Improved performance of ZnO light-emitting devices by introducing a hole-injection layer

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Abstract: ZnO *p-n* homojunction light-emitting devices (LEDs) have been fabricated, and by introducing a *p*-type GaN as the hole-injection layer, the output power of the LEDs can reach 18.5 μW when the drive current is 60 mA, which is almost three orders of magnitude larger than the pristine LEDs without the hole-injection layer. The improved performance can be attributed to the extra holes injected into the *p*-ZnO layer from the *p*-GaN hole-injection layer.

OCIS codes: (310.6845) Thin film devices and applications; (230.3670) Light-emitting diodes; (250.0250) Optoelectronics.

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1. Introduction

Zinc oxide (ZnO), which has large excitation binding energy (60 meV), has potential to be used for high-performance ultraviolet light-emitting devices (LEDs) and laser diodes [1–4]. However, the reported performance of ZnO homojunction LEDs is still far below expectation [5–11]. The main factor that limits the emission efficiency of ZnO homojunction LEDs lies on its poor *p*-type conduction of ZnO. Although many efforts have been devoted to develop high-quality *p*-type ZnO, further improvement on the corresponding hole concentration and Hall mobility is still required [11–13]. An alternative approach is to fabricate ZnO-based heterojunction devices using other available *p*-type materials to avoid the challenge *p*-type doping issue of ZnO [14–17]. Having the same crystal structure and similar lattice constant with ZnO, GaN has been considered to be the most promising candidate for ZnO-based heterojunction LEDs. However, the defect states at the GaN/ZnO interface caused by the discrepancy between these two materials act as nonradiative center, which will degrade the performance of the ZnO-based heterojunction LEDs drastically [18–20]. Therefore, to realize high-performance ZnO-based LEDs, homojunction is almost a necessarity. Recently, we have realized reproducible and stable *p*-type ZnO films via a lithium and nitrogen co-doping method, and ZnO homojunction LEDs that can operate continuously for several hours have been fabricated [8, 21, 22]. Nevertheless, the hole concentration in this *p*-type ZnO films is still limited to around 10^{16} cm^{-3}, such a low hole concentration hinders the realization of high-performance ZnO homojunction LEDs drastically [23]. Considering that the relative low hole concentration is the limiting factor that determines the performance of ZnO LEDs, if a hole source can be employed to introduce extra holes into the ZnO *p-n* homojunctions, the performance of ZnO LEDs may be improved greatly. However, none such report can be found up to date.

In this letter, we proposed to employ a *p*-type GaN layer with a hole concentration of around 4.1×10^{17} cm^{-3} as the hole-injection layer for a ZnO *p-n* homojunction LED, and the output power of the ZnO LED has been enhanced by over three orders of magnitude compared with the pristine LEDs, and the enhancement can be attributed to the extra holes injected into the ZnO *p-n* junctions from the *p*-GaN layer.

2. Experiment

In order to verify the validity of the hole-injection layer, *p*-type ZnO layer was grown on a commercial available *p*-type GaN layer (purchased from Semiconductor Wafer, Inc) by plasma-assisted molecular beam epitaxy (VG V80H). The *p*-ZnO film was realized by employing lithium and nitrogen codoping method, in which nitric oxide and oxygen were cracked in an Oxford Applied Research HD25 radio-frequency (13.56 MHz) atomic source to form the N dopant and O source, respectively. Metallic zinc and lithium contained in individual Knudsen effusion cells were used as Zn and Li sources. During the growth process, pressure in the growth chamber was fixed at 2×10^{-5} mbar, and the NO and O_2 flow rate were maintained at 0.85 and 1.10 sccm respectively. The substrate temperature was kept at 650 °C during the growth process of the *p*-type ZnO layer. For the growth of *n*-ZnO, the substrate temperature was kept at 750 °C. Metallic Ni/Au and In layer were deposited onto the *p*-type GaN and *n*-type ZnO layers acting as contacts, respectively. The diameter of these metal electrodes is about 1 mm. For comparison, *p*-ZnO/*n*-ZnO homojunction was also prepared under the same growth conditions as stated above. The crystalline properties of the films were evaluated by a Bruker-D8 Discover x-ray diffractometer with Cu Kα (λ = 1.54 Å) as the radiation source. The electrical properties of the ZnO and GaN layers were measured in a Hall system (LakeShore 7707) under van der Pauw configuration. Electroluminescence (EL) measurements of the LEDs were carried out in a Hitachi F4500 spectrometer using a CW current power source. Photoluminescence (PL) spectra of the films were recorded under the excitation from the 325 nm line of a He-Cd laser. The output power of the LEDs was recorded using an OPHIR Nova II power-meter.

