THE STUDY OF FERRO AND ANTIFERROMAGNETISM IN
DILUTED MAGNETIC SEMICONDUCTOR GaMnAs

A thesis submitted to the School of Graduate Studies
Addis Ababa University

In partial Fulfillment of the Requirements for the
Degree of Master of Science in Physics

By
Yared Solomon

Addis Ababa, Ethiopia

June 2010
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ADDIS ABABA UNIVERSITY

FACULTY OF SCIENCE
DEPARTMENT OF PHYSICS

The undersigned hereby certify that they have read and recommended to the Faculty of Science School of Graduate Studies for acceptance a thesis entitled

“THE STUDY OF FERROMAGNETISM IN DILUTED MAGNETIC SEMICONDUCTOR GaMnAs” by Yared Solomon in partial fulfillment of the requirements for the degree of Master of Science in Physics.

Name          Signature

Advisor       Prof. Singh.p

Examiner      Dr.Tesgera Beddassa

Examiner      Prof.Mal’nev
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Acknowledgements

From the very beginning, I would like to thank the Almighty God who brought me in to this picture. Wishing him a God blessing, I would like to truly express my deepest gratefulness to my advisor and instructor prof. P.Singh for his non-stopping guidance to a blend of physics, assistant and follow up, during carrying out the course and thesis work. I really admire him for his friendly approach with immeasurable and valuable contribution throughout the whole period of the research work. It gives me pleasure to acknowledge ministry of education for the sustainable financial hold up I received during my study.

My parents should take their share of my boundless gratification. I have special heart for my mom who really feels my busyness and provides me all my requirements. Eventually, thanks be to my friends for all kinds of encouragement and support I get from them.
Abstract

The prospect of a new generation of electronic devices based on the fundamental quantum property of angular momentum, known as spin, has led to the rapidly developing field of spintronics. These materials are created by using molecular beam epitaxy (MBE) to incorporate into traditional semiconductors a quantity of transition metal atoms sufficient that ferromagnetism is exhibited.

We present first principles calculations for prospective magnetic materials for future applications in spintronics. The material studied is focused on dilute magnetic semiconductors (DMS) like $Ga_{1-x}Mn_xAs$ that play a key role in semiconductor spintronics. Due to their ferromagnetic properties they can be used in magnetic sensors and as spin injectors. The basic problem for applications are, however, the relatively low Curie temperatures of these systems. We therefore focus on understanding the magnetic properties and on a reliable calculation of Curie temperatures from first principles. We have developed a theoretical framework for calculating critical temperatures by combining first principles calculations and in terms of the Ruderman–Kittel–Kasuya–Yosida quantum spin model in Green’s function approach. Random distributions of the magnetic atoms are treated by using an analytical average of magnetic configuration are performed using green function formalism using the dispersion relation for $k^2$ for antiferromagnetic case and $k$ for ferromagnetic case. Temperature dependencies of the spin wave specific heat, inverse magnetic susceptibility and reduced magnetization are determined. The result shows $T^{-3/2}$ dependence of for magnon specific heat in ferromagnetic case and $T^3$ in antiferromagnetic, the dependence of the Neel temperature on the manganese ion concentration is linear thus for our calculation the highest Neel temperature obtained $T=36.5k$ with in the concentration of 0.05, the reciprocal susceptibility increases linearly with increasing temperature above 173k,
INTRODUCTION

In recent years, considerable work has been devoted to the study of diluted magnetic semiconductors. These are semiconductors that are doped with ions that have a net spin. The interaction among these spins leads to a ferromagnetic state at low temperatures. These type of materials have a lattice structure similar to that of the undoped semiconductor, at least for the diluted magnetic case. This similarity provides a good lattice match between the doped and undoped cases, allowing for the potential preparation of devices. The field of DMS received a considerable boost when a few years ago Ohno doped Mn ions into GaAs, and found Curie temperature above 100K. This is not yet sufficient for applications, but progress in this area is rapid and room temperature ferromagnetism could be achieved even with considerable dilution[1].

The mass, charge, and spin of electrons in the solid state lay the foundation of the information technology we use today. Integrated circuits and high-frequency devices made of semiconductors, used for information processing and communications, have had great success using the charge of electrons in semiconductors. Mass storage of information indispensable for information technology is carried out by magnetic recording (hard disks, magnetic tapes, magneto-optical disks) using spin of electrons in ferromagnetic materials. It is then quite natural to ask if both the charge and spin of electrons can be used to further enhance the performance of devices. We may then be able to use the capability of mass storage and processing of information at the same time. Alternatively, we may be able to inject spin- polarized current in to semiconductors to control the spin state of carriers, which may allow us to carry out (quantum bit) operations required for quantum computing. However, there are good reasons why this has not yet been realized.

The semiconductors used for devices and integrated circuits, such as silicon (Si) and gallium arsenide (Ga As), do not contain magnetic ions and are nonmagnetic, and their magnetic Lande-g factors are generally rather small. In order for there to be a useful difference in energy between the two possible electron spin orientations, the magnetic fields that would have to be applied are too high for everyday use. Moreover, the crystal structures of magnetic materials are usually quite different from that of the semiconductors used in electronics, which makes both materials incompatible with each other [2].
CHAPTER 1

1. GENERAL INTRODUCTION TO SEMICONDUCTOR

1.1 Semiconductors Theory

Starting with the development of the transistor by Bardeen, Brattain, and Shockley in 1947, the technology of semiconductors has exploded. With the creation of integrated circuits and chips, semiconductor devices have penetrated into large parts of our lives. The modern desktop or laptop computer would be unthinkable without microelectronic semiconductor devices, and so would a myriad of other devices. One could call a semiconductor a narrow-gap insulator in the sense that its energy gap between the highest filled band (the valence band) and the lowest unfilled band (the conduction band) is typically of the order of one electron volt. The electrical conductivity of a semiconductor is consequently typically much less than that of a metal. The purity of a semiconductor is very important and controlled doping is used to vary the electrical properties. Donor impurities are added to increase the number of electrons and acceptors are added to increase the number of holes (which are caused by the absence of electrons in states normally electron occupied, holes act as positive charges). Donors are impurities that become positively ionized by contributing an electron to the conduction band, while acceptors become negatively ionized by accepting electrons from the valence band.

The electrons and holes are thermally activated and in a temperature range in which the charged carriers contributed by the impurities dominate, the semiconductor is said to be in the extrinsic temperature range, otherwise it is said to be intrinsic. Over a certain temperature range, donors can add electrons to the conduction band (and acceptors can add holes to the valence band) as temperature is increased. This can cause the electrical resistivity to decrease with increasing temperature giving a negative coefficient of resistance. This is to be contrasted with the opposite behavior in metals. Fig. 1.1 shows the three types of energy band gaps.

Fig. 1.1 energy band gap for metal, semiconductor, and insulator
1.1.2 Calculation of Electron and Hole Concentration in semiconductor

Here we give the standard calculation of carrier concentration based on (a) excitation of electrons from the valence to the conduction band leaving holes in the valence band, (b) the presence of impurity donors and acceptors (of electrons) and (c) charge neutrality. The charge-carrier population of the levels is governed by the Fermi function \( f \). The Fermi function evaluated at the Fermi energy \( E = \mu \) is \(1/2\). We have assumed \( \mu \) is near the middle of the band. The Fermi function is given by

\[
f(E) = \frac{1}{\exp\left(\frac{E - \mu}{k_B T}\right) + 1}
\]

..........................1.1

We now evaluate expressions for the electron concentration in the conduction band and the hole concentration in the valence band. We assume the non degenerate case when \( E \) in the conduction band implies \((E - \mu) >> kT\), so

\[
f(E) \approx \exp\left(-\frac{E - \mu}{kT}\right)
\]

..........................1.2

We further assume a parabolic band, so

![Fig. 1.2 Energy gaps, Fermi function, and defect levels (sketch). Direction of increase of \( D(E) \), \( f(E) \)is indicated byarrows](image)

\[
e = \frac{\hbar k^2}{2m_e} + E_c
\]

(1.3)
$E_C$ is the energy of the bottom of the conduction band where $m_e^*$ is a constant. The density of states is given by

$$D(E) = \frac{1}{2\pi^2} \left( \frac{2m_e^*}{\hbar^2} \right)^{\frac{3}{2}} \sqrt{E - E_C}$$  (1.4)

The number of electrons per unit volume in the conduction band is given by

$$n = \int_{E_c}^\infty D(E) f(E) dE$$  (1.5)

Evaluating the integral, we find

$$n = 2 \left( \frac{m_e^* KT}{2\pi \hbar^2} \right)^{\frac{3}{2}} \exp \left( \frac{\mu - E_C}{KT} \right)$$  (1.6)

For holes, we assume, following (1.3),

$$E = E_v - \frac{\hbar^2 K^2}{2m_h^*}$$  (1.7)

the density of states for holes is

$$D(E)_h = \frac{1}{2\pi^2} \left( \frac{2m_h^*}{\hbar^2} \right)^{\frac{3}{2}} \sqrt{E_v - E}$$  (1.8)

The number of holes per state is

$$f_h = 1 - f(E) = \frac{1}{\exp \left( \frac{\mu - E}{KT} \right) + 1}$$  (1.9)

Again, we make a non degeneracy assumption and assume $(\mu - E) >> kT$ for $E$ in the valence band, so

$$f(E)_h \approx \exp \left( \frac{E - \mu}{kT} \right)$$  1.10

The number of holes/volume in the valence band is then given by

$$p = \int_{-\infty}^{E_v} D_h(E) f_h(E) dE$$  (1.11)

from which we find

$$p = 2 \left( \frac{m_h^* KT}{2\pi \hbar^2} \right)^{\frac{3}{2}} \exp \left( \frac{E_v - \mu}{KT} \right)$$  (1.12)

Since the density of states in the valence and conduction bands is essentially unmodified by the presence or absence of donors and acceptors, the equations for $n$ and $p$ are valid with or without donors or acceptors. (Donors or acceptors, as we will see, modify the value of the chemical
potential, \( \mu \).) Multiplying \( n \) and \( p \), we find

\[ np = n_i^2 \] (1.13)

where

\[ n_i = 2 \left( \frac{K T}{\pi \hbar^2} \right)^{\frac{3}{2}} \left( m_e^* m_h^* \right)^{\frac{3}{4}} \exp \left( -\frac{E_g}{2 K T} \right) \] (1.14)

where \( E_g = E_c - E_V \) is the band gap and \( n_i \) is the intrinsic (without donors or acceptors) electron concentration. Equation (1.13) is sometimes called the Law of Mass Action and is generally true since it is independent of \( \mu \).15

### 1.2 Magnetism

#### 1.2.1.1 Atomistic Origins of Magnetism

In order to understand why semiconductors are traditionally not magnetic and how it is possible to introduce magnetic behavior in these materials, it is first necessary to look at the fundamental origins of magnetic behavior, and then analyze how magnetic behavior can be exploited in the development of dilute magnetic semiconductor compounds. Magnetism in materials arises from the fundamental property of electron spin, which is the intrinsic angular momentum of an electron. This intrinsic spin and the orbital angular momentum of an electron within an atom represent two of the four fundamental quantum numbers of an electron within an atom. The behavior of a materials system is not determined by individual isolated atoms. The magnetic behavior is determined by the collective behavior of all of the atoms within the system. Depending on the alignment of and interaction between atoms within a material, the observed macroscopic behavior of the system can be very different. The fundamental types of magnetism in a materials system are described below.

#### 1.2.1.2 Paramagnetism

In a paramagnet, the magnetic moments tend to be randomly orientated due to thermal fluctuations when there is no magnetic field. In an applied magnetic field these moments start to align parallel to the field such that the magnetisation of the material is proportional to the applied field.

