The origin of optical gain in cubic InGaN grown by molecular beam epitaxy

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The optical properties of cubic InGaN samples with varying In content are investigated to provide insight into the processes responsible for optical amplification. The samples were grown by molecular beam epitaxy on GaAs substrates. The structural and optical properties were studied by means of time-resolved and time-integrated photoluminescence spectroscopy and cathodoluminescence microscopy, as well as gain measurements at various temperatures. From these measurements, localized states are proposed to be responsible as recombination mechanism. The cathodoluminescence measurements evidence a direct correlation of the degree of In fluctuation and the efficiency of optical amplification of the samples. © *2000 American Institute of Physics.* [S0003-6951(00)04720-3]

The epitaxy of metastable, cubic InGaN on GaAs (001) substrates has attracted some interest in several respects.[1–3] From the technological point of view *c*-InGaN layers are well suited for the fabrication of laser cavities with cleaved facets because they have a common cleavage plane with the GaAs substrate.[1] On the other hand it was shown in high-excitation optical experiments on *c*-InGaN/GaN/GaAs (001) that the samples with higher In contents emit at long wavelengths up to 550 nm.[4] This is of special interest in comparison to the hexagonal phase where detrimental piezoelectric fields suppress the radiative recombination.[5] In addition to the lower band gap, *c*-GaInN is proposed to be a very promising basic emitting material for the green spectral range. Despite these technological and physical advantages of this material system only a few experimental data are available on the optical properties of *c*-InGaN.[6,7] Therefore, the purpose of the present letter is to reveal an insight of the recombination mechanism and to correlate the impact of the structural properties on the efficiency of optical amplification in cubic InGaN/GaAs samples.

All our InGaN/GaN heterostructures were grown on GaAs (001) substrates by molecular beam epitaxy (MBE) using a radio-frequency (rf) plasma nitrogen source, see details in Ref. 8. The In content of the $In_xGa_{1-x}N$ layers was obtained from high resolution x-ray diffraction (HRXD), ranging from 3% to 31%. For the time integrated high-excitation 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.[9] For time-dependent photoluminescence the signal was analyzed in a 0.35 m subtractive double spectrometer and detected by a multichannel plate (MCP) photomultiplier. A single photon counting setup was used with a 50 ps response to the laser pulse. Employing convolution techniques the overall time resolution is enhanced to 15 ps. 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.[10]

The low temperature photoluminescence (PL) spectra of our $In_xGa_{1-x}N$/GaN heterostructures are shown in Fig. 1 as full lines. Luminescence was exited by the 325 nm radiation of a continuous-wave (cw) HeCd laser. The linewidth [full width at half maximum (FWHM)] of the PL emission band of the $In_xGa_{1-x}N$ layers slightly increases from 200 to 450 meV with increasing x. The energy of the emission peak shifts to lower energies with higher In content and is about 100–200 meV lower than the gap energy E_g which was obtained from spectrally resolved ellipsometry and measurements of the reflectivity by Goldhahn *et al.*[11] The corresponding values of E_g are indicated by arrows in Fig. 1. The dashed curves in Fig. 1 are the spectra of light emitted from a cleaving edge of the layer. In this case the layer surface was excited by the 340 nm light of a pulsed laser with an intensity of 1 MW/cm^2. Notably the peak energy of the edge emission is close to the gap energy and clearly shifted with respect to the luminescence peak. The localized states which contribute to PL at weak excitation are filled by carriers with increasing excitation intensity. At 1 MW/cm^2 all low energy radiative recombination processes are saturated and the observed edge emission is the near band edge luminescence from the $In_xGa_{1-x}N$ layers. This suggestion is corroborated by results of time resolved PL measurements. Figure 2 shows the transient of the PL from $In_xGa_{1-x}N$ layers with $x=0.07$ and 0.1, respectively. A nonexponential decay is observed for all InGaN samples. To describe the uncommon shape of the transients the model of stretched exponentials