3. Results and discussion

The schematic diagram of the ZnO *p-n* homojunctions without the hole-injection layer is illustrated in Fig. 1(a). The chip size is marked in the diagram. The thickness of the *n*- and *p*-type ZnO layers is around 400 nm and 150 nm, respectively. The electron concentration and Hall mobility of the *n*-ZnO films are 2.1×10^{19} cm^{-3} and 47 cm^2/Vs, respectively. For the *p*-ZnO layer, the hole concentration is around 3.4×10^{16} cm^{-3} and the Hall mobility is around 1.3 cm^2/Vs. The current-voltage (*I- V*) curve of the *p-n* junction shows a noticeable rectification behavior with a turn-on voltage of around 3.0 V, as shown in Fig. 1(b). The inset of Fig. 1(b) shows the *I-V* curves for both In contact on the *n*-ZnO and Ni/Au contact on the *p*-ZnO. It is observed that both curves are nearly beeline, indicating that ohmic contacts have been formed. Hence, one can conclude that the rectification behavior observed in the *I- V* curve arises from the *p*-ZnO/*n*-ZnO homojunction.

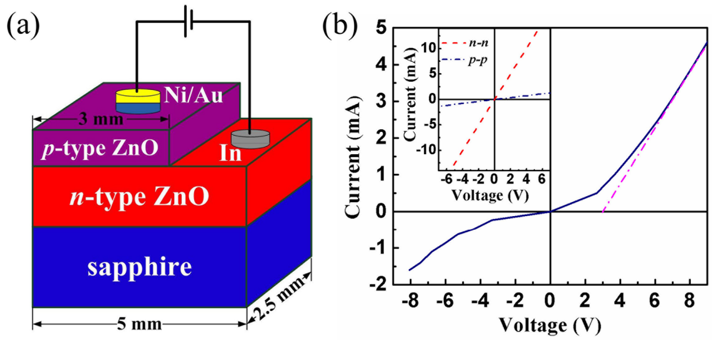

Fig. 1. (a) Schematic diagram of the *n*-ZnO/*p*-ZnO homojunction structure, and the size of the device has been marked in the diagram; (b) *I*– *V* curve of the *p*-*n* junction, and the inset shows the *I*– *V* curves of the Ni/Au contact on the *p*-ZnO layer and the In contact on the *n*-ZnO layer.

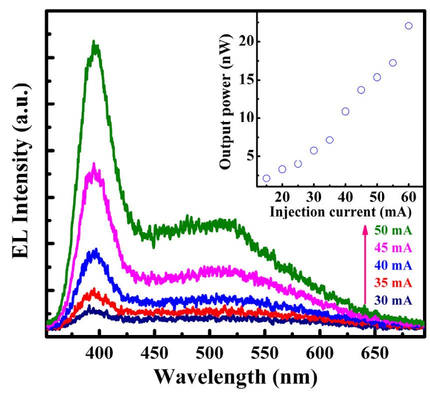

Fig. 2. Room temperature EL spectra of the *n*-ZnO/*p*-ZnO homojunction under different injection current, and the inset shows the dependence of the output power of the *p*-*n* junction on the injection current.

Figure 2 shows the room temperature EL spectra of the *p*-ZnO/*n*-ZnO homojunction. Two emission bands can be observed from the spectra – A dominant one at around 395 nm and a weak one at around 500 nm. The former has been commonly observed from ZnO-based *p*-*n* junctions, which can be attributed to the near-band-edge emission of ZnO. As for the latter, it is usually attributed to the deep-level related emissions of ZnO [24, 25]. The dependence of the output power of the *p*-ZnO/*n*-ZnO homojunction on the injection current is shown in the inset of Fig. 2. One can see that the output power increases with the injection current, and it reaches around 24 nW when the injection current is 60 mA. To increase the performance of the LED is a vital step for future applications of ZnO-based LEDs. It is speculated that the limiting factor that determines the performance of the LEDs lies in the low hole concentration in the *p*-type ZnO. Therefore, if extra holes can be injected into the *p*-type ZnO, the performance of the LEDs may be enhanced significantly.

