MYSTERIES OF GAN

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Abstract

Despite of all successes in commercialization of GaN-based devices present knowledge of light emission mechanisms in these devices is surprisingly limited. In my talk I discussed several puzzling properties of wide band gap nitrides. Some of them are briefly described below. First, I discuss correlations between structural quality of thin films of GaN and their light emission properties. Then, the mechanisms responsible for strong carrier/excitons localization effects are discussed. Finally, I discuss properties of n- and p-type doped GaN layers.

1. History of development of light emitting diodes (LEDs)

Light emitting diodes (LEDs) are presently produced on a massive scale. Their history is relatively short. Possibility of light emission from p-n junction was demonstrated in 1962 in GE Company. First, LEDs operated at fairly low power, thus their applications were very limited. In 1968 two companies (HP and Monsanto) commercialized LEDs, proposing their use instead of so-called Nixie tubes. These LEDs were based on doped GaP, emitted deep red color light and had fairly low performance - 1 mlm at 20 mA current.

A first revolution came around 1985. A new generation of LEDs with quantum well (QW) structures was commercialized. They had improved light emission performance - 1-100 lm, as compared to 900 lm of 100 W incandescent lamp. Despite still limited performance, these LEDs were good enough for several new applications. For example, matrix of 75 LEDs was used as stop lights in cars. Further progress was very fast. In 1990 GaAs/GaAlAs QW-based LEDs achieved 10 lm/W at 640 nm, whereas quaternary GaAlInP LEDs 20 lm/W at 620 nm, which was good enough for signal lights applications (see table 1).

Table 1. Performance of signal lamps LEDs, as compared to emission of filtered incandescent lamps.

Color	Incandescent bulb Performance (lm/W)	LED Performance (lm/W)
red	1-6	16
yellow	4-8	10
green	3-10	48

Second revolution in LED development came in 1993 when dr. S. Nakamura from Nichia Company introduced first efficient blue/violet color LEDs. Meanwhile, in 1995 HP Company improved their GaAlInP-based LEDs, which reached 50 lm/W light emission performance. 150 lm/W is predicted for these LEDs, which is good enough to replace e.g. Na arc lamps!

Based on GaN-based diodes white LEDs were commercialized around 2000. These diodes use so-called hybrid structure – blue color emission from LED excites yellow emission from YAG:Ce luminophor. Mixture of these two emissions gives an impression of a white color. Table 2 compares light emission properties of present white light sources with those predicted for new generations of white LEDs.

Table 2. Comparison of performance of white light sources

Lamp type	Power (W)	Performance (lm/W)	Lifetime (h)
Incandescent	135	12	1000-5000
Halogen	300	24	3000
Fluorescent	30	80	20000
White LED (2000)		20	>100000
White LED (2010)		50	>100000

Table 3. Energy and money savings due to introduction white LEDs (data for USA)

Year	2005	2010	2015	2020	2025
% of white LEDs	0.05	2	12	30	55
Energy savings (TWh/year)	2	67	330	720	1100
Money savings (mln USD/year)	200	6700	33000	72000	110000

Already introduction of LEDs as traffic signal lamps led to significant money savings, about $1O^9$ USD per year for a country of USA or Japan size! This is still rather small sum, as compared to predicted money savings related to substitution of inefficient incandescent lamps with white color LEDs. Money savings of the range of 10^{11} USD per year are predicted! Moreover, large electricity savings will lead to a significant reduction of CO_2 emission to the atmosphere by hundreds of millions ton per year. Not surprisingly a rapid development of optoelectronic industry is predicted to the size of present day electronic industry.

2. Dislocations In Nitrides

GaN-based LEDs and laser diodes (LDs) are presently produced in huge amounts. For example, more than 50 mln LED devices per month is produced at present at cost few hundred times lower than of those from the first generation of blue color LEDs.

Massive scale production normally means that we understand how the devices produce light. Surprisingly, this is not the case for GaN-based LEDs and LDs. For all other III-V materials used in optoelectronic devices density of dislocations should be less than 10^5 cm^{-2}, for efficient light emission [1]. GaN-based LEDs emit light at shockingly high density of dislocations of 10^{11} cm^{-2}. It was first suggested that dislocations are not acting as centers of nonradiative recombination in nitrides. We now know that this is not the case. It was then proposed that role of dislocations as centers of nonradiative recombination can be limited in the samples with a small diffusion length of carriers and excitons [2,3].

