

Parasitic sub-band-gap emission originating from compensating native defects in Si doped AlGa_n

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(Received 16 July 2007; accepted 30 August 2007; published online 19 September 2007)

The authors investigate the dependence of the photoluminescence intensity ratio between the near-band-edge and the parasitic blue emission as a function of silane flow in *n*-type Al_{0.3}Ga_{0.7}N. It is found that when the silane flow rate is higher than 1.13×10^{-9} mol/min, the UV-to-blue ratio is a constant. When the silane flow rate is lower than 1.13×10^{-9} mol/min, the UV-to-blue ratio increases rapidly as the silane flow rate decreases. A theoretical model is proposed, which assumes that the parasitic blue emission is caused by an acceptorlike compensating native defect. The model is fully consistent with the experimental results. © 2007 American Institute of Physics.

[DOI: 10.1063/1.2786838]

AlGa_n based ultraviolet (UV) light-emitting diodes (LEDs) are expected to have many applications such as biological agent detection, water purification, nonline-of-sight communications, and thin film curing.¹ However, a broad parasitic emission band in the blue-green region is often observed in UV LEDs.²⁻⁵ There has not been a definite conclusion on the origin of this parasitic emission. A similar situation occurred in the early days of the GaN research: it was found that the room-temperature photoluminescence (PL) spectrum of GaN samples grown by various techniques often contained a parasitic sub-band-gap yellow luminescence (YL) band emitting at 2.20–2.25 eV ($\lambda=552-565$ nm).⁶⁻¹⁰ The ratio between the near-band-edge emission and the YL intensity was reported to decrease,^{7,8} increase,⁹ or remain approximately unchanged¹⁰ with Si doping in different experiments.

In this letter, the near-band-edge and the parasitic blue emission of *n*-type Al_{0.3}Ga_{0.7}N are investigated as a function of silane flow. A detailed study is performed on the intensity ratio between the near-band-edge and parasitic blue luminescences. A theoretical model is proposed that considers an acceptorlike compensating native defect. The results obtained from the model are in excellent agreement with the experimental results.

The *n*-type Al_{0.3}Ga_{0.7}N samples are grown by using an Aixtron 200/4 RF-S metalorganic vapor-phase epitaxy (MOVPE) system. Trimethylgallium (TMGa), trimethylaluminum (TMAI), and ammonia (NH₃) are used as precursors for Ga, Al, and N, respectively. Diluted silane (SiH₄) is used as the *n*-type dopant source. In order to investigate the Si doping effect on the parasitic blue emission, several samples

are grown with the same layer structure but different Si doping levels. The sample structures are shown in Fig. 1. The details about the growth conditions can be found in Ref. 11. The top 1.35- μ m-thick Al_{0.3}Ga_{0.7}N layers are doped using different silane flow rates, varying from 0 (undoped) to 2.79×10^{-8} mol/min (heavily doped). For the room temperature PL measurement, a frequency-quadrupled neodymium doped yttrium aluminum garnet laser with emission wavelength of 266 nm is used as the excitation source.

Room-temperature photoluminescence spectra of a heavily doped and an undoped Al_{0.3}Ga_{0.7}N sample are shown in Fig. 2. Both samples show a sharp luminescence peak at 295 nm, which is the near-band-edge luminescence. This will be referred to as the “UV luminescence.” For the heavily doped Al_{0.3}Ga_{0.7}N sample, there is another broad luminescence band centered at around 450 nm, which will be denoted as the “parasitic blue emission.” The PL spectra for the Al_{0.3}Ga_{0.7}N samples doped with different silane flow rates (not shown here) are similar with that of the heavily doped sample. All Si doped samples show a sharp UV emission as well as a parasitic blue emission. However, the parasitic blue

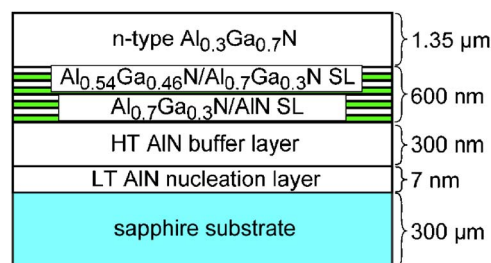


FIG. 1. (Color online) Schematic *n*-type Al_{0.3}Ga_{0.7}N sample structure grown on the (0001) plane of sapphire substrate.

