Study of (0001) InGaN SQW optical memory effect correlated to the increase of intense emission local domains by time-resolved photoluminescence and thermal and pressure dependence.

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ABSTRACT

InGaN is used in optical devices such as light emitting diodes (LED), laser diodes (LD) etc. The crystal structure has many strain caused by lattice mismatch. Also, it has Indium composition fluctuation due to the miscibility gap and this is forming quantum dots (Qds) in the crystal structure. For these reasons the photoluminescence is characterized by an inhomogeneous pattern of local domain (bright spots) that appears in the photoluminescence images. These defects in the InGaN crystal structure is considered to cause instable blinking and optical memory effect in time-resolved photoluminescence (TRPL). There is not an unified description of the physical mechanism of these phenomena. In this study, we find that the bright spots increase in number with time in certain conditions, this is related to the instability blinking and optical memory effects on InGaN single quantum well (SQW). We used TRPL and found that intense emission is localized and changes with the scale of several tens of minutes. Because this phenomena is in long time scale, we think that excitation light promote change in crystal strain and sample emission. Also, we studied the thermal and pressure dependence of the optical emission and found variation of the number of bright spots, we will give our first insights on the reasons of this phenomenon too.

Keywords: InGaN, Photoluminescence, defects, strains, Instability blinking, Optical memory effect, quantum dots

1. INTRODUCTION

1.1 InGaN

InGaN is a mixed crystal of GaN and InN. It widely covers the band gap between near-ultraviolet (3.4eV) and infrared (0.7eV) depending on Gallium and Indium composition ratio. So, in principle it is a material that is expected to produce light-emitting devices of various colors [** IF POSSIBLE reference. (Kawakami, Funato) **]. Recently, InGaN-based LED of green ~ ultraviolet region and white LED with phosphor have into practical use. However, the production methods and understanding of InGaN devices has problems. The structure of this crystal has lattice mismatch in the range of 15% between GaN and sapphire substrate and 11% between GaN and InGaN[1]. So, the threading dislocation density is 10^{8}~10^{10}cm^{-2} [2]. In spite of very large defects and strain, InGaN optical devices achieve very high internal quantum efficiency and optical emission. In general, a crystal with many strain has piezoelectric field, and the internal quantum efficiency is reduced significantly because of that. In addition, InGaN quantum well has high Indium composition area such as quantum dots known to induce local excitation emission[3] (we call those bright spots or bright points). The formation of region like quantum dots is caused by miscibility gap on mixed crystal, this is presumably the cause of confined domain of high quantum efficiency that are seen as intense luminescence center. These variety of complex optical properties remain unknown completely.

1.2 Defects in crystal structure

Recently, instability blinking[4] and optical memory effect[5] in TRPL of InGaN SQW was found. It is thought these phenomena are caused by the presence of defects and strain within the crystal. In addition to InGaN, instability blinking was found in other optical devices such as CdSe nanocrystal[6] and the epitaxial grown film[7], ZnCdSe QDs[8], GaAs QDs[9],

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InP Qds[10], and Porous silicon[11]. Instability blinking in InGaN SQW is localized around a QDs. The instable blinking in InGaN SQW is a phenomenon in which a localized regions of few μm of diameter blinks unstablily. It has thermal dependence and the flashing intensity width becomes extremely small at low temperatures of 280K or less[12]. Therefore, the phenomenon is associated to beating of slightly different thermal waves vibration creating unstable optical blinking. Instead, the so called "optical memory effect" was first found in the GaN epitaxial film[13]. Optical memory effect in InGaN is the phenomenon in which the large scale photoluminescence emission becomes stronger at the time scale of seconds to a few minutes, a slow growth of optical intensity. This growth is general all over the sample and depend also from the previous illumination history of the sample as the sample has a "memory" of what happened in the past (so the name optical memory effect). A complete and universally accepted theory to explain these two phenomena is unknown.

2. EXPERIMENTAL METHOD

2.1 Sample

We used InGaN SQW sample made by metal organic vapor chemical deposition (MOCVD). This sample is a double heterostructure consists of 3nm InGaN layer sandwiched 4μm and 5nm GaN layer on [0001] sapphire substrate (Figure1 left). Indium composition is 0.2 and the luminescence center wavelength is 540nm.

