Optical Properties of GaN Epitaxial Layers Grown by Low-Pressure Metalorganic Vapor Phase Epitaxy Under Various Growth Conditions

Sho Shirakata[1], Hideto Miyake[2] and Kazumasa Hiramatsu[2]
[1]Faculty of Engineering, Ehime University, Matsuyama, Ehime 790-8577, Japan
[2]Faculty of Engineering, Mie University, Tsu, Mie 514-8507, Japan

Much effort has been paid to the growth of the high quality GaN epilayers by means of metalorganic vapor phase epitaxy (MOVPE) which is thought to be the most promising technique. Many optical characterizations have been reported for the high quality epilayers of the MOVPE-grown GaN. These include photoluminescence (PL), reflectance and photoreflectance (PR) spectroscopies. However, very few is reported for the optical properties of the GaN epilayer in connection with the systematically changing MOVPE growth conditions.[1)]

In this work, photoluminescence (PL) and photoreflectance (PR) measurements have been carried out on the series of hexagonal GaN epilayers grown on the sapphire substrate by means of low-pressure metalorgnic vapor phase epitiaxy (LP-MOVPE) under systematically changing growth parameters such as growth temperature (T_g: 900-1050°C) and pressure (P_g: 40-500 Torr).

GaN epilayers were grown on the c-plane of the sapphire substrate by means of LP-MOVPE with a horizontal reactor using H_2-trimethylgallium-NH_3 system. Growth parameters used are 900°C≦T_g≦1050°C and P_g=40, 80, 300 and 500 Torr.[2)] PL measurements were done at 8K under the photo-excitation of the 325 nm line of the He-Cd laser (10mW). PL was analyzed by the monochromator (focal length of 50cm) equipped with a grating of 1800 line/mm brazed at 300nm and detected using a photomultiplier (Hamamatsu R-636). For PR measurement, the pump light was 325 nm line of the He-Cd laser. The white light (W-lamp) was used as the probe light, and the reflected light was analyzed. PR measurements have been carried out at 77K.

The PL spectra at 8K of the typical high quality GaN epilayer grown on the sapphire substrate (T_g=1050°C, P_g=500Torr) is dominated by the sharp PL line of the exciton bound to the neutral donor, (D^0, X), at 3.488 eV and other PL peaks are weak. The free exciton line, FX_A, at 3.494 eV and the weak PL line due to the exciton bound to the neutral acceptor (A^0, X) at 3.482 eV are observed. Weak PL lines, at 3.404 and 3.311 eV, are one and two LO phonon replicas of FX_A, respectively.[3)] The PL line at 3.389 eV is due to the LO phonon replica of (A^0, X). The PL peak at 3.28 eV is the donor-acceptor (D-A) pair emission.[4)] The weak broad PL bands centered at 2.3 and 2.8 eV are also observed which are referred to as YL and BL bands,[5,6)] respectively.

The PR spectra at 77K exhibited two strong A and B and a weak C exciton transitions. Transition energies for the typical high quality epilayer (T_g=1050°C, P_g=500Torr) are E_A=3.492, E_B=3.502 and E_C=3.530 eV, and the energies indicate that the epilayers are compressively strained. PR broadening parameters are 3.7 and 3.0 meV for A and B transitions, respectively.

Figure 1 shows PL spectra of GaN epilayers grown at various T_g (900-1110°C) for P_g=80 Torr. At T_g=1000°C, intensity of (D^0, X) is the maximum and the full-width at half-maximum (FWHM) value is the smallest. For the 3.2-3.4 eV region, phonon replicas of excitons are strong for epilayers grown at 925≦T_g≦1000°C, while the D-A pair PL peak (3.28 eV) is pronounced for those grown at high T_g (T_g≧1025°C). This shows that the incorporation of both donor and acceptor impurities is remarkable at high T_g. The YL and BL bands are strong for epilayers grown at low T_g. Their intensity decreases monotonically as T_g increases, and they are almost absent for epilayers grown high T_g (T_g>1000°C). This indicates that the concentration of the native defect (Ga-vacancy: V_{Ga}) increases as T_g decreases because these PL bands are related to V_{Ga}.[5,6)] The broadening parameter of PR spectra tends to be minimum at T_g=1000°C, which is in good agreement with the corresponding tendency of the FWHM value of the bound exciton PL. These results show that for P_g=80 Torr, the quality of epilayers tends to be the best at T_g=1000°C. The transition energy E_A (PR, 77K) changes between 3.477 and 3.487 eV depending on T_g, and the value tends to be maximum at T_g=1000°C. This result shows that the magnitude of biaxial compressive strain increases with crystal quality.

