ELECTRICAL TRANSPORT STUDIES OF MOLECULAR BEAM
EPITAXY GROWN (GA,MN)AS EPILAYERS AND
HETEROSTRUCTURES

A dissertation in
Physics
by
Meng Zhu

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Submitted in Partial Fulfillment
of the Requirements
for the Degree of

Doctor of Philosophy

December 2008
The dissertation of Meng Zhu was reviewed and approved* by the following:

Nitin Samarth  
Professor of Physics  
Dissertation Advisor, Chair of Committee

Peter Schiffer  
Professor of Physics

Julian D. Maynard  
Professor of Physics

Srinivas Tadigadapa  
Associate Professor of Electrical Engineering

Jayanth R. Banavar  
Professor of Physics  
Head of the Department of Physics

*Signatures are on file in the Graduate School.
Abstract

Diluted magnetic semiconductors (DMS) grown by molecular beam epitaxy have been drawing attention in the context of emerging spintronics, which utilizes electron spins to develop devices with new functionalities. The canonical DMS – (Ga,Mn)As – has been on center stage for almost a decade, and extensive efforts have been dedicated to understanding its hole-mediated ferromagnetism, optimizing growth and annealing conditions to achieve higher-Tc, studying the magneto-transport, exploiting its abundant magnetic anisotropy, and so on.

This dissertation focuses on three aspects of the study of (Ga,Mn)As: (1) Magneto-transport under hard magnetization reversal; (2) Electrical noise properties; and (3) Exchange-biasing and spin-dependent transport in (Ga,Mn)As/MnAs hybrid structures.

The first chapter provides the motivation for this dissertation and introduces several aspects of the current understanding of (Ga,Mn)As. Both the theoretical models and experimentally established observations are reviewed, focusing on the magnetic and transport properties of (Ga,Mn)As epilayers. Next, the hybrid ferromagnetic metal/semiconductor heterostructures are introduced. As an excellent candidate for making these heterostructures, the semi-metal MnAs is reviewed in terms of its structural and magnetic properties, which are essential for making the exchange-biased devices described in Chapter 5 and Chapter 6.

The second chapter describes the experimental techniques encompassed in the scope of this dissertation. Several important techniques, such as MBE growth, device patterning, magnetometry and transport measurements are discussed.

The third chapter reports the first experiment in this dissertation, which describes the longitudinal magnetoresistance (MR) anomalies of a (Ga,Mn)As epilayer experiencing hard axis magnetization reversal in an perpendicular magnetic field. By probing the MRs for currents running along different crystallographic directions, the origins of these
anomalies are identified, and a model based on magneto-impurity scattering is applied to simulate the MR curves at different temperatures.

The second experiment, described in the fourth chapter, focuses on low frequency electrical noise measurements in (Ga,Mn)As epilayers with different Mn concentrations. The temperature-dependent noise measurements show $1/f$-like noise spectra in all the samples, with no observation of any anomalies across the Curie temperatures. However, in the less conducting sample, we find an increase of the integrated noise (over the span 125mHz-11Hz) in the temperature range from 6K to 29K, accompanied by random telegraph noise (RTN) in the time domain. From the magnetic field dependence of the RTN, we infer the existence of nanoscale magnetic clusters, whose fluctuations modulate hole transport.

The fifth and sixth chapters are dedicated to the study of exchange-biasing and spin-dependent transport in (Ga,Mn)As/MnAs hybrid heterostructures. We demonstrate the fabrication of exchange biased devices based upon (Ga,Mn)As by using a hard ferromagnetic metal (MnAs) overlayer that is exchange coupled with a magnetically softer (Ga,Mn)As underlayer. This discovery of exchange biasing offers a new testbed for studying exchange coupling between metals and semiconductors and opens up opportunities for engineering the coercivity of ferromagnetic semiconductors for device applications. In Chapter 5, we first discuss vertical transport in MnAs/(Ga,Mn)As bilayers, demonstrating a "self-exchange biased" spin valve effect in the current-perpendicular-to-the-plane geometry. Next, in Chapter 6, we discuss the transport properties of (Ga,Mn)As-based exchange-biased MTJs, wherein the tunnel magnetoresistance shows evidence for an exchange-spring configuration in the exchange biased (Ga,Mn)As layer.
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Glossary of Acronyms

AFM: Atomic force microscopy
AHE: Anomalous Hall effect
AMR: Anisotropic magnetoresistance
BEP: Beam equivalent pressure
CMR: Colossal magnetoresistance
CPP: Current perpendicular to plane
CVD: Chemical vapor deposition
DFT: Density functional theory
DOS: Density of states
DW: Domain wall
EB: Exchange biasing
EPMA: Electron probe micro micro analyzer
FDT: Fluctuation-dissipation theory
FET: Field effect transistor
FMR: Ferromagnetic resonance
FMS: Ferromagnetic semiconductor
GMR: Giant magnetoresistance
IVC: Inner vacuum can
LED: Light emitting diode
MBE: Molecular beam epitaxy
MFT: Mean field theory
MIT: Metal-insulator transition
MR: Magnetoresistance
MRAM: Magnetic random-access memory
MTJ: Magnetic tunnel junction
PBN: Pyrolytic Boron Nitride
PDF: Probability density function
PID: Proportional-Integral-Derivative
PIXE: Particle-induce X-ray emission
PHE: Planar Hall effect
PSD: Power spectral density
PDW: Partial domain wall
QW: Quantum well
RHEED: Reflection high energy electron diffraction
RIE: Reactive ion etching
RTN: Random telegraph noise
SDM: Single domain model
SIMS: Second ion mass spectroscopy
SQUID: Superconducting quantum interference device
STM: Scanning tunneling microscopy
TAMR: Tunneling anisotropic magnetoresistance
TEM: Transmission electron microscopy
TLF: Two level fluctuator
TMR: Tunneling magnetoresistance
UHV: Ultra high vacuum
VCA: Virtual crystal approximation
XRD: X-ray diffraction
Acknowledgments

First, I would like to thank my thesis advisor, Professor Nitin Samarth, who guided me throughout the course of my Ph.D. His wide knowledge in physics, deep insights into scientific problems and enthusiasm for new ideas have encouraged me to develop the abilities essential for doing research. More personally, his guidance, patience and kindness made my time at Penn State enjoyable and memorable.

My thanks also go to my laboratory colleagues. I would like to thank Dr. Gang Xiang, Dr. Xia Li, Dr. Khalid Eid, Dr. Keh-Chiang Ku, Dr. Wen-hua Wang and Dr. Oleg Maksimov for imparting their valuable experimental skills. Thanks go to Dr. Partha Mitra, Benjamin Cooley, Andrew Balk, Bill Fadgen, Ming Yang, Joon Sue Lee, Dave Rench, Jing Liang and Krystaufeux Williams for their encouragement and help both inside and outside the lab. We spent many unforgettable times together. I am also grateful to Professor Peter Schiffer at Penn State and his group members for various collaborations: Dr. Ben-Li Sheu and Mark Wilson spent many hours measuring the magnetic properties of the samples, and Dr. Xianglin Ke and Jie Li shared many stimulating ideas. I also thank Michelle Smith at graduate writing center for helping me edit this dissertation.

Additionally, I would like to thank our external collaborators: Dr. Jens Müller, Prof. Stephan von Molnár and Prof. Peng Xiong at Florida State University (FSU) for their help on noise measurement; and Prof. David Awschalom at the University of California, Santa Barbara (UCSB) and his group members, Nathaniel Stern and Sayantani Ghosh, for the collaborations on the spin-Hall projects.

I acknowledge ONR and NSF for their generous funding support of the research in this dissertation.

Above all, I am grateful to my family and friends for their endless support over the years. My thanks also go to Liangchun Yu for her encouragement and companionship.

I dedicate this dissertation to my parents.
Chapter 1

Introduction

1.1 Emerging spin electronics

The era of semiconductor electronics and today’s information technology started with the invention of the transistor by Shockley, Bardeen and Brattain in 1947, a discovery later awarded the 1965 Nobel Prize in Physics [1]. Through the manipulation of the charge properties of electrons (or holes) in semiconductors, a variety of electronic devices have been introduced, and they play an important role in our daily life. The famous “Moore’s law” states that the number of transistors on a chip will double about every two years. Ever since Moore’s prediction in 1965, the semiconductor industry has followed this trend quite well for over 40 years. Today, a 45 nm generation processor has been put into production by Intel, utilizing a revolutionary high-k dielectric and a metal gate in the transistor to replace the problematic silicon dioxide gate dielectric and poly-silicon gate [2]. On the road map of silicon technology, 32 nm technology will be implemented in the next few years, with over two billions of transistors on a single chip [3].

In parallel with these development, the discovery of giant magnetoresistance (GMR) by Fert and Grünberg (the 2007 Nobel Prize in Physics) [4], which exploits the spin properties of electrons, has greatly boosted the capacity of magnetic storage devices, and it has also become one of the key components of information technology. In the data storage market, the capacities of hard disk drives have expanded significantly by shrinking each bit to the nanoscale. New hard disk drive architectures, such as perpendicular recording using vertically aligned magnetic bits, have to be employed to overcome the constraints imposed by the superparamagnetic effect with reducing magnetic grain size [5]. However, we may ask, is there a limit for the scaling of transistors as the feature size is approaching the regime of quantum effects? And are there any alternatives to
keep the momentum of increasing processing capability and storage density? Is it possible to utilize both the spin and charge properties of electrons in order to realize new functionalities of devices and circumvent the limits of ever-decreasing physical size?

Figure 1.1. Roadmap for emerging semiconductor spintronics

One of the many approaches to the next generation of technology is semiconductor spintronics, which harnesses the other degree of freedom – spin – of the already easily controlled carriers in conventional semiconductors. With this concept, we may be able to combine the capability of mass storage and information processing at the same time. Alternatively, the injection of a spin-polarized current into a semiconductor and control of the spin state of the carriers may offer a new direction for quantum information science, in which the computational algorithm is based on the superposition and entanglement of quantum states. A number of new devices, such as spin field effect transistors (spin-FET), magnetic bipolar transistors, spin light emitting diodes (spin-LED), spin qubits and so on, have been proposed or realized. The nature of carrier mediated magnetism makes it easy to achieve optical/electrical control of ferromagnetism. From the technology aspect of view, it can be readily integrated into the existing semiconductor technology without significant modifications.
1.2 Basics of magnetism

Magnetism has been a fascinating topic for thousands of years, dated back to about 1040 AD when the first magnetic device for navigation was mentioned in Chinese literature [13]. Today’s understanding of magnetism is based on the concept of spin which arises from the relativistic description of an electron. This concept results in the spin magnetic moment as well as the orbital magnetic moment due to the motion of electrons.

1.2.1 Fundamental terms of magnetism

In classic electromagnetism, a magnetic moment can be calculated assuming a current loop $I$ with a finite area. The magnetic moment $\mu$ is the product of current $I$ and the integration of the vector area:

$$ \mu = I \int dA $$ (1.1)

The magnetization $M$ is defined as the total magnetic moment per volume unit:

$$ M = \mu \frac{N}{V} $$ (1.2)

When a magnetic field $H$ is applied to a material, the response of the material is called magnetic induction, or magnetic flux density, $B$. The relationship between $B$ and $H$ is a characteristic property of the material. In vacuum, $B = \mu_0 H$, where $\mu_0$ is the magnetic permeability of free space. Inside a magnetic material, however, $B$ also depends on the magnetization of the material:

$$ B = \mu_0 (H + M) $$ (1.3)

The magnetization $M$ of a material is subject to change when it is exposed an external magnetic field $H$: $M = \chi H$, where $\chi$ is the magnetic susceptibility to characterize the response of $M$ to $H$. In this case,

$$ B = \mu_0 (1 + \chi)H = \mu_0 \mu_r H $$ (1.4)

with $\mu_r = 1 + \chi$ being the relative permeability.

1.2.2 Classification of magnetism

(1). Diamagnetism:

Diamagnetism arises from the induction of magnetic dipoles in an external field. The induced dipole moments are oriented anti-parallel to the field due to Lenz’s rule so the
diamagnetic susceptibility is negative: \( \chi^{\text{dia}} = \text{constant} < 0 \). Diamagnetism is a property of all materials. Superconductors below their critical temperatures are ideal diamagnets with \( \chi^{\text{dia}} = -1 \).

(2). **Paramagnetism:**

Paramagnetism occurs in materials with permanent magnetic dipoles, but they orient randomly due to thermal fluctuations (Fig. 1.2). Paramagnetic materials exhibit a small net magnetic moment when an external magnetic field is applied to align the dipole moments. The susceptibility of a paramagnetic material is positive: \( \chi^{\text{para}} = \chi^{\text{para}}(T) > 0 \), and it depends on temperature. A typical \( \chi^{\text{para}} \) is between \( 10^{-3} \sim 10^{-5} \).

![Paramagnetism, Ferromagnetism, Anti-ferromagnetism, Ferrimagnetism](image)

**Figure 1.2.** Illustrations of magnetic dipoles ordering in different magnetic materials. (a) Paramagnetism (b) Ferromagnetism (c) Anti-ferromagnetism (d) Ferrimagnetism.

The magnetic moments can be localized or itinerant in nature:

- **Localized moments**: They are usually caused by electrons of a partially filled inner shell of an atom, such as \( 4f \) electrons in the rare earth metals. The paramagnetism of this class of materials is called **Langevin paramagnetism**, in which the temperature dependent susceptibility \( \chi^{\text{Langevin}}(T) \) is described by the Langevin function. In the high temperature limit, \( \chi^{\text{Langevin}}(T) \) is approximately proportional to the
inverse of the temperature (Curie Law):

$$\chi^{Langevin}(T) = C/T$$  \hspace{1cm} (1.5)

• **Itinerant moments**: In paramagnetic metals, electrons are nearly free and each of them carries a magnetic moment of $1\mu_B$, with $\mu_B$ being the Bohr magneton. The application of an external field will change the density of states of spin-up and spin-down electrons at the Fermi energy, and resulting in an imbalance in the population of spin-up and spin-down electrons. A net magnetic moment develops proportional to the number difference of the two types of electrons. This is called *Pauli paramagnetism*. The corresponding susceptibility is independent of temperature. $\chi^{Pauli}$ is usually much smaller than $\chi^{Langevin}$.

(3). **Ferromagnetism**:

Ferromagnetism is one of the most important types of magnetic ordering. Ferromagnetism, anti-ferromagnetism and ferrimagnetism all fall into the category of collective magnetism, which comes from strong interactions between the magnetic dipole moments. In a ferromagnetic material, magnetic moments exhibit a preferential orientation below a certain critical temperature $T_C$ (Curie temperature), as shown in Fig.1.2(b). Above $T_C$, thermal fluctuations overcome the exchange coupling between magnetic moments, and the material displays paramagnetic ordering. Ferromagnets often have very large susceptibilities ($\chi^{Ferro} > 1000$), and they usually display a spontaneous magnetization below $T_C$.

(4). **Antiferromagnetism**:

In an antiferromagnetic material, the magnetic moments divide into two sublattices with anti-parallel orientations as shown in Fig.1.2(c). The critical temperature for antiferromagnetism is called the Néel temperature $T_N$. Below $T_N$, an antiferromagnet displays a vanishing net magnetic moment due to the cancellation of magnetic moments in the opposite directions.

(5). **Ferrimagnetism**:

Ferrimagnets are somewhat like antiferromagnets, in that the magnetic moments of two sublattices align antiparallel. However, in a ferrimagnet, one type of sublattice has larger moment than that of the other (Fig.1.2(d)), so the material has a net overall mag-
netic moment below the Curie temperature. Like ferromagnetic materials, ferrimagnets tend to concentrate magnetic flux in their interiors, and they have large susceptibilities.

1.3 Dilute magnetic semiconductors (DMS) (Ga,Mn)As

One of the prerequisites to make semiconductors suitable for spintronic applications is achieving magnetic ordering. The hunt for magnetic semiconductors can be traced back to the late 1960s, when studies focused on the first generation of magnetic semiconductors, such as europium chalcogenides (EuSe, EuTe and EuO) and semiconducting Cr spinels (CdCr$_2$S$_4$), which have a periodic array of magnetic elements [14]. However, despite the interesting properties resulting from the interaction between band electrons and magnetic ions, the difficulties in crystal growth impaired their further applications.

Later on, the idea of introducing a fractional population of magnetic atoms into non-magnetic semiconductor lattices, stimulated the investigations of “diluted magnetic semiconductors” (DMS). Studies of DMS began with incorporating Mn atoms into II-VI compound semiconductors, such as CdTe and ZnSe, because the valence of the cations (2+) matches that of the Mn atoms [15], making it easier for sample preparation. However, in II-VI DMS’s, the magnetic interaction is dominated by the antiferromagnetic coupling between Mn atoms, resulting in paramagnetic, antiferromagnetic or spin-glass behavior. Although ferromagnetic ordering has now been achieved in (Cd,Mn)Te [16] and (Zn,Mn)Te [17], the transition temperatures are low, and not until recently was room-temperature ferromagnetism reported in (Zn,Cr)Te [18].

The discovery of carrier-induced ferromagnetism in Mn-doped zinc-blende III-V semiconductors, such as InAs [19] and GaAs [20], opened up new areas for exploring DMS in a more promising way because of their relatively high $T_C$ and wide applications of III-V compounds in electronic and optoelectronic devices. The unprecedented opportunities for III-V DMS lie in the fact that the magnetic properties are strongly influenced by the holes in the valence band, allowing one to exploit the well-developed methods for changing the carrier concentration in semiconductors by applying an electric field or shining light.

Although the search for a new DMS with a higher transition temperature is ongoing, (Ga,Mn)As is no doubt the most extensively studied DMS, and it serves as a textbook example of a ferromagnetic semiconductor. In the following sections, I will give an overview of this canonical DMS. I will start with the lattice structure of (Ga,Mn)As and the defects therein. Then I will address the current understanding of the origin
of ferromagnetism. Finally, the experimental aspects, such as the preparation/post-growth annealing conditions, and the magnetic/electrical properties of (Ga,Mn)As will be discussed.

1.3.1 Lattice structure

(Ga,Mn)As has the zinc-blende lattice of its GaAs host. Fig.1.3 illustrates the structure of the (Ga,Mn)As lattice and two types of defects that are usually involved. Because Mn has two valence electrons, when it takes the Ga site and bonds with As, it acts as an acceptor, providing free hole carriers. At the same time, it also provides a local magnetic moment with $S = 5/2$, which comes from the strongly localized $3d^5$ electrons of Mn. The itinerant holes then mediate a ferromagnetic interaction between the local moments, resulting in a ferromagnetic ordered state. This simple yet powerful picture has been proved by many experiments, showing a strong correlation between $T_C$ and the carrier concentration [21].

![Zinc-blende lattice structure of (Ga,Mn)As](image)

Figure 1.3. The zinc-blende lattice structure of (Ga,Mn)As. Substitutional Mn atoms take Ga sites, providing magnetic moments as well as hole carriers; Two types of defects exist: As$_{Ga}$ is an As anti-site, Mn$_I$ is a Mn interstitial. Both of them serve as a double donor, compensating two holes. From Ref.[10]

In order to achieve collective ferromagnetic order, a minimum of about 2% of the substitutional Mn atoms has to be incorporated to provide sufficient hole carriers to mediate the ferromagnetic coupling [10]. To circumvent the obstacle that the solubility
of Mn in GaAs crystals at thermal equilibrium is only about 0.1%, low temperature molecular beam epitaxy (LT-MBE) was applied to drive the growth condition far from equilibrium. The low growth temperature ensures there is not enough thermal energy to form the more stable metallic MnAs clusters. However, the side effect of the LT-MBE is that a large number of point defects occur, and they compromise the ferromagnetic ordering. The most important defects are the Mn interstitials (MnI) and As anti-sites (AsGa). Both of them act as a double donor, which compensates two holes in the lattice and in turn has a severe impact on the electric and magnetic properties of (Ga,Mn)As.

Mn interstitials, as illustrated in Fig.1.3 were first identified by the combined channel Rutherford backscattering and particle-induced X-ray emission (PIXE) experiment [22]. It was found that up to 20% of Mn resides on interstitial positions in a highly doped as-grown sample. The population of these metastable MnI can be significantly reduced by thermal annealing, which drives the Mn interstitials to the free surface where they will get passivated. The diffusion of MnI depends strongly on the nature of the surface of the (Ga,Mn)As layer. The diffused MnI need to be passivated at the surface, and post-growth annealing in N2, O2 or air are employed to assist this passivation [21, 23]. However, even a few nanometers of a GaAs capping layer on top of the (Ga,Mn)As will reduce the diffusion process [24]. The outdiffusion of MnI effectively increases the free hole density and correspondingly enhances the TC.

Asenic anti-sites, however, can not be removed by post-growth annealing, and they are stable up to 450°C [25], beyond the temperature at which precipitation of MnAs clusters start to form. The existence of AsGa is mainly a combined aftermath of the non-equilibrium growth condition and As overpressure, which is used in the MBE growth to prevent the evaporation of As atoms from the substrate. AsGa will also compensate two holes and degrade the ferromagnetic ordering in (Ga,Mn)As. Care must be taken during the (Ga,Mn)As growth to minimize the incorporation of AsGa. Studies have suggested using As2 dimer instead of As4 tetramers [26, 27] or optimizing the As/Ga flux ratio to effectively suppress the formation of AsGa [28, 29].

The lattice constant of the (Ga,Mn)As layers is affected by strongly by the impurity densities. Asymmetric X-ray diffraction (XRD) showed that the (Ga,Mn)As thin films were not relaxed [3]. The lattice constant mismatch between the (Ga,Mn)As and GaAs substrate induces strains that determine the magnetic easy axes and anisotropic properties of (Ga,Mn)As. Theoretical efforts based on density-functional theory (DFT) predict that the substitutional Mn may also lead to very small reduction in the lattice constant [30], while AsGa and MnI produce a large expansion of the GaAs lattice. Several ex-
periments have confirmed that both \( \text{As}_{Ga} \) and \( Mn_I \) lead to a significant expansion of the lattice \([31, 32, 33, 34]\), and annealing results in a significant reduction of the lattice constant due to the out-diffusion of \( Mn_I \).

### 1.3.2 Theory of ferromagnetism in (Ga,Mn)As

Extensive experiments have already established that the robust ferromagnetism in (Ga,Mn)As is a result of hole-mediated exchange interactions between \( Mn^{2+} \) ions, since the incorporated Mn atoms serve as both magnetic moments and acceptors \([35, 36, 37]\). A \( Mn^{2+} \) ion has a spin \( S = \frac{5}{2} \) due to the half-filled \( d \)-shell. The \( d \) orbitals of \( Mn^{2+} \) cations strongly interact with \( sp \)-band carriers in the partially filled valence band, and as a result, localized \( Mn^{2+} \) moments are aligned at long range.

Although the detailed mechanism of this \( p-d \) interaction is still under debate, the phenomenological kinetic exchange model based on the mean-field theory is considered to be the most successful one to explain a variety of magnetic properties in (Ga,Mn)As, such as Curie temperature, magnetic anisotropy \( etc. \). This model was proposed by Dietl \textit{et.al.} \([36, 37]\), adapted from the Zener model \([38]\). This mean-field model introduces a Ginzburg-Laudau free energy density: \( F[M(r)] \). The free energy of a ferromagnetic semiconductor consists of two terms:

\[
F[M(r)] = F_S[M(r)] + F_c[M(r)]
\]  

(1.6)

where \( F_S[M(r)] \) is the free energy density of the Mn spins in the absence of the carriers, and \( F_c[M(r)] \) is the free energy density of the carriers in the presence of the Mn spins. The asymmetry in the treatment of the carriers and of the spins is based on an adiabatic approximation, in which the dynamics of the spins in the absence of the carriers is assumed to be much slower than that of the carriers.

Assuming a spatially uniform magnetization \( M \), \( F_S[M(r)] \) in a magnetic field \( H \) can be written as:

\[
F_S[M] = \int_0^M dM_0 h(M_0) - MH
\]  

(1.7)

Here, \( h(M_0) \) is the inverse function of \( M_0(h) \), where \( M_0 \) is the experimental macroscopic magnetization of the spins in the absence of carriers in the field \( h \). Near \( T_C \) and for \( H = 0 \), \( M \) is small enough to take \( M_0(T, h) = \chi(T) h \), where \( \chi(T) \) is the susceptibility
of localized spins in the absence of carriers. Now $F_S[M]$ is simplified as:

$$F_S[M] = \frac{M^2}{2\chi(T)}$$  \hspace{1cm} (1.8)$$

This equation shows that the increase of $F_S$ with $M$ due to antiferromagnetic Mn-Mn interaction slows down with lowering temperature because of the increase of $\chi(T)$. On the other hand, the assumed uniform magnetization $M$ results in a giant Zeeman spin splitting in the valence band. The energy of the carriers, $F_c[M]$, decreases with $|M|$: $F_c[M] - F_c[0] \sim -M^2$. At a certain temperature ($T_C$), the two energies may balance and a minimum total energy $F[M]$ is achieved. Further reduction of temperature leads to spontaneous spin splitting and spin polarization, signalizing the appearance of ferromagnetic order.

Figure 1.4. A simple picture of the hole-mediated ferromagnetism in (Ga,Mn)As. Localized Mn spins are antiferromagnetically coupled to free hole carriers via $p$-$d$ exchange interactions, resulting in ferromagnetic ordering between adjacent Mn spins.

A simple picture of the mean-field model is shown in Fig.1.4. Strong $p$-$d$ exchange interactions antiferromagnetically couple the Mn spins and the hole spins. This results in a parallel alignment of adjacent localized Mn spins that overcomes the antiferromagnetic coupling between them (superexchange). The hole spins tend to spread out because of the kinetic or band energy cost of localization. Long range ferromagnetic order can then be achieved by the coupling between the local Mn moments and the itinerant holes.
in the valence band. In spite of the simplicity of the mean-field model, it has been successfully used to describe the $T_C$ in (Ga,Mn)As as a function of Mn concentration and carrier density. Also because this approach focuses on the degrees of freedom that are important for magnetism, such as the orientations of Mn local moments and occupation numbers of acceptor levels near the top of the valence band, it helps in explaining many magnetic, transport and optical properties of (Ga,Mn)As.

Figure 1.5. (a) Schematic illustration of two simplifying approximations – virtual crystal approximation (VCA) and mean field theory (MFT) – that are valid for systems with long-range Mn-Mn coupling. (b) A schematic phase diagram for carrier-induced ferromagnetism in DMS as a function of exchange coupling strength relative to the band Fermi energy $\epsilon_F$ (y-axis), and the carrier concentration relative to the Mn concentration (x-axis). From Ref.[10]

Although the prediction of high-$T_C$ materials based on mean-field model asks for systems with strong exchange interactions and a large valence-band density of states, a limitation may occur when two approximations for the mean-field model fail [10]. The first approximation is called virtual crystal approximation (VCA), which replaces the random distribution of Mn ions on the host lattices by a continuum with the same density. Therefore the random alloy can be modeled as an effective perfect crystal. The second approximation is the mean-field theory (MFT), in which the fluctuations in the relative orientations of Mn ions in different part of the system are not taken into account. A schematic of these two approximations is shown in Fig.1.5. If the exchange interaction between Mn and valence band holes is too strong, the acceptor band levels are more localized, only showing coupling to an individual spin. This leads to the failure of VCA, and the carrier-induced magnetism enters the regime of strong coupling collective,
as shown in the phase diagram in Fig. 1.5. On the other hand, a large valence-band density of states implies a larger effective mass, also leading to more localized acceptor levels. Additionally, when the carrier density increases, the average distance between the carriers becomes comparable to that between the magnetic ions so the RKKY interaction dominates. The sign of the RKKY exchange interaction oscillates rapidly as a function of spatial distance, resulting in the failure of MFT (RKKY collective regime). In summary, the prediction from the mean-field model for higher $T_C$ cannot continue because strong exchange interactions will eventually localize holes, and a high density of holes will lead to RKKY interactions that vary rapidly in space. The phase diagram derived from the above analysis suggests the largest ferromagnetic transition temperature may occur near where the three regions intersect.

Recent reviews on contemporary understanding of ferromagnetism in (Ga,Mn)As are given by Macdonald et. al. [10] and Jungwirth et. al. [11].

1.3.3 Preparation of (Ga,Mn)As

As mentioned in the previous section, (Ga,Mn)As thin films have to be grown at low temperatures to overcome the low solubility of Mn in GaAs lattices. Here, I address some important parameters concerning the (Ga,Mn)As low temperature growth.

1. Growth temperature and Mn flux:

The growth temperature and the Mn flux are the two most important parameters in (Ga,Mn)As growth. Depending on how much Mn is going to be incorporated (or Mn flux value), the growth temperature varies from 170°C to 250°C. Generally speaking, the higher the Mn flux, the lower the growth temperature. Ga$_{1-x}$Mn$_x$As epilayers with Mn composition ($x$) of up to 20% have been grown with careful control of growth temperatures [39, 40]. Although lowering the growth temperature allows more Mn to be incorporated, there is a lower temperature limit below which the number of defects will increase rapidly and start to compromise the ferromagnetic order. A large portion of the incorporated Mn for $x > 10\%$ will exist as interstitials, which do not contribute to, but rather counteract ferromagnetism. On the other hand, the upper temperature is limited by the formation of MnAs clusters. A optimal temperature has to be chosen to achieve a balance between incorporating more Mn and minimizing the defect formations.

