Real-time near-field evidence of optical blinking in the photoluminescence of InGaN by scanning near-field optical microscope

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Abstract: InGaN/GaN devices are currently used for many applications, for example, full color display, white (RGB) illumination systems and for the realization of shorter wavelength emitters for optical data storage. We previously reported a blinking phenomenon in the photo-luminescence of InGaN device ready single quantum well materials. In this study we observe in high resolution this optical instability with a near-field nano-probe. The phenomenon appears only in local confined domains and does not seem to behave as a bistable state process like reported on quantum dots generated photo-luminescence. We investigated by a modified scanning near-field optical microscope (SNOM) and studied the time/intensity profile of the optical signal with a resolution in the range of 100*nm*. The dynamics of the blinking was time-resolved and its behaviour studied with Fourier analysis. Despite the intensity oscillations were found to have chaotic component (autocorrelation coefficient is about 0.63), the optical oscillations appear to include regular characteristics. Fourier analysis of the light intensity from confined domains exhibit peaks in the range of 4-5 s. The emergence of these intriguingly slow and partially regular dynamics should shed light on the inner mechanism that are involved in the fundamental processes of optical emission in these devices.

OCIS codes: (180.4243) Near-field microscopy; (230.5590) Quantum well devices; (250.5230) Photoluminescence; (160.6000) Semiconductor materials; (190.3100) Instabilities and chaos.

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Recent advances in studies for InGaN/GaN have led to high-brightness green and blue light emitting diodes (LED). InGaN/GaN are currently used for many applications, for example, full color display, white (RGB) illumination systems and for the realization of shorter wavelength devices for optical data storage [1].

In a previous research we have revealed optical instabilities (blinking phenomena) in $In_xGa_{(1-x)}N$ single quantum well systems (SQW) [2,3]. Similar fluorescence blinking has been observed in multiple quantum wells such as InGaN clusters [4]. In these samples the carriers are trapped in a three dimensional quantum box where a mechanism of Auger ionization intervene to produce intermittent blinking phenomena [5, 6]. In our samples, quantum confinement is restricted to one of the three available directions, so blinking in our case is not trivial and there is not yet an universal explanation [7].

In this paper we study a device ready $In_xGa_{(1-x)}N$ system composed by a 4μm GaN with a 3nm SQW InGaN active layer covered by a 5nm GaN capping, all grown on a sapphire substrate. We want to focus particularly on the local characteristic of the luminosity of these emitters. Confined domains appear to blink irregularly. If observed with due attention, the blinking areas are recognizable with optical fluorescence microscopy; to the eye they appear to fluctuate in intensity with a not-regular and mostly not-intermittent slow activity. Figure 1 shows the optical instability phenomena as recorded by a CCD camera. The circled point changes intensity in a fashion that appears chaotic, however FFT analysis shows a characteristic peak of a few Hz. In Fig. 2 we show a frame of the acquired original video (Media 1, Media 2) before processing.

Studying various time/intensity profiles, we noticed the presence of different types of blinking points that have different time characteristics. In a first more rare type, the dot appear to switch between two different intensity levels ('two level blinking point' or 'telegraphic blinking') [4]. In the second type, blinking points appear to have a continuous range of intensity levels, the blinking seems purely chaotic ('multi level blinking point'). We are particularly interested and we are going to focus here in these latter unstable points.

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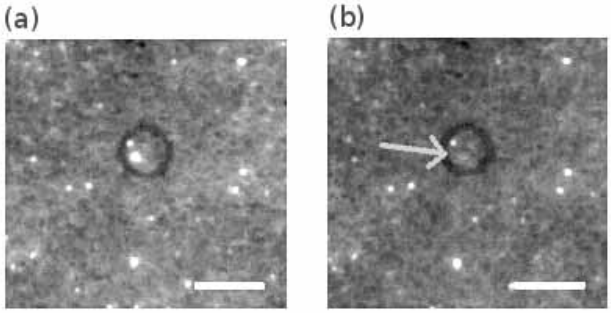

Fig. 1. Optical microscope PL images of a 510 nm $In_xGa_{(1-x)}N$ sample. An unstable blinking point is circled and indicated by an arrow. The figures (a) and (b) are gray scaled and contrast enhanced for clarity. Images are taken about a few seconds apart. The sample was excited by Ultra violet Hg lamp (about 425 nm). Bar is 10μm.

