Enhancement of the near-band-edge electroluminescence from the active ZnO layer in the ZnO/GaN-based light emitting diodes using AlN-ZnO/ZnO/AlN-ZnO double heterojunction structure

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Abstract

In this work, an AlN-ZnO/ZnO/AlN-ZnO double heterojunction (DH) structure prepared using the cosputtering technology was deposited onto the p-type GaN epitaxial layer. The indiffusion of the oxygen atoms to the p–GaN epilayer was obstructed as the cosputtered AlN-ZnO film inset between n-ZnO/p–GaN interface. The near-ultraviolet (UV) emission from this ZnO/GaN-based light emitting diode (LED) was greatly improved as compared to an n-type ZnO film directly deposited onto the p-GaN epilayer. Meanwhile, the native defects in the n-ZnO layer associated with the green luminescence was less likely to form while it was sandwiched by the cosputtered AlN-ZnO film. As the thickness of the active n-ZnO layer in the DH structure reached 10 nm, the near-band-edge (NBE) emission became the predominated luminescence over the resulting LED spectrum.

1. Introduction

Photonic devices emitting luminescence ranging from the near-ultraviolet (UV) to UV wavelength are indispensable for application to the lighting, display, information storage, secure communication, and biosensors industries [1–4]. To emit short-wavelength light, the active layer in the photonic device must possess wide and direct energy bandgap. Among the semiconductor materials, gallium nitride (GaN)-based material that performs quality crystallinity with the direct energy bandgap of 3.40 eV, which is fabricated using the metalorganic chemical vapor deposition (MOCVD) or molecular beam epitaxy (MBE) technologies, has been comprehensively employed in the photonic device and commercialized. However, the epitaxial method to achieve crystalline GaN is substrate dependent making it too complicated and expensive to popularize. Accordingly, the effort to explore other direct semiconductor with a wide energy bandgap as a substitute for the GaN-based material performs quality crystallinity with the direct energy bandgap of about 3.37 eV by using cost-effective deposition technologies, such as hydrothermal method, pulse laser deposition, and sputtering [5–7], making it a promising candidate for short-wavelength photonic device application. In addition, ZnO also has the advantages of a large exciton binding energy of 60 meV at room temperature, resistance to radiation damage, and suitability for the wet-etching process over the commercial-used GaN material. Those excellent physical and chemical properties are expected to be brighter than the current state-of-the-art GaN-based photonic device for the near UV and UV light emitting diode (LED) applications. Currently, ZnO-based homojunction diodes are fabricated for the photodetector (PD) or LED applications [7–11]. Quality and stable n- and p-type ZnO semiconductors as well as engineer their interfacial property are essential to achieve the ZnO-based homojunction diode. Unfortunately, development of the quality ZnO-based homojunction diode is obstructed because the hole carriers in the p-ZnO film are prone to be compensated for the native donor defects, such as zinc interstitials (ZnI) and/or oxygen vacancies (V_O), calling into question the reproducibility of the p-type ZnO conduction. Accordingly, the crystalline p-GaN epilayer with the structure matching to the ZnO material becomes a substitution material for
hetero-contacting to n-ZnO film. Although the resulting n-ZnO/p-GaN heterojunction diode showed better diode rectification and photo-electric conversion efficiency as compared to the ZnO-based homojunction diode [12–17], structural optimization to confine the device active layer in the n-ZnO layer as well as improvement of its crystallinity is still ongoing. For example, Long et al fabricated an MgZnO/ZnO/MgZnO double heterojunction (DH) structure on the p-GaN epilayer layer using a radio frequency (rf) magnetron sputtering system to confine the injection carriers in the ZnO layer. The resulting LED emitted green luminescence at a peak of about 530 nm, which was ascribed to the transition related to the ZnO and V_{O} defects in the active ZnO layer. The authors also showed their achievement of an LED that emitted intense UV/near UV luminescence as the active ZnO film in the double heterojunction structure replaced by the hydrothermal prepared ZnO nanorods [18]. Moreover, Long et al designed the Ta_{2}O_{5} and HfO_{2} films as the hole- and electron-blocking layers, respectively, to confine the injection carries into the active i-ZnO layer. The resulting LED constructed from the Ta_{2}O_{5}/ZnO/HfO_{2} asymmetric DH structure showed enhancement and long-time stability on the defect electroluminescence (EL) related to oxygen interstitials (O_{i}) transition in the ZnO layer at wavelengths ranging from 620 to 690 nm [19]. Kong et al grew an MgZnO/ZnO DH structure on the silicon substrate using MBE technology. They demonstrated that the resulting ZnO-based LED emitted stronger near-band-edge UV emission than that of the LED without the DH structure [20]. Pandey et al fabricated a Sb-ZnO/Cd-ZnO/Ga- ZnO DH structure using a dual ion beam sputtering deposition system. They showed that the p-i-n ZnO-based diode yielded blue EL at around 446 nm as a result of the effective confinement of the injected electron-hole pairs in the Cd-ZnO active layer [21].

