

# Photoluminescence study of blue light LED epitaxial wafer during growth by MOCVD

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In this work, an independent design photoluminescence (PL) measurement tool was used to monitor the PL spectra evolution during epitaxial growth of blue-green quantum well structures on silicon substrate by metal organic chemical vapor deposition (MOCVD). A split optical fiber and a notch OD 6 filter were employed to minimize the incoming laser beam (405nm) influence on the spectra. Using this method, the PL characterization of the layer between steps of the growth at several points were obtained by decreasing the reactor temperature. The effects of the temperature, the number of quantum wells and the p-type GaN layer on the photoluminescence are reported. We found that the presence of the p-type GaN layer significantly decreased the intensity of multiple quantum wells PL.

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## 1. Introduction

In the recent years, III-V group nitrides have been attracting great attention due to their wide applications in optoelectronic and power devices [1-3]. Photoluminescence (PL) is indispensable in semiconductor photoelectric materials characterization [4], and the researches on GaN epitaxial wafer by PL have been reported widely. Typically, PL measurement is performed after the epitaxial growth [5-6], however, as the epitaxial structure gets more and more complex, it becomes difficult to characterize individual layers of the structure by conventional ex during growth. The PL monitoring during epitaxial growth could be achieved by decreasing the temperature of metal organic chemical vapor deposition (MOCVD) reactor to a proper value. Obtaining PL spectra during growth, the property of each epitaxial layer can be characterized. As the temperature of reactor exceeds 900°C during the growth procedure [7], the weak PL of the wafer might get concealed in the strong thermal radiation [8-9]. The severe lattice vibration and susceptor rotation over 100rpm bring extra difficulty to PL measurement. Moreover, the in situ PL measurements are further challenged by many inevitable technical issues such as the introduction of excitation source, the PL signal collection, and the optical detector contamination during epitaxial growth. There are many reports about in situ PL monitoring in a molecular beam epitaxy (MBE) [10-11]. However, studies show that, at present, there are fewer researches about PL of Light emitting diode (LED) epitaxial wafer in the process growth by MOCVD.

In this paper, we report on the result of a PL measurement tool developed to investigate the PL spectra of InGaN/GaN Multiple Quantum Wells (MQW) blue light LED epitaxial wafer grown on silicon substrate in a Thomas Swan commercial reactor. The PL spectra of several layers of the structure were obtained by decreasing the reactor temperature. The effects of the temperature, the number of quantum wells and the p-type GaN layer on the PL are reported.

## 2. Experimental

In order to achieve PL spectra on the epitaxial wafer, a split optical fiber was fitted on the quartz optical temperature probe of a THOMAS SWAN CCS reactor. The quartz probe was fixed and the laser was focused onto the wafer's surface. The end of the probe was kept some distance (about 15mm) away from the wafer's surface and purged with the nitrogen gas to avoid the deposition of GaN. The probe top was shaped into a focusing lens in order to focus the excitation and collect the PL signal. One branch of the split fiber was connected to a semiconductor laser (405 nm, 200 mW) and the other was connected to an Ocean Optics USB2000+ spectrometer. The excitation laser scattered back was filtered with a 405nm notch filter (Edmund Optics, OD6 Blocking) from the PL signal. The experimental set-up is shown in Fig. 1.

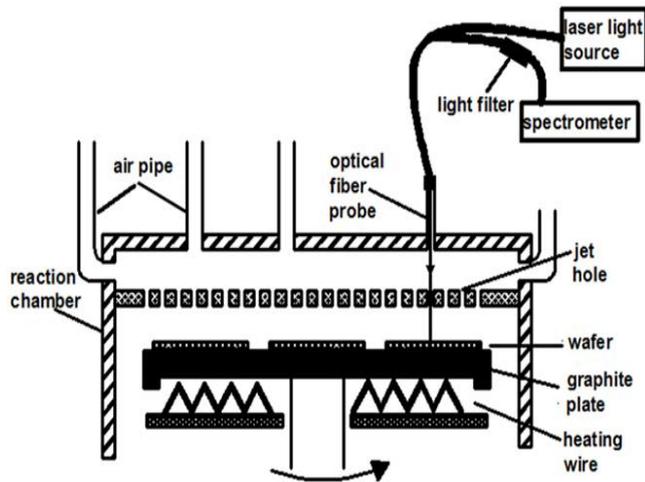


Fig. 1. Schematic layout of the experimental setup used for photoluminescence measurement

