行政院國家科學委員會補助專題研究計畫 ■成果報告 □期中進度報告

計畫名稱:

半導體量子點偏振精細光譜的研究

計畫類別：■個別型計畫　□整合型計畫
計畫編號： NSC 98-2112-M-009-011-MY2
執行期間：　98 年 8 月 1 日至　100 年 11 月 30 日

執行機構及系所：國立交通大學電子物理系

計畫主持人：鄭舜仁
共同主持人: Dr.Hanz Ramirez(博後),盧書楷(博四),陳彥廷(助理), 廖禹淮(博一), 許克銘(碩二),徐燁(碩二),曾浤鈞(碩一),廖建智(碩一),張語宸(碩一),林以理(碩一), 鄭丞偉(碩一),張書瑜(碩一),古智豪(碩一),陳力瑋(碩一)

成果報告類型(依經費核定清單規定繳交)：□精簡報告　■完整報告

本計畫除繳交成果報告外，另須繳交以下出國心得報告：
□赴國外出差或研習心得報告
□赴大陸地區出差或研習心得報告
□出席國際學術會議心得報告
□國際合作研究計畫國外研究報告

處理方式：除列管計畫及下列情形者外，得立即公開查詢
□涉及專利或其他智慧財產權，□一年■二年後可公開查詢

中　華　民　國　101 年　1　月　10　日

目錄

計畫中文摘要

半導體量子點偏振精細光譜的研究

關鍵字: 量子點 激子 精細結構 量子糾纏

量子糾纏態(quantum entanglement)自 Einstein, Podolsky 和 Rosen 提出後一直是物理界廣泛討論的議題，量子通訊與資訊的發展開啟了量子糾纏態相關的應用。在光量子傳輸中，糾纏態光子對發射器(entangled photon-pair emitters)為不可或缺的光子元件。利用半導體量子點(quantum dots)製作單一光子(single-photon)和極化光子對(polarized photon-pair)發射器早已研發多年。然而，直接從量子點中產生糾纏光子對仍尚未具體實現，這是因為在大部份量子點中存在著所謂精細結構分裂(fine structure splitting)，如此僅約$10^1-10^2\,\mu$eV 精細結構的匹裂導致單激子(single exciton)自旋態不完全簡併，而使得不同自旋的電子電洞再結合的路徑變成可以分辨而無法產生糾纏光子對。目前造成精細結構分裂背後的真實物理機制仍存有爭議。然而一般認為，精細結構分裂的存在和量子點中對稱性破壞有關，量子點中形變(shape deformation)、應變(strain)和壓電(piezoelectricity)不對稱的綜合效應引發電子和電洞對間的交換作用(electron-hole exchange interaction)導致精細結構的匹裂。因此如何有縮小甚至控制量子點的激子精細結構是在目前量子傳輸應用中的重要課題，要達成這個目標則有必要對精細結構分裂的背後的物理機制有深入的瞭解。

在這個計劃中，我們將發展一多尺度理論用來研究量子點中電子電洞對的交換作用能，此研究工作將與交大電物張文豪教授的實驗團隊密切合作，目前已有部分初步結果。研究方法如下:首先建構一考慮量子點中載子微觀性質的多激子 configuration interaction method 的理論。在這個理論中我們必須充分的考慮粒子與粒子的交互作用，包含一般的電子與電子、電洞與電洞、以及電子與電洞間的交互作用，再加上最重要的電子與電洞的交換交互作用。量子點的電子結構計算，則使用多能帶的 k.p 模型結合連續彈力模型的應力理論，並進一步計算出由應力所引發的壓電場。多激子的問題以及量子點的電子結構計算部份須大尺度矩陣對角化的技術，我們將使用 ARPACK 矩陣對角化的數值方法。此一研究可以讓我们對精細結構分裂的物理機制有更深入的瞭解進而找出有效控制量子點的激子精細結構的方法。

計畫英文摘要

Theoretical studies of the polarized fine structures of spin excitons in semiconductor quantum dots

Keywords: quantum dot, exciton, quantum entanglement, electron-hole exchange interaction, fine structure splitting

