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Complex strain distribution in individual faceted InGaN/GaN nano-columnar heterostructures

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Abstract: Selective area growth technique is very promising for the realization of optoelectronic nano-devices based on InGaN/GaN quantum disks, as it allows precise positioning of the nano-objects on the substrate. However, this fabrication method induces a pronounced pyramidal shape of the nano-columnar heterostructures. To understand how the optical properties of these heterostructures are affected by this shape, we investigated the linear polarization of the luminescence from 0-dimensional localization centers included in their active layer. Our experimental results and our simulation show that a complex strain distribution exist in the active layer and also that quantum dot-like objects can be used to probe the local strain distribution through nano-scale heterostructures.

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1. Introduction

InGa_N/Ga_N based heterostructures are among the most attractive for the realization of light-emitting devices in the visible spectral range [1,2]. Indeed, their band gap energy can be tuned from infra-red to ultra-violet just by changing the In composition of their active layer. Nevertheless, it is well known that InGa_N based optical devices suffer of luminous efficiency "droop" at high indium content. This droop is caused by the degradation of the crystal quality and the enhancement of the so called quantum confined stark effect (QCSE) [3,4]. To reduce the electron-hole separation that is induced by QCSE and thus ameliorate the internal quantum efficiency (IQE) of bi-dimensional InGa_N/Ga_N quantum well (QW) systems, various growth strategies have been proposed such as the use of non-/semi-polar InGa_N QWs [5–7], InGa_N QWs with large overlap designs [8,9] and of ternary template/substrate [10,11]. Another promising technique that attracts the attention of numerous teams aiming to fabricate high IQE InGa_N based optoelectronic nano-devices is the embedding of InGa_N/Ga_N QWs in nano-columnar objects. These nano-columnar heterostructures provide high light extraction efficiency, low dislocation density, stress free epilayers [12] and constitute nowadays one of the most valuable alternatives for the realization of high performance optoelectronic devices based on III-nitride compounds.

The last decades, continuous improvement of the nano-column fabrication techniques has opened the door to promising applications [13,14] and attractive fundamental physics matter

[15]. Among all, selective area growth (SAG) techniques constitute a major step in the fabrication techniques, because it allows accurate positioning of the nano-objects over the substrate, precise calibration of their lateral size and fine control of their emission wavelength [16]. All these characteristics are essential for the realization of optoelectronic nano-devices. However, by molecular beam epitaxy (MBE), the control of the nano-columnar morphology is a critical issue intimately linked to the interplay between diffusion and desorption of impinging atoms at the substrate surface during the growth process. It appears that in optimal growth condition, III-nitride based nano-columns elaborated by SAG techniques exhibit hexagonal cross section and pyramidal top shape composed of 6 semi-polar m-facets that form an angle of 62° to the (0001) axis [16] (see inset of Fig. 1(a)). In the following, an InGaN/GaN QW embedded in such pyramidal nano-column will be called nano-faceted quantum disk (NFQ-disk).

It is challenging to predict how the morphology of the NFQ-disks will affect the properties of the exciton formed in the InGaN layer: a priori, radiative recombination may take place either through the facets or the edges of the pyramidal heterostructure leading to various confinement degrees of the exciton and complex polarization properties of the emitted light. However, a complete understanding of the radiative recombination processes and the excitonic confinement degree through the InGaN layer in such NFQ-disks is of first importance for the realization of state of the art light emitting nano-devices. In this report, we present optical investigation of individual InGaN/GaN NFQ-disks. So far, the correlation between the pyramidal shape of InGaN/GaN NFQ-disks and their optical properties has never been discussed; we believe that such study should open new interest for application and fundamental approaches.

