

MECHANISM FOR RADIATIVE RECOMBINATION IN $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}/\text{GaN}$ MULTIPLE QUANTUM WELL STRUCTURES

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Abstract

We present a study of the radiative recombination in $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}/\text{GaN}$ multiple quantum well samples, where the conditions of growth of the InGaN quantum layers were varied. The piezoelectric field as well as short range potential fluctuations are screened via different mechanisms by donor electrons and excited electron-hole pairs. These effects account for a large part of the spectral shift with donor doping (an upward shift of the photoluminescence (PL) peak up to 0.2 eV is observed for a Si donor density of $2 \times 10^{18} \text{ cm}^{-3}$ in the well), with excitation intensity and with delay time after pulsed excitation (also shifts up to 0.2 eV). It appears like 2-dimensional screening of short range potential fluctuations is needed to fully explain the data. We suggest that excitons as well as shallow donors are at least partly impact ionized by electrons in the rather strong lateral potential fluctuations.

Introduction

InGaN/GaN multiple quantum well (MQW) structures provide the active medium for the violet III-nitride lasers [1]. The recombination processes in these structures therefore are of interest to determine. It has recently been claimed that the broad photoluminescence (PL) emission spectra in these structures are due to an inhomogeneously broadened envelope of excitonic emissions from “quantum dots (QDs)” [2]. These QDs are supposed to consist of regions of a size 3-5 nm having a bandgap much lower than the alloy matrix, due to segregation of In during growth [2]. Other recent work points towards the importance of the piezoelectric (PZ) field as the dominant mechanism governing the QW electronic structure and the recombination processes in these structures [3,4]. In the previous studies the In composition x in the QWs was typically smaller than 0.15, and the recombination processes were described in terms of QW excitons [2-4]. In this work we report on a study of the recombination dynamics in $\text{In}_x\text{Ga}_{1-x}\text{N}$ MQWs over the temperature range 2 - 300 K. We conclude that both the PZ field and potential fluctuations are important at this In composition.

Samples and experimental procedure

The $\text{In}_x\text{Ga}_{1-x}\text{N}$ MQW samples with 5 QWs were grown with MOCVD with an In fraction x about 0.15 for all samples studied in this work. The QW thickness was 3.0 nm, with a GaN barrier thickness of 6.0 nm. The detailed growth procedures have been described elsewhere [5]. The structural quality was investigated for selected samples. It was found that the interfaces of the QW were rather smooth, the lower GaN/InGaN interface was virtually atomically flat in high

resolution TEM pictures, while the upper InGaN/ GaN interface had a short range fluctuation of about 1-2 monolayers.

The spectral PL experiments discussed here involve continuous wave (cw) laser excitation, as well as more advanced transient studies. For the transient PL measurements we had several UV laser systems available, tunable both above and below the GaN bandgap. For detection we employ photon counting techniques for the time domain 100 ps to 4 μ s, and a streak camera for the faster time domain 15 ps to 2 ns. PLE spectra were obtained with a Xenon lamp and a monochromator as light source.

Transient photoluminescence spectra

We have investigated the transient behavior of the PL over a large time scale and a large dynamic range. The results for the shorter time scale will be reported separately [6]. The PL decay over a longer time scale (ns - μ s) has been measured with photon counting technique over the entire spectral range covered by the PL emission. Typical results are shown in Figs 1(a) and 1(b) for selected samples at low temperature. In Fig 1 we compare the time resolved spectra of an undoped (a) and a doped sample (b) ($n = 2 \times 10^{18} \text{ cm}^{-3}$), respectively. It is noted that the spectra for the doped samples are considerably narrower, with the impression that the lower energy part of the spectrum in Fig 1 (a) is simply missing in the spectrum 1(b). This will be discussed further below. There is also a rather strong shift downwards for the peak energy with delay time, which is expected due to screening by the excited e-h pairs, and also due to spectral diffusion from carrier hopping before recombination.

