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Surface band-bending effects on the optical properties of indium gallium nitride multiple quantum wells

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We report the use of selective wavelength excitation to examine the surface band-bending effects on the optical properties of 3.0-nm-thick indium gallium nitride (InGaN) multiple quantum wells (MQWs). Under a 355-nm excitation, the $In_{0.18}Ga_{0.82}N$ well emission exhibits a linear dependence on the injected carrier density (N_{inj}) with a coefficient of (i) 8.5×10^{-18} meV cm^3 for the spectral blueshift and (ii) 3×10^{-14} V cm^2 for the change of internal field at a density up to $N_{inj}\sim10^{19}$ cm^{-3} at 77 K. When excited by a shorter wavelength at 248 nm, the emission from the thin GaN cap layer quenches, but that from the InGaN wells prevails. These observations are attributed to the transportation of photogenerated carriers from the bent GaN surface and redistribution in the InGaN wells. By solving the rate and Poisson equations with a Fermi-level pinning in the band-structure analysis, the emission from the InGaN/GaN MQWs is shown dominant by the recombination between the high-lying subbands and the screening of internal field effects. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1583869]

From recent developments in the material growth and device processing on aluminum indium gallium nitride (AlInGaN), this material system has emerged as a promising light source spanning a wide spectral range from ultraviolet to red.[1] Commercialization of candela-class InGaN quantum-well (QW) blue/green light-emitting diodes,[2] and long-lifetime violet and blue laser diodes[3] are examples of such exciting achievements. Despite the progress that has been made in active layer design,[4] many of its peculiar optical properties still remain a great mystery to be solved.[5] A prevailing theory ascribes the emission mechanism to the localized states[6] from the growth of InGaN layer.[7] Alternatively, a characteristic internal field distribution, arisen from the discontinuity of piezoelectricity (P_{pz}) and spontaneous polarization (P_{sp}) at the nitride interface,[8] can also respond to the change of oscillator strength in spectral analysis.[9]

A general assumption is that the recombination is governed by the localized states at low temperature, whereas at high temperature, it is controlled by the extended states. A recent study,[10] however, reveals a drastic reduction in the stability of localized exciton due to the large internal field[11] and carrier screening effects[12] in the InGaN/GaN QW. This makes the treatment of field distribution inside the nitride multiple QWs (MQWs) an important consideration for the optical analysis.[13] The discontinuity of P_{sp} and P_{pz} at the cap/air interface also imposes a disturbance on the boundary condition. The resultant surface charge, which can be as large as 2×10^{13} cm^{-2}, constitutes another issue to be resolved in the band-structure analysis.[14]

In this letter, we report a series of photoluminescence (PL) experiments by selective wavelength excitation at various temperatures to examine the surface band-bending effects on the optical properties of InGaN/GaN MQWs. We find it essential to include a Fermi-level pinning in the GaN cap layer[15] to quantify the carrier redistribution effects. Our analysis indicates the emission properties of the 3.0-nm InGaN MQWs can be attributed to the dominant transitions between the high-lying subbands and the screening of internal field effects.

Samples used in this study were grown by the metalorganic chemical vapor deposition technique.[16] The sample structures consisted of, in sequence, an undoped 30- or 50-nm GaN cap layer with three pairs of silicon doped 3.0-nm (L_w) $In_xGa_{1-x}N$/GaN MQWs at a concentration of 10^{18} cm^{-3}, followed by a 1.5-μm-thick GaN buffer layer of $\sim10^{17}$-cm^{-3} background doping on the (0001) plane of sapphire substrates. The In composition ($x=0.15$ and 0.18) in the well and thickness in the GaN barrier ($L_b=10$ and 7 nm) were varied to examine the distribution of internal field effect on the optical properties. The selective wavelength excitation experiments were performed by using a third-harmonic generation (355 nm) of neodymium: yttrium-aluminum-garnet (Nd:YAG) laser (New Wave, USA) and a 248-nm KrF excimer laser (TuiLaser, Germany). The PL signals were collected from the sample surface, dispersed by a grating spectrometer, and detected by a CCD array.