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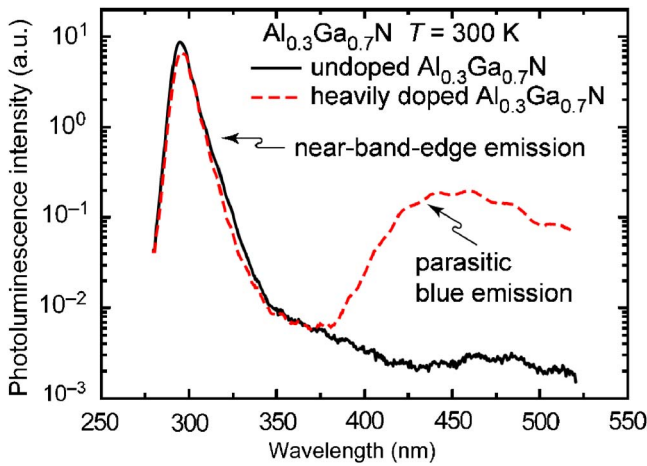


FIG. 2. (Color online) Room-temperature photoluminescence spectra of undoped and heavily doped $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$ samples.

emission for the undoped $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$ sample is two orders of magnitude lower than the heavily doped sample.

The peak intensity ratio between the UV luminescence (I_{UV}) and the blue luminescence (I_{blue}) at different silane flow rates is shown in Fig. 3. The free carrier concentrations of samples A–D are obtained from room-temperature Hall measurements, as shown in the figure. It is found that when the silane flow rate is higher than 1.13×10^{-9} mol/min, the UV-to-blue ratio is almost unchanged even though the silane flow rate changes over more than one order of magnitude. However, when the silane flow rate is lower than 1.13×10^{-9} mol/min, the UV-to-blue ratio increases rapidly as the silane flow rate decreases. Especially when the silane flow rate reaches zero (undoped), the UV-to-blue ratio for the undoped $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$ reaches 3000.

Next, a theoretical model that considers acceptorlike compensating native defects is developed to help understand the observed behavior of the UV-to-blue ratio versus silane flow. As shown in Fig. 4, the neutral (unoccupied) and negatively charged (occupied) deep level concentrations are represented as N_T^0 and N_T^- , respectively, with the total deep level concentration expressed as $N_T = N_T^0 + N_T^-$. Since not all the donors (with a doping concentration of N_D) in the n -type $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$ are ionized at room temperature, the fraction of

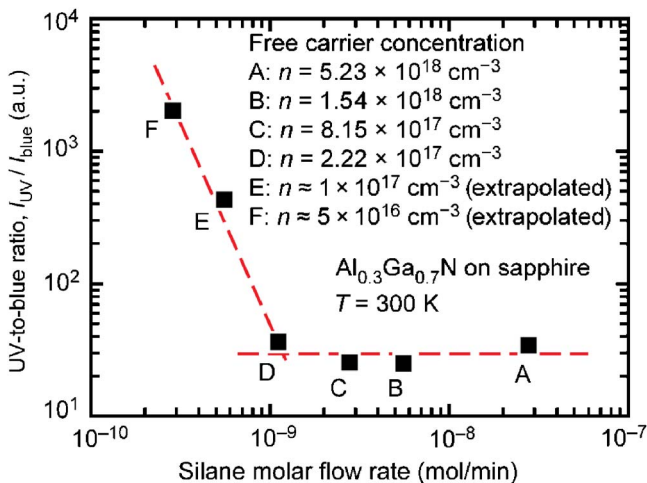


FIG. 3. (Color online) Peak intensity ratio between the UV and parasitic blue emissions, $I_{\text{UV}}/I_{\text{blue}}$, as a function of silane flow rate.