Polarization-entangled photon pair sources are one of the key photonic devices necessary in optical quantum cryptography and teleportation. Fabrication of semiconductor quantum dots (QDs) as single photon and polarization entangled photon pair sources has been under development for years. However, generating entangled photon pairs directly from QDs still remains a challenge because of the fine structure splittings (FSSs) of spin exciton states inherent in solids. Due to the FSSs, the cascade emission paths via the different spin exciton states becomes distinguishable and unable to generate photon-pairs in entanglement. Such optical FSSs are known as a consequence of e-h exchange interaction associated with the symmetry breaking of QD, a combined effect of shape deformation, strain, and piezoelectricity. How to reduce and control the FSS is essential for the realization of optical quantum information technology using dot based photonic devices. To achieve that, a full understanding of the complex underlying physics of the optical FSS and a unified theory of e-h exchange interaction is necessary.

In this project, we shall develop a unified theory of the e-h exchange interaction in self-assembled quantum dots (QDs), combining the theory of multi-exciton in and the computational techniques of the electronic structure of the dots. In the theory, the particle-particle interactions, including the conventional e-e, h-h, and e-h interactions, and the e-h exchange interactions, are fully taken into account. The multi-exciton problem is solved by using the advanced numerical technique of exact diagonalization. The electronic structure of strained self-assembled QDs is studied using a multi-band k.p model for quantum dots, combined with the theory of strain in the continuous elasticity model. The numerical technique (ARPACK or LANZOS) is employed in the numerical large-scale matrix diagonalization. The developed theory of polarized fine structures of quantum dots allows us to explore the physical origin of FSSs and propose feasible ways to reduce or even manipulate the FSSs.

前言:

Polarization-entangled photon pair sources are key photonic devices necessary in the applications of optical quantum cryptography and teleportation. Fabrication of semiconductor quantum dots (QDs) as “on-demand” single photon and polarization entangled photon pair sources has been under development for years.[1] However, generating entangled photon pairs directly from QDs is still very challenging because of the inherent fine structure splittings (FSSs) in solid-state nanostructures. Due to the FSSs, the cascade emission paths via the different spin exciton states becomes distinguishable and unable to generate photon-pairs in entanglement. Such optical FSSs are known as a consequence of e-h exchange interaction associated with the symmetry breaking of QD, a combined effect of shape deformation, strain, and piezoelectricity. Thus, how to reduce and control the FSS is desirable for the realization of optical quantum information technology using dot based photonic devices.

In this project, we have developed a unified theory of the e-h exchange interaction in self-assembled quantum dots (QDs), combining the theory of multi-exciton in and the computational techniques of the electronic structure of the dots. In the theory, the particle-particle interactions, including the conventional e-e, h-h, and e-h interactions, and the e-h exchange interactions, are fully taken into account. The exciton problem is solved by using the advanced numerical technique of exact diagonalization. Furthermore, the theory for the fine structures of excitons in strained self-assembled QDs is being extended in the scheme of a multi-band k.p model for quantum dots, combined with the theory of strain in the continuous elasticity model. The numerical technique (ARPACK) is employed in the numerical large-scale matrix diagonalization. The developed theory of polarized fine structures of quantum dots allows us to explore the physical origin of FSSs and propose feasible ways to reduce or even manipulate the FSSs. The revealed physics and developed computation techniques in this project have been summarized in more than ten scientific articles published in SCI journals in the past two years, including one paper published in the high-impact journal “Physical Review Letters”. [2]

研究目的:

Polarization-entangled photon pair sources are key photonic devices necessary in the applications of optical quantum cryptography and teleportation. Fabrication of semiconductor quantum dots (QDs) as “on-demand” single photon and polarization entangled photon pair sources has been under development for years.[1] However, generating entangled photon pairs directly from QDs is still very challenging because of the inherent fine structure splittings (FSSs) in solid-state nanostructures. Due to the FSSs, the cascade emission paths via the different spin exciton states becomes distinguishable and unable to generate photon-pairs in entanglement. Such optical FSSs are known as a consequence of e-h exchange interaction associated with the symmetry breaking of QD, a combined effect of shape deformation, strain, and piezoelectricity. Thus, how to reduce and control the FSS is desirable for the realization of optical quantum information technology using dot based photonic devices.

