

# Spatial distribution of deep level defects in crack-free AlGa<sub>0.75</sub>N grown on GaN with a high-temperature AlN interlayer

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The deep level luminescence of crack-free Al<sub>0.25</sub>Ga<sub>0.75</sub>N layers grown on a GaN template with a high-temperature grown AlN interlayer has been studied using spatially resolved cathodoluminescence (CL) spectroscopy. The CL spectra of Al<sub>0.25</sub>Ga<sub>0.75</sub>N grown on a thin AlN interlayer present a deep level aquamarine luminescence (DLAL) band at about 2.6 eV and a deep level violet luminescence (DLVL) band at about 3.17 eV. Cross-section line scan CL measurements on a cleaved sample edge clearly reveal different distributions of DLAL-related and DLVL-related defects in AlGa<sub>0.75</sub>N along the growth direction. The DLAL band of AlGa<sub>0.75</sub>N is attributed to evolve from the yellow luminescence band of GaN, and therefore has an analogous origin of a radiative transition between a shallow donor and a deep acceptor. The DLVL band is correlated with defects distributed near the GaN/AlN/AlGa<sub>0.75</sub>N interfaces. Additionally, the lateral distribution of the intensity of the DLAL band shows a domainlike feature which is accompanied by a lateral phase separation of Al composition. Such a distribution of deep level defects is probably caused by the strain field within the domains. © 2006 American Institute of Physics. [DOI: 10.1063/1.2402964]

## I. INTRODUCTION

AlGa<sub>0.75</sub>N alloy with a high AlN mole fraction is of great interest for applications in ultraviolet light-emitting diodes<sup>1,2</sup> and solar-blind photodetectors.<sup>3,4</sup> A good knowledge of the deep level defects existing in materials and their influences on optical properties is of great significance for optimizing materials and improving the performance of optoelectronic devices. A comprehensive review of the defect-associated luminescence in GaN is now available.<sup>5</sup> Compared to the considerable efforts spent on studying the famous yellow luminescence (YL) band of GaN, much less is known about the deep level luminescence of AlGa<sub>0.75</sub>N. The crystalline quality of AlGa<sub>0.75</sub>N directly deposited on sapphire substrate degrades rapidly with increasing AlN mole fraction.<sup>6</sup> Although the quality may be greatly improved by growing AlGa<sub>0.75</sub>N on a GaN template, the tensile stress in the epilayer due to the large lattice mismatch, which is up to -2.4% for AlN/GaN at room temperature, may lead to crack formation during the deposition process,<sup>7</sup> which tends to severely deteriorate material properties<sup>8</sup> and device performance.<sup>9</sup> An insertion of a high-temperature (HT) grown AlN interlayer (IL) between the AlGa<sub>0.75</sub>N overlayer and the GaN underlayer can achieve a crack-free AlGa<sub>0.75</sub>N surface.<sup>10,11</sup> In this work, spatially resolved cathodoluminescence (CL) measurements were used to investigate the deep level emissions (DLEs) from the crack-free Al<sub>0.25</sub>Ga<sub>0.75</sub>N layers grown on a GaN template with a HT-AlN IL. This detailed study of the spatial distribution of the deep level defects in AlGa<sub>0.75</sub>N will be helpful in

understanding the microstructure of materials and their influences on the performance of optoelectronic devices.

## II. EXPERIMENT

A series of samples was prepared using low pressure metal organic chemical vapor deposition. Trimethylgallium, trimethylaluminum, and ammonia were used as precursors for Ga, Al, and N, respectively. H<sub>2</sub> serves as the carrier gas. Firstly, a 2.5 μm GaN template was grown on (0001) sapphire substrate using a low-temperature GaN nucleation layer. Then a HT-AlN IL was deposited on GaN at 1040 °C for samples A, B, C, and D with nominal thicknesses of 9, 18, 26, and 40 nm, respectively. Finally, on the HT-AlN IL, a 1.1 μm Al<sub>0.25</sub>Ga<sub>0.75</sub>N layer capped with a 40 nm GaN layer was grown under an identical growth condition for all the samples.

