# 行政院國家科學委員會專題研究計畫　期中進度報告

## 自聚型半導體量子點中多激子問題的研究(2/3)

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# 行政院國家科學委員會補助專題研究計畫期中進度報告

計畫名稱:

## 自組式半導體量子點的電子結構及其多激子理論之研究

計畫類別：√ 個別型計畫　　□ 整合型計畫

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計畫主持人：鄭舜仁

計畫參與人員： 研究助理 容震軒, 碩一 陳彥廷

執行單位：國立交通大學電子物理系

中　華　民　國 九十四 年 五 月 二十七 日

# 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

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

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 investigatie 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.

報告內容

**Subject:** Theoretical investigation of multi-exciton complexes in semiconductor self-assembled quantum dots (2/3)

**Contract No.:** NSC93-2112-M-009-021-

**Investigators:** 鄭舜仁(NCTU/EP), 容震軒(NCTU/EP),陳彥廷(NCTU/EP)

**Collaborators**: Pawel Hawrylak (Natl Research Council of Canada), Weidong Sheng (NRC). 林浩雄(Natl Taiwan Univ/dept of Electrical Engineering) 吳建明(NTU/EE)

**Introduction:**

First, we sincerely acknowledge NSC for supporting this project. In the past year, we have successfully built up an advanced computation environment on the Intel Itanium 64-bit PC-workstation that was purchased using the grant for this project from NSC. The computation facility incorporated with advanced developed numerical techniques (conjugated gradient method and Lanczos eigen solver) are of vital importance in the computation of many-body physics in nanostructures—the main subject of this project. Based on those techniques, we have finished or are preceeding the following investigations: (1) the excitonic quantum Hall droplet in self-assembled quantum dots, (2) the charged exciton-complexes in quantum dots, (3) the electronic and magnetic properties of semi-magnetic quantum dots (Mn^2+-doped colloidal nanocrystals), and (4) multi-band theory for semiconductor nanostructures.

In the following, we shall give a brief introduction to each research topic, 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. Set-up of computation techniques and environment**

***Introduction***

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.

- 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 (see Fig.1).

- 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.

**2. Studies of correlated multi-excton complexes in self-assembled quantum dots**

***Introduction***

Number of interesting and fascinating physical phenomena in the quantum dots in the quantum Hall regime have been revealed and extensively studied. However, most

studied are focused on the one-component electron quantum Hall droplet, a strongly interacting few-particle complex in QD in the quantum Hall regime. Recently, the measured excitonic spectrum of self-assembled quantum dot ensemble in high magnetic field up to 28T has been demonstrated. Thus, a direct probing multi-exciton complexes in a "single" dot in the quantum Hall regime, referred as to "excitonic quantum Hall droplet", is expected in very near future.

We have developed a theory of excitonic quantum Hall droplet (EXQHD) at filling factor nu=2 in a self-assembled quantum dot (SAD) subject to strong perpendicular magnetic field B (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 it's magnetic field evolution from the EXQHD is predicted. The preliminary result has appeared in the proceedings of "the 27$^{th}$ international conference on the physics of semiconductor", held in Flagstaff, USA, in July 2004. An extended version of the work on EXQHD has been submitted to journal Physical Review B.[2]

**3. Studies of charged exiton in quantum dot**

***Introduction***

It has been shown that charged exciiton can be electrically created in a self-assembled quantum dot embedded in p-n junction by utilizing the resonant tunneling technique, and probed using emission spectroscopy.[3] For instance, a charged exciton X^2- (3e+1h) is formed by electrically injecting an electron into the dot in which a charged exciton X^- (2e+1h) exists(see Fig.2). The electrically controllable charged exciton states can be utilized in the scheme of quantum computing implemented using the quantum optical technique. The dot embedded in p-n junction is actually subjected to a high electric field. The high field separates the e-h pairs and might create an internal electrical dipole. The internal dipole indicates the some kind of symmetry breaking and might lead to different nature of the few-exciton ground states. This stimulates our interest in studying the charged exciton of dot under strong external field (Stark effect).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 drak exciton states (see Fig.3)[4]. In the studies of the charged exciton, we have found the emission pattern is a sensitive function of

charge number. The stark effect (high electric field in the direction of crystal growth) also significantly affect the energy spectrum. A manuscript for a journal paper on this subject is in preparation.