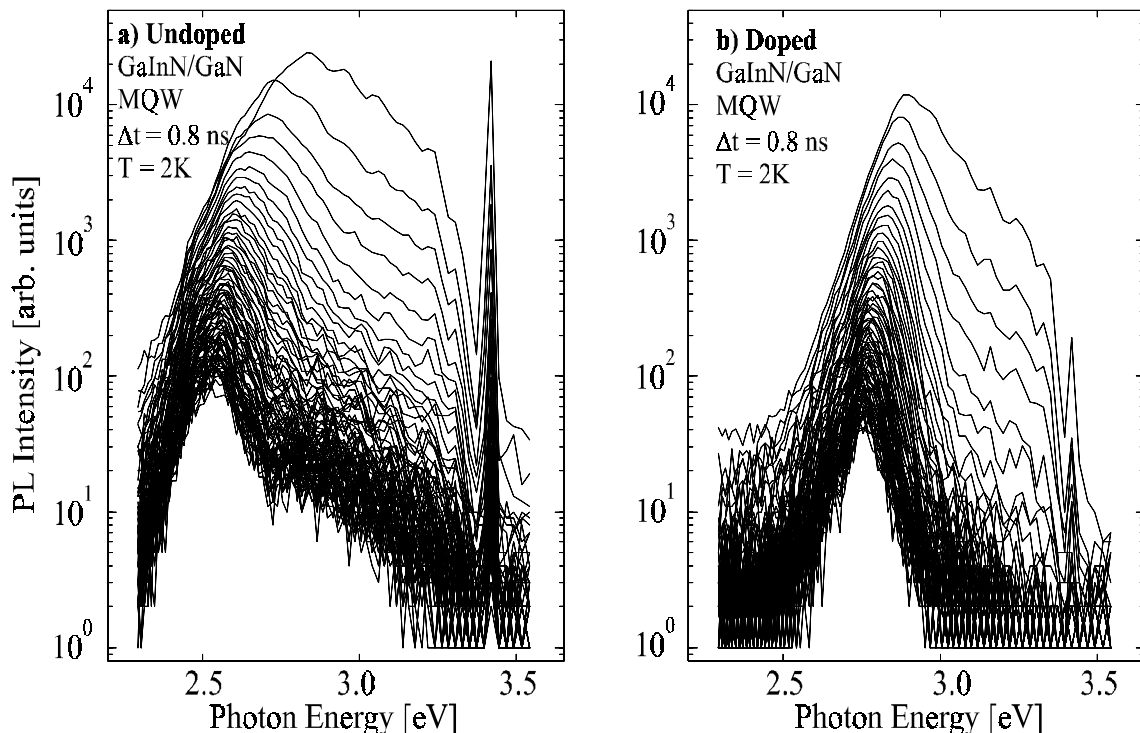


Fig. 1. Timeresolved PL spectra, with 0.8 ns between each spectrum, for two MQW samples obtained at 2 K with excitation at 3.6 eV. In a) are shown the spectra for a undoped sample while b) shows the corresponding data for a doped sample. Note the strong spectral shift between the two samples.