![Schematic showing the magnetic dipole moments randomly aligned in a paramagnetic sample.](image)
1.2.1.2 Diamagnetism

Most materials exhibit an overall magnetic behavior known as diamagnetism. In the presence of a magnetic field, the material develops an internal magnetization opposite the direction of the applied field. This is a materials analogy to Lenz’s Law, where a changing magnetic field induces a current in a coil of wire opposite the direction of the applied field. This type of behavior is prevalent in materials with no unpaired electrons and thus no net magnetic moment internally; adding an additional magnetic field cannot align the electron spins as they are already paired. Using Langevin theory, the magnetic susceptibility of a diamagnetic material is given by the expression:

The diamagnetic signal is roughly independent of temperature. Only in the case of superconductors is the diamagnetic field unity; here the magnetic field is completely expelled from the material through a phenomenon known as the Meissner effect.

1.2.1.3 Ferromagnetism

Although diamagnetism is present in many materials, due to its weak and transient nature, it is not particularly useful for most applications. In order to have a strong macroscopic signal that is useful, a material must possess not only extra spins on the atoms, but also a mechanism by which the spins collectively align in the same direction. Iron is one such material, hence the origin of the name. With ferromagnetic materials, an external magnetic field can be applied to align the extra spins on the individual atoms in the same direction. Upon removal of this field, a majority of these spins remained aligned in this direction through an energetically favorable exchange mechanism; this will be discussed later in this chapter in relation to magnetic semiconductors. In metals, the ferromagnetism is stabilized by interaction with the free electron carriers in the system; this mechanism is know as the RKKY interaction after the four scientists who proposed it in the late 1950’s. In order to align the spins in the opposite direction, an additional field must be applied with an opposite field strength. In general, the strength of a magnetic signature from a concentrated ferromagnet can be roughly 6 orders of magnitude greater than that of a typical diamagnetic. Also, because the signal remains after the external field is removed, these materials can be used in applications where non-volatility is required, such as nonvolatile magnetic data storage[48]
1.2.1.4 Antiferromagnetism and Ferrimagnetism

Antiferromagnetism is similar to ferromagnetism except that the lowest-energy state involves adjacent spins that are anti parallel rather than parallel ferrimagnetism is similar to antiferromagnetism except that the paired spins do not cancel and thus the lowest-energy state has a net spin. Ferrimagnetism requires the existence of a strong positive exchange interaction that causes the excess electronic spins to align in the same direction. It is not always the case that the most energetically favorable magnetic alignment of spins will be in the parallel direction. Often, even via the RKKY interaction, the most energetically favorable alignment is to have adjacent spins couple and align in an anti parallel direction. Like ferromagnetism, this will occur below a specific ordering temperature, above which the random paramagnetic alignment will prevail; in this case, it is called the Neel temperature. Unlike in the ferromagnetic case, there is no net magnetic moment; the alignment of the various spins will cancel out, as there are equal number of atoms with opposite spin. A schematic of the antiferromagnetic arrangement is shown in Fig 1.4a 

An alternate case of antiferromagnetic exchange which leads to a net overall magnetic moment is known as ferrimagnetism. As the name suggests, it is similar, but weaker in magnitude to ferromagnetism. In ferrimagnetism, the net nearest neighbor exchange is antiferromagnetic, such that atoms couple with opposite spins. However, either due to a difference in the either the number of sites with the opposite spins or through engineering of the compound to contain elements with different numbers of unpaired spins on each site, it is possible to have an incomplete cancellation of the unpaired spins and an overall net magnetic moment in the system.[49]

![Fig 1.4 Spin Alignment of Ferromagnetic, antiferromagnetic and ferrimagnetic Substance](image)
CHAPTER 2

2 OVERVIEW OF DILUTE MAGNETIC SEMICONDUCTORS

2.1 Introduction: A little History of Diluted magnetic semiconductors

Magnetic semiconductors are recently considered as one of the main building blocks for future replacement of conventional electronics by spin-electronics or spintronics. The latter is a field of materials research born from the need of higher speed, lower power consumption and smaller integration densities of computational devices. A model device of spintronics was proposed already in the year 1990. The field of Diluted magnetic semiconductors (DMS) was greatly pushed in the year 2000 when $Ga_{1-x}Mn_xAs$ was, both experimentally and theoretically, found to be ferromagnetic up to 110 K. The mechanism of the ferromagnetic interaction is based on the exceptional case that Mn doping introduces shallow acceptors and thus holes into the valence band of the GaAs that are antiferromagnetically coupled to the local Mn moments. T. Dietl calculated the Curie temperature of different p-type semiconductors doped with Mn ions. He found that it is proportional to the concentration of the impurities and to the square root of the p-type charge carrier concentration. Recently, the Curie temperature of $Ga_{1-x}Mn_xAs$ could be increased up to 173 K by means of increasing its crystal quality and the number of Mn impurities up to 6.8% [16].

Diluted magnetic semiconductors which are materials with some fraction of the nonmagnetic lattice atoms replaced by magnetic atoms (i.e., a fraction $x$ of Ga, In, or Ge atoms being randomly replaced by Mn atoms), have attracted a great deal of attention following the recent discovery of ferromagnetism in $Ga_{12x}Mn_xAs$, with a Curie temperature $T_c$ 100 K in the ($x \approx 0.03-0.07$) range. The subject is of considerable fundamental and technological interest. Technologically, a semiconductor, which is also a ferromagnet, raises the exciting potential of spintronic applications[1, where logic and memory operations could in principle be seamlessly integrated on a single device. From a fundamental perspective, understanding ferromagnetism in a novel material (which is also a semiconductor) is an important challenge. It is therefore not surprising that there has been a great deal of theoretical activity[34] trying to understand the ferromagnetic mechanism in GaMnAs. Although no theoretical consensus on the precise ferromagnetic mechanism has yet been reached in the literature it is now established that the interaction between the magnetic atoms, which leads to the ferromagnetic phase at low enough
temperatures, is induced by charge carriers (holes in the case of GaMnAs) in the semiconductor host. Theoretical investigation of these systems is hampered by the fact that both disorder and interactions are strong and must be taken into account non perturbatively. This problem has been approached in a number of ways, including theoretical approximations assuming charge carriers being almost free and numerical studies in the opposite limit of strongly localized charge carriers. However, a comprehensive understanding of the physics of diluted magnetic semiconductors has not been achieved yet.

The potential for a material to possess both ferromagnetic and semiconducting properties has been known and studied since the 1960s [24] Semiconductor physics and magnetism are established subfields of condensed matter physics that continue to reveal a rich variety of unusual phenomena, often in new types of solid-state materials. The properties of semiconductors are extraordinarily sensitive to impurity atoms, defects, and charges on external gates. Magnetism is a collective electronic phenomenon with an ordered state that is often stable to exceptionally high temperatures. Magnetic order, when it is present, has a large impact on other material properties including transport and optical properties. In both semiconductor and magnetic cases, sophisticated and economically important technologies have been developed to exploit the unique electronic properties, mainly for information processing in the case of semiconductors and for information storage and retrieval in the case of magnetism. The realization of materials that combine semiconducting behavior with robust magnetism has long been a dream of material physics. One strategy for creating systems that are simultaneously semiconducting and magnetic, initiated in the late 1970s is to introduce local moments into well-understood semiconductors. The result is a new class of materials now known as diluted magnetic semiconductors (DMSs). Over the past 15 years, building a series of pioneering publications in the 1990s it has been established that several (III,V) compound semiconductors become ferromagnetic when heavily doped with Mn, and that the ferromagnetic transition temperatures can be well above 100 K. In semiconductors like GaAs and InAs, Mn has been shown to act both as an acceptor and as a source of local moments. These (III,Mn)V materials are examples of ferromagnetic semiconductors, a phrase we reserve for magnetic systems in which ferromagnetism is due primarily to coupling between magnetic element moments that is mediated by conduction-band electrons or valence-band holes. This definition implies that, in ferromagnetic semiconductors, magnetic properties can be influenced by the same assortment of engineering variables that are available for other more conventional semiconductor
electronic properties. In the best-understood arsenide DMSs, semiconductor valence-band carriers participate in the magnetic order. The materials require participation of valence-band holes for the formation of a ferromagnetic state. Efforts to increase their critical temperatures further run into incompletely understood fundamental limits on the ratio of the magnetic transition temperature to the Fermi temperature of the free-carrier systems and are also affected by the role of disorder in these heavily doped materials. The tension between achieving high Curie temperatures and the desire for low, and therefore gate able, carrier densities is among the major issues in the study of these materials In this article we review the considerable theoretical progress that has been made in understanding the very broad range of properties that occur in (III,Mn)V ferromagnetic semiconductor epilayers in different regimes of Mn content and defect density. The main focus of this article is on the extensively studied (Ga,Mn)As ferromagnetic semiconductor,

The benefits of DMS-based devices include reduced power requirements compared to traditional semiconductor devices. This energy gain is due to the reduced power needed to flip an electron spin, as opposed to moving a charge in an electric field. However, before these devices can be realized, it will be necessary to determine if the injection, transport, and detection of the carrier spins can be successfully carried out at meaningful temperatures [25].

One major incentive for finding a DMS material that can operate at meaningful temperatures (i.e., above room temperature), is that one could then integrate the new technology with the existing semiconductor industry. Recent theoretical work by T. Dietl et al. [20] generated interest in the possibility of achieving room-temperature ferromagnetism in a DMS that is based on GaN, using Mn as the transition metal dopant.

2.2 Ferromagnetic semiconductors (FS)

Ferromagnetic semiconductors (FMSs) are materials that simultaneously exhibit semiconducting properties and spontaneous long-range ferromagnetic (FM) order. Classic examples, studied decades ago, include the europium chalcogenides and the chalcogenide spinels. The coexistence of these properties in a single material provides fertile ground for fundamental studies as well as a host of new applications. Development of novel devices languished for some time due to the inability to incorporate these materials with mainstream semiconductor device materials.

Interest in FMSs was rekindled with the discovery of spontaneous FM order in $In_{1-x}Mn_xAs$ in 1989 and $Ga_{1-x}Mn_xAs$ in 1996, when FM properties were realized in semiconductor hosts already widely recognized for semiconductor device applications.[26] Although these new FMS
materials have magnetic ordering, or Curie, temperatures (Tc) below room temperature, they have been closely studied for their potential in future spin-dependent semiconductor device technologies, with the expectation that further research will increase Tc. $Ga_{1-x}Mn_xAs$, for example, has been used as a source of spin-polarized carriers in both light-emitting diodes and resonant tunneling diode heterostructures.

This given name to this category may be somewhat misleading since it can be mixed-up with the next group. It refers to semiconductors in which, at least one element constituting the unit cell of the material is magnetic (fig.2.1). Materials such as EuO and GdN shows ferromagnetic order below their Curie temperature which is usually low due to the weak exchange integral between the localized electrons of the f-shell that builds the magnetic moment and the d-electrons in the conduction band. These materials were studied strongly in 1970’s and then abandoned for a certain time. The arising interest toward these materials can be seen more and more in recent years after the appearance of spintronics domain. The difficult growth process of these materials as single crystal thin films on the standard substrate, the incapability of codoping them because of the large density of intrinsic defects their low Curie temperature are among the points which should be solved before their application in spintronics domain. [23]

![Fig 2.1: Schema concerning different materials which can be applied in spintronic](image)

### 2.2.1 Ferromagnetic GaMnAs

the extension of spintronics from metallic alloys to traditional semiconductor materials such as GaAs opens the door for new applications. Working with well-established compound semiconductor technologies, control of electron spin can be incorporated into structures and devices that already capitalize on the electrical and optical advantages of III-V and II-VI materials. The incorporation of a small amount of Mn into the GaAs lattice produces ferromagnetic, $Ga_{1-x}Mn_xAs$ a promising alloy for combining spin effects with traditional electronics. Recent studies of MBE[9] growth conditions for GaMnAs have resulted in improved material
quality.

