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8th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 19–23, 2000

GaN/Al_2O_3 epilayers grown by MBE with a controllable nitrogen plasma composition

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Molecular beam epitaxy with N_2 plasma activation (PA MBE) is a promising technique to fabricate III-nitride nanostructures. The chemical activity of N_2 molecules in the PA MBE is increased due to excitation of molecular electron-vibration states as well as dissociation and ionization [1]. However, difficulties in the effective realization and control of these processes under ultra-low pressure conditions typical for MBE result in a low growth rate (much less than 1 μm/h) and imperfections in the epitaxial layers, induced by high-energy particles. To characterize material quality of GaN, photoluminescence (PL) measurements are widely used along with x-ray diffraction (XRD) studies. Particularly, the relative PL intensities of a useful band-edge line (BL) near 3.46–3.47 eV and a parasitic PL bands near 3.26–3.27 eV (DAP recombination line), as well as near 2.2 eV (YL line) are rather informative [2].

Nitrogen activators using both a radio frequency (RF) inductive discharge and that with electron cyclotron resonance excitation have been commonly employed in PA MBE [3]. In addition, we have proposed a new type of N_2 activator with an RF capacitively-coupled magnetron (RF-CCM) discharge [4, 5]. Using the RF-CCM activator with the discharge localized in the coaxial inter-electrode gap permitted us to obtain the GaN growth rate (v_g) up to 0.5 μm/h with reasonable quality. However, some growth rate lowering was observed during the exploitation of the RF-CCM operated in the "localized" mode at high RF-power ($\sim$200 W) and a short distance between the activator and a substrate ($\sim$4 cm).

In this paper, we demonstrate a new mode of the RF-CCM operation with the relatively intense plasma glowing extended into the growth chamber forwards the sample substrate. The "extended" mode shows good reproducibility and provides the GaN growth rate as high as 1.8 μm/h. Moreover, we demonstrate a possibility to control the GaN optical quality by a variation of the activated N_2 composition and a growth temperature.

GaN epitaxial films were grown on Al_2O_3(0001) substrates in a home made EP 1203 MBE setup equipped with the 13.56 MHz RF-CCM activator operating at a maximum power of 150 W and an axial magnetic field of 500 G in a coaxial design, as described in details elsewhere [5]. Unlike the "localized" mode employing a multi-hole output activator aperture, the single-hole one with a diameter of 3.5 mm was used for the "extended" mode. Typical distance between the activator aperture and the substrate was 7 cm. Nitrogen mass flow rate (Q_N) up to 30 sccm was controlled by measurements of N_2 background pressure in a growth chamber, which was lower than 1 mTorr. The MBE setup was equipped with one (in the most of the experiments) or two turbomolecular pumps with total effective pumping rates of 350 and 600 l/s, respectively. Thermal cleaning at 900°C, nitridation of the sapphire substrates at 850°C and growth of a GaN buffer layer ($\sim$20 nm) at 350°C were followed by the growth of a main layer ($\approx\mu$m) in a 500–700°C temperature range. All the processes were monitored in situ by reflection high energy electron diffraction.

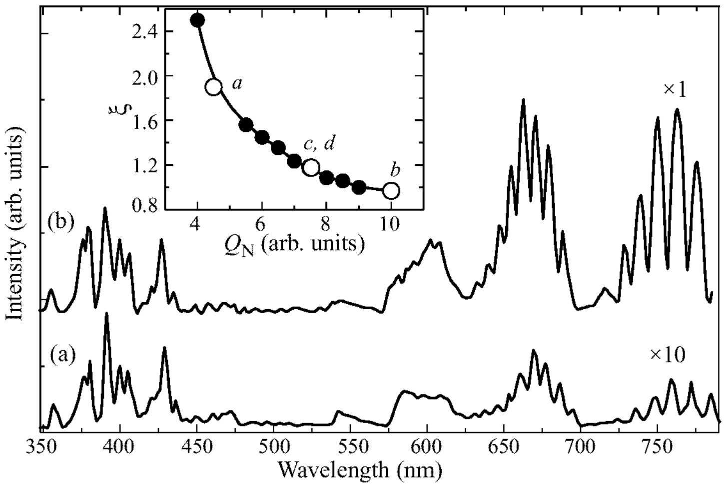

Fig. 1. Optical emission spectra (OES) of RF-CCM discharges in the "localized" (a) and "extended" (b) modes, operated at the same external parameters: RF-power of 150 W and N_2 pressure in the growth chamber of 3×10^{-4} Torr. The inset demonstrates dependence of OES intensity ratio of 391.4 and 380.4 nm lines (ξ) on N_2 mass-flow rate (Q_N) for the "extended" mode.

Optical emission spectra (OES) of the RF-CCM discharge were also recorded in situ. In the "localized" mode the emission from a discharge chamber reflected by a Si mirror plate mounted on a substrate holder was registered, while in the "extended" mode the direct measurements of the glowing plasma volume in the growth chamber were used. PL spectra of the GaN epilayers were obtained at 77 K with excitation by a 325 nm line of a 1 mW He-Cd laser. XRD measurements were perfomed using CuK_α radiation.

