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Distributed Bragg reflector assisted low-threshold ZnO nanowire random laser diode

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An electrically pumped nitrogen doped p-type ZnO nanowires/undoped n-type ZnO thin film homojunction random laser with a 10-period SiO_2/SiN_x distributed Bragg reflector is demonstrated. The formation of p-n homojunction is confirmed by the current-voltage and photocurrent characteristics. The random lasing behaviors with a low threshold of around 3 mA are observed. The output power is measured to be 220 nW at a drive current of 16 mA. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4870513]

Random lasers have attracted much attention owing to their various potential applications such as imaging, sensing, and medical diagnostics.[1–3] ZnO nanowire is not only natural Fabry-Perot cavity[4,5] but also excellent random lasing medium due to its large refractive index and exciton binding energy.[6] A great deal of work has already been spent on the optically pumped random lasers, and further development of electrically pumped random lasers is critical for practical applications. ZnO nanowire random lasers based on a p-n junction[7] and metal insulator semiconductor structures[8–10] have been demonstrated recently. Nevertheless, the threshold current of these lasers is still too high and shall be decreased.[11,12] On the other hand, the output power of the electrically pumped nanowire random laser devices also needs to be increased. To achieve these goals, one way is to reduce the loss at the light reflection or scattering. Distributed Bragg reflector (DBR) structures, which are formed from multiple layers of alternating materials with different refractive index and have high reflectivity, can significantly reduce the threshold pumping density and enhance laser performance.[13,14] Here, we report the fabrication and characterization of low-threshold DBR-assisted ZnO nanowire random laser device with a p-n homojunction. The output power is significantly enhanced with the assistance of DBR.

The device structure consists of nitrogen doped p-type ZnO nanowires/n-type ZnO thin film/SiO_2/SiN_x DBR on Si (100) substrate. The 10-period 60.9 nm SiO_2/48.7 nm SiN_x DBR structure was grown by plasma enhanced chemical vapor deposition (PECVD). Both calculation and experiment results show that the DBR gives desirable optical quality and the reflectivity at 380–390 nm reaches 95%. Details can be found elsewhere.[14] After the DBR deposition, the substrate was immediately transferred to a molecular beam epitaxy (MBE) system, and a 500 nm undoped n-type ZnO seed layer was grown. To grow nitrogen doped p-type ZnO nanowires, 200 SCCM argon diluted oxygen (0.5%) as reaction gas and 50 SCCM argon diluted N_2O (0.5%) as dopant gas were introduced into a CVD furnace. A flow of 1000 SCCM. nitrogen carrier gas was passed continuously through the furnace during the growth. Zinc (Zn) powder (99.999% Sigma Aldrich) was placed in a silica bottle and served as source material. The substrate was positioned at the center of the silica tube, which was about 1 cm away at the downstream side from the Zn silica bottle. The growth temperature is 650 °C. A piece of silicon wafer was used to cover a portion of the substrate during the growth for subsequent electrical contact formation on the ZnO thin film.

Figure 1(a) shows the top view scanning electron microscope (SEM) image of the ZnO seed layer. The film consists of compact columnar structures. Figure 1(b) shows x-ray diffraction (XRD) spectrum of the seed layer, indicating that these columns grow preferentially along the c-direction of the ZnO wurtzite lattice. The morphology of the seed layer is desirable for subsequent growth of nanowires.

Figures 1(c) and 1(d) show the top view and side view SEM images of the as-grown nanowires, respectively. Slightly tilted nanowires with different diameters from 100 nm to 600 nm are observed. The average length of the nanowires is about 9 μm, although the length of nanowires varies. The size nonuniformity of the nanowires may be due to rough surface of the columnar ZnO seed layer on DBR structure. Figure 1(e) shows x-ray photoelectron spectroscopy (XPS) result of nitrogen doped ZnO nanowires. Two N 1 s related peaks at around 397.8 eV and 406.8 eV are evident. Similar peaks were observed in the previous work,[7] which confirms the doping of nitrogen and indicates possible origin of nitrogen related shallow acceptors.[15] From the XPS result, the approximate atomic concentration of nitrogen is 0.1%.

