PHOTO INDUCED FERROMAGNETISM AND MECHANISM OF FERROMAGNETISM IN DILUTED MAGNETIC SEMICONDUCTOR

By

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Abstract

Conventional electronic devices rely on manipulating charge to produce desired functions, spintronic devices would manipulate both the charge flow and electron spin within that flow. This would add an extra degree of freedom to microelectronics and usher in the era of truly nanoelectronic devices. Research aimed at a whole new generation of electronic devices is underway by introducing electron spin as a new or additional physical variable, and semiconductor devices that exploit this new freedom will operate faster and more efficiently than conventional microelectronic devices and offer new functionality that promises to revolutionize the electronics industry.

In order to enable electronic devices in active part of operation 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.

In this thesis we study a photo induced ferromagnetism and mechanism of ferromagnetism in diluted magnetic semiconductors by solving a Hamiltonian model that consists of localized magnetic moments interacting with photoexcited carriers. The mechanism for photo-induced ferromagnetism is coherence between conduction and valence bands induced by the light which leads to an optical exchange interaction. When light is incident on the diluted magnetic semiconductors, electron and holes are created across the band gap. Photo excited carriers mediated a ferromagnetic interaction between the localized moments resulting in ferromagnetic state in the range of critical temperature. The situation is similar to the famous Rabi problem of a two state system coupled to time-dependent oscillating electric field. The time dependance of the light-matter interaction term is eliminated by a unitary transformation and the resulting Hamiltonian is solved by making a Bogoliubov-Valtain transformation. Since the system of electrons and holes in contact with the photon bath is considered in a steady state, we calculate the free energy of the system. Starting the free energy again we calculate the magnetization of the system in self-consistent mean field way. The magnetization and magnitude of T_c is determined by the photon energy incident on the system. By increasing the light coupling and frequency of the light, the transition temperature is increased.

Introduction

The integrated circuits and the high frequency devices of semiconductors use the charge of electrons for information processing while for storage of information is done by magnetic recording using spin of electrons in a ferromagnetic metal[1]. But for the future, information technology will use spin and charge combined in one device that exploits both charge and spin to process and store the information. That means we may be able to use the capability of storage and processing of information in the same devices. Such a device will be called spintronics[2]. One of the approaches to drive a semiconductor ferromagnetic is to introduce magnetic atoms like Mn, Cr, Co and Fe into non-magnetic semiconductors. In these ferromagnetic semiconductors, a part of the lattice is made up of substitution magnetic atoms. Hence they are called diluted magnetic semiconductors[3]. Diluted magnetic semiconductors have a great deal of attention for their potential in combining ferromagnetism and semiconductors in single material. There are semiconductors in nature, which have a periodic array of magnetic elements, such as Europium chalcogenides and semiconducting spinels but their crystal structure is quite different from that of other semiconductors (Si, Ge, GaAs). Also their crystal growth is very difficult. Hence they are not ideal for spintronics application[4].

In recent years there has been an extensive research towards introducing ferromagnetic property at room temperature in semiconductors to realize a new class of spintronics devices such as spin valve, transistors, spin light emitting diodes, magnetic sensors, non-volatile memory, logic devices, optical isolators and ultra-fast optical switches. The potential advantages of spintronics devices will be higher speed, greater efficiency, better stability and low energy required to flip a spin[5].

Diluted magnetic semiconductors where transition metal atoms are introduced into the cationic sites of the semiconducting host lattice, have recently attracted increasing attention because of their potential use in spintronic devices. A key factor to realize spintronic devices is to develop ferromagnetic DMSs with a Curie temperature above room temperature. Interest in ZnO-based DMSs was initially generated by the theoretical prediction that above room temperature ferromagnetism is possible in ZnO-based DMSs. One of the systems considered in the past is ZnO doped with Co. The ferromagnetic behavior with the Curie temperature higher than 280 K for films grown by the pulsed-laser deposition method are reported, but with less than 10% reproducibility. Intrinsic ferromagnetic behaviour of Co-doped ZnO films with a Curie temperature of approximately 350 K[6]. Recent reports of room temperature ferromagnetism in Co dopped to TiO_2 films have therefore spurred intense research activities. Ferromagnetism has now been observed in numerous conducting oxide semiconductors including TiO_2 and ZnO doped with various transition metal ions and with surprisingly high magnetic ordering temperature[7].

On this thesis we study photo induced ferromagnetism and mechanism of ferromagnetism in diluted magnetic semiconductors by solving a Hamiltonian model that consists of localized magnetic moments interacting with the photoexcited itinerant carriers. The spin states of the itinerant carriers are spilt due to the interaction with the localized magnetic moments, which are assumed to be in thermal equilibrium in the local magnetic field due to the carriers. When light falls on the dilute magnetic semiconductors, it creates electro-hole pairs across the band gap. These carriers interact with the impurity magnetic atoms and so induced ferromagnetic coupling among magnetic moments and thereby create a ferromagnetic state in these systems when temperature is lowered.

The first chapter of this thesis starts with importance of spintronics and a brief introduction to diluted magnetic semiconductors, the magnetic interactions that may be relevant for these materials and discuss the hole mediated ferromagnetism in more detail.

A brief overview is given of theoretical models that describe the origin of the ferromagnetism.

The second chapter discuss photo induced ferromagnetism in diluted magnetic semiconductors and interacting of localized magnetic moments with the photoexcited itinerant carriers. The photoexcited carriers mediate a ferromagnetic interaction between the localized moments resulting in a ferromagnetic state in the range of curie temperature.

Chapter three treats Hamiltonian model that account the coulomb interaction, light matter interaction and the exchange interaction between carriers and the localized magnetic moments.

chapter four discusses how to construct Bogoliubov-valatin transformation and transform our Hamiltonian of the system into this quasi-particle and calculate the free energy by digonalizing the transformed Hamiltonian.

Chapter 5 include discussion on results and chapter 6 the conclusions drawn from the thesis.

Chapter 1

SPINTRONICS AND DILUTED MAGNETIC SEMICONDUCTORS

1.1 Spintronics

Magnetic materials for future applications in spintronic combine the charge and spin degree of freedom of the electrons making it possible to create novel devices with increased functionality compared to semiconductor devices used today. One of the most promising approaches for integrating magnetic materials and semiconductors are to prepare magnetic semiconductor alloys or diluted magnetic semiconductors, in which a few atomic percent of magnetic atoms likeMn, Fe, Co and Ni are incorporated in compound semiconductors. The ferromagnetic properties of these materials can be controlled by electric field, magnetic field and light [8]. However, the ferromagnetic Curie temperature has been much lower than room temperature.

Spintronics is an emerging field of electronic devices which aims to exploit and manipulate the electrons spin in addition to its charge. A hope of using the spins and charge of electron in the same device for storing, transmitting and processing quantum information invoked a great deal of interest in spin effects and magnetism in semiconductors[9]. New electronic and optic devices such as the spin-FET (field effect transistor), spin-LED (light emitting diode), MRAM (magnetic random access memory), etc. may also offer numerous benefits such as increased processing speed, decreased power consumption and non-volatility. The best known successful spin based devices at present are the magneto-resistance (MR) sensors made of multilayer's containing metal ferromagnetism, showing giant magneto-resistance (GMR) or tunneling magneto-resistance (TMR), which is currently used in the read heads of a hard disk drive.

For the implementation of semiconductor based spintronic devices several materials challenges such as injection, transport, manipulation and detection of the spin must be overcome. A ferromagnetic material, in which the charge carriers posses a certain degree of spin polarization, is therefore an essential component of such a device. Diluted magnetic semiconductors are promising materials for spintronics because of their structural compatibility with semiconductors used in present electronics, as the epitaxial growth of ferromagnetic-semiconductor hetrostructure with well-ordered interfaces allows controlled spin injection from the ferromagnetic layer into the semiconductor [10].

1.2 Diluted magnetic semiconductor

Diluted magnetic semiconductors(DMS) are semiconductor compounds in which a few fractions of the constituent ions is replaced by magnetic atoms like Mn, Fe, Co, Ni etc. The great interest in diluted magnetic semiconductor is due to the fact that, in contrast to magnetic semiconductor, diluted magnetic semiconductor offer a possibility of studying the magnetic phenomena in crystals with a simple band structure and excellent magneto-optical and transport properties. Moreover, the well developed technology of growing diluted magnetic semiconductors allows for tuning their magnetic properties not only by an external magnetic field, but by varying the band structure or carrier, impurity and magnetic ions concentrations. When atoms like Mn, Fe, Co, Ni etc are substitution incorporated into semiconductor lattice, they do not only introduce local magnetic moments but also act as accepters, providing holes that mediate the interaction between the localized transition metal(TM) spins. These magnetic moments can originate from 3d or

4f open shells of transition metals or rare-earth elements respectively. Certain typical examples of DMS are $Cd_{1-x}Co_xSe, Hg_{1-x}Fe_xSe, Zn_{1-x}Co_xS, Ga_{1-x}Mn_xAs, In_{1-x}Mn_xAs$ etc. In most cases Mn is used as the magnetic dopant because Mn has the highest possible magnetic moment and also the first half of the d-band is full, creating a stable fully polarized state. The term DMS is usually reserved for single-phase systems to differentiate them from systems where magnetic second phases are incorporated as precipitates[11].

Dilute magnetic semiconductors are regarded with high interest because of magnetic properties that can be probed or controlled electrically and alternatively electronic properties that are influenced by magnetic interactions between localized and itinerant spins. The underlying explanation for these effects is the quantum mechanical exchange between localized spins of TM d orbitals and host valence band p orbitals(12).

Magnetic behaviour in the transition metal clusters is motivated largely by the desire to understand how magnetic properties change in the reduced dimension. Manganese clusters are particularly interesting among all 3d transition metal elements due to the $4s^2 3d^5$ electronic configuration in Mn atoms. Because of the filled 4s and half filled 3d shells and the large energy gap $\sim 8eV$ between these levels and as well as due to the high $4s^2 3d^5 \rightarrow 4s^1 3d^6$ promotion energy of 2.14 eV, Mn atoms do not bind strongly[13].

1.2.1 Methods of preparation of DMS

Preparation of DMS materials are mainly done by i) Bulk crystal growth. Ex. Bridgman method. ii) Thin film growth. Ex. a) Molecular Beam Epitaxy (MBE) b) Pulse Laser Depositions (PLD) c) Chemical Vapour Depositions (CVP).

The bulk growth of DMS materials are limited by solid solubility of magnetic ions in the lattice of the semiconductor crystals. This is because of the equilibrium growth is controlled by phase diagram.

In contrast thin film growth is non-equilibrium in nature, therefore by controlling the growth temperature a large concentration of magnetic ions can be introduced in the crystal lattice of semiconductor.

One of the approaches for integration of semiconductors and magnetic materials is the epitaxial growth. The term epitaxy is applied to processes used to grow a thin film is such a way that the layer grown has the same lattice structure as substrate.

1.2.2 II-VI based DMS

The II-VI semiconductor crystals incorporated with magnetic ions likes Mn^{+2}, Cr^{+2}, Fe^{+2}, Co^{+2} gives II-VI based DMS. In such DMS the +2 magnetic ions are easily incorporated into the host II-VI crystals by replacing group II cations. II-VI based DMSs are difficult to dope to create p- and n type, which makes the materials less attractive for applications.

The magnetic interaction in II-VI based DMS is dominated by the anti-ferromagnetic exchange among the magnetic ion spins, which results in the paramagnetic, anti-ferromagnetic, or spin glass behaviour of the materials. It was not possible until very recently to make a II-VI DMS ferromagnetic at low temperature.

Recently, the II-VI compound semiconductor ZnO, GaN have attracted revival attention since it was found that high quality epitaxial thin film display excitonic ultraviolet laser action at room temperature. In addition, the energy gap of this compound can be extended up to $\sim 4eV$ by synthesizing alloy compounds to $Mg_xZn_{1-x}O$ electro doping $> 10^{21}/cm$ was readily achieved in contrast to the II-VI compound semiconductors. Therefore, the amount of injected spin and carriers into the film can be very large.

1.2.3 III-V based DMS

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 less. A breakthrough was made by using molecular beam epitaxy (MBE), a thin-film growth technique in vacuum that allows one to work far from equilibrium. When a high concentration of magnetic elements is introduced in excess of the solubility limit, formation of the second phase occurs if conditions are near equilibrium. However, when the crystal is grown at low temperature by MBE, there is not enough thermal energy available to form the second phase, and yet there still exists a local potential landscape that allows epitaxial growth of a single-crystal alloy[14]. The effort to grow new III-V-based DMSs by low-temperature MBE was rewarded with successful epitaxial growth of uniform (In,Mn)As films on GaAs substrates in 1989, where partial ferromagnetic order was found and ferromagnetic (Ga,Mn)As in 1996 [15]. In III-V-based DMS materials, hole carriers are generated by the doping of Mn^{+2} magnetic ions, which act as acceptors in GaAs and InAs. Thus it is logical to consider that the exchange interaction between the localized d electrons of Mn^{+2} ions and the itinerant hole carriers (p-d exchange interaction) is the possible origin of the ferromagnetic order[16].

