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Below band gap photoreflectance transitions in epitaxial GaN

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A photoreflectance (PR) and photoluminescence (PL) study has been performed on a Si-doped epitaxial GaN layer that contains impurity or defect related below band gap features in its PR spectrum. In the 300 K PR spectrum, these features appear at energies of 3.26 and 3.33 eV, respectively, but below 180 K they can no longer be seen. The 3.26 eV line evidently corresponds to a donor acceptor pair transition, also seen in PL. The origin of the 3.33 eV line is uncertain, but may correspond to a transition involving the nitrogen vacancy. © 2004 American Institute of Physics. [DOI: 10.1063/1.1780602]

The photoreflectance (PR) technique has recently been demonstrated^{1,2} to be a valuable method for the characterization of compound semiconductors, heterostructures, and multiple quantum wells. In most PR experiments, the critical point energy is determined by fitting the shape of the signal at the band edge with a first or third derivative functional form, for low electric fields. However, PR spectra below the band gap have been observed^{3–8} from bulk semiconductors where experimental evidence indicates that the features derive from impurity transitions. Glembocki *et al.*⁶ performed systematic experiments on GaAs in order to identify the below band gap spectra (BLS) and also to understand the mechanisms that produce them. They compared the electrolytic electroreflectance spectra of their *p*-type GaAs samples with photoluminescence spectra and were able to show that the BLS could be attributed to acceptor impurity transitions. However, other experiments showed that BLS resulted from back surface reflection effects.^{9,10} Tober *et al.*¹¹ attributed the BLS in their GaAs samples to both impurities and back surface reflections.

In this letter, we report the observation of below bandgap spectra in GaN epitaxial layers. Photoluminescence (PL) measurements were also performed to identify the origin of the BLS. Experiments were performed on two GaN samples, A and B, grown by metalorganic vapor phase deposition (MOCVD) with Si doping, and by hydride vapor phase epitaxy (HVPE) without any deliberate doping, respectively. Hall-effect measurements yielded carrier concentration (n) and mobility (μ) values of $n_A = 1.5 \times 10^{18} \text{ cm}^{-3}$, $n_B = 4.5 \times 10^{16} \text{ cm}^{-3}$, $\mu_e^A = 209 \text{ cm}^2/\text{V s}$, and $\mu_e^B = 610 \text{ cm}^2/\text{V s}$. The thickness of samples A and B are 1.3 and 30 μm , respectively.

Our photoreflectance apparatus and technique is similar to that discussed in Ref. 12. The excitation source was the

strong 296-nm line of a 150-W Hg–Xe lamp, filtered by using a monochromator (PTI, $f=0.2 \text{ m}$). The reflectance of the sample was probed by exposing monochromatically dispersed light from a 200 W Xe lamp using a scanning monochromator (Spex, $f=0.5 \text{ m}$). The reflected light from the sample passed through another scanning monochromator (Optometric, $f=0.125 \text{ m}$) that functioned as an adjustable, narrow bandpass filter. It was used to remove spurious light such as photoluminescence or scattered light. The detector was a photomultiplier tube. The signal was preamplified and detected by the conventional lock-in technique. PL data were obtained by using the 10 mW, 325 nm line of a HeCd laser as an excitation source. Figure 1 illustrates some selected PR spectra from samples A and B at $T=300 \text{ K}$. The line shape fitting was made in the context of Pollak's first derivative

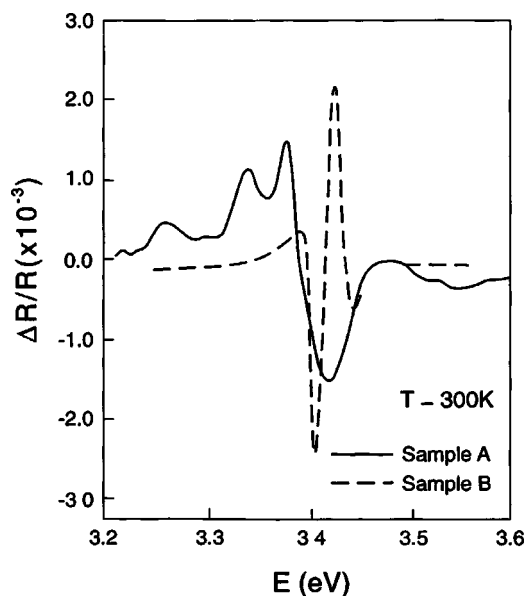


FIG. 1. PR spectra obtained at $T=300 \text{ K}$ for samples A and B.

