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Optical and structural studies of homoepitaxially grown *m*-plane GaN

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Cathodoluminescence (CL) and transmission electron microscopy studies of homoepitaxially grown *m*-plane Mg-doped GaN layers are reported. Layers contain basal plane and prismatic stacking faults (SFs) with $\sim 10^6 \text{ cm}^{-1}$ density. Broad emission peaks commonly ascribed to SFs were found to be insignificant in these samples. A set of quite strong, sharp lines were detected in the same spectral region of 3.36–3.42 eV. The observed peaks are tentatively explained as excitons bound to some impurity defects, which can also be related to SFs. Donor-acceptor pair (DAP) recombination involving Si or O and Mg was ruled out by fitting DAP energies and CL mapping. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4706258>]

At present, most GaN optoelectronic devices are based on *c*-axis oriented (polar) structures, which are strongly influenced by piezoelectric and spontaneous polarization effects due to the lack of inversion symmetry in the wurtzite structure. These effects result in very strong polarization-induced electric fields that lead to spatial separation of charge carriers in quantum well (QW) structures, and thus increased radiative lifetime, lower internal quantum efficiency of light emitters as well as a dependence of the light emission intensity, and wavelength on the injection level. To avoid this problem, much research has been concentrated lately on the growth of non-polar (i.e., *a*- and *m*-planes) GaN structures.^{1,2}

Epilayers of *m*-plane GaN have been grown by metal-organic vapor phase epitaxy (MOVPE) and molecular beam epitaxy methods on γ -LiAlO₂,³ *m*-plane SiC,⁴ *r*-plane sapphire, and native *m*-plane GaN substrates.⁵ Light-emitting diodes based on *m*-plane GaN with promising device performance have been reported as well.^{6,7}

However, all heteroepitaxial non-polar structures suffer from high stacking fault (SF) densities of up to 10^5 – 10^6 cm^{-1} (here, the density is defined as the stacking fault length per unit area). SFs in GaN layers are defects with levels in the bandgap and are known to give characteristic cathodoluminescence (CL) emissions in the region of 3.29–3.41 eV.⁸ These SF-related peaks are present in undoped (i.e., unintentionally *n*-type doped) *a*- and *m*-planes GaN grown on sapphire.^{8–11} Recently, we also observed SFs in *c*-plane homoepitaxial GaN doped with Mg, which is the only viable *p*-type dopant for this material, thus showing that Mg doping can facilitate SF creation in polar GaN. Such samples have demonstrated metastability of the acceptor bound excitons (ABEs)^{12,13} and SF-related luminescence at low temperatures.^{14,15} The relationship between Mg-doping, SF formation, and optical properties in *m*-plane GaN is still poorly understood, partly due to lack, until recently, of homoepitaxial non-polar GaN layers. In this letter, we report on optical and structural properties of high-quality Mg-doped GaN layers grown by MOVPE on *m*-plane GaN substrates.

GaN layers of thickness 400 nm doped with Mg and with concentrations between 2×10^{18} and $3 \times 10^{19} \text{ cm}^{-3}$ were grown by MOVPE, starting with an undoped 0.6 μm

GaN buffer layer on *m*-plane GaN substrates (for details, see Ref. 16). The freestanding GaN substrates with threading dislocation density of $\sim 5 \times 10^6 \text{ cm}^{-2}$ were provided by Kyma Technologies. Mg concentrations were determined by secondary ion mass spectrometry (SIMS) at Evans Analytical Group. Samples were studied before and after annealing at 800 °C in N₂ atmosphere. Cross-sectional TEM analysis was done with a high resolution FEI Tecnai G2 200 keV FEG instrument. CL spectra were measured using a MonoCL4 system integrated with a LEO 1550 Gemini scanning electron microscope (SEM) and equipped with a liquid-He-cooled stage for low-temperature experiments. The typical acceleration voltage for this study was 15 kV. Either a fast CCD detection system or a Peltier cooled photomultiplier tube (PMT) was used for spectral acquisition.

Cross-sectional TEM micrographs shown in Fig. 1 reveal that the studied *m*-plane GaN layers have a rather high density of small basal plane SFs (BSFs) with characteristic length of $\sim 10 \text{ nm}$, which is similar to homoepitaxial *c*-plane GaN doped by Mg.¹⁵ Here, the density of BSFs is $\sim 10^6 \text{ cm}^{-1}$ and $\sim 3 \times 10^6 \text{ cm}^{-1}$ for the GaN layers with Mg concentrations of 1×10^{19} and $3 \times 10^{19} \text{ cm}^{-3}$, respectively. However, unlike *c*-plane GaN samples, a number of more extended defects including both prismatic SFs (PSFs) and BSFs were formed at the interface region between the GaN substrate and the buffer layer. This is shown in Fig. 1(b) with the loop formed by two long BSFs and several shorter PSFs. The near surface region of the same sample (inset in Fig. 1(b)) contains some smaller BSFs typical for GaN doped by Mg. Thus, we suggest that the formation of the BSFs in our *m*-plane GaN is generated by both Mg doping and by a residual strain relaxation during the homoepitaxial growth procedure.

