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自聚型半導體量子點中多激子問題的研究

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中文摘要

自組式半導體量子點的電子結構及其多激子理論之研究

關鍵詞: 量子點、激子、螢光、螢光激發

由於自組式(self-assembled)半導體量子點(semiconductor quantum dots)高品質的光電特性及原子般的電子結構，人们對於利用此一量子點實現新一代光電元件深寄厚望。一般而言量子點中的光電過程主要涉及電子電洞對(electron-hole pairs)的產生或湮滅，在此奈米尺度的結構中電子電洞對緊密地束縛在一起而形成多激子聚合體(multi-exciton complexes)，此聚合體中的庫倫作用力相當強烈而且複雜，因此如何研究量子點中的多激子並發展相關理論便成為一項挑戰。本計劃的目的是對自組式量子點的電子結構及其多激子性質進行理論研究。本計劃主要由下列主題構成:

1. 量子點電子結構的計算

在本主題中我們將採用多重尺度計算的方法研究量子點的單電子電子結構:包括多能帶 k.p 法，緊密束縛法。我們發展的模擬程式將可普遍適用於各種量子點材料、形狀和結構。我們將定量地考慮真實量子點的構成元素包括其應力分布,化學成份及幾何形狀。其中 k.p 法適合較大量子點的計算並且可以很容易地與應力計算結果相結合。緊密束縛法精密地考慮電子波函數到原子尺度所以可以更有效地考慮化學成份擴散的問題，但計算量也因此變得龐大而需要更有效的高階演算法(例如 ARPACK eigensolver)和計憶體容量。我們的計算結果將與原子力顯微鏡量測和螢光(Photoluminescence)實驗結果驗證並提供多激子計算中量子點模型的依據。

2. 多激子(multi-exciton)問題研究

利用 configuration interaction (CI) 的方法及 exact diagonalization (ED)的技術我們將計算量子點中多激子的量子態及能量，由於量子點內的粒子數有限,CI 和 ED 的方法數值上得以求得幾近正解的結果並同時允許深刻的物理解析。在我們先前的研究中我們已計算出激子數達十二個的多激子基態和化學能。計算多激子問題的主要困難在於隨著激子數的增加哈密頓矩陣變得相當龐大以至於一般的個人電腦設備和數值函式庫無法解決其問題。在本計劃中我們將運用高等的ARPACK數值演算方法配合高速高容量(>2G RAM)的工作站級個人電腦計算完整的基態和激發態,掌握量子點所有激子量子態的資訊後我們便可以進一步計算量子點完整的光譜性質。

3. 單量子點(single quantum dot)光譜:非對稱量子點和激子的動態行為

最近量子點光譜實驗的快速發展引發相關理論普遍的研究興趣，包括量子點螢光(Photoluminescence)，螢光激發(Photoluminescence excitation) 和時析(time resolved)光譜學，在我們先前的研究中我們已對量子點的強磁場螢光進行深入的分析。最新的光譜實驗(Robin Williams *et. al,* NRC)已量得單量子點豐富的光譜精細結構。實驗顯示理想量子點的對稱性似乎受到某程度的破壞，我們初步的研究發現其非對稱性活化許多暗激子(dark exciton)進而造成複雜的光譜結構。但是要完整解釋測量的單量子點光譜必需對激子再結合(recombination)前的動態過程有所了解，激子的動態行為與其幅射(radiative)與非幅射(non-radiative)的鬆弛(relaxation)過程有關並直接影響量子點內激子數目的統計分佈,目前為止相關研究並未確實地考慮量子點的真實電子結構,我們將在本計劃中探討其對量子點光譜及激子數統計分佈的影響。

英文摘要

Theoretical investigation of exciton complexes in semiconductor self-assembled quantum dots

Keywords: quantum dots, exciton, photoluminescence, photoluminescence excitation

With the atomic-like energy spectrum and the high quality of the optical properties, self-assembled quantum dots (SAQD's) are considered as attractive candidates for advanced optoelectronic applications. Most optical processes in SAQD's involve either recombination or creation of electron-hole (e-h) pairs from the multi-exciton complexes in dots, strongly bound via Coulomb attraction further enhanced by dot confinement. However, a thorough theoretical study of those multi-exciton complexes in quantum dots is still a challenge due to the complication of the Coulomb interactions, which involve e-e, h-h, and e-h interactions. The objective of this project is to develop theory to investigate the electronic structure, optical spectrum and dynamical processes of multi-exciton complexes in semiconductor SAQD's.

This project consists of the following main parts:

1. Electronic structure of SAQD's

   In this part, we compute the electronic structure of SAQD's using multi-scale approaches, including the macroscopic k.p theory and the atom-scaled tight-binding theory. Realistic effects in strained SAQD's, including those of strain distribution, composition intermixing, dot shape, will be taken into account. The k.p model is a valuable way to calculate the electronic structure of large-size strained dot, and can be easily implemented using conventional. The tight-binding theory provides more precise description of electronic states in basis of atom sites of semiconductors and is a more natural way to consider the effect of composition intermixing. The tight-binding model, however, require much more numerical capacity and in usual is feasible only for small-dot calculation. To take the both advantages of the macroscopic k.p and microscopic tight-binding approaches, we shall try to develop a multi-scale theory properly combining both approaches. The simulation results provide useful information for the modeling of SAQD's and are the base to further study many-particle physics in dots.

2. Multi-exciton complexes

   In our previous studies, we have successfully computed the low-lying states and the chemical potential of multi-exciton in dots using the method of configuration interaction (CI) and exact diagonalization (ED) technique. In this project, we plan to calculate complete electronic spectrum of multi-exciton (with exciton number up to ~10), including the ground and "all" excited states, using ED technique combined with advanced numerical eigensolver (e.g. ARPACK solver) and advanced computation facility. A complete optical spectrum from dots can be calculated only if the complete electronic spectrum of the multi-exciton is calculated. The numerically calculated "exact" spectrum is compared with those analytically calculated using CI method, in which rich physical insight can be more easily captured.

3. Single-dot spectroscopy: dot deformation and dynamic processes of excitons

   The latest result of single-dot spectroscopy (by R. Williams *et al*) has successfully revealed rich aspect of fine structure in the PL spectrum from a single InAs/InP dot. The measured results indicate the fine structure mainly arisen from multi-exciton effects and the broken-symmetry of deformed dot due to strain or irregular dot shape. We shall study the both

effects of deformed SAQD's on the energy and optical spectra. To quantitatively explain the measured spectra, including the excitation power dependence of spectra and time-resolved spectroscopy, we shall study the dynamical relaxation process of the photoexcited excitons, which determine the occupation number distribution in dots and directly affect the measured spectrum feature.

目錄

報告內容

Introduction（前言）:

In the past three years, we have successfully built up a computation laboratory at Department of Electrophyscs, NCTU, equipped with advanced computation facilities financially supported by National Science Council of R.O.C. We sincerely acknowledge NSC for that. The computation facilities (mainly a 64-bit workstation PC) incorporated with advanced developed numerical techniques (conjugated gradient method and Lanczos eigen solver) are key techniques in our research in the field of computational nano-technology and physics —the main subject of this project. With the financial support, the number of my group member increases year by year and currently the group consists of two PhD and four master graduate students. Based on the facilities and human resources, we have completed the investigations on the subjects originally planned in the original projects, and published 14 SCI journal papers since 2003. Besides, we also explore the new research field of semi-magnetic semiconductors, a currently attractive and popular subject in condensed matter physics.

In the following, we will report on each research topic in this project, including

1. Electronic structure of semiconductor nanostructures

2. Multi-exciton complexes in quantum dots

3. Single-dot spectroscopy

4. Semi-magnetic semiconductor nanostrcutrures

We will describe the task that we have done, summarize the results that we have revealed or predicted, and finally shortly describe the study plan in the coming year.

1. Electronic structure of self-assembled quantum dot

Objective(研究目的):

In this topic, we intend to develop a computational tool to simulate the electronic structure of semiconductor nanostructures, including self-assembled quantum dots and colloidal nanocrystals. The tool should provide a theoretical support to physically and quantitatively illustrate experimental data and inspire new design of advanced devices. The theoretical approaches are developed towards a multi-scales version, including k.p model, tight-binding theory, and pseudopotential method, for self-assembled dots whose formation involves complex factors, including strains, composition intermixing, and the complex nature of valence bands.

