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Photoluminescence from freestanding GaN with $(10\bar{1}0)$ orientation

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ABSTRACT

Freestanding GaN templates with $(10\bar{1}0)$ orientation (M-plane) were obtained by halide vapor phase epitaxy (HVPE) on nearly lattice-matched $LiAlO_2$ and subsequent removal of the substrate by wet chemical etching. Photoluminescence (PL) spectrum from both sides of the GaN template investigated is dominated by peaks at 3.47, 3.42 and 3.36 eV, tentatively attributed to an exciton bound to the neutral shallow donor and two unidentified structural defects, respectively. The quantum efficiency of the exciton-related emission exceeds 10%, whereas that of the combined emission from the defect-related bands (red, yellow and blue) is below 0.1%. The evolution of the PL spectrum with temperature and excitation intensity is analyzed in detail. Effects of polishing and etching on the PL properties are also discussed.

INTRODUCTION

Large freestanding GaN wafers are valuable for homoepitaxial growth of advanced nitride-based devices. Growth of the M-plane GaN on nearly lattice-matched $LiAlO_2$ was originally proposed by Hellman et al. [1]. The advantage of the $(10\bar{1}0)$ oriented GaN substrate (M-plane) over the traditional (0001) orientation (C-plane) is due to significant reduction of the undesirable strain induced electric field when heterojunctions are produced [2]. A comprehensive study of thin M-plane GaN layers grown by molecular beam epitaxy (MBE) on $LiAlO_2$ revealed high concentration of stacking faults that apparently correlated with sharp PL line at 3.356 eV [3]. A significant number of structural defects, primarily stacking faults, were observed [4] also in the M-plane freestanding templates studied in this work. While the stacking faults in Ref. [3] were identified as being of the I_2 type, the experimental results of Ref. [4] indicated predominant I_1 type of stacking faults that have the lowest stacking fault energy. In this paper we report results of a PL study of freestanding GaN templates with the $(10\bar{1}0)$ orientation.

EXPERIMENTAL DETAILS

GaN films with 50 mm diameter, each about 300 microns thick, were grown on nearly lattice-matched $LiAlO_2$ substrates by HVPE. GaCl and NH_3 were used as precursors, and the growth was performed at 875 ^{o}C in a three-zone tube furnace. After removal of the $LiAlO_2$ substrate by wet chemical etching, the freestanding GaN wafers were found to have the non-polar $(10\bar{1}0)$ orientation. Front side of as-grown wafer looked smooth but with large-scale triangular-shaped features on it. The backside was featureless and rather smooth. The wafers were cut, the front side of some pieces was mechanically polished and cleaned in hot HCl. Selected samples were dipped in hot (160°C) H_3PO_4 for 1 minute to remove a surface layer.

PL experiments were carried out in the temperature range from 15 to 300 K using a closed cycle optical cryostat. The luminescence was excited with the 325 nm line of a He-Cd laser with excitation densities ranging from 3×10^{-5} to 300 W/cm^2. The luminescence signal was dispersed using a 0.5 m grating monochromator and detected with a Hamamatsu photomultiplier tube.

EXPERIMENTAL RESULTS

Figure 1 shows PL spectra from the front side of the as-grown M-plane GaN freestanding template at different temperatures.

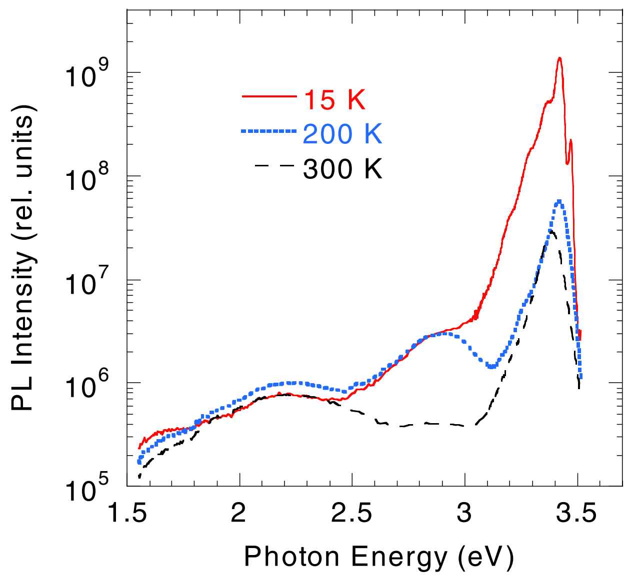

Figure 1. PL spectra from as-grown M-plane freestanding GaN template (front side) at different temperatures. PL was excited with unfocused 50 mW laser beam with diameter of 4 mm.

