附件一

行政院國家科學委員會補助專題研究計畫 □ 成果報告 v 期中進度報告

晶體材料之載子自旋與同調聲子動力學之研究(2/3)

計畫類別：v 個別型計畫　　□ 整合型計畫
計畫編號：NSC 92-2112-M-009-014-
執行期間：92 年 08 月 01 日至 93 年 07 月 31 日

計畫主持人：黃中垚
共同主持人：
計畫參與人員：

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

本成果報告包括以下應繳交之附件：
□赴國外出差或研習心得報告一份
□赴大陸地區出差或研習心得報告一份
□出席國際學術會議心得報告及發表之論文各一份
□國際合作研究計畫國外研究報告書一份

處理方式：除產學合作研究計畫、提升產業技術及人才培育研究計畫、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
□

執行單位：國立交通大學光電工程研究所

中　華　民　國 93 年 5 月 31 日

附件二

可供推廣之研發成果資料表

v 可申請專利 v 可技術移轉　　日期：2004 年 5 月 31 日

國科會補助計畫	計畫名稱：晶體材料之載子自旋與同調聲子動力學之研究 計畫主持人：黃中垚 計畫編號：NSC92-2112-M-009-014-　　學門領域：物理
技術/創作名稱	飛秒雷射波形轉換、全光場檢測、與同調控制多光子顯微光譜技術
發明人/創作人	陳明彰、李兆奎、黃中垚
技術說明	中文： 本研究計畫發展一新分析工具並在此同一平台上實現飛秒光場轉換、全光場分析(包括光場的大小和相位)、與同調控制等三項功能。我們將此同調控制技術與雙光子激發螢光光譜顯微術結合，以分析半導體微結構。達到以同調控制方式分別增強光譜差僅 20 nm 的凝態材料的雙光子螢光。 英文： An apparatus, which can offer femtosecond pulse conversion, characterization and coherent control functionalities on the same platform, has been developed and could be combined with advanced multiphoton optical microscopy for imaging enhancement, selective excitation, and coherent control purpose, *etc*.
可利用之產業及可開發之產品	1. Manufacturing industry of advanced optical microscope. Product: adaptive coherent controlled multiphoton luminescent microscope with pulse shaping and compensation; 2. Laser industry. Product: direct complete-field characterization apparatus of femtosecond optical pulse 3. Femtosecond laser instrument. Product: femtosecond pulse design and conversion
技術特點	1. New algorithm for rapid search of optimum spectral phase of optical field needed to achieve coherent control; 2. Pulse conversion, complete-field characterization and adaptive coherent control on the same apparatus; 3. Direct approach to deduce complete-field characteristics of femtosecond coherent pulse.
推廣及運用的價值	1. selective excitation optical microscopy quantum-control; 2. imaging enhancement of multiphoton optical microscopy; 3. quantum control and manipulation of optical excitation in nanostructured materials; etc.

※ 1. 每項研發成果請填寫一式二份，一份隨成果報告送繳本會，一份送　貴單位研發成果推廣單位（如技術移轉中心）。

※ **2. 本項研發成果若尚未申請專利，請勿揭露可申請專利之主要內容。**

※ 3. 本表若不敷使用，請自行影印使用。

中文摘要

此三年期計畫主要目標在應用飛秒雷射科技分析凝態材料載子自旋與同調聲子。我們使用光波合成技術希望能深入分析並操控量子侷限材料體系之電子自旋與同調聲子傳輸動力過程。

在第二年進度我們達成：(一)新分析工具之發展。我們使用此技術在同一平台上實現飛秒光場轉換、全光場分析、與同調控制等三項功能。（二）我們將此同調控制技術應用於分析半導體微結構，實現雙光子激發螢光光譜僅差 20 nm 的固態材料之同調控制增強效應。（三）為滿足量子侷限結構材料之研究需求，我們也報導以溶液成長法製備之磁性材料/貴金屬核-殼量子點結構。將用於製備週期排列自動組裝奈米結構薄膜。這些成果不但呈現我們在材料製備與分析技術方面的發展而且也展現我們的努力與為下一階段研究所做的準備。

Abstract

The major objective of this three-year research project is to apply femtosecond laser technology for probing dynamics of carrier-spin and coherent phonon in condensed materials. By employing optical pulse shaping technique, we hope to manipulate the coherence transport of the carrier spin and phonon generation in some quantum-confined systems.

