行政院國家科學委員會補助專題研究計畫成果報告

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※ 半導體材料與結構之載子動力學研究 ※
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計畫主持人：孫啟光

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行政院國家科學委員會專題研究計畫成果報告

半導體材料與結構之載子動力學研究

Studies of Carrier Dynamics in Semiconductor Materials and Structures

計畫編號：NSC87－2112－M－002－022

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一、中文摘要

本計畫研究氮化鎵及氮化銦鎵材料之載子動力與聲子動力特性。

在實驗系統架設方面，本計畫於第一年度即已完成鈦：藍寶石雷射系統之採購與架設。

在實驗結果方面，本計畫於三年執行期間完成下列成果：

(1) 電子冷卻實驗：發現氮化鎵材料之電子冷卻時間約為 0.5ps，較砷化鎵系統為快。
(2) 導帶谷間散射實驗：發現氮化鎵材料導帶之第二能帶谷底距真正能帶谷底之能量差約為 1.34 eV，遠較砷化鎵系統為大。谷間散射所造成之冷卻延遲時間為 1ps，亦較砷化鎵系統為快。
(3) 帶尾載子動力學實驗：發現氮化鎵材料之帶尾能態依能量高低而有截然不同之載子動力學表現。此行為是由於 mobility edge 低於帶底之故。
(4) 載子擴散行為實驗：發現氮化銦鎵/氮化鎵量子井中之載子擴散具有"giant ambipolar diffusion"之特性。此特性是由於量子井中巨大之壓電場所致。
(5) 同調聲學聲子震盪實驗：發現氮化銦鎵/氮化鎵多重量子井中具有極強之同調聲學聲子震盪。此特性是由於量子井中巨大之壓電場所致。
(6) 聲子震盪之同調控制實驗：利用飛秒雷射脈衝，本實驗室首次完成聲子震盪之光學同調控制實驗。聲子震盪之大小與相位均可由脈衝控制而達成。
(7) 非線性光學性質研究：本計畫完成氮化鎵材料之雙光子吸收常數、非線性折射係數、二倍頻常數、與三倍頻常數之量測。並將雙光子吸收、二倍頻、與三倍頻技術應用於氮化鎵共焦顯微鏡。

關鍵詞：電子、電洞、聲子、載子冷卻、谷間散射、擴散、同調聲子震盪、帶底能態、壓電場、非線性光學、共焦顯微鏡

Abstract

GaN has recently attracted a lot of attention due to its potential application in blue to UV light sources and high temperature and high field microwave electronics. Compared with the well-known GaN and its related material systems, GaN has heavier masses, larger optical phonon energies, a large exciton binding energy, and a wurzite crystal structure. In order to understand their influence on the material performance, we perform femtosecond pump-probe experiments in order to investigate the carrier and phonon dynamics.

For the system construction, we have built up a new femtosecond Ti:sapphire laser system under the NSC support during the first year of this project.

For the experimental part, we have obtained the following results with this project:

(1) Electron cooling experiment: Found 0.5 ps electron cooling time for GaN material system, which is faster than GaAs material system.
(2) Intervalley scattering experiment: Found 1ps delayed cooling time due to intervalley scattering. The second conduction band minimum was found to be 1.34 eV above the first conduction band minimum, which is much higher than GaAs material system.
(3) Bandtail state experiment: Found two different kinds of carrier dynamic behaviors in GaN bandtail states. These different behaviors are energetically divided by mobility edge.
(4) Carrier diffusion experiment: Found "giant ambipolar diffusion" behavior in InGaN/GaN multiple-quantum-wells. This behavior is induced by the large built-in piezoelectric field.
(5) Coherent acoustic phonon oscillation experiment: Observe extremely-large coherent acoustic phonon oscillations in InGaN/GaN multiple-quantum-wells. This large oscillation is induced and influenced by the large built-in piezoelectric field.
(6) Coherent control of phonon oscillation experiment: With an femtosecond pulse, we

demonstrate for the first time the coherent control of phonon oscillations. Both magnitude and phase control were successfully achieved.

(7) Nonlinear optical properties study: We have studied the nonlinear refractive index, two-photon absorption constant, second-harmonic generation, and third-harmonic generation behaviors in GaN. Extremely large nonlinearities were observed. Taking advantage of these large nonlinearities, we have demonstrated two-photon microscopy, three-photon microscopy, second-harmonic generation microscopy and third harmonic generation microscopy of GaN for the first time.

Keywords: electron, hole, phonon, carrier cooling, intervalley scattering, diffusion, coherent phonon oscillation, bandtail states, piezoelectric field, nonlinear optics.