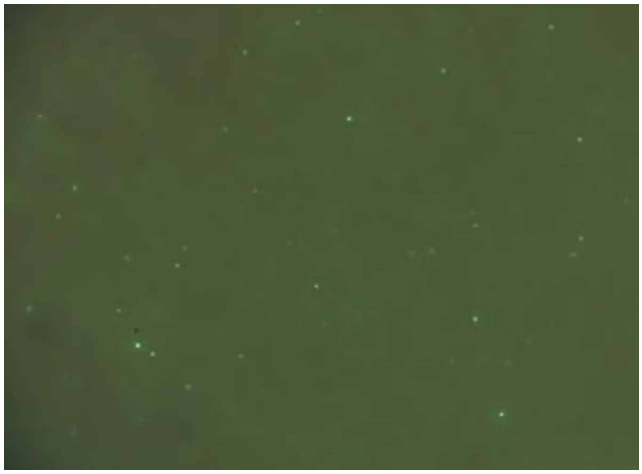

Fig. 2. The recording by CCD camera of a region containing more than one blinking point (Media 1). The sample and conditions are similar to those in the digital pictures of Fig. 1, however CCD camera media processing is different so image contrast appears different.

The described blinking phenomena can be caused by different effects. One effect is due to the presence deep traps [6]. Defects of crystal structure and doped impurities can cause deep localized states where carriers are trapped. The carrier recombination does not occur when a carrier is trapped in these confined states. The trapped carrier will finally cause thermoluminescence and escape to the ground state. This cycle suggests that observed two level blinking phenomena is related to these deep traps [8–10].

A second process that can be involved is related to quantum-confined Stark effect (QCSE) [11]. An electron caught in a deep localized state cause internal fields that modify locally the quantum well band. This alters the local wave-function matrix element that regulates the recombination rate. This results in a reduced photo-luminescence until a de-trapping phenomena intervene. Models suggest that blinking phenomena involve defects or impurities within the crystal structure. Therefore the investigation of this phenomena can be seen as a way to under-

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stand and evaluate mechanisms that relate with the crystal quality of InGaN/GaN LEDs with a pure optical methodology.

To focus on these processes with higher spatial discrimination, we used the scanning near-field optical microscopy (SNOM) [12]. Conventional microscopy have resolution limited by diffraction. SNOM is not subject to this limitation and can offer up to nanometre rage resolution and allows direct time resolved measurements.

The sample we used are grown by metal-organic chemical-vapor deposition method on a sapphire substrate. These type of samples are characterized by a higher density of threading dislocations due to epitaxial growth on mismatched sapphire substrates at high Indium concentrations. The sample has three layers, a GaN layer (4 μm), an $In_xGa_{(1-x)}N$ active layer (3 nm) and a GaN layer (5 nm). The band gap edge of GaN is about 3.4 eV (365 nm) and InGaN is generally under 2.95 eV (over 420 nm) [13]. The macroscopic photoluminescence (PL) peaks of the samples we used are located at about (a) 510 nm and (b) 540 nm at room temperature (see Fig. 4).

The SNOM system used for this experiment is a setup originally modified by us specifically for this experiment. An He-Cd laser was used as excitation source (325 nm single emission peak). We know that the 325 nm line excites both the GaN barrier and the $In_xGa_{(1-x)}N$ active layers. The fiber probe used for our study is shaped by chemical etching. The etching solution is mixed in this volume ratio: HF(50%):NH_4F(40%):H_2O = 1:2:1 [14, 15]. All measurements were performed at room temperature.