Preliminary testing of the RF-CCM has shown that the single-hole aperture enables one to excite the magnetron discharge in the "extended" mode with the OES intensity one order of magnitude higher than that in the "localized" mode (Fig. 1). Besides, higher intensities of the first and second positive series, related to excited N_2 molecules, as compared to the first negative one, related to N_2 ions, allow us to assume a lower electron temperature (E_e) in the "extended" mode [5]. Therefore, one may expect a higher content of metastable N_2 molecules and N-atoms impinging onto the substrate. In addition, a strong dependence of E_e on N_2 mass flow rate has been found. The inset in Fig. 1 shows, that the OES intensity ratio (ξ) of the ion to molecular lines at 391.4 and 380.4 nm, respectively, directly proportional to E_e, is raised towards low Q_N values. It should be noted that electrical probe measurements revealed charged particles in the activated beam in this mode in contrast to the "localized" one.

The main technological result of employing the "extended" mode of the RF-CCM is the greatly enhanced GaN growth rate, which can be regulated from 0.2 to 1.8 μm/h, depending on the Ga flux, the distance to a substrate and Q_N. In addition, this mode provides the possibility to control the material quality of GaN films by variation of activated nitrogen beam composition. To illustrate this, the set of growth runs was carried out at different Q_N and, hence, different E_e (shown by (a)–(d) points in the inset in Fig. 1). The experiments were done at moderate values of Q_N and Ga fluxes, when v_g (about 0.7 μm/h) was determined by Ga flux, i.e. the films were grown under the N-rich conditions.

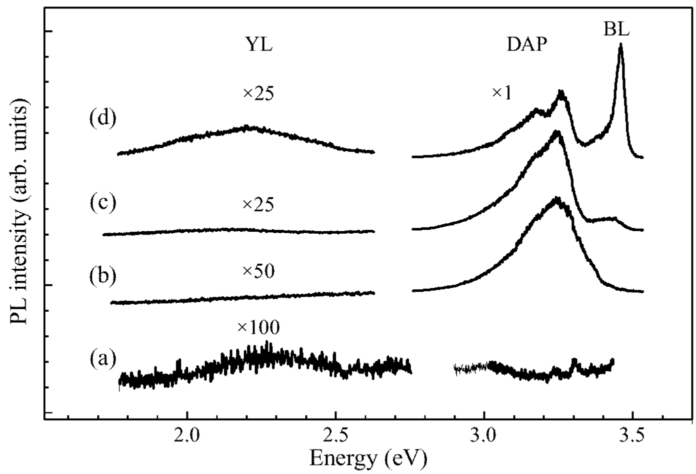

Fig. 2. Photoluminescence spectra (77 K) of GaN films grown with various composition of N_2 activated beam (see points a-d in the inset in Fig. 1): (a) $\xi = 1.9$; (b) $\xi = 1.0$; (c), (d) $\xi = 1.2$. Growth temperature: (a)–(c) 500°C, (d) 700°C.

A variation of the optical properties of the GaN epilayers is illustrated in Fig. 2. One may see that to obtain the intense band edge emission with relatively suppressed parasitic lines, the discharge must be kept at the optimal Q_N corresponding to $\xi = 1.2$ (Fig. 2(d)). Indeed, in the case of high E_e ($\xi > 1.2$, Fig. 2(a) both the content and energy of the ions in the activated beam are increased, resulting in a generation of non-radiative defect complexes [1]. Furthermore, high energy ions can be considered as a possible reason of the YL in the PL spectra, as was demonstrated in [6] and in our previous studies on the "localized" mode [7], where an absence of the YL was explained by a good separation of the high energy ions from the N_2 activated beam. Currently, the YL is considered as being related to Ga vacancies (V_{Ga}) [8], oxygen contamination and/or to point defects decorating dislocations [9, 10]. We believe that the ions serve as a trigger in the processes of the complex defect generation.

In the regime with low E_e corresponding to the inessential ion component ($\xi < 1.2$, Fig. 2(b)), the YL may be completely suppressed. However, this regime is not optimal due to the domination of the DAP band in the PL spectra. This band is attributed to a 225 meV acceptor in GaN, which has commonly been assumed to have an intrinsic origin related to the V_{Ga} [2]. Indeed, the high Q_N results in a non-stoichiometric growth with the enhanced Ga vacancies generation. Additionally, at this conditions a molecular regime of the gas flow is disturbed, resulting in three-dimensional growth.

The appearance of the BL corresponds to the medium E_e $\xi = 1.2$, Fig. 2(c). The increase in the substrate temperature from 500 to commonly used 700°C permits us to enhance the BL with the better ratio of the BL/DAP(YL) intensities (Fig. 2(d)). The BL line with relatively narrow FWHM of 36 meV dominates in this PL spectrum. The reasonable quality of the former film is confirmed by (0002)GaN XRD rocking curves with FWHM values of 994 and 67 arcsec in ω- and $\theta-2\theta$ scans, respectively. In conclusion, our experiments have shown that the discovered "extended" mode in the RF CCM can provide a high MBE GaN growth rate. The comparative analysis of the OES and PL spectra reveals the optimal composition of a N_2 activated beam, which permits us to compromise on the ratio between BL and parasitic lines.

Acknowledgements

This work has been supported in part by the RFBR grants and the Program of Ministry of Science of RF "Physics of solid-states nanostructures" (Grant 972014).

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