2.2 Experimental device

In this research, I was using the selective excitation fluorescence microscope such as Figure1 (right). The light from mercury lamp irradiates Filter A that transmits only the wavelength of 400nm or less, then the excitation light hits the sample. And by Filter B (pass through 400nm and more), the photoluminescence of sample was recorded by a 3CCD camera. If needed, a shutter can cut off the light from the lamp. Recording area is 500×365μm and frame rate is 60fps (Time resolution is about 16.7msec). In experiment of thermal and pressure dependence, we used cryostat. In addition, the emission intensity is expressed by RGB color (the figure is 0 ~ 255), it is defined as below.

$$\text{Intensity} = (\text{Red} + \text{Green} + \text{Blue}) / 3 \tag{1}$$

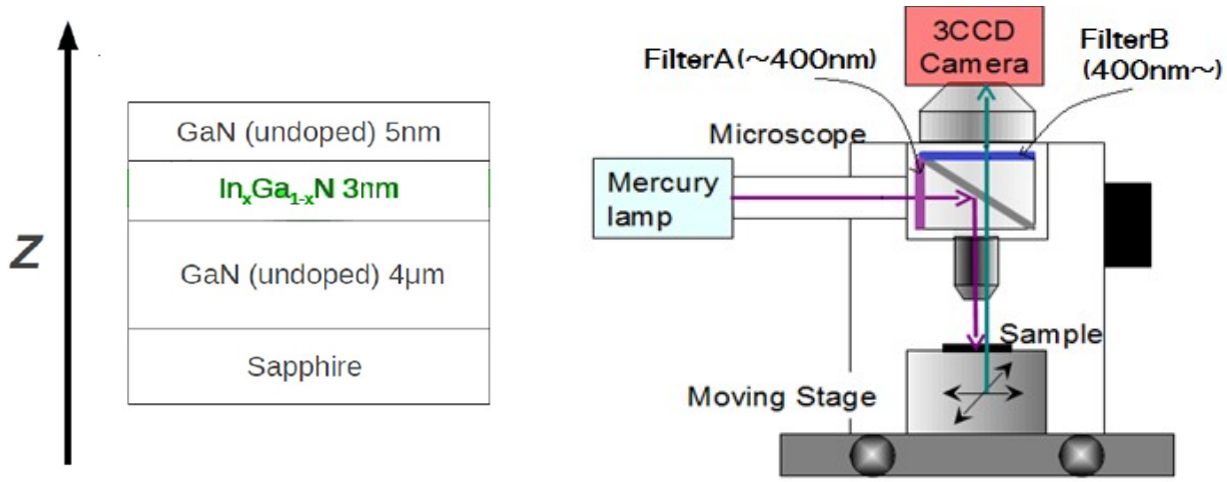

Figure2. InGaN SQW sample (left) and selective excitation fluorescence microscope (right)

3. RESULTS AND DISCUSSION

3.1 Finding and isolating the blinking points intensity from the rest of the sample

In this experiment, we observed the increase of intense emission local domains by photoluminescence for the first time. We recorded changes in emission surface by TRPL for long time such as one hour. We keep room temperature (300K) and atmosphere pressure (1.0×10^3mBar) constantly. The graph of the time variation cross average intensity of emission surface is Figure3, and picture of the time variation of sample surface in photoluminescence is Figure4. Immediately after the measurement beginning (0 second), the number of bright points is low and the average intensity of emission surface is about 46 (0-255 a.u. scale). Then until 500 seconds, the number of bright spots were greatly increased and the average emission intensity is increased to about 55 (20% gain from the beginning of measurement). Also, while the number of bright points is drastically changing, we were able to observe bright spots to blink unstably (URL1). Then, the average intensity of emission surface hardly changed, and little increase of bright points. Why the phenomenon exists? The important feature of this phenomenon is that this occurs in long time scale such as 10 minutes or more. So, we expect that crystal strain changes by similar long time TRPL in InGaN SQW. Then we considered to observe the same phenomena if we change the crystal strain by thermal expansion and Pressure change.

