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Luminescence of the InGaN/GaN blue light-emitting diodes

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Abstract

InGaN/GaN double heterostructure (DH) and multiple quantum wells(MQW) light-emitting diodes were grown by metalorganic vapor phase epitaxy(MOVPE). Band gap narrowing of the PL spectra for the InGaN/GaN MQW LEDs can be observed at room temperature. In addition, the emission wavelength of EL and PL spectra for the MQW blue LEDs exhibit a blue-shift phenomenon when increasing the injection current and laser power, respectively. This luminescence behavior can tentatively be understood as a competition between a spectral red-shift mechanism of piezoelectricity-induced quantum-confined Stark effect(PQCSE) and a blue-shift mechanism of band-filling and charge screening effects.

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I. Introduction

In the past several years, the most studied of wide bandgap semiconductors have been the III-nitrides. Among them, GaN and its alloys with InN and AlN have attracted numerous attentions since the successful commercialization of bright blue/green light-emitting diodes followed latter by the demonstration of injection lasers[1-3]. GaN-based nitride semiconductors have several advantages over other wide bandgap semiconductors such as SiC and diamond. They can be doped both p- and n-type , have direct bandgaps, and can form heterostructures conducive to device applications. Blue/green laser diodes have been also achieved in ZnSe and related materials. However, the short lifetime prevent ZnSe-based devices from commercialization at present. It is considered that the short lifetime of these ZnSe-based devises is caused by crystal defects at a density of $10^3/cm^2$, because one crystal defect would cause the propagation of other defects leading to failure of the devices. By contrast the III-V nitrides are mechanically strong and chemically rather inert and do not have any gross reliability problems, at least judging from the preliminary studies of LED and LD operation.

II. Experiment

InGaN/GaN double heterostructure(DH) and multiple quantum well(MQW) light emitting diodes(LEDs) were grown by MOVPE system using a high-speed rotating disk in a vertical growth

In *Display Technologies III*, I-Wei Wu, Heiju Uchiike, Editor,
Proceedings of SPIE Vol. 4079 (2000) • 0277-786X/00/$15.00

chamber. Briefly, trimethylgallium (TMGa) and ammonia (NH_3) were used as Ga and N precursors, respectively. Biscyclopentadienyl magnesium (CP_2Mg) and Si_2H_6 were employed as the p-type and n-type dopant, respectively. The trimethylindium (TMIn) was employed as the In precursors. The carrier gas was hydrogen, and the growth pressure was maintained at 100 torr for the growth of Si-doped GaN film. The carrier gas was nitrogen, and the growth pressure was maintained at 300 torr for the growth of InGaN layers. The typical growth procedures are described as follows: Before growing III-V nitride films, the substrates were treated by thermal baking at 1100.in hydrogen to remove surface contamination. A low-temperature GaN nucleation layer, with thickness of about 300 Å, was grown at 560.. After the growth of low-temperature GaN nucleation layer, the wafer temperature was raised to 1060 .to grow the Si-doped GaN buffer layer. Figure 1(a) and Fig. 1(b) shows the schematic structure of the MQW and DH blue LED, respectively, which consists of 300 Å-thick GaN nucleation layer grown at a low temperature of 560., a 3.5 μm-thick layer of Si-doped GaN(electron concentration of about $8x10^{17}/cm^3$), 9 period of InGaN/GaN MQW structure consisting of 30 Å-thick $In_{0.3}Ga_{0.7}N$ well layers and 70 Å-thick GaN barrier layers (or 500 μm-thick bulk $In_{0.13}Ga_{0.87}N$ layer with Si and Zn coping)grown at a temperature of 780.. Finally, a 0.3 μm-thick Mg-doped contact layer(hole concentration of about $3\sim5x10^{17}/cm^3$) were grown. For fabrication of LED chips , the processing procedures were described as follows: (a) Ni was deposited onto the epi-wafer as the etching mask before ICP dry etching[4]. (b) the p-GaN layer was partially etched until the n-GaN was exposed. (c) An ultra thin Ni/Au(2 nm/6 nm) bi-layer was evaporated onto the p-GaN layer as the transparent p-type electrode[5]. (d) A Ti/Al(50 nm/2000 nm) bi-layer was deposited onto the n-GaN surface as the n-type electrode. (e) Wafer lapping was performed until the thickness down to 80 μm. Then the wafer was polished to remove the stress which results from the large roughness difference between the lapped surface and epitaxial surface. (f) The thin wafer was cut into a squared shape (350 μm ×350 μm) by diamond scriber and cutter.