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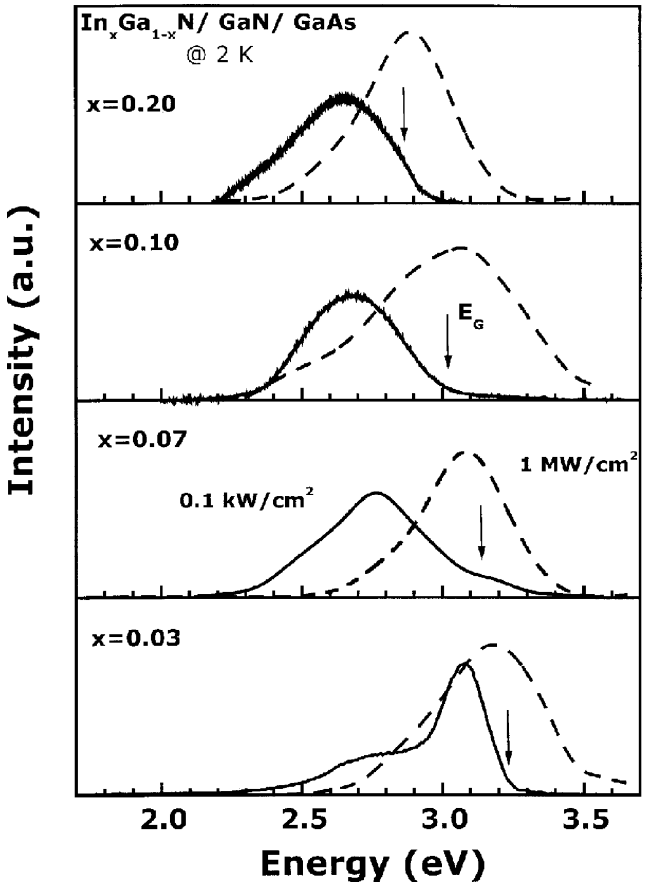

FIG. 1. Comparison of low-excitation PL (full curves) and the high-excitation edge emission (dotted curves) of c-InGaN samples with varying In content. The values of the band gap, determined by reflectivity, are indicated by arrows.

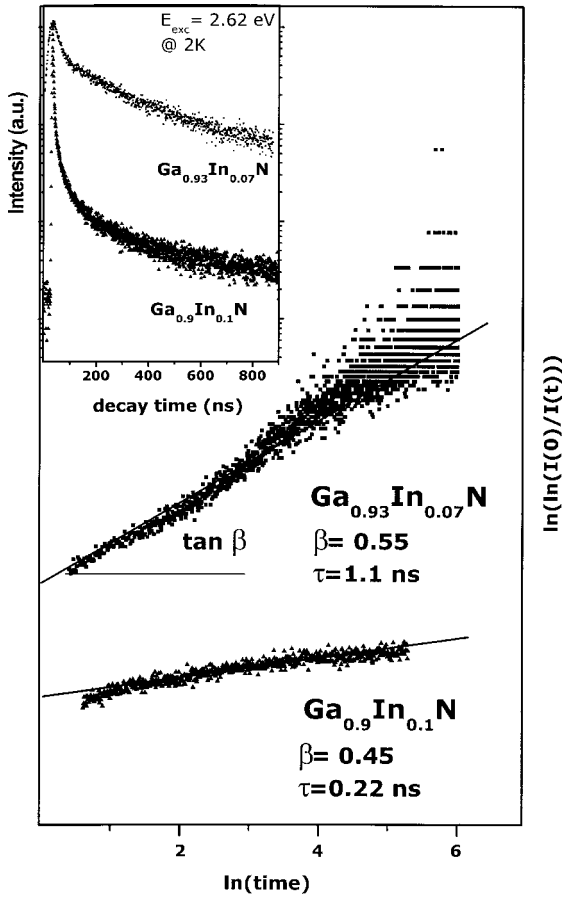

FIG. 2. The transients of the two samples with In contents of 7% and 10% plotted according to the model of stretched exponentials. The values for τ and β can be determined by plotting $\ln\{\ln[I(0)/I(t)]\}$ vs $\ln(t)$. (Inset) Luminescence decay of the two c-InGaN samples.

was used. This model was introduced to describe the radiative decay of strongly disordered systems of localized recombination centers and was applied successfully to h-InGaN,[12] II–VI quantum dots,[13] and porous Si.[14] The decay of a system of localized recombination centers can be described by the formula:

$$I(t) = I_0 \exp\left[-\left(\frac{t}{\tau}\right)^{\beta}\right],$$

where β is between 0 and 1.[15] The values for β and τ can be determined independently simply by plotting the double logarithm of the intensity versus the Ln of the time. The kinetics of the recombination can be described by the model of stretched exponentials, which indicates the strong influence of energetic and structural disorder of the system on the emission process in c-InGaN. The spatial fluctuations of In concentration[16] and the well known In segregation[17] are the reasons for such a microscopic disorder of the luminescence centers. From these observations and the reflectivity and PL data we conclude that the origin of the luminescence is due to localized carriers confined at varying In fluctuations. These localized states are not only relevant for the luminescence. This is shown in Fig. 3, where the gain spectra for excitation densities up to 5 MW/cm^2 of an $In_{0.07}Ga_{0.93}N$ sample are displayed. With increasing excitation intensity a shift of the crossover gain absorption to higher energies can be observed, indicating the band filling of the localized states. The gain structures broaden and the peak position shifts to higher energies. This behavior is a further indication that the optical amplification is due to localized states because the gain of an electron-hole plasma is expected to shift to lower energies with increasing density.[18] It should be noticed that optical gain is observed up to 500 nm, indicating

