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Studies of InGaN/GaN multiquantum-well green-light-emitting diodes grown by metalorganic chemical vapor deposition

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InGaN(3 nm)/GaN(5 nm) three period multiquantum green-light-emitting diodes (LEDs) grown by the metalorganic chemical vapor deposition technique have been studied using high-resolution transmission electron microscopy (HRTEM), double crystal high resolution x-ray diffraction (HRXRD) and low temperature photoluminescence. HRTEM analysis showed that the defect density gradually decreased in the growth direction with increasing thickness. Self-assembled quantum dot-like structures in the wells and black lumps between the well and barrier due to In segregation and strain contrast were observed, respectively. The HRXRD spectrum of the green LED structure was simulated using the kinematical theory method to obtain the composition and thickness of the well and barrier. The quantum-well (QW) green emission peak 2.557 eV at 10 K showed "S" shaped shift like a red-blue-red shift with variation of the temperature in the photoluminescence spectra due to potential fluctuations caused by inhomogeneous alloy distribution in the wells. The activation energy of 49 meV obtained from the QW green emission line indicated deepening of the localization of the carriers. © 2004 American Institute of Physics.
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Group III nitrides play a major role in optoelectronic devices despite their high density of defect levels (10^9 cm^{-2}). High brightness light-emitting diodes (LEDs) are now commercially available, however green LEDs show less efficiency than that of blue LEDs,¹ therefore investigations of green LED structures are essential in order to understand their device physics. Deposition parameters such as well, barrier widths and carrier sheet density are important criteria in optoelectronic devices,² in the case of laser diodes, to obtain a quasifield-free shaped quantum well, where the favorable sheet density is $5 \times 10^{12} \text{ cm}^{-2}$ and for lasing action the well width should be less than 5 nm.³ Group III nitrides generate large piezoelectric fields because of their large piezoelectric constants, which lead to the quantum confined Stark effect on the optical properties. The InGaN layers are active regions in the device in which In segregation or phase separation is the main drawback. The segregation of In is due to large lattice mismatch and low miscibility between GaN and InN compounds, which cause large compositional fluctuations in the InGaN layers.⁴ In this letter, we report the structural and optical properties of green-light-emitting diodes.

The green-light-emitting devices studied in this work were grown by metalorganic chemical vapor deposition (MOCVD). A GaN buffer layer with thickness of 25 nm was first grown on *c*-plane sapphire substrates at low temperature of 560°C, followed by a 2 μm thick Si doped GaN layer at higher temperature of 1060°C, on which a three period 3 nm InGaN well/5 nm GaN barrier was then grown at low tem-

perature of 780°C using N₂ only as the carrier gas, followed by a 0.1 μm *p*-AlGaIn cladding layer and finally *p*-GaIn with thickness of 0.5 μm was deposited. Metalorganic sources such as trimethylgallium, trimethylindium, trimethylaluminum, and NH₃ were used as precursors for Ga, In, Al, and radical nitrogen, respectively.

In order to study the microstructure of the layers, the back surface of the samples was mechanically polished and then thinned by an Ar⁺ ion milling. A Philips transmission electron microscope was operated at 200 kV to obtain structural data. The samples were characterized using a Philips X-Pert double crystal x-ray diffractometer (XRD) with a Ge(220) single crystal, operated at 40 kV and 50 mA with radiation of 1.540 56 Å. A He-Cd laser used at 325 nm for excitation, a GaAs based photodetector used to record excited emission, and a closed loop He cryostat for low temperature were employed.

The density of defect levels gradually decreased in the growth direction from 10^9 to 10^8 cm^{-2} and some of the nanopipe-like structures existed in the Si-doped GaN layer, and after growth of quantum wells on it the density of defects drastically reduced and propagation of some of the defect levels ended up underneath the quantum wells, as shown in Fig. 1(a). This could be due to partial reduction of strain in the layers. A transmission electron microscopy (TEM) micrograph showed a quantum dot-like structures in the wells due to segregation of In [Fig. 1(b)]. The dark regions of contrast formed between the well and barrier due to strain and In precipitation was high indicating In phase immiscibility with GaN, which continued into the cap layer of GaN deposited on the InGaN/GaN quantum wells.⁵ The segregation of In was high with increasing In concentration in the quantum wells. In order to shift emission wavelength from