Illustrated in Fig. 1 are the emission spectra measured at (a) 77 and (b) 300 K from the 3.0-nm $In_{0.18}Ga_{0.82}N$/GaN MQWs as excited by the 355-nm Nd:YAG laser. Note the peak emission energy remains nearly stationary in the low-density excitation regime ($<10^{-3}$ MW/cm^2, not shown here, but will be seen in Fig. 4) but blueshifts ($\sim$70 meV) with the increase of pump intensity until a threshold ($\sim$0.1 and 1 MW/cm^2, respectively, for the 77 and 300 K measurements) is reached. Thereafter, one resumes a spectral redshift

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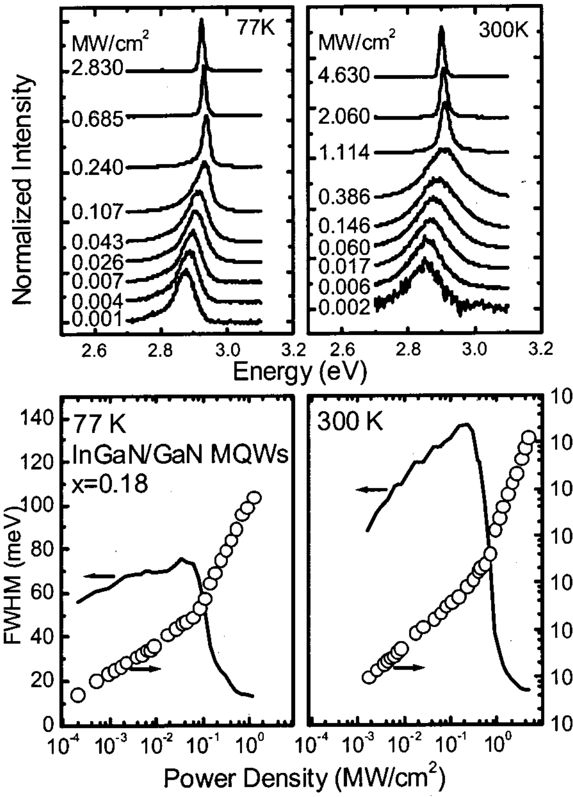

FIG. 1. PL spectra of the 3.0/10.0-nm $In_{0.18}Ga_{0.82}N$/GaN MQWs excited by a 355-nm Nd:YAG laser at (a) 77 and (b) 300 K. The spectra have been vertically shifted for comparison.

($\sim$15 meV) with further increase of carrier injection density due to a dominant band-gap renormalization effect.[17] The integrated peak emission intensity, as inferred from Fig. 1, exhibits an abrupt change from a linear into square power dependence on the excitation intensity as the latter exceeds the threshold. In concurrence with this, we observe a linewidth narrowing, which is followed by a slight broadening due to a contribution from the electron–hole plasma effect.[18]

The revealing of constant emission peak energy in the low-density excitation ($<10^{-3}$ MW/cm^2) regime, which is independent of the measurement temperature, suggests the state of localization has been minimized in the samples under study. Were this not the case, momentum space filling of the localized or band-tail states by the photogenerated carriers would quickly raise the emission energy. A similar phenomenon has also been recently reported on the room-temperature emission spectra of a wider 4-nm-thick $In_{0.15}Ga_{0.85}N$/GaN MQWs.[19] We further note the temperature dependence of the peak energy shift, as measured in the low-density excitation experiments, reaches only half of the value that would be expected from the Varshni effect. Including the localization effect, however, would enhance rather than compensate the energy difference.[20] Moreover, referring to the inset of Fig. 2, one notes the emission intensity from the GaN cap layer is two orders of magnitude lower than that from the InGaN wells when excited by a 355-nm laser. By changing the excitation source to a 248-nm laser, we find emission from the GaN cap layer ceases, but that from the InGaN well remains.