In the previous paper, Ho et al fabricated a n-ZnO/p-GaN-based LED that emitted broad near UV luminescence by optimizing the transparent electrode contact to the n-type ZnO [14]. The origin of the red, green-yellow, and blue-violet emission in the EL spectra from the n-ZnO/p-GaN-based LEDs annealed under various ambient to cause the interfacial diffusion, had also been discussed [22, 23]. In addition, Zheng et al also fabricated an AlN-ZnO/ZnO/AlN-ZnO DH structure using the rf magnetron cosputtering technology. The ZnO film sandwiched by the cosputtered AlN-ZnO film exhibited quality crystalline structure and showed enhancement of the NBE photoluminescence (PL) a consequence of the suppression of the native V_{O} defects [24]. In this study, the AlN-ZnO/ZnO/AlN-ZnO DH structure was applied to consecutively deposit onto the p-GaN epilayer. The diode performance and EL spectrum of the ZnO/GaN-based LED with this DH structure was investigated. Moreover, the evolution of the EL spectra from the ZnO/GaN-based LEDs as a function of the well thickness of the active ZnO layer in the DH structure was also discussed.

2. Experiment

The Mg-doped p-GaN epilayer employed in this study was grown on the c-plane sapphire substrate using the MOCVD technology. The Mg dopants in the p-GaN epilayer was activated to a hole concentration of \(4.8 \times 10^{17} \text{cm}^{-3}\) and a mobility of 8.8 cm^{2}/Vs, respectively, at the annealing temperature at 750 °C for 30 min under nitrogen ambient. Patterned AlN-ZnO/ZnO/AlN-ZnO DH structure (hereafter denoted as DH-ZnO), defined by the standard lift-off technology, was then consecutively deposited onto the p-GaN epilayer using the rf magnetron cosputtering system. Targets of ZnO purity of 99.99% and AlN (purity of 99.99%) were employed as the source material for preparing the cosputtered AlN-ZnO and undoped ZnO films at room temperature under the working pressure of 1.33 Pa and argon ambient of 15 sccm. The rf power supplied on the AlN target was fixed at 85 W, while the power on the ZnO target was controlled at 70 W to obtain the cosputtered AlN-ZnO film at a theoretically atomic ratio of [Al/Zn + Al] of 40%. Detailed technology to derive the theoretical atomic ratio of the Al concentration in the cosputtered AlN-ZnO film through their deposition rates had been reported elsewhere [25]. The thickness of the cosputtered AlN-ZnO film in the DH-ZnO structure was controlled at about 50 nm, while that of the undoped ZnO film was varied at 10, 25, 50, and 100 nm to study the emission efficiency. As a comparison, an undoped ZnO film with the thickness of 100 nm was directly deposited onto the p-GaN epilayer. The ZnO/p-GaN and DH-ZnO/p-GaN contact systems were then annealed in the furnace at temperature of 7000 °C for 30 min under vacuum ambient. Following, Ni/Au (5/50 nm) metallic system was deposited onto the p-GaN surface and annealed at 500 °C for 10 min under air ambient for achieving good ohmic contact behavior. A 150 nm-thick ITO-ZnO film [Zn / (Zn + In) = 33 at%] was deposited onto the patterned DH-ZnO and undoped ZnO surface, respectively, by the rf magnetron cosputtering system, using the ZnO and ITO (purity 99.99% with In_{2}O_{3} 90 wt% : SnO_{2} 10 wt%) targets. The transparent ITO-ZnO electrode ohmic contact to the n-type ZnO was processed by a rapid thermal annealing (RTA) at 400 °C for 5 min under vacuum ambient [14].