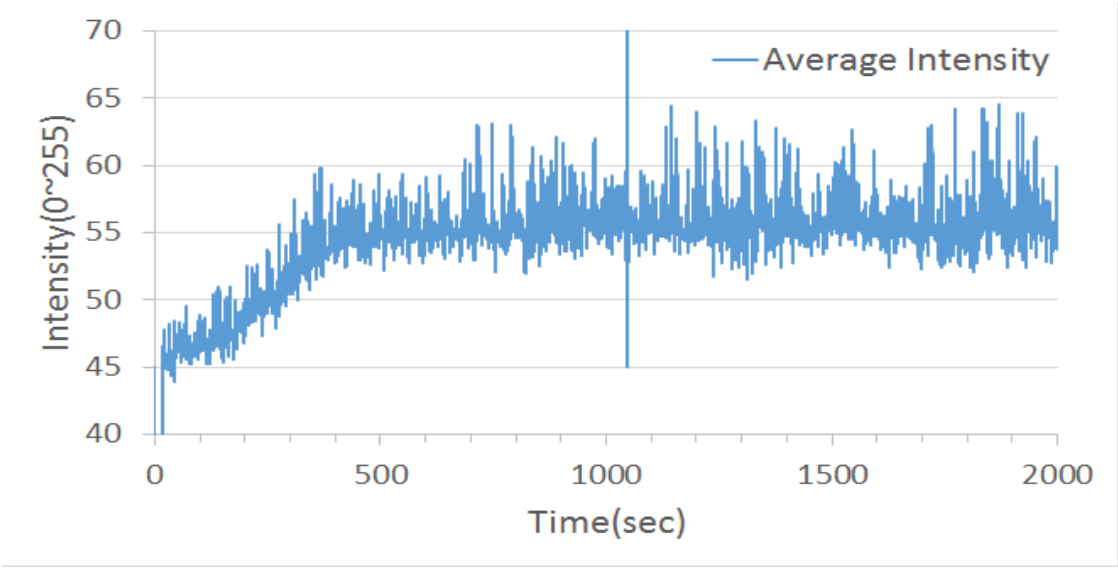

Figure3. Time variation of average intensity of InGaN SQW surface emission in long time TRPL in room temperature and atmosphere pressure. The luminescence intensity was increased excessively to 500 seconds from the measurements beginning, then average intensity hardly changed