III. Results and discussions

Figure 2 shows the electroluminescence (EL) spectra of the $In_xGa_{1-x}N$/GaN DH blue LED at forward dc currents of 5 mA, 20 mA and 60 mA. A typical peak wavelength and FWHM of the EL spectra were 468 nm and 76 nm, respectively, at current of 20 mA. The spectra exhibit blue shift as the injection current increases, as shown in Fig 2. The peak wavelength is 470 nm at 5 mA, 468 nm at 20 mA, and 462 nm at 40 mA. This blue shift of EL spectra suggests that the luminescence mechanism is donar-to-acceptor transition in the InGaN active layer codoped with both Zn, Si. At 60 mA, a shorter-wavelength peak emerges around 402 nm, as shown in Fig 2. In order to better understand the luminescence properties, the semilogarithmic EL spectra were performed for current ranging from 5 mA to 500 mA. At currents above 60 mA, 500 μs pulses at 1% duty cycle were used to prevent heating. At currents above 60 mA, the intensity of the shorter-wavelength peak at 402 nm related to blue band begins to increase more rapidly than the blue band, as shown in Fig 3. However, the position of the shorter-wavelength peak(402 nm) is fixed even though the injection current is further increased. In addition, the blue band exhibits blue shift as the injection current increases. Similar EL spectra have reported by Nakamura et al[6] and Lester et al[7] for the DH blue LEDs which with Zn, Si codoped InGaN active layer. But, as the active layers in InGaN/AlGaN DH LEDs with similar structure are not compensated, shifting peak spectra are not observed[3]. The shorter-wavelength peak could be attributed to the band-to-band transition in the InGaN active layer. In other words, the 402 nm emission peak corresponds to the energy gap for $In_{0.13}Ga_{0.87}N$ and therefore is due to radiative recombination of electrons in the conduction band and holes in the valence band. This peak becomes resolved at higher injection levels where the impurity-related recombination is saturated. Electroluminescence with shifting peak spectra has been observed and analyzed for GaAs p-n junction[8]. In GaAs , the two mechanisms for shifting peak spectra are photon-assisted tunneling and band filling. In photon-assisted tunneling, the hole and electron tunnel into the depletion region where they recombine with the emission of a photon. As shown in Fig 3, for the $In_{0.13}Ga_{0.87}N$/GaN DH blue LED, the emission intensity of the blue band does not saturate as the injection current is increased. Thus, the emission spectra shown in Fig 3. are consistent with the band filling mechanism. In band filling, the shift in the main peak results from the minority carriers(holes) filling the empty acceptor levels and valence band tails in the co-doped and therefore compensated active layer. The InGaN active layer of the blue LED is heavily doped with both the Si and Zn. Thus, the Zn acceptor level is expected to exhibit Gaussian broadening and both the valence band and the conduction band will have band tails[9-10].

For an $In_{0.3}Ga_{0.7}N$/GaN MQW LED, the emission peak should be equal to or less than 450 nm. However, the emission peak of the $In_{0.3}Ga_{0.7}N$/GaN MQW LED is around 465 nm. The energy difference between the strained-free band-edge emission peak of $In_{0.3}Ga_{0.7}N$ bulk layer, which the In content was determined by PL measurement, and the PL peak wavelength of the $In_{0.3}Ga_{0.7}N$/GaN MQW LED is approximately 80 meV. Notice that the growth condition of $In_{0.3}Ga_{0.7}N$ bulk layer is the same as the active layers (well region) of the MQW LED. Plausible causes of such a band gap narrowing effect have been tentatively proposed by Nakamura but no quantitative explanations have been made[11]. The possible explanations of this band gap narrowing of $In_xGa_{1-x}N$/GaN quantum well can be attributed to the exciton effects(Coulomb effects correlated to the electron-hole pair) of the active layer or strain effects caused by the lattice mismatch and the thermal expansion coefficients difference between well layers and barrier layers. In the former case, it might be anticipated that radiative recombination in these devices is related to the presence of highly localized excitons , localized on fluctuations of indium contents in the active layer. In other words, a continuos density of states may be present within the band gap of the active layer. Perhaps, the emission mechanism of the $In_xGa_{1-x}N$/GaN multi-quantum well LEDs results from a competition between above-mentioned effects and quantum confinement.