Film thickness of the undoped ZnO and DH-ZnO structure was measured using a surface profile system (Dektak 6M, Veeco) and confirmed by the cross-sectional transmission electron microscopy (TEM) observation. The electrical properties of the undoped ZnO and cosputtered AlN-ZnO films as well as the...
DH-ZnO structure deposited onto the p-GaN epilayer were conducted using the van der Pauw Hall measurements (Ecopia HMS-5000) at room temperature. Radiative characteristics of the undoped ZnO and cosputtered AlN-ZnO films as well as the DH-ZnO structure with various well thickness were determined from their PL spectra measured at room temperature using a He-Cd laser (λ = 325 nm) pumping source. Elemental distributions of the ZnO/p-GaN and DH-ZnO/p-GaN structures were examined by Auger electron spectroscopy (AES) depth profiles on a scanning Auger nanoprobe (Ulvac-PHI, PHI 700). The current-voltage (I-V) properties of the ZnO/p-GaN and DH-ZnO/p-GaN LEDs as well as the ITO-ZnO/DH-ZnO and Au/Ni/p-GaN ohmic contact systems were measured by a semiconductor parameter analyzer. The electroluminescence (EL) spectra related to these LEDs were obtained at room temperature under various forward injection currents.

### 3. Results and discussions

The electrical properties of the 100 nm-thick undoped ZnO, cosputtered AlN-ZnO films, and DH-ZnO structure with the active ZnO layer of 100 nm, annealed at 700 °C for 30 min under vacuum ambient, are summarized in table 1 (the electrical property of the activated p-GaN epilayer is also given in this table). The undoped ZnO film behaved as degenerated n-type conduction with the electron carriers of $2.4 \times 10^{19}$ cm$^{-3}$ (hereafter denoted as n-ZnO), while the cosputtered AlN-ZnO film acted as an insulator with electrical resistivity higher than $10^5$ Ω cm after thermal annealing treatment. The vacuum-annealed ZnO film which was abundant in the VO donors due to the outdiffusion of oxygen atoms was ascribe to be responsible for the film that possessed a high electron concentration [26, 27]. For the undoped ZnO film sandwiched by the cosputtered AlN-ZnO film, the electron carriers of the annealed sample notably increased to $7.0 \times 10^{19}$ cm$^{-3}$. It implied that donors other than VO had been activated. The room-temperature PL (hereafter denoted as RT-PL) spectra of the n-ZnO, cosputtered AlN-ZnO films, and DH-ZnO structure deposited onto the p-GaN epilayer, after annealing at 700 °C for 30 min under vacuum ambient, are depicted in figure 1 (the spectrum of the p-GaN epilayer is also shown in this figure). The spectrum of the p-GaN epilayer showed a dominant peak at about 433 nm that was ascribed to the deep-level emission from the transition of the conduction band or shallow donors to the deep Mg-acceptor level (indicated as DL emission (p-GaN)) [28]. By contrast, significant green luminescence with the

