Optimizing Light Emission from Nitride Quantum Wells

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Abstract

Time resolved and CW excitation dependent photoluminescence measurements of InGaN and AlInGaN quantum wells are presented. Contributions of piezoelectricity, phase segregation, and spontaneous polarization on the emission efficiency of III-N heterostructures are discussed.

INTRODUCTION

The development of UV and visible light emitting diodes and lasers continue to be of considerable technological interest. Significant progress has been made in the development of LEDs with emission from the UV to the red. Recently, there have been reports of high efficiency green LEDs fabricated from GaN based materials. Moreover, AlGaN/InGaN/GaN UV and blue lasers with long lifetimes are commercially available but remain expensive. Much research interest has focused on three mechanisms affecting device performance: phase-segregation resulting from In clustering in InGaN alloys, piezoelectricity in InGaN resulting from the lattice mismatch between InGaN and the GaN substrate, and spontaneous polarization of AlGaN resulting from the asymmetry in the tetrahedral sub-structure of the wurtzite lattice. It is of significant technological importance to determine the relative contribution of each of these processes to the emission efficiencies in these nitride materials. We feel that further advances in these materials that will result in the improvement of device operation and the development of new light sources, e.g., demonstrating a green laser, requires a better understanding of the emission processes and carrier dynamics in these materials.

In this abstract, we present comparative time-resolved and CW photoluminescence measurements of the emission from InGaN/GaN, InGaN/AlGaN, and AlInGaN/AlGaN heterostructures. The first studies presented are focused on understanding the carrier dynamics associated with piezoelectricity and indium segregation. This first study was limited to InGaN quantum wells with GaN barriers in which spontaneous polarization can be neglected. Time-resolved photoluminescence measurements of these structures were performed to ascertain the magnitude of the energy shifts and carrier lifetimes of these structures. The magnitude of the observed energy shifts can be explained by phenomenological models that include only piezoelectricity. However, consistent with indium segregation, quantum wells with the same nominal indium concentrations and well width are observed to have drastically differing peak emission energy and linewidths.

The second study presented was focused on understanding the effects of spontaneous polarization and indium segregation. In these studies, we performed complementary spectroscopic studies of an InGaN single quantum well (SQW) with AlGaN barriers and a strain engineered AlInGaN SQW with AlGaN barriers. For these samples, spontaneous polarization, resulting from the AlGaN barriers, must be included in any models. However, the in-well strain in the strain engineered AlInGaN SQW is significantly reduced by a careful match of the lattice constant of the substrate and well, therefore the piezoelectric field can be neglected. Excitation dependent CW photoluminescence was used to optically probe these structures. The results of these studies imply that phase segregation produces regions of radiative recombination centers that can be optically saturated on the low energy (long wavelength) side of the observed emission with long carrier lifetimes.

PIEZOELECTRICITY AND INDIUM SEGREGATION

The observation of piezoelectricity in Wurtzite $In_xGa_{1-x}N$/GaN heterostructures [1-5] has changed our understanding of the behavior of these materials under optical or electrical excitation. It has been reported that these piezoelectric fields are quite large (reported between 300 kV/cm to 1.1 MV/cm for 20 % indium) [2,3]. Additionally, the in-well piezoelectric field has been demonstrated to point toward the substrate for materials grown on a c-plane Sapphire substrate [3]. In samples that are grown with n^+ GaN substrates, the built-in electric field of a *p*-*i*-*n* structure can partially compensate for the induced piezoelectric field. For piezoelectric materials embedded within a *p*-*i*-*n* structure the results from previous detailed studies of piezoelectricity in (111) grown semiconductor materials [6,7,8] are used to calculate the electric field within the quantum well. These calculations provide the magnitude of the in-well and barrier fields and the resulting band-structures of InGaN/GaN *p*-*i*(MQW)-*n* samples in dark conditions and under optical excitation. Once the band structure is known, the quantum confined energy levels and quantum confined Stark shift can be readily calculated using a variational technique. The details of these field calculations for III-N quantum wells and the accompanying spectral shifts have been reported elsewhere [5]. These calculations can not always accurately predict the behavior of these materials, especially where differences in growth conditions drastically affect the homogeneity of the quantum well material.