1.2.4 DMSs based on oxide semiconductors

The possibility of designing a suitable spintronic material having simultaneously the properties of room-temperature ferromagnetism and 100% spin polarization or half metallicity has brightened after the advent of transition-metal-doped semiconducting oxides like ZnO and TiO_2. A wide range of contradictory experimental results, debating the success versus failure of obtaining a T_c above room temperature based on such dilute magnetic oxide systems, have injected much excitement about the origin of ferromagnetism in these systems. Among all these systems, ZnO belongs to the list of the most suitable candidates for spintronics application due to its abundance and environment-friendly nature and also due to its potential as a suitable optoelectronic material with a wide band gap ($\sim 3.3eV$) and high exciton binding energy of 60meV. Earlier first-principles electronic structure calculations suggested that transition-metal(TM) doped ZnO compounds are ferromagnetic provided the TM doping produces carriers forming a partially filled spin-split impurity band. Theoretical calculations also indicate the possibility of designing a ZnO-based room-temperature ferromagnetic obtained by n-type carrier doping ($Ga_xZn_{1-x}O$), as well as by p-type carrier doping (ZnN_yO_{1-y}). Most of the experimental results show that, among transition-metal dopants, Co doped ZnO systems are ferromagnetic in both thin films as well as bulk materials. Also, there have been few reports of ferromagnetism in other TM-doped to ZnO. The ferromagnetism in the Mn-doped system is yet to be understood, as Mn in the +2 valence state does not dope any carrier into the ZnO system and hence, by first-principles studies, must lead to an antiferromagnetism ground state. However, the experimental success with $< 4\%$ Mn doped ZnO thin film has posed a challenge to the theoretical understanding of ferromagnetism for such systems. Further experimental studies reveal the importance of defects such as interstitial zinc and oxygen vacancies for the magnetic ordering in such systems. Regarding the experimental studies, it can be commented that the presence of long-range ferromagnetic ordering, especially the magnetic moment per cation and T_c depend largely on critical details of the sample preparation. Hence, in order to understand the exact ferromagnetic mechanism in such systems, there are several important issues to be resolved. First, the existence of room-temperature ferromagnetism must be related to some intrinsic origin and it must not be due to some impurity phase or TM magnetic clusters. There are studies that advocate the presence of a segregated inverted-spinel phase in oxide-based dilute magnetic nano particle systems. This phase segregation apparently supports a magnetic network over the dimension of the crystal, resulting in a high-temperature ferrimagnetic phase[17].

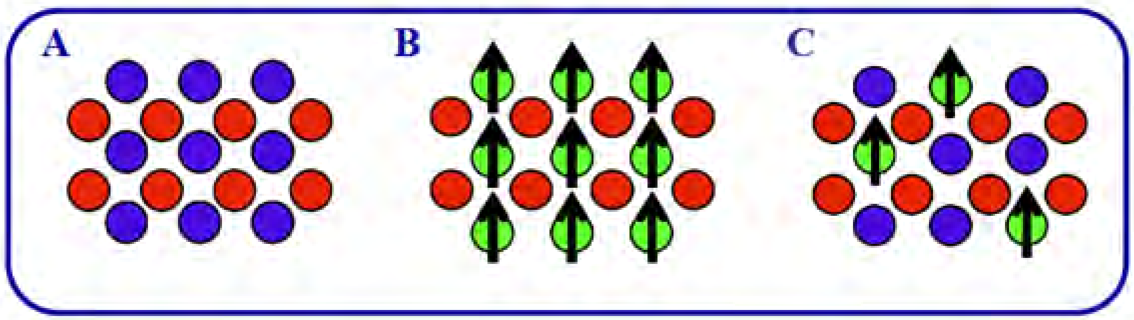

Figure 1.1: Three types of semiconductors: (A)conventional semiconductors; (B) magnetic semiconductor, in which a periodic array of magnetic element is present (C) diluted magnetic semiconductor.

1.3 Magnetic Semiconductors

Ferromagnetism and semiconducting properties coexist in magnetic semiconductors, such as Europium Chalcogenides and Semiconducting spinels that have a periodic array of magnetic elements (Fig. 1.1B). In these magnetic semiconductors, which were extensively studied in the late 1960s to early 1970s, exchange interactions between the electrons in the semiconducting band and the localized electrons at the magnetic ions lead to a number of peculiar and interesting properties, such as a red shift of band gap when ferromagnetism sets in. Unfortunately, the crystal structure of such magnetic semiconductors is quite different from that of Si and GaAs; in addition, the crystal growth of these compounds is notoriously difficult. To obtain even a small, single crystal requires weeks of preparation and growth [18].

1.4 Ferromagnetism and Mechanism of Ferromagnetism

Many materials that contain atoms with a net magnetic moment show some type of spontaneous magnetic ordering below a certain transition temperature. A material is called ferromagnetic when the moments are aligned parallel to each other, and the transition temperature is then called the Curie temperature T_c. For such a spontaneous ordering to take place, one needs a coupling or interaction between these moments, as the classical dipole interaction is typically not strong enough to provide the normal ordering mechanism[19].

Figure 1.2 shows the operative mechanisms for magnetic ordering in Mn doped to GaAs. There are two basic approaches to understand the magnetic properties of diluted magnetic semiconductors. The first one is based on mean field theory which originates in the model of Zener[20]. The theories that fall into this general model implicitly assume that the diluted magnetic semiconductor is a more or less random alloy, in other words, the doping atoms will substitute randomly for the lattice constituents. The second class of approache suggests that the magnetic atoms form small clusters that produce the ferromagnetism[21].

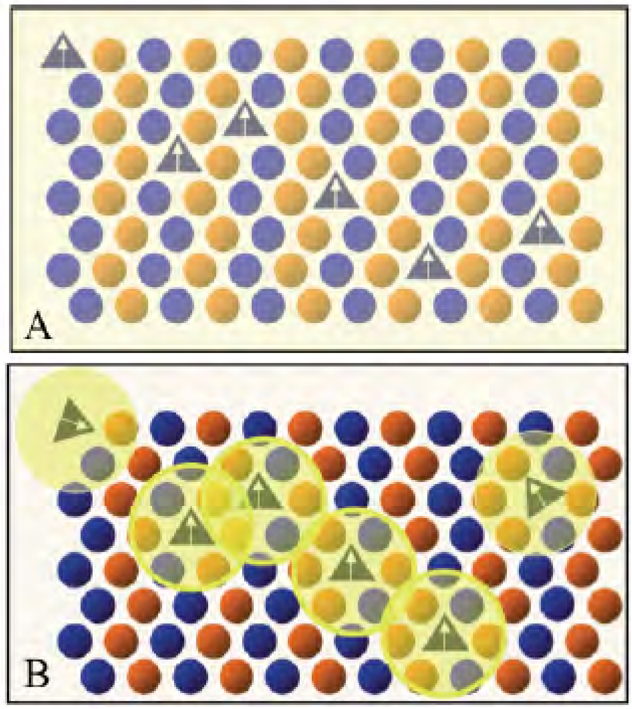

Figure 1.2: Illustration of carrier mediated ferromagnetism in diluted magnetic semiconductor(GaMn)As. Mn^{+2} ions sit on trivalent Ga sits(triangles), and therefore act as electron acceptors(providing holes) as well as producing a magnetic moment(arrows). (A) The holes are thought to mediate ferromagnetic coupling between the magnetic ions. (B)Below certain temperatures, a percolation network is formed in which clusters of the holes are delocalized and hop from site to site, which is an effective mechanism for aligning Mn moments within the cluster network[18].

1.4.1 Exchange interactions

When magnetic ions are doped into semiconductors, they will substitute part of positive ions and their local spin momentum has strong spin-spin exchange interaction with carriers, which further will lead to different behaviour of charge carriers, thus make DMS different from normal semiconductors. Spin-spin exchange interaction is the key point through which various magnetic moments are formed. In DMS, exchange interaction include interactions between s electrons in conducting band, p holes in valence band and d electrons from magnetic ions (sp-d exchange interaction) and interaction between d electrons from magnetic ions(d-d exchange interaction). The exchange interaction between magnetic ions is due to superexchange mediate by negative ions in distorted crystal lattice.

There are several origins for the interactions between the magnetic moments that can lead to a long-range ordering of the unpaired spins, called exchange interactions. The most common exchange interactions are schematically illustrated in Fig. 1.3. (a) (b) (c)

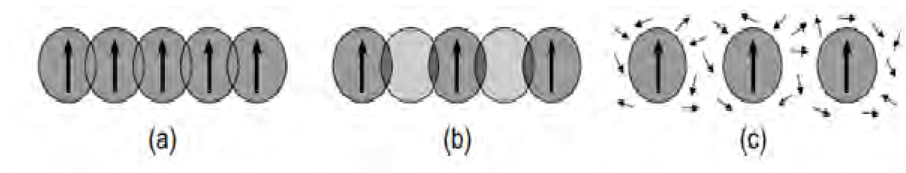

Figure 1.3: Schematic illustrations of (a) direct exchange, in which the magnetic ions interact through their overlapping charge distributions; (b) super exchange, where the interaction takes place via an intermediate non-magnetic ion and (c) indirect exchange, in which the interaction is mediated by interactions with the charge carriers [2].

- **Direct interaction** occurs when there is direct overlap of the wave functions of electrons associated with nearest-neighbor magnetic atoms. The direct exchange mechanism was formulated to describe magnetic behaviour in conventional materials like Fe, Co and Ni.

- **superexchnge** is an exchange mechanism that describes the interaction of magnetic cations via an intermediate non-magnetic anion. In the case of II-VI and III-V diluted magnetic semiconductors, the super exchange results from the spin dependent hybridization between anion p and Mn d states and leads to a short-range anti-ferromagnetic (AF) coupling among the Mn moments. In the absence of holes, Mn-based II-VI and III-V DMS are indeed found to be anti-ferromagnetism[22].

- **Indirect interaction** occurs when the direct overlap of the magnetic atoms is small, they are also able to interact through indirect exchange interactions which are mediated by the charge carriers. This indirect exchange can occur through several mechanisms such as the RKKY interaction, the double exchange or the Zener mechanism which will be discussed below. In diluted magnetic semiconductors it is exactly this indirect exchange which will lead to the ferromagnetism.

• **Double exchange** this exchange mechanism operates if the width of the carrier band is smaller than the exchange energy J_{ex}, as would be expected for narrow impurity bands formed from d states, where carrier hopping is the dominant conduction mechanism. This model was originally developed by Zener and considers carrier hopping over d states which is facilitated when the localized spins are ferromagnetically ordered. This involves the simultaneous transfer of electrons between two Mn ions with a different valency and an intermediate ion, which is therefore called double exchange. The ferromagnetic transition is then driven by the lowering of the carrier energy due to an increase in the bandwidth. Accordingly in such system spin ordering is always accompanied by a strong increase in the conductivity, which leads to the so-called colossal magneto-resistance.

This model has been successfully applied in manganite, where only d electrons in narrow bands appear to be involved. In $Ga_{1-x}Mn_xAs$ no evidence for d-band transport or the associated colossal magnetoresistance has so far been found. However, the d levels of transition metals other than Mn reside in the band gap of III-V and II-VI semiconductors. In III-V and II-VI diluted magnetic semiconductor with such magnetic dopants the double exchange may thus constitute the dominant mechanism of spin-spin interactions. In insulating diluted magnetic semiconductors the strongly localized carriers give rise to the presence of forming bound magnetic polarons. To gain Coulomb energy, these magnetic polarons are preferentially formed around close pairs of ionized acceptors. In the case of III-V DMS the Mn ions are acceptors as well as magnetic impurities, and one hole localized at two Mn ions may generate a strong ferromagnetic coupling between the magnetic polarons via Zener's double exchange[23].

• **Zener model of ferromagnetic interactions** Zener first proposed a model of ferromagnetism driven by the exchange coupling of the carriers and the localized spins. According to that model, spin polarization of the localized spins leads to spin splitting of the bands, which results in the lowering of the carrier energy. At sufficiently low temperature, this lowering overcompensates the increase of the free energy caused by the decrease of entropy that is associated with the polarization of localized spins. However, the Zener model was later abandoned, as neither the itinerant character of the magnetic electrons nor the quantum oscillations of the electron spin polarization around the localized spins were taken into account, and both of these are now established to be critical ingredients of the theory of magnetic metals. In particular, the resulting competition between ferromagnetic and antiferromagnetic interactions in metals leads rather to a spin-glass than to a ferromagnetic ground state. In the case of semiconductors, however, the mean distance between the carriers is usually much greater than that between the spins. Under such conditions, the exchange interaction mediated by the carriers is ferromagnetic for most of the spin pairs, which reduces the tendency toward spin-glass freezing. In fact, for a random distribution of the localized spins, the mean-field value of the Curie temperature T_c deduced from the Zener model is equal to that obtained from the RKKY approach[24].

• **RKKY interaction** a long range indirect exchange mechanism which is mediated by itinerant electrons was first proposed by Ruderman and Kittel to describe the coupling of nuclear magnetic moments in a metal by means of the hyper-fine interaction with the conduction electrons. Later this model was extended by Kasuya and Yosida to give the theory now generally know as the RKKY interaction. When a magnetic impurity is present in the compound, the conduction electrons attempt to screen the spin of the impurity and will spin-polarize in concentric rings around the impurity. As a result, the electron spin polarization around the localized spins will show so-called Ruderman-Kittel oscillations. This oscillatory behaviour is reflected in the RKKY interaction, as distant magnetic impurities are assumed to interact indirectly by their spin coupling to the charge carriers. The RKKY exchange coefficient, J_{RKKY}, for the interaction of two magnetic impurities at sites i and j at a distance r_{ij} is then given by

$$J_{RKKY}(r_{ij}) = \frac{2mk_f^4}{\pi h^4} J_{ex}^2 F(2k_f r_{ij}), \tag{1.4.1}$$

where m is the effective mass of the charge carriers, k_f their Fermi wave number, h is Planck's constant, J_{ex} is the exchange between the carrier spin and the local moments, and F(x) is the Ruderman-Kittel oscillation term [25]:

$$F(x) = \frac{x cos(x) - sin(x)}{x^4}. \quad (1.4.2)$$

1.4.2 Hole mediated ferromagnetism

Ferromagnetism mediated by delocalized or weakly localized holes is well established by a mean-field model taking into account a p-d exchange hybridization and a spin-orbital interaction. One of the characteristic features of magnetic semiconductors is the s(p)-d exchange interaction between the sp band of the host semiconductor and the localized d electrons associated with magnetic ions. This exchange interaction yields spectacular magneto-optical and magneto transport effects. Magneto transport and magnetic circular dichroism (MCD) measurements are extensively used for studying the s(p)-d exchange interaction of ferromagnetic semiconductors.