Approaches (研究方法):

1. Set-up of computation techniques and environment

For the studies of the many-body physics of few-particle (electron, or quasi-particle like exciton) in quantum dots, we employ the configuration interaction (CI) method and the exact diagonalization (ED) technique. The combination of the both approaches gives the possibility to calculate the eigen state of few particle in extremely high accuracy (nearly being “the exact solution”) if the number of configuration is sufficiently large. Thus, a numerical eigen solver for large matrix (typically $>>10^2 \times 10^2$) is required. Two advanced numerical eigen solvers, conjugated gradient method and Lanczos eigen solver, are being in development in this project.[1]

- Conjugated gradient method

  We have developed a numerical code using the conjugated gradient algorithm to solve the problem. The code is developed using C language and can be easily incorporated with any C program. With that, we can calculate the few number of the ground and excited states for a large Hamiltonian matrix with the size $\sim 10^4 \times 10^4$ (the computation limit of conventional eigen problem subroutine like LAPACK is less than $10^3 \times 10^3$). The eigen solver has been successfully implemented in the studies of the ground state of quantum Hall droplet in self-assembled dots with exciton number Nx<8 .[2]

- Lanczos eigen solver

  In order to calculate the full emission spectrum, we actually need the information about the "all" excited states of multi-exciton. The number of the excited states that are involved in optical spectrum is very high particularly for the cases of high exciton number and strongly interacting dots.

  The problem of finding the eigenvalues of large sparse matrices involves computer memory usage and the computation time. The Lanczos method is a very simple and yet effective algorithm for the large size sparse matrices. It is based upon Lanczos recursion for tridiagonalizing the matrix. Given a large sparse matrix A and a starting vector v1 which is generated randomly, the Lanczos recursion implements a Gram-Schmidt orthogonalization of the matrix-vector products Avi corresponding to the Lanczos vector vi generated by the recursion. Since Lanczos method needs only to access the matrix through matrix-vector multiplications, we don't need to store all the elements of the matrix. So that Lanczos algorithm reduces the computer memory usage.

  Here, we are trying to use the Lanczos eigen solver developed by Dr. K.S. Wu to solve the problem [1]. The code is freely released. Nevertheless, we have to create a interface that is user-friendly and can be well incorporate into the code of CI calculation. The code for the interface is being developed. So far, we can already solve the eigen problem of a $10^3 \times 10^3$ Hamiltonian matrix using the Lanczos solver.

Results and Discussions（結果討論與計畫成果自評）:

We develop a multi-band k.p theory to calculate the electronic structure of semiconductor nanostructures (III-V SADs and colloidal nanocrystals) with the consideration of the realistic effects, including the strain distribution, composition diffusion, and Coulomb interactions. The theory is numerically implemented in the basis of plane waves, with typical number $>10^4$ for convergence. [1,7]

The developed code of four-band k.p theory has been transferred to experimentists (prof. H.H.Lin NTU) to design and fabricate advanced nano-devices (far-infrared laser devices) based on InAs/GaAs SADs. The calculated strain distribution in pyramid shaped SADs is shown in Fig.1 (b). The calculated energy spectra of a single electron and a single hole of strained SADs as function of the dot base size are shown in Fig.2. The calculated results show the effects of quantum size and strains in SADs significantly change the effective energy gap of semiconductor. The valence hole energy spectrum of strained SADs are found not monotonically increased with decreasing the dot size. Properly doping small amount of In ion into the GaAs capping layer on SADs can engineer the effective energy gap to match the optimal wave length ~1.3 um for optical communication. Calculated results for a conduction electron in SADs support the simplified

parabolic model of the confining potential, which have widely used in the many-body calculations for SADs. The code is programmed in the manner so that more remote band can be easily included in the future.

Basically, the studies of this subject reach the goal and meet the schedule planned in the original proposal. The theoretical results are in agreement with experiments carried out by the experimental group of Prof. Hao-Hsiung Lin at NTU/EE. A journal paper based on the work, combined with the experimental results given by the group at NTU/EE, is in intensive preparation. Although multi-band theory have been previously used for the calculation of the electronic structure of SADs by several theory group around the world, we are one of the very few groups in Taiwan that are capable of simulating the electronic structure of SADs using the multi-band theory. We believe that the developed theory provide valuable tool for both theoretical and experimental studies of nanostructures and the development of nanotechnology in Taiwan.

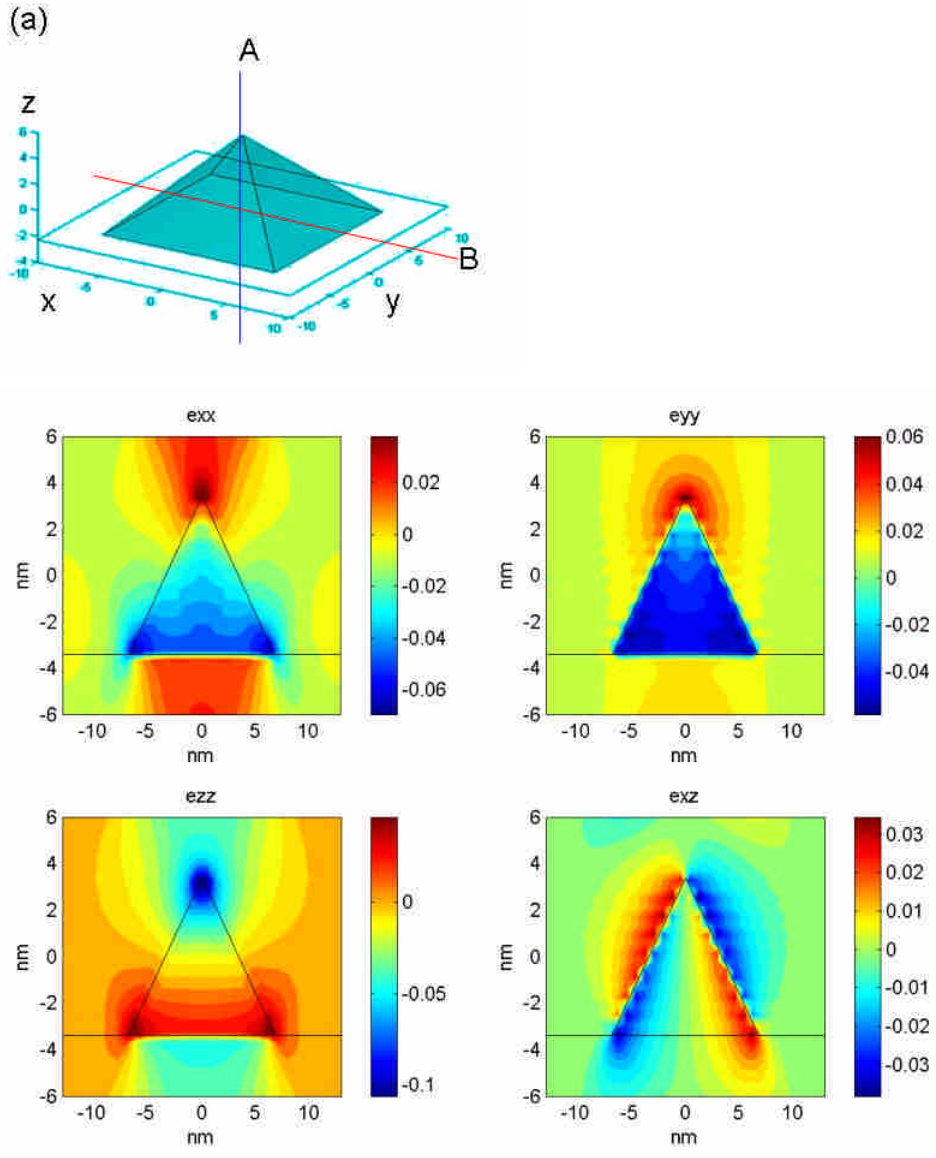

Fig.1 Calculated strain profiles in a pyramid shaped self-assembled quantum dots (13.6nmx13.6nmx6.8nm).