## 4. Studies of the electronic and magnetic properties of semi-magnetic quantum dots (Mn2+-doped colloidal semiconductor nanocrystals)

### *Introduction*

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”.

Here, we apply the developed theoretical approaches, CI and ED, to the investigation of the electronic and magnetic properties of spherical quantum dot doped with a single Mn2+ ion. (see Fig4) The motivation of the studies is stimulated by the recent measurement of the magnetization and the magnetic susceptibility of colloidal PbSe:Mn and CdSe:Mn quantum dots by Prof. Wen-Bin Jiang at the department of electrophysics, NCTU.

### *The single-particle electronic structure of chemically synthesized colloidal nanocrystals*

We take the widely used hard-wall spherical potential to model the confinement of chemically synthesized colloidal nanocrystals. The typical dameter 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 Fig5.

### *Coulomb interaction matrix element*

Based on the simple 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 numercally. We build up a big table of the values of Coulomb interaction matrix elements, serving

for other computation of interacting spherical quantum dots.

***Results***

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. The calculation is compared with the recent measurement on colloidal PbSe QDs and accounts for the observed low-field paramagnetism. 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. An abstract on the work has been accepted by the $12^{th}$ International conference on the Modulated Semiconductor Structures. A manuscript for journal paper on the subject is in preparation.

**5. Development of multi-band theory of self-assembled quantum dots: toward ab-initio approaches.**

***Introduction***

In order to more precisely model self-assembled dots whose formation involve compex factors, including the ssues of strain, composition intermixing, complex nature of valence bands,etc, we shall develop a theory for the calculation of the single-particle electronic structure, to some degree, beyond the simplified effective mass approximation. We start with the k.p theory. In spite of the macroscopic spirit of the theory, it is shown that the k.p model still capture most physics for the dot with the size on the ten-nanometer scale.[7]

We shall develop a multiband microscopic theory of many-exciton complexes in self-assembled quantum dots. We shall start from the multiband k·p method and then study using the psuedopotential method to calculate the single particle states of quantum dot. The electronic structure calculations are coupled with strain calculations via Bir-Pikus Hamiltonian. The theory shall be applied to the excitonic recombination spectrum in self-assembled quantum dots and colloidal nanocrystals. The results of the single-band effective-mass approximation are compared with those obtained by using the of k·p and psuedopotential methods. This study is in the cooperation with the group of Prof. H.H. Lin (林浩雄) at NTU/EE. So far, we have developed a simple code for quantum well by using 4x4 k.p model and plane wave expansion method. In the coming year, we shall extend the technique to the calculation of quantum dot. Meanwhile, we plan to study smaller dot system ,e.g. nanocrystals, using the psuedopotentail approaches. We shall try to find out the boundary between the applicable regime of the macroscopic k.p theory and microscopic ab ignition method.

**Summary**

For calculation of the full spectrum of multi-exciton complexes in quantum dot, we have developed a eigen solver using conjugated gradient method, which has been successfully applied in the calculation for the spectrum of excitonic quantum Hall droplet. A numerical interface for another more powerful eigen solver Lanczos is in intensive developments.

We have developed a theory of excitonic quantum Hall droplet (EXQHD) at filling factor nu=2 in a self-assembled quantum dot. We determine the ground and excited states, the stability against spin flips, and the optical emission spectrum of the nu=2 EXQHDs. The preliminary result has appeared in the proceedings of "the 27$^{th}$ international conference on the physics of semiconductor", Flagstaff, in July 2004. A manuscript on the subject for journal Physical Review B is gong to be submitted out very soon.

In the studies of the charged exciton, we have found the emission pattern is a sensitive function of charge number. The stark effect significantly affects the energy spectrum.

We have studied the spherical quantum dot with electron number Ne=1,2..8 and with a single Mn2+ magnetic impurity. We have clarified the underlying physical mechanism and the role of the sp-d coupling between carrer and Mn2+-ion. An abstract has been accepted by "the 12$^{th}$ International conference on the Modulated

Semiconductor Structures", Albuguerger, July 2005. A manuscript for journal paper on the subject is intensively in preparation.

We have started to develop a multiband microscopic theory for self-assembled quantum dots. A simple code using 4X4 k.p model is finished for quantum well system.

