Effective band gap inhomogeneity and piezoelectric field in InGaN/GaN multiquantum well structures

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The emission mechanisms of strained $In_xGa_{1-x}N$ quantum wells (QWs) were shown to vary depending on the well thickness, L, and x. The absorption edge was modulated by the quantum confined Stark effect and quantum confined Franz-Keldysh effect (QCFK) for the wells, in which, for the first approximation, the product of the piezoelectric field, F_{PZ}, and L exceed the valence band discontinuity, ΔE_V. In this case, holes are confined in the triangular potential well formed at one side of the well producing the apparent Stokes-like shift. Under the condition that $F_{PZ}\times L$ exceeds the conduction band discontinuity ΔE_C, the electron-hole pair is confined at opposite sides of the well. The QCFK further modulated the emission energy for the wells with L greater than the three dimensional free exciton Bohr radius a_B. On the other hand, effective in-plane localization of carriers in quantum disk size potential minima, which are produced by nonrandom alloy compositional fluctuation enhanced by the large bowing parameter and F_{PZ}, produces a confined electron-hole pair whose wave functions are still overlapped (quantized excitons) provided that $L < a_B$. [S0003-6951(98)02040-3]

Major developments of III-nitride semiconductors[1,2] have led to the commercial production of InGaN single quantum well (SQW) light-emitting diodes (LEDs)[1] and to the demonstration of multiquantum well (MQW) blue laser diodes (LDs).[1,3] Although they[1–3] contain InGaN in active regions, the emission mechanisms are not fully understood. Several groups have assigned the spontaneous emission from InGaN quantum wells (QWs) to the recombination of excitons localized at certain potential minima.[1,4–6] On the other hand, several groups have discussed the importance of the quantum confined Stark effect (QCSE)[7] due to the piezoelectric field (F_{PZ}) in strained wurtzite InGaN QWs.[2,5,8–10] In order to obtain an insight into what dominates the emission properties for further optimization of InGaN MQW LDs, it is necessary to investigate the effects of the effective band gap inhomogeneity[1,4–6] and the piezoelectric field[2,4,5,8,9,11] consistently.

In this letter, complementary absorption and static/time-resolved (TR) photoluminescence (PL) spectra of InGaN MQW's are shown as a function of the well thickness, L. While strong F_{PZ} exist in the QWs, effective in-plane localization of quantized excitons play an important role for the QWs with L smaller than the three dimensional (3D) free exciton (FE) Bohr radius a_B.

The samples were prepared by metalorganic chemical vapor deposition. A series of MQWs with 14 periods of undoped $In_{0.1}Ga_{0.9}N$ wells (L was varied from 1.2 to 6 nm) and 4.3-nm-thick Si-doped (2×10^{18} cm^{-3}) GaN barriers capped by 100-nm-thick $Al_{0.06}Ga_{0.94}N$ was grown on 2-μm-thick GaN:Si on (0001) sapphire. Low excitation (less than 1 W/cm^2) photoluminescence excitation (PLE) and PL spectra were taken using monochromatic light from a 80 W Xe lamp. High excitation (nearly 500 W/cm^2) PL and TR-PL were excited by a frequency-tripled 150 fs pulse from a tunable Ti:sapphire laser operating at an 80 MHz repetition rate. All measurements were carried out between 4 K and room temperature (RT).

Since the critical thickness of $In_{0.1}Ga_{0.9}N$ is reported to be greater than 40 nm,[2,8] we assume coherent growth of InGaN. This strain will cause F_{PZ} along the growth direction in wurtzite InGaN(0001).[2,4,5,8–11] On the other hand, 3D GaN exhibits FE emission[12] at RT since the exciton binding energy E_{ex} is as large as 26 meV and a_B is as small as 3.4 nm.[12–14] Thus the problem treated here is the behavior of confined energy states in QWs under high electric field, F.[7,15,16]

In QWs, exciton absorption can be observed even at RT[7,15] due to confinement of wave functions of electron-hole (e-h) pair to increase E_{ex}.[15] Miller *et al.*[7] observed an excitonic absorption in GaAs QWs, which was redshifted by 2.5 times the zero-field E_{ex} for $F=10^5$ V/cm (50 times the ionization field F_i of 3D excitons), and explained the findings in terms of field modulation of quantized energy levels (QCSE).[7] We estimated[17] E_{ex} in GaN/$Al_{0.1}Ga_{0.9}N$ QWs un-

