Stimulated Emission in InGaN/GaN Multiple Quantum Wells with Different Indium Content

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We report on high-excitation luminescence spectroscopy of In\textsubscript{x}Ga\textsubscript{1−x}N/GaN multiple quantum wells with a high indium content (\(x = 0.22\div0.30\)). High excitation conditions enabled us to achieve screening of built-in field by free carriers. This allowed for the evaluation of the influence of the band potential fluctuations due to variation in In-content on optical properties. Enhanced spontaneous emission was found for \(x \geq 0.22\) due to carrier localization within the chaotic band potential. Meanwhile the stimulated emission was found to be the highest for structures with \(x \approx 0.25--0.27\). We attribute the In-content dependence of the stimulated emission intensity to a trade-off between an increased carrier density and a decrease in the density of states.

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1. Introduction

Light-emitting and laser diodes for the green to near-UV region are based on quantum structures with the InGaN active region \cite{1}. The indium content in the wells is a technological parameter that provides a straightforward access for the change in the color of the light-emitting device. However, the redshift of the emission peak with increase in indium content is caused not only by a simple alloying, but also by carrier localization due to difference in In and Ga atomic...
size, compositional clustering of the material with higher In content, or even complete phase separation [1]. Another effect leading to an additional redshift in InGaN/GaN multiple quantum wells (MQWs) is strong built-in electric field created by large piezoelectric and spontaneous polarization and resulting in spatial separation of electrons and holes. The role of carrier localization enhances with In content [2, 3] however this impact is unclear due to correlated changes in the built-in field, especially for higher In content, when pronounced clustering is expected.

The built-in field can be completely screened by free carriers [4]. Here we present an experimental study of spontaneous and stimulated emission of highly excited InGaN/GaN MQWs with a high In content, varying from about 0.22 to 0.3. High excitation conditions (the created carrier density higher than $10^{19}$ cm$^{-3}$) enabled us to achieve complete screening of the built-in field by free carriers in a broad range of the excitation density. This allowed for the evaluation of the band potential fluctuations for high In content MQWs and for study of the recombination dynamics within the tail of the density of states.

2. Experimental

The undoped MQW structures were grown on sapphire substrates by metalorganic chemical vapor deposition. The growth temperatures of 1020 and 720°C were used for GaN and InGaN, respectively. The structures consisted of five periods of 10 nm thick GaN barriers and In$_x$Ga$_{1-x}$N wells grown over a GaN buffer. A series of MQWs with the nominal In content $x$ varying from about 0.22 to about 0.30 were investigated. The samples were excited by the fourth harmonic (photon energy 4.66 eV) of the actively-passively mode-locked YAG:Nd$^{3+}$ (yttrium aluminum garnet) laser (pulse duration 20 ps, repetition rate 2.7 Hz, maximum pump energy 40 µJ). The size of the excitation spot was approximately $1.2 \times 1.2$ mm$^2$. The emission was dispersed by a 0.4 m grating monochromator. A toluene optical Kerr shutter was used for temporal resolution (20 ps) of luminescence. The experiments were carried out at room temperature.

3. Results and discussion

The emission properties of highly excited InGaN/GaN MQWs can be interpreted by recombination of partly localized carriers under screened built-in field [5, 6]. Thus, high-power excitation spectroscopy can reveal the impact of fluctuating potential on both spontaneous and stimulated emission of InGaN/GaN MQWs with the different mean In content. Figure 1 depicts time-integrated spontaneous emission spectra of InGaN/GaN MQWs for a moderate excitation density 0.3 mJ/cm$^2$ observed in backscattering direction (a) and laterally recorded spectra of stimulated emission for 2.8 mJ/cm$^2$ excitation (b). The spontaneous emission spectra of the investigated structures show two main bands originating from the QW and barrier/buffer layers. The higher-energy band can be attributed to band-
-to-band recombination in the electron–hole system [7]. The QW related band is attributed to radiative recombination of partially localized carriers within the randomly distributed energy potential of the QW [5]. The QW related band steadily redshifts with increase in the In content, while the well/buffer related band is positioned at the same photon energy in the vicinity of the GaN band gap. At moderate excitation, the total intensity of spontaneous emission monotonously increases for $x \geq 0.22$ (Fig. 1a). Such a behavior can be related to fluctuations of the band potential in the well, since carrier localization decreases the nonradiative capture rate. This is proved by a correlation in the luminescence decay time on slow relaxation stage with In content (Fig. 2a). A similar increase in band potential fluctuations for $x \geq 0.22$ shows local states filling, which is monitored by a dynamic blueshift of the luminescence band with excitation density (Fig. 2b) as well as by a transient redshift showing depopulation within the local states (Fig. 2c).

In disordered materials, fluctuations of band potential result in a smeared density of states at the band edge. Therefore stimulated emission occurs on the high-energy side of the spontaneous band (Fig. 1b). The blueshift of the stimulated emission band in respect of the spontaneous one reflects the magnitude of the band potential fluctuations within QWs [6]. The energy difference between the peaks of stimulated and spontaneous emission shows an increase in band potential fluctuations for structures with a higher In content (Fig. 2d).

All above-mentioned experimental features indicate on a steady increase in compositional disorder for $x \geq 0.22$. It should be noted that the highest stimulated emission is observed for MQWs containing $x \approx 0.25–0.27$. It might be speculated that in disordered material, stimulated emission appears from the upper local-
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Fig. 2. Variation of the luminescence parameters with In content in InGaN/GaN MQWs. Decay time on the slow-relaxation stage (a); dynamic peak shift with a 10-fold increase in excitation intensity (b); difference between the peak position of spontaneous and stimulated emission $E_{st} - E_{sp}$ (c); transient peak shift $E_{(0ps)} - E_{int}$ (d).

ized states, when an adequate inverse population is achieved. Carrier localization enables one to reach a higher carrier density, thus the efficiency of stimulated emission enhances with $x$. However, the further increase in compositional disorder leads to a significant reduction in density of states, which decreases the optical gain coefficient and total lateral emission.

In conclusion, the emission properties were studied in InGaN/GaN MQWs with nominal In content $x$ varying from 0.22 to 0.30 under conditions of screened built-in field by free carriers. Enhanced spontaneous emission was found for $x \geq 0.22$ due to carrier localization within fluctuating band potential. Meanwhile the intensity of stimulated emission was found to have a nonmonotonous dependence on $x$. We attribute the In content dependence of the stimulated emission intensity to a trade-off between an increased carrier density and a decrease in the density of states. This implies that an optimal indium content for InGaN/GaN lasing structures can be established.

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References


