Polarization effects in $Al_xGa_{1-x}N$ / GaN superlattices

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ABSTRACT

Room temperature and low temperature photoluminescence studies of $Al_xGa_{1-x}N$/GaN superlattices reveal a red shift of the dominant transition band relative to the bulk GaN bandgap. The shift is attributed to the quantum-confined Stark effect resulting from polarization fields in the superlattices. A theoretical model for the band-to-band transition energies based on perturbation theory and a variational approach is developed. Comparison of the experimental data with this model yields a polarization field of 4.6×10^5 V/cm for room temperature $Al_{0.1}Ga_{0.9}N$/GaN and 4.5×10^5 V/cm for room temperature $Al_{0.2}Ga_{0.8}N$/GaN. At low temperatures the model yields 5.3×10^5 V/cm for $Al_{0.1}Ga_{0.9}N$/GaN and 6.3×10^5 V/cm for $Al_{0.2}Ga_{0.8}N$/GaN. The emission bands exhibit a blue shift at high excitation densities indicating screening of internal polarization fields by photo-generated free carriers.

INTRODUCTION

GaN and $Al_xGa_{1-x}N$ have a large Mg acceptor activation energy ≥ 200 meV, much larger than kT at 300 K, resulting in low acceptor activation and low p-type conductivity in pn junction devices such as LEDs, lasers, and bipolar transistors. P-type $Al_xGa_{1-x}N$/GaN doped superlattices have been proposed to have a lower acceptor activation energy due to the possibility of *tunneling* activation rather than *thermal* activation of acceptors.[1,2] Since tunnel activation is independent of temperature, a higher acceptor activation is expected. Recently, reduced activation energies and enhanced hole concentrations have indeed been reported by several groups. [3-5] There have been several theoretical [6-8] and experimental [9-16] investigations relating to polarization fields of III-V nitride heterostructures. Polarization fields as large as 5×10^6 V/cm have been predicted based on tight-binding simulations.[6]

In this paper, the presence of polarization effects in doped $Al_xGa_{1-x}N$ / GaN superlattices (SLs) and the quantitative assessment of these fields are revealed. The magnitude of polarization is inferred from a comparison of experimental transition energies with a theoretical model based on second-order perturbation theory and a variational calculation using the Fang-Howard wave function.

EXPERIMENTAL DETAILS

The Mg-doped gallium-faced superlattices used in this study were grown by MBE on a c-plane sapphire substrate. They have 20 periods, and an equal barrier and well width of 100 Å. The layers are uniformly doped at a level of $N_{Mg} \approx 10^{19}$ cm^{-3}. The SL samples have p-type conductivity at room temperature.[3] Room and low-temperature photoluminescence (PL) spectra

are taken for $Al_xGa_{1-x}N$ / GaN SLs with Al mole fractions of x = 10 % and 20 %. The 325 nm line of a 15 mW He-Cd laser is used as the excitation source. A 0.75 m Spex spectrometer disperses the luminescence and a lock-in technique is used to acquire the spectra. For low-temperature PL the samples were mounted on the cold-finger of a liquid nitrogen cryostat.

DISCUSSION

The schematic band diagram of a superlattice without and with an internal electric field due to polarization effects is shown in figure 1(a) and (b), respectively. For superlattices without any overall field, a *blue* shift is expected as compared to bulk GaN due to the quantum size effect. However, with an internal electric field, a *red* shift is expected. The different contributions to the transition energy are shown in figure 1(c). Using the energies indicated in figure 1, the optical band-to-band transition energy is given by

$$E \;=\; E_{\mathrm{g,\,GaN}} + E_{0,e} + E_{0,h} - e\mathbf{E}\;L_{\mathrm{QW}}\;, \qquad (1)$$

where $E_{\mathrm{g,GaN}}$ is the GaN bandgap energy, and $E_{0,\mathrm{e}}$ and $E_{0,\mathrm{h}}$ are the quantized energies of the lowest electron and hole state in the triangular well, respectively, and the last term of Eq. (1) describes the energy drop within the GaN well layer of thickness L_{QW} due to the polarization field. Band bending due to free carriers and impurities is much less than polarization effects and is neglected. Excitonic transitions are not taken into account in Eq. (1); the effective excitonic Bohr radius is roughly four times smaller than the GaN well widths, implying that excitons would be ionized by the large electric field present there.