2. Experiment and results

The studied InGaN/GaN NFQ-disks were fabricated by SAG techniques using a Ti nano-mask pattern. Initially, the Ti film was deposited over a MOCVD-GaN template, followed by the formation of nano-holes by focused ion beam (FIB) milling. Then, GaN nano-column of 1 μm height were grown through the nano-hole followed by the deposition of a single InGaN active layer of 3 nm finally capped by a 5 nm thick GaN barrier. Detail of the growth technique can be found in Refs. [17,18]. The experimental results discussed hereafter were obtained on individual InGaN/GaN NFQ-Disks with a diameter of 200 nm deposited in a triangular lattice array of 4 μm period (see Figs. 1(b) and 1(c)). We excited and collected the signal from individual NFQ-Disk in a parallel direction with the c-axis through a $40\times$ optical objective lens of a confocal micro-photoluminescence (μPL) setup (see Fig. 2(a)). The spatial resolution of the μPL setup is 1 μm . The sample was cooled at 8 K by a continuous helium flow cryostat. Contrarily to previous work (Ref. [16]) the growth condition of the sample was not optimized to observe a uniform emission wavelength, so that, the NFQ-disks emit in a broad blue-green region (see Fig. 1(c)). We plotted in Fig. 1(a), typical spectra obtained from a single InGaN/GaN NFQ-Disk (called NFQ-disk_i) under three different power excitation densities. As observed in the case of self-organized InGaN/GaN quantum disks, it appears that the radiative recombination processes in the NFQ-disks grown by SAG techniques take place in localization centers (LCs) constituted by In clusters in the InGaN layer [19,20]: the spectra are structured and exhibit sharp emission peaks. The emission peaks intensity in the spectrum of Fig. 1 varies linearly with the excitation power density suggesting that each emission peak is induced by excitonic recombination in different individual LCs. Moreover, in a general way, we could fit the entire spectrum of these NFQ-disks by the sum of Lorentz functions indicating that these sharp peaks are emitted by individual quantum dots like (QDs-like) nanostructures.

It is well known that the excitonic fine structure [21] of semiconductor QDs or QD-like objects (such as LCs) are intimately linked to the strain distribution in their surroundings. Therefore, to obtain physical information about the strain distribution and the recombination

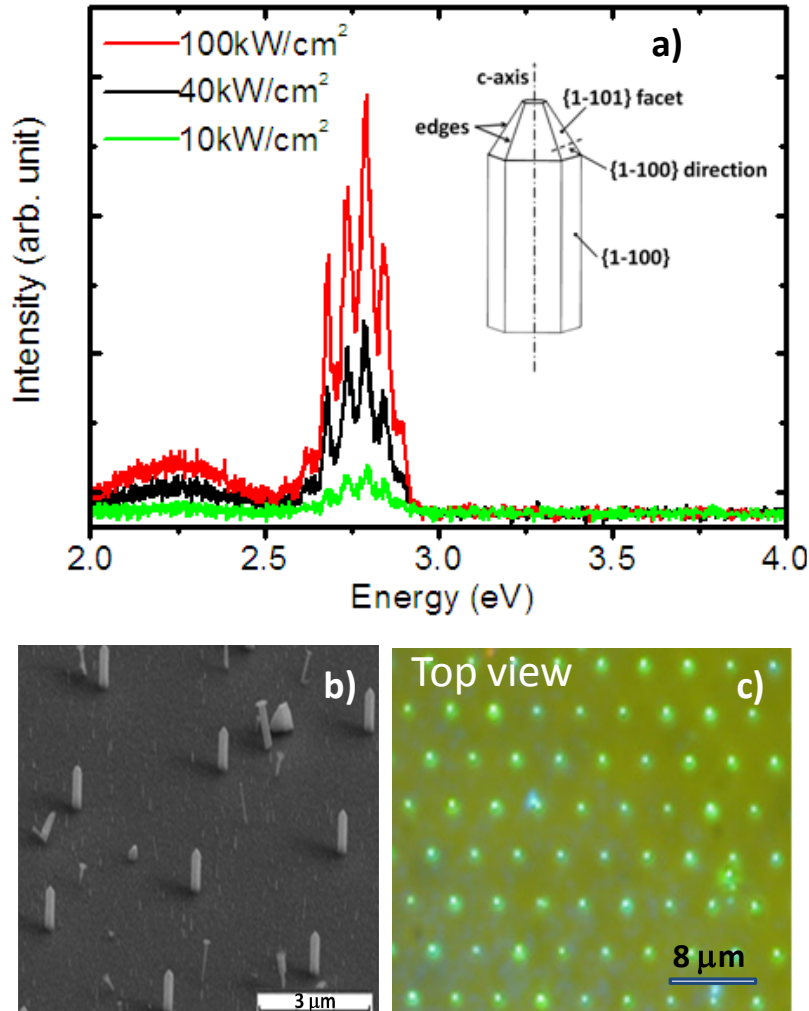


Fig. 1. (a) Schematic representation of an individual NFQ-disk. (b) μ PL spectra of an individual Q-disk1 obtained for three different excitation power densities. (b) Bird's eye view of the sample by scanning electron microscope. (c) Image of the sample acquired by luminescence microscope.

