UC Santa Barbara

UC Santa Barbara Previously Published Works

Title

Effects of Growth Temperature and Postgrowth Annealing on Inhomogeneous Luminescence Characteristics of Green-Emitting InGaN Films

Permalink

<https://escholarship.org/uc/item/4rd8k791>

Journal

Journal of Electronic Materials, 39(1)

ISSN

1543-186X

Authors

Masui, Hisashi
Melo, Thiago
Sonoda, Junichi
[et al.](#)

Publication Date

2010

DOI

10.1007/s11664-009-0969-y

Peer reviewed

Effects of Growth Temperature and Postgrowth Annealing on Inhomogeneous Luminescence Characteristics of Green-Emitting InGaN Films

HISASHI MASUI,^{1,2} THIAGO MELO,¹ JUNICHI SONODA,¹
CLAUDE WEISBUCH,¹ SHUJI NAKAMURA,¹
and STEVEN P. DENBAARS¹

1.—Solid State Lighting and Energy Center, Materials Department, College of Engineering, University of California, Santa Barbara, CA 93106-5055, USA. 2.—e-mail: masui@engineering.ucsb.edu

Microscopic photoluminescence was applied to investigate μm -order inhomogeneity of InGaN alloys. Samples had InGaN/GaN multiple-quantum-well structures grown on sapphire substrates at various temperatures, and luminescence was adjusted to be green. Luminescence morphologies of dendritic appearance were observed on as-grown samples. Bright spots luminescing at long wavelengths (green to amber) were formed at high growth temperatures. After annealing at 1000°C , the bright spots disappeared and the dendritic morphology turned into a granular morphology. Because of these μm -order inhomogeneities, it has been suggested that small-scale characterization (sub- μm or smaller) requires special attention in order not to miss effects of μm -order inhomogeneity in InGaN alloys.

Key words: InGaN, light-emitting devices, photoluminescence, dendrite-like morphology

INTRODUCTION

The foundation of solid-state lighting technology has been served by two semiconductor material systems: AlInGaP and AlInGaN. AlInGaP currently covers the infrared-amber spectral range. AlInGaN is responsible for the ultraviolet-green range; however, luminescence efficiency in the green is relatively low. InGaN-based green emitters commonly exhibit significant blue shifts in luminescence as excitation intensity (i.e., device operation current) increases. Low efficiency and the spectral blue shift in the green range have been attributed to the following nature of the InGaN quantum-well (QW) structure grown on *c*-plane GaN layers. The strained QW structure induces the quantum-confined Stark effect, which may be suppressed by selecting nonpolar and semipolar orientations. InGaN alloys experience spinodal decomposition,

which results in alloy phase separation forming In-rich domains in the low-In matrix and causes inhomogeneity of luminescence. Alloy phase separation is related to thermodynamics and material growth kinetics.

InGaN phase separation has been investigated via techniques such as transmission electron microscopy,^{1–4} scanning near-field optical microscopy,^{5,6} microphotoluminescence,^{7,8} and cathode luminescence.⁹ These techniques are mostly intended to resolve the smallest possible features of alloy inhomogeneity, while large-scale features (on the order of μm or larger) are often unexamined. Additionally, high-performance optoelectronics devices are ultimately aimed at utilizing InGaN luminescence. For example, laser diodes employ contact stripes of a few μm wide to excite gain materials, where the gain distribution in the gain material becomes a question. Hence, it is of interest and importance to investigate InGaN luminescence characteristics (inhomogeneity in particular) on the optical scale in the vicinity of μm . With these issues

(Received March 16, 2009; accepted October 6, 2009; published online November 3, 2009)

in mind, we have employed an optical microscope to “look at” InGaN luminescence directly and characterize the inhomogeneity of InGaN films, rather than indirectly generating luminescence maps or images.