The growth of (Ga,Mn)As can be clearly monitored by reflection high-energy electron diffraction (RHEED). The typical surface reconstruction pattern during the low-temperature (Ga,Mn)As growth is (1×2) as shown in Fig. 1.6(a)(b). For samples grown
just above or at the upper temperature boundary (e.g. the temperature is too high for a certain Mn flux), the RHEED shows a very clear $(1\times2)$ pattern before a certain critical thickness $t_c$ is grown and starts to change into a spotty pattern typical of 3D (three-dimensional) growth beyond that thickness. Fig.1.6(c)(d) show one example of such a spotty pattern, when the 2D (two-dimensional)-3D growth transition takes place after 2min of growth. This transition from the smooth 2D growth to the rough 3D growth indicates the onset of MnAs cluster formation. A systematic study of this MnAs clustering shows that the critical thickness depends on the substrate temperature as well as the Mn flux. For a given substrate temperature, there is a maximum concentration of Mn that can be incorporated. Excessive Mn atoms will segregate on the surface, and when the amount reaches a critical value, the 2D-3D transition occurs \cite{111}. The optimum condition is where the samples are grown close to the 2D/3D boundary, so the careful control of growth temperature, depending on the Mn flux, as well as the film thickness is important.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{rheed_patterns.png}
\caption{RHEED patterns during the growth of (Ga,Mn)As. (a) $1\times$ pattern for beam along [110] and (b) $2\times$ pattern for beam along [1\bar{1}0]. (c) and (d) show the spotty RHEED patterns when MnAs second phase occurs.}
\end{figure}
It has also been suggested that when the growth of (Ga,Mn)As is initiated, the impingement of the hot source beams on the substrate [28] and the change of absorption/emission characteristics of the growing layer may increase the real growth temperature from its standby temperature [42]. When the growth temperature is close to the upper limit, this unexpected increase of substrate temperature may also lead to the precipitation of the MnAs second phase. With the band-absorption spectroscopy capability, we can monitor the substrate temperature in real time and adjust the heater to maintain a stable growth temperature.

(2). As/Ga stoichiometry:

Arsenic antisite defects compensate holes and they can not be eliminated by post-growth annealing. The density of As$_{Ga}$ can reach up to 10$^{20}$ cm$^{-3}$ and become comparable to a typical Mn doping level. To reduce the concentration of As anti-sites, As dimer (As$_2$) can be used rather than the conventionally used As tetramer (As$_4$). Because the surface lifetime of As$_2$ is less than As$_4$, the probability of incorporating As$_{Ga}$ is much reduced [27] [28]. An alternative for inhibiting the formation of As$_{Ga}$ is to adjust the As to Ga flux ratio [43]. A systematic study of the effect of As$_{Ga}$ incorporation on the electrical and magnetic properties was given by Myers et.al. [29] where they grew low-Mn doping (1% ~ 2%) samples with different As:Ga flux conditions.

It is found that in an As-rich condition, as-grown films exhibit maximum smoothness, while showing a high density of As$_{Ga}$ and suppressed $T_C$. In the Ga-rich condition, three dimensional growth becomes dominant with the accumulation of excess Ga on the surface, leading to the increase of surface roughness. The Ga-rich surface also suppresses substitutional Mn incorporation, giving low $T_C$. When the As:Ga ratio is in the range of 9~10, the best stoichiometric condition is achieved, where the hole concentration and $T_C$ are maximized. In practice, we chose a As:Ga flux ratio between 10~12 to meet this stoichiometric condition and the requirement for surface smoothness.

(3). Post-growth annealing:

(Ga,Mn)As samples grown at moderate Mn doping level will inevitably contain Mn interstitials defects. To achieve a higher $T_C$, post-growth annealing is necessary to out-diffuse the $Mn_I$ and increase the hole density. Fig[1.7] shows the effect of annealing on a 120nm Ga$_{1-x}$Mn$_x$As ($x=8.3\%$) in Nitrogen gas at 250$^\circ$C. The $T_C$ is substantially enhanced from 65K to 110K upon only 30 minutes of annealing. The resistivity of the annealed sample with highest $T_C$ also shows a minimum value, indicating the increase
of the hole density. Longer annealing does not help enhance, but rather suppresses the ferromagnetism as the $t_{\text{anneal}} > 2$ hours. This is presumably due to the short-range clustering of Mn-As complexes with reduced ferromagnetism [44].

Annealing also changes the shapes of $M-T$ curves significantly [45]. The $M-T$ curve for as-grown sample has a kink at a certain temperature below $T_C$, and the magnetization $M(T)$ shows a relatively linear behavior below and above that characteristic temperature with different slopes. The $M(T)$ drops much faster when $T$ is approaching $T_C$. However, an annealing of 30 min or longer transforms the $M(T)$ into a smooth mean-field-like curve typical for other Heisenberg ferromagnets. This is thought to be associated with the reduction of large-scale disorder in the films.

![Figure 1.7.](image)

Figure 1.7. (a) Magnetizations and (b) resistivities as a function of temperature for 120nm (Ga,Mn)As epilayer annealed for different time in N$_2$ at 250°C. From Ref. [46]

On the other hand, the thicknesses of the (Ga,Mn)As films may also influence the effectiveness of annealing. As shown in Fig. 1.8 (a), the thinner film displays a bigger increase of $T_C$ than the thicker sample grown and annealed at identical conditions. The study of a number of samples with different thicknesses but the same doping level shows that the highest $T_C$ in both as-grown and annealed samples occurs in the range of 10nm $\leq t \leq 50$nm ($t$ is the thickness of the epilayer). This thickness dependence on $T_C$ may arise from the difference of $Mn_I$ diffusion near the surface from that in the bulk, or the difference of the kinetics for $Mn_I$ in the proximity of a free surface [47].

Moreover, it is found that a GaAs capping layer of only 10 monolayer thickness on
Figure 1.8. (a) Magnetization as a function of temperature for (Ga,Mn)As samples (Mn=8.5%) of different thicknesses as a comparison of annealing effect. $t = 15 \text{nm}$ (squares) and $t = 50 \text{nm}$ (circles). (b) Magnetization as a function of temperature for 50nm (Ga,Mn)As samples (Mn=9%) with (solid symbols) and without (open symbols) 10 monolayers of GaAs capping layer. From Ref.[46]

Top of the (Ga,Mn)As epilayers will completely suppress the enhancement of $T_C$ from annealing (See Fig.1.8 (b) [24], and annealing in a vacuum of as-grown samples typically does not help increase $T_C$. These experiments demonstrate that the surface structure strongly affects the diffusion of the defects in (Ga,Mn)As. A simple picture is that the out-diffusing Mn$_I$ have to be passivated at the surface. Once they reach the surface and form a reacted layer, an efficient barrier is formed against further out-diffusion [48]. Both the thickness dependence of optimum $T_C$ and the effect of the GaAs capping layer can be explained by this picture. Annealing in a variety of gas ambients such as Nitrogen, Oxygen [49] or air can effectively passivate the Mn$_I$. Also, by properly choosing adjacent layers as efficient sinks for Mn$_I$, such as As capping layers [48] [50] or MnAs layers in proximity [51], the $T_C$ of (Ga,Mn)As epilayers can be greatly promoted. Theoretical analysis on the mechanism of Mn$_I$ diffusion can be found in Ref.[52].

Another way to engineer the $T_C$ finds its application in nanosize devices. By patterning the (Ga,Mn)As layers into nanowires, the exposed free surface at the sidewalls provides more surface area for Mn$_I$ diffusion and leads to an enhanced $T_C$ and reduced resistivity [53]. This method implies the possibility of engineering $T_C$ as well as electrical/magnetic properties on a portion of (Ga,Mn)As-based nano-devices by controlling their free surfaces through nano-patterning.

Practically, the choice of annealing temperature depends on the Mn concentration
and the as-grown growth temperature of the (Ga,Mn)As epilayers. The annealing temperature has to be $5\sim10^\circ$C lower than the growth temperature to prevent the formation of MnAs precipitates. Apart from annealing at the temperature close to the growth temperature, low-temperature annealing ($T_{\text{anneal}} = 170 \sim 190^\circ$C) over a prolonged time (>100 hours) is also employed to optimize the effect of annealing [23]. In this way, the disadvantage of longer annealing time at higher temperatures due to the formation of Mn-As complexes is suppressed, and the maximum amount of $\text{Mn}^+$ are supposed to diffuse out. This low-temperature annealing technique is usually accompanied by in-situ resistivity monitoring. The resistivity falls monotonically with the annealing time, in contrast to the high-temperature annealing, where the resistivity is found to fall initially and then increase after 2 hours of annealing [45]. This decrease of resistivity indeed suggests the increase of free hole carriers and the reduction of disorder. When a saturated value of minimal resistivity is achieved, the highest possible $T_C$ is also obtained for that (Ga,Mn)As epilayer.

### 1.3.4 The role of strain and anisotropy

As mentioned before, the lattice constant of relaxed (Ga,Mn)As is larger than that of GaAs, especially when large quantities of interstitial Mn and As antisites are present. This leads to a compressive strain in the (Ga,Mn)As layer grown on a GaAs substrate. However, tensile-strain (Ga,Mn)As layers can be obtained by growing them on (In,Ga)As buffers [54, 55].

Fig.1.9 shows the magnetization curves for (Ga,Mn)As epilayers grown on (a) GaAs and (b) (In,Ga)As buffers. Clearly, the magnetization easy axis is in plane for the compressive-strained sample and is out of plane for the tensile-strained sample.

Another important property for magnetic materials is magnetocrystalline anisotropy, which describes the dependence of ferromagnetic energy on the magnetization direction with respect to the crystalline axes. Unlike the normal ferromagnet, where the magnetocrystalline anisotropy is caused by the spin-orbit interaction of localized electrons in magnetic $d$ or $f$ shells, the anisotropy energy in (Ga,Mn)As is believed to arise from a strong spin-orbit coupling in the valence band [56]. This leads to rich anisotropic properties in (Ga,Mn)As, such as the reorientations of easy axes as a function of hole density, temperature or strains [57, 58], and the competition of biaxial and uniaxial anisotropy [59].
1.3.5 Magnetic properties of (Ga,Mn)As epilayer

(1). Magnetization:

The magnetic properties of (Ga,Mn)As are usually measured directly using a magnetometer as shown in Fig. 1.9. On the other hand, the Mn concentration can be determined by using an electron probe micro analyzer (EPMA) or second ion mass spectroscopy (SIMS). Thus, the magnetic moment per Mn atom can be calculated. Experimentally, an apparent magnetization deficit has been observed using SQUID magnetometry, showing that the moment per Mn ion is well below the full saturation value of $5\mu_B$ for spin $5/2$ moment [31]. This deficit was found to increase monotonically with increasing Mn
doping ($x$), implying an increasing fraction of the Mn spins that do not participate in the ferromagnetism. Detailed studies using x-ray magnetic circular dichroism (XMCD) has shown that an antiferromagnetic coupling between interstitial and substitutional Mn may account for this deficit [60].

Due to the nature of hole-mediated ferromagnetism, the magnetization of (Ga,Mn)As strongly depends on the hole density. Experiments have shown that both the Curie temperature and coercivity of a (Ga,Mn)As layer can be tuned by applying an external electric field (or gating) to change the hole concentration [61]. In practice, co-doping (Ga,Mn)As with holes using Be modulation doping in a quantum well (QW) structure [62] and wavefunction engineering in heterojunction systems using Mn δ-doping [63] have been developed to increase the effectiveness of the exchange interactions between localized moments and hole carriers. Nevertheless, the simplest way to alter the magnetic properties of (Ga,Mn)As is low temperature annealing. As shown before, annealing effectively increases hole density by reducing the compensation from Mn interstitial defects, and this results in an enhancement of $T_C$. In magnetization hysteresis loops, optimally annealed samples often show an increase of the saturation magnetization as well as a decrease of the coercivity. This change is attributed to the improved ferromagnetic order and the reduction of defect pinning sites that affect the magnetic domain movements.

(2). Magnetic domain state:

Controlling and manipulating magnetic domain wall (DW) have became increasingly hot subjects in recent years due to possible applications in novel memory and logic devices [64, 65, 66]. In (Ga,Mn)As, large single domains, on the scale of hundreds of microns were observed using a magneto-optical imaging technique [67]. In tensile-strained (Ga,Mn)As, a stripe-shape domain pattern was observed using a scanning Hall probe microscope [68]. Because the saturation magnetization ($M_S$) of (Ga,Mn)As is two orders of magnitude lower than in conventional metal ferromagnets, while at the same time the magnetocrystalline anisotropy energies ($K$) and spin stiffness ($A$) are comparable to their metal counterparts [69], (Ga,Mn)As provides an excellent testbed for the study of DW dynamics. The critical current for DW switching in (Ga,Mn)As was found to be two orders of magnitude lower than that in metal ferromagnets [70].

On the other hand, the studies of the DW resistance [71, 72] and the DW motion in (Ga,Mn)As [73, 74] have also advanced our understandings of carrier and spin transport in this nonuniform magnetization background.

Another aspect of domain manipulation is the current-driven magnetization switch-
As the spin-polarized current encounters a local magnetization with an opposite spin orientation, the spins of the carriers will get re-polarized to the new direction. Due to the conservation of angular momentum, the local magnetization moment will also experience a spin-transfer torque, which may result in the reversal of the moment. Current-driven magnetization reversal has been observed in (Ga,Mn)As-based magnetic tunnel junctions (MTJs) \[75\], and efforts are being made to realize spin torque transfer in ferromagnetic metal/semiconductor hybrid devices.

1.3.6 Transport properties of (Ga,Mn)As epilayers

(1). Anomalous Hall Effect (AHE):

As is well known, the normal Hall effect refers to the transverse potential difference (Hall voltage) on the opposite sides of a conductor when a magnetic field is applied perpendicular to the current flow (Fig. 1.10(d)). It arises from the Lorentz force experienced by carriers in a perpendicular magnetic field. The Hall measurement has become a standard method for characterizing semiconductor materials, due to its sensitivity to the type and concentration of charge carriers. The Hall coefficient \( R_H \) is defined as:

\[
R_H = \frac{E_y}{j_x B} = \frac{V_{xy} d}{I_x B} = R_{xy} \frac{d}{B} \quad (1.9)
\]

\[
R_H = -\frac{1}{ne} \text{ (electrons), or } R_H = \frac{1}{pe} \text{ (holes)} \quad (1.10)
\]

where \( d \) is the thickness of the sample. The sign of \( R_H \) depends on the type of the carriers. From Eq. (1.9) and (1.10), the carrier density is determined by:

\[
n(p) = \frac{1}{|e| \text{slope}(\frac{R_{xy}}{B})} d \quad (1.11)
\]

where \( \text{slope}(\frac{R_{xy}}{B}) \) is the slope of the Hall resistance curve. Fig. 1.10(c) shows an example of normal Hall resistance of a Be-doped GaAs epilayer. The hole density calculated from the Hall measurement is \( 1E19/cm^3 \).

In ferromagnets, the Hall resistance will have an additional term due to the spin-dependent scattering between the carriers and the local magnetic moments. The Hall resistance in (Ga,Mn)As is then written as:

\[
R_{xy} = \frac{R_0}{d} B + \frac{R_A}{d} M \quad (1.12)
\]

where \( R_0 \) is the ordinary Hall coefficient, \( R_A \) is the anomalous Hall coefficient, \( d \) is the
Figure 1.10. Hall resistances with a magnetic field perpendicular to the sample plane and the current flow. (a) Anomalous Hall resistance of a compressive-strained (Ga,Mn)As epilayer with in-plane easy axes at different temperatures. (b) Anomalous Hall resistance of a tensile-strained (Ga,Mn)As epilayer with an out-of-plane easy axis. (c) Normal Hall resistance of a 200nm thick high-temperature grown Be-doped GaAs epilayer. (d) Illustration of a Hall measurement geometry: current flowing in the sample plane with a field applied in the perpendicular direction. The Hall voltage $V_{xy}$ measures the potential difference across the current flow.
sample thickness, and $M$ is the perpendicular component of the magnetization \[^{[35]}\]. The $R_A$ term arises from the spin-orbit interactions, from which the spin-up and spin-down carriers are deflected in different directions in the ferromagnet, even in absence of the external field. $R_A$ is found to be related to longitudinal resistance by the equation: $R_A = c\rho \gamma_{xx} \[^{[76]}\]$, where $c$ is a constant. The exponent $\gamma$ indicates two different scattering mechanisms: “skew scattering” for $\gamma=1$ and “side-jump scattering” for $\gamma=2$. $\gamma$ is found to be sample dependent in (Ga,Mn)As, and it can vary between 1 and 2 \[^{[77]}\]. However, for most of the samples showing “metallic” behaviors, the “skew scattering” picture is usually assumed \[^{[78]}\].

Because $R_{xy}$ strongly depends on magnetization at low fields and the anomalous Hall coefficient is proportional to $\rho_{xx}$, the temperature dependent saturation magnetization $M_S$ can be obtained from an Arrott plot, in which $(R_{xy}/R_{xx})^2$ is plotted against $\left[ B/(R_{xy}/R_{xx}) \right]$ at each temperature. $T_C$ can thus be extracted from the condition when $M_S=0 \[^{[6]}\]$. Also, in the low-field region, the dominance of the anomalous Hall term allows one to use Hall resistance as a proxy for perpendicular magnetization since $R_{xy} \propto M$. For example, in Fig.1.10(a), the magnetization of a compressive-strained (Ga,Mn)As epilayer rotates from an in-plane to out-of-plane direction upon the application of the perpendicular magnetic field. The Hall resistance ($R_{xy}$) increases gradually at low fields, like a typical hard-axis magnetization curve, and then it saturates at some points and shows a small linear dependence on $B$ at high fields. As temperature increases, the amplitude of $R_{xy}$ drops due to the weakening of (Ga,Mn)As magnetization. Above the $T_C$, the curve becomes linear, but there is still a large contribution to the Hall resistance from the paramagnetic response of $M$. For the tensile-strained (Ga,Mn)As sample, however, as shown in Fig.1.10(b), $R_{xy}$ displays very sharp transitions with a hysteresis loop, similar to an easy-axis magnetization curve. The different behaviors of the Hall resistance for samples with different easy-axis orientations further testifies that the AHE is proportional to the vertical component of the magnetization, which can be used as a powerful tool to track the magnetization motion in a perpendicular field. This property will be employed in Chapter 3 to study the hard axis magnetization reversal in (Ga,Mn)As.

On the other hand, the AHE makes it difficult to obtain the hole density from the ordinary Hall resistance term at the low field region. To get an accurate value of the hole density, one has to take measurements in the limit where the magnetization is fully saturated: at low temperature and under high magnetic fields \[^{[79]}\]. However, even at this limit, the AHE contribution is not constant, due to the dependence of $R_A$ on $\rho_{xx}$.
The negative longitudinal resistance at high fields has to be taken into account when doing the curve fitting for determining $R_0$ and, in turn, the hole density. There are also several other methods to obtain hole density, such as Raman-scattering techniques \[80\] and electrochemical capacitance-voltage profiling \[81\].

(2). Planar Hall Effect (PHE):

The planar Hall effect (PHE) refers to the transverse voltage developed in an in-plane field. A “giant” PHE was first discovered by Tang et al.\[82\] in (Ga,Mn)As epilayers subjected to a in-plane magnetic field. The basic idea for PHE is as following: as the magnetization lies with an angle to the longitudinal current flow, a spontaneous transverse voltage develops, and as the magnetization switches between the four energy minima in an external field, the transverse resistance experiences abrupt jumps that are sensitive to the magnetization directions. PHE has also been observed in metallic ferromagnets, but is very small. The PHE in single domain (Ga,Mn)As is about 4 orders of magnitude higher than that found in metallic ferromagnets, and this large magnitude presumably stems from the significant spin-orbit coupling in the valence band and the large spin polarization of holes in (Ga,Mn)As.

![Figure 1.11](image)

**Figure 1.11.** (a) Planar Hall resistance of a (Ga,Mn)As Hall bar (200\(\mu\)m wide) at 4.2K with magnetic field applied in the sample plane close to [110] direction. (b) Illustration of relative orientations of current, magnetic field and magnetization.

Fig.1.11(a) shows a typical planar Hall resistance in a (Ga,Mn)As epilayer with field applied along the in-plane [110] direction with a small misalignment angle. The trans-
verse resistance switches between +/- 100Ω at distinct fields and remains relatively constant between the switching events. Systematic study of PHE as a function of magnetic field direction indicates that PHE originates from the anisotropic magnetoresistance (AMR) and can be explained using a single domain model. The two orthogonal electric fields within a single domain can be written as [83]:

\begin{align}
E_x &= j\rho_\perp + j(\rho_\parallel - \rho_\perp)\cos^2 \phi_M = j\rho_\perp \sin^2 \phi_M + j\rho_\parallel \cos^2 \phi_M \quad (1.13) \\
E_y &= j(\rho_\parallel - \rho_\perp)\sin \phi_M \cos \phi_M \quad (1.14)
\end{align}

where \( j \) is the current density along the Hall bar, \( \phi_M \) is the angle between the magnetization and the current (See Fig.1.11(b)), and \( \rho_\parallel \) and \( \rho_\perp \) are the resistivities when the magnetization is parallel and perpendicular to the current. In most of the (Ga,Mn)As samples, strong cubic (biaxial) anisotropy dominates with a superposition of small uniaxial anisotropy. This results in four local energy minima at \(<100>(\phi \sim \pi/4, 5\pi/4)\) and \(<010>(\phi \sim 3\pi/4, 7\pi/4)\) directions. From Eq.1.13, we can clearly see that when the magnetization experiences a 90° rotation from one energy minimum to another, the polarity of the transverse voltage changes. Detailed analysis shows that \( \rho_\parallel < \rho_\perp \), which differs from conventional ferromagnets, where \( \rho_\parallel > \rho_\perp \). This is presumably due to the difference in the way holes and electrons contribute to the spin-orbit interactions in FM materials. The PHE strongly depends on the angle between applied magnetic field and the current, showing the largest switching fields when the angle is close to \(<110>(0, \pi)\) or \(<\bar{1}10>(\pi/2, 3\pi/2)\) directions. Moreover, both its magnitude and the switching fields decrease rapidly with increasing temperature.

Due to the strong AMR in (Ga,Mn)As, the PHE allows one to systematically investigate the in-plane magnetic anisotropy in (Ga,Mn)As (such as determining anisotropy fields) and monitor the magnetization switching via electrical transport measurement. The PHE has the advantage of mapping out magnetization reversal processes over conventional magnetometry measurements, such as Superconducting Quantum Interference Devices (SQUID), in which case only the projection of magnetization along the measurement direction is sensed. In the scope of this thesis, PHE is used to determine the magnetization state in field-dependent noise measurement in Chapter 4, to facilitate the study of the association between noise and magnetization.

(3). Longitudinal resistance:

The temperature dependent longitudinal resistance of (Ga,Mn)As exhibits a shoulder
in more “metallic” samples (usually high $T_C$ samples or samples after optimal annealing), as shown in Fig.1.12 curve C, and a peak in less metallic or more insulating samples (usually as-grown low $T_C$ samples or samples with lots of defects), as shown in Fig.1.12 curve A&B. In all the samples, the $T_C$ is close to the position of the resistivity peak. Thus this peak serves as a good estimation of $T_C$ from $R(T)$ measurement, although the $T_C$ obtained from the resistivity peak is usually a little larger than that obtained by a direct magnetization measurement [46]. The origin of this resistivity peak around $T_C$ is still under debate. Recent studies have shown that it can be described by a perturbative scaling theory of localization [84] or by scatterings of carriers from spin fluctuations, in which a correlation between temperature derivative of resistivity and $T_C$ is clearly identified [85].

![Figure 1.12](image)

**Figure 1.12.** Temperature dependent resistivity $\rho_{xx}$ of three samples in different conductivity regimes at zero field. The $T_C$ is determined from the resistivity peak position.

Below the $T_C$, three samples show distinct behaviors as the temperature approaches 4K. For the high $T_C$ sample (C), the resistivity drops as the temperature decreases, showing a typical metallic-like behavior. For sample A and B, however, the resistivity shows a minimum and increases again when approaching the lowest temperature. Sample A, which is more resistive, shows a rapid increase of resistivity at low temperatures, indicating that the sample is at the boundary of metal-insulator-transition (MIT). Sample B, while still metallic, has a resistivity tail at low temperatures that can be interpreted...
in terms of the Kondo effect arising from the presence of Mn interstitials \[86\].

The longitudinal resistance \(R_{xx}\) of a compressive-strained (Ga,Mn)As epilayer in an in-plane magnetic field can be modeled by Eq.1.13 when considering its single domain behavior. The longitudinal magneto-resistance (MR) exhibits rich behavior which strongly depends on the directions of the applied field and the current flow. A detailed quantitative investigation of longitudinal MR is given by Goennenwein et.al. \[87\]. In their study, since the \(j(\rho_\parallel - \rho_\perp)\cos^2 \phi_M\) term is a constant when the magnetization switches from one easy axis to another in the reverse direction, the jumps observed in \(\rho_{xx}\) are attributed to the magnetic field dependence of both \(\rho_\perp\) and \(\rho_\parallel\). The difference between \(\rho_\perp\) and \(\rho_\parallel\) is a constant, observed as the steady states in PHE. \(\rho_\perp\) and \(\rho_\parallel\) are considered to depend on the magnetic induction \(B = |\mu_0(H + M)|\), supported by the magnetoimpurity scattering model \[88, 89\]:

\[
\rho(B) = a - bB + cB^{3/2}
\]

At low fields, the linear term in Eq.1.15 is sufficient to correctly describe the longitudinal MR behavior. At high fields, the negative MR can be understood as the reduction of scattering by aligning spins with the external magnetic field.

Fig.1.13 shows the longitudinal MR for currents flowing along different crystalline directions. The data for [\(\bar{1}10\)] and [110] display similar negative MRs but distinct jumps when the magnetization switches. The spikes in Fig.1.13(a) at the switching fields are attributed to domain wall scattering when a domain wall nucleates and propagates through the sample in the vicinity of the switching fields \[82\]. The distinct jumps are due to the different angles between the magnetic field and the current, and they can be modeled in the same way as stated before using a single domain model. However, for the current running along [100] direction, big jumps are observed. This is simply because the magnetization now lies either parallel or perpendicular to the current (\(\phi_M\) switches between 0 and \(\pi/2\)). The difference in the resistivity jump is equal to \(\rho_\perp - \rho_\parallel\). This situation is similar to the determination of \(\rho_\perp - \rho_\parallel\) from the difference of Planar Hall resistance states for currents along [\(\bar{1}10\)] and [110] directions.

In Chapter 3, the study of longitudinal MR is extended to the perpendicular field geometry. By employing the models developed for in-plane geometry measurements, we successfully explain the observed longitudinal MR anomalies under the hard axis magnetization reversal.

In a tensile-strained (Ga,Mn)As, antisymmetric spikes were observed in the longitudinal MR. This is explained by the contribution of AHE to the \(R_{xx}\) due to circulating...
Figure 1.13. Longitudinal magnetoresistance $R_{xx}$ for current running through three different crystalline directions. A magnetic field is applied in the sample plane at an angle of 15° with respect to the [110] direction. (a) $I//[\bar{1}10]$. (b) $I//[110]$. (c) $I//[100]$. (d) Illustration of the measurement configuration. The Hall bar width is 200$\mu$m and the distance between the longitudinal voltage probes is 450$\mu$m.

Currents when domain walls are located in between the voltage probes [90, 91].
1.4 Hybrid magnetic semiconductor heterostructures

1.4.1 Motivation

In the context of semiconductor spintronics, heterostructures that integrate conventional semiconductors with ferromagnetic semiconductors and metals have been drawing attention recently, because they might provide new opportunities for the discovery of new physical phenomena and potentially lead to the development of new device functionalities [92]. Extensive studies of heterostructures have been carried out to combine semiconductors with magnetic materials to make devices such as ferromagnetic heterojunction bipolar transistors (FJBT) [93, 94], all semiconductor magnetic tunnel junctions [95, 96], magnetically tunable resonant tunneling diodes [97] and spin light-emitting diodes [98].

Ferromagnetic semiconductor (Ga,Mn)As is of particular importance because of the large spin polarization up to reasonably high temperatures and the easy integration with III-V semiconductors. In (Ga,Mn)As-based heterostructures, in order to successfully implement the concept of spin-controlled electronics, several goals have to be achieved, such as injecting spin-polarized currents across the hetero-surfaces, utilizing the spin-dependent transport in the multi-layers and engineering the magnetic properties of (Ga,Mn)As. A hybrid heterostructure consisting of (Ga,Mn)As and ferromagnetic metal MnAs shows several unique advantages in realizing these goals. First, being a semi-metal with a $T_C$ up to 320K, MnAs displays a very large spin-polarization at room temperature. Successful spin injections of electrons into GaAs using MnAs as an injector have been reported [99, 100]. Moreover, (Ga,Mn)As and MnAs have very different magnetocrystalline anisotropy constants ($K$) and Curie temperatures ($T_C$). For instance, at cryogenic temperatures, $K_{GaMnAs}\sim5kJ/m^3$ [101], while $K_{MnAs}\sim500kJ/m^3$ [102]. Further, the distinct Curie temperatures ($T_C=320K$ for MnAs and $T_C\leq170K$ for (Ga,Mn)As) could be exploited to systematically tune the anisotropy and magnetization of the soft FM layer relative to the hard layer. Finally, despite the difference in crystal structure, MnAs can be epitaxially grown on zinc-blende GaAs with reasonably good quality.

