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

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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/cm^2 up to 1 MW/cm^2. 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.
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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-based single or multiple quantum wells (MQWs) are the key structures for high-efficiency, long-lifetime violet, blue, and green light-emitting diodes and laser diodes.[1–3] In spite of this importance, the edge emission mechanism for these InGaN layers and quantum wells is not yet fully understood. They exhibit a remarkably strong blueshift both with an increase in temperature and in carrier injection.[4–6] In order to explain this blueshift, the idea of band-tail states was proposed with either a Gaussian-like density distribution[7] or localization[8] at quantum dots[9] or disks.[10] Attempts were also made to explain this edge emission peculiarity solely by the presence of a strain-induced piezoelectric field[11] or by a combination of the field and the localized states.[10]

In this letter, we investigate the photoluminescence (PL) of high-quality InGaN-based epilayers and MQWs in a wide dynamic range of excitations. By comparing the PL from InGaN MQWs and thick InGaN epilayers of exactly the same composition as the well material, we were able to separate an excitation-induced blueshift from the band filling of the localized states from the screening of the internal electric field by the nonequilibrium photoexcited carriers. The value of the measured spectral shifts thus enabled us to calculate both the internal electric-field strengths in InGaN MQWs and the carrier density screening of the potential (using a triangular well model).

The samples for this study consisted of either single layers of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N}$ or $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N}$/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.[12] The InGaN/GaN MQW sample consisted of six periods of 4-nm-thick quantum wells ($\mathrm{In}_{0.15}\mathrm{Ga}_{0.85}\mathrm{N}$) 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 ($\lambda = 325$ nm, maximum power $\sim$40 mW) or by a pulsed N_2 laser ($\lambda = 337$ nm, pulse duration of 0.6 ns, energy per pulse $\leqslant 5\ \mu$J). The laser beam was focused to a spot of $\sim$0.1 mm^2. We were thus able to reach excitation power densities of $\sim 10^{-3}$–40 W/cm^2 for the cw laser pump, and up to $\sim$10 MW/cm^2 for the pulsed N_2 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/cm^2 excitation the spectrum is exciton originated:[10] 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

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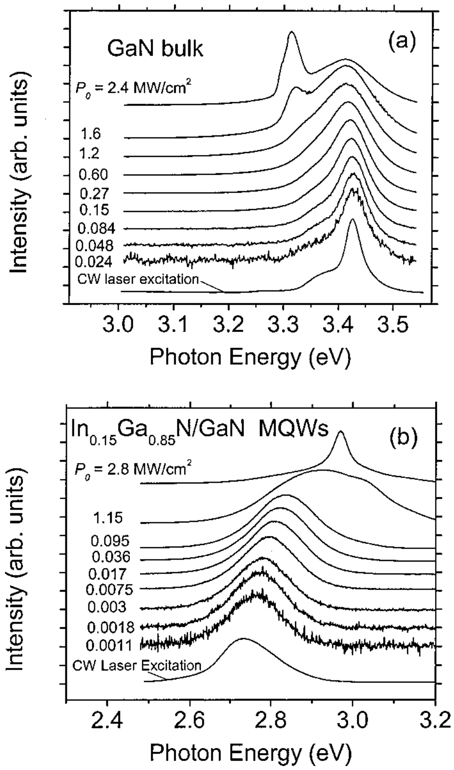

FIG. 1. Photoluminescence spectra of GaN epilayers (a) and $In_{0.15}Ga_{0.85}N$/GaN MQWs (b) under different N_2 laser excitation power densities. The lowest curves correspond to He–Cd cw laser excitation with power density <10 W/cm^2.

the PL spectra with excitation power, however, the full width at half maximum of the PL line is sufficiently broader (~180 meV) and the peak position remains unchanged unless the excitation power density exceeds 20 W/cm^2 for the InGaN MQWs, and 3 kW/cm^2 for the InGaN epilayer (see Fig. 2).

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.[13,14]

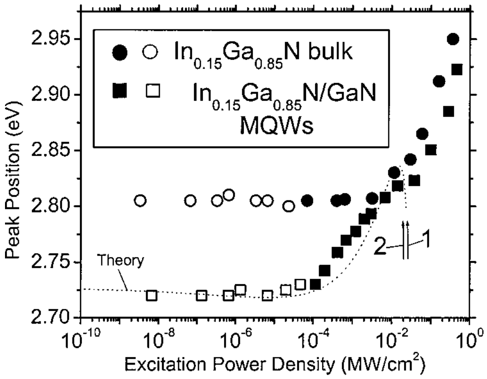

FIG. 2. Photoluminescence peak position dependences on excitation power density in $In_{0.15}Ga_{0.85}N$ epilayers (the upper dependence) and in $In_{0.15}Ga_{0.85}N$/GaN MQWs (the lower one). The open symbols correspond to He–Cd cw excitation, and the filled ones correspond to N_2 laser pulsed excitation. Arrow 1 marks the excitation power density of complete screening of the electric field, and arrow 2 shows the validity limit of the model. The dotted line demonstrates the theoretical dependence of the emission maximum on excitation.

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 blueshift 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 $>\sim 100$ kW/cm^2).

When the threshold of 1.0–1.4 MW/cm^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,[13] whereas spontaneous PL in In-containing samples may be attributed to transitions between the density-of-state tales.[7–10]

We now analyze the spontaneous PL in the InGaN samples in more detail. We explain the PL behavior differences of the bulk $In_xGa_{1-x}N$ and $In_xGa_{1-x}N$/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.[15] 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

$$h\nu = E_g(n) - edF(n) + E_e(n) + E_h(n). \tag{1}$$

Here, $Eg(n)$ is the carrier-density-dependent forbidden gap, which, taking into account band-gap renormalization, can be expressed as[14] $E_g(n) = E_g(0) - \beta n^{1/3}$ 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 ϵ, which has been taken as[5,16] 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[17]

$$E_{e,h} \cong \left(\frac{\hbar^2}{2m_{e,h}} \right)^{1/3} \left[\frac{9\pi eF(n)}{8} \right]^{2/3}. \tag{2}$$

The effective masses of electrons or holes, $m_{e,h}$, used in the calculations have been assumed to be $0.25m_0$, $m_h \cong m_0$, respectively.[5,16] 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^2), which for the case of predominantly square-law recombination are related as $n = \sqrt{P\alpha/h\nu\gamma}$, where α is the absorption coefficient for laser light and has been taken as[18] $1.5\times10^5\ \mathrm{cm}^{-1}$, and γ is the square-law recombination coefficient and has been taken as[19] $4.8\times10^{-11}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$.

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 1.87×10^6 V/cm, which is quite similar to other evaluations in similar semiconductor structures.[5,10,20] The estimation of the built-in (mainly piezoelectric) field in $\mathrm{In_{0.15}Ga_{0.85}N/GaN}$ 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 $\sim10^{20}\ \mathrm{cm}^{-3}$ (arrow 1 in Fig. 2). Note that the infinite triangular quantum well model is valid only up to a carrier density $\sim8\times10^{19}\ \mathrm{cm}^{-3}$ (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.[7–10] 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.[22]

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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