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Growth of InGaN/GaN Multiple Quantum Wells by Metalorganic Chemical Vapor Deposition and Their Structural and Optoelectronic Properties

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Abstract

InGaN/GaN multiple quantum wells (MQWs) were grown by metalorganic chemical vapor deposition and their structural and optical properties were studied. When the average In content was increased by increasing TMIn flow rate, PL measurement showed little change in PL peak position and large increase in PL intensity instead. Large changes in PL peak position could be achieved by changing growth temperature. We propose the formation of fixed In content, highly In-rich quantum dot-like phases in InGaN MQWs driven by spinodal decomposition.

1. Introduction

Nitride semiconductors have become indispensable materials of choice for visible-to-ultraviolet optical devices and high-power and high-frequency devices. Especially, for optical applications, the successful growth of InGaN/GaN multiple quantum well (MQW) structures for active layers are pivotal in realizing high-brightness blue and green light emitting diodes [1]. It is widely accepted that the high luminescence efficiency of the InGaN/GaN MQW is due to the carrier localization induced by compositional fluctuation of InGaN layers [2]. However, its detailed mechanism is still a controversal issue. Moreover, the high equilibrium vapor pressure of In makes it very difficult to incorporate high In content in InGaN layers, especially at high temperatures [3]. It is very likely that the use of preheated ammonia would provide sufficient nitrogen overpressures to keep InGaN from decomposing. In this paper, we report the successful growth of InGaN/GaN double heterostructures (DHs) and MQWs grown by metalorganic chemical vapor deposition (MOCVD) with preheated ammonia. We studied the optical properties of InGaN/GaN DHs and MQWs and the luminescence mechanism in InGaN alloys.

2. Experimental and Simulation Details

InGaN/GaN DHs and MQWs were grown by an MOCVD system (HR-21SC, Hanvac Co., Ltd.) equiped with an ammonia preheater. Trimethylgallium (TMGa), trimethylindium (TMIn) and ammonia were used as Ga, In and N sources, respectively. The details of the MOCVD system and the growth procedure and properties of GaN epilayers were described in our previous report [4].

The samples were grown on (0001) oriented sapphire substrates. The structures consist of a thick (2000 nm) buffer GaN layer, an active region, and a 70 nm thick

GaN cap layer. In the case of InGaN/GaN DH samples, thickness of the InGaN active layer was about 200 nm. The active region of InGaN/GaN MQW samples consists of 7 pairs of 3 nm InGaN well layer and 7 nm GaN barrier layer. During the growth of the active region of samples, growth temperature and input TMIn carrier gas flow rate were varied at ranges of 760~800℃ and 60~180 sccm, respectively. Other growth parameters such as reactor pressure, input ammonia flow rate and input TMGa carrier gas flow rate, ammonia preheating temperature were kept constant at 300 Torr, 3.6 slm, 2 sccm and 500 ℃, respectively. Growth conditions for thick GaN buffer layers and GaN cap layers were also fixed for all samples. Average In contents in InGaN layers were estimated by high resolution x-ray diffraction (HR-XRD). Optical properties of the samples were analyzed by photoluminescence (PL). Thickness of film was measured by scanning electron microscope (SEM), transmission electron microscope (TEM) and HR-XRD.

3. Results and Discussion

We could control the In incorporation by tuning two growth parameters; TMIn flow rate and growth temperature. When growth temperature was raised from 760 to 800 ℃ at a fixed TMIn input flow rate of 60 sccm, the average In content in the InGaN/GaN DH samples decreased from 25% InN to 13% InN. When TMIn input flow was increased from 60 sccm to 120 sccm at 800 ℃, the average In content in the InGaN DH samples increased from 13% InN to 17% InN. This result is in good agreement with the previous report [3].

InGaN/GaN MQWs were fabricated and their properties were analyzed. Fig. 1 shows PL spectra of an InGaN/GaN MQW sample and an InGaN/GaN DH sample grown at the same growth conditions. Emission intensity from the InGaN/GaN MQW sample was about 60 times larger than that from that from the InGaN/GaN DH sample, which indicates that our MQW structure was of high quality. Structural properties of our InGaN/GaN MQW sample were analyzed. Fig. 2 shows HR-XRD pattern of our InGaN/GaN MQW sample. Satellite peaks from InGaN/GaN MQW structure could be found in Fig. 2, which indicates the periodicity of InGaN wells and GaN barriers in our InGaN/GaN MQW sample. In TEM analysis, InGaN/GaN MQW structure with periodic configration could also be observed.

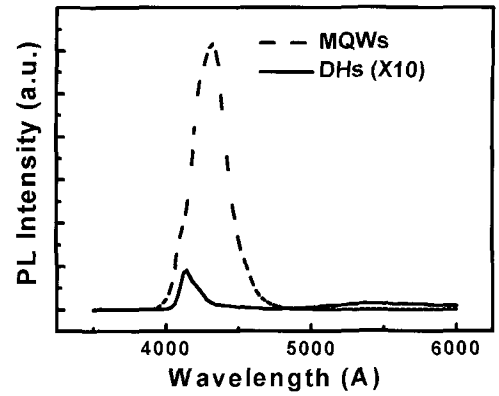

Fig. 1. PL spectra of InGaN/GaN DH and MQW grown at the same condition.

