

Impact of GaN Buffer Growth Conditions on Photoluminescence and X-ray Diffraction Characteristics of MOVPE Grown Bulk GaN

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Abstract. Properties of metalorganic vapor phase epitaxy (MOVPE) grown GaN bulk layers with varying GaN buffer growth conditions are characterized by low-temperature (6K) photoluminescence (LT-PL) and X-ray diffraction (XRD). Full width at half-maximum (FWHM) of the near-bandedge emission of undoped layers is between 4.9 and 10meV, exhibiting no distinct dependence on buffer growth conditions. PL as well as photoreflectance measurements allowed the identification of neutral-donor bound exciton (D_0X) emission at $\sim 3.48\text{eV}$, and free A and B exciton emission lines at ~ 6 and $\sim 15\text{meV}$ higher energies, respectively. UV/yellow luminescence integrated intensity ratio and XRD FWHM show clear dependence on buffer growth conditions. Decreasing buffer thickness results in increasing PL intensity ratio and decreasing XRD FWHM. For thicker buffers, increasing the temperature ramping time between buffer and bulk growth also improves optical layer quality. Si-doped GaN was grown with carrier concentrations between $9 \times 10^{17}\text{cm}^{-3}$ and $2 \times 10^{19}\text{cm}^{-3}$. The PL peak position decreases with increasing carrier concentration and its FWHM increases due to donor banding effects.

1. Introduction

III-N compound semiconductors have attracted wide attention, largely due to the demonstration of blue light emitters [1], and their potential in high-temperature and high-power applications [2]. Despite the large differences in lattice constant and thermal expansion coefficients with regard to nitrides, sapphire has been the substrate of choice for device applications, made possible only through the introduction of low-temperature grown GaN or AlN buffer layers [3,4]. A wide variety of GaN buffer growth conditions, e.g. thickness, growth temperature, etc., have been reported to result in device quality MOVPE grown GaN layers on sapphire substrates (e.g. [1,2]). This is, however, in most cases growth system specific and only few reports are published on specific trends of buffer growth conditions on bulk layer quality [5-8]. This paper addresses such issues and reports on the impact of GaN buffer growth conditions on GaN bulk quality as characterized by LT-PL and single-crystal XRD.

2. Experimental

The GaN layers were grown by low-pressure MOVPE (60torr) on c-plane (0001) sapphire in a modified EMCORE GS3200 system. TMGa and NH_3 were used as precursors, Si_2H_6 for Si-doping, and H_2 as carrier gas. Bulk growth temperature was 1040°C , bulk growth rate was $2.5\mu\text{m/hr}$

(TMGa=136 μ mol/min) at V/III=1650, and bulk layer thickness was constant 1.25 μ m. The GaN buffers (TMGa=17 μ mol/min and V/III \approx 5000) were grown at temperatures between 500 and 550 $^{\circ}$ C, growth time was varied between 150 and 630sec, corresponding to buffer thicknesses of \sim 80-400 \AA , and the temperature ramping time between buffer and bulk growth was varied between 4 and 25min. The GaN layers were characterized by single crystal (θ -2 θ) XRD. LT-PL measurements have been performed at 6K using an Argon laser by COHERENT, combined with a monochromator to select the 334nm emission line from its spectrum. Output power was kept constant at 15mW, and the samples have been mounted on the cold fingers of a liquid helium cooled cryostat equipped with a temperature controller.

3. Results

All GaN layers exhibit smooth, shiny surfaces and are clear and transparent. XRD FWHM is typically \sim 250arcsec. During this study, background carrier concentration was in the mid 10^{17}cm^{-3} range with mobilities between 30 and 110 cm^2/V s corresponding to non-optimized buffer growth conditions.

6K PL FWHM of the bandedge emission of undoped layers is between 4.9 and 10meV, exhibiting no distinct dependence on buffer growth conditions. However, the UV/yellow luminescence integrated intensity ratio shows clear dependence on buffer growth conditions. Figure 1 shows representative UV bandedge PL spectra for samples grown with constant buffer growth temperature $T_b=500^{\circ}\text{C}$ and different buffer growth times. The main peak at 3.482eV is usually identified as the neutral-donor bound exciton peak (D_0X), blue-shifted by 13meV due to strain from the 3.469eV [9] measured on very thick (and presumably completely relaxed) GaN layers. These spectra clearly exhibit 3 additional peaks besides the main peak. The shoulder on the low energy side of the exciton peak at 3.468eV is attributed to the radiative recombination of an exciton bound to an acceptor [10]. The two peaks at the high energy side at 3.488 and 3.497eV, which are separated by 6 and 15meV from the D_0X peak, respectively, are in good agreement with previous reports and can be interpreted as free A and B exciton [9-15]. In samples where the exciton lines were not well resolved in the PL spectra, photoreflectance measurements also allowed the identification of free A and B exciton lines at 5 and 14meV above D_0X energy in excellent agreement with the PL results. Free C exciton was only weakly resolved and its energy is estimated to be 30-35meV above D_0X energy.