One of the properties of interest when considering ferromagnetic materials for device applications is the Curie temperature (Tc) at which the material loses its ferromagnetic properties. This temperature is theoretically predicted to be close to 300K for GaMnAs. Earlier studies, in which the Tc did not exceed ~110 K, prompted concern that this could be a fundamental limitation in low-temperature grown GaMnAs. As an acceptor, each Mn atom (which substitutes for a Ga atom) provides one hole, but these carriers may be compensated by As antisite defects common in low-temperature grown GaAs, as well as by interstitial Mn incorporation. The current work reports the reproducible growth of GaMnAs with Curie temperatures up to 150-160 K in annealed samples. The results may be extrapolated to determine the material requirements for room temperature ferromagnetism.

2.3 DMS MATERIALS

Two major criteria are considered to select the most promising materials for semiconductor spintronics. First, the ferromagnetism should be retained to practical temperatures namely room temperature. Second, it would be a major advantage if there were already an existing technology base for the material in other applications. A very interesting family of magnetic materials can be obtained from the semiconductors already applied in micro electronics. The largely studied materials such as Si, GaAs and GaN are among the semiconductors which are widely used not only for their electrical properties (as processing unit devices) but also for their opto-electronic properties (LED). Of course these performing materials are not intrinsically magnetic. In order to introduce magnetic properties to these systems they have to be doped by magnetic ions such as the transition metal ions. In this case some of these materials present ferromagnetism (with different $T_C$) and become useful in spintronics.

The advantages of using semiconductor systems instead of the metallic systems lies in the capability of the manipulation of the magnetic state of these materials via optical, electrical and structural control (this case will be discussed in more details in following sections). The longer mean free path of the spin-polarized carriers in these materials and the stronger spin-orbit interaction are other advantages. Also the unification of the processing and storage units in information is made possible in their application.

The very important and major drawback of these materials is their low $T_C$. At present time, the critical temperatures are still far below the room temperature and prevent their direct
application. A theoretical study of the magnetic properties of different p-type semiconductors doped by manganese (Fig. 2.2). They predicted from the mean field (MF) theory, the Curie temperatures above the room temperature for the two cases of GaN and ZnO systems.

![Curie temperature graph](image)

**Fig. 2.2 Computed values of the Curie temperature $T_C$ for various p-type semiconductors containing 5% of Mn and $3.5 \times 10^{20} \text{cm}^{-3}$ holes ([9])**

Sofar, the only III-V DMS members which have shown high single-phase intrinsic ferromagnetism are GaMnAs and InMnAs. The former material has been the object of numerous studies, just as the GaAs host material. The very high compatibility of this material with the electronic technology, in addition to its Curie temperature which has increased from its first observation at $T \approx 50K$ ([11]) to $T = 180K$ in recent samples ([13]) in only 10 years, both have motivated us to investigate its magnetic properties and the $T_c$ related parameters. We were interested both in its fundamental properties and the material aspects which would allow to push the critical temperature up to room temperature. [23]

### 2.3.1 The Magnetic Elements for Doping

The magnetic elements for doping purpose are mainly; Cr, Mn, and Fe in diluted magnetic semiconductors. The table shown below gives configuration why Mn$^{2+}$ is more appropriate.

<table>
<thead>
<tr>
<th>element</th>
<th>Cr$^{24}$</th>
<th>Mn$^{25}$</th>
<th>Fe$^{26}$</th>
<th>Cr$^{2+}$</th>
<th>Mn$^{2+}$</th>
<th>Fe$^{2+}$</th>
<th>Fe$^{3+}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Configuration</td>
<td>$3d^5 4s^1$</td>
<td>$3d^5 4s^2$</td>
<td>$3d^6 4s^2$</td>
<td>$3d^4$</td>
<td>$3d^5$</td>
<td>$3d^6$</td>
<td>$3d^5$</td>
</tr>
</tbody>
</table>

In Mn$^{2+}$ doped III-V DMSs, there is a valence mismatch between that of Mn and the group III-
element with \( S=5/2 \) creating holes in the valence band. In Cr\( ^{2+} \) doped III-V the 3d electrons is less in number which is 4 pair atom and \( S=4(1/2)=2 \) where less magnetic impurity spins per atom results in weak ordering in the compound. In Fe\( ^{2+} \) the 3d electrons gives rise to \( S=4(1/2)=2 \) and Fe\( ^{3+} \) gives the valence match with group III element as in II-VI DMSs case. Therefore, Mn\( ^{2+} \) with shorter nearest neighbor distance of \( 2.24^\circ \text{Å} \) relatively, is more preferable for doping purpose in which high concentration of 3d spins is required.

### 2.3.2 GaAs, The Host Material (structural and Electrical Properties)

GaAs is one of the most largely studied semiconductors because of its very particular properties which can be found nowadays in many electronics and semiconductors handbooks. It is used to make devices such as microwave frequency integrated circuits (i.e. MMICs), infrared light-emitting diodes, laser diodes and solar cells. The crystal structure of this material is shown in fig. 2.3. It is a Zinc-blend structure with four Ga(or As) atoms per lattice. The lattice parameter of stoichiometric material is \( a = 5.653\text{Å} \). Usual techniques used for growing epitaxial GaAs layers include Molecular beam epitaxy (MBE), Low pressure epitaxy (LPE), VPE and Molecular Chemical Vapor Deposition (MOCVD). In its epitaxial form it contains both donor and acceptor impurities and the relative concentration of these impurities determine the p or n-type of GaAs. GaAs is a direct band gap semiconductor with gap energy of 1.519 eV at \( T \approx 0\text{K} \) which decreases as a function of temperature. The As has the higher contribution to the hybrid valence band. On the high pressure this band gap will change also into an indirect band gap.[23]

![Fig 2.3 The crystal structure of the semiconductor GaAs. If Mn is introduced as a dopant, three different scenarios are possible: (1) it can substitute the Ga atom; (2) it can find an interstitial position between four cations; or (3) it can find an interstitial position between four anions.](image)
GaAs are already in use in a wide variety of electronic equipment in the form of electronic and optoelectronic devices, including cellular phones, compact disks, and in many other applications. Therefore, the introduction of magnetic semiconductors based on GaAs opens up the possibility of using a variety of magnetic phenomena not present in conventional nonmagnetic GaAs material in the optical and electrical devices already established. In 1996, Ohno firstly used MBE technique to produce Ga1−x Mnx As thin film with a largely enhanced solubility of the order of 1020 cm−1. Through study of its magnetic transportation, it was found that the Curie temperature varied as a function of doping factor x, and its behavior satisfied $T_c = 2000x \pm 10K$[6]. Later F. Matsukura etc discovered that the Curie temperature of (Ga,Mn)As reached its highest 110K when $x = 0.05$, as shown in Fig. 2. When Mn concentration was reduced, Curie temperature $T_c$ would also decrease, when Mn concentration $x$ went below 0.005, ferromagnetism would disappear[14]. Moreover, as Mn concentration in (Ga,Mn)As increases, the transportation properties experienced series of changes, i.e., non-metal to metal and again to non-metal[15].

![Fig. 2.4: Curie temperature vs. Mn concentration for DMS (Ga,Mn)As][14]

The Study of origin of ferromagnetism in (Ga,Mn)As has always attracted lots of attentions. H. Ohno etc believes that it is hole charges who lead to ferromagnetism in (Ga,Mn)As[6]. They discovered that the number of Mn ions in (Ga,Mn)As is of the same order compared to those of hole charges, which supported their opinion. Nowadays, it is still not fully understood the reason of ferromagnetism in (Ga,Mn)As. Based on the study of (Ga,Mn)As, Other DMS materials with even higher Curie temperature has been discovered, for example, (Ga,Mn)N, Co: TiO2 etc. However, they are all not as good as (Ga,Mn)As in terms of experimental capability and compatibility with current semiconductor industry. Till now (Ga,Mn)As is the most promising DMS material in practice.
2.3.3 Crystal Structure and Growth

Alike the GaAs host lattice, the $Ga_{1-x}Mn_xAs$ layers exhibit a zinc blende lattice structure, where the manganese atoms are preferentially incorporated at Ga lattice sites. Here $x$ denotes the Mn concentration with respect to the Ga lattice sites. $Ga_{1-x}Mn_xAs$ layers are typically grown via molecular-beam epitaxy (MBE) on semi-insulating (001)-oriented GaAs substrates. However, at the optimal GaAs growth temperature ($\approx 600^{\circ}C$) the solubility of Mn atoms is very low ($\approx 10^{-18}cm^{-3}$), allowing a maximum $x \approx 1x10^{-4}$ without the precipitation of inter metallic MnAs clusters. To obtain $Ga_{1-x}Mn_xAs$ As layers with a Mn-content sufficient for the formation of ferromagnetism ($x > 0.01$), growth is performed beyond thermal equilibrium at substrate temperatures of $250^\circ C$ to $270^\circ C$ denoted low-temperature MBE (LT-MBE) [49] to obtain ferromagnetic films of GaMnAs, it is necessary to grow the material at low temperature and to anneal for an appropriate time at temperature similar used for growth. The anneal temperature that we have used ($\approx 190^\circ C$ for all samples) has been chosen to allow a slow anneal, during which the resistivity of the sample can be monitored in order to optimize the process. The growth temperature is necessary to prevent Mn out diffusion or surface segregation, but this intern means that arsenic anti-cites defects may be present, which act as double donor and thus compensate the holes introduced by the Mn atoms on the Ga sub-lattice. Interstitial Mn will also compensate the whole density since they also act as a double donor [35].

The major obstacle in making III-V semiconductors magnetic has been the low solubility of magnetic elements (such as Mn) in the compounds. Because the magnetic effects are roughly proportional to the concentration of the magnetic ions, one would not expect a major change in properties with limited solubility of magnetic impurities, of the order of $10^{18}cm^{-3}$ or less. A breakthrough was made by using molecular beam epitaxy (MBE), a thin film growth technique in vacuum that allows one to work far from equilibrium. When a high concentration of magnetic elements is introduced in excess of the solubility limit, formation of the second phase occurs if conditions are near equilibrium. However, when crystal is grown at low temperature by MBE, there is not enough thermal energy available to form the second phase, and yet there still exists a local potential landscape that allows epitaxial growth of a single-crystal-alloy. The effort to grow new III-V based DMSs by low-temperature MBE was rewarded with successful epitaxial growth of uniform (In,Mn)As films on GaAs substrates in 1989, where partial ferromagnetic order was found, and ferromagnetic (Ga,Mn)As in 1996.
In a ultra-high vacuum, a beam of atoms or, more general, a beam of molecules is directed towards a crystalline substrate such that the atoms or molecules stick at the substrates surface forming a new layer of deposited material, as shown in Fig. 5. The difference between MBE and other material deposition methods as e.g. thermal vacuum evaporation is as follows. MBE does not only deposit material like it is done by conventional evaporation techniques, but using the very low rates of impinging atoms, migration on the surface and subsequent surface reactions ensure the controlled epitaxial growth of a new layer. Simply spoken: Every atom reaching the surface of the heated substrate has enough time to migrate around and find his place to build up a new crystal lattice. With lots of physics applied, chemistry and engineering work done since beginning MBE in the late 60s and the early 70s to reach state of the art MBE. The typical construction of a MBE is shown in Fig. 6. At the left hand side there are the eusion cells to provide the molecular beam for either the bulk constituents or the dopants. These cells can be thermal evaporation cells (Knudsen cells), cells for gaseous media or plasma sources as well. In front of them is a shutter, this means a plate which could be brought into the beam for switching the beam on and off. Opposite to the cells and the shutters is the substrate, mounted on a heatable and rotatable substrate holder. The whole system is in ultra-high vacuum environment to guarantee formation of a molecular beam (without vacuum, the atoms or molecules leaving the eusion cells will be scattered at residual gas molecules and never form a beam directed towards substrate) and purity and therefore quality of the grown layer. Often there are instruments for in-situ analysis like RHEED attached to the growth chamber. A cryopanel around the sample and the cells absorbs residual gases and provides a clean substrate environment[9]Molecular beam epitaxy (MBE) researchers are exploring the growing field of
spintronics to couple the traditional properties of compound semiconductor materials with the spin properties of ferromagnetic materials. The quantum property of spin, which plays no role in conventional electronics, may exploited for a range of powerful new applications[53]. The precise atomic layer control and layer-by-layer deposition capabilities of MBE are ideal for producing structures to test and advance this new technology.