As stated earlier, one interest of this work is to investigate the contribution of indium segregation. For our first study we have chosen two samples with similar well widths and indium concentrations grown under different conditions. Because the piezoelectric field is a function of lattice mismatch, these samples, with similar indium concentrations, are expected to have very similar in-well piezoelectric fields. Both samples, which were provided by Xerox-Palo Alto Research Center, were grown on a c-face (0001) sapphire substrate using organometallic vapor phase epitaxy (OMVPE). Initially, a 4 micron thick layer of GaN:Si was deposited. This layer was followed by growth of the GaN barrier followed by a 2 nm thick $In_{.2}Ga_{.8}N$ quantum well region. This barrier and quantum well region was repeated to achieve the desired number of quantum wells (2 and 20). The last quantum well in each sample was capped with a GaN barrier region followed by a GaN:Mg p-contact layer. For simplicity we refer to these samples as M2 and M20.

We performed detailed temperature and excitation dependent time-resolved photoluminescence (TRPL) measurements on these samples. For the time-resolved photoluminescence, the pump source was the frequency doubled output from a tunable mode-locked Ti:Sapphire laser that typically produced 40 mW of ~200 fs pulses with a repetition rate of 80 MHz tunable from 360 to 420 nm. The tunability of the Ti:Sapphire laser allowed the wavelength of the incident light to be varied so that we could effectively pump directly within the quantum well without exciting the higher bandgap barrier material. The resulting photoluminescence was spectrally- and temporally-resolved using a Hamamatsu streak camera with a 20 ps response time. In order to maximize the intensity of the photoluminescence and to narrow the excitonic emission, all time-resolved data presented here were acquired at a temperature of 15K.

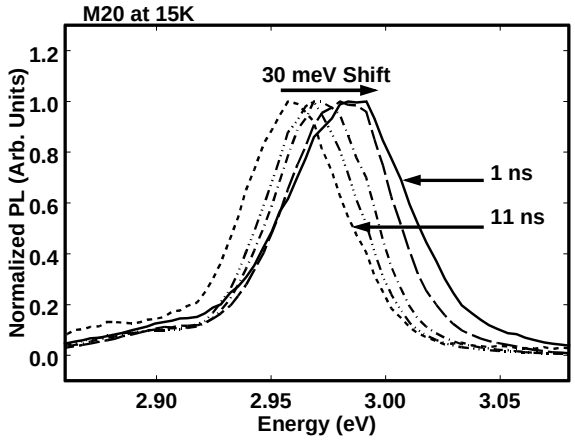

Figure 1. Time-resolved PL of M20 at 15K shows a 30 meV shift towards higher energy at short times. Peak emission energy is consistent with expected value from a quantum mechanical model.

Normalized emission spectra at various time delays following the excitation pulse are shown in Figure 1 for sample M20. A few things are worth noting. Clearly, the emission spectra blue-shifts approximately 30 meV after each optical pulse. At short times after the pump excitation, the field within the quantum well is screened by the excess carriers generated within the quantum wells. This reduction

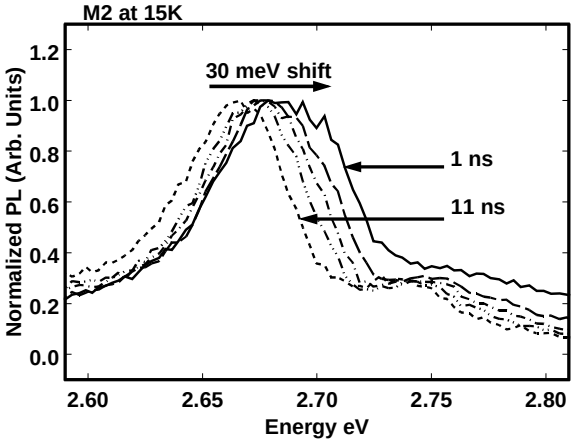

Figure 2. Time-resolved PL of M2 at 15K shows a 30 meV shift towards higher energy at short times. Peak emission energy is 300 meV below the expected emission energy using a quantum mechanical model.

of in-well field results in a shift in the emission energy through the quantum confined Stark effect. As time increases, the carriers recombine and the screening decreases. This decrease in screening is evidenced by the shift in the excitonic emission toward lower energies at later times. In addition, it should be noted that the linewidths of the emission remains relatively constant for all delay times.