EXPERIMENTAL PROCEDURES

The experiments were designed to evaluate the effects of growth temperature (T_g) and postgrowth heating (to mimic thermal processes in device fabrication) on luminescence characteristics. Seven InGaN/GaN multiple-QW (MQW) samples were grown via metalorganic chemical vapor deposition on 2-inch *c*-plane sapphire wafers at various T_g values between 780°C and 855°C. The MQW consisted of six 3-nm-thick InGaN layers separated by GaN barrier layers. The MQW structure was grown in N_2 carrier gas on a Si-doped GaN layer (4 μm thick) and capped with a GaN barrier layer on top. T_g was monitored via a thermocouple placed in the graphite wafer holder. The temperature of the wafer surface typically appears 50°C to 100°C lower than T_g in our growth reactor. InGaN luminescence was made to be green by adjusting the In precursor (trimethylindium) supply rate while the Ga precursor supply rate was maintained constant. During the growth, InGaN layers did not experience any temperature higher than their T_g . Apart from these MQW samples, another sample that had a complete light-emitting diode (LED) structure was prepared. The LED structure consisted of an identical MQW stack ($T_g = 845^\circ\text{C}$) followed by a Mg-doped *p*-type GaN layer (grown at 880°C for 10 min) without removal from the growth chamber. The LED wafer was reheated (700°C for 15 min) for Mg activation after removal from the growth chamber.

Luminescence characteristics were evaluated via a fluorescence microscope.^{10,11} The standard V-2A filter cube selects the wavelength range (380 nm to 420 nm) of illumination generated by a metal halide lamp. With a 100 $\times$ objective lens (numerical aperture 0.90), the illumination intensity was 7 W/cm² at the sample. The optical resolution of the microscope was estimated to be 0.4 μm . For repetitive observations of samples (see the “[Annealed MQW Samples: Effects of Postgrowth Heating](#)” section), the area near the center of the sample pieces was selected for characterization. Position errors during repeated observations was within a 0.5 mm $\times$ 0.5 mm area. The viewing field of the microscope was 200 μm in diameter, which provided an area of photomicrograph images of 60 μm $\times$ 80 μm . A representative area was cropped from each image for presentation purposes; otherwise, the captured images were intact to avoid artificial photographic manipulation. The exposure time of image capture was varied. Luminescence spectra were recorded via a compact spectrometer combined with an optical fiber equipped on the microscope. The integration time of spectrum capture was 1 s.

The experimental procedure was as follows:

1. As-grown MQW sample wafers were characterized via the fluorescence microscope. Bright-field (BF) imaging was also performed on the microscope. Results are presented in the “[As-Grown MQW Samples: Effects of Growth Temperature](#)” section.
2. Two of the MQW sample wafers (with the highest and lowest T_g) were cut into pieces (approximately 5 mm $\times$ 5 mm). Each sample piece was characterized via the microscope. These sample pieces were placed in a rapid thermal annealer and annealed in a N_2 flow at various annealing temperatures (T_a) for 1 min. T_a was monitored via a thermocouple placed on the sample holder. The ascent time was approximately 1 min while the descent time varied from 3 min to 10 min depending on T_a . After characterization of the 1-min annealed samples, sample pieces were subjected to an additional 6-min annealing at the same temperatures as before. The sample pieces were characterized again. Results are discussed in the “[Annealed MQW Samples: Effects of Postgrowth Heating](#)” section. Finally, the photoluminescence morphology of the LED sample was characterized; the result is described in the “[Annealed MQW Samples: Effects of Postgrowth Heating](#)” section.

RESULTS AND DISCUSSION

As-Grown MQW Samples: Effects of Growth Temperature

Figure 1 compares the luminescence of the seven samples. Luminescence morphology generally consisted of two parts: bright spots and background with dendritic appearance. Bright spots were developed at high T_g (above 815°C). As T_g became higher, (1) the number of bright spots was increased, (2) the bright spots started losing spatial correlation with the background morphology (as if the bright spots were randomly formed, above 845°C), and (3) the bright spots appeared to be yellow and amber (above 847°C). These bright spots luminescing yellow and amber are surmised to be In-rich domains: similar long-wavelength spots have been observed via microphotoluminescence.⁷ The strongest luminescence was recorded for $T_g = 845^\circ\text{C}$, where a large number of bright spots maintained green luminescence. Bright spots were not always small spots, but regions surrounded by bright peripheries (white circles in Fig. 1 indicate examples); we were not able to identify whether the bright peripheries were physically connected In-rich domains or caused by the effects of optical interference between discrete domains. The dendrite-like morphology (collections of luminescing curved stripes, represented well by images for $T_g = 780^\circ\text{C}$ and $T_g = 840^\circ\text{C}$) was an unexpected observation: we had imagined that luminescence would be more

Fig. 1. Luminescence from MQW samples. The luminescence morphologies appearing like collections of luminescing curved stripes (seen in the entire images of $T_g = 780^\circ\text{C}$ and $T_g = 840^\circ\text{C}$, for example) are called dendritic morphologies in the text. Each image has a $30\ \mu\text{m} \times 30\ \mu\text{m}$ area. Spectra are shown at the top right corner. The duration shown at the bottom of each image is the exposure time (online color only).

uniform over the area of observation. The morphology was likely generated by miscible InGa_N with relatively low In content, but no concrete evidence for dendritic growth has been obtained. The background became dark as T_g was increased above 850°C , and dendritic morphologies were barely observed.

