

n-ZnO/p-GaN/ α -Al₂O₃ HETEROJUNCTION AS A PROMISING BLUE LIGHT EMITTING SYSTEM

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It is reported on the fabrication and characterization of light emitting *n*-ZnO/*p*-GaN heterojunction. Firstly, a Mg-doped *p*-GaN layer was grown on (0001) sapphire substrate by the molecular-beam epitaxy technique, then thin Ga-doped *n*-ZnO film was deposited by the chemical vapor deposition in the low-pressure system. Diode-like rectifying behavior of this heterojunction and room temperature electroluminescence in the blue-violet region with peak wavelength 430 nm under forward bias were observed. Light-current characteristics of this structure feature a superlinear behavior in the low current range with a slope 1.9 and a sublinear behavior with a slope 0.85 in the high current range.

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1. Introduction

Zinc oxide is a direct wide bandgap semiconductor very similar in many physical properties to gallium nitride [1]. Both materials have wurtzite crystal structure, almost the same lattice parameters (crystal lattice mismatch ~1.8 %) and bandgap widths ($E_g = 3.3$ eV for ZnO and 3.4 eV for GaN). At the same time, ZnO features an intense excitonic emission at high temperatures due to the large excitonic binding energy (61 meV), and a high stability to the high-energy radiation, so it may be considered as a promising semiconductor for the fabrication of the blue and ultraviolet light emitting devices (LED) suitable for operation in various environments and high temperatures [2]. Until now there are no reports on high quality conductive *p*-type ZnO production, so fabrication of *n*-ZnO/*p*-GaN heterojunction remains the only possible way to make LED structures. There were several reports on the growth of *n*-type ZnO on *p*-type materials (ZnTe, Cu₂O, and SrCu₂O₂) [2-4] to form LEDs, but in all cases the hetero-interface had to accommodate a large lattice mismatch that had greatly impaired device performance. A number of dislocations in device interface arising from the lattice mismatch between the layers of *p*-*n* heterojunction is a key factor that controls the quantum efficiency of the LEDs. The fabrication of ZnO/GaN structures was reported earlier [5-7], however the GaN films were *n*-type in all of these works, and used primarily as buffer layers for high quality ZnO films preparation due to their close lattice parameters. We report in this work, for the first time, a fabrication of the perfect lattice-matching *n*-ZnO/*p*-GaN heterojunction with relatively intense electroluminescence (EL) in the blue-violet region under forward bias.

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2. Experimental

p-GaN:Mg $\sim 1 \mu\text{m}$ thick film was grown on (0001) α -sapphire substrate by molecular-beam epitaxy (MBE) using ammonia as the nitrogen source, and a SUMO cell for the Ga. The substrate temperature during the growth process was 800°C being measured by an optical pyrometer operating with a central wavelength of $1 \mu\text{m}$ and an emissivity setting of 0.3. The growth rate was $0.8 \mu\text{m hr}^{-1}$. After deposition of the GaN:Mg layer, the sample was placed into a separate reactor and n-ZnO:Ga layer $\sim 1 \mu\text{m}$ thick was grown by the chemical vapor deposition (CVD) technique in the low-pressure system, high-purity hydrogen being used as a carrier gas. Gallium, a Group III element was found to be a good dopant element for n-type ZnO thin layers fabrication, where Ga incorporates substitutionally on the Zn sites in the lattice, forming a donor level with activation energy of $\sim 50 \text{ meV}$ [8]. CVD system and growth technique details are described elsewhere [9]. For comparison, isotype n-ZnO/n-GaN heterostructures were also grown under the same conditions as the n-ZnO/p-GaN heterostructures.

The heterojunction devices were fabricated by masking the surface, and then using a 10 % HNO_3 aqueous solution to etch the ZnO layer down to the p-GaN layer through a $2 \times 2 \text{ mm}^2$ opening in the mask. This technique exploits the property of ZnO to be etched rapidly in acids, while GaN is very resistive to these environments. Ohmic contacts to the n-ZnO were made by applying indium with a soldering iron, and to the p-GaN by thermal deposition of gold. Current-voltage (I - V) characteristics of the area between two In-In and two Au-Au points of the ZnO:Ga and GaN:Mg layers, respectively, were measured and good Ohmic contacts were observed. The I - V characteristics of the heterostructures were measured using an oscilloscope specially adapted for this purpose. EL measurements were performed under DC-biased conditions at RT and at various injection currents. The Hall measurements of the both GaN and ZnO films were made at room temperature (RT) using the four-point van der Pauw technique. Optical properties of the films were explored using cathodoluminescence (CL) spectroscopy at room temperature, with 20 keV electron accelerating voltage and 0.1-1 μA electron-beam current range.

