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Optical Properties of Structures with Single and Multiple InGaN Insertions in a GaN Matrix

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We report on growth and optical and structural characterisation of samples with single and multiple InGaN insertions in a GaN matrix. We found that InGaN insertions decompose to arrays of In-rich nanodomains or quantum dots (QDs) with lateral sizes of 2 to 4 nm. High density of the QDs results in a possibility to achieve surface-mode lasing under photoexcitation (λ = 400 nm) in a structure with 12-fold stacked InGaN insertions even without using of Bragg mirrors. We found that the threshold excitation density is weakly affected by temperature up to 120 K, while it increases at higher temperatures, a behavior typical for QD lasers. We also demonstrate a possibility to achieve gain in the bright-blue and green spectral range using similar ultrathin insertions but with higher In content.

1. Introduction

Recently, quantum dot (QD) heterostructures [1] have attracted much interest in view of the possibility to improve significantly the basic parameters of optoelectronic devices and extend their spectral range for the same substrate material. For example, it was recently demonstrated for dense arrays of stacked II–VI quantum dots (QDs) that ultrahigh material gain of the active medium allows to achieve surface lasing even without using of highly-reflecting mirrors [2]. Formation of QD-like structures in the InGaN system has been also demonstrated by deposition of thin InGaN layers on a lattice-mismatched GaN substrate resulting in spontaneous formation of arrays of nano-islands [3], and by spinodal decomposition of thicker InGaN alloy layers [3 to 5].

In our case QDs were formed by spinodal decomposition of ultrathin InGaN insertions in a GaN matrix. In this work we investigated growth, structural and optical properties of the structures with multiple InGaN insertions. We demonstrate the advantages of these structures for nitride-based vertical-cavity surface-emitting lasers (VCSELs).

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2. Growth and Characterisation

The samples studied were grown by low pressure metalorganic chemical vapor deposition (MOCVD) employing an AlGaN nucleation layer deposited at 530 °C on (0001)-oriented sapphire substrates. Ammonia, trimethylindium (TMI), trimethylgallium (TMG) and trimethylaluminum (TMA) were applied as component precursors. Purified hydrogen and/or argon [6] were used as carrier gases. The samples consisted of a 2.5 μm thick GaN layer grown at 1050 °C in a hydrogen ambient, an InGaN active region grown in argon ambient and a 0.1 μm-thick GaN cap layer grown at 1050 °C in hydrogen. A 25 nm thick InGaN layer having a low indium content (10% In) was grown at 800 °C and served as a buffer layer for strain-compensated InGaN–GaN multilayer structures, having the same average In content. The multilayer structures were formed by temperature cycling between 730 and 860 °C, resulting in a modulated In compositional profile, as the In incorporation is strongly affected by the substrate temperature. During the growth of the active region TMI and TMG flows remained constant. The structure grown under the TMI/(TMI + TMG) mole flow ratio of 0.4 (it will be referred as "violet") demonstrated lasing in surface geometry as well as stimulated emission in edge direction. Cross-section transmission electron microscopy (TEM) image of this structure and digitally processed lattice image (DALI) of HRTEM of one period are shown in Fig. 1.

We found that by increasing the mole flow ratio up to 0.7 one can reach the bright-blue spectral range using this technique. Attempts to increase further the In content in this way resulted in accumulation of metal indium on the surface and degradation of the optical properties. However, the structure grown under mole flow ratio 0.8 applying both temperature cycling and modulation of the TMI flow demonstrated a clear-green emission, even the average thickness of the insertions revealed in transmission electron microscopy (TEM) remained fairly small (1 to 2 nm) (it will be referred later as "green").

The structural studies were performed by using TEM and X-ray diffractometry (XRD). The XRD data allowed to determine the average In content and the periodicity of the multilayer structures. The periods derived from the XRD data and the TEM cross-sectional micrographs were in good agreement with the growth rate calibrations.