In this second-year progress report, we present: (1) the developments of an apparatus which enables us to perform femtosecond pulse conversion, characterization and coherent control on the same platform. We demonstrate these functionalities with some semiconductor saturable absorber Bragg reflectors. We are able to realize a selective enhancement of two-photon excited luminescence from semiconductor microstructures with PL spectral peak differing by only 20 nm.

Solution growth technique of magnetic/noble metal core-shell quantum dots has been developed for building highly ordered self-assembling bulk film. This progress offers an opportunity to probe and control the spin transport dynamics in a low-dimensional quantum-confined system. The results reported here reflect our efforts and well preparation for the next stage research.

Progress Report

I. 3C (conversion, characterization and coherent control) strategy of femtosecond laser pulse on a single platform[1]

By using the broadband property of an ultrashort laser pulse, Silberberg *et al.*[2] had developed a coherent control scheme to demonstrate selective imaging of molecules via single-pulse coherent anti-Stokes Raman scattering. Similar enhancement with coherent control technique had also been observed in two-photon fluorescence process.[3,4] Along this study, the question remained is whether the optimal laser field contains a set of rational rules that govern the dynamics. Recent study appears to reveal that the answer could be affirmative.[5] Therefore the purpose of femtosecond coherent control study is not only to control the evolution of a complex system but also to deduce the detailed dynamic mechanism from the optimal laser field used.

In order to have the scheme widely applicable, the apparatus shall offer the so-called **3C** strategy, which includes the functionalities of **conversion** (to any desired pulse profile from a given coherent optical pulse), complete-field **characterization** (of the designed pulse to verify the characteristics needed), and **coherent control** (of the light-matter interaction) at the femtosecond scales. During the second year of this project, we have developed such a technique

to fulfill the requirement of future quantum evolution control.

The operational principle bases on the concept that views the spectral phase components of a coherent pulse as a system of interacting particles. If a temperature sensor to reveal how cold it is at zero degree and a thermal pump to control the amount of heat to be added into or taken from the system are available, information about the interacting-particles can be yielded by studying how the system responses to a thermal excitation.

Our experimental implementation of this concept includes a femtosecond laser whose output pulses are tailored by a pulse shaper. The pulse shaper consists of a pair of gratings, two concave reflectors, and a liquid crystal spatial light modulator (SLM). After reassembled by the output grating, the shaped pulse is focused onto a sample under test. The phase distortion in the reflected optical pulse can be pre-compensated by the SLM. An optical pulse with constant phase can be produced in front of the 3-mm thick type-I β-Ba_2BO_4 (BBO) second harmonic generation (SHG) crystal. We combine BBO SHG with a photodiode to offer a functionality of constant phase detection. The photodiode signal is sent to a computer for generating phase compensating pattern with the freezing-phase algorithm.

This apparatus has been applied to convert single femtosecond pulse to multiple pulses, single pulse with rectangular or triangular profile, *etc*. Our technique is fast, accurate and more immune to the noise and laser power fluctuation than that with genetic algorithm.

To demonstrate the functionality of complete-field characterization, we employed the apparatus for investigating three types of semiconductor saturable absorber Bragg reflectors (SBR). The results show that our apparatus can clearly reveal the spectral phase distortion from slight structural changes of the devices.

We first use second-harmonic generation frequency-resolved optical gating technique (SHG-FROG) to characterize the optical field reflected from the SBR devices. The SHG-FROG results are shown in Fig. 1. We then compare the results deduced from our new apparatus with SHG-FROG.