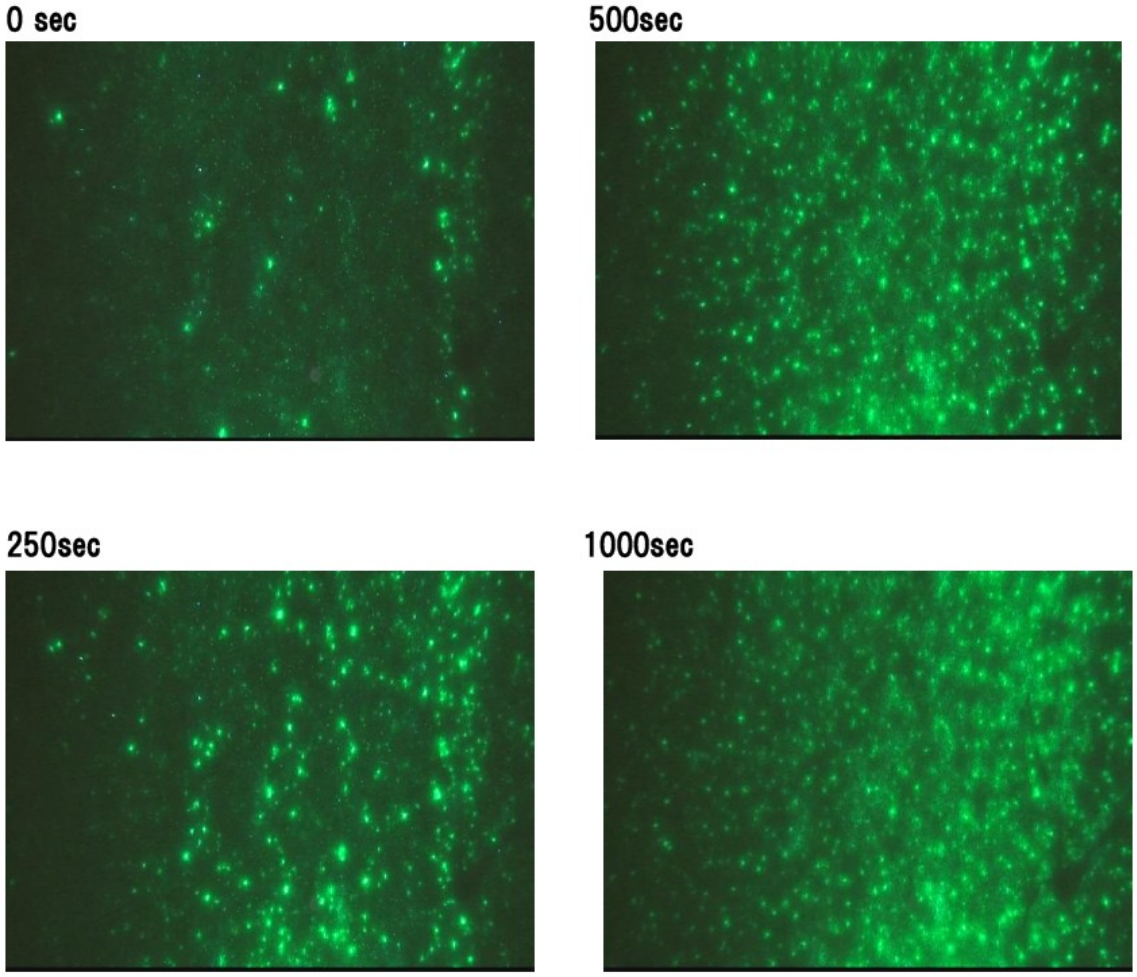

Figure4. Time variation of sample suefece emission by TRPL. We observed increase bright points to 500 seconds from the beginning and then little increase of the spot.(URL1 : http://ruggero.sci.yokohama-cu.ac.jp/data/oneHour_InGaN.avi)

3.2 Temperature dependence of surface emission

We observed temperature dependence of InGaN SQW surface emission. In this experiment, while varied between 300 ~ 320K temperature, we observed the InGaNSQW surface emission by photoluminescence. The time variation of temperature and average intensity of surface emission is shown in Figure5, the picture of time variation of sample surface emission is depicted in Figure6. In this experiment, we changed temperature of the sample from 300K to 320K for first about 170 seconds. Then, it was back to 300K over a period of about 1460 seconds. And then, we change the temperature to 300K over 150 seconds. Together with the temperature up and down, we have observed increase or decrease in the average emission intensity. We think that the cause is the change in recombination rate because the carrier density changed by thermal change. Also, in the beginning of this experiments, the sample surface emission was homogeneous with a little number of bright points. But, after 170sec the temperature was changed to 320K from the beginning of the experiment, then the bright spots had been increased significantly. And after this, even if we change again the temperature, there was little change in the emission intensity of the bright points. Thus, if we increased the bright spots once during PL measurement, it is difficult to vary the number of bright spots by the temperature change in the range of 300 ~ 320K.

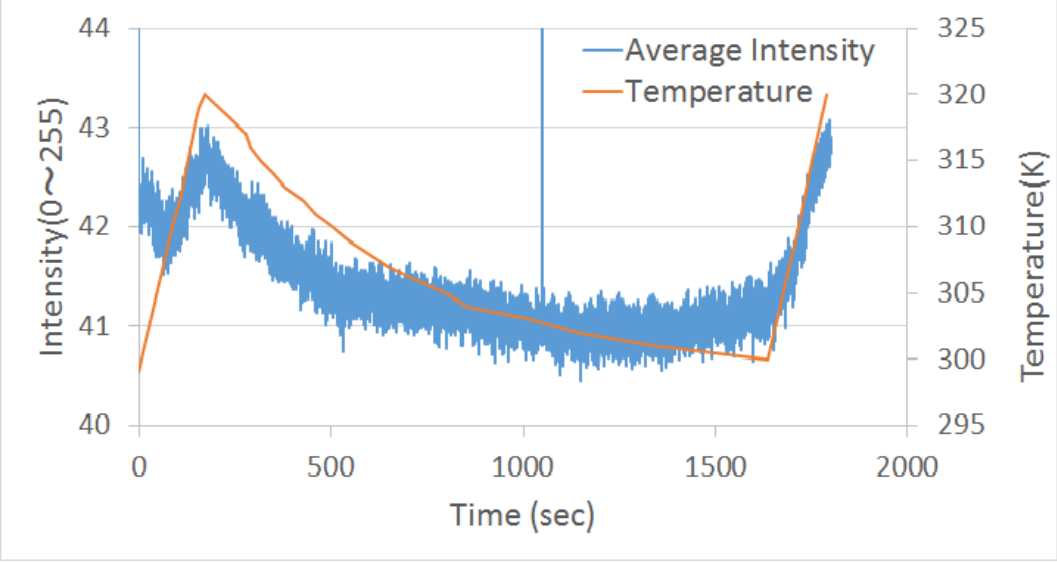

Figure5. Time variation of average emission intensity of InGaN SQW in TRPL by temperature change between 300K and 320K.By changing intentionally the temperature of the sample, the average emission intensity of the sample also changes.