二、計畫緣由與目的

半導體材料中的基本載子為帶負電之電子與帶正電之電洞。由於研究基本載子相互之間及基本載子與週遭環境，如聲子系統，間之交互作用為了解半導體現象之基礎，載子動力學因而受到高度重視。其研究動機更跟隨近日超小與超快電子元件之發展而增強。本計畫提出高能隙材料與低維結構之載子動力學研究。高能隙材料以氮化鎵/氮化銦鎵為研究對象。低維結構則以多重量子井為研究對象。

由於在藍光二極體、藍光至紫外光半導體雷射、高溫及高功率微波電子元件上的應用，氮化鎵及其相關材料的研究目前在國際上與國內均受到極高度重視。與已被充分了解之砷化鎵及其相關材料為比較對象，氮化鎵具有較重之電子與電洞質量、較高之激子鍵結能量、及六邊型的晶體結構。為了解這些不同特性所造成之影響，本計畫研究氮化鎵及氮化銦鎵材料之載子動力與聲子動力特性。

三、結果與討論

在本計畫執行期限內，因受本計畫之經費贊助，完成下列事項(部分經費由台大補助)：

(A)實驗系統架設方面:

1. 完成鈦：藍寶石雷射系統之採購與架設

   波長輸出範圍為 690-810 nm，功率為 400-550 mW，脈衝寬度約為 100 fs。經 BBO 倍頻後，在 360-400 nm 之功率約為 15-45 mW，脈衝寬度約為 150-250 fs。

2. 建置完成一 frequency resolved optical gating 系統。

(B) 實驗結果方面:

1. 電子冷卻實驗：

   材料使用為摻矽之 n-type 氮化鎵材料。利用一紅外光飛秒脈衝加熱以存在之背景電子，並利用一紫外光飛秒脈衝作為探測光源，本計畫成功量測氮化鎵材料之電子冷卻時間約為 0.5ps，較砷化鎵系統為快。結合其他實驗資料[1]，本實驗亦顯示氮化鎵材料之載子冷卻為電子系統所主宰。發表論文請見計畫成果自評：論文(A)3、(B)I2。

2. 導帶谷間散射實驗：

   在上述電子冷卻實驗中，若紅外光能量大於導帶之第二能帶谷底與真正能帶谷底之能量差，則將發生谷間散射。利用可調變波長技術，本實驗發現氮化鎵材料之第二能帶谷底與真正能帶谷底之能量差，約為 1.34 eV，遠較砷化鎵系統為大。谷間散射所造成之冷卻延遲時間為 1ps，亦較砷化鎵系統為快。發表論文請見計畫成果自評：論文(A)3、(B)I2。

3. 帶尾載子動力學實驗：

   本實驗室利用 pump-probe 技術，在純氮化鎵材料中，發現氮化鎵材料之帶尾能態依能量高低而有截然不同之載子動力學表現。較近帶底之能態其載子動力學表現與能帶載子行為相同，而較遠帶底之帶尾能態則呈現 single-exponential 衰減。此行為是由於 mobility edge 低於帶底之故。發表論文請見計畫成果自評：論文(A)18、(B)I10、(B)I11。

4. 載子擴散行為實驗：

   本實驗室利用光學 pump-probe 技術，成功發展出量測二維載子擴散係數之量測方法，並應用於氮化銦鎵/氮化鎵量子井中之二維載子擴散係數量測。量測結果顯示，在氮化銦鎵/氮化鎵量子井中之二維載子(ambipolar)擴散係數極大，且隨量子井增後而急速增加。在 62Å 量子井中，其 ambipolar diffusion coefficient 為 $2700cm^2/s$，較 bulk 值高出至少 1000 倍以上。實驗顯示氮化銦鎵/氮化鎵量子井中，具有"giant ambipolar diffusion"之特性。此特性是由於量子井中巨大之壓電場所致。發表論文請見計畫成果自評：論文(A)19。

5. 同調聲學聲子震盪實驗：

發現氮化銦鎵/氮化鎵多重量子井中具有極強之同調聲學聲子震盪。此特性是由週期載子分布而偶合至聲子系統，並經由於量子井中巨大之壓電場所激發。其 dephasing 發現是由週期不確定性所決定。發表論文請見計畫成果自評：論文(A)4、(A)8、(A)15、(B)I5、(B)I7、(B)I8、(B)I13。

6. 聲子震盪之同調控制實驗

利用飛秒雷射脈衝，本實驗室首次完成聲子震盪之光學同調控制實驗。聲子震盪之大小與相位均可由脈衝控制而達成。發表論文請見計畫成果自評：論文(A)17、(B)I15。

7. 非線性光學性質研究

本計畫完成氮化鎵材料之雙光子吸收常數、非線性折射係數、二倍頻常數、與三倍頻常數之量測。並將雙光子吸收、二倍頻、與三倍頻技術應用於氮化鎵共焦顯微鏡。在雙光子吸收常數方面，發表論文請見計畫成果自評：論文(A)9、(A)11、(B)I4、(B)I6。在非線性折射係數方面，發表論文請見計畫成果自評：論文(A)6、(A)11 、(B)I6。在雙光子顯微技術方面，發表論文請見計畫成果自評：論文(A)5、(A)9、(A)11、(B)I12。在二倍頻與三倍頻顯微技術方面，發表論文請見計畫成果自評：論文(A)14、(A)16、(B)I14、(B)I17、(B)I21。

四、計畫成果自評

本計畫已完成當初預期之目標。在論文方面則已發表期刊論文共 14 篇，國際會議論文 22 篇，國內會議論文 15 篇，另有兩篇論文已被接受，兩篇論文已投稿尚在審查中。

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(B) 研討會論文

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