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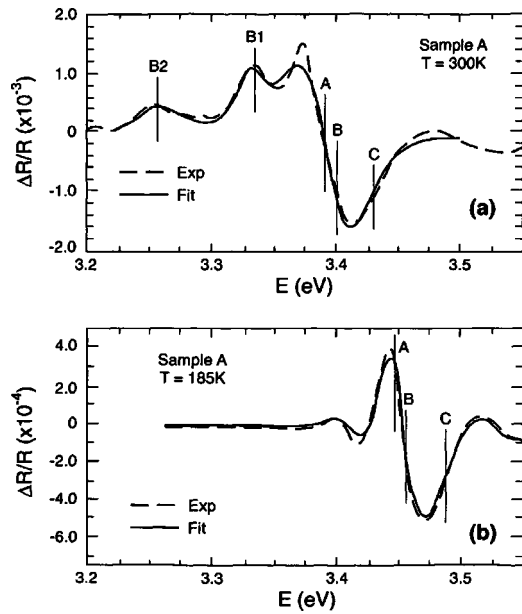


FIG. 2. PR spectra obtained at $T=300\text{ K}$ (a) and $T=185\text{ K}$ (b) for sample A.

line shape method.¹³ The solid lines of Fig. 2 are least-squares fitted to the first derivative of a Lorentzian functional form, which is appropriate for an excitonic transition¹⁴

$$\Delta R/R = \text{Re} \left[\sum_{j=1}^3 A_j e^{i\theta_j} (E - E_j + i\Gamma_j)^{-2} \right], \quad (1)$$

where A_j , θ_j , E_j , and Γ_j are the amplitude, phase, energy, and broadening parameter, respectively, of the j th feature and E is the photon energy. Vertical lines in the figure designate the fitted values of the energies.

The PR spectra of sample A can be fit with three resonances as indicated by vertical lines, which correspond to the A, B, and C excitons of the direct band gap¹⁵ and are related to the $\Gamma_9^v - \Gamma_7^c$, Γ_7^v (upper band) $-\Gamma_7^c$, and Γ_7^v (lower band) $-\Gamma_7^c$ interband transitions in the wurzite lattice. However, sample A shows extra features below the A exciton in Figs. 1 and 2. The extra features are designated as B1 and B2 in Fig. 2. The vertical lines denote transition energies obtained from the fit of Eq. (1). Note that sample B does not show the below band gap features, which is indeed the case for most GaN samples. The energy differences between A and B1 and B2 are $\Delta(A - B1) = 63\text{ meV}$ and $\Delta(A - B2) = 141\text{ meV}$.

PR spectra for sample A (Fig. 2), shows that lower temperatures result in higher energies for the A, B, and C excitons, which are expected from the band gap temperature dependence. But the B1 and B2 transitions also experience change, gradually decreasing in amplitude with decreasing temperature. At 185 K they completely disappear [Fig. 2(b)]. Figure 3 displays the photoluminescence results in the range 2.9–3.5 eV. Prominent lines include a strong near band edge emission at 3.466 eV, and a weaker 3.275 eV no-phonon line, with three longitudinal optical (LO) phonon replicas. The 3.466-eV peak can be decomposed into two peaks at 3.467 eV and 3.455 eV. We attributed these peaks to a neutral donor bound exciton¹² and a neutral acceptor bound exciton,¹¹ respectively. The neutral donor is likely Si_{Ga} ,^{16,17} since the sample was doped with Si. The I_2 energy of 3.467 eV is less than the 3.473 eV value observed in a less Si doped sample,¹⁶ probably because of increased stress re-

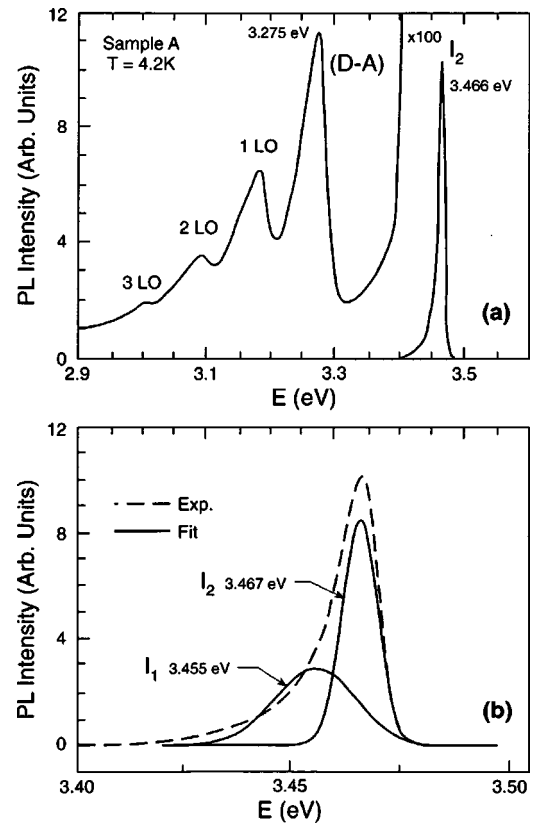


FIG. 3. PL spectrum obtained at $T=4.2\text{ K}$ for sample A. (b) Shows that the 3.466 eV peak is decomposed into two peaks at 3.467 and 3.455 eV.

laxation due to the higher Si doping,¹⁸ and also more band gap renormalization,¹⁹ due to the higher electron concentration. The acceptor may be due to Si_N , or unintentional Mg doping.

