration in the sample implies that material's ferromagnetism results from the
interplay between the magnetic moment of the rare earth dopants' magnetic moment with the surrounding carriers. This may be modeled as a polarization effect of the dopants on the surrounding carriers. In this model the effective range and magnitude of the dopants' ability to polarize carriers, the density of dopants, and the polarizability of the surrounding matrix of magnetic moments determines the overall magnetization density of the material. This mechanism of ferromagnetism allows for control of ferromagnetic state of the material both during materials deposition and during device operation. During deposition, the crystalline quality of the material and doping concentration can be varied to change the polarizability and density of polarizing dopants, respectively. During operation, the carrier concentration may be modified through the gate voltage as in standard MOSFET transistor operation. This control is technologically useful.

The search for room temperature DMS materials advanced in 2002 with the report of ferromagnetism in gadolinium doped gallium nitride layers grown by molecular beam epitaxy (MBE)[25]. In 2005 ferromagnetic MBE-grown Gd:GaN films with magnetic moments of up to 4000 Bohr magnetons ($\mu_B$) per dopant atom were reported[26]. However, their extraordinary report of ferromagnetism could not be explained by the same mechanism as for GaMnAs, due to the more complex variation of the saturation magnetization of the films. Further investigations GaGdN showed that the saturation magnetization may range from ~100 $\mu_B$ to ~1000 $\mu_B$. Studies of ion-implanted films showed that at the same Gd-doping density, gadolinium ion-implanted films show stronger ferromagnetism than films grown by MBE. Also, films co-implanted with silicon and gadolinium show a greater saturation magnetizations than those implanted
only with gadolinium[27]. One possible explanation for the variation in the saturation magnetization is that the magnetization is mediated through defects. Then the silicon doping results can be understood as an increase in the number of defects, leading to increased magnetization. This also explains why MBE-grown films exhibit lower saturation magnetizations than for ion-implanted films; the energetic ions induce more defects than are produced during incorporation of gadolinium into the epitaxially grown films. Finally, a defect-induced mechanism for magnetization could explain the variations in the saturation magnetizations of films grown by different group as different growth procedures can dramatically affect the quality of deposited material.

2.2 Foundational concepts in spin physics

Before addressing the technological challenges faced in using spin as a computational variable, some basic concepts and definitions are developed. First a quantitative measure of the spin polarization of a system is defined. Then the decay of spin ensembles in a solid state material is discussed. Finally, the electronic method of spin polarization is discussed at a basic theoretical level.

Spin Polarization

The efficiency of spin injection into a material can be quantified by defining a spin polarization of the spins in a system or a current entering it. Because many possible definitions of spin polarization exist, care should be taken in interpreting this term in the literature. A general definition of the spin polarization, \( P_{\uparrow} \), is
where $X^\uparrow$ measures the number of spin up particles or current and $X^\downarrow$ measures the number of spin down particles or current. In a common definition of the spin polarization, the variable $X^\uparrow(\downarrow)$ stands for the total number of charges with a spin up (down) magnetic state. This definition measures the spin accumulation density a collection of charges and is sometimes referred to as the density spin polarization. If the spin polarization of the current is of more interest in a system, a spin polarization of the charge current may be defined where $X^\uparrow(\downarrow)$ stands for the spin up (down) current density, defining a spin polarization of the charge current, sometimes referred to as the current spin polarization. Even in the absence of net charge flow, a spin polarization may be defined where the $X^\uparrow(\downarrow)$ stands for the spin up (down) current moving in a particular direction plus the spin down (up) current moving in the opposite direction. Such a definition may be useful in a case where charges inter-diffuse between spin polarized and spin un-polarized material regions. As a further complication, the system under consideration must be carefully defined by determining its size and scope. Thus defining the spin polarization requires two choices of convention for implementation to any given system: a choice of the system under consideration and a convention for the spin up direction. This freedom of convention can lead to values of the spin polarization that cannot be directly compared.

Take as an example the optical excitation of carriers in a semiconductor by a circularly polarized laser. The spin polarization of the incident beam is typically defined such that $N$ is the number of photons within the beam and the spin up direction is the
majority spin direction. The spin polarization inside the semiconductor might be defined by the excited carriers induced by the laser. When these carriers recombine, the emitted light’s spin polarization could be defined by the number of photons in a certain photoluminescence peak. The photoluminescence spin polarization, in turn is used as a lower bound (exact in the limit $\frac{\tau_{\text{exciton}}}{\tau_{\text{spin}}} \rightarrow 0$, where $\tau_x$ represents the lifetime of $x$) on the spin polarization of the excited carriers [19, 20].

**Spin-dependent chemical potential**

An alternative way to quantify the spin polarization of the system is to examine the net spin angular momentum density (net spin angular momentum divided by volume) of a material. Of particular use in visualizing the spin polarization of a semiconductor are spin-dependent density of state (DOS) diagrams. These diagrams indicate the density of spin up and spin down electrons at varying energy levels, with completely filled states typically omitted. In the DOS diagrams used here, the shaded region on the left (right) represents the density of electrons in the spin up (down) state for valence electrons. The area of the shaded region for the spin up (down) electrons is the total number density of valence electrons in the up (down) spin state. Thus the total spin density in the semiconductor is the difference between the area on the right and left of the diagram.
The equilibrium spin polarization of a material is determined by how the spin up and spin down states fill. When the semiconductor’s electrons are in equilibrium and ignoring thermal effects, electrons fill the lowest available states, regardless of spin state. In magnetic materials, the asymmetry of the availability of states with respect to spin results in more electrons filling states with one spin orientation than the other, as shown in figure (1). The ferromagnet depicted has a net magnetization in the spin down direction because there are more total spin down (majority spin) than spin up (minority spin) electrons. Interestingly, at the Fermi level, there are more spin up than spin up electrons. Thus carriers injected from this idealized ferromagnet at certain biases will be dominated by the minority spin-state carriers, as is further discussed in the section on electronic spin injection.

When the available states for electrons have the same distribution in energy for spin up and spin down states, the total numbers of spin up and spin down electrons at equilibrium are equal; the net spin density is zero, as shown in figure (2a).
Figure 2. The filling of states in a non-magnetic semiconductor. In equilibrium (a), there is no net spin polarization; both spin states are equally filled. A non-equilibrium density of states as shown in (b) induces a net spin polarization in the semiconductor, despite its lack of intrinsic magnetism.

In such a case, the Fermi level is simply defined as the energy equilibrium electrochemical potential, $\mu_0$. In such cases, no spin polarization is intrinsic to the material. However, when the electron distribution is driven out of equilibrium, electrons may be excited into states with a higher energy and a different spin orientation, as shown in figure (2b). This results in a spin-dependent chemical potential, $\Delta \mu$, as shown in figure (2b); it is defined to be $\Delta \mu = \mu_\uparrow - \mu_\downarrow$, where $\mu_\uparrow(\downarrow)$ is the chemical potential of the Fermi energy of spin up (down) electrons. This value is also referred to as the non-equilibrium magnetization [28]. The spin-dependent chemical potential can give rise to a spin dependent voltage which can be utilized to electrically detect certain systems’ spin states, as in the non-local measurements discussed later.
Spin transport phenomena in non-magnetic materials

The steady-state spin dependent chemical potential, or spin accumulation, in non-magnetic materials is determined by the balance of spin injection with effects that decrease the spin polarization: spin transport, and spin decay and dephasing. Electronic spin injection occurs when spin is injected from a collection of charges with a net spin-polarization, such as at the Fermi level of a ferromagnet, into another material, such as a normal semiconductor. The polarization of the interfacial current depends on the spin polarization of the injector, the applied bias and direction, and the interfacial tunnel barrier profile.