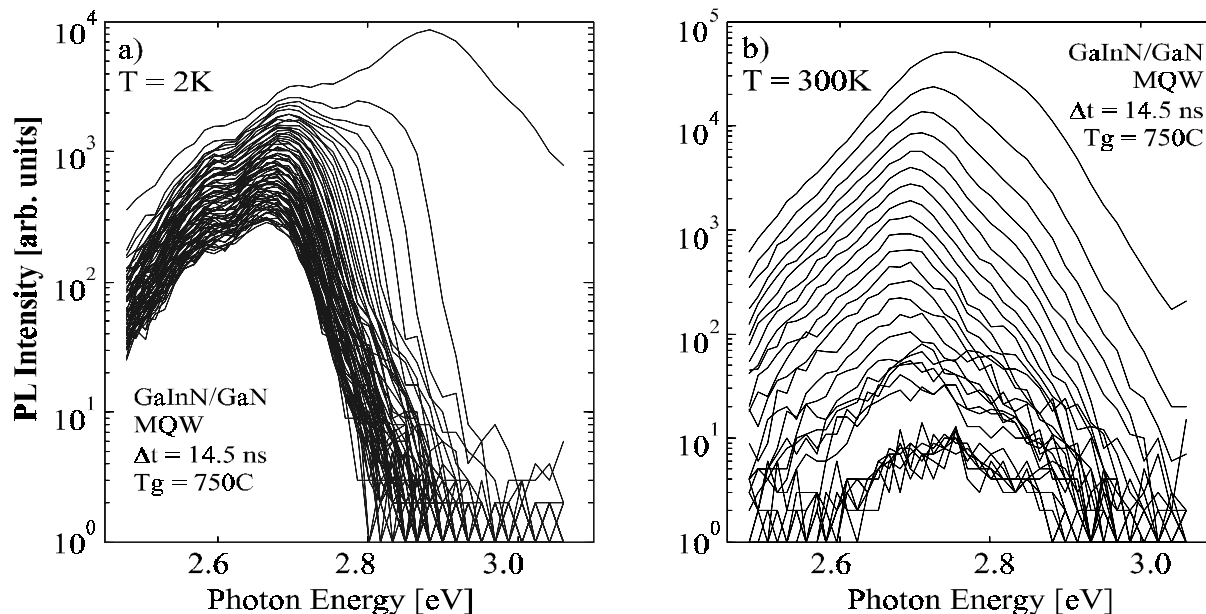


Fig. 2. Timeresolved PL spectra for an MQW sample with the InGaN layer grown at 750 C, obtained at 2 K (a) and at 300 K (b), respectively. The spectra at 300 K are broadened towards higher energy, and also have shorter decay time.

In Fig 2 we show a comparison of the timeresolved spectra at 2 K and 300 K for an MQW sample with the InGaN layers grown at 750 °C. The spectra are quite similar concerning downward shift with delay time, but the 300 K spectrum is broadened considerably and selectively on the high energy side, even at long delay times.

This may be understood by the presence of strong localization potentials in the system. Also, the decay is faster at 300 K, presumably due to the presence of a nonradiative recombination component at this temperature. The low temperature decay times are identified as the radiative decay times in the MQWs, since they are independent of the excitation power.

The values of the decay times are not welldefined at any temperature, since the decay curves are not exponential. Defining an effective decay time (measured as the time for a decrease of intensity by a factor 1/e of the initial value) we obtain a value about 150 ns at 2 K for the sample with the InGaN layer grown at 780 C [6]. The effective decay times are illustrated at three different temperatures in Fig 3. The temperature dependence of the decay times is not strong; in fact it seems like the radiative decay times are rather independent of temperature, so that the faster decay at 300 K is due to the presence of a nonradiative component at this temperature.

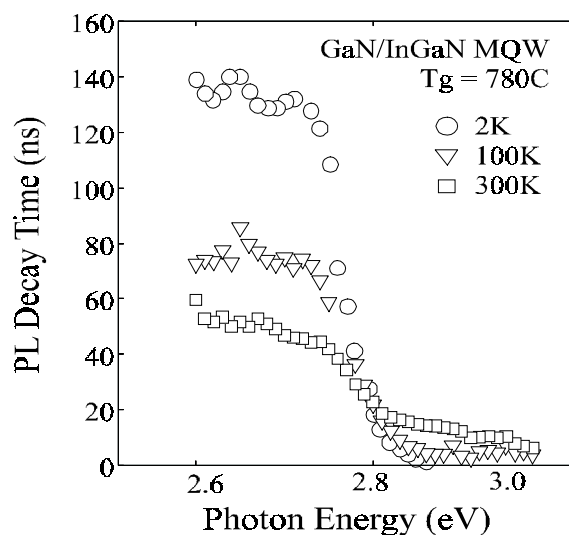


Fig. 3. Effective decay times as a function of detection photon energy for an undoped sample with the InGaN growth temperature 780 °C.

Discussion.

The shape of the broad PL spectrum from these InGaN MQWs does not give any direct information on the recombination mechanism. The considerably narrower PL peaks observed in similar systems with lower In composition can more consistently be ascribed to QW excitons [2,4,7]. We suggested earlier that due to the rather low fields necessary for impact ionization of excitons in these QWs, excitons may be largely transformed into separated electron-hole pairs when they encounter the strong local lateral fields due to the potential fluctuations in the QW [8]. Further, the importance of the piezoelectric field effects vs localization in potential fluctuations for the PL lineshape has to be determined.

The energy of the QW bandgap in our samples is estimated as 3.05 eV at 2 K [6]. The energy position of the broad PL peak is consistent with a downshift expected from a strong piezoelectric field across the QW [3]. The peak position observed for undoped MQW samples in low intensity stationary PL spectra (about 2.65 eV [6,8]), as well as in the timeresolved spectra at long time delay after the excitation pulse, are in good agreement with Ref. 3 for MQW samples with a similar In composition. The downshift from the estimated QW bandgap energy is about 0.4 eV. We therefore conclude that the PL peak position is consistent with a major effect of the piezoelectric field across the QW. The value of the PZ field necessary to explain the large downshift is about 1 MV/cm [3,4]. The width of the peak strongly indicates the presence of substantial potential fluctuations in the QWs, however.

