Transient memory effect in the photoluminescence of InGaN single quantum wells

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Abstract: The transition to maximum photoluminescence of InGaN single quantum wells is a phenomena that has time constants in the range of few seconds. Using a systematic illumination/darkening procedure we found that these characteristics are related to previous stimulations as if the sample has a memory of past illumination events. Choosing opportune time sequences, time constants were observed to vary more than 100%. These facts suggest the presence of carrier trapping/de-trapping processes that act beyond the single illumination event, accumulating over time in a complex effect.

OCIS codes: (250.5230) Photoluminescence; (310.5448) Other Optical properties; (160.6000) Semiconductor Materials; (230.5590) Quantum well devices

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Wide-band-gap GaN based quantum wells[1] are the essential part of many electro-optical devices, such as light-emitting-diodes (LEDs) or laser diodes, the precise nature of the light emitting process has not fully been understood due to the complex material physics and InGaN engineering involved.[2, 3, 4, 5, 6, 7] A particular feature of InGaN photoluminescence (PL) is its tendency to slowly alter its PL emission, a phenomena that has been noticed by various authors[8, 9, 10, 11, 12, 13, 14] and often called ”fatigue”, ”degradation” or with similar expressions.

In this study we want to focus on this phenomenon and present systematic tests concerning the PL time dependence of the main peak of InGaN single quantum wells (SQW). Our measurements confirm that, right after starting to illuminate the sample, the PL coming off the GaN/InGaN single quantum well rises or falls exponentially depending on conditions with time constants in the range of seconds. Using systematic illumination/darkening sequences we could also prove that these time constants change depending on the illumination history. The device behaves as if it has a memory of previous stimulations.

The samples used in this study were grown on an (0001) oriented sapphire substrate by metallorganic chemical vapor deposition technique developed at Nichia Chemical Industries Ltd. The layers are $In_xGa_{(1-x)}N$ single quantum well structures composed of an undoped GaN layer (4 μm), an $In_xGa_{(1-x)}N$ active layer (3 nm) and a GaN layer (5 nm). We used samples with different Indium concentrations resulting in different main emission peaks located at about 420, 460, 510, 540 nm at room temperature.

Figure 1 shows the experimental setup. It consists of a photoluminescence microscope coupled with a monochromator. We excited the samples with an ultraviolet Hg lamp (365nm and 405nm emission lines). Considering that the band gap of InGaN is about 3.44 eV (365 nm) and the one of InGaN is under 2.95 eV (over 420 nm)[15], we know that the 365 nm line excites both the GaN and InGaN layers. On the other hand, the 405 nm line only excites the $In_xGa_{(1-x)}N$ active layer.

First of all, we observed that the PL spatial pattern is not stable after starting the illumination. Figure 2 shows the PL pattern of the sample right after starting the illumination and after one minute of continuous illumination with the 365 nm line. The contrast and general luminosity appeared increased in the second map, smaller domains seems to disappear and darker regions become more visible suggesting that major spatial reorganization in the carrier recombination process is occurring.

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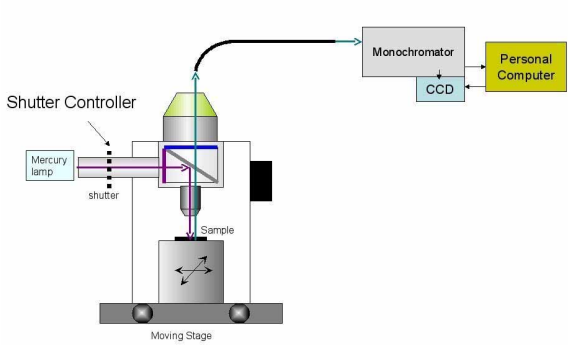

Fig. 1. The apparatus used to perform selective photoluminescence consists of a UV microscope coupled with a monochromator to observe the intensity-time profiles of the photoluminescence at single wavelengths

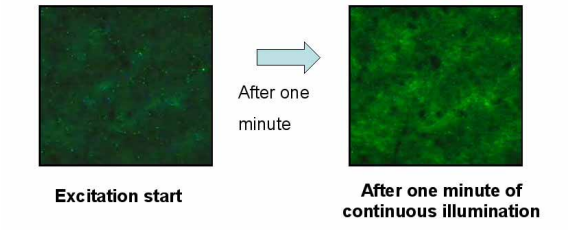

Fig. 2. The difference in the photoluminescence pattern right after starting the illumination (left picture) and after one minute of illumination (right). The contrast and granularity appear enhanced in the latter case, smaller bright domains disappear and some other are modified. Image taken with an Olympus BX51-W1 microscope, with 40x objective lens. Digital camera parameters and integration constants are identical.

We observed the intensity-time-profiles of the yellow band and the main emission peak of a 460 nm centered sample (Fig. 3) . Figure 4 shows the temporal behaviour of the intensity after starting to illuminate the sample with 365 nm light. The yellow band immediately reaches the maximum intensity, whereas the main emission peak rises exponentially with slower time constant.

For further investigation of the behaviour of the main emission peak we exited the samples selectively with 365 nm and 405 nm (Fig. 5). The left graph shows the intensity of the main peak excited with 365 nm. The right graph shows the time-intensity profile of the main peak (same sample, same place and conditions) now excited with the 405 nm line. The intensity reaches a maximal value and then falls exponentially. We fitted the 365 nm graph with $Y_0 - A_1 exp(-t/\tau_1) - A_2 exp(-t/\tau_2)$ and the 405 nm excited one with $Y_0 + A_1 exp(-t/\tau_1)$.

After noticing irregular behaviour in the tests, we decided to verify if the time profile changes for different illumination history. For this purpose we arranged an illumination interval sequence and did a series of measurements as shown in Fig. 6. The measurement and illumination time was fixed to 40 s whereas the darktime was varied beginning from 5 min up to 60 min.

