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Impact of Structural Properties on the Mechanisms of Optical Amplification in Cubic GaInN

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The structural and the optical properties of cubic GaInN MBE-grown on GaAs substrates are investigated using scanning electron- and cathodoluminescence microscopy, time-resolved and time-integrated photoluminescence spectroscopy as well as gain measurements at 2 K and 300 K. The In content is ranging from 3% to 30%. From the carrier dynamics localized states are proposed to be responsible as recombination mechanism. From temperature- and intensity-dependent gain measurements, the identification of the gain processes was possible. Optical gain values up to 60 cm^{-1} were observed at wavelengths up to 500 nm, indicating the advantage of this material system due to the lack of detrimental pyro- and piezoelectric fields. The degree of In fluctuations directly determines the optical quality and the efficiency of optical amplification of the samples.

1. Introduction

Recently the epitaxy of metastable, cubic GaInN on GaAs(001) substrates has attracted some interest since c-GaInN layers and the GaAs substrate have a common cleavage plane. Consequently they are considered to be well suited for the fabrication of laser cavities with cleaved facets [1 to 6]. In high-excitation optical experiments on c-GaInN/GaN/GaAs(001) it was shown that the samples with In contents up to 30% emit at long wavelengths up to 550 nm [7]. This can be explained by the absence of detrimental piezoelectric fields which suppress the radiative recombination in the hexagonal phase. Therefore, c-GaInN is very promising as basic emitting material for the green spectral range. However, it was found that the optical gain is reduced for In contents >7% [7]. This was explained by the increasing impact of In fluctuations on the radiative efficiency. It is the purpose of the present paper to analyze the correlation of structural properties on the mechanisms of optical amplification in cubic GaInN/GaAs samples with In contents up to 30%.

2. Experiment

The GaInN/GaN/GaAs(001) heterostructures were grown by plasma assisted MBE. The GaN buffer layers with a typical thickness in the range of 100 to 200 nm were grown at

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T = 720 °C under carefully controlled stoichiometric conditions, exploiting Reflection High Energy Diffraction (RHEED) measurements of the surface reconstruction as an *in situ* control of the composition of the layer surface during growth. The GaInN layers have a thickness between 200 and 300 nm and were deposited at lower temperatures (T = 610 to 680 °C). During the GaInN epitaxy we used a Ga flux which was reduced by about 20% as compared to that of the GaN layer. The In flux was adjusted to establish a metal-rich surface taking into account the extremely small and strongly temperature dependent sticking coefficient of In. In concentrations ranging from 3% to 30% are realized.

The low excitation photoluminescence (PL) spectra were recorded from the top of the sample with a continuous-wave (cw) helium–cadmium laser. To obtain the high excitation density necessary for our investigations we used a dye laser pumped by an excimer laser, providing pulses with a duration of 15 ns at a rate of 30 Hz and a total energy of up to 20 μJ at 340 nm. The samples were mounted in a bath cryostat at 1.8 K. Gain measurements were performed using the variable-stripe-length method [8].

The cathodoluminescence (CL) measurements were performed at liquid He temperature (5 K) in a fully computer-controlled modified scanning electron microscope (SEM). A spatial resolution better than 40 nm is achieved under optimum conditions. Details and applications of the CL technique have been described elsewhere [9].

3. Results

Fig. 1 shows the photoluminescence spectra of a c-GaInN/GaN/GaAs sample with an In content of 10% at different excitation densities up to 5 MW/cm^2. The spectra broaden