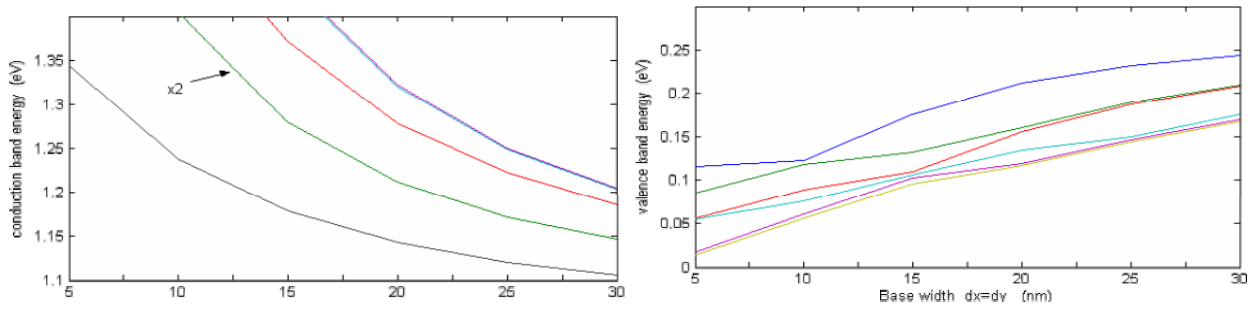

Fig.2 Calculated energy spectra of a single electron (left) and a single valence hole (right) in the strained SADs of various base size (height is set to 5nm).

2.Multi-exciton complexes in quantum dots

In this subject, we develop the theory and the computation technique to study the multi-exciton physics and optical spectra of self-assembled quantum dots (SADs). Multi-exciton states and their spectrum are calculated by using the method of configuration interaction (CI) and the technique of exact diagonalization (ED).

(a) Magneto-Exciton complexes

Our theory has previously accounted for the recent magneto-PL measurement on SAD ensembles by *S. Raymond et al.* We pointed out that the correlation in multi-exciton complexes turns out to be pronounced as the shell electronic structure of dot is formed at special strength of magnetic fields. Beside, we also find the signatures in the experiment associated with *hidden symmetry*, an important nature of multi-exciton complexes.

(b) Excitonic quantum Hall droplets

It is known that number of interesting and fascinating physical phenomena have been revealed in the quantum dots in the quantum Hall regime. In this work, we present the first theory for excitonic quantum Hall droplets (EXQHD) in a self-assembled quantum dot (SAD) subject
to strong magnetic field B (up to few tens Tesla). Using the ED technique implemented with the conjugated gradient method we determine the ground and excited states, the stability against spin flips, and the optical emission spectrum of the nu=2 EXQHDs (see Fig.1). In contrast with one component electronic droplets, the singlet-singlet nu=2 EXQHD is found to be intrinsically correlated, and stable even in high magnetic fields due to the neutrality of exciton. The characteristic spin related emission spectrum and its magnetic field evolution from the EXQHD is predicted. We predict that, unlike the one-component Hall droplets, an EXQHD possess different nature of the ground state. Owing to the intrinsic correlation interactions in the neutral quasi-particles, the EXQHD at the filling factor $\nu=2$ (a spin-unpolarized state) is stable as the ground state against fields increasing. The studies provide a important tool to interpret the magneto-optical experiments on SADs in high magnetic fields.[2]

3. Single-dot spectroscopy: charged dots and symmetry breaking

It has been experimentally demonstrated that charged exciton X^{-n} can be *electrically* created in a self-assembled quantum dot embedded in p-n junction. With long coherence time, the spin charged exciton states of quantum dots are promising candidate to represent qubit in quantum

computing. Thus, the exists the need to identify the spin character of charged exciton states of SADs.[3]

In this work, we calculate the spin characteristic pattern of optical absorption and emission in charge tunable quantum dots using the CI method.(see Fig.3) The calculated results account for the recent measurement of photoluminescence excitation on those charge tunable dots by *E. Ware et al*.. We have found the asymmetry quantum dot, deformed by strain or irregularity of shape, exhibit the complex feature of emission spectrum, caused by deformation-induced activation of dark exciton states (see Fig.3)[4]. The theory also explains the recent measurement on single deformed InAs/InP SAD by *D. Chithrani et al.* We find that the dot-shape deformation selectively activate spin-singlet *dark* exciton and creates number of additional fine structure in the PL spectrum from an asymmetric dot. [4]

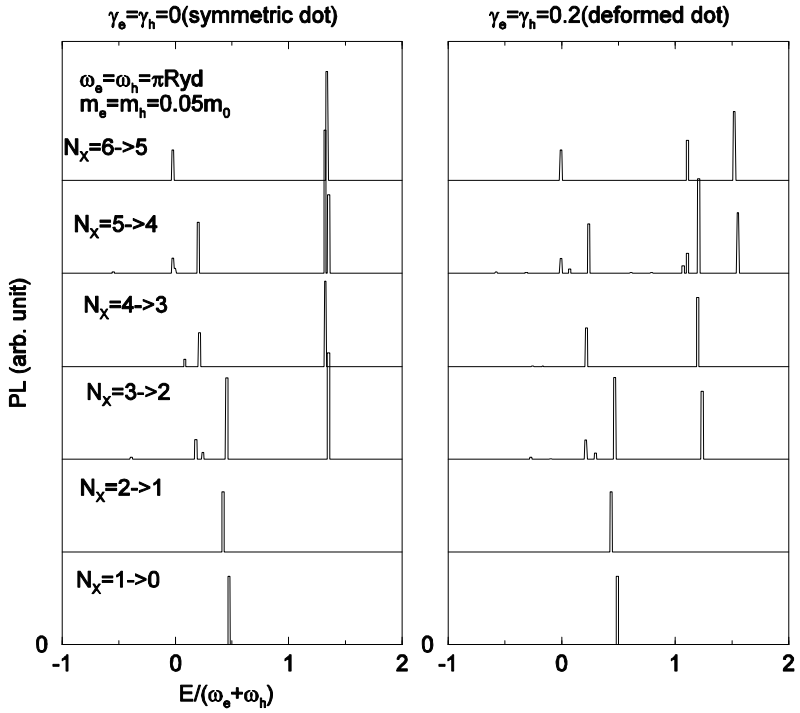

Fig.3 Emission spectra of symmetry dot (left) and asymmetry dot (right) as function of exciton number (excitation power)

4. Semi-magnetic semiconductor nanostrcututres

Objective(研究目的):

In recent years, magnetic ions (typically Mn2+) have been successfully incorporated into both self-assembled semiconductor quantum dot and chemically synthesized dots.[5] The studies of optical spectroscopy on the (semi-) magnetic nanostructures have explored and revealed number of interesting physics. With those techniques, fabricating magnetic nanostructures with controllable individual spin of magnetic ions becomes possible, and has triggered a series of concepts to apply those magnetic nanostructures in “spintronics”.

Approaches (研究方法):

In the work, we present the theoretical results on the electronic structure and magnetic properties of II-VI semiconductor nanocrystals containing few electrons and a single magnetic ion impurity. To study the magnetic properties and particle-particle interaction of the semi-magnetic

nanocrystals, we develop the CI method based on hard-wall sphere model for nanocrystals.[3] We derive the explicit formulation of the Coulomb matrix elements in the basis of single-particle states of quantum sphere and calculate the electron-Mn energy spectra. In fact, the developed theory is applicable for any other kinds of sphere-like nanostructures and can be straightforward extended to perform exact diagonalization calculation.

Model 1: hard-wall sphere

We take the widely used hard-wall spherical potential to model the confinement of chemically synthesized colloidal nanocrystals. The typical diameter of the quantum dots is about 3-10nm depending on the condition of synthesis and material. In the model, the energy spectrum and wave function are known and explicitly given in terms of special Bessel function and spherical harmonic function.

The single-particle energy spectrum vs the quantum number m(the z-component of angular momentum) is show in Fig4.