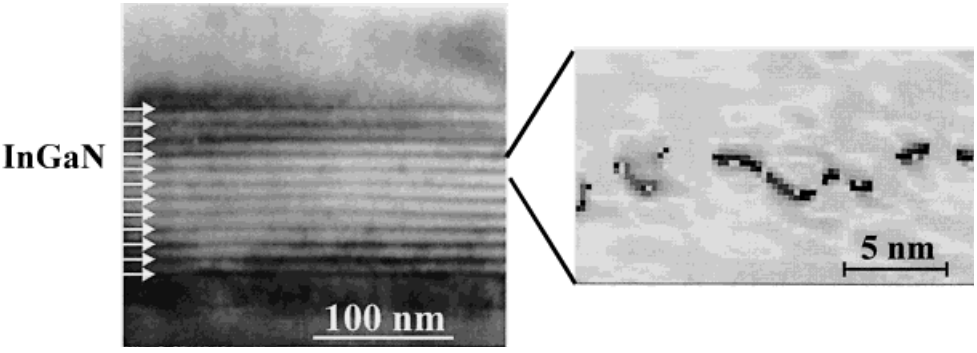

Fig. 1. a) Cross-sectional TEM image for the "violet" structure and b) DALI processed image of one InGaN insertion (black color represents higher In content)

The photoluminescence (PL) study was performed in the temperature range 14 to 300 K using a continuous wave He–Cd laser or a pulsed N_2 laser for excitation. Gain studies were performed at 2 K using dye laser pumped by excimer laser.

3. Results and Discussion

Low-temperature photoluminescence (PL) spectra of the "violet" structure recorded from the sample surface are shown in Fig. 2a. The PL spectrum at low excitation densities shows a single relatively broad peak with extended tails on both high and low energy sides. This shape of the spectrum is not typical for QWs, where the high-energy side of the spectrum above the exciton mobility edge usually demonstrates a temperature-sensitive exponential decay. On the contrary, the PL shape agrees with the formation of In-rich nanodomains, or QDs, with significant dispersion in size and In concentration. Recent studies using high-resolution transmission electron microscopy (HRTEM) allowed to determine the shape of the nanodomains. Processed HRTEM images revealed flat In-rich nanodomains with lateral size of 2 to 4 nm and a height of one or several monolayers (Fig. 1a). The area concentration of the domains was about $10^{12}\,\mathrm{cm}^{-2}$ [7]. With increase in the excitation density, the high energy side of the PL spectrum increases its intensity, and a new narrow peak evolves in the spectrum. The intensity of this peak increases superlinearly with the excitation density. These effects point to observation of stimulated emission in surface geometry. As it will be shown later, the analysis of the mode structure clearly indicates surface lasing.

Fig. 2b shows the temperature dependence of the InGaN-related PL peak energy and its full width at half maximum (FWHM) for the violet structure. As it follows from Fig. 2, the temperature shift of the PL peak energy does not follow the band gap. This behavior is typical for structures with extended density of states. Initially, temperature increase results in population of higher energy states, and the PL peak energy shifts

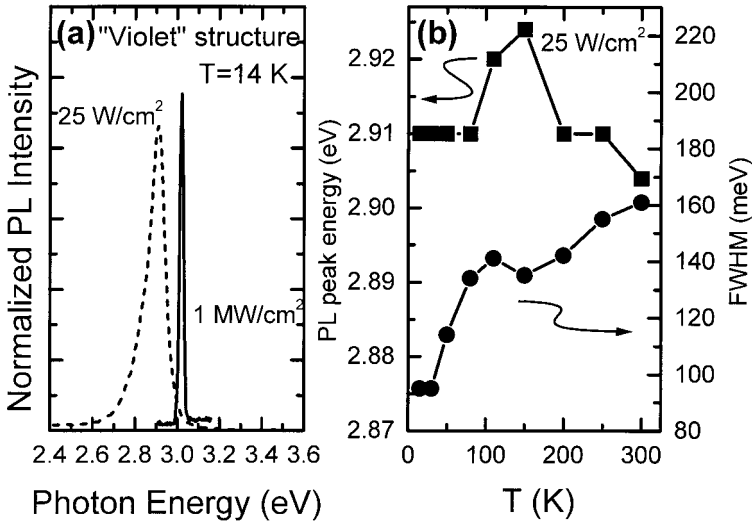

Fig. 2. a) Low temperature photoluminescence spectra at low and high excitation densities. b) Temperature dependence of the PL peak energy and the FWHM for the "violet" structure evaluated at low excitation densities

