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Optical Spectroscopic Investigation of InGaN/GaN Multiple Quantum Well Light Emitting Diode Wafers Grown on Sapphire by Metalorganic Chemical Vapor Deposition

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Abstract. InGaN/GaN multiple quantum well structures have been grown on sapphire substrates by metalorganic chemical vapor deposition, for wide range visible light emitting diode application. The compositions and sizes within quantum wells were designed according to the requirements on the LED performance. Samples were investigated by a variety of characterization techniques. Optimization of the growth parameters and process was realized and evidenced by high resolution X-ray diffraction measurements. Optical spectroscopic properties were further studied and quantum confined stokes shift was observed from room temperature photoluminescence and photoluminescence excitation measurements.

1. Introduction

Research and developments on GaN-based compound semiconductors and structures for optoelectronic and electronic applications have been very active in recent years. GaN and related III-nitride semiconductors possess large direct band gaps, large heterojunction offsets, and high thermal conductivity and have been well-developed in recent years for their materials growth and device manufactures in applications in blue-UV light emitting diode (LED), laser diode (LD) and other optoelectronic and electronic devices [1]. InGaN-based LEDs and LDs grown on sapphire are now available commercially. InGaN/GaN multiple quantum wells (MQWs) are attracting much research interest currently, acting as the active layer in above high brightness III-Nitride LED and cw blue-green laser diode LD. InGaN/GaN heterostructures can exhibit intense photoluminescence (PL) and electroluminescence (EL) despite of a high dislocation and defects density existed [2]. More research efforts have been made on InGaN/GaN MQW structures, among which, most of InGaN/GaN MQWs were prepared by the metalorganic chemical vapor deposition (MOCVD) technique, that has been approved a powerful technology for R&D and industry production of III-nitride materials and devices. Since mid-1990s, a turbo-disk technology has been employed in manufacturing MOCVD
systems on III-Nitrides. It uses a vertical growth configuration with a high speed rotation platter for multiple sample wafers. This paper reports on the optical spectroscopic investigation on the MOCVD-grown InGaN-GaN MQW blue-green LED structures on sapphire substrates. Analytical techniques of photoluminescence (PL), photoluminescence excitation (PLE), high-resolution (HR) X-ray diffraction (XRD) have been employed to investigate their optical and structural properties.

2. Experimental
High quality InGaN-GaN MQW LED wafers were grown on (0001)-plane sapphire substrates by low pressure (LP) MOCVD using several Emcore systems with the vertical growth configuration and a high speed rotation disk, i.e. the so-called Turbo-disc technology. Trimethylgallium (TMGa), Trimethylindium (TMIn), and ammonia NH₃ were used as precursors. In this study, typical experimental samples were investigated. Their structures were: (0001) sapphire substrate, 30 nm low temperature (520°C) grown GaN, 2 μm thick high temperature (HT) (1020°C) grown GaN, a 800 nm InGaN layer followed by 5 (or 8, or 10)-period InGaN-GaN MQWs grown at 800°C, 50 nm p-type AlGaN and 150 nm HT-grown GaN cap. The barrier material in MQWs was GaN with a thickness of 10-40 nm while the well was InGaN with thickness of 2-4 nm and varied In-composition. From sample-to-sample, there were different values of layer parameters. A Philips MRD HR-XRD including a five-crystal monochromator and a Renishaw micro-PL system equipped with a 325 nm He-Cd laser were used for characterization, measured at room temperature (RT). The low-T PL measurements were performed by a detection system with the 0.73 m Spex spectrometer-lock in amplifier-photomultiplier. RT PL and PLE measurements were also performed using a Jobin Yvon Fluorolog-3 system with a Xe-lamp as the excitation source.

3. Results
3.1 Room and Low Temperature Photoluminescence

![Fig. 1. Two RT PL spectra from MOCVD-grown InGaN-GaN MQWs.](image1)

![Fig. 2. T-varied (7-300 K) PL spectra from a MOCVD-grown InGaN-GaN MQW.](image2)
Figure 1 shows two typical RT PL spectra from MOCVD-grown green InGaN-GaN MQW LED samples containing five and ten QWs, respectively. The peak at 3.4 eV is from the GaN layers and the broad emissions in the range of 2.6-3.2 eV and with the peak near 2.9 eV are due to the InGaN/GaN MQW structures. The modulation of the broad emissions should be from the multiple reflectance induced interference. As the composition of InGaN well changes, different color of LED emissions from green to blue and to UV can be achieved.

Figure 2 shows temperature (T) varied (7-300 K) PL spectra from a MOCVD-grown blue InGaN-GaN MQW LED sample, consisting of five QWs. The major MQW emission band is located at 3.02 eV in 7-160 K and is shifted to 3.0 eV at RT. The emissions in the lower energy side of the major peak were developed with the increase of temperature.