1.4.2 Structural properties of MnAs

In bulk MnAs, it is found that below the critical temperature ($\sim40^\circ C$), MnAs shows a hexagonal NiAs structure with a ferromagnetic $\alpha$-MnAs phase, while above $40^\circ C$, it transforms to a paramagnetic (or antiferromagnetic) $\beta$-MnAs phase, which has an orthorhombic MnP-type structure [103]. This phase transition is first order, characterized by the abrupt change of the magnetization.
In the growth of MnAs thin films, in principle, if one could keep replacing Ga with Mn atoms in a Ga$_{1-x}$Mn$_x$As alloy without limit, the end result would be zinc-blende MnAs. However, this zinc-blende MnAs phase is not stable, even with non-equilibrium epitaxial growth. Instead, more stable mixed phases with the co-existence of hexagonal \(\alpha\)-MnAs and orthorhombic \(\beta\)-MnAs are often obtained in the MBE growth. This co-existence of two phases is a result of the combination of two types of stress: phase transition stress, due to an abrupt volume change at the phase transition and thermal stress, due to large thermal expansion of MnAs \cite{105}. During the growth of MnAs, most of the lattice mismatch between MnAs and GaAs is compensated by introducing misfit dislocations. As they cool down towards the room temperature, the MnAs films are tensile strained because the thermal expansion of MnAs is much larger than that of GaAs. When the temperature is below \(T_C\), however, the MnAs suddenly experiences additional compressive strain, as the phase transition from the \(\beta\)-phase to the \(\alpha\)-phase involves a volume expansion. The sudden change of the lattice constants of MnAs occurs in the [1120] and [\bar{1}100] directions, whereas the lattice constant in the [0001] direction barely changes. As a consequence, the strain is relieved by forming a periodic stripe structure of mixed phases, as shown in Fig. 1.14. The alternating stripes are oriented along the \(c\)-axis, and the strain along the \(a\)-axis is relaxed. The fractions of these two
phases vary continuously with temperature and also depend on the strain induced by the substrates. These co-existing phases can result in very unusual behaviors, such as “self-exchange biasing” within a single layer of MnAs [106].

![Diagram of MnAs and GaAs crystals and epitaxial relationships](image)

**Figure 1.15.** Crystal structure (unit cells) of MnAs and GaAs, and their epitaxial relationships of (a) type-A and (b) type-B MnAs. The growth directions are [1100] for type-A and [1101] for type-B. From Ref. [107]

Extensive studies have been carried out on the MBE growth of MnAs on GaAs substrates. The growth kinetics of MnAs on GaAs (001) has been studied in real time by *in situ* RHEED [108] and Scanning Tunneling Microscope (STM) [109]. A phase diagram showing the dependence of the reconstructions on the growth conditions has also been presented [110], giving a guideline for choosing growth parameters.

Tanaka *et.al.* first showed that MnAs grown by MBE on GaAs (001) substrates can take two different epitaxial orientations – type-A and type-B – depending on the growth procedure [111][112]. Fig.1.15 shows the epitaxial directions of these two types of hexagonal MnAs lattices. The growth planes for type-A and type-B are (1100) and (1101) respectively. The epitaxial relationships in the sample plane are: [1120]MnAs//[110]GaAs and [0001]MnAs//[110]GaAs for type-A; [1120]MnAs//[110]GaAs and [1102]MnAs // [110]GaAs for type-B. Note that the [1120] axis of MnAs rotates by 90° between type-A and type-B. This is related to the difference in the atomic structures of $c(4 \times 4)$ and...
It was found that the starting monolayer template formation and the surface reconstruction of the GaAs surface prior to the growth of MnAs play a critical role in determining the epitaxial orientation. When an As flux was first supplied and MnAs was then grown on the resulting disordered \( c(4\times4) \) \( d(4\times4) \) GaAs surface, the growth direction was \( [\overline{1}100] \), giving type-A MnAs growth. On the other hand, when one monolayer of Mn was first deposited on the \( c(4\times4) \) GaAs surface, the growth direction was \( [\overline{1}101] \), resulting type-B MnAs. This template effect suggests the essential role of the starting surface reconstruction and interface stoichiometry in epitaxial relationships. Because the magnetic properties of MnAs strongly depend on the epitaxial structure, care must be taken during the MnAs growth to ensure the proper epitaxial relationship is obtained.

1.4.3 Magnetic properties of MnAs

![Figure 1.16. Room temperature magnetization characteristics of a type-A (a)(b) and a type-B (c)(d) MnAs thin film. The magnetic field was applied in plane along [110] GaAs (a)(c), and along [\overline{1}10] GaAs (b)(d). From Ref. 107](image-url)
MnAs displays robust ferromagnetism up to room temperature. Magnetization measurements show that MnAs thin films have a strong magnetic anisotropy, and the easy magnetization axis is the [11\overline{2}0] direction. Fig. 1.16 shows the M-H characteristics for both 50nm type-A and type-B MnAs thin films measured by vibrating sample magnetometry. The type-A MnAs shows perfectly square hysteresis when the field is applied along GaAs[1\overline{1}0], which is also MnAs[\overline{1}1020] (Fig. 1.16(a)), while when the field is along GaAs[1\overline{1}0]/\[\overline{0}001]MnAs, no hysteresis loop was observed (Fig. 1.16(b)). On the other hand, for type-B MnAs, the easy axis rotates 90° to GaAs[\overline{1}10] direction because the [11\overline{2}0] axis of MnAs now lies parallel to GaAs[\overline{1}10]. In general, the in-plane a- and c-axes are the magnetic easy and hard axes of MnAs grown on (001)GaAs, respectively. In hybrid magnetic semiconductor heterostructures, type-A MnAs is favored because it often displays a better epitaxial growth quality and a square hysteresis loop.

1.4.4 MnAs/(Ga,Mn)As hybrid heterojunctions

MnAs/III-V heterostructures have been demonstrated in MnAs/GaAs lateral spin valves [113], MnAs/GaAs/MnAs [114] and MnAs/AlAs/MnAs tunnel junctions [115]. In the context of (Ga,Mn)As-based heterojunctions, hybrid metal/semiconductor heterostructures have been first explored to study spin polarized tunneling in a single barrier MTJs comprised of a ferromagnetic metal MnAs and a ferromagnetic semiconductor (Ga,Mn)As separated by an AlAs barrier [116]. Later on, a MnAs/AlAs/GaAs/AlAs/(Ga,Mn)As double barrier MTJ was also studied [117]. Unlike traditional metallic MTJs, or all magnetic semiconductor based MTJs, the two ferromagnetic components in these hybrid heterostructures have conductivities differing by four orders of magnitude (\(\sim 1\mu\Omega cm\) for MnAs and \(\sim 10 m\Omega cm\) for (Ga,Mn)As).

Fig. 1.17 shows the magnetization hysteresis loops and the tunneling magnetoresistances (TMR) of the hybrid junctions. The TMR shows a non-monotonic dependence on the AlAs barrier thickness, with the highest effect around 30% for the 5nm barrier sample. With conservative estimates for the spin polarization in each ferromagnetic layers (\(P(GaMnAs)\sim 85\% [118]\) and \(P(MnAs)\sim 40\%) [119], the observed TMR is \(\sim 30\%\) of the ideal TMR calculated based on the Julliere’s model [120]. This indicates that the spin-dependent tunneling through the AlAs barrier is fairly efficient.

Although MnAs is a naturally attractive material in fabricating hybrid structures with (Ga,Mn)As, the fabrication of MnAs-based devices requires special care due to the particular chemical properties of MnAs. The device patterning often involves wet chemical etching or dry etching processes. MnAs is found to react with most of the
Figure 1.17. (a) Hysteresis loops at 5K for a type-B MnAs-(Ga,Mn)As hybrid junction and for the same sample after the MnAs is etched away. (b) Magnetoresistances of hybrid junctions with a different AlAs barrier thickness (T=4.2K). The data are shifted for clarity. From Ref.[116]

acid and alkaline. However, reactive ion etching using Cl\textsubscript{2} gas virtually does not etch MnAs, and Ar ion milling is often employed to fabricate nano-scale MnAs structures [121]. Moreover, MnAs is found to be chemically unstable, and even the exposure of MnAs to water results in a certain degree of chemical reaction [122]. Fig.1.18 shows the AFM images of a freshly grown MnAs sample and a sample sitting in the air for one month. The as-grown sample shows a relatively smooth surface with some trenches, similar to the observations using STM [109]. However, the exposure of MnAs surface to the air results in a rougher surface with the formation of segregated islands. This deterioration is presumably due to the reaction of MnAs with the water vapor or oxygen in the air. To keep a good shelf life of our MnAs samples, we put the samples in vacuum sealing bags to minimize MnAs degradation.

1.5 Closing remarks

Ferromagnetic semiconductors allow the possibility of engineering spin-orbit couplings in ways that are unavailable in metallic systems. Although the technological application of (Ga,Mn)As is limited by its cryogenic transition temperature, the search for other room-temperature ferromagnetic semiconductors continues. Several criteria were proposed as calibers for an ideal ferromagnetic semiconductor [10]:

(1) The material should possess ferromagnetism that is induced by a low-density car-
Figure 1.18. Atomic force microscope (AFM) images of MnAs surfaces of (a) a freshly grown sample and (b) a sample exposed to the air for one month. The freshly grown sample shows a rms roughness of $\sim 1 \text{ nm}$, while the aged sample shows a rougher surface with a rms of $\sim 3 \text{ nm}$

Carrier system so that magnetic properties can be modulated over a broad range by modifying the carriers through doping, photo injection, gating, or heterojunction band-structure engineering.

(2) The material should have a transition temperature above 500K so that it can be used in a wide range of temperatures.
(3) The material’s magnetic properties should be insensitive to the fluctuation of magnetic ion distribution to allow good reproducibility.

(4) The mean exchange field of the free carriers should be large enough to yield strong MR and TMR effects.

(5) The material should have strong magneto-optical effects so that the magnetization can be optically sensed.

(6) Collective magnetic damping should be weak enough to allow optical and electrical spin-transfer for magnetization manipulation.

Based on these criteria, the search for new magnetic semiconductors will keep going, and new physical phenomena may emerge as the advances in growth and defect control are achieved.
Chapter 2

Experimental techniques

2.1 Introduction

The fabrication of magnetic semiconductors often involves crystal growth under non-equilibrium conditions. Molecular beam epitaxy (MBE) provides a unique advantage in this context, allowing access to a variety of material composition and crystal structures not attainable by other crystal growth techniques. MBE grown samples are then patterned into micro/nano devices using a variety of semiconductor processing tools at the Penn State Nano-fabrication facility. The transport measurements are carried out using different measurement schemes depending on applications. This chapter gives the general overview of the experiment techniques concerning growth, patterning and transport measurements that are used in this thesis.

2.2 Molecular Beam Epitaxy (MBE)

2.2.1 System overview

Molecular beam epitaxy (MBE) is one of the several methods used for growing epitaxial single crystals. As suggested by its name, in an MBE system, the ultra-pure constituent elements, such as gallium and arsenic, are heated in Knudsen effusion cells to form molecular beams which are directed towards a heated substrate and react with each other on the substrate to form a single crystal. The technique was invented in the late 1960s at Bell Laboratories by J. R. Arthur and Alfred Y. Cho [123, 124]. A summary of the historical development of MBE and new aspects of this technique can be found in references [125, 126].
MBE is very useful for growing high-quality heterostructures due to its layer-by-layer growth mechanism, low growth rate (\(\sim 1\) monolayer/s), precise control of doping profile and the ability to obtain abrupt interfaces. Compared to the commonly used Chemical Vapor Deposition (CVD) technique, MBE’s has a relatively low yield, but it is nevertheless a powerful tool in developing sophisticated electronic and optoelectronic devices such as laser diodes. A schematic of a typical MBE system is shown in Fig. 2.1(a), and Fig 2.1(b) shows a picture of the MBE system in our group.

A typical MBE system consists of a deposition chamber with vacuum pumps, effusion cells, shutters, sample transfer mechanism, sample heater and rotation assembly, and usually in situ characterization equipment such as reflection high energy electron diffraction (RHEED). The growth chamber is often inter-connected with a buffer chamber for the propose of loading/unloading samples or pre-growth desorption. Other in situ characterization tools, such as scanning probe microscopy (SPM), low energy electron diffraction (LEED), can also be connected to the growth chamber without breaking the vacuum so that the surface properties of the sample can be studied without exposure to air. The samples are transferred between chambers using a magnetically coupled transfer mechanism. Each chamber is separately pumped and isolated by gate valves.

The ultra high vacuum (UHV) condition is essential in MBE systems since the mean free path of the molecules or atoms from the cells has to be larger than the geometrical size of the chamber. Thus, the arrival rates of the molecules or atoms are not affected by the collisions with residue gas molecules and the impurity incorporation is minimized. A background pressure of \(10^{-9}\) \(\sim 10^{-11}\) torr is usually needed for MBE growth. Apart from vacuum pumps used to maintain the UHV condition, a liquid nitrogen shroud is located on the inside wall of the chamber to act as an effective “cryopump” that absorbs residual gases and brings the pressure even lower during the growth. Another cryopanel is also located between effusion cells as a thermal isolation between them. The pressures of the chambers are measured by Bayard-Alpert style ion gauges. One ion gauge with a retractable arm is mounted in front of the substrate holder for the measurement of beam equivalent pressure (BEP) of the source materials.

A computer controlled pneumatic shutter is positioned in front of each cell so that sample composition and thickness can be precisely controlled. During the growth, the substrate is heated up to a selected growth temperature and rotates continuously to improve the uniformity of the films. RHEED is used to determine the layer-by-layer growth rate as well as to monitor the quality of the growth. We also have a KSA BandiT system to accurately monitor the growth temperature in real time.
In our MBE system, two UHV chambers are inter-connected. One is dedicated to III-V semiconductors with III-V elements: Ga, As, Al, In, Mn and Be, Si for doping. The other is dedicated to II-VI semiconductors with II-VI elements: Zn, Se, Cd, Te, Mn and Cl for doping. High purity sources (from 5N to 7N) are used to ensure the quality of the samples. The common maintenance of the chambers includes reloading.
source materials, regenerating the cryopumps, replacing broken ion gauge filaments, leak checking, etc. After the chamber is vented to air, a bake-out at \(180^\circ C\) for an extended period of time is often necessary to restore the UHV condition.

### 2.2.2 Vacuum systems

The UHV condition in MBE systems is achieved by careful designing of the vacuum chamber with a good selection of vacuum pumps. All internal materials of the chambers are chosen for minimal background gas contribution and \(200^\circ C\) backout temperature compatibility. Vacuum chambers are normally fabricated from stainless steel, and are often electropolished and passivated to minimize the surface outgassing. The viewports of the chamber are often made from quartz and all chamber ports are sealed with metal gaskets on conflat (CF) flanges.

MBE systems rely on a family of pumps, the selection of which depends on the growth materials and on the purpose of the chambers. Generally speaking, vacuum pumps can be broadly categorized according to three pumping mechanisms: positive displacement, momentum transfer and entrapment. 

Positive displacement pumps use a mechanism to repeatedly expand the volume of a cavity, allow gases to flow in, seal off the cavity, and exhaust it to the atmosphere. Typical examples of positive displacement pumps are the rotary vane pump and the scroll pump. They are often used as roughing pumps or backup pumps for UHV pumps to bring the chamber pressure down to a few millitorr (\(10^{-3}\) torr).

Momentum transfer pumps use high speed jets of dense fluid (diffusion pump) or high speed rotating blades (turbo molecular pump) to push gaseous molecules out of the chamber. They operate in the free molecular flow regime as compared to displacement pumps which operate in the viscous flow regime. Diffusion pumps are generally not favored in MBE systems because of the possible contamination from the backstreaming of the pump oil, while turbo molecular pumps are widely used in small vacuum chambers such as loadlocks. Since the momentum transfer depends on the mass of the gas molecules, turbo pumps are very good at pumping heavy gases such as N\(_2\), but they are less effective for light gases such as H\(_2\) and He. A pressure of \(10^{-6} \sim 10^{-8}\) torr can often be achieved by using a turbo pump.

When the ultimate purity is required, gas entrapment mechanism is used. Gases are entrapped in an absorbed or a solid state by using cold heads or reactive metals. Two most commonly used entrapment pumps for UHV are the cryopump and the ion pump. A cryopump consists of a closed cycle refrigerator in which Helium gas is compressed and
expanded to cool a cold head inserted into the vacuum chamber. Gases with relatively high boiling points are then effectively condensed or trapped on a cold surface. The temperature of the cold head is usually only a few Kelvin, and sometimes a two-stage cold head with two cold plates at different temperatures (12K and 65K in our cryopump) is used to improve the pumping efficiency. Eventually, the cold head becomes saturated with the absorbed gases and needs to go through a “regeneration” cycle, in which the cold head is isolated from the vacuum chamber, heated up and pumped to remove the collected gases by a roughing pump. Cryopumps are less effective in pumping gases with low boiling points such as Helium and also work at moderate noise/vibration level because of the compressor. Ion pumps, on the other hand, operate in a quieter and more reliable way in which no solid moving parts are involved. An ion pump works by ionizing the residual gases and trapping them inside magnetically confined cathodes. The ionized gas molecules move in helical trajectories towards the cathodes and are either chemically combined with the sputtered/sublimated reactive metal (usually Titanium) or buried into the cathodes. Ion pumps are clean, bakeable, vibration free, and UHV as low as $10^{-9} \sim 10^{-11}$ torr can be achieved. The drawback of ion pumps includes relatively low pumping speed, especially for light gases such as hydrogen. Neither of these pumps can pump all the way from the atmosphere to UHV, so roughing pumps such as scroll pumps or turbo pumps are often used to pump the system down to a level ($10^{-5} \sim 10^{-6}$ torr) where the UHV pumps kick in.

Specifically, in our MBE systems, the EPI 620 system uses a cryopump as a UHV pump connected to the growth chamber, while a turbo pump is attached to the buffer chamber which is also used as a load-lock for sample loading/reloading. A scroll pump is connected to the 620 buffer chamber to serve as a roughing pump. The EPI 930 system has two ion pumps for the growth and buffer chamber respectively.

### 2.2.3 Effusion cells and shutters

In MBE growth, the composition of the film and the growth rate strongly depend on the fluxes of the source molecules. Thus precise control of the outgassing of the source materials is essential. Knudsen Cells are often used as source evaporators for MBE growth.

A typical Knudsen cell (Fig. 2.2) contains a crucible (usually made of Pyrolytic Boron Nitride (PBN)), heating elements (often made of metal tantalum), a water/cryogen cooling system, thermocouples and electrical feedthroughs. The source charge is loaded into the crucible and is radiantly heated by the heater. Effusion flux is determined by Knud-
sen cell temperature which is controlled by a feedback loop from a tungsten-rhenium thermocouple and a 3-term (PID) temperature controller to the heater power supply. In order to achieve a flux control within $\pm 1\%$, the cell temperature must be held within $\pm 0.5^\circ$C. In some cases, special cells with additional filaments are used to generate temperature gradient in the crucible to form “hot lip” or “cold lip” cells. “Hot lip” cells are used for sources with low melting points such as Gallium so that the condensation at the the lips of the crucibles is reduced. For aluminum, a “cold lip” cell is often employed to prevent the melting aluminum from creeping out the crucible and cracking the cell due to its large thermal expansion at the melting point.

Another important element for precise control of the flux is the fast moving shutters positioned in front of the effusion cells. These pneumatic shutters can open/close within a fraction of a second, allowing monolayer film growth control and nearly atomically abrupt transitions from one structure to another.

2.2.4 Bandgap thermometry

In MBE growth, the substrates are indium bonded onto the molybdenum block holders that are transferred to the manipulator in the growth chamber. The manipulator rotates the block holder with a heater assembly positioned on the back of the holder. Due to the gap between the block holder and the heater assembly, the real temperature of the sample is much lower ($\geq 100^\circ$C lower) than the thermocouple readings. The

Figure 2.2. Picture of a typical effusion cell, from www.Riber.com
temperature of the substrate can be measured by a pyrometer which measures thermal radiation directly from the wafer. However, for (Ga,Mn)As, low growth temperatures have to be employed to drive the growth condition far from thermal equilibrium. At such low temperatures (∼250°C), pyrometry fails due to the diminishing thermal radiation.

To obtain accurate temperature measurements at low temperatures, a KSA Bandit system was installed in our III-V system. By measuring the temperature-dependent optical absorption edge of semiconductor material and comparing this with a calibration file, the BandiT can provide accurate temperature monitoring at low temperatures. It can also be used for infrared (IR) transparent substrates and films.

In our system, the Bandit is installed in the reflection mode, where a light source is mounted on one port and the detector is mounted on a second, non-specular port. A schematic diagram is shown in Fig. 2.3. A Quartz-Halogen lamp is used to shed light on the sample through focusing lenses. Then the diffusely scattered light component from the wafer is collected through the collection optics on the detector and fed back to the solid state spectrometer via a dual fiber optic cable. By fitting the light absorption spectrum, the band gap of the semiconductor can be calculated and is directly associated with the real time temperature of the wafer. A laser beam is also used through the dual fiber optic for alignment of the detector.

Band-edge thermometry enables accurate temperature measurement in a broad range
of temperature (room temperature up to the wafer oxide desorption temperature) and it is insensitive to change of view port transmission, stray light sources and signal from substrate heaters. However, it is found that for heavily doped substrates, such as p-type GaAs wafers, band-edge thermometry fails due to excessive light absorption. A way to circumvent this problem is by putting an undoped substrate side-by-side with the doped substrate on the block holder as the temperature calibrator.

### 2.2.5 RHEED

One of the most important in-situ characterization techniques for real-time sample growth monitoring is RHEED, and it has become a standard component of an MBE system. It can be used to calibrate growth rate, observe the oxide desorption, give information about surface morphology and provide evidence of surface reconstruction.

![RHEED patterns](image)

**Figure 2.4.** Images of RHEED patterns with electron beam pointed along the (a) [110] (2×) and (b) [110] (4×) directions during the high-temperature (~580°C) GaAs growth.

In this method, a beam of high-energy electrons (12KeV in our system) is emitted from an electron gun, and strikes the sample surface at a grazing angle (1° ~ 3°). Incident electrons diffract from atoms at the surface of the sample and then impinge on a phosphor screen to produce a diffraction pattern, which is captured by a CCD camera and is fed back to the computer for analysis. Due to the strong interactions of the electrons and crystal lattice and the small incident angle, the penetration depth is limited to a few Angstroms. Therefore the diffraction patterns only show the periodic information from the topmost surface of the sample. Detailed analysis shows that a “streaky” pattern indicates a flat surface, while a “spotty” pattern results from transmission of electrons
through surface islands, implying the surface is not smooth.

The distance between the streaks in the diffraction pattern reflects the lattice spacing of the surface atoms in reciprocal space. In UHV, the atoms on the surface do not arrange them in the same way as they do in the bulk because of lacking of bonding from the vacuum side. The rearrangement of atoms at the surface is called “surface reconstruction” and it can be characterized using RHEED. For instance, Fig. 2.4 shows the $2\times$ and $4\times$ patterns of GaAs for electron beam pointed along [110] and [-110] direction respectively. The $2\times$ pattern has secondary lines with a spacing of 1/2 the primary line and the $4\times$ pattern has additional lines with 1/4 of the primary line spacing. The smaller the spacing in the diffraction pattern, which represents the reciprocal space of the real space, the larger the spacing in the real lattice. The $(2\times4)$ reconstruction pattern is often observed in high-temperature GaAs growth under As rich condition, indicating the lattice spacings on the surface are 2 times by 4 times those of the underlying bulk material.

In the case of layer-by-layer growth, RHEED intensity oscillation is used to accurately measure the growth rate. When the growth is initiated on a smooth sample surface, the intensity of the specular spot starts to oscillate, as illustrated in Fig. 2.5. The intensity is high when the surface is smooth and starts to dim as small islands nucleate. The intensity
reaches minimum when the surface coverage is about 50%. As the islands coalesce into a flat layer, the intensity increases, reaching peak value again when a monolayer growth finishes. The oscillation frequency of the intensity corresponds to the monolayer growth rate. By carrying out a Fourier transform of the oscillation curve or counting the period of the oscillation, the growth rate can be accurately determined. RHEED oscillation
is found to be dependent on the material, the growth crystalline direction and electron incident angles.

RHEED oscillations for the growth of GaAs, AlAs and AlGaAs are shown in Fig. 2.6. The insets show the Fourier transform of the oscillation curves and the growth rates are determined from the peak frequencies. At the start of the growth, the intensity may either increase or decrease depending on the initial layer coverage. The overall magnitude of RHEED oscillation is damped as the growth progresses. This is because some islands nucleate before the previous layer is finished. The median intensity of the oscillation may also decrease with time, which is caused by the electron beam drifting due to a charging effect [130]. The growth rate obtained from RHEED oscillations can also be used to determine the composition of ternary alloys, such as Al\(_x\)Ga\(_{1-x}\)As. The composition \(x\) is calculated by the difference in growth rate for binary and ternary materials, given by:

\[
x = \frac{R(\text{ternary}) - R(\text{binary})}{R(\text{ternary})}
\]

where \(R\) represents the growth rate. For instance, in Fig. 2.6(c), the Al composition calculated from the growth rate of GaAs and AlGaAs is 0.7.

### 2.3 Superconducting Quantum Interference Device Magnetometry (SQUID)

Superconducting Quantum Interference Device (SQUID) magnetometry is one of the most sensitive methods for measuring magnetic properties. The core part of SQUID consists of one or more Josephson junction(s) in which two superconductors are separated by a thin insulating layer. The Josephson junction has a unique property called the Josephson effect, which is a characteristic of tunneling of Cooper pairs of electrons through the junction. In the DC Josephson effect, a current proportional to the phase difference of the wavefunctions across the insulator can flow in the junction in the absence of an external voltage. In the AC Josephson effect, a fixed DC voltage is applied across the junction and the tunneling current will oscillate with a characteristic frequency \(f = 2eV_{DC}/h\), which is proportional to the applied voltage [131].

The great sensitivity of the SQUID comes from the ability to detect the change in one flux quantum \(\Phi_0 = h/2e\), where \(h\) is the Planck constant and \(e\) is the electron charge. As illustrated in Fig. 2.7, a steadily increase in magnetic flux will develop a constant voltage across the junctions. Then the measured voltage (current) oscillates with one period of
Figure 2.7. Illustration of measurement of one flux quantum in SQUID [131].

oscillation corresponding to an increase of one flux quantum. Counting the oscillations allows one to evaluate the flux change which has occurred [131]. In a more realistic way, the magnetic flux change induces the phase difference between the current on two sides of the ring and by detecting the phase change of the voltage oscillation, the magnetic moments can be accurately measured.

In a real SQUID system, the SQUID loop is shielded from the external magnetic field by a superconducting enclosure within the liquid helium bath. A flux transformer is used to couple the signal from the external environment to the SQUID loop. By stepping the sample vertically through the pickup coils, the change of the magnetic flux from the sample is inductively coupled to the SQUID loop. A second derivative gradiometer, which consists of two parts of coils winding in opposite direction, is often used as the flux transformer to cancel any uniform magnetic field. In this way, the measurement is only sensitive to the gradient of flux change from the sample moving in the vertical direction [132][133].

All the magnetometry measurements in this thesis are done using a Quantum Design RF SQUID. All the samples are field-cooled in a 1T field and the temperature dependent remnant magnetization is measured upon warming up in a small field (20Oe ∼ 50Oe).
The major or minor hysteresis loops are measured by sweeping the external magnetic field in a certain range, usually after saturating the magnetization in one direction.

### 2.4 Device fabrication

After the samples are grown, they are fabricated into micro/nano devices using a variety of processing techniques. Depending on the structure of the devices, different processing steps are used. More detailed processing recipes for specific applications are listed in the Appendix section. Here only the general fabrication techniques are discussed.

Modern micro/nano patterning usually involves lithography. Depending on the way of exposure, lithography can often be divided into two categories: lithography using a mask and direct write lithography. The sources for exposure includes ultraviolet (UV) light, laser beams, X-ray and electron beams due to the wave nature of electrons.

Two typical photolithography steps are illustrated in Fig.2.8. In the standard photolithography (Fig.2.8(a)), a sample is spin coated with photoresist (Shipley 1813 in this case) at a speed of 5000rpm for 40 seconds, giving a uniform thickness of \( \sim 1000\text{nm} \). It is found that a high acceleration rate is crucial to get good uniformity and avoid edge beads. After spinning, the resist must undergo a softbake process (115 \(^\circ\)C for 70s), in which the solvent in the resist is driven off and the resist is hardened. Then the sample is exposed in 365nm (i-line) UV light at an energy density of 12mW/cm\(^2\) using a contact aligner. A mask is positioned in close contact to the top of the sample so that the patterns on the mask determine the exposure area on the photoresist. The exposed resist is developed using MF CD-26 developer. The exposed part on the resist (positive tone) is removed and the patterns are transferred onto the photoresist. Subsequent wet chemical etching or dry etching (reactive ion etching) processes etch the sample into the resist-defined patterns, and after the removal of the photoresist after etching, patterns such as these circular mesas are obtained. Sometimes, a second photolithography step is used to open holes on the top of the mesas for making electrical contacts, with the rest of sample covered by a layer of photoresist as an insulating layer.

