Ultraviolet light emissions in MgZnO/ZnO double heterojunction diodes by molecular beam epitaxy

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ZnO double heterojunction structure was grown by molecular beam epitaxy. 100 nm MgZnO/ZnO/MgZnO well was inserted between Ga-doped ZnO and Sb-doped ZnO layers. X-ray diffraction spectrum confirmed the preferential growth along *c*-direction and secondary ion mass spectroscopy measurements showed a clear double heterojunction profile of this structure. Thin MgZnO layers made no difficulties for electrons and holes to get into active intrinsic ZnO layer. Dominant ultraviolet electroluminescence was observed at the injection currents from 40 to 80 mA at room temperature. The output power was 7.3 times as that from *p-n* homojunction diode at the same driving current due to a good confinement of electrons and holes in the intrinsic ZnO layer. © *2010 American Vacuum Society.* [DOI: 10.1116/1.3374436]

Ultraviolet (UV) light emitters are in strong demand for use in a variety of applications, such as solid-state lighting, displays, information storage, secure communications, and biosensors.[1,2] Among the available wide band gap materials such as diamond and GaN, ZnO has direct band gap of 3.37 eV and a large exciton binding energy of 60 meV, making it a candidate in the area of UV LED and laser diode.[3,4] The availability of large-area ZnO substrates and the possibility of wet etching also favor the fabrication of ZnO-based optical devices.[5,6] However, intrinsic defects such as Zn interstitials, oxygen vacancies, H incorporation, etc. evolve in the ZnO material during growth, making *p*-type doping of ZnO become the main hindrance in the development of ZnO based optoelectronic devices.[7] Several groups have reported *p-n* homojunction ZnO LEDs by various doping elements such as N,[8–10] P,[11–13] As,[14,15] and ZnO *p-n* heterojunctions which are mostly made from *n*-type ZnO and other *p*-type semiconductors such as SiC, GaN, and $SrCu_2O_2$.[16–18] They showed rectifying I–V characteristics and electroluminescence (EL) at room temperature, however, the near band edge emissions were not dominant and the efficiency was low.

Our group successfully developed a process to produce reliable *p*-type ZnO layers on Si with good optical properties by Sb doping.[19,20] We also successfully developed good Ohmic contacts to *n*-type and *p*-type ZnO films, which showed linear current-voltage (I–V) characteristics with low specific contact resistances.[21,22] We reported a *p-n* homojunction ZnO LED with Sb doped *p*-type layer grown on Si (100) substrate by molecular beam epitaxy (MBE).[23] The device produced a fairly good current rectifying behavior with an EL emission peak at 380 nm. The strain from the lattice mismatch between ZnO and Si substrate is relaxed by the formation of ZnO columnar structures with grain size from 100 to 400 nm, whose sidewalls terminate a great deal of threading dislocations glided from column/substrate interface. With lower dislocation density in the upper diode portion of each column, nonradiative recombination rates are reduced and UV near band edge emission is dominant.

The simplest type of *p-n* LED design is the first step to demonstrate the potential of a material for lighting application, however, is rarely used in current LED technology for lighting and illumination applications since there are some drawbacks which limit the device efficiency.[24] For improvement of efficiency, it is important that the region in which recombination occurs has a high carrier concentration. Designing of double heterojunctions is an excellent way to achieve such high carrier concentrations in active layer.

For ZnO based LEDs, a larger band gap barrier can be realized by incorporation of Mg into ZnO. However, *p*-type doping for MgZnO films is difficult to achieve,[25,26] so thin MgZnO layers with intrinsic ZnO layer in the middle were inserted between *n*-type and *p*-type ZnO layers to confine the carrier recombination process.[12] The two cladding or confinement $Mg_{0.54}Zn_{0.46}O$ layers have a larger band gap than the ZnO active region.

