THE STUDY OF ANTIFERROMAGNETISM IN DILUTED MAGNETIC SEMICONDUCTOR "CDMNTE"

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A THESIS PRESENTED TO
THE SCHOOL OF GRADUATE STUDIES
ADDIS ABABA UNIVERSITY
IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF

MASTER OF SCIENCE in PHYSICS

ADDIS ABABA, ETHIOPIA
JULY 2006

ADDIS ABABA UNIVERSITY

DEAPRMENT OF PHYSICS

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Department: **Deaprment of Physics**

Degree: **M.Sc.** Convocation: **July** Year: **2006**

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This work is dedicated to my father Tadesse Zerihun and my mother Asrate Muche

Table of Contents

Table of Contents **v**

List of Figures **vii**

Abstract **ix**

Introduction **1**

1 General introductions to semiconductors and magnetism **2**
1.1 Semiconductors 2
1.1.1 Introduction 2
1.1.2 Intrinsic semiconductors 3
1.1.3 Extrinsic semiconductors 6
1.1.4 n-type doping 6
1.1.5 p-type doping 7
1.2 Magnetism 8
1.2.1 Introduction 8
1.2.2 Diamagnetism 9
1.2.3 Paramagnetism 10
1.2.4 Curie's law 11
1.2.5 Ferromagnetism 12
1.2.6 Spontaneous magnetization 14
1.2.7 Antiferromagnetism 15
1.2.8 Ferrimagnetism 17
1.2.9 Spin glass 18
1.3 Antiferromagnetic spin waves 19
1.3.1 Exchange interaction 20

2 A review on the study of DMS **22**
2.1 Introduction 22
2.2 Alloy of II-VI semiconductors with magnetic materials 25
2.3 Alloys of III-V semiconductors with ferromagnetic properties 28
2.4 The magnetic elements for doping 29
2.5 Structure of CdTe 30

2.6 Synthesis of dilute magnetic semiconductors 31

3 Mathematical techniques for spin wave excitations 32
3.1 Formulation of the problem 32
3.2 Green's function formalism 40
3.3 Magnetization and transition temperature for antiferromagnets 44
3.4 Magnon heat capacity in antiferromagnets 45

4 Results and discussion 47
4.1 Dependence of magnetization on temperature 47
4.2 Dependence of Neel temperature on concentration 48
4.3 Dependence of specific heat on temperature 50

5 Conclusion 52

Bibliography 53

List of Figures

1.1 *Types of semiconductors: A) a magnetic semiconductor B) a diluted magnetic semiconductor C) a non-magnetic semiconductor* 3
1.2 *Band scheme for intrinsic conductivity in a semiconductor* 4
1.3 *a) a semiconductor doped with donor atom. b) a semiconductor doped with acceptor atoms* 7
1.4 *Magnetization VS magnetic field and susceptibility VS temperature* 10
1.5 *Magnetization VS magnetic field and susceptibility VS temperature* 12
1.6 *spin alignment for ferromagnets* 13
1.7 *Susceptibility VS temperature for ferromagnets* 14
1.8 *Spin alignment for antiferromagnetism* 15
1.9 *Susceptibility VS temperature for antiferromagnets* 16
1.10 *Spin alignment in canted antiferromagnetism* 17
1.11 *Spin alignment in ferrimagnetism* 17

2.1 *The zinc blende structure of CdTe* 30

4.1 *Reduced magnetization VS temperature when values of x are kept constant* 48
4.2 *Neel temperature versus concentration* 49
4.3 *Heat capacity per unit volume VS temperature when values of x are kept constant* 50

Acknowledgements

First of all, I would like to express my sincere thanks to my advisor and instructor professor P. Singh for his limitless and invaluable effort in guiding, assisting and supervising this work. His rich in knowledge and experience in the field, without which the thesis would not have the present shape, facilitated the progress of my work and motivated me to think of further research activities in this field. I appreciate him also for that he provided me almost all references and allowed me to use all his facilities with out any limit. To work with him is really a great pleasure.

I gratefully acknowledge the department of physics, AAU, for the assistance and the facilities received in the course of the research work ingeneral and my instructors in particular. I also wish to thank to all my friends who provided me continuing support and special encouragement at crucial moments.

In addition, I would like to express my heart felt thanks to my families specially Tiringo Tadesse for her constant encouragement and moral support during the process of completing the two years of study.

Finally my greatest thanks are forwarded to city government of Addis Ababa Education Bureau for sponsoring my education and technical support.

Gebru Tadesse

Addis Ababa University

June, 2006

Abstract

Most of the semiconductor materials are diamagnetic by nature and therefore cannot take active part in the operation of the magneto-electronic devices. In order to enable them to be useful for such devices efforts have been made to develop diluted magnetic semiconductors (DMS) in which small quantity of magnetic ion is introduced in to normal semiconductors. The first known such DMS are II-VI and III-V semiconductors diluted with magnetic ions like Mn, Fe, Co, Ni, etc. Most of these DMS exhibit very high electron and hole mobility and thus useful for high speed electronic devices.

CdMnTe systems with face centered cubic structure and long-range antiferromagnetic ordering is investigated with in the frame work of the Heisenberg model. The Hamiltonian includes exchange interactions with the nearest neighbors and Zeeman terms. Calculations are performed using Holstein-Primakoff transformation and Green's function formalism. Temperature dependencies of the spin-wave specific heat and reduced magnetization are determined. In addition to these linear concentration dependence of Neel temperature is also predicted.

The spin-wave energy shows T^4 temperature dependence, where as the predicted magnon part the heat capacity is proportional to T^3 which is similar to the Debye phonon heat capacity. The sublattice magnetization decreases quadratically with temperature in the low-temperature region. But the dependence of the Neel temperature on the manganese ion concentration is linear. All the findings are in broad agreement with experimental observations.

Introduction

Diluted magnetic semiconductors (DMS, known also as semi-magnetic semiconductors) are compounds based on typical semiconductors (like CdTe or InAs), for which a fraction of nonmagnetic cations has been replaced by magnetic ions (typically transition metal ions like Mn,Fe or rare earth metal ions) [1]. DMS bridge the physics of semiconductors and magnetics since they show typical semiconductor behavior and they also reveal pronounced magnetic properties. The most relevant feature of DMS, which attracted considerable interest, is the coexistence and interaction of two different electronic subsystems: delocalized conduction (s-type) and valence (p-type) band electrons and localized (d or f-type) electrons of magnetic ions [1]. In particular the s, p-d exchange interaction leads to strong band splitting, which results in giant magneto-optical effects. On the other hand the d-d exchange interaction, coupling randomly distributed magnetic ions, triggers formation of spin-glass and antiferromagnetic phases, depending on magnetic ion concentration and temperature range [2,3].

$Cd_{1-x}Mn_xTe$ alloys are the most thoroughly investigated diluted magnetic semiconductors [1]. It is known that the single phase zinc blende crystals of $Cd_{1-x}Mn_xTe$ can be obtained by equilibrium growth techniques up to $x \approx 0.7$. The recent diluted magnetic semiconductor materials reported are (CdMn)Te, (GaMn)As, (GaMn)Sb, ZnMn (or Co)O, TiMn(or Co)O, etc.

Chapter 1

General introductions to semiconductors and magnetism

1.1 Semiconductors

1.1.1 Introduction

A semiconductor is a material that is an insulator at very low temperature, but which has a sizable electrical conductivity at room temperature. The distinction between a semiconductor and an insulator is not very well-defined, but roughly, a semiconductor is an insulator with a band gap small enough that its conduction band is appreciably thermally populated at room temperature.

In semiconductors at 0k (and with out excitations) the uppermost band of occupied energy states is completely full. It is well-known from solid state physics that electrical conduction in solids occurs only via electrons in partially-filled bands, so conduction in pure semiconductors occurs only when electrons have been excited -thermally,optically,etc...in to higher unfilled bands.

At room temperature, a proportion (generally very small, but not negligible) of electrons in a semiconductor have been thermally excited from the valence band, the band filled at 0k, to the conduction band, the next higher band. The ease with which electrons can be excited from the valence band to the conduction band depends on the energy gap between the bands, and it is the size of this energy band gap that serves as an arbitrary dividing line between semiconductors and insulators.

When electrons are excited from the valence band to the conduction band in a semiconductor, both bands contribute to conduction, because electrical conduction can occur in any partially-filled energy band. The current carrying electrons in the conduction band are known as free electrons, though often they are simply called electrons. The free energy states in the valence band are known as holes. It can be shown that holes behave very much like positively-charged counter parts of electrons, and they are usually treated as if they are real charged particles. There are three types of semiconductors [4]:

¡) a magnetic semiconductor, in which a periodic array of magnetic element is present.

¡¡) a diluted magnetic semiconductor, an alloy between magnetic semiconductor and magnetic element.

¡¡¡) a nonmagnetic semiconductor, which contains no magnetic ions.

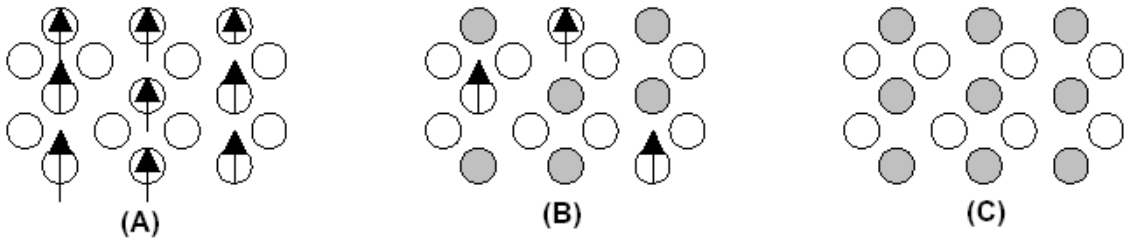

Figure 1.1: *Types of semiconductors: A) a magnetic semiconductor B) a diluted magnetic semiconductor C) a non-magnetic semiconductor* .

1.1.2 Intrinsic semiconductors

An intrinsic semiconductor is one which is pure enough that impurities do not appreciably affect its electrical behavior. In this case, all carriers are created by thermally or optically exciting electrons from the full valence band in to the empty conduction band. Thus equal numbers of electrons and holes are present in an intrinsic semiconductor. Electrons and holes flow in opposite directions in an electric field, though they contribute to current in the same direction since they are oppositely

charged. Hole current and electron current are not necessarily equal in an intrinsic semiconductor, however, because electrons and holes have different effective masses.

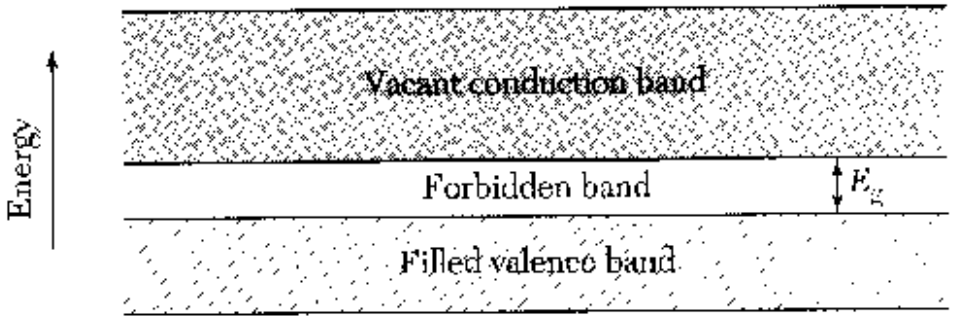

Figure 1.2: *Band scheme for intrinsic conductivity in a semiconductor* .

The concentration of carriers is strongly dependent on the temperature. At low temperatures, the valence band is completely full, making the material an insulator. The concentration of electrons in the conduction band at a temperature T is given by

$$n = \int_{E_c}^{\infty} D_e(\varepsilon) f_e(\varepsilon, T) d\varepsilon. \tag{1.1}$$

The Fermi Dirac distribution function is

$$f_e(\varepsilon) = \frac{1}{exp[(\varepsilon - \varepsilon_F)\beta] + 1} \tag{1.2}$$

where

$$\beta = \frac{1}{k_B T}.$$

This is the probability that a conduction electron orbital is occupied. The energy of

an electron in the conduction band is

$$\varepsilon_k = E_c + \frac{\hbar^2 k^2}{2m_e} \tag{1.3}$$

where E_c is the energy at the conduction band edge.
Thus the density of states at ε is

$$D_e(\varepsilon) = \frac{1}{2\pi^2}(\frac{2m_e}{\hbar^2})^{\frac{3}{2}}(\varepsilon - E_c)^{\frac{1}{2}}. \tag{1.4}$$

Hence the concentration of electrons in the conduction band is

$$n = 2(\frac{m_e k_B T}{2\pi\hbar^2})^{\frac{3}{2}} exp[(\varepsilon_F - E_c)\beta]. \tag{1.5}$$

The equilibrium concentration of holes (p) is calculated as follows. The distribution function for holes is related to the electron distribution function by $f_h = 1 - f_e$, because a hole is the absence of an electron.

$$f_h = \frac{1}{exp[(\varepsilon_F - \varepsilon)\beta] + 1}. \tag{1.6}$$

If the holes near the top of the valence band behave as particles with effective mass m_h, the density of hole states is given by

$$D_h(\varepsilon) = \frac{1}{2\pi^2}(\frac{2m_h}{\hbar^2})^{\frac{3}{2}}(E_v - \varepsilon)^{\frac{1}{2}} \tag{1.7}$$

where E_v is the energy at the valence band edge. Therefore the concentration of holes in the valence band is

$$p = \int_{-\infty}^{E_v} D_h(\varepsilon) f_h(\varepsilon, T) d\varepsilon \tag{1.8}$$

and

$$p = 2(\frac{m_h k_B T}{2\pi\hbar^2})^{\frac{3}{2}} exp[(E_v - \varepsilon_F)\beta]. \tag{1.9}$$

Multiplying together the expressions for n and p we get the equilibrium relation:

$$np = 4(\frac{k_B T}{2\pi\hbar^2})^3 (m_e m_h)^{\frac{3}{2}} exp(\frac{-E_g}{k_B T}) \tag{1.10}$$

where E_g is the energy gap which is given by

$$E_g = E_c - E_v.$$

In an intrinsic semiconductor the number of electrons is equal to the number of holes [5], because the thermal excitation of an electron leaves behind a hole in the valence band.
Thus from equation (1.10) we have

$$n_i = p_i = 2(\frac{k_B T}{2\pi\hbar^2})^{\frac{3}{2}}(m_e m_h)^{\frac{3}{4}} exp(\frac{-E_g}{2k_B T}). \tag{1.11}$$

Hence the intrinsic carrier depends exponentially on $\frac{E_g}{2k_B T}$.
To determine the fermi energy we set (1.5) equal to (1.9). From this we will get

$$\varepsilon_F = \frac{1}{2}E_g + \frac{3}{4}k_B T ln(\frac{m_h}{m_e}). \tag{1.12}$$

1.1.3 Extrinsic semiconductors

An extrinsic semiconductor is one that has been doped with impurities to modify the number and type of free charge carriers. One of the main reasons that semiconductors are useful in electronics is that their electronic properties can be greatly altered in a controllable way by adding small amounts of impurities [6]. The process of adding an impurity is called doping. These impurities, called dopants, add extra electrons or holes.