The epitaxial structure of the wafer is schematically shown in the inset of Fig. 2. When the MOCVD reactor's temperature reaches 1000 °C during epitaxial growth, the PL signal may be small compared to the thermal radiation, in order to alleviate the influence of thermal radiation, the temperature was decreased slowly (about 1°C/s) to approximately 290°C from the growth temperature after the growth of the n-type GaN layer. The temperature was also decreased to 180°C after the second, the fourth and the sixth quantum well growth were completed, respectively. When the epitaxial growth was finished, the temperature decreasing speed was deliberately slowed down to observe the PL continuously from the growth temperature to 60°C. Details of these processes are shown in Fig. 2. During each measurement, the laser was switched off momentarily to evaluate the influence of thermal radiation. While decreasing the temperature of the reactor, we let  $\text{NH}_3$  flow continuously. A GaN cap layer had been added on the quantum well before cooling down. We took these two measures to avoid the decomposition of the epilayer, hence our PL measurement should in principle not affect the normal growth process and the quality of the LED wafers. At each test point, the temperature was kept stable 5 minutes before the data was recorded.

In order to compare the PL measurement in the MOCVD reactor to ex situ PL measurement, the same wafer was placed on a electronic hot plate and measured at the same distance using of the new optical fiber probe. The temperature of the plate was increased from 60°C to 140°C at the interval of 20°C. The result was compared with the one obtained from the MOCVD reactor. Moreover, the PL properties of the wafers with the same GaN epitaxial structure in MQW and p-type on Si and sapphire substrates were also compared.

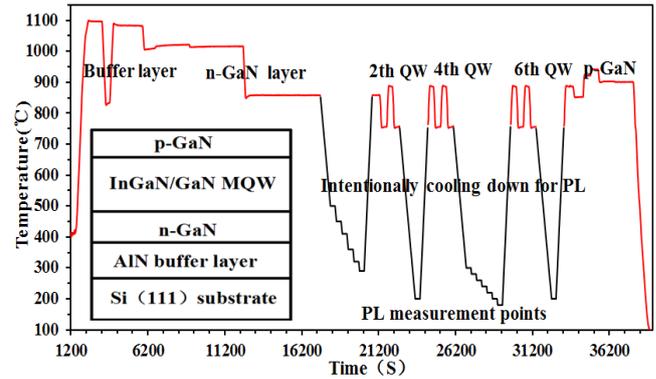


Fig. 2. The diagram of the temperature levels over time (The red lines are growth procedures, the black lines are intentionally cooling down and PL measurement points). The inset shows the structure of blue light LED epitaxial wafer

### 3. Results and discussion

The PL spectra of the epitaxial wafers on Si and on sapphire substrates, are shown in Fig. 3. The PL signal of the MQW or the yellow band on the sapphire wafer is much stronger than that on the silicon substrate, which means that it is very difficult to monitor the PL of the GaN layer grown on silicon. It should be pointed out that we could easily get more than 500 mW from this LED structure on silicon after applying a current of 350 mA, which demonstrates that our these wafers have high quality, even though their PL are so weak.

