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Nature of low-energy optical emission in doped AlGaN/GaN heterostructures

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Photoluminescence (PL) in modulation-doped and nominally undoped $Al_xGa_{1-x}N/GaN$ heterostructures was studied and compared with PL spectra of GaN films grown on sapphire substrates. It is demonstrated that optical emission in the energy range of 3.3–3.46 eV related to the two-dimensional electron gas radiative processes can be completely suppressed in modulation-doped $Al_xGa_{1-x}N/GaN$ heterostructures. Instead of this, an intense broad long-wavelength emission attributed to the recombination of donor-acceptor pairs in the lower energy range of 2.7–3.3 eV is revealed. This spectral transformation is explained by the presence of deep-level defect-related acceptor centers in $Al_xGa_{1-x}N/GaN$ heterostructures introduced at the modulation doping of the $Al_xGa_{1-x}N$ barrier layer. © *2007 American Institute of Physics*.
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I. INTRODUCTION

In recent years, GaN-based heterostructures have been widely used in high-power, high-voltage, and high-frequency electronics.[1] In particular, AlGaN/GaN heterostructures are attractive for various field-effect transistors with high electron mobility transistors (HEMTs), metal-semiconductor field-effect transistors (MESFETs).[2] In these devices a two-dimensional electron gas (2DEG) of large carrier density ($>10^{13}$ cm^{-2}) at the AlGaN–GaN interface can be formed even in nominally undoped heterostructures. Due to specific nitride properties, *c*-axis wurtzite GaN-based heterostructures possess strong built-in piezoelectric fields ($\sim 10^6$ V/cm) thus affecting the behavior of the confined 2DEG. The induced fields pull the electrons into the quantum well (QW) at the heterointerface and push the holes out of the QW area. As a result, photoluminescence (PL) related to the 2DEG-free holes recombination in AlGaN/GaN heterostructures is weak, and special techniques are needed to increase PL efficiency. Possible approaches include increasing the Al content ($x>0.2$) in the $Al_xGa_{1-x}N$ barrier for better 2DEG confinement,[3] incorporation of a thin $Al_xGa_{1-x}N$ ($x\approx 0.1$) layer at the bottom of the GaN buffer for double 2DEG confinement,[4] use of a nucleation AlN layer prior to *i*-GaN growth on the substrate to improve the structure stoichiometry,[5] and additional *n*-type doping in an unintentionally doped (UID) AlGaN barrier to enhance 2DEG radiative recombination.[6] A number of optical studies of modulation-doped (MD) AlGaN/GaN heterostructures have been performed during the past decade. Bergman *et al.*[7] provided insights about a broad PL emission in a spectral range of 50 meV below the GaN band gap (E_G=3.503 eV at 4.2 K) associated with recombination between the 2DEG electrons accumulated at the heterointerface band potential and the photoexcited holes located near the flatband of the valence band. Fang *et al.*[8] studied the low-temperature PL spectra in A1GaN/GaN HEMTs with various δ-doping densities and spacer thicknesses, and found different interband transitions from the 2DEG subbands to the valence band corresponding to the energy range of 3.486–3.312 eV. Shields *et al.*[9] reported about a magnetoresonance PL phenomenon caused by 2DEG recombination at a single AlGaN/GaN heterojunction. Nevertheless, many issues related to the PL of 2DEG in modulation-doped AlGaN/GaN heterostructures are still not resolved. This paper reports on features of the PL behavior in AlGaN/GaN heterostructures caused by Si doping in the AlGaN barrier in comparison with an unintentionally doped GaN-based heterostructures and GaN films grown on sapphire substrates.

II. EXPERIMENT

All samples were grown by low-pressure metal-organic chemical vapor deposition (MOCVD) on monocrystalline (0001) Al_2O_3 430-μm-thick substrates. The heterostructures

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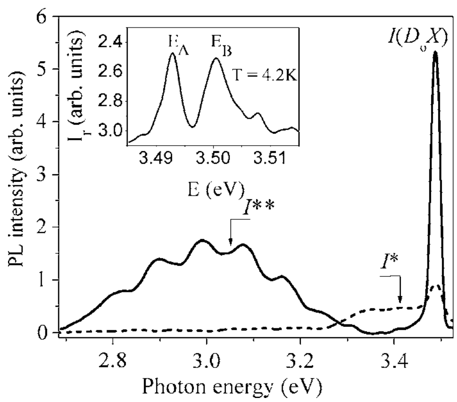