1.1.4 n-type doping

A semiconductor with extra electrons is called an n-type semiconductor. The purpose of n-type doping is to produce an abundance of carrier electrons in the material. Consider Si atoms which have four valence electrons, each of which is covalently bonded with one of four adjacent Si atoms. If an atom with five valence electrons, such as those from group V A of the periodic table (e.g phosphorus, arsenic, antimony) is incorporated in to the crystal lattice in place of a Si atom, then that atom will have four covalent bonds and one unbounded electron. This extra electron is only

weakly bound to the atom and can easily be excited in to the conduction band.

At normal temperatures, virtually all such electrons are excited in to the conduction band. Since excitation of these electrons does not result in the formation of a hole, the number of electrons in such a material far exceeds the number of holes. In this case the electrons are the majority carriers and the holes are the minority carriers. Because the five electron atoms have an extra electron to donate, they are called donor atoms.

1.1.5 p-type doping

A semiconductor with extra holes is called a p-type semiconductor [6]. The purpose of p-type doping is to create an abundance of holes. In the case of silicon a trivalent atom, such as boron, is substituted in to the crystal lattice. The result is that an electron is missing from one of the four possible covalent bonds. Thus the atom can accept an electron from the valence band to complete the fourth band, resulting in the formation of a hole. Such dopants are called acceptors. When a sufficiently large number of acceptors are added, the holes greatly out number the excited electrons. Thus, the holes are the majority carriers, while electrons are the minority carriers in p-type materials.

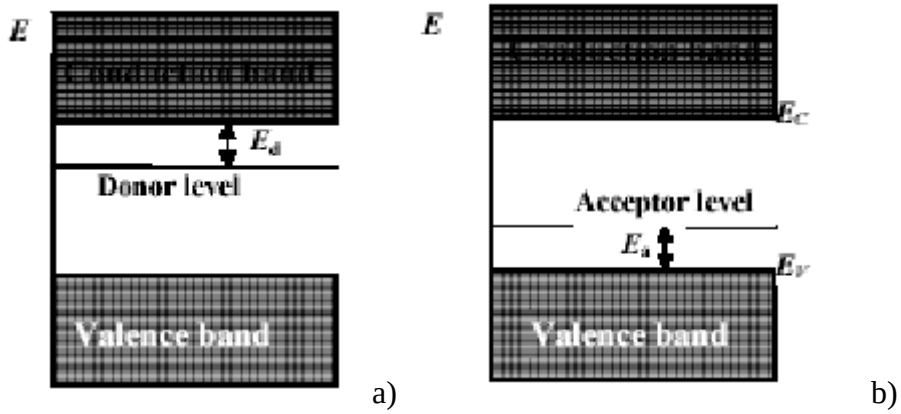

Figure 1.3: *a) a semiconductor doped with donor atom. b) a semiconductor doped with acceptor atoms .*

If there are no acceptors present, then the concentration of conduction electrons is

$$n \equiv (n_o N_d)^{\frac{1}{2}} exp(\frac{-E_d}{2k_B T}) \tag{1.13}$$

where $n_o \equiv 2(\frac{m_e k_B T}{2\pi\hbar^2})^{\frac{3}{2}}$ and N_d is the concentration of donors. Identical results hold for acceptors, under the assumption of no donor atoms.

$$p \equiv (p_o N_a)^{\frac{1}{2}} exp(\frac{-E_a}{2k_B T}) \tag{1.14}$$

where $p_o \equiv 2(\frac{m_h k_B T}{2\pi\hbar^2})^{\frac{3}{2}}$ and N_a is the concentration of acceptors.

1.2 Magnetism

1.2.1 Introduction

Magnetism is one of the phenomena by which materials exert an attractive or repulsive force on other materials. Magnetic forces are fundamental forces that arise from the movement of electrical charge. Thus, magnetism is seen whenever electrically charged particles are in motion. This can arise either from movement of electrons in an electric current, resulting in "electromagnetism", or from the quantum mechanical spin and orbital motion of electrons, resulting in what are known as "permanent magnets". Electron spin is the dominant effect with in atoms. The so called "orbital motion" of electrons around the nucleus is a secondary effect that slightly modifies the magnetic field created by spin.

The physical cause of the magnetism of objects, as distinct from electrical currents, is the atomic magnetic dipole. Magnetic dipoles, or magnetic moments, result on the atomic scale from the two kinds of movement of electrons. The first is the orbital motion of the electron around the nucleus, this motion can be considered as a current loop, resulting in an orbital dipole magnetic moment along the axis of the nucleus. The second, much stronger, source of electronic magnetic moment is due to a quantum mechanical property called the spin dipole magnetic moment.

The overall magnetic moment of the atom is the net sum of all of the magnetic moments of the individual electrons. Because of the tendency of magnetic dipoles

to oppose each other to reduce the net energy, in an atom the opposing magnetic moments of some pairs of electrons cancel each other, both in orbital motion and in spin magnetic moment. Thus, in the case of an atom with a completely filled electron shell or sub shell, the magnetic moments normally completely cancel each other out and only atoms with partially-filled electron shells have a magnetic moment, whose strength depends on the number of unpaired electrons.

The differences in configuration of the electrons in various elements thus determine the nature and magnitude of the atomic magnetic moments, which in turn determine the differing magnetic properties of various materials. Several forms of magnetic behavior have been observed in different materials, including diamagnetism, paramagnetism, ferromagnetism, ferrimagnetism, antiferromagnetism and spin glasses. Materials in the first two groups are those that exhibit no collective magnetic interactions and are not magnetically ordered. Materials in the next three groups exhibit long range magnetic order below a certain critical temperature.

1.2.2 Diamagnetism

Diamagnetism is a form of magnetism which is only exhibited by a substance in the presence of an externally applied magnetic field. It is the result of changes in the orbital motion of electrons due to the application of an externally applied magnetic field [7]. Applying a magnetic field creates a magnetic force on a moving electron in the form of $\overrightarrow{F} = q\overrightarrow{V}X\overrightarrow{B}$. This force changes the centripetal force on the electron, causing it to either speed up or slow down in its orbital motion. This changed electron speed modifies the magnetic moment of the orbital in a direction against the external field.

Diamagnetic materials include water, DNA, quartz(SiO_2), calcite($CaCO_3$), most organic compounds such as oil and plastic, many metals such as Gold and Bismuth and noble gases. Diamagnets are repelled by magnetic fields. Superconductors may be considered to be perfect diamagnets ($susceptibility(\chi) = -1$), since they expel all field from their interior due to the Meissner effect. Diamagnetic materials have a relative magnetic permeability that is less than one, and a magnetic susceptibility less than zero. Susceptibility is a measure of how magnetizable a substance can become in the presence of a magnetic field and can be used in a general way to describe the

various classes of magnetic materials [8].

Diamagnetism is a fundamental property of all matter, although it is usually very weak. It is due to the non-cooperative behavior of orbiting electrons when exposed to an applied magnetic field. Diamagnetic substances are composed of atoms which have no net magnetic moments (i.e., all the orbital shells are filled and there are no unpaired electrons). However, when exposed to a field, a negative magnetization is produced and thus the susceptibility is negative. In atoms with no permanent dipole moment, e.g., for filled electron shells, a weak dipole moment can be induced in a direction antiparallel to an applied field, an effect called diamagnetism. If we plot magnetization (M) versus magnetic field (H), we see:

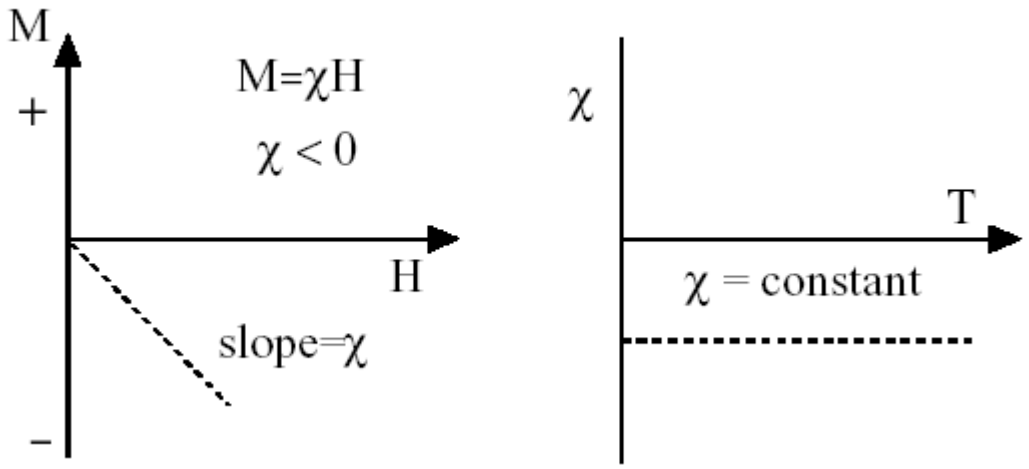

Figure 1.4: *Magnetization VS magnetic field and susceptibility VS temperature* .

Note that when the field is zero the magnetization is zero. The other characteristic behavior of diamagnetic materials is that the susceptibility is temperature independent.

1.2.3 Paramagnetism

Paramagnetism is the tendency of the atomic magnetic dipoles to align with an external magnetic field. This effect occurs due to quantum-mechanical spin as well as electron orbital angular momentum. Paramagnetic materials attract and repel

like normal magnets when subject to a magnetic field. This alignment of the atomic dipoles with the magnetic field tends to strengthen it and is described by a relative magnetic permeability greater than unity (or, equivalently, a small positive magnetic susceptibility).

Paramagnetism requires that the atoms individually have permanent dipole moments even with out an applied field [7], which typically implies a partially filled electron shell. In pure paramagnetism, these atomic dipoles do not interact with one another and are randomly oriented in the absence of an external field, resulting in zero net moment. If they do interact, they can spontaneously align or anti-align, resulting in ferromagnetism (permanent magnets) or antiferromagnetism, respectively. Paramagnetic behavior can also be observed in ferromagnetic materials that are above their Curie temperature, and in antiferromagnets above their Neel temperature. Paramagnetic materials in magnetic fields will act like magnets but when the field is removed, thermal motion will quickly disrupt the magnetic alignment.

In this class of materials, some of the atoms or ions in the material have a net magnetic moment due to unpaired electrons in partially filled orbitals. However, the individual magnetic moments do not interact magnetically, and like diamagnetism, the magnetization is zero when the field is removed. In the presence of a field, there is now a partial alignment of the atomic moments in the direction of the field, resulting in a net positive magnetization and positive susceptibility [8].
At normal temperature and in moderate fields, the paramagnetic susceptibility is small (but larger than the diamagnetic contribution). Unless the temperature is very low ($\ll 100K$) or the field is very high paramagnetic susceptibility is independent of the applied filled. Some examples of paramagnetic materials include clay, Iron-rich clay, silicate, carbonate and sulfide. In addition, the efficiency of the field in aligning the moments is opposed by the randomizing effects of temperature. This results in a temperature dependent susceptibility, known as the Curie law.