2.3.4 Functional (III,Mn)V Material Requirements

III-V materials are among the most widely used semiconductors. There is little doubt that ferromagnetism in these materials would enable a host of new microelectronics device applications if the following criteria were met: (i) the ferromagnetic transition temperature should safely exceed room temperature, (ii) the mobile charge carriers should respond strongly to changes in the ordered magnetic state, and (iii) the material should retain fundamental semiconductor characteristics, including sensitivity to doping and light, and electric fields produced by gate charges. For more than a decade these three key issues have been the focus of intense experimental and theoretical research into the material properties of Mn-doped III-V compounds. At first sight, fundamental obstacles appear to make the simultaneous achievement of these objectives unlikely. Nevertheless, interest in this quest remains high because of the surprising progress that has been achieved. Highlights of this scientific endeavor are briefly reviewed in this introductory section.[16].

2.4 .SPINTRONIC MATERIALS & DEVICE

2.4.1 Introduction to spintronics

Spintronics is an emergent technology that exploits the quantum feature of the electron to spin and makes use of its charge state [1,2]. The spin itself is manifested as a detectable weak magnetic energy state characterized as spin up or spin down. Conventional electronic devices rely on the transport of electrical charge carriers – electrons - to a semiconductor such silicon or gallium arsenide. Now, however, device engineers and physicists are inevitably dealing with the looming presence of quantum mechanics and are trying to exploit the spin of the electron rather than its charge. Devices that rely on the electron spin to perform their function form the foundations of spintronics (short of spin- based electronics), also known as magneto electronics [3]. spintronics devices are smaller than 100 nm in size, more versatile and faster than those commercially used for making up processing chips and circuit elements. The potential market is worth hundreds of billions of Euros per annum [53].

While conventional semiconductor devices operate by controlling the flow of electrons and
holes based on their charge, the rapidly growing field of spintronics utilizes the fundamental property of electron spin for novel device applications. Spin is an intrinsic quantum property of electrons and is closely related to magnetism. In the simplest model, an electron can be depicted as a charged spinning ball which produces a small magnetic field along the direction of the spin axis. Since the spin can be either clockwise or counterclockwise around a given axis, the electron is either “spin up” or “spin down”, with the two states having different energy levels in an applied magnetic field. In traditional electronics, the electron spins are randomly oriented and have no effect on the operation and performance of a device. The novelty of spintronic devices is that the currents are spin polarized and the spin is used to control current flow or store data. Spintronic devices fall into three major categories.

• Ferromagnetic metallic alloys for read heads and magnetic random-access memory capitalize on the spintronic effect of giant magneto resistance (GMR). Such data storage techniques, while still advancing, are already in use and well-developed.

• Semiconductor structures which utilize spin-polarized currents can be integrated with existing microelectronics technology to produce ultra-fast switches and spintronic microprocessors. Such materials are currently being investigated in numerous MBE laboratories. Researchers are exploring how best to combine ferromagnetic and semiconductor layers, how to extend the ferromagnetic behavior of semiconductors to room temperature, and how to efficiently inject and retain spin-polarized currents in semiconductors and across junctions of different semiconductor materials. A variety of such issues and technological barriers remain to be solved before semiconductor spintronic devices are commercially viable.

• Finally, the more distant goal of creating devices that manipulate. Electrons is also under investigation for applications in large-scale quantum computing. A variety of systems are being explored for this application, including the MBE-grown solid state semiconductor materials discussed in this note. [53]

The field of magnetism has enchanted humans over a span of several centuries. It all started with the observation of the attraction of lodestone to a piece of iron. Mankind has come a long way from then to understand magnetism as one of quantum mechanical phenomena and also to control and commercialize the processes to produce state of the art devices like hard disks and magnetic random access memories which are based on metallic multilayers. Presently, magnetic devices utilize only the spin degree of freedom of the electrons to bring about these functionalities.
Conventional electronics utilizes only the charge degree of freedom of the carrier. Recently, a new field named Spintronics has emerged wherein it is envisioned that novel functional devices can be fabricated if the charge as well as the spin degrees of freedom can be combined [33].

A new approach to electronics, called 'Spintronics' (a short for spin electronics), is now emerging, and it is based on the up and down spin of the charge carrier rather than on electrons charge as in traditional semiconductor electronics. Spintronics, also called magnetoelectronics including all the electronic devices where ferromagnetic thin films play an essential role, is today one of the most rapidly growing fields in electronics. Spin electronics (spintronics) is a young interdisciplinary field of nanoscience. Its rapid development, like that of competing new branches of electronics –. The main goal of spintronics is to gain knowledge on spin-dependent phenomena, and to exploit them for new functionalities. All the devices in spintronics made so far are based on metallic thin films. However, it is anticipated that if ferromagnetism and electroThe Curie temperature itself is a critical point, where the magnetic susceptibility is theoretically infinite and, although there is no net magnetization, domain-like spin correlations fluctuate at all length scales. The effect of magnetism would be significantly stronger than the phenomena observed in metals. Indeed, a recently found new ferromagnetic semiconductor thin film (Ga,Mn)As has opened a completely new road to combine magnetism with charge transport in well known semiconductor device structures. Ferromagnetic semiconductors such as EuO and $CdCr_2S_4$ have been known already since the 1960's, but an extreme difficulty in growing these crystals has hindered the study and utilization of these materials. On the other hand, ferromagnetism has not been a part of the activity in Ga As-based devices because of the lack of material technologies that allow us to introduce magnetic cooperative phenomena into compound semiconductor heterostructure. However, now the situation has changed completely, and the success in synthesis of III-V materials and transition elements like Mn in (Ga,Mn)As and (In,Mn)As has introduced a new and unexplored degree of freedom related to ferromagnetism, which can be combined with all the other degrees of freedom we already enjoy today in Ga As device technology. Indeed, recent experimental studies have shown that charge carriers in Ga As can preserve their spin states over long distances, which may eventually enable even the realization of quantum computers in solid state. The highest critical temperatures observed so far in (Ga,Mn)As have been in the range about 170 K and the increase in the critical temperature has stopped so far.[15]
2.4.2 Advantage of Spintronics

One of the alternative options is the application of another degree of freedom, the spin of the carriers. Already spintronic plates are used in the field of mass-storage devices and have led to compressing massive amounts of data into a small area, at approximately one trillion bits per square inch (1.5Gbit/mm²)[23].

Conventional electronics is based on moving around the electron’s charge and manipulate it using electric fields. The electrical charge of electrons is used to process and store digital information. However, as the device size shrinks and miniaturizes to the nano scale, the devices operate at higher speed, driving an exponential growth in the amount of power needed to move electrons around. This results in a serious heat dissipation problem. Scientists have realized that the answer may lie in the spin of the electron. Rather than moving charges around, spin-based devices that would operate by flipping the electron’s spin orientation would use less energy, generate less heat and, would be faster than conventional charge-based devices. This has resulted in a new field of research spin electronics or spintronics that offers a new paradigm for the development of novel devices for use in the post-CMOS era. In spin-based digital information processing, the two orientations of the electron spin - “up” or “down” - can be used, instead of charge accumulation, to represent or encode the bits 1s and 0s, used by all computers. Spin-based computing will be much faster[17].

The advantage of spin over charge is that spin can be easily manipulated by externally applied magnetic fields, a property already in use in magnetic storage technology. Another significant property of spin is its long coherence, or relaxation, time (nanoseconds, compared to tens of femto seconds during which electron momentum decays) once created it tends to stay that way for a long time, unlike charge states, which are easily destroyed by scattering or collision with defects, impurities or recombination [21]. These characteristics open the possibility of developing devices that could be much smaller, consume less power and will be more powerful for certain types of computations which is not possible with electron-charge based systems. It is widely expected that new functionalities for electronics and photonics can be derived if injection, transfer and detection of carrier spin can be controlled over room temperature[22].

The four main investigation areas in spintronics are:

I. Understanding the fundamental physics, such as spin-dependent transports across the magnetic/semiconductor interfaces and spin coherence length in semiconductors
II. Synthesizing suitable spintronics materials with as higher as possible Curie Temperature, large spin polarization at the Fermi level and matching conductivity between the magnetic and semiconductor materials;

III. Fabricating devices with nanometers feature sizes and developing new techniques for mass production;

IV. Integrating spin-devices with current microelectronics
CHAPTER 3

3.THEORETICAL APPROACH TO EXPRESS DILUTE MAGNETIC SEMICONDUCTOR

3.1 Theoretical Models for Explaining Ferromagnetism in DMS

Although a number of different theories have been proposed, the mechanisms responsible for the origin of ferromagnetism in diluted magnetic semiconductor is not fully understood and explained yet. In this chapter, the theoretical models that have been most commonly used to explain ferromagnetism in diluted magnetic semiconductors are summarized.

3.1.1 The Zener Model

The theory proposed by Zener [39] indicates that the direct super-exchange interaction [82] between half filled d-shell electrons of TM cations and completely filled p-orbitals of anion is antiferromagnetic (Fig. 3.1). Since the d-shell electrons from both adjacent TM atoms occupy the same p-level, their spins must be opposite according to Pauli exclusion principle. This leads to an anti ferromagnetic coupling of nearest-neighbor TM cations through a shared anion. On the other hand, the indirect super exchange interaction between localized d-shell electrons of TM cations mediated by the delocalised band carriers tends to align the spin of the partially filled d-shells in a ferromagnetic manner (Fig. 3.2). In the Zener model, ferromagnetism is only possible when the indirect exchange interaction dominates over the direct superexchange interaction.

![Fig 3.1: Direct superexchange interaction: Antiferromagnetic coupling of adjacent TM cations through shared anion.](image)

3.1.2 The Mean-Field Model

In the mean-field model, the interaction between the localized Mn atoms through free holes in the material leads to a ferromagnetic alignment of Mn atoms (Fig. 3.3). Due to the possibility of direct superexchange Mn-Mn interactions, there is always a competition between the ferromagnetic and antiferromagnetic interactions.
3.1.3 The RKKY Model

The RKKY interaction was first proposed by Ruderman and Kittel [49] and later extended by Kasuya and Yoshida. It has originally been developed for nuclear magnetic moments in a metal. It is an indirect interaction of local moments via conduction electrons. If one puts an impurity into the free electron gas, slowly decaying oscillations of the electron charge density centered around the perturbation are found. These so-called Friedel oscillations are a consequence of screening. If the oscillations are different for spin-up and spin-down electrons one expects oscillations of the magnetization density. The mutual interaction of two impurities via the oscillations of their corresponding magnetization densities is then called RKKY interaction. For an impurity within a simple cubic lattice assuming the free electron dispersion and zero temperature, the interaction is of the form

\[ I(R_i - R_j) = -\frac{j^2}{\pi^2} \frac{a_o^6}{2} \frac{\sin 2K_F R_{ij} - 2K_F R_{ij} \cos 2K_F R_{ij}}{(2k_F R_{ij})^4} \]  

and the corresponding Hamiltonian is given by

\[ H_{RKKY} = \sum_{K \alpha} \epsilon_k n_{k,\alpha}^+ + \frac{1}{2} \sum_{i \neq j} I(R_i - R_j) S_i S_j \]  

The argument of \( I \), \( R_{ij} = R_i - R_j \), denotes the distance between the magnetic impurities, \( a_o \) the simple cubic lattice constant and \( J \) the direct exchange constant between the impurity and the conduction electrons. The period of the oscillation is determined by the Fermi-surface diameter \( K_F \).