In order to better understand the luminescence properties of the $In_xGa_{1-x}N$/GaN multi-quantum wells, the excitation power dependence of the PL spectra were performed at room temperature. Figure 4. shows the PL spectra of $In_{0.3}Ga_{0.7}N$/GaN multi-quantum well LED measured at different excitation power. In the MQW blue LEDs, the well region of the active layer is $In_{0.3}Ga_{0.7}N$, and its band-edge emission peak is around 450 nm if there were strained-free in the QW structure. The emission peak of the MQW blue LED should be shorter than 450 nm when the quantum size effect is in action. However, our $In_{0.3}Ga_{0.7}N$/GaN MQW LED exhibits the peak wavelength of around 465 nm. This unexpected observation may be caused by the piezoelectricity-induced quantum-confined Stark effect(PQCSE)[12-13]. In other words, it may tentatively be understood as an effect of the QW potential, which was severely distorted by the piezoelectric field . In addition, it is clear that the PL peak shifts toward shorter wavelength when the laser power is increased. Between $I=I_o$ and $I=I_o/20$ of excitation intensity, the shift can be as large as 8.5 nm (50 meV). Basically, the transition energy of the InGaN strained QWs was smaller than that of unstrained QWs since the band alignment of InGaN strained well layer was tilted by piezoelectric fields. When the samples were pumped with the excitation sources, the piezoelectric fields in the InGaN strained well layer were screened by generated carriers, thus weakening the PQCSE. Increasing the excitation intensity further weakened the PQCSE and increased the transition energy, that is , blueshift occurred.

To further explore the origin of the band gap shifting effect of $In_{0.3}Ga_{0.7}N$/GaN quantum well, the EL measurements were performed at room temperature with various driving current. Figure 5 shows the EL spectra for current ranging from 1 mA to 500 mA. At currents above 60 mA, 500 μs pulses at 1% duty cycle were used to prevent heating. The EL peak energy of the MQW blue LED exhibits blueshift when the injection current is increasied. The blueshifts are about 140 meV as the forward current are increased from 1 mA to 500 mA., In addition to the description mentioned-above, these blue shifts may also be due to a band-tail filling effect. According to the report of Narukawa et. al[14], the peak of the spontaneous emission shifts toward the high energy side with increasing excitation intensity. This emission behavior is only observed in the LED which have heavily doped and compensated active layer[14]. However, the active layer of the MQW blue LED which used in this study is undoped. The blueshifts could be attributed to a filling of band-tail states(i.e., localized states) where carrier or excitons are recombined for emission with increasing injection current. The localized states may be formed by indium composition fluctuation in the $In_{0.3}Ga_{0.7}N$ well layer due to a phase separation or indium segregation of the $In_{0.3}Ga_{0.7}N$ during growth. In other words, the radiative recombination may be attributed to excitons localized at deep traps which probably originate from the In-rich region in the well as quantum dots[14]. Recently, Peng et. al [13] reported that a spectral blueshift was observed as the injection current increases from 1mA to 1A. According to their repotrs, the emission spectrum of the InGaN QW is determined by a competition between a spectral redshifting mechanism of PQCSE and a blueshifting mechanism of band-filling and charge screening effects.

IV. Conclusions

In summary, the InGaN/GaN DH and MQW LEDs were fabricated. The peak wavelength of the typical EL spectra at 20 mA is about 465 nm. The FWHM of the typical EL spectra at 20 mA are about 70 nm and 30 nm for DH and MQW LEDs, respectively. The electroluminescence for the Si, Zn codoped InGaN active layers demonstrated spectra whose emission peak shifted to shorter wavelength as the injection current was increased which may be attributed to band filling. In addition, the blue shift and band gap narrowing of the EL and PL spectra for the InGaN/GaN MQW may tentatively be understood as a competition between a spectral redshifting mechanism of PQCSE and a blueshifting mechanism of band-filling and charge screening effects.

