Two Mechanisms of Blueshift of Edge Emission in InGaN-Based Epilayers and Multiple Quantum Wells

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“Blue” temperature-induced shift and band-tail emission in InGaN-based light sources
Two mechanisms of blueshift of edge emission in InGaN-based epilayers and multiple quantum wells

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We present the results of a comparative photoluminescence (PL) study of GaN and InGaN-based epilayers, and InGaN/GaN multiple quantum wells (MQWs). Room-temperature PL spectra were measured for a very broad range of optical excitation from 10 mW/cm² up to 1 MW/cm². In contrast to GaN epilayers, all In-containing samples exhibited an excitation-induced blueshift of the peak emission. In addition, the blueshift of the emission in the InGaN epilayers with the same composition as the quantum well was significantly smaller. The comparison of the blueshift in the “bulk” InGaN and in the MQWs allowed us to separate two different mechanisms responsible for this effect: (i) filling of the localized states in In-rich areas and (ii) screening of the polarization electric field in strained MQW structures. © 2002 American Institute of Physics. [DOI: 10.1063/1.1433164]

Indium-based III-nitride semiconductors are of great interest for applications in nearly all commercial light-emission systems since they allow band-gap tunability from the UV to the visible region of the spectrum. Ternary InGaN-emission systems since they allow band-gap tunability from interest for applications in nearly all commercial light-

ers of InGa1−xN or InGa1−xN/GaN MQWs. They were deposited on a 1-μm-thick GaN layer on a basal plane (0001) sapphire substrate using metalorganic chemical-vapor deposition. The 1-μm-thick GaN epilayer was grown at 980 °C under a pressure of 76 Torr. More detailed information on the growth conditions may be found elsewhere. The InGaN/GaN MQW sample consisted of six periods of 4-nm-thick quantum wells (In0.12Ga0.88N) and 6-nm-thick GaN barriers, which were deposited at 731 °C and capped by a 40-nm-thick GaN layer. The PL was excited using a cw He–Cd laser (λ = 325 nm, maximum power ~40 mW) or by a pulsed N2 laser (λ = 337 nm, pulse duration of 0.6 ns, energy per pulse ≤5 μJ). The laser beam was focused to a spot of ~0.1 mm². We were thus able to reach excitation power densities of ~10⁻³–40 W/cm² for the cw laser pump, and up to ~10 MW/cm² for the pulsed N2 laser excitation. The emitted light was collected perpendicular to the sample surface in a backscattering direction. The luminescence signal was analyzed by a SPEX550 monochromator with an UV-enhanced liquid-nitrogen-cooled charge-coupled-device array. All the PL measurements were performed at room temperature.

At low excitation, PL spectra shapes are not excitation sensitive. Figure 1 shows typical PL spectra for the GaN epilayer (a) and InGaN MQWs (b) under different excitation power densities. At first, the increase of excitation power density does not change either peak position, or shape of the PL spectra for all the samples. In GaN under up to 50 kW/cm² excitation the spectrum is exciton originated; the predominant line corresponds to free excitons, whereas the low-energy wing structure of the spectrum could be explained by the LO phonon replicas. The InGaN samples (both epilayers and MQWs) demonstrate similar stability of
InGaN samples are quite different. When the excitation density in In\textsubscript{0.15}Ga\textsubscript{0.85}N epilayers exceeds 20 W/cm\textsuperscript{2} for the InGaN at half maximum of the PL line is sufficiently broader ~180 meV, and 3 kW/cm\textsuperscript{2} for the InGaN epilayer resolved. This PL spectra behavior for GaN epilayers under He–Cd cw excitation, and the filled ones correspond to N\textsubscript{2} laser pulsed excitation. Arrow 1 marks the excitation power density of complete screening, and arrow 2 shows the validity limit of the model.

At higher excitation the PL properties of the GaN and InGaN samples are quite different. When the excitation power increases, the emission peak in GaN shifts towards lower energies (the maximum shift is ~15 meV), the line broadens, and the structure of the LO phonons is no longer resolved. This PL spectra behavior for GaN epilayers under strong excitation may be explained by a dense electron–hole plasma model, which has been successfully used for a number of highly pumped direct-gap semiconductors.

In contrast to GaN, the PL position both in the InGaN epilayers and InGaN MQWs shifts towards the high-energy side. However, the features of the excitation-induced blue-shift in the bulk InGaN and in the MQW samples are different. As shown in Fig. 2, the total blueshift of the PL peak in bulk InGaN is about 150 meV, whereas it is more than 200 meV for the MQWs. For the MQW sample the blueshift occurs at the excitation power density, which is nearly two orders lower in comparison with the bulk InGaN. The blueshift at very high excitation in both cases demonstrates a remarkably similar behavior (see Fig. 2, experimental points at excitation > ~100 kW/cm\textsuperscript{2}).

When the threshold of 1.0–1.4 MW/cm\textsuperscript{2} is exceeded the stimulated emission line appears, however, for GaN it is located on the long-wavelength side of the spontaneous PL spectrum, while for all InGaN samples it is located on the short-wavelength side [Figs. 1(a) and 1(b), upper curves]. Stimulated emission in all the samples can be explained by band-to-band recombination, whereas spontaneous PL in In-containing samples may be attributed to transitions between the density-of-state tails.

