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Photoluminescence spectroscopy for the evaluation of band potential roughness of InGaN active layers

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Photoluminescence spectroscopy in combination with Monte Carlo simulation of exciton hopping is demonstrated to be a valuable tool for quantitative analysis of the band potential profile in active layers for InGaN-based light emitters. Recently proposed double-scaled potential profile model is used to reveal the scale of potential fluctuations in the individual In-rich regions as well as the dispersion of the average exciton localization energy in these regions. The influence of the different potential fluctuation scales on the stimulated emission threshold and luminescence decay time of highly excited InGaN active layers is studied.

Keywords: photoluminescence, InGaN quantum wells, Monte Carlo simulation, exciton hopping.

1. Introduction

InGaN quantum wells serve as the active layers of blue-UV light-emitting diodes (LEDs) and semiconductor laser diodes (LDs) available on the market. The major factor enhancing the output efficiency of InGaN-based LEDs is carrier/exciton localization in the band potential fluctuations induced by inhomogeneous distribution of indium. The localization restricts carrier motion, and, consequently, the probability of reaching nonradiative recombination sites by the carriers. Although not completely unveiled, the role of localization in InGaN LDs can be significant too. Optimization of the output efficiency of the InGaN light-emitting devices requires an accurate quantitative analysis of the potential fluctuations of their active layers.

2. Proposed treatment

In this work, we evaluate the band potential roughness in InGaN QWs of different thickness and study its influence on the luminescence decay time and the threshold of stimulated emission.

The $In_{0.12}Ga_{0.88}N/In_{0.03}Ga_{0.97}N$ QWs under study were deposited by plasma-assisted molecular beam epitaxy (MBE) on pressure-grown bulk GaN substrates with a dislocation density of about 10^2 cm^{-2}. Three samples consisting of seven 2.5 nm, 4.0 nm and 6.0 nm thick InGaN QWs, respectively, separated by 9 nm thick InGaN barriers, were chosen for this study. The barriers were doped with Si ($\sim 1\times10^{19}$ cm^{-3}) to screen the electric field in the polarized QWs.

Photoluminescence (PL) spectroscopy of the QWs was performed under low-intensity excitation by 10 mW continuous wave He-Cd laser and under high excitation by pulsed megawatt-power YAG:Nd lasers (pulse duration 10 ns and 20 ps). The picosecond pulses were used to measure luminescence decay transients. The PL signal was dispersed and detected by means of a double monochromator and a UV-enhanced photomultiplier.

The quantitative analysis of the band potential roughness in InGaN QWs was accomplished by measuring temperature evolution of the linewidth of the near-band-edge PL. Typically for ternary nitride alloys containing In, the PL band peak features a characteristic S shape shown by the dashed line in Fig. 1**a** for 4-nm-thick QWs.

