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Photoluminescence and Gain of MBE Grown Cubic $In_xGa_{1-x}N$/GaN Heterostructures

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We report on the molecular beam epitaxy of cubic $In_xGa_{1-x}N$/GaN heterostructures with different In contents. High resolution X-ray diffraction (HRXRD) measurements gave the In content in the layers. Reciprocal space mapping of the symmetrical (002) and asymmetrical (113) reflection reveal that cubic $In_xGa_{1-x}N$ with low In content ($x < 0.2$) is single phase whereas layers with higher In content show evidence of phase separation. The photoluminescence (PL) of the $In_xGa_{1-x}N$ layers was measured at 2 K. The full width at half maximum of the PL is as low as 200 meV for single phase layers whereas the emission is significantly broadened for phase separated $In_xGa_{1-x}N$. Optical gain measurements revealed a maximum gain of about 70 cm^{-1} from $In_xGa_{1-x}N$ ($x = 0.07$) and a decrease of the gain with increasing In content.

1. Introduction

The recent observation of optically stimulated emission at room temperature from cubic GaN/GaAs (001) [1] has demonstrated the potential of III-nitrides with cubic crystal structure for the realisation of laser structures with cleaved facets. However, the bulk of experience with hexagonal III-nitride laser structures shows that all working diode lasers contain InGaN in the active region. The growth of c-InGaN layers may therefore be considered as a major issue on the way towards a c-III nitride based laser diode. Epitaxial growth of c-$In_xGa_{1-x}N$/GaN/GaAs(001) heterostructures ($x = 0.17$ and 0.4) has been reported recently [2, 3]. The low temperature photoluminescence (PL) from these InGaN layers consisted of one emission band with a full width of half maximum (FWHM) of about 400 to 550 meV. In this paper, we report on the growth of c-$In_xGa_{1-x}N$/GaN heterostructures ($0.02 < x < 0.33$) and their low temperature PL as well as measurements of their optical gain.

2. Results and Discussion

All our InGaN/GaN heterostructures were grown on GaAs (001) substrates by molecular beam epitaxy (MBE) using an rf plasma nitrogen source. The GaN buffer layers were grown at a substrate temperature of 720 °C under carefully controlled stoichiometric conditions [4]. InGaN layers with a thickness between 50 and 330 nm were depos-

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ited at lower growth temperatures (610 to 680 °C) and an N_2 background pressure of 5×10^{-3} Pa. During the InGaN epitaxy the Ga flux was about 20% less than that used during the GaN layer deposition and the In flux was adjusted to establish a metal-rich surface composition. The growth was continuously monitored by RHEED. The diffraction patterns did not show any reflections originating from hexagonal or misoriented cubic crystals and exhibited a cubic symmetry along all major azimuths.

The X-ray diffraction measurements were performed with a high resolution diffractometer. We measured the distribution of the scattered X-ray intensity in reciprocal space (reciprocal space maps, RSM) and ω–2θ scans of the symmetric (002) and the asymmetric (113) Bragg reflection of the $In_xGa_{1-x}N$/GaN heterostructure. We find that the FWHM of the reflections in the direction of the scattering wave vector parallel to the sample surface ($q_{||}$) was almost identical for GaN and the InGaN layers, indicating that the density of extended defects in both layers is approximately equal.

The intensity maxima of the asymmetrical (113) reflections are located on a line which points from the GaN (113) reciprocal lattice point towards the origin of the reciprocal space (000) indicating that the lattice of the InGaN layers is fully relaxed. The relaxation of the InGaN lattice parameter was also observed during growth from the separation of the RHEED reflections. The alloy composition of the InGaN layers was calculated from the measured lattice parameter assuming a linear variation of the lattice parameter between $a_{(GaN)} = 0.452$ nm and $a_{(InN)} = 0.497$ nm [5].

Fig. 1 depicts the RSM of the (002) and (113) X-ray reflection from sample No. 294. We find three reflections in the vicinity of the GaN Bragg peak. The isointensity contours of the reflection with the highest intensity (centered at $q_{||} = 0.02$ Å^{-1}, $q_\perp = 2.7$ Å^{-1} and $q_{||} = 1.89$ Å^{-1}, $q_\perp = 4.18$ Å^{-1}, respectively) are quite similar to the reflections from other InGaN layers with a lower In content. These reflections are due to a fully relaxed $In_xGa_{1-x}N$ layer with $x = 0.33$. The structure of the other two reflections is significantly different. The spread of intensity in the $q_{||}$ direction indicates that a large number of small crystals which are not perfectly aligned along the growth axis contribute to the observed reflections. The localisation of the maximum intensity in the (113) RSM