3. Results and discussion

The free carriers density and mobility are found to be $p = 3.5 \times 10^{17} \text{ cm}^{-3}$ and $\mu_p = 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for acceptors in GaN:Mg, and $n = 4.5 \times 10^{18} \text{ cm}^{-3}$ and $\mu_n = 40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for donors in ZnO:Ga.

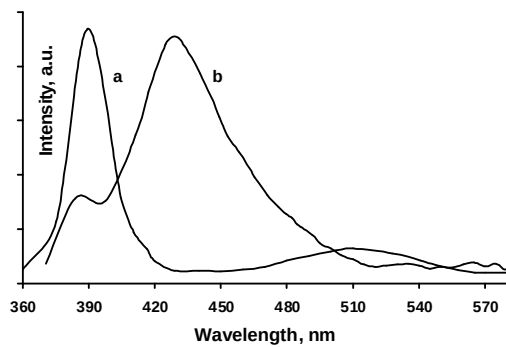


Fig. 1. Room temperature cathodoluminescence spectra of (a) ZnO:Ga and (b) GaN:Mg films.

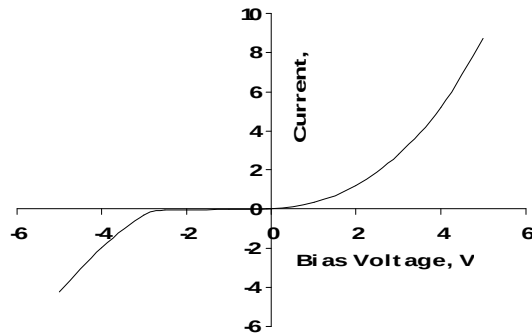


Fig. 2. Current-voltage characteristics of n-ZnO/p-GaN heterostructure.

Fig. 1 shows the CL spectra of the ZnO and GaN layers. The CL spectrum of the ZnO film (Fig. 1a) consists of intense, near-band-edge (NBE) ultraviolet emission with a wavelength maximum λ_{max} at 390 nm and a full width at half maximum (FWHM) of 118 meV. A broad defect-related green band with much lower intensity near $\lambda_{\text{max}} \sim 510 \text{ nm}$ with a FWHM of 330 meV is also shown in this figure. In contrast, the CL spectrum of the GaN:Mg film (Fig. 1b) consists of a weak NBE band with λ_{max} at 383 nm, and a more intense broad band with λ_{max} at 430 nm. This is typical for Mg-doped GaN

films, and is generally attributed to transitions from the conduction band or shallow donors to deep Mg acceptor levels [10-11].

The I - V dependence of the fabricated heterostructure at RT are presented in Fig. 2. As can be seen, the fabricated n -ZnO/ p -GaN heterostructure exhibits non-linear and rectifying behavior with a threshold voltage of ~ 3 V and a leakage current of $\sim 5 \times 10^{-6}$ A, very similar to that of p - n homojunctions. In contrast to this, the I - V dependences of the n -ZnO/ n -GaN heterostructure was Ohmic and nearly linear. Based on this fact, we have concluded that the rectifying diode-like behavior of the n -ZnO/ p -GaN junction results from the electrical nature of components of the n - p heterojunction, rather than from some interface or contact related phenomena. Furthermore, the $\sim I$ - V^2 relationship is clearly observed over a wide forward bias range. Such a dependence is very usual for wide band-gap materials like ZnO, GaN and SiC [12-13], and is attributed to space charge limited current. The relatively low threshold voltage of our n - p heterostructure (~ 3 V) is probably conditioned by the defects, especially by the extended defects in the GaN because of the large lattice mismatch between GaN and sapphire (~ 16 %).

