Photoluminescence and recombination mechanisms in GaN/Al$_{0.2}$Ga$_{0.8}$N superlattice

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(Received 10 December 1999; accepted for publication 7 February 2000)

A detailed study of photoluminescence (PL) of GaN(1 nm)/Al$_{0.2}$Ga$_{0.8}$N(3.3 nm) twenty periods superlattice grown via metal-organic chemical vapor deposition is presented. The dependence of the PL emission energy, linewidth, and intensity on temperature, in the low temperature regime, is consistent with recombination mechanisms involving bandtail states attributed to a small degree of interfacial disorder. The activation energy of the nonradiative centers in our superlattice agrees well with the value we derive for the width of the tail-state distribution. Moreover, we find that the average phonon energy of the phonons that control the interband PL energy at high temperatures is larger for the superlattice than for a high-quality GaN film. This observation is consistent with model calculations predicting the phonon mode properties of GaN–AlN-based wurtzite heterostructures. © 2000 American Institute of Physics. [S0003-6951(00)00915-3]

Anomalous temperature dependence of the photoluminescence (PL) emission energy from InGaN quantum wells that does not follow at low temperatures, the accepted Varshni$^1$ or Bose–Einstein$^{2,3}$ type behavior of a semiconductor band gap has been reported.$^4$ Such behavior has been observed as well in quantum wells and superlattices of the zincblende structure.$^{5-8}$ The PL energy, instead of being approximately constant at low temperatures, exhibits a red-shift followed by a blue-shift. In the above studies the anomalous dependence was attributed to the effect of the interface defect density of states (DOS), on the PL energy.

The presence of defect density in an heterostructure device is crucial to the device performance, especially in structures where multiple interfaces exist. In this letter, we present a detailed study of the photoluminescence from a GaN/Al$_{0.2}$Ga$_{0.8}$N superlattice of wurtzite structure. We establish the presence of a defect density of states, which controls the PL energy at low temperatures and acts as a nonradiative channel to the PL at higher temperatures. Moreover, the electron–phonon interaction that affects the PL at higher temperatures is stronger in the superlattice than in a GaN film indicating the contributions of phonons of higher energies.

The luminescence measurements were acquired utilizing the 244 nm (5.08 eV) line from the second harmonic generation of the 488 nm line of Ar ion Coherent laser and the Triplemate ISA monochromator in conjunction with a liquid nitrogen-cooled charge-coupled device (CCD) detector. The cold stage consists of a closed-cycle cryostat, which enables measurements from 10 K up to room temperature. The superlattice consists of twenty periods of (1 nm)GaN/(3.3 nm)Al$_{0.2}$Ga$_{0.8}$N grown on 2 μm GaN with a 25 nm GaN buffer layer on a (0001) sapphire substrate. The nitride materials described in this work are grown by metal-organic chemical vapor deposition (MOCVD) in an EMCORE D125 UTM rotating-disk reactor. The growth temperature is $\approx$1050 °C for the thick undoped GaN layer and the Al$_{0.2}$Ga$_{0.8}$N superlattice layers.

In the following, we present our study focusing on the photoluminescence behavior as a function of temperature of the GaN/Al$_{0.2}$Ga$_{0.8}$N superlattice. Figure 1 depicts the PL spectrum of the superlattice (SL) acquired at 10 K. The peak at 3.795 eV with a full width at half maximum of 32 meV is attributed to the emission from the GaN wells. The inset shows the PL intensity as a function of the laser excitation power.

FIG. 1. The PL from the superlattice at 10 K. The peak at 3.795 is due the emission from the GaN wells. The inset shows the PL intensity as a function of the laser excitation power.
layer, as well as a weak peak at \( \sim 3.7 \text{ eV} \), the origin of which will be addressed in a later study. The inset to Fig. 1 depicts the intensity of the luminescence from the superlattice as a function of excitation intensity at 10 K. A linear relation between the two intensities was ascertained which is indicative that the PL emission of the SL at 10 K is due to excitonic recombination. Figure 2 shows the luminescence peak position as a function of temperature compared to that of a high quality GaN thin film. The inset in Fig. 2 plots the PL linewidths of the superlattice sample.

It has been established that the temperature dependence of an interband transition energy, \( E(T) \), in semiconductors can be approximated via the relation:

\[
E(T) = E(0) - \frac{2\alpha}{\exp(\Theta/T) - 1}.
\]  

(1)

This relation represents the modification of a band gap of a semiconductor due to the electron–phonon interaction at thermal equilibrium, and may be derived from the Bose–Einstein approximation for the lattice vibration energy: \( \langle E \rangle = \langle 1/2 + (n) \rangle \hbar \omega \), where \( (n) \) is the average phonon occupation number. In Eq. (1), \( E(0) \) is the transition energy at 0 K, \( \alpha \) is a measure of the strength of the electron–average phonon interaction, and \( \Theta \) corresponds to a mean temperature value of the longitudinal acoustic as well as of optical phonons taking place in the interaction.\(^{2,3,10–12}\) The average phonon temperature \( \Theta \) is related to the average phonon energy \( E_p \) via the Boltzmann constant \( k_B \): \( \Theta = E_p/k_B \). The model presented in Eq. (1) exhibits a good fit to our PL experimental data of the GaN film, as can be seen in Fig. 2. The fit resulted in the following values: \( E(0) = 3.4866 \text{ eV} \), \( \alpha = 55 \text{ meV} \), and \( \Theta = 309 \text{ K} \) and thus \( E_p = 27 \text{ meV} \). These values agree with the ones obtained previously for GaN films via the same model utilizing optical absorption\(^{13}\) and contactless electroreflectance.\(^{10}\)