There exist two important limits. In the limit of high carrier concentrations and low concentrations of magnetic impurities ( \( 2K_F R_{ij} \gg 1 \) ) the RKKY interaction changes sign and spin-glass behavior can be found. In the other limit, namely low carrier concentrations and high concentrations of magnetic impurities ( \( 2K_F R_{ij} \ll 1 \) ), the RKKY interaction is always FM. In undoped semiconductors the lack of free carriers leads to an exponential decay of the RKKY
interaction and it becomes insignificant. For systems with a higher concentration of carriers the role of the RKKY interaction (e.g. as part of the mean-field Zener model) is still under debate [53]. The RKKY (Ruderman-Kittel-Kasuya-Yosida) model [40] describes the magnetic interaction between a single localized magnetic ion and the delocalized conduction band electrons. Due to this interaction the conduction electron close to the magnetic ion is magnetized and act as an effective field to influence the polarization of nearby magnetic ions with the polarization decaying in an oscillatory manner. This oscillation mediates either ferro- magnetic or anti ferromagnetic exchange coupling depending upon the separation between magnetic ions. This model is efficient when a high concentration of delocalized carriers is present in the host material.

**3.2 Exchange Interaction** are several origins for the interactions between the magnetic moments that can lead to a long-range ordering of the unpaired spins, called exchange interactions. The most common exchange interactions are schematically illustrated in Fig. 3.3

![Fig 3.3 Schematic illustration of (a) super exchange, where the interaction takes place via an intermediate non-magnetic ion; (b) direct exchange, in which the magnetic ions interact through their overlapping charge distributions; (c) indirect exchange, in which the interaction is mediated by interactions with the charge carriers.](image)

For spontaneous magnetization electron-electron (called ”magnetic interaction” not because of the origin of the force rather because of the effect of the force) is a must. Now there could be different types of interactions. It could either be dipolar interaction (interaction between two magnetic dipoles) which is magnetic in origin or it could be exchange interactions due to Pauli’s exclusion principle which is purely electrostatic or coulombic in origin. Qualitatively speaking, Pauli’s exclusion principle prohibits two electrons with same spin from taking same state and hence on an average they would be farther apart and therefore coulombic interaction energy would be lowered compared to the case when two electrons have opposite spin and hence can occupy same state.
Usually exchange interaction is much stronger compared to the dipolar interaction. There could be many types of exchange interactions as well:-

3.2.1 Direct Exchange Interaction:- it arises from the direct coulomb interaction among electrons from the two ions [9]. When paramagnetic ion/atom cores are next to each other on the lattice sites and if they are not sufficiently tightly bound .

3.2.2 Super-Exchange Interaction The Curie temperature itself is a critical point, where the magnetic susceptibility is theoretically infinite and, although there is no net magnetization, domain-like spin correlations fluctuate at all length scales. ction:- two magnetic ions interact with the mediation of non magnetic ion. In such cases when paramagnetic ion/atom electron clouds don’t overlap and there is another non paramagnetic ion/atom sitting in between we call such interactions as superexchange interaction.

3.2.3. Indirect Exchange Interaction:- In the sea of conduction electrons two paramagnetic ions/atoms can interact with each other by the mediation of free electrons. This type of exchange interaction is called indirect exchange interaction. One of the main mechanisms for indirect exchange interactions in DMS is RKKY mechanism.

3.2.4 Itinerant Exchange Interaction :- For those electrons that aren’t well localized and are shared by the entire crystal, such electrons can have exchange interaction among them selves. Such electron- electron interactions are called itinerant exchange interaction[9].

The singlet-triplet energy splitting measures the extent to which the anti-parallel (S=0) spin alignment of two electrons is more favorable than the parallel (S=1). Since \( E_s - E_t \) is the difference between eigenvalues of a Hamiltonian containing only electrostatic interactions, this energy should be of the order of electrostatic energy differences, and therefore quite capable of being the dominant source of magnetic interaction, even when explicitly spin dependent interactions are added to the Hamiltonian.

The singlet-triplet energy splitting is then \( E_s - E_t \) which is consistent with the general theorem \( E_s < E_t \) for two electron systems. The singlet-triplet energy difference is referred to as an exchange splitting or, when viewed as a source of magnetic interaction, an exchange interaction . For two atoms with two-electron systems at sites i and j in which the exchange integral is expressed as:

\[
J_{ij} = \frac{1}{2} \left| E_s - E_t \right|
\]
Equation (3.1) shows that half the energy difference between the singlet (symmetric) and the triplet (antisymmetric) state gives the exchange integral. The spin Hamiltonian for a two electron system is given by

$$H = -2j_{ij} \vec{S}_i \cdot \vec{S}_j$$  \hspace{1cm} (3.4)

For many cases of interest the form of the spin Hamiltonian is simply that for the two spin-case, summed over all pairs of ions:

$$H = -2 \sum J_{ij} \langle \vec{S}_i \rangle \langle \vec{S}_j \rangle$$  \hspace{1cm} (3.5)

The Curie temperature itself is a critical point, where the magnetic susceptibility is theoretically infinite and, although there is no net magnetization, domain-like spin correlations fluctuate at all length scales. The spin Hamiltonian (3.3) is known as the Heisenberg Hamiltonian and the $J_{ij}$ are known as the exchange coupling constants. [18]

### 3.3 Spin Glasses

Spin Glasses are dilute magnetic alloys where the interactions between spins are randomly ferromagnetic or anti-ferromagnetic, and are considered as paradigmatic examples of frozen disorder. The presence of disorder (the random interactions) induces frustration and a greater difficulty for the system to find optimal configurations. As a consequence, these systems exhibit non trivial thermodynamic and dynamic properties, different and richer than those observed in their non disordered counterpart.

Spin glasses can be modeled using Ising-like Hamiltonians where the bonds between spins can be positive or negative at random. Due to the heterogeneity of the couplings, there are many triples or loops of spin sequences which are frustrated, that is for which there is no way of choosing the orientations of the spins without frustrating at least one bond. As a consequence, even the best possible arrangement of the spins comprises for a large proportion of frustrated bonds. More importantly, since there are many configurations with similar degree of frustration one may expect the existence of many local minimal of the free energy[56]
CHAPTER 4

4 mathematical technique to express DMS

4.4.1 Green's function approach to the RKKY model

We use the standard spin RKKY Hamiltonian

\[
H = -\frac{1}{2} \sum J_{ij} \hat{S}_i \hat{S}_j ,
\]

(4.52)

where \( J_{ij} \) is the RKKY effective spin exchange interaction between local spin \( \hat{S}_i \) and \( \hat{S}_j \) (\( \hat{S}_i \) and \( \hat{S}_j \) are both quantum spin operators), formula

\[
J_{ij} = -J_{pd} m^* \frac{k_F^3}{2\pi^3 \hbar^2} F(2K_F R_{ij})
\]

(4.53)

where \( F(x) \) is an oscillating function defined by The oscillatory nature of the RKKY Hamiltonian thus allows a coexistence of ferromagnetic and antiferromagnetic interactions in the system. [13]

\[
F(x) = \frac{x \cos|x| - \sin|x|}{x^4}
\]

The coupling constant \( J_{pd} \) in eq.(4.53) is the local coupling of the itinerant spin to the local magnetic moment, \( m^* \) is the effective mass of the carrier, \( k_F \) is the Fermi wave vector \((K_F = 3 \pi^2 p)\) under free electron gas approximation) \( R_{ij} \) is the distance between local magnetic moments at Ri and Rj. Here, the magnetic atoms are distributed randomly in the full lattice of the undoped semiconductor. The prime for the summation in eq.(4.52) means that the summation is done only over the sites occupied by magnetic impurities. Hence, we use Green’s function approach[25] to treat the model eq.(1). In this scheme one uses a double-time Green’s function

\[ \ll \hat{A}(t); \hat{B}(t^\prime) \gg \]

which satisfies the following equation of motion:

\[
i \hbar \frac{d}{dt} \ll \hat{A}(t); \hat{B}(t^\prime) \gg = \delta(t-t^\prime) \ll [\hat{A}(t), \hat{B}(t^\prime)] + \ll [\hat{A}(t), \hat{H}]; \hat{B}(t^\prime) \gg \]

(4.54)

This approach proves successful for various Heisenberg spin models on regular lattices. Usually,
one needs more than one Green’s function when spin $S$ is larger than $1/2$. For our random spin system, we construct double-time spin Green’s functions between $1/2$ spin operators at two magnetic sites $R_i$ and $R_j$.

$$G_{ij}^{n}(t,t') = \langle \hat{S}_i^+(t); (\hat{S}_j^-(t'))^n (\hat{S}_j^+(t'))^{n-1} \rangle$$  \hspace{1cm} (4.55)

where $n$ takes 1, 2, 3, ... , 2$S$. As a rule, we need $2S$ independent Green’s functions for spin $S$. They can be expressed as

$$G_{ij}^{n}(t,t') = \frac{1}{2\pi \hbar} \int G_{ij}^n(\omega) e^{-\frac{i\omega(t-t')}{\hbar}} d\omega$$

### 4.1.1 The Green’s Function Formalism

The Green functions play the most important part in the field theoretical treatment of the many-body problem, and are especially useful for summing over the restricted classes of perturbation theory of diagrams, and also are very powerful when combined with spectral representations. Also they are flexible enough to describe the effects of retarded interactions and all quantities of physical interest like thermodynamic properties can be derived from them. There are different types of Green functions, or propagators: one particle, two-particle... $n$ particles, advanced, retard, The Curie temperature itself is a critical point, where the magnetic susceptibility is theoretically infinite and, although there is no net magnetization, domain-like spin correlations fluctuate at all length scales. $d$, zero temperature, finite temperature, real-time, imaginary time [40],

$$G_{kk}(t-t') = \langle b_k(t); b_k^+(t') \rangle = -i\theta(t-t') - \langle b_k(t), b_k^+(t') \rangle$$  \hspace{1cm} (4.1)

denotes a Havi side step function, and given by [41]

$$\theta(t-t') = \begin{cases} 1 & \text{if } t > t' \\ 0 & \text{if } t < t' \end{cases}$$  \hspace{1cm} (4.2)

$$G_{kk}(t-t') = \langle b_k(t), b_k^+(t') \rangle = \langle b_k(t), b_k^+(t') \rangle$$  \hspace{1cm} (4.3)

In order to obtain equation of motion we differentiate equ. (4.1) With respect to $t$ and multiplying both sides of 4.3 by $i$

$$i \frac{d}{dt} G_{kk}(t,t') = i \frac{d}{dt} \langle b_k(t), b_k^+(t') \rangle = \delta(t-t') \langle b_k(t), b_k^+(t') \rangle + \langle b_k(t), H, b_k^+(t') \rangle$$  \hspace{1cm} (4.4)

To solve equ. (4.4) it is convenient to work with the Fourier transformation of this equation.