mechanism of exciton in these pyramidal structures, we analyzed the linear polarization of the emission peaks emitted by LCs in the InGaN layer of the NFQ-disks. By analyzing systematically the linear polarization of the sharp emission peaks in the μ PL spectra of about 10 NFQ-Disks, we aimed to correlate the polarization properties of the LCs to the local strain distribution through the facets of the pyramidal heterostructure. In other words, we want to use the LCs as a nano-scale optical tool to probe the local strain distribution through the facets of the pyramidal structure.

Figure 2(b) presents the spectrum obtained from another NFQ-Disk₂ of the same sample, exhibiting a larger number of LCs that participate to excitonic radiative recombination. We could fit the entire spectrum by a sum of Lorentzian functions, confirming that these sharp peaks are emitted by individual 0-dimensional objects: LCs. Given that the photoluminescence in bi-dimensional InGaN/GaN QWs grown on {1-101} facet is linearly polarized along the {1-100} direction [22], one could expect that the polarization orientation of the LCs emission peaks would be statistically distributed along the {1-100} direction of

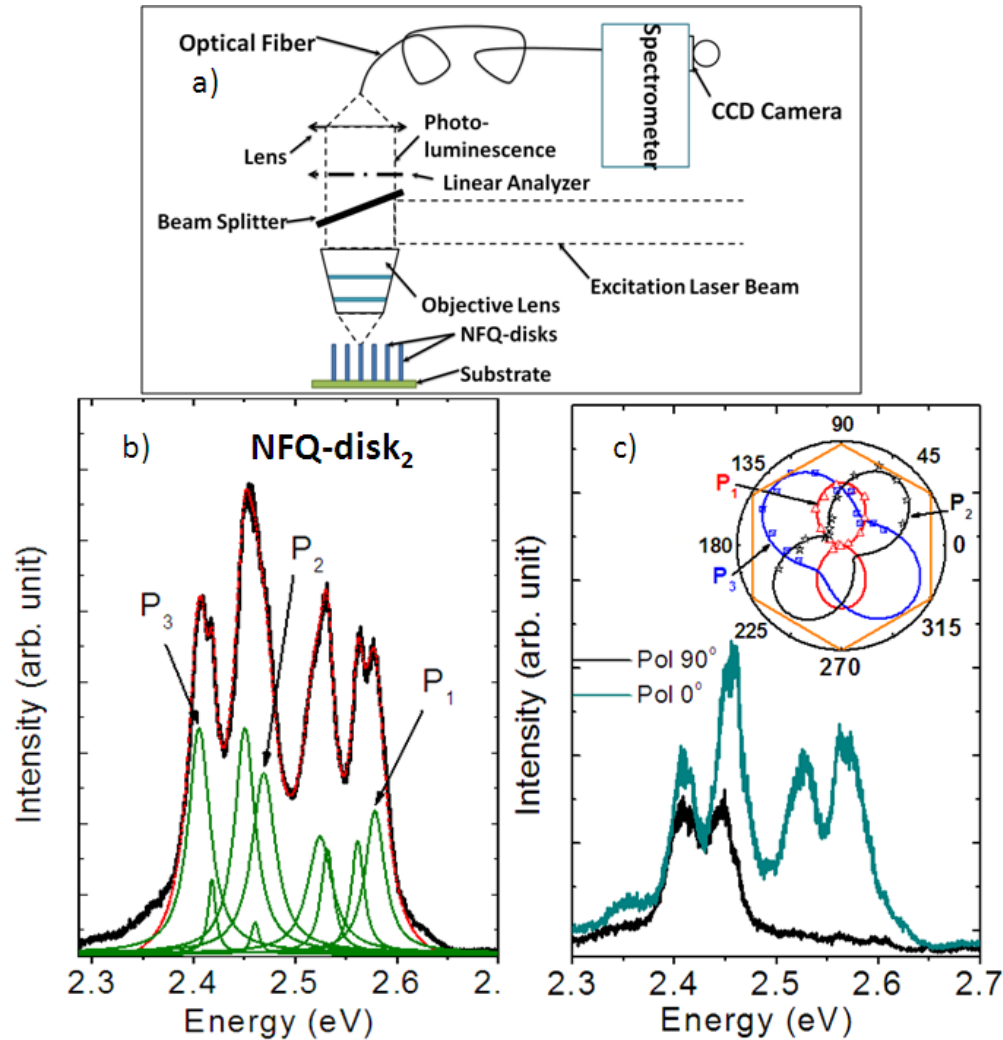


Fig. 2. (a) Simplified schematic view of the experimental setup use in polarization-resolved μ PL measurement. (b) In black line, μ PL spectrum of the NFQ-disk₂; in red dash line sum of the Lorentz function fit plotted in green lines. (b) Spectra of the NFQ-disk₂ acquired for two cross position of the linear polarizer. In the inset is plotted the variation of the integrated intensity of the peaks P₁, P₂, and P₃ defined in (c). The orange hexagon represents the shape of the NFQ-disk.

each one of the six m-facets (see inset of Fig. 1). Thus, we expected that the distribution of the polarization orientation of the LCs emission peaks would be defined by three principal direction separated by an angle of 60° . On about 10 NFQ-Disks investigated, we could not extract such behavior: the polarization orientation of the LCs emission peaks is uniformly distributed. As an example, in inset of Fig. 2(b) we plotted in polar coordinate the dependency of the integrated intensity of the emission peaks P₁, P₂ and P₃ function of the polarizer angle. The angle between the polarization