Another important process is called “lift-off”. In Fig.2.8(b), electron beam lithography is used as an example for lift-off. Lift-off can also be used for optical lithography. After spin-coating two layers of e-beam resist (\( \sim 200\text{nm} \) 950K PMMA and \( \sim 400\text{nm} \) P(MMA-MAA) copolymer) and prebake procedure, the sample is exposed by direct write of electron beam. Due to the sensitivity difference of the two resist layers, after developing, the P(MMA-MAA) layer is dissolved more than the PMMA layer and “undercuts”
are produced in the bottom layer. During the metal evaporation, this overhang profile prevents the sidewall deposition so that the metal deposited on the sample surface and the metal on the resist are not connected. Finally the sample is immersed into a resist removal solvent (such as Microposit 1165) to dissolve all the photoresist so the metal deposited on the resist is lifted off, leaving only the defined metal patterns on the sample surface. The lift-off process is very useful for depositing metal contacts onto existing
devices or it can be used to define a metal hard mask for subsequent etching processes.

2.5 Low temperature transport measurement

2.5.1 Ohmic contact

Achieving ohmic contact on semiconductor devices is essential for electrical measurement. The ohmic contact depends on the work functions of the metal and the semiconductor and it strongly depends on the semiconductor doping. A list of ohmic contacts on semiconductor can be found in the book [134]. In most cases, Indium can serve as a “universal” ohmic contact on II-VI and III-V semiconductors after careful thermal annealing treatment.

For (Ga,Mn)As, however, because it is highly doped with a carrier concentration up to $10^{20}\text{cm}^{-3}$, the ohmic contact can often be obtained by directly putting indium on the sample surface. We use a thin Teflon film to hold a 80$\mu$m diameter indium dot by static charge. By maneuvering the transparent Teflon film under the optical microscope, the dot can be precisely pressed onto the contact pad. Gold wires are positioned on the flattened dot and then another indium dot is put atop in the same way to sandwich the gold wires. This method avoids the heating of the sample when a soldering iron is often used to make indium contacts.

Although an ohmic contact can be made by directly putting indium on (Ga,Mn)As, the indium contact often has a finite contact resistance. It is highly desirable to minimize the contact resistance when the measured device resistance is very small, such as in the current-perpendicular-to-plane (CPP) transport measurement of spin-valve structures. To achieve a lower contact resistance, a thin layer of Au (50nm $\sim$ 100nm) is evaporated on the device to serve as a contact pad. To improve the adhesion of Au, a layer of Ti or Cr (5nm $\sim$ 10nm) is deposited before the Au evaporation.

2.5.2 Cryostat

We carry out electrical transport measurements using a cryostat inserted in a liquid-Helium dewar fitted with a superconducting magnet. Two cryostats were used for the work in this thesis: an Oxford Heliox Helium-3 cryostat and a Janis Helium-4 cryostat. We now discuss some basic principles of cryogenics using the He-3 cryostat as an example [135]. Detailed operation instructions are enclosed in Appendix B.

A schematic diagram of the Oxford He-3 cryostat is shown in Fig[2.9] The sample is mounted on the base of the $^3\text{He}$ pot with a cold finger touching the bottom of the sample.
as a good thermal contact. The cryostat is inserted into a liquid Helium dewar to cool down to 4.2K. The inner vacuum chamber (IVC) is sealed by a greased cone seal, and pumped down to $< 10^{-5}$ torr to give a good thermal isolation from the Helium bath.

![Diagram of Oxford Heliox Helium-3 cryostat](image)

**Figure 2.9.** A schematic illustration of an Oxford Heliox Helium-3 cryostat

The valuable $^3$He gas is stored in a storage vessel at the top of the insert. A 1K pot is connected to the $^4$He bath via two pickup tubes and the $^4$He flow is adjusted by a needle valve on the 1K pot. When cooling down to temperatures below 4.2K, the $^4$He is allowed to flow into the 1K pot and the pot is also pumped through the 1K pot pumping line. As the vapor pressure of the $^4$He in the 1K pot is reduced, the temperature drops to $\sim 1.5$K. During this process, the sorb (a sorption pump) is heated to 45K so that it will not absorb the $^3$He gas when it condenses. The $^3$He gas is free to leave the storage dump and condenses on the 1K pot assembly and runs down into the $^3$He pot. After most of the $^3$He is condensed at $\sim 1.5$K, the heater on the sorb is turned off and the sorb is cooled by letting liquid helium flow through the sorb heat exchanger by pumping the sorb heat exchanger outlet. The cooling of the sorb will pump the $^3$He gas and reduce the vapor pressure above liquid $^3$He. The base temperature of $\sim 350$mK can then be achieved. For temperature control between 0.35K and 1.5K, a heater on the sorb is used to vary
the pumping ability of the sorb and in turn adjusts the $^3$He vapor pressure. For the intermediate region between 1.5K and 4.2K, the temperature is controlled by a heater on the $^3$He pot, while keeping the 1K pot pumped and the sorb warm. Above 4.2K, the 1K pot and the $^3$He system are not taking effect and temperature is adjusted solely by the heater on the $^3$He pot. The proportion-integral-derivative (PID) settings for the feedback loop of the temperature controller have to be tuned accordingly in different temperature regimes.

2.5.3 Superconducting magnet

A superconducting magnet is fitted in the liquid Helium dewar, capable of generating magnetic field up to 8 Tesla in the vertical direction. The magnet consists of a number of concentric solenoid sections wound from Niobium Titanium (NbTi) wires under careful design so that the magnetic field is uniform over a certain range in the coil [136]. The whole magnet is submerged into liquid Helium to achieve a superconducting state.

![Circuit diagram of a superconducting magnet fitted with a persistent switch.](image)

**Figure 2.10.** Circuit diagram of a superconducting magnet fitted with a persistent switch.

A persistent switch is used to control the energization of the magnet by shunting the current from the power supply. As shown in Fig.2.10, the switch consists of a length of superconducting wire non-inductively wound with an electrical heater. This superconducting wire is connected in parallel with the superconducting magnet. To energize/de-energize the magnet, the superconducting wire is made resistive by raising
its temperature through the switch heater. Then the switch is in “open” state in which the current from the magnet power supply will flow in the superconducting magnet. The switch is in its “closed” state when the switch heater is turned off, the wire segment becomes superconductive again and shorts the magnet.

The persistent mode of the magnet can be obtained by the following steps: open the switch to energize the magnet to the desired field, then close the switch and reduce the current from the power supply to zero, leaving the magnet in its previously energized state. The current flowing in the magnet then remains constant without external power. To take the magnet out of the persistent mode, the current from the power supply has to be ramped up to match the current in the magnet so that the switch can be opened, and then the current in the magnet can be controlled by the external power leads.

2.5.4 AC and DC measurement

In the magneto-transport measurements, both DC and AC schemes are used, depending on the specific requirements. Generally speaking, AC offers the advantage of higher signal to noise ratio when detecting small signals. It is useful when using a low current at low temperatures to avoid sample heating. While DC is helpful when the measured resistance is large or when parasite capacitances exist in the measured device. Four-probe technique is often used in the resistance measurement to eliminate the contributions from the contact resistances. In this configuration, the current is sourced through two leads and the voltage is sensed through another two separate leads. Since the voltmeters usually have high impedances, the current flowing through the portion of the circuit comprising the voltmeter is negligible. Thus the potential drops across the contact resistances are minimized and only the sample resistance between the two voltage probes is accurately measured.

The illustrations of different measurement setups are shown in Fig. 2.11. In DC measurement (a), a programmable current source provides a constant current across a Hall bar, and then both the longitudinal (R_{xx}) and transverse (R_{xy}) resistances are measured by digital multimeters (DMMs). In AC measurement (b), the excitation current is obtained by placing a large limiting resistor (several orders larger than the sample resistance) at the voltage output of the lock-in, mimicking a constant current source. The excitation frequency is often chosen to be low (17Hz) so that the effects of parasite capacitances are minimized. The applied AC excitation current is drained to an electrical ground which is also connected to the chassis ground of the instrument.

For the measurements of the tunneling resistances of magnetic tunnel junctions, a
source meter is used to provide a constant bias voltage and to measure the tunneling current, as shown in Fig. 2.11(c). Due to the small size of the mesas, a pseudo four probe scheme is often used by wiring separate voltage and current leads on one contact. It may not eliminate contact resistances, but it removes the resistance contributions from the sensing wires.

For different experiments, specific Labview programs are designed to meet the measurement requirements. A variety of parameters including magnetic field control, temperature control, time delay, voltmeter reading and averaging can be set in the programs to customize each magneto-transport scan. Instruments are automatically controlled and the taken data are saved for later analysis.
Chapter 3

Magnetoresistance anomalies during hard axis magnetization reversal

3.1 Introduction

Recent studies have found that the magnetic anisotropy of (Ga,Mn)As plays an important role in determining its electrical [82] as well as optical [137] properties. In-plane magnetotransport studies have mapped out a systematic picture of magnetization reversal in compressive-strained samples [82, 87, 138, 139], which have their easy axes lying in the film plane. On the other hand, the perpendicular anisotropy was brought to attention by the observation of magnetoresistance (MR) anomalies in tensile-strained samples with easy axes perpendicular to the film plane in a planar geometry [90]. A detailed analysis of the perpendicular magnetization reversal on samples with in-plane easy axes has been given by X.Liu et al. using ferromagnetic resonance (FMR) measurements [101]. Coherent rotation originating from Stoner and Wohlfarth’s single domain model (SDM) [140] and non-coherent switching from domain wall nucleation and propagation were used to explain the Hall resistance (transverse MR) in the low and high field regimes respectively [101]. However, the behavior of the longitudinal MR has not been quantitatively studied in this situation.

By measuring both longitudinal and transverse MRs as a function of temperature and magnetic field, we probe the hard axis magnetization reversal in (Ga,Mn)As thin films grown on (001) GaAs substrates. We measured MRs in Hall bars oriented along the
three principal crystalline directions ([100], [1¯10] and [¯1¯10]) over a temperature range from \(0.35 \text{ mK} \leq T \leq 120 \text{ K}\), with the magnetic field normal to the sample plane. We find that the longitudinal MR exhibits distinct signatures corresponding to different types of magnetization reversal processes: while domain wall nucleation and propagation produces large MR anomalies (\(\sim 50 - 100 \Omega\)), coherent domain rotation is accompanied by a surprisingly weak MR. Hysteretic behaviors found in longitudinal MR for current along [100] are attributed to the misalignment of external magnetic fields. From comparing data taken with an in-plane field geometry, this misalignment is calculated. Finally, we compare our experimental measurements with analytical simulations that use SQUID magnetization data as additional input.

3.2 Experimental details

The 30nm \(\text{Ga}_{1-x}\text{Mn}_x\)As thin film (050323B) was grown on (001) semi-insulating, epi-ready GaAs substrate at \(\sim 250^\circ\text{C}\). Growth was monitored \textit{in situ} with reflection high energy electron diffraction (RHEED) at 12KeV. Prior to the growth of \((\text{Ga},\text{Mn})\)As, a buffer of a 170nm GaAs epilayer grown at a standard high temperature \((580^\circ\text{C})\) was prepared. The Mn concentration \(x\) measured from the second ion mass spectroscopy (SIMS) is approximately 0.06. A clear \((1\times2)\) reconstruction pattern was observed during the growth indicating the high quality of the film.

![Figure 3.1. Schematic depiction of the 3-way Hall bar orientations. The field is applied normal to the sample plane, \textit{i.e.} [001] direction.](image)

The as-grown wafer was cleaved into small pieces, and some of them were treated with a post-growth annealing process in nitrogen gas at \(250^\circ\text{C}\) for 90 min. These optimal annealing parameters are known to enhance the \(T_C\) by increasing the free hole density.
The sample pieces were patterned with a specially designed 3-way Hall bar mask using conventional photolithography followed by a chemical wet etching. Each Hall bar pattern has three arms oriented along three principal crystalline directions: [100], [110] and [110] (See Fig.3.1).

By sending a small AC current (50nA-80nA) through any two of the three arms, we are able to monitor the longitudinal and transverse MR in both two branches simultaneously, using conventional low frequency lock-in techniques. The samples are mounted on the $^3$He cryostat, capable of carrying measurement from 360mK to 120K. The applied magnetic field from a superconducting magnet points perpendicular to the sample plane. We also measure magnetization as a function of temperature in SQUID as an additional input.

### 3.3 Longitudinal MR in a perpendicular field

The resistivity of samples with moderate Mn concentration (3% ~ 6%) typically show “metallic” behavior: as the temperature decreases from room temperature, the resistivity first increases to its peak value around $T_C$ and then decreases as the temperature drops below $T_C$. In our sample, the temperature-dependent resistivity measurement shows exactly the same feature, from which we conclude that our samples are “metallic”. The $T_C$ determined from the $\rho - T$ curve is 55K for the as-grown sample and 90K for the annealed sample (Fig.3.2).

![Figure 3.2. Resistivity versus temperature for both the as-grown and annealed sample.](image)
Anisotropic magnetoresistance (AMR) in (Ga,Mn)As has been studied in great detail as a function of Mn concentration, temperature and annealing conditions [139, 141]. Contrary to the conventional ferromagnetic metals, the resistivity of (Ga,Mn)As is greater when the magnetization is oriented perpendicular to the current than when it lies in parallel to the current, i.e. $\rho_\| - \rho_\perp < 0$. The anisotropic MR is often measured in three orthogonal directions of the external magnetic field, while keeping the orientation of the Hall bar fixed. In a recent paper [139], it is found that the in-plane longitudinal resistivity depends not only on the relative angle between the magnetization and the current, but also on the crystalline axis the current is sent through. However, the latter dependence has not been explored in a systematic way, especially in an out-of-plane field geometry.

### 3.3.1 Determination of easy axis

By using a 3-way Hall bar in a perpendicular magnetic field, we are able to observe the dependence of longitudinal resistivity on crystalline orientations and map out the picture of hard-axis magnetization reversal. The longitudinal MR data of the as-grown sample for three orientations are shown in Fig.3.3(a)(b)(c). The data are offset for different temperatures. For clarity, the scales of the three plots are kept the same so that we can compare the relative amplitude of the MR curves. The scale bar gives a resistivity of 0.3mΩcm.

In all the orientations, the MR data show a negative MR at high fields, a peak at a critical field, followed by a sharp positive MR and a relatively flat region below that field. The negative MR at high fields is understood as the field-induced reduction of spin-dependent scattering by aligning the spins along the external field [35]. The small positive MR observed in Ref.[35] is attributed to the rotation of magnetization from its original in-plane direction to the out-of-plane direction, because $\rho_\| - \rho_\perp < 0$. However, both the longitudinal and the transverse MR may not be as smooth as depicted in Ref.[35]. As illustrated in Fig.3.3 abrupt changes in both $\rho_{\text{sheet}}$ and $\rho_{\text{Hall}}$ may occur, and they are accompanied by hysteretic behaviors during these switching processes, especially at low temperatures.

Considering the anomalous Hall effect (AHE) from skew scattering, where the perpendicular magnetization (denoted as $M_z$) is proportional to $\rho_{\text{Hall}}/\rho_{\text{sheet}}$ [35], the relative magnitudes of $M_z$ as a function of magnetic field at different temperatures can be obtained, as shown in Fig.3.3(d). It should be noted that the hysteretic behavior of AHE has been reported [101, 142], but it may not be universal because of its depen-
Figure 3.3. Longitudinal magnetoresistivity at various temperatures (data are offset for clarity) with current along (a) [110] (b) [110] (c) [100] direction. Note the similarity between (a) and (b) and their difference from (c). The left axis of (d) represents the $R_{\text{Hall}}/R_{\text{sheet}}$, which is proportional to perpendicular component of magnetization. The regions of coherent and incoherent magnetization rotation are indicated by the dashed circles.
dence on Mn concentration [139]. Nevertheless, it provides a good template to study the perpendicular magnetization reversal process. These double hysteresis loops (Fig.3.3(d)) in Hall (and also magnetization) data are explained in terms of coherent spin rotation due to single domain rotation at low fields and noncoherent spin switching from domain wall nucleation and expansion at the switching fields [101]. We will further explore the anomalies shown in the longitudinal MR that display a plateau during the coherent domain rotation in the next section.

As shown in Fig.3.3, the longitudinal MR for current running along the cubic axis [100] is distinct from the MR in the other two directions. At the first glance, the shape of the curves for [110] and [110] directions is almost identical from 4.2K up to $T_C$, while the MR curve for [100] direction gives a larger resistivity change during the incoherent spin switching. This feature can be understood simply by the AMR characteristic of (Ga,Mn)As. (Ga,Mn)As films under compressive strain often show a very large in-plane cubic magnetic anisotropy field $H_{4\parallel}$ biased by a small in-plane uniaxial anisotropy $H_{2\parallel}$ [143]. The easy axes of these films often point along $\langle 100 \rangle$ directions in the film plane [82, 87].

In a large magnetic field, the magnetic moment $M$ is saturated along [001], and it is perpendicular to the in-plane current, giving a large resistivity. As the applied field gets smaller, the in-plane anisotropy overcomes the Zeeman energy term, and $M$ tilts toward the sample plane, seeking the magnetic easy axis. At zero field, the magnetization should be aligned along one of the easy axes: [100] or [010]. Because of the negative value of $\rho_{\parallel} - \rho_{\perp}$ and the reduction of resistivity for [100] direction, we conclude that the magnetization points along [100] easy axis in the plane, parallel to the current flow. Since the in-plane magnetization at zero field still has an angle of $\approx 45^\circ$ with respect to [110] and [110] directions, the reduction in resistivity in those two directions due to magnetization rotating into the sample plane are not as prominent as the one in [100]. Combining with the temperature-dependent MR curves, we see that the easy axes remain along the cubic axes even up to $T_C$. The temperature dependent transition of the easy axis [67] is not seen in this case.

The fact that $M$ favors one particular in-plane easy axis to the other can be explained by assuming a slight misalignment of the perpendicular magnetic field. This misalignment results in a small in-plane component of magnetic field which fixes the in-plane magnetization along [100]. Another striking character can also be explained by this misalignment. As seen in Fig.3.3, the longitudinal MR curve for current along [100] starts to show huge hysteretic spikes/jumps when the temperature is raised above 10K.
At the fields of these jumps, only small MR disturbances are seen for currents along the other two directions.

### 3.3.2 Calculation of the field misalignment

To confirm the origin of these jumps and calculate the misalignment angles, we measure the MR with an in-plane magnetic field as a comparison. Fig. 3.4 illustrates the scheme of this measurement. A, B and C denote for the three arms of the Hall bar. An in-plane magnetic field is applied at an angle of $\phi_H \approx 15^\circ$ with respect to $[\overline{1}10]$. Figures 3.4(a) and 3.4(b) show the schematic configuration of a planar geometry and a perpendicular geometry, respectively.

The MR data along $[\overline{1}10]$ and $[100]$ are shown in Fig. 3.5 for 4.2K, 15K and 20K. For $[100]$, when the field decreases from positive direction to zero, the magnetization first rotates toward $[\overline{1}00]$ direction, where the $M$ is denoted as $M_1$. After the field is reversed, magnetization reorients itself toward $[010]$ via the nucleation and expansion of $90^\circ$ domains, giving the first jump as shown in the graph. The switching field is defined as $H_{sw1}$ and the $M$ at $[010]$ is denoted as $M_2$. Further increase of field in the negative direction will finally align the $M$ back to the field direction $[100]$ and result in the second jump. This second switching field is labeled as $H_{sw2}$ and the $M$ at $[100]$ is denoted as $M_3$. For current along $[\overline{1}10]$ direction, only small resistivity spikes exist at the points of magnetization switching. These quickly suppressed spikes are evidence...
of the 90° domain wall scattering upon switching [82]. From the analysis of the free energy density of four single domain states, the switching fields are associated with a domain wall pinning energy $\epsilon$ by the following equation: $H_{\text{sw}} \cdot (M_f - M_i) = \epsilon$, where $M_f$ and $M_i$ are the final and initial magnetization [82]. Therefore, the two switching events for in-plane MR are described by:

$$H_{\text{sw}1} \parallel M [\cos(45^\circ + \phi_H) - \cos(135^\circ + \phi_H)] = \epsilon_{[110]}$$  

(3.1a)

$$H_{\text{sw}2} \parallel M [\cos(45^\circ - \phi_H) - \cos(45^\circ + \phi_H)] = \epsilon_{[110]}$$  

(3.1b)

---

**Figure 3.5.** (a) (b) (c) panels show the zoom-in longitudinal resistivity data at 4.2K, 15K and 20K in a perpendicular magnetic field. In each panel, the resistivity curves for current along three different directions are labeled by different colors. The arrows indicate the direction of sweeping field. (d) (e) (f) panels show resistivity data at three temperatures with an in-plane field. The switching fields at which the magnetization rotates from $[010]$ plane to $[100]$ plane and back to $[010]$ plane are marked as $H_{\text{sw}1}$ and $H_{\text{sw}2}$.

In the case of a perpendicular magnetic field (Fig.3.5(a)(b)(c)), there is little change in resistivity at 4.2K when the magnetization is in the coherent rotation state. This indicates that the in-plane shadow field stemmed from the misalignment is too small to rotate the in-plane component of $M$ (denoted as $M_{xy}$). The rotation of $M$ is mainly
restricted in the (010) plane.

As the temperature is raised to 10K, the rotation of $M_{xy}$ becomes evident. When the field decreases from a positive high value to zero, the magnetization first rotates in the (010) plane from [001] to [100] direction through a series of noncoherent spin switching and coherent rotation. Then, after the field is reversed, $M$ jumps to (100) plane at $H_{sw1}^\perp$, with majority of the magnetization lying in the sample plane. At $H_{sw2}^\perp$, $M$ switches back to (010) plane and continues rotating in that plane until it aligns with [001] direction. It should be noted that throughout the rotation process from (010) plane to (100) plane, $M_z$ is constantly increasing, while in-plane $M$ component decreases. However, from the magnetization data extracted from the Hall resistivity, we know $M_z$ is small compared to $M_{xy}$ at low fields so that we use $M$ in stead of $M_{xy}$ to calculate the domain wall pinning energy. A zenith angle $\theta$ and an azimuth angle $\phi$ are used to describe the misalignment (see Fig.3.4(b)). Now the energy equations are:

\begin{align}
H_{sw1}^\perp \sin \theta M [\cos(45^\circ + \phi) - \cos(135^\circ + \phi)] &= \epsilon_{[110]} \tag{3.2a} \\
H_{sw2}^\perp \sin \theta M [\cos(45^\circ - \phi) - \cos(45^\circ + \phi)] &= \epsilon_{[110]} \tag{3.2b}
\end{align}

By equating Eqs. (3.1a) (3.2a) and Eqs. (3.1b) (3.2b), we calculate the misalignment angles to be $\theta = 6^\circ$, $\phi = 29^\circ$ from 15K data and $\theta = 6^\circ$, $\phi = 25^\circ$ from 20K data. We estimate the misalignment angles to be the average of these results: $\theta = 6^\circ$, $\phi = 27^\circ$.

Considering an in-plane magnetic field only, for a single ferromagnetic domain, the longitudinal resistivity from anisotropic magnetoresistance (AMR) is written as [83]:

$$\rho_{\text{sheet}} = \rho_\perp + (\rho_\parallel - \rho_\perp) \cos^2 \phi_M = \rho_\perp \sin^2 \phi_M + \rho_\parallel \cos^2 \phi_M$$ \hspace{1cm} (3.3)

When the magnetization $M$ rotates in the plane, the change of resistivity is described with this equation. For [100], $\phi_M$ changes from $\sim 180^\circ$ to $\sim 90^\circ$ and to $\sim 0^\circ$. The resultant change in resistivity is $\Delta \rho = \rho_\perp \text{in-plane} - \rho_\parallel \text{in-plane}$. Here we discriminate the case where $M$ is pointing out of the plane and the case where $M$ is in the plane but is perpendicular to the current by the superscripts. The resistivities for the other two directions, on the other hand, barely change. $\Delta \rho$ obtained from field-out-of-plane data and from field-in-plane data are 0.42mΩcm and 0.43mΩcm for T=15K; 0.38mΩcm and 0.37mΩcm for T=20K respectively. Comparing them not only verifies the origin of these low field jumps, but also indicates another possible way to obtain the difference between $\rho_\perp \text{in-plane}$ and $\rho_\parallel \text{in-plane}$.

It should be pointed out that in a certain temperature range, the MR with the field
along the hard axis shows two abrupt changes within only one field sweep (Fig. 3.6), e.g., as the field sweeps from zero up to a large negative field (sweeping down), the resistivity experiences a first change due to the in-plane magnetization switching (3) and a second change due to the rotation of magnetization to perpendicular direction (2). On the reversed field path (sweeping up), however, only the second change exists. This feature can be exploited in applications like magnetic memory or logic devices.

We also notice that at low temperature, the slope of negative MR at high fields is larger for [100] configuration than the other two. It indicates the reduction of spin-dependent scattering by the external field may also depend on the crystallographic directions. The mechanism of this dependence needs further investigations in a higher field region.

3.4 Application of the single domain model (SDM)

We note when the field is sweeping down from the out-of-plane direction to zero, the MR shows an abrupt change at a certain critical field and gives a relatively flat MR region afterwards (Fig. 3.6(4)). In this process, we assume that \( M \) only rotates from [001] to [100] in (010) plane (See Fig. 3.7(a)). From the calculation of the resistivity

![Figure 3.6. Illustration of longitudinal MR response to the direction of the field sweep for I//[100] at 20K. Four regions of MR with different mechanisms are labeled.](image-url)
tensor \[\mathbf{S}\], we find, analogous to the in-plane AMR, the out-of-plane \( \mathbf{M} \) rotation gives the expression of longitudinal resistivity for current along [100] direction in the same fashion:

\[
\rho_{\text{sheet}} = \rho_{\text{out-of-plane}} \cos^2 \theta_M + \rho_{\text{in-plane}} \sin^2 \theta_M
\]  

By normalizing the magnetization data in Fig.3.7(b), we get \( \cos \theta_M \) and \( \sin \theta_M \) as a function of field, from which we can picture out the path of \( \mathbf{M} \) as it rotates.

\[\begin{align*}
\rho_{\perp}^\text{out-of-plane} &= a_{\perp} - bB_{\perp} + cB_{\perp}^{3/2} \\
\rho_{\parallel}^\text{in-plane} &= a_{\parallel} - bB_{\parallel} + cB_{\parallel}^{3/2}
\end{align*}\]  

**Figure 3.7.** (a) Schematic illustration of the magnetization rotation in (010) plane. (b) Determination of \( \theta_M \) as a function of field from the Hall data.

The magneto-impurity scattering model predicts that the negative MR from the field suppression of spin-dependent scattering has the form \( \rho(B) = a - bB + cB^{3/2} \) \[89, 88\]. Experiments on temperature dependence of resistivity \[144\] and a quantitative study of longitudinal resistivity with an in-plane field \[87\] have given supporting evidence for this model. Here, we exploit this idea to explain the magnetotransport during perpendicular magnetization reversal.

Because the applied field required for perpendicular magnetization reversal is much larger than the magnetization itself (\( \sim 10 \) times larger), we decompose magnetic induction \( \mathbf{B} \) into two orthogonal parts: \( B_{\perp} \) and \( B_{\parallel} \). Then the two resistivity terms depend on the corresponding magnetic induction, \( i.e.\):
Figure 3.8. Comparison between the fitting curves and the experimental data at 4.2K (a), 15K (b) and 25K (c) (current along [100] direction). Reasonable agreement is achieved between the theoretical prediction and experiment.

Here we consider both resistivity terms have the same field-dependent coefficients except that they have different constant terms. Taking into account the misalignment angles in the previous section ($\theta = 6^\circ$, $\varphi = 27^\circ$), $B_\perp$ and $B_\parallel$ are written as:

$$B_\perp = \mu_0 M \cos \theta + \mu_0 H \cos \theta$$

$$B_\parallel = \mu_0 M \sin \theta + \mu_0 H \sin \theta \cos (45^\circ - \varphi)$$

Due to the small magnitude of $M$ compared to $H$, the second term in Eq. (3.6a) is the dominant at a high field, even the magnetization is in the same direction as the field. On the other hand, in Eq. (3.6b), the two terms are comparable at a low field when the magnetization is mostly in the sample plane. Consequently, involvement of the misalignment angles is essential for the accuracy of this simulation.

From SQUID measurements, we estimate the magnitude of the saturation magnetization in [001] direction to be 22.5 emu/cm$^3$ at 4.2K, 19 emu/cm$^3$ at 15K and 16 emu/cm$^3$ at 25K. The relative ratios of the saturation magnetization at different temperatures are confirmed from the $\rho_{Hall}/\rho_{sheet}$ data shown in Fig. 3.7(b).
Table 3.1. Fitting parameters at three temperatures.

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>(a_{\perp})</th>
<th>(a_{\parallel})</th>
<th>(b)</th>
<th>(c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.2K</td>
<td>19.4</td>
<td>18.4</td>
<td>3.0</td>
<td>1.4</td>
</tr>
<tr>
<td>15K</td>
<td>17.5</td>
<td>16.7</td>
<td>1.7</td>
<td>0.5</td>
</tr>
<tr>
<td>25K</td>
<td>18.0</td>
<td>17.3</td>
<td>1.6</td>
<td>0.37</td>
</tr>
</tbody>
</table>

Taking the 4.2K data as an example, at high fields, where the resistivity curve shows a pure negative MR, \(\theta_M=0^\circ\), Eq. (3.4) becomes

\[
\rho_{\text{sheet}} = \rho_{\perp}^{\text{out-of-plane}} = \rho_{\parallel}^{\text{in-plane}} + \mu_0 H \cos \theta.
\]

By fitting high field data up to 0.8T using Eq. (3.5a), we get \(\rho_{\perp}^{\text{out-of-plane}} = 19.4 - 3.0 B_{\perp} + 1.4 B_{\perp}^{3/2}\) in \(\text{m\Omega cm}\).

