行政院國家科學委員會專題研究計畫成果報告

氮化鎵系列材料生長、製程及光電特性研究 – 總計畫 (I)
Growth, Process and Optical Properties Characterization of GaN-Series Materials (I)

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摘要：

本整合計畫結合了五位教授，針對氮化鎵系列材料之生長、製程技術及光電與材料特性分析進行研究。

主要研究成果及發表著作：

子計畫一：以分子束磊晶法成長氮化鎵系列材料(I)

主持人：林浩雄

In this study, the growth technologies of GSMBE and RF plasma atomic nitrogen source for wurtzite GaN, InGaN and $In_{0.062}Ga_{0.938}N$/GaN multiple quantum well (MQW) grown on (0001) sapphire substrates were studied. The main objective of this work is to optimize the growth conditions. For wurtzite GaN, the growth was preceded by the nitridation of the sapphire at substrate temperature 780°C, then a thin (~200 Å) GaN buffer layer was grown at 550°C. Subsequently, GaN bulk layer was overgrown at 760°C. During the growth, in situ RHEED measurement was used to monitor the growth condition. By adjusting the temperature of Ga cell to turn the RHEED pattern from (1×1) to (2×2), we could find a near stoichiometric condition for the growth. The InGaN/GaN MQW was grown on the sapphire (0001) substrate with the same initial procedures. However, after the growth of GaN bufferlayer, due to the volatility of In and the lower thermal stability of InN, a lower temperature of 600°C was used for the growth of the 10 pairs InGaN/GaN MQW.

The GaN sample shows high quality room temperature and low temperature PL spectra. At room temperature, there is only the near-band-edge transition at 3.41eV, and no yellow band or other broad band transitions are detectable in this measurement. Another study on the 10K PL reveals that the yellow band emission is below two thousands of the near-band-edge emission. These all indicate a good quality of the grown sample. There are two major transitions in the 10 K PL spectrum; one is corresponding to the bounding exciton peak at 3.48eV, the other may be due to the D-A transition at 3.44 eV. The 10K PL of the InGaN/GaN MQW shows a emission at ~2.7 eV. From its X-ray data with clear satellite peaks, the structure parameters were found as follows. The In composition of the InGaN wells is about 6.2%, and the thickness of InGaN and GaN are about 8 nm and 10.9 nm, respectively.

子計畫二:氮化鎵系列緩衝層生長及材料特性研究

主持人：張毅、馬廣仁

早期成長 GaN 薄膜大都著重於藉由改基板材料或中間緩衝層來降低 GaN 材料內缺陷;然最近幾年許多研究指出若在 GaN 材料製程中成長單層或多層的 InGaN 量子井結構，即使差排密度高，其發光強度及效率上仍有非常好的結果。目前已有一些研究指出此現象係因當 In 含量高於某個臨界值後 InGaN 材料會產生增幅分解反應(spinodal decomposition)而形成 InN 相分離，或產生成份變化之 InGaN 固溶相析出反應。這些組成的不均勻處形成局部的低位勢(potential minimum) ，將有助於捕獲載子形成激子而使發光效率改善。本計畫主要將藉由高解析穿透式電子顯微鏡(HRTEM)、X-ray 繞射及能譜分析(EDX)等儀器，研究以有機金屬化學氣相沈積法(MOCVD)成長 InGaN 多層量

子井(MQWs)之顯微結構，並探討 InN 相分離現象。

由穿透式電子顯微鏡明視野晶格影像可清晰觀察到 InGaN 量子井結構，並發現顆粒狀的析出物及不均勻的明暗區域分佈。由擇區繞射圖形(SAD)發現其呈現之繞射點皆分離成兩點以上，且 In 含量愈大其繞射點之分離現象將更顯著，由此即可判定發生相分離。當 In 含量較高時 (25%)，其 X 光繞射圖形除了出現 InN(0002)的繞射峰外，更可清楚的觀察到成份分布很廣的 $In_xGa_{1-x}N$ 的固溶相繞射峰，意味著 $In_xGa_{1-x}N$ 相中 In 組成極不均勻。當 In 含量較低(20%)時雖出現明顯相分離，但組成不均勻的增幅分解現象則較不明顯。所有在 InGaN 薄膜中之 In 濃度是依據 Vegard's 定理及沿(0002)方向之 InGaN 繞射峰相對於 GaN 繞射峰之偏移距離計算得之。利用能譜分析儀分析量子井附近 In 含量，發現組成在 2%-12%之間變化，且晶格影像中黑色區域 In 含量較周圍的亮區為高。以上這些結果都清楚說明了 InGaN/GaN 量子井結構中發生了明顯的相分離及組成不均勻的增幅分解現象，且 In 含量愈高這種現象愈明顯。如何藉由薄膜成長製程及熱處理來控制相分離或偏析物的尺寸大小、形狀及分佈，並探討其對光電性質的影響將是未來研究方向。

子計畫三：氮化鎵系列材料光學特性及製程技術 (I)

主持人：楊志忠、莊東榮

Due to the large lattice constant difference between GaN and InN, indium aggregation and phase separation have been discovered. Such indium composition fluctuations in InGaN compounds were supposed to be very crucial for efficient light emission. It was believed that such indium-rich nano-scale structures formed the localized states, similar to those of quantum dots. When the density of the indium-rich structures is higher than the defect density, the localized states can trap a significant amount of carriers for radiative recombination, leading to efficient light emission. Therefore, it was widely accepted that the measured photoluminescence (PL) in InGaN samples and hence the output from a light-emitting diode came from the recombination of localized excitons. Meanwhile, several stimulated emission (SE) studies have led to the conclusion that their measured SE also originated from band-filled localized states. Although other research groups reported non-localized state laser spectrum, it was believed that the gain in laser oscillation was strongly related to the structure of indium composition fluctuation. With such a crucial implication, indium composition fluctuations in InGaN/GaN quantum well structures have received special attention in material and optical studies.

Here, we report the results of our optical and material studies on InGaN/GaN multiple quantum well samples. Two SE spectral components were always observed with their relative intensity varied with temperature and pump fluence. Also, the spectral positions of the SE features, corresponding to the localized states, changed little with the two parameters.

發表之期刊論文：

Chi-Chih Liao, Shih-Wei Feng, C. C. Yang, Yen-Sheng Lin, Kung-Jen Ma, Chang-Cheng Chuo, Chia-Ming Lee, and Jen-Inn Chyi, "Stimulated Emission Study of InGaN/GaN Multiple Quantum Well Structures," *Applied Physics Letters*, January 17, 2000.