It is crucial to realize that the dimensions of the bright spots and dendritic morphologies were on the order of μm . It was also evident that the luminescence intensity varied significantly over areas of tens of μm (e.g., cases of $T_g = 850^\circ\text{C}$ and $T_g = 855^\circ\text{C}$). These in-plane luminescence inhomogeneities are strongly influenced by growth temperature; photoluminescence spectral measurements are insufficient to reveal morphological changes. Broad spectral peaks are typically attributed to “inferior” materials, but they can be associated with different luminescence morphologies, as in the case of $T_g = 780^\circ\text{C}$ and $T_g = 855^\circ\text{C}$, and potentially with different alloy phases. These results lead to the statement that, when small-scale characterization (sub- μm or smaller) is performed on InGa_N alloys, it is essential to have information on the thermodynamic states of the InGa_N alloys (related to growth conditions) and the location on the alloy being characterized (as a small drift may change luminescence from a bright spot to a dark gap in the dendritic morphology).

BF microscopic observation is insufficient to find the dendritic morphologies. Figure 2 compares BF and luminescence images of the same location on the MQW sample grown at 840°C . The bright spots were recognizable in the BF image as dark spots as a result of high optical absorption (low reflection),

Fig. 2. Photomicrographs comparing photoluminescence (PL) and bright-field (BF) images of the same location on the MQW sample. $T_g = 840^\circ\text{C}$. The area is $30\ \mu\text{m} \times 30\ \mu\text{m}$ (online color only).

which was caused by the smaller bandgap of the In-rich domains. BF images showed a faint trace of dendritic luminescence morphologies, but an observer would be unlikely to find such a trace unless guided by a luminescence image showing dendritic morphologies.

Annealed MQW Samples: Effects of Postgrowth Heating

The results of luminescence characterization on the samples grown at 855°C and 780°C are summarized in Figs. 3 and 4, respectively, where changes in luminescence morphologies caused by high T_a are found. Dendritic morphologies were barely affected by annealing in both samples (Figs. 3 and 4), indicating that miscible InGa_N matrices were reasonably stable. An exception in both samples was the 7-min annealing at 1000°C , for which the

Fig. 3. Luminescence characteristics of the InGaN MQW sample grown at 855°C. T_a is shown on the left. The annealing duration is shown at the top. The exposure time is shown in each image. Each image has a $15\ \mu\text{m} \times 15\ \mu\text{m}$ area. Luminescence spectra are shown on the right (luminescence intensity on the ordinate, in arbitrary units) (online color only).

dendritic morphology turned into a granular morphology. Bright spots were barely affected by annealing for $T_a = T_g$ (note that T_g and T_a are thermocouple readout temperatures) in Fig. 3, while they merged into the background for $T_a = 1000^\circ\text{C}$. The tendency for bright spots to disappear was also confirmed at intermediate temperature, $T_a = 955^\circ\text{C}$, over 1 min. The luminescence spectra indicated that the $T_a = T_g$ condition in general generated a broad spectra with an intense long-wavelength luminescence component ($\sim 530\ \text{nm}$) relative to the short-wavelength component ($\sim 500\ \text{nm}$) due to phase separation. Higher T_a tended to enhance the short-wavelength component, resulting in blue-shifted spectra. Well-barrier intermixing⁸ was proposed as the reason for the blue shift, in addition to InN thermal dissociation.

Despite the observations of bright spots and dendritic morphologies in the MQW samples, we noticed that the photoluminescence morphologies of LED structures typically appear granular.¹¹ Figure 5 shows an example of the granular luminescence morphology of the LED wafer along with that of the corresponding MQW sample ($T_g = 845^\circ\text{C}$). It is apparent that postgrowth heating changes the InGaN luminescence morphologies and that granular morphologies are developed after

sufficient thermal treatment. Although the granular morphology may be considered to be reasonably uniform over a large scale (sub-mm), e.g., for LED chip fabrication, luminescence inhomogeneity still deserves attention in small-scale characterization of laser diode fabrication. Postgrowth thermal treatment induces changes in luminescence inhomogeneity; this fact also requires special attention when performing luminescence characterization.