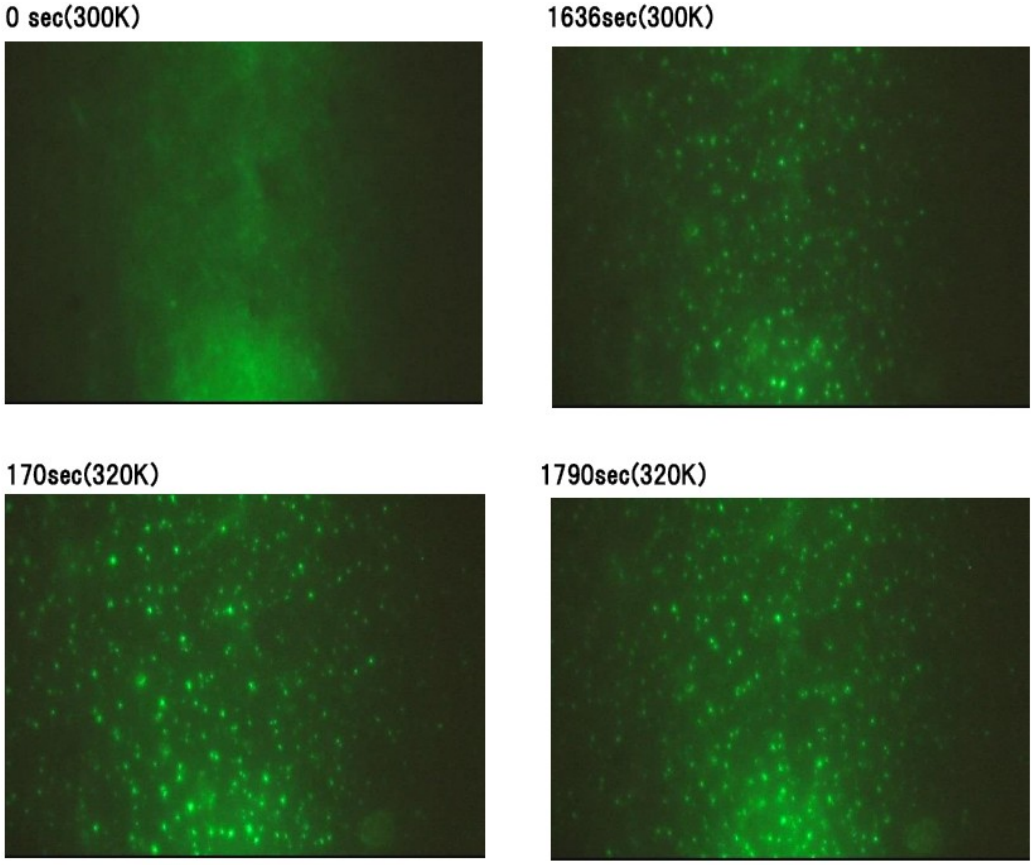

Figure6. Surface emission change of InGaN SQW by temperature change in TRPL. In the beginning of the experiment, surface emission was homogeneous, but bright spot is increased by raising the temperature to 320K. Then, the state of the bright point is changed slightly by a change in temperature. (URL2 : https://www.dropbox.com/s/x8v9zq61jxefnf7/ThermDep.avi?dl=0)

3.3 Pressure dependence of surface emission

In this experiment, we observed the surface emission of InGaN SQW in TRPL while changing the pressure between high vacuum (1.0×10^{-4}mBar) and atmospheric pressure (1.0×10^{3}mBar). The time variation of pressure and average intensity of surface emission is shown in Figure7, and the picture of time variation of sample surface emission is in Figure8. We had repeated air suction and release in this eexperiment three times. Immediately after the start of TRPL in the vacuum suction for the first time, we evacuated by a high vacuum (1.0×10^{-3}mBar) pump from atmospheric pressure (1.0×10^{3}mBar) over 500seconds and we observed that the bright spots were distinguished and sample surface emission was homogeneous. Then by returning to atmospheric pressure, average emission intensity of the sample surface change from about 70 to about 50. It is a decrease of intensity of about 32%. Also, we looked many blinking points while we have changed in pressure (URL3). Next, we had irradiated light under the atmospheric pressure for 1690seconds. And we observed increase of intense emission confined, local domains. After the number of bright points is increased, we evacuated again a second time. It took about 3000 seconds to obtain a homogeneous emission. It is quite a lot of time compared to the vacuum suction for the first time. Then we returned to atmospheric pressure, average emission intensity is decrease again about 27%, and we observed instability blinking while we release the vacuum. In the vacuum suction third time, we change in pressure to 1.0×10^{-4}mBar before the bright points appeared again around atmospheric pressure. And we observed increase bright spots while we evacuated to high vacuum. Then, as it got closer to 1.0×10^{-4}mBar, the surface emission was uniform again. And then, by vacuum release the average intensity of the sample surface is decreased by about 27%.