Several experimental results indicate that the ferromagnetic interaction in diluted magnetic semiconductors is mediated by the charge carriers, which are in most cases valence band holes. Ferromagnetic ordering in II-VI based DMS for instance can only be obtained with sufficient p-type doping, indicating that the anti-ferromagnetic superexchange in these samples can be overcompensated by ferromagnetic interactions mediated by band holes. A similar phenomenon is observed in III-V DMS, since the magnetic interaction among Mn has been shown to be anti-ferromagnetic in n-type $In_{1-x}Mn_xAs$ and in fully carrier compensated $Ga_{1-x}Mn_xAs$ using Sn as a donor, indicating the critical role of the holes for the magnetic coupling. The carrier-induced nature of the ferromagnetic state in III-V DMS has been compellingly demonstrated by field-effect experiments on $In_{1-x}Mn_xAs$, where the carrier concentration was tuned by a gate and the critical temperature was modified accordingly. The hole mediated origin of the ferromagnetism in

DMS is further evidenced by the correlation between its transport and magnetic properties [10].

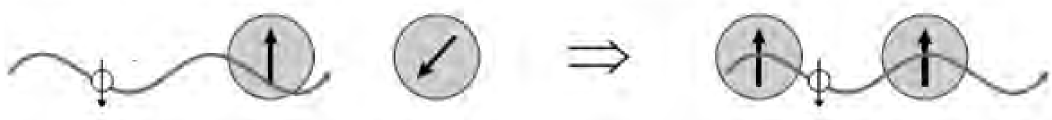

Figure 1.4: Schematic illustration of the hole mediated exchange interaction in diluted magnetic semiconductors. A long range ferromagnetic interaction of the TM spins results from a strong anti-ferromagnetic coupling of the itinerant or weakly bound holes to the localized spins.

The ferromagnetic properties of diluted magnetic semiconductors result from the presence of a spin-dependent interaction between the electrons or holes in the sp bands of the semiconductor host and those residing on the d shells of the magnetic impurities, as illustrated schematically in Fig. 1.4. This interaction assumes a form of the Heisenberg exchange coupling as:

$$H = -J_{ex}\vec{s}_i.\vec{S}_j, \tag{1.4.3}$$

where J_{ex} describes the strength of the interaction between the carriers spin $\vec{s}$ and the transition-metal spin $\vec{S}$. If J_{ex} is positive then the lowest energy state corresponds to a parallel spin alignment, while an anti-ferromagnetic alignment is favored when J_{ex} is negative. In $Ga_{1-x}Mn_xAs$, the hybridization of the valence band holes (with p-orbital character) and the Mn d states that results in spin-dependent interaction, the so called kinetic exchange, which is characterized by a rather large exchange energy $J_{pd} \sim -1eV$. The negative sign of J_{pd} reflects an anti-ferromagnetic coupling between the hole spin and the localized Mn spin. These sp d exchange couplings give rise to a spin-splitting of the bands proportional to the sample magnetization.

The anti-ferromagnetic coupling between the hole spin and the localized Mn spin leads to a long range ferromagnetic coupling between neighboring Mn spins (see Fig. 1.4), which eventually results in a ferromagnetic state at low temperatures. In principle various mechanisms for the hole mediated exchange coupling in diluted magnetic semiconductors[24].

1.4.3 The mean-field theory of ferromagnetism

In a ferromagnetic ordered state the mean value of the magnetization is the same at each lattice point. Making use of the relation M = N $< \mu_i >$ between magnetization and magnetic moment[26].

The magnetization due to the spin(S) in a magnetic field is

$$M = \frac{N\sum_{-S}^{S} -g\mu_B S exp(-\beta g\mu_B HS)}{\sum_{-S}^{S} exp(-\beta g\mu_B HS)}, \tag{1.4.4}$$

$$M = Ng\mu_B SB_s(\beta g\mu_B HS), \tag{1.4.5}$$

$$M = Ng\mu_B SB_s(x), \tag{1.4.6}$$

$$x = \beta g\mu_B HS. \tag{1.4.7}$$

In the mean field theory this expression has to be modified by replacing the magnetic field(B) by the effective field(H_{eff}), leading to

$$M = Ng\mu_B SB_s(\beta g\mu_B S(H + \eta M)), \tag{1.4.8}$$

$$H_{eff} = H + \frac{2}{Ng^2\mu_B^2}\sum_j J_{ij}M, \tag{1.4.9}$$

$$\sum_j J_{ij} = J, \tag{1.4.10}$$

$$H_{eff} = H + \frac{2}{Ng^2\mu_B^2}JM, \tag{1.4.11}$$

where J is the average value of various exchanging interaction constant.

$$H_{eff} = H + \eta M, \tag{1.4.12}$$

$$\eta = \frac{2J}{Ng^2\mu_B^2}, \tag{1.4.13}$$

where η specifies the relation between the internal field and magnetization in the Weiss theory.

In the absence of an external magnetic field the argument of the Brillouin function is

$$x = \beta g \mu_B S \eta M = \frac{2JSM}{Ng\mu_B k_B T}, \tag{1.4.14}$$

where B_s(x) is the Brillouin function for spin, which is given by

$$B_s(x) = \frac{2S+1}{2S} coth(\frac{2S+1}{2S})x - \frac{1}{2S} coth(\frac{x}{2S}), \tag{1.4.15}$$

$$cothx = \frac{1}{x} + \frac{x}{3} - \frac{1}{45}x^3. \tag{1.4.16}$$

Then

$$coth(\frac{2S+1}{2S})x = \frac{2S}{(2S+1)x} + \frac{2S+1}{6S}x - \frac{1}{45}(\frac{2S+1}{2S}x)^3, \tag{1.4.17}$$

$$coth(\frac{x}{2S}) = \frac{2S}{x} + \frac{x}{6S} - \frac{1}{45}(\frac{x}{2S})^3. \tag{1.4.18}$$

Neglecting the higher ordered

$$B_s(x) = \frac{2S+1}{2S}(\frac{2S}{(2S+1)x} + \frac{2S+1}{6S}x) - \frac{1}{2S}(\frac{2S}{x} + \frac{x}{6S}), \tag{1.4.19}$$

$$B_s(x) = \frac{1}{x} + \frac{(2S+1)^2}{12S^2}x - \frac{1}{x} - \frac{x}{12S^2}, \tag{1.4.20}$$

$$B_s(x) = \frac{x}{12S^2}(4S^2 + 4S + 1 - 1), \tag{1.4.21}$$

$$B_s(x) = \frac{x(S+1)}{3S}. \tag{1.4.22}$$

Using Eq.(1.4.6) and Eq.(1.4.14)

$$\frac{k_B T N g\mu_B}{2J_0 S}x = Ng\mu_B S B_s(x), \tag{1.4.23}$$

$$\frac{k_B T}{2J}x = S^2 B_s(x). \tag{1.4.24}$$

Substituting Eq.(1.4.22)in Eq.(1.4.24)and rearranging

$$k_B T = \frac{2JS(S+1)}{3}. \tag{1.4.25}$$

From this equation,the curie temperature is

$$T_c = \frac{2JS(S+1)}{3k_B}. \tag{1.4.26}$$

Let us return to Eq.(1.4.6) to calculate magnetic susceptibility;

$$M = Ng\mu_B S B_s(x), \tag{1.4.27}$$

$$M = Ng\mu_B S\frac{(S+1)}{3S}x. \tag{1.4.28}$$

In the presence of external field

$$x = \frac{g\mu_B S}{k_B T}(H + \eta M). \tag{1.4.29}$$

Therefore

$$M = Ng^2\mu_B^2 S\frac{(S+1)}{3k_B T}H + Ng^2\mu_B^2 S\frac{(S+1)}{3k_B T}\eta M. \tag{1.4.30}$$

Substituting Eq.(1.4.13)in Eq.(1.4.30)in term of η

$$M = Ng^2\mu_B^2 S\frac{(S+1)}{3k_B T}H + \frac{M}{k_B T}(\frac{2JS(S+1)}{3}), \tag{1.4.31}$$

using Eq.(1.4.26)

$$M = Ng^2\mu_B^2 S\frac{(S+1)}{3k_B T}H + \frac{k_B T_c}{k_B T}M, \tag{1.4.32}$$

$$M(1 - \frac{T_c}{T}) = Ng^2\mu_B^2 S\frac{(S+1)}{3k_B T}H, \tag{1.4.33}$$

solving this equation for magnetization

$$M = \frac{Ng^2\mu_B^2 S(S+1)}{3k(T-T_c)}H. \tag{1.4.34}$$

Then the magnetic susceptibility becomes

$$\chi = \frac{\partial M}{\partial H} = \frac{Ng^2\mu_B^2 S(S+1)}{3k(T-T_c)} = \frac{C}{T-T_c}. \tag{1.4.35}$$

Chapter 2

Photo induced ferromagnetism in diluted magnetic semiconductors

When light falls on the DMS, it creates electron-hole pairs across the band gap. These carriers interact with the impurity magnetic atoms and so induced ferromagnetic coupling among magnetic moments and thereby create a ferromagnetic state in these semiconductors when temperature is lowered.

2.1 Photo induced ferromagnetism

Information processing in electronic devices is based on control of charge flow in semiconductor materials, whereas nonvolatile information storage exploits ferromagnetism, spontaneous alignment of the spins of many electrons. Spintronics aims to achieve a merger of these technologies, motivating interest in diluted magnetic semiconductors (III,Mn)V compounds like (Ga,Mn)As and II,Mn)VI materials like (Cd,Mn)Te with free moments that can be aligned by external fields. The optical properties of these semiconductors are particularly interesting in this respect because laser radiation can control both charge and spin dynamics. The influence of the laser field on the electronic system is reactive rather than dissipative and follows ultimately from a change in the effective electronic Hamiltonian.

In doped systems, band electrons mediate RKKY interactions between Mn spins. On the other hand, virtual fluctuations in the Mn valence lead to short range superexchange antiferromagnetic interactions which couple only nearest neighbors. In a sample with a fraction x of Mn randomly located at the cation sites is the fraction of sites containing Mn atoms without a magnetic first neighbor. Consequently, samples with a low Mn concentration ($x \simeq 0.01$) and no band carriers are paramagnetic. It has been shown that two localized spins in a semiconductor can interact via the virtual carriers created in the (otherwise empty) bands by a laser of frequency smaller than the band gap. The effective interaction is a ferromagnetic Heisenberg coupling which we shall call optical RKKY. It is our contention that the optical RKKY interaction can drive a diluted magnetic semiconductor into a ferromagnetic phase at low temperatures[27].

In this section we see how light can induce ferromagnetism in diluted magnetic semiconductor materials. The idea is that the hole produced by the dopant magnetic atoms can interact with polarized light and be spin polarized and mediate ferromagnetic order between magnetic ions moments. However, it is recently that a great deal of attention has been focused on the photo induced magnetization, where ferromagnetism is induced or enhanced by the interaction with the incident light. The subject has acquired added importance in view of the search for materials with spin-polarized electrons to be relevant in the emerging area of spintronics.

Photo induced magnetization was first demonstrated by Krenn et al.[28] who found a magnetization signal in $Hg_{1-x}Mn_xTe$ which depends characteristically on the degree of polarization of exciting radiation. The measured photomagnetization increase linearly with photon power to certain value of photo power, but saturates on increasing the photon power. Later on, the method extended to observation of time resolved phenomena in the photo induced magnetization of $Cd_{1-x}Mn_xTe$. Photoinduced magnetic effects have been extensively studied in a variety of systems, including cyanometalate-based magnets, spin crossover complexes, diluted magnetic semiconductors, doped manganite and spinel

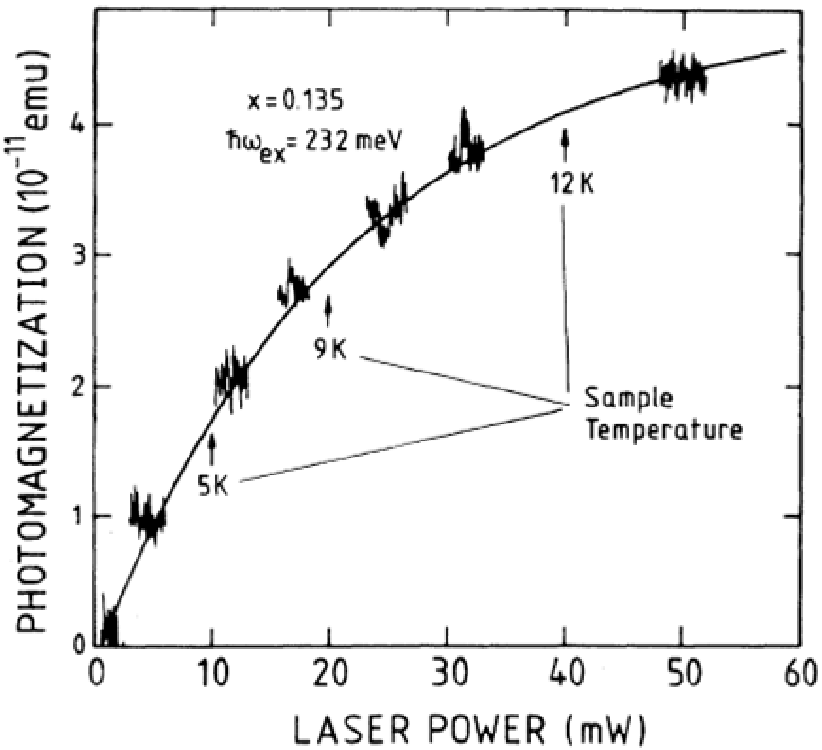

Figure 2.1: The dependence of photo induced magnetization in $Hg_{1-x}Mn_xTe$ (x=0.135) on the impinging laser power.

ferrite films. One of the challenges in this area of research is design of materials that exhibit both photoinduced magnetization and long-range magnetic order at relatively high temperatures[29].

