Multisubband photoluminescence in \( p \)-type modulation-doped \( \text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN} \) superlattices

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Photoluminescence spectra from \( p \)-type modulation-doped \( \text{Al}_{0.20}\text{Ga}_{0.80}\text{N}/\text{GaN} \) superlattices with 10-nm well width show multiple, well resolved, interband transitions between quantum-confined states. In addition to the ground-state transition, a number of excited-state transitions are observed. The observation of multiple peaks is attributed to the inverse dependence of subband population and oscillator strength on energy. The relative strength of the peaks strongly changes with excitation intensity. At low excitation intensity, the spectra display only the ground-state transition. At higher excitation intensity, excited-state transitions become dominant. At high excitation intensities, the dominant transition occurs at energies about 500 meV above the electron ground-state to hole ground-state transition. Self-consistent calculations are used to assign transition energies, lifetimes, and rates to each photoluminescence line. Theoretical and experimental transition energies are in excellent agreement. We attribute the excellent optical properties to the modulated doping of the structure, which consists of doped barriers and undoped well layers. Our calculations also show an average recombinination lifetime of 50 ns at high excitation intensities, despite the large quantum-confined Stark effect. The changes of the photoluminescence spectra can be explained via the effects of band filling and oscillator strengths at higher excitation intensity.

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The role of spontaneous and piezoelectric fields\(^1\)\(^–\)\(^3\) in the \( \text{Al}_x\text{Ga}_{1-x}\text{N} \) material system continues to receive great interest due to the influence of these fields on both the optical\(^4\)\(^–\)\(^5\) and electronic\(^6\)\(^–\)\(^8\) properties of \( \text{Al}_x\text{Ga}_{1-x}\text{N} \) heterostructures. In particular, as III-nitride optical devices approach the ultraviolet (UV) range, the use of \( \text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN} \) and \( \text{Al}_x\text{Ga}_{1-x}\text{N}/\text{Al}_x\text{Ga}_{1-x}\text{N} \) quantum wells (QW’s) is becoming a necessity. However, the relaxation efficiency of these QW’s has been shown to be very inefficient for wider well sizes due to the spatial separation of the electron and hole wave functions.\(^9\)\(^–\)\(^10\) This separation is due to the strong polarization fields present in \( \text{Al}_x\text{Ga}_{1-x}\text{N} \) QW’s. The luminescence emitted from these QW’s is also strongly redshifted due to the quantum-confined Stark effect (QCSE).\(^11\)

\( \text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN} \) \( p \)-type superlattices (SL’s) are of great interest because of their enhanced \( p \)-type doping properties.\(^12\)\(^–\)\(^13\) Specifically, modulation-doped SL’s demonstrate low-temperature electrical properties superior to any \( p \)-type \( \text{Al}_x\text{Ga}_{1-x}\text{N}/\text{GaN} \) material reported to date.\(^9\) The modulation-doped SL investigated presently, which is the same modulation-doped SL as in Ref. 6, has a resistivity of 0.068 \( \Omega \) cm, a mobility of 36 cm\(^2\)/Vs, and a free hole concentration of 2.5 \( \times \) 10\(^{19} \) cm\(^{-3} \), all measured at 90 K. Because of the effects of superlattice and piezoelectric doping, the free carrier concentration is nearly independent of temperature.

In this work we report the first experimental results on multisubband photoluminescence (PL) spectra of \( p \)-type modulation-doped \( \text{Al}_{0.20}\text{Ga}_{0.80}\text{N}/\text{GaN} \) SL’s. We observe clearly resolved multisubband luminescence peaks whose relative intensity changes with excitation intensity. The luminescence peak energies are compared to the transition energies calculated from a self-consistent Poisson-Schrödinger one-dimensional (1D) solver using the method of finite differences. In addition, spontaneous recombination rates and lifetimes are compared between experiment and theory by using the calculated energies and envelope functions. The theoretical and experimental results agree very well. The oscillator strength and population distribution between subbands have opposite dependencies on energy. As the electronic eigenstate energy increases so does the overlap integral between the electronic eigenstate and the ground hole eigenstate. However, the carrier population in the higher energy electronic subbands decreases. As a result, spontaneous transitions between sparsely populated electronic excited states and the ground hole state become observable at high excitation intensity due to the larger overlap integral between higher-energy electron-hole pair states. These characteristics are due to the large polarization fields present in the SL. The polarization fields also destroy the symmetry of the QW’s and therefore remove all parity selection rules between transitions.