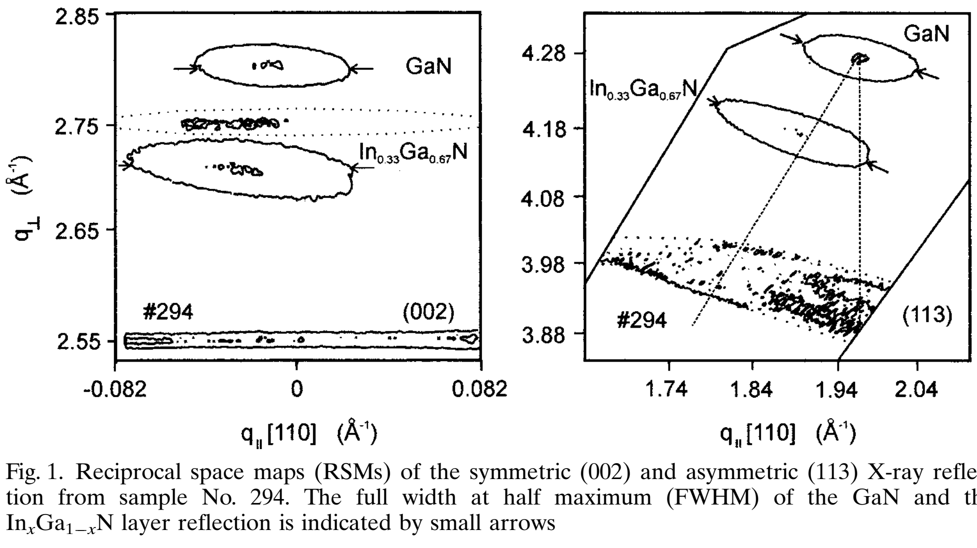

Fig. 1. Reciprocal space maps (RSMs) of the symmetric (002) and asymmetric (113) X-ray reflection from sample No. 294. The full width at half maximum (FWHM) of the GaN and the $In_xGa_{1-x}N$ layer reflection is indicated by small arrows

shows clearly that the unit cells of these crystallites are tetragonally distorted. In this case the Poisson ratio ν enters the calculation of the composition of the alloy. We performed an approximate calculation of the In content using the value $\nu = 0.366$ of c-GaN [6] obtaining $x = 0.2$ and $x = 0.75 \pm 0.05$ for the In content of the respective crystals. We suppose that the formation of these inclusions is due to spinodal decomposition [7] of the c-InGaN ($x = 0.33$) layer. Evidence for phase separation has also been found in a recently reported X-ray diffraction spectrum of a c-InGaN layer with $x = 0.4$ [3].

The low temperature PL spectra of our $In_xGa_{1-x}N/GaN$ heterostructures are shown in Fig. 2. The PL spectra of $In_xGa_{1-x}N$ layers with $x \leq 0.2$ consist of one emission band. The FWHM of this band is in the order of 200 to 450 meV and slightly increases with increasing x. The energy of the emission peak is about 100 to 200 meV lower than the gap energy E_g which was obtained from spectrally resolved ellipsometry and measurements of the reflectivity of the $x = 0.02$, 0.07, 0.10 and 0.2 GaInN layer by Goldhahn et al. [8]. The corresponding values are indicated by arrows in Fig. 2. In the PL spectrum of sample No. 294 we find strong emission with photon energies clearly exceeding 2.55 eV. We suppose that the luminescence peak at 2.4 eV stems from the InGaN ($x = 0.33$) layer while the emission at higher energies is due to microcrystalline inclusions with an approximate In content of $x = 0.2$.

The gain of our $In_xGa_{1-x}N/GaN$ heterostructures was measured by the variable stripe length method. Low temperature gain spectra of sample 302 obtained at different excitation intensities are depicted in Fig. 3. Gain close to the band gap energy of the $In_xGa_{1-x}N$ layer at about 3.05 eV was already observed with an excitation density of about 0.5 MW/cm^2. Increasing the excitation density results in a shift of