A number of early experiments regarding the photo induced magnetization have been reported for the diluted magnetic semiconductors system. A ferromagnetism order by photo generated carriers in the hetrostructure (In Mn)As/GaSb was reported. At low temperature, samples preserved the ferromagnetism order even after light is switched off. They attribute the effect to hole transfer from GaSb to InMnAs in the hetrostructure, which enhances a ferromagnetism spin exchange among Mn ions in the InMnAs layer. photo induced magnetization in ferromagnetic (Ga,Mn)As thin films caused by spin polarized holes generated optically also observed. The observed result suggest that a small amount of non-equilibrium carrier spins can cause collective rotation of Mn spins presumably p-d exchange interaction. The realization of ferromagnetism in a semiconductor results in many unique phenomena which have no analogy in the standard of magnetism. For instance, the hole-mediated ferromagnetic exchange that occurs in this semiconductor opens the possibility to change the magnetic properties of (Ga,Mn)As by modulation of the hole concentration and a huge photoinduced magnetization of about 15% of the saturation magnetization in ferromagnetic (Ga,Mn)As films. This opens the way to ultra fast manipulation of the magnetic state of (Ga,Mn)As. The photoexcited non equilibrium spin polarization in (Ga,Mn)As relaxes with a decay time of 30 ps, which is related to the spin relaxation of electrons in the conduction band. So weak interaction between the spins of the electrons in the conduction band and the spins of the Mn ions, which may have consequences for the application of (Ga,Mn)As in spintronic devices[30].

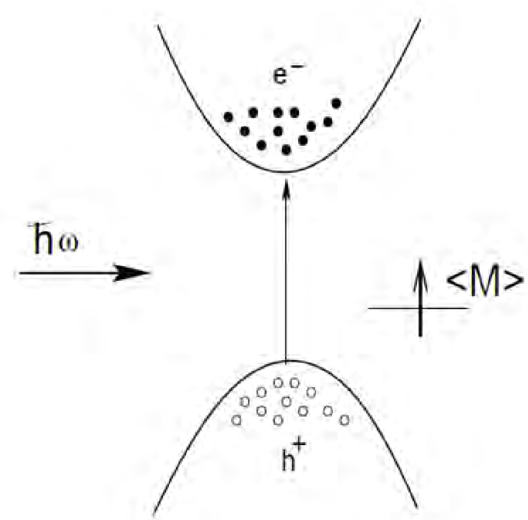

Figure 2.2: A schematic picture of light induced ferromagnetic in diluted magnetic semiconductor. The electrons and holes that are created across the band gap interact with the magnetic moments and when temperature is lowered, systems changes from paramagnetic to ferromagnetic phase.

While rotation occurs instantaneously with generation of hole spins, with relaxation takes places within some tens of picoseconds resulting from strong damping[31]. Very recently, photo magnetism has been observed in GaAs(Mn), where a transient increase of T_c by about 0.5K has been observed with laser irradiation [32].

2.2 Optical control of ferromagnetism

Because carriers mediate ferromagnetic interaction, photo-generated carriers change their magnetic properties and thus optical control of ferromagnetism in these materials is possible. So optical control of ferromagnetism by light irradiation was also observed in II-VI Mn-doped semiconductors, indicating that the effects are not limited to IIIV ferromagnetic semiconductors. By designing the structure, it was shown that one could either enhance or destroy ferromagnetism in (Cd,Mn)Te quantum well (QW) with (Cd,Zn,Mg)Te barriers. Illumination of light enhances the ferromagnetism in QW of an unhoped region of a p-i-n structure, whereas illumination destroys that in a p-i-p structure. In p-i-n structure, photogenerated holes, separated by internal electric-field from electrons, migrate toward the QW to enhance the ferromagnetism. By contrast in the p-i-p structure, the photo-generated electrons are collected by the QW due to band bending, where they diminish hole-induced ferromagnetism[33].

The mechanism of coupling of spins localized in neighboring quantum dots by virtual excitation of delocalized exciton states in the host materials by a light field. This indirect exchange interaction termed as the ORKKY interaction is analogous to the RKKY interaction; the difference from the usual one is that the intermediate electron-hole pair (carrier mediating the magnetic interaction) are produced by the external light. The optical quantum control of a single exciton in a semiconductor QD has been recently reported in GaAs QDs generated by monolayer fluctuations and InGaAs self-assembled QDs. The short radiative recombination lifetime of the exciton (of the order of 100 ps) gives a severe limitation for the application to quantum computation, even with the help of shaping techniques. This can be avoided by doping QDs each with a single conduction electron and by encoding the quantum information in the spin degrees of freedom. Optical control by virtual excitation avoids the fast optical decoherence. Thus, the advantages of a very long spin coherence time in QDs and fast optical control can be combined[34].

Chapter 3

HAMILTONIAN

3.1 The Hamiltonian of the problem

The Hamiltonian describing diluted magnetic semiconductor where electrons and holes generated by a light field are spin-split through an exchange interaction with the magnetic ions within mean field approximation. The Coulomb interaction includes electron-electron, hole-hole and electron-hole interactions. The semiconductor-light interaction is considered such that an electron-hole pair is created by absorption of light or an electron-hole pair recombines by emitting light. The light field is treated as a classical one. The Hamiltonian H is written as

$$H = H_{kin} + H_c + H_L(t), \tag{3.1.1}$$

where

$$H_{kin} = \sum_{k\sigma}(\epsilon_{ck\sigma} c^+_{k\sigma} c_{k\sigma} + \epsilon_{vk\sigma} d^+_{k\sigma} d_{k\sigma}). \tag{3.1.2}$$

The single particle energy including the magnetic moment interaction, which is taken here in a mean field way can be written for the conduction band electron and valance band hole as

$$\epsilon_{ck\uparrow(\downarrow)} = E_c(k) \pm J_e\langle M\rangle, \tag{3.1.3}$$

$$\epsilon_{vk\uparrow(\downarrow)} = E_v(k) \pm J_h\langle M\rangle. \tag{3.1.4}$$

Here J_e and J_h is the electron and hole exchange interaction coupling with the localized magnetic moments, $< M >$ is the average magnetization. The kinetic energy of the conduction band electron and that of valence band hole can be written as

$$E_c(k) = E_g + \frac{\hbar^2 k^2}{2m_e} \tag{3.1.5}$$

and

$$E_v(k) = \frac{\hbar^2 k^2}{2m_h}, \tag{3.1.6}$$

where E_g is the band gap and m_e and m_h are electron and hole effective masses respectively. The combined of these single particle energies of spin up electron and spin down hole and spin down electron and spin up hole shown in Fig.3.1

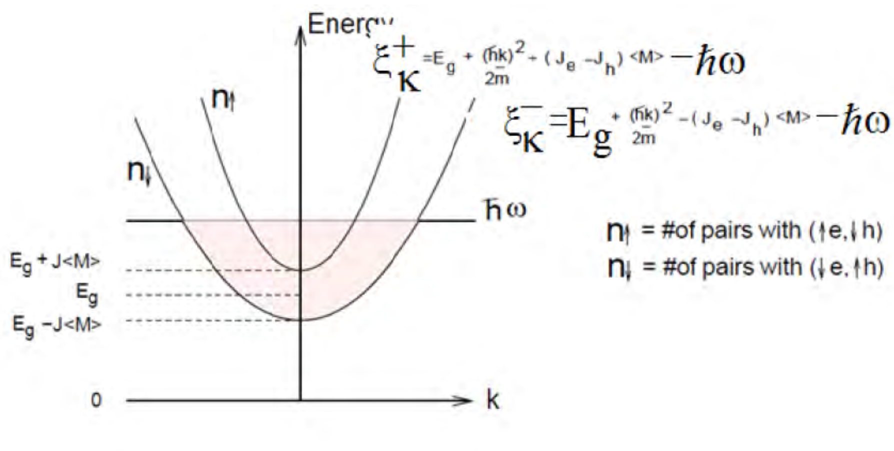

Figure 3.1: Energy of the electron-hole pairs of two different spin types produced by photoexcitation. ξ_k^+ and ξ_k^- are the two particle energies measured with respect to the chemical potential.

$$H_c = H_{ee} + H_{hh} + H_{eh}, \tag{3.1.7}$$

$$H_{ee} = \frac{1}{2} \sum_{k,k',\sigma,\sigma'} V_q c^+_{k+q\sigma} c^+_{k'-q\sigma'} c_{k'\sigma'} c_{k\sigma}, \tag{3.1.8}$$

$$H_{hh} = \frac{1}{2} \sum_{k,k',\sigma,\sigma'} V_q d^+_{k+q\sigma} d^+_{k'-q\sigma'} d_{k'\sigma'} d_{k\sigma}, \tag{3.1.9}$$

$$H_{eh} = -\sum_{k,k',\sigma,\sigma'} V_q c^+_{k+q\sigma} d^+_{k'-q\sigma'} d_{k'\sigma'} c_{k\sigma}, \tag{3.1.10}$$

$$H_L(t) = \lambda \sum_k \rho_k [c^+_{k\uparrow} d^+_{-k\downarrow} + c^+_{k\downarrow} d^+_{-k\uparrow}) e^{-i\omega t} + H.C]. \tag{3.1.11}$$

In the above equations c^+ and c are electron creation and destruction operators, d^+ and d are the corresponding operators for holes. V_q denotes the particle -particle interaction. The negative sign in E.q (3.1.10) denotes attractive interaction between electron and hole. We have parameterized the interaction of light with the particles. So we do not consider explicitly the matrix element of light-interaction which comes from the λ_k $\sim \langle \Psi_i | \hat{e}.\vec{p} | \Psi_f \rangle$ term when light interaction is treated in a semi-classical way. $\hat{e}$ is the polarisation vector of the light field. We take the $\lambda_k = \lambda \rho_k$ and λ is the interaction strength of light-semiconductor coupling and momentum dependence of coupling is contained in ρ_k. σ and σ' denotes the spin indices.

3.2 Photon bath and Free energy

The system of electrons and holes is in contact with a light bath. This is shown pictorially in Fig.(3.2). The chemical potential is the energy to add a particle to the system. Here $\mu = \hbar\omega$ is required to create a hole and an electron. So the free energy to be minimized is given as:

$$F = H - \mu N_h, \tag{3.2.1}$$

$= H - \mu \sum_{k\sigma} d^+_{k\sigma} d_{k\sigma},$

$= H - \frac{\mu}{2} \sum_{k\sigma} c^+_{k\sigma} c_{k\sigma} - \frac{\mu}{2} \sum_{k\sigma} d^+_{k\sigma} d_{k\sigma}.$

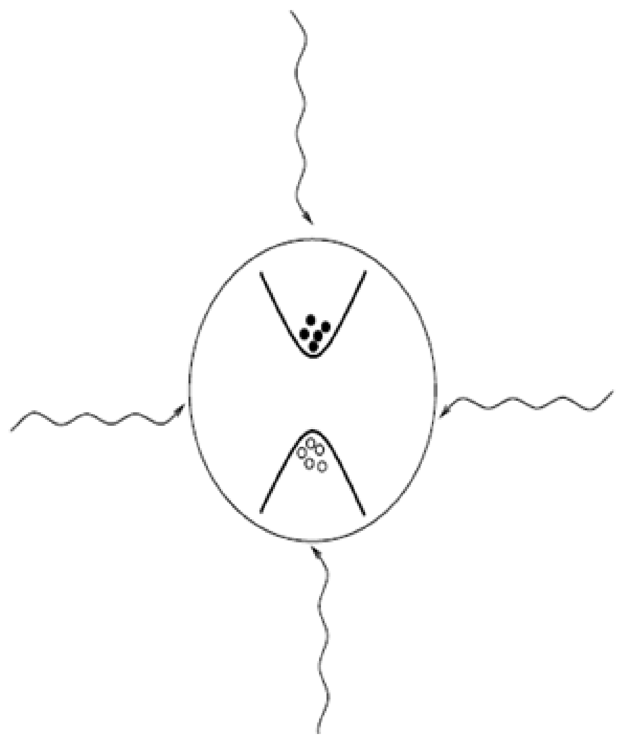

Figure 3.2: The system of electrons in the conduction band and holes in the valence band inside a light bath[35].

3.3 Unitary transformations and elimination of time of Hamiltonian

Let us use a unitary transformation to eliminate time in our original Hamiltonian Eq.(3.1.1-3.1.11), we first study simpler model Hamiltonian to illustrate the transformation which is related to our many particle Hamiltonian. Also by diagonalzing these original time dependant Hamiltonians, we calculate the energy eigenvalues of these particular Hamiltonians. The effective Hamiltonian of the problem gives the time evolution of the system. One can do a unitary transformation on time-independent or time-dependent Hamiltonians. However, if the unitary transformation itself is time-dependent, the transformed Schrödinger equation takes a modified form. Sometimes the time-dependent original Hamiltonian transforms into a time-independent.