The first SBR device to be studied comprises of two $Ga_{0.47}In_{0.53}As$ quantum wells, which are embedded in an $Al_{0.48}In_{0.52}As$ quarter wave layer on a distributed Bragg reflector (DBR) stack (hereafter is abbreviated as *d*-QW). The DBR stack is formed with 25 pairs GaAs/AlAs designed to yield a Bragg wavelength at λ_B=1.23 μm. The other is self-assembled InAs quantum-dots layer embedded in a quarter-wave-thick (QD-λ/4) or half-wave-thick (QD-λ/2) GaAs layer on a DBR stack. The DBR of the two devices is identical and contains 21-periods stack of 97 nm/112 nm GaAs/$Al_{0.92}Ga_{0.08}As$. The DBR was designed to yield high reflection at 1.3 μm. The experimental SHG-FROG trace was retrieved with an error of 0.0025. The retrieved spectral phase profiles for the three SBR devices are presented in Fig. 2(c) and are found to overlap with each other near the central region but significant difference can be observed at the spectral tails of QD-λ/4 and QD-λ/2. The field strength experienced by the InAs QDs in QD-λ/2 is smaller and therefore we expect to observe weaker pulse shaping effect and therefore larger phase distortion in QD-λ/2.

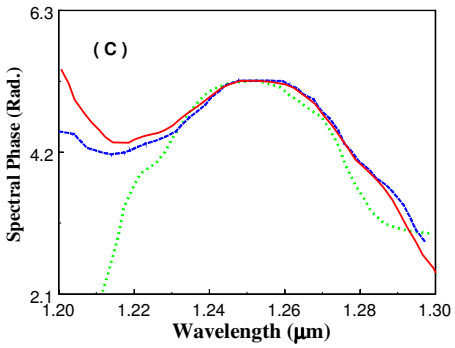

Fig. 1. The retrieved spectral phase profiles from *d*-QW (solid curve), QD-λ/4 (long dashed) and QD-λ/2 (short dashed line) SHG-FROG traces.

After SHG-FROG analysis, we proceed to perform the complete-field characterization with our adaptive phase compensation apparatus. The results are summarized in Fig. 2, where in 2(a); a direct comparison of the deduced spectral profile of the optical pulse reflected from the *d*-QW sample to that measured with Fourier-transformed infrared spectroscopy (FTIR) is presented. An excellent agreement was found, indicating that our adaptive phase compensation scheme not only be able to yield the spectral phase profile but also the amplitude of a coherent optical pulse. We then present the measured spectral phase profiles with FA for the three SBR devices in Fig. 2(b). The global features of the measured spectral phase profiles are similar to that obtained with SHG-FROG technique. The most deviations occur at the regions with small spectral amplitude where retrieving with SHG-FROG algorithm is usually less reliable. The slight shift of the QD-λ/2 spectral phase profile (short dashed curve) from *d*-QW (solid curve) and QD-λ/4 (long dashed) also appears in the FTIR spectra. Note that the device structure of QD-λ/2 is very similar to QD-λ/4 except a twice thicker QDs embedded layer employed in QD-λ/2. The clearly distinguishable differences in the spectral phase profiles ensure that our new complete-field characterization scheme is sensitive and accurate to reveal influence on femtosecond optical pulse with a slight change in SBR structure. Furthermore, unlike SHG-FROG where the pulse characteristics are retrieved with sophisticated mathematical procedure, our method belongs to a direct measurement approach.

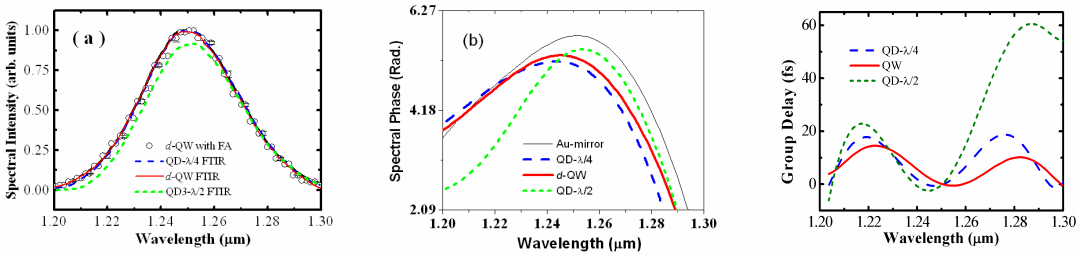

Fig. 2. (a) Spectral profiles of optical pulses reflected from *d*-QW (open circles) measured with freezing-phase algorithm (FA) and from *d*-QW (solid curve), QD-λ/4 (long dashed), QD-λ/2 (short dashed) with Fourier-transformed infrared spectroscopy (FTIR) (b) Spectral phase profiles from Au-mirror (thin solid curve), *d*-QW (thick solid curve), InAs QD-λ/4 (long dashed), and InAs QD-λ/2 (short dashed) deduced with phase freezing scheme; (c) group delay time of the three SBR devices over the entire pulse spectral range.