The 3.275 eV emission is likely a donor acceptor pair (DAP) recombination,²⁰ an assignment supported by the higher energy shift of the peak position with higher excitation intensity. If we assume a 4 K band gap of 3.503 eV, donor energy of 30 meV, and an average Coulomb coupling energy of 10 meV, then the acceptor energy must be about 220 meV, typical for acceptors such as Mg. Note that B2 has a 300 K energy of about 3.26 eV, about the same as that of the DAP line considering the temperature difference in the spectra. Thus, we believe that B2 in the PR spectrum may represent a DAP transition. As the temperature is lowered, the DAP transitions become weaker with respect to the excitonic transitions. In the more sensitive PL spectra, the DAP lines are still observable at low temperature, but this is evidently not the case for the PR spectra. More PL and PR spectra were taken on 5 additional samples to clarify the origin of the B2 in the PR spectra. Two of the samples of GaN were grown by MOCVD and three samples were grown by molecular beam epitaxy (MBE) techniques. Samples C and D were grown by MOCVD with room temperature carrier concentrations of $n_C = 2.8 \times 10^{17}\text{ cm}^{-3}$ and $n_D = 5.0 \times 10^{17}\text{ cm}^{-3}$. The sample E was grown with $n_E = 5.3 \times 10^{17}\text{ cm}^{-3}$ using the MBE method. Note that Fig. 4 shows two below band gap PR features for samples C and D while sample E does not show the below band gap feature. In fact three MBE samples up to $n = 1.6 \times 10^{18}\text{ cm}^{-3}$ do not show the below band gap PR features nor the $\sim 3.27\text{ eV}$ (DAP) transitions. It is evident that the $\sim 3.275\text{-eV}$ (DAP) transitions

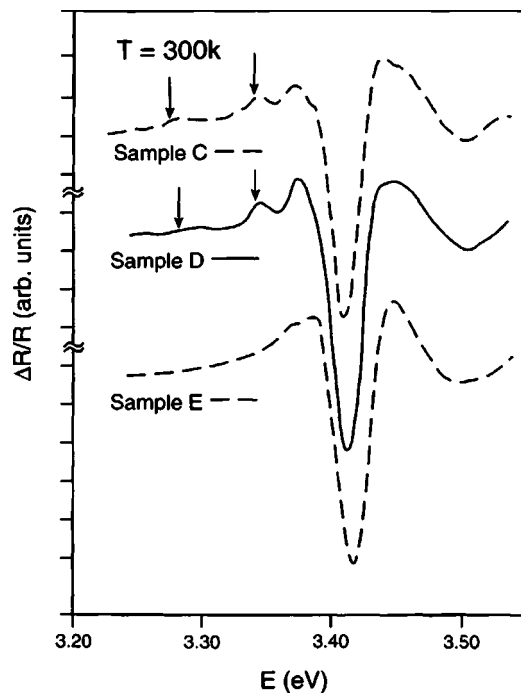


FIG. 4. PR spectra obtained at $T=300$ K for samples C, D, and E. The arrows indicate the presence of below band gap transitions which are seen in samples C and D but not in sample E.

can be correlated with the below band gap transition B2.

The origin of B1 is somewhat more uncertain, because no PL analog is observed. If B1 also has the nature of a DAP line, then the acceptor would have to be very shallow, only about 80 meV. Although such a scenario is unlikely, still it must be noted that two-electron satellite lines have been used to identify an acceptor nearly this shallow (Reynolds *et al.*).²¹ Another possibility might be a free hole to nitrogen vacancy (V_N) donor transition, since the V_N energy is in the range 70–80 meV.²² However, at this time, the identity of B1 remains a mystery.

In summary, we have observed below band gap photo-reflectance spectra at 300 K in Si-doped GaN samples grown by MOCVD. One of the PR lines, at 3.26 eV, appears to correspond to a donor acceptor pair line, also seen in low-temperature photoluminescence spectra. The line is not ob-

servable by PR at the lower temperatures, evidently due to the dominance of excitonic transitions.

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