On the other hand, at zero field, \(\theta_M=90^\circ\) and \(H = 0\). \(\rho_{\text{sheet}} = \rho_{\parallel}^{\text{in-plane}}\) and \(B_{\parallel} = \mu_0 M\). This gives fitting parameter \(a_{\parallel} = 18.4\). The final fitting data for \(\rho_{\text{sheet}}\) is calculated by Eq. (3.4).

Follow the same procedure, the fitting equations for 15K are found to be

\[
\begin{align*}
\rho_{\perp}^{\text{out-of-plane}} &= 17.5 - 1.7 B_{\perp} + 0.5 B_{\perp}^{3/2} \\
\rho_{\parallel}^{\text{in-plane}} &= 16.7 - 1.7 B_{\parallel} + 0.5 B_{\parallel}^{3/2}
\end{align*}
\]

and for 25K are

\[
\begin{align*}
\rho_{\perp}^{\text{out-of-plane}} &= 18.0 - 1.66 B_{\perp} + 0.37 B_{\perp}^{3/2} \\
\rho_{\parallel}^{\text{in-plane}} &= 17.3 - 1.66 B_{\parallel} + 0.37 B_{\parallel}^{3/2}
\end{align*}
\]

The fitting parameters are listed in Table 3.1 for the three temperatures. The fitting curves and experimental data are shown for comparison in Fig. 3.8.

From the fitting graphs, we see a good agreement between the simulated curves and the experimental data, especially at low temperatures. The relatively flat MR region during coherent magnetization rotation can be viewed as the resultant contribution of one decreasing term \(\rho_{\perp}^{\text{out-of-plane}} \cos^2 \theta_M\) and one increasing term \(\rho_{\parallel}^{\text{in-plane}} \sin^2 \theta_M\) as the field ramps down. As a result of the resistivity minimum of \(\rho - T\) curve at around 15K, the fitting parameters \(a_{\perp}\) and \(a_{\parallel}\) do not change monotonically with temperature. The difference between \(a_{\perp}\) and \(a_{\parallel}\), however, constantly decreases with temperature, indicating the weakening of AMR effect.

Parameter \(b\), which represents the efficiency of field-suppressed spin-dependent scattering (the slope of the negative MR at high fields), is seen to decrease as \(T\) increases.
However, from Fig.3.3 we see this coefficient increases again as $T$ approaching $T_C$. We also observe that the magnitude of the negative MR slope drops again when $T$ is above $T_C$. This change of slope may indicate the different mechanisms of spin-dependent scattering between ferromagnetic, paramagnetic phases and the stage where the phase transition takes place. At low temperatures, the magneto-impurity scattering is the dominant mechanism; while as $T$ is getting close to $T_C$, spin fluctuation scattering may become dominant and result in a resistivity peak on the $\rho - T$ curve; when $T$ is well above $T_C$, other mechanism like spin-disorder scattering will probably take over \[35\]. This also explains why the fitting curves based on magnetoimpurity scattering model deviate from the experimental data as $T$ increases.

### 3.5 Temperature-dependent magnetic anisotropy

A variety of anisotropy terms contribute to the free energy density $F$ of a single homogeneous magnetic domain. In this case, where the applied magnetic field is perpendicular to the sample plane, $F$ is written as \[142\] \[143\]:

$$F = -\frac{1}{2}M(H \cos \theta_M - 4\pi M \cos^2 \theta_M + H_{2\perp} \cos^2 \theta_M + \frac{1}{2}H_{4\perp} \cos^4 \theta_M + \frac{1}{2}H_{4\parallel} \sin^2 \theta_M + \frac{1}{2}H_{4\parallel} \sin^4 \theta_M)$$

Here, the first term is the Zeeman energy; the second term is the demagnetization energy (or shape anisotropy); $H_{2\perp}$ is the perpendicular uniaxial anisotropy field which arises from the tetragonal distortion of the lattice during the MBE growth; $H_{2\parallel}$ is the in-plane uniaxial anisotropy field which represents the small difference between [110] and [110] directions; $H_{4\perp}$ and $H_{4\parallel}$ are the perpendicular and in-plane cubic anisotropy field respectively, indicating the difference between (100) and (110) directions. In ideal cubic system, $H_{4\perp}$ and $H_{4\parallel}$ should be identical. However, in a tetragonally-distorted crystal grown on a mismatched substrate, their difference should be distinguished. From the analyses in the previous sections, we see little difference between [110] and [110] in our sample, and the easy axes remain to be cubic up to $T_C$. As a result, the in-plane uniaxial term $H_{2\parallel}$ is small and negligible. In addition, the compressive strain makes the contribution from the perpendicular cubic anisotropy field $H_{4\perp}$ overshadowed by the effect of its uniaxial counterpart $H_{2\perp}$ and also may be ignored. The assumption of $|H_{2\parallel}| \ll |H_{4\parallel}|$ and $|H_{4\perp}| \ll |H_{2\perp}|$ is confirmed by the FMR study of (Ga,Mn)As sample with the same Mn concentration \[143\]. And since $4\pi M \cos^2 \theta_M - H_{2\perp}$ always
 occur together, they are simplified as a single term $4\pi M_{\text{eff}}$.

![Graph showing $H_T$ and $H_N$](image)

**Figure 3.9.** Depiction of how to define $H_T$ and $H_N$ (indicated by vertical bars) on the longitudinal MR data for current along [100] direction. (Data are offset for clarity)

By minimizing the free energy density as a function of magnetization angle $\theta_M$, a complete picture of magnetization reversal could be mapped out. Here we use the equations derived by Liu *et al.* [101] to calculate the two unknown anisotropy fields $4\pi M_{\text{eff}}$ and $H_{4\parallel}$. The first equation states, at low field, the ratio of $M_z/M$ increases proportionally to the applied field with the slope:

$$\frac{dM_z/M}{dH} = \frac{1}{4\pi M_{\text{eff}} + H_{4\parallel}}$$  \hspace{1cm} (3.7)

The second equation calculates the field $H_T$, where the magnetization takes sharp jumps when it is approaching [001] direction. The value of $H_T$ is taken as the field in the middle of the hysteresis loop as shown in Fig.3.9

$$H_T \approx \left(4\pi M_{\text{eff}} + H_{4\parallel}\right) - \sqrt{H_{4\parallel}(4\pi M_{\text{eff}} + H_{4\parallel})}$$  \hspace{1cm} (3.8)

The effect of domain nucleation and expansion is prominent at low temperatures due to large pinning energies and small thermal excitations. However, at high temperatures, the value of $H_T$ is not well defined. We use another equation giving the field $H_N$, where $M_z$ is saturated:

$$H_N = 4\pi M_{\text{eff}} - H_{4\parallel}$$  \hspace{1cm} (3.9)
Figure 3.10. The calculated anisotropy field $H_{4\parallel}$ and $M_{eff}$ as a function of temperature.

From the magnetization data (Fig.3.7(b)) we can extract $\frac{dM_z}{dH}$, and the values of $H_T$ or $H_N$ can be picked out from the longitudinal MR curves as illustrated in Fig.3.9. Solving these equations gives the values of anisotropy fields as a function of temperature as plotted in Fig.3.10.

3.6 Effect of annealing

We also measured the annealed sample in the same manner. The longitudinal resistivities for annealed sample, however, show distinct features from the as-grown ones (Fig.3.11). First, The change of resistivity upon magnetization reversal is very small, ~10 times smaller than the as-grown sample. Post-growth annealing has been found to increase the hole density and in turn enhance $T_C$ by out-diffusion of compensating Mn interstitials defects [22]. The change of hole density is predicted to influence the magnetic anisotropy [37] and easy axes. From the MR data of the annealed sample, the rotation of easy axes from cubic towards uniaxial is confirmed. The annealing is also seen to weaken the difference between $\rho_\perp$ and $\rho_\parallel$.

Second, the shape of the resistivity for [100] direction changes completely, showing two minima as the magnetization rotates from [001] to the sample plane. Instead of a flat MR region as seen in the as-grown sample, the resistivity first decrease as $\mathbf{M}$ rotates from the field direction, and then at a certain point, it reaches a minimum value and starts to increase as the field approaching zero. The combination of two negative MR with a positive MR in between is considered to come from the interplay of the external
magnetic field and in-plane anisotropic field. The magnetization is initially aligned along [001] direction, and it tilts towards the sample plane as the field decreases. However, as \( \mathbf{M} \) rotates, it does not stay in the (010) plane, but due to a strong in-plane uniaxial anisotropy, it tends to align itself along \( [\bar{1}10] \) direction. On the trace of \( \mathbf{M} \), there is a place where the angle between the magnetization and the current is the smallest, giving rise to the minimum of resistivity for current along [100] direction.

### 3.7 Summary and conclusions

In summary, the unusual shapes of MRs along three different crystalline directions under a perpendicular magnetic field are identified. The abrupt jumps of MR for current along [100] direction after the field is reversed are considered to come from the rotation of in-plane magnetization component due to a misalignment of the external field. By comparing switching fields in a planar geometry and using domain wall pinning energy formula, this misalignment is calculated.

Other aspects of the anomalies are one abrupt and one relatively weak MR regions during the perpendicular magnetization reversal. The SDM model is applied to explain
these anomalies, and by fitting high-field MR using magneto-impurity scattering model, good agreement is achieved between the simulated curves and the experimental data. This indicates the possibility of simulating longitudinal resistivity in a more complicated system where the magnetization rotates in three dimensions, as seen in the annealed sample.
Chapter 4

Electrical noise in (Ga,Mn)As

4.1 Introduction

Noise or fluctuations are spontaneous stochastic variations of physical quantities in time. These fluctuations come from thermal motion of matter and exist in a variety of systems such as metal, semiconductor, superconductor etc. The importance of studying fluctuations, in one aspect, lies in the fact that the ultimate accuracy of measurement of any physical quantity is limited by its inherent fluctuations and hence the sensitivities of many devices are affected. On the other hand, fluctuations in matter reflect the kinetic processes therein. The so-called “fluctuation spectroscopy” is a very informative tool used to study the microscopic motion and its dynamics in matter. In an equilibrium system, the fluctuations and the response of the system to external perturbation are related by the renowned fluctuation-dissipation theory (FDT). Therefore, the measurement of fluctuations provides abundant information on the kinetic properties of solids [145].

In magnetic materials, due to the coupling between the electronic transport and magnetic properties, the resistance noise measurement provides an informative way to study magnetic instabilities, as well as their relations with charge carriers via spin dependent scattering. Extensive research has been carried out to study electronic noise in Colossal Magnetoresistance (CMR) perovskites, magnetic tunnel junctions (MTJs) and Giant Magnetoreistance (GMR)-based magnetic read head. The ferromagnetic semiconductors and perovskite manganites share many similarities, such as a resistivity peak around ferromagnetic (FM) transition, existence of spin-charge interactions. Driven by these similarities, we took detailed measurement of electrical noise in (Ga,Mn)As samples with different Mn concentrations from the one that is nearly metallic to the one close to metal-insulator transition (MIT). In this chapter, I will first discuss
some basic noise measurement background and techniques, followed by the temperature dependent and magnetic field dependent measurements of resistance fluctuations in two (Ga,Mn)As samples.

### 4.2 Introduction to noise

#### 4.2.1 Mathematical

Generally speaking, noise refers to a physical quantity fluctuating around some mean value. The fluctuating quantity, $V(t)$, shows stochastic behavior in the time domain, which means the instantaneous value of the quantity can not be predicted. Several fundamental concepts regarding time domain fluctuations are:

- **The average value:**

  $$
  \overline{V} = \lim_{T \to \infty} \frac{1}{T} \int_{-T/2}^{T/2} V(t)dt
  $$

  where $T$ is the observation time.

- **The variance:**

  $$
  \sigma^2 = (V(t) - \overline{V})^2 = \delta V^2 = \overline{V^2} - \overline{V}^2
  $$

- **The probability density function (PDF):**

  The PDF is used to calculate the probability of a continuous variable falling into some range of values. If $V(t)$ has a PDF $P(V)$, then the probability for $V(t)$ to take a value in an infinitesimal interval $[V, V + dV]$ is $P(V)dV$. In most cases, when a large number of independent effects and random events contribute to the overall fluctuations, the PDF is Gaussian distribution function:

  $$
  P(V) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left[-\frac{(V - \overline{V})^2}{2\sigma^2}\right]
  $$

  For a Gaussian fluctuation, 68% of the $V(t)$ lies between $V \pm \sigma$.

- **The auto-correlation function:**

  $$
  \psi_V(\tau) = \overline{V(t)V(t+\tau)} = \lim_{T \to \infty} \frac{1}{T} \int_{-T/2}^{T/2} V(t)V(t + \tau)dt
  $$

  The autocorrelation function of a random process describes the correlation between
the processes at different points in time. It provides a measure of the memory of the process. \( \tau \) is the interval between two time points as the continuous variable changes with time. At \( \tau = 0 \), the auto-correlation function gives the mean square value of the fluctuations: \( \psi_V(0) = \overline{V(t)^2} \).

An alternative yet more informative way to characterize fluctuations is to study them in the frequency domain. The quantity in frequency domain \( V_T(\omega) \) is related to time domain signal \( V(t) \) via Fourier transform and inverse Fourier transform:

\[
V_T(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-T/2}^{T/2} V(t) e^{i\omega t} dt \\
V(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} V_T(\omega) e^{-i\omega t} d\omega
\]

(4.5)

(4.6)

The total energy of the signal, \( E \), can thus be presented in the frequency domain showing the distribution of the energy of the signal at different frequencies:

\[
E = \lim_{T \to \infty} \int_{-T/2}^{T/2} V(t)^2 dt = \frac{1}{2\pi} \int_{-\infty}^{\infty} |V_T(\omega)|^2 d\omega
\]

(4.7)

The average power of the fluctuation is therefore written as (Parseval Theorem):

\[
P = \lim_{T \to \infty} \frac{1}{T} \int_{-T/2}^{T/2} V(t)^2 dt = \lim_{T \to \infty} \frac{1}{2\pi} \int_0^{\infty} \frac{2 |V_T(\omega)|^2}{T} d\omega = \frac{1}{2\pi} \int_0^{\infty} S_V(\omega) d\omega
\]

(4.8)

where the power spectral density (PSD) \( S_V(\omega) \) of the fluctuating quantity \( V(t) \) is defined by:

\[
S_V(\omega) = \lim_{T \to \infty} \frac{2 |V_T(\omega)|^2}{T}
\]

(4.9)

In noise analysis, the PSD represents the amount of energy in each normalized frequency band and is usually used to characterize the noise level. If the fluctuating quantity is voltage, the unit of \( S_V(\omega) \) is \( V^2/\text{Hz} \). The Parseval Theorem shows that the integration of PSD over all frequencies is equal to the variance of the signal, provided the mean value of the signal is zero.

\[
\overline{\delta V^2} = \int_0^{\infty} S_V(f) df
\]

(4.10)

where \( f = \omega/2\pi \).
4.2.2 Types of noise

In solids, several types of noise may exist and contribute independently to the overall fluctuations. Although the origins of the noise can be diverse and even specific to samples, a clear classification of noise can still be made according to the frequency dependence of its PSD. Four common types of electrical noises are: thermal noise, shot noise, flicker noise (or $1/f$ noise) and random telegraph noise (RTN).

Thermal noise exists in almost all kinds of electronic devices as a background noise. It is first observed by Johnson in 1927 [146] [147] and the thermodynamic-based theoretical calculation for thermal noise was given by Nyquist in 1928 [148]. Also referred to as Johnson-Nyquist noise, thermal noise falls into a category called “white noise” since its PSD contains all the frequencies and does not vary with frequency, just as white light has all frequencies of colors. For a device with resistance $R$ at temperature $T$, the PSD of thermal voltage noise is $S_V(f) = 4k_BT R$, and the mean square voltage fluctuation is $\overline{V_n^2} = 4k_BT R \Delta f$. Here, $\Delta f$ is the frequency bandwidth in which the voltage is measured. Thus, shrinking and matching the measurement frequency bandwidth with the frequency of the desired signal is an effective way to improve the signal-to-noise ratio. Narrow-band measurements such as phase sensitive detection in lock-in amplifiers is a good example of such a reduction of thermal noise.

Another important white noise is shot noise, which is first observed by Schottky in a thermionic tube in 1918 [149]. Shot noise is caused by the random and discrete nature of the carrier motion. An intuitive picture is to think of electrons moving in an applied potential running across an energy barrier. The potential energy of electrons builds until they have enough energy to cross the barrier. When they pass the barrier, potential energy is released abruptly and transformed into kinetic energy, creating fluctuations of current. The PSD of shot noise is also a flat spectrum expressed as current noise: $S_I(f) = 2qI$. The corresponding fluctuating current is: $I_{sh} = \sqrt{2qI \Delta f}$ in $A/\sqrt{Hz}$. Here $q$ is the charge of the carriers, $I$ is the average current and $\Delta f$ is the frequency bandwidth. Shot noise has been observed in electronic devices such as p-n junctions [150] and magnetic tunnel junctions [151]. It does not depend on temperature and is always associated with current flow and the charge of the carrier. The signal-to-noise ratio of shot noise in devices is $I/\sqrt{2qI} \propto \sqrt{I}$, which suggests that applying a larger current will improve the measurement sensitivity.

Apart from white noise, noise can have other colors based on its dependence on frequency. $1/f$ noise, or flicker noise, has the color of pink, since its PSD follows the $1/f$ dependence, and if you mix frequency components of light according to the “$1/f$”
law, you will get a pink color. $1/f$ noise becomes especially prominent at low frequencies and is more likely to be obscured by thermal noise or shot noise at higher frequencies. Low-frequency $1/f$ noise is found in a wide variety of solid state systems, such as metals, semiconductors, tunnel junctions, spin glasses, GMR sensors, carbon nanotubes and even superconducting quantum interference devices (SQUID) [152]. It has also been observed in many other systems such as earthquakes, water levels and even human heartbeats. Extensive efforts have been devoted to understanding the ubiquitous nature of the $1/f$ noise. However, it is found that despite the generality of the $1/f$ noise, there is no satisfying universal explanation and it is advised that the $1/f$ noise should be studied in specific physical systems.

The $1/f$ noise has the PSD showing $S_V \propto 1/f^\alpha$, where the exponent $\alpha$ is close to 1. In the scope of our study on electrical noise in solids, the $1/f$ noise is thought to be caused by fluctuations in the mobility and/or in the number of carriers as a result of defects and impurities.

![Figure 4.1](image.png)

**Figure 4.1.** Illustration of a two level system separated by an energy barrier.

Another type of noise in solids that has aroused great interest in recent years is random telegraph noise (RTN). It is a non-Gaussian noise, which means it comes from a small number of independent events, or “fluctuators”. Although often found in sub-micron samples, RTN is not limited to small size samples. RTN arising from magnetization reversal of small magnetic clusters has also been observed in macroscopic samples [153] [154]. The simplest case of RTN is the fluctuation between two distinct states. As shown in Fig.4.1, two well defined states are separated by an energy barrier. The barrier heights seen from the two states are $E \pm \Delta E$. Each state couples to the charge carriers in a different way so that their contributions to the conductivity are different. The switching process between the two states thus results in discrete jumps in sample resistance.
Although the switching takes place randomly, the mean time occupied in each state is determined by statistics.

Considering thermal activated processes only, the average time $\tau_i(T)$ spent in the $i^{th}$ state is $\tau_i = \tau_0 \exp\left(\frac{E_i^{\pm} \Delta E}{k_B T}\right)$ ("+" for $i=1$ and "−" for $i=2$), where $\tau_0$ is the attempting time. The calculated PSD of the noise is given by [145]:

$$S_V = \frac{S_V^0(0)}{\cosh(\Delta E/k_B T)[\cosh^2(\Delta E/k_B T) + \omega^2 \tau_0^2]}$$

(4.11)

where $\tau^{-1} = \tau_1^{-1} + \tau_2^{-1}$ and $S_V^0(0)$ is the zero-frequency PSD at $\Delta E = 0$. The PSD is a Lorentzian spectrum with a corner frequency equal to $\tau^{-1}$. As the $\Delta E$ increases, the double-well becomes less symmetric and the noise becomes lower since the lower energy state is more stable. In the limit of $\Delta E = 0$, it is easy to obtain:

$$S_V(f) \propto \frac{\tau}{1 + 4\pi^2 f^2 \tau^2} = \frac{f_c}{f_c^2 + 4\pi^2 f^2 \tau^2}$$

(4.12)

Two characteristic regions can be identified: (1) $f \gg f_c$, $S_V \propto 1/f^2$, where $f_c$ is the corner frequency defined as $f_c = 1/\tau$; (2) $f \ll f_c$, the spectrum is almost flat. Usually, it is more convenient to plot $f \times S_V(f)$ as a function of frequency because it shows a peak at which the corresponding corner frequency can be easily obtained. Observing the corner frequency shift with temperature or external field is a very effective means to study the energies and associated dynamics of the system.

### 4.3 General noise measurement techniques

The simplest scheme to measure noise is the 4-terminal DC technique, as shown in Fig.4.2(a). It basically resembles the setup of a 4-probe resistance measurement, except that a capacitor $C$ is used as a high pass filter to block the DC offset. However, the measurement frequency range is limited to above $f_{min} \sim 1/R_i C$, where $R_i$ is the input resistance of the preamplifier. This setup also suffers from fluctuations in external sources such as voltage source, temperature instabilities etc..

An improvement over the 4-terminal DC setup is the 5-terminal scheme shown in Fig.4.2(b). The two branches of the Hall bar, along with two balancing resistors $R_1$ and $R_2$, form a bridge-type circuit to eliminate the DC offset. Since the external factors are minimized due to the bridge balancing, the intrinsic sample fluctuations are less sensitive to the external sources, and this technique does not have a limitation of measurement frequency.
Figure 4.2. (a) 4-terminal DC noise measurement setup. (b) 5-terminal DC setup using a bridge-type design.

An AC method was also introduced by Scofield [155] to avoid the dominant 1/f noise from the preamplifier at low frequencies. A circuit diagram of this technique is shown in Fig. 4.3. The sensitivity of this technique is not limited by the low frequency preamplifier noise, which increases significantly when frequency is lowered, but, rather, by the preamplifier noise at the modulation frequency. The employment of the phase-sensitive detection (PSD) technique enables the shifting of driving frequency to higher frequency band where the preamplifier has lower noise, and the five-probe Wheatstone bridge geometry provides the advantages of eliminating contact noises and external perturbations. In real experiments, a lock-in amplifier provides the excitation current, and amplifies and demodulates noise sidebands produced by fluctuation $\delta V(t)$.

4.4 Noise spectroscopy in (Ga,Mn)As

Point defects – specifically Mn interstitials ($\text{Mn}_I$) and As antisites – play an important role in the complex interplay between ferromagnetism and electronic structure in (Ga,Mn)As. Both these defects are donors that compensate for the hole-mediated ferromagnetic exchange and hence suppress ferromagnetic order [6]. Theoretical calculations have also predicted that $\text{Mn}_I$, in addition to compensating holes, may also quench ferromagnetism because of antiferromagnetic coupling with neighboring substitutional Mn atoms [156]. However, there is no explicit experimental evidence for such a coupling or for any magnetic activity of $\text{Mn}_I$.

On the other hand, it is well known that electrical noise provides a powerful probe
into the physics underlying the coupling between spin and charge transport in complex magnetic materials such as the manganites [157] [158] [159] and spin glasses [160]. The similarity of $R$ vs. $T$ curves between (Ga,Mn)As and manganites, in which resistivity peaks are associated with $T_C$, and the spin-charge interaction in (Ga,Mn)As piques the interest in using electrical noise measurement as a probe to glimpse the interplay between spin transport, hole localization and Mn interstitials in the ferromagnetic semiconductor (Ga,Mn)As.

4.4.1 Sample preparation and experimental details

(Ga,Mn)As epilayers are fabricated using low temperature molecular beam epitaxy (MBE) on (001) semi-insulating, epiready GaAs substrates following the growth of a 170 nm thick high-temperature buffer of GaAs epilayer.

Three samples are selected for noise measurements, with the following (as grown) characteristics (Table 4.1) determined using secondary ion mass spectrometry (SIMS), room temperature Hall effect (up to 14 T) and superconducting quantum interference device magnetometry (SQUID).

These data indicate that samples A and B have similar net Mn concentrations, but that the latter has a larger Mn$_I$ density. The higher level of disorder in sample B results in significantly more localization at low temperatures: the resistivity of samples A and B at 4.2 K is 12 m$\Omega$cm and 55 m$\Omega$cm, respectively. We also measured a control sample
Table 4.1. Characteristics of samples for noise measurement

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>d (nm)</th>
<th>Mn Concentration</th>
<th>Hole Density ($cm^{-3}$)</th>
<th>$T_C$(K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>050323B (Sample A)</td>
<td>30</td>
<td>0.06</td>
<td>$p \sim 5.6 \times 10^{19}$</td>
<td>55</td>
</tr>
<tr>
<td>060215A (Sample B)</td>
<td>30</td>
<td>0.066</td>
<td>$p \sim 4.4 \times 10^{19}$</td>
<td>50</td>
</tr>
<tr>
<td>020827A (Sample C)</td>
<td>50</td>
<td>0.08</td>
<td>$p \sim 1 \times 10^{21}$</td>
<td>100</td>
</tr>
</tbody>
</table>

consisting of 100 nm thick epilayer of p-GaAs (doped with Be) with $p \sim 10^{21} cm^{-3}$ at 300 K. Samples are patterned using a wet mesa etch into a 50 µm wide Hall-bar-like geometry along the [110] crystalline direction, with a spacing of 200 µm between adjacent longitudinal voltage probes (contacted with In).

Resistance noise measurements are carried out using a five-probe ac scheme [155], with a lock-in amplifier (SR830) providing the excitation current and demodulating the fluctuation signal. The output of SR830 is fed into a SR785 spectrum analyzer to get power spectrum. The resistances of the balancing resistors are chosen to be at least 10 times larger than the sample resistance to minimize their contributions to the overall noise. In typical ac noise measurements, the excitation frequency is chosen to lie in the eye of the noise figure of the preamplifier (100Hz-1KHz). However, in (Ga,Mn)As, we selected an excitation frequency $f_0=43Hz$ because of a strong frequency dependence of resistance at high frequencies.

The resistance fluctuations modulate the carriers to produce noise sidebands, which are demodulated by phase-sensitive detection on the lock-in amplifier. Thus, the validity of the AC measurements is limited to $f < f_0/2$, where $f$ is the frequency of the noise spectrum [155]. By shorting the inputs of the lock-in, we measured the intrinsic noise from the instrument under different settings of the low-pass filter. Fig.4.4(a) shows the PSD with different time constants of the low-pass filter. It is found that 3ms time constant gives a flat band response up to 12Hz, setting the upper limit of our spectra. Then the output low-pass filter of the lock-in is set to have a 3ms time constant with a rolloff of 24dB/Oct. A similar parameter setting was also reported in noise measurement using SR830 lock-in by another group [161].

Another important parameter that can affect the intrinsic noise from the instrument is the line filter on SR830. Fig.4.4(b) shows that the application of line filters indeed raises the noise floor. Since the measured spectra is far below 60Hz, the application of line filters is not necessary. Under these conditions – ($f_0=43Hz$, time constant=3ms with a rolloff of 24dB/Oct and no line filter)– the intrinsic noise background from the
instrument is measured to be $2.7 \times 10^{-16} \text{V}^2/\text{Hz}$ at room temperature.

For all the samples, a quadratic dependence of PSD on the applied bias voltage is first confirmed, indicating that the voltage fluctuations we measured indeed stem from the fluctuations of sample resistance [162]. Fig. 4.5 illustrates one of the examples of the quadratic bias dependent PSD from sample B.

Later on, the output voltage from the lock-in is set in the range 3V-5V rms, which gives a current of 5µA-10µA, depending on balancing resistors and sample resistances.
Figure 4.5. (a) PSD spectra with different bias voltages. (b) PSD at 1Hz plotted as a function of bias voltage, showing a quadratic voltage dependence.

A reasonable current density of $\sim 10^2$ A/cm$^2$ ensures the excess noise is higher enough to stand out from background noise, but low enough to avoid substantial heating.

4.4.2 Temperature dependence of noise spectra

Noise measurements in the higher conductivity control sample and the high $T_C$ sample (C) do not yield significant noise. This is most likely a limitation of our measurement setup which limits us to a maximum excitation current of 10$\mu$A: this is too low to yield measurable voltage fluctuations from these low-resistivity samples. We note that, with the highest possible current, measurements on sample C do show a 1/f tail below 1 Hz, but the spectrum above 1 Hz is overwhelmed by background noise. Hence, we focus our discussion on the lower conductivity samples A and B where the noise signal is readily measured.

