

## Optical characterization of AlN/GaN heterostructures

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AlN/GaN/sapphire heterostructures with AlN gate film thickness of 3–35 nm are characterized using photorefectivity (PR) and photoluminescence (PL) spectroscopy. Under a critical AlN film thickness, the luminescence from the GaN channel layer near the interface proves to be excitonic. No luminescence related to the recombination of the two-dimensional electron gas (2DEG) is observed, in spite of high 2DEG parameters indicated by Hall-effect measurements. The increase of the AlN gate film thickness beyond a critical value leads to a sharp decrease in exciton resonance in PR and PL spectra as well as to the emergence of a PL band in the 3.40–3.45 eV spectral range. These findings are explained taking into account the formation of defects in the GaN channel layer as a result of strain-induced AlN film cracking. A model of electronic transitions responsible for the emission band involved is proposed. © 2003 American Institute of Physics.

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### INTRODUCTION

Electronic devices based on III–V nitride materials have recently shown great promise for high-frequency/high-power applications.<sup>1–3</sup> For the most part, these devices are based on low Al-composition, AlGa<sub>x</sub>N/GaN heterostructure field-effect transistors (FETs). However, the gate barrier in the devices involved is based on Schottky contacts which have not yet proven to be stable at high temperatures. In contrast, metal–insulator–semiconductor (MIS)-based FETs can use high-temperature stable insulators at the interface. The use of an insulator at the gate can reduce gate leakage current in a device leading to improved low noise performance. In addition, a thin insulator, by placing the gate much closer to the two-dimensional electron gas (2DEG) channel, can help to improve the intrinsic transconductance of the device. AlN, with its relatively high dielectric constant (8.5) and wide band gap (6.2 eV), has the potential to be an excellent choice for the gate dielectric in GaN based MISFET devices.<sup>4,5</sup>

However, from the growth standpoint, it is difficult to grow high-quality AlGa<sub>x</sub>N layers with high Al content. The problems of lattice mismatch (2.47% for AlN on GaN), pre-reactions between trimethylaluminum and ammonia, and three-dimensional growth must be overcome for effective high-quality AlN/GaN-based MISFET devices.<sup>6,7</sup> As a result, the number of reports on transport properties of Al<sub>x</sub>Ga<sub>1-x</sub>N/GaN structures with  $x > 0.5$  is very limited.<sup>4–7</sup> Nevertheless, the recently achieved values of room-temperature electron mobility ( $\sim 900$  cm<sup>2</sup>/V s) in AlN/GaN heterostructures<sup>7</sup> are very close to the typical values in AlGa<sub>x</sub>N/GaN structures (1000–1600 cm<sup>2</sup>/V s). At the same time, due to the much larger polarization induced electric

fields in AlN/GaN structures, the 2DEG density in this case is several times higher compared to that in AlGa<sub>x</sub>N/GaN heterostructures.<sup>6,7</sup>

While the role of the 2DEG channel in the transport properties of Al<sub>x</sub>Ga<sub>1-x</sub>N/GaN heterostructures is well understood, the data related to the radiative recombination of 2DEG in these structures are scarce and contradictory.<sup>8–11</sup> This is in contrast with Al<sub>x</sub>Ga<sub>1-x</sub>As/GaAs heterostructures where optical properties related to the recombination of the 2DEG are well documented.<sup>12–14</sup> The respective differences between Al<sub>x</sub>Ga<sub>1-x</sub>As/GaAs and Al<sub>x</sub>Ga<sub>1-x</sub>N/GaN heterostructures can be attributed to the very strong piezoelectric polarization in the AlGa<sub>x</sub>N layer on GaN<sup>15,16</sup> that leads to rapid diffusion of photoexcited holes into the flatband region of GaN in Al<sub>x</sub>Ga<sub>1-x</sub>N/GaN heterostructures. Therefore, the probability of recombination between the 2DEG and photoexcited holes is much lower in Al<sub>x</sub>Ga<sub>1-x</sub>N/GaN heterostructures than in Al<sub>x</sub>Ga<sub>1-x</sub>As/GaAs ones. The higher the Al content, the stronger the piezoelectric field, thus reducing the chance to observe radiative recombination of the 2DEG. On the other hand, the higher the Al content, the larger the lattice parameter and thermal expansion coefficient mismatches between AlGa<sub>x</sub>N and GaN. Hence, the probability of defect formation at the Al<sub>x</sub>Ga<sub>1-x</sub>N/GaN interface increases with Al content and, consequently, recombination mechanisms alternative to the 2DEG recombination are expected.