FIG. 1. PL spectra of the AlGaN/GaN heterostructures measured with the photoexcitation from the front-face side: the dashed curve corresponds to UID heterostructure (U), the solid curve corresponds to MD heterostructure (D). Inset: photoreflectance spectrum of the MD heterostructure (D), T =4.2 K.

of type I (marked U for "undoped") consisted of a 28 nm $Al_xGa_{1-x}N$ (x=0.33) UID barrier and a 1.5 μm GaN UID layer. The heterostructures of type II (marked D for "doped") have a 10 nm $Al_xGa_{1-x}N$ cap (x=0.25), a 15 nm $Al_xGa_{1-x}N$ Si-doped layer (x=0.25; $N_D \approx 2 \times 10^{18}$ cm^{-3}), a 3 nm $Al_xGa_{1-x}N$ UID spacer (x=0.25), and a 2 μm GaN UID base layer. In addition, a test sample of a 1 μm GaN epitaxial film (marked S for "single" layer) was used for comparison. It is known that the background density of impurities N_D-N_A in the nominally undoped heterostructures strongly depends on the growth regime used as well as on the precursors purity, in particular, the purity of ammonia.[10] In our case, as follows from the data obtained in the Hall effect measurements on GaN and AlGaN epitaxial layers, the background density was at the level of 10^{16} cm^{-3}.

Photoluminescence spectra were measured under two excitation conditions: front face and back face for three types of GaN-based structures. A steady-state PL was excited by a 325 nm He–Cd laser radiation (continuous-wave power of 7 mW) and detected by a sensitive photomultiplier tube using a scanning grating monochromator with a spectral resolution better than 0.02 meV. Photoreflectance spectra were measured using the excitation of a Xe lamp. Optical measurements were performed at a cryogenic temperature T =4.2 K under similar conditions in order to provide correct relations between PL spectral intensities and thus to get comparative information about the photoexcited carriers and the native and impurity-defect-related states involved in the PL process.

III. RESULTS

Figure 1 shows the PL spectra for AlGaN/GaN heterostructures (U and D) measured at T=4.2 K. The inset in Fig. 1 presents a typical photoreflectance spectrum (T=4.2 K) for the sample D, which reveals two well-resolved features, E_A =3.492 eV and E_B=3.501 eV. These values coincide within ±1.0 meV accuracy with similar resonance energies detected in samples U and S, and are consistent with the free exciton (FE) ground states X_A and X_B (n=1) for good epitaxial GaN/Al_2O_3 films.[7] Though the full width at half maximum (FWHM) of the photoreflectance bands in the heterostructures is larger by a factor of 2 than that in the reference GaN films, we tentatively assume the good structural quality of our heterostructures.

The dominating feature in the near band-edge region in the PL spectra for samples U and D in Fig. 1 is located at the energy E_{D_0X}=3.487 eV (FWHM $\approx$22 meV), which is slightly below the FE transition with energy E_A. This feature, labeled $I(D_0X)$, is well known and is naturally assigned to the donor-bound exciton D_0X band, i.e., recombination of the exciton bound to a neutral shallow donor in GaN followed by electron capture on the donor in its ground state. The binding energy ε^* is estimated to be $\varepsilon^* = E_A - E_{D_0X}$ $=5 \pm 1$ meV for both heterostructures. A low-energy side with respect to the donor-bound exciton D_0X feature of the PL spectrum for the UID heterostructures (U) demonstrates a broad plateau I^* with a weakly developed structure, which spans the range from 3.25 to 3.46 eV (FWHM$\approx$150 meV). We attribute this I^* feature to the recombination between the 2DEG electrons at the AlGaN–GaN interface and the photoexcited holes in the GaN layer, as in Ref. 7. It should be mentioned that the sample U is unintentionally doped, and the 2DEG channel here is formed only due to the polarization effects. According to the Hall measurement data for undoped AlGaN/GaN heterostructures,[11] a sheet carrier density n_s of the 2DEG is about 1×10^{13} cm^{-2}. No other resolvable features are detected below the energy of 3.25 eV in the PL spectrum of the undoped heterostructure (U in Fig. 1).

The PL radiative recombination of the MD heterostructures (D in Fig. 1) demonstrates completely different behavior. It was expected that doping the $Al_{0.25}Ga_{0.75}N$ barrier by Si (N_D=2×10^{18} cm^{-3}) would strengthen the below-bandgap PL due to the increased 2DEG concentration in comparison to the undoped heterostructure. Indeed, the donor-bound exciton D_0X peak became five times stronger but surprisingly a plateau I^* below the donor-bound exciton D_0X in the range of 3.3–3.46 eV completely disappears. Instead of this, an intense broad, long-wavelength feature I^{**} superimposed by interference fringes with a maximum at ~3.0 eV appears in the lower energy range from 2.7 to 3.3 eV (FWHM $\approx$350 meV). The integral intensity of this I^{**} emission is 20 times higher than for the $I(D_0X)$ band, and the total quantum yield of PL in the sample D is greater by an order of magnitude than in U. Such significant modification of PL in the doped heterostructure is the evidence of the presence of a high concentration of additional recombination centers located in the GaN layer near the interface between the GaN and AlGaN barrier layer with characteristic energies in the range corresponding to the long-wavelength emission of the I^{**} band.