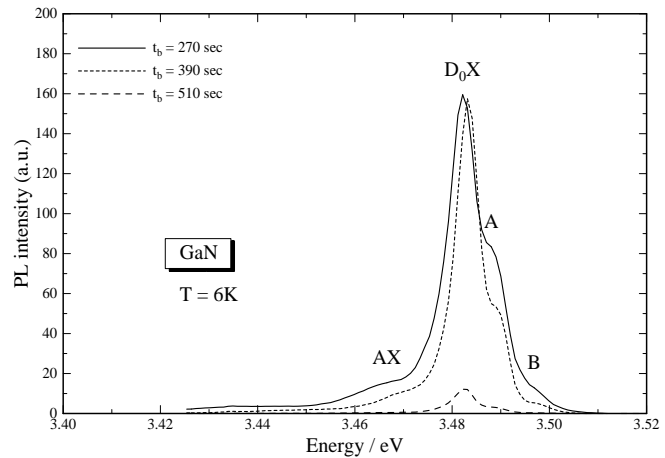


Fig. 1: PL spectra at 6K showing acceptor bound, neutral-donor bound and free A and B exciton peaks.

3.1 Impact of buffer growth time

Samples have been grown with different buffer growth times to determine the effect of buffer thickness. In Fig. 2, the UV/yellow luminescence integrated intensity ratio and the XRD FWHM are plotted as a function of buffer growth time with buffer growth temperature and ramping time

between buffer and bulk growth as parameters. (Intensity ratios between different series are not to scale.) This figure clearly indicates improved layer quality with decreasing buffer layer thickness. Except for very thin buffer and long ramping time (25min), the UV/yellow luminescence integrated intensity ratio increases and the XRD FWHM decreases as the buffer thickness is reduced. This dependence is in general independent of buffer growth temperature and temperature ramping time between buffer and bulk growth. However, for thin buffers, a shorter ramping time seems to be favorable (circles vs. squares in Fig. 2). This might indicate, that the optima of ramping time and buffer thickness depend on each other. For a thicker buffer layer, the optical layer quality improves clearly with longer ramping times as can be seen in Fig. 3.

3.2 PL measurements of highly Si-doped samples

Si-doped GaN has been grown with carrier concentrations between $9 \times 10^{17} \text{ cm}^{-3}$ and $2 \times 10^{19} \text{ cm}^{-3}$. Hall mobility for these layers is between $182 \text{ cm}^2/\text{Vs}$ at $n=10^{18} \text{ cm}^{-3}$ and $133 \text{ cm}^2/\text{Vs}$ at $n=2 \times 10^{19} \text{ cm}^{-3}$, respectively, Carrier concentrations as high as $n=5 \times 10^{19} \text{ cm}^{-3}$ have been achieved, however, surface morphology and mobility start degrading for carrier concentrations higher than $2 \times 10^{19} \text{ cm}^{-3}$. A recent publication for MBE-grown Si-doped GaN on GaAs reports drastic broadening of the PL linewidths up to 105 meV at 6 K for $n=10^{19} \text{ cm}^{-3}$ [15]. Figure 4 shows the shift of the main PL peak position and its FWHM as a function of the Si_2H_6 flow. In the semi-logarithmic plot,

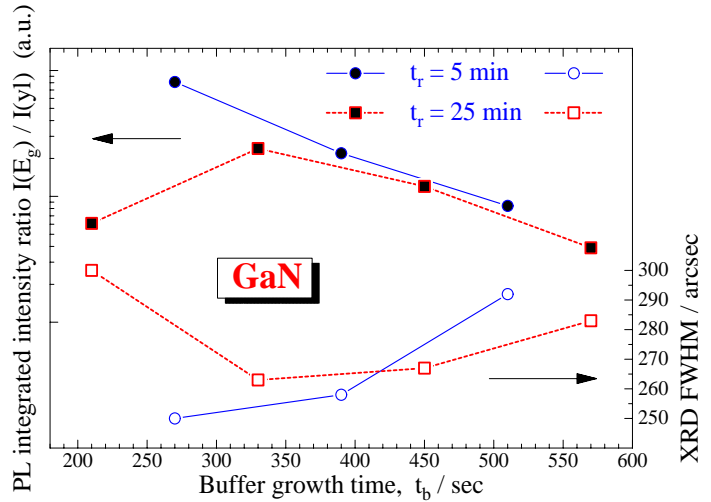


Fig. 2: UV/yellow luminescence integrated intensity ratio and XRD FWHM as a function of buffer growth time. (Intensity ratio between different series not to scale.)

