



# Fabrication and characterization of high quality n-ZnO/p-GaN heterojunction light emission diodes

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## ARTICLE INFO

### Article history:

Received 24 September 2009

Received in revised form 4 May 2011

Accepted 14 June 2011

Available online 21 June 2011

### Keywords:

Zinc oxide

Gallium nitride

Heterojunctions

Electroluminescence

## ABSTRACT

High quality single crystalline n-type ZnO film was grown on p-type GaN substrate using molecular beam epitaxy. Transmission electron microscopy reveals a sharp ZnO/GaN interface. Light-emitting diode was fabricated from this heterostructure, and a turn-on voltage of ~3.4 V was demonstrated. We found that the emission peak shifts from violet (430 nm) to near-ultraviolet (375 nm) when the driving current increases from 0.38 mA to 3.08 mA. This intriguing phenomenon can be understood by charged carrier's radical recombination occurring at both sides of the device, and the current enhancement of ZnO emission efficiency.

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## 1. Introduction

GaN-based short wavelength optoelectronic devices, such as blue-light-emitting diode, solid ultraviolet detector and laser diode [1–3], have been fabricated with marvelous commercial success, after the breakthrough in stable p-doping [4]. In terms of its large exciton binding energy (60 meV, compared to 26 meV for GaN at room temperature), available bulk material and permission for wet chemical etching [5–8], ZnO seems to become the next focus of research after GaN in wide bandgap semiconductor materials. The main challenge in ZnO is p-type doping. To avoid the problem, several groups used p-type thin films, like diamond [9], ZnRh<sub>2</sub>O<sub>4</sub> [10] and SrCu<sub>2</sub>O<sub>2</sub> [11] as substrates for n-ZnO growth to fabricate p–n junctions for light emitting diodes (LEDs). However, it incurs another problem: different crystal structures and large lattice mismatch make preparation of high quality films very difficult. GaN has the same wurtzite structure and a close lattice constant with ZnO, the n-ZnO/p-GaN heterostructure is probably among the best choices for fabricating ZnO light emitting devices along this line [12].

In previous work, different material growth methods were applied to produce n-ZnO/p-GaN heterojunction. The devices showed large diversity in their optical and electrical properties, especially the main emission peak of electroluminescence (EL) [13–16]. It is probably due to the poor quality of the ZnO films and heterostructures, which may

induce unexpected scattering and recombination. In this letter, we prepared the n-ZnO/p-GaN junction by radio-frequency plasma assisted molecular beam epitaxy (rf-MBE). With a better controlled junction structure, turn-on voltage of 3.4 V, which is identified with the ZnO (GaN) band gap, was observed on the current-voltage (I-V) characteristics. The main emission peak was found to shift from 430 nm to 375 nm with increased injection current. Compared with the room temperature (RT) photoluminescence (PL) result, we interpret the violet emission as originating from the p-GaN side, while the near ultraviolet peak from the n-ZnO side. We show that the emission wavelength blue-shift on driving current in the EL spectra can be explained by the higher light emitting efficiency of ZnO than GaN.

## 2. Experimental details

The schematic diagram of our n-ZnO/p-GaN heterojunction device is shown in Fig. 1. The 1 μm-thick Mg-doped GaN film was deposited on c-plane sapphire by metal-organic chemical vapor deposition. RT Hall measurement shows that the bulk hole density of the GaN film is about  $3 \times 10^{17} \text{ cm}^{-3}$ , and the mobility is  $5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ . On this substrate, a Ga-doped ZnO film with 1 μm thickness was grown by rf-MBE. Zn and Ga were evaporated from two Knudsen cells (Createch Fisher & Co. GmbH) at 430 °C and 300 °C, respectively. Oxygen was supplied by an rf-plasma source (SVT Associates, Inc.) with a gas flux of 2.0 sccm and an rf power of 400 W. The electron concentration of the n-ZnO sample is  $5 \times 10^{19} \text{ cm}^{-3}$ , and the mobility is  $30 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  at RT. After standard photolithography and wet chemical etching, 10 nm

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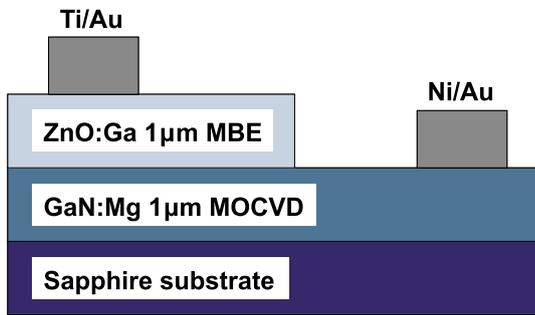


Fig. 1. Schematic diagram of cross-section view of the n-ZnO/p-GaN heterojunction structure.

