Intensity dependence of photoluminescence in GaN thin films

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We report the intensity dependence of band-gap and midgap photoluminescence in GaN films grown by electron cyclotron resonance (ECR) microwave plasma-assisted molecular beam epitaxy. We find that the band-gap luminescence depends linearly while the midgap luminescence has a nonlinear dependence on the incident light intensity. These data were compared with a simple recombination model which assumes a density of recombination centers 2.2 eV below the conduction band edge. The concentration of these centers is higher in films grown at higher microwave power in the ECR plasma.

Gallium nitride (GaN) is a wide direct band-gap semiconductor ($E_g = 3.4$ eV at 300 K), which is currently being investigated in many laboratories for its potential in optical devices (light emitting diodes, lasers, detectors) operating in the blue-violet ultraviolet part of the electromagnetic spectrum.[1] The performance and reliability of such devices depends critically on the type and concentration of electronic defects, whose origin is the heteroepitaxial growth (misfit dislocations and polarity related defects), the formation of native defects (vacancies, interstitials, and antisite defects) and the incorporation of intentional and unintentional impurities.

Defects in semiconductors can be studied by a variety of techniques, principal among which is photoluminescence (PL), a method which has been used extensively for the study of GaN films.[2,3] Photoluminescence spectra of undoped GaN films generally show a sharp peak close to the energy gap of the semiconductor, attributed to excitons bound to shallow donors[3] and a broader peak centered around 2.2 eV.[4,5] Various models have been advanced to account for the high shallow donor concentration in unintentionally doped GaN. The prevailing view is that the donors are due to nitrogen vacancies[6] although there is some evidence that oxygen impurities can also act as substitutional donors.[7] The broad yellow luminescence was attributed to electron transitions from the conduction band to a band of deep acceptor states placed 860 meV above the valence band edge.[8] These authors presented arguments that such states are introduced by carbon impurities in the films while Pankove *et al.*,[4] who observed the same photoluminescence band in ion implanted GaN samples, attributed the deep states to ion implantation processes. In general, the existence and magnitude of the yellow luminescence is associated with defective GaN films and the ratio of the yellow luminescence peak to band-gap luminescence was employed as a criterion of the carbon doping effects on the films[8] or the ion implantation related damage.[4]

In this letter, we investigated the excitation intensity dependence of PL from undoped GaN films and found that the band-gap luminescence depends linearly while the yellow luminescence has a nonlinear dependence on the light intensity. Thus the use of the ratio of yellow to band-gap luminescence as a criterion of the quality of the GaN films is meaningless unless one specifies the magnitude of the employed excitation light intensity. A simple recombination model has been proposed to qualitatively account for the observed light intensity dependence of the two luminescence bands.

The GaN films used in this study were grown by the electron cyclotron resonance microwave plasma-assisted molecular beam epitaxy (ECR-MBE), which was described in detail in a number of recent papers.[9,10] Here, we present only a brief description of the growth process. All the films were deposited on (0001) sapphire substrates, whose surface after chemical cleaning and thermal outgassing, is converted to AlN by exposing it to an ECR activated nitrogen plasma.[9] The films were deposited by the two-step growth process in which a GaN buffer of about 300 Å thick is deposited at 500 °C and the rest of the film, 1 to 2 μm thick, is deposited at 800 °C. This two-step growth method was found to lead to a low two-dimensional nucleation rate and a high lateral growth rate leading to films with relatively smooth surface morphology.[9,10] Power in the ECR discharge was the main variable during the growth of the films reported in this letter. More specifically type I samples were grown at a total microwave power of 20 W while type II samples were grown at 35 W. The transport coefficients of two representative samples are shown in Table I.

The photoluminescence was excited by a pulsed N_2 laser as the excitation source. The laser has a photon energy of 3.678 eV (337.1 nm), pulse, width of 10 ns, repetition rate of 40 Hz and a listed peak power of 250 kW. A circular aperture was used in front of the rectangular collimated beam and the resulting output was focused on the sample using a lens. A set of neutral density filters was used to attenuate the incident laser intensity for studying the excitation dependence of PL from the sample. The sample was held on a cold finger with a colloidal suspension of graphite which also acts as a

TABLE I. Typical values for the room temperature carrier concentration and mobility for representative type I and type II samples.