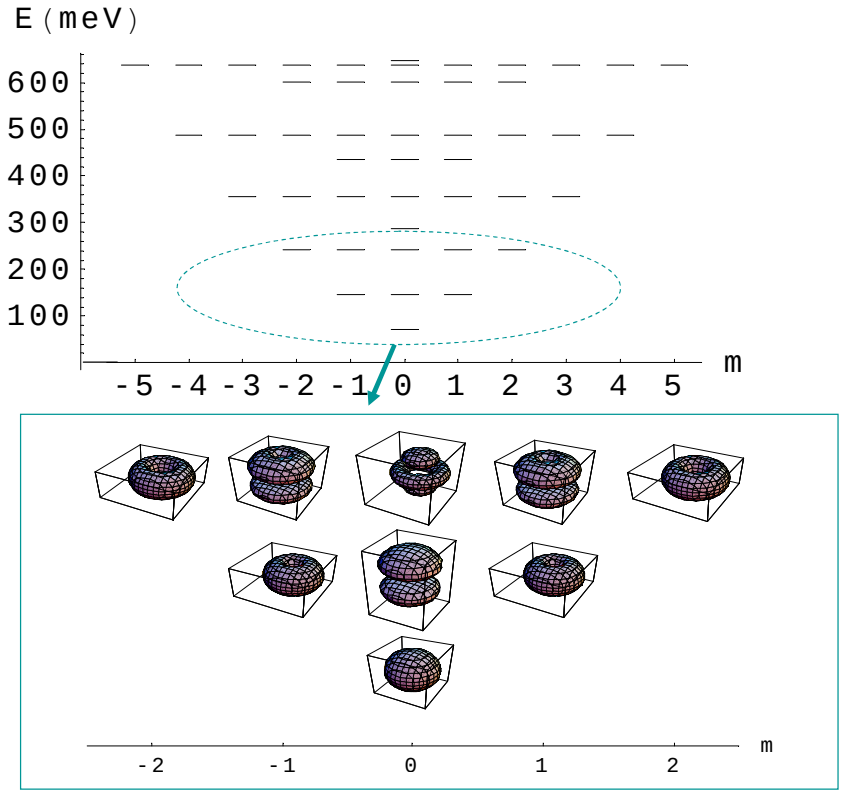

Fig.4 The single-particle energy spectrum versus the magnetic quantum number m(upper). The wave function density of the states on the three lowest shells (lower).

Coulomb interaction matrix elements

Based on the simplehard-wall spherical model, the explicit derivation of the Coulomb interaction matrix elements is possible. Although that has been done and published in several literature, [6] in which the expression is usually ambiguous and not straightforward to use. We spent few months on deriving the analytical formulation for the Coulomb interaction matrix elements whenever possible and partly carrying out the calculation numerically. We build up a big table of the values of Coulomb interaction matrix elements, serving for other computation of interacting spherical quantum dots.

Model 2: 3D parabolic model

Many physical bounded systems can be approximated well by the harmonic oscillator, such as ion traps [7], hydrogen atoms in van der Waals potential [8], and semiconductor nanostructures[9]. The Fock-Darwin model has provided an essential and simple analytic concept for the analysis of 2D harmonic oscillator systems. To our knowledge, however, in 3D systems, there are still only asymptotic approximated and numerical solutions for some specific parameters (the anisotropy of confining strength and external fields). The task of our work is to give an analytic solution of the wave functions for charged particles in a 3D parabolic potential (not necessarily isotropic) with uniform magnetic field.

First we explore the single particle behavior of a charged oscillator in a magnetic field. In Fig.5, we show a few lowest-lying energies as a function of the magnetic field. At zero magnetic field we have $\omega_+ = \omega_- = \omega_0$ and

$$E_{B=0} = (\frac{3}{2} + n + m + s)\hbar\omega_0,$$

It means that the energy of all eigenstates with the same value of $n+m+s$ is the same. These are the degenerate shells; let us name some of them here. The lowest one, called the s shell, consists of only one state $n = m = 0$. (We neglect the effect due to spin here.) The second one, called the p shell, consists of states $(n, m, s) = (1,0,0), (0,1,0)$ and $(0,0,1)$. Subsequent states can be generated in analogous manner.

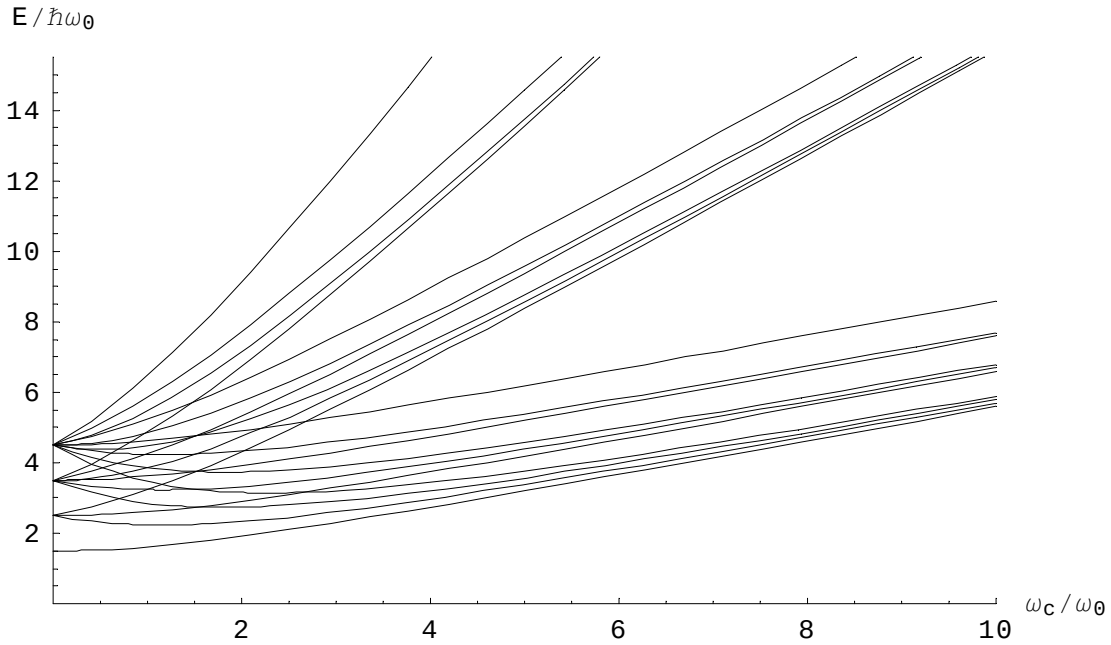

Fig.5 Energy spectrum of a single particle in a three-dimensional parabolic potential versus magnetic field.

Results and discussions（結果討論與計畫成果自評）:

In our theoretical investigation, we reveal the importance of the effects of particle-particle interaction and the sp-d coupling between carries and Mn2+ magnetic impurity . We study the spherical quantum dot with electron number Ne=1,2..8 and with a single Mn2+ magnetic impurity. Within the effective mass approximation, the energy spectrum, magnetization, and magnetic susceptibility x of the semi-magnetic QDs are investigated by using the configuration interaction method. In the absence of impurity, the quantum confinement of the small QD with radius R=5nm gives rise to paramagnetism (x>0) at low field. Without particle-particle

interaction, the dots with partially filled shell are expected to have strong paramagnetism. However, due to particle-particle interaction, the ground state of the dot with Ne=5 undergoes a spin state transition (S=3/2 to S=1/2) with magnetic field and a significant suppression of paramagnetism results. A single magnetic ion in dot can significantly affects the feature of the paramagnetism of the semi-magnetic QD as a function of Ne via the sp-d coupling between the electron carriers and the magnetic ion. The coupling leads to suppression or enhancement of paramagnetism, depending on electron number and the position of the ion site. We have clarified the underlying physical mechanism and the role of the sp-d coupling, which is particularly of importance in the understandings of carrier-mediated ferromagnetism in diluted magnetic semiconductors.