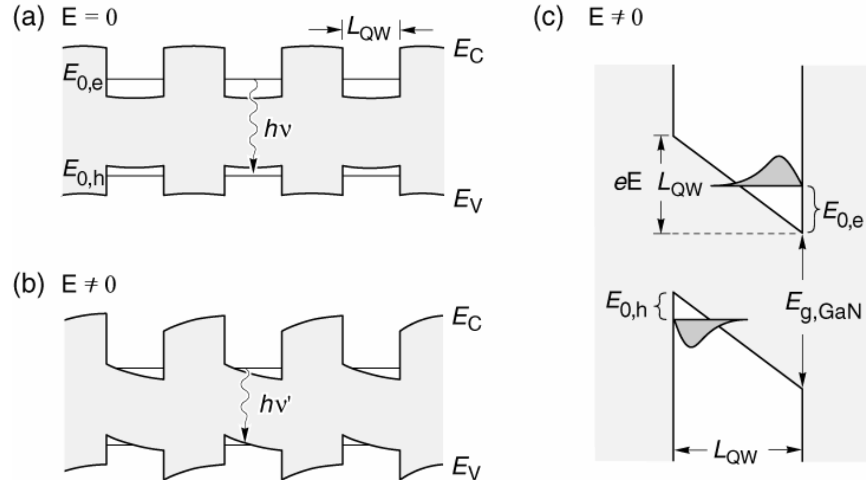

Figure 1. *Energy band diagram of $Al_xGa_{1-x}N/GaN$ superlattices (a) without and (b) with internal polarization fields. (c) Detailed schematic showing the different energies taken into account for the calculation of the optical transition energy.*

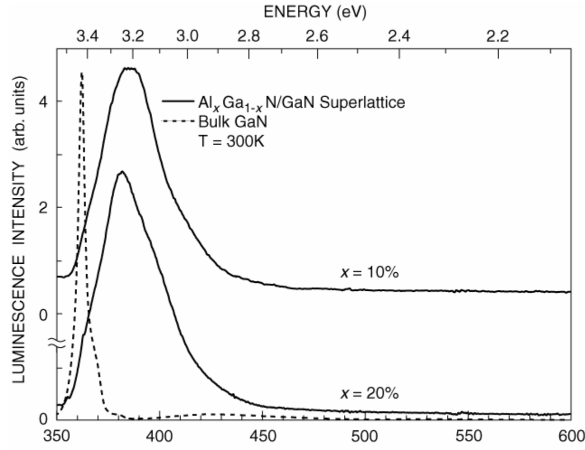

***Figure 2**. Room temperature photoluminescence spectra of $Al_xGa_{1-x}N$ / GaN doped superlattices with an Al content of 10 % and 20 %. Bulk GaN provided for comparison.*

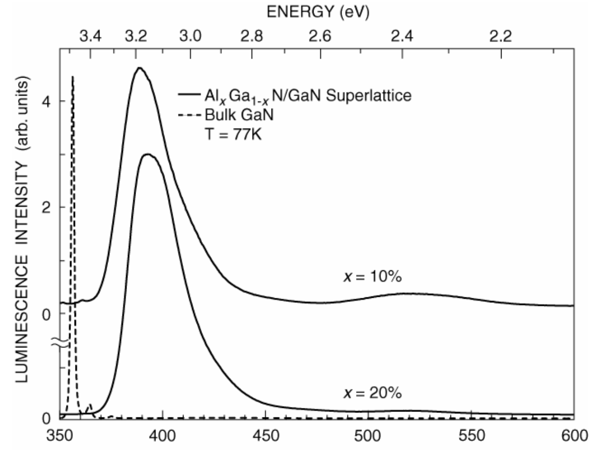

***Figure 3**. Low temperature photoluminescence spectra of $Al_xGa_{1-x}N$ / GaN doped superlattices with an Al content of 10 % and 20 %. Bulk GaN provided for comparison.*

Room temperature photoluminescence spectra of the two superlattices are shown in figure 2. A luminescence spectrum of bulk GaN with the main emission line at 362.0 nm is also included for comparison. Inspection of the figure reveals a main emission band at 383.5 nm and 382.3 nm for the $Al_{0.1}Ga_{0.9}N$/GaN and the $Al_{0.2}Ga_{0.8}N$/GaN superlattice, respectively. Both superlattice samples exhibit a strong red shift as compared to the bulk GaN sample.

Low temperature photoluminescence spectra of the two superlattices as well as bulk GaN are shown in figure 3. Inspection of the figure reveals a main emission band at 388.5 nm and 394.7 nm for the $Al_{0.1}Ga_{0.9}N$/GaN and the $Al_{0.2}Ga_{0.8}N$/GaN superlattice, respectively. Similar to the room-temperature results, the superlattice samples display a strong red shift as compared to bulk GaN.

We attribute the red shift of the main emission band with respect to bulk GaN to polarization effects occurring in the superlattices. Note that the changes in peak position with Al content and temperature variation are relatively minor. The full-width at half maximum of the PL spectra are 230 meV to 300 meV, probably due to well width variations. The relatively broad nature of the emission line is not indicative of excitonic transitions.