Low-temperature CL investigations were performed in a scanning electron microscope (SEM) equipped with an Oxford mono-CL2 and He-cooling stage operating at 9 K. A grating monochromator and a cooled charge-coupled device array were used to disperse and detect the CL signal, respectively. The acceleration voltage of the incident electron beam,  $V_b$ , was adjusted to vary the penetration depth of the incident electrons into the samples and to obtain depth-dependent CL spectra. A series of CL spectra was obtained from line scan measurements by moving the focused electron beam (e beam) step by step along a straight line either on the surface or on the cleaved edge of the samples. Taking into account the electron scattering and the small diffusion length of the excited carriers in AlGa<sub>0.75</sub>N, the diameter of the CL excitation volume, and hence the spatial resolution of the CL, can be estimated to be about 150 nm in a thin AlGa<sub>0.75</sub>N

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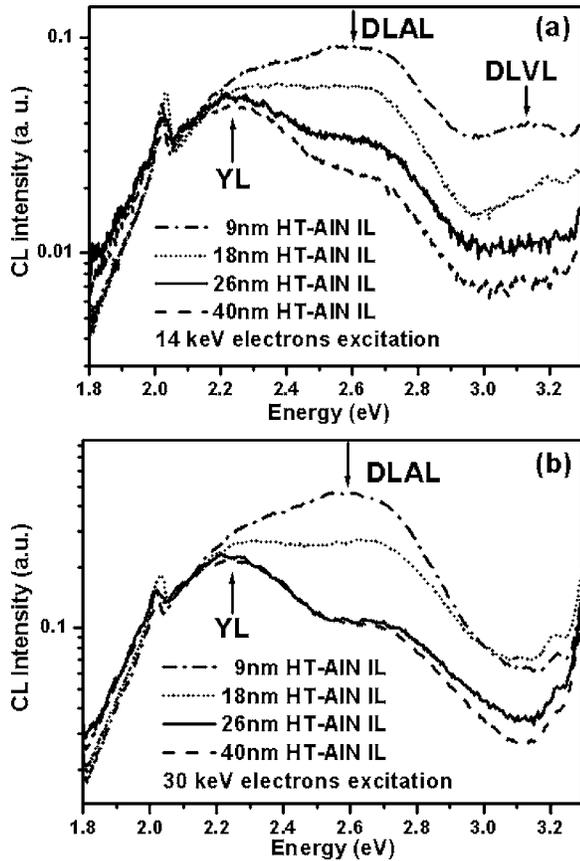


FIG. 1. Low-temperature CL spectra of samples A, B, C, and D using a HT-AIN IL with nominal thicknesses of 9, 18, 26, and 40 nm, respectively. The acceleration voltage of the incident e beam is (a) 14 kV or (b) 30 kV.

film. Monochromatic CL images were obtained by scanning the electron beam over an area of the samples. The crack-free surface morphology of all the samples was observed by SEM.

### III. EXPERIMENTAL RESULTS

Figure 1 shows the low-temperature CL spectra of all the samples, covering a spectral range from 1.8 to 3.3 eV. In each figure, the CL spectra are normalized to maintain the same integral intensity of the near-band-edge (NBE) emissions centered at about 4 eV (not shown) from the  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$  layer of various samples. A sharp peak appears at about 2 eV in all the spectra, which is ascribed to the second order line of the AlGa<sub>N</sub> NBE emission. The penetration depth of 14 keV incident electrons into nitride materials is about 770 nm, according to Monte Carlo simulations using a single scattering model, which falls within the 1.1  $\mu\text{m}$   $\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$  layer. But the GaN underlayer can also be excited by the AlGa<sub>N</sub> NBE emission. In fact, a sharp and intense peak at 3.48 eV and a broad deep level emission at about 2.25 eV were detected from all the samples, which are attributed to be the NBE and the YL band of GaN, respectively.

It is noted that there are two other broad bands at about 2.6 and 3.17 eV in the CL spectra of sample A with the thinnest HT-AIN IL [Fig. 1(a)], which are referred as deep level aquamarine luminescence (DLAL) and violet lumines-

cence (DLVL), respectively. When increasing the HT-AIN IL thickness for samples B, C, and D, the DLVL band is quickly reduced and finally disappears, while the DLAL intensity decreases gradually. The intensity of the DLAL from sample A is higher than that of the YL of the GaN layer. For sample B with an 18 nm HT-AIN IL, however, the DLAL band is weakened to the YL level of the GaN layer. When the thickness of HT-AIN ILs increases beyond 18 nm (i.e., for samples C and D), the DLAL intensity is finally reduced to be smaller than that of the GaN YL. When  $V_b$  is elevated from 14 to 30 kV for CL measurements, the incident electrons may get deeper into the underlying GaN template. Although the DLAL band keeps a similar characteristic, nearly no DLVL band appears in any CL spectra of the samples [Fig. 1(b)], indicating that it does not stem from the GaN underlayer but from the  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$  layer. However, the source of the 2.6 eV DLAL band can hardly be determined by the CL measurements with the normal incidence of the e beam onto the sample surface.

