

Nonpolar light emitting diode made by m-plane n-ZnO/p-GaN heterostructure

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Abstract: Nonpolar (100) m-plane n-ZnO/p-GaN light-emitting-diodes (LEDs) were grown by chemical vapor deposition on p-GaN templates which was grown by metalorganic chemical vapor deposition on LiAlO₂(100) substrate. Direct current (DC) electroluminescence (EL) measurements yielded a peak at 458nm. The EL peak position was independent of drive current and a full width of half maximum (FWHM) of 21.8 nm was realized at 20mA. The current-voltage characteristics of these diodes showed a forward voltage (V_f) of 6V with a series resistance of $2.2 \times 10^5 \Omega$.

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The current commercial technology for III-nitride-based and ZnO based LEDs employs films grown in the polar c direction [1,2]. However, conventional c-plane III-nitride-based and ZnO based LEDs suffer from the quantum-confined Stark effect [3,4] due to the existence of piezoelectric and spontaneous polarization [5]. The growth of nonpolar (Al, Ga, In)N and ZnO have attracted great interest for its potential use in the fabrication of nonpolar electronic and optoelectronic devices [6,7]. The built-in electric fields along the c-plane direction cause spatial separation of electron and holes that in turn gives rise to restricted carrier recombination efficiency, reduced oscillator strength and red-shifted emission [8–21]. In the last few years, a lot of research group have also attempted different approaches to avoid the built-in electric field by using non-polar QW [8–12], c-plane QW with larger optical matrix element [13–21], valence subband rearrangement [22–25] approaches in III-Nitride semiconductors.

In this study, we report on the growth and electronic and luminescence characteristics of CVD-regrown nonpolar m-plane n-ZnO/m-plane p-GaN heterojunction LEDs using MOCVD-grown p-GaN/LiAlO₂(100) template. In particular, the effect of emission wavelength and linewidth in the electroluminescence (EL) emission spectra were investigated.

The LED structure, as shown in Fig. 1(A), was regrown by CVD on p-GaN/LiAlO₂ template fabricated by metal organic vapor phase deposition (MOCVD). GaN epilayer with p-type dopant were grown on LAO (1 0 0) substrates in a Thomas Swan MOCVD system. This system used a 3 × 2" Close- Coupled Showerhead reactor. TMGa, CP₂Mg, and NH₃ were used as the sources of Ga, Mg, and N, respectively. Before growth, the substrate loaded in the growth chamber was initially treated at 1000°C for 120 s for further out-gassing in N₂ ambient. A two- step growth method was then applied to grow p-GaN epilayer. First, an AlN nucleation layer was grown on the LAO substrate at 660, 680, and 700°C in N₂ ambient at 200 mbar for 30 s. The ambient was then changed to H₂ and the substrate temperature and pressure were changed to 1050 °C and 100 mbar, respectively, to grow a 0.6-mm-thick p-GaN layer at a growth rate of 1 μm/h [26]. In situ normal incidence reflectance was measured throughout the growth process by illuminating the samples with a tungsten lamp. Light reflected through a spectrometer was collected by a photodiode [27]. The regrowth of ZnO, carried out in a horizontal CVD reactor, Zinc shots(99.9999%) were placed in a quartz boat as the Zn source in the center of a quartz tube in a furnace. The quartz tube was kept at 1 atmosphere pressure by flowing high purity Ar (99.99%) with a flow rate of 100 sccm and heated up to 800°C. When the temperature was reached up to 800°C, high purity oxygen (99.9999%) was introduced with a flow rate of 5 sccm for the growth of the ZnO films. The thickness of n-ZnO layer is independent of the amount of Zn shots because we do believed that the Zn were evaporated in a very short time when the saturated vapor pressure reached. The morphology of the as grown ZnO film on GaN/LiAlO₂ template was examined by a JEOL JSM-7000F field emission scanning electron microscopy (SEM). X-ray diffraction (XRD) experiments were carried out on a Bede triple-axis diffractometer system, using Cu Kα1 (λ = 1.5406 Å) radiation. Photoluminescence (PL) was measured with a continuous wave He-Cd 325 nm laser with power density incident on the samples ~1 W/cm². The carrier concentration of n-ZnO is 5 × 10¹⁹ cm⁻³ and mobility of n-ZnO is 12 cm²/(V-s), which were measured by Hall measurement system (Bio-Rad HL5500PC). Diode mesas were defined by reactive ion etching (ICP). Ni and Cr/Au were used as n-ZnO and p-GaN contacts, respectively. The electrical and luminescence characteristics of the diode were measured by on-wafer probing of the devices. The I-V measurements were performed with Agilent 4156C semiconductor parameter analyzer.