Once injected, the spin ensemble decays to its equilibrium through spin decay or precession. In spin decay processes, spin scattering mechanisms such as momentum scattering coupled to a spin flip act to randomize the orientation of each spin in the spin ensemble. In spin decay, individual spins do not flip but are coherently rotated by an effective magnetic field. Because the net spin polarization of spin ensembles is dependent on the relative phase of its spins, non-uniformity of the electromagnetic fields within the materials, both intrinsic and extrinsic, leads to spin decay. For a comprehensive overview of this somewhat involved topic, see [6]. In general, low atomic number materials, and crystals with inversion symmetry should have longer spin decay and dephasing times.

The decay of the spin polarization of a system causes the spin polarization of a system to decay within a characteristic length, the spin decay length, $\lambda_s$. In the absence of an electric field, the spin decay length is given by $\lambda_s = \sqrt{D\tau_s}$, where $D$ is the electron
diffusion constant and $\tau_s$ is the spin lifetime. In experiments and devices where the injection and detection areas are spatially separated, the distance should be less than $\lambda_s$ in order for a spin signal transmission.

The non-equilibrium spin state in a semiconductor can be manipulated through the application of a transverse magnetic field, as in the Hanle effect. As basic quantum mechanics describes, the orientation of the expectation value of an ensemble of electron spins (the vector spin polarization in the sample) will precess at the Larmour frequency, $\omega_L$, around the vector of the applied magnetic field. If a single electron experiences precession for a time “$t$”, its phase will change by $\Delta \varphi = \omega_L t$. By changing either the Larmour frequency or the time of precession, the phase change of an injected spin can be controlled. Utilizing this effect allows a class of Hanle-type experiments which can be used to characterize the spin lifetime or the transit time in a material system.

To model the time evolution of the spin density, $s$, in a material, one may follow a theoretical procedure similar to that used in obtaining the continuity equation for charge current. With the additional consideration of spin decay, the differential form of the spin density drift-diffusion equation becomes

$$\frac{\partial s}{\partial t} = D \frac{\partial^2 s}{\partial x^2} - v_d \frac{\partial s}{\partial x} - \frac{s}{\tau_s} \tag{2}$$

where $D$ is the diffusion constant, $v_d$ is the drift velocity, $\tau_s$ is the spin decay time, and $x$ is the spatial variable. When this differential equation is solved through use of Green’s function solution for a point spin density in both position and time, and the spin precession is accounted for, the drift-diffusion equation describing the spin polarization at a point $x_2$ due to spin injected at point $x_1$ becomes:
\[ s(x_1, x_2, B) = \int_0^\infty \frac{s_0}{\sqrt{4\pi Dt}} e^{(x_1 - x_2 + v_d t)^2 / 4Dt} \cdot e^{-t / \tau_s} \cdot \cos \left( \frac{g\mu_B B}{\hbar} t \right) dt \quad (3) \]

where \( s(x) \) is the spin polarization at point \( x_2 \), \( s_0 \) is the initial spin polarization at \( x_1 \), \( B \) is the effective magnetic field in the material, \( g \) is the electron g factor in the material, and \( \mu_B \) is the Bohr magneton. By numerical integration the integral spin drift-diffusion equation over the width of the detector and injector contacts and over all time, this equation has been successfully used to model experimental spin dynamics using only one free parameter, \( s_0 \)[29]. For experiments where the injection and detection sites are the very large and located in the same location in space, the diffusion term, \( 1 / \sqrt{4\pi Dt} \), and the drift term, \( e^{(x_1 - x_2 + v_d t)^2 / 4Dt} \), become unimportant, since spins do not move away from the injection/detection areas. By integrating the position independent form of equation three, \( s(B) = \int_0^\infty s_0 \cdot e^{-\frac{L}{\tau_s}} \cdot \cos \left( \frac{g\mu_B B}{\hbar} t \right) dt \) over all time, one obtains a Lorentzian curve \( s(B) = \frac{s_0}{1 + (g\mu_B B \tau_s / \hbar)} \) which has been found to explain spin dephasing in a wide array of spin dephasing experiments[30].

Electronic spin injection

The conductivity and spin polarization of interfacial charge transport across an FM/N junction is strongly influenced by the barrier formed at the surface. For ferromagnet / semiconductor (FM/SC) junctions, a Schottky barrier and an interfacial dipole arises. In ferromagnet/insulator/non-magnetic material (FM/I/N) junctions, the insulator itself serves as a barrier in addition to any interfacial dipoles and built-in
barriers which may form. Ferromagnet/non-magnetic metal junctions (FM/M) may be viewed as a tunnel junction with vanishing barrier height. Then the transport properties of the interface may be analyzed in the framework of tunneling theory. Because spin polarization is preserved in tunneling, tunneling theory can be extended to spin-polarized tunneling by analyzing the tunneling conductance as a sum of conductances from two spin channels. FM/I/M, FM/I/S, and possibly FM/I/degenerately-doped SC tunnel junctions, the WKB approximation may be applied and the tunneling current is proportional to the barrier height and the width of the barrier: 

$$I \propto \exp\left\{-\frac{2i}{\hbar} \int dx \sqrt{2m(E - V(x))}\right\}$$

[31], where the integral is taken over the width of the barrier. As a result, virtually all tunneling occurs for electrons lying at or near the Fermi level and tunneling current through an ideal half metal/insulator/metal stack is almost completely spin-polarized for low biases.

For injection from FM across a barrier to a semiconductor with a relatively low density of states, a Fermi Golden Rule analysis should be applied. For the case of spin-polarized scanning tunneling microscopy (STM) with a spherical tip, the Fermi Golden Rule analysis shows that the tunneling current is then proportional to

$$I \propto V \sum |\psi(r_0)|^2 e^{-kr}$$

[32], where $V$ is the voltage between the tip and the sample, $k$ is the electron wave vector, $r$ is the distance from tip to sample, and $\psi(r_0)$ is the amplitude of a semiconductor state. A spin-polarized STM tip can be used to probe the spin-dependent local density of states on the surface and image the spin of single adatoms on a surface [33].
Figure 3. The polarization of spin injected into a semiconductor through a tunnel barrier in as a function of applied voltage. Adapted from [34]. Shown here for various values of the Schottky barrier height.

Tunneling across real F/I/SC interfaces is complicated by a host of effects, including dislocations in the junction region, interfacial compounds, and interfacial roughness [32]. A calculation of the effects of Schottky barrier height, built in voltage, and semiconductor doping levels on tunneling across a FM/I/SC stack was performed using a tight binding model, non-equilibrium Greens functions, and the Keldysh formalism [34]. The calculation found that the spin polarization of the injected current was highest for low SC doping levels, low Schottky barrier height, and high built-in voltages. Their calculations also showed that the spin polarization of the injected current is not symmetric around zero bias, as shown in figure (1) [34]. Non-idealities can reduce the spin polarization of injected carriers in FM/I/SC junctions. Defect states within the barrier can lead to hopping transport. For finite-temperature measurements, thermionic emission also contributes to the interfacial current. Furthermore, if the tunneling resistance is higher for one of the spin channels, the Johnson voltage noise for that
channel will be higher than for the other, as $\langle V^2 \rangle \propto R$ [35]. All these effects can lower the spin polarization of the injected current.

The spin polarization of an injected current is also adversely affected by the resistance mismatch between the injector and channel materials. When a more conductive material, such as a ferromagnetic metal, is used as a spin injector into a less conductive material, such as a semiconductor, the efficiency of spin injection can be quite low; this is the resistance mismatch problem. The two channel model of electrical conduction explains this phenomenon. First used by Mott to explain low temperature conduction in ferromagnets, the two channel model treats spin up and spin down electrons as separate carrier types flowing through parallel resistors. As a specific example, take the ferromagnet/semiconductor junction, as shown in figure (4).