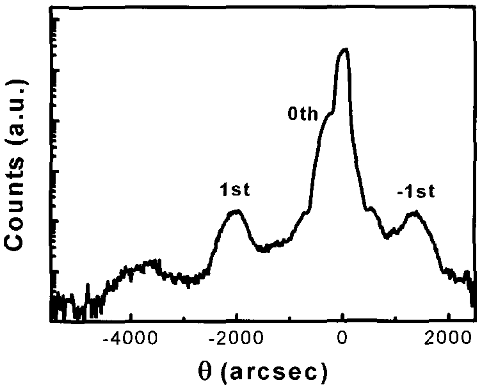

Fig. 2. HR-XRD pattern of the InGaN/GaN MQW.

PL properties of the samples with different average In contents were analyzed. At first, samples with different average In contents were obtained by varying TMIn flow rate during growth. Fig. 3 shows PL spectra of the InGaN/GaN MQW samples grown at different TMIn flow rates. When average In content

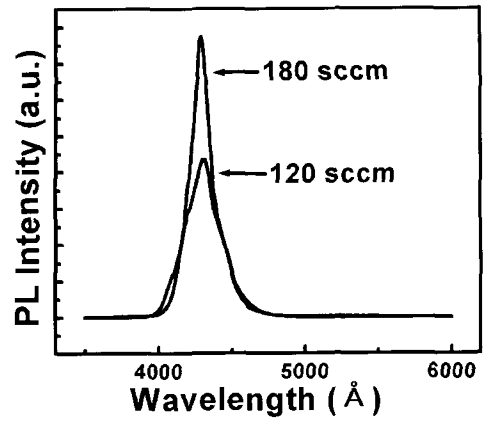

Fig. 3. PL spectra of InGaN/GaN MQWs grown with different TMIn flow rates.

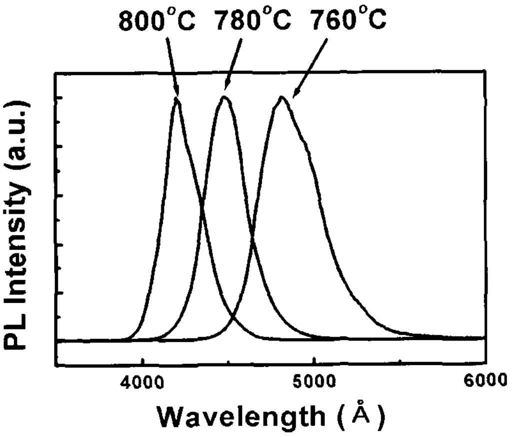

Fig. 4. PL spectra of InGaN/GaN MQWs grown at different temperatures (normalized).

was increased by increasing TMIn flow rate from 120 sccm to 180 sccm, however, in this case little change in emission peak position was observed. In contrast, about two-fold increase of PL intensity was observed at the same time. Similar results could be obtained in PL measurements of the InGaN/GaN DH samples.

Then, the samples with different average In contents were obtained by varying growth temperature. Fig. 4 shows PL spectra of the InGaN/GaN MQW samples grown at different temperatures. Here, the TMIn flow rate was kept constant at 120 sccm. As shown in Fig. 2, large peak shift ranging from 430 nm to 490 nm could be obtained when growth temperature was decreased slightly from 800 to 760 ℃. Similar results could be obtained in PL measurements of the InGaN/GaN DH samples grown at different temperatures.

Large PL peak shift could be obtained only by changing growth temperature, whereas the increase in TMIn flow rate resulted in increase in PL intensity. From these results, we can deduce that there is no direct relation between average In contents and PL peak positions in both InGaN/GaN DHs and MQWs. It seems to be due to the carrier localization effect induced by the formation of highly In-rich, quantum dot-like phases. Ho and Stringfellow predicted the spinodal decomposition in InGaN alloys induced by the existence of immiscibility gap in InN-GaN system [6]. Following their prediction, highly In-rich InGaN nano-phase would precipitate by spinodal decomposition in InGaN alloy. The precipitated highly In-rich InGaN nano-phases would act as quantum dot-like structures and form carrier localization centers. O'Donnell et al. proposed the existence of InN-like quantum discs in InGaN light emitting diodes [8]. In this case, increased In incorporation into InGaN alloy would result in increase in volume fraction of highly In-rich InGaN nano-phase, and the number of carrier localization centers for radiative recombination, leading to the increase in PL intensity from InGaN without changing PL peak position.

In contrast, when In incorporation into InGaN sample was increased by decreasing growth temperature, the In content of highly In-rich nano-phase would increase. Changes in growth temperature might also affect the kinetics of spinodal decomposition in InGaN, leading to changes in the microstructure of InGaN and changes in localization state and eventually the large PL peak shift.

4. Conclusions

In summary, InGaN/GaN DHs and MQWs were

grown by MOCVD. Especially, InGaN/GaN MQWs were successfully fabricated to show enhanced optical properties. Average In contents in InGaN samples were controlled by changing two growth parameters; growth temperature and input TMIn carrier gas flow rate. However, when average In content was increased by increasing TMIn flow rate, PL measurement showed little change in PL peak position and large increase in PL intensity. Large changes in PL peak position could be achieved by changing growth temperature.

Negligible change in PL peak position with increased average In content in InGaN can be attributed to the formation of fixed In content, highly In-rich quantum dot-like phases driven by spinodal decomposition. In contrast, increases in average In content might lead to the increase in volume fraction of highly In-rich quantum dot-like phases, resulting in increase in PL intensity. In this point of view, our observation strongly suggests that the carrier localization by highly In-rich quantum dot-like phase in InGaN is the major mechanism for PL in InGaN.

Acknowledgements

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