AIP Applied Physics Letters

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Citation: Applied Physics Letters **80**, 231 (2002); doi: 10.1063/1.1432751
View online: http://dx.doi.org/10.1063/1.1432751
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/80/2?ver=pdfcov
Published by the AIP Publishing

Strong localization in InGaN layers with high In content grown by molecular-beam epitaxy

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(Received 30 July 2001; accepted for publication 13 November 2001)

The effect of the III/V ratio and growth temperature on the In incorporation has been studied in thick (>300 nm) InGaN layers, with In mole fractions from 19% to 37%, grown by molecular-beam epitaxy on sapphire and on GaN templates. Significant desorption of In occurs at growth temperatures above 550 °C. Symmetric and asymmetric reflections from high resolution X-ray diffraction reveals that the layers are not fully relaxed. A bowing parameter of 3.6 eV is calculated from optical absorption data, once corrected for strain-free band gap values. The increase of both, the absorption band-edge broadening and the photoluminescence full width at half maximum at room temperature with the In content, is discussed in terms of a strong In localization effect. This localization effect is further evidenced by the S-shaped temperature dependence of the emission energy. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1432751]

InGaN-based light emitting diodes and lasers, grown by metalorganic vapor phase epitaxy (MOVPE), have already been demonstrated.[1] Yet, several important issues of the InGaN growth, like its thermodynamic instability,[2,3] and the In segregation during the growth,[4,5] are not well understood. Molecular-beam epitaxy (MBE) growth at low temperatures is a suitable way to obtain InGaN alloys with a high In content and simultaneously avoiding decomposition.[6] Since the crystal quality of MBE-grown GaN on sapphire is generally poorer than MOVPE-grown layers, the use of high quality MOVPE-GaN templates as pseudosubstrates brings MBE growth closer to homoepitaxy, leading to a substantial material quality improvement.[7]

We study the effect of the III/V ratio and the growth temperature on the In incorporation in InGaN layers grown by MBE on sapphire and on MOVPE-GaN templates. The increase of the optical absorption band-edge broadening and the photoluminescence (PL) full width at half maximum (FWHM) with the In content is explained in terms of an In-dependent localization effect. High resolution x-ray diffraction (HRXRD), optical transmission, and PL measurements were used to assess the structural material quality and its optical properties.

InGaN layers were grown by plasma-assisted MBE on Al_2O_3 and on GaN templates consisting of 2 μm-thick GaN/Al_2O_3 grown by MOVPE. Details about the MBE system configuration are found in Ref. 8. In all cases, the amount of active nitrogen was kept constant with a rf power of 400 W and a nitrogen flow rate of 1.0 sccm. PL was excited with the 325 nm line of a He–Cd laser with 1 mW power.

For bare Al_2O_3 substrates, a 10 nm AlN buffer followed by a 400 nm GaN layer (both at 700 °C) preceded the growth of 300 nm InGaN layers at lower temperature (500 °C–600 °C). The first set of InGaN layers (set A) was grown varying the Ga flux (ϕ_{Ga}) from 4×10^{-7} to 8×10^{-7} Torr, and keeping the growth temperature and In flux (ϕ_{In}) constant at 540 °C and 2×10^{-7} Torr, respectively. The second set of layers (set B) was grown at temperatures from 500 °C to 600 °C, while both ϕ_{In} and ϕ_{Ga} were set at 2×10^{-7} Torr and 6×10^{-7} Torr, respectively. The third set of layers (set C) was grown on GaN templates at temperatures from 520 °C to 585 °C with the same fluxes as in set B. In this case, a 300 nm thick GaN buffer layer was grown on the template at 700 °C, prior to the InGaN layer.

The strain state and the In content of the layers were assessed by HRXRD from symmetric (0002), (0004), and asymmetric (105) reflections, assuming the validity of Vegard's law for the lattice parameter.[9,10] Figure 1 shows a plot of the lattice parameter c versus a for different InGaN layers, indicating that, even for the highest In content (37%), the

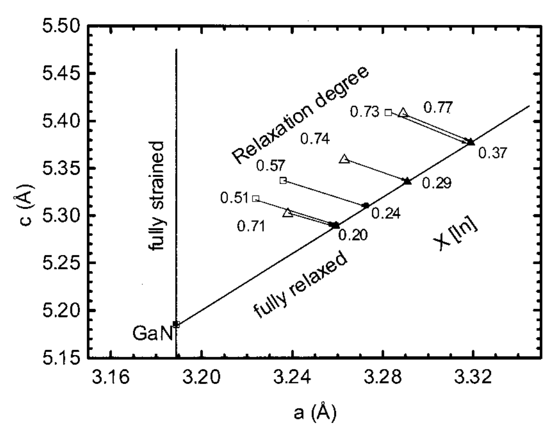

FIG. 1. c and a lattice parameters obtained from HRXRD measurements of samples grown on sapphire (open triangles) and templates (open squares). Black symbols indicate the calculated c and a lattice parameters for unstrained InGaN for each composition.