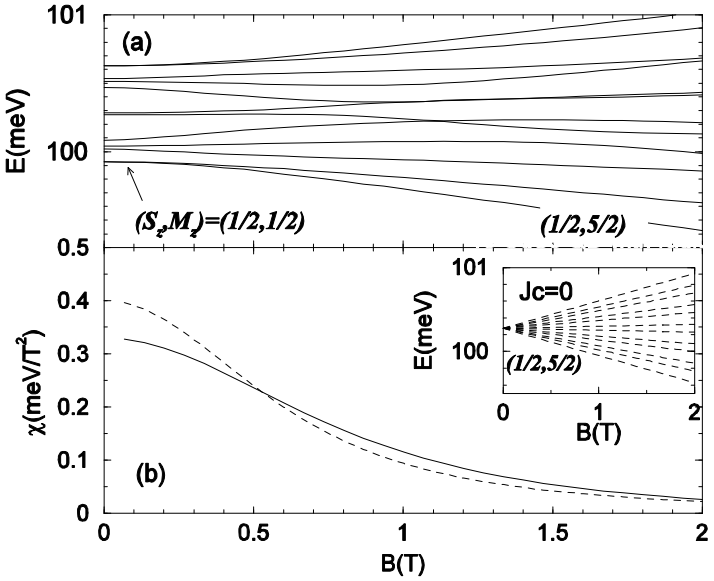

Fig.6 (a)The energy spectrum of a single electron in the dot with a single Mn2+ ion located at the dot center versus B. (b) the corresponding magnetic susceptibility (paramagnetism) versus B.

Our theoretical results successfully account for the low-field paramagnetism observed in the recent measurement of magnetization on colloidal CdSe:Mn and PbSe nanocrystal ensembles recently carried out by Wen-Bin Jiang *et. al.*. Furthermore, we explore the ground state properties of a Mn-doped NC and reveal the importance of the *sp-d* coupling between quantum-confined carries and Mn^{2+} magnetic impurity in magnetic response. It is known that the *sp-d* coupling triggers the ferromagnetism in III-VI GaMnAs semiconductors and is a crucial issue to study in condensed matters. We found that the *sp-d* coupling interaction significantly affects the low-field paramagnetism of NCs, depending on electron number and the location of the ion. The competition between electron-electron interaction and the *sp-d* coupling leads to the pronounced anisotropy of magnetic properties, ground state transitions in magnetic field, and the violation of the Hund's second rule. Our studies of the subject is particularly timely since the fabrication and spectroscopy of semi-magnetic quantum dots have been realized very recently. The developed theory is valuable since it is beyond the widely used mean field approximation, that is suitable for large scaled systems but not proper for quantum dots.

We have published two journal articles based on the work in Phys. Rev. B (2005) and Physica E(2006), respectively. Here we show a paragraph in the report of one of the PRB referees for this article" ... *These results are certainly interesting and timely, because most of the previous*

theoretical works dealing with diluted magnetic semiconductor quantum dots use mean field approximations to account for a high concentrations of magnetic impurities in the dots, but recent experimental developments allow for single ion doping..."

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Phys. Rev. B. **73**, 035326 (SCI)

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研討會論文:

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2. Shun-Jen Cheng, Weidong Sheng, and Pawel Hawrylak.

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第 27 屆國際半導體物理會議 (icps-27)心得報告

一、會議名稱：the 27th international conference on physics of semiconductor (icps-27)

二、會議日期：7/25-7/30, 2004.

三、會議地點：Flagstaff, AZ.US

四、報告者：國立交通大學電子物理學系鄭舜仁助理教授

五、方式 ： 壁報(Poster)

六、題目：Theory of excitonic filling factor nu=2 quantum Hall droplet in self-assembled quantum dots.

七、過程與討論：

七月二十五日由加拿大 Ottawa 飛抵 Phoenix，再搭乘 Shuttle Bus 順利抵達 Flagstaff，旅館 check in 後已是深夜，隔日在 Northern Arizona University 大會堂註冊。第一天的大會演講，大會邀請世界重量級半導體大師 Prof. M. Dresselhaus, Prof. K. von Klitzing, Dr. L.N.Pfeiffer 等人演講最新半導體物理發展。

再接下來四天之活動中，令我印象深刻的是，單量子點光譜技術(Single-dot spectroscopy)之快速發展，這一、二年來不僅越來越多之研究團隊已發展出成熟之單量子點光譜技術，更可利用單量子點發展先進之量子計算元件。在半導體奈米結構理論方面，Microscopic scale 之計算技術(pseudo-potential, tight-binding methods) 日益成熟，使今天我們對半導體奈米結構得以了解的更爲透徹。

我的壁報展覽被排在七月二十九日下午，在展覽會場中，Prof. J.L.Merz, Dr.A.M.Mintairov (University of Notre Dame) 對我的理論研究非常感興趣，因他們最近剛觀測到 InP 單量子點之磁光譜，因該量子點摻雜大量雜質，該磁光譜隨磁場有特殊之變化關係，會後我們互留名片，希望可以保

持未來之合作關係。第三天巧遇過去在法國 Grenoble,Max-Plack Institute 之朋友 Prof. M. Potemski，他提及他們最近在法國 Grenoble 強磁場中心進行的單量子點在超強磁場(>30Tesla)下光譜之研究，我們進行約半個鐘頭之深入討論，該討論激發我許多未來理論研究之靈感。

八、心得與感想：

(1) 分別與從事實驗與理論工作的學者進行了深入的討論，除觸發了許多研究上的想法，也建立了初步合作的關係。

(2) 感覺上目前在凝態物理的研究上，Quantum dots、Quantum optics 以及量子計算是比較熱門的領域。

八、此次報告之內容(見附件)：

國際合作計畫赴國外研究心得報告

計畫編號	94-2112-M-009-011-
計畫名稱	自聚型半導體量子點中多激子問題的研究(3/3)
出國人員姓名 服務機關及職稱	**鄭舜仁 助理教授** 國立交通大學電子物理學系(所)
出國時間及地點	July 11, 2006 – Sep.10 2006, Ottawa, Canada
合作研究機構	Institute for Microstructural Sciences, National Research Council.
合作計畫名稱	自聚型半導體量子點中多激子問題的研究(3/3)
合作計畫主持人	Prof. Pawel Hawrylak
出國事由	執行國際合作計劃

內容及成果：(見附件)

Report for the joint project between EP/NCTU and IMS/NRC in 2006: Spin-Characteristic Photoluminescence from Semi-magnetic Quantum Dots

Shun-Jen Cheng

Joint project of National Science Coucil of Taiwan with National Research Council of Canada and Department of Electrophysics, National Chiao Tung University, Hsinchu 30050, Taiwan, R. O. C.

(Dated: November 15, 2006)

Abstract

In this report, we present the research results of the joint project between my research group at Department of Electrophysics, National Chiao Tung University, Hsinchu, and Quantum Theory group at Institute for Microstructural Sciences (IMS) of National Research Council, Ottawa. The project is partly carried out during the period of my visiting of IMS in this summer (July.2006-Sep.2006) under the support of NSC project No.NSC94-2112-M-009-011. In the studies, we reveal the characteristic emission spectra from a photoexcted semi-magnetic quantum dot containing one or two excitons interacting with two Mn^{2+} ions, calculated by using configuration interaction method. A journal paper based on the work is being in preparation.

PACS numbers:

I. INTRODUCTION

In this work, we consider a 2D quantum dot conataining two Mn^{2+} impurities under photoexcitation and study the characteistic emission spectra from 1X-2Mn, and 2X-2Mn complexes in the semi-magnetic dots. Before performing the calculation of emission spectra, we first calculate the energy spectra and the eigen states of 0X-2Mn, 1X-2Mn, and 2X-2Mn complexes in dots.

In the calculations, we take the following parameters for II-VI SC QDs throughout this work: $m_e = 0.106m_0$, $m_h = 0.424m_0$, $\epsilon = 10.6$, $\Delta^0 = 1$meV, $J_{eM}^{(0)} = 15\text{meV}\cdot\text{nm}^3$, $J_{hM}^{(0)} = 60\text{meV}\cdot\text{nm}^3$, $\omega_e = 4Ry^*$, $\omega_h = Ry^*$, $l_e = l_h = l_0 = \sqrt{Ry^*/\hbar\omega_e}\cdot a_B^* = 2.65$nm , where $Ry^* = 12.8$meV, $a_B^* = 5.29$nm are the effective Rydberg and the effective Bohr radius for electron, respectively. The parameters of e-h exchange interaction: The energy splitting between the dark and bright X's: $\delta_0 = \frac{3}{2}(\Delta^0 + \frac{9}{4}\Delta_z^1) = 1.0$meV. The energy splitting between the bright X's, arisen from deformation: $\delta_1 = \frac{3}{4}(\Delta_x^1 - \Delta_y^1) = 0$ (symmetric dot considered). The energy splitting between the dark X's: $\delta_2 = \frac{3}{4}(\Delta_x^1 + \Delta_y^1) = 0.1$meV.