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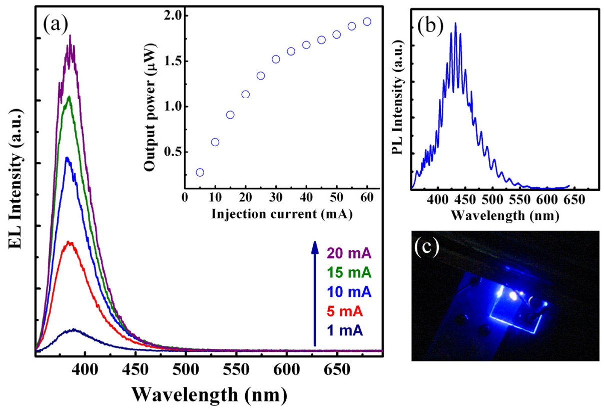

Fig. 3. (a) Room temperature EL spectra of the *p*-GaN fastened ZnO *p-n* junctions via direct contact method under different injection current, and the inset shows the dependence of the output power on the injection current; (b) Room temperature PL spectrum of the *p*-GaN layers; (c) A typical emission image of the *p*-GaN coated ZnO *p-n* junction under an injection current of 20 mA.

To test the above idea, a commercial available *p*-type GaN films was fastened onto the *p*-type ZnO layer via a direct contact method by a clip, and the hole concentration and Hall mobility of the *p*-type GaN layer is 4.1×10^{17} cm^{-3} and 14 cm^2/Vs. The thickness of the *p*-GaN layer is 2 μm. The emission spectra of the *p*-GaN fastened ZnO *p-n* junctions under different injection current are shown in Fig. 3(a). A dominant band at around 385 nm can be observed from the spectra, the intensity of which increases with the injection current. For the *p*-GaN fastened ZnO *p-n* junctions, it is important to explore the origin of the emission since intense emission may also be emitted from the *p*-GaN layer. To explore the origin of such emission, the PL spectrum of the *p*-GaN layer was investigated, as shown in Fig. 3(b). One can find from the figure that there appears only one broad emission at around 430 nm. Note that the fringes in the spectrum may come from the interference of the emitted light. Since the EL spectrum of the *p*-type GaN fastened ZnO *p-n* junctions is distinctly different from the PL of the *p*-type GaN layer, and its position (385 nm) is close to that of the ZnO *p-n* junctions shown in Fig. 2 (395 nm), one can deduce that the emission should not come from the *p*-type GaN layer but from the ZnO *p-n* junction. Note that the slight blueshift of the emission compared with the pristine *p-n* junction may be due to the following reasons: Since the *p*-type GaN layer has been fastened onto the *p*-type ZnO layer via a direct contact method, the contact between these two layers is nonuniform. Because of the non-uniform contact, the emission of the *p*-GaN coated ZnO *p-n* junctions comes from several points, as indicated by the emission image shown in Fig. 3(c), then the current density at these points should be very large. The high injection current density may lead to the occupancy of carriers on higher energy levels, thus the emission peak (385 nm) shows slight blueshift compared with the pristine ZnO *p-n* junctions (395 nm) [26]. The output power of the *p*-GaN fastened ZnO *p-n* junctions was shown in the inset of Fig. 3(a), from which one can see that the output power increases monotonically with the injection current, and it reaches 1.9 μW when the injection current is 60 mA. The output power of the *p*-GaN fastened ZnO *p-n* junction has been significantly enhanced compared with that of the pristine structure, and this enhancement confirms the validity of the above idea that by introducing a hole-injection layer, the performance of ZnO *p-n* junction LEDs can be increased greatly.

Nevertheless, one can see from Fig. 3(c) that the emitted light comes only from several points, which is caused by the non-uniform contact of the *p*-GaN and *p*-ZnO layer as stated above. It is accepted that the non-uniformity is adverse to the performance of the LEDs. To improve the performance of the ZnO LEDs further, *p*-type GaN layer grown on sapphire substrate was employed as a template for the growth of ZnO *p-n* junctions, and *p*-type ZnO

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layer was firstly deposited onto the *p*-GaN layer, then *n*-ZnO layer was deposited onto the *p*-ZnO layer. The schematic diagram of the structure is shown in Fig. 4(a). The *I*- *V* characteristic of the ZnO *p*-*n* junction grown on *p*-GaN layer shows obvious rectification behaviors with a turn-on voltage of around 4.7 V, as shown in Fig. 4(b). The inset of Fig. 4(b) shows the *I*- *V* curves for both In contact on the *n*-ZnO and Ni/Au contact on the *p*-GaN. It is observed that both curves are nearly beeline, indicating that ohmic contacts have been formed. Note that this device has larger turn-on voltage than that of the *p*-*n* junction without the hole-injection layer. This is because the lateral distance between the two electrodes is much larger than the thickness of the devices, which means that the resistivity of the devices was mainly determined by the lateral distance between the *p*-type and *n*-type electrode. Thus the resistance in the *p*-*n* junction without the hole-injection layer is mainly determined by the sheet resistance of the *n*-ZnO layer, while that of the structure with the hole-injection layer is mainly determined by the sheet resistance of the *p*-GaN layer. The resistance of the *p*-GaN (~1.1 Ωcm) is larger than the *n*-ZnO (~6.3 × 10^{-3} Ωcm). As a result, the series resistance of this device grown on the *p*-GaN hole-injection layer is larger than that of the pristine *p*-*n* junction, which explains the larger turn-on voltage observed in Fig. 4(b).