Note that we can also determine the group delay times of the SBR devices by first taking

a difference between the spectral phase profiles of SBR and Au-mirror and then differentiating the phase difference profiles with respect to angular frequency. The results are presented in Fig. 2 (c), which show that the *d*-QW SBR exhibits much weaker wavelength-dependent group delay within the entire spectral range of the optical pulse. Indeed this device had been designed for passively mode locking femtosecond laser at 1.25 μm and was confirmed experimentally to be able to generate femtosecond laser pulse with pulse duration less than 60 *fs*. As expected, among the three SBR structures the QD-λ/2 shows largest variation in group delay within the spectral range, especially for the spectral components with wavelength longer than 1.27 μm. The larger variation in group-delay by QD-λ/2 can originate from weaker field strength being experienced by the InAs QDs and therefore weaker pulse shaping effect is yielded.

II. Coherent-controlled two-photon luminescent microscopy of semiconductor microstructure with freezing algorithm of spectral phases [6]

The core issue about this research is to develop a technology platform with improved control capability on light-matter interaction. With this control power in hand, we shall be able to steer photon energy into specific degrees of freedom of materials, to create new materials, to generate new functionality from a device, or even to probe specific molecule within an ensemble of species.

In particular, we are interested in new phenomena resulting from an enhanced coupling between electrons and photons at nanometer scales. Photon energy with wavelength of hundreds nanometers had been predicted to localize to a few nanometers by using a proper control of transient optical phase. If the technique is verified, an approach to enhance coupling between photon and electron at nanometer scales shall be possible and brand new concepts for photonics are expected. Along this direction, we first explore the possibility of selectively enhancing the two-photon excited photoluminescence from a semiconductor microstructure. The experimental setup is depicted in Fig. 3(a). The sample used is self-assembled InAs quantum dots embedded inside a GaAs epilayer. The device structure and related optical transitions are shown in Fig. 3(b).

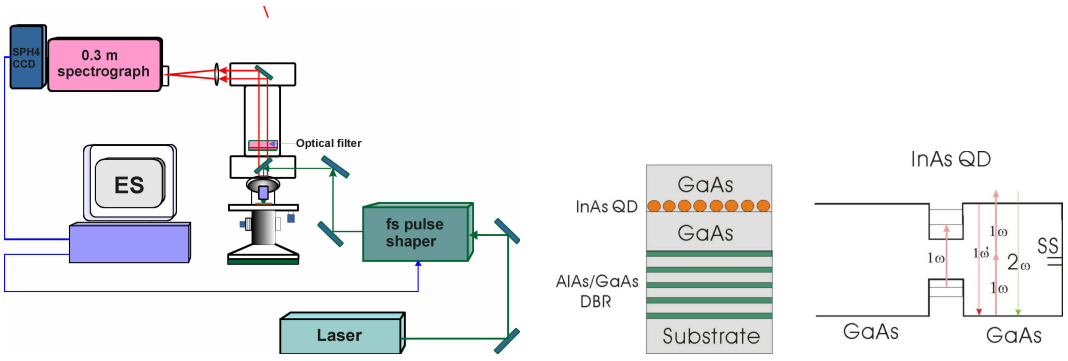

Fig. 3 (a) Schematic of the adaptive coherent control multiphoton luminescence microscope, (b) the related optical transitions and electronic structure of semiconductor microstructure used in this study.

Two-photon excited photoluminescence near 880 nm can be observed and is attributed mainly due to the GaAs epilayer. The two-photon excited PL image sensitively reflects the film quality and the in-plane material inhomogeneity. A typical two-photon excited micro-PL image obtained with femtosecond laser at 1.25 μm is presented in Fig. 4(a). By using our coherent

control multiphoton luminescence microscope, we can enhance the PL signal at least three times to yield an image with higher contrast (see Fig. 4(b)).