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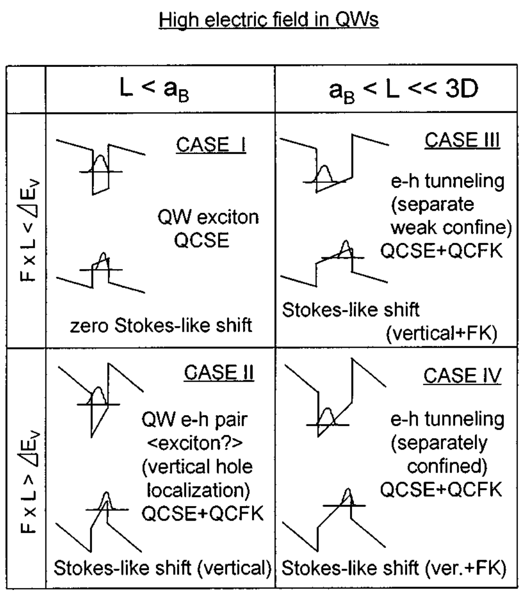

FIG. 1. Schematic band diagrams of GaN/InGaN QWs under the electric field *F*.

der zero field as a function of L to obtained $E_{ex}=47$ meV for $L=3$ nm, and $F_i=6.0\times10^5$ V/cm.

Taking the 1:4 relation between the valence and conduction band discontinuities (ΔE_V and ΔE_C) into account,[10] schematic band diagrams of InGaN/GaN QWs are drawn in Fig. 1. Since the restriction $F\times L>\Delta E_V$ breaks before breaking $F\times L>\Delta E_C$ with increasing F or L, restrictions between $F\times L$ and ΔE_V and between L and a_B are drawn. Note that in-plane band gap fluctuation is omitted. In CASE I, both electron and hole wave functions are confined in the well, and they have unique quantized energy levels where zero Stokes-like shift is expected. In CASE II, at least the hole wave function drops into the triangular shape potential well at one side of the well, and continuum states are formed at the rest of the potential slope inside the QW region. Therefore a vertical component Stokes-like shift is produced. In CASES III and IV, the absorption tail would be broadened due to QCSE and quantum confined Franz-Keldysh effect (QCFK)[16] where the originally ''forbidden'' QW transitions become strong. To study the in-plane effective band gap fluctuation and effects of F_{PZ} on the Stokes-like shift, we prepared the samples belonging to CASES I, II, and IV assuming constant F_{PZ}.

Low excitation PL spectra exhibited broad peaks peculiar to InGaN QWs with full width at half maximum (FWHM) of nearly 120 meV, as shown in Fig. 2. The PL peak showed a redshift by 360 meV with increasing L from 1.2 to 6.2 nm, and the intensity decreased for $L>3.6$ nm ($L>a_B$). The PLE spectra exhibited broadened absorption tail except for $L=1.2$ nm, and the broadening was pronounced for the wells with $L>a_B$.

The apparent band gap energy determined as the energy where the PLE signal intensity drops to half the maximum, low and high excitation PL peak energies, and apparent Stokes-like shifts are plotted as a function of L in Fig. 3. As expected, Stokes-like shift increased from nearly 50 to 220 meV. Similar results were obtained at 4 K where the in-plane Stokes-like shift ($L<3.6$ nm) was nearly 100 meV.

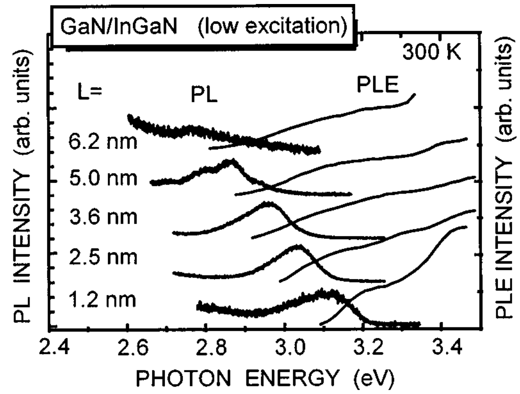

FIG. 2. Low excitation PL and PLE spectra at RT of $In_{0.1}Ga_{0.9}N$ GaN:Si for different well thickness, *L*.

In order to estimate the electric field F across the QW, confined energy levels and wave functions in the $In_{0.1}Ga_{0.9}N$ QWs are calculated as functions of F, L, and doping density in the barrier by variational method neglecting E_{ex} within Hartree approximation by solving the Schrödinger equation and Poisson equation simultaneously self-consistently.[18] We fit the relation between the low excitation PL peak energy and L ($L<3.6$ nm) in Fig. 3. As a result, the zero-field band gap of the 3D InGaN is obtained to be 2.92 eV and ΔE_C and ΔE_V are estimated to be 380 and 120 meV, respectively. F is estimated to be nearly 3.5×10^5 V/cm which gives a Stark shift of nearly 45 meV in 3-nm-thick QW. The estimated F nearly agrees with previously reported F_{PZ} for $In_{0.1}Ga_{0.9}N$.[19] Under $F=3.5\times10^5$ V/cm, $F\times L$ exceeds ΔE_V for $L>3.4$ nm. The hole confined level would already be formed in the triangular potential (CASE II) between 2.5 and 3.6 nm. Beyond this, the system belongs to CASE IV where the hole (and electron) could tunnel through the Coulomb potential barrier, resulting in the separate *e-h* in opposite sides of the well. This may explain the extremely long decay times, τ, for $L=5.0$ and 6.2 nm at 4 K, which exceed 35 ns, in terms of reduction of the oscillator strength.[11] On the other hand,