To resolve these problems, we adapt a finite difference

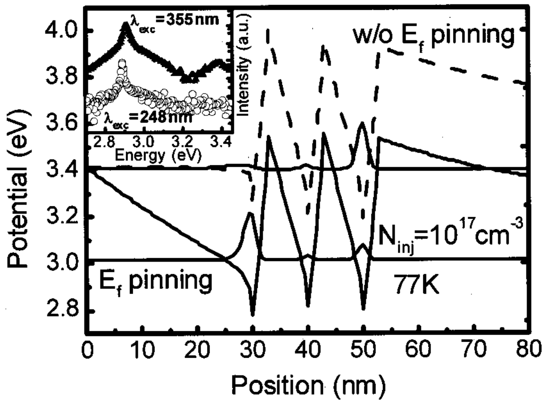

FIG. 2. Band-edge profiles of the InGaN/GaN MQWs analyzed with and without Fermi-level pinning at the GaN cap layer. Inset: emission spectra of the InGaN MQWs under an excitation of a 248-nm and a 355-nm laser.

method in the band structure analysis[21] that incorporates solutions from the rate and Poisson equations to describe the photo-excitation and carrier redistribution processes at the nitride interface. Since Fermi-level (E_F) pinning represents a genuine situation occurred to the undoped n^--GaN surface,[22] this phenomenon is assessed by a donor-like surface state with density $\sim 10^{13}$ cm^{-2} to ensure a pinning of E_F at 0.4 eV below the conduction band edge of n^--GaN.[23] We let the Roosbroeck–Shockley radiative recombination coefficient of GaN be the only fitting parameter, with a temperature dependence following that in Ref. 24. The rest of material parameters are taken as linear interpolations from those in Ref. 25, and a bowing factor of 2.05 eV is used for calculating the band-gap energy of InGaN.

The band-edge profiles shown in Fig. 2 correspond to the boundary condition of (a) periodical field distribution (F_w) $\propto (P_b - P_w) \cdot L_b / (L_w + L_b)$[26] with a flat surface, and (b) surface Fermi-level pinning with polarization-induced charge of $-\nabla \cdot (P_{sp} + P_{pz})$ at each of the nitride interface, respectively. In thermal equilibrium charge neutrality associated with the lineup of Fermi level in (b) determines the amount of free carrier that could be released from the ionized surface state. A nonzero surface electric field in the GaN cap layer (-0.17 MV/cm) reflects one such effect and can facilitate the transportation of photogenerated carriers into the adjacent InGaN wells.

We further note the field strength ($\sim$2 MV/cm) in the nitride QW, as inferred from the slope of the potential profile shown in Fig. 2, far surpasses that required for the dissociation of localized exciton.[27] Under a strong field action, not only can one encounter a reduced oscillator strength, but also a relaxation of parity-selection rule; that is, $\Delta n \neq 0$ among the interband transitions. As a result, one would expect the emission spectra of InGaN MQWs dominated by the (i) high-lying subband transitions whose wave-function overlap in space is less susceptible to the field action and thus leads to larger momentum matrix elements, and (ii) screening of internal field due to the Fermi-level pinning and carrier redistribution effects. We illustrate in Fig. 3 the normalized emission spectra of (a) $x=0.15$ and (b) $x=0.18$ $In_xGa_{1-x}N$/GaN MQWs to elucidate such a principle. The calculation is based upon absorption of photogenerated car-