Figure 1(f) shows low-temperature photoluminescence (PL) spectrum of the nitrogen doped ZnO nanowires at 20 K. An He-Cd laser with an excitation wavelength of 325 nm was used in this experiment. The acceptor bound exciton (*AX*), free electron to acceptor (*FA*) emission, and donor-acceptor-pair (*DAP*) emission peaks are observed at 3.359 eV, 3.305 eV, and 3.232 eV, respectively. These emissions arguably indicate the formation of shallow acceptors in the nanowires. Comparing to the reported PL spectrum of nitrogen doped p-type ZnO nanowires,[7] additional peaks are observed in the PL spectrum of this sample, which originate from the PL oscillations due to the DBR structure.[16]

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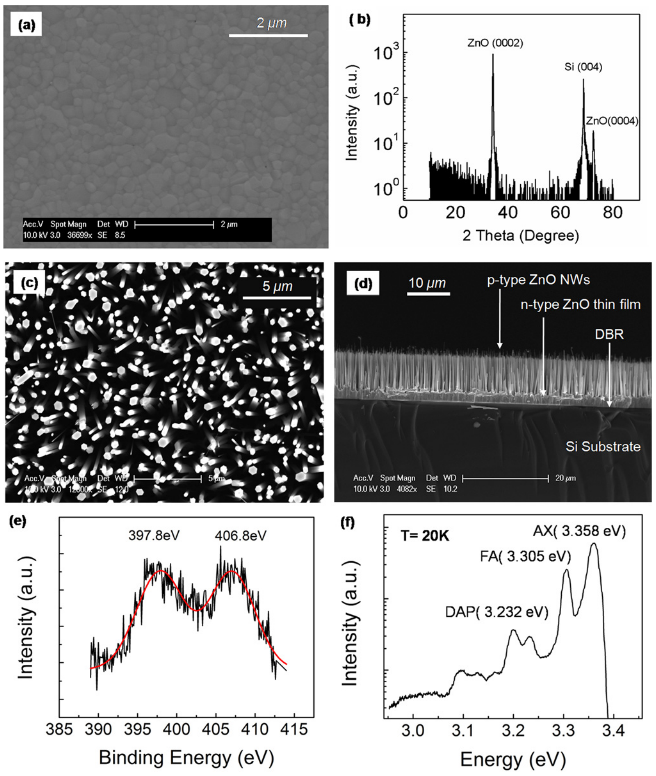

FIG. 1. (a) SEM image of ZnO seed layer on DBR. (b) XRD spectrum of the seed layer. (c) Top view and (d) cross-sectional SEM images of nitrogen doped ZnO nanowires on ZnO seed layer on DBR. (e) XPS result showing nitrogen related peaks for nitrogen doped ZnO nanowires. (f) Low-temperature PL (20 K) spectrum of nitrogen doped ZnO nanowires.

The schematic of the device is shown as an inset of Figure 2. The ITO glass was clamped on the top end of the nanowires as the top electrode. Ti/Au (10 nm/100 nm) was deposited on the undoped n-type ZnO thin film as the ground electrode using e-beam evaporation. During the Ti/Au metal evaporation, the nanowires were protected by an aluminum foil. Figure 2 shows current-voltage (I-V) characteristics of the ZnO p-n junction device with and without UV illumination. The UV illumination was carried out using an Oriel Xe arc lamp. I-V curve in dark exhibits rectifying behavior with a turn-on voltage of about 5.5 V, which confirms the formation of nitrogen doped ZnO nanowire/undoped ZnO thin film p-n homojunction. Very good UV response in the reverse biased regime is observed. UV response in the forward bias region, which is significantly smaller than that in the reverse bias region, is also observed. It should be noted that if the device is an ideal p-n junction photodetector, the total current under forward bias shall instead decrease with respect to the dark current. In addition, the short-circuit photocurrent at 0 V is only on the order of about 2 nA, which is much smaller than photocurrents under other forward and reverse biases. These results suggest that the increased current under forward bias as well as current change in the reverse bias region may have originated from other sources, namely, the p-n photodiode is most possibly in series with a photoconductor. The photoconductor characteristic is mainly due to the photo-carrier generation and collection in the n-type thin film. Other factors include the emission from the surface trap states and electron-hole pair generation in the nanowires away from the junction.[17]

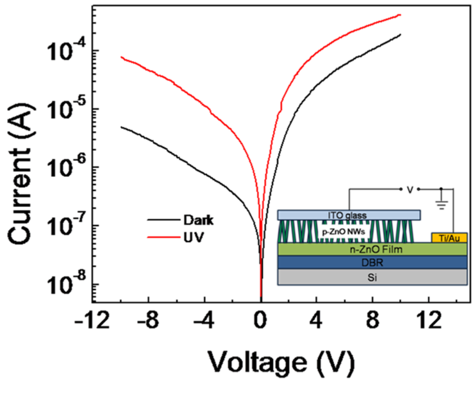

FIG. 2. I-V characteristics of the ZnO nanowire/ZnO thin film structure in dark and under UV light. The Ti/Au contact on thin film is grounded. The inset is a schematic of the laser device.