No data are reported to date on radiative recombination mechanisms at an AlN/GaN interface. The goal of this work is to make use of optical methods such as photorefectivity (PR) and photoluminescence (PL) for the characterization of AlN/GaN interfaces in an attempt to shed light upon the radiative recombination channels in Al<sub>x</sub>Ga<sub>1-x</sub>N/GaN heterostructures at high Al content.

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## EXPERIMENTAL DETAILS

The GaN and AlN layers were grown at the University of Michigan by low-pressure (60–110 Torr) metalorganic chemical vapor deposition on *c*-plane (0001) sapphire substrates. Standard precursors of trimethylgallium, trimethylaluminum, and ammonia were used as alkyl and hydride sources. The alkyl and hydride sources were kept separately until just before the quartz reactor. The carrier gas was Pd-cell purified hydrogen ( $H_2$ ). Heating was accomplished by rf induction of the graphite susceptor. All valves and manifolds switching was done by using computer control.

The sapphire substrates were initially cleaned in TCE, ACE, IPA, and  $H_2SO_4/H_3PO_4$ . After a high-temperature (1200 °C) cleaning in  $H_2$ , the growth temperature was lowered to 500 °C. Nitridation was performed and then a  $\sim 20$ -nm-thick GaN nucleation layer was grown. After ramping the temperature to 1100 °C, a 1.3  $\mu m$  unintentionally doped GaN channel layer was grown. The AlN films were grown with thickness ranging from 3 to 35 nm.

The quality of the GaN layer and the presence of AlN thin film were confirmed using a high-resolution x-ray diffractometer (XRD). The XRD scans have shown distinct (0002) GaN and AlN peaks. The full width at half maximum of the GaN peak was  $\sim 120$  arc sec, indicating good quality of the GaN channel layer. An average 4% residual Ga concentration was measured in AlN films using a VG Scientific ESCA Lab II x-ray photoelectron spectroscopy (XPS) system. This was the result of Ga diffusion into the AlN layer, leading to a less sharp interface. However, by changing the growth conditions, it was possible to reduce the Ga diffusion and improve the interface.<sup>7</sup>

The AlN/GaN heterostructures involved were previously characterized by Hall-effect measurements, atomic force microscopy (AFM), and cathodoluminescence (CL) microanalysis.<sup>17</sup> The 2DEG Hall mobility was found to decrease with increasing AlN film thickness. This decrease proved to correlate with the increase of the red-yellow CL intensity.

The PR was measured using the light from a halogen lamp. The reflected white light from the sample was analyzed through a double spectrometer with 1200 lines/mm gratings assuring a linear dispersion of 0.8 nm/mm. The signal from a FEU-106 photomultiplier with SbKNaCs photocathode working in a photon counting mode was connected to an IBM computer via IEEE-488 interface. The PL was excited by the 334 nm line of an  $Ar^+$  SpectraPhysics laser and analyzed with the same experimental setup. The resolution was better than 0.5 meV in both PR and PL experiments. The samples were mounted on the cold station of a LTS-22-C-330 workhorse-type optical cryogenic system.