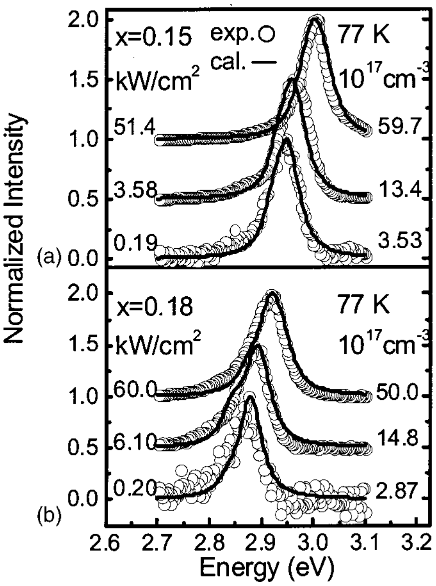

FIG. 3. Measured and calculated emission spectra of the 3.0-nm (a) $x=0.15$, and (b) $x=0.18$ In$_x$Ga$_{1-x}$N/GaN MQWs at 77 K. The spectra have been vertically shifted for comparison.

riers in the thin GaN cap layer and their transportation and redistribution into the InGaN MQWs by substituting a Roosbroeck–Shockley coefficient of 2.73×10^{-9} cm^3 s^{-1} at 77 K and solutions from the rate and Poisson equations to the band structure analysis. Note the agreement with experimental data can extend over three decades of power excitation up to ~0.1 MW/cm^2 ($N_{\mathrm{inj}}\sim10^{19}$ cm^{-3}) and cover a spectral range of 0.2 eV.

Shown in Fig. 4 are data of the measured and calculated peak emission energy of the 3.0 nm (a) $x=0.15$ and (b) $x=0.18$ In$_x$Ga$_{1-x}$N/GaN MQWs, revealing the evolution of spectral blueshift with carrier injection. Further information can be gathered from the inset of Fig. 4, from which one can resolve a linear dependence of the (i) peak energy shift and (ii) decrease in the internal field (F_w) of sample (b) on the carrier injection density (N_{inj}). The underlying physics is that the originally saw-tooth-like band-edge profile can provide a nonvanishing, *first-order* correction in the interband transition energy when a perturbation, such as carrier injection, to the internal field occurs. A linear dependence ($\sim3\times10^{-14}$ V cm^2) of F_w can thus be obtained over two decades of density variation in N_{inj}. Although this coefficient is half of what would be obtained by a simplified slab-like charge model analysis, that is, $\delta F_w\sim\delta n\cdot L_w/\varepsilon_0\varepsilon_r$, it does reveal the sensitivity of field screening due to the combined effects of surface Fermi-level pinning and carrier redistribution. Such a linear relationship, in turn, translates into a peak energy shift with $\sim8.5\times10^{-18}$ meV cm^3 on N_{inj} due to the nonzero first-order perturbation on the field screening effect.

In summary, we report the use of low-temperature, selective wavelength excitation technique to examine the surface bending effects on the emission properties of InGaN/GaN MQWs. The linear dependence of spectral blueshift and emission intensity can be correlated with the decrease of internal field with carrier injection. These observations are ascribed to the GaN surface Fermi-level pinning and the carrier transportation and redistribution effects in the InGaN wells.

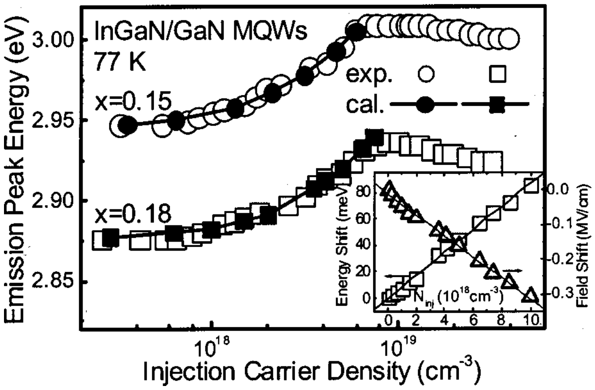

FIG. 4. Measured and calculated peak emission energy of the (a) $x=0.15$, and (b) $x=0.18$ In$_x$Ga$_{1-x}$N/GaN MQWs at 77 K. Inset: a linear dependence of the peak energy shift and change of internal field of sample (b) with carrier injection.

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