The Schrödinger equation is given as

$$i\hbar\frac{\partial}{\partial t}\Psi(t) = H(t)\Psi(t), \tag{3.3.1}$$

where H(t) is a time dependent Hamiltonian. Let us now consider the time dependent unitary operator,

$$U(t) = e^{iS(t)}, \tag{3.3.2}$$

where S(t) is Hermitian at all time. Then the transformed state vectors and operators are given by

$$\tilde{\Psi}(t) = e^{-iS(t)}\Psi(t), \tag{3.3.3}$$

$$\tilde{H}(t) = e^{-iS(t)}H(t)e^{iS(t)}. \tag{3.3.4}$$

The rotation of operators and state vectors in Hilbert space does not change the eigenvalues so that the eigenvalues of H and $\tilde{H}$ are the same. The Schrödinger equation that defines the evolution of $\tilde{\Psi}(t)$ does not follow $\tilde{H}$, but follow $\tilde{H}$ with an extra term added to it. Let us compute how $\tilde{\Psi}(t)$changes with time[36] using Eq. (3.3.3),

$$i\hbar\frac{\partial}{\partial t}\tilde{\Psi}(t) = i\hbar(\frac{\partial}{\partial t}e^{-iS(t)})\Psi(t) + i\hbar e^{-iS(t)}\frac{\partial}{\partial t}\Psi(t), \tag{3.3.5}$$

$= i\hbar(\frac{\partial}{\partial t}e^{-iS(t)})e^{iS(t)}\tilde{\Psi}(t) + \tilde{H}()\tilde{\Psi}(t),$

$=(\tilde{H}(t) + i\hbar(\frac{\partial}{\partial t}e^{-iS(t)})e^{iS(t)})\tilde{\Psi}(t),$

$=\tilde{H}_{eff}\tilde{\Psi}(t).$

Thus the effective transformed Hamiltonian (the transformed Hamiltonian which describes the evolution of $\tilde{\Psi}(t)$ is given as

$$\tilde{H}_{eff} = \tilde{H}(t) + i\hbar(\frac{\partial}{\partial t}e^{-iS(t)})e^{iS(t)}. \tag{3.3.6}$$

One-level system interacting with a light field

Let us make a unitary transformation of the form e^{iS}, where S is Hermitian operators ($S^{+} = S$) and it is given as

$$S = \omega t c^{+} c. \tag{3.3.7}$$

The Schrödinger equations obeyed by H and $\tilde{H}_{eff}$ for time independents are

$$i\hbar \frac{\partial}{\partial t} \Psi = H\Psi, \tag{3.3.8}$$

$$i\hbar \frac{\partial}{\partial t} \tilde{\Psi} = \tilde{H}_{eff} \tilde{\Psi}. \tag{3.3.9}$$

For one-level system interacting with a light field, the Hamiltonian is given by

$$H = \alpha c^{+} c + \lambda(e^{-i\omega t} + e^{i\omega t}) c^{+} c, \tag{3.3.10}$$

where $c^{+}(c)$ is the particle creation(annihilation) operator, λ is the coupling strength of the classical light field with frequency ω and we have two terms due to the Hermitian conjugation. The eigenvalue of H is given as

$$Eigenvalue = \alpha + \lambda(e^{-i\omega t} + e^{i\omega} t) = \alpha + 2\lambda cos(\omega t), \tag{3.3.11}$$

which oscillate between α + 2λ and α-2λ.

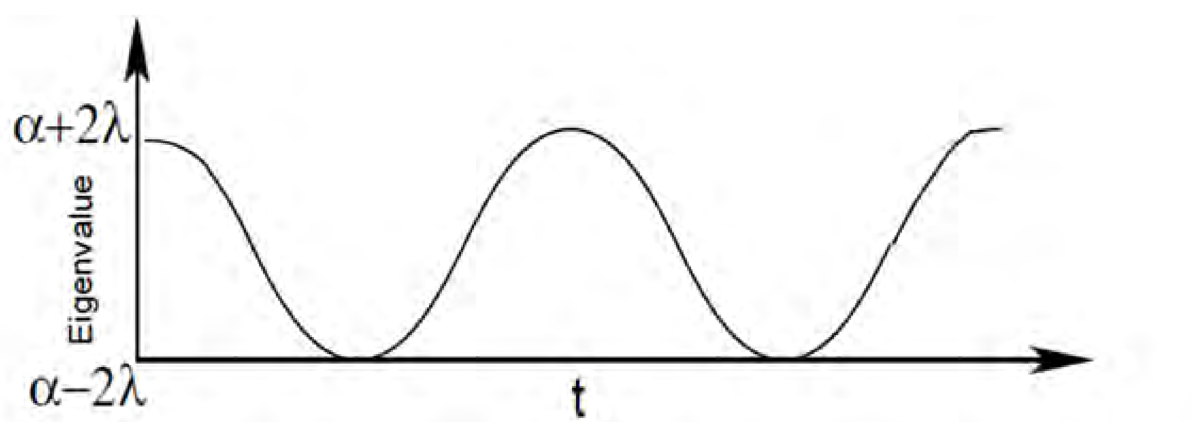

Figure 3.3: Eigenvalue as a function of time t as given by Eq.(3.3.11). The graph shows the variation of the eigenvalue between the two limits α + 2λ and α - 2λ

Using Eq.(3.3.7), the transformed Hamiltonian is given as

$$\tilde{H} = e^{-iS} H e^{iS}, \tag{3.3.12}$$

$= \alpha c^+ c + e^{-i\omega t c^+ c}[\lambda c^+ c(e^{-i\omega t} + e^{i\omega t})]e^{i\omega t c^+ c},$

$= \alpha c^+ c + \lambda c^+ c(e^{-i\omega t} + e^{i\omega t}).$

Eigenvalues of H and $\tilde{H}$ are the same as expected because the unitary transformation is a rotation in Hilbert space at any specific time t.

$$\tilde{H}_{eff} = \tilde{H}(t) + i\hbar(\frac{\partial}{\partial t} e^{-iS})e^{iS}, \tag{3.3.13}$$

$$\tilde{H}_{eff} = \alpha c^+ c + \hbar\omega c^+ c + \lambda c^+ c(e^{-i\omega t} + e^{i\omega t}). \tag{3.3.14}$$

Note that $\tilde{H}_{eff}$ has a different eigenvalues than that of $\tilde{H}$. The eigenvalue is given as

$$eigenvalue = \alpha + \hbar\omega + \lambda(e^{-i\omega t} + e^{i\omega t}) = \alpha + \hbar\omega + 2\lambda cos(\omega t). \tag{3.3.15}$$

The eigenvalue oscillates from maximum $\alpha + \hbar\omega + 2\lambda$ to $\alpha + \hbar\omega - 2\lambda$.

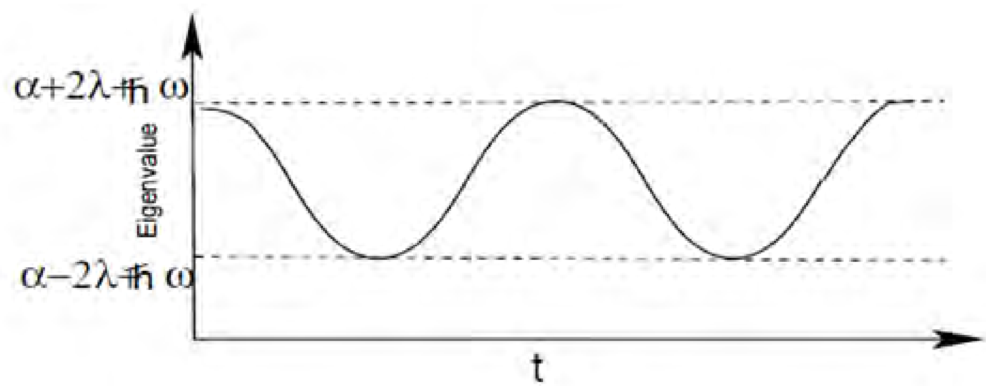

Figure 3.4: Eigenvalue of $\tilde{H}_{eff}$ as a function of time t as given by Eq. (3.3.15). The graph shows the variation of the eigenvalue between the two limits $\alpha+$ $\hbar\omega$+2λ and $\alpha+$ $\hbar\omega$-2λ.

3.3.1 The Rabi problem

A particle in a two state system when coupled to a light field oscillates between these states. This is the famous Rabi problem[37]. Now we consider a Hamiltonian which describes a two state problem coupled to a light field. The electron creation operator c^+ is in one level and hole creation d^+ is in other level. The coupling of light to electron and hole is denoted by λ.

$$H = \alpha c^+ c + \beta d^+ d + \lambda(c^+ d e^{i\omega t} + d^+ c e^{-i\omega t}). \tag{3.3.16}$$

We have taken just two electron states here, which are coupled to light. The Hamiltonian can be written in matrix form as

$$H = \begin{pmatrix} \alpha & \lambda e^{i\omega t} \\ \lambda e^{-i\omega t} & \beta \end{pmatrix} \tag{3.3.17}$$

By digonalizing the Hamiltonian the eigenvalues is

$$eigenvalue = \frac{\alpha + \beta}{2} \pm \sqrt[2]{(\frac{\alpha - \beta}{2}^2) + \lambda^2}. \tag{3.3.18}$$

Note energy of the two-level system is shifted by the coupling λ. Now observe that even though the Hamiltonian is time dependent, the eigenvalues are not.

Unitary transformation and elimination of time

Let us use a unitary transformation on the Hamiltonian H, in two particle system

$$\tilde{H} = e^{-iS} H e^{iS}, \tag{3.3.19}$$

where,

$$S = \omega_1 t c^+ c + \omega_2 t d^+ d. \tag{3.3.20}$$

The Hamiltonian of the two particle system written as:

$$H = H_1 + H_2 + H_3. \tag{3.3.21}$$

So the transformed Hamiltonian for each are given below

$$\tilde{H}_1 = e^{-i\omega_1 t c^+ c - i\omega_2 t d^+ d}(\alpha c^+ c) e^{i\omega_1 t c^+ c + i\omega_2 t d^+ d} = \alpha c^+ c. \quad (3.3.22)$$

Similarly for hole state

$$\tilde{H}_2 = \beta d^+ d. \quad (3.3.23)$$

To compute $\tilde{H}_3$, let us write

$$H_3 = T_1 + T_2, \quad (3.3.24)$$

where $T_1 = \lambda c^+ d e^{i\omega t}$ and T_2 is its Hermitian conjugate.

$$\tilde{T}_1 = e^{-i\omega_1 t c^+ c - i\omega_2 t d^+ d}(\lambda c^+ d e^{i\omega t}) e^{i\omega_1 t c^+ c + i\omega_2 t d^+ d}. \quad (3.3.25)$$

Eq.(3.3.25) can be written in the form of $e^{iBt} A e^{-iBt}$ where $B = -\omega_1 c^+ c - \omega_2 d^+ d$ and $A = \lambda c^+ d e^{i\omega t}$. We know that $[c^+ c, c^+] = c^+$ and $[d^+ d, d] = -d$.

Hence

$$[B, A] = [-\omega_1 c^+ c - \omega_2 d^+ d, c^+ d]\lambda e^{i\omega t}, \quad (3.3.26)$$

$$= (-\omega_1 [c^+ c, c^+] d - \omega_2 c^+ [d^+ d, d])\lambda e^{i\omega t}, \quad (3.3.27)$$

$$= (-\omega_1 + \omega_2)\lambda c^+ d e^{i\omega t}, \quad (3.3.28)$$

$$[B, [B, A]] = (-\omega_1 + \omega_2)^2 c^+ d \lambda e^{i\omega t}. \quad (3.3.29)$$

Then

$$e^{iBt} A e^{-iBt} = A + it[B, A] + \frac{(it)^2}{2!}[B, [B, A]] + ... \quad (3.3.30)$$

So using the expression of Eq.(3.3.30)

$$\tilde{T}_1 = \lambda c^+ d e^{i\omega t} + it(-\omega_1 + \omega_2)\lambda c^+ d e^{i\omega t} + \frac{(it)^2}{2!}(-\omega_1 + \omega_2)^2 \lambda c^+ d e^{i\omega t}, \quad (3.3.31)$$

$= \lambda c^+ d e^{i\omega t}[1 + it(-\omega_1 + \omega_2) + \frac{(it)^2}{2!}(-\omega_1 + \omega_2)^2],$

$=\lambda c^+ d e^{i\omega t} e^{-it(\omega_1 - \omega_2)}.$

When $\omega = \omega_1 - \omega_2$, then

$$\tilde{T}_1 = \lambda c^+ d. \tag{3.3.32}$$

Similarly

$$\tilde{T}_2 = \lambda d^+ c. \tag{3.3.33}$$

So the total transformed Hamiltonian under the condition that $\omega = \omega_1 - \omega_2$ can be written as:

$$\tilde{H} = \tilde{H}_1 + \tilde{H}_2 + \tilde{H}_3 = \alpha c^+ c + \beta d^+ d + \lambda(c^+ d + d^+ c). \tag{3.3.34}$$

Note that $\tilde{H}$ became independent of time unlike H. $\tilde{H}$ has the same energy spectrum as H. The $\tilde{H}_{eff}$ becomes

$$\tilde{H}_{eff} = \tilde{H} + i\hbar(\frac{\partial}{\partial t} e^{-iS}) e^{iS}, \tag{3.3.35}$$

$$\tilde{H}_{eff} = (\alpha + \hbar\omega_1) c^+ c + (\beta + \hbar\omega_2) d^+ d + \lambda(c^+ d + d^+ c). \tag{3.3.36}$$

3.3.2 Hamiltonian for a single particle-hole pair

Let us see the Hamiltonian which describes an electron-hole pair interacting with the classical light field. c^+ is the creation operator for the electron in the conduction band and d^+ the creation hole in valence band. This Hamiltonian resembles a single particle picture of our full Hamiltonian for describing the light induced particle-hole creation in diluted magnetic semiconductor.

$$H = \alpha c^+ c + \beta d^+ d + \lambda(c^+ d^+ e^{i\omega t} + dc e^{-i\omega t}). \tag{3.3.37}$$

This Hamiltonian allows for creation and destruction of one electron and one hole simultaneously. The one hole and one electron state here are coupled to light. For the basis set

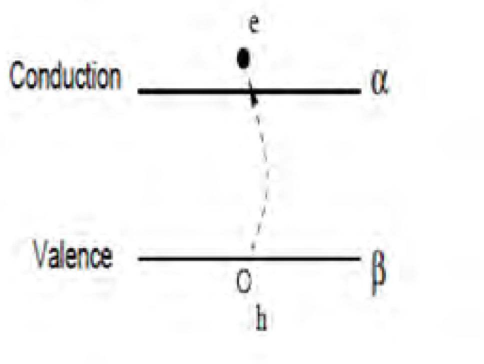

Figure 3.5: Plot of electron(e) in the conduction band and hole(h) in the valence band.