towards higher energies and the FWHM of the PL emission increases. However, at about 140 K a significant redistribution of carriers to different kinds of localized states occurs. At this temperature, weaker localized carriers can be recaptured by larger QDs (or by QDs having higher In content) and a significant long-wavelength shift of the PL peak is observed. One may conclude that the potential barriers between QDs are high enough. Above this temperature the energy distribution of excitons becomes narrower, and the FWHM decreases. This anomalous behavior (so called S-shaped dependence) was also observed in [8] and attributed to the presence of localized states.

PL spectra of the "violet" structure recorded at 150 K at different excitation densities are shown in Fig. 3a. The reduced differential efficiency of the stimulated emission at higher temperatures makes the spectral changes related to increase in excitation density more evident. It can be clearly seen that all the PL spectra are modulated by the modes of the Fabry-Perot microcavity formed by the GaN/Al_2O_3 interface and the GaN/air interface. It can be seen that at high excitation densities one of the cavity modes starts to dominate in the PL spectra and its peak intensity grows superlinearly. Single-mode emission together with a strong increase in the slope efficiency indicates the key role of the feedback in the system despite of the remarkably low finesse of the cavity. The threshold modal gain (g_{th}) necessary to overcome external losses (α_{ext}) and achieve surface lasing can be calculated as

$$g_{th} = \alpha_{ext} = \frac{1}{2L} \ln\left(\frac{1}{R_1 R_2}\right), \tag{1}$$

where R_1 and R_2 are the reflectivity coefficients for both interfaces forming the cavity, and L is the cavity length. We estimate the reflectivity coefficients to be 2.4% for GaN/sapphire and 17% for GaN/air interfaces. Taking into account that the active region, responsible for gain, has a thickness of 0.15 μm, one can obtain a value of 2×10^5 cm^{-1} for the threshold gain in the active medium to overcome losses.

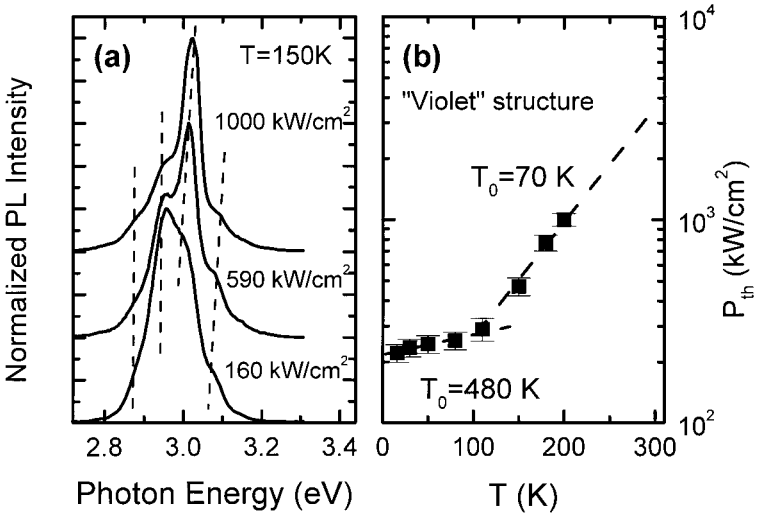

Fig. 3. a) Photoluminescence spectra recorded at different excitation densities (150 K). b) Temperature dependence of the threshold excitation density for surface lasing

In Fig. 3a one can see a short wavelength shift of the cavity modes (marked with the dashed lines) with increase in the excitation density. The larger shift (up to 3.2 nm) was observed for the high-energy modes, while the low energy modes did not shift. This effect results from a strong modulation of the absorption/gain curve in the vicinity of the lasing energy. To explain the observed shift one needs to assume a value of the change of refractive index in the active region of 0.4. This giant value agrees, however, with the estimated gain value necessary to achieve surface lasing. The effect of interaction of gain spectrum and cavity modes has been reported for QD based lasers in InGaAs/GaAs [9] and II–VI systems [3, 5].