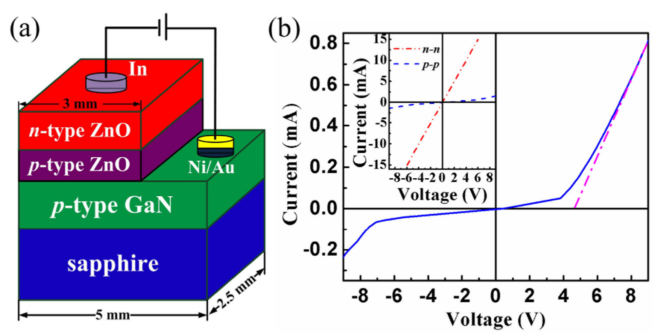

Fig. 4. (a) Schematic diagram of the *n*-ZnO/*p*-ZnO homojunction grown on *p*-GaN hole-injection layer, and the size of the device has been marked in the diagram. (b) *I*– *V* curve of the structure, and the inset shows the *I*– *V* curves of the Ni/Au contact on the *p*-GaN layer and the In contact on the *n*-ZnO layer.

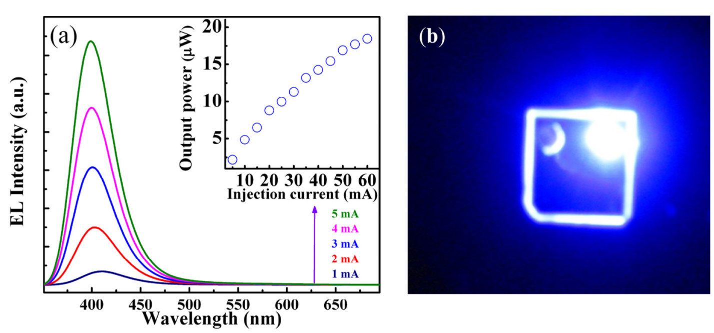

Fig. 5. (a) Room temperature EL spectra of the *p*-*n* junction grown on *p*-GaN hole-injection layer at different injection current, and the inset shows the dependence of the output power on the injection current; (b) A typical emission image of the structure under an injection currents of 20 mA.

The emission spectra of the ZnO *p*-*n* junction grown on the *p*-GaN hole-injection layer are shown in Fig. 5(a). An emission at around 400 nm dominates the spectra of the LED, while the deep-level emission at around 500 nm is negligible. The dependence of the output power of the LED on the injection current is illustrated in the inset of Fig. 5(a). One can see that the

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emission of the LED increases with the injection current, and the output power can reach around 18.5 μW when the drive current is 60 mA, which is amongst the best value ever reported in ZnO-based LEDs [27–29]. The output power is almost three orders of magnitude larger than that of the pristine ZnO homojunction LEDs. A typical emission image of the LEDs is shown in Fig. 5(b). Intense emission can be observed from the structure, note that the emission can be observed easily by naked eyes under normal indoor lighting conditions. Also one can see that the emission image is much more uniform than that of the *p*-GaN fastened ZnO *p-n* homojunction shown in Fig. 3(c). Since the emission peak wavelength of the *n*-ZnO/*p*-ZnO homojunction with the hole-injection layer (400 nm) is in good agreement with that of the device without the hole-injection layer (395 nm), one can speculate that the emission comes from the *n*-ZnO/*p*-ZnO homojunction.

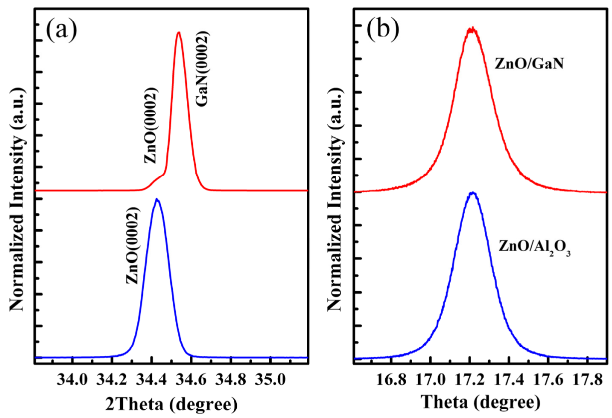

Fig. 6. (a) θ-2θ XRD pattern of the ZnO films grown on sapphire substrate (Bottom) and *p*-GaN layer (Top) in the same growth process. (b) X-ray rocking curves for the ZnO films grown on sapphire substrate and the *p*-GaN hole-injection layer.