1.2.4 Curie's law

Under relatively low magnetic field saturation when the majority of the atomic dipoles are not aligned with the field, paramagnetic materials exhibit magnetization

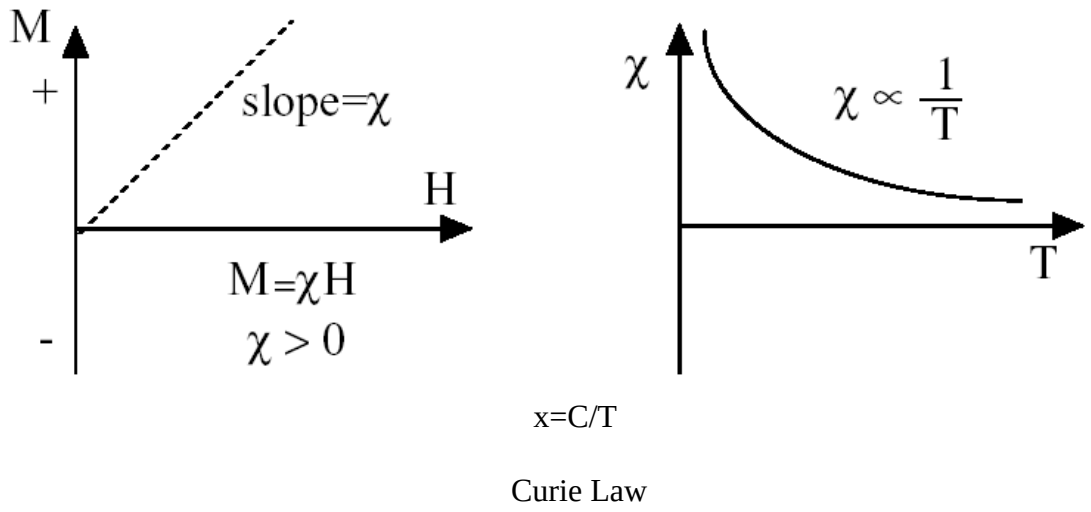

Figure 1.5: *Magnetization VS magnetic field and susceptibility VS temperature* .

according to Curie's law:

$$M = C(\frac{B}{T}) \tag{1.15}$$

where M-the resulting magnetization.

B-the magnetic flux density of the applied field measured in tesla.

T-the absolute temperature measured in kelvin.

C-is a material-specific Curie constant.

This law indicates that paramagnetic materials tend to become increasingly magnetic as the applied magnetic field is increased, but less magnetic as temperature is increased. Curie's law is incomplete because it fails to predict the saturation that occurs when most of the atomic dipoles are aligned (after every thing is aligned, increasing the external field will not increase the total magnetization) [8].

1.2.5 Ferromagnetism

Ferromagnetism is a phenomenon by which a material can exhibit a spontaneous magnetization, and is one of the strongest forms of magnetism. It is responsible for most of the magnetic behavior encountered in every day life and is the basis for all

permanent magnets (as well as the metals that are noticeably attracted to them). In particular, a material is ferromagnetic in narrower sense only if all of its magnetic ions add a positive contribution to the net magnetization. If some of the magnetic ions subtract from the net magnetization (if they are partially anti-aligned), then the material is ferrimagnetic. If the ions anti-align completely so as to have zero net magnetization, despite the magnetic ordering, then it is an antiferromagnet. All of these alignment effects only occur at temperatures below a certain critical temperature, called the Curie temperature (for ferromagnets and ferrimagnets) or the Neel temperature (for antiferromagnets). Curie temperature is the temperature above which the material cease to exhibit spontaneous magnetization.

If the crystal symmetry is such that the localized dipole moment alignment at different lattice sites results in a net macroscopic magnetization at $T = 0K$ even in the absence of an external magnetic field then such a solid is referred to as ferromagnetically ordered.

Unlike paramagnetic materials, the atomic moments in these materials exhibit very strong interactions. These interactions are produced by electronic exchange forces and result in a parallel or antiparallel alignment of atomic moments. The exchange force is a quantum mechanical phenomenon due to the relative orientation of the spins of two electrons. Ferromagnetic materials exhibit parallel alignment of moments resulting in large net magnetization even in the absence of a magnetic field.

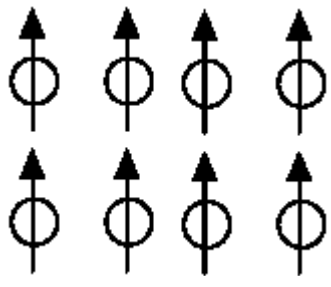

Figure 1.6: *spin alignment for ferromagnets* .

The susceptibility of a ferromagnet above the Curie temperature has the form

$$\chi = \frac{c}{T - T_C}. \tag{1.16}$$

This is the Curie-Weiss law in the mean field approximation for $(T > T_c)$.
Two distinct characteristics of ferromagnetic materials are their:-
1.spontaneous magnetization and
2.the existence of magnetic ordering temperature.

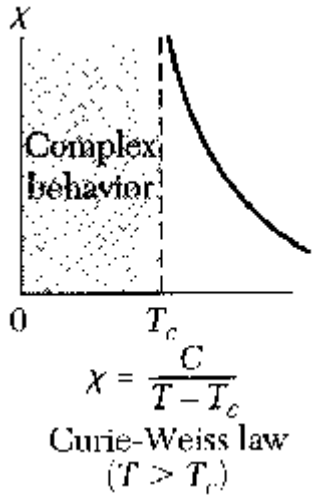

Figure 1.7: *Susceptibility VS temperature for ferromagnets* .

1.2.6 Spontaneous magnetization

Spontaneous magnetization is the net magnetization that exists inside a uniformly magnetized microscopic volume in the absence of a field. The magnitude of this magnetization, at 0k, is dependent on the spin magnetic moments of electrons. There can be two types of spontaneous magnetism-localized magnetism and itinerant magnetism. Localized magnetism occurs due to the interaction between sufficiently localized and tightly bounded inner shell electrons of atoms or ions at close by lattice sites (still some wave function overlap is must). Where as itinerant magnetism occurs because of the "free" electrons which are shared by the entire crystal.

A related term is the saturation magnetization which we can measure in the laboratory. The saturation magnetization is the maximum induced magnetic moment that can be obtained in a magnetic field (H_{sat}); beyond this field no further increase in magnetization occurs. Saturation magnetization is an intrinsic property, independent of particle size but dependent on temperature. Fe, Co, Ni, MnAs etc are few examples of ferromagnetically ordered crystals.

1.2.7 Antiferromagnetism

A more common situation is when the crystal symmetry is such that the localized dipole moment alignment at different lattice sites results in no net macroscopic magnetization at $T = 0K$ in the absence of an external magnetic field, such a solid is referred to as antiferromagnetically ordered and resultant magnetization (at higher temperatures) is referred to as antiferromagnetism.

In materials that exhibit antiferromagnetism, the spins of magnetic electrons align in a regular pattern with neighboring spins pointing in opposite directions. This is the opposite of ferromagnetism. Generally, antiferromagnetic materials exhibit antiferromagnetism at a low temperature, and become disordered above a certain temperature; the transition temperature is called the Neel temperature. Above the Neel temperature, the material is typically paramagnetic.

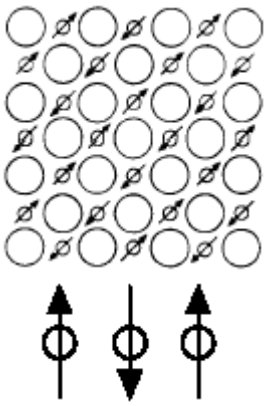

Figure 1.8: *Spin alignment for antiferromagnetism* .

The antiferromagnetic behavior at low temperature usually results in diamagnetic

properties, but can sometimes display ferrimagnetic behavior, which in many physically observable properties is more similar to ferromagnetic interactions. The magnetic susceptibility of an antiferromagnetic material will appear to go through a maximum as the temperature is lowered; in contrast, that of a paramagnet will continually increase with decreasing temperature.

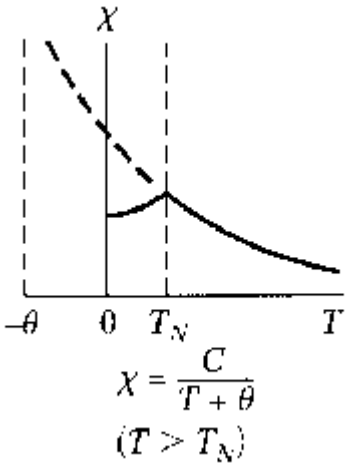

Figure 1.9: *Susceptibility VS temperature for antiferromagnets* .

If the two sublattice moments are exactly equal but opposite, then the net moment is zero. This type of magnetic ordering is called antiferromagnetism. Antiferromagnetic materials also have zero remanence, no hysteresis, but a small positive susceptibility that varies in a peculiar way with temperature. An antiferromagnet is a special case of a ferrimagnet for which both sublattices a and b have equal saturation magnetization. Above T_N, the susceptibility obeys the Curie-Weiss law for paramagnets but with a negative intercept indicating negative exchange interactions.

Slight deviations from ideal antiferromagnetism can exist if the antiparallelism is not exact. If neighboring spins are slightly tilted $(< 1^0)$ or canted, a very small net magnetization can be produced. This is called canted antiferromagnetism and hematite is a well known example. Canted antiferromagnets exhibit many of the typical magnetic characteristics of ferro-and ferrimagnets (e.g., hysteresis, remanence, Curie temperature) [8].

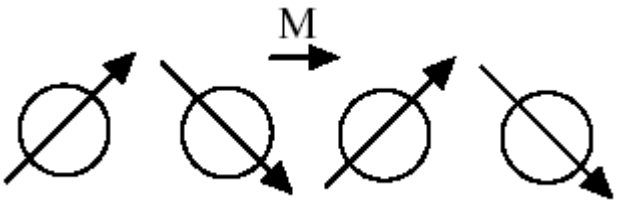

Figure 1.10: *Spin alignment in canted antiferromagnetism* .

1.2.8 Ferrimagnetism

A ferrimagnetic material is one in which the magnetic moment of the atoms on different sublattices oppose as in antiferromagnetism but the opposing moments are unequal and a spontaneous magnetization remains. This happens when the sublattices consist of different materials or ions (such as Fe^{2+} and Fe^{3+}) [7]. Ferrimagnetic materials are like paramagnets in that they show no magnetic order above the Curie temperature, and are like ferromagnets in that they can hold a spontaneous magnetization below. However, there is some times a temperature below the Curie temperature at which the two sublattices have equal moments, resulting in a net magnetic moment of zero; this is called the compensation point.

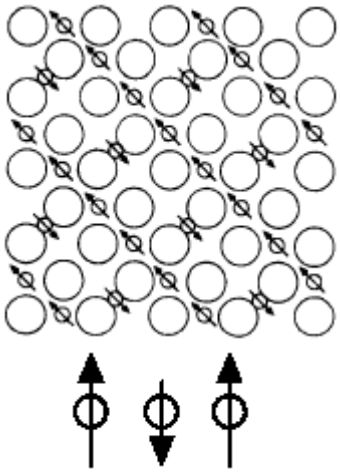

Figure 1.11: *Spin alignment in ferrimagnetism* .

Strictly, ferrimagnetism is only a special case of ferromagnetism. If the mare symmetry consideration predicts an antiferromagnetic ordered but the crystal turns out to have net resultant macroscopic magnetization due to the different magnitude of exchange interactions on different lattice sites we reffered to such a solid as ferrimagnetically ordered and the resultant magnetization as ferrimagnetism. Fe_3O_4 is one of the most famous examples of ferrimagnetically ordered solids [9].

The magnetic structure in ferrimagnets is composed of two magnetic sublattices. The magnetic moments of the two sublattices are not equal and result in a net magnetic moment. Ferrimagnetism is therefore similar to ferromagnetism. It exhibits all the hall marks of ferromagnetic behavior-spontaneous magnetization, Curie temperature, hyste- resis and remanence. However, ferro-and ferrimagnets have very different magnetic ordering.

1.2.9 Spin glass

A spin glass is a disordered material exhibiting high magnetic frustration. The origin of the behavior can be either a disordered structure (such as that of a conventional, chemical glass) or a disordered magnetic doping in an otherwise regular structure. Frustration refers to the inability of the system to remain in a single lowest energy state (the ground state). The spin glasses have many ground states which are never explored on experimental time scales.

A spin glass has been defined as a random, metallic, magnetic system in a non magnetic host characterized by a random freezing of moments without long range order at a rather well-defined temperature T_f [10].

It is the time dependence which distinguishes spin glasses from other magnetic systems. Beginning above the spin glass transition temperature, T_f, where the spin glass exhibits more typical magnetic behavior, (such as paramagnetism as discussed here but other kinds of magnetism are possible), if an external magnetic field is applied and the magnetization is plotted versus temperature, it follows the typical Curie law (in which magnetization is inversely proportional to temperature) until T_f is reached, at which point the magnetization becomes virtually constant (this value is called the field cooled magnetization). This is the onset of the spin glass phase. When the external field is removed, the spin glass has a rapid decrease of magnetization to a

value called the remanent magnetization, and then a slow decay as the magnetization approaches zero. This slow decay is particular to spin glasses.

For spin glass we have a long range interaction via the conduction electrons, which causes the freezing process, and hence a cooperative behavior would be expected [10]. Experimentally one observes in a spin glass a sharp, cusp like maximum in the very low (or even zero) field susceptibility at the freezing temperature T_f, which becomes greatly broadened for external fields of only a few hundred Oersteds-spin glass can be strongly magnetized by applying magnetic fields, but below T_f there is large hysteresis.

1.3 Antiferromagnetic spin waves

Here we will consider the nature of the elementary excitations over the ground state at a non-zero temperature or with the application of a small perturbation on the system. Any local deviation from perfect alignment of the spin system will not in general remain confined in the region, but, owing to exchange coupling, will propagate like a wave. These low-lying excitations are called spin waves and were first introduced by Bloch (1930). The spin wave can be thought of as one spin reversal spread coherently over the entire crystal. When spin waves are quantized, we refer to the state of excitation interms of a certain number of magnons in the particular mode. They behave like field quanta and obey Bose statistics. It should be noted that the concept of magnons as non-interacting quasiparticles is valid in very low temperature regions, i.e. far below the characteristic temperature beyond which long range magnetic order breaks down. At elevated temperatures certain kinematic and dynamic interactions have to be considered.