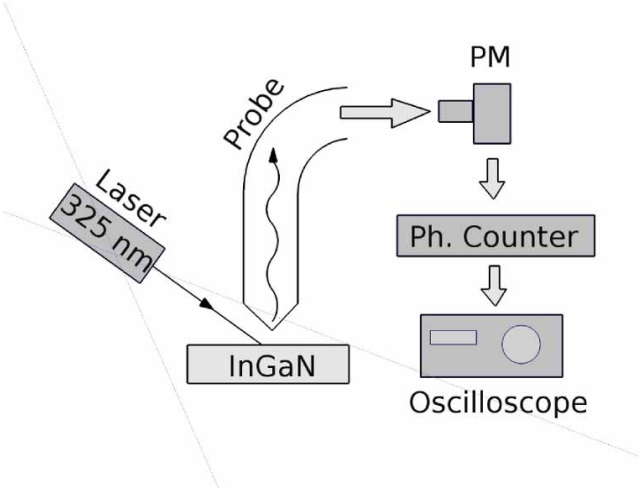

Fig. 3. A schematic explanation of detection mechanism: A photomultiplier tube (PM) collects the photoluminescence signal from confined 100nm area. Then a photon counter integrates each photon spike over 30ms and the digital signal obtained is fed to the oscilloscope that maps the entire sample or plots the time/intensity profile shown in Figs. 5 and 6.

Figure 3 shows a simplified scheme of the setup. The integration time of the photoncounter is 30 msec, to have a margin of redundancy the oscilloscope is collecting the data at about 100ms per pixel. During SNOM mapping the probe is maintained in the proximity of the surface (few nanometer distance) by a shear force controller [12]. We acquire the intensity map of the evanescent optical field in the sample surface, called here 'SNOM PL map'. The light is generated by a He-Cd laser, is then reflected by a mirror and from there reaches the sample

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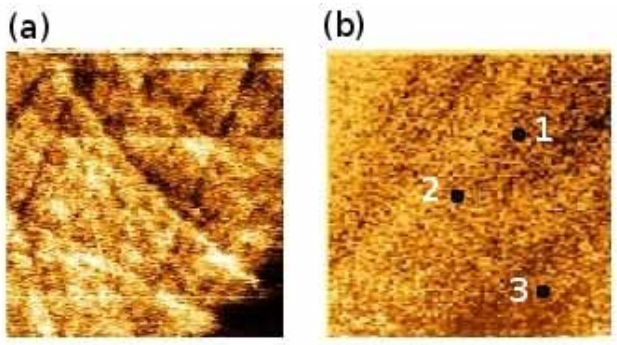

Fig. 4. The PL mapping of (a) 510 nm and (b) 540 nm samples taken in near-field. Image size is $15\times15\ \mu m^2$ composed of 128×128 pixels. The position of three domains where the photoluminescence time-profile was recorded are indicated by the dark spots and named by numbers.

surface without passing through other optical elements. The sample surface is scanned over an area of about 15x15 μm^2, while the photoncounter detects the photo-luminescence generated from the sample surface through the fiber probe. Figure 4 shows SNOM PL mapping of the (a) the 510 nm sample and (b) the 540 nm one.

First of all, these results show that characteristics inhomogeneous patters are present also at this smaller spacial scales, and compare well with already observed PL maps obtained by conventional optical microscopy [2].

To study in real-time the photo-luminescence time dependence, the fiber probe was held on a single pixel for the whole time of the measure. We recorded the optical intensity for 20 sec using a digital oscilloscope. This confined PL was converted to a .txt file and then processed off-line to analyze the temporal PL changes of InGaN/GaN SQW for different local domains (see Fig. 4(b)). We plotted a single pixel time/intensity profile in Fig. 5(a). The signal suggests that the single pixel is a 'multi level blinking point'. In the time/intensity profile no discrete intensity levels can be identified. To test regularity of the signal, we calculated the autocorrelation function and found a r coefficient of about 0.63, indicating that the behaviour is not pure chaos.

Despite the apparent random behaviour, we did a Fourier study in order to find any regular frequency embedded in the data. Figure 5(b) shows a characteristic peak (5 Hz) in the signal analysed by Fast Fourier Transform (FFT).

