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Strong room temperature 510 nm emission from cubic InGaN/GaN multiple quantum wells

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ABSTRACT

Cubic InGaN/GaN double heterostructures and multi-quantum-wells have been grown by Molecular Beam Epitaxy on cubic 3C-SiC. We find that the room temperature photoluminescence spectra of our samples has two emission peaks at 2.4 eV and 2.6 eV, respectively. The intensity of the 2.6 eV decreases and that of the 2.4 eV peak increases when the In mol ratio is varied between $x = 0.04$ and 0.16. However, for all samples the peak energy is far below the bandgap energy measured by photoluminescence excitation spectra, revealing a large Stokes-like shift of the InGaN emission. The temperature variation of the photoluminescence intensity yields an activation energy of 21 meV of the 2.6 eV emission and 67 meV of the 2.4 eV emission, respectively. The room temperature photoluminescence of fully strained multi quantum wells ($x = 0.16$) is a single line with a peak wavelength at about 510 nm.

INTRODUCTION

Low cost short-haul communications systems using polymethyl methacrylate (PMMA) plastic optical fibers (POFs) require inexpensive light sources emitting around the PMMA absorption minimum at 510 nm (around 70 dB/km). For the realization of these devices group III-nitride wide-band-gap semiconductors are the material of choice. Due to their large direct band gap they are well suited for a wide range of applications, e.g. as light emitter in the green to ultraviolet range or as detectors [1].

Group III-nitrides can be produced in the thermodynamic stable configuration with hexagonal (wurtzite) crystal structure and in a metastable modification with cubic (zincblende) structure. The main difference between these two modifications is the absence of piezoelectric and spontaneous polarization fields in the cubic modification. Today high-efficiency LEDs which have InGaN/GaN quantum wells in the active zones are produced in large quantities. However, these quantum wells, which are mainly grown in a (0001) growth direction, have

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strong built-in electric fields due to the piezoelectric effect and spontaneous polarization. For that reason only nm-wide quantum wells yield large radiative recombination efficiency. Due to the higher symmetries polarization fields are absent in cubic III-nitrides [2]. Furthermore, due to the slightly smaller energy gap of the cubic nitrides (200meV lower than the hexagonal counterpart), smaller mol fractions of In in the well of InGaN/GaN quantum wells are necessary to reach emission wavelengths beyond 510 nm. Therefore, group III-nitrides seem to be the material choice for the realization of resonant cavity light emitting diode (RC-LED) for the 500–570 nm spectral range [3].

In this work, we report on the optical properties of cubic InGaN double heterostructures (DH) and multi quantum well (MQW) structures for room temperature 510nm emission which were grown by RF plasma-assisted molecular beam epitaxy (MBE). We show that cubic InGaN DHs emit at two distinct emission energies which vary with increasing In molar fraction between 2.6 and 2.4eV. In contrast to hexagonal InGaN the photoluminescence (PL) intensity of the InGaN/GaN quantum well increases with increasing well thickness, which enables high efficient green emission of cubic InGaN/GaN quantum wells in the green spectral region.

EXPERIMENTS

All samples were grown on 3C-SiC substrates. Our Riber 32 MBE system is equipped with an RF plasma source for activated N atoms. The In and Ga atoms were evaporated from conventional Knudsen cells and the metal fluxes were adjusted by changing the source temperatures. Prior to growth, the 3C-SiC substrates were chemically etched and annealed for 10 hours at 500°C. First a 700 nm thick cubic GaN buffer layer was grown on the 3C-SiC substrate at 720°C under stoichiometric growth conditions. On top of this c-GaN buffer layers $In_xGa_{1-x}N$ double heterostructure (DH) and multi quantum wells (MQW) have been grown. For the DH structure the substrate temperature (T_{sub}) has been reduced to 620°C first and 40 to 60 nm thick $In_xGa_{1-x}N$ were grown with an In-content varying between $x = 0.045$ and 0.19. Then these layers were capped with 50 nm GaN grown at $T_{sub} = 720°C$.

The MQW consists of 6 $In_xGa_{1-x}N$/GaN QWs with a barrier and well thickness of 12 nm and 5 nm, respectively. The $In_xGa_{1-x}N$ layers were deposited under In-rich conditions at a growth temperature of 610°C with an In content of 0.16.