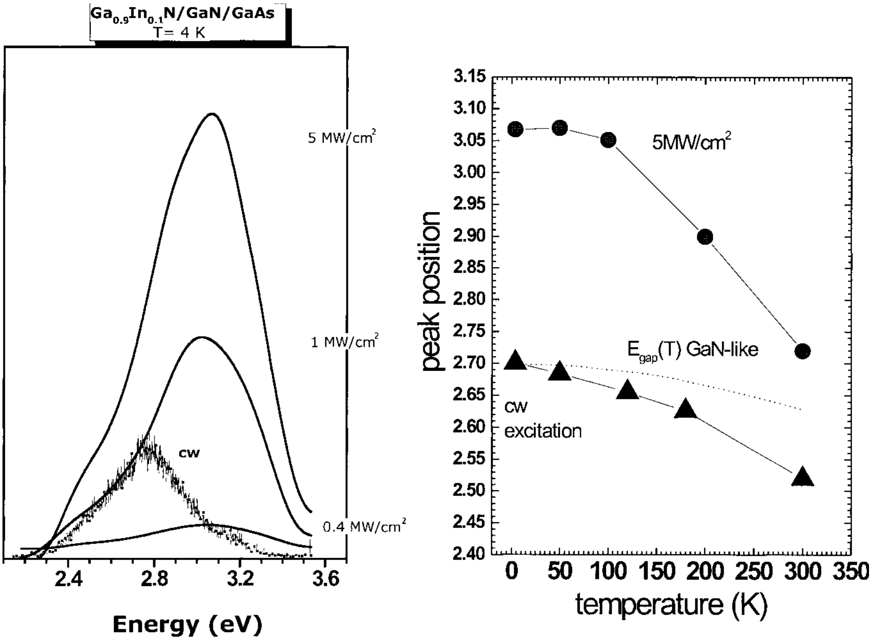

Fig. 1. Left: High-excitation spectra of $Ga_{0.9}In_{0.1}N$ at 4 K for different excitation densities (the cw spectrum is plotted for comparison). Right: peak position of low excitation (triangles) and high-excitation (circles) spectra at various temperatures between 4 K and 300 K

and the peak position is slightly shifted into the blue spectral range with increasing excitation density. The low excitation PL spectrum is displayed for comparison, resembling the strong blue-shift of the high-excitation emission. This can be explained in terms of a band fluctuation model by consecutive filling of the high-energy states with increasing excitation. On the right hand side of Fig. 1 the temperature dependence of the low excitation cw PL is compared with high-excitation measurements for the 7% In sample. The triangles represent the peak position at low excitation and the circles at high-excitation for temperatures from 4 to 300 K. The expected theoretical energy shift extrapolated from the band gap of GaN according to Varhni's formula is plotted for comparison. Obviously, the temperature induced red-shift of the InGaN peak is significant in both cases, i.e. at the low and much more pronounced at high excitation densities. This temperature-dependent behavior is similar to observations of localized excitons in CdSe islands [10] in a ZnSe matrix. Therefore, we believe that localized carriers in In rich islands are responsible for the observed behavior. This model is strongly supported by our time-dependent measurements where a strongly non-exponential decay of the luminescence at 4 K and 300 K is found. Concluding our time-resolved and time-integrated PL measurements we believe that localized carriers trapped at potential fluctuations are the origin of the luminescence in c-GaInN.

To understand the correlation of structural and optical properties we performed cathodoluminescence microscopy and results are presented in Fig. 2 for a sample with [In] = 7%. A CL wavelength image (CLWI), i.e. the mapping of the local emission wavelength is depicted in Fig. 2a together with the laterally integrated CL spectrum b) and a set of local CL spectra c) to f). The local CL spectra in this area (c, d) exhibit broad emission bands, but the peak position changes according to the local In content.