Now let $G_{kk}(\omega)$ be the Fourier transform of $G_{kk}(t-t')$ such that
\[ G_{kk'}(t-t') = \int G_{kk'}(\omega) e^{-i\omega|t-t'|} d\omega \] (4.5)

and

\[ G_{kk}(\omega) = \int G_{kk'}(t-t') e^{-i\omega|t-t'|} d|t-t'| \] (4.6)

In addition, the delta function can be defined as

\[ \delta(t-t') = \int e^{-i\omega(t-t')} d\omega \] ................................................(4.7)

Using equation (4.5), (4.6) and (4.7) in equation 4.4 we have got,

\[ \omega G_{kk'}(\omega) = \langle b_k(t), b_{k'}(t') \rangle > \omega + \ll \langle b_k(t), H \rangle ; b_{k'}(t') \gg \omega \] (4.8)

where

\[ i \frac{d}{dt} b_k(t) = [b_k(t), H] \] and \( H = \langle H_{\text{magnon}} \rangle = \omega_k \langle b_k^+ ; b_k \rangle \)

we will write \( b_k \) dropping \( t \)

where \( \ll \cdots \gg \) is the abbreviated notation for the Fourier transform of the corresponding Green function, and \( \ll \cdots \gg \) denotes averaging over a grand canonical ensemble[24]. The commuting and anti commuting relation for the two operators is also given by

\[ [b_k b_{k'}] = b_k b_{k'} - \eta b_k b_{k'} \]

where \( \eta = 1 \) for Boson operators, and \( \eta = -1 \) for fermions operators. That is

\[ [b_k, b_{k'}^+] = b_{k^0} b_{k'^0} - b_{k^0}^+ b_{k'^0} \] (4.9)

For two boson operators and

\[ b_{k^0} b_{k'^0} = 0 \] (4.10)

for two fermions operator

\[ b_{k^0} b_{k'^0} = b_{k^0} b_{k'^0}^+ + b_{k'^0}^+ b_{k^0} = \delta_{kk'} \delta_{\alpha\alpha'} \] (4.11)

\[ \{ b_{k^0}, b_{k'^0} \} = \{ b_{k^0}^+, b_{k'^0}^+ \} = 0 \] (4.12)

the correlation function \( \langle b_{k'}(t), b_k(t) \rangle \) is related to the analytic property of Green’s function by
Taking the Fourier transformation (2.6), we get

\[
\omega G_{kk} = \frac{\delta_{kk}}{2\pi} + \ll [b_k, H], b_k^\dagger \gg \omega
\]  

(4.13)

\[
[ b_k, H ]
\]

is the Commutation relation of \( b_k \) with the magnon Hamiltonian, \( H \) magnon where

\[
[ b_k, H_{magnon} ] = \omega_k b_k \quad \text{and for } k=k' \quad \delta_{kk'} = 1 \quad \text{these gives}
\]

\[
\ldots \omega G_{kk}(\omega) = \frac{1}{2\pi} + \ll \omega_k b_k, b_k^\dagger \gg \omega
\]  

(4.14)

where

\[
\ll \omega_k b_k, b_k^\dagger \gg \omega = \omega_k \ll b_k, b_k^\dagger \gg = \omega_k G_{kk}(\omega)
\]

from which we obtain

\[
(\omega = \omega_k) G_{kk}(\omega) = \frac{1}{2\pi}, \quad G_{kk}(\omega) = \frac{1}{2\pi(\omega - \omega_k)}
\]  

(4.15)

Poles of the Green function will be given by \( \omega - \omega_k \) from which we find dispersion. Again considering equ (4.13) and taking \( t = t' \) equal time correlation gives the number operator;

\[
\ll b_k, b_k^\dagger \gg + i \delta = \frac{1}{2\pi(\omega + i \delta) - \omega_k} = \frac{1}{2\omega} \left[ \frac{p}{(\omega - \omega_k)} - i\delta \omega(\omega - \omega_k) \right]
\]  

(4.16)

\[
\ll b_k, b_k^\dagger \gg - i \delta = \frac{1}{2\pi(\omega - i \delta) - \omega_k} = \frac{1}{2\omega} \left[ \frac{p}{(\omega - \omega_k)} + i\delta \omega(\omega - \omega_k) \right]
\]  

(4.17)

Where \( p \) is the principal part of the integral …finally,

\[
\ll b_k b_k \gg = \lim_{\delta \to 0} i \int \delta(\omega - \omega_\delta) \frac{d\omega}{e^{\beta\omega} - 1}
\]

this can be expressed as
\[ \bar{n}_k = \frac{1}{e^{\beta \omega} - 1} \]  

(4.18)

where \( \bar{n}_k = \langle b_k^+ b_k \rangle \) is the number of magnons in state \( k \).

### 4.2 Magnon Distributions Function

A magnon is a quantized spin wave[11]. The ground state of a simple ferromagnet has all spins parallel. At long wave lengths \( ka << 1 \) so that, the frequency of magnon \( \omega_k \) is proportional to \( k^2 \); in the same limit the frequency of a phonon is proportional to \( k \). The quantization of spin waves proceeds exactly as for photons and phonons. The energy of a mode of frequency \( \omega_k \) with \( \bar{n}_k \) magnons can also be equated to,

\[ E_k = (n + \frac{1}{2}) \hbar \omega_k \]  

(4.19)

The excitation of magnon of corresponds to the reversal of one spin \( \frac{1}{2} \) eqn. (4.18) is the Bose-Einstein distribution that magnons also obey. The total number of magnons in all modes excited at temperature \( T \) can be calculated as:

\[ \sum_k \bar{n}_k = \int D(\omega) n(\omega) d\omega \]  

(4.20)

Where \( D(\omega) \) is the number of magnon modes per unit frequency range. The integral is taken over the allowed range of \( k \), which is the …first Brillouin zone. At sufficiently low temperature we may carry the integral between 0 and \( \infty \) because \( n(\omega) \rightarrow 0 \) exponentially as \( \omega \rightarrow \infty \). Magnons have a single polarization for each value of \( k \) [33]. In three dimensions the number of modes of wave vector less than \( k \) is \( \left( \frac{1}{2\Pi} \right)^3 \left( \frac{4\Pi k^2}{3} \right) \) per-unit volume, whence the number of magnons with frequency in \( d\omega \) at \( \omega \) is

\[ \left( \frac{1}{2\Pi} \right)^3 \left( \frac{4\Pi k^2}{3} \right) \left( \frac{dk}{d\omega} \right) \]  

(4.21)

Under these assumptions, the magnon part of the Hamiltonian can simply be written like a harmonic oscillator or phonon type Hamiltonian:

\[ \langle H_{\text{magnon}} \rangle = \sum_k \bar{n}_k \omega_k \]  

(4.22)

where is the average number of magnons in state \( k \) and \( \omega_k = 2\times J_S a^2 k^2 \) is the long wave length.
magnon dispersion. Substituting equation \( \omega_k \) into equation (4.22) for \( \omega_k \) we get

\[
< b_k^+ b_k > = \frac{1}{e^{2xJSa^2/k_BT}} - 1
\]  \hspace{1cm} (4.23)

\[
\sum_k < \tilde{n}_k > = \frac{1}{2\pi} \int_0^\infty \frac{4\pi k^2 dk}{e^{2xJSa^2/k_BT} - 1}
\]  \hspace{1cm} (4.24)

\[
\sum_k < \tilde{n}_k > = \frac{1}{2\pi^2} \int_0^\infty \frac{k^2 dk}{e^{2xJSa^2/k_BT} - 1}
\]  \hspace{1cm} (4.25)

let \( y = \frac{2xJSa^2k_B^2}{k_BT} \) and \( dy = 4xJSa^2k_B^2dk \) solving \( k \)

\[
k = \left( \frac{K_B T}{2X JS a^2} \right)^{\frac{1}{2}} y^{\frac{1}{2}}
\]  \hspace{1cm} (4.26)

and

\[
k^2 dk = \left( \frac{K_B T}{2X JS a^2} \right)^{\frac{3}{2}} y^{\frac{1}{2}}\frac{1}{2}
\]  \hspace{1cm} (4.27)

substituting equation (4.27) into equ. (4.25) we get

\[
\sum_k \tilde{n}_k = \frac{1}{2\pi^2} \int_0^\infty \left( \frac{k_B T}{2X JS a^2} \right)^{\frac{3}{2}} \int_0^\infty \frac{y^{\frac{1}{2}}}{e^y - 1} \frac{1}{2}
\]  \hspace{1cm} (4.28)

where the integration \( \int_0^\infty \frac{1}{e^y - 1} = 2.3174 \)

hence

\[
\sum_k \tilde{n}_k = (2.3174) \left( \frac{1}{4\pi^2} \right) \left( \frac{k_B T}{2xJSa^2} \right)^{\frac{3}{2}}
\]  \hspace{1cm} (4.29)

\[
\sum_k \tilde{n}_k = (0.0587) \left( \frac{K_B T}{2x JS a^2} \right)^{\frac{3}{2}}
\]  \hspace{1cm} (4.30)

Where equ. (4.30) gives us the number of reversed spins given by the ensemble average of the spin wave occupancy numbers. The average magnons excitation energy at low temperature is given by:

\[
\langle H_{magnon} \rangle = \sum_k \langle n_k \rangle \omega_k
\]  \hspace{1cm} (4.31)
4.3 Magnon Heat capacity in Ferromagnetism

Magnons are another important type of energy excitation and they occur in magnetically ordered solids [15] the internal energy of unit volume of magnon gas in thermal equilibrium at a temperature $T_i$ neglecting magnon magnon interaction, at very low external field, and considering $ka << 1$ is given by:

$$U = \sum_k \omega_k \langle \bar{n}_k \rangle_T = \sum_k \frac{\omega_k}{e^{\beta \omega} - 1}$$  \hspace{1cm} (4.32)

Taking $\omega_k = 2xJSa^2 k^2$  \hspace{1cm} (4.34)

$$U = \frac{1}{(2\pi)^3} \int_0^\infty \omega_k \frac{4 \omega k^2 dk}{e^{\frac{2xJSa^2 k^2}{k_BT}} - 1}$$  \hspace{1cm} (4.35)

Again from equ.(3.49) $d \omega_k = 2xJSa^2 (2k) dk$ and $dk^2 = \frac{\omega_k}{2xJSa^2}$ we get

$$dk^2 = (\frac{1}{8xJSa^2}) \omega_k^2 d \omega_k$$  \hspace{1cm} (4.36)

$$U = \frac{1}{(2xJSa^2)^\frac{3}{2}} \frac{1}{4\pi^2} \int_0^\infty \frac{\omega_k^\frac{3}{2}}{e^{\frac{2xJSa^2 k^2}{k_BT}} - 1}$$

Let $y = \frac{\omega_k}{k_BT}$ and

$$d \frac{\omega_k}{k_BT} = dy \Rightarrow dy \frac{k_BT}{d \omega_k} = d \omega_k \text{ and } k_BT = \frac{d \omega_k}{dy}$$  \hspace{1cm} (4.37)

where $\frac{1}{4\pi^2} \int_0^\infty \frac{y^{\frac{3}{2}} dy}{e^y - 1} = 0.0456$ then

$$U = 0.0456(\frac{1}{2xJSa^2})^\frac{3}{2} k_BT^\frac{5}{2} \hspace{1cm} (4.38)$$

Specific heat capacity of magnons will be calculated as:

$$C_{\text{magnon}} = \frac{\partial U}{\partial T} = \frac{\partial}{\partial T} \sum_k \omega_k \langle \bar{n}_k \rangle_T$$  \hspace{1cm} (4.39)
\[
C^{\text{magnon}} = \frac{\partial}{\partial T} [0.0456 \left( \frac{1}{2xJSa^2} \right)^{3/2} k_B^{5/2} T^{5/2}]
\] (4.40)

\[
C^{\text{magnon}} = 0.113 \left( \frac{1}{2xJSa^2} \right)^{3/2} k_B^{5/2} T^{3/2}
\] (4.41)

This shows that \(C^{\text{magnon}} \sim T^{3/2}\)

Fig 4.1 Heat capacity per unit volume VS temperature when values of x are kept constant

**4.4 Magnon Heat capacity in Antiferromagnetism**

The internal magnon energy for antiferromagnets can be written as

\[
\omega_k = 4\sqrt{3} Jkx\alpha
\]

\[
U = \sum_k \omega_k \langle n_k \rangle
\] (4.42)

Which can be written as

\[
U = \sum_k \frac{\omega_k}{\exp(\omega_k \beta) - 1} = \frac{1}{(2\pi)^3} \int_0^{k_{\text{max}}} \omega_k \frac{d^3k}{\exp(\omega_k \beta) - 1}
\] (4.43)

\[
U = \frac{1}{(2\pi)^3} \int_0^{k_{\text{max}}} \omega_k 4\pi k^2 \frac{dk}{\exp(\omega_k \beta) - 1} = \frac{1}{2\pi^2} \int_0^{k_{\text{max}}} \omega_k k^2 \frac{dk}{\exp(\omega_k \beta) - 1}
\] (4.44)
\[ U = \frac{1}{2 \pi^2} \int_0^{k_{\text{max}}} 4 \sqrt{3} J x a k^3 \frac{dk}{\exp \left( \frac{4 \sqrt{3} JS K x a}{k_B T} \right) - 1} \]  