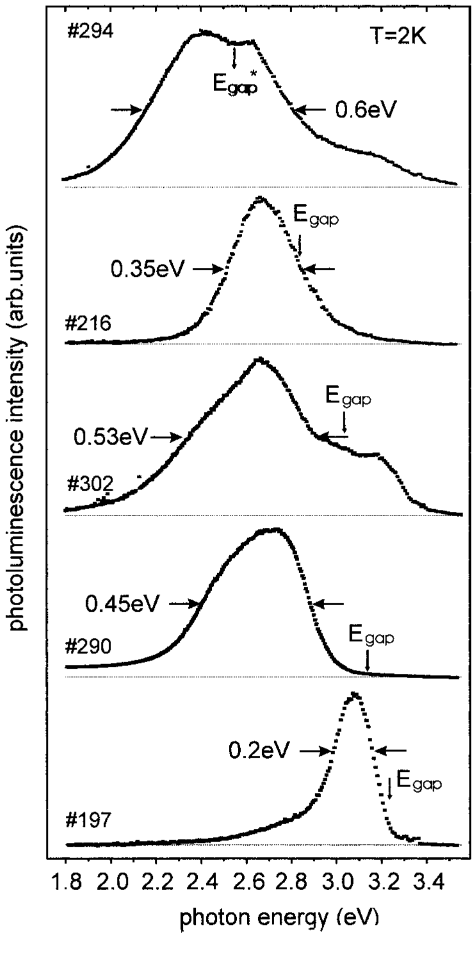

Fig. 2. Low temperature photoluminescence of $In_xGa_{1-x}N/GaN$ heterostructures with different In contents. (# 197, $x = 0.02$; # 290, $x = 0.07$; # 302, $x = 0.10$; # 216, $x = 0.20$; # 294, $x = 0.33$). The full width at half maximum (FWHM) of the PL emission as well as the corresponding gap energy E_g from spectrally resolved ellipsometry and reflectivity measurements is indicated. E_g^* is the extrapolated gap of $In_xGa_{1-x}N$ ($x = 0.33$)

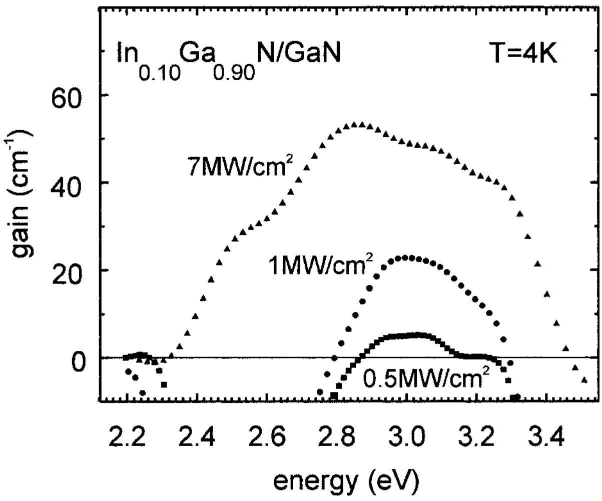

Fig. 3. Low temperature gain spectra of sample No. 302 ($In_xGa_{1-x}N$, $x = 0.1$) measured at different excitation densities

the gain maximum to lower energies (from its energy position the gain at 3.23 eV is attributed to emission from the c-GaN layer) and a broadening of the gain spectrum due to the increased number of excited carriers. This behaviour can be explained by the In-fluctuation model. With increasing carrier density the probability of a non-radiative decay into the energetically lower fluctuation states is enhanced and the amplification from these states is increased. We observed optical gain even at 2.5 eV indicating a strong shift of the emission into the green spectral range at significantly lower In contents than in the hexagonal phase. This is due to the fact that a quantum-confined Stark effect due to piezoelectric fields is *not* expected in cubic $In_xGa_{1-x}N$/GaN/GaAs(001) heterostructures. The absorption on the low energy side of the gain structures can be explained by the inhomogeneous distribution of In in the InGaN layers, which causes an increased number of defects.

The maximum gain measured at an energy close to the band gap energy of the $In_xGa_{1-x}N$ layers is depicted in Fig. 4. With increasing In content the gain values are

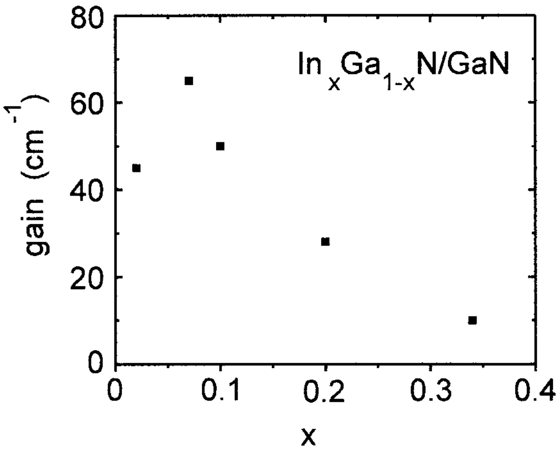

Fig. 4. Low temperature maximum gain of different $In_xGa_{1-x}N$/GaN heterostructures measured at the gap energy of the $In_xGa_{1-x}N$ layer as a function of the In content x

increasing up to an In content of about 7%. For higher In concentrations the gain is decreasing. We believe that the decrease of the gain in In-rich samples is due to the InN–GaN miscibility gap [7] and the enhanced tendency for the formation of In clusters.

3. Conclusions

In conclusion, we have grown $In_xGa_{1-x}N/GaN$ heterostructures with cubic crystal structure. The In content in the $In_xGa_{1-x}N$ layers ($0 < x < 0.4$) was measured by X-ray diffraction and Rutherford backscattering spectroscopy. For $x > 0.2$ we find clear evidence for In-rich inclusions being most likely formed by spinodal decomposition of the growing layer. The effect of phase separation is also observed in the broadening of luminescence and gain spectra of $In_xGa_{1-x}N$ layers with $x > 0.2$. $In_xGa_{1-x}N/GaN$ heterostructures with $x < 0.1$ have a maximum low temperature gain of about 70 cm^{-1}.

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