Sample	Power (W)	Carrier conc. (cm^{-3})	Mobility ($cm^2 V^{-1} s^{-1}$)
Type I	20	1.0×10^{18}	50
Type II	35	1.90×10^{17}	13.8

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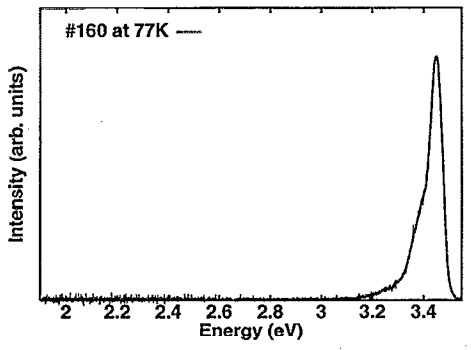

FIG. 1. PL spectra from a type I sample.

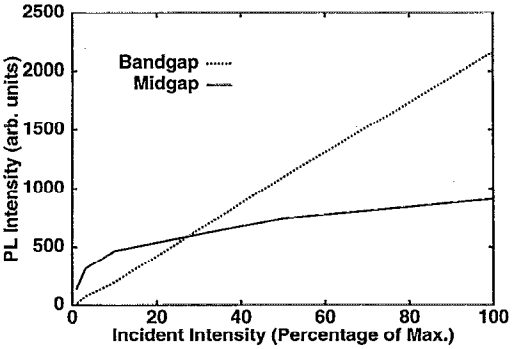

FIG. 3. Plot of the PL intensity vs the percentage excitation for the band-gap and midgap luminescence.

good heat conductor, hence preventing the sample from being heated by the laser. PL was collected by a lens and focused on the entrance slits of a 0.25-m grating spectrometer. A Hamamatsu photomultiplier (R-928) was used as the detector and its output was read by a lock-in amplifier. The spectra were not corrected for the spectral response of the system.

Figure 1 shows the photoluminescence spectrum at 77 K for a type I GaN sample. This spectrum shows only the band-gap luminescence at 3.47 eV. At the maximum excitation intensity we see no evidence of yellow luminescence, suggesting that the concentration of midgap defects is very low. The band-gap luminescence was found to vary linearly with light intensity over two orders of magnitude, a result consistent with excitonic recombination.[11]

The photoluminescence spectra of a type II GaN sample measured at 100% and 1% of the incident laser light is shown in Fig. 2. It is apparent from these data that characterizing the quality of the material by the ratio of the magnitude of the band-gap luminescence to the midgap luminescence is not correct, since the ratio depends on the excitation intensity. Thus, it is quite important to take the excitation intensity into account when analyzing any PL spectra for this material.

Figure 3 illustrates the intensity dependence of the band-gap and midgap luminescence peaks of the type II GaN sample. The band-gap luminescence depends linearly on light intensity as for type I samples. However, the midgap luminescence was found to have a light intensity dependence as shown in the figure. It initially increases with light intensity at low excitation levels and then tends to saturate as the excitation intensity is increased further.

The observed dependence on light intensity can be qualitatively accounted for in the simple recombination model illustrated in Fig. 4. We assume that the states responsible for the yellow luminescence form a broad band of a total density $N_{1\max}$. Shown in the figure are also the lifetimes for the various recombination paths and the densities N_1 and N_2 of the occupied states in the defect and conduction bands, respectively. If G ($\mathrm{cm^{-3}\,s^{-1}}$) is the generation rate then the rate of change of N_1 and N_2 are given by the equations

$$\frac{dN_2}{dt}=G-\frac{N_2}{\tau_{20}}-\frac{N_2}{\tau_{21}},\quad \frac{dN_1}{dt}=\frac{N_2}{\tau_{21}}-\frac{N_1}{\tau_{10}}. \tag{1}$$

Under steady-state conditions, i.e., when the recombination lifetimes are smaller or comparable to the duration of the excitation pulse, Eqs. (1) become

$$G=\frac{N_2}{\tau_{20}}+\frac{N_2}{\tau_{21}},\quad \frac{N_2}{\tau_{21}}=\frac{N_1}{\tau_{10}}. \tag{2}$$

The band-gap luminescence (I_{gap}) and the midgap luminescence (I_{yel}) are given by the expressions:

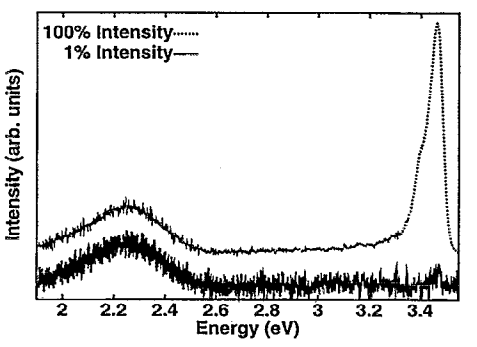

FIG. 2. PL spectra from type II samples at two different excitation levels.