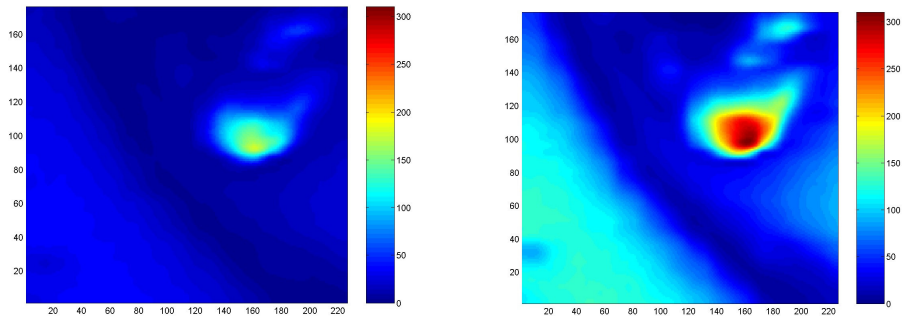

Fig. 4 (a) Two-photon excited micro-PL image of a sample of self-assembled InAs quantum dots embedded in a GaAs epilayer obtained with femtosecond laser at 1.25 μm; and (b) the coherent control enhanced multiphoton μ-PL image on the same area.

In Fig. 5(a), the two-photon excited PL spectrum from position A exhibits about 20 nm red-shift from that emitted at position B. The micro PL spectra are presented in Fig. 5(b). This red-shift is presumably originated from a localized compositional difference in the epilayer.

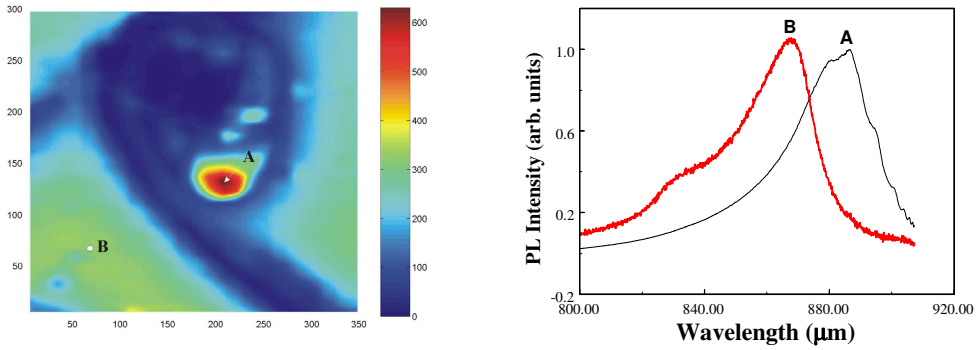

Fig. 5(a) Coherent control enhanced multiphoton μ-PL image of the same sample as Fig. 4; and (b) the two-photon excited PL spectra from position A and B marked in Fig. 5(a).

It is interesting to train the adaptive control multiphoton luminescence microscope to yield higher signal from position A and B, respectively. For each searching, the time to reach the optimal phase profile usually takes less than 5 minutes, which demonstrates that our coherent control apparatus has achieved a status of practical usage. The optimum phase profiles for enhancing the PL signals at position A and B are shown in Fig. 6(a) for comparison. The resulting coherent-control difference map of PL is presented in Fig. 6(b). We can clearly see fine feature developing near position A, indicating coherent control via femtosecond spectral phase leads to excitation redistribution within the semiconductor microstructure. The underlying mechanism can be explained by using the electronic model shown in Fig. 3(b) and will be detailed in a separate publication.