A relatively sharp peak [full width at half maximum (FWHM) varied from 12 to 25 meV in different samples] at about 3.47 eV is attributed to exciton bound to a neutral donor. The strongest band with the main peak at 3.42 eV followed by a few shoulders has not been clearly identified and will be analyzed in detail below. The PL spectrum contains also the blue luminescence (BL) band at 2.9 eV that is often seen in GaN grown by MOCVD [5] and HVPE [6] and apparently related to Zn contamination [6], although its attribution to a native defect [5] cannot be ruled out. The characteristic feature of the BL in undoped GaN quenches rapidly at temperatures above 200 K [5], consistent with our experiments. A very weak yellow luminescence (YL) band peaking at 2.2 eV and a shoulder below 1.8 eV corresponding to the red luminescence (RL) band can also be seen in these spectra. After polishing and cleaning in HCl, the intensity of the 3.47 and 3.42 eV peaks increased by about 3 times, whereas the defect-related broad bands (especially the BL band) quenched. Dipping in hot H_3PO_4 resulted in increase of PL intensity by about a factor 3 without changing the spectrum. The quantum efficiency of PL after polishing and cleaning or after etching in H_3PO_4 reached 15%.
Figure 2 shows the PL spectra from backside of the same sample (as-received). While the intensity of the BL increased, intensities of other PL bands decreased in the spectra from the backside. An additional PL band appeared at 3.29 eV. This band is attributed to transitions from the conduction band (or shallow donors) to an unidentified shallow acceptor. The characteristic LO phonon replica, as well as fast quenching of its intensity above 100 K support the above assignment.

The PL intensity and spectrum are very uniform across the 50 mm wafer when PL is excited with an unfocused laser beam (with a diameter of 4 mm). When the PL is excited with a focused laser beam (0.1 mm diameter), periodic nonuniformity could be detected. Figure 3

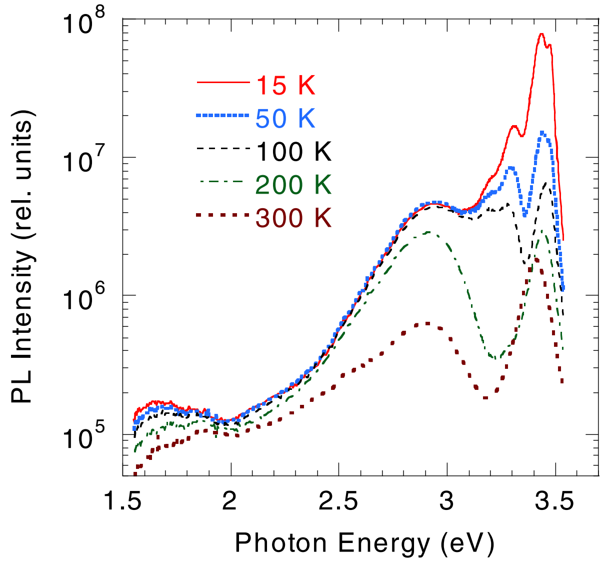

Figure 2. Same as in Fig. 1 for the backside of the sample.