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附圖

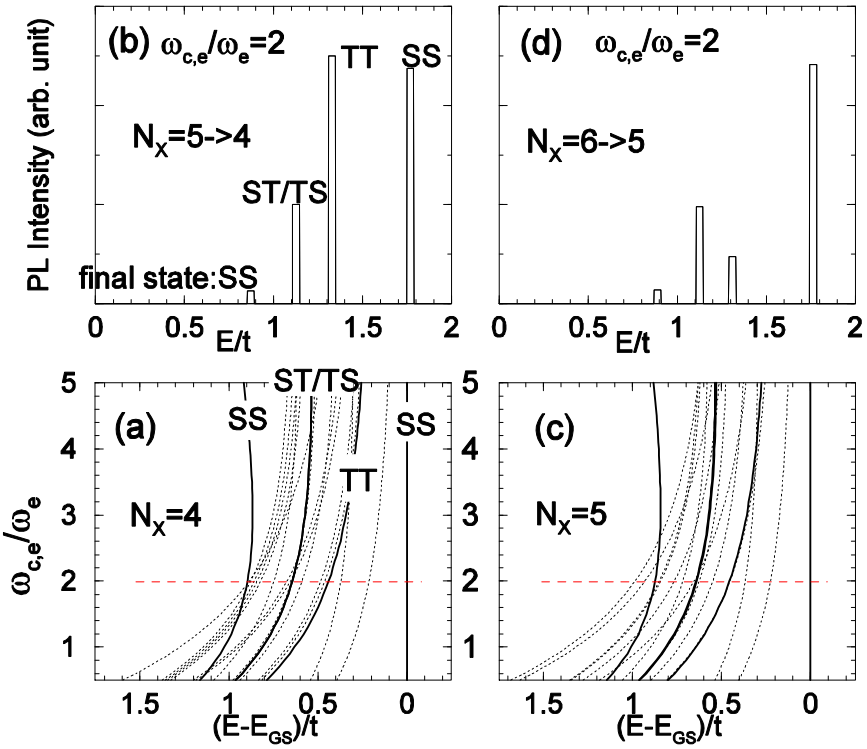



Fig1. The energy spectra of the ground and excited states of excitonic quantum Hall droplet versus magnetic field for exciton number (a) Nx=4 (c) Nx=5. The spectra at B~17Tesla (corresponding to omega_c/omega_e=2) are mapped out by the emission spectra from a (b) Nx=5 (d) Nx=6 droplet.[2]

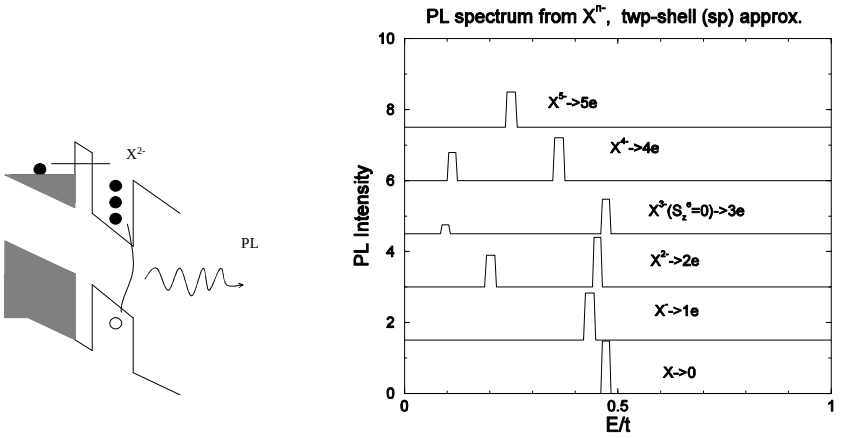



Fig.2 The schematic diagram of the charged exciton in self-assembled quantum dot created by using the resonant tunneling technique(left panel). The emission spectrum from the charged symmetry dot as function of charge number of the exciton (right panel).