![Figure 1. PL spectra of the n-ZnO, cosputtered AlN-ZnO films, and DH-ZnO structure deposited onto the p-GaN epilayer, after annealing at 700 °C for 30 min under vacuum ambient (the PL spectrum of the p-GaN epilayer is also shown).](image)
peak at about 520 nm ascribed to the deep-level emission emerging from the \( n \)-ZnO film (denoted as DL emission (\( n \)-ZnO)) was predominate over the \( n \)-ZnO/\( p \)-GaN heterojunction structure. As the reports indicated [29, 30], the native defects of the vacancies of zinc (\( V_{\text{Zn}} \)) and oxygen (\( V_{\text{O}} \)) as well as the interstitial of zinc atoms (\( \text{ZnI} \)) were responsible for the radiative transition of this broad DL emission. The intense DL emission emerged mainly from the transition of \( V_{\text{O}} \) donor also provided evidence of the vacuum-annealed ZnO film with a high electron concentration. In addition, another weak UV luminescence associated with the NBE transition from the \( n \)-ZnO film (assigned as NBE emission (\( n \)-ZnO)) appeared at about 382 nm. Compared to the \( n \)-ZnO film, the cosputtered AlN-ZnO film emitted very weak DL luminescence with undistinguished NBE emission. For the DH-ZnO structure deposited on the \( p \)-GaN epilayer, the luminescence from the DL transitions was markedly depressed while the NBE emission was apparently enhanced. It concluded that the formation of the native defects in the ZnO film could be effectively suppressed as it was sandwiched by the cosputtered AlN-ZnO films.

Figure 2. \( I-V \) characteristics of the \( n \)-ZnO/\( p \)-GaN and DH-ZnO/\( p \)-GaN contact systems (the \( I-V \) curves of the \( p \)-type and \( n \)-type ohmic contacts are shown in the inset figure).

Figure 3. (a) EL spectra of the \( n \)-ZnO/\( p \)-GaN heterojunction LED under forward-bias current and (b) the corresponding coordinate in the CIE color system while operated at an injection current of 20 mA (the light emission image is also shown in the inset figure).

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Furthermore, the $n$-ZnO film in the double heterojunction structure emitted intense NBE luminescence also could be ascribe to the improvement in the crystalline structure. As discussed in the previous work, the improvement in the ZnO film’s crystallinity was demonstrated to be the diffusion of the Al atoms in the cosputtered AlN-ZnO film into the ZnO layer to suppress the formation of the native oxygen vacancies. These indiffused Al atoms to act as the AlZn donor also led to the red-shift of the ultraviolet luminescence to about 388 nm, with a wider full width at half maximum (FWHM)\[^{24}\]. Moreover, as listed in table 1, the activation of the AlZn donor in the DH-ZnO structure resulted in a very high electron concentration in spite of the suppression of the VO donor. Figure 2 shows the current-voltage ($I$-$V$) characteristics of the $n$-ZnO/$p$-GaN and DH-ZnO/$p$-GaN contact systems (the $I$-$V$ curves of the $p$-type and $n$-type ohmic contacts are shown in the inset figure). Both curves exhibited diode-like rectifying behavior. An apparent current drop was observed from the DH-ZnO/$p$-GaN diode to result in the increase of the turn-on voltage with a low forward-bias current. The notable increase in the series resistance was attributed to the inferior conduction of the cosputtered AlN-ZnO film with a low ohmic contact current as shown in the inset figure. The room temperature EL (RT-EL) spectra of the $n$-ZnO/$p$-GaN heterojunction LED emitted yellowish light at the coordinate of (0.468, 0.380) in the CIE color system, as shown in figure 3(b). By contrast, the red luminescence associated with Ga-O transition was almost absent in the EL spectrum of the LED constructed from the DH-ZnO/$p$-GaN structure, as shown in figure 3(a). The near UV luminescence became the dominant emission with a broad green-yellow luminescence. As referred to the previous reports\[^{23, 29, 31–33}\], the native defect transitions in the $n$-ZnO and $p$-GaN layers, such as oxygen ($V_O$) and/or gallium ($V_Ga$) vacancies, could be