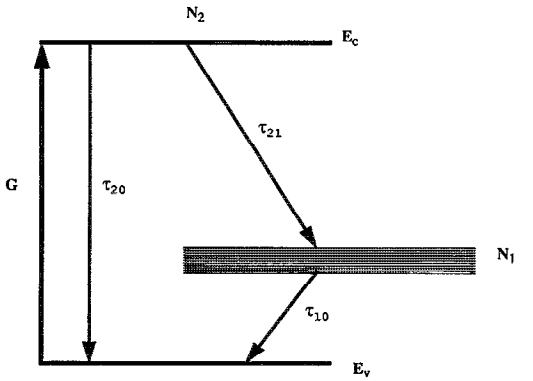

FIG. 4. Schematic of the proposed recombination model.

$$I_{\text{gap}} \propto \frac{N_2}{\tau_{20}} = \frac{G\tau_{21}}{(\tau_{21}+\tau_{20})}, \quad I_{\text{yel}} \propto \frac{N_2}{\tau_{21}} = \frac{G\tau_{20}}{(\tau_{21}+\tau_{20})}. \tag{3}$$

We assume that the recombination lifetimes τ_{20} and τ_{10} are constants while τ_{21} depends on light intensity. More specifically the value of τ_{21} can be expressed in terms of the low light intensity recombination lifetime τ'_{21} as[11]

$$\tau_{21} = \frac{N_{1\max}}{N_{1\max} - N_1}\,\tau'_{21}. \tag{4}$$

From Eqs. (3) and (4) we get

$$I_{\text{gap}} \propto \frac{GN_{1\max}\tau'_{21}}{N_{1\max}\tau'_{21} + (N_{1\max} - N_1)\tau_{20}},$$
$$I_{\text{yel}} \propto \frac{G\tau_{20}(N_{1\max} - N_1)}{N_{1\max}\tau'_{21} + \tau_{20}(N_{1\max} - N_1)}. \tag{5}$$

Case I: Low light intensity, i.e., $N_1 \ll N_{1\max}$

$$I_{\text{gap}} \propto \frac{G\tau'_{21}}{\tau_{20}+\tau'_{21}}, \quad I_{\text{yel}} \propto \frac{G\tau_{20}}{\tau_{20}+\tau'_{21}}. \tag{6}$$

Hence, in this case the yellow and the band-gap luminescence increase proportionally to the incident light intensity.

Case II: High light intensity, i.e., $N_1 \cong N_{1\max}$, the first of Eqs. (5) gives

$$I_{\text{gap}} \propto G. \tag{7a}$$

On the other hand, the yellow luminescence is governed by the recombination time, τ_{10}. Thus from Eqs. (2) and (3),

$$I_{\text{yel}} \propto \frac{N_{1\max}}{\tau_{10}} \rightarrow \text{Const.} \tag{7b}$$

Equations (6) and (7) account qualitatively for the experimental results for Fig. 3.

Although this model does not take into account the change in the number of traps and recombination centers due to the shift in the quasi-Fermi levels as excitation intensity is changed,[12] it is sufficient to explain the saturation of the yellow PL from GaN. It would help further to have information about the recombination lifetimes of carriers to compute the exact number of midgap states.

In conclusion, we have determined the light intensity dependence of the band-gap and midgap luminescence in GaN films grown by electron cyclotron resonance assisted molecular beam epitaxy. As expected the band-gap luminescence varies linearly with the light intensity but the midgap luminescence initially increases and then saturates at higher light intensities. The data show the importance of relating $I_{\text{yel}}/I_{\text{gap}}$ to the generation rate employed during the measurement for the purposes of comparing samples on the basis of PL spectra. It further illustrates that samples grown at higher microwave power have higher concentration of midgap defects, which accounts for the observed compensation in the transport coefficients.

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