The relevant mechanism of localization limiting diffusion length was proposed by S. Nakamura and coworkers [2,3]. InGaN is used as QW material in all commercialized nitride-based LEDs and LDs. Fluctuations of In fraction may result in potential fluctuations in a QW plane. Quantum dot like structure of this region was proposed with strong carrier/excitons localization effects and thus with reduced diffusion lengths.

If the model is correct, localization should be weak (weaker than in InGaN) in GaN layers or QWs. We recently tested this possibility by performing cathodoluminescence (CL) investigations of a series of GaN thin films grown with metal organic vapor phase epitaxy (MOVPE) [4]. Growth process was modified in the way to grow films with GaN grains of a different size, i.e. with different density of edge dislocations, since these dislocations are present at grain boundaries.

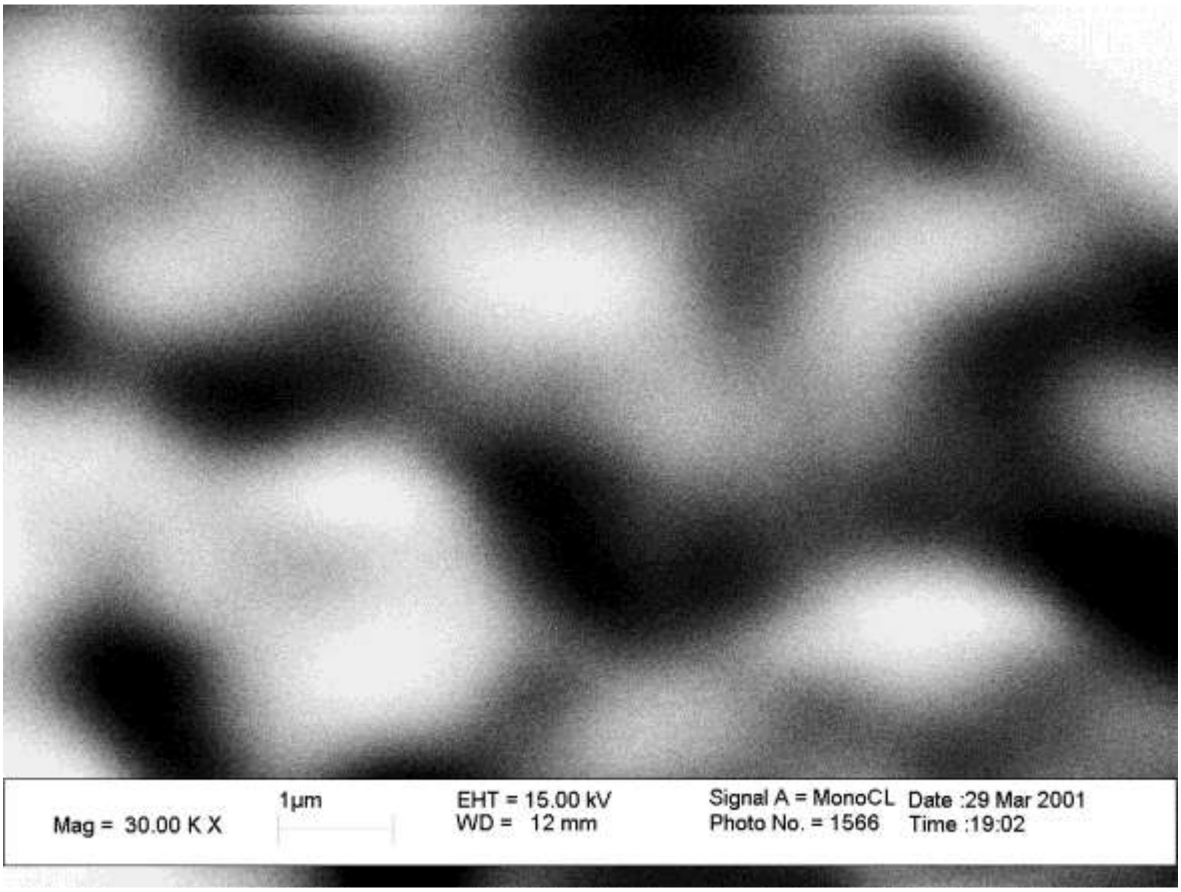
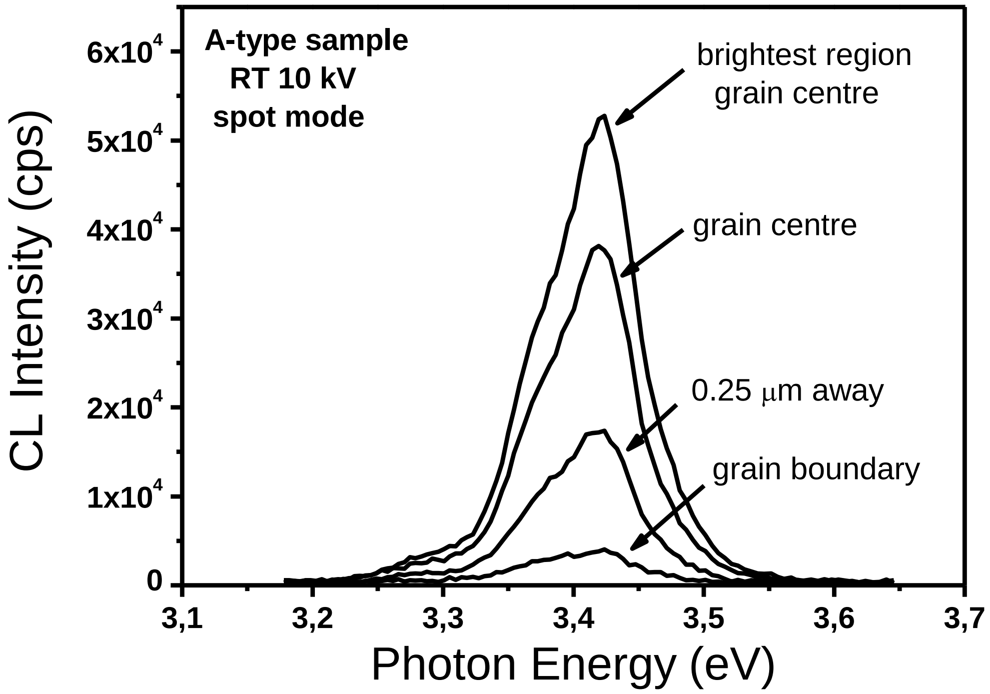
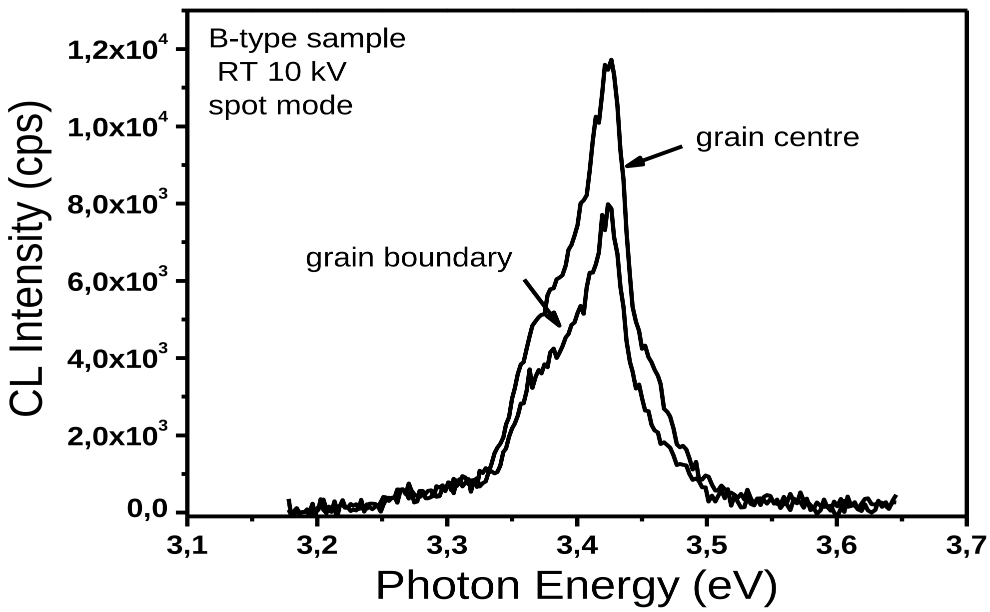


Fig. 1. Scanning CL image obtained with detection set at GaN excitonic emission, taken at 30000 times magnification for GaN film with large grains.