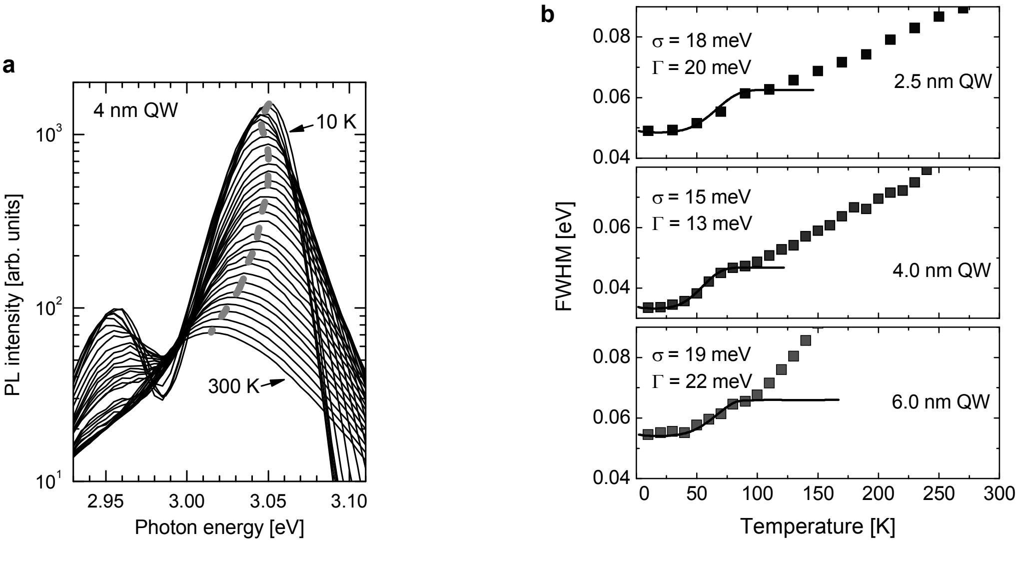

Fig. 1. Temperature evolution of PL spectrum of 4 nm thick InGaN QWs (dashed line indicates PL peak and serves as a guide for the eye) – **a**. PL linewidth (points) as a function of temperature measured in 2.5, 4.0 and 6.0 nm thick MBE grown $In_{0.12}Ga_{0.88}N$ QWs (the lines show results obtained by Monte Carlo simulation of exciton hopping within band potential fluctuations in individual In-rich regions σ that are dispersed in the average exciton energy Γ) – **b**.

The kink at low temperatures indicates localized exciton “hop-like” motion through the minima of potential fluctuations formed by indium clustering and interface roughness [1, 2]. The W-shaped temperature dependence of the PL linewidth (shown by points in Fig. 1**b**) is also of the same origin. The temperature for which a plateau occurs (~80 K) in this dependence might serve as a rough estimate of the scale of potential fluctuations. However, a more sophisticated approach, like Monte Carlo simulation of exciton hopping, is required to describe the linewidth evolution at low temperatures quantitatively [1]. The lines in Fig. 1**b** show the results of the Monte Carlo simulation obtained by taking potential fluctuation profile to be composed of two scales. We stress that the double-scaled profile model is prerequisite to obtain a fair correspondence between the simulations and experiment. This model implies localized exciton hopping within band potential fluctuations in individual In-rich regions (on the scale σ) that are dispersed in the average exciton localization energy Γ [1, 2]. In the simulation, the exciton hopping rate was defined by the Miller–Abrahams expression generally used to describe hop-like exciton motion between the localized states in disordered compounds. The potential profile roughness in terms of σ and Γ attained the values of 18 meV and 20 meV in 2.5 nm QWs, 15 meV and 13 meV in 4.0 nm QWs, and 19 meV and 22 meV in 6.0 nm QWs, respectively.

The impact of the potential roughness on the stimulated optical transitions in our InGaN QWs was examined by measuring excitation power dependence of the PL spectra using the thin-stripe technique. Thin excitation stripe (30×1000 μm) was focused by a cylindrical lens onto the front surface of the sample near its edge, whereas the lateral emission propagating along the stripe was collected. Spectrally integrated PL intensity is shown as a function of the pump density for different-thickness QWs in Fig. 2. The inset of Fig. 2 displays excitation-power-induced changes to the spectrum, which undergoes characteristic narrowing indicating the lasing. Surprisingly, the stimulation threshold (30±5 kW/cm^2) was found to be insensitive to the different potential roughness present in QWs with different thickness. This result contradicts numerous reports on a strong dependence of the threshold pump density on the QW thickness [3] and the degree of potential fluctuations [4]. However, this contradiction can be explained by assuming that the lasing in our MBE grown InGaN QWs is caused by extended states rather than by localized ones. Our estimation based on the excitation-power induced PL blueshifting [5] yielded the localized states density of about 1×10^{18} cm^{-3} that is one order of magnitude lower than the carrier density at the lasing threshold. Luminescence intensity decay in the QWs measured at high -intensity photoexcitation, which ensured conditions similar to those in operating LDs, is shown by points in Fig. 3. The initial fast carrier relaxation (0–300 ps) in all transients is dominated by stimulated optical transitions [6], whereas the slow relaxation at a later decay stage is governed by spontaneous emission. The decay time of spontaneous luminescence extracted from a single-exponent fit (lines) was found to be quite similar (ranging from 650 to 720 ps) in all QW structures studied. The luminescence decay time is somewhat larger in the 4 nm thick sample exhibiting the lowest potential fluctuations. It is worth noting that these decay times of free

