InGaN/GaN multi-quantum dot light-emitting diodes

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Abstract

It has been demonstrated that InGaN/GaN blue light-emitting diodes (LEDs) with multiple quantum dot (MQD) were successfully fabricated by metal-organic chemical vapor deposition (MOCVD). We have formed nanoscale InGaN self-assembled QDs in the well layers of the active region with a typical 3-nm height and 10-nm lateral dimension. With a 20-mA DC injection current, the forward voltage was 3.1 V and 3.5 V for MQD LED and conventional nitride-based multi-quantum well (MQW) LED with the same structure, respectively. It was also found that EL peak position of the MQD LED is more sensitive to the amount of injection current, as compared to the conventional MQW LEDs.

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1. Introduction

Heteroepitaxial growth of highly strained material systems has been quite attractive as it offers the possibility of producing low-dimensional carrier confinement nanostructures, such as quantum wells (QWs) and quantum dots (QDs) [1]. They present the utmost challenge to semicon-
ductor technology, making possible fascinating novel devices. III-nitride semiconductor materials have a wurtzite crystal structure and a direct energy band gap. We could also achieve nitride-based heteroepitaxial growth easily. At room temperature, the band gap energy of AlInGaN varies from 0.7 to 6.2 eV depends on its composition. Therefore, III-nitride semiconductors are particularly useful for light-emitting diodes (LEDs) and laser diodes (LDs) in this wavelength region [2-5]. Typical high-brightness LEDs have a multiple quantum well (MQW) active region. The MQW LED is a kind of heterojunction device, in which electrons and holes are confined in the well layers. Thus, one can achieve high quantum efficiency from the MQW LEDs since carrier can recombine easily in the confined well layers [6-9]. Although high brightness InGaN-GaN MQW LEDs are already commercially available, it can be theoretically predicted that the realization of LEDs with QDs in the active layer would improve the performance of LEDs.

Recently, it has been shown that nitride nanostructures can be self-assembled using the strain-induced Stranski-Krastanov (S-K) growth mode without any substrate patterning process [10-12]. It has also been shown that nitride nanostructures can be self-assembled using growth interruption during the metal-organic chemical vapor deposition (MOCVD) growth [13]. Although the size fluctuations of self-assembled QDs could result in inhomogeneous optical and electrical characteristics, the self-assembly of strain-induced islands provides the means for creating zero-dimensional quantum structures without having to overcome the current limitations of lithography. These self-assembled QDs could also be used to study novel device physics [14-16]. In this work, we report the successful fabrication of blue LEDs with multiple InGaN dots-in-a-well (DWELL) structure, i.e. InGaN-GaN multiple quantum dot (MQD) LEDs, grown on 2-inch sapphire substrates using a growth interruption method.
in MOCVD system. Details for the formation of QDs are reported in this study. In addition, we discuss the optoelectronic characteristics (including electro-luminescence and current-voltage measurements) of the In-GaN-GaN MQD LEDs compared with conventional MQW LEDs.

2. Experiments

Samples used in this study were grown on (0001)-oriented 2-inch sapphire (Al₂O₃) substrates in a vertical low-pressure MOCVD reactor with a high-speed rotation disk [6-9]. Briefly, the gallium, indium and nitrogen sources were trimethylgallium (TMGa), trimethylindium (TMIn), and ammonia (NH₃), respectively. Biscyclopentadienyl magnesium (CP₂Mg) and disilane (Si₂H₆) were used as the p-type and n-type doping sources, respectively.

InGaN-GaN LEDs with multiple DWE LL were then fabricated as Fig. 1. Prior to growth, sapphire substrates were thermally baked at 1100°C in hydrogen gas to remove surface contamination. After a 30-nm-thick low-temperature GaN nucleation layer was deposited onto the sapphire substrate at 500°C, the temperature was raised to 1000°C to grow the Si:GaN buffer layer. Subsequently, the temperature was ramped down to 730°C to grow the InGaN-GaN MQW active region with InGaN QDs. It should be noted that we introduced an interrupted growth method [13] so as to achieve InGaN DWE LL layers. In other words, we first deposited a 1.2-nm-thick InGaN layer, stopped the growth for 12 seconds, and then deposited another 1.2-nm-thick InGaN layer so as to achieve a 2.4-nm nominal thickness of InGaN. In the active region, each InGaN-GaN pair consists a 2.4-nm-thick InGaN DWE LL layer and a 15-nm-thick GaN barrier layer. The InGaN DWE LL layers were unintentionally doped. On the other hand, the GaN barrier layers were Si-doped with a doping concentration of 3 × 10¹⁷ cm⁻³. After growth of active region, the substrate
temperature was elevated to 1060°C again to grow the Mg-doped AlGaN cladding layer and Mg-doped GaN contact layer. In order to increase the indium incorporation rate, nitrogen was used as the carrier gas when we grew the InGaN-GaN MQD active regions. On the other hand, hydrogen was used as the carrier gas when we grew other parts of the samples. The growth pressure was kept at 350 mtorr throughout the growth. The as-grown samples were then annealed at 760°C for 25 min in N\textsubscript{2} ambient to activate the Mg-doped p-type layers. With this thermal annealing process, we could achieve uniformly doped highly conductive p-type layers. Conventional MQW LEDs in the same structure were also subsequently fabricated for comparison.