In our experiments, ZnO double heterojunction was grown on *n*-type Si (100) substrate ($1-20\ \Omega$ cm) using MBE system. A thin magnesium oxide (MgO) buffer layer was deposited at 350 °C to reduce the lattice mismatch between Si and ZnO, followed by 450 nm Ga-doped *n*-ZnO at 550 °C. A thin MgZnO layer of about 1.5 nm was then deposited, followed by a 100 nm thick intrinsic ZnO, and on top is another 1.5 nm thick MgZnO layer. Then the temperature was increased to 600 °C to grow 450 nm Sb-doped *p*-ZnO. This diode sample with defined mesa size $800 \times 800\ \mu m^2$ was fabricated by employing standard photolithography techniques and HCl etching. Au/Ni (200/10 nm) and Au/Ti (200/10 nm) were then deposited on *p*-type ZnO and *n*-type ZnO, respectively, by lift-off process. The contacts were annealed at 800 °C for Au/Ni and 700 °C for Au/Ti, respectively, to become Ohmic. The device was packaged onto TO5 can using conductive epoxy resin.

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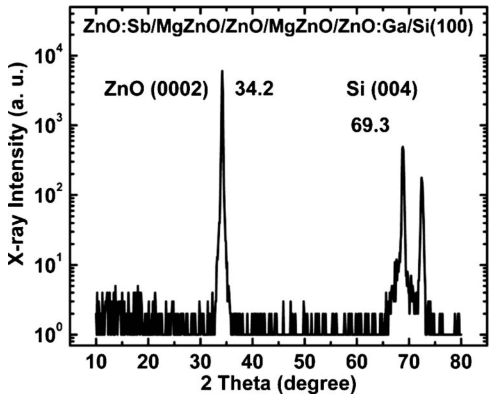

FIG. 1. XRD spectrum of MgZnO/ZnO/MgZnO double heterojunction on Si (100) substrate.

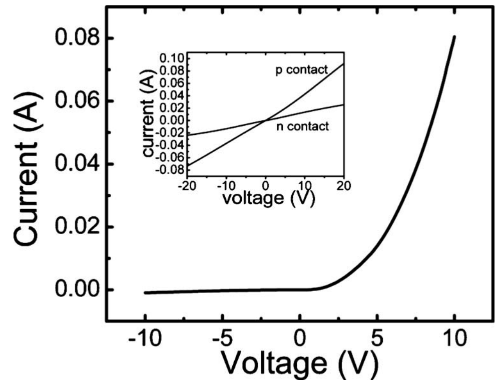

FIG. 3. I–V characteristics of the double heterojunction diode, showing typical rectifying behavior. The inset shows the I–V curve of both *n*-type and *p*-type contacts.

X-ray diffraction (XRD) $\theta/2\theta$ scans show that the diode film grows preferentially along the *c*-direction of the ZnO wurtzite lattice (Fig. 1). The elemental distribution of Zn, O, Mg, Sb, Ga, and Si was obtained by performing secondary ion mass spectroscopy (SIMS) measurements, as shown in Fig. 2. Dominant distribution of Sb and Ga dopants are in *p*-type and *n*-type layers, respectively. Mg profile shows that the MgZnO/ZnO/MgZnO double heterojunction is formed in between Ga–ZnO and Sb–ZnO layers.

Current-voltage (I–V) characteristics were measured using Agilent 4155C semiconductor parameter analyzer and Signatone probe station, as shown in Fig. 3.The inset figure shows fairly good Ohmic contact made by Au/Ti for *n*-layer and Au/Ni for *p*-layer. The diode shows fairly good rectification behavior with a threshold voltage of about 2.5 V, comparable to the threshold voltage of a *p*-*n* homojunction diode without MgZnO/ZnO/MgZnO double heterostructure.[23] These results suggest that the thin MgZnO layers do not make difficulties to electrons and holes injecting to the well area.

Electroluminescence (EL) measurements were performed using an Oriel monochromator and photomultiplier tube at room temperature. The diode was biased under dc forward voltages. Figure 4 shows the EL spectra obtained at different injection current. Near band edge emission at 3.2 eV appears when the current is 40 mA, while in ZnO homojunction device it starts to appear when the current is larger than 60 mA.[23] Afterward, the intensity of this emission increases as the injection current increases from 40 to 80 mA. There is an abnormal decrease in intensity when the current increases from 50 to 60 mA and the intensity almost has no change as the current changes from 70 to 80 mA. This is partly due to the heat effect as a result of the increasing current through the diode, and partly due to the degradation of device with short life time. Because of poor film quality, EL emission intensity degraded even with the same driving current as the previous measurement. The near band edge emissions show redshifts with increasing current due to heat induced band gap reconstruction.