Fig. 1 shows CL image measured for the sample with large grains of 1-2 micrometers size. The image was taken at room temperature with detection set at GaN excitonic emission. Intensity variations are observed which correlate with a granular micro-structure of the film. Similar correlation of intensity variations we observed for the film with much smaller grains (100-200 nm), i.e. film with a significantly increased dislocation density (10^{10} cm^{-2} instead of $2x10^{8}$ cm^{-2}) [4].

We could quantify the observed intensity variations by measuring so-called spot-mode CL spectra for our films. Instead of scanning a given area with electron beam and following CL intensity variations, we set excitation at different points selected from scanning CL images. We measured CL spectra at spots of white or black contrast in scanning CL images and compared intensity and spectral shape of the CL spectra. The relevant results are shown in Figs. 2 and 3.





Figs. 2 and 3. CL spectra measured in spot-mode CL for two types of GaN films with grains of a different size (after [4]).

The following conclusions were drawn from the spot-mode CL study. First of all, for both types of the films diffusion length must be smaller than grain size. Secondly, diffusion length is increased in the sample with larger grains. Apparently there is a direct correlation between micro-structure of the film and diffusion length of carriers/excitons. The observed diffusion lengths are not much larger than those reported for InGaN films, which were related to In fluctuations, once more opening the discussion on the origin(s) of strong localization effects in nitrides [4].

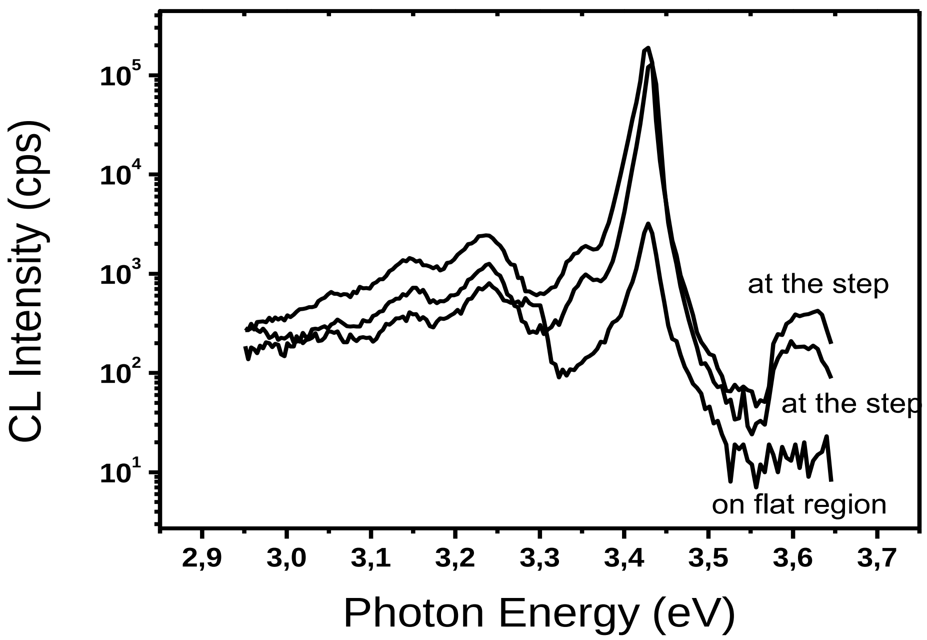


Fig. 4: CL spectra measured in spot-mode CL for homoepitaxial GaN film grown by MOVPE (after [5]).