Figure 4. A two channel model for spin transport through a ferromagnet/semiconductor junction.

In the language of the spin polarization, the current flowing through the upper (lower) branch would be $X_{\uparrow}(\downarrow)$, and the spin polarization is determined by the relative
difference in the current flowing through the two pathways. Thus the spin polarization of the current can be expressed as

$$P_\uparrow = \frac{I_\uparrow - I_\downarrow}{I_\uparrow + I_\downarrow} = \frac{R_\uparrow - R_\downarrow}{R_\uparrow + R_\downarrow} = \frac{R_{FM,\uparrow} - R_{FM,\downarrow} + R_{SC,\downarrow} - R_{SC,\uparrow}}{R_{FM,\uparrow} + R_{FM,\downarrow} + R_{SC,\downarrow} + R_{SC,\uparrow}}$$

(4)

Assuming that the resistance in the semiconductor is much larger than the resistance in the ferromagnet and that the resistance in the ferromagnet is spin independent, this equation approximates to

$$P_\uparrow \approx \frac{R_{FM,\uparrow} - R_{FM,\downarrow}}{2R_{SC}}$$

(5)

Since $R_{FM} \ll R_{SC}$ for both signs of spin, the spin polarization is much less than one.

This inefficiency in spin injection has been overcome by the insertion of a tunnel barrier (TB) with a spin-dependent resistance, schematically shown in figure (5).

Figure 5. A two channel model for spin transport through a ferromagnet/tunnel barrier/semiconductor junction.

If the tunnel barrier resistance for the spin down channel is much larger than for the spin up channel and is approximately equal to the semiconductor resistance, then the spin polarization equation becomes

$$P_\uparrow = \frac{R_{FM,\uparrow} - R_{FM,\downarrow} + R_{TB,\downarrow} - R_{TB,\uparrow} + R_{SC,\downarrow} - R_{SC,\uparrow}}{R_{FM,\uparrow} + R_{FM,\downarrow} + R_{TB,\downarrow} + R_{TB,\uparrow} + R_{SC,\downarrow} + R_{SC,\uparrow}} \approx \frac{R_{TB,\downarrow}}{R_{TB,\downarrow} + R_{SC,\downarrow} + R_{SC,\uparrow}}$$

(6)
which may be an appreciable spin current polarization. Because the numerator is controlled by the difference in the tunnel barrier resistances for the spin up and spin down channels, solving the resistance mismatch problem with a spin-dependent tunnel barrier is effective only if the tunnel barrier has a high resistance for the relevant spin channel (spin down in the previous discussion). If the tunnel barrier is compromised through pinholes or interfacial states which facilitate hopping through the barrier, the resistance mismatch problem again becomes important.

2.3 Previous Work in Spin Injection

Spin-polarized injection of electrons has been demonstrated using both optical and electronic methods. Similarly, detection of spin-polarization in a material can also be done optically or electronically. To elucidate the experimental progress made in spin injection, specific experiments are analyzed in some detail.

In experiments done by Crooker, et al. [19], the spin polarization in an n-GaAs channel was examined both electronically and optically. The experimental device is show in figure (6). In addition, a polarized laser for spin injection and a Kerr-rotation measurement setup were available for the experiment. In Kerr-rotation spectroscopy, the magnetic fields of the material from which a polarized laser reflect affect the polarization of the reflected beam. Thus the angle of Kerr-rotation tracks the magnetization of a sample, as shown in the figure (7) [30].
In their first measurement, a current was injected from the iron contact on the left and drifted toward the other contact (in the positive x direction), as shown in figure (8). After applying a very weak magnetic field to precess the spins to an angle where they could be detected, the spin density in the channel was imaged using Kerr-rotation spectroscopy. It reveals that the injected current is initially spin polarized, but the polarization decays rapidly away from the injector. By switching the magnetization of the iron contacts and showing that the Kerr-rotation angle also changes sign, the Kerr
rotation was confirmed to result from the spin-polarized contacts. Near the other contact, the channel is also spin polarized, but to a much lesser extent. To determine the origin of this polarization, the authors apply stress to the device, changing the semiconductor crystal’s built-in electric polarization field. Because the electric fields in the frame of the electrons transform into an effective magnetic field dependent on the electron wavevector, \( \vec{k} \), the authors were able to show that the spin-polarized electrons near the right contact were diffusing away from the electrode (against the direction of electron drift) by examining the effects of strain on the spin polarization in the channel. If the spin polarized electrons are drifting away from the left electrode, this implies that the electrons in the channel are polarized by reflection from the electrode.

Figure 8. Kerr-rotation spectroscopy showing spin polarizatioin in the semiconductor channel.
[19]

In another experiment reported in the same paper, a polarized laser was used to optically induce a spin polarization in the channel. This optical polarization was imaged using Kerr rotation spectroscopy, and the normalized change in the conductance of the
contact, $\Delta G/G$, is measured, as shown in figure (9). The effect is shown to be due to spin-dependent tunneling by reversing the magnetization of the ferromagnetic contact and observing that the normalized change in conductance also changes sign. When the injected spin is oriented parallel (antiparallel) to the magnetization of the contact after drifting toward it, the conductance is high (low). To summarize, these experiments illustrate the electronic spin injection, optical spin injection, electrical spin detection, and optical spin detection.

Another method of optical detection of the spin polarization in a semiconductor is the measurement of the polarization of light emitted from a light emitting diode. When spin polarized carriers are injected into the recombination region of an LED, the electron spins undergo precession and decay until they recombine with holes and emit a photon. The angular momentum of the carriers at the time of recombination is preserved as the
circular polarization of the emitted photons. If the spin decay time is much longer than the recombination time, the circularly polarization may yield an accurate measure of the spin polarization of the excited carriers in the device’s active region. Motsnyi et al. used this type of device, termed a spin-LED to demonstrate electronic spin-injection across an insulating tunnel barrier into an AlGaAs/GaAs quantum well [21]. Their device structure is shown in figure 10 (a). To demonstrate that the polarization of the incident light resulted from the spin polarization of the device’s excited carriers, a magnetic field was applied and the oblique Hanle effect was observed (fig. 10 (b)). The applied field initially rotates the spins to align in such a way (with a component along the z-direction) as to induce a circular polarization in the light which is able to escape from the top of the device. As the field is increased, the carriers’ spins may precess multiple times before recombination, leading to a saturation of the spin ensemble’s polarization along the z-direction. This is reflected in the optical polarization of the emitted light, as shown in fig. 10 (c).
Figure 10. Spin LED
(a) A spin LED device demonstrating electrical spin injection and optical spin detection. (b) The oblique Hanle effect is used to orient the spins so that they can be detected. (c) optical polarization of emitted light as a function of applied magnetic field. [21]

The non-local spin injection-detection experiment, which Johnson and Silsbee used to show spin injection and detection into a metal in 1985, is shown in figure (11) [36]. In the non-local spin injection-detection experiment, a spin polarized current is run from the injector to ground and the voltage from a nearby detector to ground is recorded. Although the net current flows from the injector to ground, the spin polarization underneath the injector is able to diffuse underneath the detector when the distance between the detector and injector is less than the spin diffusion length. Under these circumstances, a non-equilibrium spin density is present underneath the detector.
electrode and a voltage $V_d$, proportional to the spin-dependent chemical potential, arises. The non-local geometry has two main advantages over the local geometry: (1) because there is no net current flowing between the injector and detector, there is no voltage drop over this distance and (2) because the injector and detector are separate electrodes, one does not need to account for magnetoreistive effects of spin injection. To further ensure that the signal indeed arises from the spin-dependent chemical potential, the Hanle effect on $V_d$ was measured. Their results are shown in figure (12).