## RESULTS AND DISCUSSION

Figure 1 illustrates typical PR and PL spectra of GaN channel layers. The reflectivity spectrum consists of two major features related to the  $X_A^{n=1}$  and  $X_B^{n=1}$  exciton ground states as well as a weak feature associated with the  $X_A^{n=2}$  excited state. The luminescence spectrum is predominated by the  $D^0X$  bound exciton,<sup>18</sup>  $X_A^{n=1}$  and  $X_B^{n=1}$  excitonic emis-

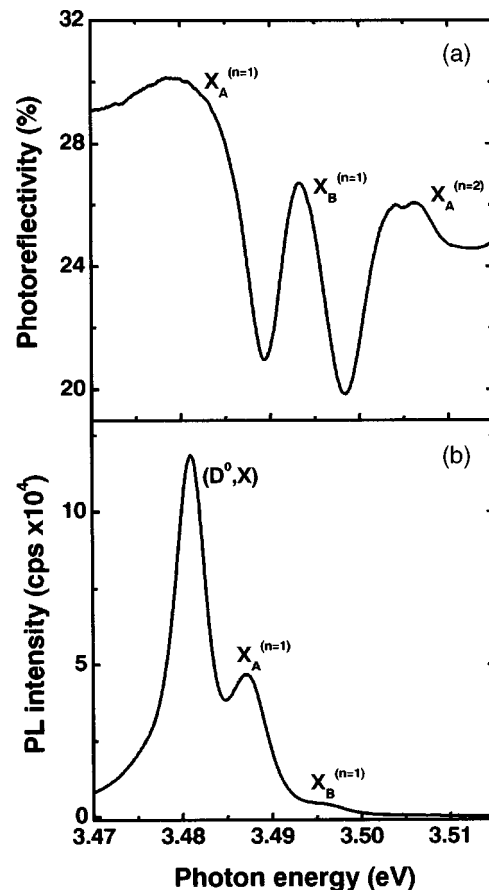


FIG. 1. Reflectivity (a) and PL (b) spectra of GaN channel layer measured at 10 K.

sion. This luminescence is at least three orders of magnitude more intensive than 3.29 eV and yellow luminescence. The position of the  $D^0X$  (3.481 eV),  $X_A^{n=1}$  (3.487 eV) and  $X_B^{n=1}$  (3.495 eV) peaks in the luminescence spectrum is shifted by  $\sim 10$  meV towards high energies in comparison with the position of respective excitons in nonstressed GaN layers.<sup>19</sup> This means that considerable strain remains in the GaN channel layer due to the mismatches between the layer and sapphire substrate. Strains of 0.5 GPa are estimated using the previously reported rates of the exciton line shifts with the biaxial stress in GaN layers.<sup>19</sup>

The exciton resonance amplitude starts to decrease when increasing the AlN gate film thickness up to 12 nm. The excitonic features disappear completely in the reflectivity spectrum of the GaN layer if the AlN film is as thick as 30 nm (Fig. 2). Analogous changes are realized in the PL spectra of GaN layers in relation to the thickness of the top AlN film (Fig. 3). The intensity of the exciton luminescence peaks decreases sharply at AlN film thickness more than 12 nm. At the same time, a new luminescence band at 3.40–3.45 eV emerges. The 3.40–3.45 eV PL band is one of the commonly occurring bands in  $Al_xGa_{1-x}N/GaN$  heterostructures<sup>8–11</sup> and GaN layers.<sup>20–38</sup> In  $Al_xGa_{1-x}N/GaN$  heterostructures this band is believed to be related to the recombination of the 2DEG with photoexcited holes. Different recombination channels have been considered as the origin of this band in GaN layers: (i) recombination of excitons bound to structural

