Effect of p-NiO interlayer on efficiency of p-GaN/n-ZnO LED devices

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***Abstract* — We report on numerical study of the characteristics of p-GaN/n-ZnO light-emitting diodes with p-NiO interlayer, and on LED design optimization which includes bandgap engineering, thickness and doping of constituent layers. The current-voltage dependences of investigated LEDs show a threshold voltage of 3.1 V and 5.4 V for the LED devices without- and with presence of p-NiO interlayer, respectively. It is found that p-NiO layer act as electron blocking layer, that lead to the enhance of charge carriers confinement in active region, and to the increasing of internal quantum efficiency (IQE) of LED devices in four times up to 0.5%.**

***Index Terms* — light-emitting diode, zinc oxide, gallium nitride, nickel oxide, internal quantum efficiency.**

I. INTRODUCTION

The III-V and II-VI wide bandgap semiconductors like gallium nitride (GaN) and zinc oxide (ZnO) was attracted significant interest of many researchers as promising materials for optoelectronic and photonic applications. GaN and ZnO materials have a similar room temperature bandgap of 3.43 eV and 3.37 eV, respectively. Both materials exhibit piezo- and spontaneous polarizations, have a similar crystal wurtzite structure, and lattice mismatch ~ 1.8% [1]. In compare with GaN, zinc oxide has several advantages such as like a direct wide bandgap, large exciton binding energy of ~ 60 meV (~ 25 meV for GaN), high electron mobility and good transparency in the visible spectral range, higher radiation hardness, and relative low material costs [2]. Thus, these properties make ZnO attractive for fabrication ZnO-based light-emitting diodes (LEDs). Moreover, zinc oxide could be fabricated as nanotubes [3] and as water-soluble nanocrystals [4,5], that open wide perspective to produce cheaper all-inorganic quantum dot LEDs [6] using spun-casting or print inkjet technique.

In order to improve the light emitting properties of ZnO-based LEDs, an interlayer of any other suitable material acting as buffer medium is highly required for significant improvement of the LED structure because the interlayer provides a stable charge environment during hole and electron injections in the light emitting part of LED device. Recently, were reported studies of LEDs with different interlayers such as like WO_3, TiO_2, Ag, MoS_2, MgO, MoO_3 and NiO. It was established that these interlayer materials has improved the performance of polymer LED significantly, it has brought the change in the barriers for electrodes and also increases the hole injection which in result lower the threshold and working voltage [7]. The use of MoS_2, NiO, and TiO_2 nanocrystals as inorganic hole transport layers in organic- and quantum dot LEDs has been reported [8].

In this paper, we report the simulation results for p-GaN/n-ZnO light-emitting diodes with inserting of p-NiO interlayer as hole transport- and electron blocking layers. This results will be used to identify the most important parameters for achieving high performance and high efficiency LEDs.

II. MODEL

For the LED device simulations in this study, the 1D-DDCC one-dimensional solver software was used [9]. This software based on the modified ABC model [10] coupled with one-dimensional Schrödinger-Poisson and drift-diffusion equations, which are solved self-consistently. The modified ABC model describe the radiative and non-radiative recombination, which occur in the LED device, and accounts the radiative bimolecular recombination [11], Shockley-Read-Hall (SRH) non-radiative recombination [12], Auger non-radiative recombination [13], carrier leakage [14], and polarization effects [15]. The internal quantum efficiency (IQE) of LED device can be defined as ratio of radiative recombination rate to the sum of all recombination rate terms, integrated in a whole area of LED structure.

III. RESULTS AND DISCUSSION

We consider first a conventional p-GaN/n-ZnO LED structure according to published design specifications [16]. The conventional reference LED device consist of a 500 nm thick Mg-doped p-GaN layer ($N_A = 1\cdot10^{17}$ cm^{-3}, E_A = 170 meV) grown on c-plane sapphire and a 200 nm thick Al-doped n-ZnO ($N_D = 5\cdot10^{17}$ cm^{-3}, E_D = 50 meV) cap

layer. The p-GaN/n-ZnO heterojunction form a type-II band alignment [17] and its schematic structure and energy bandgap diagram is shown in Figs. 1(a) and 1(b), respectively. The band diagram of this ideal heterojunction is constructed according Anderson's model [18] and using material parameters collected from Refs. [16, 19-21]. Using energy band diagram from Figs. 1(a) and 1(b) we can estimate the energy barriers ΔE_C and ΔE_V for an electron and hole respectively:

$$\Delta E_C = \chi(\text{GaN}) - \chi(\text{ZnO}) = -0.15 \text{ eV},$$
$$\Delta E_V = E_g(\text{GaN}) + \Delta E_C - E_g(\text{ZnO}) = -0.083 \text{ eV}.$$