![Figure 11. Non-local device geometry.](image)

In this geometry, the relevant time “t” for Hanle precession is the time required for a single spin at the injector to drift to the detector and be detected. During this time, the spin phase changes by $\Delta \varphi = \omega_L t$. Assuming the injector and detector are aligned, a precession of the spin phase by $\Delta \varphi = \pi$ will result in the electron spin to be anti-aligned.
from the dominant spin tunneling channel. The decay of $V_d$ with the applied field at low bias can be qualitatively understood in this manner. At higher applied magnetic fields, the signal approaches a baseline value instead of oscillating with a phase $\omega_L t$. To explain this signal decay, the spin-ensemble picture is more enlightening. For an ensemble of charges diffusing through random walks from the injector to the detector, the time $t$ required for each electron to diffuse to the detection electrode and be detected differs. This leads to an inhomogeneous change in the spin phase, leading to dephasing of the spin ensemble and decay of the spin signal through the Hanle effect.

![Figure 12. Hanle dephasing effect on the non-local voltage $V_d$.](image)

In their measurements of all-electronic spin injection and detection into n-GaAs, Lou et al. used an extension of the non-local measurement to further confirm that the non-local voltage signal arises from the spin polarization of injected spins [29]. Their devices utilized the Schottky barrier as a tunnel barrier for efficient spin injection. They performed the non-local measurement of $V_d$ and the decay of $V_d$ with applied field.
through the Hanle effect, but they also performed a magnetic field sweep along the
direction of the magnetization of the injector and detector. Because the injector and
detector were fabricated to have different coercive fields, this allows the injector and
detector to be switched from parallel to anti-parallel to the applied field. The change in
conductivity of the channel, similar to the magnetic tunnel junction’s magnetoresistance,
results in a hysteretic voltage change tracking the alignment state of the contacts, as
shown in figure (13 b). In the lower half of the graph, the background has been
subtracted. In figure (13 c), the Hanle effect on the non-local voltage is displayed. The
oscillation around zero voltage change corresponds to the semi-coherent rotation of the
spins over an angle of $\pi$ before the decoherence of the spin ensemble.
Figure 13. Non-local measurement geometry.
(a) Device schematic. (b) Field sweep showing magnetoresistive effects. (c) Hanle spin dephasing curves in the non-local geometry. From [29].
Chapter 3:
Modeling III-Nitride Heterojunction Band Structure with BandEng

3.1 BandEng Modeling of III-V Polar Heterojunctions

Band diagram and carrier concentration calculations were carried out using BandEng software [37] in order to engineer 2DEG heterojunctions for electron spin injection and magnetic heterojunction studies. The modeling takes into account experimental semiconductor band parameters, including the band materials band gap, the electron affinity, the height of the barrier formed at the interface (Schottky barriers to metal contacts, etc.), and the effective masses of carriers in the material. Given these parameters and a given heterojunction structure, the band structure is determined through self-consistent iterations of solutions to the Poisson equation. This calculation includes unbound charges such as the charge buildup at the interface of polar faces of nitrides. After solving the Poisson equation to obtain the band structure and a corresponding carrier concentration profile, the band structure is used to iterate solution to the Schrodinger equation to find the wavefunction of the carriers. The total carrier
concentration is the sum of the individual carrier probabilities, given by the wavefunctions.

Although the nitride materials modeled are relatively well studied, the experimental band parameters required to calculate the band profiles are not available for every alloy concentration. In order to overcome this limitation, an extension of Vegard’s law was used to interpolate between available experimental results. Using this approximation, band parameters such as band gap or Schottky barrier heights for allows are estimated as $E_{Al_xGa_{1-x}N} = xE_{AlN} + (1 - x)E_{GaN}$. Although Vegard’s law is only an approximation, its use allows modeling of heterostructures which would be impossible using only the available experimental band parameters.

### 3.2 Modeling Gadolinium Doped Heterojunctions

Because the mechanism of ferromagnetism in GaMnAs is carrier-mediated, the possibility that carrier-mediated ferromagnetism is important in the similar material GaGdN should be considered. In the experiments done with gadolinium doped AlGaN/GaN 2DEG and two-dimensional hole gas (2DHG) heterostructures, modeling is used to determine the expected overlap between the polarization induced carriers and the dopants. The results of these modeling efforts are presented along with the heterojunction structures in chapter 4.
3.3 Modeling for Spin Injection into 2DEG Heterojunctions

In order for successful spin injection and detection into semiconductors, the width and height of the tunnel barrier into the conduction channel in the semiconductor should be chosen to optimize the spin polarization of carriers in the channel. In the two channel model this equates into choosing the tunnel barrier width and height so that the difference in tunneling resistances for the two channels is significant compared to the resistance of the semiconductor channel, as can be seen from equation 6. In the experiments described herein, the conducting channel is a 2DEG formed at the polar interface between an AlGaN capping layer and a buried GaN. The tunnel barrier is comprised of the surface barrier to the AlGaN capping layer and the capping layer itself. In these experiments, the AlGaN capping layer serves both as the source of polarization doping in the conducting channel and as a portion of the tunnel barrier. As the concentration of aluminum in the capping layer is increased, the tunnel barrier height and the polarization doping both increase. As the thickness of the capping layer increases, the width of the tunnel barrier and the polarization doping both increase. This effect is shown in figure 14. For this reason, the AlGaN capping layer thickness and composition should be carefully chosen if the expected polarization of carrier injection into the semiconductor channel is to be optimized.
Figure 14. The AlGaN capping layer as a tunnel barrier. The tunnel barrier is as seen by an electron with an energy equal to the lowest energy state in the GaN conduction band is reduced as the thickness and aluminum composition of the capping layer is reduced.

For efficiency of spin injection, maximizing the difference of spin dependent tunneling resistances is the only concern. However, for the injected current to be detected, the spin polarization in the channel and the tunnel barrier resistance must be addressed. The carrier density and mobility help determine the spin polarization in the channel. The tunnel barrier resistance (not the difference between the spin polarized resistances) determines the level of Johnson noise in the semiconductor.

The carrier density and mobility within the channel are crucial for spin transport. If the carrier concentration and mobility are too low, the resistance of the semiconductor channel will be very large and the spin polarization in the ensemble will low (see equation 6). On the other hand, if the carrier concentration is too high, the spin lifetime
of the carriers in the channel will be reduced by electron-electron interactions. When
using modeling to decide which heterojunctions were to be fabricated, the desired carrier
density was assumed to be the same as that which leads to the longest spin lifetimes in
bulk GaN, a density on the order of $10^{16}/cm^2$ [38].