Next, we calculate the red shift of the emission energy using second-order perturbation theory for low fields and a variational approach for high fields. At low fields, the band-to-band transition energy is given by

$$E = E_{\mathrm{g,GaN}} + E_{0,e}^{(0)} + E_{0,h}^{(0)} + \Delta E_{0,e} + \Delta E_{0,h}, \qquad (2)$$

where $E_{0,e}^{(0)}$ and $E_{0,h}^{(0)}$ are the unperturbed ground-state quantized energies of electrons and holes respectively. $\Delta E_{0,e}$ and $\Delta E_{0,h}$ are the respective second order correction terms to the energy. The transition energy in Eq. (2) is approximated by the GaN bulk energy gap plus the unperturbed ground state energies of the electron and hole in GaN, plus the second-order correction terms. We use $E_{g,GaN}$ = 3.4 eV, an electron mass of $0.2 \times m_0$ and a hole mass of $0.8 \times m_0$. Also, we treat each quantum well in the SL as a single QW which is reasonable due to the large SL barrier thickness of 100 Å. For smaller barrier thicknesses the coupling between adjacent quantum wells must be taken into consideration and would cause a small decrease in

confinement energy.[9] The infinite well approximation is used for $E_{0,\mathrm{e}}$ and $E_{0,\mathrm{h}}$. We obtain a minor difference of 7.5 meV and 4.5 meV for the transition energy between the exact finite barrier calculation and the infinite well approximation using band discontinuities of ΔE_C = 3/4 ($E_{g,\mathrm{AlGaN}} - E_{g,\mathrm{GaN}}$) and ΔE_V = 1/4 ($E_{g,\mathrm{AlGaN}} - E_{g,\mathrm{GaN}}$) at an Al content of x = 10 % and x = 20 % respectively.

To calculate the ground state energy shifts near the origin we treat the electric field as a second-order correction to the infinite square well Hamiltonian leaving

$$\Delta E_0 \quad = \quad E_0 - E_0^{(0)} \quad = \quad e^2 \left|\vec{\mathsf{E}}\right|^2 \sum_{k \neq 0} \frac{\left|\left\langle \psi_k^{(0)} \right| x \left| \psi_0^{(0)} \right\rangle\right|^2}{E_0^{(0)} - E_k^{(0)}} \quad . \tag{3}$$

Taking only the first four terms of the sum in Eq. (3) yields

$$\begin{aligned} E(\mathsf{E}, L_{QW}) \cong E_{g,GaN} + \frac{1}{L_{QW}^2}\left(\frac{\hbar^2\pi^2}{2m_e^*} + \frac{\hbar^2\pi^2}{2m_h^*}\right) - 4e^2\left|\vec{\mathsf{E}}\right|^2 \left(\frac{2m_e^*}{\hbar^2\pi^2} + \frac{2m_h^*}{\hbar^2\pi^2}\right) \\ \times \left(\frac{|V_{01}|^2}{3} + \frac{|V_{03}|^2}{15} + \frac{|V_{05}|^2}{35} + \frac{|V_{07}|^2}{63}\right) \end{aligned} \tag{4}$$

where $|V_{0k}|^2 \equiv \left|\left\langle \psi_o^{(0)} \right| x \left| \psi_k^{(0)} \right\rangle\right|^2$. To get a value for the transition energy at high polarization fields we use the variational approach and the Fang-Howard wave function of the form $\Psi(x) = 2\alpha^{3/2} x e^{-\alpha x}$. One obtains

$$E_{0,e} \quad = \quad \frac{3}{2} \cdot \left[\frac{3e\left|\vec{\mathsf{E}}\right|\hbar}{2\sqrt{m_e^*}}\right]^{\frac{2}{3}}, \tag{5}$$

and a similar result for $E_{0,h}$. The result of the calculation is shown in figure 4.

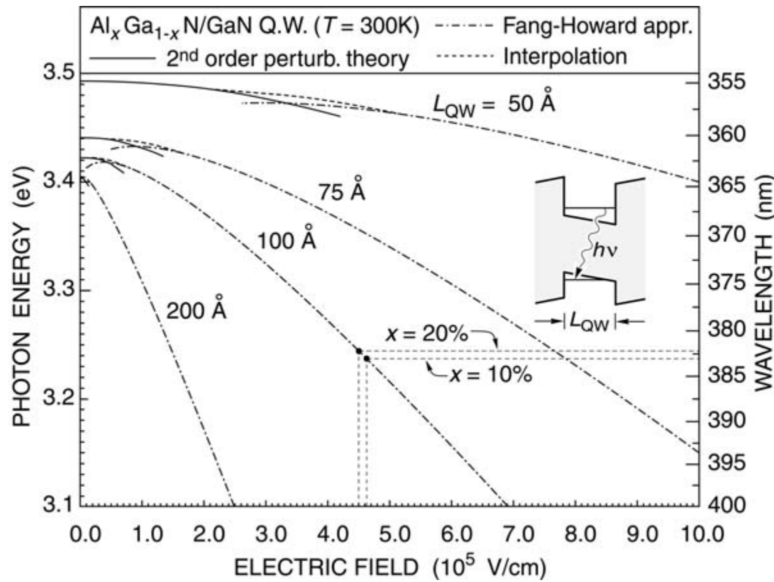

Figure 4. *Calculated transition energy versus internal electric field in the GaN well layers as a function of the well width, L_{QW}. The theoretical predictions are based on second-order perturbation theory and a variational approach using the Fang-Howard wave function.*