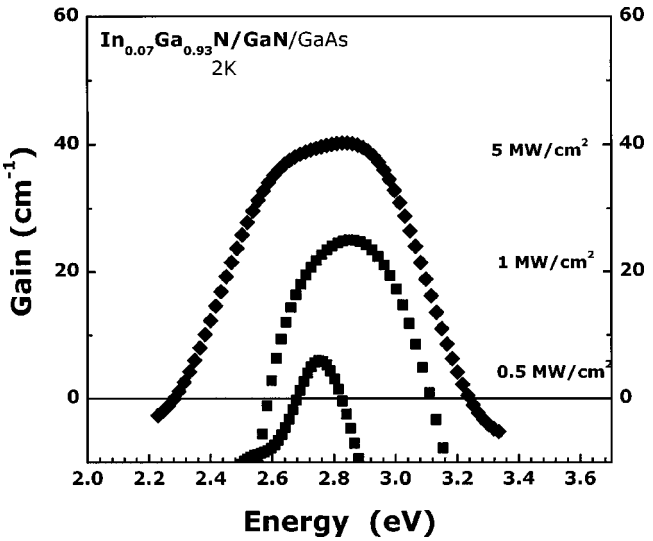

FIG. 3. Gain measurements at various excitation densities of an $In_{0.07}Ga_{0.93}N$ sample.

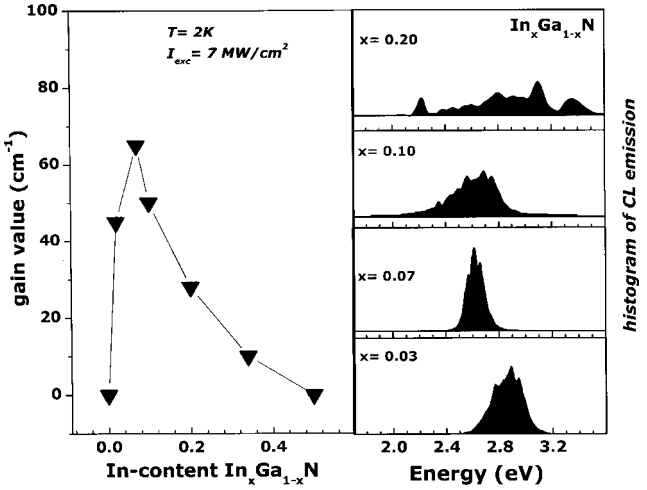

FIG. 4. (right hand side) Comparison of the peak gain values at an excitation density of 7 MW/cm^2 for *c*-InGaN samples with different In content. (left hand side) Results of the cathodoluminescence measurements. Displayed are the histograms of the CLWI for *c*-InGaN samples with varying In content.

the advantage of this material system for device applications in the green spectral range due to the lack of detrimental pyro- and piezoelectric fields.

To investigate the correlation of structural and optical properties we performed cathodoluminescence microscopy on the *c*-InGaN samples. The local CL spectra of the samples exhibit broad emission bands, but the peak position changes according to local In content. This is a typical feature for all samples investigated. 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 CL wavelength images (CLWI), where the frequency of the local emissions wavelength is accumulated. The left hand side of Fig. 4 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 in the range of 170 meV in the 7% sample, while for the 20% sample a strongly inhomogeneous energetic distribution of luminescence centers 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. To monitor the impact of the degree of In fluctuations on the optical amplification, gain spectra at a fixed excitation density of 7 MW/cm^2 are measured and the results are shown for samples with varying In content in the right hand side of Fig. 4. The sample with an In content of 7% exhibit the highest gain values, while for increasing In contents above 20% the optical amplification is suppressed. This can be understood by the theoretically pronounced immiscibility gap of InGaN. A phase separation is expected at In contents higher than 7%[17] and above this value nonradiative processes suppress the optical gain.

In summary, we used time-integrated and time-resolved PL, cathodoluminescence and gain spectroscopy to examine the origin of the optical recombination processes of *c*-InGaN samples with different In contents. From the optical investigations a strong experimental evidence was verified that the luminescence and the optical gain are due to localized carriers which are confined in band fluctuations. In these *c*-InGaN layers the degree of In fluctuations strongly determines the efficiency of optical amplification. The insufficient incorporation and solubility of In in the *c*-InGaN layers seems to limit the optical gain in the green spectral range.

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