In order to shed light on the respective source of these DLEs, we have performed a cross-section line scan CL measurement on the cleaved edge of sample A along the growth direction from the sapphire/GaN interface towards the Al-GaN layer surface [Fig. 2(a)], which corresponds to the CL spectra sets from the top towards the bottom in Fig. 2(b). Each step of the cross-section line scan covers a distance of about 130 nm. When the incident e beam is focused on the GaN layer, the GaN NBE peak at 3.48 eV and its YL band at about 2.25 eV are clearly observed. No other broad DLE peak appears in the CL spectra of the GaN layer. When the e beam reaches the AlGa<sub>N</sub> layer, both the GaN NBE peak and the corresponding YL band (at 2.25 eV) are replaced by the AlGa<sub>N</sub> NBE peak (at 4.04 eV) and by the DLAL (at 2.6 eV)/DLVL (at 3.17 eV) bands, respectively. The results distinctly demonstrate that the latter two broad emissions originate exclusively from the AlGa<sub>N</sub> layer.

A careful study of the cross-section line scan CL spectra (from step 22 to step 30) reveals that the integral intensity of the AlGa<sub>N</sub> NBE peak, as well as that of the 2.6 eV DLAL band, gradually increases from the GaN/AIN/AlGa<sub>N</sub> interfaces and finally reaches a constant maximum value in the inner region of the AlGa<sub>N</sub> layer, as shown in Fig. 2(c). The intensity of the 3.17 eV DLVL band, however, exhibits a broad maximum centered at step 25, which corresponds roughly to the region near the interfaces. Its intensity decreases gradually when the e beam is moved away from the interfaces towards the AlGa<sub>N</sub> surface. It is noted that at step 24, the DLVL band in the spectra is superimposed by a small and sharp peak at 3.31 eV, which is the LO phonon replica of the donor-acceptor pair recombination line centered at about 3.4 eV. The integral intensity of the 3.31 eV peak is less than 1% of the DLVL, whose decrease cannot contribute to the great reduction (~67%) of the DLVL intensity from step 25 to step 30. In fact, this result implies that the density of the DLVL-related defects decreases from the bottom of the AlGa<sub>N</sub> layer towards the surface, while the concentration of the DLAL-related defects seems to be more uniform in the growth direction. Combined with these findings, the depth-dependent CL spectra (Fig. 1) achieved by varying the accel-