On the other hand for high growth pressure (P_g=300 and 500 Torr), quality of the epilayer is improved monotonically as T_g increases. Figure 2 shows PL spectra of GaN epilayers grown at P_g=300 Torr. Intensity of

exciton emissions increases and FWHM value of (D^0, X) decreases monotonically as T_g increases. Intensities of YL and BL bands observed at low T_g as 950℃ decrease as T_g increases. For P_g=300 and 500 Torr, almost no D-A pair PL at 3.28 eV has been observed. The PR spectra are strongly dependent on T_g. PR spectra of the epilayers grown at low T_g as 950℃ are represented by the weak A transition peaks and the strong broad C transition peaks. This may be due to the degradation of the c-axis orientation of epilayers.

As for P_g, the general tendencies observed for 950≦T_g≦1050℃ is as follows: (i) intensity of the (D^0, X) line increases and FWHM value of the (D^0, X) line decreases as P_g increases, (ii) Intensity of the D-A pair PL (3.28 eV), BL and YL bands decreases as P_g increases. This result shows that the epilayer quality is improved with increasing P_g.

In this study, the best quality epilayer in view of the PL and PR property is grown at T_g=1050℃ and P_g=500 Torr, which are the highest T_g and P_g.

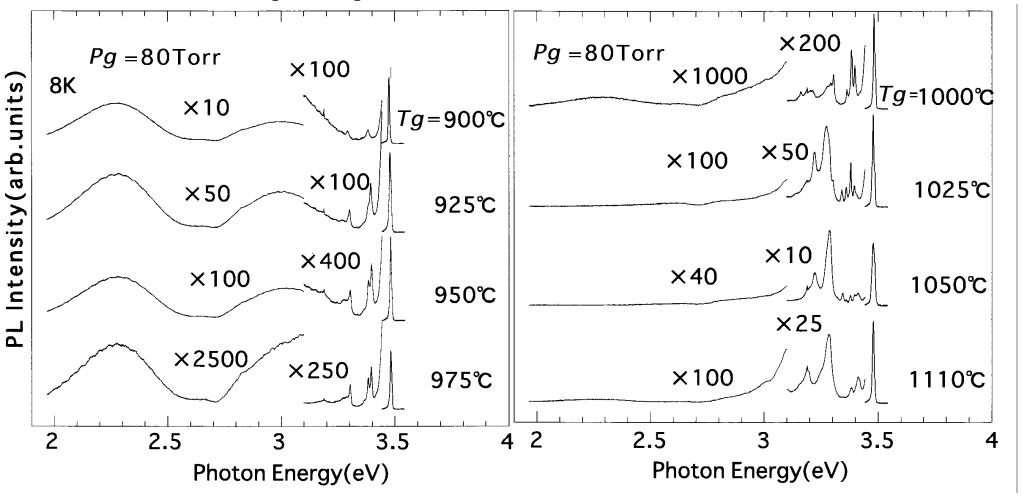

Fig. 1 PL spectra of GaN epilayes for P_g=80 Torr.

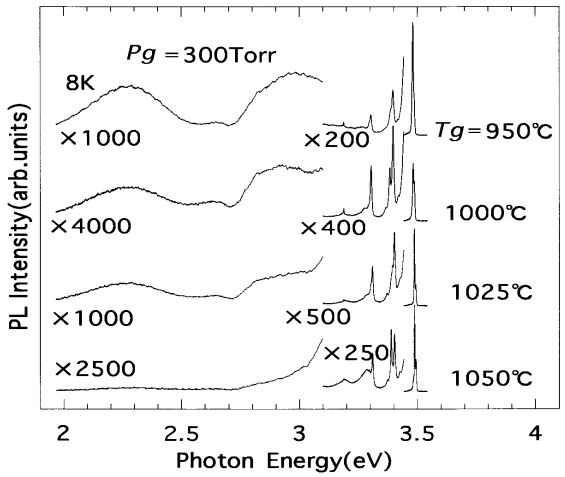

Fig. 2 PL spectra of GaN epilayes for P_g=300 Torr.

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