directions of these three peaks differs from a multiple of 60° . Moreover, we could identify two types of emission peaks considering both the polarization orientation and the polarization degree $D = [I_{\max} - I_{\min}] / [I_{\max} + I_{\min}]$ (with I_{\max} (I_{\min}) the maximum (minimum) of the integrated intensity of the emission peaks): (i) below 2.5 eV, the emission peaks are polarized with a random orientation and exhibit a polarization degree of about 60% as observed in self-organized Q-disks sample [19] – (ii) the peaks emitted above

2.5 eV are nearly totally polarized (i.e., $D \approx 1$) and contrary to what is observed below 2.5 eV, they have the same polarization orientation θ_0 . We could identify the polarization angle θ_0 as parallel to one of the edge of the pyramidal structure with an angle error of $\pm 5^\circ$. In bi-dimensional $\{1-101\}$ InGaN/GaN quantum well, the fluctuation of the thickness as well as the In composition of the active layer can affect the polarization properties of the emitted light at the macroscopic scale. However, while in bi-dimensional QWs the luminescence collected in common photoluminescence experiment arise from a macroscopic number of LCs, through μ PL experiment on single NFQ-disk, we have access to the luminescence coming from few individual LCs. Consequently, the randomness of the polarization direction of the sharp emission peaks below 2.5 eV should be related to very local effect through each facet of the pyramidal structure. Because of the parallelepiped shape and the nano-scale of the facet, two LCs placed at two different position of a same facet must undergo a completely different strain environment and thus exhibit two different polarization orientations. Furthermore, the total polarization of the group of emission peaks observed above 2.5 eV, should be related to a huge strain gradient and/or an important shape elongation of the LCs [21]. Such huge strain distribution or shape elongation could be induced by the edges of the pyramidal structure. As all these peaks at upper energy (>2.5 eV) are all orientated along the same direction θ_0 , we believe that this group of totally polarized emission peaks (such as P_3) must be assigned to a chain of LCs [23] formed in the vicinity of one and same edge of the structure.