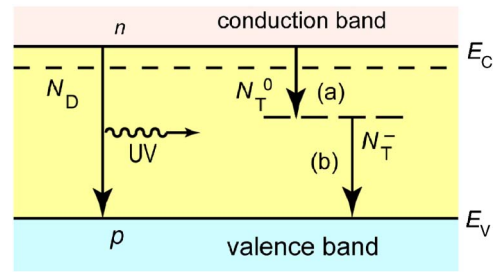


FIG. 4. (Color online) Band diagram of n -type $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$ illustrating near-band-edge UV emission and parasitic blue emission coming from a deep-level transition.

the ionized donors is expressed as $\alpha_D N_D$, where α_D is a constant representing the ratio of ionized donors to the total doping concentration. Under optical excitation, the electron and hole concentrations in the n -type $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$ are $n \approx \alpha_D N_D + \Delta n$ and $p \approx \Delta p$ ($\Delta n \approx \Delta p$), where Δn and Δp are the excess carrier concentrations induced by optical excitation.

The intensity of the UV luminescence can be expressed by the bimolecular recombination with the recombination coefficient B ,

$$I_{\text{UV}} = Bnp = B(\alpha_D N_D + \Delta n)\Delta p. \quad (1)$$

The deep level transition consists of two sequential transitions, (a) and (b), whose transition rates are given by

$$I_{(a)} = C_1 n N_T^0, \quad (2)$$

$$I_{(b)} = C_2 N_T^- p. \quad (3)$$

Here C_1 and C_2 are the recombination coefficients for transitions (a) and (b), respectively. Under steady state, the transition rates of these two sequential transitions are equal, that is,

$$C_1 n N_T^0 = C_2 N_T^- p. \quad (4)$$

It is known that the abundance of the compensating native defects increases with doping concentrations.^{10,12,13} The total formation energy for the defects continuously decreases as the n -type conductivity increases.¹⁴ Therefore, the dependence of the deep level concentration on the doping concentration can be expressed according to the power law,¹⁰

$$N_T^0 + N_T^- = N_T \propto (N_D)^i \quad (i = 0, 1, 2, \dots). \quad (5)$$

For typical compensating native defects, i is equal to 1 or 2, that is, the concentration of the defects increases with doping concentration. In our model, it is also assumed that the parasitic blue emission comes from the recombination between electrons and neutral deep levels, $I_{\text{blue}} = I_{(a)}$. One could also assume that the parasitic blue emission comes from the recombination between holes and negatively charged deep levels, $I_{\text{blue}} = I_{(b)}$. Since the transition rates of these two sequential transitions are equal under steady state, there will be no difference for the final results of the model, no matter which of the two assumptions is used.

Next, two situations will be considered. In the *low-doping regime*, $\alpha_D N_D \ll \Delta n$, the ionized donor concentration is smaller than the excess carrier concentration induced by optical excitation. In this situation, both the electron and hole concentrations are approximately equal to the optically ex-

cited carrier concentration, i.e., $n \approx p \approx \Delta n$. Equation (4) then yields $N_T^- \approx (C_1/C_2) \times N_T^0$. Inserting this expression of N_T^- into Eq. (5), one obtains $N_T^0 = C(N_D)^i$, where C is a constant. Thus, the ratio between the UV luminescence and the parasitic blue luminescence in the low-doping regime is

$$\frac{I_{UV}}{I_{blue}} = \frac{I_{UV}}{I_{(a)}} = \frac{Bnp}{C_1 n N_T^0} \propto \frac{\Delta n}{C(N_D)^i}. \quad (6)$$