In the spin valve and four point spin precession measurements, the spin
polarization in the channel is indirectly sampled by measuring the voltage between spin
polarized leads. One lead is over an area with a non-zero spin polarization while the
other is over an area far away from any spin injection but still electrically connected. The
lead over the spin polarized carrier ensemble samples the spin polarization state of the
carrier ensemble. The other lead is connected to a spin ground; it samples unpolarized
carriers. By varying the spin state either of the polarized carrier ensemble or the lead
over it, the resistance of the spin-dependent tunnel barrier changes. This change in tunnel
barrier resistance results in a change in the voltage measured from the lead over the
ensemble to the lead over ground. In effect, the measurement probes the spin dependent
chemical potential shown in figure 2. Since the measurement is a voltage measurement,
sources of voltage noise, including Johnson noise, need to be mitigated. Since Johnson
noise increases with increasing tunnel barrier resistances, trade-offs must be made
between maximizing the spin polarization of injected currents and reducing the Johnson
noise in the measurements.
Chapter 4: 
Gadolinium-Doped III-N Quantum Well Heterostructures Experiment and Modeling

4.1 Initial gadolinium-doped III-N quantum well structure design

A series of gadolinium doped III-N quantum well heterostructures were grown to investigate how the magnetism of the heterostructures depends on overlap between the quantum confined carriers in the heterojunction as well as on the sign of these carriers. In the first set of samples, GaN/AlGaN 2DEG heterostructures were grown with three different gadolinium doping profiles: δ-doped at the 2DEG, δ-doped away from 2DEG, and three-dimensionally doped at the interface. The delta doping profile is a sheet of gadolinium, the 3D profile is a layer of GaN in which gadolinium is co-deposited in the hope of obtaining GaGdN. In the second set of samples, 2DHG structures were grown with a δ-doped layer of gadolinium either overlapping the hole gas or spatially separated from the hole gas. If the mechanism of ferromagnetism in Gd:GaN is electron-mediated, the doping profiles with the most overlap with the 2DEG should exhibit the greatest ferromagnetism. If the mechanism of ferromagnetism in Gd:GaN is hole-mediated, the
doping profiles with the most overlap with the 2DHG should exhibit the most ferromagnetism. These samples are designed to test the dependence of the ferromagnetism of gadolinium doped GaN on the carrier density around the dopant atoms.

The target structures for the three Gd-doped 2DEG heterojunctions discussed herein are shown in figure 15. The estimated carrier density as a function of depth for these structures is based on a gallium-polar heterostructure with a 5 nm AlN capping layer on top of a relaxed GaN layer. This structure’s band diagram and carrier density profile was simulated using BandEng and the results are shown in figure 16. Based on these predictions, sample 091211A is designed so that the gadolinium doping overlaps the 2DEG near the region of greatest electron density. Sample 091211B is designed so that the gadolinium doping is separated from the region of greatest electron density. Sample 100125C is designed for overlap between the gadolinium doped region and the carriers; the layer of GaGdN is deposited where the 2DEG would be if that layer were simply GaN. The black lines through the structures for 091211A and 091211B indicate the depth at which the gadolinium δ-doping layer was deposited. The layers of GaN directly on top of the GaN templates were deposited with MBE. These buffer layers serve to lower the defect density native to the gallium nitride substrates.

Overlap between charge carriers and the dopant atoms also determined the target structures for the 2DHG samples; these are shown in figure 16. In sample 100628A, the doping layer is 0.3 nm away from the interface, with maximum predicted overlap with
the hole gas. In sample 100628B, the doping layer is 2 nm away from the interface, with reduced overlap with the hole gas as compared to 100628A.

![Diagram of target structures for Gd-doped 2DEG heterojunctions.](image1)

Figure 15. Target structures for Gd-doped 2DEG heterojunctions.

![Diagram of target structures for Gd-doped 2DHG heterojunctions.](image2)

Figure 16. Target structures for Gd-doped 2DHG heterojunctions.

In order to determine the location and extent of the sheet of carriers in the quantum wells, BandEng simulations of the heterostructures were carried out. The presence of gadolinium is ignored in this calculation; gadolinium doped GaN is assumed to have the same electronic properties as undoped GaN. This should slightly overestimate the sheet carrier density. However, the effect should not be large, especially in the delta-doping case where less than a monolayer of gadolinium is deposited. The results of the BandEng calculations are shown in figures 17 (2DEG heterostructures) and 18 (2DHG heterostructures).
Figure 17. Calculated target structure carrier profile and band diagram for Gd-doped 2DEG heterostructures.

Figure 18. Calculated target structure carrier profile and band diagram for Gd-doped 2DHG heterostructures.
4.2 Gadolinium doped III-N heterostructures characterization methods

The quantum well structures discussed herein were deposited at The Ohio State University on gallium nitride templates by Jing Yang and Dr. Roberto Myers through molecular beam epitaxy (MBE) in a system dedicated to III-nitride growth. Structural characteristics of the heterojunctions were undertaken using a Bruker D8 Discover High-Resolution instrument for x-ray diffraction (XRD) to characterize crystalline structure and alloy composition, a Veeco atomic force microscopy (AFM) to characterize surface morphology, and secondary ion mass spectroscopy (SIMS) performed by Evans Analytical Group for gadolinium doping profile characterization. Electronic characterization of the heterojunctions was undertaken using a Lake Shore Cryotronics Hall Effect Measurement System. Magnetic characterization was undertaken using a Quantum Design MPMS superconducting quantum interference device (SQUID) Magnetometer.

4.3 Gadolinium doped GaN/AlGaN 2DEG characterization

In order to quantify the Gd-doping levels and verify that expected heterostructures were grown, SIMS analysis was carried out by Evans Analytical Laboratories on the initial set of samples grown. These results were used to verify the calibration of the MBE doping process for all subsequent depositions. In the case of sample 091211A, the proximity of the dopant layer to the surface of the sample made an accurate determination of the doping density using SIMS difficult, so in this case the spatial doping profile was determined by SIMS while the overall density was determined using Rutherford backscattering (RBS). The doping profiles are shown in figure 19.
Using the data shown in figure 19, the three-dimensional density was integrated over depth to obtain the two dimensional density of the dopant atoms. The depth of the delta-doped gadolinium layers was taken to be the concentration peak depth, while the uncertainty in depth was taken to be half of the full-width at half maximum (FWHM) of the peak. The full-width at half maximum may be due to line broadening associated with the SIMS measurement itself. SIMS measures the ion density by ion milling material from the material and measuring the concentration of released ions. If material is not milled in the shape of a perfect cylindrical well or if material is sputtered from buried layers, ions of that material are released when the bulk of the sputtering is occurring at a different depth, broadening the apparent depth at which the material is present. Thus the figure 19 should be taken as an indication of the distance between the interface and the center of the doping layer, not as a measure of the abruptness of the interface or the thickness of the gadolinium doping layer. However, the data is still useful in examining the overlap of dopant atoms with the carrier gas and the
2D density of dopants. The aggregate data SIMS is summarized in tables 1 and 2. The SIMS and RBS data verify that the doping levels are accurate within 10%. The doping calibration was assumed to be stable over the course of growth of the samples when the doping density is presented.

<table>
<thead>
<tr>
<th></th>
<th>Value</th>
<th>Uncertainty</th>
<th>Expected value from growth parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gd peak depth (nm)</td>
<td>7.2</td>
<td>2.7</td>
<td>5.4</td>
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<tr>
<td>Interface depth (nm)</td>
<td>5</td>
<td>1</td>
<td>5</td>
</tr>
<tr>
<td>Gd concentration (atoms/cm²)</td>
<td>2.5 E14</td>
<td>0.5 E14</td>
<td>2.3 E14</td>
</tr>
</tbody>
</table>

Table 1. Doping profile of Gd-doped 2DEG 091211A.

<table>
<thead>
<tr>
<th></th>
<th>Value</th>
<th>Uncertainty</th>
<th>Expected value from growth parameters</th>
</tr>
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<td>Gd peak depth (nm)</td>
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<td>2.9</td>
<td>15</td>
</tr>
<tr>
<td>Interface depth (nm)</td>
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<td>1</td>
<td>5</td>
</tr>
<tr>
<td>SIMS Gd concentration (atoms/cm²)</td>
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<td>0.2 E14</td>
<td>2.3 E14</td>
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<tr>
<td>Rutherford Backscattering Gd concentration (atoms/cm²)</td>
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<td>0.6 E14</td>
<td>2.3 E14</td>
</tr>
</tbody>
</table>

Table 2. Doping profile of Gd-doped 2DEG 091211B.