It is accepted that substrate may have a huge impact on the crystalline quality of the films grown on it. Then to explore the mechanism of the enhanced emission from the ZnO LEDs, the crystalline properties of the ZnO films grown on the *p*-GaN and sapphire substrate in the same growth process have been measured. One can find from the θ- 2θ pattern of the ZnO films shown in Fig. 6(a) that both the films grown on *p*-GaN and sapphire shows wurtzite structure with (0002) preferred orientations. The XRD rocking curves of both films show symmetric Gaussian lineshape. It is accepted that the full width at half maximum (FWHM) of the rocking curve can symbols the crystalline quality of the films to some extent. The FWHM for the ZnO films grown on *p*-GaN is 880 arcsec, and that of the films grown on sapphire is 846 arcsec. The very similar FWHM reveals that the substrate does not affect the crystalline quality of the ZnO films in our case greatly. Thus the enhanced EL output power for the ZnO LEDs grown on *p*-GaN may not be caused by the improved crystalline quality of the ZnO films. Then the enhanced emission from the ZnO LEDs should come from the extra hole injection into the ZnO *p-n* homojunction from the *p*-GaN layer, and the hole injection process from the *p*-GaN layer is depicted below.

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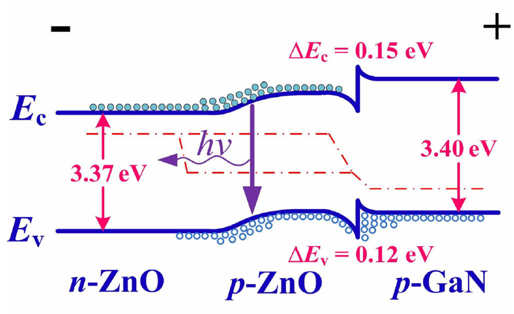

Fig. 7. Schematic bandgap diagram of the *n*-ZnO/*p*-ZnO homojunction deposited on the *p*-GaN hole-injection layer.

For the *n*-ZnO/*p*-ZnO homojunction without the *p*-GaN hole-injection layer, since the electrons concentration in the *n*-type ZnO layer is much higher than that in the *p*-ZnO layer and the mobility of the electrons is larger than that of holes, radiative recombination between electrons and holes will mainly occur in the *p*-type ZnO layer. It is accepted that the output power of the LEDs is determined by the product of electrons and holes, but the number of electrons is much larger than that of holes, thus the hole concentration in the *p*-ZnO layer is the limiting factor that hinders the output power of the LEDs. Figure 7 shows the schematic energy band diagram of the *n*-ZnO/*p*-ZnO homojunction with the hole-injection layer. The bandgap diagram was constructed by considering that the electron affinity of ZnO and GaN are 4.35 and 4.2 eV, and the bandgap of ZnO and GaN is 3.37 and 3.40 eV, respectively. Hence, the conduction band offset and valence band offset at the *p*-GaN/*p*-ZnO interface can be deduced to be 0.15 eV and 0.12 eV, respectively [30, 31]. Under forward bias, some holes can pass across this barrier formed by the valence band offset at the *p*-GaN/*p*-ZnO interface, and inject from the *p*-GaN layer into the *p*-ZnO layer, thus more holes and electrons can be recombined radiatively inside the *p*-ZnO. Consequently, the emission of the LEDs has been enhanced greatly. Note that although extra holes have been injected into the *p*-ZnO layer from the *p*-GaN layer, the depletion region of the device locates still mainly inside the *p*-type ZnO layer just as that in the pristine *n*-ZnO/*p*-ZnO homojunction. This is the reason why the emission wavelength of the LED with the hole-injection layer (400 nm) is in such a good agreement with that of the pristine device (395 nm).

4. Conclusion

In conclusion, *n*-ZnO/*p*-ZnO homojunction LEDs have been fabricated, and by introducing a *p*-GaN as the hole-injection layer, the output power of the LEDs can be improved by three orders of magnitude. The improved performance can be attributed to the extra holes injected into the *p*-ZnO layer from the *p*-GaN layer. Hence, the results reported in this letter may provide a route to high-performance ZnO-based LEDs.

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