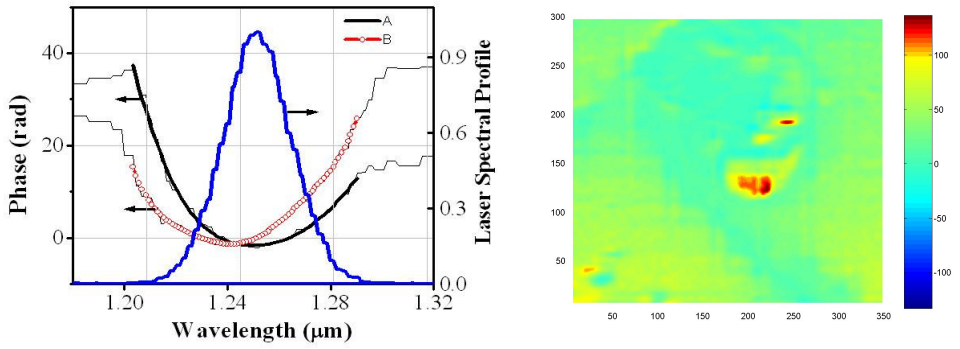

Fig. 6(a) The optimum phase profiles for enhancing the PL signals at position A and B; and (b) the resulting coherent-control difference map of PL signal from the semiconductor microstructure.

III. Preparation of self-assembled film of magnetic-metal core-shell quantum dots

One of the major objectives of this research is to probe and control the carrier spin transport in a low-dimensional quantum system. Magnetic quantum dots and their periodic arrays are the ideal advanced material system to be studied.

Magnetic quantum-dots (QD) such as Fe, Co, and Ni with an outer shell of noble metal can be prepared from solution growth technique. Furthermore, their size distribution is very narrow and controllable with growth duration. A sample-growing technique has been developed for preparing the core-shell magnetic nanocrystals and a bulk film with regular nano structure. The preparation schematic is depicted in Fig.7.

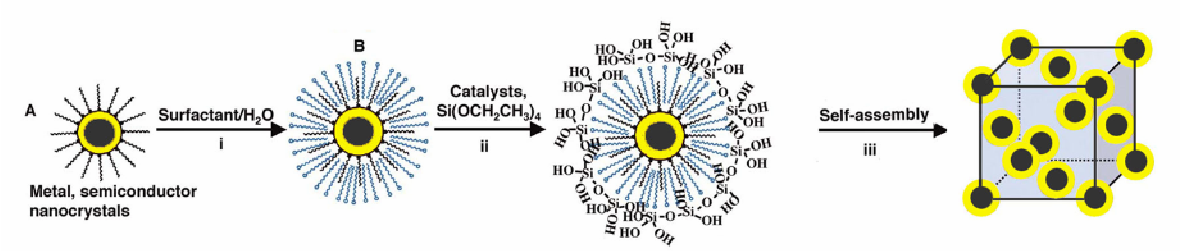

Fig. 7 Schematic for preparing core-shell magnetic nanocrystals and a plausible route for self-assembling into a bulk film with high structural regularity.

TEM and XRD measurement shows these solution-grown Fe/Au core-shell QDs have uniform particle size and high crystal quality. We are currently endeavored to develop a method for fabricating a highly regular structure of the nanoparticles in silica matrix formed by the TEOS surfactant. The resulting sample shall be unique for probing the quantum-confined spin transport dynamics.

Selected References

[1] M. Z. Chen, Qiantso Yang, and Jung Y. Huang, *Freezing phase scheme for fast adaptive control and its application to characterization of femtosecond coherent optical pulses*, J. Opt. Soc. Am. B (2004) **under revision**.
[2] N. Dudovich, B. Orion, and Y. Silberberg, Nature **418**, 512-515 (2002).
[3] T. Brixner, N. H. Damrauer, P. Niklaus, and G. Gerber, "Photoselective adaptive femtosecond quantum control in the liquid phase," Nature **414**, 57-60 (2001).
[4] Igor Pastirk, Johanna M. Dela Cruz, Katherine A. Walowicz, Vadim V. Lozovoy, and Marcos Dantus, "Selective two-photon microscopy with shaped femtosecond pulses," Opt. Express **11**, 1695 (2003).
[5] C. Daniel, *et al*., "Deciphering the reaction dynamics underlying optical control laser fields," Science **299**, 536 (2003).
[6] M.Z.Chen, and Jung Y. Huang, *Coherent controlled two-photon luminescent microscopy of semiconductor microstructure with freezing algorithm of spectral phases*, **in preparation**.