Ni and 50 nm Au were deposited by rf-sputtering on GaN as the p-electrode, followed by thermal annealing to 500 °C for 10 min in air to facilitate the Ohmic contact formation [17]. 10 nm Ti and 50 nm Au were sputtered on the ZnO surface as the n-electrode [18,19]. The samples were investigated by cross-sectional transmission electron microscopy (TEM) at 200 kV in a Philips CM200 field emission gun. TEM specimens were prepared by the conventional mechanical grinding and polishing, followed by the ion beam thinning in a Gatan precision ion polisher system. I-V measurement and RT PL and EL were used to investigate the electrical and optical properties of the device. PL spectra measurement used 325 nm line of a He–Cd continuous wave laser. Emitted light was dispersed by a triple grating monochromator and detected by a GaAs photomultiplier tube.

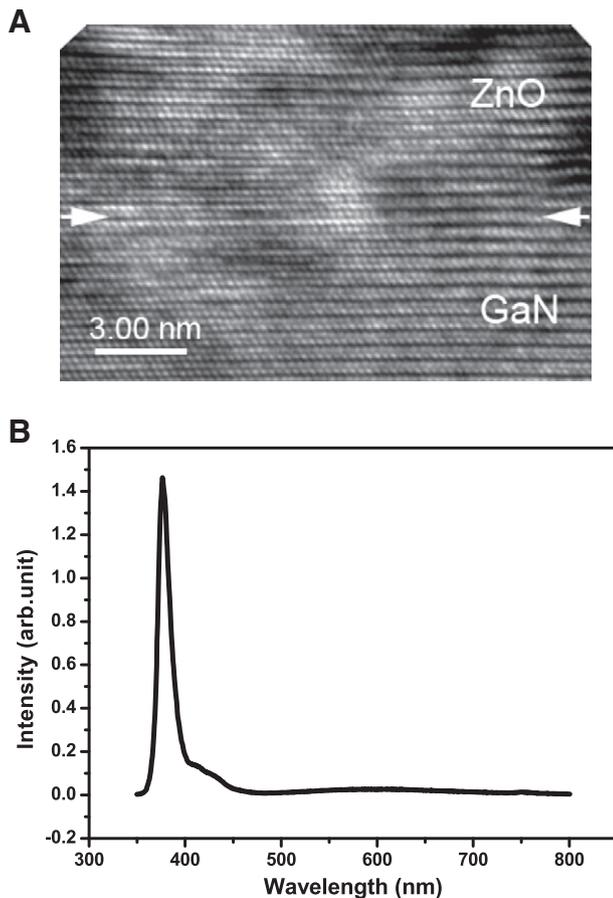


Fig. 2. (A) Cross-sectional TEM image of the n-ZnO/p-GaN heterostructure taken along  $\langle 11\bar{2}0 \rangle_{\text{ZnO(GaN)}}$  direction. (B) RT PL taken from the top ZnO layer.

### 3. Results and discussion

Fig. 2(A) shows a cross-section TEM image of the interface of the n-ZnO/p-GaN heterojunction with the electron beam parallel to  $\langle 11\bar{2}0 \rangle_{\text{ZnO(GaN)}}$  direction. Contrast to the case of ZnO deposited on other p-type materials, there is no obvious transition region at this heterostructure, which will scatter the charge carrier less severe than in the other structures in the references. RT PL (Fig. 2(B)) shows clean ultra-violet emission with peak located in 375 nm, free from green-yellow emission band, which comes from the impurity and defects. The high crystal quality of our ZnO film is believed to be a result of the lattice matched epitaxial growth, as well as the similarity of crystal structures and physical properties between ZnO and GaN. With such sharp interface indicated by TEM images and high quality ZnO film characterized by PL spectrum, our n-ZnO/p-GaN LED is expected to have a good device performance. As seen in Fig. 3(A), a nearly identical rectified I-V curve to homojunction is obtained from our LED. The turn-on voltage is approximately 3.4 V, which is very similar to the bandgaps of ZnO and GaN. The relatively low current is attributed to the big series resistance, which is caused by the large separation between the electrodes on the device ( $\sim 1$  mm) and the high resistance of p-type layer. The improvement of I-V characteristic is expected when the device is small.