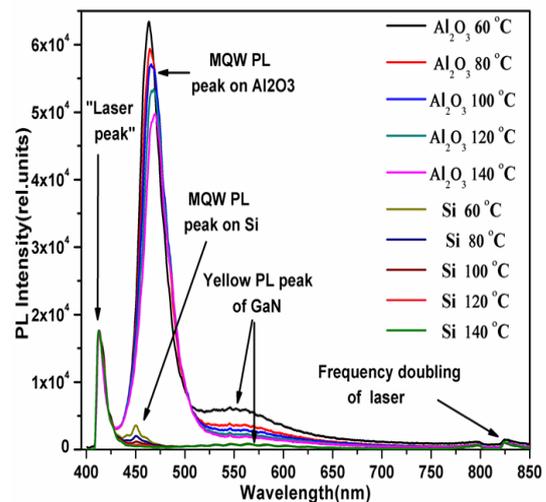


Fig. 3. Comparison of the PL spectra of blue LED epitaxial wafer sample grown by MOCVD on Si and sapphire at different temperatures by the electronic hot plate heating

The oscillations shown in Fig. 3 were induced by thin film interference, which could also affect the measurement. For the wafers grown both on silicon and sapphire substrates, the PL intensity gradually became weaker with the rise in temperature and the PL of the MQW on silicon substrate couldn't be measured by our monitor for temperatures above 140°C. At the end of the epitaxial growth, the PL results obtained from the MOCVD reactor were similar compared to ex situ results measured on an electronic hot plate using of the new optical fiber probe. This result shows that the optical fiber probe not be contaminated during epitaxial growth, the PL spectra could be accurately obtained from MOCVD reactor by the techniques described above.

After the growth of the n-GaN layer, PL spectra measured at several temperatures in the range between 500°C~290°C are shown in Fig. 4. A broad yellow band peak at about 560nm was observed [5]. This yellow band is a commonly observed impurity band in the PL spectrum of n-type GaN. Ga vacancy and its complexity with  $O_N$  donors have been claimed responsible for the yellow band, as well as carbon impurities [5]. In our experiment, intensity of the yellow band gradually increases with a rise in temperature as reported elsewhere [12]. The enhancement can be attributed to the increase in the transition via deep levels associated with impurities [13-15]. For comparison, the thermal radiation spectrum at 800°C and the background signal at 500°C are also shown in Fig. 4.

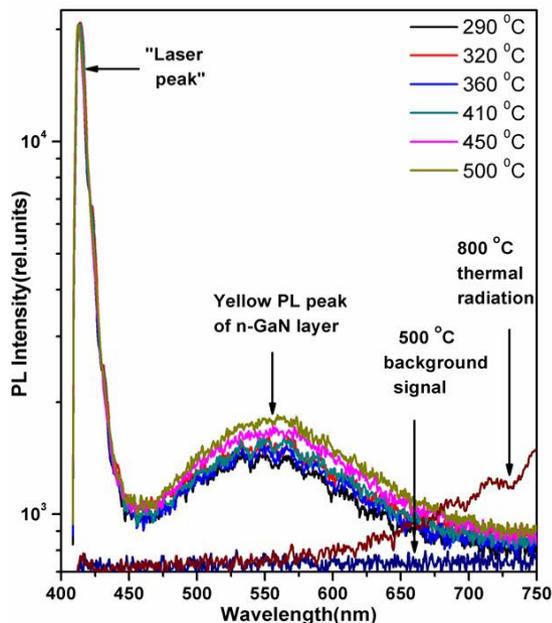


Fig. 4. Yellow band spectra of n-GaN layer grown by MOCVD on Si (111) in the temperature range from 500 °C to 290 °C, the thermal radiation spectrum at 800 °C and the background signal at 500 °C

Fig. 5 shows the temperature dependence of PL spectra from quantum wells before the growth of the p-GaN layer. The PL spectra could not be detected until

the temperature fell to 300°C after the fourth quantum well growth was established. The intensity of PL gradually increased and the full width at half maximum (FWHM) shrunk with the decrease in temperature, this could be explained by the decrease of non-radiative recombination via deep-level. Table 1 shows the FWHM varying with temperature by fitting the spectra data in Fig. 5. The inset in Fig. 5 clearly shows that the growth of p-type GaN layer greatly decreased the PL intensity of MQW and the PL intensity of MQW became stronger with the number of quantum wells.