![Figure 4. (a) EL spectra of the DH-ZnO/$p$-GaN LED under forward-bias current and (b) the corresponding coordinate in the CIE color system while operated at an injection current of 20 mA (the light emission image is also shown in the inset figure).](image-url)
responsible for the green-yellow emission, with a peak at around 520 nm. Moreover, it was noted that the peak of the near UV luminescence emerged from the DH-ZnO/p-GaN LED shifted to a shorter wavelength of 412 nm as compared to that of the n-ZnO/p-GaN heterojunction LED. Incorporating the origin of the short-wavelength luminescence, the reason responsible for the blue-shift could be linked to the enhancement of the portion of the NBE emission from the n-ZnO active layer. The EL spectrum dominated by the near UV luminescence with a distinct shoulder extending to the green-yellow luminescence emitted white light that corresponded to a coordinate of (0.351, 0.340) in the CIE system, as

![Figure 5](image-url)  
*Figure 5. Elemental depth profiles, obtained from the AES measurements, for the (a) n-ZnO film and (b) DH-ZnO structure contact to the p-GaN epilayer.*

![Figure 6](image-url)  
*Figure 6. PL spectra as a function of the ZnO well thickness in the DH-ZnO structure.*
shown in figure 4(b). The elemental depth profiles for the n-ZnO film and DH-ZnO structure contacting the p-GaN epilayer obtained from the AES measurements are illustrated in figures 5(a) and (b). As can be seen in figure 5(a), the signal of the oxygen element was detected at a deeper depth (i.e., a higher sputter time) than that of the zinc element. This implied that more oxygen atoms in the n-ZnO film were diffused into the p-GaN epilayer. An interfacial layer composed mainly of the gallium and oxygen atoms (Ga-O interfacial layer) thus could be determined through the decrease in the counts of half of each element. This interfacial layer originating from the indiffusion of the oxygen atoms into the p-GaN epilayer also provided evidence of the red luminescence from the n-ZnO/p-GaN LED. For the DH-ZnO structure contacting system. Thus, the diffusion of the oxygen atoms into the p-GaN epilayer could be effectively suppressed, and
thereby the red luminescence from the Ga-O interfacial layer was absent in the EL spectrum of the DH-ZnO/p-GaN LED.

Based on the discussion thus far, the Ga-O interfacial layer formed at the n-ZnO/p-GaN interface to result in the red luminescence was suppressed as the n-ZnO sandwiched by the cosputtered AlN-ZnO film. The EL spectrum of the DH-ZnO/p-GaN LED therefore purely emerged from the active n-ZnO layer and p-GaN epilayer. In order to purify the EL spectrum without the defects-related luminescence (i.e., green-yellow luminescence), well thickness of the active n-ZnO layer in the DH-ZnO structure was engineered. Figure 6 shows the RT-PL spectra as a function of the ZnO well thickness in the DH-ZnO structure. All these spectra showed dominated NBE luminescence with very weak DL emission. In addition, the intensity of the NBE emission gradually decreased as the well thickness decreased. Current-voltage (I-V) characteristics of the DH-ZnO/p-GaN contact systems as a function of the well thickness of the active n-ZnO layer are plotted in Figure 7. All these curves show the rectifying characteristic, with the turn-on current increasing as the well thickness decreased in the DH-ZnO structure. The series resistance, $R_s$, in these diodes, could be evaluated from the following equation developed by Cheung and Cheung [34]:

$$\frac{dV}{dI} = R_s + \frac{\eta k_B T}{q l}$$

where, $\eta$ is a measure of the conformity of the diode to pure thermionic emission and $k_B$ is the Boltzmann constant. The experimental $dV/dI$ versus $1/I$ plots obtained from the forward bias of the DH-ZnO/p-GaN contact systems are shown in the inset figure of Figure 7. The series resistance of the DH-ZnO/p-GaN contact system was evaluated from the intercept of these curves. According to the evolution of the series resistance, the increase in the turn-on current of the DH-ZnO/p-GaN diodes was closely linked to the reduction in the series resistance. The series resistance was decreased from 14.2 to 9.2 kΩ as the well thickness in the DH-ZnO structure was thinned from 100 to 10 nm. Meanwhile, a lower turn-on voltage of 2.15 V was also obtained from the well thickness of 10 nm as compared to that of the well thickness of 100 nm ($\sim$2.74 V). Figure 8(a) depicted the RT-EL spectra of the DH-ZnO/p-GaN diodes with the active ZnO layer of 100, 50, and 10 nm under the injected forward current of 20 mA. The intensity of the near UV emission was gradually decreased as the well thickness decreased. Interestingly, although the near UV emission was degraded, the green-yellow luminescence was almost absent in the EL spectrum of the DH-ZnO/p-GaN LED with the active n-ZnO layer thickness of 10 nm.
This indicated that the defect-related transitions in the active \textit{n}-ZnO layer could be further suppressed as designed the well thickness in the DH structure. The peak position and FWHM of the near UV emission as a function of the well thickness in the DH-ZnO structure is given in figure 8(b). The peak position of the near UV emission shifted toward shorter wavelength, and its FWHM was also narrowed as the well thickness decreased. Figure 9(a) and (b), respectively, show the deconvolution of the near UV emission from the well thickness of 100 and 10 nm in the DH-ZnO/p-GaN LED. The DH-ZnO/p-GaN LED with the well thickness of 100 nm emitted the dominant luminescence from both the \textit{p}-GaN epilayer and the \textit{n}-ZnO/p-GaN interfacial recombination with a weak signal from the \textit{n}-ZnO active layer. By contrast, as the well thickness in the DH-ZnO/p-GaN LED decreased to 10 nm, the emission from the active \textit{n}-ZnO layer was significantly enhanced and became stronger than the DL emission from the \textit{p}-GaN epilayer. The peak of the near UV emission therefore shifted toward a short wavelength, as shown in figure 8(b). The evolution of the coordinate in the CIE color system from the emission spectra of the \textit{n}-ZnO/p-GaN LED and the DH-ZnO/p-GaN LEDs as a function of the well thickness is shown in figure 10 (the corresponding emission lights are shown in the inset figure). The yellowish light emitted from the \textit{n}-ZnO/p-GaN LED due to the dominant luminescence from the Ga-O deep-level transition changed to whitish light ((0.351, 0.340) in the CIE color system), composed mainly of the near UV and green-yellow luminescence, thus was observed from the DH-ZnO/p-GaN LED as the thickness of the active \textit{n}-ZnO layer of 100 nm. The DH-ZnO/p-GaN LED emitted purely the near UV emission ((0.322, 0.263) in the CIE color system) could be further achieved as thinned the active layer to a thickness of 10 nm. Such a DH-ZnO structure was beneficial for both the confinement of the injection carriers and avoiding the defect transitions in the active \textit{n}-ZnO layer.

4. Conclusions

An undoped ZnO layer sandwiched by the cosputtered AlN-ZnO film was consecutively deposited onto the \textit{p}-GaN epilayer. The cosputtered AlN-ZnO film functioned as a barrier layer to obstruct the interdiffusion between the \textit{n}-ZnO/p-GaN interface. The red luminescence emerged from the Ga-O interfacial layer was almost absent in the resulting EL spectrum of the DH-ZnO/p-GaN LED. Whitish light ((0.351, 0.340) in the CIE color system), composed mainly of the near UV and green-yellow luminescence, thus was observed from the DH-ZnO/p-GaN LED as the thickness of the active \textit{n}-ZnO layer of 100 nm. The DH-ZnO/p-GaN LED emitted purely the near UV emission ((0.322, 0.263) in the CIE color system) could be further achieved as thinned the active layer to a thickness of 10 nm. Such a DH-ZnO structure was beneficial for both the confinement of the injection carriers and avoiding the defect transitions in the active \textit{n}-ZnO layer.

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Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

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