Figure 1(B) shows photoluminescence spectra measured at 10 K of the m-plane ZnO regrown on m-GaN/LiAlO₂ template. A near band-edge emission at 370.5 nm with a full width at half maximum (FWHM) of 6.5 nm without green emission was observed. This implied that the crystal quality of n-ZnO/p-GaN/ LiAlO₂ were good and intrinsic defects and oxygen vacancy inside are low. Figure 1(C) and Fig. 1(D) show the top view and cross section view of SEM images of n-ZnO thin film regrown on p-GaN template under 800°C by CVD with growth time is 1 hour, respectively. As shown in the Fig. 1(C), the n-ZnO thin film showed a smooth surface with roughness is around 16 nm, which is measured by AFM. As shown in Fig. 1(D), the thickness of n-ZnO thin film is around 100 nm, which is so thin and the defect density in the interface between the regrown n-ZnO and p-GaN template is high that the EL intensity will become lower when the driving current excess than 20 mA which will cause some heat in the interface.

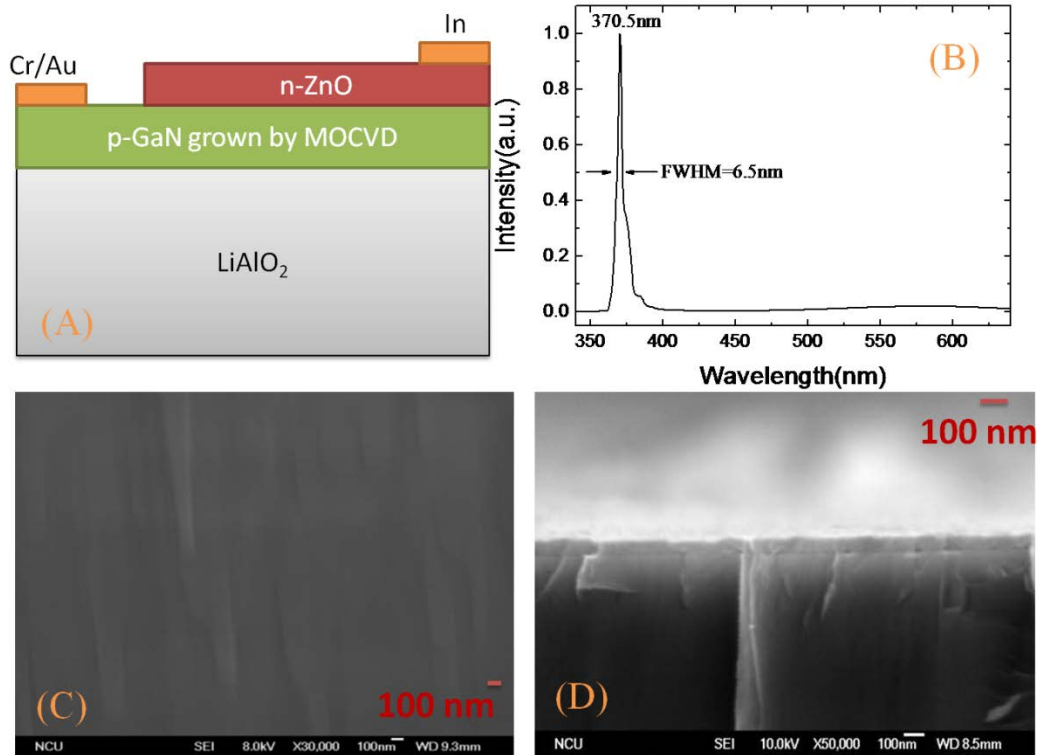


Fig. 1. (A) Schematic of m-plane n-ZnO/m-plane p-GaN/LiAlO₂ heterostructure LEDs, (B) Room-temperature PL spectra of ZnO thin film on m-plane p-type GaN, (C) Top-view FE-SEM image of as-grown ZnO thin films on m-plane p-GaN film, (D) Cross-sectional FE-SEM image of as-grown ZnO films on p-GaN film.

