Nano-structures and luminescence mechanisms of InGaN/GaN multiple quantum well light emitting diodes

By

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Abstract:

A series of InGaN/GaN MQW LEDs were prepared by low pressure metalorganic chemical vapor deposition (MOCVD) and studied on the nano-structural features correlated with optical properties, and luminescence emission mechanisms by analytical techniques of photoluminescence (PL), PL excitation (PLE), time resolved PL (TRPL), high-resolution (HR) X-ray diffraction (XRD) and HR transmission electron microscopy (TEM). They have shown the excellent optical and structural properties, evidenced by HRXRD, HRTEM and optical measurements. The quantum dot like structure features, unique T-behaviors of PL spectra, quantum confined Stokes effect, TRPL exploration with the variation of detecting energy and temperature and modeling analyses are studied and discussed.

Keywords:

InGaN/GaN, MQW, LED, MOCVD, XRD, TEM, photoluminescence, time-resolved

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1. Introduction:

Energy-efficient and environmentally friendly solid-state light sources, in particular GaN-based light emitting diodes (LEDs), are currently revolutionizing an increasing number of applications, and bring apparent benefits to vast areas of development, such as lighting, communications, biotechnology, imaging, and medicine. It is expecting that LEDs may replace traditional light bulbs and tubes to achieve a new lighting era [1-3].

InGaN/GaN multiple quantum wells are the key components of these commercial devices emitting UV-green and white light, acting as the active layer, which can exhibit intense luminescence despite of a high dislocation and defects density existed. However, despite an impressive commercial success, the mechanism of luminescence from InGaN/GaN is not yet well-understood and the physical origin of efficient light generation is unveiled incompletely. More active researches are carrying on this scientific issue [3-14].

In this paper, we study the nano-structural features and correlation with optical properties and emission mechanisms in InGaN/GaN MQW LEDs, prepared by metalorganic chemical vapor deposition (MOCVD).

2. Experiment:

High quality InGaN/GaN MQW LEDs were grown on (0001)-plane (c-face) sapphire substrate by low pressure MOCVD. Trimethylgallium (TMGa), Trimethyl-lindium (TMIn), and ammonia NH₃ were used as precursors for Ga, In, and N, with carrier gas of H₂ and N₂, respectively. The substrates were initially treated in H₂ at 1173°C, and 30 nm thick GaN buffer layer was grown on sapphire at 520°C. Followed, 2 µm thick GaN was grown at 1020°C, and 800 nm InGaN layer and 5-8 periods of InGaN/GaN QWs were grown at 800°C. Different designed MQW structures lead to different color LEDs. Analytical techniques of photoluminescence (PL), PL excitation (PLE), high-resolution (HR) X-ray diffraction (XRD) and HR transmission electron microscopy (TEM) have been used to investigate these LEDs.
3. Results and Discussion:

Figure 1 shows a HRTEM image from a MOCVD-grown InGaN-GaN MQW LEDs. The interfaces between GaN and InGaN layers in QWs are abrupt. The In-rich precipitates can be seen, which are caused by alloy fluctuation appeared in the regions with a dark contrast. The HR image shows no threading dislocation at this area, but some strain field around the well.

*Figure 1. HRTEM images from an InGaN- MQW LED (scale: 50-nm).*

Figure 2 shows the TEM digital analysis of lattice images (DALI), obtained from cross-section HRTEM. To avoid specimen degradation during electron-beam irradiation, the exposure time before image recording was kept smaller than 1 min. They exhibit the color-coded map of the local In-concentration inside QW structure, and the inhomogeneity of the QW layer, thickness, indium composition and well-to-well. The quantum dot (QD) - like structures around the In-rich areas are formed, which is the cause of strong luminescence from InGaN/GaN MQWs [6].

*Fig. 2. TEM DALI images of an InGaN MQW with left pattern from QW1 just next to the capping layer and the right one is from QW5 at the bottom of the active layer.*

Figure 3 exhibits a HRTEM from another LED on a QW area, with the In-clustering seen clearly. These nano-structural or QDs features are closely correlated with results from HRXRD, PL/PLE. Figure 4 shows a XRD pattern for an InGaN MQW with 8-QWs, with multiple narrow satellite bands.

*Fig. 3. HRTEM image from an InGaN MQW LED (scale: 2-nm).*
More fine structures are seen between satellite peaks in Fig. 4, characterizing the multi-layer thickness flatness variation less than 1-nm. The simulation gives precisely layer parameters of thickness and composition.

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

Figure 5 shows the temperature (T)-dependent PL spectra of this MQW. The MQW emission peak seems to vary little with T, quite unusual.

**Fig. 5.** Temperature dependent PL spectra for the InGaN/GaN 8-QWs structure from 9 to 300 K.

Figure 6 shows the comparison of RT PL and PLE. A large energy shift of 216-meV between PL and band-edge absorption was observed, due to the quantum confined Stocks shift (QCSS) effect [4, 5]. Our PLE-QCSS was measured at RT, in contrast to others below 20 K [4], indicating the high quality of our MQWs with excellent characteristics.

**Fig. 6.** Comparison of RT PL and PLE spectra from a MOCVD-grown InGaN/GaN MQW (8-QWs) sample.
There appears a hot discussion on the mechanism or origin of luminescence from InGaN/GaN [1, 5, 7, 8]. C. J. Humphreys and co-authors have suggested [7] that the contrast observed in many HRTEM images is to a large extent a consequence of exposure to the electron beam and is not due to the presence of In-clustering, that strong luminescence from the carrier localization is possible due to monolayer well width fluctuations with a lateral scale of about 2 nm. They also postulate [8], by observing the gross well-width fluctuations in single InGaNGaN QWs and interlinking strips of InGaN with In-rich centers, that excitons are localized at these In-rich regions preventing them reaching threading dislocations.

Apparently, our work provides new insights of physics on the luminescence mechanism of InGaN/GaN MQWs. Our HRXRD data showed fine oscillation corresponding to atomic layer variation of ~3Å, which might question on the mechanisms from Humphreys et al [6,7].

4. Conclusions:

In conclusion, a series of InGaN/GaN MQW LEDs have been grown by low pressure metalorganic chemical vapor deposition (MOCVD). They were studied on the nano-structural features correlated with optical properties, and luminescence emission mechanisms by various analytical techniques, including photoluminescence (PL), PL excitation (PLE), time resolved PL (TRPL), high-resolution (HR) X-ray diffraction (XRD) and HR transmission electron microscopy (TEM). These LED wafers have shown the excellent optical and structural properties, evidenced by HRXRD with high order (some up to 10th order) QW XRD satellite peaks and fine fringes and by HRTEM with sharp MQW structures and V-shaped defects. The quantum dot like structure features are revealed to exist within the MQW structures, which leads to unique T-behaviors of PL spectra. Quantum confined Stokes effect was observed from the comparison of PL and PLE measurements, even at room temperature (RT). TRPL exploration with the variation of detecting energy and temperature and modeling analyses together with above other analytical investigation have provided new insights of physics on the luminescence mechanism of InGaN/GaN MQWs.

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