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Citation: Appl. Phys. Lett. **75**, 2241 (1999); doi: 10.1063/1.124977
View online: http://dx.doi.org/10.1063/1.124977
View Table of Contents: http://aip.scitation.org/toc/apl/75/15
Published by the American Institute of Physics

APPLIED PHYSICS LETTERS VOLUME 75, NUMBER 15 11 OCTOBER 1999

Determination of photoluminescence mechanism in InGaN quantum wells

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(Received 3 May 1999; accepted for publication 10 August 1999)

We report on the unambiguous experimental determination of the photoluminescence mechanism in a set of $In_{0.25}Ga_{0.75}N$ quantum wells. Instead of studying the photoluminescence for different In contents, we have investigated it as a function of the quantum well width in combination with a similar study performed on GaN quantum wells. In this way, we show that the photoluminescence is not coming from quantum dots or very localized states in the quantum well, but from the quantum well itself under the influence of a piezoelectric field induced by strain. The previously reported abnormal photoluminescence shifts and temperature dependencies can thus be explained. © *1999 American Institute of Physics.* [S0003-6951(99)00241-7]

GaN and related materials are currently of great interest for the realization of blue-green light-emitting diodes and laser diodes.[1] However, the origin of the gain in these devices is still a matter of controversy. This stems from the fact that the InGaN active layer is a ternary semiconductor which presents some composition fluctuations,[2–4] and it has been argued that the luminescent centers in these devices were originating from localized states or quantum dots in the InGaN quantum wells (QWs) due to the presence of In rich fluctuations[5–8] or even to the self-formation of nearly pure InN quantum dots.[9] This model relies mainly on the abnormal photoluminescence (PL) temperature dependencies[10,11] as well as the observation of a large shift between the absorption edge and the band edge related PL (often called the Stokes shift) which increases with higher In content. Simultaneously a blueshift of the PL has been observed when the optical or electrical excitation power was increased and this was interpreted as a state filling effect from the localized states or the quantum dots. However, all these data can also be explained by the presence of a piezoelectric field (PEF) in the QWs induced by the strain arising in the QWs. When the In content is increased the strain and therefore the PEF is increased, resulting in a larger electro-absorption and a larger Stokes shift. Furthermore, when the carrier density is increased, a screening of the PEF will induce a blueshift of the PL in the QWs.[12,13] In this model, the gain origin can be explained from the usual two-dimensional exciton–plasma transition in a QW semiconductor.

In this letter we present an experimental method that enables one to determine which model is relevant for a set of samples. Instead of studying the optical characteristics of the QWs as a function of the In content, we have studied them as a function of the well width in combination with a similar analysis performed on GaN QWs. In contrast to InGaN, the optical properties in GaN cannot be explained by a quantum-dot-type model.

The structures were grown at 76 Torr on the Si face of an on-axis 6H–SiC(0001) substrate by a conventional horizontal-type metalorganic chemical vapor deposition system. All samples were nominally undoped. First a 2-nm-thick AlN layer followed by a 300-nm-thick AlGaN buffer layer were deposited on the 6H–SiC substrate. The Al concentration was 15% for the GaN single quantum well (SQW) samples and 10% for the InGaN double quantum well (DQW) samples. For GaN SQW, well widths between 2.5 and 5 nm were grown at 1080 °C with a capping layer made in the same conditions as the AlGaN buffer. For InGaN DQW, well widths between 0.5 and 5 nm were grown at 750 °C embedded in 15-nm-thick $In_{0.02}Ga_{0.98}N$ barrier layers. The wells were separated by 25 nm, thus avoiding any coupling. We estimated the In concentration in the wells to be 25% by comparing the PL position of the bulk material grown in similar conditions with values given in Ref. 14.

The advantage of investigating the well-width dependence for a fixed In content is shown in Fig. 1. On one hand, when the well width decreases and for the same PEF, the screening effect as well as the Stokes shift should be reduced in relation to the well thickness [Fig. 1(a)]. On the other hand, if one consider a quantum-dot-type model, it has been suggested[9] and shown experimentally in GaN quantum dots[15] that in order to observe blueshifts of the order of a few hundreds of millielectron volts due to state filling, very small quantum dots of about 1 nm in effective diameter are required which is below usual quantum-well thickness. Therefore the localization effect should be increased with reduced well widths, or at best remain at a similar level, inducing a similar or larger state filling effect [Fig. 1(b)] when the well width is decreased. This assumption can be confirmed experimentally with the low-intensity PL position as a function of the layer thickness because, in this model, the PL is coming from quantum dots in the layer. The observation of a low-intensity PL blueshift when the layer thickness decreases would therefore show that the localized states experience a stronger three-dimensional confinement. In our InGaN samples, the low-intensity PL position peaks were observed to be blueshifted when the well width was de-