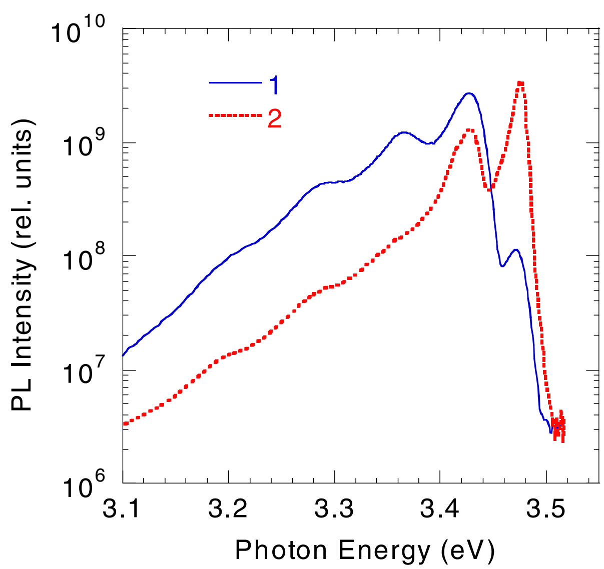

Figure 3. PL spectra from M-plane freestanding GaN template (front side) in two characteristic regions after etching in H_3PO_4. PL was excited with a focused 50 mW laser beam with 0.1 mm diameter.

shows the ultraviolet (UV) part of the PL spectrum at two regions where the spectrum shape and intensity differed to the greatest degree. On the majority of the surface area, the PL spectrum (curve 1 in Fig. 3) was nearly the same as that obtained with unfocused beam (such as in Fig. 1). In contrast, in selected small areas, the 3.47 eV peak increased substantially, whereas the 3.42 eV peak and especially the peaks at ~3.36 eV and ~3.29 eV decreased (curve 2 in Fig. 3). Comparison of the two spectra in Fig. 3 leads us to conclusion that at least the 3.47, 3.42, and 3.36 eV peaks are independent, and their varying contribution indicates nonuniform distribution of some defects in the sample.

With increasing excitation intensity from 3×10^{-5} to 300 W/cm^2 no shift of the 3.36 eV, 3.42 eV, and 3.47 eV peaks was detected to an accuracy of 2 meV. Intensity of these peaks increased linearly with the excitation intensity, whereas the BL, YL and RL bands began to saturate above 1 W/cm^2. Note however, that the energy positions of the peaks in the UV portion

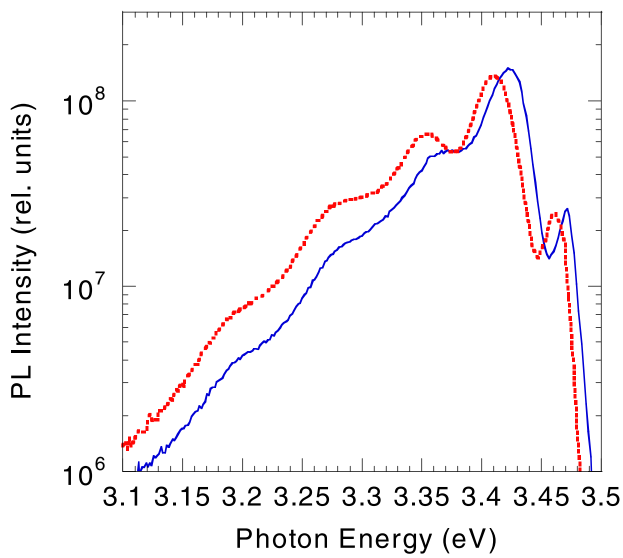

Figure 4. PL spectrum from as-grown M-plane freestanding GaN template (front side) in two points. PL was excited with focused beam.

of the spectrum varied across the wafer, and the maximal shift amounted to 14 meV (Fig. 4). The shift points to the existence of nonuniform strains in the sample and explains at least partially the broadness of PL peaks.