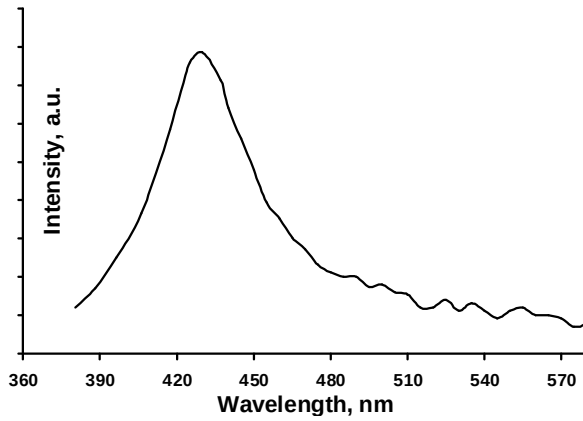


Fig. 3. Electroluminescence spectrum of the n -ZnO/ p -GaN heterostructure.

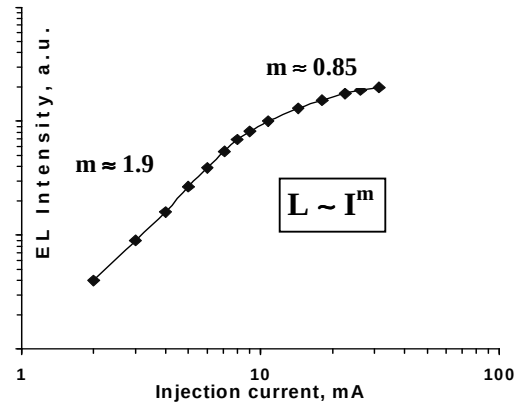


Fig. 4. Room temperature light-current (L - I) characteristics of n -ZnO/ p -GaN heterostructure.

Electroluminescence emission of the n -ZnO/ p -GaN heterostructure was observed under forward bias and is shown in Fig. 3. The EL spectra of the forward biased LED consists of a broad emission band with a λ_{max} at about 430 nm and a FWHM of about 25 nm. The peak emission at 430 nm lies in the violet spectrum region, and there is a tail extending to longer wavelengths. Our results are in agreement with the previous works that showed that 430 nm radiation is emitted whenever electrons are injected from the n region into a Mg doped p region of GaN [10, 14, 15]. A comparison of Fig. 3 with the CL spectra of ZnO and GaN shown in Fig. 1a and 1b allows us to suggest that the EL emission emerges from the GaN side of the n -ZnO/ p -GaN heterostructure. Thus, the EL properties of the formed n -ZnO/ p -GaN heterostructure LED are determined primarily by electron injection from the n -ZnO side of the n -ZnO/ p -GaN to the p -GaN side, where radiative recombination occurs. The probability of this scenario increases taking into consideration the fact that the donor density in ZnO:Ga is almost one order higher than the acceptor density in GaN:Mg ($4.5 \times 10^{18} \text{ cm}^{-3}$ and $3.5 \times 10^{17} \text{ cm}^{-3}$, respectively). As can be seen from the EL spectra in Fig. 3, there is no 383 nm peak as in the case of the GaN CL spectrum (Fig. 1b). This can possibly be explained by self-absorption effects in the upper ZnO layer because the 383 nm emission wavelength of GaN is shorter than the 390 nm wavelength associated with excitonic emission of ZnO [16].

The EL spectra of the n -ZnO/ p -GaN heterostructure were measured at various injection currents. The measurements show that the shape of EL spectrum does not change significantly with the injection current, except for the FWHM increases slightly. The light-current characteristics (L - I) have been obtained from a direct measurement of the peak emission intensity at 430 nm (see Fig. 4). Being in full agreement with the spectral evolution, the L - I curve shows a superlinear dependence at

low currents (<10 mA) with a slope 1.9 ($L \sim I^m$, $m = 1.9$) and becomes sublinear ($m = 0.85$) at higher currents (>10 mA). We believe, the break in the slope corresponds to the current value at which the intensity of the blue-violet emission at 430 nm is saturated. However, it can also be caused by Auger recombination at high injection current densities and by heating effects or series resistances, the quantification of which would require further investigation.

4. Conclusions

We have successfully fabricated on (0001) Al_2O_3 substrate the light emitting device based on $n\text{-ZnO}/p\text{-GaN}$ heterojunction. The prepared LED features highly intensive room-temperature blue-violet electroluminescence under the forward bias and diode-like I - V dependence behavior.

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