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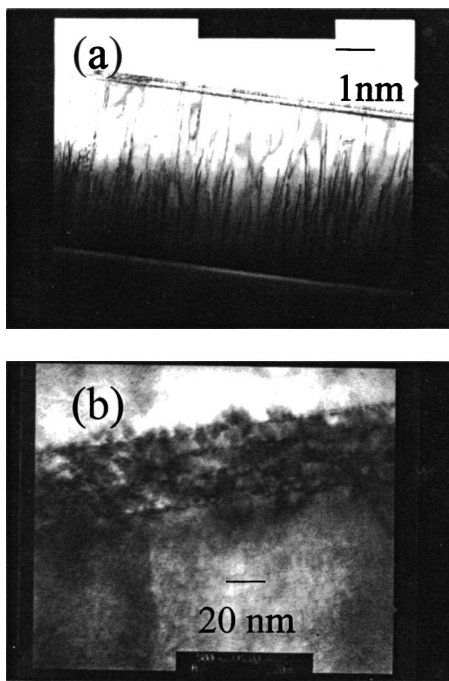


FIG. 1. (a) High resolution transmission electron micrograph bright field images, showing a decrease of the density of defect levels towards the growth direction. (b) Black contrast due to strain between the well and barrier, and quantum dot-like structures due to segregation of In from the InGaN wells.

blue to yellow, In percentage has to be increased, as shown explicitly from our results.⁶ The well and barrier period width was found to be ~ 9 nm.

XRD analysis revealed that the In composition and period were found to be 13.5% and 8.6 nm from simulation of a $\omega/2\theta$ scan of the (0004) curve, as shown in Fig. 2; the In composition obtained from XRD is quite low compared to that in photoluminescence (PL) analysis (In of 24%). The reason why is that the PL emission is basically from the active layers of quantum wells, which contained not only the quantum dot-like structures, which provided efficient radiative recombination centers rather than ordinary layers, the In-rich GaN matrix favors longer wavelength emission but also the quantum confined Stark effect whereas XRD analysis showed an average approximate In composition in the well layers.⁶

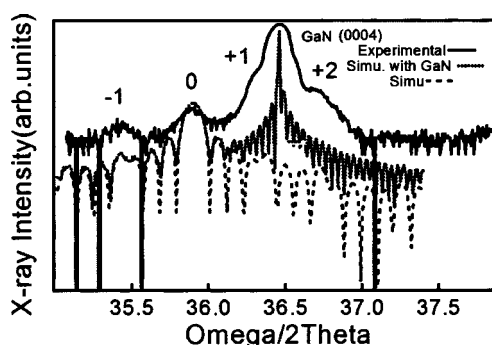


FIG. 2. Double crystal x-ray diffraction spectrum of a $\omega/2\theta$ scan of (0004) for an InGaN/GaN multiquantum-well green LED structure: (a) experimental data (solid line), (b) simulation (dashed line), giving In composition of 13.5% in the wells and period of 8.6 nm obtained from satellite peak positions $-1, 0, +1$ etc., taking into account the GaN thick base layer, and (c) simulation (dotted line) without taking into account the GaN layer.

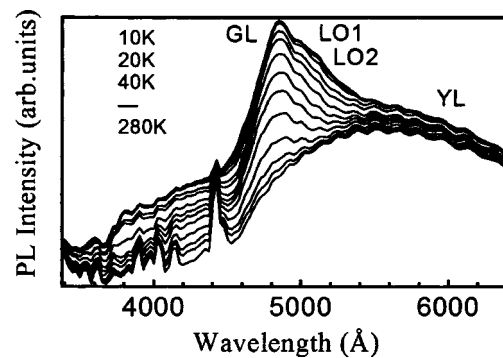


FIG. 3. PL spectra of InGaN/GaN multiquantum-well green LEDs measured at different low temperatures: the quantum-well resonance peak (GL) shows “S” shaped shift, i.e., red-blue-red shift with variation of the temperature.