II. RELEVANT STUDIES IN LITERATURE

To date, we found the following papers relevant to the subject.

Experimental works:

1. the papers by the group of L. Besombes, Y. Leger, H.Mariette. Since 2000, the group demonstrate a series of experimental works of the emission spetra from II-VI self-assembled dots containing *single Mn* in magnetic fields. The characteristic spectrum of a single Mn interacting with single X, charged X, and bi-X are clearly identified, in comparison with the calculation based on simple effective spin Hamiltonian consisting of e-h exchnage, e-Mn, h-Mn spin-spin interactions. The features of the emission spectra differ from dot to dot, sensitively depending on the geometry of dot and the position of Mn. Only in the symmetric cases, six main lines with nearly equal energy spacing are observed in a spectrum. In asymmetric cases, emssion lines fall into two groups, whose separation created by the ansiotropic part of e-h exchange interaction associated with the dot deformation. In the newly published PRL paper in 2006 [PRL 97,107401(2006)], they demonstarte the electrical control of the carrier number in dot and the measured characteristic emission spectrum of

X, X^+, X^-, and XX, interacting with a single Mn, in a dot. Theory based on simple effective spin Hamiltonian well interpret the experimental results.

2. the pre-print by P.Wojnar, entitled "magneic QDs containing only few Mn ions". In the work, micro-PL spectroscopy is carried out on individual II-V CdMnTe/ZnCdTe QDs in magnetic fields ranging from $B = -6$T to 6T. They observed *the giant Zeeman effect* in the PL spectra, i.e. the significant red shift of PL peaks with respect to increasing magnetic field. They proposed a model Hamiltonian to simulate the evolution of energyies, degree of circular polarization, and line width of the PL peaks with magnetic field. In the model, the ensemble of Mn ions is modeled as Mn cluster with a total effective spin $\vec{S}$ (it is however not very clear yet to me how they determine the spin value S). The interaction between carriers (electron and hole) and the model Mn cluster follows the Heisenberg model Hamiltonian. The anti-ferromagnetc interaction between Mn's is neglected here. The emission spectra are calculated by using the formulation of the Fermi's golden rule. Comparing the simulation with the experimental results, it was estimated that a large (small) dot in the dot ensemble contains the number of Mn $N_{Mn} \sim 25$ ($N_{Mn} \sim 5$).

Theoretical works:

1. papers by A. O. Govorov et al (e.g. PRB, 70, 035321 (2004), "Optical probong of the spin state of a single magnetic impurity in a self-assembled quantum dot".) In the first part of the paper, the authors calculate the energy spectrum of a 1X-1Mn asymmetric dot for different location of Mn by diagonalizing the spin Hamiltonian (both isotropic and anisotropic parts of e-h exchange interaction are considered). Then they solve the master equation to study the dynamic processes (pumping, emission, and relaxation) of 1Mn dot subject to a short pumping pulse. They showed that, with the resonant pumping technique, one can manipulate selectively individual spin of a Mn impurity, and a Mn impurity can act as qubit.

2. PRB, 73, 045301 (2006), J. Fernandez-Rossier, "Single-exciton spectroscopy of semi-magnetic quantum dots". The paper presents the theoretical studies of the emission spectra from a single X in dot, interacting with few Mn's($N_{Mn} = 1, 2, 3, 4$). The authors took the hard-wall quantum box model for dot confining potental. For valence hole states, the (slight) HH-LH intermixing is considered.Although the X-Mn Hamiltonian is partly expressed in second quantization, the single X itself (to my understanding) is treated in single-particle picture (no higher shell scatterings due to e-e, h-h, e-h, e-Mn, and h-Mn interactions are

considered). In the e-h exchange interaction, only the isotropic part of the e-h interaction is taken into account. The e-Mn and h-Mn interactions are modeled by spin-spin Heisenberg Hamiltonian. The interaction between Mn's is neglected because the sufficiently long distances between Mn's are assumed for diluted magnetic dots (small number of Mn in each dot). Emission spectra are calculated using the Fermi's golden rule. In the case of X-2Mn dots, the emission spectra versus the adjustable parameter $r \equiv |\psi_0(r_2)|^2/|\psi_0(r_1)| = 0.1 \sim 1$ are discussed, where $r_1(r_2)$ is the position of the Mn#1 (Mn#2). In symmetry case $r = 1$, 11 lines are observed in the calculated spectrum.

III. ENERGY SPECTRA

A. Two Mn's QDs

We start with a carrier-free dot with two Mn^{2+} impurities located at postions $\vec{R_1}$ and $\vec{R_2}$ in a dot, respectively. The interactions between Mn's and between Mn and carriers in quantum dots are modeled by the spin-spin exchange Hamiltonian (For II-VI semi-magnetic semiconductors (SCs), Mn^{2+} impurities substitute the divalent cations of SC and the induced electrostatic potential is negigible.) The Hamiltonian of a two-Mn dot system is thus written as

$$H_{MM} = +|J_{MM}(R_{12})|\vec{M}_1 \cdot \vec{M}_2\,, \tag{1}$$

where $\vec{M}_i$ denote the spin of the ith Mn, $R_{12} \equiv |\vec{R_2} - \vec{R_1}|$ is the distance between the two Mn^{2+} ions, and $J_{MM}(R_{12}) = J^{(0)}_{MM}\exp\{-\lambda[R_{12}/a_0 - 1]\}$ is the coupling constant of the anti-ferromagnetic Mn-Mn interaction, where a_0 is the lattice constant and $J^{(0)}_{MM}$ is a nearest-neighbor Mn-Mn interaction.[F.Qu PRL2006] The positive sign before the coupling constant in Eq.(1) indicates the anti-ferromangetic nature of the Mn-Mn interaction.

One can derive the energy spectrum of the 2Mn system with the Hamiltonian Eq.(1) analytically, which is given by

$$E_{M,M_z} = \frac{|J_{MM}(R_{12})|}{2}[M(M+1) - \frac{35}{2}]\,, \tag{2}$$

where $M = 0, 1, ..5$ is the total spin of the two-Mn system ($\vec{M} \equiv \vec{M}_1 + \vec{M}_2$) and M_z is its z-component. Accordingly, the ground state (GS) is that one with zero total spin of Mn's $M = 0$, an anti-ferromagnetic (AF) state. Fig. 1 shows the calculated energy spectra of