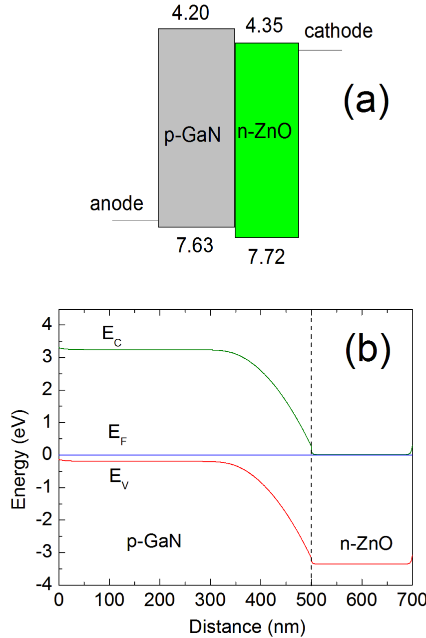

Fig. 1. The schematic structure (a) and energy band diagram (b) of the p-GaN/n-ZnO LED.

Due to the conduction and valence band offsets the free electrons and holes can be accumulated near the interface between p-GaN and n-ZnO. Both materials GaN and ZnO have the wurtzite crystal symmetry and exhibit polarization effects (spontaneous and electric polarization) that can contribute to the interface charges. When we consider polarization effects contribution in the LED structure, the small triangular well is arising in the interface between p-GaN and n-ZnO. It is found that the triangular well act as electron blocking layer and lead to enhance the electron accumulation and confinement in the p-GaN/n-ZnO interface, that resulting to increase the internal quantum efficiency (IQE) of LED device. We found that the threshold voltage for p-GaN/n-ZnO LED device is about of 3.1 V.

In order to optimize the doping level of p-GaN and n-ZnO layers as well as IQE efficiency of LED device, we have simulated the set of p-GaN/n-ZnO LED structures with different doping concentration of acceptors in p-GaN and donors in n-ZnO layers. The polarization contribution to the charge interfaces of LED structure was considered. We found that the IQE efficiency of p-GaN/n-ZnO LED devices is strongly dependent on p- and n-doping of p-GaN and n-ZnO epi-layers, respectively. The optimization of p- and n- doping of p-GaN and n-ZnO epi-layers of LED structure lead to the enhancing the balance between injected electrons and holes in active region, as well as IQE efficiency, and also shifts the maximum of IQE(j) dependence to the higher current density values. As result, the p-GaN/n-ZnO LED device can operate at higher current densities. It is found that optimal p- and n-doping of p-GaN and n-ZnO epi-layer allow to fabricate the p-GaN/n-ZnO LED devices with IQE efficiency up to 0.5%. In general, the increasing of both p- and n-doping of p-GaN and n-ZnO layers, respectively, lead to the enhancing the IQE efficiency of LED structure.

Figure 2 shows the schematic structure of the p-GaN/p-NiO/n-ZnO LED device and corresponding energy band diagram simulated for this heterostructure. The values of material parameters for the simulations is were take from Refs [16, 19-21]. The LED structure consist of 500 nm thick p-GaN (Mg-doped, $N_A = 1\cdot10^{17}$ cm^{-3}, E_A = 170 meV) epi-layer grown on c-plane sapphire, a 10 ÷ 1000 nm thick p-NiO (Mg-doped, $N_A = 1\cdot10^{16} \div 1\cdot10^{20}$ cm^{-3}, E_A = 170 meV) interlayer, and a 200 nm thick n-ZnO cap layer (Al-doped, $N_D = 5\cdot10^{17}$ cm^{-3}, E_D = 50 meV). This double heterojunction form a type-II band alignment.

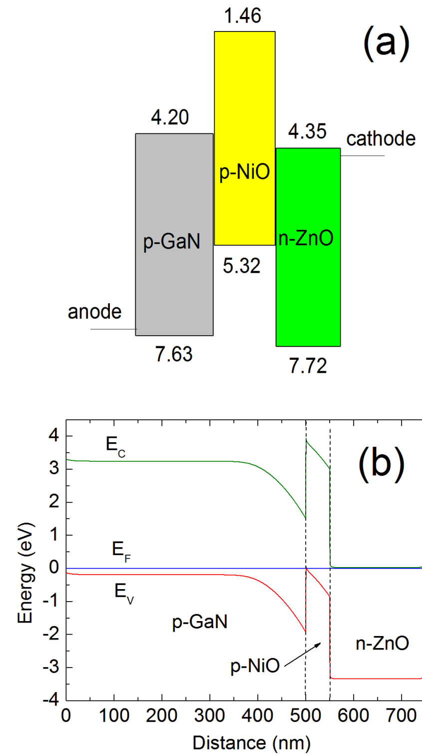

Fig. 2. The schematic structure (a) and energy band diagram (b) of the p-GaN/p-NiO/n-ZnO LED.