The green luminescence (GL) peak due to quantum-well resonance at 485 nm (2.557 eV) showed “S” shaped shift with a decrease in temperature from room temperature to 10 K, as shown in Fig. 3. Two phonon replicas (LO1 and LO2) existed with a difference in energy of about 71 meV, which coincides with the reported value of 73 meV (588 cm^{-1}), whereas in GaN the phonon separation energy is 91 meV.⁷ The quantum-well resonance emission peak showed a redshift at lower temperature of 10–60 K because the (electron hole pairs) carriers, which are randomly distributed in the potential minima, do not have sufficient thermal energy so that they can relax down to lower energy level states by reducing higher emission energy. In the temperature range of 80–140 K, the emission peak showed a blueshift, indicating the carriers do have sufficient thermal energy to occupy higher energy level states by emitting higher energy radiation. Again the emission peak showed a redshift due to the dominance of nonradiative recombination at near room temperature.^{6,8} The activation energy was found to be 49 meV from the PL intensity versus inverse temperature, which is higher than the reported value of the blue-light-emitting diode, because, as expected, the In alloy fluctuations increased in the InGaN layers with increasing In composition, therefore the activation energy is also high.⁹ The other wide yellow luminescence (YL) peaked at 2.45–2.41 eV is due to secondary phase of InGaN, and the activation energy was found to be 10 meV. Note that the higher activation energy of 60 meV was reported for yellow luminescence of bulk GaN,¹⁰ therefore the presence of yellow luminescence is due to InGaN not GaN. A similar kind of strong parasitic emission with different peak position and activation energy for the InGaN secondary phase in the green-light-emitting diodes was reported in the literature.¹¹

In the InGaN/GaN quantum-well (QW) structure, the amount of piezoelectric polarization (P^{Pz}) and spontaneous polarization (P^{Sp}) in the direction of the c axis can be found by using the following equations.¹²

$$P^{Pz} = 2 \frac{a - a_0}{a_0} \left(e_{31} - e_{33} \frac{C_{13}}{C_{33}} \right) \quad \text{and} \quad (1)$$

$$P^{Sp}(x) = -\{0.032(1-x) + 0.029x\} C/m^2,$$

where a/a_0 is the lattice constant of either the well or the barrier, e_{31} and e_{33} the piezoelectric constants, and C_{13} and C_{33} the elastic constants, respectively. The samples are con-

sidered Ga-face layers. The electric field in the well and barrier can be determined by using the expressions,

$$F_W = \frac{L_B}{\epsilon_B L_W + \epsilon_W L_B} [|P_B^{pz}| + |P_B^{sp}| - |P_W^{sp}| - |P_W^{pz}|], \quad (2)$$

$$F_B = \frac{L_W}{\epsilon_B L_W + \epsilon_W L_B} [|P_W^{sp}| + |P_W^{pz}| - |P_B^{pz}| - |P_B^{sp}|], \quad (3)$$

where ϵ is the dielectric constant, L is the width, and $B(W)$ represents the barrier (well).

The QW ground level transition energy can be estimated as

$$E_{1e1h} = E_g - eF_W L_W + \left(\frac{9\pi\hbar e F_W}{8\sqrt{2}} \right)^{2/3} \left(\frac{1}{m_e} + \frac{1}{m_h} \right)^{1/3}, \quad (4)$$

where E_g is the band gap of the quantum well, m_e the electron effective mass, and m_h the hole effective mass. In Eq. (4), the second term represents the redshift due to the electric field in the well and the third term is for the blueshift due to electron and hole confined states. The quantum-well ground level emission (E_{1e1h}) value was found to be 2.557 eV, E_g value of 2.523 eV and electrostatic field strength of 1.0×10^5 V/cm for the $\text{In}_{0.24}\text{Ga}_{0.76}\text{N}/\text{GaN}$ quantum well, considering InN $E_g=0.77$ eV. The QW ground level emission shift of 34 meV (2.557–2.523 eV) was due to the dominance of the quantum confined Stark effect.¹³

In summary, the decrease of defect density propagation towards the growth direction, quantum dot-like structures due to In phase separation in green LEDs, were observed and the well and barrier widths obtained from TEM analysis coincided with those of the XRD results. The low temperature PL studies showed “S” shaped shift, i.e., red–blue–red shift for quantum-well emission and it is attributed to potential

fluctuations caused by the InGaN alloy inhomogeneity. The higher activation energy of 49 meV for green LEDs is an indication of the deep localization energies of excitons.

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