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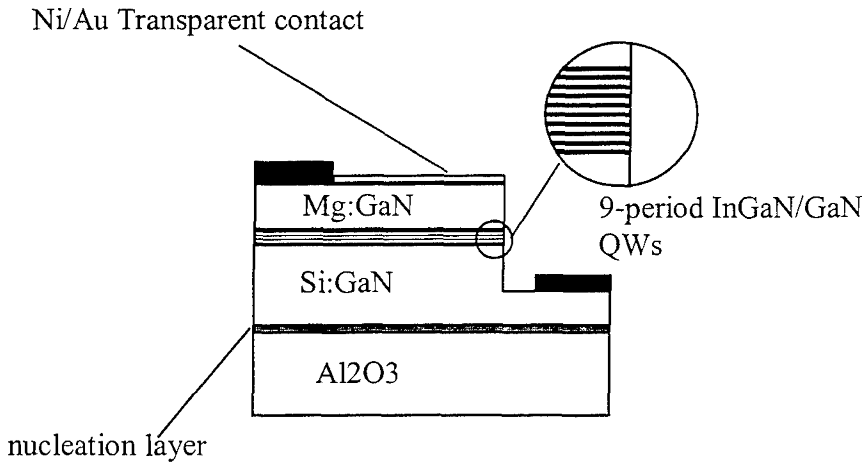
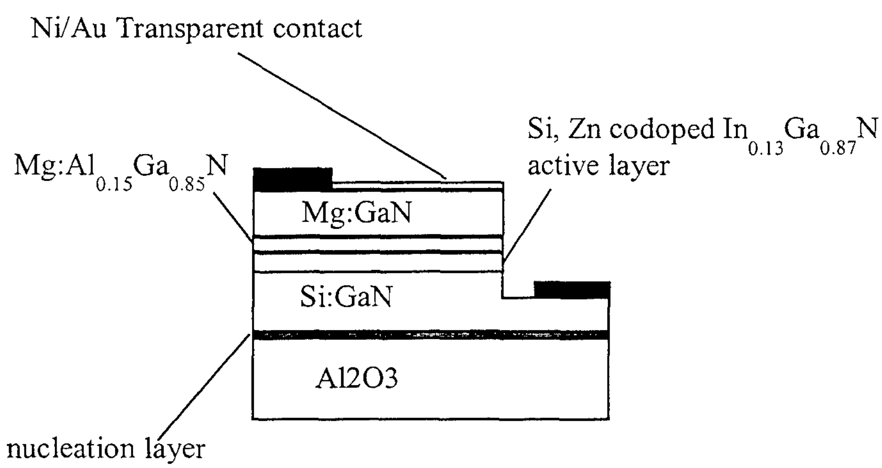


(a)



(b)

Figure 1. The schematic structures of the (a) MQW and (b) DH LED.

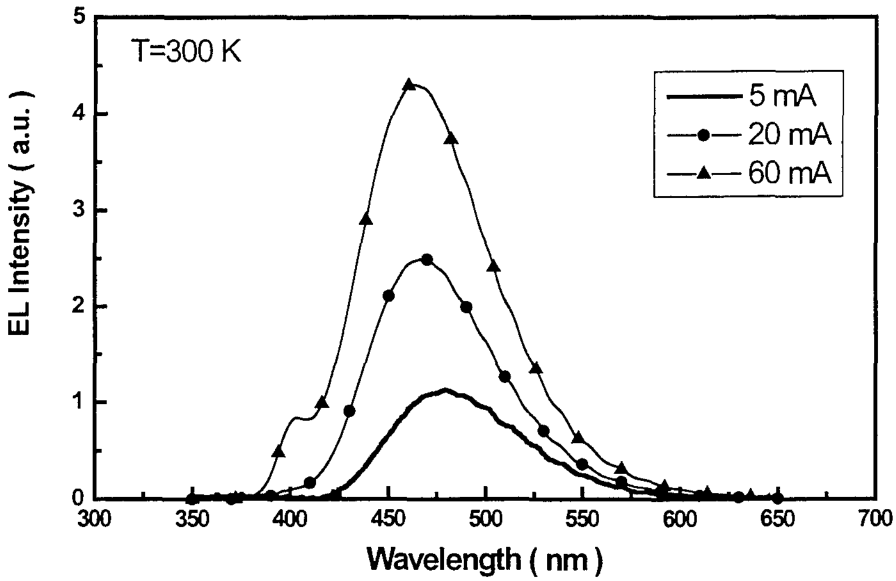


Figure 2. Electroluminescence spectra of the Si and Zn codoped InGaN/GaN DH blue LED at forward currents of 5 mA, 20 mA and 60 mA.