Using schematic structure of LED device and its simulated energy band diagram (Fig. 2(a) and 2(b)) we can estimate energy discontinues ΔE_C and ΔE_C for holes and electrons, respectively. For p-GaN/p-NiO heterojunction we can obtain the values of energy discontinues as follows:

$$\Delta E_C = \chi(\text{GaN}) - \chi(\text{NiO}) = 2.74 \text{ eV};$$
$$\Delta E_V = E_g(\text{GaN}) + \Delta E_C - E_g(\text{NiO}) = 2.317 \text{ eV}.$$

For p-NiO/n-ZnO heterojunction the values of energy discontinues can be calculated as follows:

$$\Delta E_C = \chi(\text{NiO}) - \chi(\text{ZnO}) = 2.89 \text{ eV},$$
$$\Delta E_V = E_g(\text{NiO}) + \Delta E_C - E_g(\text{ZnO}) = 2.40 \text{ eV}.$$

Thus, the energy barrier for the injected holes and electrons at the p-GaN/p-NiO and p-NiO/n-ZnO interfaces are found

to be 2.317 eV and 2.89 eV, respectively. It is found from Figs. 1(a), 1(b), 2(a), and 2(b) that the energy band discontinues in the p-GaN/p-NiO/n-ZnO LED structure is higher in compare with that for p-GaN/n-ZnO LED structure. Thus, presence of p-NiO interlayer can act as electron blocking layer for electrons injected from n-ZnO layer, and also lead to enhance the hole injection to active region from p-GaN epi-layer. The free electrons and holes will be accumulated near the n-ZnO/p-NiO interface in the n-ZnO and p-NiO sides, respectively. The radiative recombination of free electrons and holes will be occurring in the p-NiO interlayer and in the interface p-NiO/n-ZnO.

Figures 3(a), 3(b), and 3(c) shows the energy band diagram of p-GaN/p-NiO/n-ZnO LED operated under forward bias (at fixed current density j = 100 A/cm^2), the depth distribution of electron and hole concentrations, and the depth distribution of radiative recombination rate, respectively. It is seen that the free electrons and holes will be accumulated at the p-GaN/p-NiO and p-NiO/n-ZnO interfaces, respectively.

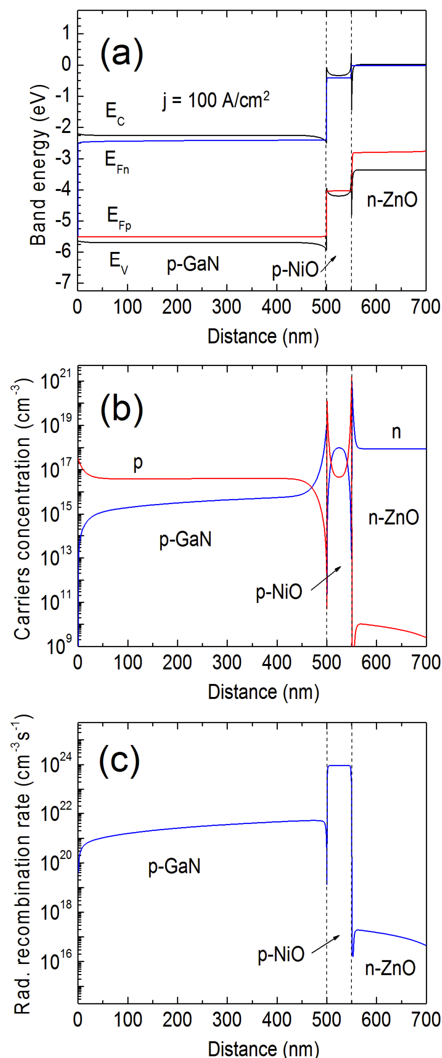

Fig. 3. The energy band diagram (a), the depth distribution of charge carriers concentration (b), and the depth distribution of radiative recombination rate (c) for p-GaN/p-NiO/n-ZnO LED device operated under forward bias voltage. The current density was kept constant to be j = 100 A/cm^2.

It is seen that the values of radiative recombination rate in the p-NiO interlayer at least in two- three- orders of magnitude higher than that in the p-GaN and n-ZnO epi-layers. Thus, p-NiO interlayer effectively blocks the free electrons injected from n-ZnO layer and enhance holes injection in the active region of LED device.

Figure 4(4) shows current-voltage characteristics of two p-GaN/n-ZnO LED devices with presence and absence of p-NiO interlayer. It is seen that the presence of p-NiO interlayer lead to the higher values of threhold voltage, which was found to be ~ 5.4 V in compare with value of ~ 3.1 V for p-GaN/n-ZnO LED device without of p-NiO interlayer. The observed increased values of threshold voltage can be attributed to the low electron affinity (1.46 eV) and large bangap energy (3.86 eV) of p-NiO interlayer, that create additional energy barrier for injected charge carriers.