(4.45)

The upper limit can be taken as infinity for temperatures small compared with the Neel temperature. Thus

\[ U = \frac{1}{2 \pi^2} \int_0^{\infty} 4 \sqrt{3} J S x k^3 \frac{dk}{\exp \left( \frac{4 \sqrt{3} JS K x a}{K_B T} \right) - 1} = \frac{4 \sqrt{3} JS x a}{2 \pi^2} \int_0^{\infty} k^3 \frac{dk}{\exp \left( \frac{4 \sqrt{3} JS K x a}{k_B T} \right) - 1} \]  

(4.46)

let \( y = \frac{4 \sqrt{3} JS K x a}{K_B T} \) and \( dy = \frac{4 \sqrt{3} JS x a dk}{K_B T} \)

substituting the above equation we get

\[ U = \frac{K_B T}{2 \pi^2} \left( \frac{T}{4 \sqrt{3} JS x a} \right)^3 \int_0^{\infty} y^3 \frac{dy}{e^y - 1} \]  

(4.47)

but the value of the integral

\[ \int_0^{\infty} \frac{y^3}{e^y - 1} = \frac{\pi^4}{15} \]

there for the internal magnon energy for anti ferromagnet is

\[ U = \frac{\pi^2}{30} \frac{(K_B T)^4}{(4 \sqrt{3} JS x a)^3} \]  

(4.48)

\[ c_{\text{magnon}} = \frac{\partial U}{\partial T} = \frac{4 \pi^2}{30} \frac{K_B (k_B T)^3}{(4 \sqrt{3} JS x a)^3} \]  

(4.49)

which is the magnon specific heat capacity for anti ferromagnetc case follows

**Fig4.2** Heat capacity per unit volume VS temperature when values of x are kept constant
the anti ferromagnetic magnon specific capacity per unit volume versus temperature $T$ for GaMnAs will be shown in this work with the temperature range of (0k-50k) to derive the specific heat we should neglect magnon -magnon interaction

4.5 FERROMAGNETIC TRANSISTION TEMPERATURE

Ferromagnetism refers to solids that are magnetized without an applied magnetic field. These solids are said to be spontaneously magnetized. Ferromagnetism occurs when paramagnetic ions in a solid “lock” together in such a way that their magnetic moments all point (on the average) in the same direction. At high enough temperatures, this “locking” breaks down and ferromagnetic materials become paramagnetic. The temperature at which this transition occurs is called the Curie temperature. In the ferromagnetic state at low temperatures, the spins on the various atoms are aligned parallel. There are several other types of ordered magnetic structures. These structures order for the same physical reason that ferromagnetic structures do. The Weiss theory is a mean field theory and is perhaps the simplest way of discussing the appearance of the ferromagnetic state. Thus the basic equation for ferromagnetic materials is

$$M = NSg \mu_B B_s(a') \quad \text{.................................................................} \quad (4.50)$$

$\text{where} \quad a' = \frac{\mu_o S g \mu_B (H + \xi) M}{K T} \quad \text{.................................................................} \quad (4.51)$

That is, the basic equations of the molecular field theory are the same as the paramagnetic case plus the $H \rightarrow H + \gamma M$ replacement in which For the case of a simple paramagnet

$$M = NSg \mu_B B_s(a)$$

where $B_s$ is defined by it is easy to recover the high-temperature results of $B_s$

$$B_s(y) = \frac{s + 1}{3s} y \quad \text{........ if } y \ll 1 \quad \text{where}$$

$$a = \frac{\mu_o S g \mu_B H}{K T}$$

Let us now indicate how this predicts a phase transition. By a phase transition, we mean that spontaneous magnetization ($M \neq 0$ with $H = 0$) will occur for all temperatures below a certain temperature $T_c$ called the ferromagnetic Curie temperature.

At the Curie temperature, for a consistent solution of (1.15) and (1.16) we require that the following two equations shall be identical as $a' \rightarrow 0$ and $H = 0$:
\[ M_1 = NSg \mu_B B_s(a') \]  
\[ M_2 = \frac{KT a'}{sg \mu_B \gamma \mu_o} \quad \text{with} \quad H \to 0 \]

where \( \mu_b = \frac{e \hbar}{2m} \) is the Bohr magneton, and \( g \) is sometimes called simply the g-factor, \( N \) is the number of particles per unit volume. If these (eq 1.17 and eq 1.18) are identical, then they must have the same slope as \( a' \to 0 \).

That is, we require

\[ \left( \frac{dM_1}{da'} \right)_{a' \to 0} = \left( \frac{dM_2}{da'} \right)_{a' \to 0} \]  
(4.54)

Using the known behavior of \( B_s(a') \) as \( a' \to 0 \) we find that condition (1.19) gives

\[ T_c = \frac{\mu_0 N g^2 s(s+1)\mu_b^2 \gamma}{3K} \]  
(4.55)

Equation (1.20) provides the relationship between the Curie constant \( T_c \) and the Weiss molecular field constant \( \gamma \). Note that, as expected, if \( \gamma = 0 \), then \( T_c = 0 \) (i.e. if \( \gamma \to 0 \), there is no phase transition). Further, numerical evaluation shows that if \( T > T_c \), (7.21) and (7.22) with \( H = 0 \) have a common solution for \( M \) only if \( M = 0 \). However, for \( T < T_c \), numerical evaluation shows that they have a common solution \( M \neq 0 \), corresponding to the spontaneous magnetization that occurs when the molecular field overwhels thermal effects. [15] The Curie temperature itself is a critical point, where the magnetic susceptibility is theoretically infinite and, although there is no net magnetization, domain-like spin correlations fluctuate at all length scales.

The study of ferromagnetic phase transitions, especially via the simplified Ising spin model, had an important impact on the development of statistical physics. There, it was first clearly shown that mean field theory approaches failed to predict the correct behavior at the critical point (which was found to fall under a universality class that includes many other systems, such as liquid-gas transitions), and had to be replaced by renormalization group theory.[10]

When we vary the external magnetic field \( H \) acting on solid its change of magnetization \( M \) as a function of temperature \( T \) hence the susceptibility \( \chi(T) \) is given by

\[ \chi(T) = \lim_{H \to 0} \left( \frac{\partial M}{\partial H} \right) \]  
thus the magnetic susceptibility of a solid \( \chi(T) \) is measured and it is used to categorize different kinds of solid like ferromagnetism in which all magnetic moments are in parallel alignment, the effective ensures the effect of ordering of the system experienced by
each atomic site and there for is given by $H_{\text{eff}} = H + \lambda M$

where $\lambda > 0$ is a constant that parameterize the strength of molecular field as a function of the magnetization. Then the mean magnetic moment or magnetization for $N$ atoms is given by

$$M = N \sum_i \mu_i p_i = N \frac{\sum_{-s} (-m_s g \mu_B) e^{-\beta m_s g H_{\text{eff}}}}{\sum_{-s} e^{-\beta m_s g H_{\text{eff}}}}$$  \tag{4.56}

$N g \mu_B B_s(x)$ where $B_s(x)$ is the Brillouin function given by the expression

$$B_s(x) = \frac{1}{S}[(S + \frac{1}{2}) \coth((S + \frac{1}{2}) x) - \frac{1}{2} \coth \frac{x}{2}]$$  \tag{4.57}

and $x = \frac{g \mu_B H_{\text{eff}}}{K_B T}$ and $g$ is the land factor which depends on the magnitude of $L$ and $S$ for $x \ll 1$

and using $H_{\text{eff}} = H + \lambda M$ we have a self-consistent equation for $M$ as

$$M = \frac{N g^2 \mu_B^2 S(S+1)(H+\lambda M)}{3 K_B T}$$  \tag{4.58}

equation can be written as

$$M[1 - \frac{C \lambda}{T}] = \left(\frac{C}{T}\right) H$$  \tag{4.59}

but $M = \chi H$, so that the magnetic susceptibility is given

$$\chi = \frac{C}{T - C \lambda}$$  \tag{4.60}

and obtaining the curie Weiss law

$$\chi = \frac{C}{T - T_c}$$ which is valid for $T \gg T_c$ and the constant $C \lambda$ which is independent of $T$ and the curie temperature is given by $T_c = C \lambda$

the curie temperature $T_c$ is the temperature above which the spontaneous magnetization vanishes. It separates the disorder paramagnetic phase at $T > T_c$ from the ordered ferromagnetic phase at $T < T_c$. The mean field constant

$$\lambda = \frac{T_c}{C} = \frac{3 K_B T_c}{N g^2 S(S+1) \mu_B^2}$$  \tag{4.61}

where $C$ is the curie constant

the high temperature susceptibility is generally taken as is evidence for paramagnetic spins, thus
\( \chi \) becomes infinite when \( T \rightarrow T_c \) at the curie temperature where the substance GaMnAs becomes ferromagnetic and the magnetic susceptibility increases with transition temperature decreases then this equation will becomes

\[
\chi = \frac{C}{T - T_c}
\]

\[
\frac{1}{\chi} \text{ (inverse susceptibility)}
\]

![Graph showing the reciprocal of susceptibility versus temperature in (K)](image)

*Fig 4.3 the reciprocal of susceptibility versus temperature in (K)*

the curve is for some slight departure at high temperature looks a straight line and then the material becomes ferromagnetic below 173k magnetic susceptibility infinite when \( T \rightarrow T_c \) at the curie temperature where the substance becomes ferromagnetic also can see from the graph that

\[
\frac{1}{\chi} = T - T_c = 0 \quad \text{at the point } \quad T = T_c
\]

let's summarizes the general trends of magnetic properties for solid-state materials. the magnetic susceptibility is the degree of magnetization of a material, \( M \) is the magnetic dipole moment per unit volume in response to a magnetic field \( H \), as

\[
\chi(T) = \lim_{H \to 0} \left( \frac{\partial M}{\partial H} \right)
\]

If \( \chi_m \) is positive, the material is called paramagnetic and the magnetic field is strengthened by the presence of the material. If \( \chi_m \) is negative, then the material is diamagnetic and the magnetic field is weakened in the presence of the material. Figure 1 displays the temperature dependence of the magnetic susceptibility of paramagnetic, ferromagnetic, and anti ferromagnetic solids [57]
Paramagnetic materials attract and repel like normal magnets when subjected to a magnetic field. For thermal equilibrium, the magnetization of paramagnets is treated using the Brillouin formalism. Under relatively low magnetic field saturation when the majority of atomic dipoles are not aligned with the field, paramagnetic materials exhibit magnetization according to the well known Curie–Weiss law, which treats the interaction between spins and molecular field

\[ \chi_m = \frac{C_p}{T} \]

where \( C_p \) is the curie constant, the Weiss constant \( \theta_p \) moments below the Curie or Neel temperature for uncorrelated spins.

