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Emission studies of InGaN layers and LEDs grown by plasma-assisted MBE

Pekka Laukkanen[a,*], Sami Lehkonen[a], Petteri Uusimaa[a], Markus Pessa[a], Anni Seppälä[b], Tommy Ahlgren[b], Eero Rauhala[b]

[a] *Optoelectronics Research Centre, Tampere University of Technology, P.O. Box 692, FIN-33101 Tampere, Finland*
[b] *Department of Physics, University of Helsinki, P.O. Box 9, FIN 00014 Helsinki, Finland*

Abstract

We prepared InGaN layers on GaN/sapphire substrates using rf-MBE. Photoluminescence (PL) from these layers, grown at different temperatures T_S, shows that there is a strong tendency of GaN to form a separate phase as T_S is increased from 600°C to 650°C. Concomitant with the phase separation, the PL from the InGaN phase broadens, which indicates that indium composition in this phase becomes increasingly non-uniform. Indium compositions measured by Rutherford backscattering (RBS) are consistent with these results. We also observed an increase in PL intensity for InGaN layers grown at higher temperatures. In this paper, we also report on preparing a top-contact InGaN/GaN light emitting diode. The device was operated at 447 nm and had the emission line width of 37 nm with no observable impurity related features. The turn-on voltage was 3.0 V. The output power was 20 μW at 60 mA drive current.

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1. Introduction

MOCVD technology has a leading edge in the GaN studies [1–3]. However, growth by MBE is evolving, due to the introduction of efficient nitrogen sources and a homoepitaxial growth approach. Radio-frequency (rf) MBE growth has some advantages over the MOCVD method. These include the use of a hydrogen-free growth atmosphere and a possibility of p-type doping of GaN without a post-growth heat treatment. Furthermore, atomic nitrogen species produced from an rf-source for incorporation into the growing front are independent of the substrate temperature.

InGaN alloys act as active regions for ultraviolet (UV), blue, and green light emitting diodes (LEDs) and laser diodes (LDs). Properties of the InGaN layer differ from those of GaN. InGaN layers must be grown at low temperature, due to its low decomposition temperature (~650°C).

In this work, InGaN layers were grown by MBE at different temperatures T_S. A temperature dependent phase separation and broadening of

*Corresponding author. Fax: +358-3-365-3400.
E-mail address: pekka.laukkanen@orc.tut.fi (P. Laukkanen).

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InGaN related emission were observed in photoluminescence (PL) experiments. InGaN PL exhibited an intensity peak at a longer wavelength than what was expected from the indium content as seen by RBS. Finally, an electroluminescence of InGaN/GaN MQW-LED is presented.

2. Experimental details

InGaN layers were grown by rf-MBE on HVPE grown GaN on sapphire (0 0 0 1) substrates. The group-III elements were produced from effusion cells. The dopants were Si and Mg. Nitridation of samples was carried out at 650°C, prior to growth. The films were grown at 600–650°C under 7×10^{-5} mbar nitrogen backround pressure at an rf-power of 400 W. Thickness of the layers was varied between 0.2 and 0.4 μm, and the growth rate was 0.1–0.2 μm/h. Surfaces of the films were studied by high energy electron diffraction (RHEED) during growth: (1×1) patterns were observed indicating two-dimensional growth mode. Room temperature PL was measured using an HeCd laser oscillating at 325 nm for excitation.

InGaN/GaN LEDs consisted of a 0.7 μm Si doped layer, 3 periods of InGaN (40 Å)/GaN (100 Å) MQWs and a 0.3 μm Mg doped layer. Doping levels in the n- and p-type GaN layers were 2×10^{19} and $2 \times 10^{18}\,cm^{-3}$, respectively, as deduced from separate Hall and capacitance–voltage ($C-V$) measurements for single GaN layers. Streaky (1×1) RHEED patterns were observed during the growth of layers.

To prepare circular mesa structures of 300 μm diameter, LED samples were dry-etched by CH_4/H_2 to a depth of 0.8 μm to expose the n-GaN layer. Circular Ni/Au contacts with a diameter of 150 μm were evaporated on the p-GaN layer. Circular Ti/Al/Ti/Ni/Au contacts with an inner diameter of 330 μm were evaporated on the n-GaN layer. Finally, the LED array was bonded to a heat carrier and electroluminescence was measured.

The Rutherford backscattering spectrometry (RBS) experiments were performed at the University of Helsinki. The 2.2 MeV ^{4}He ion beam was generated by the 2.5 MV Van de Graaff accelerator. The scattered ions were detected by a standard silicon surface barrier detector placed at an angle of 170°. The data analysis was accomplished by using a spectrum simulation code [4].