The dashed line is an interpolation between the second order perturbation and the variational calculation. Comparing the theoretical data shown in figure 4 with the experimental peak energies shown in figure 2 and figure 3 yields an internal polarization field of 4.6×10^5 V/cm for $Al_{0.1}Ga_{0.9}N$/GaN and 4.5×10^5 V/cm for $Al_{0.2}Ga_{0.8}N$/GaN at room temperature. At liquid nitrogen temperatures we get 5.3×10^5 V/cm for $Al_{0.1}Ga_{0.9}N$/GaN and 6.3×10^5 V/cm for $Al_{0.2}Ga_{0.8}N$/GaN. The increase in electric field for the sample with higher Al content at low temperature is consistent with the electric field caused in part by the piezoelectric effect.[16, 17]

The low temperature data presented here allows for the assessment of the relative contributions of the spontaneous and the piezoelectric polarization. Linear extrapolation of the electric field to $x = 0$ % yields a value of E = 4.3 x 10^5 V/cm for GaN. Thus about 45% of the field in the $x = 20$ % SL is caused by piezoelectric polarization. This is comparable to recently published data.[6]

The variation of optical transition energy with excitation density is shown in figure 5. The excitation power was varied over two orders of magnitude, from 15 mW to 0.15 mW using neutral density filters. Figure 5 reveals a blue shift of the optical transition energy at high excitation densities. This is in agreement with free carriers screening the internal field thereby shifting the emission peak to higher energies. However, the blue shift is rather small consistent with a short radiative lifetime found in GaN materials.

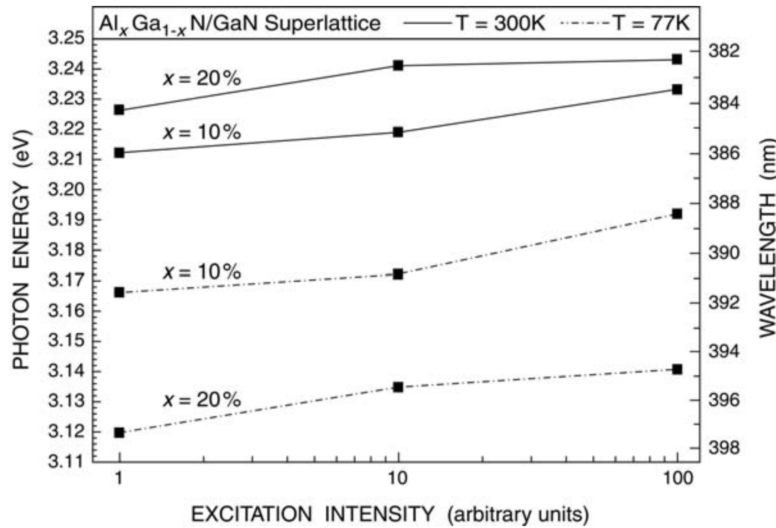

Figure 5. *Room and low temperature transition energy in $Al_xGa_{1-x}N$ / GaN doped superlattices versus excitation*

CONCLUSIONS

In conclusion, we have shown evidence of, and measured, polarization fields in Mg doped $Al_xGa_{1-x}N$/GaN superlattices. We attribute the red shift of the photoluminescence transition peaks to the quantum confined Stark effect arising from large polarization fields. Comparison of experimental luminescence data with a theoretical model based on perturbation theory and a variational approach yields electric field strengths of 4.6×10^5 V/cm for $Al_{0.1}Ga_{0.9}N$/GaN and 4.5

$\times 10^5$ V/cm for $Al_{0.2}Ga_{0.8}N/GaN$ at room temperature. Low temperature studies reveal 5.3×10^5 V/cm for $Al_{0.1}Ga_{0.9}N/GaN$ and 6.3×10^5 V/cm for $Al_{0.2}Ga_{0.8}N/GaN$. The emission bands exhibit a blue shift at high excitation densities indicating screening of internal polarization fields by photo-generated free carriers. This work was supported by the Office of Naval Research and monitored by Dr. C. E. C. Wood.

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17. It is well known that the oscillator strength is reduced due to piezoelectric effects in $Al_xGa_{1-x}N$ / GaN quantum wells. Because the oscillator strengths do not affect the main photon transition *energies*, we do not consider them.