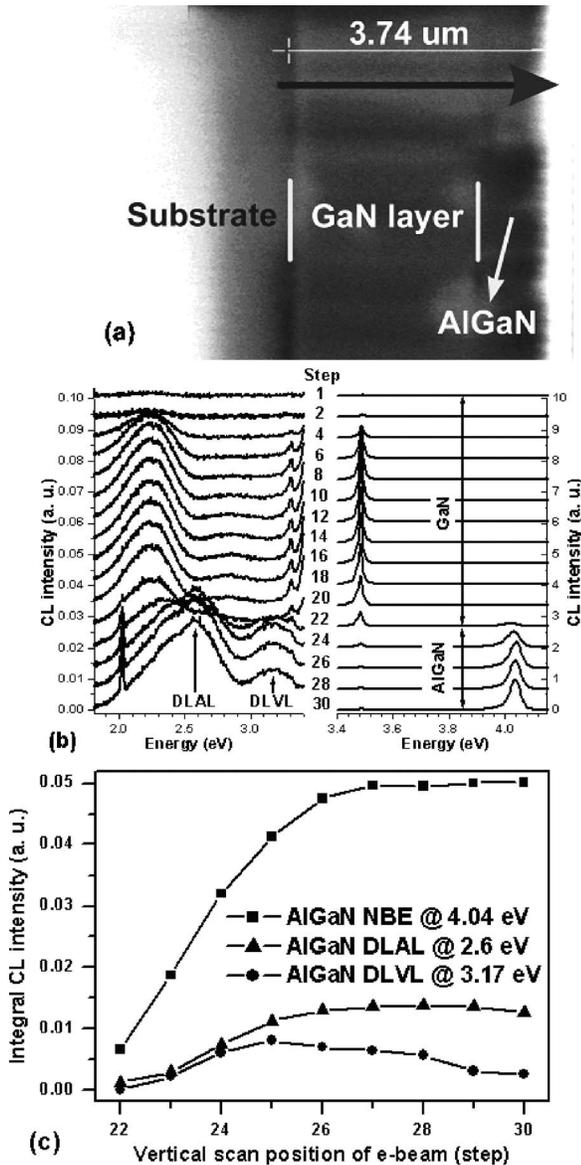


FIG. 2. (a) SEM cross-section image of sample A, where the black arrow indicates the direction of the line scan. (b) Series of CL spectra obtained by the line scan. The curves are vertically shifted for clarity. (c) Integrated intensity of the 4.04 eV AlGaIn NBE peak (square), 3.17 eV DLVL band (circle), and 2.6 eV DLAL band (triangle) as a function of the vertical position (with respect to the layer) of the focused e beam, respectively. Each step of the cross-section line scan covers a distance of 130 nm.

eration voltage of the e beam can be easily understood. Under the excitation of 30 keV electrons, many excited electron-hole (e-h) pairs could recombine around the DLAL-related defects within the AlGaIn layer along the path of incident electrons and emit photons at about 2.6 eV. In contrast, fewer e-h pairs are captured by the DLVL-related defect due to its lower density and inhomogeneous distribution on the trace of incident electrons. Only when the penetration depth of incident electrons is near the region rich in DLVL-related defects near the interfaces can a sufficiently large number of e-h pairs recombine through these defects and make an appreciable contribution to the 3.17 eV DLVL band.

Our recent CL investigations<sup>12</sup> have shown that there is a lateral phase separation in the AlGaIn layer grown on a relatively thick HT-AIN IL. The lateral phase separation shows a

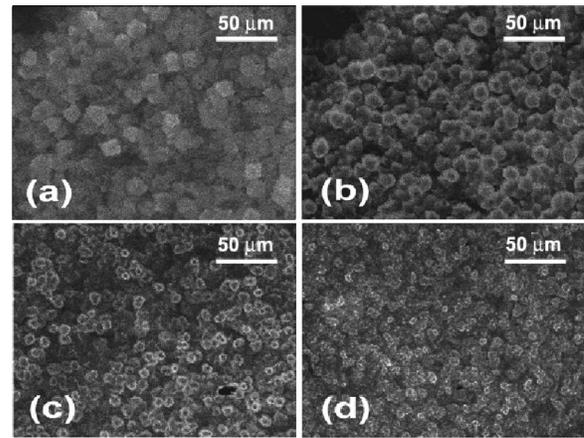


FIG. 3.  $233 \times 173 \mu\text{m}^2$  monochromatic CL images of samples (a) A, (b) B, (c) C, and (d) D taken at 4.008, 4.036, 3.995, and 3.981 eV, respectively.

domainlike feature, as shown in Fig. 3 and the domain size decreases significantly when the thickness of the HT-AIN IL increases. The monochromatic CL image of Fig. 4(a), taken at 4.008 eV, shows a typical domain in sample B. A lateral line scan CL measurement was performed along the line across the domain, as indicated in Fig. 4(a). It is found that the peak energy of the AlGaIn NBE is a bit lower at the periphery ( $\sim 3.99$  eV) compared with the central region ( $\sim 4.04$  eV) of the domain. The CL spectra of steps 7 and 10 represented by a dotted and a solid line in Fig. 4(b), respectively, show that the intensity of the 2.6 eV DLAL band is much stronger in the central region as compared with that in the periphery of the domain. This is more clearly shown in the intensity profiles of Fig. 4(c), where the CL intensity of the DLAL band is indicated by stars. Due to the broad characteristic of the DLAL band, its integrated intensity should include all the contributions from the AlGaIn alloys regardless of  $x_{\text{Al}}$ . We found that during the lateral line scan process, the 2.6 eV DLAL band and the 4.04 eV AlGaIn NBE peak increase in intensity synchronously from the periphery to the center of the domain, while the intensity of the 3.99 eV AlGaIn NBE peak alters in an opposite phase, as shown in Fig. 4(c). However, the sum of the integrated CL intensities of the two AlGaIn NBE peaks remains nearly constant. Consequently, the fluctuation of the DLAL band is not due to an inhomogeneous distribution of nonradiative recombination centers, but due to an inhomogeneous distribution of the DLAL-related defects. The lateral line scan CL spectra across a domain of samples C and D present similar features. It is concluded that a relatively thick HT-AIN IL may cause a lateral domainlike distribution of deep level defects in AlGaIn.<sup>10</sup>