Figure 4.6(a) shows the normalized PSD of the voltage fluctuations ($S_V/V^2$) for sample A at different temperatures, demonstrating 1/f-resistance noise over a wide temperature range from 4.2K to 65K. The magnitude of 1/f noise is characterized using the phenomenological equation [163]:

$$S_V(f) = \frac{\gamma}{n_c \Omega f^\alpha}$$

where $\gamma$ is the Hooge parameter, $V$ is the voltage across the sample, $n_c$ is the carrier density and $\Omega$ is the sample volume. Using $n_c \sim 1 \times 10^{20}$/cm$^3$, we obtain $\gamma = 3 \times 10^{-2}$, comparable to typical values reported in metals ($\sim 10^{-2}$-$10^{-3}$) [145]. The exponent $\alpha$
varies from 0.96 to 1.18 over the temperature range $4.2K < T < 65K$.

![Normalized PSD’s of sample A at four temperatures drawn in the same scale. Dashed line shows $1/f$ relation. The $\alpha$ exponent of $1/f^\alpha$ varies from 0.96 to 1.11 within fitting accuracy. (b) Temperature dependence of the integration of normalized PSD (left) and longitudinal resistance versus temperature (right) for sample A (lines are guides for eyes). Inset displays the $\rho_{xx}$ vs. $T$ curve as cooling down the sample, showing a metallic behavior.](image)

**Figure 4.6.** (a) Normalized PSD’s of sample A at four temperatures drawn in the same scale. Dashed line shows $1/f$ relation. The $\alpha$ exponent of $1/f^\alpha$ varies from 0.96 to 1.11 within fitting accuracy. (b) Temperature dependence of the integration of normalized PSD (left) and longitudinal resistance versus temperature (right) for sample A (lines are guides for eyes). Inset displays the $\rho_{xx}$ vs. $T$ curve as cooling down the sample, showing a metallic behavior.

The temperature-dependence of the relative fluctuation variance $\langle \delta V^2/V^2 \rangle$ is obtained by integrating the normalized noise $S_V/V^2$ over the range 125 mHz – 11 Hz. This is shown in Fig.4.6(b), along with the temperature dependence of the longitudinal resistivity ($\rho_{xx}$). Our measurements do not indicate any detectable correlation between
the noise amplitude and $T_C$ (where $\rho_{xx}$ has a peak). This contrasts with the man-
ganites where the integrated noise has a peak at $T_C$ [158], attributed to a percolative
phase transition between the charge-ordered and FM phases [159]. We speculate that in
(Ga,Mn)As, contributions to resistance fluctuations from possible mixed phases may be
overshadowed by the effect of disorder. At low temperatures where $\rho_{xx}$ goes through
a minimum, we observe an increase in noise amplitude presumably from the increased
localization of holes.

![Figure 4.7](a) A superposition of relaxation processes with a broad distribution of relaxation
times gives $1/f$-like behavior. (b) Illustrative cartoon of hopping conduction (red circle – Mn
acceptor sites; white circle – holes)

To investigate the origin of the $1/f$ noise, we recall that a single fluctuator with
a relaxation time $\tau$ yields a Lorentzian PSD with the form $S(f) \propto 2\tau/1 + (2\pi f \tau)^2$.
An ensemble of fluctuators with a broad distribution of relaxation times then produces
a $1/f$ spectrum due to the superposition of many Lorentzians [162], as illustrated in
Fig.4.7(a). $1/f$ noise is generally considered to be specific to the system studied. In
solid state systems, there are several possible sources of $1/f$ noise [145 163 164]. One
is charge trapping/de-trapping at the surface/interface, referred to as the McWhorter
Model. The other is the DDH model which describes thermally activated processes
with a broad distribution of transition energies. Recently, studies have also found that
donor/acceptor traps in hopping transport can give rise to $1/f$ noise [165 166]. Con-
sidering the high carrier concentration and low mobility in (Ga,Mn)As, the calculated
mean free path is very small ($\sim 1$nm), putting (Ga,Mn)As into the category of dirty
metals. Recent evidence of impurity band conduction in (Ga,Mn)As further suggests
that hopping conduction in Mn impurity band may be a plausible reason for $1/f$ noise.
Figure 4.8. (a) Normalized PSD of sample B at different temperatures. Dashed line shows $1/f$ and $1/f^2$ relations. Strong deviations of PSD from $1/f$ are clear over range $6 \, K \lesssim T \lesssim 29 \, K$. (b) Temperature dependence of the integrated normalized PSD (left) and $\rho_{xx}$ (right) for sample B (lines are guides for eyes). Inset shows the $\rho_{xx}(T)$ over an expanded temperature range.
Holes hop between Mn acceptor sites, and these Mn acceptors can act as traps on the holes’ hopping path to trap and release them with a wide spectrum of relaxation times (Fig.4.7(b)). The increase of noise level at low temperatures may be due to the strengthening of localization.

A different scenario was found on sample B, which has a $T_C$ and net Mn concentration similar to that of sample A, but has a higher Mn interstitial density. Similar to what is observed in sample A, there is no evidence for a correlation between noise characteristics and $T_C$, and there is a marked enhancement of noise at low temperatures. In contrast with sample A, however, we find clear deviations from $1/f$-noise over the temperature range from 6 K to 29 K (Fig.4.8(a)).

Further, the enhancement of noise at low temperatures is far more pronounced compared with sample A: as shown in Fig.4.8(b), the relative rms fluctuation peaks at 10 K, reaching a value that is almost two orders of magnitude higher than that above 30 K. At the lowest temperature measured (4.2 K), $1/f$ behavior is restored, accompanied by a clear drop in the noise amplitude. We attribute the deviation of PSD from $1/f$ to the dominance of a few fluctuators, each of which has a different relaxation time. The $1/f^2$ tails observed at higher frequencies in Fig.4.8(a) are evidences of such Lorentzian spectra and is corroborated by time-domain measurements (Fig.4.9(a)) that show RTN, with $\delta V/V \sim 0.01\%$.

We now focus on the temperature range over which we clearly observe RTN (~6K to ~16K). Note that the deviation from $1/f$ persists to ~29K, but this may simply arise from a shift of the corner frequency to values larger than our upper measurement limit (11 Hz). The observed RTN can be modeled as a simple two-level fluctuator (TLF) in which the barrier heights to escape from one state to another are $E+\delta E$ and $E-\delta E$ respectively. The average time spent in each state is given by $\tau_i = (2\pi f_0,i)^{-1}\exp(E \pm \delta E/k_BT)$. The overall relaxation time of the whole system is $1/\tau_c = 1/\tau_1 + 1/\tau_2$. The normalized PSD is then given by a Lorentzian term plus a small $1/f$ background:

$$\frac{S_V(f)}{V^2} = \frac{2\pi \tau_c A}{1 + (2\pi \tau_c)^2 f^2} + \frac{B}{f} = \frac{f_c A}{f_c^2 + f^2} + \frac{B}{f},$$

(4.14)

where A and B are the relative strengths of the $1/f$ and $1/f^2$ contributions, respectively, and $f_c = 1/(2\pi \tau_c)$ is the corner frequency of the Lorentzian. These parameters can be extracted by fitting the frequency dependence of $fS_V/V^2$ [167] (Fig.4.9(b)).

Figure 4.9(c) shows the temperature dependence of the fitting parameters A, B and $f_c$, demonstrating that the temperature variation of the noise spectrum in Fig.4.8(b)
Figure 4.9. (a) Time-dependent voltage fluctuations in sample B, showing RTN. (b) $fS_V(f)/V^2$ at different temperatures in sample B. Solid lines are fits to Lorentzian spectra with a superposition of a $1/f$ background (Eq. 4.14). (c) Relative strengths of the Lorentzian term (A) and the $1/f$ term (B) obtained by fits to Eq 4.14. Inset shows the corner frequency $f_c$ vs. $T$. 
is dominated by that of the Lorentzian contribution. $f_c$ shifts monotonically toward higher frequencies with increasing temperature, indicating phonon activated processes. Further determination of thermally attempt frequency $f_0$ and barrier heights is found to be difficult due to the limited temperature range and exponential dependence of $f_0$ on $1/T$.

4.4.3 Magnetic field dependent noise measurement

To further explore the origin of the RTN, we measure the magnetic field dependence of the noise spectra with an in-plane magnetic field $\vec{H}||[110]$ (Fig.4.10(a)). These measurements are compared with the switching of the magnetization ($\vec{M}$), tracked using the giant planar Hall effect (GPHE) [82]. Prior to the noise measurement at positive fields, the magnetization was first saturated at a large negative field ($< -0.5T$), then the field was reduced to zero and ramped to a positive setpoint to start noise measurement.

From the field dependent transverse resistance measured at 9K (blue and red circles in Fig.4.10(a)), the 90 degree magnetization switching can be clearly identified [82]. The biaxial anisotropy in (Ga,Mn)As results in easy axes close to the [100] and [010] directions: at $H = 0$, $\vec{M}||[\bar{1}00]$, corresponding to one of the four energy minima. The enhancement and subsequent sudden quenching of noise at low field ($0 < H \lesssim 900\text{Oe}$) signals the nucleation of small domains, followed by their coalescence into a single macroscopic domain with $\vec{M}||[010]$. The second switching event, when $\vec{M}$ rotates from [010] to [100], is also accompanied by the enhancement and subsequent suppression of noise, although the changes are less abrupt because the magnetization reversal process is dominated by coherent rotation rather than domain nucleation (also supported by the less sharp change in Hall resistance).

At higher fields ($0.5T < H < 2T$), $\vec{M}$ gradually rotates coherently towards field direction $\vec{H}$, accompanied by a striking enhancement in integrated noise and RTN, before the PSD reverts to $1/f$-behavior at $H > 3T$. A fit to Eq.4.14 shows that the Lorentzian contribution peaks at $H = 1.2T$. Further, in time domain, the dwelling times of RTN for up and down states change systematically with field (Fig.4.10(b)), implying field-dependent energy barriers. The field dependence of the noise amplitude and energy barriers both suggest a magnetic origin of the TLF, rather than domain fluctuations, because a single domain state should have been achieved at high fields.
Figure 4.10. (a) Magnetic field-dependence of the integrated normalized PSD (solid squares) at $T = 9K$. For comparison, we also plot the GPHE with red (blue) circles for up (down) field sweeps. (b) Time traces of RTN at different fields, also at $T = 9K$.

4.4.4 Discussion of origins of the RTN

RTN is often attributed to trapping or de-trapping events of a single electron, in which case it only occurs in small scale samples, usually submicron [167]. The observation of RTN in our 50um wide sample suggests that the fluctuator comes from macro-scale long range interactions.

One possible explanation is the existence of small ferromagnetic clusters with two magnetic states. The energy barrier between the two states is a product of the difference in the projection of the magnetic moment along the field direction and the magnetic field: $\Delta E = \Delta mH$ [153]. In this case, the barrier heights may be written in terms of a field-independent and a field-dependent contribution. The average time in each states is: $\tau_i = (2\pi f_0)^{-1}\exp(\pm \Delta E + E_i(H)/k_BT)$. The ratio of dwelling time is then:
Figure 4.11. (a) Magnetic field dependence of energy difference $\Delta E$ between the two states of the TLF in sample B at $T = 6K$. (b) Comparison between RTN at $T = 6K$ in as-grown and annealed pieces of sample B.

$\tau_1/\tau_2 = \exp(2\delta E + \Delta E(H)/k_B T)$, where $\tau_1$ and $\tau_2$ is the average time spent in the “spin up” and “spin down” orientations, respectively. For a cluster with magnetic moment $m$, we expect $\Delta E(H) = mH$. By measuring the mean dwelling time in each of the two states (averaged over 10 minutes) at different magnetic fields and plotting $k_B T \ln \tau_1/\tau_2$ as a function of magnetic field, we obtain a linear field dependence, from which $m \sim 20\mu_B$ is extracted (Fig.4.11(a)).

The HRTEM images and electron diffraction pattern (Fig.4.12), as well as SQUID data (not shown) have no proofs of MnAs second phase, indicating sample’s homogeneity. The small amplitude of the magnetic moment also suggests evidence for nanoscale magnetic clusters with a few Mn atoms. Since the RTN decreases significantly upon annealing (Fig.4.11(b)), we infer that Mn interstitials must play a direct role in these clusters. The prediction of an antiferromagnetic interaction between interstitial Mn and substitutional Mn leads us to speculate that the Coulomb interaction between these
may result in nanoscale antiferromagnetic clusters with uncompensated spins. These clusters may coherently influence the hopping conduction of holes occupying Mn impurity band, possibly achieving two states with different conductivities. The onset of magnetic correlation in such clusters at low temperature would explain the pronounced enhancement of noise at $T \sim 10K$. At even lower temperatures ($T \lesssim 10 K$), the fluctuations of these clusters are frozen out and carrier hopping conduction is not affected. Equivalently, at higher temperatures, spin flip fluctuations of these clusters are frozen out by the application of an external magnetic field, quenching the RTN as a result of the increased barrier heights.

![Figure 4.12](image)

Figure 4.12. (a) High resolution cross sectional TEM image of sample B (b) Electron diffraction pattern of sample B.

### 4.5 Summary and conclusions

In summary, electrical noise measurements can shed new light into the interplay between transport, localization and magnetism in (Ga,Mn)As. We have studied the noise properties of (Ga,Mn)As epilayer samples of different localizations. For moderate localization, we find a $1/f$ normalized power spectrum density over the entire range of temperatures studied ($4.2K < T < 70K$). However, for stronger localization and a high density of Mn interstitials, we observe Lorentzian noise spectra accompanied by random telegraph noise. Magnetic field dependence and annealing studies suggest that interstitial Mn defects possibly couple with substitutional Mn atoms to form nanoscale magnetic clusters characterized by a net moment of $\sim 20\mu_B$ whose fluctuations modulate hole transport. Surprisingly, we do not observe any change in noise in the vicinity of the Curie temperature, ruling out a percolative transition with coexisting phases as in the
manganites.
Chapter 5

Hybrid magnetic metal/semiconductor bilayers

5.1 Introduction

Fundamental studies of spin-dependent transport and exchange coupling in ferromagnetic (FM) multilayers have played a central role in spintronics [168, 169], with key emphasis on phenomena such as tunneling magnetoresistance [170], the spin valve effect [171, 172, 173], and exchange biasing [174]. A number of spin-electronic devices such as hard drive read heads, magnetic sensors [175] and magnetic random-access memories (MRAMs) [176] have been introduced, and they are playing an important role in today’s information technology.

Within the particular context of semiconductor-based spintronics [177, 8, 7], such studies have centered on heterostructures derived from the ferromagnetic semiconductor (FMS) (Ga,Mn)As [10, 11], aiming at FMS versions of conventional metal spintronics phenomena such as the spin valve effect in trilayers [178], exchange biasing by an antiferromagnetic (AFM) pinning layer [179, 180], tunneling magnetoresistance [95, 116] and current-driven magnetization control [75, 73].

In the following chapters, we report the observation of exchange biasing in hybrid ferromagnetic metal/semiconductor heterostructures and its application. In Chapter 5, we discuss magnetization and magnetoresistance measurements in MnAs/(Ga,Mn)As bilayers and MnAs/Be-GaAs/(Ga,Mn)As trilayers, showing a spin-valve effect in these self-exchange biased heterostructures. In Chapter 6, I will discuss one application of this exchange coupling: namely, pinning one (Ga,Mn)As layer in a magnetic tunnel junction. We observe a large and quasi-reversible tunnel magnetoresistance that allows...
us to probe the exchange-spring configuration within the exchange-biased (Ga,Mn)As layer.

5.1.1 Introduction to spin valve effect

The giant magnetoresistance (GMR) effect describes the phenomenon where the resistance of a thin film structure, composed of alternating ferromagnetic and nonmagnetic metal layers, changes in an external magnetic field. As the magnetizations of the ferromagnetic layers align in parallel, the resistance drops significantly. A spin valve, which consists of two FM layers separated by a non-magnetic conducting layer, is a simplified structure to exploit GMR. The two FM layers often have different coercivities so that the two layers can be aligned in parallel or anti-parallel configurations in an external field, giving two resistance states.

The spin valve effect can be qualitatively explained using the Mott’s model [181, 182]. There are two important points that should be taken into consideration in Mott’s model. First, the electrical conductivity in metals should be able to described by two largely independent conducting channels in parallel. This requires that the probability of spin flip scattering in these metals is much smaller than the probability of the scattering processes in which the spin is conserved. The up and down spins do not mix with each
other over long distances. Second, the scattering rates of up-spin and down-spin electrons are quite different in ferromagnetic metals. The electrical current is mainly carried by electrons from \( sp \) valence band due to their low effective mass and high mobility, while the \( d \) band provides final states for the scattering of the \( sp \) electrons. In ferromagnets, the \( d \) bands are exchange-split, which means the density of states (DOS) of up-spin and down-spin electrons are not equal at the Fermi energy. Because the probability of scattering into these states is proportional to their DOS, the scattering becomes spin dependent, so the two channels of spin experience different scattering rates in ferromagnets [173].

Mott’s theory gives an intuitive model to explain GMR in metallic magnetic multilayers. We assume that scattering is strong for electrons with spin anti-parallel to the magnetization and is weak for electrons with spin parallel to the magnetization. In Fig.5.1(a), the spin-up electrons experience little scattering because their spins are parallel to the magnetization of the layers, while the spin-down electrons experience strong scattering. The total resistance of the two independently spin conducting channels is obtained by putting the two channel resistances in parallel: 
\[
R_P = \frac{2R_{up}R_{down}}{R_{up} + R_{down}}.
\]

\( R_P \) is low due to the high conductivity in one channel. On the other hand, in the anti-parallel configuration as shown in Fig.5.1(b), both the spin-up and spin-down electrons experience strong scattering within one of the layers, giving a high overall resistance: 
\[
R_{AP} = \frac{(R_{up} + R_{down})}{2}.
\]

The mean free path, or the relaxation time, is determined by both the scattering potentials (impurity, defects, etc) which could be spin-dependent or independent and by the DOS at the Fermi energy. However, in real spin valve devices, the spin-dependent scattering potentials can be disguised by spin-independent scattering from defects such as grain boundaries and misfit dislocations, so that the average value of the scattering potentials can be spin-independent. In this case, the role of spin-polarized band structure becomes decisive.

### 5.1.2 Exchange bias

Exchange bias in antiferromagnetic (AFM)-ferromagnetic (FM) structures has been extensively studied and it has became one of the key elements for developing spin-valve devices [174]. To obtain exchange biasing experimentally, the AFM/FM structure is cooled in a magnetic field through the Néel temperature of the AFM. During the cooling process, the AFM order is established, and the exchange anisotropy is created at the interface. The resultant exchange coupling causes the hysteresis loop of the FM layer to shift by an amount known as exchange field (\( H_e \)) to the opposite direction of the cooling
field, and it is usually accompanied by the broadening of the hysteresis (denoted by the coercivity $H_C$) (Fig. 5.2(a)).

The exchange biasing (EB) in (Ga,Mn)As has been demonstrated using a MnO overlayer as an AFM layer by our group [179, 183]. Fig. 5.2(b) shows the exchange biasing of (Ga,Mn)As layer upon different cooling procedures. Several follow-up studies on this system have focused on the annealing-dependence of EB [184], the temperature dependence of EB [185], the anisotropy using ferromagnetic resonance [180] and the magnetization reversal probed by planar Hall effect [186]. To obtain the MnO layer on the (Ga,Mn)As surface, a thin Mn layer is first deposited in vacuum and then subject to subsequent oxidization processes. The oxidization of Mn to form MnO grains is hard to control by annealing, resulting a poor yield [179], and over-annealing may degrade the EB due to the formation of other oxides of manganese [185]. Moreover, the insulating nature of MnO precludes the fabrication of vertical transport devices for applications.

On the other hand, the exchange biasing has also been observed in a FM/FM system,
which consists of one hard ferromagnetic layer and one soft ferromagnetic layer with distinct coercivities. This configuration often results in an “exchange spring” effect, where the magnetization of the soft layer can be reversibly wound (or unwound) with respect to the hard layer by sweeping the magnetic field [188, 189]. For example, a positive exchange bias was found in pervoskite manganites [187, 190] in the sense that the hysteresis loop is shifted to the same direction as the biasing field (Fig.5.3). This indicates the exchange interaction between the two layers are antiferromagnetic. The type of the exchange coupling is usually determined by the exchange constant $J_{ex}$. When the exchange constant is zero, the two layers are completely decoupled. When $J_{ex}$ is negative, the anti-parallel configuration is favored, giving positive exchange biasing. If $J_{ex}$ is positive, then the parallel configuration is favored, resulting in negative exchange biasing.

In traditional metallic spin-valve devices, exchange biasing and spin dependent scattering often work together to achieve device functionalities. As illustrated in Fig.5.4, an AFM layer is used to pin one of the FM layers, leaving the top FM layer free to switch...
in a small magnetic field. This gives distinct coercivities for the two FM layers, as well as two resistance states depending on the alignment of the two layers, as seen in the magnetization and magnetoresistance curves in Fig. 5.4(b). In our study, however, we show that the exchange biasing and spin valve effect are simultaneously achieved in all ferromagnetic metal/semiconductor heterostructures, avoiding the complicity of using an AFM layer for (Ga,Mn)As based devices.

5.2 Sample preparation and device fabrication

MnAs/(Ga,Mn)As heterostructures were grown using low temperature molecular beam epitaxy (LT-MBE) on either semi-insulating (SI) or p-doped (001) GaAs substrates. On SI-substrates, a 170 nm thick high temperature buffer of GaAs was grown prior to the growth of the bilayers. For the samples grown on p-type substrates, the GaAs buffer layer was doped with Be to achieve the similar carrier concentration as the substrates (p~1E19/cm³). (Ga,Mn)As layers were grown at 235°C to 250°C depending on the Mn concentrations. After the growth of (Ga,Mn)As, the substrate temperature
Figure 5.5. 1× (a) and 2× (b) RHEED pattern for MnAs

was lowered to ∼190°C-200°C with As shutter open. Tanaka et.al. showed that the epitaxial orientation of MnAs depends on the growth of the first few monolayers. Type-A MnAs is generally obtained under the As-rich condition, while if a monolayer of Mn is first deposited, the grown MnAs shows type-B configuration. The difference between type-A and type-B MnAs has already been reviewed in Chapter 1.

In our case, a few monolayers of MnAs (∼2min) were first grown at this low temperature (∼200°C) to form a template for type-A MnAs. Then the substrate temperature was raised to ∼250°C to resume the growth of MnAs layer. We find that this protocol consistently produces high quality type-A MnAs as the RHEED shows streaky (1x2) patterns after the MnAs growth (Fig.5.5). Since the RHEED oscillations of MnAs were hard to measure due to the low growth rate, and the growth rate of MnAs strongly depends on the Mn flux and the growth temperature, we determined the thickness of the MnAs layer using high resolution cross sectional transmission electron microscopy (HRTEM) (Fig.5.6).

Here, we focus on two different sets of samples. Series A (Fig.5.7(a)) consists of five bilayer samples with 10nm type-A MnAs on top of (Ga,Mn)As (Mn concentration ∼6% \( t_{gma} = 20, 30, 50, 80, 120 \text{nm} \)). Series B (Fig.5.7(b)) are MnAs/p-GaAs/(Ga,Mn)As trilayer structures with a fixed (Ga,Mn)As thickness (30nm) and Mn composition (6%), and with varying thickness of the p-GaAs spacer layer (\( t_{\text{spacer}} = 1, 2, 3, 4, 6, 8 \text{ nm} \)). The p doping in the spacer is nominally designed to be similar to that in the buffer layer.

For the current-perpendicular-to-plane (CPP) transport measurements, the samples
grown on p-GaAs substrates are patterned into cylindrical mesas of 100 μm diameter using conventional photolithography and wet etching down to the p-substrates. After the etching, the samples are transferred into an E-beam evaporator with the photoresist on top of the mesas. A layer of SiO₂ is then deposited on the patterned mesa with a thickness larger than the heights of the mesas. A window is opened up by lifting-off SiO₂ in acetone when the photoresist on the mesa is dissolved. A second photolithography step is used to deposit 5nm Ti/100nm Au through the window on top of the mesa as an Ohmic contact. The bottom (side) contact is formed by evaporating Ti/Au directly onto the etched substrates. A pseudo-four-probe scheme is used with one current lead and one voltage probe on top of the mesa and separated current and voltage probes on the bottom contact (Fig.5.7(c)).

Magnetoresistance measurements are carried out in a CPP geometry using a DC source and digital multimeters. Since the resistance of the mesa is small (∼ 300mΩ), a current of 4 mA is used to achieve good signal-to-noise ratio and avoid sample heating. We have checked that the I-V characteristics are linear during all our measurements. Finally, we note that in all the magnetization and magnetoresistance measurements discussed here, the external magnetic field is applied along the [110] direction of the GaAs substrate. This corresponds to the [1120] direction of the MnAs layer and is the easy axis for type-A MnAs.
5.3 Magnetometry measurement

The magnetic properties of the samples are characterized using a DC SQUID magnetometer. We first discuss the temperature- and magnetic-field dependent magnetization of MnAs/ (Ga,Mn)As bilayer samples (Series A). Fig. 5.8 shows the temperature dependent remanent magnetization \( M(T) \) in four bilayer samples with \( t_{\text{GaMnAs}} = 30, 50, 80, 120 \) nm, measured in a field of 30 Oe after cooling down from room temperature in a 1T field. The data clearly reveal the two different Curie temperatures for the MnAs (\( T_C \approx 318 \)K) and (Ga,Mn)As (\( T_C \approx 75 \)K) layers. The major magnetization hysteresis loops are measured after first saturating the MnAs layer in a 20 kOe field. An example is shown in Fig. 5.8(b) for the sample with 50nm (Ga,Mn)As, revealing two distinct transitions representing the magnetization switching of (Ga,Mn)As (~100 Oe) and MnAs (~2 kOe). Similar data are obtained for all the bilayers with \( t_{\text{GaMnAs}} \geq 20 \) nm. However, SQUID measurements on the sample with \( t_{\text{GaMnAs}} = 15 \) nm are unable to resolve the temperature-
**Figure 5.8.** (a) Temperature-dependent magnetization $M(T)$ for bilayer samples with different (Ga,Mn)As thicknesses (070710-070713). The magnetization is shown per unit area. (b) Major magnetization loop $M(H)$ for a MnAs/(Ga,Mn)As (50nm) sample (070705A), showing two distinct coercive fields for the two FM layers ($T=4.2$K).

and field-dependent magnetization contribution from the (Ga,Mn)As layer.

Fig. 5.9 shows the minor hysteresis loop for the same sample. The minor loops are measured over a field range of $-1$ kOe $\leq H \leq +1$ kOe, after first saturating the MnAs layer in a positive(a) or negative 20 kOe field. Note the minor loop of the bilayer is actually the major loop (complete loop) of the (Ga,Mn)As layer. The displacement of the center of the hysteresis loop from zero field is unambiguous evidence for a “negative exchange biasing” of (Ga,Mn)As layer. In other words, the shift of the hysteresis loop is opposite to the magnetization of the MnAs layer. This indicates a FM coupling between the two layers, where the parallel alignment of the two layer is favored. Similar behavior is observed in all the bilayer samples with $t_{gma} \geq 30$ nm.

In Fig 5.10 we show that $H_e$ decreases monotonically with the thickness $t_{gma}$ of the (Ga,Mn)As layer. We find that $H_e$ is proportional to $1/t_{gma}$, qualitatively consistent with an analytical model for exchange-coupled FM/FM bilayers wherein a partial domain wall forms in the softer side of the interface [192]. Our data are inconsistent with the inverse square prediction of an earlier idealized model for an exchange-coupled FM/FM bilayer [193]. Finally, we note that magnetostatic “orange peel” coupling is unlikely to be responsible for the observed biasing effect since these epitaxial bilayers have relatively
Figure 5.9. Minor hysteresis loop (red circles) for the MnAs/(Ga,Mn)As (50nm) sample (070705A) after saturating the MnAs at +20 kOe (a) and -20 kOe (b). Part of the minor loops coincide with the major loops depending on the initializing condition. The arrows indicate the field sweeping direction. The minor loops are seen to shift to the opposite direction of the MnAs magnetization. The orientations of the two layers are also depicted.

smooth surfaces (with typical rms roughness $\sim 0.5$-2nm). While the inverse dependence of $H_e$ with $t_{gma}$ is superficially consistent with orange peel coupling, quantitative analysis [194] shows that the observed values of $H_e$ would require unphysical values for the wavelength of the roughness ($\sim 0.5$ nm). Fig.5.10 also shows that the coercivity $H_c$ of the (Ga,Mn)As layer (or the window of the minor loop) does not vary much with $t_{gma}$. This contrasts with observations in exchange biased FM/AFM bilayers where a larger $H_e$ is typically accompanied by a larger $H_c$.