CONCLUSIONS

Inhomogeneity of photoluminescence morphology was studied on green-luminescing InGaN MQW samples prepared at various temperatures. Luminescence morphologies clearly indicated μm -order inhomogeneities which were represented by bright spots and morphologies with dendrite-like appearance. Postgrowth heating of the samples induced changes in luminescence morphology and spectrum; sufficient thermal treatment (7 min at 1000°C and longer/higher) turned the dendritic morphologies into granular morphologies, which are typically found in luminescence morphologies of InGaN-based LED structures. Selection of postgrowth process temperatures in relation to T_g is crucial to InGaN luminescence characteristics, especially

Fig. 4. Luminescence characteristics of InGaN MQW sample grown at 780°C. T_a is shown on the left. The annealing duration is shown at the top. The exposure time is shown in each image. Each image has a $15 \mu\text{m} \times 15 \mu\text{m}$ area. Luminescence spectra are shown on the right (luminescence intensity on the ordinate, in arbitrary units) (online color only).

Fig. 5. Comparison of photoluminescence morphology between the MQW and LED samples. The MQW stack was grown identically at $T_g = 845^\circ\text{C}$ for both samples. Each image has a $30 \mu\text{m} \times 30 \mu\text{m}$ area. The exposure time was 6 ms (online color only).

near 1000°C. It is suggested that, for small-scale (sub- μm or smaller) characterization of InGaN alloys, it is essential to acquire information on the thermodynamic states of the alloys (growth conditions and postgrowth treatments) and the location on the alloy being characterized (because of μm -order alloy inhomogeneity).

OPEN ACCESS

This article is distributed under the terms of the Creative Commons Attribution Noncommercial License which permits any noncommercial use, distribution, and reproduction in any medium, provided the original author(s) and source are credited.

REFERENCES

1. L.T. Romano, M.D. McCluskey, B.S. Krusor, D.P. Bour, C. Chua, S. Brennan, and K.M. Yu, *J. Cryst. Growth* 189/190, 33 (1998).
2. Q. Wang, T. Wang, J. Bai, A.G. Cullis, P.J. Parbrook, and F. Ranalli, *Appl. Phys. Lett.* 93, 081915 (2008).
3. Y.-C. Cheng, S.-W. Feng, C.C. Yang, C.-T. Kuo, J.-S. Tsang, S. Jursėnas, S. Miasojedovas, and A. Žukauskas, *Lithuanian J. Phys.* 46, 311 (2006).
4. Y.-Y. Chung, Y.-S. Lin, S.-W. Feng, Y.-C. Cheng, E.-C. Lin, C.C. Yang, K.-J. Ma, C. Hsu, H.-W. Chuang, C.-T. Kuo, and J.-S. Tsang, *J. Appl. Phys.* 93, 9693 (2003).
5. A. Kaneta, K. Okamoto, Sg. Fujita, G. Marutsuki, Y. Narukawa, and T. Mukai, *Appl. Phys. Lett.* 81, 4353 (2002).
6. H. Itoh, S. Watanabe, M. Goto, N. Yamada, M. Misra, A.Y. Kim, and S.A. Stockman, *Jpn. J. Appl. Phys.* 42, L1244 (2003).
7. J. Danhof, C. Vierheilig, U.T. Schwarz, T. Meyer, M. Peter, B. Hahn, M. Maier, and J. Wagner, *Phys. Status Solidi C* 6, S747 (2009).
8. D. Queren, M. Schillgalies, A. Avramescu, G. Brüderl, A. Laubsch, S. Lutgen, and U. Strauß, *J. Cryst. Growth* 311, 2933 (2009).
9. S.-W. Feng, T.-Y. Tang, Y.-C. Lu, S.-J. Liu, E.-C. Lin, C.C. Yang, K.-J. Ma, C.-H. Shen, L.C. Chen, K.H. Kim, J.Y. Lin, and H.X. Jiang, *J. Appl. Phys.* 95, 5388 (2004).
10. H. Masui, H. Yamada, K. Iso, H. Hirasawa, N.N. Fellows, J.S. Speck, S. Nakamura, and S.P. DenBaars, *Phys. Status Solidi A* 205, 1203 (2008).
11. H. Masui, S. Nakamura, and S.P. DenBaars, *Semicond. Sci. Technol.* 23, 085018 (2008).