$|1,0>, |0,1>, |1,1>$ and $|0,0>$ the above Hamiltonian can be written in matrix form as

$$H = \begin{pmatrix} \alpha & 0 & 0 & 0 \\ 0 & \beta & 0 & 0 \\ 0 & 0 & \alpha+\beta & \lambda e^{-i\omega t} \\ 0 & 0 & \lambda e^{i\omega t} & 0 \end{pmatrix} \tag{3.3.38}$$

By diagonalizing the above Hamiltonian, the eigenvalues E_i given as

$$E_1, E_2 = \alpha, \beta, \tag{3.3.39}$$

$$E_3, E_4 = \frac{\alpha+\beta}{2} \pm \sqrt{(\frac{\alpha-\beta}{2})^2 + \lambda^2}. \tag{3.3.40}$$

Energy of the two level system is shifted by coupling λ. Even though the Hamiltonian is time dependent, the eigenvalues are not.

Using unitary transformation e^{iS}, the transformed Hamiltonian of single electron-hole pair is given as

$$\tilde{H} = e^{-iS} H e^{iS}, \tag{3.3.41}$$

where,

$$S = \omega_1 t c^+ c + \omega_2 t d^+ d. \tag{3.3.42}$$

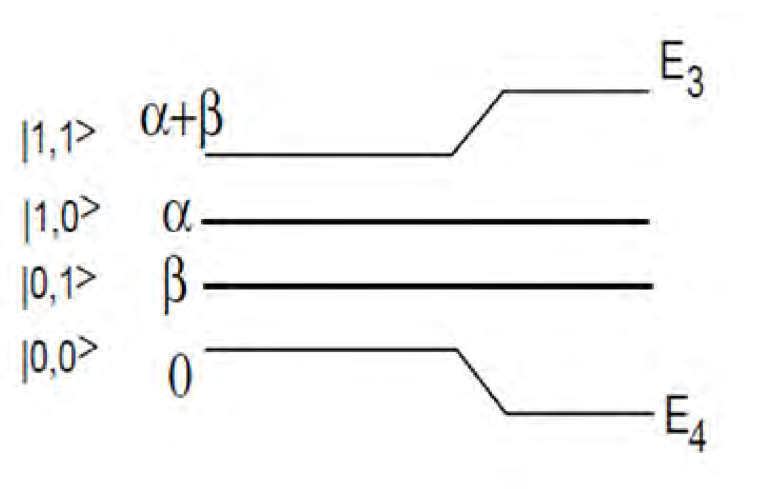

Figure 3.6: Eigne energies of Hamiltonian Eq.(3.3.38). $|n_e, n_h>$ denotes the occupation number state of the electron and hole.

When

$$H = H_1 + H_2 + H_3. \tag{3.3.43}$$

So the transformed Hamiltonian for each are given below

$$\tilde{H}_1 = e^{-i\omega_1 tc^+c - i\omega_2 td^+d}(\alpha c^+c)e^{i\omega_1 tc^+c + i\omega_2 td^+d} = \alpha c^+c. \tag{3.3.44}$$

Similarly for hole state

$$\tilde{H}_2 = \beta d^+d. \tag{3.3.45}$$

To compute $\tilde{H}_3$,let us write

$$H_3 = T_1 + T_2, \tag{3.3.46}$$

where $T_1 = \lambda c^+d^+e^{i\omega t}$ and T_2 is its Hermitian conjugate.

$$\tilde{T}_1 = e^{-i\omega_1 tc^+c - i\omega_2 td^+d}(\lambda c^+d^+e^{i\omega_1 t})e^{i\omega tc^+c + i\omega_2 td^+d}, \tag{3.3.47}$$

$$\tilde{T}_1 = A + it[B, A] + \frac{(it)^2}{2!}[B, [B, A]] + ... \tag{3.3.48}$$

$= \lambda c^+d^+e^{i\omega t} + it(-\omega_1 + \omega_2)\lambda c^+d^+e^{i\omega t} + \frac{(it)^2}{2!}(-\omega_1 + \omega_2)^2\lambda c^+d^+e^{i\omega t},$

$= \lambda c^+d^+e^{i\omega t}[1 + it(-\omega_1 + \omega_2) + \frac{(it)^2}{2!}(-\omega_1 + \omega_2)^2,$

$=\lambda c^+d^+e^{i\omega t}e^{-it(\omega_1 - \omega_2)}.$

When $\omega = \omega_1 - \omega_2$,then

$$\tilde{T}_1 = \lambda c^+ d^+. \tag{3.3.49}$$

Similarly

$$\tilde{T}_2 = \lambda dc. \tag{3.3.50}$$

For $\omega = \omega_1 - \omega_2$, the transformed Hamiltonian is

$$\tilde{H} = \tilde{H}_1 + \tilde{H}_2 + \tilde{H}_3 = \alpha c^+ c + \beta d^+ d + \lambda(c^+ d^+ + dc). \tag{3.3.51}$$

$\tilde{H}$ is independent of time and it has the same energy spectrum as H.

The effective Hamiltonian calculated as

$$\tilde{H}_{eff} = \tilde{H} + i\hbar(\frac{\partial}{\partial t} e^{-iS})e^{iS}, \tag{3.3.52}$$

$$\tilde{H}_{eff} = (\alpha + \hbar\omega_1)c^+ c + (\beta + \hbar\omega_2)d^+ d + \lambda(c^+ d^+ + dc). \tag{3.3.53}$$

3.4 Unitary transformation of the full Hamiltonian

In this section we will perform a unitary transformation on our many particles Hamiltonian of electrons and holes ($Eq.(3.1.1 - 3.1.11)$) interacting among themselves and with a time-dependent classical light field to get rid of the time.

$$H = H_{kin} + H_{ee} + H_{hh} + H_{eh} + H_L(t), \tag{3.4.1}$$

where,

$$H_{kin} = \sum_{k\sigma} (\epsilon_{ck\sigma} c^+_{k\sigma} c_{k\sigma} + \epsilon_{vk\sigma} d^+_{k\sigma} d_{k\sigma}). \tag{3.4.2}$$

Using unitary transformation the transformed Hamiltonian of kinetic energy is given as

$$\tilde{H}_{kin} = e^{it(\Sigma_{k\sigma}(\omega_1 c^+_{k\sigma}c_{k\sigma}+\omega_2 d^+_{k\sigma}d_{k\sigma}))}(\sum_{k\sigma}(\epsilon_{ck\sigma}c^+_{k\sigma}c_{k\sigma} + \epsilon_{vk\sigma}d^+d))e^{-it(\Sigma_{k\sigma}(\omega_1 c^+_{k\sigma}c_{k\sigma}+\omega 2 d^+_{k\sigma}d_{k\sigma})}). \tag{3.4.3}$$

Using $Eq.$3.3.48 properties we can write $Eq.$3.4.3 as

$$\omega_1 \sum_{k\sigma}[c^+_{k\sigma}c_{k\sigma}, c^+_{k\sigma}c_{k\sigma}] + \omega_2 \sum_{k\sigma}[d^+_{k\sigma}d_{k\sigma}, d^+_{k\sigma}d_{k\sigma}]. \tag{3.4.4}$$

The other terms are zero because they refers different particles.

$=c^+_{k\sigma}[c^+_{k\sigma}c_{k\sigma}, c_{k\sigma}] + [c^+_{k\sigma}c_{k\sigma}, c^+_{k\sigma}]c_{k\sigma}+ d^+_{k\sigma}[d^+_{k\sigma}d_{k\sigma}, d_{k\sigma}] + [d^+_{k\sigma}d_{k\sigma}, d^+_{k\sigma}]d_{k\sigma},$

$= -c^+_{k\sigma}c_{k\sigma} + c^+_{k\sigma}c_{k\sigma} - d^+_{k\sigma}d_{k\sigma} + d^+_{k\sigma}d_{k\sigma},$

$=0.$

So [B,A]=0 and also [B,[B,A]] = 0..., Hence

$$\tilde{H}_{kin} = \sum_{k\sigma}(\epsilon_{ck\sigma}c^+_{k\sigma} + \epsilon_{vk\sigma}d^+_{k\sigma}d_{k\sigma}) = H_{kin}. \tag{3.4.5}$$

The transformation of electron-electron part of the Hamiltonian, where,

$$H_{ee} = \frac{1}{2}\sum_{k,k',\sigma,\sigma'} V_q c^+_{k+q\sigma}c^+_{k'-q\sigma'}c_{k'\sigma'}c_{k\sigma}, \tag{3.4.6}$$

$$\tilde{H}_{ee} = e^{it\Sigma_{k\sigma}(\omega_1 c^+_{k\sigma}c_{k\sigma}+\omega_2 d^+_{k\sigma}d_{k\sigma})}(H_{ee})e^{-it\Sigma_{k\sigma}(\omega_1 c^+_{k\sigma}c_{k\sigma}+\omega_2 d^+_{k\sigma}d_{k\sigma})}, \tag{3.4.7}$$

$$[B, A] = [\omega_1 \sum_{K\sigma} c^+_{k\sigma}c_{k\sigma} + \omega_2 \sum_{k\sigma} d^+_{k\sigma}d_{k\sigma}, c^+_{k+q,\sigma}c^+_{k'-q,\sigma'}c_{k'\sigma}c_{k\sigma}]. \tag{3.4.8}$$

The other part is zero because they refers different particles. So

$$[B, A] = \omega_1 \sum_{k\sigma}[c^+_{k\sigma}c_{k\sigma}, c^+_{k+q,\sigma}c^+_{k'-q,\sigma'}c_{k'\sigma}c_{k\sigma}], \tag{3.4.9}$$

$=\omega_1 \sum_k([c^+_{k\sigma}c_{k\sigma}, c^+_{k+q,\sigma}]c^+_{k'-q,\sigma'}c_{k'\sigma'}c_{k\sigma} + c^+_{k+q,\sigma}[c^+_{k\sigma}c_{k\sigma}, c^+_{k'-q,\sigma}]c_{k'\sigma'}c_{k\sigma}$

$+ c^+_{k+q,\sigma}c^+_{k'-q,\sigma}[c^+_{k\sigma}c_{k\sigma}, c_{k'\sigma'}]c_{k\sigma} + c^+_{k+q,\sigma}c^+_{k'-q,\sigma'}c_{k'\sigma'}[c^+_{k\sigma}c_{k\sigma}, c_{k\sigma}],$

$$\begin{aligned}[B, A] = \omega_1 \sum_{k\sigma}(&c^+_{k+q\sigma}c^+_{k'-q\sigma'}c_{k'\sigma'}c_{k\sigma} + c^+_{k+q\sigma}c^+_{k'-q\sigma'}c_{k'\sigma'}c_{k\sigma} \\ &- (c^+_{k+q\sigma}c^+_{k'-q\sigma'}c_{k'\sigma'}c_{k\sigma} + c^+_{k+q\sigma}c^+_{k'-q\sigma'}c_{k'\sigma'}c_{k\sigma})),\end{aligned} \tag{3.4.10}$$

$= 0.$

Hence [B,[B,A]] and other higher order commutators are zero. Therefore

$$\tilde{H}_{ee} = H_{ee}. \tag{3.4.11}$$

Similarly it can be shown that

$$\tilde{H}_{hh} = H_{hh}. \tag{3.4.12}$$

The transformation of the electron-hole part of the Hamiltonian

$$\tilde{H}_{eh} = e^{-iS} H_{eh} e^{iS}. \tag{3.4.13}$$

As before

$$[B, A] = [c^{+}_{k\sigma} c_{k\sigma}, c^{+}_{k+q,\sigma} d^{+}_{k'-q,\sigma'} d_{k'\sigma'} c_{k\sigma}] + [d^{+}_{k\sigma} d_{k\sigma}, c^{+}_{k+q,\sigma} d^{+}_{k'-q,\sigma'} d_{k'\sigma'} c_{k\sigma}], \tag{3.4.14}$$

$= [c^{+}_{k\sigma} c_{k\sigma}, c^{+}_{k+q,\sigma}] d^{+}_{k'-q,\sigma'} d_{k'\sigma'} c_{k\sigma} + c^{+}_{k+q,\sigma} d^{+}_{k'-q,\sigma'} d_{k'\sigma'} [c^{+}_{k\sigma} c_{k\sigma}, c_{k\sigma}]$

$+ c^{+}_{k+q,\sigma} [d^{+}_{k\sigma} d_{k\sigma}, d^{+}_{k'-q,\sigma'}] d_{k'\sigma'} c_{k\sigma} + c^{+}_{k+q,\sigma} d^{+}_{k'-q,\sigma'} [d^{+}_{k\sigma} d_{k\sigma}, d_{k'\sigma'}] c_{k\sigma},$

$= c^{+}_{k+q,\sigma} d^{+}_{k'-q,\sigma'} d_{k'\sigma'} c_{k\sigma} - c^{+}_{k+q,\sigma} d^{+}_{k'-q,\sigma'} d_{k'\sigma'} c_{k\sigma}$

$+ c^{+}_{k+q,\sigma} d^{+}_{k'-q,\sigma'} d_{k'\sigma'} c_{k\sigma} - c^{+}_{k+q,\sigma} d^{+}_{k'-q,\sigma'} d_{k',\sigma'} c_{k,\sigma},$

$=0.$

So it can be shown that $[B, [B, A]]$ and all other higher order commutators vanish. So only the zero order term survives.

$$\tilde{H}_{eh} = H_{eh}. \tag{3.4.15}$$

The transformed light term,

$$[B, A] = [c^{+}_{k\uparrow} c_{k\uparrow}, c^{+}_{k'\uparrow} d^{+}_{-k\downarrow}], \tag{3.4.16}$$

$=[c^{+}_{k\uparrow} c_{k\uparrow}, c^{+}_{k'\uparrow}] d^{+}_{-k\downarrow},$

$= c^{+}_{k\uparrow} d^{+}_{-k\downarrow},$

$=$ A.

