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HTML ABSTRACT * LINKS

MgZnO/AlGaN heterostructure light-emitting diodes

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We report on $p-n$ junction light-emitting diodes fabricated from MgZnO/ZnO/AlGaN/GaN triple heterostructures. Energy band diagrams of the light-emitting diode structure incorporating piezoelectric and spontaneous polarization fields were simulated, revealing a strong hole confinement near the n-ZnO/p-AlGaN interface with a hole sheet density as large as 1.82×10^{13} cm^{-2} for strained structures. The measured current–voltage (IV) characteristics of the triple heterostructure $p-n$ junctions have rectifying characteristics with a turn-on voltage of ~3.2 V. Electron-beam-induced current measurements confirmed the presence of a $p-n$ junction located at the n-ZnO/p-AlGaN interface. Strong optical emission was observed at ~390 nm as expected for excitonic optical transitions in these structures. Experimental spectral dependence of the photocurrent confirmed the excitonic origin of the optical transition at 390 nm. Light emission was measured up to 650 K, providing additional confirmation of the excitonic nature of the optical transitions in the devices. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1815377]

The great potential of ZnO-based materials for solid-state lighting applications derives from the high excitonic binding energy of 60 meV,[1,2] enabling efficient excitonic optical transitions in ZnO to take place at elevated temperatures. The high excitonic binding energy is expected to prevent thermal dissociation of excitons at temperatures as high as 400 °C, leading to the design and fabrication of solid-state optical light emitters operating at fairly high temperatures without additional cooling. Thus far, the main difficulties in fabricating ZnO-based emitters has been the lack of a reliable p-type doping process necessary for the formation of $p-n$ junction diodes. Recently, light-emitting diodes (LEDs) were fabricated from wurtzite n-ZnO/p-AlGaN single heterostructure (SHS).[3] The LEDs fabricated from n-ZnO/p-AlGaN heterostructures emitted UV light at 389 nm at 300 K and operated up to 500 K.[3]

We report on the demonstration of $p-n$ junction LEDs fabricated from $p-n$ junction MgZnO/ZnO/AlGaN/GaN triple heterostructures (THS). The THS design is expected to improve optical and thermal characteristics of LEDs with ZnO-based active regions. In this work we also present theoretical analysis of energy band diagrams for SHS and THS, accounting for polarization effects. Experimental measurements of IV characteristics and electroluminescence (EL) are reported for THS LED devices operating up to 650 K.

To illustrate the design concept, the band diagram and free carrier distribution in $Mg_xZn_{1-x}O/Al_yGa_{1-y}N$ heterostructures were simulated using a self-consistent one-dimensional Schrödinger–Poisson solver.[4] The parameters used in the calculation are given in Ref. 5. Ga(Al) polarity and Zn polarity were assumed for GaN (AlGaN) and ZnO (MgZnO) layers, respectively. The contacts were assumed to be ohmic. A type-II band alignment was used for ZnO/AlGaN SHS.[3] In our model the AlGaN valence band (VB) maxima was located above ZnO by $\sim[1-\Delta E_v(\text{AlGaN})]$ eV, where ΔE_V(AlGaN) is the VB discontinuity between $Al_xGa_{1-x}N$ and GaN. The piezoelectric (P_{PE}) and spontaneous (P_{SP}) polarizations were incorporated into our model to account for the polar nature of the materials. The polarization charge was calculated using the relation $\sigma(P_{SP}+P_{PE})=[P_{SP}(\text{bottom})+P_{PE}(\text{bottom})]-[P_{SP}(\text{top})+P_{PE}(\text{top})]$.[6]