We have performed the identical TRPL experiment on sample M2 with typical emission spectra at various delay times shown in Figure 2. Although, M2 shows an identical blue-shift of approximately 30 meV consistent with a similar piezoelectric field, the peak PL intensity is located some 300 meV lower in energy that that of sample M20. In addition, the observed linewidth of the M2 is two times larger than the linewidth of M20.

A quantum mechanical calculation of the expected excitonic emission within the well is in excellent agreement with the peak emission energy of sample M20. However, the peak emission energy from sample M2 suggests that sample M2 is not behaving as a traditional quantum well at all. Moreover, emission at this energy would require either a much higher indium concentration or the presence of quantum "dots" due to phase segregation [9,10]. The indium concentration within the quantum wells of M2 and M20 have both been measured using X-ray diffraction and estimated to be approximately 20 %. Moreover, sample M2 shows much longer carrier lifetimes than sample M20 (> 10 ns for M2 compared to 5 ns for M20). These increased lifetimes are also consistent with stronger confinement due to localization of carriers in nanometer scale regions [11,12,13].

STRAIN ENGINEERING TO OPTIMIZE LONGER WAVELENGTHS

Indium segregation and the corresponding quantum dot formation may successfully push the wavelength of emission to long wavelengths but, so far, in these III-N materials it simultaneously increases the emission linewidth. These large linewidths are consistent with a non-uniform distribution of indium concentration and fluctuations in indium rich region size within these materials. An alternative method to produce longer wavelength emitters is to use high concentrations of indium in the wells and to somehow decrease the formation of these indium rich regions. At high indium concentrations the large lattice mismatch between InGaN and GaN produces a biaxial compressive strain, this strain increases proportionally to the indium concentration and results in the aforementioned piezoelectric field. Consequently, threading dislocations easily propagate in these wells and provide non-radiative recombination centers for carriers and reduce the overall efficiency of the device. Recently, much attention has been placed on reducing the strain in these materials through the introduction of aluminum. Specifically, quaternary well materials of AlInGaN can be engineered (through the appropriate choices of Al and In concentrations) to produce completely lattice matched quantum wells, therefore allowing

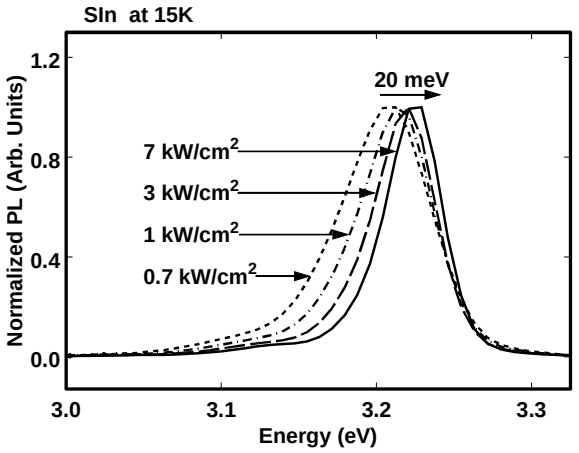

Figure 3. Time-integrated PL of SIn (InGaN/AlGaN) at 15K shows a 20 meV Stark shift and a reduction in emission linewidth, predominantly on the low energy side, as excitation pump power is increased.

for the incorporation of much higher indium concentrations and drastically reduce defect dislocations [14].