Photoluminescence (PL) was excited by a 325 nm He-Cd laser and measured with a standard PL set-up at temperatures between 4K and 300K. High resolution X-ray diffraction (HRXRD) and reciprocal space mapping have been performed to determine the In molar fraction and the strain in the $In_xGa_{1-x}N$ layers.

RESULTS AND DISCUSSION

Room temperature PL spectra of $In_xGa_{1-x}N$/GaN DH samples with different In content and a InGaN layer thickness of 60nm are shown in Fig.1. The $In_xGa_{1-x}N$ emission dominates the spectra and only a weak 3.2eV c-GaN emission is observed, indicating a high recombination

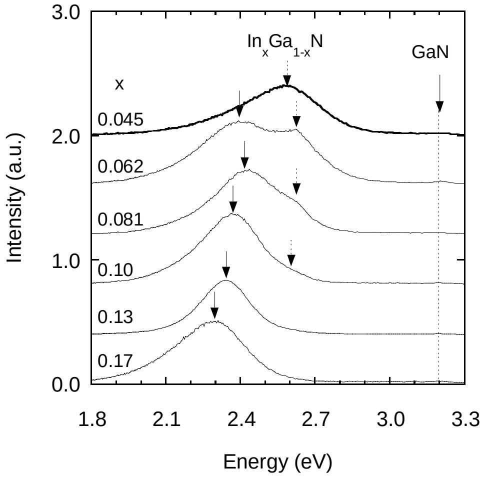

Figure 1. *Room temperature PL spectra of InGaN DH samples with different In composition, the full and dashed arrows mark the peak position.*

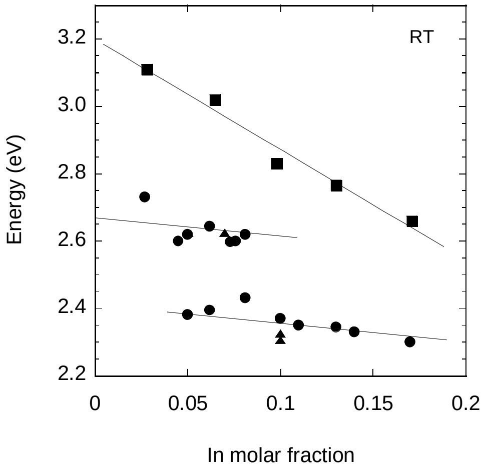

Figure 2. *PL peak energy (full circles) and band gap energy (squares) versus In composition. Triangles are data from literature [2, 5].*

efficiency of the $In_xGa_{1-x}N$ layers. The PL-spectra of $In_xGa_{1-x}N$ samples in the lowest ($x = 0.045$) and highest ($x > 0.13$) In concentration ranges (as measured by XRD) consists of a single Gaussian-like peak at photon energies of 2.4 eV and 2.6 eV, respectively. For intermediate In concentrations however, the spectra show two distinct peaks, one around 2.4 eV and another around 2.6 eV. These peaks move to lower photon energies as the In concentration increases, as shown by the arrows in Fig.1.

Figure 2 shows the $In_xGa_{1-x}N$ related peak energy of our DH structures plotted versus In composition (full circles). Also included in this diagram are the PL peak energies of cubic $In_xGa_{1-x}N$ layers grown on 3C-SiC substrates and results from other groups (black triangles) [2, 4]. These data reveal clearly that the PL emission of c-InGaN does not shift monotonically with increasing In content, but seems to be dominated by two well defined emission bands. The full squares are the $In_xGa_{1-x}N$ band gap energies measured by photoluminescence excitation (PLE) spectroscopy [5]. These data are in excellent agreement with spectral ellipsometry measurements performed on c-$In_xGa_{1-x}N$ grown on GaAs substrates [6]. The black line shows the $In_xGa_{1-x}N$ band gap versus In composition. The PL peak energy is about 300 meV lower than the band gap and does not exactly follow the band gap energy dependence of $In_xGa_{1-x}N$. This large stokes shift suggests that localized structure in $In_xGa_{1-x}N$ may be responsible for the PL emission [2] and the existence of two peaks in the PL spectra indicate that at least two different $In_xGa_{1-x}N$ phases are formed which coexist simultaneously in the intermediate In-molar fraction range.