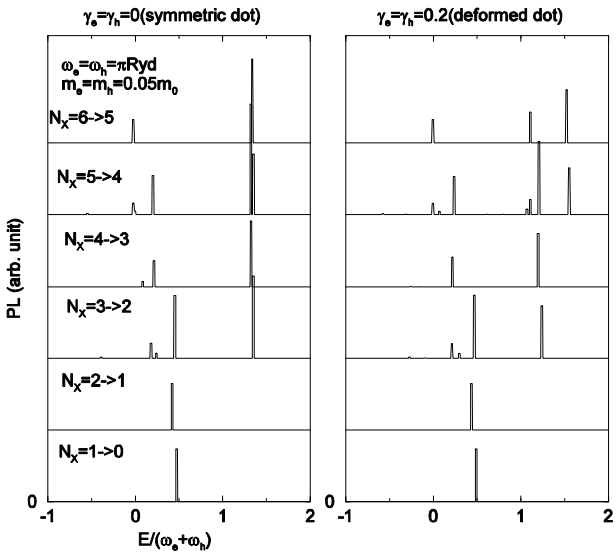
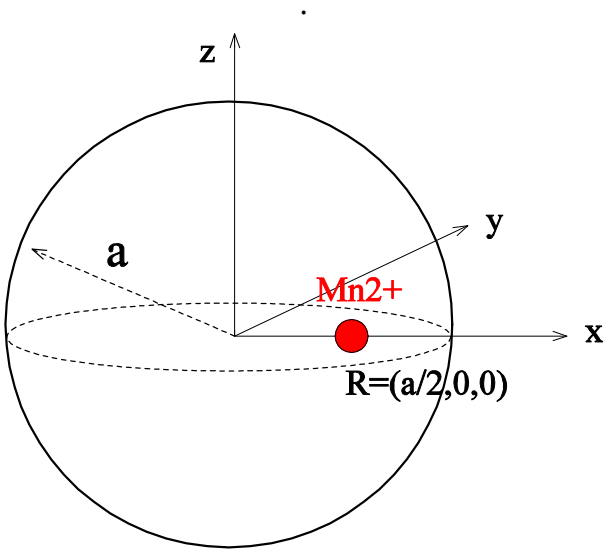



Fig.3 The emission spectrum of symmetry dot (left) and asymmetry dot (right) as function of exciton number (excitation power)




Fg.4 Schematic diagram of a spherical quantum dot with a single Mn2+ ion impurity.

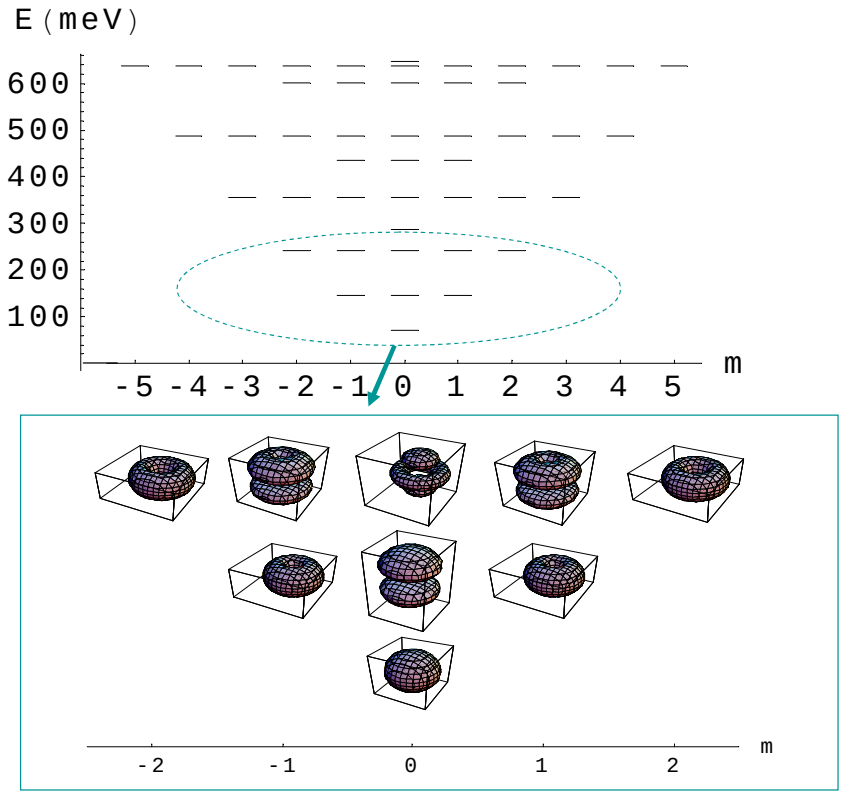



Fig.5 The single-partcle energy spectrum versus the quantum number m(upper). The corresponding wave function density of the states on the three lowest shells(lower).