5.4 A model for exchange bias

To calculate the exchange bias field, we use a partial domain wall (PDW) model analogous to the one used in AFM/FM systems [195, 192]. The magnetization of MnAs is considered to be fixed to the positive field direction along [110], and the bottom (Ga,Mn)As is free to switch in magnetic field as illustrated in Fig.5.11. The directions of magnetization in (Ga,Mn)As are described with respect to the MnAs magnetization. A PDW of thickness $t_1$ is formed in the (Ga,Mn)As near the interface with twisted spins. Assuming a strong coupling at the interface, where $\phi_1 \approx 0$ and $t_2 \approx t$, the energy density per unit area can be written as:
Figure 5.10. Dependence of the exchange bias field $H_e$ (circles) and the coercivity of (Ga,Mn)As (windows of the hysteresis loops) $H_c$ (squares) on $t_{GaMnAs}$ at 4.2K. The dashed line shows the fit to $1/t$ dependence.

\[
E = 2\sqrt{AK}(1 - \cos\varphi_2) - A_{ex} + K_u t \sin^2 \varphi_2 + 1/4K_c t \cos^2 2\varphi_2 - HMt \cos \varphi_2
\]  

(5.1)

The first term is the energy of the PDW, where $A$ is the spin stiffness of (Ga,Mn)As, and $K$ is the effective anisotropy constant, the second term the exchange coupling at the interface, the third and fourth two terms are the uniaxial and biaxial anisotropy energy in (Ga,Mn)As, and the last term is the Zeeman energy. When considering a strong cubic anisotropy, the energy minimum occurs at 45 and 135 degrees. Then the two switching field is determined by making the second derivative of the energy with respect to $\varphi$ greater than zero at 45° and 135°:

\[
\frac{\partial^2 E}{\partial \varphi_2^2} (\varphi_2 = \pi/4) > 0, \quad \frac{\partial^2 E}{\partial \varphi_2^2} (\varphi_2 = 3\pi/4) > 0
\]  

(5.2)

The above two equations give the two switching fields at these two energy minima:

\[
H > H_{C1} = \frac{-2\sqrt{2AK} - 4K_c t}{\sqrt{2Mt}}
\]  

(5.3)
The exchange biasing field is then found to be $H > H_{C2} = \frac{-2\sqrt{2AK} + 4K_c t}{\sqrt{2M}}$, showing a 1/t dependence. With typical parameters of a GaMnAs sample ($t=30$nm, $A \sim 0.4$ pJ/m, $K \sim 0.3$KJ/m$^3$, $M \sim 16$emu/cm$^3$ [69]), we calculated an exchange field of about 440 Oe for the 30nm (Ga,Mn)As, which is reasonably close to the experimental value of 375 Oe.

### 5.5 CPP transport measurement

In CPP magnetoresistance (MR) measurements, we observe a spin-valve effect as shown in Fig.5.12 for a bilayer sample with $t_{gma}=30$nm. The relative MR is characterized as $(R(H) - R(0))/R(0)$ in percentage, normalized to the resistance at zero field. In the major hysteresis loops (a), where both the MnAs and (Ga,Mn)As layers switch, a comparison between the transport data and SQUID data provides evidence for the spin valve effect, showing a high (low) resistance state when the (Ga,Mn)As and MnAs layers...
have antiparallel (parallel) alignment. The magnitude of the MR is small, ∼0.1%. As expected, the magnitude of the spin valve effect decreases with increasing temperature and eventually disappears above the $T_C$ of (Ga,Mn)As. Also at low temperatures, a positive MR background is seen, which is attributed to the MR contribution from the Be-doped GaAs buffer.

![Figure 5.12](image)

**Figure 5.12.** (a) Normalized CPP MR (in percentage) in a bilayer sample (070416A $t_{gma}$=30nm) in (a) major and (b) minor hysteresis loops at different temperatures. The data are offset vertically for clarity. The scale bars show the relative magnitude of the MR.

Fig5.12(b) shows the exchange biased spin valve effect when the external field is swept over a range corresponding to a minor hysteresis loop. Again, the effect decreases with increasing temperature, but interestingly reveals reversible behavior (hysteresis loop diminishes) well before reaching $T_C$, even though the exchange biasing persists up to $T_C$. This transition to reversible behavior at higher temperatures is also observed in SQUID measurements of the field-dependent magnetization on the sample sample. We speculate that, at higher temperatures still below the Curie temperature of (Ga,Mn)As, the coercivity of the (Ga,Mn)As layer becomes small enough to allow exchange spring behavior \[189\]. In this temperature range, the (Ga,Mn)As layer shows no hysteresis because its magnetization adiabatically follows the net field comprised of the external field and the local exchange field. It should be noted that not all the bilayer samples we measured
show the complete reversible behavior, which may be due to the anisotropy energy of the specific samples. There have been several reports on the giant magnetoresistance (GMR) effect in an exchange-spring configuration \[196, 197\]. However, those MR measurements were done in a current-in-plane (CIP) configuration, which usually includes a contribution from the anisotropy magnetoresistance (AMR). The CPP measurement here unambiguously provides the evidence for MR in an exchange-spring configuration.

The MR ratio given by the two channel model depends on the spin asymmetry in the two ferromagnetic layers and is written as \[173\]:

$$\frac{\delta R}{R} = \frac{(\alpha_1 - 1)(\alpha_2 - 1)}{\alpha_1(1 + q) + \alpha_2(1 + q^{-1})}$$  \hspace{1cm} (5.5)

where $\alpha_1 = \rho_{1\downarrow}/\rho_{1\uparrow}$ and $\alpha_2 = \rho_{2\downarrow}/\rho_{2\uparrow}$ are the asymmetry parameters for the two ferromagnetic layers. And $q = \rho_{1\uparrow}/\rho_{2\uparrow}$ is the ratio of the up-spin resistivities in the two ferromagnets. However, there are two main limitations for the two channel model: first, the mean free path should be longer than the layer thickness; second, the $sp$ band and $d$ band should be relatively independent. However, in (Ga,Mn)As, the mean free path is quite small ($\sim 1$nm), and there is a strong $pd$ hybridization. So the carrier scattering is strong in bulk (Ga,Mn)As for both majority and minority spins. We speculate that the spin asymmetry is more likely to happen at the interface. We cannot rule out the possible small tunneling barrier at the interface, although we observed a linear I-V characteristic in our samples. A detailed study has to be based on the calculation of band structures and density of states (DOS) at the Fermi energy in both (Ga,Mn)As and MnAs.

5.6 Trilayer structures with doped spacers

We also measured the magnetization and CPP MR in trilayer samples from series B in order to examine whether the interfacial exchange coupling can be mediated by a spacer layer. We note that previous studies of (Ga,Mn)As/AlAs/MnAs tunneling structures have suggested a weak coupling even in the presence of a tunnel barrier \[116\]. Fig.5.13 shows both magnetization and CPP MR measurements in a trilayer sample with a 2 nm spacer, confirming that the exchange coupling is mediated by holes in the spacer layer and that an exchange biased spin valve effect persists in such devices.

To further investigate the hole-mediated exchange coupling, exchange field as a function of spacer thickness is measured (Fig.5.14). We find that the exchange biasing is very weak when the spacer is larger than 6nm (around the anticipated spin diffusion length in Be-doped GaAs). Also the trilayer sample with 3nm undoped spacer shows
Figure 5.13. (a) Major and (b) minor hysteresis loops for a trilayer sample (070413A) with 2nm spacer layer at 4.2K, showing that exchange biasing persists even with a spacer layer. (c) Major and (d) minor MR loops for the same sample, showing the spin valve effect in the trilayer.

no exchange biasing in contrast to the sample with 3nm doped spacer (Fig.5.15). This suggests that the interaction between the two FM layers is RKKY type, in which the exchange is mediated by the free carriers in the spacer. A more detailed investigation of this phenomenon is beyond the scope of this thesis.

5.7 Summary and conclusions

In summary, we have demonstrated the CPP spin valve effect in self-exchange biased ferromagnetic MnAs/ (Ga,Mn)As bilayers, as well as in MnAs/ p-GaAs/ (Ga,Mn)As trilayers. The spin valve effect in these systems likely arises from spin-dependent scattering at the interfaces, but a detailed knowledge of the band structure, interfaces, and impurity scattering is required for a quantitative understanding. Nonetheless, the results reported here bring to light a new model system that can help address fundamental questions about the nature of spin-dependent transport and scattering at FM
Figure 5.14. Exchange bias field as a function of p-doped spacer thickness.

Figure 5.15. (a) The trilayer sample with a 3nm undoped GaAs spacer (071026B) shows no exchange biasing, as opposite to (b) the trilayer sample with a 3nm Be-doped GaAs spacer (071026C), which shows exchange biasing.

metal/semiconductor interfaces. Furthermore, the observation of exchange coupling between a metallic and a semiconducting ferromagnet suggests novel approaches for the epitaxial and lithographic engineering of magnetic properties such as the coercivity and the Curie temperature of FMS such as (Ga,Mn)As. Finally, we note that the exchange biasing and spin valve effect behavior reported here also appear to occur in recent studies of hybrid trilayer structures that use phase-separated GaAs:MnAs instead of single phase (Ga,Mn)As as the FMS layer [198].
Chapter 6

Exchange-biased magnetic tunnel junctions

6.1 Introduction

One of the applications of the exchange-biasing in (Ga,Mn)As using a FM MnAs layer is the exchange-biased magnetic tunnel junction (MTJ). MTJs exploit the tunnel magnetoresistance (TMR) arising from spin-dependent tunneling between two ferromagnets that are separated by a tunnel barrier. The TMR is sensitive to the relative magnetization orientations of the two ferromagnetic layers and also depends on the details of the electronic structure (such as the spin-dependent density of states). Apart from providing a wealth of fundamental insights into spin-dependent tunneling, MTJs now form a key component of contemporary magnetic recording technologies [170].

Much of our basic understanding of MTJs is derived from studies of metallic ferromagnets and is already at a very advanced stage. In contrast, ferromagnetic semiconductor (FMS) based MTJs have drawn attention in the context of emerging semiconductor-based spintronics [168] and are just beginning to be understood. MTJs based on diluted FMS (Ga,Mn)As with different types of spacer barriers have been fabricated and studied, revealing unique behaviors due to the nature of hole-mediated ferromagnetism and the intimate connection between magnetic anisotropy and electronic bandstructures [95, 96, 199, 200, 201, 202]. An example is the tunneling anisotropic magnetoresistance (TAMR), wherein the TMR also depends on the orientation of the magnetization with respect to crystal axes [201, 203].

(Ga,Mn)As based MTJs have several advantages: (1) The high quality single crystal (Ga,Mn)As can be epitaxially grown on III-V semiconductors semiconductors, allowing
easy integration into existing III-V based devices; (2) The band structure and anisotropy are controllable in (Ga,Mn)As by changing the Mn doping level or through the engineering of strains; (3) Novel quantum heterostructures such as quantum well (QW) or resonant tunnel junctions can be readily introduced. (4) All-semiconductor MTJs may provide new insights into the spin-dependent tunneling phenomena.

In (Ga,Mn)As based MTJs, different coercivities of the two (Ga,Mn)As layers are usually obtained by controlling the Mn concentration [95, 96], using p-type modulation doping [62] or post-growth annealing processes [202]. However, the coercivities of the two (Ga,Mn)As layers are often close and the interlayer interactions between them has been found [204] to couple the switching of the two layers [75].

For device application, it is desired to have one ferromagnetic (FM) layer pinned or magnetically hardened while keeping the other FM layer easy to switch. This is often achieved by using the exchange coupling from an antiferromagnetic (AFM) layer in close contact with the FM layer [205], similar to exchange-biased spin-valve devices. As mentioned in the last chapter, the widely reported AFM material for (Ga,Mn)As is MnO, whose insulating nature makes it unsuitable for MTJ devices, because in MTJs, the current flows perpendicular to the layers.

In this chapter, we report the TMR in (Ga,Mn)As MTJs exchange biased by MnAs layers. The top (Ga,Mn)As layer is pinned, due to the strong ferromagnetic coupling from the magnetically harder MnAs layer in proximity [206], and an “exchange spring” configuration is formed in the soft (Ga,Mn)As layer. In such exchange spring bilayers, the magnetization of the softer ferromagnetic layer is twisted about an axis normal to the layer plane, akin to a partial domain wall [188]. We show that the spin-dependent tunneling in our exchange-spring MTJ devices is very sensitive to the detailed magnetic configuration, yielding a direct picture of the winding and unwinding of the exchange spring.

6.1.1 Introduction to TMR

A magnetic tunnel junction (MTJ), which consists of two ferromagnetic (FM) layers separated by a thin insulating layer as a tunneling barrier, exhibits tunneling magnetoresistance (TMR), due to spin-dependent electron tunneling. MTJ is of great technological importance because of its big MR ratio and possible applications in magnetic sensor devices, such as read heads or magnetoresistive random access memory (MRAM).

Since the discovery of room temperature TMR using 3d ferromagnetic metal electrodes and amorphous aluminum oxide tunnel barriers, extensive efforts have been de-
voted to increasing the TMR ratio. Up to 70% of TMR has been achieved in Al-O based MTJs. To meet the continuous demands for higher MR for spintronics devices, new tunneling barriers, such as MgO, have received great attention, and a MR ratio up to 500% has been demonstrated [207].

An illustrative view of TMR is shown in Fig. 6.1. The resistance of a MTJ depends on the relative magnetic alignment of the two ferromagnetic layers. The tunneling resistance is lower when the magnetizations are parallel than when the magnetizations are anti-parallel. The relative change of resistance as compared to the lower resistance is called the tunneling magnetoresistance ratio (TMR ratio): \( \frac{R_{AP} - R_P}{R_P} \), which is the quantitative measure of the TMR effect.

![Figure 6.1. Schematic illustration of the TMR effect in a MTJ with a constant voltage bias.](image)

(a) The tunneling current is large (small) when the two FM layers are parallel (anti-parallel).

(b) Density of states of the two electrodes and the tunneling process for up and down spins.

An intuitive picture of TMR is provided by the famous Julliere model [120], which is based on two assumptions [170]. First, it assumes that the spin of electrons is conserved in the tunneling process. Similar to the two channel model for the GMR spin valve structure stated in Chapter 5, the tunneling processes for up and down spin electrons are considered to be independent. In this assumption, electrons originating from one spin state of the first FM electrode will only tunnel to the unfilled states of the same
spin of the second electrode. If the two FM electrodes are in parallel (P) configuration, then the majority spins tunnel to the majority states and the minority spins tunnel to the minority states, giving a large tunneling current. However, in an anti-parallel (AP) configuration, the majority spins in the first electrode are actually the minority spins in the second electrode (and vice versa), so the tunneling probability is drastically reduced, due to the mismatch of the density of states (DOS) for both up and down spins (Fig.6.1). The second assumption of Julliere’s model is that the conductance for a particular spin orientation is proportional to the product of the effective DOS of the two FM electrodes. Based on these two assumptions, the conductance for the P and AP states can be written as:

\[
G_P \propto D_1^\uparrow D_2^\downarrow + D_1^\downarrow D_2^\uparrow
\]

\[
G_{AP} \propto D_1^\uparrow D_2^\downarrow + D_1^\downarrow D_2^\uparrow
\]

where \( D_i^\uparrow \) and \( D_i^\downarrow \) are the tunneling DOS of the two FM electrodes \( (i=1,2) \) for the majority and minority spins respectively (note the arrows here do not represent up-spin or down-spin, but rather majority and minority). Then the TMR is obtained using Eq(6.2)

\[
TMR = \frac{(R_{AP} - R_P)}{R_P} = \frac{(G_P - G_{AP})}{G_{AP}} = \frac{2P_1P_2}{1-P_1P_2},
\]

\[
P_i = \frac{D_i^\uparrow - D_i^\downarrow}{D_i^\uparrow + D_i^\downarrow} \quad (i = 1, 2)
\]

where \( P_i \) is the effective spin polarization of the two FM electrodes.

It should be noticed that the validity of the Julliere’s model is still under debate, and it has been suggested that for (Ga,Mn)As based MTJs, where strong spin-orbit interactions occur, the TMR may also depends on the barrier thickness and the angle between the current and the magnetization \cite{208}. Nevertheless, this simple model still provides a quantitative picture of the TMR that can be easily correlated with the experiments.

6.2 Sample preparation and device fabrication

The MTJ devices studied here are fabricated from heterostructure samples that are grown by low temperature molecular beam epitaxy on \( p \)-type (001) GaAs substrates (Fig.6.2). We first deposit a 150 nm thick Be-doped p-GaAs buffer layer (with \( p \sim 1 \times 10^{19} \text{cm}^{-3} \)). This is followed by a heterostructure comprised of two nominally iden-
tical (Ga,Mn)As layers (with 30 nm thickness and a Mn content of ∼6%) separated by a tunnel barrier. In sample A (071201A), the tunnel barrier consists of 1 nm GaAs/4 nm AlAs / 1 nm GaAs spacer; the GaAs spacers prevent diffusion of Mn into the AlAs barrier. In samples B (071201B) and C (071201C), the tunnel barriers consist of 4 nm GaAs and 8 nm GaAs, respectively. The top (Ga,Mn)As layer has an epitaxial overlayer of type-A MnAs (with thickness 10±2 nm). The latter is a semimetallic ferromagnet that provides an exchange bias for the top (Ga,Mn)As layer [206], making it harder to switch, while the bottom (Ga,Mn)As layer is free to rotate in a small magnetic field. The function of the ferromagnetic MnAs layer is analogous to that of the antiferromagnetic layer commonly used in metallic MTJ devices as a pinning layer [205].

![Figure 6.2. Schematic illustration of the biased-MTJ structure.](image)

To protect the top MnAs during the processing, Ti(5 nm)/Au(30 nm) were first evaporated onto fresh sample pieces to serve as a protecting layer as well as an ohmic contact. Then 50 um or 100 um diameter round-shape mesas were defined by photolithography followed by chemical removal of the uncovered Ti/Au and MnAs layer. A chlorine-based reactive ion etching (RIE) was used to etch (Ga,Mn)As to form mesas with heights around 600 nm. A pseudo-four-probe scheme is used by wiring separate voltage and current leads on the top of the mesas for top contacts and on back of the substrate to form bottom contacts. The magnetic properties of the samples are measured using a Quantum Design superconducting quantum interference device (SQUID) magnetometer. TMR is measured by applying constant bias voltages and measuring the tunneling current using a Keithley 2410 source meter. In the magnetization and magnetotransport measure-
ment, the external field is applied along the [110] direction of the GaAs substrate. This direction corresponds to the [1120] direction where the easy axis lies in type-A MnAs.

### 6.3 Magnetometry measurement

![Graphs showing magnetometry measurement](image)

**Figure 6.3.** (a) Temperature-dependent magnetization $M(T)$ for sample C. (b) Major magnetization loop $M(H)$ for sample C, showing distinct switching fields for the bottom (Ga,Mn)As layer, the top (Ga,Mn)As layer and the MnAs layer ($\sim 1.8$ kOe). (c) Minor magnetization loops for the same sample, measured after saturating the MnAs layer in a field of $-20$ kOe. Solid triangles show the minor loop when only the bottom (Ga,Mn)As is switched. Hollow circles show the minor loop when both (Ga,Mn)As layers are switched. Note the hysteresis loop of top layer is shifted while the that of the bottom layer is intact. (d) $M(T)$ and $M(H)$ (inset) of the same sample after chemical removal of the MnAs layer. All the hysteresis loops are measured at 4.2K.

Fig. 6.3 shows the SQUID data for sample C. The other two samples show similar behavior. The temperature-dependent remanent magnetization $M(T)$ (Fig 6.3 a)) show that the two (Ga,Mn)As layers have different $T_C$, although they have nominally identical
compositions. The \(~20\)K difference in \(T_C\) can be readily seen on \(M(T)\) curve when the top MnAs layer is chemically removed (Fig.6.3(d)). We found that during the growth of MnAs, the diffusion of Mn interstitials in the \((\text{Ga,Mn})\text{As}\) layer is promoted so that the \(T_C\) is enhanced due to less compensation of the holes from Mn interstitials. This enhancement of \(T_C\) is found in all our MnAs/(Ga,Mn)As bilayer samples and MnAs/p-GaAs/(Ga,Mn)As trilayer samples with thin doped spacer layers. The \(T_C\) of the bottom \((\text{Ga,Mn})\text{As}\) layer is 65K and the \(T_C\) of the top \((\text{Ga,Mn})\text{As}\) is 85K. Fig.6.3(b) shows the major magnetization curve \(M(H)\) of sample C, revealing different switching behaviors for the top and bottom \((\text{Ga,Mn})\text{As}\) layers: the bottom layer experiences a sharp transition while the magnetization of the top one gradually changes in external field due to the exchange coupling with the MnAs overlayer.

From the minor loops of the MTJ shown in Fig.6.3(c), we can clearly identify the distinct switching behaviors for the two \((\text{Ga,Mn})\text{As}\) layers. We obtain these minor loops by sweeping the magnetic field in a small field range to switch either the bottom \((\text{Ga,Mn})\text{As}\) layer (-300 Oe \(\leq H \leq 300\) Oe) or both layers (-1kOe \(\leq H \leq 1\)kOe), after first saturating the magnetization of MnAs layer in a -20 kOe field. The bottom \((\text{Ga,Mn})\text{As}\) layer shows a small coercivity with a square hysteresis loop that is symmetric around zero field. In contrast, the top \((\text{Ga,Mn})\text{As}\) layer exhibits a large displacement \((\sim 600\) Oe\) of the hysteresis loop due to the ferromagnetic exchange coupling to the MnAs layer [206]. As a control experiment, we chemically etched away the MnAs layer on the same sample and measured \(M(T)\) and \(M(H)\) (Fig.6.3(d)). The distinct \(T_C\) are unambiguously revealed and the coercivity of the top \((\text{Ga,Mn})\text{As}\) layer is drastically reduced to \(~50\)Oe with a much sharper transition. Note that in both Fig.6.3(c) and the inset of Fig.6.3(d), the magnetization per unit area for each \((\text{Ga,Mn})\text{As}\) layer is \(~1 \times 10^{-6}\)emu/mm\(^2\), except for an offset in Fig.1(c) due to the magnetization of MnAs layer.

### 6.4 Bias voltage dependence of TMR

The three MTJ samples show distinct I-V characteristics (Fig.6.4), due to different barrier heights and widths. Devices derived from sample A has the largest differential resistance (several k\(\Omega\) for a 100\(\mu\)m diameter mesa) and a strong non-linearity, as anticipated for an AlAs barrier with a height of \(~550\) meV in the valence band [95]. In contrast, since the height of the GaAs barrier is small \((\sim 100\)meV\)) [96], devices made from sample B (4nm wide barrier) display almost linear I-V characteristics (Fig.6.4(b)), while those from sample C (8nm wide barrier) show non-linear I-V behaviors of a good
tunnel barrier (Fig. 6.4(c)). The dependence of the differential resistance of sample C on the bias voltage at different temperatures is shown in Fig. 6.4(c), indicating a strong tunneling behavior at low temperatures. In the rest of the chapter, we focus on the TMR in two devices fabricated from sample A (100µm diameter) and sample C (50µm) where we have taken an extensive set of data. Measurements on other devices (of varying diameter) fabricated from these samples show qualitatively similar results.

![Graphs showing I-V and dV/dI characteristics for samples A, B, and C.](image)

Figure 6.4. (a) Bias voltage dependence of the current (left) and differential resistance (right) for sample A (100µm). (b) Linear I-V characteristic for sample B (100µm) and its linear fit (solid curve). (c) Bias voltage dependence of the current (left) and differential resistance (right) for sample C (50µm). (d) Differential resistance of sample C as a function of bias voltage at different temperatures.

Fig. 6.5 shows the bias voltage dependence of the major loop TMR for the two samples. The TMR ratio shows a monotonic decay with increasing bias and is symmetric in bias voltage polarity. The voltage where the normalized TMR ratio becomes 0.5, \( V_{\text{half}} \), is \( \sim 30\text{mV} \), which is consistent with what have been reported in (Ga,Mn)As MTJs with GaAs and AlAs barriers [96, 95]. In (Ga,Mn)As based MTJs, \( V_{\text{half}} \) is much smaller than that of the metal-based MTJs (300-400 mV) [209]. This smaller \( V_{\text{half}} \) is believed to arise
Figure 6.5. Applied bias voltage dependence of major loop TMR for sample A (a) and sample C (b).

from the high carrier spin polarization of (Ga,Mn)As and rapid increase of the empty DOS for both spin-up and spin-down bands at the Fermi energy under even a small bias voltage [96]. Thus, a considerable current for both spin directions flows through the barrier even in the anti-parallel magnetization configuration under a small bias, and the difference in the spin-dependent tunneling for parallel and anti-parallel configurations diminishes drastically, resulting in the small $V_{half}$.

6.5 TMR measurement

Figs 6.6(a)(b) show the major loop TMR in samples A and C as a function of in-plane magnetic field at temperatures ranging from 4.2 K to 20 K. The magnetic field is applied along [110], parallel to the easy axis of the MnAs layer. The data shown here are all taken at a constant 4 mV dc bias. Compared with the magnetometry data, the TMR data show three distinct jumps in resistance, each corresponding to the independent switching of the three ferromagnetic layers. The sharp increase in resistance at low field corresponds to the switching of the lower (Ga,Mn)As layer which creates an angle with respect to the upper (Ga,Mn)As layer. The second jump in resistance occurs when the magnetization of the upper (Ga,Mn)As layer switches direction and aligns with that of the lower (Ga,Mn)As layer, creating a low resistance state. As we will discuss in more detail later, this process is more complex, resulting in a gradual decrease in TMR as the
two layers come into alignment.

![Graphs showing TMR ratio vs. H (Oe) and temperature]

**Figure 6.6.** Major loop TMR at different temperatures in (a) a 100µm diameter mesa device patterned from sample A and (b) a 50µm device patterned from sample C. Temperature dependent major loop TMR ratio for sample A (c) and sample C (d).

For sample A, we find a peak TMR ratio as high as ~11% at 4.2 K, comparable to earlier reports in (Ga,Mn)As-based MTJs with thinner AlAs barriers [95]. There, the TMR ratio of 10% was obtained on MTJ with ~2nm AlAs spacer. In our case, because the growth of MnAs usually takes longer time, Mn atoms could possibly diffuse into
AlAs barrier even with 1nm GaAs spacer on both sides of the AlAs, cutting the effective barrier thickness. Sample C shows a much larger peak TMR (∼ 45%) at 4.2 K, consistent with more coherent tunneling through the GaAs barrier [96]. For sample C, the effective barrier thickness could also be less than 8nm due to the diffusion of Mn atoms during the growth. This reduced effective barrier thickness is also suggested by the fact that in the 4nm GaAs spacer sample (sample B), the tunneling barrier is so small that the I-V shows linear characteristic and the measured TMR is only about 0.6%.

The third jump in resistance (indicated by arrows in Fig.6.6(a)(b)) occurs when the MnAs layer finally switches (at ∼ 1800 Oe), aligning its magnetization with that of the (Ga,Mn)As layers. This additional lowering of the overall device resistance is accompanied by the closing of the major hysteresis loop. We observe this small decrease in resistance in devices derived from all three samples and attribute it to a spin valve effect due to spin dependent scattering at the MnAs/(Ga,Mn)As interface [206]. We note that once all the three ferromagnetic layers are aligned, the low resistance state is stable, and the TMR follows an irreversible path as the magnetic field is swept down.

For sample A, the peak TMR decreases as expected with increasing temperature (Fig.6.6(c)), due to the decreasing spin polarization of the (Ga,Mn)As layers. On the other hand, the peak TMR for Sample C shows a maximum value around 10K, accompanied with the shape change of the TMR plateau. This is due to the complex magnetization switching behavior stemmed from the cubic anisotropy of (Ga,Mn)As. The increase of the temperature will drastically change the relative strength of the two anisotropy terms – biaxial (cubic) anisotropy and uniaxial anisotropy – and in turn affects the TMR. Details about how the anisotropy is related to the TMR will be discussed in the following section.

We now address the minor loop TMR shown as solid lines in Fig.6.7 These sweeps are obtained by following consistent protocol: we first saturate the magnetization of all three magnetic layers in a field of −3 kOe; we then sweep to a starting field +H₀ and then sweep the field down in magnitude. The minor loop TMR has a complex behavior whose exact form is sensitive to the value of H₀. When the minor loop is initiated at values of H₀ just above the coercive field of the lower (Ga,Mn)As layer, the TMR shows a standard irreversible minor hysteresis loop, with the high resistance state remaining stable as the field is swept down. However, if H₀ is in a range within which the top (Ga,Mn)As layer has begun to switch direction (indicated within the dashed ellipses in Fig.6.7), the minor loop TMR is “quasi-reversible.” In both samples, we observed a gradual shrinking of the minor hysteresis loops when H₀ is in that range.
Figure 6.7. Major loop TMR (circles for sweep up and triangles for sweep down) and minor loop TMR (lines) at 4.2 K for devices fabricated from (a) sample A and (b) sample C. The arrows indicate the direction of minor loops starting from different fields. The insets show the minor loop TMR ratio as a function of starting field at different temperatures for the two samples. Groups of blue and red arrows illustrate the magnetization configuration of the layers in different field regions.
In sample A, the TMR retraces a path whose shape resembles that observed while the field is swept up, but with a field-offset whose value depends on $H_0$. In sample C, both the shape of the TMR and its amplitude are sensitive to $H_0$; most surprisingly, the peak value of the TMR increases dramatically during the down sweep, almost doubling in value compared with the peak TMR obtained during the sweep up. The behavior in both samples suggests that the magnetization of the upper (Ga,Mn)As layer does not remain in a stable state while the magnetic field is swept down.

