phys. stat. sol. (a) **192**, No. 1, 49–53 (2002)

Time-Resolved Studies of InGaN/GaN Quantum Dots

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(Received February 15, 2002; in revised form April 16, 2002; accepted April 20, 2002)

PACS: 73.21.La; 78.47.+p; 78.55.Cr; 78.67.Hc

We report on structural and optical properties of InGaN inclusions in a GaN matrix. High-resolution transmission electron microscopy (HRTEM) studies and HRTEM image processing indicate the spontaneous formation of ultra-dense arrays of InGaN disc-like nano-inclusions. Non-resonant pulsed excitation results in a broad photoluminescence (PL) peak, which shifts towards longer wavelengths with time, demonstrating also an increase in decay time at the peak maximum. In contrast to that, resonant excitation into the non-resonant PL maximum results in an evolution of a sharp resonant PL peak, having a spectral shape defined by the excitation laser pulse and a radiative decay time close to that revealed for PL under non-resonant excitation. No spectral diffusion or PL peak energy shift caused by reduced piezoelectric screening at lower overall concentrations of non-equilibrium carriers was found. These observations give a clear proof of the quantum dot nature of PL in our InGaN/GaN samples.

Introduction Recently there is a strong interest in physics of radiative recombination processes in InGaN/GaN heterostructures stimulated by the development of InGaN-based high-brightness light emitting diodes (LEDs) [1] and lasers [2] operating in the violet to visible spectral ranges. In spite of extensive research in this area in recent years (see e.g. [3] and references therein), up to now there still exist conflicting interpretations of the optical processes in these structures.

Some researchers [4–6] proposed, that InGaN insertions represent arrays of quantum dots (QDs). These QDs were proposed to be responsible for the good luminescence properties of InGaN-insertions [2], as they suppress carrier diffusion towards defects and were claimed to be crucial for lasing in InGaN-based structures. Indeed, HRTEM studies confirmed the existence of ultradense arrays of small (3–8 nm) In-rich disc-like inclusions [7]. The nature of the radiative recombination mechanism crucially affects all the basic laser device parameters: transparency current density, material and differential gain, importance of k-selection rules, stability of the threshold current, etc., and it is crucial for adequate device design. Nevertheless there exists a conflicting set of optical data, which allowed many researchers to conclude, that photoluminescence (PL) properties of wurtzite InGaN insertions may be well-explained by a QW-like material distri-

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 0031-8965/02/19207-0049 $ 17.50+.50/0

bution [8, 9]. Indeed, many groups reported a long wavelength shift of InGaN PL after non-resonant excitation accompanied by a significant increase in the PL decay time [10–12]. Screening of the piezoelectric potential separating electrons and holes at higher excitation densities was proposed to be responsible for this behavior.

In this work we performed high-resolution transmission electron microscopy (HRTEM) and time-resolved (TR) PL studies for InGaN/GaN structures. Our data fully confirm the QD nature of InGaN insertions and point to ultrasmal size and very high density of the QDs.

Experimental The structures studied in this work were grown on a (0001) *c*-plane sapphire sustrate by metal-organic chemical vapour deposition (MOCVD). The structure consists of a 2.5 μm GaN buffer layer, an active region with 10 periods InGaN/GaN superlattice and a 0.07 μm GaN cap layer. The formation of the 3 nm InGaN/7 nm GaN superlattice was achieved by periodic substrate temperature variations from 790 °C to 885 °C and from 820 °C to 915 °C for two different structures [13]. The TR PL was measured at 2 K under tunable dye laser excitation with a repetition rate of 3.79 MHz at wavelengths of 291 nm and 424 nm with an excitation density of 600 W/cm^2. Registration of TR PL was performed using a fast response multi-channel photomultiplier. The HRTEM studies were carried out using a Philips CM20 FEG/ST electron microscope operating at 200 kV.

Results The insertion in Fig. 1 presents a grayscale-coded map of the local indium content obtained by DALI processing of a HRTEM image [14] for the structure, which InGaN insertions formed at 820 °C. The lateral size of the nanoislands is about 3 nm and the local In content in these regions is at least twice higher than the average In content. The dot shape was found to be similar for both samples, while the dot density and average In composition in the QDs were lower for the sample grown at a higher temperature. There is a high spread in size and in composition of the InGaN nanodomains, which is consistent with a broadening of the PL line up to 200 meV. More details on structural properties of nanodomains are given in [15]. The PL decay process under nonresonant excitation was highly nonexponential and could be easily fitted by a stretched exponent:

$$I(t) = I_0 \exp\left[-\left(\frac{t}{\tau^*}\right)^{\beta}\right], \tag{1}$$

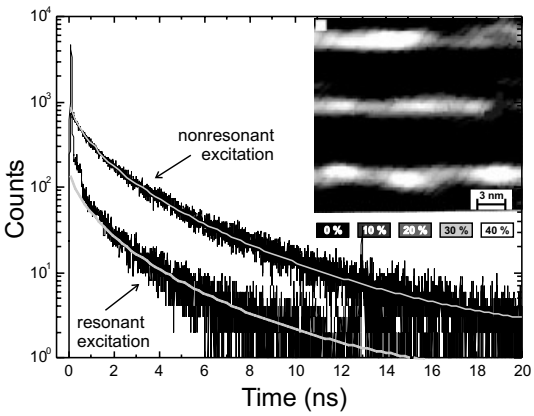

Fig. 1. Time-resolved photoluminescence (PL) for a structure grown at 820 °C under nonresonant and resonant excitation at 428 nm. Gray lines are corresponding to the fitting by Eq. (1). The inserts show a grayscale-coded map of local indium content in the same structure

where $I(t)$ is the PL intensity as a function of time, and β and τ^* are the stretching parameter and the time constant, respectively. The stretched exponential behaviour is usually used in a heavily disordered media [16, 17]. Relaxation processes have the same nature either under nonresonant or under resonant excitation as the PL decays have essentially the same shape at different excitation wavelengths.