Figure 1 shows the band diagram and schematic charge

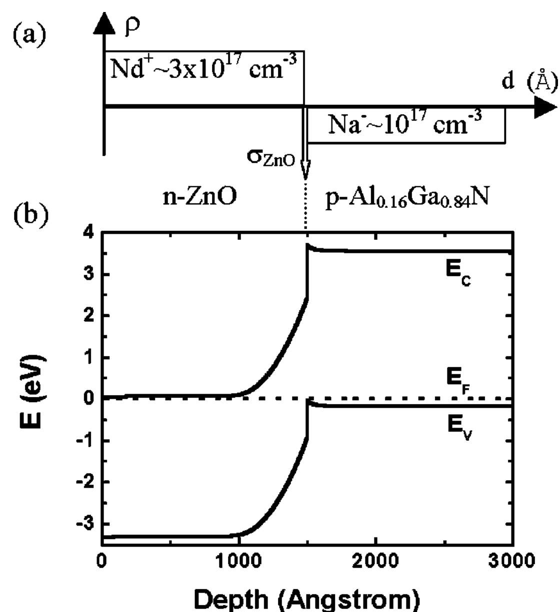

FIG. 1. (a) Charge distribution and (b) band diagram of a n-ZnO/p-AlGaN SHS considering polarization effect ($-\sigma_{ZnO}$).

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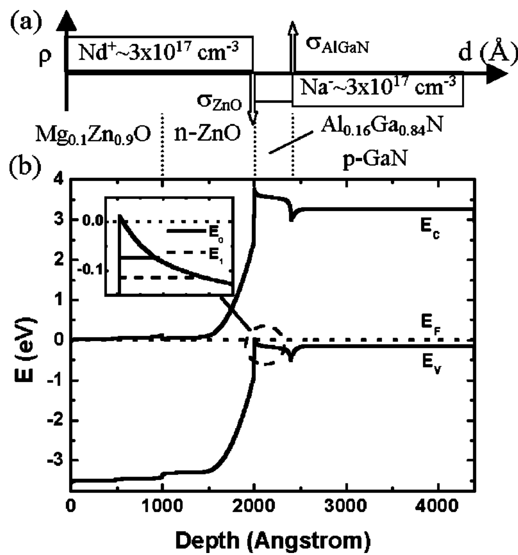

FIG. 2. (a) Charge distribution and (b) band diagram of the $p-n$ junction MgZnO/ZnO/AlGaN/GaN heterostructure. The layer thicknesses are 100, 100, 40, and 200 nm, respectively.

density for a typical ZnO/$Al_{0.16}Ga_{0.84}N$ heterostructure. Both layers were assumed to be 150 nm thick with free carrier electron and hole concentrations of about 3×10^{17} cm^{-3} and 1×10^{17} cm^{-3}, respectively. To account for the P_{PE}, the AlGaN layer was assumed to be partially relaxed with in-plane tensile strain of 1.95×10^{-3}. In accordance with the standard sign convention P_{PE} and P_{SP} in AlGaN are aligned in same direction and point towards the substrate. The ZnO layer on the AlGaN was also assumed to be partially relaxed with a compressive in-plane strain of 8.9×10^{-3}. In this case, P_{PE} and P_{SP} are aligned in opposite directions. Using the above relationship, a net negative polarization charge of $\sigma_{ZnO}=-6\times10^{12}$ cm^{-2} at the ZnO/AlGaN interface was calculated. The energy band structure reveals the presence of a hole accumulation layer at the ZnO/AlGaN interface due to the strong polarization effects.

The radiative recombination rate in a heterojunction is $R_{rad}\propto C_{Opt}np$, where C_{Opt} is the radiative recombination coefficient and *n and p* are the excess electron and hole concentrations near the $p-n$ junction interface, respectively. Based on this relation, R_{rad} near ZnO/AlGaN interface can be optimized by increasing the hole and electron concentrations. To enhance the hole and electron confinement near the interface, we examined a THS consisting of n-$Mg_{0.1}Zn_{0.9}O$/n-ZnO/p-$Al_{0.16}Ga_{0.84}N$/p-GaN. The free carrier electron concentration in MgZnO and ZnO was assumed to be $\sim3\times10^{17}$ cm^{-3}, with free hole concentrations in AlGaN and GaN assumed to be 1×10^{17} and 3×10^{17} cm^{-3}, respectively [Fig. 2(a)]. The total polarization mismatch between $Mg_{0.1}Zn_{0.9}O$ and ZnO layers at the interface was assumed negligible.[7] Taking the GaN layer to be relaxed, a net positive polarization charge of $\sigma_{AlGaN}=7\times10^{12}$ cm^{-2} was calculated for the AlGaN/GaN interface. Thus, addition of the GaN layer contributes a $+\sigma_{AlGaN}$ charge resulting in the formation of a triangular quantum well (TQW) for holes near the ZnO/AlGaN interface with quantized energy levels as shown in the inset to Fig. 2. The ground state and first excited state were calculated to be $E_0=-75.1$ meV and $E_1=-115.4$ meV, respectively, below the Fermi level. The TQW ensures strong confinement of holes near the ZnO/AlGaN interface and increases the probability of radiative recombination under forward bias. Additionally, the TQW with high free hole density aids the lateral conductivity in the structure, reducing the access resistance. The sheet concentration of holes in the TQW was calculated to be 3.5×10^{12} cm^{-2} when both ZnO and AlGaN were assumed partially relaxed, and 1.82×10^{13} cm^{-2} for the fully strained structure. In contrast to the SHS structure presented in Ref. 3, in which the carriers were spread throughout the thick p-AlGaN layer, the THS described here gives rise to strong hole confinement in the TQW enhancing R_{rad} at the interface. Due to conduction band offsets, the $Mg_{0.1}Zn_{0.9}O$ layer forms a barrier of ~100 meV for electrons injected into the ZnO providing additional confinement.