SQUID magnetometry was performed to characterize the magnetic properties of the doped heterojunctions. Unless otherwise noted, all samples have a diamagnetic background subtraction, which is obtained by taking high field data (> 1T) at 300 K, performing a linear fit for positive and negative field values, then averaging the slopes. Data taken by the SQUID magnetometer in emu is converted to Bohr magnetons per
gadolinium dopant ($\mu_B$/Gd) based on a caliper measurement of sample area and the two-dimensional density of gadolinium dopants provided by SIMS and RBS. Magnetization data for the three dimensionally doped GaN/AlGaN 2DEG is not presented because the magnetic polarization of the sample was too low to effectively center the sample and obtain good-quality data.

![Saturation Magnetization vs. Temperature](image)

Figure 20. Saturation magnetization verses temperature for two 2DEG

As can be seen from figure 20, both of these samples display the giant magnetic moment effect, with $\mu_B$ greatly exceeding the number of Gd-electrons in unfilled shells available for magnetic alignment, seven. As expected, the saturation magnetization declines as a function of temperature as thermal fluctuations decrease the magnetization. The saturation magnetization is consistently lower for the sample with greater overlap between the electron density. This suggests that the giant magnetic moment observed in these samples is not mediated by electrons.
Since the mechanisms of magnetism have characteristic temperature dependences, the saturation magnetization was measured as a function of temperature for the sample exhibiting a greater giant moment effect: sample 091211B, doped 10 nm from the interface. This data along with the coercive field is plotted in figure 21. The functional form of the decline does not match a standard theoretical prediction such as the $1/T$ dependence expected for paramagnetism or the temperature insensitivity of diamagnetism.

![Coercive Field and Saturation Magnetization vs. Temperature](image)

**Figure 21.** In-plane saturation magnetization and coercive field and vs. temperature for sample 091211B.

In addition to exhibiting the striking giant magnetic moment effect, these samples also display unusual hysteresis loops at low temperature. The unusual non-monotonic behavior of the hysteresis loops at low temperature is shown in figure 22. Possible explanations for the anomalous hysteresis loop include secondary phase formation. As can be seen in figures 23 and 24, the anomalous hysteresis loop disappears at temperatures of 60 K or higher and also when the magnetization is measured out of plane.
Figure 22. Anomalous low temperature hysteresis loops for samples 091211A and B.

Figure 23. Disappearance of anomalous hysteresis loop with increasing temperature.
To examine the possibility that the samples morphology affects the magnetization, the surface morphology was characterized using AFM and the crystal structure was characterized using XRD. Atomic force microscopy (figure 25) shows that samples 091211A and 091211B have surfaces which are as smooth as the substrates on which they are grown, as measured by surface rms roughness. This indicates that the growth was undertaken in a region where growth of films occurs by layers, not three dimensionally and that the layer structure that was predicted was actually grown over a large surface instead of in isolated patches. The layered growth mode is also indicated by the thickness fringes in the XRD pattern, which originate from coherent Bragg reflection from the heterostructures interfaces (figure 26). Thickness fringes do not appear for films on which blocky, three dimensional depositions occur. The XRD also fail to show evidence of any secondary phases that may be expected if the gadolinium doping resulted
in precipitating crystallites within the GaN matrix. This indicates that the ferromagnetism is a property of the gadolinium doped GaN layer within the heterostructures, not a property of a gadolinium precipitate such as GdN.

Figure 25. AFM images of heterojunction surfaces for 2DEG heterostructures δ-doped with Gd.

Figure 26. XRD data showing AlN and GaN peaks only.

The electronic properties of the samples are important for computing; magnetic semiconductors are only useful for alternative computing schema based on charge flow if
the magnetic properties of the samples interact with the charge carriers. For this reason, the electronic properties of the samples were characterized using Hall effect measurements on samples with Van der Paw geometry. Contact to the 2DEG was attempted using both hot indium soldered contacts and Ohmic contacts deposited using established metal layer stacks and annealing techniques. Both methods gave comparable results, shown in table 3. The carrier densities are higher than expected for a 2DEG while the mobilities are lower than expected. This is attributed to parallel conduction through charges with lower mobilities than found in the 2DEG. The parallel conduction may be the result of unintentional doping of the GaN material or of charge layers present at interfaces within the templates.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Carrier Density (cm$^{-2}$)</th>
<th>Hall Mobility (cm$^2$/V∙s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>091211A</td>
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<td>230</td>
</tr>
<tr>
<td>091211B</td>
<td>1.9e14</td>
<td>320</td>
</tr>
</tbody>
</table>

Table 3. Electronic characteristics of gadolinium doped 2DEG heterostructures.

4.4 Gadolinium doped GaN/AlGaN 2DHG characterization

While the aforementioned gadolinium doped 2DEG heterostructures were ferromagnetic and showed the dramatic giant moment effect, the gadolinium doped 2DHG structures showed paramagnetism but no ferromagnetism. As shown in figure 27, the field sweep for sample 100628B does not show the hysteresis characteristic of ferromagnetism; sample 100628A similarly shows no ferromagnetism. The lack of
ferromagnetism in these samples suggest that the mechanism of ferromagnetism in Gd:GaN is not hole mediated, as it is with GaMnAs.

![Graph](image)

**Figure 27.** Magnetic field sweep of a representative gadolinium-doped 2DHG

To help determine the significance of the lack of ferromagnetism in these samples, the morphology of the samples was characterized using XRD and AFM. The AFM showed a surface morphology which was smoother for the deposited heterojunctions than for the underlying substrate. This indicates that the growth mechanism was planar, but also reflects the general trend in III-nitride growth that the higher the aluminum percentage of an AlGaN alloy, the more difficult it becomes to grow smooth films of high crystalline quality. The XRD did not show any evidence of GdN or other gadolinium alloy formation.
In contrast to the 2DEG samples, the 2DHG samples were insulating. Van der Paw samples exhibited giga-ohm level resistances for two point contacts from corner to corner. This may indicate poor electrical contact to the expected carrier gas; making contact to 2DHG layers is more difficult than making electrical contact to 2DEG layers. It could also reflect some compensation of the hole gas by the gadolinium dopants, though this seems unlikely given that gadolinium is an isoelectronic with the group III elements in III-nitride compounds. A more likely cause is oxygen contamination of the samples. Unintentional doping with oxygen was discovered in other samples grown around this time during a dopant calibration of the III-nitride MBE system. Since oxygen is an electron donor to GaN, this could have compensated the hole gas charge.

4.5 Conclusion

After a routine check of impurity levels in III-nitride MBE growth uncovered the presence of unintentional oxygen doping in the gadolinium-doped 2DHG samples, a set of GaN/AlGaN 2DEGs with the same basic design as samples 091211A and 091211B was deposited on insulating GaN templates. These structures exhibited no ferromagnetism. Their surface morphology was smooth, while their XRD analysis revealed no evidence of GdN₃ precipitates. Since the only substantive difference between the two sets of samples should have been a lowered level of oxygen defects, the lack of ferromagnetism in the re-grown gadolinium doped 2DEG samples is consistent with a defect-mediated mechanism of ferromagnetism.
Chapter 5:
All electronic spin injection and detection devices on III-N 2DEG heterostructures

5.1 2DEG Heterostructure modeling for electronic spin injection

In this project, all electronic spin injection and detection devices for 3 terminal Hanle spin precession measurements and 4 terminal spin valve and Hanle measurements are designed. In this design, the AlGaN capping layer of the heterojunction serves as both the source of polarization doping and as a tunnel barrier to mitigate the resistance mismatch problem discussed in chapter 2. The key parameters in heterostructure design for spin injection are the tunnel barrier resistance, electron density in the 2DEG, and a lack of parallel conduction paths. The tunnel barrier resistance depends on the contact area as well as the barrier height and thickness, which can be controlled by varying the aluminum percentage in the capping layer and the capping layer thickness. As described earlier, the longest spin lifetimes are expected for carrier concentrations around \(10^{17}/\text{cm}^3\); the capping layer is designed to ensure that a 2DEG will be formed while attempting to generate a carrier concentration of approximately \(10^{17}/\text{cm}^3\). The general approach to
modeling is described in Chapter 3. The specific results for this project are displayed in Figures 28 and 29 for 1.5 nm thick AlGaN capping layers.