The rather large upward shifts of the PL peak position with Si donor doping in the QWs [8], as well as with excitation intensity, points towards strong effects of carrier screening of the PZ field. We have simulated to what extent the donors or the excited carriers can screen out most of the PZ field. The most extreme screening effect would occur if one puts all the donor ions on one side and the electrons on the other side of the QW (this would be a gross overestimation of the ability of the carriers to screen the field). This screening would only reduce the potential drop across the well with 12%, for the highest Si doping concentration we have studied, $2 \times 10^{18} \text{ cm}^{-3}$. A similar modeling with ordered electron-hole pairs at this density, as required to simulate the spectral shifts with intensity and delay time, gives a similar result. A doping (or excited electron-hole pair) concentration of the order 10^{19} cm^{-3} would be needed to reproduce the observed shifts (up to 200 meV upshift with Si doping at $2 \times 10^{18} \text{ cm}^{-3}$), if these shifts were due to screening of the PZ field alone. To explain the screening effects in this case we need to assume rather strong potential fluctuations in the QWs, in addition to the PZ field.

We have made some simple modeling to find out if the doping induced blue shift of the PL spectrum [8] could be reproduced from lateral screening of potential fluctuations in the QW. In a 3D system we know that the screening increases with carrier concentration; the Thomas-Fermi screening length decreases. In a 2D system the Thomas-Fermi screening length is unaffected by a change in carrier concentration. However, the Thomas-Fermi screening length only describes the screening of a slowly varying potential or the resulting potential far away from the center of the potential. The higher the carrier concentration the better the system screens a rapidly varying potential or the core of the potential. This means that the effective depth of a potential well caused by fluctuations can vary with doping or carrier concentration. (In the present discussion we assume that the shallow donor electrons are ionized, e g by the strong fluctuating electric field in the QW).

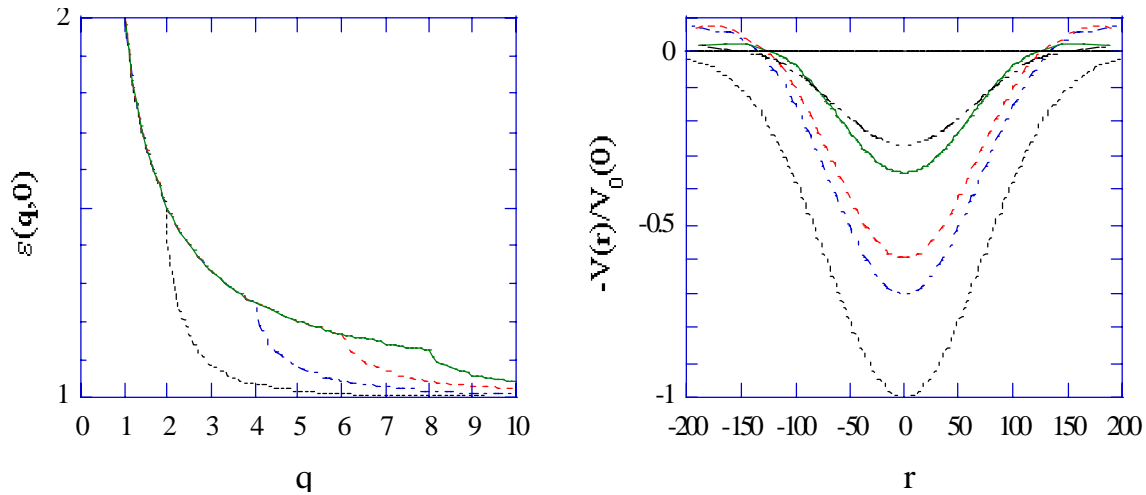


Fig. 4 (a). The general behavior of the static 2D dielectric function. Each curve is for a different carrier concentration. The dotted, dash-dotted, dashed and solid curves are for progressively higher carrier concentrations. All curves coincide for small momentum. Fig. 4 (b). The screening effect on a Gaussian potential of width 100 Å in one of the quantum wells. The dotted curve is the bare potential. The dash-dotted, dashed, full and dash-triple-dotted curves are for the carrier concentrations $2 \times 10^{17} \text{ cm}^{-3}$, $4 \times 10^{17} \text{ cm}^{-3}$, $2 \times 10^{18} \text{ cm}^{-3}$ and $1 \times 10^{19} \text{ cm}^{-3}$, respectively. All curves have been scaled to the value at the center of the unscreened potential.