3.2 High Resolution X-ray Diffraction

HR-XRD is a sensitive technology to characterize the fine structural features of MQW samples. Figure 3 shows the HR-XRD 2θ-ω scans of (0002), (0004) and (0006) GaN patterns, from two MOCVD-grown InGaN-GaN MQW samples on sapphire. One is for an initially grown sample with 5-QWs which exhibits poor XRD scanned patterns with only 1-2 satellite bands. The right figure shows the development from a 10-QW sample after the improvements of growth conditions. The 1\textsuperscript{st}-order pattern exhibits a broad GaN 2θ peak at 34.6° and only two MQW satellite bands while the 2\textsuperscript{nd}-order shows three satellite bands. The 3\textsuperscript{rd} order (0006) patterns exhibits five satellite bands with the n=0 observable. Indeed, this is from an old sample grown a few years ago.

![HR-XRD patterns](image)

**Fig. 3.** HR-XRD (0002), (0004) and (0006) patterns from two MOCVD-grown InGaN-GaN MQWs on sapphire, (left) 5-QWs in the initial growth and (right) 10-QWs from the improved growth.

The continual efforts have been made on further improvements of the MOCVD growth and parameter optimization for growth parameters, including growth temperature, pressure, V/III ratio, indium source flow, carrier gas flow etc. Good improvements have been realized. Figure 4 provides such evidence, where the GaN peak is very sharp and all satellite bands are narrow. More fine structures are seen between satellite (SL) peaks. These indicate the excellent layer crystalline perfection and sharp interfaces between all multiple layers. The computer simulation leads to the
precise determination of layer parameters of the thickness and composition, as shown in the right side of the table for this sample. The average InN mole fraction in the InGaN MQW is 15%. The well and barrier thicknesses are calculated to 4 and 41.5 nm, which nearly agrees with the transmission electron microscopy (TEM) results of 4 and 40nm. These also show that great improvements have been achieved on the MOCVD growth of high quality InGaN-GaN MQW LED wafers.

| Material | Thickness | Composition |
|----------|-----------|-------------|
| GaN      | 41.5nm    | In: 0%      |
| InGaN    | 4.0nm     | In: 15%     |

Fig. 4. HR-XRD pattern and simulation from a MOCVD-grown InGaN-GaN MQW on sapphire.

4. Discussion

Further studies and discussion on the optical properties of a high quality InGaN/GaN MQW blue LED wafer, with HR-XRD data shown in Fig. 4, are given in this section. The penetrating investigation may help us to better understand the luminescent mechanisms and find ways to further improve the material design and growth.

4.1 Temperature and Excitation Dependent Photoluminescence

Figure 5 shows the T-dependent (left) and excitation power dependent (right) PL spectra of this MOCVD-grown InGaN-GaN MQW (8-QWs) on sapphire. At 9K the broad peak at 2.686 eV is interpreted from the InGaN quantum well and the narrow peak at 3.478 eV from the GaN barrier.
The full width at half maximum (FWHM) of the QW peak for this LED structure is 177 meV at 9 K. Such large inhomogeneous broadening was due to the compositional fluctuation which is inherently present in the InGaN ternary system.

4.2 PL band shifts and quantum efficiency

Figure 6 shows the T-dependent PL peak shifts of both the GaN and QW emissions, and the integrated PL intensity of blue emission peak versus inverse temperature, obtained from Fig. 5. The T-dependent PL peak shifts for GaN band follow the variation of the GaN energy gap versus T. But the T-shifts of the QW band exhibit a special behavior: from 9 K to 150 K, it keeps almost a constant, and from 150-300 K, it decreases in energy only very slightly. This unique T-behavior of PL spectra differs from the previously reported phenomena, i.e. the so-called the S-shape variation [3-5]. This anomalous behavior can be attributed to the effect of the localized states caused by potential fluctuations. The activation energy, $E_A$, is deduced to be 80 meV from the slope of data in the temperature range of 140-300 K from Fig. 6. This large value of $E_A$ indicates the strong localization of the carriers in this MQW structure [6]. Assuming that the internal quantum efficiency equals unity at 9 K, we obtained the internal quantum efficiency as high as 34.8% at RT.

4.3 Stokes shift

Figure 7. Comparison of RT PL and PLE spectra from a MOCVD-grown InGaN-GaN MQW (8-QWs) sample.
Figure 7 shows the comparison of RT PL and PLE spectra on this MOCVD-grown InGaN-GaN MQW (8-QWs) sample. A large energy difference, so called Stocks shift (SS), between the band-edge absorption and emission was observed from this QW LED structure. Setting the turning point of PLE spectrum in figure 7 as the reference point [7-9], we obtained the Stocks shift as high as 216 meV, which is consistent with the large value of $E_A$ associated with the strong localization of the carriers in this MQW structure.

5. Conclusions

In conclusion, we have employed MOCVD to grow a series of InGaN/GaN MQWs blue-green LED structures on sapphire substrates. Analytical investigation by PL, PLE, HR-XRD has shown the excellent optical and structural properties. Detailed studies of luminescent properties have been performed via temperature and excitation dependent PL plus PLE measurements. Quantum efficiency was determined. The unique T-behavior of PL spectra is observed for the QW-related emission band, which is due to the quantum dot like structure features within the MQW structures. Quantum confined Stokes effect was observed from the comparison of PL and PLE measurements. A further penetrating investigation is helpful to better understand the luminescent mechanisms and find ways to further improve the material design and growth of InGaN/GaN MQWs for the wide spectral LED applications.

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