Figure 1 shows a schematic and the self-consistently calculated band diagram of the Ga-faced \( \text{Al}_{0.20}\text{Ga}_{0.80}\text{N}/\text{GaN} \) SL sample under study. The SL samples were grown on a c-plane sapphire substrate by molecular beam epitaxy (MBE). The SL’s are doped with Mg at a level of \( N_{\text{Mg}} \approx 1 \times 10^{19} \) cm\(^{-3} \). One of the SL’s is uniformly doped and the other is modulation doped. The modulation-doped SL has only the barrier layers doped whereas the well layers are undoped. The uniformly doped sample displays one broad PL line that we attribute to electron subband to neutral acceptor transitions. The free holes in the modulation-doped SL
are measured at \( \approx 2.5 \times 10^{18} \text{ cm}^{-3} \) at low temperature. The modulation-doped SL has 20 periods with equal barrier and well widths of 100 Å. The SL layers are below the critical thickness and were grown pseudomorphically on a GaN buffer layer, resulting in biaxial tensile strain in the barrier layers.\(^{14}\) The 325-nm (3.815 eV) line of a He-Cd laser was used for excitation and the PL was dispersed with a 0.75-m spectrometer in the Czerny-Turner configuration and a Raleigh resolving power of \( \sim 2000 \). The SL was cooled using the coldfinger of a liquid-nitrogen cryostat. The detection used standard lock-in techniques in conjunction with a GaAs photodetector.

SL structures use semiconductors with different band gaps and, in the case of the III-nitrides, large internal polarization fields.\(^{15,16}\) A bound surface charge results when \( \mathbf{P} \cdot \mathbf{n} \neq 0 \), where \( \mathbf{P} \) is the total polarization at the surface and \( \mathbf{n} \) is the surface normal unit vector. A bound interface charge results from a bound liquid-nitrogen cryostat. The detection used standard lock-in techniques in conjunction with a GaAs photodetector.

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**FIG. 1.** (a) Schematic of the Al\(_{0.20}\)Ga\(_{0.80}\)N/GaN superlattice used in this study. (b) Self-consistently calculated band diagram.
the concentration of ionized donors, \(N_A\), is less than 0.6 MV/cm. This field causes the electron and hole face charges. The internal electric field is slightly greater than the polarization field resulting primarily from the bound interfacial charge which is present at each SL interface, and the relative dielectric constant along \(z\), \(\varepsilon_r = 10.4\), is taken as constant throughout the structure. The free-electron and free hole concentrations are given by Fermi-Dirac statistics,

\[
n_f(z) = N_f \sum_i |\psi_i^c(z)|^2 \ln \left[ 1 + e^{(\varepsilon_f - \varepsilon_i)/(kT)} \right],
\]

where \(\varepsilon_f\) is the Fermi energy, and \(N_f = m_f^* kT/\pi \hbar^2\).

Figure 2 shows the self-consistently calculated band diagram as well as the eigenfunctions used for the quantum-confined transition data. We choose the first five electronic eigen-states and the ground hole eigenstate for our transition data. The transitions, labeled on the right side of the figure, between these chosen eigenfunctions cover the energy range of experimentally observed transitions with excellent agreement. The figure also shows the band tilting due to the large polarization field resulting primarily from the bound interface charges. The internal electric field is slightly greater than 0.6 MV/cm. This field causes the electron and hole eigenfunctions to separate markedly along the growth direction (QCSE). It can also be seen in the figure that the higher energy electron states have a greater overlap with the ground hole state \(h_0\) as compared to the lower energy electron states. This overlap dependence on energy will be quantified below.

It should be noted that our one-dimensional model is based on an ideal SL with perfectly abrupt interfaces. There are, however, many physical effects that induce disorder in AlGaN/GaN multiple quantum well structures. These effects include impurity disorder, well-thickness variations, lattice imperfections, compositional fluctuations, strain fluctuations, and the resulting piezoelectric field fluctuations, and other effects. Capacitance-voltage (C-V) measurements on our SL samples have clearly shown multiple hole gases. Individual peaks of the C-V profiles show a full width at half maximum of only 30 Å. Such narrow profiles indicate a high superlattice interface quality and planar growth over a large area. Additionally, the hole mobilities and resistivities of our AlGaN/GaN heterostructures are 36 cm²/V s and 0.068 Ω cm at 90 K, indicating excellent interfacial quality. This is consistent with the results of other groups who concluded that the interfacial quality of their AlGaN/GaN heterostructures was excellent based on PL data. We therefore attribute the multiple peaks found in PL measurements to multisubband transitions. Disorder effects, although indisputably present, are believed to be secondary.