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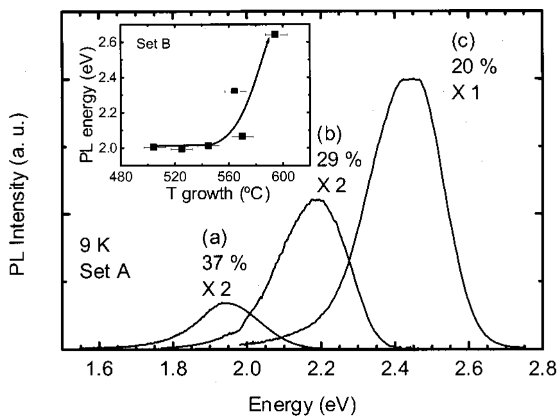

FIG. 2. Low temperature PL spectra of InGaN layers from set A, grown on sapphire at 540 °C with Ga-beam equivalent pressure of (a) 4×10^{-7} Torr, (b) 6×10^{-7} Torr, and (c) 8×10^{-7} Torr. The indium content is specified for each sample. Insert: Low temperature PL emission peak of InGaN layers from set B (ϕ_{Ga} and ϕ_{In} fixed at 6×10^{-7} Torr and 2×10^{-7} Torr, respectively) as a function of growth temperature.

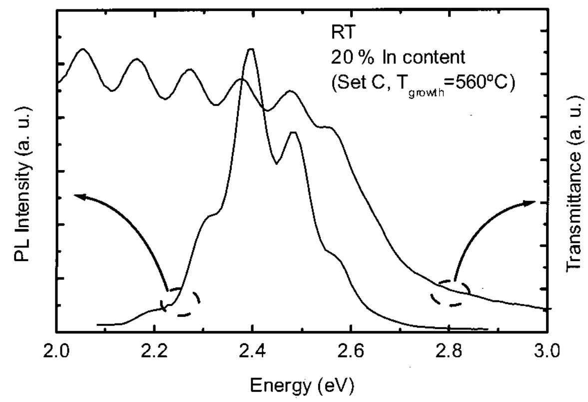

FIG. 3. RT PL and transmittance spectra of an InGaN layer with 20% of In from set C, grown at 560 °C on a GaN template. Fabry–Perot fringes can be observed in both spectra.

degree of relaxation is not 100%. This relaxation degree depends slightly on the substrate used, and it increases with In content and layer thickness.

Figure 2 shows low temperature PL spectra of the layers from set A. A single near band-gap emission is observed in all layers. The In content, estimated from HRXRD data, decreases with increasing ϕ_{Ga} (37% to 20% for ϕ_{Ga} from 4×10^{-7} to 8×10^{-7} Torr), while metal droplets develop on the surface. This decrease of In content with increasing ϕ_{Ga} corresponds to the alloy linear dependence with the III-element flux ratios. On the other hand, the PL intensity increases with ϕ_{Ga}, while the reflection high-energy electron diffraction (RHEED) pattern changes from spotty to streaky. In addition, the morphology of the layers, analyzed by scanning electron microscopy, also improves when ϕ_{Ga} increases. This layer quality improvement with increasing ϕ_{Ga} is due to changes in the stoichometric conditions (III/V ratio), from N rich to matching stoichiometry (III/V=1).

The set B layers, grown from 500 °C to 600 °C, show PL spectra with a blueshift for growth temperatures above 550 °C, due to the thermal In desorption process[11–13] (insert in Fig. 2). Below 540 °C, the mobility of the In adatoms at the growth front is reduced and the RHEED pattern becomes spotty. The degradation of the optical quality of the InGaN layers grown at low temperature is observed as an increase of the PL FWHM (from 180 to 368 meV for samples grown at 565 °C and 505 °C, respectively). A similar temperature effect on the In content is observed in InGaN layers grown on GaN templates (set C). These layers show a much higher PL intensity than similar InGaN layers grown an Al_2O_3, as expected. The Fabry–Perot oscillations in both PL and transmittance spectra (Fig. 3) give an estimate of the resonant cavity length, in very good agreement with the total structure thickness (GaN template+GaN buffer+InGaN layer=2.72 μm).