We now analyze the spontaneous PL in the InGaN samples in more detail. We explain the PL behavior differences of the bulk In\textsubscript{x}Ga\textsubscript{1−x}N and In\textsubscript{x}Ga\textsubscript{1−x}N/GaN MQWs by quantum confinement and the predominant strong piezoelectric field in thin quantum wells. Indeed, in hexagonal-nitride MQWs, the quantum-confined Stark effect arises due to the piezoelectric field as well as due to spontaneous polarization.

We analyzed the influence of nonequilibrium carriers on the position of PL spectra in the InGaN/GaN MQWs using a triangular well model. The photoexcited carriers screen the internal field. For an idealized case, neglecting thermal distribution in the bands, the emitted quantum energy \(h\nu\) for band-to-band recombination in a quantum well in the presence of nonequilibrium electron–hole pair density \(n\) can be expressed as

\[
h\nu = E_g(n) - edF(n) + E_e(n) + E_h(n).
\]

Here, \(E_g(n)\) is the carrier-density-dependent forbidden gap, which, taking into account band-gap renormalization, can be expressed as \(E_g(n) = E_g(0) - n\beta^{1/3}\) with \(\beta = 2 \times 10^{-8}\) eV cm; \(d\) is the well width; \(F(n)\) is the internal electric field strength, which can be expressed as \(F(n) \approx F_0 - ned\epsilon\epsilon_0\) with maximum field strength \(F_0\) (in the unexcited sample at limit \(n \rightarrow 0\)) and static relative dielectric constant \(\epsilon\), which has been taken as \(8.5\) and \(10\); and \(E_{e,h}\) is the difference of the lowest-energy level from the triangular well bottom for electrons and holes, respectively, and can be calculated from

\[
E_{e,h} = \left(\frac{\hbar^2}{2m_{e,h}}\right)^{1/3} \left[9\pi\epsilon F(n)\right]^{2/3}.
\]

The effective masses of electrons or holes, \(m_{e,h}\), used in the calculations have been assumed to be 0.25\(m_0\), \(m_h \approx m_0\), respectively. In order to compare our experimental results with the calculations, we expressed \(h\nu\) as a function of excitation power density \(P\) (in MW/cm\textsuperscript{2}), which for the case of
predominantly square-law recombination are related as $n = \sqrt{P \alpha h \nu \gamma}$, where $\alpha$ is the absorption coefficient for laser light and has been taken as $18 \times 10^5$ cm$^{-1}$, and $\gamma$ is the square-law recombination coefficient and has been taken as $19 \times 10^{-11}$ cm$^3$ s$^{-1}$.

The results of these calculations for InGaN/GaN MQWs are illustrated in Fig. 2 by the dotted line. This rather crude model gives remarkably good agreement between experimental data and theoretical estimations for more than six orders of excitation power density (up to $10$ kW/cm$^2$). Note that a number of other effects were not included in our model (we neglected the two-dimensional nature of the system, carrier distribution in the barriers and the wells, excitonic and nonradiative recombination channels, possible recombination coefficient change due to separation of the carriers in wells, etc.). These effects can mainly change the concentration of electron–hole pairs corresponding to a given excitation power density, which results in just the “horizontal” shift of the theoretical curve in Fig. 2. However, this shift is quite small due to a logarithmic scale of the power density axis. In these calculations we have adjusted just two parameters: the maximum electric-field strength $F_0$ and the initial PL peak position. The value of $F_0$ was found to be equal to $1.87 \times 10^8$ V/cm, which is quite similar to other evaluations in similar semiconductor structures.

The estimation of the built-in (mainly piezoelectric) field in In$_{0.15}$Ga$_{0.85}$N/GaN MQWs following Refs. 15 and 21 yields the value of $1.73$ MV/cm, which is close to that obtained experimentally. Complete screening of the field is achieved at carrier densities $\sim 10^{20}$ cm$^{-3}$ (arrow 1 in Fig. 2). Note that the infinite triangular quantum well model is valid only up to a carrier density $\sim 8 \times 10^{19}$ cm$^{-3}$ (arrow 2 in Fig. 2). As can be seen from Fig. 2, the region of excitation power density around $10$ kW/cm$^2$ is the starting point for the blueshift of the PL spectrum of bulk InGaN material, as well as some “slowing” of the blueshift in MQWs. Beyond this pumping level, a very similar PL maximum shift is observed for both the bulk and the MQW samples. This blueshift in In-containing III–N samples may be attributed to band-tail filling by carriers. Recently, we have also observed the clearly resolved blueshift maximum [as a result of the field screening leading to the competition between the second and the last two terms in formula (1)] in quaternary AlInGaN MQWs with a very low fraction of In, and which thus have no localized In-related potentials and tails.

In summary, we have shown that an excitation-induced blueshift of PL spectra is observed in bulk InGaN epilayers, as well as in InGaN/GaN MQWs. The comparison of PL data in GaN, InGaN, and InGaN/GaN MQW samples, as well as the theoretical estimations using the triangular potential-well model have shown that the blueshift observed in bulk materials is caused by filling of band-tail states, which result from potential fluctuations due to an inhomogeneous distribution of In-rich dots. In InGaN/GaN MQWs, with increased excitation levels, the blueshift at first is from screening of the internal electric field (by photoinduced carriers) and later by filling of band-tail states.

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