The spin wave excitation energy, incontrast to the ferromagnetic case, is linear in k at long wave lengths [11]. But in ferromagnets the dependence of spin wave excitation energy on k is quadratic (k^2). In an antiferromagnet the spins are ordered in an antiparallel arrangement with zero net moment at temperatures below the ordering or Neel temperature [5].

1.3.1 Exchange interaction

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:-

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 so that their electron clouds do overlap then they can have direct exchange.
2. Superexchange interaction:- 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 nonparamagnentic ion/atom sitting in between we call such interactions as superexchange interaction.
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.
4. Itinerant exchange:- For those electrons that aren't well localized and are shared by the entire crystal, such electrons can have exchange interaction among themselves.Such electron- electron interactions are called itinerant exchange interaction[9].

The singlet-triplet energy splitting measures the extent to which the antiparallel (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 [11]. For two atoms with two-electron systems at sites i and j in which the exchange integral is expressed as:

$$J_{ij} = \frac{1}{2}(E_s - E_t) \tag{1.17}$$

Equation (1.17) 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

$$H = -2J_{ij}\overrightarrow{S_i}.\overrightarrow{S_j} \tag{1.18}$$

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:

$$H = -2\sum_{ij} J_{ij}\overrightarrow{S_i}.\overrightarrow{S_j} \tag{1.19}$$

The spin Hamiltonian (1.19) is known as the Heisenberg Hamiltonian and the J_{ij} are known as the exchange coupling constants.

Chapter 2

A review on the study of DMS

2.1 Introduction

Diluted magnetic semiconductors (DMS) are composed of an inert host semiconductor doped with both localized spins and carriers (electrons or holes) that are either itinerant, or localized on a much longer length scale [12]. The name diluted magnetic semiconductors implies that the system has only a small percentage of localized spins, they are at the opposite extreme of the dilute magnetic alloys, the canonical systems involving itinerant fermion and localized spin degrees of freedom, which have been studied extensively [13].

In the dilute magnetic metallic alloys, the law of density of spins are a perturbation on the Fermi liquid representing the non magnetic host metal, so depending on the concentration of the localized moments, they may be studied interms of diluted Kondo systems, or amorphous magnetic system with a spin-spin coupling mediated by the fermi sea of conduction electrons (RKKY coupling), which lead often to spin glass behavior [14]. DMS's, which are made by substitution of small amount of 3d magnetic transition-metal (TM) atoms such as Mn, Fe, and Co ions in to cation sites in the II-VI or III-V compound semiconductors, involve both semiconductor physics and magnetism [15].

In the 1980s, research in DMS focused mainly on II-VI semiconductor such as (Cd, Mn)Te and (Zn, Mn)Se. In a II-VI semiconductor, the II element is substituted by an equivalent valence of magnetic transition metal atoms, making it possible to

achieve a high density of magnetic atoms and to fabricate quantum structures. In a II-VI DMS, the optical characteristics such as Faraday effect are greatly modulated by magnetic fields due to the exchange interaction (sp-d interaction) between the s and p orbitals of non magnetic atoms and the d orbitals of the magnetic atoms. However, it is generally difficult to control conduction by doping in II-VI semiconductors, which is one of the major obstacles for the use of II-VI semiconductors as electronic materials.

Although II-VI DMS generally exhibit antiferromagnetism, spin glass and/or paramagnetic properties, as we will discuss later, it was recently reported that a p-doped II-VI DMS undergoes a ferromagnetic transition at low temperatures. The most relevant feature of DMS, which attracted considerable interest, is the coexistence and interaction of two different electronic subsystems: delocalized conduction (s-type) and valence (p-type) band electrons and localized (d or f-type) electrons of magnetic ions [16].

Diluted magnetic semiconductors are expected to play an important role in interdisciplinary material science and future electronics because charge and spin degrees of freedom accommodated in to a single material exhibits interesting magnetic, magneto-optical, magneto-electronic and other properties [4]. DMS are semiconductors in which a fraction of the component ions are replaced by those of transition metals or rare earths. Most importantly, state of magnetization changes the electronic properties and vice versa through the spin exchange interaction between local magnetic moments and carriers.

The usefulness of semiconductors resides in the ability to dope them with impurities to change their properties, usually to p- or n-type. This approach can be followed to introduce magnetic elements in to nonmagnetic semiconductors to make them magnetic. This category of semiconductors, called diluted magnetic semiconductors (DMSs), are alloys of nonmagnetic semiconductor and magnetic elements. Diluted antiferromagnets represent an interesting class of materials whose crystallographic structure is closely related to their magnetic properties [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. This can be done by achieving information processing and data storage facilities in a single crystal that gives increased transistor density making use of spin of electrons in magnetic materials [17].

Diluted magnetic semiconductors are prepared by introducing magnetic elements in to nonmagnetic semiconductors to make them magnetic. Therefore, they are alloys of semiconductors and magnetic ions of transition metals or rare earths which exhibit a variety of cooperative effects via spin exchange interactions not originally present in nonmagnetic semiconductors. The transport, optical, and magnetic properties of the host semiconductors will be altered due to the exchange interaction between the conduction carriers and the localized moments of the magnetic ions. In these magnetic semiconductors exchange interactions between the electrons in the semiconducting band and the localized electrons at the magnetic ions lead to a number of unusual and interesting properties, such as a red shift of band gap when ferromagnetism or antiferromagnetism sets in. The crystal structure of such magnetic semiconductors is quite different from that of nonmagnetic semiconductors.

In a new branch of electronics known as spintronics or spin electronics or sometimes magneto-electronics, there is need for manipulating of spin and charge degrees of freedom of electrons to yield a desired electronic out come. All spintronic devices act in order that:-

1) information is stored (written) in to spins as a particular spin orientation (up or down).
2) the spins, being attached to mobile electrons, carry the information along a wire.
3) the information is read at a terminal.

Spin orientation of conduction electrons survives for a relatively long time (nanoseconds compared to tens of femtoseconds during which electron momentum and energy decay), which make spintronic devices particularly attractive for memory, storage and magnetic sensors application, and potentially for quantum computing where electron

spin would represent a bit (called qubit) of information.

What is expected from these materials is to have a large commercial and economical impact in nonvolatile memories (the information stays in the memory even if the electric power is switched off, in contrast to semiconductor memories that we use today). In general the advantage of these new devices would be nonvolatility, increased data processing speed, decreased electric power consumption, and increased transistor density compared with conventional semiconductor devices.

Within the next few years, it is expected that magnetoelectronic chips will be used in quantum computers. An inherent advantage of magnetoelectronics over electronics is the fact that magnet tend to stay magnetized for long. Hence this arises interest in industries to replace the semiconductor-based components of computer with magnetic ones, starting from RAM. The new magnetic RAM will retain data even when the computer is turned off.

2.2 Alloy of II-VI semiconductors with magnetic materials

Diluted magnetic semiconductors (DMS) are compound of alloy semiconductors containing a large fraction of magnetic ions $(Mn^{+2}, Cr^{+2}, Fe^{+2}, Co^{+2})$ and are studied mainly on II-VI based materials such as CdTe and ZnSe etc. This is because such +2 magnetic ions are easily incorporated in to the host II-VI crystals by replacing group II cations. In such II-VI based DMS such as (Cd, Mn)Se, magneto-optic properties were extensively studied,and optical isolators were recently fabricated using their large Faraday effect. Although this phenomenon makes these DMS relatively easy to prepare in bulk form as well as thin epitaxial layers, II-VI based DMS are difficult to dope to create p- and n-type, which makes the material less attractive for applications.

The magnetic interaction in II-VI DMSs is dominated by the antiferromagnetic exchange among the Mn spins, which results in the paramagnetic, antiferromagnetic, or spin glass behavior of the material. It was not possible until very recently to make a II-VI DMS ferromagnetic at low temperature $(< 2K)$. The zinc-blende ternary compound $Zn_{1-x}Mn_xTe$ with $x < 0.65$ is a typical (diluted) magnetic semiconductor [18]. This compound is also a typical frustrated antiferromagnet with 5/2 spins (Mn^{2+}) randomly arranged on face centered cubic (f.c.c) sublattice points [19]. This compound

undergoes spinglass transition below 50k in the wide manganese concentration range, $x = 0.20 - 0.65$ [20].We recall that for all II-VI dilute magnetic semiconductors with Mn, Co or Fe p-d exchange is antiferromagnetic (AF) [1,2].

A semiconductor AB where A and B are, respectively, group II and VI elements, upon being alloyed with a compound MB where M is an element of the iron group, forms part of a class of materials usually called diluted magnetic semiconductors (DMS). If x is the atomic concentration of M atoms replacing atoms of type A, the formula of a DMS is written $A_{1-x}M_xB$. A typical example is $Cd_{1-x}Mn_xTe$ which can form homogeneous solid solutions for x ranging from 0 to 0.75. These materials have a unique combination of semiconducting and magnetic properties. $Cd_{1-x}Mn_xTe$ crystalizes in the zinc-blend structure for $0 \leq x \leq 0.75$ in which at random cation sites Cd is replaced by Mn atoms. For $x < 0.17$, $Cd_{1-x}Mn_xTe$ is paramagnetic for all temperatures [21]. If $x > 0.17$, below a critical temperature $T_c(x)$, there is a magnetically ordered phase due to antiferromagnetic coupling between neighboring Mn^{2+} ions. For example $Cd_{1-x}Mn_xTe$ with $x = 0.75$ is in antiferromagnetic phase below 40k [21]. It has been experimentally established that CdMnTe and ZnMnTe compounds with zinc-blende (ZB) crystallographic structures reveal long range antiferromagnetic ordering when the concentration of Mn is relatively high [22]. Detailed investigations performed with the use of the neutron diffraction show that the antiferromagnetic phase of type III appears for concentrations as high as 0.8 [23].

The II-VI DMS are based on semiconductors AB, where A is a group II element and B is a group VI element (such as CdTe or ZnSe) [12]. In the II-VI DMS, some of the divalent sites (Cd/Zn) are substituted by a magnetic element, typically Mn. This fraction is denoted by x, so the DMS we consider is $A_{1-x}Mn_xB$. Mn is also a group II element, but in addition it has a half filled 3d shell, with a total spin given by Hund's rule: S=5/2. In the absence of other types of dopants, the system $A_{1-x}Mn_xB$ is an insulator which exhibits antiferromagnetic (AFM) tendencies at low temperatures. This is seen, for instance, from measurements of the susceptibility which is found to depend on temperature as $\chi(T) \sim 1/(T + T_N)$, with a Neel temperature of a few kelvin [1]. The origin of this AFM tendency is the (expected) antiferromagnetic exchange between the Mn spins. However, for low doping concentrations x, the average distance between Mn spins is large and this AFM direct exchange is rather small. In $CdTe/Cd_{1-x}Mn_xTe$,

the dilution decreases the influence of ion-ion interaction, which favors an antiparallel alignment of Mn^{2+} spins at low temperatures [24]. When a Mn^{2+} ion occupies a substitutional Cd site in the cubic CdTe crystal, its free-ion terms split due to the cubic crystal field [25].

When a low density of charged dopants, such as group-V phosphorus (P) substituting for the group-VI element, is introduced in the system, each of them binds a hole (or electron) in a shallow hydrogenic 1S state $\phi(r) \sim exp(-r/a_B)$, characterized by a Bohr radius $a_B \sim 10 - 20\mathring{A}$. Exchange interactions arise between the spins of these charge carriers and the Mn spins, and are described by the Hamiltonian

$$H = \sum_{i,j} J(\overrightarrow{r_i}, \overrightarrow{R_j}) \overrightarrow{S_i}.\overrightarrow{S_j} \tag{2.1}$$

Here, $\overrightarrow{S_j}$ is the spin of the Mn at position $\overrightarrow{R_j}$ and $\overrightarrow{S_i}$ is the spin of the electron/hole centered at $\overrightarrow{r}_i$. The exchange interaction $J(\overrightarrow{r_i}, \overrightarrow{R_j})$ is dependent on the overlap between the orbital $\phi(\overrightarrow{r} - \overrightarrow{r_i})$ of the charge carrier and the orbitals $\psi_d(\overrightarrow{r} - \overrightarrow{R_j})$ of the 3d electrons responsible for the Mn spin. Since these 3d orbitals are localized on a scale of a few $\mathring{A}$ around the Mn nucleus, the exchange is proportional to the carrier charge density at the Mn site, i.e.,

$$J(\overrightarrow{r_i}, \overrightarrow{R_j}) = J_o |\phi(\overrightarrow{R_j} - \overrightarrow{r_i})|^2 = J_o exp(-2|\overrightarrow{r_i} - \overrightarrow{R_j}|/a_B) \tag{2.2}$$

where J_o characterizes the strength of the exchange. Typically for electron $J_o < 0$, while for holes $J_o > 0$.

As mentioned earlier, $Cd_{1-x}Mn_xTe$ exhibits a magnetically ordered low temperature phase for $x > 0.17$. The transition from the paramagnetic to the magnetically ordered phase is accompanied by the appearance of a new Raman feature at low temperatures. Since this excitation is associated with magnetic order, it is attributed to a magnon. A distinct magnon feature was observed in $Cd_{1-x}Mn_xTe$ for the composition range $0.40 \leq x \leq 0.70$. The magnon feature is absent when the incident and the scattered polarizations are parallel. In an antiferromagnetic system these excitations can be described by classifying the spins in to those which in the state of equilibrium point in one direction and those pointing in an antiparallel direction. This classification gives rise to magnetizations $\overrightarrow{M_1}$ and $\overrightarrow{M_2}$ where $\overrightarrow{M_1}$ is the magnetic moment per

unit volume of the spins of the first class and $\overrightarrow{M_2}$ that of the second. In equilibrium $\overrightarrow{M_1} + \overrightarrow{M_2} = 0$. Now, the agents responsible for the preferential orientation of a spin of type 1 are those of type 2 and conversely. The best fit to the data for $x = 0.70$ yields $T_N = 40K$ [21].