The bowing parameter of the InGaN system was estimated from transmission data and HRXRD results. The "effective" band gap, $E_{G,\mathrm{eff}}$ and the absorption edge broadening ΔE of the alloy were derived from a sigmoidal absorption approximation given by:[14,15]

$$\alpha(E)=\frac{\alpha_0}{1+\exp\dfrac{E_{G,\mathrm{eff}}-E}{\Delta E}}, \tag{1}$$

where absorption is derived from transmission through the relation $\alpha(E)\propto-\ln(T)$ which neglects interface reflection losses and optical scatter. The gap energy is corrected to relaxed InGaN by using $dE_G/d\epsilon_{\parallel}=14.5$ eV (Ref. 16) and considering the strain measured in the layers. The energy difference between $E_{G,\mathrm{eff}}$ and PL emissions [both at room temperature (RT)] increases with In content, likely due to an enhanced In fluctuation that induces strong localization for In contents above 15%.[14] The obtained bowing parameter value, 3.6 eV, agrees well with previous estimates of Wetzel *et al.*[17] (3.8 eV) in samples grown by MOVPE. The calculated absorption edge broadening (ΔE) correlates with the PL FWHM at RT, both increasing with the In content, giving 153±5 (ΔE) and 186±10 meV for 20% In, and 210±5 (ΔE) and 257±10 meV for 27% In, in good agreement with previous data.[15,18]

Figure 4 shows the temperature evolution of the PL emission energy and FWHM of an $In_{0.2}Ga_{0.8}N$ layer grown on a GaN template. For temperatures below 200 K, a redshift of the PL peak is observed, followed by a blueshift above 200 K ("S-shaped" emission shift). This behavior can be explained by assuming a distribution of potential minima for electrons and holes due to different In compositions (local fluctuations). At low T, the photogenerated electron–hole pairs are randomly distributed among the potential minima, without enough thermal energy to thermalize to the lowest energy states. As the temperature increases (25–200 K), the thermally activated carriers move to the lowest potential minima, and the emission redshifts while the FWHM decreases. Further temperature increases (200–300 K) provide enough energy for the electron–hole pairs to populate higher energy states, leading to a blueshift and to an increase of the FWHM.[15,18] This effect, together with the increase of the absorption band edge and the FWHM of the RT-PL emission with the In content, supports the hypothesis of the existence of a strong In dependent localization in these layers. This localization effect has been modeled as arrays of InN quantum dots embedded in a low In-content InGaN or GaN

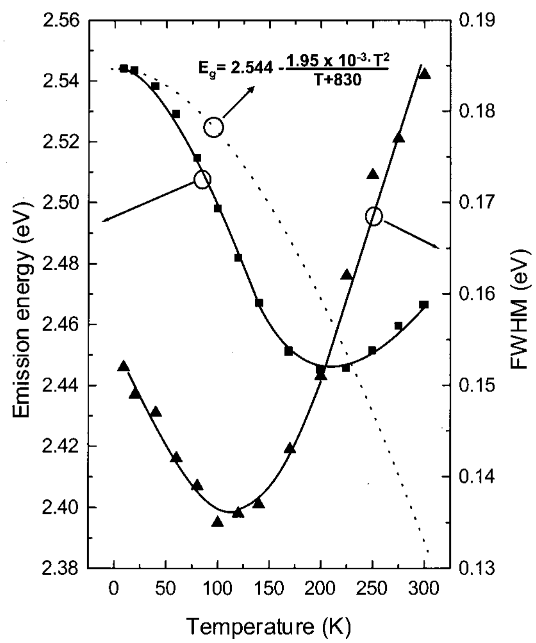

FIG. 4. Temperature evolution of the PL emission energy and FWHM for the same sample as in Fig. 3 InGaN layer with 20% In grown at 560 °C on GaN template. The theoretical band-gap evolution with temperature (dotted line), using Varshini's formula, is shown for comparison, the parameters used were taken from Ref. 21.

matrix,[18,19] or in terms of a nonuniform chemical composition.[20]

Figure 4 also shows that the FWHM and emission energy minima do not happen at the same temperature. This may be understood assuming that the emission comes from different In-rich regions with different weight in the total PL emission. This means a rather inhomogeneous distribution of the In atoms. Indeed, the occurrence of both minima at the same temperature has been observed in a sample with higher In content. This behavior indicates that the specific inhomogeneous distribution of In atoms depends also on the In content.

In summary, high In-content InGaN layers were grown by MBE using a low growth temperature. A significant In desorption occurs at growth temperatures above 550 °C in InGaN layers grown on sapphire and GaN templates. For both substrates, the layers are not fully relaxed despite their thickness (300 nm) and In-content (20%<In%<37%), but they tend towards full relaxation with increasing In content. From optical absorption data at RT, a meaningful downward band-gap bowing parameter of 3.6 eV is derived for this In composition range. The increase of both the absorption band edge broadening and the FWHM of the RT-PL emission with In content, together with the "S-shaped" temperature dependence of the PL, is attributed to a strong local fluctuation of the In composition that induces localization of carriers. The inhomogeneous distribution of In is found to depend on the specific In content.

Thanks are due to N. Herres, J. Hernando, A. Jiménez, and S. Fernández for valuable suggestions and discussions. Partial financial support was provided by IST ESPRIT 1999-10292 "AGETHA" Project.

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