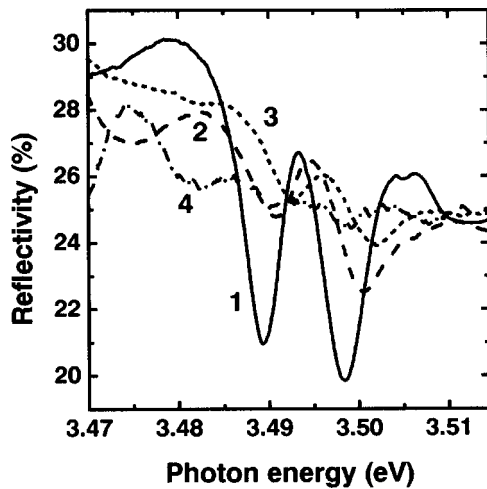


FIG. 2. The reflectivity spectrum of GaN channel layer measured at 10 K for different values of the AlN gate film thickness, nm: 1–5; 2–12; 3–15; 4–30.

defects or to stacking faults;<sup>20–28</sup> (ii) recombination between electrons bound to a donor and free holes;<sup>29–35</sup> (iii) donor acceptor pair (DAP)-type transitions involving a very shallow acceptor.<sup>36–38</sup> Taking this into account, it is reasonable to assume that the origin of luminescence in the 3.40–3.45 eV spectral range is different for different samples, depending on the material processing.

The decrease and the disappearance of exciton features in the PR and PL spectra of GaN layers is indicative of serious damages induced by the AlN film at the AlN/GaN interface. These findings correlate with the decrease in the 2DEG Hall mobility observed in the structures with AlN film thicker than 12 nm.<sup>17</sup> In light of these observations we believe that the luminescence in the 3.40–3.45 eV spectral

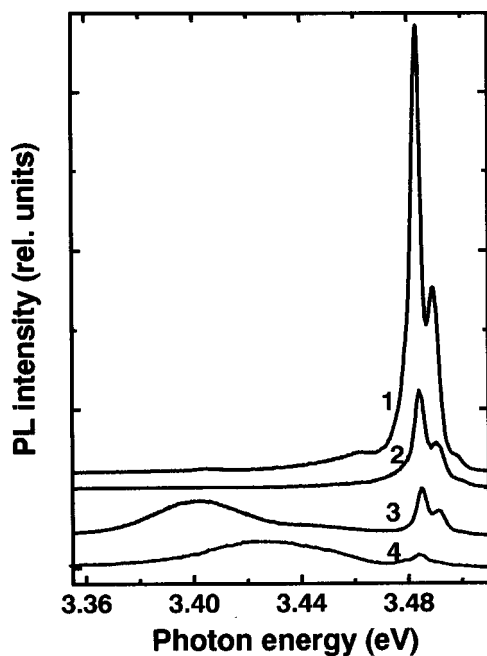


FIG. 3. PL spectra taken from GaN layers covered by AlN gate film with the thickness, nm: 1–5; 2–12; 3–15; 4–30.  $T = 10$  K.

range in our AlN/GaN heterostructures is doubtful to come from the recombination of the 2DEG. Apart from that, no features characteristic of the radiative recombination of the 2DEG were observed in the luminescence spectra at undercritical AlN layer thickness. At the same time, electrical characteristics point out to electron confinement effects. The room-temperature electron mobility reaches values over  $500 \text{ cm}^2/\text{V s}$  at undercritical AlN film thickness, while the bulk GaN mobility for our samples is  $\sim 80 \text{ cm}^2/\text{V s}$ . The existence of the 2DEG near the AlN/GaN interface is also clearly indicated by the temperature dependence of electrical characteristics. For instance, the sample with 5 nm AlN film thickness shows mobility saturation at a value over  $3000 \text{ cm}^2/\text{V s}$  at low temperatures.<sup>7</sup>