The temperature dependence of the threshold excitation density for surface lasing is shown in Fig. 3b ("violet" structure). In a temperature range between 16 and 120 K the threshold excitation density remains weakly affected, while it increases at higher temperatures. Characteristic temperatures (T_0) of 480 K and 70 K are measured in the low and high temperature ranges, respectively. Increase in the threshold excitation density at higher temperatures can be attributed to thermal evaporation of carriers from QDs and their subsequent nonradiative decay. The critical point for the change in the slope of the threshold current density vs. temperature dependence is about 120 K. This temperature fits to the characteristic temperature for the strong spectral changes of the PL emission, recorded at low excitation densities (see Fig. 2b).

Gain spectra measured using the standard variable excitation length method for the "green" structure are shown on Fig. 4. The structure contained essentially no external waveguide, and the intrinsic resonant waveguiding effect [10] due to resonant enhancement of the refractive index due to ultrahigh gain/absorption coefficients in quantum dots is believed to be small in this case because of the very broad spectral width of the spectra. This resulted in propagation of light in the defect-rich interface region where significant absorption and scattering of light occurs, and, consequently, in high internal optical losses. As it can be seen in Fig. 4, a significant relative gain has been achieved in the green spectral range for this structure. The gain saturation value in this range approaches $\approx 50\,\mathrm{cm}^{-1}$ and further increase in excitation density leads to appearance of a new peak on the high-energy side of the spectrum and evolution of stimulated emis-

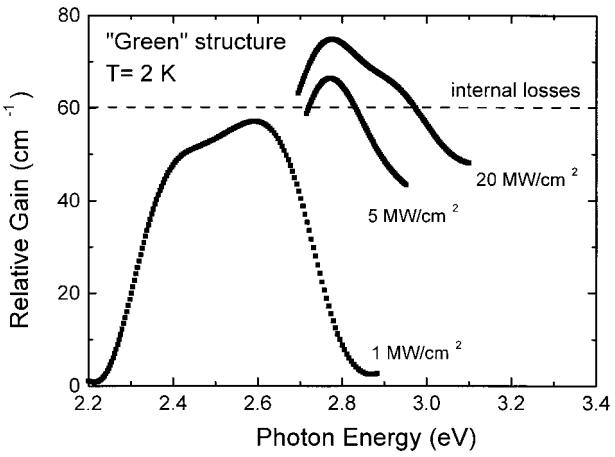

Fig. 4. Gain spectra for the "green" structure recorded at different excitation densities

sion. This result shows the principal possibility to achieve lasing in the green spectral range using ultrathin InGaN insertions in a GaN matrix, if further optimization of technology and device layout will be applied.

4. Conclusions

To conclude, we show that deposition of ultrathin InGaN insertions may result in spontaneous formation of dense arrays of In-rich nanodomains having lateral sizes of 2 to 4 nm. Structures with multiple InGaN insertions emitting in the UV-to-blue spectral ranges can be formed by the substrate temperature cycling during the InGaN deposition. The PL peak energy is strongly affected by the growth parameters in this case and the gain is high enough to overcome huge optical losses ($>2 \times 10^5\ cm^{-1}$) to achieve surface lasing even in the case of low finesse cavities. This makes the structures promising for VCSEL applications.

The structure with ultrathin InGaN insertions formed by combination of temperature cycling and In flow modulation allows to realize significant gain in the blue and green spectral range.

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