EL spectra under different dc forward driving currents are shown in Fig. 3(B). When the current is 0.38 mA, a broad emission with a

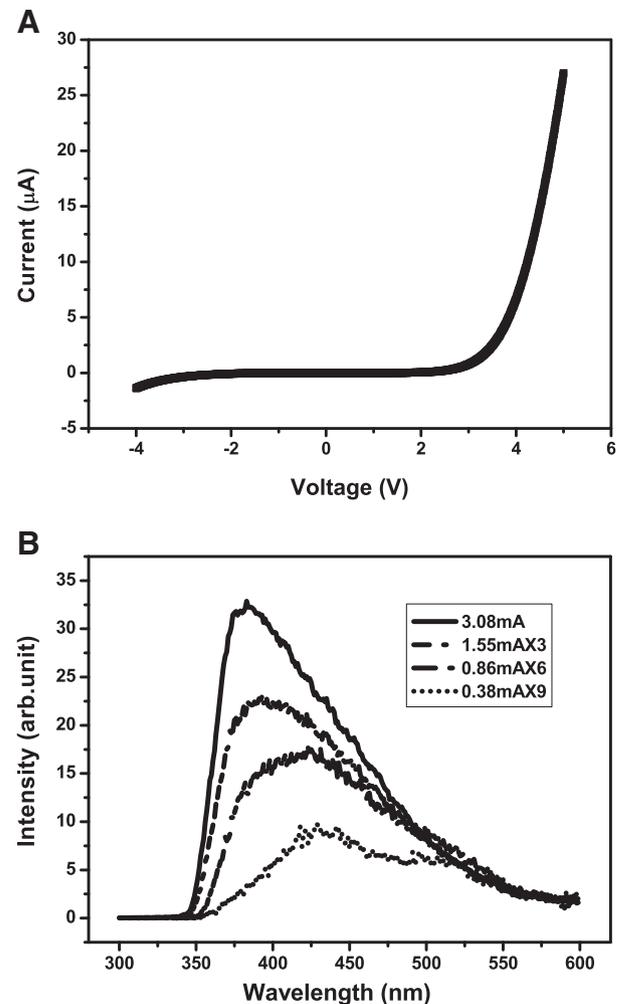


Fig. 3. (A) Current-voltage characteristic of the n-ZnO/p-GaN heterojunction LED measured at room temperature. (B) RT EL spectra of the device under different forward dc driving currents.

weak peak at 430 nm is presented. As the current is increased to 0.86 mA, a shoulder peak at 375 nm is observed. When a larger current (1.55 mA) is applied, the 375 nm peak becomes stronger than the peak at 430 nm. The dominated emission peak remains at 375 nm, when the driving current of LED is greater than 3.08 mA.

The EL emission with a low current is considered to be from the p-type side of the device, because of much higher electron concentration in the n-region by one or two orders of magnitude than the hole concentration in the p-region. This is in agreement with that the homojunction GaN LEDs exhibit a max wavelength at 430 nm [1,20], due to the transition from conduction band edge (CBE) to the Mg acceptor energy level in p-type side of the diodes.

In higher current, the emission spectra were proposed to contain two components: one is from the p-GaN region, and the other from the n-ZnO region. As aforementioned, the emission from p-GaN is located at 430 nm, whereas the recombination in the Ga-doped n-ZnO generates 375 nm near-ultraviolet light according to our RT PL result. At low current, the EL is determined by the emission from the p-GaN layer. In this case, the peak at 430 nm is strong. When the forward current increases, the emission from the n-ZnO layer becomes more important. Since the interface is atomically sharp and the interface scattering can be reasonably ignored, the hole injection from p-GaN side to n-ZnO side would play an important role. The recombination in ZnO side increases rapidly with increasing current, and eventually dominates the emission at certain high driving current. The current-enhanced light emitting from the ZnO side can be understood by the higher light emitting efficiency of ZnO than GaN due to its larger exciton binding energy. This phenomenon can be attributed to the high quality films and sharp interface, which allow the charge carrier pass through more freely and radical recombinant in both sides of the device.

#### 4. Conclusion

In summary, we have grown single-crystalline Ga-doped ZnO film on p-GaN/sapphire by rf-MBE. Due to the atomically sharp interface and high film quality, the turn-on voltage of the device is only 3.4 V. The main peak of the EL spectra shifts from 430 nm to 375 nm with

increasing driving current. We interpret it as a current-enhanced emission in the n-ZnO layer.

#### Acknowledgement

The work was financially supported by National Science Foundation of China (50532090, 60606023, 60621091) and the Ministry of Science and Technology of China (2007CB936203, 2009CB929400, 2009CB623700).

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