Previously, the effect of the sharp mobility decrease in AlN/GaN structures was realized at AlN barrier thickness larger than 5 nm.<sup>39</sup> AFM study undertaken in these AlN/GaN structures allows for identification of one of the factors causing sharp mobility decrease in specimens with thick AlN films. In structures with  $d_{\text{AlN}} > 5$  nm, lines corresponding to cracks in the AlN film were found to appear along different crystallographic planes, pointing out the beginning of the tensile strain relaxation process in the AlN film. Gradual relaxation processes in GaN–AlN–GaN structures were shown to start at the AlN film thickness of 3 nm.<sup>40</sup> The thickness of a dislocation free layer in  $\text{GaN}_m\text{AlN}_n$  superlattices (SLs) was estimated as  $2.5 \text{ nm} < L < 3.7 \text{ nm}$ <sup>41</sup> which is in good agreement with experimental data. It was also shown that the relaxation in GaN–AlN SLs fully develops when the layer thickness approaches 10 nm.<sup>41</sup>

The earlier results indicate that in our samples the relaxation processes start at AlN gate thickness around 12 nm, and fully develop at 30 nm thickness. According to AFM data,<sup>17</sup> the root-mean-square roughness for the 5- and 15-nm-thick films is 0.25 and 0.57 nm, respectively. The 15-nm-thick film exhibits defects of 100–200 nm in size. The different values of the critical AlN thickness in our experiments and studies carried out earlier<sup>40,41</sup> may be explained by 4% residual Ga concentration in our AlN films as evidenced by XPS.<sup>17</sup> Obviously, the critical film thickness increases with the Ga content increase. Substantially larger critical thickness was achieved in  $\text{GaN}-\text{Al}_x\text{Ga}_{1-x}\text{N}$  superlattices.<sup>42</sup> The higher the Ga content in  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  barriers, the higher the critical thickness beyond which the barriers relax via crack channels.<sup>42</sup> Partial relaxation of the AlGaN layers accompanied by deterioration of the transport properties in AlGaN/GaN structures was observed when the  $\text{Al}_{0.22}\text{Ga}_{0.78}\text{N}$  film thickness exceeded 60 nm.<sup>43</sup> One should mention, in this regard, that practically in all cases when the observation of the luminescence related to the 2DEG was claimed, the  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  film thickness was near-critical or overcritical.<sup>8–11</sup> Therefore, the defect nature of the luminescence should not be excluded from consideration.

Obviously, cracking of the AlN film initiates the formation of different structural and point defects in the GaN layer. The temperature and excitation power density dependence of the luminescence was analyzed for the purpose of getting additional information about the nature of PL band in the 3.40–3.45 eV spectral range. Figure 4 compares the PL spec-

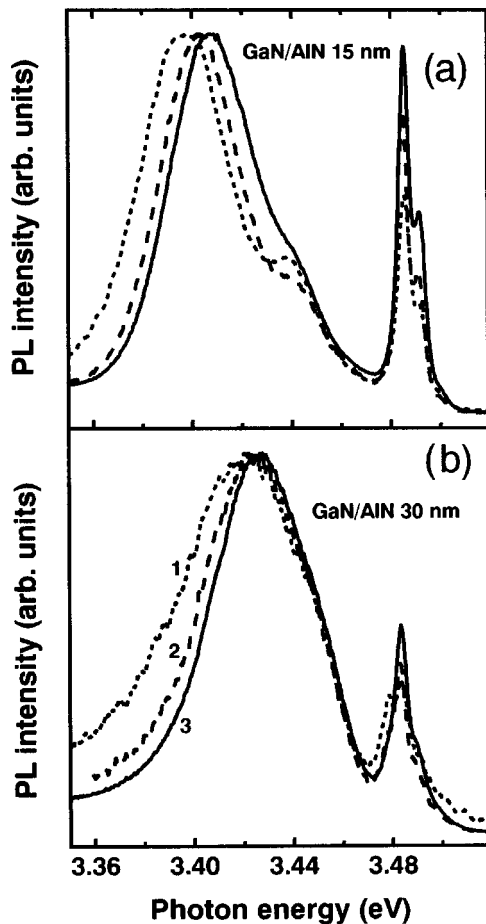


FIG. 4. PL spectra of GaN layers covered by 15- (a) and 30-nm- (b) thick AlN film.  $T = 10$  K. The excitation power density,  $\text{W}/\text{cm}^2$ : 1–1; 2–5; 3–25. The spectra are normalized to the intensity of the low energy PL band.