3. Results

Photoluminescence from the InGaN layers prepared at 600°C (Sample A), 640°C (Sample B) and 650°C (Sample C) are shown in Fig. 1. Sample D was also grown at 650°C, but the growth rate was twice that for Sample C. Emission from the GaN phase appears at 365 nm, as T_S, and the growth rate are increased. The full width at half maximum (FWHM) of PL from the InGaN phase is increased as a function of T_S. FWHMs are 127, 202, and 210 meV for Sample A, B, and C, respectively. FWHM of PL from Sample D is 213 meV. This broadening of InGaN PL emission can be accounted for indium fluctuation in the

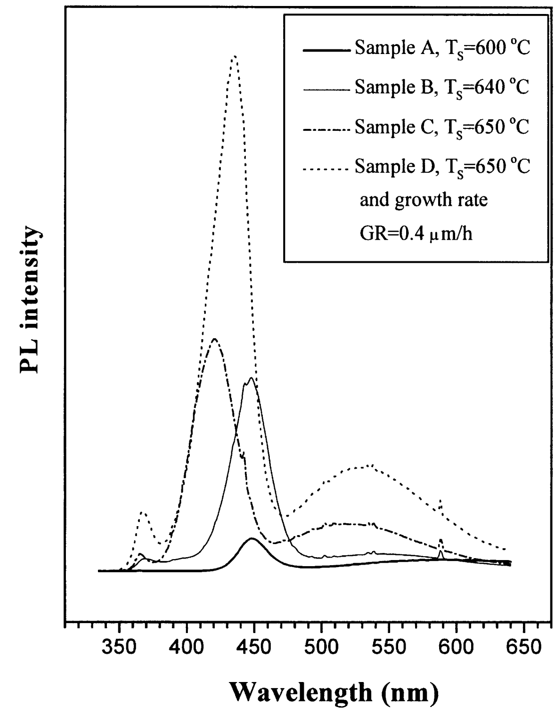

Fig. 1. Photoluminescence from InGaN layers prepared at different substrate temperatures or at a higher growth rate.

InGaN phase. We observed a clear increase in intensity when grown at high T_S and when using a higher growth rate at 650°C.

From the RBS spectrum illustrated in Fig. 2 we determine the maximum In concentration at the surface as 6.5 at% (± 0.5 at%). This corresponds to the atomic ratio of Ga/In = 6.8. The In concentration slowly decreases with depth. The depth concentration distribution obtained from this spectrum and for three other samples having lower In contents are displayed in the inset. The depth distributions were calculated from the measured areal densities (at/cm^2) by assuming a mass density of 6.1 g/cm^3.

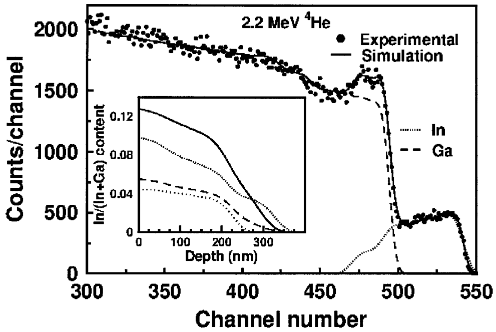

Fig. 2. Experimental and simulated (solid curve) RBS spectra for the sample with the highest In content. The simulated spectrum is composed of the theoretical backscattered signals from In and Ga atoms (dotted and dashed curves). The inset shows the In/(In + Ga) content as a function of depth obtained by RBS for four samples measured. The solid curve in the inset corresponds to the spectrum illustrated.

Fig. 3 shows the peak wavelengths of InGaN phase for layers grown at 650°C as a function of indium composition (RBS) measured at depth of 100 nm. It also shows theoretical InGaN band-edge wavelengths. The theoretical values were determined from the equation

$$E_{\mathrm{InGaN}}(x) = (1 - x)E_{\mathrm{GaN}} + xE_{\mathrm{InN}} - bx(1 - x).$$

The PL and RBS results could be explained only by assuming a bowing parameter $b = 5.4$ eV, which is somewhat larger than what is reported earlier ($b = 3.8-4.8$ eV) [5–7]. The peaking of PL emission at a longer wavelength than that expected from the RBS measurements and a typical band