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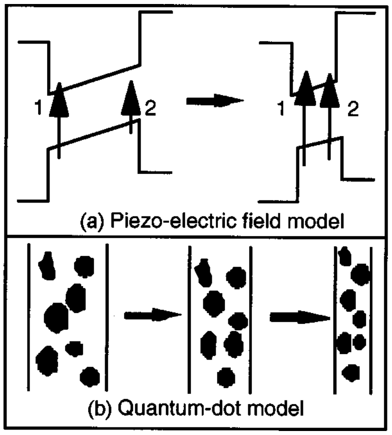

FIG. 1. Illustration of the well width's influence for the two possible PL mechanisms. (a) In the piezoelectric field model, arrow 1 symbolizes the PL energy in the absence of a PEF (after screening) whereas arrow 2 corresponds to the PL energy in the presence of the PEF for two different well widths. Because the electron and hole wave functions in the larger wells are more separated than in the narrower wells the difference between arrow 1 and 2 is larger for larger wells. (b) In the case of a quantum-dot model, the In-rich or pure InN quantum dots (closed areas) are the sources of the PL. A decrease in the layer thickness will not change or will increase the three-dimensional confinement, inducing a stronger state-filling effect and a blueshift of the low-intensity PL.

creased. In the case of the quantum-dot-type model it is explained by an increased three-dimensional confinement, whereas in the case of PEF model it is the result of an increased two-dimensional confinement.

The PL measurements were performed at 10 K with an excimer laser delivering pulses of 8 ns of duration at 308 nm. The PL was detected with an 0.5 m grating spectrometer and a photomultiplier tube. The intensity was varied between 0.4 kW/cm^2 up to 2 MW/cm^2. Some low-intensity measurements were also made with an He–Cd laser and temperature dependence studies were carried out.

Figure 2 shows the maximum blueshift when increasing the optical excitation measured for our samples at 10 K as a function of the well width. In most of the samples the blueshift reached a maximum around 500 kW/cm^2, which corresponds to a carrier density in the order of 10^{20} cm^{-3}. The maximum blueshift is clearly reduced as the well width is reduced for both GaN SQW and InGaN DQW, indicating

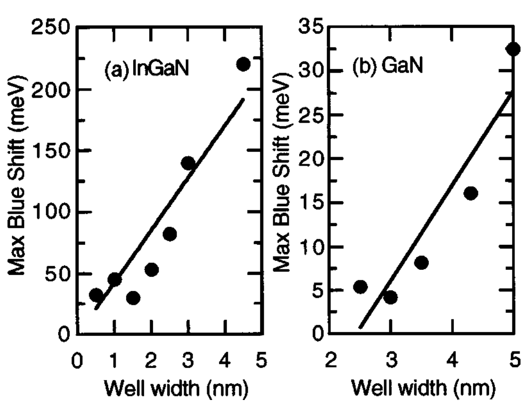

FIG. 2. Maximum blueshift of the PL as a function of the well width for (a) $In_{0.25}Ga_{0.75}N$ DQW (with $In_{0.02}Ga_{0.98}N$ barriers) and (b) GaN SQW (with $Al_{0.15}Ga_{0.85}N$ barriers). The maximum blueshift is obtained for an optical excitation intensity of about 500 kW/cm^2. The solid lines are linear adjustments.

that in our samples the screening of the PEF is the dominant mechanism. In the absence of a static electric field it has been shown that in other quantum well semiconductors the two-dimensional state-filling effect and the band-gap renormalization nonlinearity compensate each other in a similar carrier density range,[16] inducing only a change in shape of the PL spectra[17] and no significant shift. The only mechanism which can induce a significant PL blueshift in QWs must come from the existence of a PEF. Indeed no other static electric field is present in the material as we induced the carrier generation optically. In the case of electrical pumping, the built-in electric field has to be taken into account which can lead to more complicated interpretations.[18,19] Furthermore, the similar blueshift reduction with the well width for GaN and InGaN QWs reinforces the interpretation that the PEF screening model is the main mechanism as there are no composition fluctuations or phase separation in the binary material. In addition, it should be noted that the maximum blueshifts that we observed for the larger InGaN DQWs (up to 220 meV) are similar to most of the reported Stokes shifts, which indicates that the direct current-quantum-Stark effect is probably the main contributor to the Stokes shift.