So it can be shown that

$$[B,[B,A]] = [B,A] \tag{3.4.17}$$

That means all other higher order commutators. Similarly it holds for the other terms. Therefore, the transformed light part of the Hamiltonian for $c_k^+ \uparrow$ and $d_{-k}^+ \downarrow$ can be written to all orders as

$$\tilde{H}_L = H_L + \lambda \sum_k \rho_k c_{k\uparrow}^+ d_{-k\downarrow}^+ (it\omega_1 + it\omega_2) e^{-i\omega t} + \lambda \sum_k \rho_k c_{k\uparrow}^+ d_{-k\downarrow}^+ \frac{(it\omega_1 + it\omega_2)^2}{2!} e^{-i\omega t} + ..., \tag{3.4.18}$$

$=\lambda \sum_k \rho_k c_{k\uparrow}^+ d_{-k\downarrow}^+ e^{-i\omega t}(1 + it(\omega_1 + \omega_2) + \frac{(it(\omega_1+\omega_2))^2}{2!} + ...,$

$= \lambda \sum_k \rho_k c_{k\uparrow}^+ d_{-k\downarrow}^+ e^{-i\omega t} e^{i(\omega_1+\omega_2)t}.$

If $\omega = \omega_1 + \omega_2$ all the light interaction Hamiltonian are independent of time. So the full light part of the transformed Hamiltonian can be written as:

$$\tilde{H}_L = \lambda \sum_k \rho_k [(c_{k\uparrow}^+ d_{-k\downarrow}^+ + c_{k\downarrow}^+ d_{-k\uparrow}^+) + H.C]. \tag{3.4.19}$$

Then the total transformed Hamiltonian is give as:

$$\tilde{H} = H_{kin} + H_{ee} + H_{hh} + H_{eh} + \tilde{H}_L. \tag{3.4.20}$$

Note that the transformation did not change any other part of the Hamiltonian except the $\tilde{H}_L$ making it time independent.

The system of electrons and holes are in a light bath. As discussed in Eq. (3.2.1) the free energy to be minimized is given as

$$F = \tilde{H} - \frac{\mu}{2} \sum_{K\sigma} c_{k\sigma}^+ c_{k\sigma} - \frac{\mu}{2} \sum_{k\sigma} d_{k\sigma}^+ d_{k\sigma}. \tag{3.4.21}$$

Chapter 4

Solution by Bogoliubov-Valatin transformation

In this chapter let us use Bogoliubov-Valatin (BV) transformation [38-40] on our Hamiltonian to treat the system of electrons and holes in terms of BV quasi particles, so that we can study the system in a mean field way. We need to find ground state energy and the excited state energy of the system. Bogoliubov and Valatin independently applied a quasi-particle picture to study the BCS superconductivity and also Bogoliubov [40] studied Bose-Einstein Condensation in dilute Bose gas. We construct Bogoliubov-Valatin [38, 39] type quasi particles by constructing a canonical unitary transformation and transforming the total time-independent Hamiltonian of electrons and holes to that of quasi-particles.

4.1 construction of the Bogoliubov-Valatin transformation

The Hamiltonian consists of electron creation ($c^+_{k\uparrow}, c^+_{k\downarrow}$) and electron destruction operators ($c_{k\uparrow}, c_{k\downarrow}$) and also consists of hole creation ($d^+_{-k\uparrow}, d^+_{-k\downarrow}$) and hole destruction operators

$(d_{-k\uparrow}, d_{-k\downarrow})$. Let us define four new operators α, β, δ and γ in terms of the above operators using Bogoliubov-Valatin.

$$\alpha_k = u_k c_{k\uparrow} + v_k d^+_{-k\downarrow}, \tag{4.1.1}$$

$$\delta_k = u_k d_{k\downarrow} - v_k c^+_{-k\uparrow}, \tag{4.1.2}$$

$$\beta_k = \bar{u}_k d_{k\uparrow} - \bar{v}_k c^+_{-k\downarrow}, \tag{4.1.3}$$

$$\gamma_k = \bar{u}_k c_{k\downarrow} + \bar{v}_k d^+_{-k\uparrow}. \tag{4.1.4}$$

Here the annihilation operator for a BV quasi particle α_k is a line combination of annihilation operator for an electron with momentum $\vec{k}$ and spin up $(c_{k\uparrow})$ in the conduction band with probability amplitude u_k and a creation operator for a hole $(d^+_{-k\downarrow})$ within the valence band with opposite momentum and spin with probability amplitude v_k. Similarly one can interpret other three Bogoliubov operators. These are the new operators satisfying fermionic commutation relations provided the normalization conditions are satisfied. The normalization conditions are

$$u_k^2 + v_k^2 = 1, \tag{4.1.5}$$

$$\bar{u}_k^2 + \bar{v}_k^2 = 1. \tag{4.1.6}$$

Here $u_k(\bar{u}_k)$ and $v_k(\bar{v}_k)$ are real.

The fermionic canonical commutation relations are given as

$$\delta_{k,k'} = \{\alpha_k, \alpha_k^+\} = \{\delta_k, \delta_k^+\} = \{\beta, \beta_k^+\} = \{\gamma_k, \gamma_k^+\}. \tag{4.1.7}$$

And also

$$0 = \{\alpha_k, \alpha_{k'}\} = \{\delta_k, \delta_{k'}\} = \{\beta_k, \beta_{k'}\} = \{\gamma_k, \gamma_{k'}\} \tag{4.1.8}$$

and special attention should be given to the following two commutators.

$$0 = \{\alpha_k, \delta_{k'}\} = \{\beta_k, \gamma_{k'}\}. \tag{4.1.9}$$

This is true since $u_k = u_{-k}$ and $v_k = v_{-k}$. The operators defined in Eq. (4.1.1-4.1.4) are considered to destroy an excited state of the system(which consists of a correlated electron-hole pair). The hermitian conjugates of the corresponding operators create an excited state of the aforementioned system. By inverting the Eq. (4.1.1-4.1.4), we arrive at relations of electrons and holes in terms of the quasi particle operators. The new relations are given as

$$c_{k\uparrow} = u_k\alpha_k - v_k\delta^+_{-k}, \tag{4.1.10}$$

$$c_{k\downarrow} = \bar{u}_k\gamma_k - \bar{v}_k\beta^+_{-k}, \tag{4.1.11}$$

$$d_{k\uparrow} = \bar{u}_k\beta_k + \bar{v}_k\gamma^+_{-k}, \tag{4.1.12}$$

$$d_{k\downarrow} = u_k\delta_k + v_k\alpha^+_{-k}. \tag{4.1.13}$$

4.2 Transformed Free energy

Using Eq. (4.1.10-4.1.13) and their Hermitian in the Hamiltonian described by Eq. (3.4.21), the transformed free energy is given as

$$\begin{aligned} F_T = &\sum_k(\xi^+_k v^2_k + \xi^-_k \bar{v}^2_k) - \sum_{k,k'} V_{k-k'}\{u_k v_k u_{k'} v_{k'} + \bar{u}_k\bar{v}_k\bar{u}_{k'}\bar{v}_{k'} + v^2_k v^2_{k'} + \bar{v}^2_k\bar{v}^2_{k'}\} \\ &- 2\sum_k \lambda_k\rho_k(u_k v_k + \bar{u}_k\bar{v}_k) + \sum_k[\{\epsilon_{ck\omega} + J_e\langle M\rangle + A_k\}\alpha^+_k\alpha_k \\ &+ \{\epsilon_{vk\omega} - J_h\langle M\rangle + A_k\}\delta^+_k\delta_k + \{\epsilon_{ck\omega} - J_e\langle M\rangle + \bar{A}_k\}\gamma^+_k\gamma_k \\ &+ \{\epsilon_{vk\omega} + J_h\langle M\rangle + \bar{A}_k\}\beta^+_k\beta_k + B_k(\alpha^+_k\delta^+_{-k} + \delta_{-k}\alpha_k) \\ &+ \bar{B}_k(\gamma^+_k\beta^+_{-k} + \beta_{-k}\gamma_k) + O(\alpha^+_k\beta^+_k\gamma^+_k\delta^+_k)], \end{aligned}$$

$$F_T = F_O + F_2 + F_4, \tag{4.2.1}$$

where F_4 contains many terms; each one is a product of four Bogoliubov operators, F_2 describes the sum of the terms containing two operators and F_O is the constant terms involving u_k and v_k. In the transformed free energy of Eq.(4.2.1)

$$A_k = \{\frac{1}{2}\Omega_k - (\xi^+_k + \Omega_k)v^2_k\} + \Delta_k u_k v_k, \tag{4.2.2}$$

$$B_k = \{\frac{1}{2}\Delta_k(u^2_k - v^2_k) - u_k v_k(\xi^+_k + \Omega_k)\} \tag{4.2.3}$$

and similarly for $\bar{A}_k$ and $\bar{B}_k$ the expressions where all unbarred quantities are replaced by barred quantities and ξ_k^+ by ξ_k^-. The symbols used in the equations are as follows.

$$\xi^{\pm} = \xi_k^0 \pm J\langle M\rangle, \tag{4.2.4}$$

$$\xi_0 = E_g + \frac{\hbar^2 k^2}{2m^*}, \tag{4.2.5}$$

$$J = J_e - J_h, \tag{4.2.6}$$

$$\frac{1}{m*} = \frac{1}{m_e} + \frac{1}{m_h}, \tag{4.2.7}$$

$$\epsilon_{ck\omega} = E_c(k) - \frac{\hbar\omega}{2}, \tag{4.2.8}$$

$$\epsilon_{vk\omega} = E_v(k) - \frac{\hbar\omega}{2}, \tag{4.2.9}$$

$$\Delta_k = \Delta_k^0 + 2\lambda\rho_k, \tag{4.2.10}$$

$$\Delta_k^0 = 2\sum_{k'} V_{k-k'} u_{k'} v_{k'}, \tag{4.2.11}$$

$$\bar{\Delta}_k = \bar{\Delta}_k^0 + 2\lambda\rho_k, \tag{4.2.12}$$

$$\bar{\Delta}_k^0 = 2\sum_{k'} V_{k-k'} \bar{u}_{k'} \bar{v}_{k'}, \tag{4.2.13}$$

$$\Omega_k = -2\sum_{k'} V_{k-k'} v_{k'}^2, \tag{4.2.14}$$

$$\bar{\Omega}_k = -2\sum_{k'} V_{k-k'} \bar{v}_{k'}^2. \tag{4.2.15}$$

4.3 The Free energy(F_0)

Since the interested one is the ground state energy of the system, after neglecting F_4 which is small [38, 41] and the condition that the off diagonal terms in F_2 vanish so that the free energy is digonalized, then ground free energy is given as

$$F_0 = \sum_k (\xi_k^+ v_k^2 + \xi_k^- \bar{v}_k^2) - \sum_{k,k'} V_{k-k'} \{u_k v_k u_{k'} v_{k'} + \bar{u}_k \bar{v}_k \bar{u}_{k'} \bar{v}_{k'} + v_k^2 v_{k'}^2 + \bar{v}_k^2 \bar{v}_{k'}^2\}$$
$$- 2\sum_k \lambda_k \rho_k (u_k v_k + \bar{u}_k \bar{v}_k). \quad (4.3.1)$$

By minimizing the free energy with respect to v_k

$$\frac{\partial F_0}{\partial v_k} = 0, \quad (4.3.2)$$

$$0 = \sum_k 2\xi_k^+ v_k - 2\sum_{kk'} V_{k,k'} [\sqrt{v_{k'}^2 - v_{k'}^4} \frac{1}{2} (v^2 - v_k^4)^{-1/2} (2v_k - 4v_k^3)$$
$$+ \sqrt{v_k^2 - v_k^4} \frac{1}{2} (v_k^2 - v_k^4)^{-1/2} (2v_{k'} - 4v_{k'}^3)$$
$$+ 2v_k v_{k'}^2 + 2v_k^2 v_{k'}] - 2\sum_k \lambda_k \rho_k \frac{1}{2} (v_k^2 - v_k^4)^{-1/2} (2v_k - 4v_k^3), \quad (4.3.3)$$

$$= \sum_k 2\xi_k^+ v_k - 2\sum_{kk'} V_{k,k'} [v_{k'} \sqrt{1 - v_{k'}^2} \frac{1 - 2v_k^2}{\sqrt{1 - v_k^2}} + 2v_k v_{k'}^2] - 2\sum_k \lambda_k \rho_k \frac{1 - 2v_k^2}{\sqrt{1 - v_k^2}}, \quad (4.3.4)$$

$$= \sum_k 2\xi_k^+ v_k - 2\sum_{kk'} V_{k,k'} [u_{k'} v_{k'} (\frac{u_k^2 - v_k^2}{u_k}) + 2u_k u_{k'}^2] - 2\sum_k \lambda_k \rho_k \frac{(u_k^2 - v_k^2)}{u_k}. \quad (4.3.5)$$