On the other hand, in the high-doping regime, $\alpha_D N_D \gg \Delta n$, the ionized donor concentration is larger than the optically excited carrier concentration. The electron and hole concentrations are $n \approx \alpha_D N_D$ and $p \approx \Delta p \approx \Delta n$ with $n \gg p$. This condition leads to the inequality $N_T^0 \ll N_T^-$ in Eq. (4). Using this inequality in Eq. (5) yields

$$N_T^- \approx N_T (N_D)^i \Rightarrow N_T^- = C'(N_D)^i. \quad (7)$$

Inserting this result into Eq. (4) yields

$$N_T^0 = \frac{C'' \Delta n}{\alpha_D} (N_D)^{i-1}, \quad (8)$$

where C' and C'' are constants. Therefore, the UV-to-blue ratio in the high-doping regime is given by

$$\frac{I_{UV}}{I_{blue}} = \frac{I_{UV}}{I_{(a)}} = \frac{Bnp}{C_1 n N_T^0} \propto \frac{\alpha_D}{C''} (N_D)^{1-i}. \quad (9)$$

In Fig. 3, our experimental results show that the UV-to-blue ratio is a constant in the high-doping regime. Therefore, the exponent in Eq. (9) is given by $1-i=0$ or $i=1$. This implies that the deep level concentration increases linearly with the doping concentration. The dependence of $N_T^- \propto N_D$ is the typical signature of compensating native defects.¹⁰ Furthermore, by inserting $i=1$ into Eq. (6), the UV-to-blue ratio in the low doping regime becomes

$$\frac{I_{UV}}{I_{blue}} \propto \frac{\Delta n}{CN_D} \propto \frac{1}{N_D}. \quad (10)$$

That is, the UV-to-blue ratio is inversely proportional to the doping concentration. This is fully consistent with our experimental result shown in Fig. 3, which shows that the UV-to-blue ratio rapidly increases as the silane flow rate decreases. Especially for the undoped $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$, the UV-to-blue ratio reaches as high as 3000.

The above model shows that an acceptorlike compensating native defect is the likely origin of the parasitic blue emission. The parasitic blue emission in AlGaIn thus has the same physical origin as the yellow luminescence in GaN.¹⁰ In order to suppress the parasitic blue emission, the defect density needs to be reduced. The dependence of $N_T^- \propto N_D$ implies that one may reduce the compensating native defect density by reducing the Si doping concentration. However, due to the large donor ionization energy and the self-compensation effect, a reasonably high Si doping concentration is required to reach good n -type conductivity in AlGaIn.¹⁵ Therefore, it is not a practical choice to suppress the parasitic blue emission by reducing the Si doping con-

centration. An alternative choice to suppress the parasitic blue emission would be reducing the dislocation density, especially the edge dislocations. In their study of yellow luminescence in GaN, Elsner *et al.*¹⁶ showed that the most favorable position for gallium vacancy-oxygen complexes is at the edge dislocation core. The complexes are electrically active and increase the intensity of the yellow luminescence in GaN. Our study of the dependence of the parasitic blue emission in AlGaIn samples on the edge dislocation density is currently underway. The preliminary results indicate that parasitic blue emission can indeed be strongly reduced or even suppressed by reducing the edge dislocation density.

In summary, the UV-to-blue luminescence ratio in n -type $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$ grown by MOVPE is investigated as a function of silane flow rate. Experimental results show that in the high doping regime, the UV-to-blue ratio is almost unchanged even though the silane flow rate changes over more than one order of magnitude. However, when the silane flow rate is lower than 1.13×10^{-9} mol/min, the UV-to-blue ratio increases rapidly as the silane flow rate decreases. A theoretical model is developed, which reveals that the parasitic blue emission originates from acceptorlike compensating native defects.

Support from Crystal IS, DOE, ARO, NSF, Samsung Advanced Institute of Technology, Sandia National Laboratories, and New York State is gratefully acknowledged. The authors would like to acknowledge valuable contributions of Dr. Y. A. Xi of GE Global Research Center (Schenectady, NY).

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