文獻探討:

In the past years, researchers have demonstrated controlled FSS reductions of single QDs using strain and

post-annealing techniques, and the application of electric and magnetic fields[3-9]. In most experiments, however, it is not clear if the reduction of FSS is caused by the undoing of symmetry breaking or the reduction of e-h wave function overlap. The latter effect reduces not only the FSS but also the oscillator strength of e-h recombination, yielding narrow intrinsic radiative broadening and actually inhibiting the generation of entangled photon pairs. To search for more efficient means of controlling the optical fine-structure properties of quantum dots, it is necessary to understand the real underlying physics. Nevertheless, up to date the theoretical exploration into the subject is still limited. Around the world, only few advanced research groups are capable of studying the delicate properties of photo-excited quantum dots. In this project, we intend to study the optical fine-structure properties of single and double quantum dots (DQDs). The theoretical approaches that we use are based on the multi-band k.p theory and configuration interaction method. Our study provides the useful suggestion for the optimal design of dot-based entangled photon pairs.

研究方法:

A partial configuration interaction (PCI) method, beyond the widely used simplified “rigid orbital” model, is developed with full consideration of the Coulomb matrix elements of both e-h direct and exchange interactions, based on the s- and p-orbitals of individual quantum dots (QDs). To the best of our knowledge, the developed PCI theory is herein developed for the first time as a useful tool for studying the excitonic fine structures of QD molecules.The CI calculations were numerically implemented with full consideration of all possible intra- and inter-dot Coulomb matrix elements of both e-h direct and exchange interactions based on 12 single particle orbitals. The required Coulomb matrix elements were numerically calculated using 3D trapezoidal integration technique and then confirmed by analytical solutions whenever they are available.

Furthermore, the theory for the fine structures of excitons in strained self-assembled QDs is being extended in a multi-band k.p model combined with configuration interaction method. The numerical technique (ARPACK or LANZOS) is employed in the numerical large-scale matrix diagonalization.

結果與討論:

With the excellent optical and quantum properties, self-assembled quantum dots (QDs) have been considered as promising nanomaterial for advanced photonic applications. The optical processes in QD’s usually involve either recombination or creation of electron-hole (e-h) pairs in strongly bound multi-exciton complexes. Remarkably, besides the *direct* inter-particle Coulomb attractions, the electron-hole *“exchange”* interactions in an exciton are essential in the optical fine structures and the application of on-demand entangled photon-pair emitters made of QDs. The objective of this project is to develop a unified theory for the investigation of the spin properties, optical fine structures and dynamical processes of multi-exciton complexes in self-assembled QDs. The following theories and numerical approaches have been employed in the studies:

- Multi-band $k \cdot p$ theory for the electronic structure of self-assembled QDs subject to strain and composition inter-diffusion.

- 3D finite difference method.
- Large scale sparse matrix diagonalization.
- Finite element package for strain calculations (Multi-physics Comsol package).
- Green's function method for strain calculations.
- Multi-exciton configuration interaction method (including both electron-hole direct and exchange interactions)
- Parallelized Coulomb integration techniques.

In this project, we focus the study on the optical fine-structure properties of single and coupled double quantum dots (DQDs).

For single dots, we present a theoretical model for the electron-hole exchange interaction in three-dimensionally confining semiconductor nanostructures and use it to successfully explain the observed decreasing tendency of the fine-structure splitting of small InGaAs/GaAs self-assembled quantum dots _QDs_ with increasing the emission energies. The experimentally revealed FSS reduction is shown to be highly associated with the significant 3D spreading of electronic orbitals and reduced overlap of electron and hole wave functions in small and/or Ga-diffused QDs.[10]

In the study of double quantum dots, we predict that the optical FSSs in the DQDs in the strong tunneling regime are significantly diminished, without any decrease in the optical oscillator strength. Our study suggest that strongly coupled vertical quantum-dot molecules with high tunneling rate are better sources of entangled photon pairs than single dots. Tunneling is a remarkable quantum property of microscopic particles that has no classical counterpart, which allows coupling between two objects spatially separated by a finite potential barrier. Currently, extending the analogy between atoms and 0D solid state systems, coupled quantum dots (QDs) are widely studied as artificial molecules where important properties of single dots are improved for optimization and scalability of applications. Recent examples of interesting and useful tunnel effects in coupled dot systems include the tunability of fluctuations in Kondo currents, reduction of electronic spin decoherence by interaction with nuclear spin, conditional dynamics of transitions and bias control of g-factors .