#### IV. DISCUSSION

There is still much controversy in the discussion about the origin of the deep level luminescence of GaN and AlGaIn. Based on the above results, we can confirm that two deep level bands are observed in the  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$  samples. The DLAL band stems from the interior of the AlGaIn layer, while the DLVL is caused mainly by defects near the AlGaIn/AIN/GaN interfacial region. The DLAL band can be

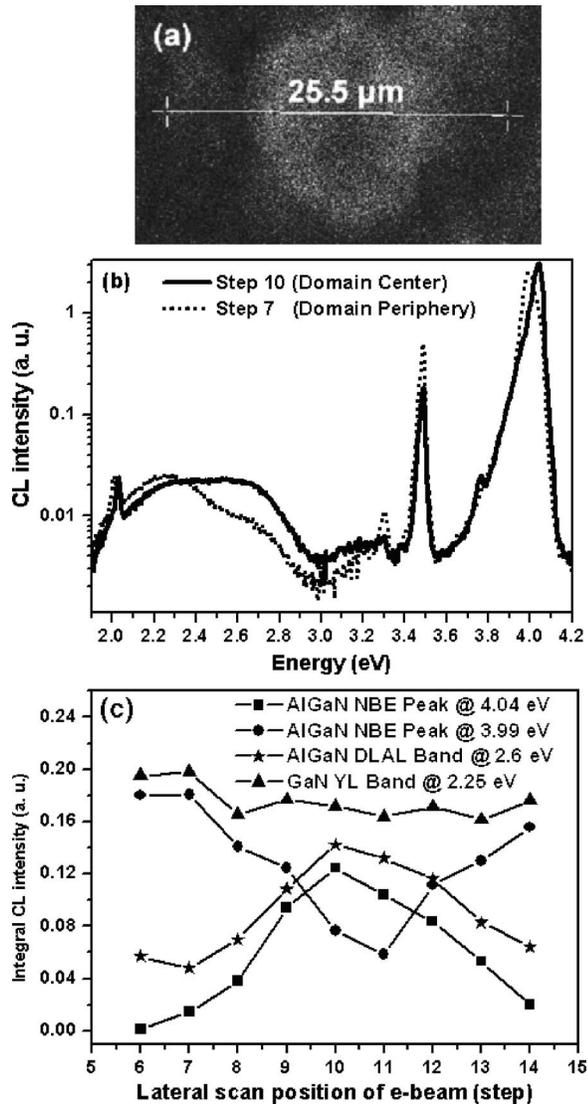


FIG. 4. (a) Monochromatic CL image of a domain structure of sample B taken at 4.01 eV. The horizontal line depicts the path for the lateral line scan CL measurement across the domain. (b) Two CL spectra taken from the periphery (dotted line) and central region (solid line) of the domain, respectively. (c) Integrated CL intensity of the 4.04 eV peak (square), 3.99 eV peak (circle), 2.6 eV DLAL band (star), and 2.25 eV YL band (triangle) as a function of the lateral position of the focused e beam. The integrated intensity of DLAL and YL bands is multiplied by factors of 30 and 20, respectively. Each step of the lateral line scan covers a lateral distance of 1.34  $\mu\text{m}$ .