2.3 Alloys of III-V semiconductors with ferromagnetic properties

An approach compatible with the semiconductors used in present-day electronics is to make nonmagnetic III-V semiconductors magnetic, and even ferromagnetic, by introducing a high concentration of magnetic ions. The III-V semiconductors such as GaAs are already in use in a wide variety of electronic equipment in the form of electronic and optoelectronic devices, including cellular phones (microwave transistors), compact disks (semiconductor lasers), and in many other applications. Therefore, the introduction of magnetic III-V semiconductors opens up the possibility of using a variety of magnetic phenomena not present in conventional nonmagnetic III-V semiconductors in the optical and electrical devices already established.

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 the absence of holes, the magnetic interaction among Mn has been shown to be antiferromagnetic in n-type (In, Mn)As and in fully carrier compensated (Ga, Mn)As. These results show that the ferromagnetic interaction is hole induced.

Magnetic III-V semiconductors show ferromagnetism, whose Curie temperature, T_c, is relatively high as predicted theoretically only when the carrier concentration is effectively raised. However, the highest Curie temperature, T_c, obtained to date is about 110k in $Ga_{1-x}Mn_xAs$ [26].ZnO, TiO_2 doped with 3d transition metals Co, Ni, Fe etc are promising DMS materials showing ferromagnetism with T_c above room temperature. The large band gap may open up transparent ferromagnetic materials in oxide DMS for application in the visible region.

When Mn is doped in a III-V semiconductor, such as GaAs, the major difference with respect to the II-VI DMS is that the Mn atom provides both the S=5/2 spin and the dopant charge carrier (a hole, since divalent Mn substitutes for trivalent Ga). While this implies nominally equal concentrations of holes and Mn spins, experimentally it is found that the hole concentration is only p=10-30 percent of the Mn concentration [27]. As in the (II, Mn)VI systems, the main magnetic interaction in the (III, Mn)V DMS is the exchange between the Mn spins and the hole spins, which is known to be AFM[28]. The DMS materials based on III-V compounds (GaAs, InAs) were always a challenging problem due to their possible application for optoelectronic devices [29,30].

The III-V diluted magnetic semiconductors are materials which exhibit spontaneous ferromagnetism mediated by holes in the valence band of the host semiconductor and thus represent new materials with promising applications in spintronics. The Mn-doped III-V magnetic semiconductors are the most intensively studied DMS materials of current interest [31,32]. These materials are expressed in the form of $(III_{1-x}Mn_x)V$; examples of which are $(Ga_{1-x}Mn_x)As$, $(Ga_{1-x}Mn_x)P$, $(Ga_{1-x}Mn_x)N$, $(Ga_{1-x}Mn_x)Sb$, $(In_{1-x}Mn_x)As$ and so on; where the carriers are typically holes.

In Mn^{2+} doped III-V compound semiconductors Mn has a different valence to group III element and introduces a spin-5/2 moment, leading to both local moment formation (playing role of magnetic ions) and acts as acceptors introducing valence band holes due to valence mismatch [31] in semiconductor host; where these holes mediate interactions between the Mn moments correlating their orientations and making ferromagnetism possible [33,35].

2.4 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.

element	Cr^{24}	Mn^{25}	Fe^{26}	Cr^{2+}	Mn^{2+}	Fe^{2+}	Fe^{3+}
configuration	$3d^5 4S^1$	$3d^5 4S^2$	$3d^6 4S^2$	$3d^4$	$3d^5$	$3d^6$	$3d^5$

In Mn^{2+} doped III-V DMS, 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 $2.24\mathring{A}$ [5] relatively, is more preferable for doping purpose in which high concentration of 3d spins is required.

2.5 Structure of CdTe

If the two atoms of the basis are identical, the structure is called diamond. Semiconductors such as Si, Ge, C, etc fall in this category. If the two atoms are different, the structure is called the zinc blende structure.

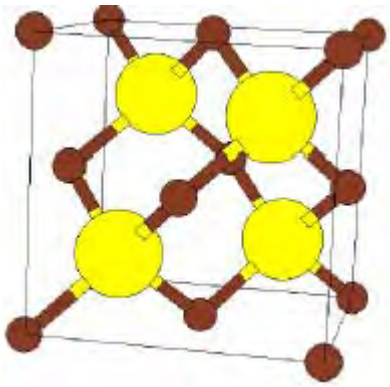

Figure 2.1: *The zinc blende structure of CdTe* .

Semiconductors such as GaAs, AlAs, CdS, CdTe, etc, fall in this category. Semiconductors with diamond structure are often called elemental semiconductors, while the zinc blende semiconductors are called compound semiconductors. The compound semiconductors are also denoted by the position of the atoms in the periodic chart, e.g, GaAs, AlAs, InP are called III-V (three-five) semiconductors while CdTe, CdS, HgTe, etc., are called II-VI (two-six) semiconductors.

2.6 Synthesis of dilute magnetic semiconductors

DMSs are semiconductors containing magnetic ions and normally prepared by doping magnetic atoms (Fe, Ni, Co, Mn, Cr) in to the compound semiconductors (CdTe, InSb). DMSs have high interaction between magnetic spins and conducting carriers because DMSs possess magnetic properties in addition to semiconducting. It is expected that DMSs can yield industrially important and high technological new materials whose functions are controlled by changing the interaction. One such example is that the DMS material will cause magnetic field when it absorbs either optical or electrical power, or the wave length of semiconductive laser will change by varying the external magnetic field. However, most of the DMSs have not been examined enough. There are many unknown factors, such as limitation on doping concentration of magnetic ions or effect of dopant on the crystal structure and the properties. We would like to synthesize high quality DMS materials and elucidate the fundamental properties. In future there will be a new productive technique to synthesize high quality DMS materials for industrial use.

Chapter 3

Mathematical techniques for spin wave excitations

3.1 Formulation of the problem

We assume the spin structure of the crystal may be divided in to two interpenetrating sublattices a and b with the property that all nearest neighbors of an atom on a lie on b, and vise versa.

The Heisenberg exchange Hamiltonian is given by

$$H = -\sum_{i \neq j} J_{ij} \overrightarrow{S_i} . \overrightarrow{S_j} \tag{3.1}$$

where the summation is over all the magnetic ions in the lattice, and J_{ij} is the exchange integral between the i^{th} and the j^{th} ion. Assume that J_{ij} is non vanishing only for the nearest neighbors and it is taken to be isotropic. When J_{ij} is positive, the lowest energy states corresponds to the situation where the spins of all the ions are parallel. This is the case of ferromagnetism. But when J_{ij} is negative the neighboring spins tend to be antiparallel, resulting in an antiferromagnetic substance.

When we are interested in the magnetic properties, we require the response of the system to the external magnetic field. The Hamiltonian including the magnetic

perturbation is given by

$$H = \sum_{ij} J_{(\overrightarrow{r}_{ij})} \overrightarrow{S_i}.\overrightarrow{S_j} - g\mu_B H_A \sum_i S_{iz}^a + g\mu_B H_A \sum_j S_{jz}^b \tag{3.2}$$

where g-the Lande g-factor for the ion.

H_A-the applied magnetic field, we take $H_A > 0$ to make the magnetic moments line up along the positive z-axis when the system is in the ground state.

$r_{ij} = |r_i - r_j| = l$-is the relative position vector of spins S_i and S_j.

l- connects the atom i with its nearest neighbors on a bravis lattice.

Here we use the Holstein-Primakoff (H-P) transformation. It is a transformation from the spin operators S_i to the spin deviation creation and annihilation operators a_i^+ and a_i, which have the following commutation relations

$$[a_i, a_j^+] = \delta_{ij} \quad and \quad [a_i, a_j] = [a_i^+, a_j^+] = 0. \tag{3.3}$$

The spin occupation number n_i, which is the eigen value of $a_i^+ a_i$, is defined by the relation

$$n_i = S - S_{iz}. \tag{3.4}$$

It is very important to note that n_i is the reduction in the z-component of the i^{th} spin from its maximum possible value of S. Thus the operator a_i creates a quantum of spin deviation, that is, it reduces S_{iz} by one unit.

To derive the H-P transformation, we use

$$S_i^{\pm} = S_{ix} \pm iS_{iy} \tag{3.5}$$

as the two independent spin operators, simply because they have the convenient raising and lowering properties. We already know from the theory of atomic spectra that:

$$S_i^{\pm}|S_{iz}\rangle = [S(S+1) - S_{iz}(S_{iz} \pm 1)]^{\frac{1}{2}}|S_{iz} \pm 1\rangle. \tag{3.6}$$

Now,taking the first relation from (3.6), we have, dropping for the moment the subscript,

$S^+|S_z\rangle = [S(S+1) - S_z(S_z+1)]^{\frac{1}{2}}|S_z+1\rangle$

$= (S^2 + S + SS_z - SS_z - S_z^2 - S_z)^{\frac{1}{2}}|S_z + 1\rangle$

$= (S + S_z + 1)^{\frac{1}{2}}(S - S_z)^{\frac{1}{2}}|S_z + 1\rangle$.

Hence we get

$$S^+|S_z\rangle = (2S)^{\frac{1}{2}}(1 - \frac{S - S_z - 1}{2S})^{\frac{1}{2}}(S - S_z)^{\frac{1}{2}}|S_z + 1\rangle \tag{3.7}$$

or,restricting the subscript i, we get, using (3.4),

$$S_i^+|n_i\rangle = (2S)^{\frac{1}{2}}(1 - \frac{n_i - 1}{2S})^{\frac{1}{2}}(n_i)^{\frac{1}{2}}|n_i - 1\rangle. \tag{3.8}$$

The fact that S^+ operating on S_z gives rise to a state having $(S_z + 1)$ means that the occupation number n_i decreases by one unit, becoming $(n_i - 1)$.

Now using the relations:

$$a|n\rangle = (n)^{\frac{1}{2}}|n - 1\rangle \tag{3.9}$$

$$a^+|n\rangle = (n + 1)^{\frac{1}{2}}|n + 1\rangle \tag{3.10}$$

$$a^+a|n\rangle = n|n\rangle \tag{3.11}$$

we find that (3.8) reduces to

$$S_i^+ = (2S)^{\frac{1}{2}}(1 - \frac{a_i^+a_i}{2S})^{\frac{1}{2}}a_i. \tag{3.12}$$

In (3.12) we have written $a_i^+a_i$ in place of $(n_i - 1)$ because, when a_i operates on n_i, we get the state $(n_i - 1)$ and also from (3.11)

$a_i^+a_i|n_i - 1\rangle = (n_i - 1)|n_i - 1\rangle$.

Thus, in (3.12), we have replaced the eigenvalue $(n_i - 1)$ by the corresponding (number) operator $a_i^+a_i$, as is possible since $\frac{n_i}{2S} \ll 1$, that is $[1 - \frac{n_i}{2S}]^{\frac{1}{2}}$ converges.

Similarly, taking the second relation from (3.6), we get

$S_i^-|n_i\rangle = (2S)^{\frac{1}{2}}(n_i + 1)^{\frac{1}{2}}(1 - \frac{n_i}{2S})^{\frac{1}{2}}|n_i + 1\rangle \ = (2S)^{\frac{1}{2}}a_i^+(1 - \frac{n_i}{2S})^{\frac{1}{2}}|n_i\rangle$

$$S_i^- = (2S)^{\frac{1}{2}}a_i^+(1 - \frac{a_i^+a_i}{2S})^{\frac{1}{2}}. \tag{3.13}$$

This is the H-P transformation, that is, the relation ship of the spin operators with the spin deviation creation and annihilation operators, a_i^+ and a_i.