As can be seen from the figures, the aluminum percentages which correspond to the desired three-dimensional electron density also correspond to exceedingly low sheet densities of electrons. A low sheet density of electrons in the 2DEG results in a high sheet resistance of the 2DEG itself, increasing measurement difficulties associated with Johnson noise. Of even more importance, as the polarization doping drops, the effects of slight differences between the target deposition and the actual deposition, as well as the effects of accidental oxidation of the AlGaN capping layer become increasingly important. Since 1.5 nm of AlGaN is the equivalent of 6 monolayers, the 2DEG may disappear entirely if small differences between the desired and the actual heterostructure occur for low polarization doping. This was taken into account when deciding on growth parameters. The general trends are consistent with expectations: as the concentration of aluminum in the capping layer increases, the electron density in the 2DEG increases and becomes more confined.
Figure 28. The 2DEG sheet carrier density and maximum 3D charge density for 1.5nm AlGaN capping layers.

Figure 29. 2DEG electron density as a function of depth.
5.2 Device design and processing for electronic spin injection

Once the heterostructures have been deposited and characterized, the spin valve measurement device must be fabricated using the heterostructure. Due to the sensitivity and high temperature nature of MBE deposition, deposition of the heterostructure on top of a device would be infeasible. The spin valve device fabrication requires definition of a mesa, deposition of contacts to the 2DEG, and deposition of large-area contacts for connection to electronic equipment.

The design of the heterojunction is most constrained by the spin diffusion length in the material. This can be calculated given the spin lifetime and the mobility. The diffusion length is (generically) related to the time elapsed by the diffusion equation, 
\[ l = \sqrt{D t}, \]
where \( l \) is the diffusion length, \( D \) is the diffusion constant, and \( t \) is the time elapsed. In the present case, \( l \) becomes the spin diffusion length when \( D \) is the diffusion constant of the carriers and \( t \) is the spin lifetime of the carriers. Using the Einstein relation, \( \mu = eD/k_B T \), the spin diffusion length can be estimated based on the carrier mobility and the spin lifetime. Under realistic assumptions at room temperature, a spin diffusion length of approximately 0.5 μm is possible[39]. The spin diffusion length sets an upper limit for the separation between the injection and detector contacts in 4-terminal spin injection-detection devices. Since this is pushing the fundamental limits of optical lithography (without designing photolithography masks to create exact interference patterns or using higher-energy exposure patterns), the injector and detector contacts are defined using e-beam lithography.
The measurement set-up also requires the definition of an electronically isolated mesa. In order to simply analyze experimental results, all electronic conduction must occur through the mesa. Since MBE-grown GaN is highly insulating, the mesa may be defined by etching away the capping layer away from the mesa. A process flow schematic is shown in figure 30. The first step, at top, shows the mesa definition using a reactive ion etch (RIE) to remove material not covered by a photolithography-patterned polymer mask. The second step shows the deposition of the ferromagnetic injector and detector contacts, which must be defined using e-beam lithography due to their submicron lateral separation. The contacts must be thick enough to be continuous over the mesa edge. In the final step, at bottom, non-ferromagnetic contacts are added to define the ground contacts at far right and left as well as to make contact to the ferromagnetic injector and detector contacts.
The shape of the ferromagnetic bars is crucial for controlling the magnetization state of the injector and detector contacts through the manipulation of shape anisotropy.

For thin-film ferromagnetic layers, magnetization states aligned in plane are energetically favorable. If the planar shape is non-isotropic, the magnetization will tend to align along the longer axis of the shape. The energetic favorability of this alignment increases with increasing aspect ratio (height/width) of the contact. The ferromagnetic contacts should be designed with different aspect ratios so that the direction of their magnetization states will switch at different applied fields; their coercive fields should be different. Under
certain circumstances, differences due to processing imperfections may be sufficient to
differentiate the coercive fields of the injector and detector electrodes.

5.3 2DEG heterojunction characterization

Based on the design considerations mentioned in the first section of this chapter,
two different 2DEG heterojunctions were deposited using MBE. The capping layers
were designed to be 1.5 nm thick layers of Al\textsubscript{x}Ga\textsubscript{1-x}N with x = 0.65 for sample 100707A
and x = 0.52 for sample 100707B. Atomic force microscopy images of the sample show
very clear hillock structures typical of high-quality layer by layer growth of nitrides, as
shown in figure 31. The electronic properties of the 2DEG heterojunctions were
characterized with Hall effect measurements at room temperature. The AFM RMS
roughness as well as the electronic properties from room temperature Hall effect
measurement are displayed in table 4. The sheet carrier densities are higher than
expected; if x were between 0.7 and 0.8, one would expect these carrier densities. The
Hall mobilities are lower than typical for GaN/AlGaN 2DEG heterojunctions by a factor
of 3, but since the capping layers are extremely thin relative to standard 2DEG
heterostructures used in conventional electronics, the existence of such a difference is
unsurprising.
Table 4. Sample properties of 2DEG heterostructures deposited for spin injection.

<table>
<thead>
<tr>
<th>Sample</th>
<th>x for Al\textsubscript{x}Ga\textsubscript{1-x}N capping layer</th>
<th>RMS Roughness</th>
<th>Sheet Carrier Density (cm\textsuperscript{-2})</th>
<th>Hall Mobility cm\textsuperscript{2}/Vs</th>
</tr>
</thead>
<tbody>
<tr>
<td>100707A</td>
<td>0.65</td>
<td>0.9 nm</td>
<td>5 E12</td>
<td>400</td>
</tr>
<tr>
<td>100707B</td>
<td>0.52</td>
<td>1.5 nm</td>
<td>3.2 E12</td>
<td>470</td>
</tr>
</tbody>
</table>

The sheet carrier density and Hall mobilities were also measured as a function of temperature, as shown in figures 32 and 33. For typical 2DEG heterojunctions, the sheet carrier density is roughly independent of temperature while the mobility increases as temperature (and the number of phonons available for electron scattering) decreases. In contrast, for doped semiconductor systems, the number of carriers drops as temperature decreases as carriers freeze out. Doped semiconductor carrier mobilities typically fall off at very high temperatures due to phonon scattering and at very low temperatures due to trapping in defect states inherent in impurity doped semiconductors. In contrast, carriers
in 2DEG heterojunctions are polarization doped and generally do have no lower energy levels into which they can be trapped. Thus one expects that the mobilities for 2DEG heterostructure carriers will rise as temperature falls. The 2DEG heterojunctions studied here show more complicated behavior. From room temperature the 2DEG carrier densities fall by less than an order of magnitude before experiencing a small increase at very low temperatures. The decline may be explained by the existence of defect induced traps or unintentional doping which would provide carriers conducting in parallel with the polarization doped carriers. This hypothesis cannot be tested with the available data, but great care was taken to minimize unintentional doping in the heterostructures, including band design as outlined in the appendix. The Hall mobility data also differs from typical 2DEG behavior. The shape of the mobility verses temperature plot is more typical of impurity doping than to polarization doping. The data may indicate the presence of impurity or defect doping in the samples or at a heterojunction interface.