assigned to evolved from the YL band of GaN. It is well known that the YL band from undoped GaN is located at about 2.25 eV.<sup>5</sup> We have observed an intense and broad peak located at about 2.4 eV in the photoluminescence spectra of  $\text{Al}_{0.1}\text{Ga}_{0.9}\text{N}$  (not shown) and the 2.6 eV DLAL band in the CL spectra of  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$ . The peak position of this band seems to be correlated with the Al composition of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  and shifts to a higher energy value when  $x_{\text{Al}}$  is elevated to a higher level. In fact, the DLE energy of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  has been reported<sup>13,14</sup> to shift gradually from 2.25 to 2.55 eV when increasing  $x_{\text{Al}}$  from zero to 0.25, and track the position of the conduction band bottom.<sup>18</sup> The nature of the YL band from GaN is widely assigned to the radiative transition between a shallow donor and a deep acceptor.<sup>5,16,17</sup> The energy level of

the shallow donor in AlGa<sub>N</sub> alloys follows the bottom of the conduction band, and thus the DLE peak energy increases synchronously together with the band gap of AlGa<sub>N</sub> when  $x_{\text{Al}}$  increases.<sup>18</sup> The nature of the DLAL band from  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$  can be assigned to be analogous to that of the YL band from GaN. The shallow donors and deep acceptors in AlGa<sub>N</sub> are assumed to be formed in the same way as in GaN. The shallow donors are probably formed by unintentional oxygen doping (oxygen on the substitutional nitrogen site) or nitrogen vacancies, while the group III vacancies and their complexes are often assumed to serve as deep acceptors.<sup>5,18</sup>

The DLAL-related defects present a lateral domainlike distribution in the AlGa<sub>N</sub> layer grown on a thick HT-AlN IL. Ponce *et al.*<sup>15</sup> also found that the inner part of the hexagonal GaN domain appears much brighter than the outer region in the monochromatic CL image at 2.22 eV. They assumed that the YL of GaN is probably associated with the clustering of native point defects and/or impurity atoms due to the strain field in the domain. Our previous cross-section transmission electron microscopy study has revealed the existence of V trenches in the HT-AlN IL, which get filled up during the subsequent AlGa<sub>N</sub> layer growth.<sup>12</sup> This feature of mesalike islands separated by V trenches in the HT-AlN IL grown on GaN (Refs. 19 and 20) may lead to an inhomogeneous distribution of strain field. The inner region of domains grown on mesas is more strained than the periphery which mainly consists of the regions over V trenches. It is possible that point defects and/or impurities get accumulated in the strained inner region of domains, which may account for the lateral domainlike distribution of the DLAL-related defects. Moreover, the DLAL band intensity decreases when the HT-AlN IL gets thicker (Fig. 1). This is because the reduction of domain size (Fig. 3) due to the increase of HT-AlN IL thickness results in an area expansion of the peripheral region of domains, and hence a shrinkage of the total area of the strained inner part of domains, which is the region that is rich in point defects and/or impurities and gives DLAL emission.

The exact nature of the DLVL band from AlGa<sub>N</sub> seems still unclear, although there are also some publications concerning this luminescence band. Its energy position was reported either to be independent of  $x_{\text{Al}}$  (Ref. 14) or shift with increasing  $x_{\text{Al}}$ .<sup>13,17</sup> There must exist more than one source that can contribute to this emission band. Taking into account the fact that its intensity decreases from the bottom of the AlGa<sub>N</sub> layer to the surface in our samples, we tentatively attribute the observed DLVL band to interface-related defects.

## V. CONCLUSIONS

We have investigated in detail the spatial distribution of the 2.6 eV DLAL and the 3.17 eV DLVL-related deep level defects in crack-free  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$  layers grown on a GaN template with a HT-AlN IL. When increasing the thickness of HT-AlN ILs, the DLAL intensity decreases gradually, while the DLVL band quenches quickly. By means of a CL line scan at the cross section of the layers, we have demon-

strated that both the DLAL and the DLVL originate exclusively from the AlGa<sub>N</sub> layer. Moreover, we found that the distribution of the DLAL-related defects is relatively uniform in the AlGa<sub>N</sub> layer along the growth direction, but the concentration of the DLVL-related defects decreases gradually from the bottom of the AlGa<sub>N</sub> layer towards the surface. The DLAL band of AlGa<sub>N</sub> is assigned to evolve from the YL band of GaN and is due to a radiative transition between a shallow donor and a deep acceptor, while the DLVL band is related to defects near the GaN/AlN/AlGa<sub>N</sub> interfaces. Additionally, the lateral line scan CL spectra and monochromatic CL imaging reveal a domainlike lateral distribution of the DLAL-related defects in AlGa<sub>N</sub> grown on a relatively thick HT-AlN IL, which is probably induced by the strain field in the domains.

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