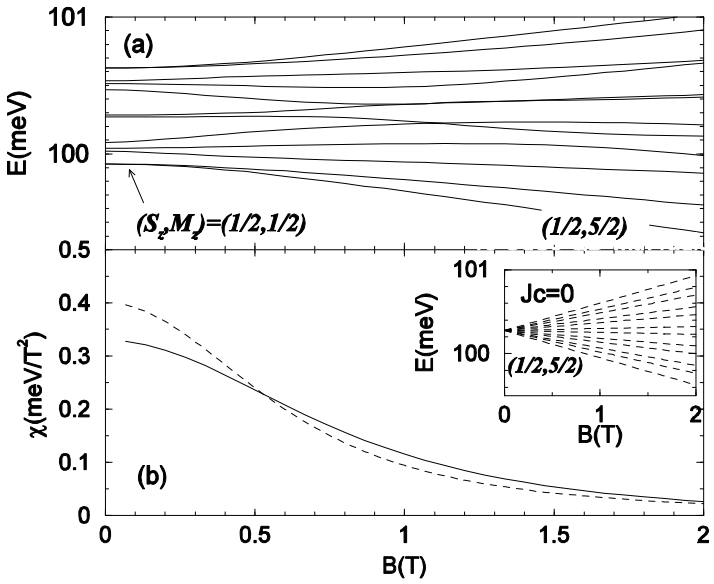



Fig.6 (a)The energy spectrum of a single electron in the dot with a single Mn2+ ion located at the dot center versus B. (b) the corresponding magnetic susceptibility (paramagnetism) versus B.

計畫成果自評部份，請就研究內容與原計畫相符程度、達成預期目標情況、研究成果之學術或應用價值、是否適合在學術期刊發表或申請專利、主要發現或其他有關價值等，作一綜合評估。

The subjects of this project mainly includes (1) development of the powerful eigen solver for large SPARS matrix (2) the excitonic quantum Hall droplet in self-assembled quantum dots, (3) the charged exciton-complexes in quantum dots, (4) the electronic and magnetic properties of semi-magnetic quantum dots (Mn^2+-doped colloidal nanocrystals), and (5) multi-band theory for semiconductor nanostructures. We have almost finished the reaserch work of (1) (2) and (3). One journal paper on (2) has been submitted to Phys. Rev. B. We are intensively working on another journal paper for (4). There remains some work for (3) and the work on (5) is just in the beginning. We shall try the best to work out (3) and (5) within the coming year.

Here, we summarize our investigation results and the schedule for the subjects:.
1. For calculation of the full spectrum of multi-exciton complexes in quantum dot, we have developed a eigen solver using conjugated gradient method, which has been successfully applied in the calculation for the spectrum of excitonic quantum Hall droplet. A numerical interface for another more powerful eigen solver Lanczos is in developments. That is expected to be finished within 1-2 months, and be capable of dealing with SPARS matrix with the size $>10^5 \times 10^5$.

2. We have developed a theory of excitonic quantum Hall droplet (EXQHD) at filling factor nu=2 in a self-assembled quantum dot. We determine the ground and excited states, the stability against spin flips, and the optical emission spectrum of the nu=2 EXQHDs, using the developed technique .of (1)
*publication*
A short-version paper on the subject has appeared in *the proceedings of "the 27$^{th}$ international conference on the physics of semiconductor"*, Flagstaff, in July 2004. An extended version paper on the subject has been submitted to journal Physical Review B. [2]

3. In the studies of the charged exciton, we have found the emission pattern is a sensitive function of charge number. The stark effect significantly affects the energy spectrum. The result is based on CI calculation. In the coming year, we shall carry out the k.p calculation for the electronic structure of the quantum dots. We shall make a comparison for the results obtained using the two approaches.

*publication*
A manuscript for a journal paper on this subject is in preparation.

4. We have studied the spherical quantum dot with electron number Ne=1,2..8 and with a single Mn2+ magnetic impurity. We have clarified the underlying physical mechanism and the role of the sp-d coupling between carrer and Mn2+-ion.
*publication*
An abstract on the subject has been accepted by "the 12$^{th}$ International conference on the Modulated Semiconductor Structures", Albuguerger, July 2005.
A manuscript for journal paper on the subject is in intensive preparation.

5. We have just started to develop a multiband microscopic theory for self-assembled quantum dots. A simple code using 4X4 k.p model is finished for quantum well system. We plan to spend one year on finishing the code for quantum dot system.