Collecting the terms from (3.4),(3.12) and (3.13), we have

$$S_i z = S - a_i^+ a_i \tag{3.14}$$

$$S_i^+ = (2S)^{\frac{1}{2}}(1 - \frac{a_i^+ a_i}{2S})^{\frac{1}{2}} a_i \tag{3.15}$$

$$S_i^- = (2S)^{\frac{1}{2}} a_i^+ (1 - \frac{a_i^+ a_i}{2S})^{\frac{1}{2}}. \tag{3.16}$$

In (3.2) the first term is the familiar Heisenberg exchange energy, expressed in terms of the atomic spin operators and the last two terms are Zeeman interaction (i.e, the interaction of spins with the applied magnetic field, H_A). The Hamiltonian involves the three components S_{ix}, S_{iy}, S_{iz} of each spin S_i, and similarly for S_j, that is

$$\overrightarrow{S_i} = S_{ix} i + S_{iy} j + S_{iz} k \qquad and \qquad \overrightarrow{S_j} = S_{jx} i + S_{jy} j + S_{jz} k. \tag{3.17}$$

From equation (3.5) we can find S_{ix} and S_{iy}:

$$S_{ix} = \frac{S_i^+ + S_i^-}{2} \qquad and \qquad S_{iy} = \frac{S_i^+ - S_i^-}{2i} \tag{3.18}$$

Similarly

$$S_{jx} = \frac{S_j^+ + S_j^-}{2} \qquad and \qquad S_{jy} = \frac{S_j^+ - S_j^-}{2i} \tag{3.19}$$

Hence,

$$\overrightarrow{S_i} = (\frac{S_i^+ + S_i^-}{2})i + (\frac{S_i^+ - S_i^-}{2i})j + S_{iz} k \quad and \quad \overrightarrow{S_j} = (\frac{S_j^+ + S_j^-}{2})i + (\frac{S_j^+ - S_j^-}{2i})j + S_{jz} k \tag{3.20}$$

In our case since we have two sublattices a and b, we can write equations (3.14), (3.15) and (3.16) as follows:

$$S_{ai}^+ = (2S)^{\frac{1}{2}}(1 - \frac{a_i^+ a_i}{2S})^{\frac{1}{2}} a_i \tag{3.21}$$

$$S_{ai}^- = (2S)^{\frac{1}{2}} a_i^+ (1 - \frac{a_i^+ a_i}{2S})^{\frac{1}{2}} \tag{3.22}$$

$$S_{bj}^+ = (2S)^{\frac{1}{2}} b_j^+ (1 - \frac{b_j^+ b_j}{2S})^{\frac{1}{2}} \tag{3.23}$$

$$S^{-}_{bj} = (2S)^{\frac{1}{2}}(1 - \frac{b^{+}_{j}b_{j}}{2S})^{\frac{1}{2}}b_{j} \tag{3.24}$$

$$S^{a}_{iz} = S - a^{+}_{i}a_{i} \tag{3.25}$$

$$-S^{b}_{jz} = S - b^{+}_{j}b_{j} \tag{3.26}$$

where b^{+}_{j}, b_{j} are creation and annihilation operators which refer to the j^{th} atom on sublattice b, they are not magnon variables. It is convenient to make a transition from the atomic a^{+}_{i}, a_{i} and b^{+}_{j}, b_{j} to the magnon variables c^{+}_{k}, c_{k} and d^{+}_{k}, d_{k} for the two sublattices a and b respectively, which is defined by

$$c^{+}_{k} = N^{\frac{-1}{2}} \sum_{i} exp(-ik.r_{i})a^{+}_{i} \quad and \quad c_{k} = N^{\frac{-1}{2}} \sum_{i} exp(ik.r_{i})a_{i} \tag{3.27}$$

and

$$d^{+}_{k'} = N^{\frac{-1}{2}} \sum_{j} exp(ik'.r_{j})b^{+}_{j} \quad and \quad d_{k'} = N^{\frac{-1}{2}} \sum_{j} exp(-ik'.r_{j})b_{j} \tag{3.28}$$

here r_{i} and r_{j} are the position vectors of the atoms i and j in the sublattice a and b respectively. The inverse transformation is then given by:

$$a_{i} = N^{\frac{-1}{2}} \sum_{k} exp(-ik.r_{i})c_{k} \quad and \quad a^{+}_{i} = N^{\frac{-1}{2}} \sum_{k} exp(ik.r_{i})c^{+}_{k} \tag{3.29}$$

and

$$b_{j} = N^{\frac{-1}{2}} \sum_{k'} exp(ik'.r_{j})d_{k'} \quad and \quad b^{+}_{j} = N^{\frac{-1}{2}} \sum_{k'} exp(-ik'.r_{j})d^{+}_{k'}. \tag{3.30}$$

Then,

$$\begin{aligned}
\overrightarrow{S_{i}}.\overrightarrow{S_{j}} &= \overrightarrow{S_{ai}}.\overrightarrow{S_{bj}} \\
&= (S^{a}_{ix}i + S^{a}_{iy}j + S^{a}_{iz}k).(S^{b}_{jx}i + S^{b}_{jy}j + S^{b}_{jz}k) \\
&= [(\tfrac{S^{+}_{ai}+S^{-}_{ai}}{2})i + (\tfrac{S^{+}_{ai}-S^{-}_{ai}}{2i})j + S^{a}_{iz}k].[(\tfrac{S^{+}_{bj}+S^{-}_{bj}}{2})i + (\tfrac{S^{+}_{bj}-S^{-}_{bj}}{2i}) + S^{b}_{jz}k] \\
&= (\tfrac{S^{+}_{ai}+S^{-}_{ai}}{2})(\tfrac{S^{+}_{bj}+S^{-}_{bj}}{2}) + (\tfrac{S^{+}_{ai}-S^{-}_{ai}}{2i})(\tfrac{S^{+}_{bj}-S^{-}_{bj}}{2i}) + S^{a}_{iz}S^{b}_{jz} \ .
\end{aligned}$$

Therefore,

$$\overrightarrow{S_{i}}.\overrightarrow{S_{j}} = \frac{1}{2}(S^{+}_{ai}S^{-}_{bj} + S^{-}_{ai}S^{+}_{bj}) + S^{a}_{iz}S^{b}_{jz}. \tag{3.31}$$

Now using the first transformation given by (3.21-3.26), we find that the first term of

the Hamiltonian (3.2) reduces to:

$$H_{ex} = \sum_{i,j} J(\overrightarrow{r}_{ij})[\frac{1}{2}(S^{+}_{ai}S^{-}_{bj} + S^{-}_{ai}S^{+}_{bj}) + S^{a}_{iz}S^{b}_{jz}] \tag{3.32}$$

and substituting the values for S^{a}_{iz} and S^{b}_{jz} the next two terms of equation (3.2) becomes;

$$H_{zeeman} = -g\mu_B H_A \sum_{i}(S - a^{+}_{i}a_i) - g\mu_B H_A \sum_{j}(S - b^{+}_{j}b_j). \tag{3.33}$$

To reduce expressions (3.32) and (3.33) to a much simpler, and thus solvable form, we made the following approximations: If the magnetization is near saturation, we expect that

$\frac{1}{2}\frac{M_o - M(T,H)}{M_o} \ll 1$ or $\frac{S - S_z}{2S} \ll 1$ or $\frac{\ll n_i \gg}{2S} = \frac{\ll a^{+}a_i \gg}{2S} \ll 1$.

Hence

1,$(1 - \frac{a^{+}_{i}a_i}{2S})^{\frac{1}{2}} \simeq 1$ similarlly $(1 - \frac{b^{+}_{j}b_j}{2S})^{\frac{1}{2}} \simeq 1$

2,$a^{+}_{i}a_i b^{+}_{j}b_j = n_i n_j \simeq 0$

3,$(2S)^{\frac{1}{2}}a^{+}_{i}a_i b_j = (2S)^{\frac{1}{2}}n_i b_j \ll 1$.

With these approximations equations (3.21-3.26) become

$S^{+}_{ai} \simeq (2S)^{\frac{1}{2}}a_i$; $S^{-}_{ai} \simeq (2S)^{\frac{1}{2}}a^{+}_{i}$ and $S^{+}_{bj} \simeq (2S)^{\frac{1}{2}}b^{+}_{j}$; $S^{-}_{bj} \simeq (2S)^{\frac{1}{2}}b_j$ and $S^{a}_{iz} = S - a^{+}_{i}a_i$; $-S^{b}_{jz} = S - b^{+}_{j}b_j$.

Substituting these in equation (3.32) we get

$$\begin{aligned}
H_{ex} &= \textstyle\sum J(\overrightarrow{r}_{ij})[\frac{1}{2}(S^{+}_{ai}S^{-}_{bj} + S^{-}_{ai}S^{+}_{bj}) + S^{a}_{iz}S^{b}_{jz}] \\
&= \textstyle\sum J(\overrightarrow{r}_{ij})[\frac{1}{2}((2S)^{\frac{1}{2}}a_i(2S)^{\frac{1}{2}}b_j + (2S)^{\frac{1}{2}}a^{+}_{i}(2S)^{\frac{1}{2}}b^{+}_{j}) - (S - a^{+}_{i}a_i)(S - b^{+}_{j}b_j)] \\
&= \textstyle\sum J(\overrightarrow{r}_{ij})[\frac{1}{2}(2Sa_i b_j + 2Sa^{+}_{i}b^{+}_{j}) - (S^2 - Sb^{+}_{j}b_j - Sa^{+}_{i}a_i + a^{+}_{i}a_i b^{+}_{j}b_j)] \\
&= \textstyle\sum J(\overrightarrow{r}_{ij})(Sa_i b_j + Sa^{+}_{i}b^{+}_{j} - S^2 + Sb^{+}_{J}b_j + Sa^{+}_{i}a_i - a^{+}_{i}a_i b^{+}_{j}b_j) \\
&= -\textstyle\sum J(\overrightarrow{r}_{ij})S^2 + \sum J(\overrightarrow{r}_{ij})S(a_i b_j + a^{+}_{i}b^{+}_{j} + b^{+}_{j}b_j + a^{+}_{i}a_i) - \sum J(\overrightarrow{r}_{ij})a^{+}_{i}a_i b^{+}_{j}b_j \ .
\end{aligned}$$

We can ignore the third term by the reason (2) above. That is when the excitation is low. Then the above equation becomes

$$H_{ex} = -\sum_{i,j} J(\overrightarrow{r}_{ij})S^2 + \sum_{i,j} J(\overrightarrow{r}_{ij})S(a_i b_j + a^{+}_{i}b^{+}_{j} + b^{+}_{j}b_j + a^{+}_{i}a_i). \tag{3.34}$$

Now we can write equation (3.33) in the following form

$$H_{zeeman} = -g\mu_B H_A \sum_i S_i - g\mu_B H_A \sum_j S_j + g\mu_B H_A \sum_i a_i^+ a_i + g\mu_B H_A \sum_j b_j^+ b_j. \quad (3.35)$$

Hence the total Hamiltonian can be written as

$H = H_o + H_{mag}$

where H_o and H_{mag} (Hamiltonian of the magnons) are given as follows

$$H_o = -\sum_{i,j} J(r_{ij})S^2 - g\mu_B H_A \sum_i S_i - g\mu_B H_A \sum_j S_j \quad (3.36)$$

$$and \quad H_{mag} = \sum_{i,j} J(l)S\{a_i b_j + a_i^+ b_j^+ + b_j^+ b_j + a_i^+ a_i\} + g\mu_B H_A\{\sum_i a_i^+ a_i + \sum_j b_j^+ b_j\}. \quad (3.37)$$

Substituting equations (3.29) and (3.30) on equation (3.37) we obtain

$$\begin{aligned} H_{mag} = \sum_{i,l} \frac{J(l)S}{N} [\sum_{k,k'} exp\{-i(k-k').r_i\} exp(ik'.l) c_k d_{k'} + \sum_{k,k'} exp\{i(k-k').r_i\} exp(-ik'.l) c_k^+ d_{k'}^+ \\ + \sum_{k'} exp\{i(k'-k').(r_i+l)\} d_{k'}^+ d_{k'} + \sum_k exp\{i(k-k).r_i\} c_k^+ c_k] \\ + g\mu_B H_A [\sum_{i,k} exp\{i(k-k).r_i\} c_k^+ c_k + \sum_{j,k'} exp\{i(k'-k').r_j\} d_{k'}^+ d_{k'}]. \end{aligned} \quad (3.38)$$

But we have

$$\sum_{kk'} exp\{-i(k-k').r_i\} = \delta_{kk'}$$

where $\delta_{kk'} = \begin{cases} 1, & \text{if } k = k'; \\ 0, & \text{otherwise.} \end{cases}$

Hence the Hamiltonian of the magnon becomes

$$\begin{aligned} H_{mag} = 2\sum_l J(l) Sz \sum_k [exp(ik.l) c_k d_k + exp(-ik.l) c_k^+ d_k^+ + d_k^+ d_k + c_k^+ c_k] \\ + g\mu_B H_A \sum_k [c_k^+ c_k + d_k^+ d_k]. \end{aligned} \quad (3.39)$$

Now taking the term $\sum J(l)$ and considering contact type of interaction we get the

following

$$\sum_l J(l) = \sum_j J(\overrightarrow{r}_{ij}) = J\sum_j \delta(\overrightarrow{r}_{ij}) = J\sum_j \delta(\overrightarrow{r}_i - \overrightarrow{r}_j). \tag{3.40}$$

Taking the average over j we obtain

$$< \sum_j \delta(\overrightarrow{r}_i - \overrightarrow{r}_j) >= x \tag{3.41}$$

where x is the concentration of the impurities added on the semiconductor. Therefore equation (3.39) becomes

$$H_{mag} = 2JSxz\sum_k [\gamma_k c_k d_k + \gamma_{-k} c_k^+ d_k^+ + d_k^+ d_k + c_k^+ c_k] + g\mu_B H_A \sum_k [c_k^+ c_k + d_k^+ d_k] \tag{3.42}$$

where z is the number of nearest neighbors. The above equation can be written as

$$H_{mag} = 2JSxz\sum_k [\gamma_k (c_k d_k + c_k^+ d_k^+) + c_k^+ c_k + d_k^+ d_k] + g\mu_B H_A \sum_k (c_k^+ c_k + d_k^+ d_k) \tag{3.43}$$

where

$$\gamma_k = \frac{1}{z}\sum_l exp(ik.l) = \gamma_{-k} \tag{3.44}$$

assuming a center of symmetry. We now look for a transformation to diagonalize H_{mag}. We transform to new creation and annihilation operators $\alpha^+, \alpha; \beta^+, \beta$ with $[\alpha_k, \alpha_k^+] = 1; [\beta_k, \beta_k^+] = 1; [\alpha_k, \beta_k] = 0$, etc. Then the transformation is defined by

$$\alpha_k = \upsilon_k c_k - \nu_k d_k^+; \qquad \alpha_k^+ = \upsilon_k c_k^+ - \nu_k d_k \tag{3.45}$$

and

$$\beta_k = \upsilon_k d_k - \nu_k c_k^+; \qquad \beta_k^+ = \upsilon_k d_k^+ - \nu_k c_k \tag{3.46}$$

here υ_k, ν_k are real and satisfy $\upsilon_k^2 - \nu_k^2 = 1$. Therefore the magnon eigenfrequencies ω_k are given by

$$\omega_k^2 = (\omega_e + \omega_A)^2 - \omega_e^2 \gamma_k^2 \tag{3.47}$$

with $\omega_e = 2JSxz; \quad \omega_A = g\mu_B H_A$.
There are thus two degenerate branches for each k, one associated with the α operator and the other with the β operators. That is each value of k is to be counted twice because of the double degeneracy. If we neglect ω_A and take $l = a$,

$$\omega_k^2 = \omega_e^2 - \omega_e^2\gamma_k^2 = \omega_e^2(1 - \gamma_k^2)$$

$$\omega_k = \omega_e(1 - \gamma_k^2)^{\frac{1}{2}}. \tag{3.48}$$

Assuming that $ka \ll 1$, then $(1 - \gamma_k^2)^{\frac{1}{2}} \approx \frac{ka}{\sqrt{3}}$; for a simple cubic lattice (z=6), and assuming $\hbar = 1$ we have finally

$$\omega_k \approx 4\sqrt{3}JSkax. \tag{3.49}$$

If we consider the external magnetic field, the dispersion relation in general case is given by

$$\hbar\omega_k^{\pm} = 2\sqrt{2z}JSkax \pm g\mu_B H_A. \tag{3.50}$$

This is the dispersion law for antiferromagnets in the long wave length limit.