Multiplying Eq.(4.3.5) by u_k,we get

$$0 = \sum_k 2\xi_k^+ u_k v_k - 2\sum_{kk'} V_{kk'} [u_{k'} v_{k'} (u_k^2 - v_k^2) + 2u_k v_k v_{k'}^2] - 2\sum_k \lambda_k \rho_k (u_k^2 - v_k^2) \quad (4.3.6)$$

$=2\{\sum_k u_k v_k (\xi_k^+ - 2\sum_{kk'} V_{kk'} v_{k'}^2) - \sum_{kk'} V_{kk'} u_{k'} v_{k'} (u_k^2 - v_k^2) - \sum_k \lambda_k \rho_k (u_k^2 - v_k^2)\}$,

$=2\{\sum_k u_k v_k (\xi_k^+ + \Omega_k) - \frac{1}{2} \sum_k (u_k^2 - v_k^2)(2\lambda_k \rho_k + 2\sum_k V_{kk'} u_{k'} v_{k'})\}$,

$$0 = 2\{\sum_k u_k v_k (\xi_k^+ + \Omega_k) - \frac{1}{2} \Delta_k \sum_k (u_k^2 - v_k^2)\}. \quad (4.3.7)$$

Using the normalization condition

$$v_k^2 = \frac{1}{2}[1 - \frac{\xi_k^+ + \Omega_k}{\sqrt{\Delta_k^2 + (\xi_k^+ + \Omega_k)^2}}]. \tag{4.3.8}$$

Similar if we minimize the free energy with respect to u_k, $\bar{u}_k$ and $\bar{v}_k$ and the normalization condition we get the condition as

$$u_k^2 = \frac{1}{2}[1 + \frac{\xi_k^+ + \Omega_k}{\sqrt{\Delta_k^2 + (\xi_k^+ + \Omega_k)^2}}], \tag{4.3.9}$$

$$\bar{u}_k^2 = \frac{1}{2}[1 + \frac{\xi_k^- + \bar{\Omega}_k}{\sqrt{\bar{\Delta}_k^2 + (\xi_k^- + \bar{\Omega}_k)^2}}] \tag{4.3.10}$$

and

$$\bar{v}_k^2 = \frac{1}{2}[1 - \frac{\xi_k^- + \bar{\Omega}_k}{\sqrt{\bar{\Delta}_k^2 + (\xi^- + \bar{\Omega}_k)^2}}], \tag{4.3.11}$$

respectively.

The ground free energy ($< F_0 >$), in the Eq.(4.2.1) can be written as:

$$\begin{aligned} F_0(< M >) = \sum_k [(\xi_k^+ + \frac{\Omega_k}{2})v_k^2 + (\xi_k^- + \frac{\bar{\Omega}_k}{2})\bar{v}_k^2 - \frac{\Delta_k u_k v_k + \bar{\Delta}\bar{u}_k\bar{v}_k}{2} \\ - \lambda\rho_k(u_k v_k + \bar{u}_k\bar{v}_k). \end{aligned} \tag{4.3.12}$$

Using the unitary condition to solve for u_k, v_k, $\bar{u}_k$, $\bar{v}_k$ and solve self constantly for Δ_k, $\bar{\Delta}_k$, Ω_k and$\bar{\Omega}_k$ to calculate Free energy.

4.4 Contact interaction

4.4.1 Case(i):$\rho_k = 1$

The amplitude ρ_k is assumed to be independent of $\vec{k}$ and is taken as $\rho_0 = 1$ and $\lambda = \lambda_0$. Within this approximation, we calculate the free energy.

4.4.2 Case(ii): $\rho(k) = \frac{1}{(1+(a_{ex}k)^2)^2}$

The k-dependent $\rho(k)$ is due to the fact that this is the fourier transform[42] of the 1s hydrogen wave function which describes an exciton of electron and hole of the system. For GaAs $a_{ex} \approx 100a_B, a_B = 0.529\dot{A}$. For $\rho(k) = \frac{1}{(1+(a_{ex}k)^2)^2}$ the free energy given in Eq.(4.3.16) evaluate numerically by determining $\Delta, \bar{\Delta}, \Omega, \bar{\Omega}, u, v, \bar{u}$ and $\bar{v}$ for each k.

Chapter 5

Results and Discussions

In this Chapter we study para to ferro-magnetic transition of magnetic moments by the incident light by calculating the magnetization in a mean-field way by using the free energy expression Eq.4.3.1 or (4.3.14) of chapter 4 and study different scenarios of dependence of transition on various physical quantities. We generally consider a contact interaction between electrons and holes and k-independent ρ (probability of light coupling to particles). We have also studied more general case of a k-dependent $\rho(k)$.

5.1 Calculation of magnetization in a mean-field way

In our system the light induced free carriers such as electrons and holes interact with the localized moments antiferromagnetically and there by induce a ferromagnetic interaction between impurity magnetic moments. These moments themselves constitute a many-body magnetic system. We replace this interacting magnetic moments in a mean-field way. The main idea is to replace all many body interactions to any one body with an average or effective interaction. This reduces the many-body problem into an effective one-body problem of magnetic moments. Each magnetic moment feels a mean-field of the other magnetic moments. In Fig.5.1, we give the picture of mean-field representation of a magnetic moment.

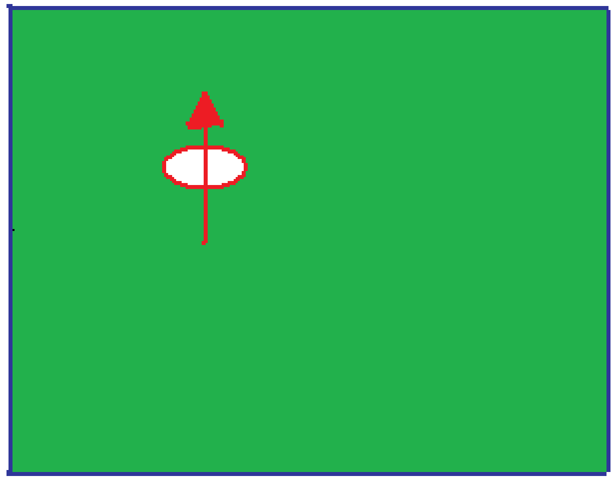

Figure 5.1: Mean field representation of interaction of impurity moment. The figure shows that the many body system is represented by a single body in an average way.

The effective magnetic field experienced by a single magnetic moment M due to the rest of the moments is given as

$$H_{eff} = -\frac{\partial F_0(M)}{\partial M}. \quad (5.1.1)$$

Thus the self-consistent equation for magnetization to be solved is

$$< M(T) >= g\mu_B(\sum_{S=-5/2}^{+5/2} Se^{-\beta H_{eff}S})/(\sum_{-5/2}^{+5/2} e^{-\beta H_{eff}S}), \quad (5.1.2)$$

where $\beta = (k_\beta T)^{-1}$ and $< M(S) > = g\mu_B < S >$. The above two equations are solved self-consistently for a given temperature using the free energy expression of Eq.(4.3.14) and calculated magnetic field.

By expanding the magnetization near T_c, the analytical expression of T_c in the limit of $V_O \rightarrow 0$ is given by

$$k_\beta T_c = \frac{4J^2cS(S+1)}{3g\mu_B} \quad (5.1.3)$$

Interaction parameters: the following parameters are taken in my calculation[27]

μ = 1.8eV,

J = 34meV $\dot{A}^3$,

a =5.65$\dot{A}$,

c = $\frac{x}{a^3/4}$ = 0.014x/$\dot{A}^3$,

$m^* = 0.1m_e$ and

U_0=1200eV$\dot{A}^3$.

The effect of light coupling(λ) on free energy as function of magnetization is shown in fig.5.2.

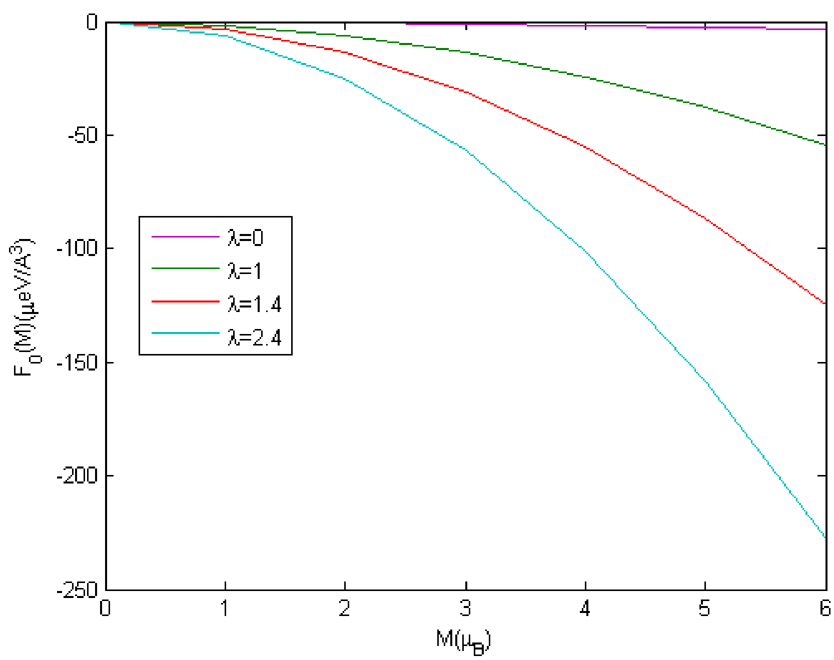

Figure 5.2: Free energy as a function of magnetic moments(M)for different values of light coupling λ(in units of eV)

The figure shows the variation of free energy as function of magnetization for different values of light coupling(λ). For particular light coupling the free energy decrease as a function of the average Mn magnetic moment and free energy increased as function of magnetic moment if the light coupling decreased. When $\lambda = 0$(no coupling of light to particles) or when energy of light is below the energy gap there is no excitation of electron and hole.

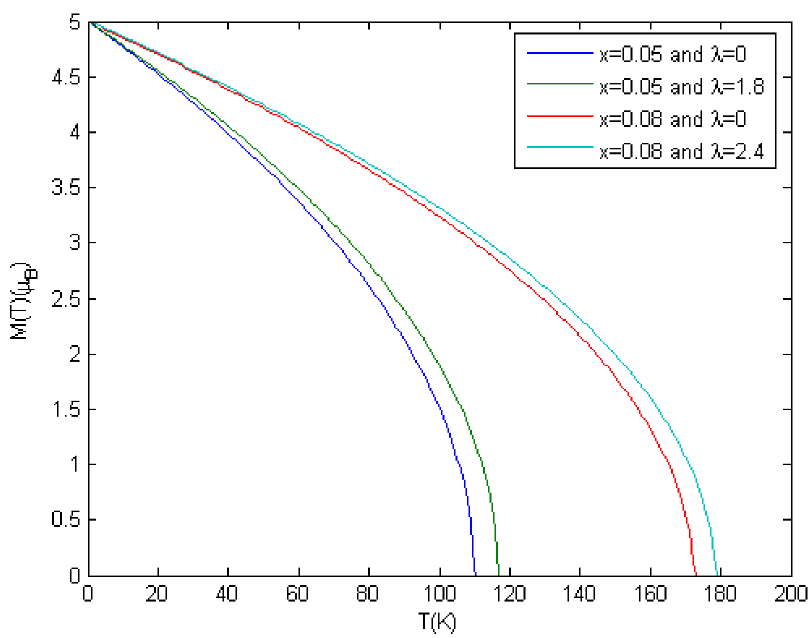

Figure 5.3: Magnetization M(T) as a function of temperature T for different values of light coupling(in units of eV) and concentration of magnetic impurity

The variation of the magnetic moment as a function of temperature for different values of λ and magnetic concentration as keeping other parameter fixed shown in fig 5.3. The figure shows a phase transition from paramagnetic to ferromagnetic state as temperature decrease. With increase in the light coupling more and more electron- hole pairs are created, thereby increasing the magnetization and hence the critical temperature is increased. But more increasing light coupling there is decrease in the critical temperature. This is due to the competition between particle-particle interaction and the light energy of the system

Chapter 6

CONCLUSION

The paper focused on the study of the photoinduced magnetization in the dilute magnetic semiconductors in a model Hamiltonian which takes into account the Coulomb interaction, light-matter interaction, and the exchange interactions between carriers and the localized magnetic moments. The model assumed no direct interaction between the localized moments and no free carriers without the incident light, so that the system is paramagnetic without the presence of light and can be ferromagnetic only via the indirect interaction with the photoinduced carriers.

The time dependence in the Hamiltonian was eliminated by a unitary transformation and the transformed Hamiltonian was then diagonalized by the Bogoliubov-Valatin transformation. From the calculated free energy, we computed the magnetization of the system as a function of temperature. There is a para- to ferromagnetic transition as temperature is decreased with the transition temperature T_c depending on the various parameters. The magnetization and critical temperature is dependent on the photon energy incident on the system. By increasing the light coupling the transition temperature increase. Also by increasing the frequency, the number of photoexcited electron-hole pairs is increased, which in turn results in a stronger magnetic interaction between the localized moments and a higher value for the T_c. For typical parameters, for the dilute magnetic semiconductors T_c is predicted in the presence of the photoexcited carriers. For the dilute magnetic semiconductors such as GaAs(Mn), where the carriers are already present even without the incident light, the additional photoexcited carriers by the laser radiation should increase the magnitude of T_c[32].

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Declaration

This thesis is my original work, has not been presented for a degree in any other University and that all the sources of material used for the thesis have been dully acknowledged.

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

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