Figure 32. Sheet carrier density for 2DEG samples 100707A and 100707B.
Figure 33. Carrier Hall mobility for 2DEG samples 100707A and 100707B.

5.4 Fabricated devices

Four terminal spin injection-detection devices were fabricated on samples 100707A and 10707B. Device fabrication required calibration or adaptation of various processing recipes, including AlGaN RIE etch rates and e-beam lithography parameters. The processing recipes with preliminary measurements are outlined in laboratory notebooks[40]. Since preliminary measurements do not provide conclusive evidence of spin injection, these results are not included here. A scanning electron microscope (SEM) image of the injector and detector portions of the device are shown in figure 34 with a schematic of electron flow in the 4-terminal device; figure 35 shows a detail of the image indicating showing the submicron widths and separation of the injector and detector contacts. Off-screen to the right and left are the large area contacts required for 4-terminal measurements. The dark area indicates un-etched areas, the grey backgrounds etched GaN, and the shapes deposited overtop are the injector and detector with their contacts.
Figure 34. SEM image of a 4-terminal all-electronic spin injection and detection device fabricated on a GaN/AlGaN heterojunction. Current flows from the left electrode to the left large area contact. The non-local voltage is measured from the right contact to the right large area contact.

Figure 35. Detail from an SEM image of a 4-terminal all-electronic spin injection and detection device fabricated on a GaN/AlGaN heterojunction.
5.5 Conclusions and future directions

III-Nitride heterojunctions for all-electronic spin injection and detection in a 4-terminal device were modeled and deposited using MBE. After characterization of their growth quality using AFM to study the surface morphology and characterization of their electronic properties with Hall effect measurements, 4-terminal spin injection devices were deposited by using reactive ion etching to define a mesa, depositing ferromagnetic contacts over the mesa, and finally depositing non-ferromagnetic metal contacts which could be used to provide electrical contact to electronic systems. Initial findings do not show clear signs of spin injection and detection in these devices. Further research on spin injection into GaN/AlGaN heterojunctions might be used to clarify the electronic properties of thin-capping layer 2DEGs, vary the capping layer thickness and composition to alter the 2DEG electronic properties and tunnel barrier profile, and fabricate and measure devices on other heterostructures.
Chapter 5: Conclusion

As the role of computing in everyday life becomes more ubiquitous, demand for more energy efficient computational schema has spurred interest in novel fundamental logic architectures. Of these, computation through the manipulation of the spin degree of freedom of electrons, spintronics, offers great potential for integration with the highly-developed conventional CMOS technology dominant today. The work in this thesis addresses two major challenges in spintronics: the development of high-quality ferromagnetic semiconductors capable of working a room temperature and the development of all-electron spin injection and detection in various semiconductor systems. The first work presented in this thesis investigates the magnetic properties of gadolinium doped GaN heterojunctions. The giant magnetic moment effect is observed in some, but not all samples and the results are consistent with a defect-mediated mechanism of ferromagnetism. The second work presented involves the design of GaN/AlGaN heterostructures for spin injection and detection and the fabrication of four-terminal, all-electronic spin injection and detection devices on these heterostructures.
These devices were fabricated and initial testing was carried out, although inconclusive.

Directions for further research in GaN/AlGaN spin injection are proposed.
Appendix

*Leakage in Electronic Measurements of 2DEG Heterojunctions*

Initial attempts at performing Hall Effect measurements of the electronic properties of two-dimensional 2DEG heterojunctions demonstrated the perils of naively interpreting Hall Effect results. The Hall mobilities initially reported were much smaller and the sheet densities much higher than reported in literature. An analysis of the discrepancy showed the difficulty of isolating the electronic properties of the 2DEG heterojunction from parallel conduction paths. Although the 2DEG region exhibits an exceptionally high volume density of electrons (number of electrons divided by the 2DEG volume), the sheet density (number of electrons divided by the area of the 2DEG) of the quantum well electrons is much lower than potential parallel conduction paths. The presence of such parallel conduction paths prevents the accurate measurement of the electronic properties in the same way that the presence of a parallel resistance path in a circuit prevents the independent measurement of the resistance of one of the paths. This
is illustrated in figure 36, where a typical voltage divider has been constructed to measure the resistance of $R_{\text{measured}}$. In figure 36a, there is no parallel conduction, so that a measurement of the voltage over the known resistor, $R_{\text{known}}$, allows the resistances of of $R_{\text{measured}}$ to be calculated in the standard way. In figure 36b, the presence of of $R_{\text{parallel}}$ complicates the measurement. In this case the resistance which is calculated using standard techniques will not give the value of $R_{\text{measured}}$, but rather the parallel resistance of $R_{\text{measured}} \times R_{\text{parallel}}/(R_{\text{measured}} + R_{\text{parallel}})$. If the value of $R_{\text{parallel}}$ is unknown, the true value of $R_{\text{measured}}$ cannot be determined. When the value of $R_{\text{parallel}}$ is much larger than $R_{\text{measured}}$, this problem contributes little error to the intended measurement, but otherwise parallel conduction is a major issue. Although the Hall measurement technique is more complicated, the essential problem remains the same.

![Diagram](image)

Figure 36. A parallel resistor interferes with a classic voltage divider measurement.

In the case of III-Nitride heterojunctions, several sources of parallel conduction may be suspected. For example, since the thickness of nominally undoped buffer layers
(often ~ 100 μm) can easily be two orders of magnitude greater than the 2DEG thickness (~ 1 μm according to modeling). Thus, even relatively low levels of unintentional doping due to incorporation of oxygen or other defects into the buffer layer material may become significant. Conductive layers at the heterojunction interfaces may also prevent the accurate determination of the 2DEG electronic properties. Possible conductive layers may also occur at buried polar heterojunctions (2DEG and 2DHG structures incorporated into the growth structure), at interfaces between the alumina wafers typically used as substrates for nitride templates, and on the surface of the film. These buried conductive layers may very well be contacted is using soldered indium contacts or the eutectic spiking contacts commonly used to contact buried 2DEGs in conventional AlN/GaN heterojunctions. Since the depth of penetration of these metal diffusion contacts is often poorly characterized, buried layers may be unintentionally contacted. Finally, conductive layers on the surface of the sample may occur if photolithography has been used to define contacts for Hall and residue from the photoresist has not been adequately cleaned. An awareness of the problem combined with a thorough solvent clean has been enough to minimize this problem in the author’s experience.

A reliable approach to accurately determine 2DEG electronic properties should be to deposit contacts on the surface without annealing to form a eutectic. The decreased penetration of the contact into the sample minimizes contact to buried layers of charge not in the 2DEG. Sputter deposition or electron beam evaporation of standard eutectic layer contacts without the usual annealing have proved effective. In addition to minimizing the depth of contact diffusion into the heterojunctions, the problem of parallel
conduction may be partially eliminated through careful band engineering of the buffer layer growth. For example, in a AlN/GaN 2DEG, one may for a layer largely free of carriers by inserting an AlN back barrier approximately 100 nm into the sample. The insertion of this layer causes the semiconductor bands to tilt, providing a driving force to remove carriers from the region between the 2DEG and the back barrier, as shown in figure 37. Unless deposited contacts penetrate to the back barrier, the 2DEG should be effectively electronically isolated from any carriers present in the buffer.

![BandEng Simulation of a GaN/AlN 2DEG with AlN Back Barrier](image)

Figure 37. An AlN/GaN 2DEG heterojunction with a 1 nm AlN back barrier at a depth of 115 nm.
References