Recombination of free excitons in polar and nonpolar nitride quantum wells

T. Langer, H. Jönen, D. Fuhrmann, U. Rossow, and A. Hangleiter
Institute of Applied Physics, University of Braunschweig
Mendelssohnstr. 2, 38106 Braunschweig, Germany
E-mail: a.hangleiter@tu-bs.de

Abstract. We compare radiative and nonradiative recombination in polar and nonpolar GaInN/GaN quantum wells and demonstrate that purely radiative recombination can be achieved even at room temperature. The radiative recombination is due to free excitons, which partially dissociate at elevated temperatures. Nonradiative recombination is strongly inhibited in the polar case but not in the nonpolar case.

1. Introduction
The dominant radiative recombination channel in III-nitride quantum wells has been subject to quite some debate in recent years. This question was linked to the highly unexpected large quantum efficiency of such structures, despite the extremely large density of threading dislocations typically found in heteroepitaxial nitride layer structures. Early work indicated that localized excitons may manage to escape nonradiative recombination [1]. More recently, however, strong evidence for energy barriers surrounding dislocations [2, 3, 4] led to the conclusion that excitons get anti-localized rather than localized.

Here, we demonstrate that in high quality GaInN/GaN quantum wells, grown either on c-plane or on m-plane GaN buffers, recombination at room temperature is excitonic and can be dominated by radiative recombination of free excitons.

2. Experimental
Our samples were grown using low pressure metal-organic vapour phase epitaxy on two types of substrates. C-plane oriented structures were grown on c-plane sapphire substrates, whereas m-plane SiC substrates were used for m-plane structures. On sapphire, an (In)GaN nucleation layer was followed by a 1.7 µm thick GaN buffer layer. On SiC, a thin AlN layer was used to initiate growth, followed by a GaN buffer layer (grown at 1190 °C). The Ga0.8In0.2N/GaN single or multiple quantum wells (MQWs) were then grown at temperatures between 700 °C and 800 °C. The well width was in the range 1 - 2 nm.

The structural characterization of the samples was performed by means of X-ray diffraction (for MQWs) and transmission electron microscopy. The optical properties were analyzed by resonant and nonresonant photoluminescence (PL) measurements. In addition the temperature and excitation power dependent PL was used in order to determine the internal quantum efficiency. Details of this method can be found elsewhere [5].

Detailed measurements were done using time-resolved photoluminescence spectroscopy. 5 ps pulses at 375 nm from a frequency-doubled cavity-dumped synchronously mode-locked dye laser, pumped
by a frequency-doubled mode-locked Nd-YAG laser, were used to excite the samples (~ 1 pJ). The luminescence was dispersed by a 30 cm subtractive double monochromator, detected by a fast micro-channel-plate photomultiplier and processed using time-correlated photon counting. The overall time resolution was about 20 ps. The samples were mounted in a variable temperature cryostat (4 - 300 K).

3. Results and discussion

In this study we compare two types of quantum wells (QWs). Quantum wells grown on nonpolar crystal planes such as the m-plane hold the promise of faster radiative recombination as compared to those on the polar c-plane, due to the lack of spatial separation of electron and hole wavefunctions [6, 7].

In order to study the mechanism of radiative recombination, high quality samples with almost purely radiative decay of excited carriers are required. A fairly good measure of quality is therefore the internal quantum efficiency estimated from temperature dependent PL measurements.

As shown in Fig. 1, state-of-the-art c-plane Ga$_x$In$_{1-x}$N/GaN quantum wells achieve room temperature internal efficiencies (IQE) of about 80 % and more. On the other hand, the nonpolar m-plane QW reaches about 45 % IQE, indicating a stronger influence of nonradiative processes.

From temperature dependent measurements of the photoluminescence decay we find the effective decay time $\tau_{\text{eff}}$. This can be separated into the radiative lifetime $\tau_r$ and the nonradiative lifetime $\tau_{\text{nr}}$ taking advantage of the measured quantum efficiency $\eta$:

$$
\tau_r = \eta \cdot \tau_{\text{eff}} \quad \text{and} \quad \tau_{\text{nr}} = \frac{1}{1 - \eta} \cdot \tau_{\text{eff}} \quad (1)
$$

As can be seen from Fig. 2, the nonpolar MQW structure indeed features fast radiative decay with about 1 ns radiative lifetime at room temperature. However, as the nonradiative lifetime gets even shorter at this temperature (0.4 ns), the effective lifetime is dominated by the nonradiative decay and thus the quantum efficiency is considerably lower than unity. One should note here that the IQE in the pulsed experiments is somewhat lower than under CW excitation as used for IQE measurements (c.f. Fig. 1) due to lower excitation level under pulsed conditions. The temperature dependence of the nonradiative lifetime is described by a rather small single thermal activation energy of the nonradiative process of 21 meV.

The radiative lifetime exhibits two distinct regions. It is constant at low temperatures below about 100 K and increases approximately linearly at higher temperatures. This behavior is well known from

![Figure 1. Temperature dependence of the internal quantum efficiency for two Ga$_x$In$_{1-x}$N/GaN samples emitting at similar wavelengths. The c-plane sample is a 1.1 nm QW with 35 % In, the m-plane sample is a 5 x 2 nm MQW with 25 % In.](image1.png)

![Figure 2. Temperature dependence of the decay times of a nonpolar Ga$_x$In$_{1-x}$N/GaN sample. While the nonradiative lifetime is thermally activated, the radiative lifetime increases with temperature as typical for 2D free excitons.](image2.png)
other materials [8] and relates to “localization” at low temperature and k-space occupation at higher temperatures. A linear increase then corresponds to the linear temperature dependence of the effective density of states in a two-dimensional system. Obviously, localization takes place at temperatures below 100 K, whereas excitons behave like free excitons at higher temperature.