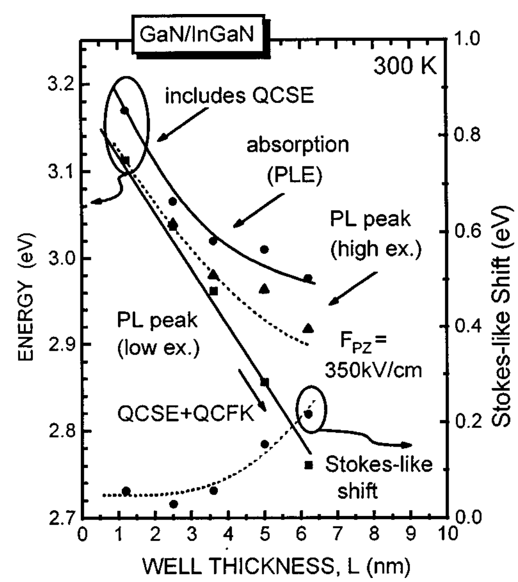

FIG. 3. High and low excitation PL peak energies, apparent band gap energy estimated from PLE spectra, and apparent Stokes-like shift of $In_{0.1}Ga_{0.9}N$ wells at RT as a function of *L*.

shorter τ (0.97–4.7 ns) for $L<2.5$ nm at 4 K indicates that the overlap of the e-h wave function is still large because L is smaller than a_B. Note that the estimated F_i (6.0×10^5 V/cm) is larger than F_{PZ} (3.5×10^5 V/cm), which implies that Coulomb interaction between the e-h pair still remains.

In-plane effective band gap inhomogeneity plays an important role in the QW emission. If there is no in-plane carrier localization, the PL peak energy should agree with the PLE resonance energy for CASE I i.e., 1.2 nm in our samples. However, we observed the in-plane net Stokes-like shift nearly 50 meV, which is a source of lateral localization of carriers. Previous cathodoluminescence (CL) measurements revealed that the broad CL band (FWHM~100 meV) consists of many sharp peaks (FWHM<20 meV)[4,5] at 10 K. Regions emitting the sharp CL peak have a lateral size of 20–300 nm.[4,5,20] This means that there exists submicrometer length scale (<300 nm)[4,5] effective band gap inhomogeneity. One of the possible origins of these is an inhomogeneous distribution of F_{PZ} caused by strain fluctuations.[5] If we attribute the in-plane Stokes-like shift to this, the potential fluctuation should be leveled by filling carriers to screen F_{PZ}. However, high and low excitation PL peak energy was nearly unchanged for CASE I ($L=1.2$ nm), and the gain spectra of InGaN MQWs were broad.[1,5,10,21]

For LDs, this effective band gap inhomogeneity is too large to obtain a uniform electron-hole plasma (EHP).[5,10,21] Indeed, some InGaN MQW LDs showed EHP lasing in tail states,[5,10,21] which indicate the presence of nonrandom alloy compositional fluctuation. Since the change in the composition simultaneously changes the strain that causes change in F_{PZ},[5] quantum disk (Q-disk) size[22] potential fluctuations are considered to be formed by nonrandom alloy compositional fluctuation enhanced[5] by the large bowing parameter[2,8] and the inhomogeneous piezoelectric[8] QCSE.[7]

In summary, the importance of in-plane submicrometer scale potential fluctuation was shown as well as strong effects of F_{PZ} in the InGaN QWs. This potential fluctuation accumulates carriers to Q-disk size potential minima, and effectively keeps carriers away from nonradiative pathways. Strong piezoelectric fields separate the e-h pair into triangular potential wells formed at opposite sides of the well, and apparent Stokes-like shift is dominated by F_{PZ} for the CASES II, III, and IV. The oscillator strength of the separate e-h pair is very small for CASES III and IV. State-of-the-art blue, green, and amber InGaN LEDs and LDs generally have QWs with $L=2.5$–3.5 nm, and they may belong to CASE II or pronounced CASE II. Therefore in-plane localized e-h pair has strong Coulomb interaction since the vertical e-h distance is still smaller than a_B, resulting in overlapping of their wave functions in the QW. In order to understand precise mechanisms of the band gap inhomogeneity and carrier localization, and to obtain long wavelength LDs expanding from pure blue to red wavelengths, fabrication and investigation of cubic InGaN QWs are mandatory to eliminate the spontaneous and strain-induced piezoelectricity.

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