For DWELL structural characterization, the cross-sectional transmission electron microscopy (XTEM) specimen for TEM observation was prepared by the conventional method including cutting, gluing, mechanical polishing, dimpling procedures and then followed by the Ar+ ion-beam milling to perforation. A Philips CM200 field emission gun TEM equipped with a Gatan image filtering (GIF) system was operated at 200 kV to carry out the experiments. The high resolution XTEM image was taken along the GaN [11\textsubscript{2}0] direction and the dark-field image was obtained under two beam condition with g=[0002]. After the growth, the surfaces of the as-grown samples were partially etched until the n-type GaN layers were exposed. Ni-Au contacts were subsequently evaporated onto the p-type GaN surfaces to serve as the p-electrodes. On the other hand, Ti-Al-Ti-Au contacts were deposited onto the exposed n-type GaN layers to serve as the n-type electrodes, to complete the fabrication of the blue LEDs. Room temperature (RT) electroluminescence (EL) characteristics were then measured by injecting different amount of DC current into the fabricated LEDs on wafer without
polishing and package. The current-voltage (I-V) measurements were also performed at RT by using an HP4156 semiconductor parameter analyzer.

3. Results and discussion

Fig. 2(a) shows the XTEM image of DWELL structures in MQD LEDs. We can achieve nanoscale self-assembled QDs by using the growth interruption method during MOCVD growth [13], it can be seen that there are obvious DWELL structures in active region. The density of these QDs was estimated to range between $10^{10}$ and $10^{11}$ cm$^{-2}$ [13]. Since the all QDs exist in the well layer, the formation mechanism of QDs in this specimen should be strain-induced S-K epitaxial growth, not phase separation. Fig. 2(b) shows the high-resolution XTEM image for an In-GaN QD embedded in InGaN-GaN quantum well of the MQD LED structure, this picture reveals that typical dot is pyramidal with a 3-nm height and a 10-nm lateral dimension. The InGaN dot height (3 nm) observed from the high-resolution XTEM image of MQD structure very approach the exiton Bohr radius (2.8 nm for GaN [17], but it can be > 2.8 nm for InGaN with the smaller effective mass), note that the exciton binding energy considerably increases even when the dot lateral size (10 nm) is three times larger than the Bohr radius [18]. As a result, we can predict such a DWELL structure will reveal strong quantum localization effect.

Fig. 3(a) shows current-voltage (I-V) characteristics of the MQD LEDs and conventional MQW LEDs in the same structure, the 20-mA forward voltage is 3.1 V and 3.5 V respectively. The smaller forward voltage 3.1V shows better electrical performance on the fabricated MQD LEDs, as a result can be attributed to QDs are effective in reducing the forward voltage due to the 3-D spatial confinement effect of carriers. Fig. 3(b) illustrates the EL peak position as a function of injection current for
the MQD LED. It can be seen that the EL peak position blue shifts toward short wavelength side as the injection current increases. Such a blue shift in EL wavelength could be attributed to the band-filling effect of localized energy states [5, 19]. It should be noted that we observed a huge 68.4-meV EL blue shift as the injection current was increased from 3 to 50 mA for the MQD LED. Such a value is much more than the 38-meV EL blue shift observed from the conventional nitride-based MQW LED with a similar structure in the same current range. It is well-known that III-V nitride materials have a large piezoelectric effect, especially in strain-induced SAQD (self-assembled quantum dot) structures. The large piezoelectric field will induce quantum-confined Stark effect (QCSE) [20, 21]. QCSE results in a spatial separation of electrons and holes, and thus, the carrier recombination energy will become smaller. In this study, since QD structures have stronger QCSE, the large EL blueshift reveals that deep localization of excitons (or carriers) originates from QDs will strengthen band-filling effect as the injection current increases. Such a result also suggests that EL peak position of MQD LED is more sensitive to the amount of injection current as compared to conventional MQW LED.

4. Conclusion

In summary, InGaN-GaN blue LEDs with multiple DWELL structures have been successfully fabricated. With a 12 second growth interruption, we formed nanoscale InGaN self-assembled QDs in the well layers of the active region. It was found that typical dot is pyramidal with a 10-nm diameter and a 3-nm height. It was also found that EL position of the MQD LED is more sensitive to the amount of injection current, as compared to conventional MQW LEDs.
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References


**Fig. 1** Schematic for blue LED with DWELL structure. Note that In-GaN/GaN MQD as the active layers of LED.

**Fig. 2** (a) XTEM image of multiple DWELL structures in active region. (b) high-resolution XTEM image of a single QD in QW taken along the [1120] axis.
Fig. 3 (a) Current-voltage ($I$-$V$) characteristics of the MQD LEDs and MQW LEDs, the forward voltage and EL intensity is 3.1 V and 3.5 V at 20-mA injection current, respectively. (b) Dominated wavelength of EL spectra depends on injection current for the MQD LED and conventional MQW LEDs.