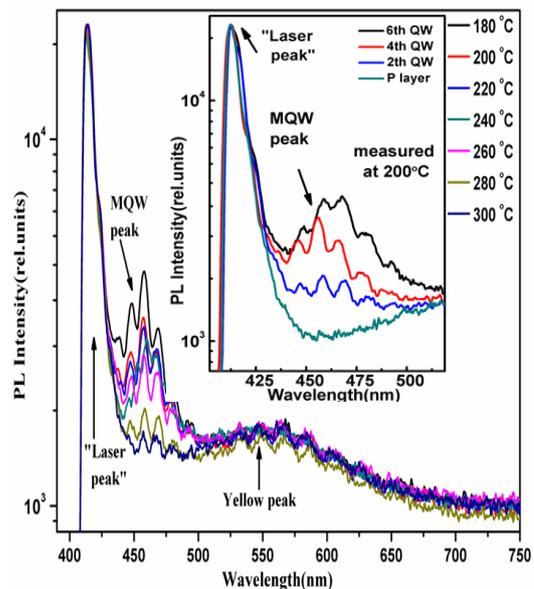


Fig. 5. The PL spectra of epitaxial wafer grown by MOCVD on Si (111) in the temperature range from 300 °C to 180 °C when fourth quantum well growth was established. The inset shows PL spectrum of second QW, fourth QW, sixth QW and P-type layer growth were completed

Table 1. The PL FWHM of the MQW varying with temperature (180~300 °C) when fourth quantum well growth was established on silicon

| t (°C)   | 180 | 200 | 220 | 240 | 260 | 280 | 300 |
|----------|-----|-----|-----|-----|-----|-----|-----|
| FWHM(nm) | 32  | 38  | 42  | 50  | 60  | 75  | 82  |

After the growth of p-type GaN layer, during the cooling procedure from 825°C to 60°C, PL spectra of epitaxial wafer were measured before taking the wafers out of the MOCVD reactor, as shown in Fig. 6. Compared with Fig. 5, the intensity of PL after p-type GaN layer growth was much weaker than the measured result after the four quantum wells were finished without p-layer. Moreover, the decrease of PL intensity with cooling was faster than for quantum wells without p-layer. The peak wavelength of yellow band was almost unaffected by temperature variations whether p-layer was present or not.

However, the intensity of the yellow band was weaker for measurements after the p-type GaN growth. (The yellow band intensity in Fig. 5 was about  $2 \times 10^3$ , but in Fig. 6 it was about  $1 \times 10^3$ ).

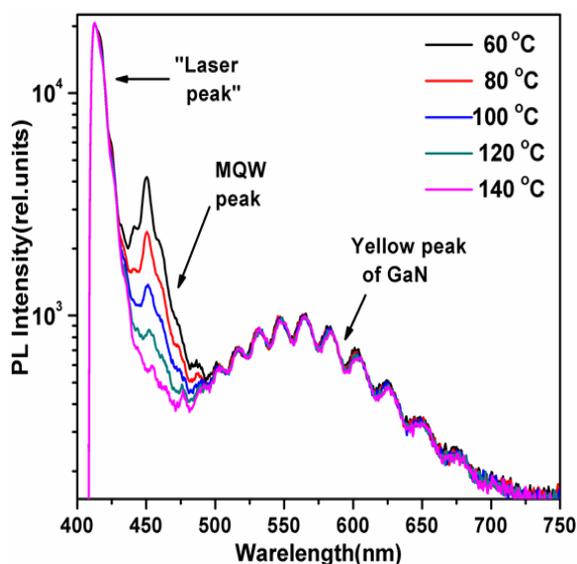


Fig. 6. The PL spectra of epitaxial wafer when p-type layer growth is established. in the temperature range from 140 °C to 60 °C