tra taken with different excitation power densities from the GaN layer for 15 (a) and 30 nm (b) AlN film thickness. The spectra are normalized to the intensity of the low energy PL band for convenience. The PL spectra measured on the same samples at different temperatures and normalized to the intensity of the excitonic luminescence are shown in Fig. 5. The analysis of the PL spectra in Figs. 4 and 5 indicates that the PL band at 3.40–3.45 eV is not elementary and the position of the maximum is different for samples with 15 nm and 30 nm AlN film thickness.

The energy position of the band involved shifts to higher energies with increasing the incident excitation power density (Figs. 4 and 6). This is consistent with the saturation of distant DAPs under increasing excitation level. This behavior is opposite to that observed previously for the PL band located at 3.40–3.42 eV and associated with excitons bound to structural defects.<sup>20–28</sup> It is also different from that inherent to band-to-impurity ( $D^0, h$ ) transitions.<sup>30,31</sup> Indeed, there is a possibility for shifting of the band-to-impurity transitions towards high energies with increasing excitation power density, as a result of band filling. In such a case, however, the measured shift is about 5 meV for three orders of magnitude variation of the excitation power density.<sup>31</sup> We observe a shift of 15 meV with the excitation density variation by a factor of 100 (Fig. 6) which is consistent with DAP

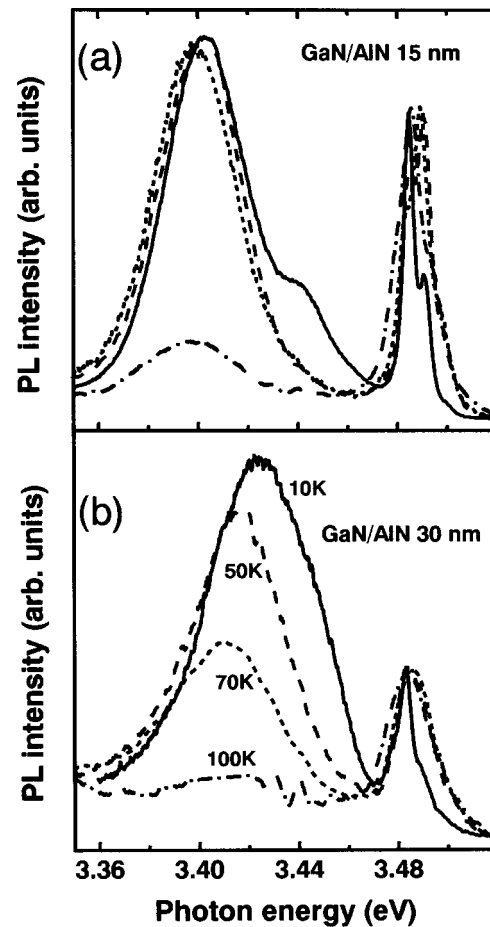


FIG. 5. PL spectra of GaN layers covered by 15- (a) and 30-nm- (b) thick AlN film measured at different temperatures under the excitation power density of  $5 \text{ W}/\text{cm}^2$ . The spectra are normalized to the intensity of the excitonic luminescence.

recombination.<sup>36,38</sup> Apart from that, in the case of band-to-impurity transitions the intensity of ( $D^0, h$ ) luminescence increases with the excitation power density slightly superlinearly and faster than the intensity of excitonic luminescence.<sup>31</sup> As one can see from Fig. 7, in our samples