The behavior of the 2D, low temperature, RPA dielectric function is shown in figure 4 (a), which represents the dielectric function for different electron concentrations. We see that all curves are equal for small momentum but deviates for large momentum. They start to deviate when the momentum exceeds the diameter of the Fermi-disk. The analytical expression for the dielectric function is:

$$\varepsilon(\mathbf{q}, 0) = \begin{cases} 1 + \frac{2me^2}{\hbar^2 \kappa q} & ; q \leq 2k_F \\ 1 + \frac{2me^2}{\hbar^2 \kappa q} \left(1 - \sqrt{1 - (2k_F/q)^2} \right) & ; q > 2k_F \end{cases}$$

, as derived in Ref. [9]. We have modeled a potential of the form $V(r) = V(0)e^{-\beta^2 r^2}$, i.e., a Gaussian potential. We chose the parameter β to be 0.01 \AA^{-1} , which means that the potential has a spatial extent of 100 Å. If we choose the unscreened depth of the potential as $V_0(0) = 386 \text{ meV}$ we get the well depths equal to 272 meV, 232 meV and 134 meV, respectively for the densities $2 \times 10^{17} \text{ cm}^{-3}$, $4 \times 10^{17} \text{ cm}^{-3}$ and $2 \times 10^{18} \text{ cm}^{-3}$. (Fig. 4 (b). This corresponds to the blue shifts 38 meV and 100 meV as compared to the experimental values 40 meV and up to 200 meV, respectively, in going from $2 \times 10^{17} \text{ cm}^{-3}$ to $4 \times 10^{17} \text{ cm}^{-3}$ and from $4 \times 10^{17} \text{ cm}^{-3}$ to $2 \times 10^{18} \text{ cm}^{-3}$ [6,8].

In order to explain the large spectral shifts observed with doping we therefore need to assume that the unscreened potential fluctuations $V_0(r)$ in the QW are of a typical size 0.4 eV over a short distance of 100 Å. The partly screened potential seen in experiment may be considerably weaker, however. A typical fluctuation strength of about 0.2 eV seems to be needed to explain the observed strong shifts with doping in the QWs.

A major effect not taken into account in the simple screening model above is carrier transfer, which needs to be considered in a realistic model to explain the experimental data. Above the average electron concentration from the donors was considered. In practice, assuming the presence of rather short range potential fluctuations, carrier transfer processes (hopping) between different potentials may easily occur before recombination. The carriers will then have a tendency to be transferred to the lowest energy parts of the fluctuating potentials, selectively screening these. These potentials will experience a higher electron density, and thus be screened more effectively than by an average electron concentration. This transfer effect would explain part of the upshift of the PL peak with doping [8], and likewise the apparent absence of the low energy part of the PL spectrum in the timeresolved spectra in Fig 1 (b).

An argument for the dominance of free carriers in the recombination process is derived from the observed radiative decay times. For an excitonic process in a 30 Å QW the PZ field has been estimated to reduce the oscillator strength by about a factor 3 compared to the case of no PZ field [10]. Assuming an excitonic radiative lifetime (localized QW excitons) of < 1 ns at 2 K, the exciton decay time would not be more than 3 ns. Our observed values are of the order 100 ns, i.e. more than one order of magnitude larger, as expected if recombination between free (or separately localized) carriers dominate. It should be pointed out that in QWs with considerably smaller In composition, and consequently smaller potential fluctuations, the recombination is clearly dominated by excitons [3,4]. Also, the PL linewidth is then much smaller than reported in this work [3,4].

Summary

In $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}/\text{GaN}$ MQWs with a well thickness of 3 nm the peak PL photon energy is mainly determined by the piezoelectric field, while the width of the peak reflects the size of the screened potential fluctuations. We suggest that excitons and shallow donors in the QWs are largely impact ionized in the fluctuating lateral potential, so that the observed recombination is to a large extent due to separately localized electrons and holes up to room temperature. 2D screening needs to be considered to explain the PL peak shifts in donor doped MQWs.

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