Similar to the case of Mg-doped *c*-plane GaN, the near band-gap luminescence in our *m*-plane GaN:Mg layers was metastable under electron irradiation for moderate Mg concentrations, even though the effect was much weaker in this case. This is illustrated in Fig. 2 where the low-temperature CL spectra are shown for three annealed GaN layers with different Mg doping densities. The CL emission is dominated by a no-phonon donor-acceptor pair (DAP) recombination band at $\sim 3.26 \text{ eV}$ with its two LO phonon replicas. Two

FIG. 1. Cross-sectional TEM images of *m*-plane GaN samples doped by Mg with concentrations (a) $3 \times 10^{19} \text{ cm}^{-3}$ and (b) $1 \times 10^{19} \text{ cm}^{-3}$. The interface region between substrate and buffer is shown in (b), while the region close to the surface is shown as the inset. Thick arrows show SFs, and the crystallographic and surface $[1\bar{1}00]$ directions are indicated by thin arrows.

peaks at $\sim 3.46 \text{ eV}$ and $\sim 3.45 \text{ eV}$ denoted as ABE1 and ABE2, respectively, are acceptor bound excitons associated with the Mg-related acceptors A1 and A2.¹³ The most evident change in the spectral shape after a 30 min long electron irradiation (dashed lines) was observed for the samples with Mg concentration of about $1 \times 10^{19} \text{ cm}^{-3}$, where the intensity of both ABE1 and ABE2 decreases slightly with time while some sharp line features appear between 3.3 and 3.4 eV, i.e., in the region where SF-related luminescence is typically observed. We will consider this spectral region in more detail for samples where, surprisingly, we have observed several (at least 10) strong and sharp lines.

Fig. 3 shows the temporal evolution of CL spectra taken at 5 K for the (a) annealed and (b) as-grown *m*-plane GaN sample with an average Mg doping density of $\sim 1 \times 10^{19} \text{ cm}^{-3}$. The delay time after the start of the electron irradiation is indicated for each spectrum. We should mention that annealing has a marginal effect on the near-band gap luminescence and results in slightly stronger ABE-related peak intensities compared to the donor bound exciton (DBE) line. This observation may be explained by a specific growth process with a slow post-growth cooling of the samples for activating the Mg acceptors. CL spectra for the

FIG. 2. Low-temperature CL spectra of three annealed GaN layers with Mg concentration indicated at the left side. The electron beam acceleration voltage is 15 kV. Spectra taken directly after the start of electron irradiation are shown by solid lines, while spectra taken after irradiation time of 30 min are shown by the dashed lines.

annealed *c*-plane GaN sample doped with similar Mg concentration are shown by the dashed lines for comparison. It is obvious from Fig. 3(a) that both the spectral shape and the temporal behavior of the CL are significantly different for the *m*- and *c*-plane GaN:Mg layers, even for the annealed samples which are generally more stable. Three distinctive features are noted: (i) the stability of the ABE-related lines for *m*-plane GaN:Mg vs. their instability in the *c*-plane GaN:Mg samples, (ii) the presence of the broad lines (also metastable) at 3.31–3.42 eV in the CL spectrum for the *c*-plane GaN, denoted in Fig. 3 as S1–S3, and related to structural defects such as BSFs, PSFs, and partial dislocations, and (iii) CL for the *m*-plane GaN demonstrates a number of rather strong sharp lines at 3.36–3.42 eV with relative intensities comparable to the near bandgap bound exciton signal, which are less likely to be associated with SFs. One would expect broadened recombination lines related to SFs, since strain in the vicinity of each SF may differently affect its localization energy. Similar lines at $\sim 3.4 \text{ eV}$, though much weaker (by a factor of ~ 30), were observed in the photoluminescence (PL) for low-doped *c*-plane homoepitaxial GaN:Mg layers where the origin of these lines was interpreted as recombination of separated donor-acceptor pairs.¹⁷

We have estimated the possible energies of the separated DAPs as shown in Fig. 3(b). The DAP luminescence has a peak position determined by the donor and acceptor levels, i.e., $E_{DAP(\infty)} = E_g - E_D - E_A$, and this energy increases due to the Coulomb interaction between the ionized donor and the acceptor in the final state,

$$E_{DAP} = E_{DAP(\infty)} + \frac{e^2}{4\pi\epsilon_0\epsilon R_i}, \quad (1)$$

where R_i is the donor-acceptor distance, the band-gap energy for wurtzite GaN is $E_g = 3.5 \text{ eV}$, the energies for shallow donor and Mg acceptor levels are $E_D \sim 0.03 \text{ eV}$ and $E_A \sim 0.224 \text{ eV}$,