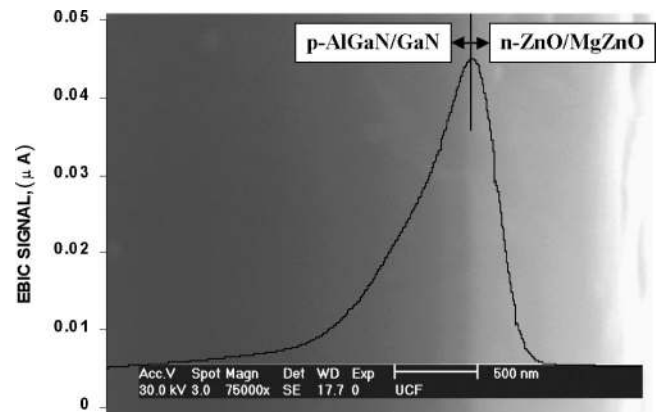

FIG. 3. Cross-sectional image of MgZnO/ZnO/AlGaN/GaN THS in secondary electrons (SE), EBIC line scan is superimposed with the SE image.

The THS LED structures considered above were grown by rf plasma-assisted molecular-beam epitaxy technique. A rf plasma source (model SVT-45) was used for producing atomic nitrogen and oxygen fluxes. Mesa geometry vertical devices were fabricated and characterized. The details on growth and fabrication are presented elsewhere.[5] The active region of the THS LED device is a $\sim$50–100-nm-thick unintentionally doped ZnO layer, sandwiched between a 150-nm-thick Ga-doped n-$Mg_{0.1}Zn_{0.9}O$ electron emitter and a $\sim$40-nm-thick p-$Al_{0.16}Ga_{0.84}N$ barrier that serves as a source for holes. The energy band diagram of the fabricated THS LED is essentially the same as that shown in Fig. 2.

Electron-beam-induced current (EBIC) measurements[8] were carried out at the $p-n$ junction cross section. Figure 3 shows the secondary electron image for the cleaved edge of the LED device superimposed with the EBIC line scan taken across the same structure. EBIC scans across $p-n$ junction allow the determination of the junction position. A pronounced peak in the EBIC signal coincides with the physical junction between n-MgZnO/n-ZnO (light on the picture) and p-AlGaN/p-GaN (dark on the picture) layers, indicating the presence of a $p-n$ junction at the interface. The diffusion lengths of minority carriers, extracted from the EBIC line scan, are 120 nm for holes in n-ZnO and 890 nm for electrons in p-AlGaN/GaN regions.

The characteristics of the fabricated ZnO/AlGaN-based $p-n$ diodes were measured at the wafer level using standard electrical and optical characterization methods. The measurements were conducted in the temperature range 300–650 K for LEDs operating in pulsed and cw modes. Figure 4 shows typical *IV* characteristics of forward biased THS LED devices measured at different temperatures. Rectifying *IV* char-