In conclusion we summarize the features of the sample surface emission in this experiment. First, we evacuate to high vacuum and the sample surface emission becomes of high intensity and relatively homogeneous. Then we release the vacuum, the emission intensity is greatly decreased. And we evacuated again, the emission goes to high intensity again and homogeneous. Also, while we change the pressure, many bright spots become instable and the blinking appears (URL3). When we change the pressure, it is considered that the InGaN SQW crystal structure is expanded and contracted. From the result of these experiments, we think that the change in the pressure causes the crystal expansion and contraction, and this causes change in crystal strain and piezo electricfield, and this provoke a change in optical emission. Also, under the vacuum suction, it is necessary to consider the influence of surface-adsorbed material.

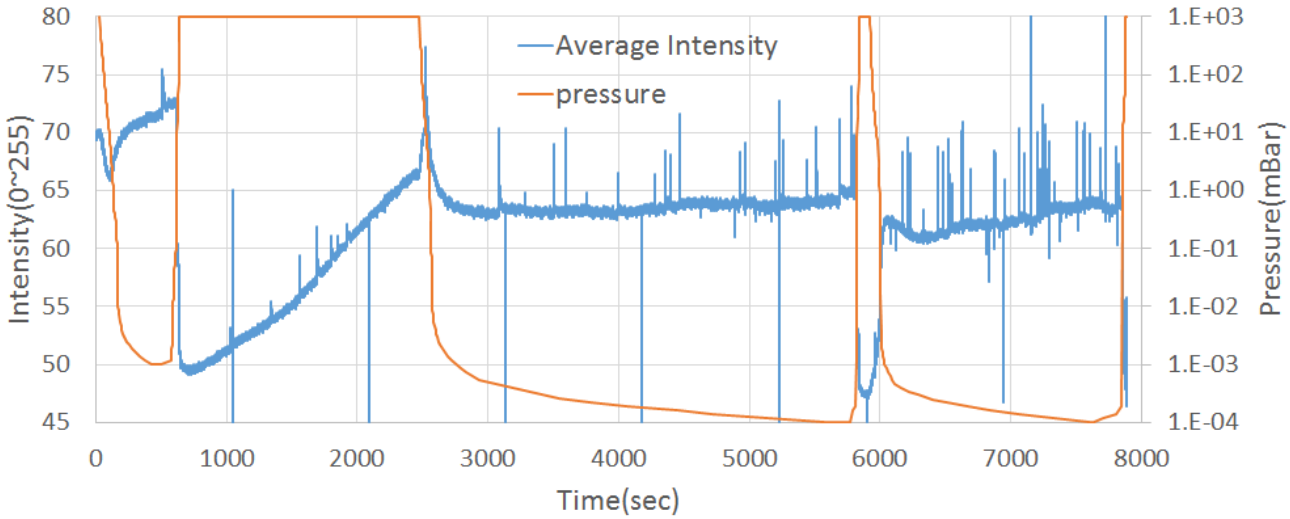

Figure7. In time-resolved PL measurements of InGaN, the time variation of the average emission intensity due to the pressure change (1.0×10^{-4}~1.0×10^{3}mBar). When you return to the atmospheric pressure from high vacuum sample, can be seen a decrease in extreme average emission intensity.