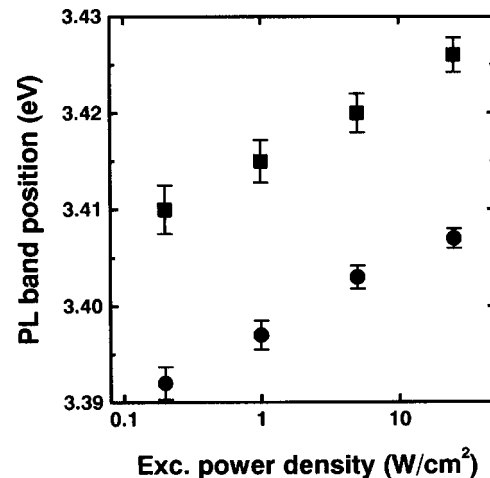


FIG. 6. The spectral peak position of the DAP emission as a function of the excitation power density for GaN layer covered by AlN film of 15 nm (circles) and 30 nm thickness (squares).  $T = 10$  K.

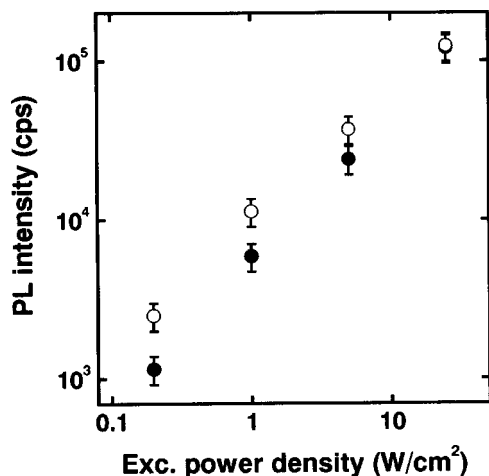


FIG. 7. The integrated PL intensities for the  $D^0X$  (full symbols) and DAP (open symbols) emissions as a function of the excitation power density.  $T = 10$  K.

the excitonic luminescence increases nearly linearly with excitation power density, while the dependence of DAP luminescence is sublinear, exhibiting a tendency to saturation. All these features are indicative of the DAP origin of the involved PL band in our samples.

However, in our experiments the temperature dependence of luminescence is unexpected for both band-to-impurity and DAP transitions. The luminescence line should move towards the band edge with increasing temperature in both cases,<sup>31,36</sup> the rate of approach being roughly  $kT/2 - 2kT$ , depending upon the nature of transition. In contrast to this, with increasing temperature the intensity of PL in our samples sharply decreases (much faster than the intensity of the excitonic luminescence) and the maximum shifts to lower energies (see Fig. 5). A possible explanation of this contradiction is the nonelementary character of the PL band involved. Obviously, different donor and acceptor-like native defects, or residual impurity-native defect complexes may be created in the GaN layer as a result of AlN film cracking.

Let us now compare our data with those reported on radiative recombination of the 2DEG.<sup>8-11</sup> A blueshift of the 2DEG peak is also expected with increasing excitation power density since the enhanced concentration of excess carriers results in reduced bending of the conduction band at the heterointerface due to increased screening. However, for Al content in  $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$  heterostructures less than 30%, the blueshift of the 2DEG peak with increasing excitation power density is either not observed<sup>8</sup> or is insignificant (a few milli-electron-volts per two orders of magnitude change in the excitation power density).<sup>9,10</sup> Taking into account the near-critical or overcritical AlGaN film thickness in the papers under consideration, it is not easy to distinguish between the 2DEG and band-to-impurity recombination.