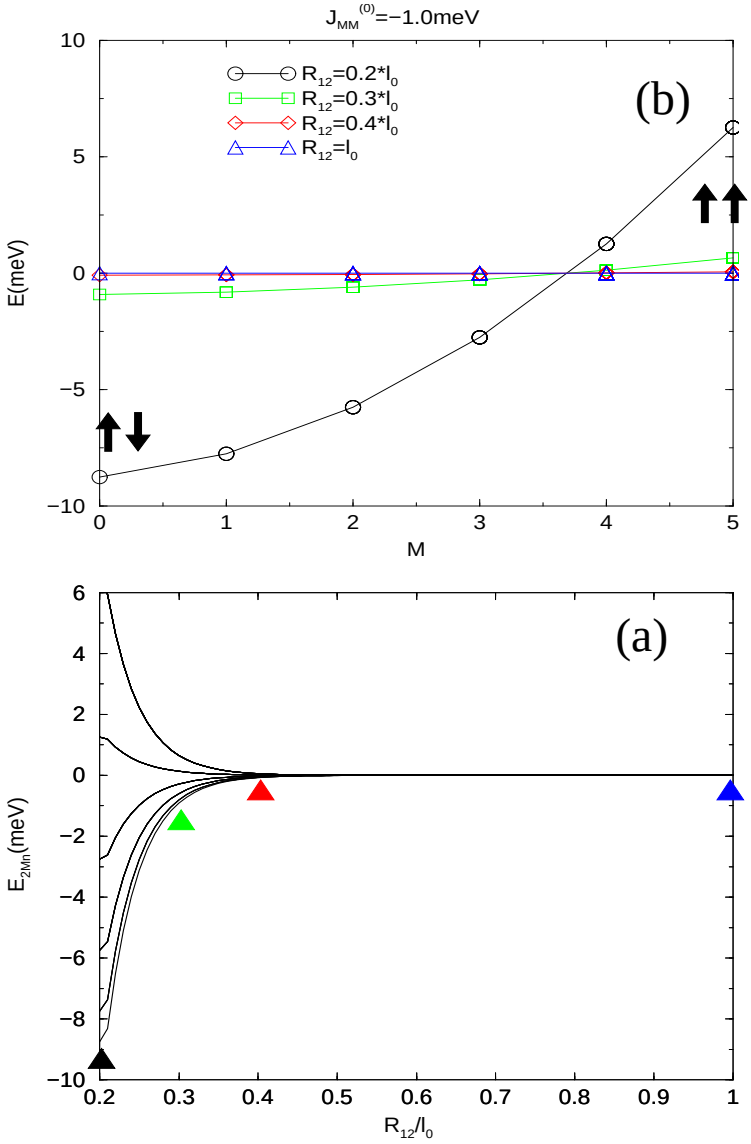

FIG. 1: The energy spectra of a quantum dot containing two Mn ions (a) vs the distance between Mn's R_{12} (b) vs the total spin of the Mn's for four different values ofR_{12} [marked with colored triangles in (a)]. See text for dot parameters.

the two-Mn systems with different R_{12}. With increasing R_{12}, the energy difference between states with spin M and $(M+1)$ decreases (E.g., the energy difference between the GS $(M=0)$ and the first excited state $(M=1)$ is given by $|J_{MM}(R_{12})|$, which decreases with increasing with R_{12}). For long $R_{12} \gtrsim 0.4l_0$, all of the 2Mn states becomes nearly degenerate.

B. Single exciton interacting with two Mn's

Under photoexcitation, excitons are created in the semi-magnetic QD and interact with Mn's via the *sp-d* spin-spin exchange interaction. Experimetally, the number of exciton can be controlled by adjusting the power of excitation. Here, we consider a semi-magnetic 2D quantum dot with a single exciton and two Mn^{2+} ions. The Hmiltonian is expressed as

$$H_{1X2Mn} = H_X^0 + H_{eM} + H_{hM} + H_{MM} \ , \tag{3}$$

where $H_X^0 = T_e + T_h - V_c(\vec{r_e} - \vec{r_h}) + H_{eh}^{ex}$ is the total energy of a single exciton in a *non-magnetic* (Mn-free) dot, the e-h exchange interaction is given by

$$H_{eh}^{ex} = -|\Delta^0|\vec{s}\cdot\vec{j} - \sum_{i=x,y,z} |\Delta_i^1| j_i^3 s_i \, , \tag{4}$$

and the e-Mn and h-Mn interaction are given by

$$H_{eM} = -|J_{eM}^{2D}|\vec{s}\cdot\vec{M}_1\delta(\vec{r_e} - \vec{R_1}) - |J_{eM}^{2D}|\vec{s}\cdot\vec{M}_2\delta(\vec{r_e} - \vec{R_2}) \, , \tag{5}$$

and

$$H_{hM} = +|J_{hM}^{2D}|\vec{j}\cdot\vec{M}_1\delta(\vec{r_h} - \vec{R_1}) + |J_{hM}^{2D}|\vec{j}\cdot\vec{M}_2\delta(\vec{r_h} - \vec{R_2}) \, , \tag{6}$$

respectively, where the coupling constant are given by $J_{eM/hM}^{2D} \equiv J_{eM/hM}^{(0)} 2/d$ with d the thickness of quantum dot. To seek for the eigen solutions of the 1X-2Mn system, we take the four 1X-configuration $|s_z = \pm 1/2; j_z = \pm 3/2\rangle$, combined with 36 possible 2Mn configurations $|M_1^z; M_2^z\rangle$, as basis and diagonalize the corresponding Hamiltonian matrix. Here we consider the pure heavy-hole states in quantum dots and hole spin has only two possible values $j_z = \pm 3/2$. We also assume that the electrons and holes are frozen in their lowest orbital (s-orbit) state of quantum dot (The assuption will be released as we consider the RKKY effect in bi-exciton states of Mn-doped quantum dots). In the approximation, we can omit the constant kinetic energies$T_{e/h}$ and direct Coulomb interaction V_c terms for brevity. In the basis, the matrix elements of the Hamiltonian are given by

$$\begin{aligned}
&\langle \ M_1^{z\prime}; M_2^{z\prime}|\langle s_z{}'; j_z{}'|H_{1X2Mn}|s_z; j_z\rangle|M_1^z; M_2^z\rangle \\
&= \langle M_1^{z\prime}; M_2^{z\prime}|\langle s_z{}'; j_z{}'|H_{eh}^{ex} - |J_{eM}(1)|\vec{s}\cdot\vec{M}_1 - |J_{eM}(2)|\vec{s}\cdot\vec{M}_2 \\
&\quad + |J_{hM}(1)|j_z M_1^z + |J_{hM}(2)|j_z M_2^z + |J_{MM}(R_{12})|\vec{M}_1\cdot\vec{M}_2|s_z; j_z\rangle|M_1^z; M_2^z\rangle \, ,
\end{aligned}$$

where $J_{eM/hM}(I) = J^{2D}_{eM/hM}|\psi_0(\vec{R}_I)|^2$ $(I = 1, 2)$ is the coupling constant for electron/hole interacting with the Ith Mn. In the model of parabolic confinement potential, the wave function ψ_0 is givn by $\psi_{m=0,n=0}(x, y) = \psi_0(x)\psi_0(y)$, where $\psi_0(\widetilde{x}) = e^{-\widetilde{x^2}/4}/(2\pi l_0^2)^{1/4}$ with $\widetilde{x} = x/l_0$ and $l_0 = \sqrt{Ry^*/\hbar\omega_0}a_B^*$, where ω_0 is the confinement frequency of parabolic potential.

1. Ground states (GS's)

The 1X-2Mn Hamiltonian contains different types of carrier-carrier interactions: the ferromagnetic (FM) e-Mn interaction, the anti-ferromagnetic (AF) h-Mn interaction, and the AF Mn-Mn interaction. For bright X with opposite spins of hole and electron, a FM state of 2Mn is favored, but in the competition with the AF interactions between Mn's. The spin properties of 1X-2Mn GS's depend on the relative strengths of e-Mn, h-Mn, Mn-Mn, and e-h exchange interactions, determined by the positions of Mn's.

Let us consider QDs containing the two Mn's: one (Mn#1) fixed at the position $\vec{R}_1 = l_0(1, 0)$ and the other one's (Mn#2) position movable on the x-y plane. Fig. 2(a) shows the calculated GS energy of the 1X-2Mn dot as a function of the second Mn's position $\vec{R}_2$. Fig. 3(a) shows the mean value of total Mn spin $\langle M^2\rangle_{GS} \equiv \langle 1X2Mn; GS\ |M^2|1X2Mn; GS\rangle$ of the 1X-2Mn GS's of the same dot as a function of $\vec{R}_2$.

In Fig. 2(a), we see that the GS energy has a dip as the Mn#2 is placed in the region around $\vec{R}_1$ with radius $R_{12} \lesssim 0.3l_0$. In Fig. 3(a), we see the corresponding $\langle M^2\rangle_{GS} \sim 0$ i.e. vanishes, indicating the AF GS of the 2Mn's. The two Mn's are in the AF GS, regardless of the existence of spin exciton, because of the strong Mn-Mn interaction in the short range. As Mn#2 is placed far from Mn#1 ($R_{12} \gtrsim 0.3l_0$), the 2Mn GS will be in the FM phase ($\langle M^2\rangle_{GS} \sim 30$) because of strong X-Mn FM interaction and weak Mn-Mn AF interaction. The absolute value of the GS energy $|E^{1X2Mn}_{GS}| \sim E^{1X2Mn}_b$ is approximately equal to the binding energy of 1X-2Mn complex, i.e. exciton magnetic polaron.