![Figure 6.8. Typical TMRs for a hard-soft MTJ (a) and an exchange-biased MTJ (b). The red arrows indicate the parallel (P) and anti-parallel (AP) magnetization configurations of the two layers. From Ref. [170]](image)

To compare the TMR of our devices with that of traditional metallic MTJs, we show the typical TMRs for two types of metallic MTJs in Fig. 6.8: (a) a hard-soft MTJ and (b) an exchange-biased MTJ. In a hard-soft MTJ, two ferromagnetic layers have different coercive fields, so the AP alignment is reached when the field is between the coercivities of the soft and hard layers. In the exchange-biased MTJ, an AFM layer is put in direct contact with one of the FM layers and shifts the magnetization loop of the FM layer away from zero field. Technologically, an exchange-biased MTJ is advantageous because the resistance change takes place near zero field, and the AP state is more stable in this case. However, in metallic exchange-biased MTJs, the biasing of the FM layer is unidirectional, in the sense that the exchange biasing on the pinned FM layer is always pointing to one direction and cannot be altered in an external field [205]. In our devices, where a FM layer is used to achieve exchange biasing, the biasing of the top (Ga,Mn)As
layer can be bi-directional, depending on the magnetization of the MnAs layer. We see in our samples that the TMR minor loops just before the MnAs switches are very much like those in metallic exchange-biased MTJs. However, once the MnAs switches (major loop), the TMR is turned off in the down-sweep because now the top (Ga,Mn)As layer is pinned to the opposite direction and the TMR will only be turned on again when the field reverses (similar to TMR in a hard-soft MTJ). Thus, the TMR behaviors of an all FM exchange-biased MTJ resemble the combination of both the hard-soft MTJ and the exchange-biased MTJ. This feature can be used in achieving control function in MTJ-based devices, such as MRAM.

6.6 Probing the exchange spring configuration

We now develop a qualitative argument that links the minor loop TMR in these devices to the unwinding of an exchange spring in the upper (Ga,Mn)As layer. We begin with a picture of the magnetization in the upper (Ga,Mn)As layer. When the magnetic field is swept above a characteristic coercive field, we speculate that the exchange coupling of this layer with the MnAs layer results in an unusual exchange spring configuration that consists of a partial domain wall of width $d_1(H)$ in the vertical direction and a complete domain of width $d_2(H)$ produced by the anisotropy of the (Ga,Mn)As layer, similar to what we proposed in Chapter 5 (Fig. 6.9). It is reasonable to assume that both these domain widths ($d_1$ and $d_2$) are field-dependent as a result of minimizing total energy. The width of the complete domain $d_2$ increases with magnetic field while the width of the partial domain wall $d_1$ diminishes, accompanied with the enhanced twisting of the spins near the interface.

Unlike complete exchange springs that exhibit fully reversible behavior with a disappearance of hysteresis [188], the system studied here can follow a more complex quasi-reversible return path: when the magnetic field is swept down, the complete domain state can remain with a fixed width $d_2$ and rotate as a single domain, while the exchange spring unwinds. This model explains the fact that the shape of the minor hysteresis loops are between reversible and irreversible. As discussed earlier, as long as the MnAs does not reverse ($H < 1800\text{Oe}$ for sweeping up), most part of the top (Ga,Mn)As magnetization can always recover back in alignment with the MnAs layer. This observation further supports the existence of spring-like magnetization within the top (Ga,Mn)As layer due to exchange coupling to the MnAs. A similar observation of the depth-dependent magnetization in FM layer has been made in metallic AFM/FM systems using magnetometry.
Figure 6.9. (a-c) Illustrations of the magnetization switching in three different field ranges. The red (green) arrows represent the top (bottom) (Ga,Mn)As layer respectively. The twisted spins in the top (Ga,Mn)As layer are also depicted, forming a spring-like PDW with a variable depth. (d-f) Illustration of magnetization switching when the field ramps down from the initial states in (a-c) and then reverses.
and magneto-optical Kerr effect (MOKE) [210]. We demonstrated the spring-like spin structure in the FM layer can also be probed by measuring TMR in such a structure, where one of the FM layers participating the spin-dependent tunneling is biased by a metallic FM layer.

The modified Julliere's model to describe TMR where two FM electrodes are aligned in an angle is often written as: [120]

\[
TMR = \frac{P_1 P_2 (1 - \cos \Delta \theta)}{1 + P_1 P_2 \cos \Delta \theta}
\]

(6.5)

where \( P_1, P_2 \) is the spin polarization of the two FM layers, and \( \Delta \theta \) is the relative angle between the two magnetizations. In our case, the polarization of the top (Ga,Mn)As layer is more complicated due to the formation of the PDW. The effective polarization depends on the resultant magnetization from the the twisted PDW \( d_1 \) and the uncoupled single domain \( d_2 \).

When combined with the cubic and uniaxial magnetic anisotropy of (Ga,Mn)-As, as well as the TAMR effect [201], this exchange-spring configuration results in minor loop TMR that can differ greatly in magnitude and shape from the major loop TMR. This is best illustrated by the dramatic dependence of the minor loop peak TMR ratio on \( H_0 \) in sample C at the lowest temperatures (inset of Fig.6.7(a)).

![Figure 6.10.](image)

**Figure 6.10.** (Major loop (circles) and minor loops (lines) of sample C starting from different representing field regions (a,b,c) as shown in Fig.6.9)

In Figs.6.9(a)-(f), we illustrate the switching of magnetization for three different values of \( H_0 \); the respective TMR for these three minor loops is shown in Fig.6.10. The external field is applied along [110] with a small misalignment angle. Since the direction
of the MnAs magnetization is fixed after saturating at \(-3\) kOe, the angles of the magnetization of (Ga,Mn)As layers are described with respect to the MnAs magnetization. When the field is swept up after reversal, the first abrupt jump in TMR corresponds to the nucleation and propagation of a \(\sim 90^\circ\) domain wall within the bottom (Ga,Mn)As layer. This leads to an abrupt switching of its magnetization from \(\sim 45^\circ\) to \(\sim 135^\circ\), as indicated by the green arrows in Fig. 6.9. The magnetization of the top layer also rotates inhomogeneously through a small angle, forming a relatively wide partial domain wall.

When a minor loop starts from this configuration (sweeping down), the \(M\) of the bottom layer simply switches back to \(\sim 45^\circ\) (Fig. 6.9(d)), giving a square hysteresis loop (green curve in Fig. 6.10). In this regime, the TMR magnitude does not vary much with the reversible unwinding of the slightly twisted exchange spring in the upper layer.

For larger values of \(H_0\) that lie in the major loop TMR plateau (200 Oe \(\lesssim H_0 \lesssim 700\) Oe), the magnetization of the bottom layer gradually rotates to the second energy minimum aligning itself at an angle of \(\sim 225^\circ\) (Fig. 6.9(b)). The presence of a uniaxial anisotropy in (Ga,Mn)As (in addition to the cubic anisotropy) causes this second 90\(^\circ\) switching through coherent rotation, rather than domain wall nucleation. This is accompanied by an additional twisting of the exchange spring in the upper layer. We speculate that the complex plateau in the major loop over that field range is a result of the interplay of both top and bottom (Ga,Mn)As layers rotating in the same direction. When the minor loop is initiated with \(H_0\) in this regime, the magnetization of the lower layer is pinned in its energy minimum at \(\sim 225^\circ\) until the field reverses. In the upper layer, however, the complete domain rotates coherently to an energy minimum at \(\sim 45^\circ\) (Fig. 6.9(e)). Because of the TAMR effect and the large relative angle between the two magnetizations, it yields a larger peak TMR (yellow curve in Fig. 6.10) than that seen during the major loop. Further, the width \(d_2(\bar{H})\) of the complete domain – and correspondingly the magnetization along [\(\bar{1}00\)] – increases with \(H_0\). Using the heuristic Julliere model (Eq. 6.5), this increase in magnetization (and thus spin polarization) explains the increase in the TMR peak value with increasing \(H_0\) (inset in Fig. 6.7(b)).

The increase in peak TMR with \(H_0\) is however not monotonic and reaches a maximum value for \(H_0 \sim 700\) Oe, beyond which we observe a slight decrease in the peak TMR (inset in Fig. 6.7(b)), which is attributed to existence of small domain pinning sites. We speculate that even though the width \(d_2\) of the complete domain increases with \(H_0\), only a portion of this complete domain can rotate coherently, limited to a critical thickness \(d_c\), as illustrated in Fig. 6.9(f). The peak TMR is maximum when \(d_2 = d_c\) (blue curve in Fig. 6.10).
Our qualitative model is consistent with the observed temperature dependence of the minor loop TMR (see inset to Fig. 6.7(b)). With increasing temperature, the peak value of the minor loop TMR of sample C becomes less dependent on the starting field, similar to the behavior observed at all temperatures in sample A (inset to Fig. 6.7(a)). We note that – at these higher temperatures – the overall character of the minor loop TMR in sample C still remains quasi-reversible (data not shown) and resembles that obtained for sample A at all temperatures. We attribute these observations to a rapid decrease in the cubic anisotropy with increasing temperature relative to the change in the uniaxial anisotropy [57]: this reduces TAMR contributions by removing the lack of symmetry between the two easy axes.

We also notice in the control sample where MnAs is etched away, this quasi-reversible behavior disappears, showing a minor TMR similar to that reported by Higo et al. [199]. A puzzling aspect of our data is the contrast in minor loop TMR between samples A and C at the lowest temperature, particularly given that the only structural difference between the two samples is the tunnel barrier. We do not currently have a convincing explanation for this difference, but note that the magnetic anisotropy of (Ga,Mn)As is a sensitive function of hole density. Studies of epitaxially grown heterostructures containing (Ga,Mn)As have shown a large variability in hole-compensating Mn interstitial concentrations that depends on the details of the heterostructure. It is thus possible that the lack of TAMR in sample A may arise from a lack of cubic anisotropy in the lower (Ga,Mn)As, resulting from Fermi-level driven differences in Mn interstitial diffusion during the overgrowth of AlAs. It is also possible that contributions of TAMR are sensitive to the nature of the tunnel barrier, although calculations of TAMR for GaAs and AlAs tunnel barriers do not suggest an obvious explanation [211].

6.7 Summary and conclusions

In summary, we have demonstrated all ferromagnetic exchange-biased MTJs which show bi-directional exchange-biasing using MnAs as a pinning layer. The TMR observed in these devices is consistent with the formation of a novel exchange spring consisting of a partial domain wall and a complete domain, both of which have field-dependent widths. The large TMRs in the minor loops show evidence of TAMR due to the anisotropic density of states in (Ga,Mn)As. The devices developed in this work could provide interesting model systems for detailed studies of spin-dependent tunneling in non-uniform magnetization configurations and for developing proof-of-concept magnetic devices with
new functionalities.
Appendix A

Device patterning recipes

The first step of device fabrication is designing the patterns. Using computed-aided design (CAD) software (such as L-Edit), the patterns of the devices are created and exported as GDSII files. Then we can either use a laser writer to make a photolithography mask or directly write the patterns using an electron-beam writer. In the following sections, I list some recipes for fabricating devices used in this dissertation.

A.1 Processing recipe for making photolithography masks

We use a Heidelberg DLW66 laser writer to make a photolithography mask. The steps are listed as following:

1. First obtain a 4" × 4" blank mask, pre-coated with a layer of photoresist.

2. Expose the photoresist using the laser writer. Choose the appropriate write head size according to the minimum feature size. Make sure to use the right “expose mode” (“inverted” or “non-inverted”) depending on the CAD design. The ‘non-inverted” mode will expose all the features in the CAD, while the “inverted” mode will expose everything within the border except for the features drawn in the CAD.

3. Develop the photoresist in CD-26 for 1 minute, then stop developing in de-ionized (DI) water.

4. Etch the unprotected chromium (Cr) using a Cr-etchant for 90s. Rinse in water and dry with N₂ gas.

5. Finally stripe the photoresist in EKC830 for 5 minutes. Rinse in water and dry with N₂ gas.
A.2 Processing recipe for making Hall bars

We use different sizes of Hall bars in our experiments, from 5µm to 400µm in width. We use wet chemical etching to make these Hall bars.

1. Clean the sample in acetone and then isopropanol (IPA), if necessary. Dehydrate the sample on a hot plate for 2 minutes.

2. Spin on photoresist (Shipley 1813). Use the recipe – S20.50/45 – on the programmable spinner. (“S20.50/45” means static dispense, 20s accelerating time, 5000 rpm for 45s). This gives you a layer of photoresist with a thickness of ∼1.1µm.

3. Bake the sample at 110°C for 70s. Cool down on a metal plate.

4. Expose the sample using the MA6 aligner (7s exposure time with 365nm wavelength light at a power of 8mW).

5. Develop with CD-26 developer for 50s. Stop developing in DI water. Dry with N₂ gas.

6. Use III-V etchant (potassium dichromate: 2g; hydrobromic acid: 174.5ml; DI water: 500ml) to etch the sample. Typical etch rate is: ∼300nm/min. Rinse in the water. Dry with N₂ gas.

7. Inspect under the optical microscope.

A.3 Processing recipe for making mesas for CPP spin valve
(Fig. A.1)

1. Perform the 1st photolithography (step 1 to step 5, as stated above) to define the mesa on the photoresist.

2. Etch the sample using III-V etchant to a depth of \( d \sim 160\text{nm} \).

3. Evaporate SiO₂ using the Semicore E-beam evaporator. Make sure the thickness of the SiO₂ is larger than the depth of the mesa \( d_{\text{SiO}} \sim 330\text{nm} \).

4. Lift-off the SiO₂ on top of the mesa by removing the photoresist in acetone or other photoresist stripper such as 1615 PR remover. May use sonicator to facilitate this lift-off process.
5. Check the sample under the optical microscope and the profilometer.

6. Perform the 2nd photolithography to define a window on top of the mesa for metal contact evaporation.

7. Evaporate Ti(5nm)/Au(50-100nm) (Ti serves as an adhesion layer) using the Semi-core E-beam evaporator.

8. Lift-off the metals in acetone. The metals left on the top and the side of the mesa will serve as the top and the bottom contact, respectively.

### A.4 Processing recipe for making mesas for MTJ (Fig.A.2)

1. Evaporate Ti(5nm)/Au(30nm) (Ti serves as an adhesion layer) onto the fresh sample piece to protect the top MnAs during the processing.

2. Perform the 1st photolithography to define the mesa on the photoresist.

3. Use Au etch TFA (Iodine complex and potassium iodide) to remove the Au, Ti and MnAs layer. Au etch rate is $\sim 28\text{Å/s}$. After 10 $\sim 12$s, the Au layer is etched.
1. Evaporate Ti/Au contact layer.

2. Photolitho step 1: Define a mesa. Etch Ti / Au layer and MnAs using Au etchant.

3. Etch the sample using reactive ion etching.

4. Remove the photoresist.

5. Etch Ti / Au layer and MnAs using Au etchant.

6. Photoresist.

Figure A.2. An illustrative picture of the fabrication process for MTJ devices.

away. 1-1.5 minutes etching shows dirty stuff and cracks on the sample surface, suggesting that the Ti and MnAs layers are peeling off. After 2 minutes etching, the surface looks smooth again, indicating that Au, Ti and MnAs layers are all removed.

4. Using the photoresist on the sample as a mask, perform reactive ion etching (RIE). First run the process recipe on a dummy wafer for 3 minutes to prepare the chamber. Then load the sample, run RIE process using the following parameters: Cl₂ flow rate: 20 sccm; pressure: 10 mtorr; power: 150W; etching time: 2-3 minutes; etching rate: ∼300nm/min.

5. Remove the photoresist in acetone. Check the mesa height using the profilometer.

6. Perform the 2nd photolithography. Define a circular hole pattern with a size slightly smaller than the previously defined mesa. After developing, a window is opened up on top of the mesa, while other part of the sample is covered by the photoresist as an insulating layer.

7. The top indium contact can be put on top of the mesa through the window, and
the bottom contact is put on the back of the substrate.

A.5 Recipe for removing MnAs layer

MnAs layer can be removed in a diluted hydrochloric acid solution (HCl:H₂O = 2:1). 10-15 nm of MnAs is etched away in 1-1.5 minutes. You can monitor the color change of the sample to determine if the MnAs has been completely etched. MnAs has a silver-like shiny surface.
Appendix B

Procedures for operating $^3$He cryostat

I. Sample Mounting

1. Both the “perpendicular-field” and “parallel-field” sample stages can be mounted. Make sure the wire connections are right (The top pin of the stage corresponds to the 3rd pin on the insert, and the bottom pin of the stage is void).

2. Mount the sample on the sample stage. Make sure there is a good thermal contact between the $^3$He pot and the sample. Usually, a piece of cooper is used on the sample carrier to make a thermal link.

3. Arrange the LED to the right position so that it can illuminate the sample. Check electrical connection of the sample by running a I-V test. Test the LED if necessary. (*Pictures of the Heliox $^3$He cryostat insert and its components are shown in Fig. B.2*)

II. Pumping IVC (Inner Vacuum Can)

1. Connect the pump to the $^3$He insert via the IVC pumping line.

2. Wash your hands. Clean the IVC flange and the cone seal with wipers.

3. Apply Dow Corning vacuum grease evenly on the inner side of the IVC as well as the cone seal. Usually a thin layer of grease is enough.

4. Carefully mount the IVC to the cone seal and start pumping using the mechanical pump. Wipe out excessive grease on the IVC.
<table>
<thead>
<tr>
<th>Sensor number</th>
<th>Sensor position</th>
<th>Sensor type</th>
<th>Sensor range (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sensor 1</td>
<td>Sorb</td>
<td>270Ω Allen Bradley</td>
<td>4.0-250</td>
</tr>
<tr>
<td>Sensor 2</td>
<td>3He pot</td>
<td>470Ω Speer</td>
<td>0.25-10</td>
</tr>
<tr>
<td>Sensor 3</td>
<td>3He pot</td>
<td>1KΩ Carbon glass resistor</td>
<td>1.0-300</td>
</tr>
</tbody>
</table>

Table B.1. Three types of temperature sensors on the 3He cryostat insert

5. When the pressure < 10 torr, turn on the turbo pump (initially I $\sim$ 5.8A, when stable, I $\sim$ 0.4-0.5A, 1200rpm). Pump down to $10^{-5}$ Torr to test IVC vacuum (it usually takes $\sim$ 2 hours).

6. Secure the helium pickup tubes to the IVC exterior with a piece of dental floss.

III. Cooling down to 77K

1. Prepare a liquid nitrogen container and place it on a stool.

2. Turn off the pumps (first close the valve on the IVC pumping line, then turn off the turbo pump. When the light on the turbo pump goes off, turn off the mechanical pump). Remove the pumping line.

3. Put the cryostat insert into the liquid nitrogen. Make sure the IVC is completely soaked in the liquid nitrogen.

4. Fill the IVC with a small amount of helium gas as exchange gas to cool the insert down to 77K. (Connect the helium gas pipe to the IVC pumping line, first open the green valve to admit a small amount of helium gas ($\sim 10cm^3$) and then close it. Then open the IVC valve to let the helium exchange gas flow into the IVC).

5. Monitor the temperature controller (ITC-503). Temperature should drop to $\sim 77K$ within 30 minutes. A summary of temperature sensors on the 3He cryostat insert is given in Table B.1 (Sensor 3 is more accurate in this temperature range.)

6. Once the temperature is $\sim 77K$, flush helium gas through the 1K pot pumping line to make sure that they are not blocked. You should see bubbles rising in the liquid nitrogen when you open the 1K pot needle valve.

IV. Cooling down to 4.2K

1. Lift the insert and put it into the liquid helium dewar. Slightly rotate it to fit the dewar. (Note the pumping lines are on the left side). \textit{Note:} Do not force it when
you feel resistance! Lift it a little bit and rotate it gently to find the right position to fit.

2. The temperature should drop to 4.2K in 10 minutes.

3. Connect the pump to the IVC pumping line to pump out the exchange gas. The pressure in the IVC can be as low as $10^{-7}$ torr. Also connect a pump to the 1K pot pumping line.

V. Condensing $^3$He

1. A flow of $^4$He through the sorb heat exchanger should be set by pumping the sorb heat exchanger pumping line. The flow of $^4$He serves as a thermal isolation between the hot sorb and the cold 1K pot.

2. Raise the temperature of the sorb (Sensor 1) to 45K. Hold “Set” button and use “raise” button to set the temperature setpoint. (Make sure that the correct heater is used, and the “heater” is set to “Auto”; Also make sure correct P.I.D settings are chosen: P=10, I=2.0, D=0 for $T>10$K.) You should see the sorb temperature increases to 45K quickly.

3. Pump the 1K pot pumping line, open the valves and adjust the needle valve to control the $^4$He flow. (Initially the helium gas pressure is 15-20 mbar.)

4. Adjust the 1K pot needle valve to tune the $^4$He flow so that the temperature (sensor 2 reading) is lowered by pumping the liquid $^4$He in the 1K pot. This step requires adjusting the needle valve to balance the $^4$He admission and consumption in 1K pot (needle valve is sensitive!) If the 1K pot runs out of helium, open the needle valve to admit a burst of liquid helium.

5. Monitor the sensor 2 reading. When $T < 3$K, $^3$He begins to condense, as illustrated in Fig[B.1][a]. It takes $\sim 30$-45 minutes to reach the lowest 1K pot temperature of $\sim 1.4$K.

VI. Pumping $^3$He

1. Set the temperature of the sorb to 4K. (Make sure to change P.I.D to 2.5, 0.5, 0). You should see the temperature of the sorb decrease to 4K in 10-15 minutes.
2. The temperature reading on sensor 2 will decrease as the sorb is pumping the $^3$He (Fig. B.1(b)). Monitor sensor 2 until it reaches the base temperature of $\sim 360$ mK. Wait a certain amount of time to let it stabilized.

**VII. Adjusting temperature**

1. Below the 1K pot temperature (1.5K $\sim$ 1.6K), sensor 2 is set for feedback. The temperature is controlled by heating the sorb. The sorb heat exchanger has to be pumped. Set sensor 2 to the desired value, then set “heater” to “Auto”. Adjust P.I.D. if necessary. *Note:* Do not set sensor 2 setpoint higher than the 1K pot temperature. Otherwise, the sorb will continue to be heated – “thermal runaway”.

2. Above the 1K pot temperature, use sensor 3 for feedback. The system will heat the $^3$He pot directly. Set sensor 3 to the desired value, then set “heater” to “Auto”. Adjust P.I.D. accordingly (P: 5$\sim$10, I: 0.3$\sim$1, D: 0$\sim$0.1).

**VIII. Warming up**
1. Close the 1K pot needle valve and make sure there is no liquid helium left in the 1K pot. Close the sorb heat exchanger needle valve as well. Turn off all the pumps (turbo first, then the mechanic pump) and disconnect all the pumping lines.

2. Take out the insert from the helium dewar and put it on the rack. The insert is then warmed up in the air bath.

3. To avoid water condensation, do not open the IVC valve until sensor 3 reading is close to the room temperature. Watch out when $T > 270$K, the IVC may fall.

IX. Removing IVC

1. Make sure there is enough space below the insert to remove the IVC.

2. Vent the IVC by opening the IVC valve. Screw slide hammer on the bottom of the IVC. Hold the insert firmly and slide the moving part of the hammer to knock off the IVC.

3. Wipe off the grease on the IVC flange and the cone seal to prevent dust accumulation. Place the IVC in a safe place. Remove your sample.
Figure B.2. Pictures of the Heliox $^3$He cryostat insert.
Appendix C

Procedures for MBE chamber opening and baking (III-V)

C.1 Opening preparation

1. Make sure there is no liquid nitrogen left in the chamber shroud (wait at least 24 hours after the growth).

2. Lower all the cell temperatures to 50°C @ 1°C/min (StpLow = 50°C, stp1 & stp2 = 50°C). For the Al cell, first lower it down to 670°C @ 1°C/min (Al melting temp = 660°C); then manually lower the Al cell temperature from 670°C to 650°C @ 1°C/3min; then lower down to 50°C @ 1°C/min.

3. Checklist for chamber opening:
   - Nitrogen gas
   - Source materials for reloading; Tweezers for reloading
   - All the gaskets for the CF-flanges
   - Personal Protection Equipment (masks, gowns, gloves)

C.2 Chamber opening procedure

I. Pre-opening

1. Check all the cells at 50°C.
2. Disconnect the water lines (blue – “out”, gray – “in”) from the chiller. Connect the hoses (blue – “out” – up position, gray – “in” – down position) from the manipulator to the purge connectors on the manifold. Direct the blue drain hose to the sink, then toggle the purge valve to purge the water out of the manipulator.

3. Shut down all the cell power and temperature controllers, substrate temperature controller, substrate power supply, heated viewport power and all the ion gauges.

II. Venting the chamber

1. Purge the nitrogen gas line (red) and adjust the nitrogen flow. Connect one end of the line to the nitrogen cylinder, and the other end to the growth chamber venting port.

2. Open the gate valve between III-V buffer and growth chambers; close the valve between the growth chamber and “growth” ion pump; make sure the valve between II-VI and III-V buffers is closed.

3. Put “buffer” ion pump into “standby” (press “9” – high voltage operation, select “off”, then press “enter”). Note: There is a small leak between the growth chamber and the “growth” ion pump. So when venting the growth chamber, you will see the pressure of the “growth” ion pump go up. If the pressure is too high, you may need to put the “growth” ion pump into “standby”.

4. Put on protective gowns and masks.

5. Put the small metal block on the venting valve, use a wrench to open the valve and start venting the chamber.

III. Source material reloading

1. Remove the cell from the chamber and add new materials (perform reloading in the fume hood). Do not overfill. Clean the port flange and the gasket with methanol. Mount the cell back on the chamber, make a continuity check of the heater filament and the thermocouple before tightening the screws. Do the As cell last. Note: Do not use aluminum foil when reloading the arsenic.

2. Check the Al crucible for cracks if necessary.

IV. Pumping down
1. Connect the scroll pump to the III-V venting port and start pumping.

2. When the pressure is \( \sim 22 \) millitorr, close and tighten the metal block valve on III-V venting port.

3. Open the gate valve between the growth chamber and the “growth” ion pump slowly, keeping the pressure of the “growth” ion pump \(< 10^{-6} \) torr. At the same time, open the gate valve between III-V and II-VI buffer chamber slowly, keeping the pressure of II-VI buffer \(< 10^{-6} \) torr.

4. When both the gate valves are fully open, turn on the “buffer” ion pump.

5. Close the gate valve between the III-V growth and buffer chamber, close the gate valve between III-V and II-VI buffers. Let each chamber pump down individually.

6. Turn on both “growth” and “buffer” ion gauges. Turn on all the cells.

C.3 Baking procedures

A baking process is often required for the III-V system after the chambers have been exposed to the atmosphere for maintenance. The baking at \( \sim 180^\circ \)C will effectively remove the water vapor adsorbed on the chamber walls.

I. Pre-baking preparation

1. After opening the chambers, pump the system down to \( \sim 10^{-7} \) torr.

2. Remove all the un-bakable parts:
   - Liquid nitrogen lines.
   - RHEED gun high voltage cables. Put a metal cap on the connector pins.
   - RHEED camera box and cables; RHEED gun high voltage cables.
   - Cables for retractable flux ion gauge. See Fig.C.1(a) for pin diagram.
   - SRS residue gas analyzer (RGA).
   - Bandit pyrometry system (both the light source and the detector).
   - Substrate motor and manipulator magnetic coupler. Unscrew three screws to remove the motor and pull the coupler out.
   - Cooling water lines (blue and grey).
Figure C.1. (a) Diagram of the ion gauge pins. (b) Diagram of the air-hose quick coupling for the shutter actuators.

- The handle on the gate valve between the ion pump and the growth chamber and the handle on the retractable flux ion gauge module.
- Magnetic handle on the transfer arm.

3. Dissemble the cell shutter actuators:
   (a) Open the shutters
   (b) Put stop-pins on the shutter shafts
   (c) Shut off the compressed air
   (d) Disconnect the air-hose quick coupling from the frame (Fig. C.1(b))
   (e) For the Ga shutter, first turn off the shutter heater
   (f) Rotate the shutter actuators CCW and remove the actuators

4. Clean the chamber surface and the manifolds.

5. Set the substrate temperature \( T_S = 300^\circ C @ 5^\circ C/\text{min} \), and all the cell (except for As) temperatures to \( 380^\circ C @ 5^\circ C/\text{min} \). For As cell, set temperature to \( 225^\circ C @ 5^\circ C/\text{min} \). (The raising of temperatures may take multiple days depending on the chamber pressure.)

6. Setup the baking panels and insulating jackets. Connect the baking panel power cables.

II. Starting baking
1. Start raising the baking panel temperature $T_B$ from 50°C. Gradually increase $T_B$ to 180°C and monitor the chamber pressure at the same time (Do not let the chamber pressure exceed $10^{-6}$ torr. It may take several days to reach 180°C.

2. Once $T_B = 180°C$, bake at this temperature for one more week. Record chamber pressure and baking panel temperatures every day.

III. Finishing baking

1. Finish bake-out. Set $T_B$ and $T_S$ to 20°C; Keep $T_{Al,Ga,In}$ at 380°C; Set $T_{As,Mn,Si,Be}$ to 110°C.

2. Wait one day to let the panel cool down. Remove the baking panels. Put all the parts back on the chamber. Do an alignment check on the transfer arm at $T_S = 20°C$ and 200°C. Test the Bandit system and align the detector.

3. Set $T_{Ga}$ to 500°C @ 10°C/min. For Al cell, set $T_{Al}$ to 650°C @ 1°C/min, then manually raise the temperature from 650°C to 670°C @ 1°C/2min, above 670°C, set $T_{Al}$ to 780°C @ 1°C/min.
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Vita

Meng Zhu

Education

• Ph.D. in Physics, The Pennsylvania State University, University Park, PA, 2008
• B.S. in Physics, Nanjing University, Nanjing, China, 2002

Honors and Awards

• 2003, 2005-2007, David C. Duncan Graduate Fellowship in Physics
• 2006, Honorable mention, Penn State International Programs Photo Contest

Publications

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