Figure 2 shows the XRD $\theta/2\theta$ scan results of the n-ZnO/p-GaN/LiAlO₂ heterojunction structure. The XRD result of the p-GaN/LiAlO₂ template exhibits only one diffraction peak corresponding to the m-plane (100) p-GaN. For the n-ZnO regrown on m-plane p-GaN template, two distinct diffraction peaks were observed. One diffraction peak corresponds to (100) m-plane GaN, the other diffraction peak corresponds to the n-ZnO layer, which is (100) m-plane n-ZnO, respectively. Hence, the n-ZnO layer was grown with a relationship of (100) n-ZnO || (100) p-GaN. The rocking curve data of m-plane n-ZnO thin films with full width at half-maximum (FWHM) of 504 arcsec was also shown in Fig. 2. The rocking curve data of m-plane p-GaN on LAO substrate is larger than c-plane p-GaN on sapphire, so, unexpected defects are present. Compared to the results of the nonpolar n-ZnO film grown on r-plane sapphire [22], the FWHM of rocking curve was about 1000 arcsec. The nonpolar n-ZnO grown on (100) p-GaN had better crystal quality because the ZnO and GaN have a very low lattice mismatch (1.8%).

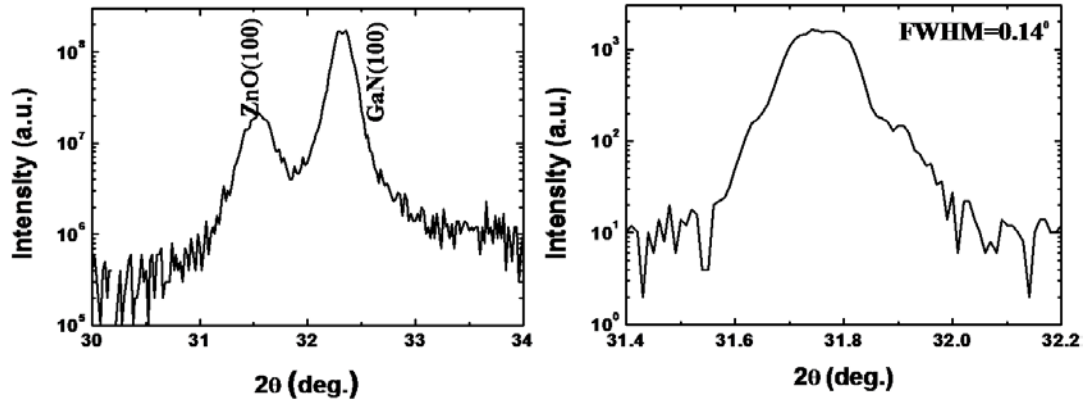


Fig. 2. Left: XRD pattern of as-grown nonpolar ZnO. Right: X-ray rocking curve of nonpolar ZnO.

The RT I-V characteristics of the n-ZnO/p-GaN heterojunction were shown in Fig. 3. The forward current is approximately 12 μ A at 10 V, and the turn-on voltage is 6 V. The breakdown voltage of our result is much smaller than the reported value, the reason should be because the high defect density in the interface between regrown n-ZnO and p-GaN template, which might cause some leakage current when applied even small reversed voltage. The turn-on voltage and power consumption exceed those of conventional LEDs, the relatively low forward and reverse threshold voltages are probably due to the existence of interface defects [28,29].

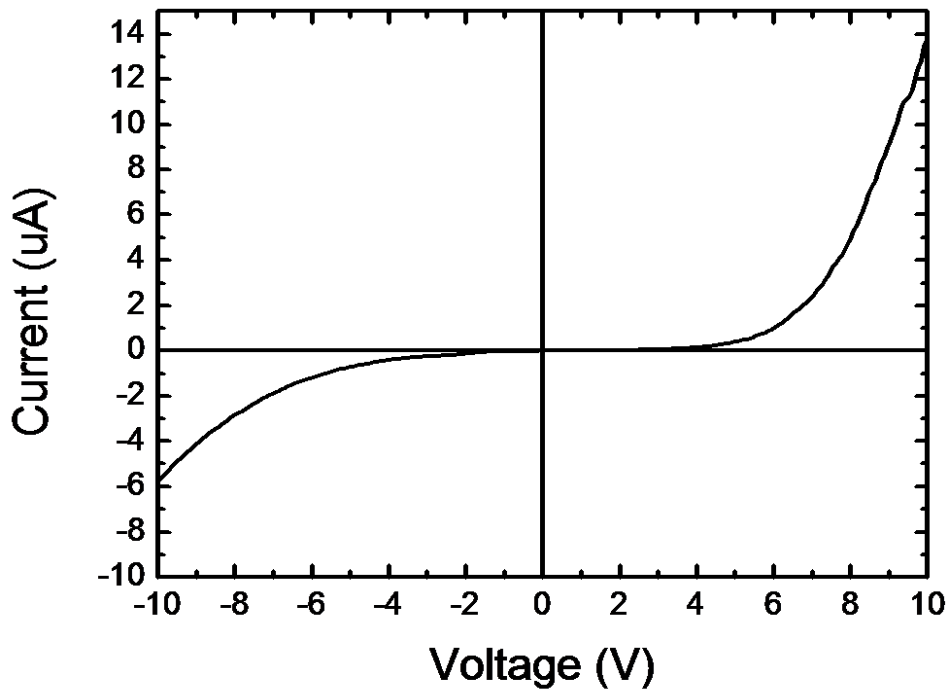


Fig. 3. Current-voltage characteristics of m-plane n-ZnO/m-plane p-GaN/LiAlO₂ heterojunction LED at room temperature.