Summarizing the results of the CL investigations we concluded that in-plane changes of CL intensity correlate with a micro-structure of a sample and that improved sample morphology results in an increased diffusion length. The resulting CL intensity depends on a comparison between diffusion length of carriers/excitons and inter dislocation distance [4].

How it is in homoepitaxial films? In Fig. 4 we show the results of spot-mode CL study taken for homoepitaxial GaN film (after [5]). Diffusion length of carriers and excitons must be in these films much larger than in heteroepitaxial films! We observed micrometer scale migration of carriers and/or excitons to the growth steps and their trapping and decay there. These growth steps are most likely decorated with donor type impurities, explaining a very bright bound excitonic emission coming from these areas of the film.

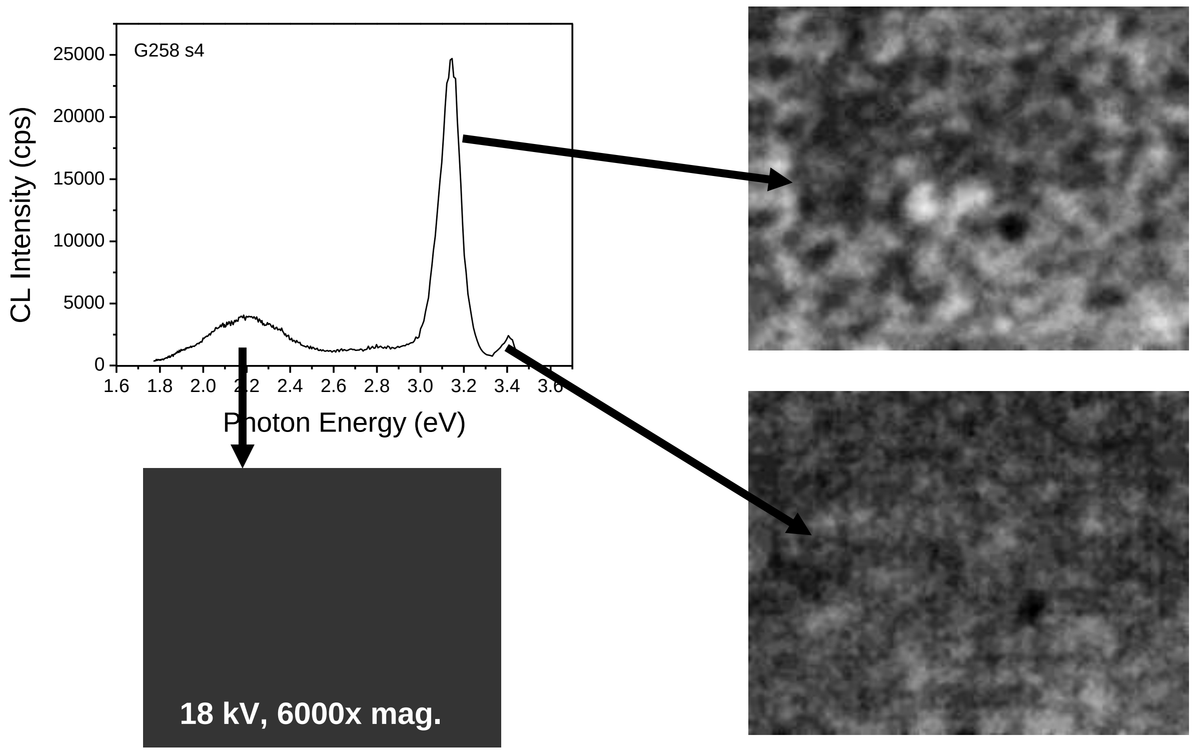


Fig. 5: CL spectrum of a single QW InGaN/GaN QW structure grown by MOVPE on sapphire together with scanning CL images taken at room temperature with detection set at either QW (top right), GaN barrier (bottom right) or parasitic yellow (bottom left) emission.

In Fig. 5 we compare the results of scanning CL investigations for InGaN/GaN QW structure. Both for a QW (InGaN) and barrier (GaN) emission we observed CL intensity variations. They do not in-plane correlate, but are of a similar scale. Only the parasitic yellow emission do not in-plane fluctuates. Properties of the latter emission are described in [6].