3.2 Green's function formalism

The retarded and advanced Green's functions $G_r(t, t')$ is defined as

$$G_r(t, t') = \ll A(t), B(t') \gg_r = -i\theta(t - t') < [A(t), B(t')] > \tag{3.51}$$

where $\ll ... \gg_r$ is the abbreviated notations for the corresponding Green's function, and $< ... >$ denotes averaging over a ground canonical ensemble. This is appropriate since the number of particles is not constant and $\theta(t)$ is the step function, that is,

$$\theta(t) = \begin{cases} 0, & \text{for } t < 0; \\ 1, & \text{for } t > 0. \end{cases} \tag{3.52}$$

Also $[A, B]$ is a commutator or anticommutator, that is,

$$[A, B] = AB - \eta BA \quad , \quad \eta = \pm 1. \tag{3.53}$$

The sign of η is positive if A and B are both Bose operators and negative if they are Fermi operators. In general, A and B are neither Bose nor Fermi operators since products of operators can satisfy more complicated commutation relations. The sign of η is chosen by considering what is most convenient for the problem.Using equation (3.53),we can write equation (3.51) as

$$G_r(t,t') = -i\theta(t,t')[< A(t)B(t') > -\eta < B(t')A(t) >]. \tag{3.54}$$

We note from (3.52) and (3.54) that $G_r(t,t') \neq 0$ when $t' < t$, $G_r(t,t') = 0$ when $t' > t$, and $G_r(t,t')$ is not defined when $t = t'$, because of the discontinuity of $\theta(t)$ at t=0. One important property of $G_r(t,t')$ is that it depends only on the difference $(t - t')$ in the case of statistical equilibrium. Now taking the operators $A = a_k$ and $B = a_k^+$, to relate with our problem we obtain

$$G_{kk'}(t,t') = \ll a_k, a_k^+ \gg = -i\theta(t-t') < [a_k(t), a_{k'}^+(t')] > . \tag{3.55}$$

Differentiating the above equation with respect to t, we get

$$\frac{d}{dt}G_{kk'}(t,t') = -i\delta(t-t') < [a_k(t), a_{k'}^+(t')] > -i\theta(t-t') < [\frac{d}{dt}a_k(t), a_{k'}^+(t')] > \tag{3.56}$$

and multiplying both sides of the above equation by i it becomes

$$i\frac{d}{dt}G_{kk'}(t,t') = \delta(t-t') < [a_k(t), a_{k'}^+(t')] > -i\theta(t-t') < [i\frac{d}{dt}a_k(t), a_{k'}^+(t')] > \tag{3.57}$$

where $i\frac{d}{dt}a_k(t) = [a_k(t), \hat{H}]$,here $\hat{H} = \hat{H}_{mag}$.
Taking

$$\hat{H} = \sum_k \omega_k a_k^+ a_k$$

and for our convenience let us drop the time (t,t') from $a_k(t)$ and $a_{k'}^+(t')$ then we will have

$$i\frac{d}{dt}a_k = [a_k, H] = [a_k, \sum_p \omega_p a_p^+ a_p]$$

$$i\frac{d}{dt}a_k = \sum_p \omega_p [a_k, a_p^+ a_p] = \sum_p \omega_p \{[a_k, a_p^+]a_p + a_p^+[a_k, a_p]\}$$

but the second term becomes zero. Then we have only

$$i\frac{d}{dt}a_k = \sum_p \omega_p [a_k, a_p^+] a_p = \sum_p \omega_p \delta_{kp} a_p.$$

But remember that $\delta_{kp} = \begin{cases} 1, & \text{if } k = p; \\ 0, & \text{otherwise.} \end{cases}$ Then we will get

$$i\frac{d}{dt}a_k = \omega_k a_k. \tag{3.58}$$

Substituting equation (3.58) in to equation (3.57), we obtain

$i\frac{d}{dt}G_{kk'} = \delta(t-t') < [a_k(t), a_{k'}^+(t')] > -i\theta(t-t') < [\omega_k a_k(t), a_{k'}^+(t')] >$

$= \delta(t-t') < [a_k(t), a_{k'}^+(t')] > -i\theta(t-t')\omega_k < [a_k(t), a_{k'}^+(t')] >.$

Using the Fourier transformation we get

$$\omega G_{kk'} = \frac{\delta_{kk'}}{2\pi} + \ll [a_k, H], a_{k'}^+ \gg_\omega \tag{3.59}$$

equation (3.59) follows since the Fourier transform of the δ function is $\frac{1}{2\pi}$. But we have got $[a_k, H] = \omega_k a_k$.

Hence $\omega G_{kk'} = \frac{\delta_{kk'}}{2\pi} + \ll \omega_k a_k, a_{k'}^+ \gg_\omega$.

For $k = k'$, we have $\delta_{kk'} = 1$, and then the above equation becomes

$$\omega G_{kk'} = \frac{1}{2\pi} + \omega_k \ll a_k, a_{k'}^+ \gg_\omega . \tag{3.60}$$

But from equation (3.51) we have the following relation

$\ll a_k, a_{k'}^+ \gg = G_{kk'}$.

Therefore equation (3.60) becomes

$\omega G_{kk'} = \frac{1}{2\pi} + \omega_k G_{kk'}$.

From this we obtain

$$G_{kk'} = \frac{1}{2\pi(\omega - \omega_k)} \tag{3.61}$$

$\omega - \omega_k = 0$ gives us the dispersion relation. From the analytical properties of the Green functions it follows that the correlation function $< a_k^+ a_k >$ can be obtained from the

equation

$$< a_k^+ a_k >= lim_{\delta \to 0} i \int_{-\infty}^{\infty} (\frac{\ll a_k, a_k^+ \gg_{\omega + i\delta} - \ll a_k, a_k^+ \gg_{\omega - i\delta}}{exp(\beta\omega) - 1}) exp(-i\omega\{t - t'\}) d\omega. \quad (3.62)$$

Taking $t = t'$, equal time correlation gives the number operator;

$$\ll a_k, a_k^+ \gg_{\omega + i\delta} = \frac{1}{2\pi[(\omega + i\delta) - \omega_k]} = \frac{1}{2\pi}[\frac{p}{\omega - \omega_k} - i\pi\delta(\omega - \omega_k)] \quad (3.63)$$

$$\ll a_k, a_k^+ \gg_{\omega - i\delta} = \frac{1}{2\pi[(\omega - i\delta) - \omega_k]} = \frac{1}{2\pi}[\frac{p}{\omega - \omega_k} + i\pi\delta(\omega - \omega_k)] \quad (3.64)$$

where p is the principal part of the integral. Therefore equation (3.62) becomes,
$< a_k^+ a_k >= lim_{\delta \to 0} i \int \frac{\delta(\omega - \omega_k)}{exp(\omega\beta) - 1} d\omega$

$$< a_k^+ a_k >= \frac{1}{exp(\omega_k \beta) - 1}. \quad (3.65)$$

Substituting equation (3.49) in to equation (3.65) for ω_k and taking $\beta = \frac{1}{k_B T}$ we get:

$$< a_k^+ a_k >= \frac{1}{exp(\frac{4\sqrt{3}JSaxk}{k_B T}) - 1}. \quad (3.66)$$

Since magnons are bosons, the above equation is the Bose-Einstein distribution which magnons obey. Now let us determine the total number of magnons that are excited at a temperature T, which is given by

$$\sum_k < n_k >= \frac{1}{(2\pi)^3} \int_0^{\infty} \frac{4\pi k^2 dk}{exp(\frac{4\sqrt{3}JSxka}{k_B T}) - 1}$$

$$\sum_k < n_k >= \frac{1}{2\pi^2} \int_0^{\infty} \frac{k^2}{exp(\frac{4\sqrt{3}JSxka}{k_B T}) - 1} dk \quad (3.67)$$

Let $y = \frac{4\sqrt{3}JSxka}{k_B T}$ and $dy = \frac{4\sqrt{3}JSxa}{k_B T} dk$ solving for k, we get $k = \frac{k_B T}{4\sqrt{3}JSxa} y$;
$dk = \frac{k_B T}{4\sqrt{3}JSxa} dy$.
Substituting these in equation (3.67) we obtain

$$\sum_k < n_k >= \frac{1}{2\pi^2} (\frac{k_B T}{4\sqrt{3}JSxa})^3 \int_0^{\infty} \frac{y^2}{e^y - 1} dy.$$

The integration of $\int_0^\infty \frac{y^2}{e^y-1}dy = 2 \times zeta(3) \approx 2 \times 1.202 \approx 2.4$.
Therefore the total number of magnons is

$$\sum_k < n_k >= 0.122(\frac{k_BT}{4\sqrt{3}JSxa})^3. \tag{3.68}$$

3.3 Magnetization and transition temperature for antiferromagnets

The sublattice magnetization at temperature T is given by

$$M(T) = g\mu_B \sum_l (S - a_l^+ a_l). \tag{3.69}$$

Using the spin-wave canonical transformation we get the following

$$M(T) = g\mu_B(nS - \sum_k \sinh^2\theta_k - \sum_k < \alpha_k^+ \alpha_k > \cosh 2\theta_k). \tag{3.70}$$

The second term of equation (3.70) gives the zero-point contribution to the sublattice spin deviation. It can be shown, on neglecting the Zeeman term, that

$$\Delta M_{zeropoint} = g\mu_B \sum_k \sinh^2\theta_k = \frac{1}{2} g\mu_B \sum_k \{(1-\gamma_k^2)^{-\frac{1}{2}} - 1\}. \tag{3.71}$$

For a simple cubic lattice this turns out to be $0.078 g\mu_B n$. The third term of equation (3.70) gives the temperature dependent part. Taking $M(0) = g\mu_B nS$ which is ground state magnetization and neglecting the second term of equation (3.70) we obtain

$$\begin{aligned} M(T) &= M(0)(1 - \tfrac{1}{nS}\textstyle\sum_k < n_k > \cosh 2\theta_k) \\ &= M(0)(1 - \tfrac{1}{nS}\textstyle\sum_k \frac{1}{exp(\frac{\omega_k}{K_BT})-1}\cosh 2\theta_k) \\ &= M(0)[1 - \tfrac{1}{nS}\textstyle\sum_k (1-\gamma_k^2)^{-\frac{1}{2}}(\frac{1}{exp(\frac{\omega_k}{K_BT})-1})]. \end{aligned}$$

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

$$\frac{M(T)}{M(0)} = 1 - \frac{\sqrt{3}}{naS} \sum_k \frac{1}{k[exp(\frac{\omega_k}{k_BT}) - 1]}. \tag{3.72}$$

Now we use

$$\sum_k \frac{1}{k[exp(\frac{\omega_k}{k_BT})-1]} = \frac{1}{(2\pi)^3}\int_0^{k_{max}} \frac{d^3k}{k[exp(\frac{\omega_k}{k_BT})-1]}$$

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

$$\sum_k \frac{1}{k[exp(\frac{\omega_k}{k_BT})-1]} = \frac{1}{(2\pi)^3}\int_0^{\infty} \frac{4\pi k^2 dk}{k[exp(\frac{\omega_k}{k_BT})-1]} = \frac{1}{2\pi^2}\int_0^{\infty} \frac{kdk}{exp(\frac{4\sqrt{3}JSaxk}{k_BT})-1}$$

let $y = \frac{4\sqrt{3}JSaxk}{k_BT}$; $\qquad k = \frac{k_BT}{4\sqrt{3}JSax}y$; $\qquad dk = \frac{k_BT}{4\sqrt{3}JSax}dy$

$$\sum_k \frac{1}{k[exp(\frac{\omega_k}{k_BT})-1]} = \frac{1}{2\pi^2}(\frac{k_BT}{4\sqrt{3}JSax})^2\int_0^{\infty} \frac{ydy}{e^y-1}.$$

The value of the integration $\int_0^{\infty} \frac{ydy}{e^y-1} = \frac{\pi^2}{6}$.
Hence substituting all these values in equation (3.72) we will get

$$\frac{M(T)}{M(0)} = 1 - \frac{\sqrt{3}}{12NS}(\frac{k_BT}{4\sqrt{3}JSx})^2. \tag{3.73}$$