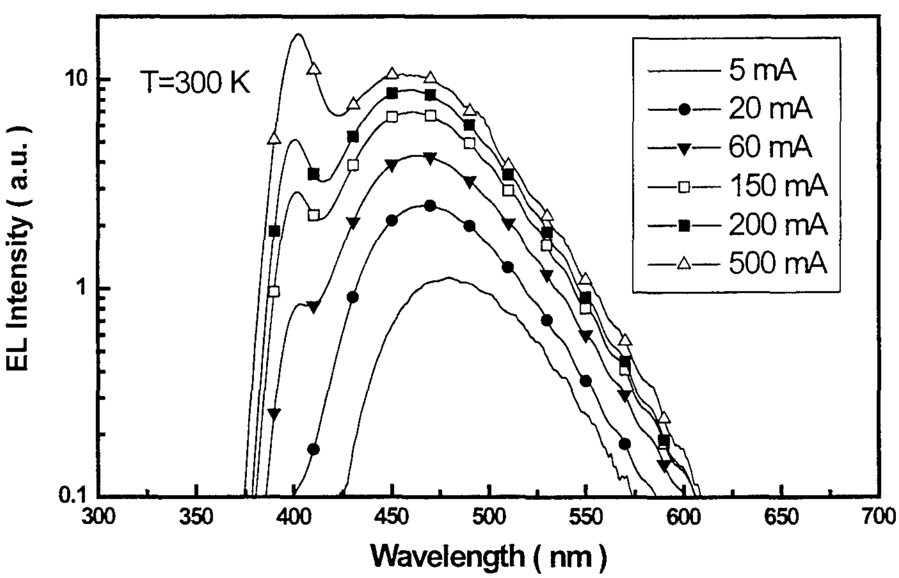


Figure 3. Semilogorithmic electroluminescence spectra of the Si and Zn codoped InGaN/GaN DH blue LED for current ranging from 5 mA to 500 mA.

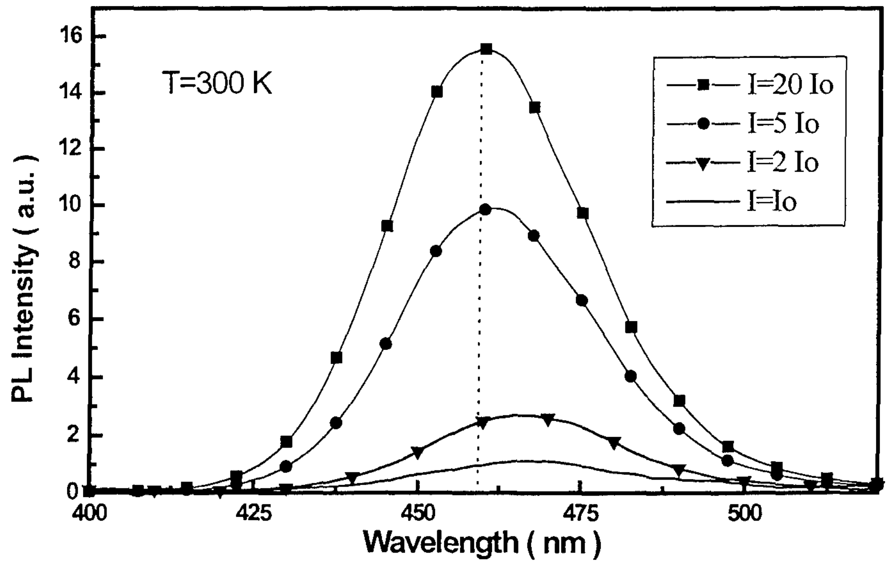


Figure 4. The PL spectra of $In_{0.3}Ga_{0.7}N$/GaN multi-quantum well LED measured at different exciting intensity.

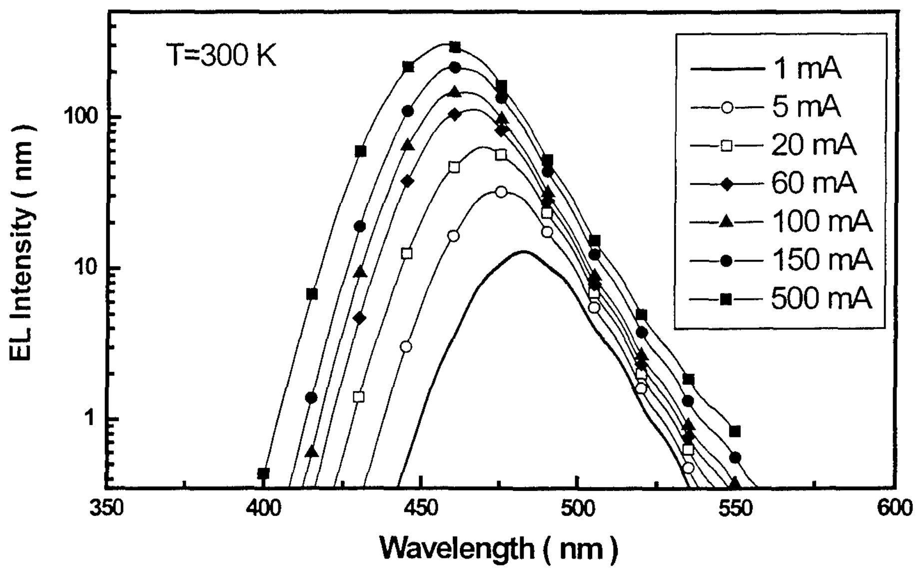


Figure 5. The EL spectra of InGaN/GaN MQW blue LED at different injection current.