In order to get a quantitative description, one needs to take into account that excitons get thermally dissociated at elevated temperatures in addition to k-space occupation. Thus

\[ \tau_r \propto -\Delta \cdot \left(1 + cT \exp\left(-E_x/kT\right)\right), \]

where \( \Delta \) describes the localization [8] and \( E_x \) is the free exciton binding energy, and \( c \) is a constant including the densities of state. From the fit (full line in Fig 2) we find a free exciton binding energy of 45 meV which is in reasonable agreement with calculations and with experimental estimates [9].

The results from nonpolar m-plane structures have to be compared to state-of-the-art c-plane quantum wells. Fig. 3 shows the effective exciton lifetime and the internal quantum efficiency as a function of temperature for a very narrow c-plane quantum well. The IQE is larger than 80 % over the full temperature range, corresponding to virtually unity efficiency. At the same time, the effective lifetime behaves like the radiative lifetime in the previous example, i.e. is almost constant at low temperature and increases approximately linearly at higher temperatures. Again, localization is present at temperatures below about 120 K, while free exciton like behavior takes over at higher temperatures. Once again, this is quantitatively described by the 2D exciton model including thermal dissociation and happens to yield the same exciton binding energy of 38 meV.

Applying eq. 1 we see that the nonradiative lifetime at room temperature is about 13 ns or larger, while the radiative lifetime is only about 1.3 ns. This shows a negligible influence of nonradiative transitions even at room temperature and is consistent with a purely radiative excitonic behavior of the measured effective lifetime as shown before.

Clearly, free excitons dominate at elevated temperatures, which are anti-localized thanks to energy barriers decorating dislocations and thus have rather large nonradiative lifetimes [2] of more than 10 ns. On the other hand, nonradiative recombination is rather fast (0.4 ns) in the nonpolar case, indicating that nonradiative defects are not disabled like they are in the polar c-plane quantum wells.

Free excitons have a finite binding energy and thus dissociate at elevated temperatures. They actually dissociate into free electrons and holes (binding energy \( E_b \))

\[ x \leftrightarrow e + h - E_b \]

Therefore, a law of mass-action holds \((n, p = \text{electron and hole densities, } x = \text{exciton density, } C(T) \text{ is an equilibrium constant})\)

\[ \frac{n \cdot p}{x} = C(T) \]

or, in the case of optical excitation with \( n = p \)

\[ \frac{n^2}{x} = C(T) \]
On the other hand, the total density $N$ of excited carriers is

$$ N = n + x \quad (6) $$

Recombination of the excited carriers is described by ($\tau_x =$ radiative lifetime of excitons, $B =$ radiative coefficient, $\tau_{nr} =$ nonradiative lifetime)

$$ \frac{dN}{dt} = - \frac{x}{\tau_x} - B \cdot n \cdot p - \frac{N}{\tau_{nr}} \approx - \frac{x}{\tau_x} - \frac{N}{\tau_{nr}} \quad (7) $$

We actually measure the luminescence intensity $I$, which comes from radiative recombination of excitons, thus

$$ I \propto \frac{x}{\tau_x} \quad (8) $$

The solution of eq. 5 and 6 actually defines two regions of carrier density: (i) a low density regime, where the exciton density (and thus the radiative rate) is proportional to the square of the total carrier density, and (ii) a high density regime, where the exciton density is proportional to the total carrier density. The decay following from eq. 7 is thus expected to be exponential at higher carrier densities and hyperbolic for the lower ones.

Experimentally, we find non-exponential decay curves up to room temperature. As shown in Fig. 4, these are well fitted by the model outlined above. Due to a limited dynamic range in the experiments, the transition between exponential and hyperbolic can not be seen with the naked eye. Nevertheless, the dissociation of free excitons is clearly evident from the decay behavior.

4. Conclusions

We have demonstrated that high quality Ga$_x$In$_{1-x}$N/GaN quantum wells can exhibit purely radiative recombination at room temperature. The recombination is due to free excitons, which are subject to partial dissociation at elevated temperatures. Both the temperature dependence and the decay kinetics are quantitatively described in the free exciton picture. Even though nonpolar quantum wells feature faster radiative recombination, polar c-plane QWs are still superior due to inhibited nonradiative recombination.

Support of this work by the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

References

[1] Chichibu S, Azuhata T, Sota T and Nakamura S 1996 Appl. Phys. Lett. 69 4188
[2] Hangleiter A, Hitzel F, Netzel C, Fuhrmann D, Rosso u U, Ade G and Hinze P 2005 Phys. Rev. Lett. 95 127402
[3] Sonderegger S, Feltin E, Merano M, Crottini A, Carlin J F, Sachot R, Deveaud B, Grandjean N and Ganiere J D 2006 Appl. Phys. Lett. 89 232109
[4] van der Laak N K, Oliver R A, Kappers M J and Humphreys C J 2007 Appl. Phys. Lett. 90 121911
[5] Hangleiter A, Fuhrmann D, Grewe M, Hitzel F, Klewer G, Lahmann S, Netzel C, Riedel N and Rosso u U 2004 phys. stat. sol. (a) 201 2808
[6] Im J S, Kollmer H, Off J, Sohmer A, Scholz F and Hangleiter A 1998 Phys. Rev. B 57 R9435
[7] Waltereit P, Brandt O, Trampert A, Grahn H T, Menniger J, Ramsteiner M, Reiche M and Ploog K H 2000 Nature 406 865
[8] Feldmann J, Peter G, Göbel E, Dawson P, Moore K, Foxon C and Elliott R 1987 Phys. Rev. Lett. 59 2337
[9] Lahmann S, Hitzel F, Rosso u U and Hangleiter A 2003 phys. stat. sol. (c) 0 2202