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 –
子計畫三：氮化鎵系列材料光學特性及製程技術 (I)
Optical Characterization and Process Techniques of GaN-Series Materials (I)

計畫編號：NSC 88-2215-E-002-040
執行期限：87/08/01 – 88/10/31
主持人：楊志忠　　　共同主持人：莊東榮
執行單位：國立台灣大學光電所

中文摘要：

本子計畫針對氮化鎵系列材料之光電特性進行分析及製程技術進行研究。我們發現氮化銦鎵量子井結構的激發放射有兩個互為消長之峰值，其變化隨溫度及激發強度而定。此現象乃歸因於該結構內之銦結塊及析出。另外，我們也首次以雷射來使 p 型氮化鎵活化。

英文摘要：

This branch project is responsible for analyzing the optical properties of GaN-based materials and developing novel process techniques for related device fabrication. We found that the stimulated emission of InGaN quantum well structures had two peaks with their relative intensity controlled by the temperature and pumping intensity. This phenomenon is attributed to the indium aggregation and phase separation in such a structure. Meanwhile, we have for the first time activated p-type GaN with laser illumination.

主要研究成果及發表著作：

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. The sample studied in this work was grown in a low-pressure metal-organic chemical vapor deposition reactor. The InGaN/GaN multiple quantum well (QW) structure consisted of five periods of silicon doped InGaN well with 3 nm in thickness. The nominal indium composition was 25 %. The silicon doping concentration was 10^{18} cm^{-3}. The barrier was 7-nm GaN. The QW layers were sandwiched with a 1.5-μm GaN buffer layer on a sapphire substrate and a 50-nm capping GaN layer. The growth temperatures were 1050 and 740 °C for GaN and InGaN, respectively.

The X-ray diffraction result of the sample, in which InN, GaN and broadened InGaN features was clearly seen. From this figure, one can see quite prominent composition fluctuation in the InGaN alloy. The SE spectral variation vs. temperature of the sample was observed from 13 K through room temperature (RT). The pump fluence was fixed at 32 mJ/cm^2. For clarity, we label the observed three peaks as A, B, and C from the long-wavelength side. The 13 K result corresponds to the highest peak A, the very left peak B, and the highest peak C. As temperature rises, the spectral position of peak A is almost unchanged and its intensity decreases. Also, peak B red-shifts and becomes stronger with increasing temperature. Meanwhile, peak C red-shits and becomes weaker with rising temperature. When temperature is lower than 200 K, there exists a feature between peaks B and C. The normalized PL spectra at 13 K and RT are compared. Peak A, which partially overlaps (on the high energy side) with the low-temperature (LT) and RT PL spectra, is supposed to come from carrier recombination of localized states. Peaks B and C are supposed to originate from free carrier recombination, corresponding to two different quantum well sub-bands. The origin of the feature between peaks B and C requires further investigation.

The red-shifts of PL peak and peak B with rising temperature are expected due to band-gap shrinkage. However, peak A position moves little with temperature. Meanwhile, one can see the decreasing and increasing trends of peaks A and B intensities, respectively, with temperature. It is believed that as temperature rises, carriers are thermally excited to move away from the localized states and may occupy free-carrier states (similar to the case of quantum dots). This leads to the decreasing trend of localized state recombination and the increasing trend of free-carrier recombination. The almost unchanged spectral position of localized state recombination (peak A) can also be interpreted for the same reason. Although the global band-gap of the material shrinks with temperature, as shown with the PL spectra, mobile carriers with thermal excitation result in almost the same spectral position for the maximum gain. Besides, the weaker strain at a higher temperature in the five quantum well structure, which leads to a blue-shift of band-gap, may contribute to cancel the global band-gap shrinkage.

The SE spectra of the sample with various 266 nm pump fluences at 13 K were also recorded for the highest pump fluence at 80 mJ/cm^2 and the lowest pump fluence at 36 mJ/cm^2. Both peaks A and B rise with increasing pump level. However, peak A seems to saturate at high pump fluences. At low pump levels, peak A from localized state recombination dominates the emission; however, at high pump levels, peak B from free-carrier recombination becomes dominant. The red-shift trend of peak B with increasing pump level, which is due to band gap re-normalization, was commonly observed. The almost unchanged spectral position of peak A can be attributed to the mutual cancellation between the blue shift, which is due to the filling-up of the low band-gap regions and the shielding of piezoelectric field (by strain) with increasing carrier density, and the red-shift, which is due to band-gap re-normalization. The small change of peak A intensity reflects the filling-up of the localized states. The increasing peak B intensity results from the combined effect of increasing carrier supply and the overflow of carriers from localized states as the pump level increases. Although a two-hump SE feature was also observed in an InGaN/GaN multiple quantum well structure, what reported was the dominance of the short-wavelength hump at low pump levels and the dominance of the long-wavelength hump (due to localized state recombination) at higher pump levels. This is in contrary to what we observed. Besides, their localized state feature was relatively sharper, compared with ours.

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.