DISCUSSION

The M-plane freestanding GaN wafers are characterized with very high quantum efficiency of PL (more than 10%) and apparently very high contribution of bound excitons in radiative recombination (99%). Linear increase of the PL intensity and absence of peak shift in a wide range of applied excitation intensities imply excitonic origin of the peaks above 3.1 eV. Sharp and intense lines in the UV part of the PL spectrum of undoped GaN were often reported in the literature and attributed to excitons bound to structural defects [7]. Thus, the 3.36 eV and 3.42 eV peaks in the studied GaN wafers resemble, respectively, the Y_2 and Y_4 lines in undoped GaN grown on C-plane sapphire [7]. However, caution is in order with analogies because the properties of the 3.36 eV and 3.42 eV lines are apparently different from those of the Y_2 and Y_4 lines. Indeed, in contrast to the above-presented results, the Y_2 line slightly blue-shifted with increasing excitation intensity and always disappeared after a brief etch in hot H_3PO_4 [7]. What this means is that either the defects responsible for the Y_2 line are present only in the top defective layer of GaN layers grown by MBE [7] and uniformly distributed in depth of the M-plane freestanding GaN, or the 3.42 eV peak and the Y_2 line have different origin. The Y_4 line observed at 3.35 – 3.36 eV in PL spectrum of the MBE-layers is usually as narrow as the donor-bound-exciton line and is characterized with a very strong electron-phonon coupling (the Huang-Rhys factor is just 0.01) [7]. The 3.36 eV peak on M-plane GaN is always broad and apparently has much larger Huang-Rhys factor. Indeed, the peaks or shoulders at the low-energy side of the 3.36 eV peak seem to be its LO phonon replicas since their separation within the accuracy of ~10 meV is multiple of the LO phonon energy in GaN (91 meV), and the contribution of the replicas is nearly the same in all spectra. We doubt that the peaks/shoulders at ~3.28 and 3.19 eV are independent peaks, e.g. related to the zero-phonon line and its first LO phonon replica for transitions from the conduction band (or shallow donors) to a shallow acceptor. Indeed, these

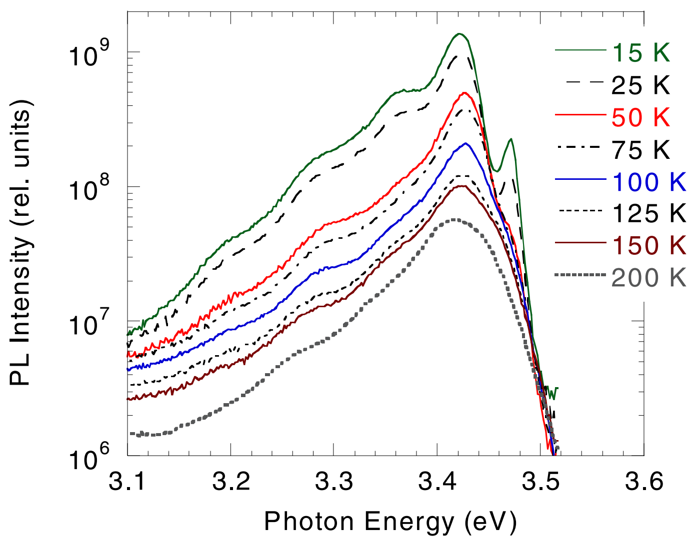

Figure 5. Temperature dependence of the PL spectrum from as-grown M-plane freestanding GaN template (front side, unfocused beam).

peaks quenched in a very similar fashion to the 3.36 eV peak (Fig. 5). The activation energy for the quenching of the 3.42 eV, 3.36 eV, 3.28 eV and 3.19 eV peaks was nearly the same (~30 meV) in the range of 50 – 200 K. The obtained activation energy and temperature range of quenching are drastically different from the values for the quenching of the shallow acceptor-related transition [8].

Compared to GaN layers grown on C-plane sapphire, either exhibiting *Y* lines or not, the studied M-plane freestanding GaN contained lower density of threading dislocations (10^8-10^9 cm^{-2} [4] versus ~10^9-10^{10} cm^{-2} [9]) and high concentration of the I_1 type stacking faults [4] (not observed in GaN grown on C-plane sapphire [9]). Therefore, predominance of the 3.36 and 3.42 eV lines in the PL spectrum of the M-plane GaN can be attributed to the stacking faults. Remarkably, Sun *et al*. [3] have also observed a strong PL line at about 3.36 eV in their M-plane GaN layers with high density of stacking faults. However the question which type of structural defects is responsible for which PL line and even whether just structural defects or some point defects trapped at the structural defects efficiently bind excitons in this material is still an open question.

CONCLUSION

We observed strong PL lines at 3.42 and 3.36 eV in M-plane freestanding GaN wafers. The intensity of these lines increased linearly with the excitation intensity, and no shift was observed in a wide range of excitation intensities. With increasing temperature, the PL intensity quenched with activation energy of about 30 meV in the range of 3.2 – 3.42 eV . The 3.42 and 3.36 eV peaks are attributed to excitons bound to unidentified structural defects.

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