The temperature dependence of the peak energies of the 2.4 eV and 2.6 eV lines show an “S” shape behaviour, a features which is also found with hexagonal InGaN [7], indicating that the emission is due to localized structures. Figure 3 shows an Arrhenius plot of the peak intensities, giving activation energies of 67 meV for the 2.4 eV peak and 21 meV for 2.6 eV peak, respectively. We assume that in our c-InGaN there are localized structures with two preferential

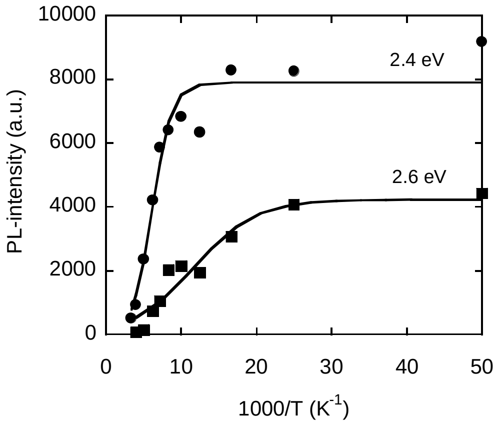

Figure 3. *Arrenhius plot of the InGaN PL intensities for the 2.4 and 2.6 eV emission.*

In compositions, which emit light in energy close to 2.4 eV and 2.6 eV in the In composition range of 0-18%. The localized structures which emit at 2.6 eV are dominant in c-InGaN with low In content (x < 0.05) and localized structures with 2.4eV emission are dominant in c-$In_xGa_{1-x}N$ with x > 0.1.

Teles et al. [8] have calculated the energetic and thermodynamic properties of cubic $In_xGa_{1-x}N$ using first principle total energy calculation and Monte Carlo simulations. They found that the formation of ordered phases is energetically favourable in cubic InGaN under biaxial strain. We assume that the observed luminescence may be due to recombination of electron hole pairs in clusters of this phases since the 2.4 eV emission is close to the band gap of c-$In_{0.25}Ga_{0.75}N$, the $[InN]_1[GaN]_3$ ordered phase. The 2.6 eV emission may be due to clusters in the disordered phase.

Cubic InGaN/GaN quantum wells

Spontaneous and piezoelectric polarization field in hexagonal III nitride can effectively separate electrons and holes in QWs and result in low PL intensity and red shift of the emission energy with increasing well thickness, which is called quantum confined stark effect (QCSE). For that reason, the optimal thickness of h-InGaN well is around 2-3 nm in the h-InGaN/GaN system. In cubic InGaN, due to the high crystal symmetry, no polarization field exists and therefore high PL intensity will be expected also with thick InGaN wells.

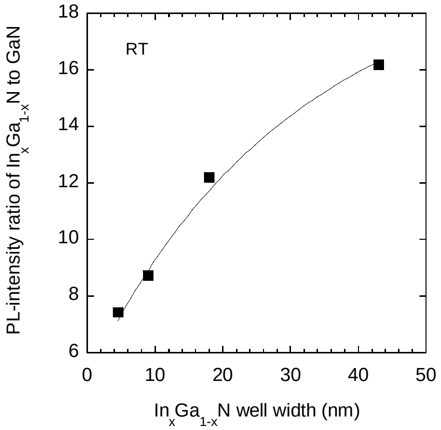

Figure 4. *Intensity ratio of InGaN peak to GaN peak versus well thickness in the InGaN/GaN single quantum well samples.*

We have grown InGaN/GaN single QWs with varying thickness of the InGaN layer. Room temperature PL spectra are measured under the same laser power. Figure 4 shows the ratio of integrated intensity of the InGaN and GaN emission as a function of well width. We find an increase of the well luminescence with increasing well width. This behavior is in contrast to what is known from h-InGaN/GaN QWs [9], where the PL intensity decreases with well thickness due to the separation of electrons and holes by spontaneous and piezoelectric polarization field. Our results reveal that the thickness of c-InGaN/GaN QW can be varied in a wide range in order to yield an optimum recombination efficiency.