#### 4. Conclusions

The independent design PL measurement equipment was used to monitor PL spectra during growth of InGaN/GaN MQW LED epitaxial wafer on silicon (111) substrate by MOCVD. We found that the PL intensity of n-type GaN layer gradually increased with a rise of temperature in the range from 290 °C to 500 °C. The peak wavelength of the yellow band was almost unaffected by temperature variations whether p-layer was present or not. However, the presence of the p-type GaN layer significantly decreased the intensity of both the yellow band and MQW PL. Some findings in this paper may be helpful to understand the mechanism of yellow impurity band.

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#### References

- [1] Michael A. ReshchiKov, Hadis MorKoc, Journal of Applied Physics, **97**, 1301 (2005).
- [2] G. Devaraju, A. P. Pathak, N. Srinivasa Rao, V. Saikiran, Francesco Enrichi, Enrico Trave, Nuclear Instruments and Methods in Physic Research, **269**, 1925 (2011).
- [3] Li Shutu, Wu, Qibao, Guanghan Fan, Tianming Zhou, Yong Zhang, Yin Yian, Miao He, Jianxing Cao, Jun Su, Semiconductor Science and Technology, **24**, 5016 (2009).
- [4] Z. Chine, B. Piriou, M. Oueslati, T. Boufaden, B. El Jani, Journal of Luminescence, **82**, 81 (1999).
- [5] H. Y. Huang, C. H. Chuang, C. K. Shu, Y. C. Pan, W. H. Lee, W. K. Chen, W. H. Chen, M. C. Lee, Applied Physics Letters. **80**, 18, (2002).
- [6] C. Prall, M. Ruebesam, C. Weber, M. Reufer, D. Rueter. Journal of Crystal Growth, <http://dx.doi.org/10.1016/j.jcrysgro.2014.04.001>
- [7] G. P. Yablonskii, V. N. Pavlovskii, E. V. Lutsenko, V. Z. Zubialevich, A. L. Gurskii, H. Kalisch, A. Szymakowski, R. H. Jansen, A. Alam, B. Schineller, M. Heuken, Appl. Phys. Lett, **85**, 5158 (2004).
- [8] J. Creighton, D. Koleske, C. Mitchell, Journal of Crystal Growth, **287**, 572 (2006).
- [9] K. Mizuguchi, N. Hayafuji, S. Ochi, T. Murotani, K. Fujikawa, Journal of Crystal Growth **77**, 509 (1986).
- [10] C. J. Sandroff, F. S. TurcoSandroff, L. T. Florez, J. P. Harbison, Applied Physics Letters, **59**, 1215(1991).
- [11] Y. Wu, Y. C. Zhou, H. R. Wu, Y. Q. Zhan, J. Zhou, S. T. Zhang, J. M. Zhao, Z. J. Wang, X. M. Ding, X. Y. Hou, Applied Physics Letters **87**, 044104 (2005).
- [12] K. Subba Ramaiah, Y. K. Su, S.J. Chang, F.S. Juang, C.H. Chen, Journal of Crystal Growth, **220**, 405 (2000).
- [13] M Esmaeili, H Haratizadeh, B Monemar, P P Paskov, P O Holtz, P Bergman, M Iwaya, S Kamiyama, H Amano, I Akasaki, Nanotechnology **18**, 025401 (2007).
- [14] Bo Monemar, Plamen Paskov, Galia Pozina, Carl Hemmingsson, Peder Bergman, David Lindgren, Lars Samuelson, Xianfeng Ni, Hadis Morkoc, Tanya Paskova, Zhangxia Bi, Jonas Ohisson, Phys. Status Solidi A, **208**, 1532 (2011)
- [15] Wang Ming-yue, Yuan Jin-she, Yu Guo-hao, Chinese Journal of Luminescence, **30**, 73 (2009).

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