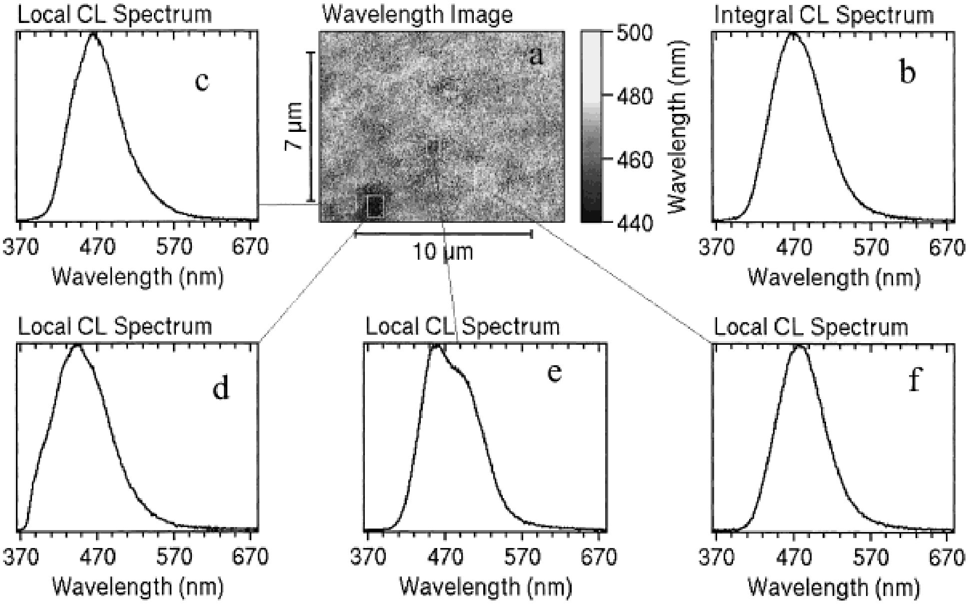

Fig. 2. Cathodoluminescence spectra of $Ga_{0.93}In_{0.07}N$. a) CLWI, b) integral CL spectrum and c) to f) local CL spectra

This is a typical feature for all samples investigated. The local compositional fluctuations are directly monitored in the CL mappings. For samples with higher In contents ([In] >10%) the degree of In fluctuations is strongly enhanced. This can be seen from the histograms of the CLWIs, where the frequency of the local emissions wavelength is accumulated. Fig. 3a displays the histograms for samples with different In contents. The width of the histogram directly monitors the degree of In fluctuation in the samples and is found to be low in the 7% sample, while for the 20% sample a strongly inhomogeneous distribution of In over the sample can be seen. Moreover, for [In] > 7% the histograms are no longer given by a statistical Gaussian distribution but become multimodal with several relative maxima directly visualizing a phase separation.

In Fig. 3b the gain spectra at a fixed excitation density of 5 MW/cm^2 are shown for GaInN samples with In contents from 3% to 20%. With increasing In content up to 7% the gain profiles are broadened, indicating the higher effective carrier concentration and maximum gain values of 60 cm^{-1} are obtained. However, for higher In concentration the optical gain decreases and is totally suppressed in the sample with [In] = 20%. This directly shows the negative impact of phase separation on quantum efficiency.