These investigations indicate that diffusion lengths in GaN films are similar to those in InGaN films. Diffusion lengths are small enough to limit nonradiative recombination at dislocations, thus radiative decay should dominate in photoluminescence (PL) of both InGaN and GaN layers. This opens several new questions, such as: why practical devices require InGaN QWs? Do we have one or two mechanisms of localization?

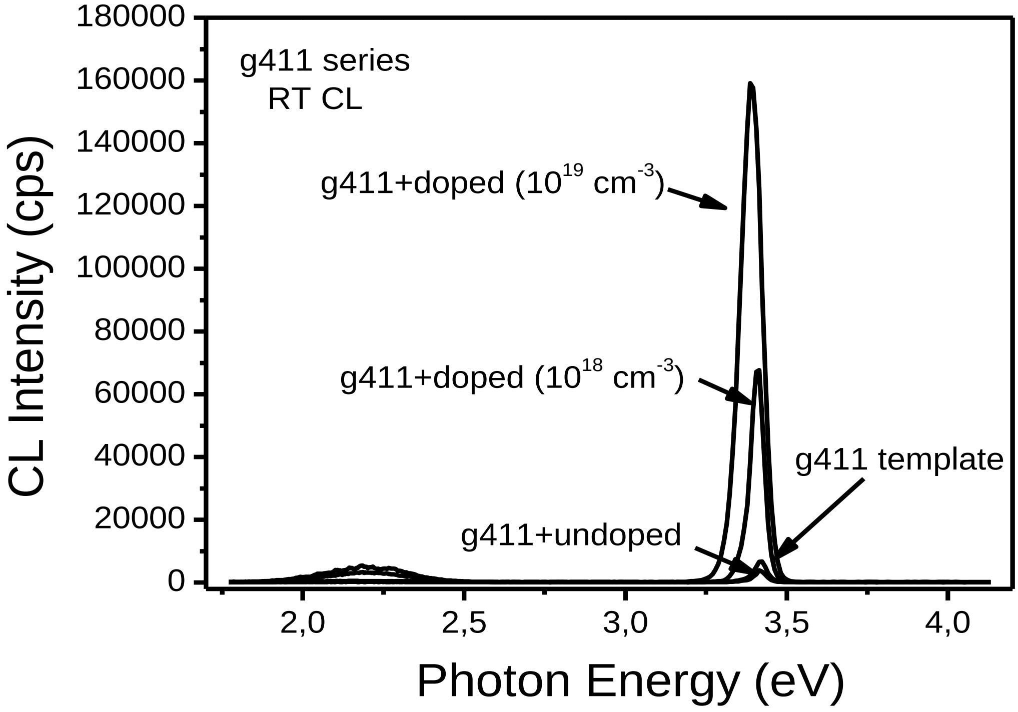


Fig. 6. CL spectra of four GaN films of a different doping level with silicon (after [7]).

3. Do we understand n-type doping?

Estimated diffusion length, compared to inter-dislocation distances, indicated that decay in GaN films should be mostly radiative. Surprisingly PL or CL is significantly (up to 100 times!) enhanced upon n-type doping, as we show in Fig. 6. Details of these studies can be found in the Ref. [7].

Several models were proposed to account for the observed PL/CL enhancement (as discussed in [7]). The most likely model related intensity enhancement to a screening of electric field. Such screening should result in a blue shift (up-in-energy) of the emission. Why emission shifts down in energy, as is shown in Fig. 7?

We can thus only speculate why n-type doping enhances rate of radiative recombination. Kelvin probe investigations [7] confirm that potential fluctuations are reduced upon n-type doping. In the consequence, diffusion length of carriers and excitons should be increased, resulting in an increased chance that carriers and excitons will approach dislocations and decay there nonradiatively. Formally, we could thus predict that emission should be deactivated upon n-type doping. Instead, emission is enhanced!