As can be observed in Fig. 2 the temperature dependence of the PL emission of the GaN/Al\(_{0.2}\)Ga\(_{0.8}\)N superlattice does not follow the monotonic variation of Eq. (1) for the band edge to lower energies with increasing temperature. Similar behavior has previously been observed in the quantum well structures of AlGaN/InGaN/GaN,\(^4\) InGaAs–InP,\(^5\) and GaInAs/AlInAs,\(^6\) as well as in superlattices of ZnSe/MgS\(^7\) and GaAs/AlAs.\(^8\) This anomalous emission characteristic has been attributed to the presence of density of states (DOS) which is due to a certain degree of disorder occurring mainly at interfaces, which may be of compositional and/or structural origin.\(^5\) An explanation of the anomalous behavior of the excitonic luminescence was given in Ref. 5 in terms of this disorder; we apply this explanation to our results presented in Fig. 2. As the temperature rises from 10 to 50 K, the excitons gain just enough thermal energy to become trapped at the lower energy levels of the DOS, which results in the red-shift of the PL emission. From 50 K, a thermal equilibrium is achieved and the excitons populate the higher levels of the DOS (blue-shift) up to a maximum energy level, in this case \( \sim 130 \text{ K} \) corresponding to an emission energy of 3.8 eV. From that point on, a “normal” band gap emission takes over which can be described in terms of Eq. (1).

A model has been recently proposed to explain the blue temperature-induced shift in InGaN-based quantum well luminescence.\(^4\) The model is based on band-tail filling of a Gaussian DOS with a parameter \( \sigma \) which describes the dispersion of the DOS (i.e., its width). The model calculation is given by:

\[
E(T) = E_D - \frac{\sigma^2}{k_B T},
\]  

(2)

where \( E_D \) is attributed to the energy range between the center of the Gaussian DOS of the electrons and that of the holes. In addition, the model assumes that \( E_D \) depends on temperature in the same manner as the gap energy, and thus \( E_D \) can be described via Eq. (1). This model is valid in the temperature range where thermal equilibrium has been established: in our case \( \sim 50 \text{ K} \) and above. We use this model to analyze our data in order to estimate the extent of the DOS in the superlattice; the fit is presented in Fig. 2. We obtain the value for \( \sigma \) to be \( \sim 11 \text{ meV} \), which is a relatively small compared to the value of the superlattice emission (\( \sim 3.8 \text{ eV} \)) indicating the high quality of the SL interfaces. Additionally, the inset to Fig. 2 shows the PL linewidth as a function of temperature. The line width characteristic can also be separated into three different temperature regimes similar to those observed for the PL emission; the broadening mechanisms will be addressed at a later study. Equation (2) yields the values of the average electron–phonon interaction strength \( \alpha = 135 \text{ meV} \) and of the mean phonon temperature \( \Theta = 573 \text{ K} \), the latter which corresponds to a mean phonon energy \( E_p = 50 \text{ meV} \). This higher value of \( E_p \) obtained for the SL relative to that of GaN (\( E_p = 27 \text{ meV} \)) may be explained in view of the model calculations developed for polar-optical phonons in GaN/AlGaN-based quantum wells of wurtzite structure.\(^{14,15}\) In that study, Komirenko et al. have shown that phonon modes in the heterolayers of GaN–AlGaN-based wurtzite heterostructures can exhibit the higher energies of the modes of the AlGaN material.\(^{14,15}\) In light of the above
model, the higher average phonon energy, $E_p$, found in our GaN wells are expected based on the predicted phonon mode structure in GaN/Al$_{0.2}$Ga$_{0.8}$N barriers.

In order to gain further insight into the mechanisms determining the recombination dynamics of the superlattice, the luminescence intensity, $I(T)$, as a function of temperature was investigated. Figure 3 presents the experimental data of that study and the curve fit to the data obtained from the relation which describes the PL intensity in the presence of a nonradiative channel with thermal activation energy $E_A$:

$$\frac{I(T)}{I_0} = \frac{1}{1 + C \exp(-E_A/k_BT)}.$$  

(3)

In this relation $C$ is a constant and $I_0$ is the PL intensity at zero absolute temperature. An activation energy $E_A = 16$ meV was ascertained from the fit. This value is in the same order of magnitude and agrees with that obtained for the energy width of the DOS (11 meV), the small difference being attributed to an experimental error and the approximate nature of the above models. Thus at higher temperatures the tail states, due to interface disorder, are the nonradiative centers in the superlattice emission.

The temperature dependence of the luminescence blue shift in InGaN based quantum wells has been discussed in terms of the screening of the piezoelectric field. In that model, as the temperature increases free carriers are thermally activated which in turn causes the screening of the field. As a result, modification of the band gap takes place that manifests itself in the blue-shift of the emission. Although this effect may be a possible additional mechanism causing the temperature dependent blue-shift of the luminescence, it does not explain the initial red-shift of the emission energy. As mentioned previously, similar luminescence behavior has been observed in the zincblende III–V quantum wells which have considerably smaller piezoelectric fields. We do not however preclude the screening effect of the PEF as an additional mechanism for the blue shift of the PL.

In conclusion, the temperature behavior of the luminescence in a GaN/Al$_{0.2}$Ga$_{0.8}$N superlattice was investigated. Our analysis verifies the existence of a defect density of states, in the recombination of excitons at low temperatures. We also find that at high temperatures, the density of states acts as a nonradiative channel for the luminescence. The higher value of the average energy of the phonons involved in the superlattice scattering is consistent with the predictions for the phonon modes of GaN/AlGaN heterolayers.

L.B. acknowledges the Army Research Office via a National Research Council postdoctoral associateship for supporting this research. The work at UT-Austin was partially supported by the Office of Naval Research under N00014-95-1-1302 (monitor J. C. Zolper) and the NSF STC program under Grant No. CHE-89-20120.