As it can be clearly seen from Fig. 2, the values of the stretching parameter depend only weakly on the registration wavelength, while the time constants strongly decrease at the shorter wavelength side of the PL peak with a characteristic onset energy of about 40 meV for both structures. Such behaviour is typical for systems with efficient energy relaxation of carriers from a higher to a lower energy states. This relaxation may be attributed to the hopping of nonequilibrium carriers through the band of localized states due to QW disorder. However, this would mean a very slow evolution of the longer wavelength PL. In our case the PL rise time at any wavelength occurs within the accuracy of the experiment, which is only 5 ps. Thus, a hopping model can be hardly applied in our case. One can attribute the observed behavior to the population of the higher-lying QD excited states followed by an ultrafast transfer towards the ground QD states.

The key experiment, which clarifies unambiguously the nature of the processes involved, is the resonant PL excitation. In the case, when the QW is excited, the carrier transfer towards the states having a lower energy and the process is accompanied by a significant spectral diffusion of the luminescence peak to lower energy. In case of wurtzite-type strained quantum wells, a reduced carrier concentration upon the radiative recombination of non-equilibrium carriers must result in a reduced screening of the piezoelectric potential, further shifting PL towards the longer wavelength side of the spectra and strongly increasing the PL decay time. The long-wavelength PL shift with time is observed in the case of nonresonant excitation (Fig. 3a).

However, neither of these effects was revealed in the case of resonant excitation. Most of the PL intensity, after the exciting laser light is gone, exactly resembles the spectral shape of the excitation source, even the decay time of this signal corresponds to the radiative decay times after nonresonant excitation. This unambiguously shows

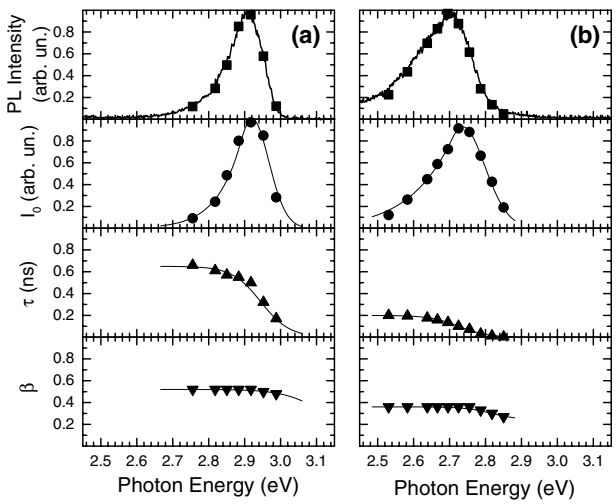

Fig. 2. Time-integrated PL spectra as well as spectral dependencies of fitting parameters from Eq. (1) for structures grown at a) 820 °C and b) 790 °C

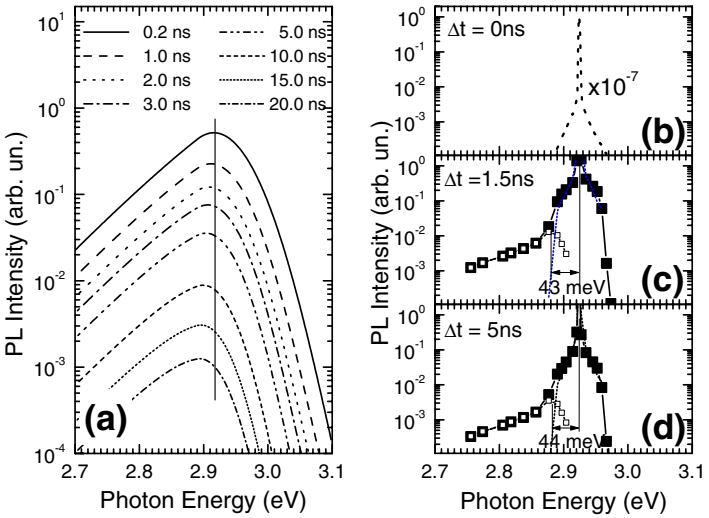

Fig. 3 (online colour). Time-delayed PL spectra of a sample grown at 820 °C at different time delays after a laser pulse under nonresonant (a) and resonant (b–d) excitations. For resonant excitation time delays are 0 ns (b), 1.5 ns (c) and 5 ns (d). At zero moment only scattered laser light (dotted line) can be seen under the resonant excitation. The spectrum shown by the open squares was obtained by subtraction of a normalized laser peak spectrum from the PL spectrum

the QD origin of the signal. The PL peak position does not shift in time and no spectral broadening was observed (Fig. 3b–d) (the appearance of an additional weak shoulder on the longer wavelength side of the PL spectrum is discussed elsewhere [18]). The observed behavior fits exactly to the behavior of resonantly-excited QDs having a delta-function-like density of states, small size enabling significant energy separation between sublevels, and no exciton or carrier transfer at low temperatures.

Conclusion Our results clearly demonstrate a quantum dot origin of InGaN luminescence in our samples. The size and the shape of the InGaN domains revealed in electron microscopy agree well with the PL spectral features. No signs of carrier diffusion to lower states or piezoelectric potential screening were revealed in our structures.

Acknowledgements This work was supported by the DFG Graduiertenkolleg "Collective phenomena in solids", RFBR and grant "NATO for Peace". I.L.K. thanks the Alexander von Humboldt Foundation. N.N.L. acknowledges DAAD Guest Professorship and equipment donation from Alexander von Humboldt Foundation.

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