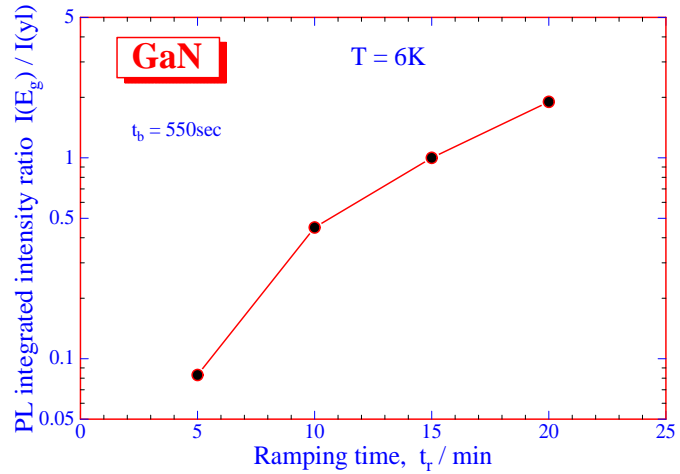


Fig. 3: UV/yellow luminescence integrated intensity ratio as a function of ramping time between buffer and bulk growth.

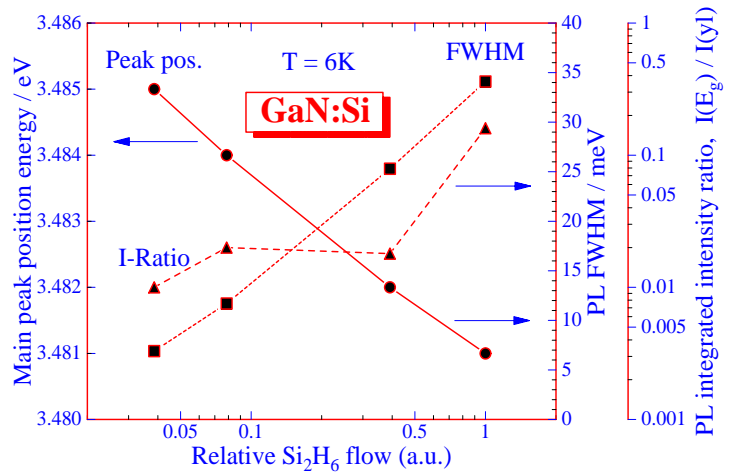


Fig. 4: Main PL peak position and its FWHM as a function of the Si_2H_6 flow.

the peak position decreases linearly with increasing Si flow, which could be attributed to a gradual shift from donor-bound exciton transitions to free hole-to-donor transitions [15]. The line broadening with increased doping level is in our case much less severe (34meV at $n=1.9\times 10^{19}\text{cm}^{-3}$) than reported in [15] and is obviously associated with donor banding effects that will begin to occur for $N_d>10^{18}\text{cm}^{-3}$ due to the Bohr radius of electrons in GaN of $\sim 23\text{\AA}$ [15].

4. Conclusions

Overall, we report on trends of PL and XRD of GaN bulk layers with buffer growth conditions. GaN characteristics are studied under various buffer growth conditions, e.g. thickness, growth temperature, and ramping time between buffer and bulk GaN growth. 6K PL FWHM of the bandedge photoluminescence is usually around 4.9-10meV, exhibiting no distinct dependence on buffer growth conditions. Neutral-donor bound exciton, free A and B exciton peaks could be identified in PL and photoreflectance measurements. Layer quality as estimated by XRD FWHM and UV/yellow luminescence integrated intensity ratio improves with thinner buffers and/or longer temperature ramping times between bulk and buffer growth. Si-doped GaN layers have been grown with carrier concentrations between $9\times 10^{17}\text{cm}^{-3}$ and $2\times 10^{19}\text{cm}^{-3}$. The peak position decreases linearly with increasing Si flow, which could be attributed to a gradual shift from donor-bound exciton transitions to free hole-to-donor transitions. The line broadening with increased doping level is obviously associated with donor banding effects.

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