Results of PL spectra measurements, shown in Figs. 2 and 3, provide further confirmation of our findings. These figures represent data for heterostructures U and D, measured in two different geometries. The PL was excited by laser illumination either from the front-face side or from the back face. In the case of a front-face excitation, the laser radiation was transmitted via the AlGaN barrier, through the AlGaN–GaN interface, and was absorbed by the GaN heterolayer. With back-face excitation, the laser light passed via the

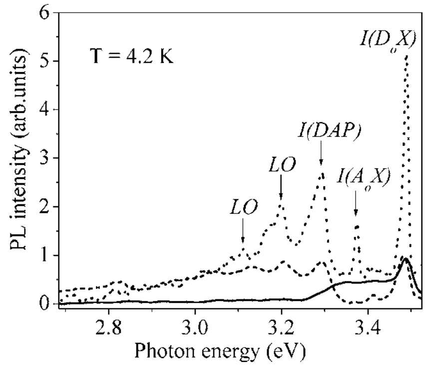

FIG. 2. Low-temperature (T=4.2 K) PL spectra of UID AlGaN/GaN heterostructure (U) measured with the photoexcitation from the front-face side (solid curve) and back-face side (dashed curve). PL spectrum of GaN film (dotted curve) is shown for comparison.

transparent Al_2O_3 substrate, through the $GaN-Al_2O_3$ interface and was absorbed in the GaN buffer. The PL spectrum recorded for the GaN/Al_2O_3 film (S) is presented for comparison in the figures.

The features observed in the PL spectrum of the reference GaN film (S) are typical of the bulk GaN (Refs. 7 and 12) and are assigned to (1) donor-bound exciton D_0X (E_{DBE}=3.489 eV, FWHM≈14 meV), (2) acceptor-bound exciton A_0X (E_{ABE}=3.374 eV, FWHM≈11 meV), and (3) donor-acceptor pairs (DAP) (E_{DAP}=3.29 eV, FWHM ≈46 meV) and their longitudinal optical (LO) phonon replicas ($\hbar\omega_{LO}\approx 90$ meV) at 3.20 and 3.11 eV. In addition, the spectrum demonstrates a few weak lines of excitonic emission at 3.41–3.43 eV due to shallow impurity-related complexes.[13] It should be noted that the PL spectra recorded with illumination from the back face of AlGaN/GaN heterostructures (U and D) reproduce in detail the PL spectrum of the GaN film except for the $I(A_0X)$ peak that disappeared in the spectra corresponding to both samples U and D. The relative intensity and spectral position of the dominant $I(D_0X)$ peak for D and S are nearly the same, only the strength of the donor-bound exciton D_0X band in the PL spectra of U is significantly reduced with respect to the D heterostructure. The FWHM of the $I(D_0X)$ peak in the spectrum of S is smaller than in the U and D spectra due to the more perfect structure of the epitaxial GaN film in comparison with AlGaN/GaN heterostructures. The PL features related to the states at the AlGaN/GaN interface in the heterostructures are not resolved in the PL spectra, taken under illumination from the back-face side.

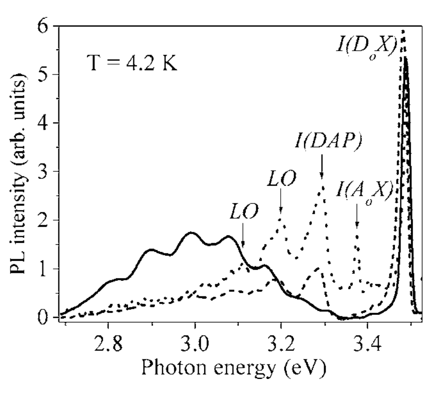

FIG. 3. Low-temperature (T=4.2 K) PL spectra of MD AlGaN/GaN heterostructure (D) measured with the photoexcitation from the front-face side (solid curve) and back-face side (dashed curve). PL spectrum of GaN film (dotted curve) is shown for comparison.