Figure 5 shows a comparison of EL emission spectrum between double heterojunction and homojunction devices under the same injection current of 70 mA. The homojunction consists of the same *n*-ZnO and *p*-ZnO layers without the middle MgZnO/ZnO/MgZnO double heterostructure.[23] The two devices were measured under the same optical system at room temperature. Relative output power was calculated by integrating intensity with step of energy. The calcu-

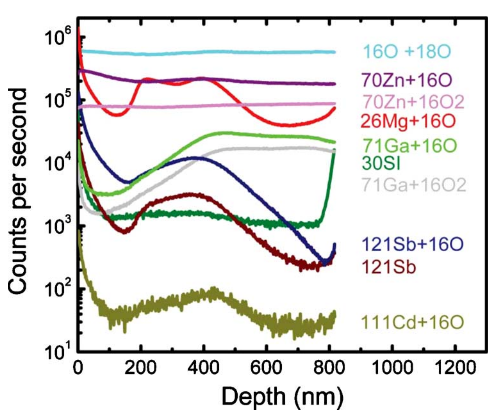

FIG. 2. (Color online) SIMS result of MgZnO/ZnO/MgZnO double heterojunction on Si (100) substrate. The elemental profiles of Sb, Ga dopants, Zn, O, Mg, and Si substrate can be seen.

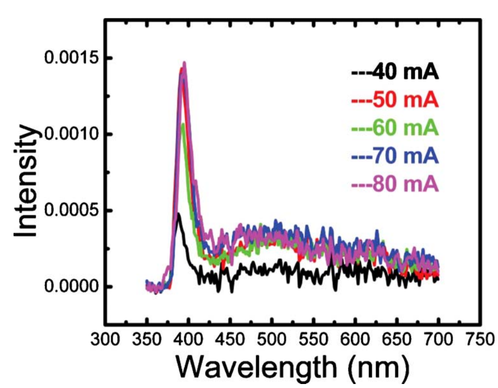

FIG. 4. (Color online) Electroluminescence spectra of ZnO double heterojunction diode at room temperature with increasing injection current from 40 to 80 mA.

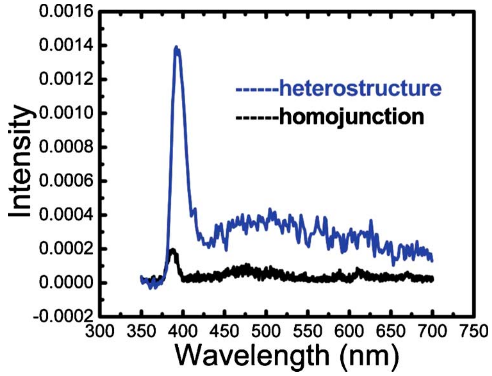

FIG. 5. (Color online) Electroluminescence spectra of MgZnO/ZnO/MgZnO double heterojunction diode and ZnO *p-n* homojunction diode at the same drive current of 70 mA.

lated output power of heterostructure device is 7.3 times stronger than that from the *p-n* homojunction device. The improvement shall come from the confinement of electrons and holes made by MgZnO barrier layers, leading to an enhanced radiative recombination rate in the active ZnO layer.

In summary, UV MgZnO/ZnO/MgZnO double heterojunction light-emitting diode on Si (100) substrate has been realized. Au/Ni and Au/Ti make good Ohmic contacts to *p*-type and *n*-type ZnO layers. I–V measurement shows good rectification behavior and EL experiments demonstrate dominant UV emissions. Compared to ZnO homojunction diode, the output power of this device is 7.3 times stronger at the same injection current, which demonstrates a better confinement of electrons and holes to recombine in an intrinsic ZnO layer.

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