In Fig. 2 the well widths are determined from the bulk growth rate. However, the excitonic PL positions after screening of the PEF in the case of GaN SQW were compared with a band structure calculation[20] including the enhanced exciton binding energy in QWs[21] and it was found that the well widths estimated from the bulk growth rate were larger than in our calculations for QWs thinner than 4.5 nm. This can be explained by a different growth rate during the first deposited layers as well as by some uncertainties in the shortest growth times which differ only by a few seconds for consecutive widths. On the other hand, for InGaN, the uncertainty of the In content determination does not allow a safe determination of the well widths by a band calculation. From these results we can estimate roughly the PEF in our samples assuming a linear decrease of the maximum blueshift with the well width to be about 60–70 kV/cm in GaN and 400–500 kV/cm in $In_{0.25}Ga_{0.75}N$. Higher values both for GaN[22,23] and InGaN QWs[12,13] have been reported. However, they are difficult to compare with the theoretical values and other samples grown in different conditions with different substrates and different buffer layers because the QWs are assumed to be fully coherently grown and the stiffness coefficients and the piezoelectric constant necessary for the determination of the PEF are not well established yet for GaN[12,24] and unknown for InGaN. They will also depend on the crystal quality and the number of dislocations in the samples. Nonetheless, the PEF is expected to be larger in InGaN QWs than GaN QWs because the strain in the GaN QWs used here is only 0.3% as compared with 2.5% for $In_{0.25}Ga_{0.75}N$. In this set of samples, the ratio of the strain in GaN SQW and InGaN DQW is comparable with the PEF ratio.

Finally, Fig. 3 shows the PL position for the 1-nm-thick InGaN DQW and the 5-nm-thick GaN SQW as a function of the temperature under high and low optical excitation. Under high excitation, the PEF is screened and the PL temperature behavior is similar to that of bulk material. This is what is

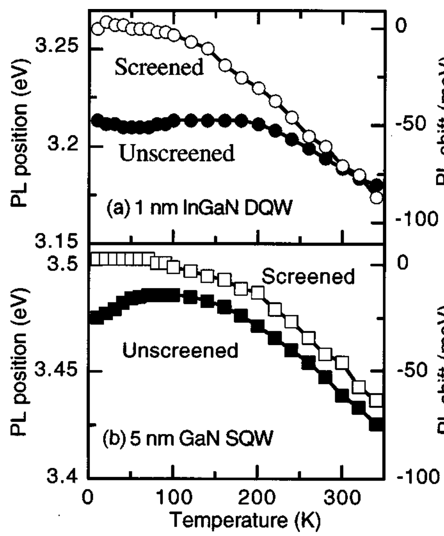

FIG. 3. PL position for (a) the 1-nm-thick $In_{0.25}Ga_{0.85}N$ DQW and (b) the 5-nm-thick GaN SQW as a function of the temperature under high (open circles and squares) and low (closed circles and squares) optical excitation.

expected for a QW without PEF as the phonon couplings responsible for the band-gap redshift are similar for bulk and QWs.[25] On the other hand, a PL blueshift around 100–150 K is observed in the case of a low optical excitation both for InGaN DQW and GaN SQW. In the light of the abovementioned results, we can understand the abnormal PL blueshift as a function of the temperature by a partial screening of the PEF induced thermally. In fact, many carriers which were frozen out in nonradiative centers at low temperature will become thermally activated as the temperature increases, inducing some partial screening of the PEF. The PL temperature dependence thus reflects a competition between a blueshift induced by a thermal screening of the PEF and a redshift induced by the phonon interactions.

In conclusion, we have shown that it was possible to determine unambiguously the origin of the previously reported blueshifts observed in InGaN QWs at high carrier excitation by studying this shift as function of the well width under high optical excitation in combination with a similar study performed in GaN QWs. We have found that in the case of our set of samples, the model involving quantum-dot-like structures could be ruled out and instead the main mechanism was found to be due to the presence of a PEF in the QWs. This model can also explain the large observed Stokes shifts in these structures and the abnormal PL temperature dependencies. A similar investigation should be done for laser devices and for different In contents in order to determine the origin of their gain.

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