IV. DISCUSSION

It is known that the lattice mismatch between GaN and Al_2O_3 (~15%) in GaN structures on sapphire substrates results in a high density of threading dislocations (from 10^8 to 10^{10} cm^{-2} depending on the growth conditions). The dislocation density slowly reduces with increasing distance from the interface toward the GaN layer.[1] The penetration of dislocations is considerable and can influence the properties of excitonic complexes and donor-acceptor pairs. In particular, they may introduce deep ($\varepsilon_a \gg kT$) acceptorlike centers, which may capture photoexcited carriers and thus result in changes of the PL spectra.[12] More extensive defects are formed in strained AlGaN/GaN heterostructures,[2] especially in the case of modulation doping.[7] Additionally, the piezoelectric polarization fields are also of great importance. Therefore, changes in intensities, bandwidths, and peaks of PL spectra reflect the structural modification of the heterostructures.

Indeed, the energy position of the DAP feature at 3.29 eV is quite similar in the PL spectra of the good quality GaN film grown on Al_2O_3 substrate (S) and of $AlGaN/GaN/Al_2O_3$ heterostructures illuminated from the back face, (U and D). It is known that the DAP emission in GaN layers arises due to the availability of shallow donors and shallow acceptors (A_1^0) caused by the point lattice defects and residual impurities located at the mismatch dislocations.[7] In this case, the electrons recombine with holes, which after the photoexcitation and the relaxation occur in states of shallow acceptor centers in GaN. The shallow acceptors reveal themselves in all samples studied—U, D, and S. However, in D the DAP emission spreads (Fig. 1) on the long-wavelength side in comparison to that of U and S samples. This spectral spreading can be caused by additional acceptor centers (A_2^0) created during the doping of the heterostructure barrier layer. In this case, the DAP emission is formed by two recombination processes in accordance with two kinds of acceptor centers: $\langle e(D^0)-h(A_1^0)\rangle$ and $\langle e(D^0)-h(A_2^0)\rangle$. This model is consistent with the experimental data,[12] which showed that the DAP recombination weakly observed in undoped GaN epilayers at 3.27 eV becomes more intensive in the Si-doped GaN.

The most striking features were found in the PL spectra of $AlGaN/GaN/Al_2O_3$ heterostructures excited from the front-face side in comparison to the back-face excitation of MD doped heterostructures (Fig. 3). The observed difference in the PL spectra together with an analysis of self-consistently calculated potential profiles for the U and D heterostructures allows us to draw a conclusion about the nature of the low-energy PL emission observed in the doped heterostructures. The results of the theoretical self-consistent calculation of the Schrödinger and Poisson equations for the energy band potential and energy levels at the AlGaN–GaN interface are shown in Figs. 4 and 5, taking into account the

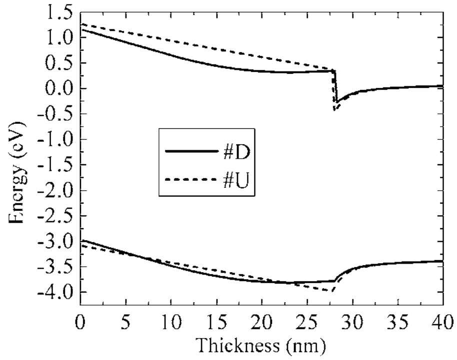

FIG. 4. Self-consistently calculated potential profile for UID (U) and MD (D) heterostructures.

relevant AlGaN and GaN material parameters (effective masses, conduction and valence band offsets, spontaneous and piezoelectric fields, etc.).[1] The sheet charge densities were measured as $\sim 1\times 10^{13}$ and $\sim 6\times 10^{12}$ cm^{-2} for U and D samples, respectively, resulting in a V-shape quantum well with 2DEG at the heterointerface. For analysis and comparison, we plot the two profiles for both types of heterostructures together with the calculated electron wave functions in Fig. 5 on an enlarged scale. The width of the potential well is estimated to be about $L_{QW}\approx 4$ nm, the energies of the ground subband are about ~ 0.15 and ~ 0.07 eV, for U and D samples, respectively. As can be seen from the figure, the extension of the electron wave function in the case of the MD structure is larger and one should expect an increase of the penetration of the wave function into the GaN layer. At the same time, the holes due to the induced electric fields drift away from the heterojunction and are located in the flatband region of the valence band. Indeed, recombination between the confined 2DEG electrons and the photoexcited holes results in the appearance of a broad low-energy shoulder below the donor-bound exciton D_0X, as defined in the PL spectra for the UID heterostructures (U in Fig. 1). The density of photoexcited holes is much lower than the density of 2DEG, therefore a magnitude of the 2DEG PL shoulder is determined mainly by the free holes available. Since the PL feature related to the 2DEG electron-hole recombination in the MD heterostructures is absent (D in Fig. 1), we conclude that there is a very small quantity of free holes in this case. This testifies to the presence of additional trapping centers