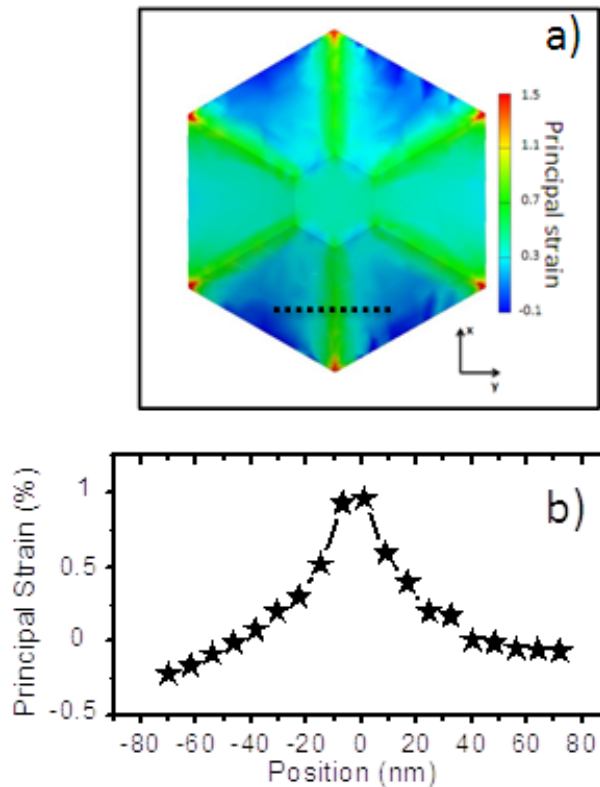


Fig. 3. (a) Principal strain distribution through the InGaN layer of a NFQ-disk. (b) Plot of the principal strain distribution extracted along the black dashed line in Fig. 3(a).

3. Finite element method calculation

To reinforce our interpretation, we calculated the strain distribution through the InGaN active layer of an individual Q-disk by 3-dimensional finite element method (FEM) calculation. In this simulation, we considered a 3 nm thick $\text{In}_{25}\text{Ga}_{75}\text{N}$ layer grown on a completely relaxed GaN nano-column with a lateral diameter of 250 nm. To ensure an optimum adaptation with the 3-dimensional pyramid shape of the InGaN layer in the NFQ-disks, we considered a meshing structure based on tetrahedral unit volume. The calculation was performed by meshing the entire nano-column from the GaN base to the GaN cap layer, by adapting smoothly the mesh structure from the bottom to the top of the NFQ-disk. The size and the local density of the tetrahedral meshes vary depending of the position in the layers. In average, the density of mesh in the InGaN layer was about $2 \times 10^8 \mu\text{m}^{-3}$. To visualize the strain distribution through the pyramidal InGaN layer, we plotted in Fig. 3(a) the principal strain distribution obtains from our FEM simulation. In accordance with our experimental results, it appears that a strong strain gradient take place in the vicinity of the edges of the pyramidal layer. As seen in Fig. 3(b), the strain increases from the inner part to the facets perpendicularly to the edges. This result suggests that the optical properties of the NFQ-disks cannot simply be deduced from what is already known for bi-dimensional $\{1-101\}$ InGaN/GaN QW structures; the effect of the shape of the nanostructure on the strain distribution in the active layer of the heterostructures should be considered carefully. Supplementary investigation such as cathodo-luminescence studies such as in the work realized by M. Merano *et al.* [24] on micrometric scale pyramidal InGaAs/AlGaAs heterostructure, should be performed to obtain accurate information on the optical properties of through InGaN/GaN NFQ-Disks. To confirm our FEM calculation of the strain distribution through the facets of the NFQ-disks and to emphasize the link with our experimental more accurate and sophisticated calculation based on theoretical model should be conducted to show how the strain distribution linked to the hexagonal shape affects the excitonic bound states of the LCs.

4. Conclusion

In conclusion, we reported for the first time linear polarization analysis of individual InGaN/GaN NFQ-disks. Other factors such as the fluctuation of the thickness and the In composition of the InGaN layer can influence the optical properties of such NFQ-disks [25], however our results suggest that the morphology of the latter induce a complex strain distribution in the active layer. Thus, the excitonic optical properties of the InGaN active layer in the NFQ-disks cannot be simply deduced from what has been reported from bi-dimensional $\{1-101\}$ InGaN/GaN QW. The particular morphology of NFQ-disks calls for deeper experimental and theoretical investigation in order to clarify the intrinsic relation between the shape and the strain distribution of the NFQ-disks. Moreover, our experimental study shows that QD or QD-like nano-objects can be considered as optical nano-tools to probe the very local structural properties through nano-scale heterostructures.

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