We have performed a comparative study of a strained single quantum well (SQW) and a strain engineered SQW that were designed to emit at the same energy. The samples, provided Bedair's group at North Carolina State University, were grown on Sapphire substrates with a buffer layer of GaN. Sample SIn (SQW of InGaN) has a 9nm $In_{0.08}Ga_{0.92}N$ quantum well with a 35nm cap layer of $Al_{0.15}Ga_{0.85}N$ and a 120nm $Al_{0.15}Ga_{0.85}N$ barrier that was grown directly on the GaN base. Similarly, sample SAl (SQW of AlInGaN) has a 9nm $Al_{0.18}In_{0.08}Ga_{0.74}N$ quantum well with a 35nm $Al_{0.15}Ga_{0.85}N$ cap layer and a 120nm $Al_{0.15}Ga_{0.85}N$ barrier grown directly on the GaN base. The $Al_{0.15}Ga_{0.85}N$ forces us to include the spontaneous polarization effects produced by the introduction of aluminum [15,16]. Moreover, because the corresponding in-well spontaneous field is so large compared to any piezoelectric field (estimated to be 3 MV/cm compared to 300 kV/cm for these particular structures), we can neglect the effects of the piezoelectric field in the InGaN SQW sample.

These samples were optically probed using CW excitation dependent photoluminescence measurements performed at 15K. In these measurements, the pump source for the time-integrated photoluminescence was a multi-line UV Argon ion laser. The resulting photoluminescence from the sample was collected and focused onto the entrance slit of a computer controlled scanning monochromator. The output from the monochromator was detected using a photodiode and standard lock-in techniques, to improve signal to noise ratio.

The normalized time-integrated emission spectra are plotted as a function of excitation power for sample SIn in Figure 3. Here, as the pump power is increased the emission spectra blue-shifts approximately 20 meV. This Stark shift is

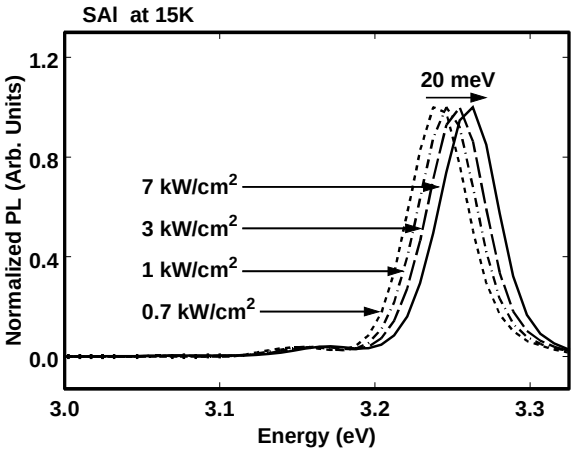

Figure 4. Time-integrated PL of SAl (AlInGaN/AlGaN) at 15K shows a 20 meV Stark shift and no reduction in emission linewidth as excitation pump power is increased.

similar to the one observed in sample M2 and M20 and results from excess photogenerated carriers screening the field within the quantum well. In addition, the linewidth of the emission is narrowed as the excitation power is increased. More importantly, the line narrowing is predominantly on the low energy side of the emission. We believe that this behavior is consistent with the optical saturation of indium rich regions resulting from indium segregation.

As shown in Figure 4, sample SAl shows an identical 20 meV shift in peak emission, but the reduction in linewidth is clearly absent. More importantly, the emission linewidth of both samples is comparable at high excitation powers. Narrower linewidths are generally associated with higher quality material (more homogenous). These two samples are nominally identical except for the presence of the Al. This improvement in crystalline quality, and subsequent reduction of linewidth, can be explained by a selective substitution of Al for In that could reduce the In clustering.

SUMMARY

These studies are consistent with phase segregation producing regions of radiative recombination centers at lower emission energies. In other words, in InGaN the presence of random fluctuations of indium rich regions, which reduces crystalline quality, provides an efficient alternate, radiative, trap for carriers in the quantum well [9,10].

Improving device efficiency and optimizing longer wavelength emitters requires a detailed understanding of the three mechanisms affecting device performance: phase-segregation, piezoelectricity, and spontaneous polarization. Each of these contributes to changes in emission energy, linewidth, and the lifetimes of the carriers. The development of quaternary structures (with varying well widths, indium concentration, etc.) that maximize the carrier lifetimes and emission efficiency for laser and LED applications should result in better long wavelength devices. Moreover, the development of composite structures that can accurately control the in-well field should result in device geometries for visible spatial light modulators for the visible wavelength range.

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