2. Excited states (ES's)

Assuming $J_{eM/hM}(1) = J_{eM/hM}(2) = \bar{J}_{eM/hM}$, the 1X-2Mn Hamitonian is written as

$$H_{1X2Mn} = -|\Delta^0|s_z \cdot j_z - \sum_{i=x,y,z} |\Delta^1_i|j^3_i s_i - |\bar{J}_{eM}|\vec{s}\cdot\vec{M} + |\bar{J}_{hM}|j_z M_z + |J_{MM}|\vec{M}_1\cdot\vec{M}_2\,. \quad (7)$$

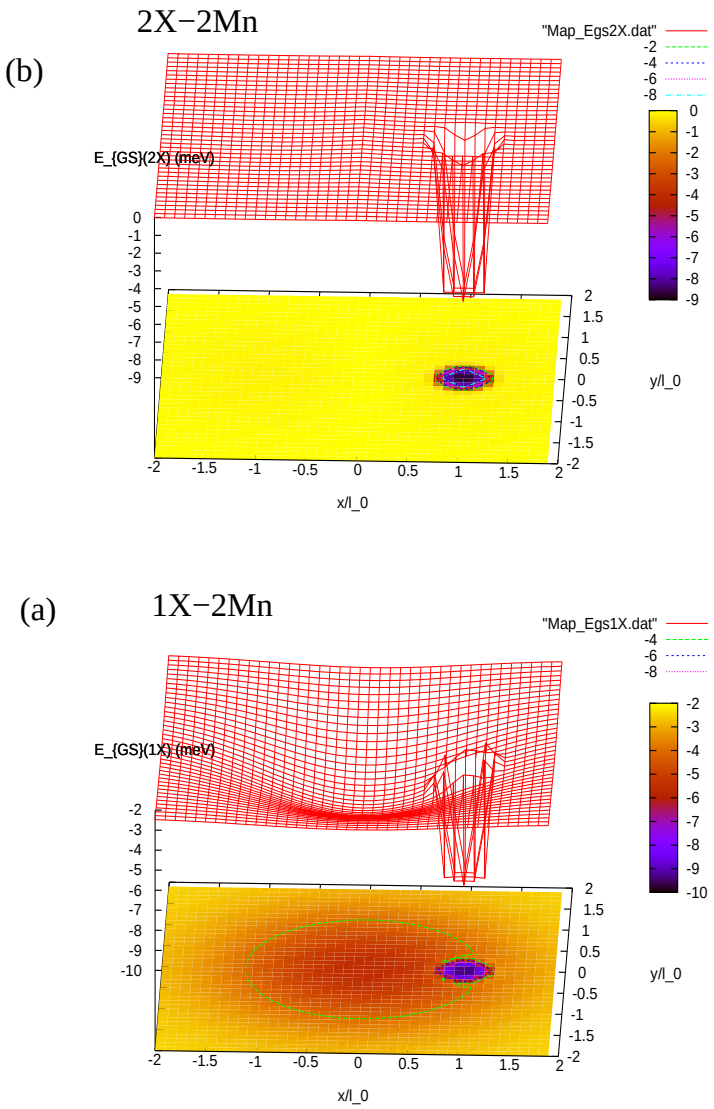

FIG. 2: Ground state energy spectra of photoexcied quantum dots with two Mn ions. Here we consider the case that one of the two Mn's (Mn#1)is located at the fixed position $\vec{R_1} = (l_0, 0)$ while the other one (Mn#2) can be moved to any position $\vec{R_2}$ on the x-y plane. The GS energy vs $\vec{R_2}$ of (a) a single exciton (b) a bi-exciton in the Mn-doped quantum dots.

The typical vaues of the parameters for II-VI semimagnetic semconductors: $|\Delta^0| \sim |\bar{J}_{hM}| > |\bar{J}_{eM}| \sim |\Delta_i^1|$.

Let us consider two special cases:

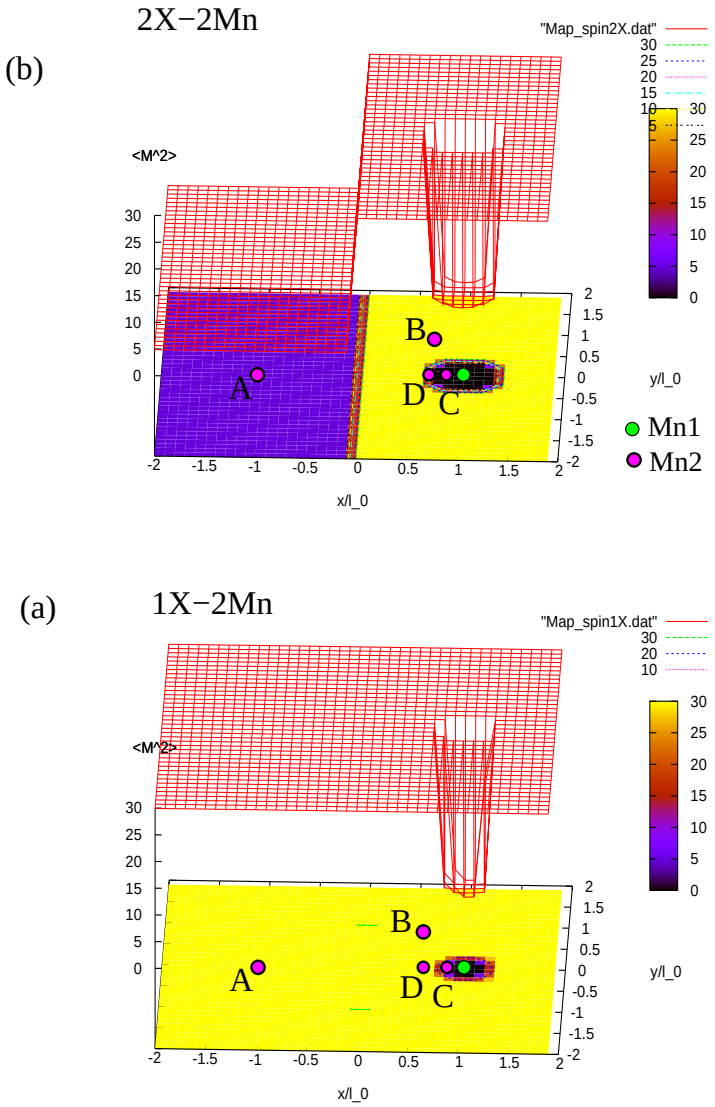

FIG. 3: The average total spin ($\langle GS|\hat{M}^2|GS\rangle$) of the GS's vs $\vec{R}_2$ of (a) a single exciton (b) a bi-exciton in the same Mn-doped quantum dots as in Fig.2. The charcteristic emission patterns of four cases for different $\vec{R}_2$, labled by capital letter A,B, C, and D, are studied in this work.

1. $J_{MM} \to 0$: The Hamiltonian is approximated to

$$H_{1X2Mn} \approx -|\Delta^0|s_z \cdot j_z + |\bar{J}_{hM}|j_z M_z - |\bar{J}_{eM}|\vec{s}\cdot\vec{M} + ... \tag{8}$$

Notable is that, due to the e-h exchange interaction, the 1X GS is supposed to be the dark X state. The carrier-Mn interaction affect the GS properties because the 2Mn in the GS's is FM ($M_z = 5$). Due to the e-Mn interaction, the dark X GS's are (slightly) intermixed with

the bright X states, and becomes optically active. Moreover, the e-Mn interaction reduce the energy separation (~ 0.15meV) between the dark GS's and the lowest bright X excited states.

2. $d \sim a_0$ and J_{MM} large:

$$H_{1X2Mn} \approx -|\Delta^0| s_z \cdot j_z + |J_{MM}| \vec{M}_1 \cdot \vec{M}_2 + |\bar{J}_{hM}| j_z M_z - |\bar{J}_{eM}| \vec{s} \cdot \vec{M} + ..., \qquad (9)$$

Fig. **??**(b) gives a schematic illustration of the energy spectrum of a 1X-2Mn dot under the condition. Because the 2Mn GS's are the AF states ($M = 0$), the carrier-Mn interaction vanishes for the GS's and the dark GS's is not intermixed with the bright excited states. The GS's remain optically inactive as those of the Mn-free dots. The energy separation between the dark GS's and the lowest bright X excited states remains the same (= 1meV) as that for Mn-free dots.