This study discusses the effects of quantum tunneling on polarized photon emission from spin excitons in vertically stacked double quantum dots. Results show that an increase in the optically active volume and charge delocalization via quantum tunneling inhibits the optical FSS of coupled QDs in the strong tunneling regime without any decrease in the optical oscillation strength. This tunneling-driven FSS reduction is robust against the bias-controlled detuning from resonance, making strongly coupled vertical quantum dot molecules better cascade decay sources of entangled photon pairs than single dots.

Moreover, we have studied the spin relaxation of an exciton confined in QDs. Spin relaxation of exciton in self-assembled quantum dots (QDs) has drawn considerable attention in recent years because of its essential role in various dot-based quantum photonic applications. In the applications, the spin relaxation between two nearby exciton states has a determinate influence on the performance of the devices, and control over it would

be desirable. For instance, the spin dynamical properties of a bright exciton (BX) with total spin M=+1/-1 in a SQD could be substantially affected by the long-lived but optically inactive dark exciton (DX) states of different spin M=+2/-2, which are usually energetically separated from the BX ones by some hundreds of micro-eV by e-h exchange interaction.

With the assistance of phonon coupling, the DX states are allow to transit to BX ones via the spin-scattering mechanisms including spin-orbital couplings or hyperfine interactions, and vice versa. In this work, we theoretically evaluate the rate of spin-state transition from a DX state to BX states via acoustic phonon interaction in self-assemble quantum dots. We present a comprehensive study of various possible involved spin flip mechanisms, including electron hyperfine interaction, electron-Rashba and -Dresselhaus spin-orbit interaction, hole-linear and hole-Dresselhaus spin-orbit interaction. In our studies, it is shown that the hole-Dresselhaus spin-orbit coupling makes a dominant contribution to the DX-to-BX spin relaxation, leading to spin relaxation time as fast as 10^-2ns consistent with recent observation. Moreover, an extraordinary size-dependence of the spin relaxation rate of a dot-confined exciton due to the combined effect of hole-Dresselhaus spin-orbit coupling and e-h exchange interaction is revealed in this work.[11]

Selected articles on the research subjects published in the past two years are listed below :

(1). Optical fine structures of quantum dot

- Chia-Hsien Lin,Wen-Ting You, Hsiang-Yu Chou, Shun-Jen Cheng, Sheng-Di Lin, and Wen-Hao Chang*, “Anticorrelation between the splitting and polarization of the exciton fine structure in single self-assembled InAs/GaAs quantum dots”, Phys. Rev. B **83**, 075317 (2011).
- H. Y. Ramirez, C. H. Lin, C. C. Chao, Y. Hsu, W. T. You, S. Y. Huang, Y. T. Chen, H. C. Tseng, W. H. Chang, S. D. Lin and S. J. Cheng*, “Optical fine structures of highly quantized InGaAs/GaAs self-assembled quantum dots”, Phys. Rev. B **81**, 245324 (2010) (**IF:3.322**)
- Hanz Y. Ramirez and Shun-Jen Cheng*,”Tunneling effects on fine structure splitting in quantum dot molecules”, Phys. Rev. Lett. **104**, 206402 (2010) (**IF:7.18**)
- Hanz Y. Ramirez, Chia-Hsien Lin, Wen Ting You, Shan-Yu Huang, Wen-Hao Chang, Sheng-Di Lin, Shun-Jen Cheng*, “Electron–hole symmetry breakings in optical fine structures of single self-assembled quantum dots“, Physica E: Low-dimensional Systems and Nanostructures, 42, 1155 (2010) (**IF:1.23**)
- H. Lin, S.-Y. Wang, C.-H. Lin, W.-H. Chang*, S.-J. Cheng, M.-C. Lee, W.-Y. Chen, T. M. Hsu, T.-P. Hsieh and J.-I. Chyi, “Exciton fine structures and energy transfer in single InGaAs quantum-dot molecules”, Physica Status Solidi. (c) **6,** 860 (2009) (SCI)
- Hanz Y. Ramirez, Shun-Jen Cheng*, and Chih-Pin Chang “Theory of Electron-Hole Exchange Interaction in Double Quantum Dots”, Physica Status Solidi. (b) **264**, 837 (2009) (SCI) (**IF:1.071**)
- Wen-Hao Chang*, Hsuan Lin, Sheng-Yun Wang, Chia-Hsien Lin, Shun-Jen Cheng, Ming-Chih Lee, Wen-Yen Chen, Tzu-Min Hsu, Tung-Po Hsieh, Jen-Inn Chyi (2008) “Nonresonant carrier transfer in single InGaAs/GaAs quantum dot molecules”, Phys. Rev. B , **77**, 245314 (SCI) (**IF:3.172**)