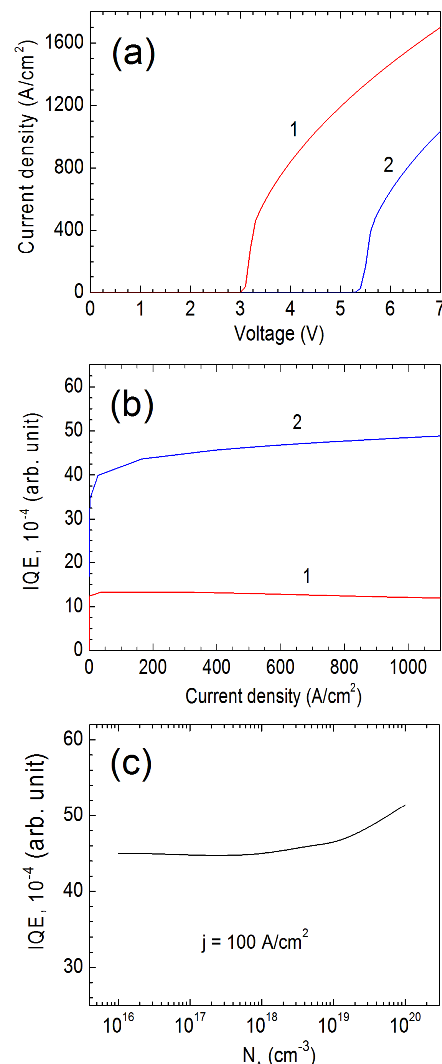

Fig. 4. The current-voltage characteristics (a) and the dependences of IQE vs. current density (b) for p-GaN/n-ZnO (curve 1) and p-GaN/p-NiO/n-ZnO (curve 2) LED devices, respectively. (c) The influence of p-doping of p-NiO interlayer on IQE efficiency of p-GaN/p-NiO/n-ZnO LED device.

Figure 4(b) shows the dependence of IQE vs. current density for both above mentioned p-GaN/n-ZnO LED structures with presence- and absence of p-NiO interlayer,

respectively. It is seen that the presence of p-NiO lead to the enhancing of the IQE efficiency of p-GaN/p-NiO/n-ZnO LED device about of four times of magnitude practically in all range of current densities, in compare with the IQE efficiency of original p-GaN/n-ZnO LED device.

In order to study effect of p-NiO interlayer on LED efficiency and optimization structure of LED devices, we have simulated a set of p-GaN/p-NiO/n-ZnO LED devices with varied width and p-doping of p-NiO interlayer. The doping and width of p-GaN and n-ZnO were kept constant and chosen to the optimal from the simulation results of p-GaN/n-ZnO LEDs described above. In the first, we fixed the width of p-NiO interlayer to be 50 nm and investigated the influence of p-doping of p-NiO interlayer on IQE efficiency of LED structure. Figure 4(c) show the dependence of IQE efficiency vs. N_A concentration in p-NiO interlayer. It is seen that IQE of LED device with p-NiO interlayer practically independent from the p-doping level of p-NiO layer in wide range of doping concentrations.

It is found also that the increasing of width of p-NiO interlayer from 10 nm to 1000 nm lead to the slightly increasing of threshold voltage values from 5.3 to 5.5 V, that can be attributed to the increasing series resistance of p-NiO interlayer. It is established that the increasing of width of p-NiO interlayer lead to the increasing of IQE efficiency of LED device due to the increasing of width of active media layer. These results confirms our suggestion that p-NiO interlayer act as active media layer, and increasing of its depth lead to the enhancing of the charge carriers confinement in active region, decrease current leakage and result to increasing of IQE efficiency of p-GaN/n-ZnO LED devices.

IV. CONCLUSION

We have numerically investigated p-GaN/n-ZnO LED devices without and with presence of p-NiO interlayer. A spatially resolved modified ABC model, that additionally includes the polarization effects contribution and carriers leakage terms, coupled with one-dimensional Schrödinger-Poisson and drift-diffusion equations and solved self-consistently, were used. It is established that inserting of p-NiO layer between p-GaN and n-ZnO lead to the increasing of threshold voltage up to 5.4 V and increasing IQE efficiency of LED device up to 0.5%. p-NiO interlayer enhance the hole injection from p-GaN to the active media and effectively blocks the electrons injected from n-ZnO epi-layer due to the their large bandgap energy (3.86 eV) and low electron affinity (1.46 eV). It is found, that the IQE efficiency of p-GaN/p-NiO/n-ZnO LEDs strongly depends on width of p-NiO interlayer, and practically independent on its doping level.

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