We have shown that c-InGaN/GaN DHs have a strong emission at about 2.4eV. The emission wavelength depends only weakly on the In content and is close to the absorption minimum at 510 nm of POFs revealing the potential of cubic InGaN for the green spectral region. Therefore, we have grown a 6-fold c-$In_xGa_{1-x}N$/GaN MQW with optimized green emission. Figure 5 shows a typical XRD reciprocal space map around the (-1-13) reflex on c-$In_xGa_{1-x}N$/GaN MQW. The superlattice peaks are clearly resolved up to 5th order. The $In_xGa_{1-x}N$ wells are pseudomorphically grown on GaN barrier layers. The InGaN well thickness is 5.6 nm, and the GaN barrier thickness is 12.1 nm as obtained from the X-ray diffraction data. The In mole fraction is about 0.16. The strong and dominant room temperature 2.4 eV (510nm) MQW emission is shown in Fig.6. The GaN emission can not be detected, revealing a high radiative recombination efficiency of the InGaN quantum wells.

Our results demonstrate that we can grow high quality c-InGaN/GaN MQWs with room temperature PL emission of 2.4 eV, which is suitable to be the active region of the green light resonant cavity LED.

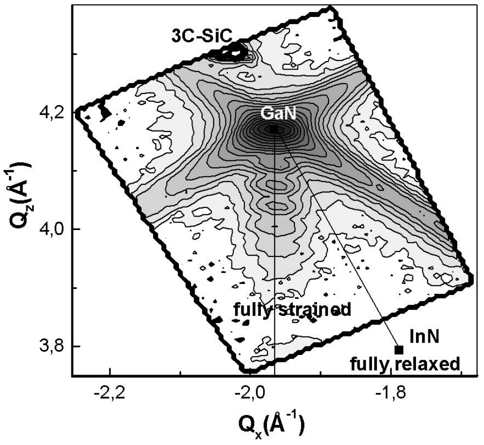

Figure 5. *High resolution XRD reciprocal space map near (-1-13) reflex of a 6 folds cubic $In_{0.16}Ga_{0.84}N$/GaN MQWs.*

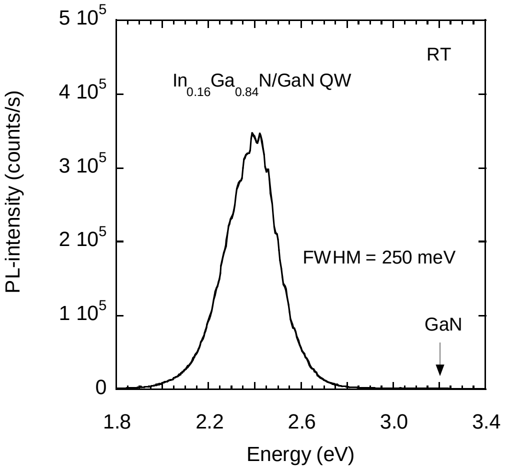

Figure 6. *Room temperature PL spectrum of a 6 folds cubic $In_{0.16}Ga_{0.84}N$/GaN MQWs.*

CONCLUSIONS

The investigation of the PL from cubic InGaN/GaN double heterojunctions and quantum wells which were grown by MBE on 3C-SiC substrates reveals that two distinct emission lines at about 2.6 eV and 2.4 eV dominate the room temperature spectra of structures with different In content. The 2.6 eV line is found with samples with an In mol ratio of x = 0.04-0.08, the 2.4 eV emission dominates the PL of structure with an higher In content (x = 0.1-0.18). The PL of all structures is significantly red shifted relative to the InGaN gap energy which was measured by photoluminescence excitation spectroscopy, indicating that the PL stems from localized structures within the InGaN layers, most likely In rich clusters. The temperature variation of the PL intensity yields the thermal activation energy of electron-hole pairs in these clusters which is 67 meV for the 2.4 eV emission and 21 meV for the 2.6eV emission, respectively. In order to demonstrate that cubic InGaN/GaN quantum wells may be used in the active region of resonant cavity LEDs for green (510nm) light we have demonstrated strong room temperature PL at about this wavelength from a strained 6-fold cubic InGaN/GaN MQW.

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