Not all the blinking pixels have the same Fourier spectrum, however many of them presents higher peaks in the range of 5-6 s. This characteristic was checked against instrumental artefacts. It is a real optical feature coming from the sample emission process, it disappears in not blinking regions and appears over different points at slight different values.

For comparison we give another example blinking point chosen form the scan in Fig. 4, with its real time blinking profile plotted in Fig. 6. A complex distribution of peaks is visible, with again a prominent peak in the 5-6 Hz range. At this stage we cannot give a complete explanation on the nature of the observed blinking behaviour. The fact that the optical instability is not purely chaotic and the presence of regular components in the range of few Hertz suggests that the phenomenon convey information related to a slow transitional process presumably involving trapping and de-trapping of carriers within the quantum well. Further investigations are currently under design.

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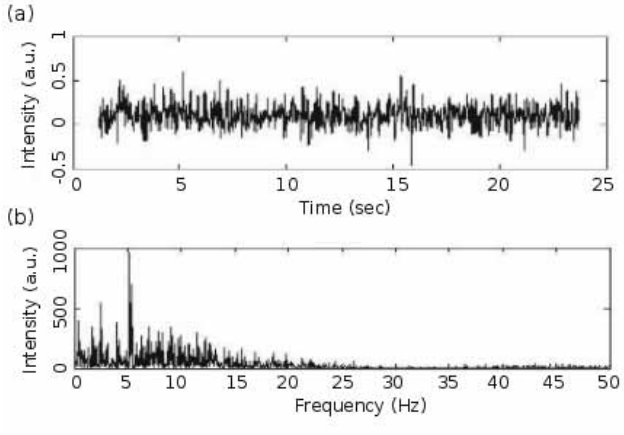

Fig. 5. (a) PL intensity trace of $In_xGa_{(1-x)}N$ SQW as a function of time. Recorded at spot "1" in figure 4(b); the point is blinking, but this fact is not apparent from the time-domain plot (plot is 2250 points). In panel (b) the frequency/intensity profile reveals the blinking characteristics of the signal (panel (a) a.u. fluctuations correspond to about 5% absolute intensity differences). An evident peak is noticeable around 5 Hz. We cannot clarify the nature of this peak at this stage, however it is presumably related to slow trapping and de-trapping processes within the quantum well.

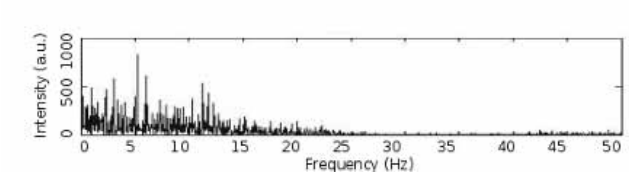

Fig. 6. Fourier trasform of signal taken from point "2" in Fig. 4. The frequency/intensity profile shows again a regular oscillation of about 5-6 Hz. This behaviour is compatible to what observed in other blinking points on the same sample.

We demonstrated that blinking phenomena happens also in the photo-luminescence of $In_xGa_{(1-x)}N$ Single Quantum Well systems. In these crystals the quantum confinement is limited to one direction, so differently from quantum dot structures like InGaN clusters, the explanation for the optical instability is not straightforward.

We presented high resolution photo-luminescence maps of 540nm and 510nm emitters and studied point by point the optical time profile using a nano-probe located in the proximity of the actual luminescence center. The experiments were performed by the use of a modified optical near-field microscope. Intriguingly, we found that despite the chaotic appearance of the optical signal, Fourier analysis demonstrated the presence of characteristic frequencies of oscillations of about 5-6 Hz embedded in the signal. This suggests the existence of multiple regular oscillations of similar frequencies that interfere in a pseudo-chaotic way resulting in the observed slow intermittent signals. The study and clarification of the physical mechanisms producing this phenomena is important for the understanding of the fundamental processes of optical emission in InGaN materials and may help to the effort of producing new and better optical emitting devices.

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