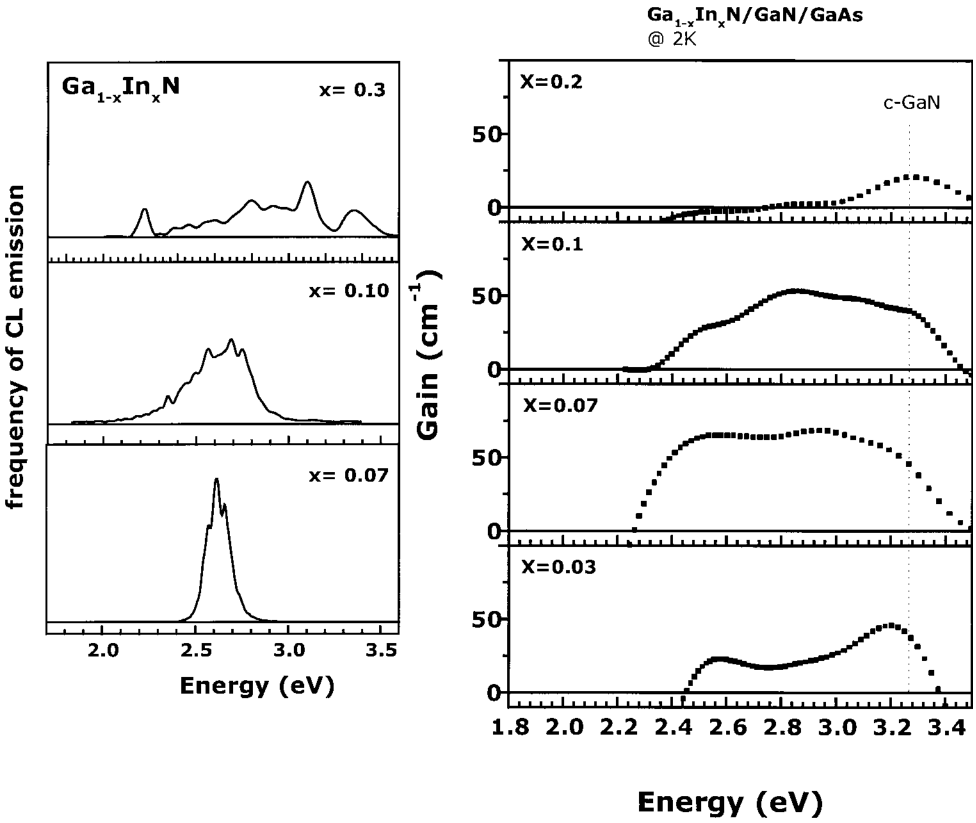

Fig. 3. Left: Histograms of Ga$_{1-x}$In$_x$N samples for various x-values. Right: gain spectra at a fixed excitation density of 5 MW/cm^2 for samples with varying In content

4. Discussion

The observed high-excitation luminescence and the emission dynamics can be understood in terms of a band fluctuation model. Localized states in the minima of potential fluctuations are proposed to be responsible as main luminescence source. The excitation density dependence results from band filling effects, both in *k*-space resulting in recombination on the high-energy side of the spectrum. The recombination dynamics can be interpreted by the model of Pophristic et al. [11] which accounts for strongly localized disordered systems and was found to be descriptive for the hexagonal GaInN system. By applying this model to our measurements we found a good agreement with the predictions. The sample with a higher degree of In fluctuations (10%) exhibits an even more non-exponential decay than the 7% sample and a recombination time of about 2 ns, pointing to an enhanced degree of disorder and localization in samples with higher In content.

The results of the CL measurements can be directly correlated to the optical properties found by high-excitation and gain measurements. The degree of In fluctuations is enhanced with increasing In content, as can be see from the histograms. At the theoretically pronounced maximal solid composition of GaInN of only 7% [12] the lowest degree of In fluctuation is observed and the same sample exhibits the highest gain values as well as the most effective emission output. At increasing In concentrations a phase separation occurs, reducing the gain values, i.e. the efficiency of optical amplification. It is interesting to note that the phase separation in cubic GaInN is not accompanied by strain-induced piezoelectric fields which are detrimental to the radiative recombination [13]. In contrast to findings in the hexagonal GaInN system, in our samples optical gain is observed at 500 nm, indicating the advantage of this material system for light emitters in the green spectral range.

5. Conclusion

Time-resolved photoluminescence, temperature-dependent high-excitation and intensity-dependent gain measurements were performed and correlated to spatially-resolved cathodoluminescence investigations on GaInN samples with varying In content. We observed a non-exponential decay of the luminescence in the ns time range, indicating the influence of localized states as origin of the recombination process. From the CL investigations the degree of In fluctuations can be directly monitored. At the maximal solubility of In in GaN which is given by [In] = 7% the lowest degree of In fluctuation is found. In correlation with the gain measurements it was evidenced that stronger In fluctuations decrease the gain values. Since no detrimental piezoelectric fields can be expected in the cubic phase we observed emission and optical gain up to wavelengths of 500 nm, indicating the interesting properties of c-GaInN as emitter in the green spectral range. All our results are consistently explained in terms of the model that carriers localized in band fluctuations are responsible for the radiative recombination in c-GaInN. Further investigations are needed to prove this and to reveal a basic understanding of the mechanisms of recombination and optical gain in the c-GaInN system.

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