FIG. 3. CL spectra (solid line) at 5 K for annealed (a) and as-grown (b) *m*-plane GaN layer doped with Mg concentration of $\sim 1 \times 10^{19} \text{ cm}^{-3}$. For comparison, CL spectra for *c*-plane GaN layer doped with Mg concentration of $\sim 1 \times 10^{19} \text{ cm}^{-3}$ (see Ref. 12) taken at similar conditions are shown by dashed lines. DAP energies calculated for Ga-sublattice acceptors and for donor atoms in Ga- and N-sublattices are shown by short black and grey vertical lines, respectively.

respectively.^{18–20} The DAP energies, calculated for atoms located at some close positions in the wurtzite Ga sublattice, cover the energy range of the observed sharp features at $\sim 3.4 \text{ eV}$, but do not account for all the transitions. We also have calculated the DAP energies for atoms placed in Ga and N sublattices (shown in Fig. 3(b) by grey lines). Still the DAP energies cannot be assigned to all sharp lines at 3.4 eV . Therefore, the origin of these features is unlikely to be associated with DAP recombination involving the Mg_{Ga} acceptor.

The following supplementary experiment was performed to validate if the sharp features at 3.4 eV are related to the DAP or not. A small stripe of the sample surface was irradiated during longer time by the electron beam with an acceleration voltage of 15 kV , which locally activated additional Mg acceptors. Fig. 4 shows a SEM image over this region together with monochromatic CL micrographs taken

FIG. 4. (a) SEM image of the *m*-plane GaN sample with Mg concentration of $1 \times 10^{19} \text{ cm}^{-3}$ showing the surface region with a stripe extra treated by electron beam. MonoCL images taken over the same place as in (a) at few specific photon energies: (b) 3.46 eV corresponding to the ABE peak, (c) 3.39 eV corresponding the sharp-line features, and (d) 3.26 eV corresponding the DAP maximum.

at different photon energies: (b) at the ABE peak, i.e., 3.46 eV , (c) in the region of the features at $\sim 3.39 \text{ eV}$, and (d) at 3.26 eV , i.e., at the DAP maximum. Such activation of acceptors in the chosen region should result in enhanced intensity of all luminescence related to the acceptor states. Indeed, as can be seen from Fig. 4, the CL signal corresponding to the ABE or DAP emissions became much stronger in the treated region (bright contrast in CL images, Fig. 4(b) and 4(d)), while the CL signal related to the fine features at 3.4 eV practically vanished (dark contrast in Fig. 4(c)). Thus, we confirm that the sharp lines at about 3.4 eV are not likely to be associated with donor-acceptor pair recombination, i.e., with the conventional Mg-related DAP peaking at $\sim 3.26\text{--}3.27 \text{ eV}$. This is also consistent with our transient PL data where the PL decay time for these lines is of the same order as for bound excitons. The latter is only possible for the closest neighbored pairs. However, as mentioned above, transitions between the closest atoms would not describe the whole family of these 3.4 eV lines.

An alternative, albeit tentative, argument can be suggested as elaborated below. The sharp features at $\sim 3.4 \text{ eV}$ are unlikely to be due to the LO phonon replicas of the higher valence subband excitons, even considering the very strong interaction between excitons and LO phonons in GaN.²¹

Toropov *et al.*²² have studied optical phonon-assisted transitions of A and B excitons bound to impurities in bulk GaN. Although the lines observed in our experiments are in the similar spectral region of 3.36–3.42 eV, replicas of higher states are very weak, because the excitons mainly relax to the lowest DBE and ABE levels before they recombine. In our case, the relative intensity of the ~3.4 eV features is very strong compared to the no-phonon exciton lines. We have only observed these peaks in Mg-doped GaN. An analogy can be made with the case of p-type GaAs, where a series of sharp exciton-like lines were observed below the acceptor bound exciton lines.²³ The origin of the features was connected with axial defects resulting from acceptor complexes.²⁴

In conclusion, homoepitaxial *m*-plane GaN samples doped by Mg contain structural defects, specifically BSFs and PSFs. Despite a rather high BSF density of 10^5 – 10^7 cm⁻², the corresponding emissions in CL spectra were insignificant. Instead, a number of sharp lines at ~3.4 eV were observed. We have shown that the origin of these lines is unlikely to be associated with the conventional DAP recombination including Si or oxygen as donors and Mg as substitutional acceptor. However, the origin of these lines is still unclear. These features, by analogy with p-type GaAs case, may have excitonic character and be related to some acceptor defect centers, possibly also SFs.

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