The only significant blue shift of the PL band with increasing excitation power density, supposed to be due to the 2DEG, was recently reported in  $\text{Al}_{0.5}\text{Ga}_{0.5}\text{N}/\text{GaN}$  heterostructures.<sup>11</sup> The PL spectra presented in Ref. 11 are very similar to our spectra. As mentioned earlier, in Al-rich  $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$  heterostructures the piezoelectric field in-

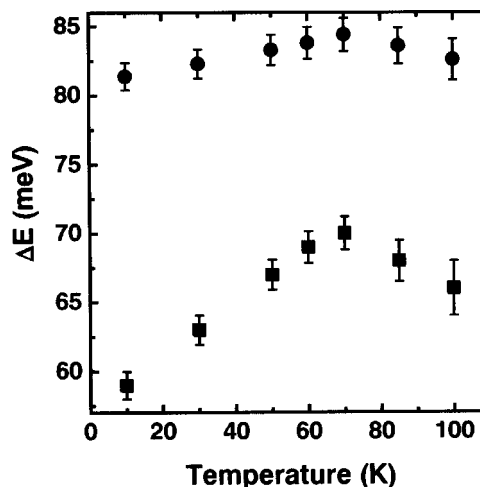


FIG. 8. The temperature dependence of the energy separation between the  $D^0X$  and DAP emission peaks,  $\Delta E$ , in GaN layer covered by AlN film of 15 (circles) and 30 nm (squares) thickness.

duced photoexcited hole diffusion is a major problem. Even at Al content as low as 22% the incorporation of an additional AlGaN layer was necessary to increase the 2DEG luminescence.<sup>9</sup> In spite of this, the 2DEG peak in Ref. 9 is several orders of magnitude weaker than the corresponding PL band in Ref. 11 where no care was taken to suppress the hole diffusion. Therefore, the previously reported data on 2DEG luminescence in  $\text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$  heterostructures are contradictory and the contribution of the recombination related to defects cannot be excluded.

The experimental data described earlier suggest a model for the broad emission at 3.40–3.45 eV in AlN/GaN heterostructures that is based on carrier recombination between a band of shallow donors and a shallow acceptor. Shallow residual impurities like Mg, Be, H,<sup>37,44</sup> or  $\text{V}_{\text{Ga}}$ -impurity complex may play the role of acceptor. As concerns the donor band, it can be caused by different defect complexes involving the nitrogen vacancy.<sup>34,35</sup> The stronger the damages in the GaN layer, the broader the donor energy band, it spreading out closer to the conduction band (CB). This explains the PL band shift to high energies in samples with 30 nm AlN film in comparison with samples with 15 nm AlN film. It is the luminescence related to the transitions from energy levels situated closer to the CB that is first thermally quenched with increasing temperature (see Fig. 5). As a result the energy separation between the  $D^0X$  and DAP emission peaks,  $\Delta E$ , initially increases with temperature (see Fig. 8), in contrast with the expected behavior. The disappearance of the high-energy wing in the PL spectrum at low temperatures suggests a very close approach of the donor energy band to the CB. If one assumes that the upper limit of the PL band position (about 3.443 eV as deduced from Figs. 4 and 5) is due to the transitions from the donor energy levels situated close to the CB and taking the band gap value of 3.503 eV, one can estimate the acceptor level to be situated about 60 meV above the valence band. This value is expected for the hydrogen-like acceptors in GaN.<sup>37,44</sup> Unfortunately, it is difficult at this stage to establish reliably the nature of the defects involved. Additional investigations using complemen-

tary methods are necessary to throw light upon the nature of these defects.

## CONCLUSION

In the investigated AlN/GaN/sapphire heterostructures the luminescence of GaN channel layer proves to be excitonic for undercritical AlN film thickness. An emission band at 3.40–3.45 eV predominates the PL spectrum of GaN layers for overcritical AlN gate film thickness. Taking into account the temperature and excitation power density dependence of this band, we suggest that it corresponds to electron transitions from a donor-type energy band to a defect-related acceptor level. The defects involved are created in the GaN layer as a result of AlN film cracking at overcritical thickness. This model is confirmed by the decrease of excitonic features in the PL and PR spectra as well as by the previously observed decrease of the two-dimensional electron gas Hall mobility and the increase of the red-yellow cathodoluminescence.

## ACKNOWLEDGMENT

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