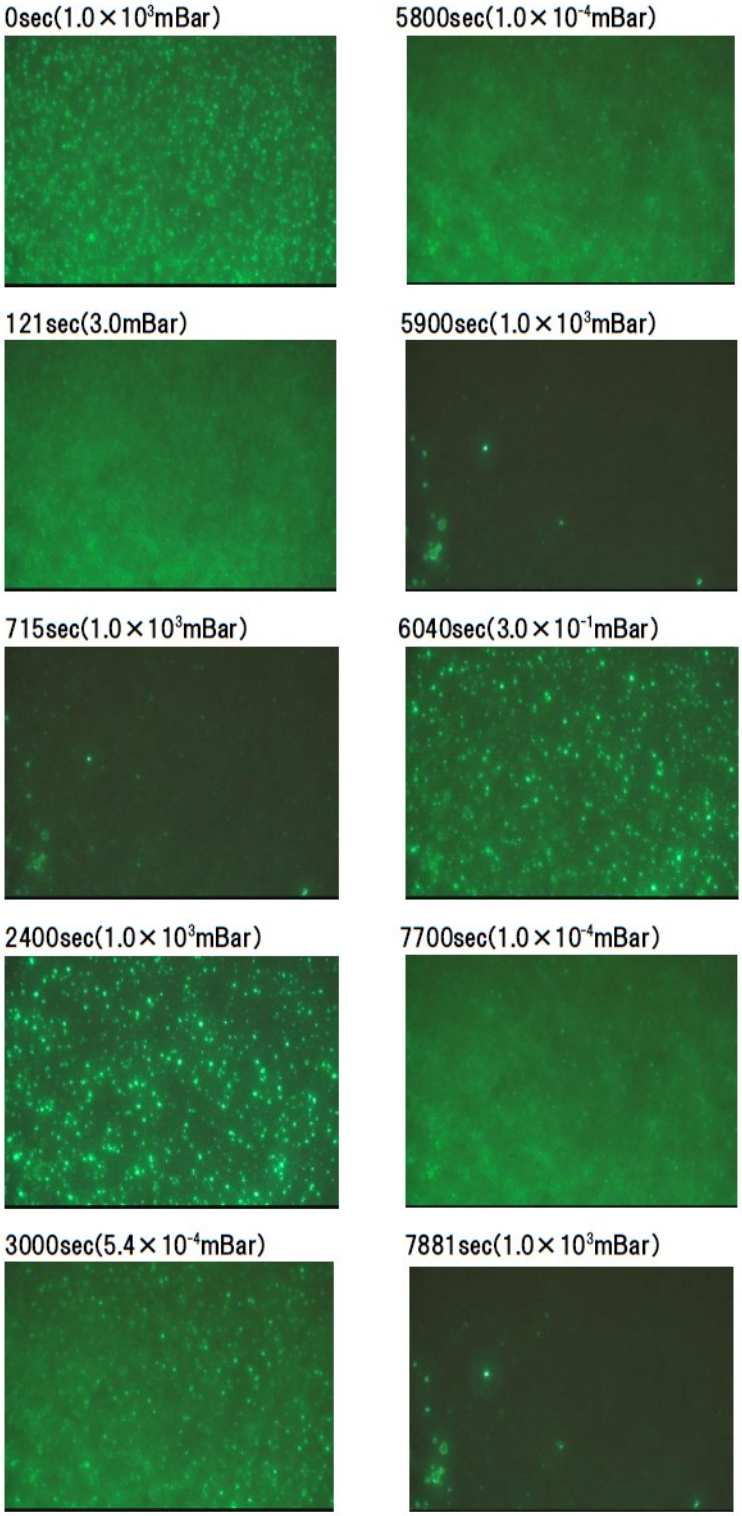

Figure8. Sample surface emission of InGaN SQW change the dynamics by pressure change.
(URL3 : https://www.dropbox.com/s/83fj9wqb2ps4j5l/pressreDep.avi?dl=0)

4. CONCLUSIONS

In this study, we find optical memory effect correlated to the increase of intense emission local domains in InGaN SQW by time-resolved photoluminescence and in order to investigate the cause, we researched the thermal and pressure dependence of photoluminescence. Through both change in temperature and pressure, we observed surface emission change of InGaN SQW samples. When we change the temperature around the sample, average PL intensity is changed, and the status and number of bright points is not or little changed. Also, when we change the pressure, the number of bright points and average emission intensity were suddenly and greatly varied. We think that increases of bright points by long time TRPL is physically related to the change in pressure. And it is considered that the temperature and pressure changes promote crystal compression and expansion. For these reasons, we think that crystal strain change greatly affect InGaN SQW surface emission. Also, in the experiment of pressure dependence, because it was considered that desorption of adsorbed material on the surface occur at the same time as the pressure change, it is necessary to examine also the influence of surface gases that are trapped or absorbed by superficial forces.

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