Using the 1D band diagram we calculate the spontaneous recombination rates taking place in the quantum well. We begin with the general expression for direct transitions between states in a III-V semiconductor,

\[
R_{\text{total}} = \sum_{ij} R_{ij}(h_v)
\]

\[
= \sum_{ij} \int_{E_{ij}}^\infty Z(h_v)B_{ij}(h_v)f_c(h_v)[1 - f_p(h_v)]\rho_{\text{red}}(h_v)\,d(h_v),
\]

where

\[
Z(h_v) = \frac{8 \pi}{(\hbar c)^3} \mu^3(h_v)^2,
\]

\[
B_{ij}(h_v) = \frac{e^2}{2m_0^* \varepsilon_0 \mu^2} |M_{ij}|^2,
\]

\[
|M_{ij}|^2 \approx \frac{m_e^2 E_{ij}}{3m_c} \frac{1 + \Delta/E_{ij}}{1 + 2\Delta/3E_{ij}} \int_{-\infty}^\infty \phi^{*}_{c,i}(z) \phi_{c,i}(z) \,dz,
\]

\[
f_c\) is the probability that the higher energy state is occupied, \(f_p\) is the probability that the lower energy state is occupied. The reduced density of states is given by

\[
\rho_{\text{red}} = \left( \frac{1}{\rho_c} + \frac{1}{\rho_p} \right)^{-1},
\]

where

\[
\rho_c = \frac{m_e}{\pi \hbar^2 L_z}
\]

and

\[
\rho_p = \frac{2m_p}{\pi \hbar^2 L_z}.
\]
band as $G$ is increased. The $R_{i0}$ increase sharply as the electron quasi-Fermi level approaches the $i$th electronic subband energy from below. Once the electron quasi-Fermi level gets $\approx 2kT$ above the $i$th electronic subband energy, $R_{i0}$ begins to level off. Thus the particular value of $G$ at which $R_{i0}$ begins to level off corresponds to the electron quasi-Fermi level rising into the $i$th electronic subband. This agrees qualitatively with our PL data.

Figure 3(b) displays the carrier subband concentrations as a function of $G$. At small values of $G$ the total number of holes $p_{\text{total}}$ approaches the equilibrium value due to doping, $p_{\text{eq}} = 2.5 \times 10^{18}$ cm$^{-3}$. As $G$ is increased the electronic subbands begin to populate commensurate with their energies. At large values of $G$ where $p_{\text{total}} > p_{\text{eq}}$, $n_{\text{total}}$ approaches $p_{\text{total}}$.

Figure 3(c) clearly shows the decrease in the average spontaneous recombination lifetime as the electron-hole pair generation rate is increased. The average spontaneous recombination lifetime is given by

$$\tau_{\text{ave}}(G) = \frac{\sum_i n_i}{\sum_i \tau_{i0}} = \frac{n_{\text{total}}}{\sum_i \tau_{i0}} = \frac{n_{\text{total}}}{\sum_i R_{i0}},$$

where $n_i$ is the number of free electrons per unit volume in subband $i$, $\tau_{i0}$ is the electron subband to ground hole subband recombination lifetime, and $R_{i0}$ is the electron subband to ground hole subband recombination rate given by Eq. (8).

As $G$ is increased, $\tau_{\text{ave}}$ decreases from $\tau_{\text{ave}} = 4.9 \times 10^{-3}$ sec at $G = 1 \times 10^{17}$ cm$^{-3}$ cm$^{-3}$ sec$^{-1}$, down to $\tau_{\text{ave}} = 5.9 \times 10^{-8}$ sec at $G = 1 \times 10^{27}$ cm$^{-3}$ cm$^{-3}$ sec$^{-1}$. Therefore as one increases $G$, a much smaller average recombination lifetime can be recovered despite the influence of the QCSE. Nonradiative transitions are neglected because of the low temperature.

Further inspection of Fig. 3(c) shows a slight dependence of the $\tau_{i0}$ on $G$, $\tau_{i0} = \tau_{i0}(G)$. This is due mainly to the dependence of $\tau$ on the Fermi occupation functions and the energy

$$\tau = n/R \int \frac{f_e(h\nu)d(h\nu)}{f(h\nu)(1-f_e(h\nu))d(h\nu)},$$

which follows from Eqs. (8) and (15).