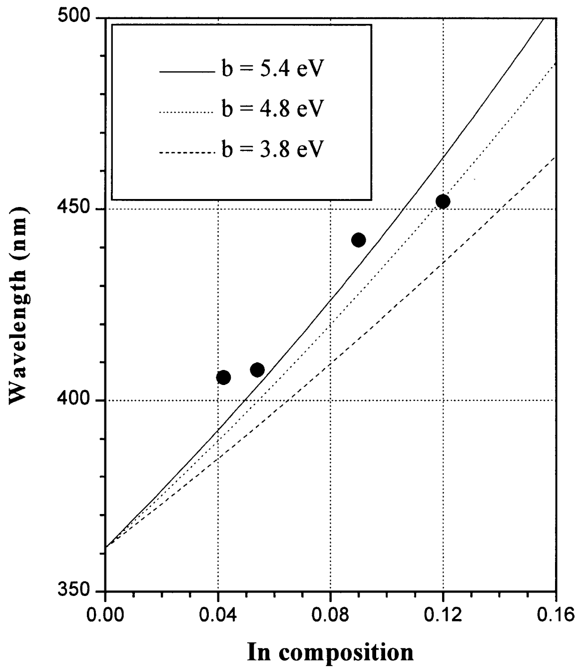

Fig. 3. Wavelength of InGaN peak emission (dots) as a function of indium content measured by RBS. The solid curve is calculated for band-edge emission assuming the bowing parameter of 5.4 eV. The dashed lines correspond the experimental limits for b [4–6].

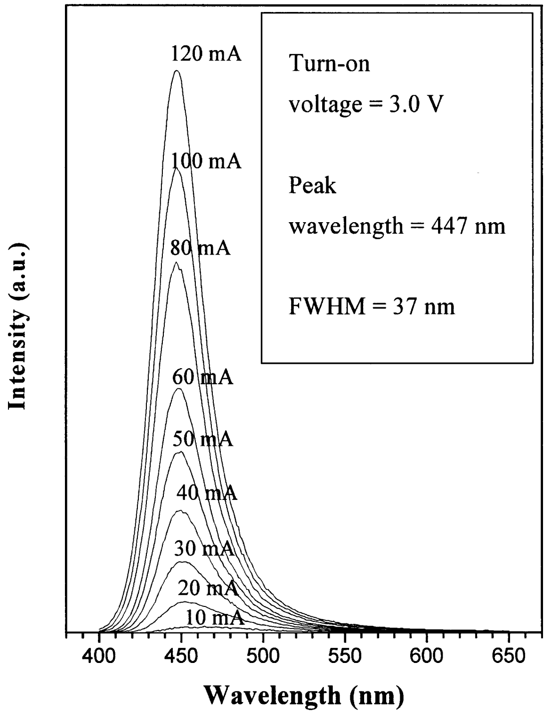

Fig. 4. Electroluminescence spectra of MQW-LED at drive currents ranging from 10 to 120 mA.

bowing, could be explained by the phase separation or the indium fluctuation. Formation of GaN phase regions in the lattice leads to formation of InGaN regions with a higher indium content than the average In content measured by RBS.

InGaN/GaN MQW-LEDs on a HVPE grown GaN substrate had the turn-on voltage of 3.0 V and FWHM of 37 nm at 60 mA driving current. Electroluminescence spectra of LED taken at 10–120 mA are shown in Fig. 4. The device emitted at 447 nm with no observable impurity related emission. The output power was 20 μW at 60 mA drive current.

4. Conclusions

In conclusion, we compared the luminescence properties of InGaN layers grown on GaN/sapphire substrates by MBE. When the growth temperature was increased over 600°C we observed the increase in GaN phase separation and increase in PL intensity and in broadening of PL emission of InGaN phase. We also observed increase in PL intensity for layers grown at the higher growth rate. InGaN/GaN MQW-LEDs showed 20 μW EL emission at 447 nm with 60 mA drive current.

References

[1] S. Nakamura, T. Mukai, M. Senoh, J. Appl. Phys. 76 (1994) 8189.
[2] S. Nakamura, M. Senoh, N. Iwasa, S. Nagahama, Appl. Phys. Lett. 67 (1995) 1868.
[3] S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, Y. Sugimoto, H. Kiyoku, Appl. Phys. Lett. 69 (1996) 4056.
[4] J. Saarilahti, E. Rauhala, Nucl. Instrum. and Meth. B 64 (1992) 734.
[5] C. Wetzel, T. Takeuchi, S. Yamaguchi, H. Katoh, H. Amano, I. Akasaki, Appl. Phys. Lett. 73 (1998) 1994.
[6] M.D. McCluskey, C.G. Van de Walle, C.P. Master, L.T. Romano, N.M. Johnson, Appl. Phys. Lett. 72 (1998) 2725.
[7] C.A. Parker, J.C. Roberts, S.M. Bedair, M.J. Reed, S.X. Liu, N.A. El-Masry, L.H. Robins, Appl. Phys. Lett. 75 (1999) 2566.