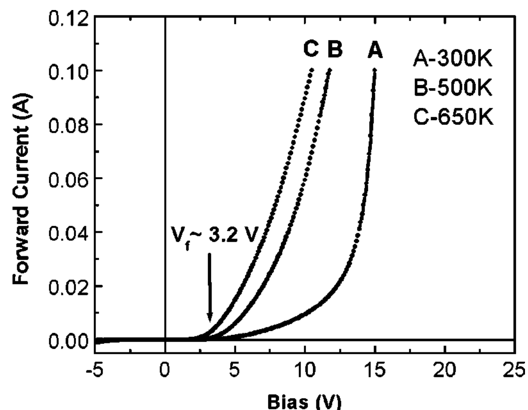

FIG. 4. Forward bias *IV* characteristics of THS UV LED device measured at different temperatures. Curves A, B, and C correspond to measurements at 300, 500, and 650 K, respectively. At 100 mA current, the series resistance is ~150 Ω, and is ~105 Ω for 300 to 650 K.

acteristics were measured up to 680 K. The turn-on voltage for these devices at 300 and 500 K was in ~3.2–3.6 V range, consistent with the ZnO bandgap. A reduction in the turn-on voltage to ~2.8 V was observed for LEDs at T =650 K. Room-temperature measurements suggest that there are two different mechanisms contributing to the current flow. Besides the exponential increase in current with forward bias near the turn-on voltage, we observe an additional rapid increase in the current at forward biases ⩾13 V. The mechanism responsible for this is not well understood, but one possibility is field induced tunneling of holes through the barrier at the AlGaN/GaN interface and another is the contact resistance to the *p*-layer. At higher temperatures this secondary current source becomes less apparent as the device series resistance decreases. Thermal activation of the Mg acceptors in *p*-type AlGaN/GaN part of the LED structure leads to lowering of the device series resistance at elevated temperatures.

EL spectra measured at different temperatures and driving currents are shown in Fig. 5. The emission band originates from the near interband transition in the ZnO active region near the *p*-AlGaN interface. The peak position of the emission band red-shifts from 390 nm (curve A) to ~391 nm (curve B) as the temperature increases from 300 to 500 K, which is consistent with the redshift observed for SHS.[3] A more significant redshift in the emission peak to 401 nm (curve C) occurs at 650 K. The full width at half-maximum of the spectra also narrows from 40 to 28 nm as the temperature is raised from 300 K to above 500 K. The observed broadening of the optical emission band at 300 K may be caused by spatial strain inhomogeneities in the ZnO active region. Another possible mechanism is optical recombination through different energy states localized in the TQW of the VB [inset in Fig. 2(b)]. The origin of the emission is the annihilation of excitons as suggested by optical absorption, photocurrent, and cathodoluminescence. A band, at zero bias, with a peak ~389 nm was measured in the photocurrent spectrum (curve D in Fig. 5). This band arises from excitonic absorption and the subsequent field-induced dissociation of the excitons in the ZnO layer by the built-in field ($\sim 8 \times 10^5$ V/cm) of the junction depletion region. A significant redshift to peak emission at 409 nm was observed for LEDs driven at 300 mA at 650 K, suggesting significant internal heating in the active region. Optical emission power increased linearly with forward current, providing additional evidence that the optical emission is interband related rather than impurity related. The optical power of a test device was ~10 μW at 300 K.

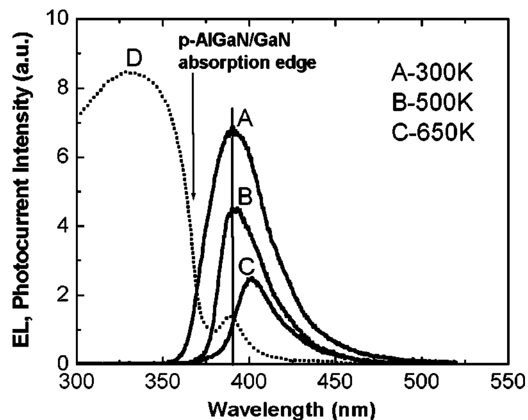

FIG. 5. The first three curves are EL spectra of UV LEDs measured, respectively, at 300, 500, and 650 K. The driving currents were 60, 150, and 160 mA, respectively. Curve D is the photoresponse spectrum at 300 K.

In conclusion, we have presented results for THS LEDs fabricated from MgZnO- and AlGaN-based materials. EL transitions were observed from a *p*–*n* device that were excitonic in nature. These optical emissions persisted to 650 K. ZnO-based emitters clearly show great potential for applications at high temperature.

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