To determine the quasi-Fermi levels, and thus $f_e$ and $f_v$, we assume unity quantum absorption efficiency at the pump laser energy, require charge neutrality, and equate the laser generation rate to the sum of subband recombination rates.
where $\tau_0$ corresponds to the minimum lifetime of a vertical transition, we see that since the [overlap] covers roughly six orders of magnitude, so too do the spontaneous lifetimes of the various transitions. The longest average electron subband to hole ground state lifetime, $\tau_{i0,\text{ave}}$, is $9.3$ ms for the ground-state to ground-state transition. However, even at the low electron-hole generation rate of $\approx 7 \times 10^{24} \text{ cm}^{-3} \text{ sec}^{-1}$, the average spontaneous lifetime $\tau_{\text{ave}}$, has a value of $\approx 3$ $\mu$s. This is in excellent agreement with typical values in the literature $^{31}$ for a similar $100 \text{ Å} \text{Al}_{0.15} \text{Ga}_{0.85} \text{N}/\text{GaN}$ QW structure. The shortest average electron subband to hole ground-state lifetime we calculate is $\tau_{d0,\text{ave}}=9.1$ ns. Using Eq. (18), this translates into a lifetime of $\approx 628$ ps for transitions whose overlap integral approaches unity, as is the case for a much thinner (~15–25 Å) QW. This is also in good agreement$^{10,21}$ with typical lifetimes reported in the literature.

Figure 4 shows low-temperature PL spectra of the modulation-doped $\text{Al}_{0.20} \text{Ga}_{0.80} \text{N}/\text{GaN}$ SL covering five orders of magnitude in pump intensity. Five clearly resolved PL peaks are observed at $E=3.11, 3.21, 3.30, 3.42$, and $3.46$ eV. The figure has a rich structure displaying four main features. First, it can be clearly seen that there are multiple, well-resolved PL peaks which change in relative intensity as the pump intensity (maximum pump intensity $I_{\text{max}}=I_0=8.3 \times 10^3$ W/cm$^2$) is changed. Second, not only are transitions well above the ground-state to ground-state transition seen, but remarkably, the high-energy transitions actually dominate the spectrum at high pump intensities. Third, the higher energy PL peaks increase in intensity relative to the lower energy PL peaks as the pump intensity is increased, in excellent agreement with the theory described earlier. Last, there is very little blueshifting of individual PL peaks as the pump intensity is increased.

We attribute the PL peaks to the sum of the individual Lorentzian broadened transitions between quantum-confined subband states in the conduction and valence band. The lack of appreciable blueshifting of individual PL peaks is consistent with a lack of screening. The lack of screening can be attributed to the fact that as the laser pump intensity is increased over our experimentally available range covering five orders of magnitude, the calculated average spontaneous lifetime decreases by roughly four orders of magnitude. Thus the steady-state number of electron-hole pairs per unit volume increases by only about one order of magnitude over our entire experimental pump intensity range. Therefore the amount of charge available to screen the internal polarization field begins to saturate at high pump intensities. This is due to two reasons. First, as the laser pump intensity is increased the average spontaneous lifetime decreases, tending to limit the number of electron-hole pairs generated. Second, as higher energy electron-hole pair states become populated their overlap integrals approach unity and they can no longer screen the internal polarization fields effectively. Both the
relatively small magnitude and the saturation of the blueshift has recently been reported in the literature for a similar QW structure.27

The expected strength of the GaN buffer layer peak is weak due to the much larger SL thickness (0.4 μm) as compared to the SL absorption length ($\alpha^{-1} \approx 0.083$ μm, assuming $\alpha = 1.2 \times 10^5$ cm$^{-1}$).23 We disregard any electron sub-band to neutral acceptor transition in the modulation-doped structure because the overlap integral of a relevant quantum-well electron envelope function with a neutral acceptor wave function is much less than the electron ground-state to hole ground-state transition.

Time-resolved measurements on these samples could provide additional experimental evidence of the different radiative lifetimes associated with the quantized optical transitions. Time-resolved measurements may also shed light on any nonradiative processes. Time-resolved measurements on AlGaN/GaN structures have been reported by several groups.21,22 These reports show that narrow AlGaN/GaN quantum wells have shorter spontaneous radiative lifetimes than wider quantum wells due to the larger spatial overlap of the electron and hole functions in the narrow quantum wells.

It was recently shown that LO-phonon replicas can give rise to multiple peaks in the emission spectra in AlGaN/GaN.28 However, one would expect that individual emission lines would be separated by the GaN LO-phonon energy which is approximately 90 meV.29,30 In our experiments, the various PL peak-energy splittings are not 90 meV or integral multiples of 90 meV. Furthermore, the PL intensity vs energy dependence, i.e., the shape of the spectra, seen in our PL spectra is different from what one would expect from LO-phonon replicas. Replica lines become weaker with the order of the replica. We therefore do not attribute the multiline luminescence to LO phonon replicas.