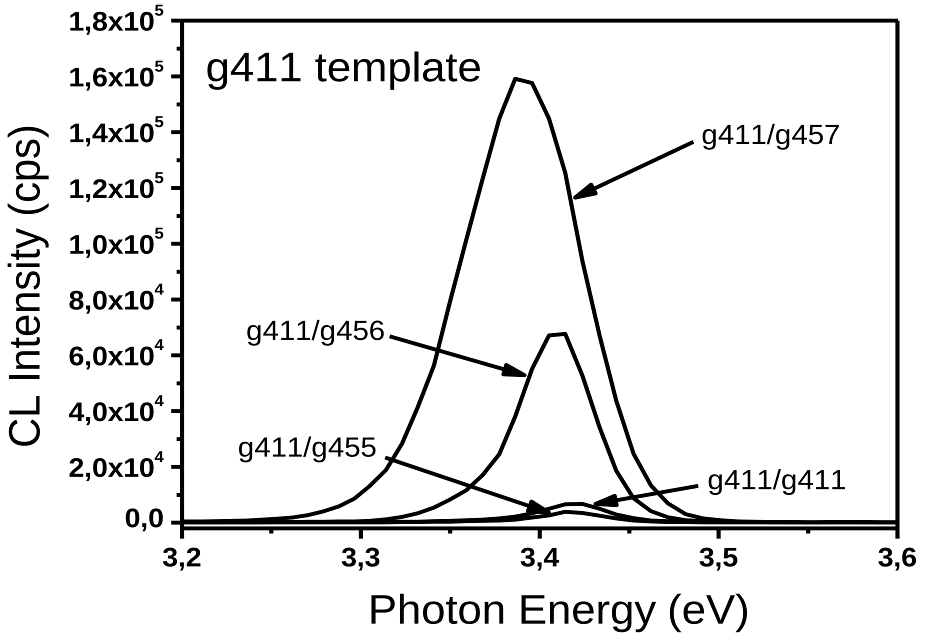


Fig. 7. Edge part of the CL spectra of four GaN films of a different doping level with silicon (after [7]).

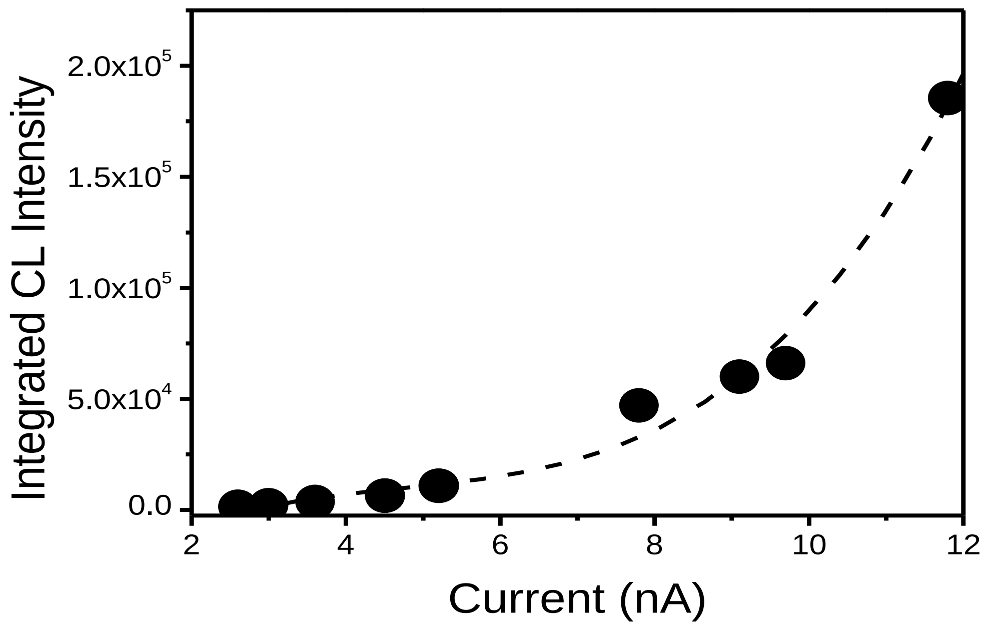


Fig. 8. Dependence of CL intensity on excitation density measured for heteroepitaxial GaN film grown by MOVPE.

Maybe decoration of grain boundaries and dislocations with impurities is here important or change of charge at dislocations? Especially the latter effect can be of importance. For example, we observed that GaN emission shows a highly nonlinear increase at increased excitation density in the CL study. Further investigations are required to clear this puzzling property of GaN layers.