But when $\frac{M(T)}{M(0)}$ goes to zero, T approaches to T_N which gives

$$T_N = (48)^{\frac{3}{4}}(\frac{JS}{k_B})(NS)^{\frac{1}{2}}x. \tag{3.74}$$

3.4 Magnon heat capacity in antiferromagnets

We have seen that in the absence of an external magnetic field the two magnon branches are degenerate, i.e,
$\omega_k^{\pm} = 2\sqrt{2z}JSkxa$.
Hence the internal magnon energy for antiferromagnets can be written as

$$U = \sum_k \omega_k < n_k > \tag{3.75}$$

which can be written as

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

$$U = \frac{1}{(2\pi)^3}\int_0^{k_{max}} \frac{\omega_k 4\pi k^2 dk}{exp(\omega_k\beta) - 1} = \frac{1}{2\pi^2}\int_0^{k_{max}} \frac{\omega_k k^2 dk}{exp(\omega_k\beta) - 1}$$

$$U = \frac{1}{2\pi^2}\int_0^{k_{max}} \frac{4\sqrt{3}JSxak^3 dk}{exp(\frac{4\sqrt{3}JSkxa}{k_BT}) - 1}.$$

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

$$U = \frac{1}{2\pi^2}\int_0^{\infty} \frac{4\sqrt{3}JSxak^3 dk}{exp(\frac{4\sqrt{3}JSkxa}{k_BT}) - 1} = \frac{4\sqrt{3}JSxa}{2\pi^2}\int_0^{\infty} \frac{k^3 dk}{exp(\frac{4\sqrt{3}JSkxa}{k_BT}) - 1}. \tag{3.76}$$

Let $y = \frac{4\sqrt{3}JSkxa}{k_BT}$ and $dy = \frac{4\sqrt{3}JSxa}{k_BT}dk$
solving for k, we get $k = \frac{k_BT}{4\sqrt{3}JSxa}y$ and $dk = \frac{k_BT}{4\sqrt{3}JSxa}dy$.
Substituting these in equation (3.72) we get

$$U = \frac{k_BT}{2\pi^2}(\frac{k_BT}{4\sqrt{3}JSxa})^3\int_0^{\infty} \frac{y^3}{e^y - 1}dy \tag{3.77}$$

but the value of the integral $\int_0^{\infty} \frac{y^3}{e^y-1} = \frac{\pi^4}{15}$.
Therefore the internal magnon energy for antiferromagnets is

$$U = \frac{\pi^2}{30}\frac{(k_BT)^4}{(4\sqrt{3}JSxa)^3}. \tag{3.78}$$

Hence the magnon heat capacity is given by

$$C_m = \frac{\partial U}{\partial T} = \frac{4\pi^2}{30}\frac{k_B(k_BT)^3}{(4\sqrt{3}JSxa)^3}. \tag{3.79}$$

Thus the predicted magnon part of the heat capacity is proportional to T^3 which is similar to the Debye phonon heat capacity.

Chapter 4

Results and discussion

In the previous chapter we have obtained expressions of magnetization, specific heat and transition temperature for antiferromagnetic magnons based on spin wave theory using the Holstein-Primakoff transformations. In addition to these we have also calculated the number of magnons excited in the mode k at a temperature T using Green's function. To calculate all of the above quantities first we have got the dispersion relation for magnons in an antiferromagnet. The dispersion relation for magnons in an antiferromagnet is linear in k which is quite different from that of magnons in a ferromagnet which is quadratic in k.

In this chapter we will try to generalize the main results obtained in this work and investigate the results with the help of figures.

4.1 Dependence of magnetization on temperature

The reduced magnetization as a function of temperature can be computed by noting that each spin-wave mode leads to one spin reversal distributed coherently through out the entire lattice. We have got the theoretical formalism describing the dependence of reduced magnetization due to thermal spin-wave excitations on temperature as follows:

$$\frac{M(T)}{M(0)} = 1 - \frac{\sqrt{3}}{12NS}(\frac{k_B}{4\sqrt{3}JSx})^2T^2$$

which shows that the sublattice magnetization decreases quadratically with temperature in the low temperature region and it agrees with the expression obtained by

Sinha et.al., 1980 [36].

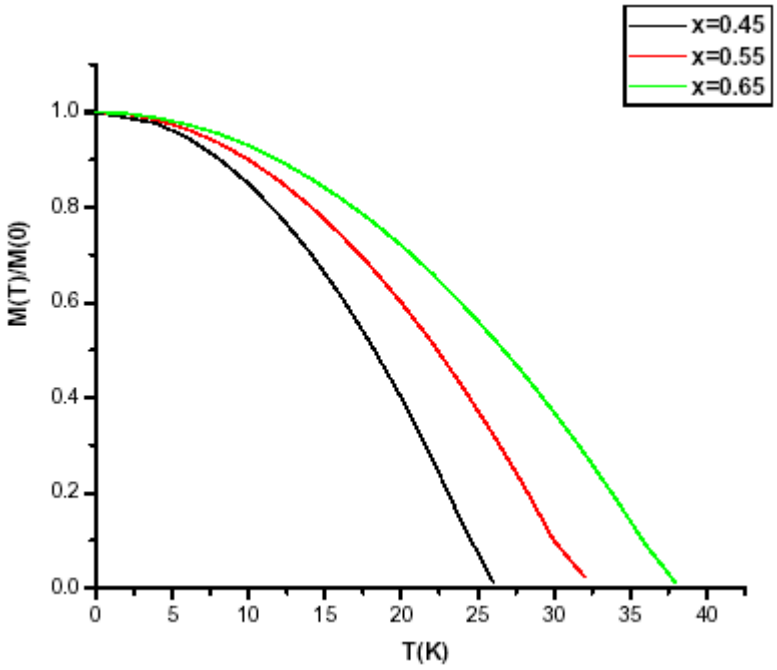

Figure 4.1: *Reduced magnetization VS temperature when values of x are kept constant* .

As we have seen from the figure Curves are slightly different for different values of the concentration (x) of the manganese ion, however, they all have a convex upward shape. The reduced magnetization versus temperature curve is obtained for $x = 0.45,\ 0.55\ and\ 0.65$. The calculated curves fit excellently well to the experimental results obtained by C. Marino et.al., 2000 [37].

4.2 Dependence of Neel temperature on concentration

A distinct magnon feature is observed in $Cd_{1-x}Mn_xTe$ for the composition range $0.40 \leq x \leq 0.70$. That is, there is a magnetically ordered phase for this range, which is antiferromagnetic ordering. In the previous chapter we have obtained the expression

for the Neel temperature which is given by:

$$T_N = (48)^{\frac{3}{4}} \frac{JS}{k_B} (NS)^{\frac{1}{2}} x$$

Using the above formula and taking the constants S=5/2 for Mn^{2+}, N=4 nearest neighbors, the exchange integral for antiferromagnets is around $|J| \approx 10^{-3} ev$, but in our case it is calculated using equation (2.2) taking the lattice constant for CdTe $a = 6.48 \mathring{A}$ and the manganese concentration is 0.70. Hence the theoretical value of the Neel temperature for CdMnTe is 42K. However, the highest Neel temperature, T_N, obtained to date is about 40K in $Cd_{1-x}Mn_xTe$ for $x = 0.70$ [21].

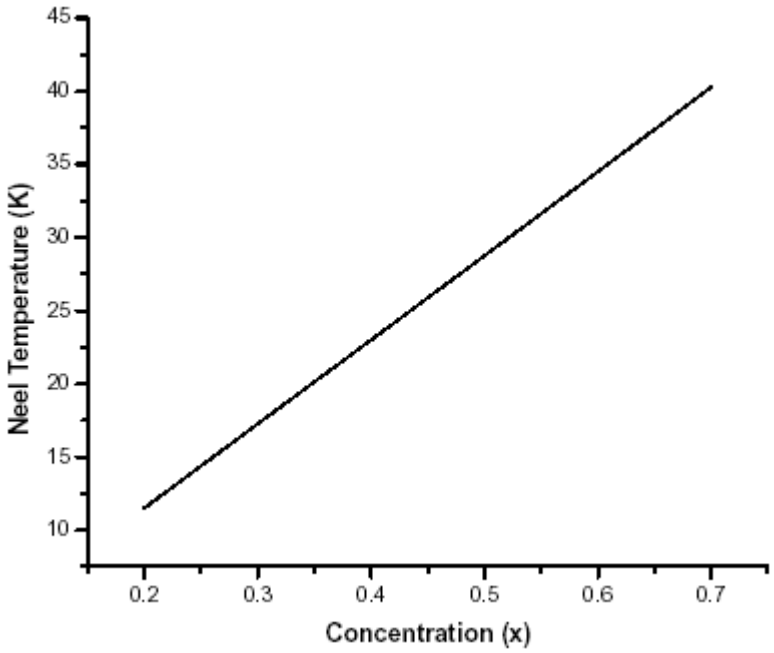

Figure 4.2: *Neel temperature versus concentration* .

As we have seen from the figure the plot of Neel temperature versus manganese ion concentration is linear. But antiferromagnetic ordering is observed when the composition is in the range of $0.4 \leq x \leq 0.7$. This shows the Neel temperature for CdMnTe increases when the concentration of manganese ion increases in the range given above. The calculated curve fits excellently to the experimental result obtained by A.Twardowski [16].

4.3 Dependence of specific heat on temperature

The detailed antiferromagnetic magnon specific heat capacity per unit volume versus temperature T for CdMnTe will be shown in this work with in the temperature range of 0K and 40K. To derive the specific heat we neglect magnon-magnon interactions, which is justifiable in the very-low-temperature regions.We have seen that magnons behave like Bose particles. Thus the internal energy of a system of magnons in thermal equilibrium at temperature T using equation (3.75) is given by

$$U = \frac{\pi^2}{30} \frac{(k_B T)^4}{(4\sqrt{3} J S x a)^3}$$

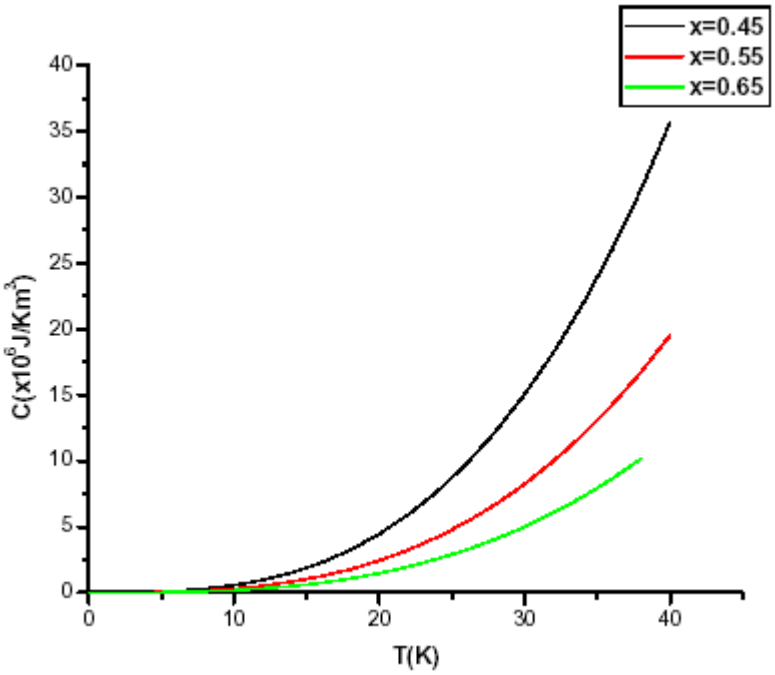

Figure 4.3: *Heat capacity per unit volume VS temperature when values of x are kept constant* .

The Zeeman contribution to the internal energy is relatively small compared with the exchange term. Taking the derivative of the above equation with respect to T, we get

the specific heat capacity per unit volume, which is

$$C = \frac{4\pi^2}{30} \frac{k_B (k_B T)^3}{(4\sqrt{3} JSxa)^3}$$

Because U goes to zero just at T_N and vanishes at all temperatures above it, there is no latent heat associated with the disappearance of the magnetization [38]. Thus the heat capacity for an antiferromagnet in the low-temperature region arising mainly from magnons is proportional to T^3, which is the same as the expression found in QTS [39].

What we have seen from the figure is that specific heat increases when temperature increases keeping the concentration of manganese ion constant. However, the specific heat of the magnons decreases as the manganese ion concentration increases.

Chapter 5

Conclusion

The theoretical results obtained compared with the experimental results (reduced magnetization and Neel temperature) and the graph of these quantities matches with the experimental graphs. The magnetization increases rapidly with decreasing temperature if the concentration is kept constant and it reaches the saturation sublattice magnetization when the temperature goes to zero. But above T_N the sublattice magnetization vanishes. Generally from the mathematical expression we can see that the sublattice magnetization decreases quadratically with temperature in the low-temperature region.

Magnetic II-VI semiconductors show antiferromagnetism, whose Neel temperature, T_N, is relatively high as predicted theoretically only when the concentration is effectively raised. The Neel temperature for CdMnTe increases when the concentration of manganese ion increases in the range $0.4 \leq x \leq 0.7$. Hence $Cd_{1-x}Mn_xTe$ is found to be antiferromagnetic with the antiferromagnetic transition temperature, T_N=23-40K for $x = 0.4 - 0.7$ where the values of x corresponding the highest T_N is 0.7.

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. The specific heat capacity vanishes above the Neel temperature. However, the heat capacity per unit volume decreases when manganese ion concentration increases.

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Declaration

I, the undersigned, declare that this thesis is my original work and has not been presented for a degree in any other university and all sources of material used for the thesis have been duly acknowledged.

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This thesis has been submitted for examination with my approval as university advisor.

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