To quantify the magnitude of the screening charge we compare the maximum magnitude of the optically generated areal charge $\sigma_{\text{opt}}$ to the bound interface charge $\sigma_b$ due to the change in total polarization across a heterointerface. Using the full width at half maximum (FWHM) of the ground-state hole wave function squared yields $\sigma_{\text{opt}} = |e| \cdot \tau_{\text{avg}} (G_{\text{max}} \cdot G_{\text{max}}) \cdot |\phi_{v,0}^{\text{FWHM}}| \approx 1.4 \times 10^{-3}$ C/m$^2$, while $\sigma_b \approx 1.9 \times 10^{-2}$ C/m$^2$ for our Al$_{0.20}$Ga$_{0.80}$N/GaN SL. Therefore since $\sigma_b \gg \sigma_{\text{opt}}$, we may reasonably use the same band diagram and eigensystem for calculations at all pump intensities used in this study.

In order to verify the assignment of luminescence peaks to transitions between the quantum-confined subband states, we show in Fig. 5 the calculated transition energies relative to one of the PL spectra. The calculated transition energies range from 3.082 eV ($e_0 \rightarrow h_0$) to 3.524 eV ($e_4 \rightarrow h_0$) and clearly fall in the same energy range as the PL lines. The onset of the PL begins precisely where our calculated ground-state to ground-state transition begins. It is striking not only that luminescence occurs well above the QCSE $e_0 \rightarrow h_0$ prediction of 3.082 eV, but that the total energy spread is nearly 500 meV.

In conclusion, we have experimentally and theoretically investigated the photoluminescence properties of Al$_{0.20}$Ga$_{0.80}$N/GaN SL’s. The PL spectra clearly show multiple, well resolved, interband transitions between quantum-confined states. We solve the Poisson-Schrödinger system of equations self-consistently to obtain the eigenfunctions, eigenenergies, and 1D band diagram in the SL. We then use this data to calculate the lifetimes, transition energies, overlap integrals squared, and the spontaneous recombination rates per unit volume for each transition between quantum-confined states. Theoretically calculated transition energies between quantum-confined states agree very well with our experimental data. Theoretically calculated spontaneous recombination rates indicate that the higher energy PL peaks should increase in intensity relative to the lower energy PL peaks as the pump intensity is increased, also in excellent agreement with our experimental data. We demonstrate that new, high-energy, transitions become observable at high pump intensities because of the increased oscillator strength of the high-energy transitions. Therefore, despite the higher energy quantum-confined states being sparsely populated compared to the lower energy states, their larger oscillator strengths allow the higher energy transitions to dominate over the lower energy transitions at high pump intensities. Our simulations also show that at high pump intensities an average spontaneous recombination lifetime in the 50 ns range can be attained even for SL’s with large well and barrier widths less than 100 Å.

FIG. 5. Selected PL spectrum showing theoretical transition energies using theory described above. The spectrum shows clearly resolved subband transitions in excellent agreement with calculated transition energies given in Table I.

<table>
<thead>
<tr>
<th>Wavelength $\lambda$ (nm)</th>
<th>Energy $E$ (eV)</th>
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<tbody>
<tr>
<td>420</td>
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<tr>
<td>430</td>
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</tr>
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<td>500</td>
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TABLE I. Theoretical and experimental transition energies for Al$_{0.20}$Ga$_{0.80}$N/GaN SL’s.

<table>
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<tr>
<th>Transition</th>
<th>Experimental</th>
<th>Theoretical</th>
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<tbody>
<tr>
<td>$e_0 \rightarrow h_0$</td>
<td>3.082 eV</td>
<td>3.082 eV</td>
</tr>
<tr>
<td>$e_1 \rightarrow h_0$</td>
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<tr>
<td>$e_3 \rightarrow h_0$</td>
<td>3.457 eV</td>
<td>3.457 eV</td>
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<tr>
<td>$e_4 \rightarrow h_0$</td>
<td>3.524 eV</td>
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<table>
<thead>
<tr>
<th>Pump Intensity (W/cm$^2$)</th>
<th>Theoretical</th>
</tr>
</thead>
<tbody>
<tr>
<td>83</td>
<td></td>
</tr>
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</table>

Al$_{0.20}$Ga$_{0.80}$N/GaN SL, 100 Å / 100 Å, $T = 77.4$ K, pump intensity $I = 83$ W/cm$^2$, arrows denote theoretical quantum-confined transition energies.
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