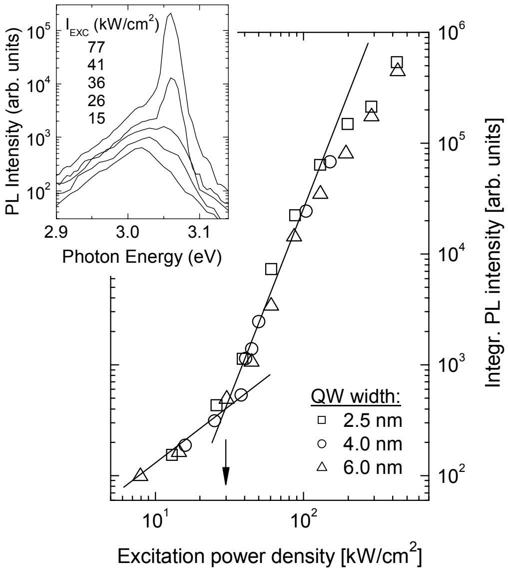

Fig. 2. Integrated PL intensity as a function of excitation power density in 2.5, 4.0 and 6.0 nm thick $In_{0.12}Ga_{0.88}N$ QWs with the threshold of stimulated emission at 30 kW/cm^{-2} (arrow). Inset displays the excitation power dependence of PL spectrum in 4 nm QWs measured in the stripe-pump configuration at room-temperature.

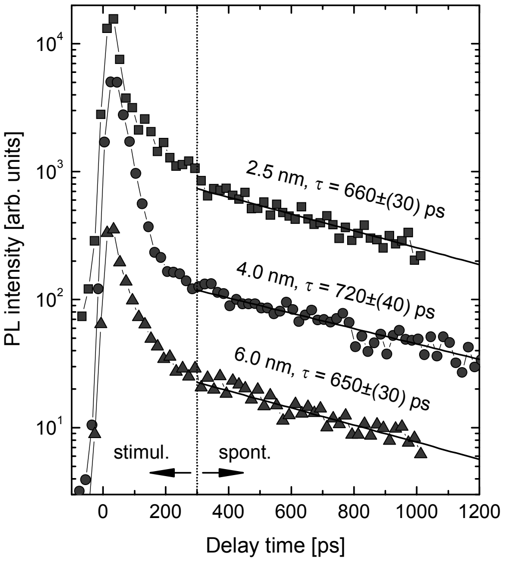

Fig. 3. Temporal evolution of PL in MBE grown $In_{0.12}Ga_{0.88}N$ QWs of different well thickness (indicated) at room temperature. The transients are arbitrarily shifted in vertical direction for clarity. Lines are single-exponent fits of spontaneous luminescence kinetics with the characteristic decay time constants τ. The vertical dashed line at 300 ps separates the fast initial relaxation stage of PL transient from the slow one at the later stage caused by stimulated and spontaneous optical transitions, respectively.

carriers are among the longest ever measured at high excitations in InGaN QWs. Such slow luminescence decay is possibly caused by significant reduction of nonradiative recombination centers in the high structural quality QWs grown by MBE on bulk GaN substrates.

3. Conclusions

In summary, PL spectroscopy combined with Monte Carlo simulation of exciton hopping was shown to be useful in quantifying the roughness of band potential profile in InGaN active layers. The deduced scales of potential fluctuations (13–22 meV) in MBE grown QWs are different in QWs of different thickness (2.5–6.0 nm). However, this difference has almost no impact on the stimulation threshold (30 kW/cm^2) and luminescence decay time (~700 ps). This insensitivity is most likely caused by the domination of the extended states over localized ones in the carrier radiative recombination at high photoexcitation conditions.

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