4. GaN- p-type doping

There are several unexplained effects related to p-type doping of GaN layers [6,8,9]. Low doping efficiency, doping-induced compensation effects, origin of acceptor centers and acceptor-related complexes are still discussed. Our recent studies [9] indicated also the presence of much enhanced potential fluctuations in p-type doped films, as concluded from Kelvin probe measurements [9].

A characteristic property of our p-type doped GaN films was the appearance of sharp peaks in PL spectra separated by LO phonon energy (Fig. 9). These peaks, so-called hot PL peaks, we relate to strong potential fluctuations in the films induced by p-type doping [9,10].

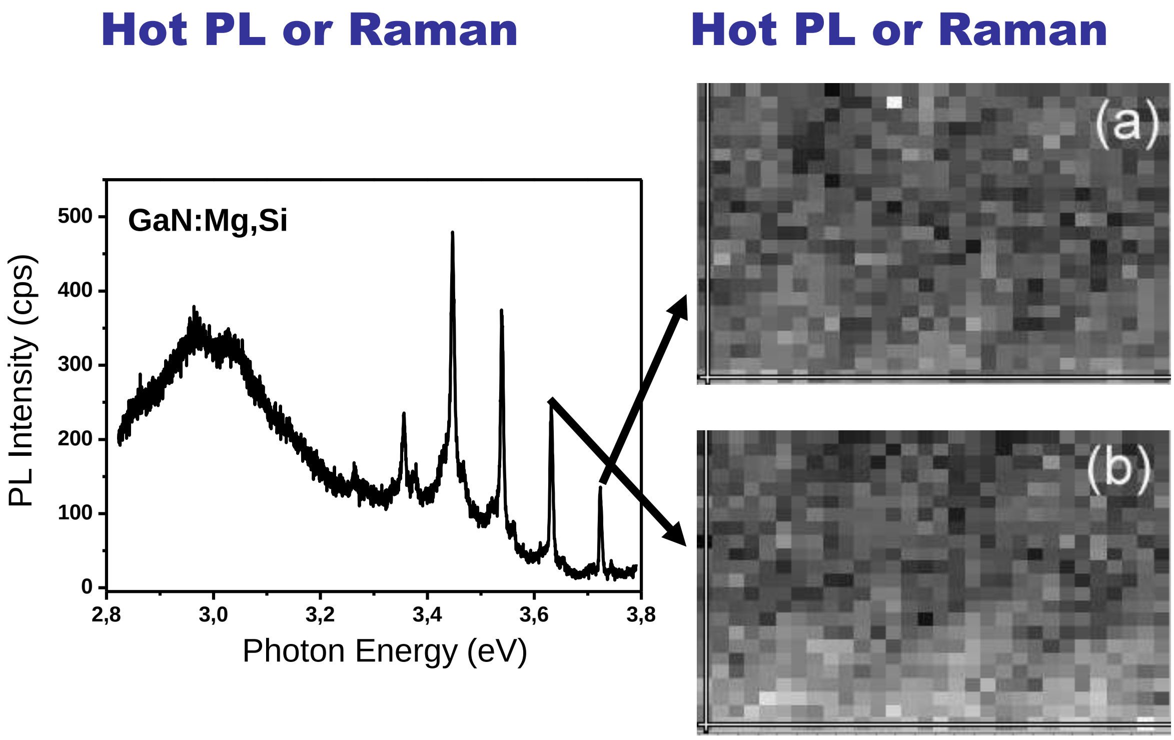


Fig. 9. Characteristic PL spectrum observed in p-type GaN samples shown together with in-plane variations of PL (a, b), as measured in the micro-PL study.

Conclusions

Summarizing, we show that diffusion lengths of carriers/excitons correlate with sample quality. These diffusion lengths are often similar in GaN and InGaN layers, once more opening the question on the origin of the observed strong localization effects in nitrides. Further on, we show that CL/PL is strongly enhanced upon n-type doping. In turn, strong compensation effects in p-type doped layers result in enhanced potential fluctuations and in the observation of the so-called hot PL spectra.

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