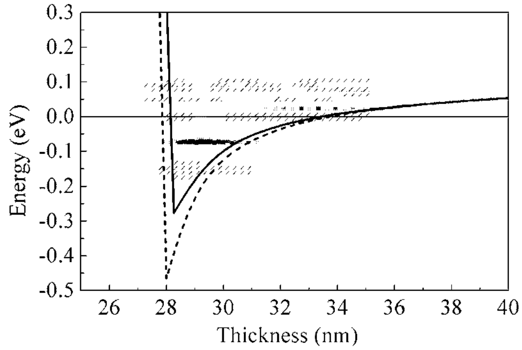

FIG. 5. Enlarged potential profile and electron wave functions for UID (U) and MD (D) heterostructures for the region of the triangular quantum well with 2DEG.

that reduce the lifetime of the free holes. The decrease of the 2DEG PL in the energy range of 3.3–3.46 eV below the donor-bound exciton D_0X is accompanied by the simultaneous increase of the DAP emission in the lower energy range of 2.7–3.3 eV. Obviously, this modification is related to the increase of the density of the photoexcited holes bound to the deep acceptors in the GaN layer.

The PL features related to transitions in the AlGaN barrier are not resolved because the layer is too thin ($\delta = 28$ nm) to hold the photoexcited carriers within the recombination time, and its band gap ($E_G\approx 3.97$ eV) is above the photon energy of the laser source ($h\nu_l = 3.815$ eV). The laser light is absorbed mainly in the GaN layer [the absorption coefficient at 325 nm is $\alpha = 10^5$ cm^{-1} (Ref. 1)]. The effective absorption length ($\ell_a\approx 0.1$ μm) is 10–20 times smaller than the thickness of the GaN layers studied ($L = 1.5-2$ μm), therefore one can expect different behavior of the PL upon the excitation of the sample from the front-face and back-face side. In the case of front-face excitation the localized states at the AlGaN–GaN interface are dominant in the radiative processes, whereas with the back-face excitation the states manifest themselves at the $GaN-Al_2O_3$ interface. These arguments allow us to conclude that the deep-level defect-related acceptors in GaN are introduced from the side of the doped AlGaN barrier, giving rise to the DAP recombination enhancement in the low-energy range from 2.7 to 3.3 eV (D in Fig. 1).

The low-energy PL band at 3.0 eV can be explained by transitions from a donor-type level located close to the conduction band, and the estimated acceptor energy level is about ~ 0.5 eV above the valence band. Indeed, the measured spectrum contains a broadband resulting from a number of overlapping elementary bands related to the recombination emission of pairs with different energies. The energy positions of DAP features at 3.17, 3.08, 2.99, 2.90, and 2.81 eV in Fig. 1 correspond to LO-phonon replicas with the energy of $\hbar\omega_{LO}\approx 90$ meV. Additionally, our results are in good agreement with the recently reported data[13] about the acceptor-type mechanism as the dominant DAP recombination in the PL spectra with the participation of deep-level defect-related complexes abundantly produced in mass-transport-grown GaN structures.

V. CONCLUSION

In summary, low-temperature PL spectroscopy of modulation-doped and nominally undoped AlGaN/GaN HEMT heterostructures as well as epitaxial GaN films grown on sapphire substrates was studied. A steady-state PL related to the recombination of the 2DEG at the AlGaN–GaN interface is found to be strongly dependent on the excitation conditions and structural morphology of the investigated samples. It is found that a below-band-gap feature of the PL attributed to 2DEG electron-hole recombination in the energy range of 3.3–3.46 eV can be weaken in the doped AlGaN/GaN. Instead of this, intense broad long-wavelength emission is formed due to the recombination of the donor-acceptor pairs in the energy range of 2.7–3.3 eV with a maximum at 3.0 eV. This spectral transformation is explained as a result of the formation of an additional radiative mechanism due to the deep-level acceptor centers in GaN introduced during the doping of the AlGaN barrier layer. These centers capture the photoexcited holes and strengthen the probability of donor-acceptor recombination. 2DEG electron-hole recombination emission decreases due to the reduction of free holes, and DAP recombination is dominant due to the additional deep acceptors in the low-energy range. The resultant PL modification caused by modulation doping in AlGaN/GaN is established and depends on the doping density and on the position of the doping layer with respect to the AlGaN/GaN interface.

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