**4.6 Magnetization Reversal for Ferromagnetism**

the number of reversal spins is given by the ensemble average of spin wave occupancy number thus for saturation magnetization with the same assumption for the heat capacity and taking unit volume the magnetization M(T) as a function of temperature can be computed by noting that each spin wave mode leads to one spin wave reversal thus the spontaneous magnetization at temperature T is given by

\[ M_s = 2 \mu_o S_z = 2 \mu_o (NS - \sum b_k^+ b_k) \]

(O.65)

and
\[ M_s(0) - M_s(T) = M = 2 \mu_0 \sum_K \langle n_k \rangle = \frac{2 \mu_0}{(2\pi)^3} \int dk^3 \frac{1}{e^{\frac{Dk^2}{T}} - 1} \]  
\[ (4.66) \]

where \( M(0) = g \mu_B Ns \) is the saturation magnetization.

where \( D = 2JSx^2 \) at low temperature such that \( DK_{\text{max}}^2 \gg \tau \)

\[ \Delta M = \frac{\mu_0}{2} \pi^2 \left( \frac{T}{D} \right)^{\frac{3}{2}} \int_0^\infty dx \frac{e^{\frac{a}{x}} - 1}{e^{\frac{a}{x}} - 1} \]  
\[ (4.67) \]

where \[ \int_0^\infty dx \frac{e^{\frac{a}{x}} - 1}{e^{\frac{a}{x}} - 1} = \Gamma \left( \frac{3}{2} \right) \xi \left( \frac{3}{2}, 1 \right) \]
therefore

\[ \Delta M = 0.117 \mu_0 \left( \frac{K_B T}{D} \right)^{\frac{3}{2}} \]  
\[ (4.68) \]

thus the reduction in magnetization due to thermal spin wave excitation is

\[ \Delta M = 0.117 \frac{\mu_0}{2a^3} \left( \frac{K_B T}{2JSa} \right)^{\frac{3}{2}} \]  
\[ (4.69) \]

we can use the relation of

\[ M(T) = M(0) \left( 1 - \frac{1}{ns} \right) (0.0587) \left( \frac{K_B T}{2XJSa} \right)^{\frac{3}{2}} \]  
\[ (4.70) \]

\[ \frac{M(T)}{M(0)} = 1 - \left( 0.0587 \right) \left( \frac{K_B T}{2XJSa} \right)^{\frac{3}{2}} \]  
\[ (4.71) \]

where \( N \) is the number of atoms per unit volume \( N = na^3 \) the reduced magnetization \( \frac{M(T)}{M(0)} \)
versus \( \frac{T}{T_c} \) (reduced temperature) graph drawn for \( X=0.08 \) and \( x=0.05 \) the magnetization decreases smoothly to zero at \( T = T_c \) this behavior classifies the usual ferromagnetic/paramagnetic using the above equation the equation reduce to

\[ \frac{M(T)}{M(0)} = 1 - \left( \frac{T}{T_c} \right)^{\frac{3}{2}} \]  
the value of \( T_c \) is equivalent to

\[ T_c = x \left( \frac{2JS}{K_B} \right) \left( \frac{na^3 s}{0.0587} \right)^{\frac{2}{3}} \]  
\[ (4.72) \]
Fig 4.5 The reduced magnetization versus reduced temperature (the spontaneous magnetization)

\[
\frac{M(T)}{M(0)} \rightarrow 0 \quad \text{then} \quad T \rightarrow T_c \quad \text{which is}
\]

\[
T_c = x \left( \frac{2J_s}{k_B} \right) \left( \frac{n a^3 s}{0.0587} \right)^{\frac{2}{3}}
\]

thus there exists the possibility of ferromagnet below a certain critical temperature \( T_c \) called the curie temperature which ids directly proportional to the concentration of the impurity atom \( T_c \approx x \)

Fig 4.6 Curie temperature, \( T_c \) versus concentration of the impurity semiconductor GaAs in GaMnAs
4.7 Magnetization Reversal for Anti-Ferromagnetism

the sub lattice magnetization at temperature $T$ is given by

$$M(T) = g \mu_B \sum_i (s - a_i^+ a_i)$$  \hspace{1cm} (4.73)$$

using the spin wave canonical transformation we get the following

$$M(T) = g \mu_B (n s - \sum_k \sinh^2 \theta_k - \sum_k \langle \alpha_k^+ \alpha \rangle \cosh 2 \theta_k)$$  \hspace{1cm} (4.74)$$

the second term of equation (4.74) gives zero point contribution to the sub lattice spin deviation. It can be shown. On neglecting the zeeman term, that

$$\Delta M_{zero\ point} = g \mu_B \sum_k \sinh^2 \theta_k = \frac{1}{2} g \mu_B \sum_k \left[ (1 - \gamma_k^2) \frac{1}{2} - 1 \right]$$  \hspace{1cm} (4.75)$$

for a simple cubic lattice this turns out to be $0.078 g \mu_B n$. the third term of equation (4.74) gives the temperature dependent part. Taking $M(O) = g \mu_B n s$ which is ground state magnetization and neglecting the second term of equation (4.74) we obtain

$$M(T) = M(0)[(1 - \frac{1}{ns} \sum_k \langle n_k \rangle \cosh 2 \theta_k)]$$  \hspace{1cm} (4.76)$$

$$M(T) = M(0)[(1 - \frac{1}{ns} \sum_k \exp(\frac{\omega_k}{k_B T}) - 1)]$$  \hspace{1cm} (4.77)$$

$$M(T) = M(0)[(1 - \frac{1}{ns} \sum_k \exp(\frac{\omega_k}{k_B T}) - 1)]$$  \hspace{1cm} (4.78)$$

But the value of $(1 - \gamma_k^2)^{\frac{1}{2}} = \frac{\sqrt{3}}{k a}$ hence

$$\frac{M(T)}{M(0)} = 1 - \frac{\sqrt{3}}{n s a} \sum_k \frac{1}{k \left[ \exp(\frac{\omega_k}{k_B T}) - 1 \right]}$$  \hspace{1cm} (4.79)$$

now we can use

$$\sum_k \frac{1}{k \left[ \exp(\frac{\omega_k}{k_B T}) - 1 \right]} = \frac{1}{2 \pi^3} \int_0^{k_{\omega}} d^3K \frac{\frac{d^3K}{k \left[ \exp(\frac{\omega_k}{k_B T}) - 1 \right]}}{k \left[ \exp(\frac{\omega_k}{k_B T}) - 1 \right]}$$  \hspace{1cm} (4.80)$$

The upper limit can be taken as infinity for temperatures small compared with the Neel temperature. Thus
\[
\sum_k \frac{1}{k \left[ \exp \left( \frac{\omega_k}{k_B T} \right) - 1 \right]} = \frac{1}{2 \pi^2} \int_0^\infty d^3 K \frac{\frac{d^3 K}{K \left[ \exp \left( \frac{\omega_k}{k_B T} \right) - 1 \right]} = \frac{1}{2 \pi^2} \int_0^\infty k dk \frac{\exp(\frac{4 \sqrt{3} J_saxk}{k_B T}) - 1}{\exp(4 \sqrt{3} J_saxk)} - 1}
(4.81)
\]

Let \( y = \frac{4 \sqrt{3} J_sax}{k_BT} \); \( K = \frac{K_B T y}{4 \sqrt{3} J_sax} \); \( dk = \frac{k_B T dy}{4 \sqrt{3} J_sax} \)

\[
\sum_k \frac{1}{k \left[ \exp \left( \frac{\omega_k}{k_B T} \right) - 1 \right]} = \frac{1}{2 \pi^2} \left( \frac{k_B T y}{4 \sqrt{3} J_sax} \right)^2 \int_0^\infty y dy \frac{y dy}{e^y - 1}
(4.82)
\]

the value of the integral \( \int_0^\infty \frac{y dy}{e^y - 1} = \frac{\pi^2}{k_B T} \)

Hence substituting the value of \( y \) in the equation (4.82) we will get

\[
\frac{M(T)}{M(0)} = 1 - \frac{\sqrt{3}}{12 NS} \left( \frac{K_B T}{4 \sqrt{3} J_sax} \right)^2
\]

which shows that the sub-lattice magnetization decreases quadratically with temperature in the low temperature region but when \( \frac{M(T)}{M(0)} \) go to zero, \( T \) approaches to \( T_N \) which gives

\[
T_N = (48)^{\frac{3}{2}} \left( \frac{J_s}{k_B} \right) (NS)^{\frac{1}{2}} x
\]

(4.83)

the dependence reduced magnetization due to thermal spin wave excitation on temperature as follows which shows that the sublattice magnetization decreases quadratically with temperature in the low temperature region and it agrees with the expression obtained. the reduced magnetization versus temperature curve as shown in the figure below below in such a case we can reduce the equation to in the form of

\[
\frac{M(T)}{M(0)} = 1 - \left( \frac{T}{T_N} \right)^2
\]

the figure curves are slightly different for different values of the concentration \( (x) \) of the manganese ion. however they all have a convex up ward shape
the dependence of neel temperature on concentration is observed for GaMnAs for the range (0.05-0.1) the figure below shows that the Neel temperature for GaMnAs increases linearly with increasing the concentration of manganese ion increases in the range given below from the above equation and taking the constants for GaMnAs $T_N \approx 731.3x$ thus for our calculation the highest neel temperature obtained $T=36.5k$ with in the concentration of 0.05 as shown in the figure below.

![Figure 4.8: Neel temperature versus concentration.](image)

the following table shows the Dispersion relation, magnetization and magnon specific heat for ferromagnet and antiferromagnet.

<table>
<thead>
<tr>
<th>Concentration (x)</th>
<th>Neel Temperature (T_N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.04</td>
<td>10</td>
</tr>
<tr>
<td>0.05</td>
<td>20</td>
</tr>
<tr>
<td>0.06</td>
<td>30</td>
</tr>
<tr>
<td>0.07</td>
<td>40</td>
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<td>0.08</td>
<td>50</td>
</tr>
<tr>
<td>0.09</td>
<td>60</td>
</tr>
<tr>
<td>0.1</td>
<td>70</td>
</tr>
<tr>
<td>0.11</td>
<td>80</td>
</tr>
<tr>
<td></td>
<td>Dispersion relation</td>
</tr>
<tr>
<td>----------------------</td>
<td>---------------------</td>
</tr>
<tr>
<td><strong>Ferromagnet</strong></td>
<td>$\omega = A_1 K^2$</td>
</tr>
<tr>
<td><strong>Antiferromagnet</strong></td>
<td>$\omega = A_2 K$</td>
</tr>
</tbody>
</table>

Table 4.1 summary of spin wave properties (at low energy and at low temperature)
CHAPTER 5

Conclusion

$Ga_{1-x}Mn_xAs$ can be considered as a model system for III-V diluted magnetic semiconductors, which are promising materials for future spintronics applications. In view of future applications, a proper characterization and understanding of the physical properties of $Ga_{1-x}Mn_xAs$ is essential, and techniques to increase the ferromagnetic transition temperature like post-growth annealing must be explored.

While dilute magnetic semiconductors could be a useful class of materials and assume a crucial role in enabling the semiconductor spintronic devices, much remains unanswered with regards to the mechanism of magnetism in these materials. Traditional III-V candidates such as $GaMnAs$, which are typically p-type, have been studied in great detail. Optical as well as electrical measurements have established carrier-mediated ferromagnetism in these materials. However, the highest $T_c$ obtained in these materials still remains under 200K, making them undesirable for technological applications and from magnon distribution function the total number of magnons in all modes excited at temperature T can be calculated as

$$\sum_k \bar{n}_k = (0.0587) \left( \frac{K_B T}{2x J S a^2} \right)^{\frac{3}{2}}$$

which follows $T^\frac{3}{2}$ relation and the Neel temperature for GaMnAs increases when the concentration of manganese ion increases in the range $0.05 \leq x \leq 0.1$. Hence $Ga_{1-x}Mn_xAs$ is found to be antiferromagnetism in the Neel temperature of $T_N = 36.5 - 73$ and thus for our calculation the highest neel temperature obtained $T=36.5k$ within the concentration of 0.05.

The heat capacity for antiferromagnets in the low temperature region arising from magnons is proportional to $T^3$ which is similar to the Debye phonon heat capacity where as the heat capacity of ferromagnets is proportional to $T^\frac{3}{2}$ and the specific heat decreases with concentration keeping the temperatuer constant $C_{\text{magnon}} \approx \frac{1}{x^3}$, the magnetization reversal follows the relation $\approx T^\frac{3}{2}$, $\approx T^2$ respectively for ferromagnetic and antiferromagnetic but vanishes as $T \rightarrow T_C$ for Ferromagnetism and $T \rightarrow T_N$ for antiferromagnetism.
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