(2). Magneto-excitons in quantum dots

- Y. J. Fu, S. D. Lin*, M. F. Tsai H. Lin, C. H. Lin, S. Y. Wang, S. J. Cheng, W. H. Chang, “Anomalous diamagnetic

shift for negative trions in single self-assembled InAs/GaAs quantum dots“, Phys. Rev. B **81**, 113307 (2010). (**IF:3.322**)

- Wen-Hao Chang*, Chia-Hsien Lin,Ying-Jhe Fu, Ta-Chun Lin, Hsuan Lin, Shun-Jen Cheng, Sheng-Di Lin, Chien-Ping Lee. “Impacts of Coulomb Interactions on the Magnetic Responses of Excitonic Complexes in Single Semiconductor Nanostructures“, Nanoscale Res. Lett. **5** 680 (2010). (**IF:1.731**)

(3) spin relaxation of exciton

- Yu-Huai Liao, Juan I. Climente, and Shun-Jen Cheng*, “Dominant channels of exciton spin relaxation in photoexcited self-assembled (In,Ga)As quantum dots”, Phys. Rev. B **83,** 165317 (2011) .(IF: 3.475)

成果自評:

研究內容與原計畫相符程度:

在這個計劃中，我們完成一多尺度理論用來研究量子點中電子電洞對的交換作用能，並利用此理論方法對單一量子點及雙量子點的經細結構進行計算，並且與交大電物張文豪教授的實驗團隊的實驗結果有充份的比對，研究內容與原計畫完全相符。

達成預期目標情況:

本計劃已達成下列預期目標

理論方法:已完成一量子點中多激子的 configuration interaction (CI) method 理論。在這個理論中我們完整的考慮粒子與粒子的交互作用，包含一般的電子與電子、電洞與電洞、以及電子與電洞間的交互作用，再加上最重要的電子與電洞的交換交互作用。量子點的電子結構計算，則使用多能帶的 k.p 模型結合連續彈力模型的應力理論，並進一步計算出由應力所引發的壓電場。

數值技術: 已完成大尺度矩陣對角化的技術，我們使用 ARPACK 矩陣對角化的數值方法計算量子點的電子結構。並完成利用 GPU 技術對庫倫積分進行平行運算。

研究結果:我們的理論計算首度指出若利用雙量子點製作光子對發射器可大幅提高發產生糾纏態光子對的成功率，因為雙量子點間的量子穿隧(tunneling)效應可降低電子和電洞對間的交換作用力進而縮小量子點精細結構匹裂，尤其值得注意的是此穿隧效應並不會降低雙量子點的發光強度反而微幅提高電子和電洞的再結合率(electron-hole recombination rate)，並進一步提高產生糾纏態光子對的成功率。近年來實驗上已可製作高品質的量子點分子因此製作我們所提出的雙量子點量子光源具相當可行性。