The tests confirmed that the time profile time constants τ_1 and τ_2 depend on the previous illumination history (Fig. 7). The first graph shows the time constants τ_1 and τ_2 against the dark time, where the darktime was varied form long (60 min) to short (5 min). The second one shows

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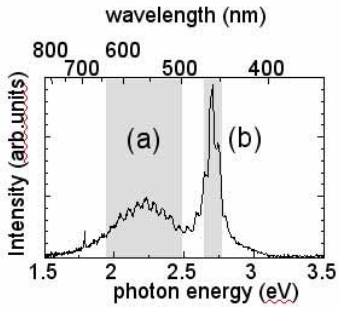

Fig. 3. The spectrum of the sample used for the measurements with the main emission peak located at 460 nm (b) and its yellow band (a).

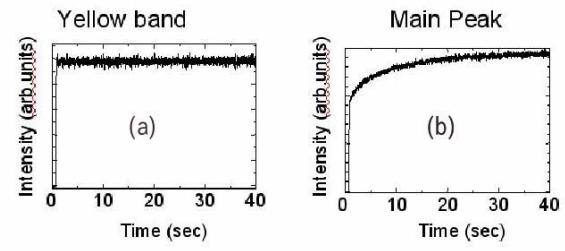

Fig. 4. (a) shows the time intensity profile of the yellow band after starting the illumination, (b) the time intensity profile of the main emission peak

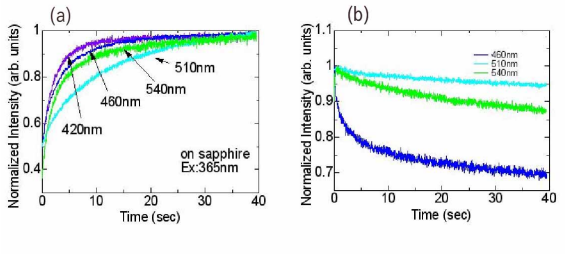

Fig. 5. Selective excitation results in different rising/decreasing photoluminescence behaviour. (a) Excitation with 365nm (a) produces a PL rise, whereas with 405nm (b) produces a decreased luminescence. The PL rise profile is fitted with a double exponential, in the case of PL decrease the best fitting is a single exponential.

the same illumination darktime sequence in reverse time order. The increase of the dark-time provokes a longer time constant. At corresponding times the time constants in the second graph are almost double than in the first graph.

We also tested the temporal behaviour of the PL at other wavelengths outside the main emission peak. Using the monochromator we selected different wavelengths located around the main emission peak of the 460 nm sample and analysed their time-intensity profile with the fitting mentioned above. We found that the time constants are different for different wavelengths of the spectrum. Figure 8 shows the spectrum of the 460nm sample and the time constants τ_1 and

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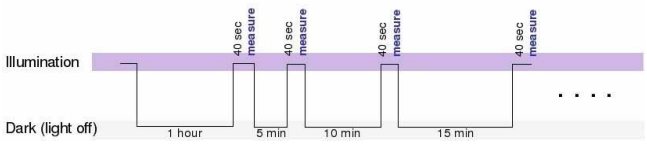

Fig. 6. The illumination interval sequence. Measurements with 40s of illumination were done starting after a dark time of 1 hour, then after increasing darktimes from 5 min up to 60 min (5 min steps). A similar series was done in reverse time order (from 60 min darktime to 5 min)

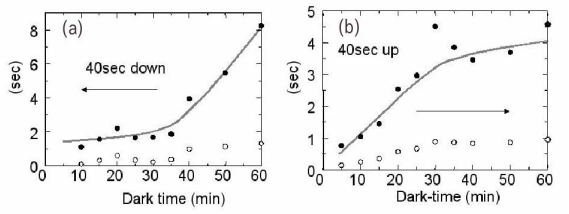

Fig. 7. A systematic on/off illumination sequence reveals a variation of PL rise/fall time constants. In the left graph (a) the dark-time was varied from 60 min down to 5 min, in the right one from 5 min up to 60 min.

τ_2 of the time profiles for different wavelengths. The time constants increase with increasing photon energy.

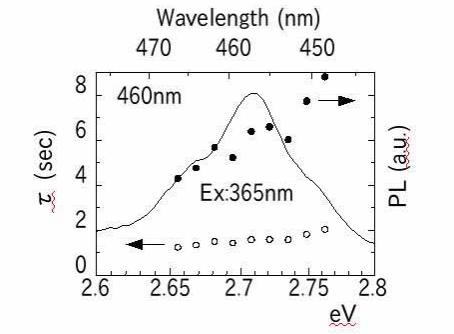

Fig. 8. This panel shows the spectrum of the 460 nm centered sample and the time constants for the photoluminescence at wavelengths near the main emission peak.

We confirmed that the PL of InGaN single quantum wells does not appear as a quasi-instantaneous process, but rises or falls exponentially with slow time constants. For excitation with 405 nm light, which only excites electrons in the InGaN layer of the sample, the emission falls. For excitation with 365 nm light, which excites both electrons in InGaN and GaN layer, the emission rises. Furthermore, it has been demonstrated that these samples remember past illumination events; different photoluminescence rise and fall behaviour is observed depending on previous illumination sequences. This fact suggests that processes related to carrier trapping and de-trapping is taking place and these act beyond the single illumination event, but cumu-

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lating over multiple on-off sequences. We believe that the slow processes in these phenomena are somehow connected with the phenomenon of photoluminescence blinking, another slow process characterized by long time constants and observed on similar samples [16, 17]. These tests may be related to the general light emitting process in InGaN structures, and help to pave the way for the realization of better and more efficient light emitting devices.

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