The EL spectra of the devices were studied as a function of the dc drive current, as shown in Fig. 4. Emission spectra were measured at drive currents ranging from 10, 20, and 40mA,

respectively. The devices emitted in the violet spectral range at 458 nm for all drive currents with minimal linewidth broadening. For our samples, the blue emission located at around 458 nm nearly merges with the UV emission peak, showing a broad emission band. Jin et al suggest that the broad blue luminescence is due to radiative defects related to the interface traps existing at the ZnO grain boundaries [30]. The absence of blueshift in the emission peak with increasing drive current is in contrast to the commonly observed phenomenon of blueshift in c-plane LEDs [31]. We believed the absence of blueshift emission is because of the absence of polarization-induced electric field in the nonpolar m-ZnO thin films [32]. The EL intensity will become lower when the drive current is exceed than 20 mA, this should because the defect density in this device are so high so when the drive current increased, the junction temperature of this device in the interface between n-ZnO and p-GaN template will increase apparently [33].

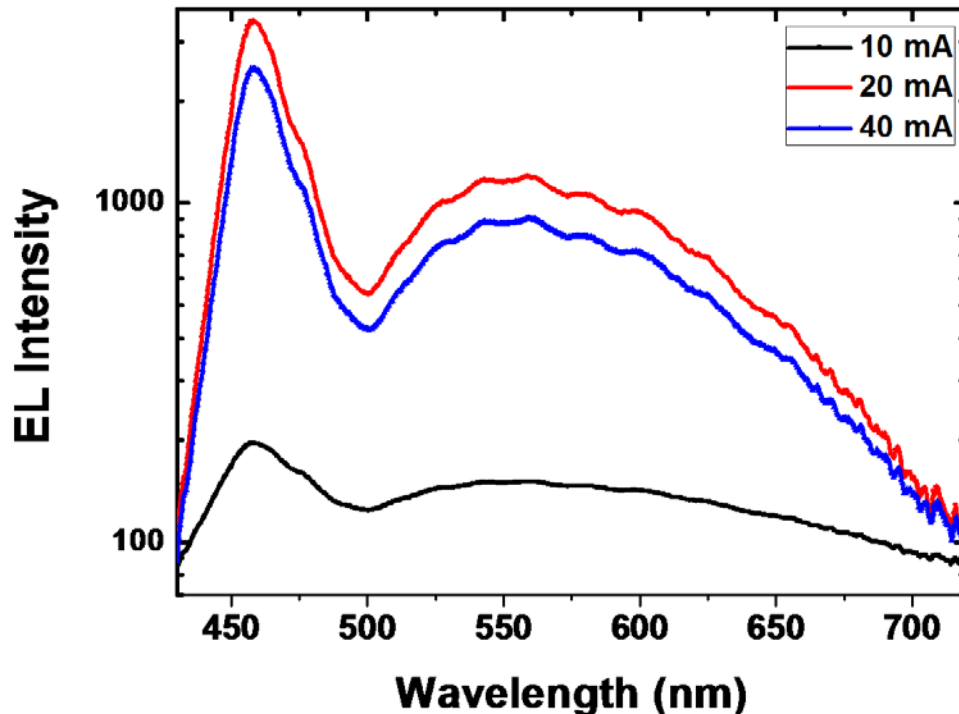


Fig. 4. EL spectra of nonpolar ZnO /p-GaN heterostructure at various drive DC current at 10, 20, and 40mA, respectively.

In conclusion, nonpolar m-plane n-ZnO thin films were grown on m-plane p-GaN templates by chemical vapor deposition (CVD) without employing a catalyst. The ZnO thin film has a heteroepitaxial relationship of (100)n-ZnO|| (100)p-GaN. The p-GaN/n-ZnO heterojunction, the forward turn-on and reverse breakdown voltages was: ~6V and ~-6V, respectively. The relatively low forward and reverse threshold voltages are probably due to the existence of interface defects, as shown in the SEM image in Fig. 1(D). Under forward bias, the EL peak at 458 nm could be attributed to radiative recombination at defects related to the interface traps existing at the ZnO grain boundaries.

Acknowledgments

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