學術合作:密切與交大電物張文豪教授,日本國家材料研究所 Prof. Takashi Kuroda 的實驗團隊合作，並有具體研究結果。這些研究結果均已整理發表國際知名期刊。

學術成就: 自 2009 至今計劃執行期間研究結果已整理發表國際知名期刊約近 20 篇，其中包含一篇

發表於 Phys. Rev.Lett.的高 IF 的論文。並與國內外的研究團對進行實質的合作，過去五年中總計共發表純理論工作 9 篇及實驗理論合作工作 11 篇。並多次接受國內外各大專院校物理系或會議的邀請演講 及學術訪問,包括

學術訪問:

- National Research Council, Ottawa, Canada (07-09/2004, 08/2005, 07-09/2006, 01/2007)
- 上海復旦大學 (08/2008)

演講:

- Yu-Huai Liao1, Juan I. Climente2 and Shun-Jen Cheng*

"Inherently fast spin relaxation of exciton in photo-excited self-assembled quantum dots"
2011 International Conference on Solid State Devices and Materials (SSDM2011), September 28-30, 2011Nagoya, Japan

- Shun-Jen Cheng*,

"Crucial excitonic properties of quantum dots as advanced light sources: spin relaxation, optical fine structure and polarization anisotropy" BIT's 1st Annual World Congress of Nano-S&T, October 23-26, 2011, Dalian, China

- "Manipulation of optical fine structure of spin excitons in self-assembled quantum dot molecules",International Workshop "Computational approaches to semiconductor, carbon and magnetic nanostructures", Ryon, France, June 16-19 2008 .
- "Electronic and magnetic properties of semi-magnetic quantum dots",INTERNATIONAL WORKSHOP ON NOVEL PHENOMENA AT NANOSCALE INTERFACES, Hsinchu, December 11–13, 2008.

技術創新: 在理論工作方面已發展的相關理論及數值技術如下：

＊大型稀疏矩陣對角化(Arpack)

＊有限差分法解三維薛丁格方程式

＊多能帶 $k \cdot p$ 法計算量子點電子結構

＊有限元素法計算應力應變(Comsol package)

＊多激子/多電子組態交互作用法(configuration interaction method)

社會影響: 利用半導體量子點製作糾纏態光子對發射器早已研發多年。然而，直接從量子點中產生糾纏光子至今仍是一極大的挑戰。我們的理論計算首度指出若利用雙量子點製作光子對發射器可大幅提高發產生糾纏態光子對的成功率。此突破有助於實現光子密碼遠傳等應用。

參考文獻

[1] O. Benson *et al.* Phys. Rev. Lett. **84**, 2513(2000).

[2] [H. Y. Ramirez, *et al.*, *"Optical fine structures of highly quantized InGaAs/GaAs self-assembled quantum dots"*, Phys. Rev. B **81**, 245324.(2010)]

[3] D. T. McClure *et al.*, Phys. Rev. Lett. **98**, 056801 (2007).

[4] D. J. Reilly *et al*., Phys. Rev. Lett. **101**, 236803 (2008).

[5] L. Robledo *et al*., Science **320**, 772 (2008).

[6] T. Andlauer *et al.*, Phys. Rev. B. **79**, 045307 (2009).

[7] E. Margapoti *et al*. Appl. Phys. Lett. **90**, 181927 (2007).

[8] B.D.Gerardot *et al.* Appl. Phys. Lett. **90**, 041101 (2007).

[9] R. M. Stevenson *et al.* Phys. Rev. B. **73**, 033306 (2006).

[10] H. Y. Ramirez, C. H. Lin, C. C. Chao, Y. Hsu, W. T. You, S. Y. Huang, Y. T. Chen, H. C. Tseng, W. H. Chang, S. D. Lin and S. J. Cheng*, "Optical fine structures of highly quantized InGaAs/GaAs self-assembled quantum dots", Phys. Rev. B **81**, 245324 (2010)

[11] Yu-Huai Liao, Juan I. Climente, and Shun-Jen Cheng*, "Dominant channels of exciton spin relaxation in photoexcited self-assembled (In,Ga)As quantum dots", Phys. Rev. B **83,** 165317 (2011) .