Photocurrent (PC) at different wavelengths was measured using a homebuilt system. The light from the Oriel Xe arc lamp passed through an Oriel 0.25 m monochromator and a specific wavelength light came out from its output port. The light was then cast on the device after chopping. The generated PC signal was collected by a lock-in amplifier. Figure 3 shows the PC spectra of the device operated at 0 V and different reverse biases. The PC peaks are at about 380 nm (3.26 eV), which correlates to the band gap of ZnO. The PC is a result of the photocarriers generated by the absorption of light in the space-charge region. The PC increases with the increase of the reverse bias due to the enhancement of carrier generation from increased space-charge region. These results, in particular the existence of photocurrent under photovoltaic mode, further confirm the formation of the p-n junction.

Figure 4 shows electroluminescence (EL) spectra of the device at different forward biases. At a drive current of 2 mA, only background noise is observed in the EL spectra. Weak EL spontaneous emission shows up when the current is increased to 2.5 mA. The intensity of the spontaneous emission from around 370 nm to 410 nm increases at a higher current of 3 mA. At the same time several lasing peaks appear at 3 mA although they are relatively weak and broad. The lasing peaks become stronger and sharper with a linewidth of ~1.5 nm at 3.5 mA. More lasing peaks are evident at 4 mA. The spacing between the adjacent lasing peaks varies and the peak positions between different measurements are different. These results indicate that the lasing peaks in the EL spectra are related to the random lasing

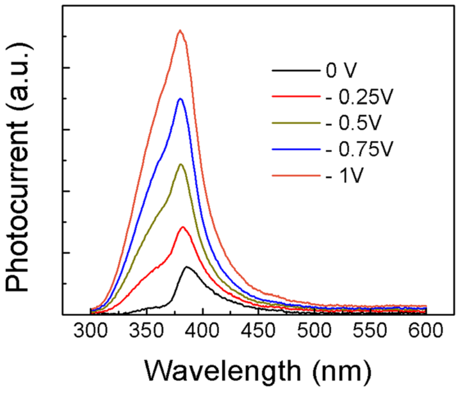

FIG. 3. Photocurrent spectra under zero and reverse biases.

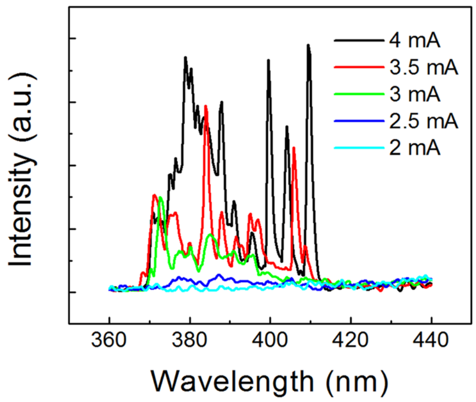

FIG. 4. EL spectra of the laser device operated at the drive current from 2 mA to 4 mA.

instead of F-P lasing. As seen from the SEM images, the nanowires are slightly tilted, thus the DBR reflector and nanowires top facets are not parallel. Therefore, the parallel reflectors for F-P laser do not exist in our device. The random lasing phenomena are related to random light scattering among the nanowires.

The output power of the device was measured using a Thorlabs PM100 Optical Power Meter and Figure 5 shows the result. The dashed lines are the guide to the eye, showing a threshold current of ~3 mA. The integrated intensity of the EL spectra (Figure 5 inset) as a function of injection current also shows a threshold current at around 2.8 mA. The output power is about 220 nW at a drive current of 16 mA. The output power is much higher while the drive current is much lower than our previous device without DBR structure with the same device size of 1 cm by 1 cm, where a threshold current is 40 mA and only 70 nW output power is obtained at 70 mA under the same measurement setup.[7]

For a random laser, different random cavities emit lasing signals in different directions due to random scattering. A detector placed at a certain angle to the sample can only collect lasing signals along the direction. In our measurement setup, the detector was placed at the normal direction to the sample surface. The enhanced signals should be related to the reflection from the DBR because the nanowires are slightly titled with respect to the normal of the substrate surface. In addition, the light initially emitted from the diode propagates all directions and a large portion is toward the substrate direction. In the device without DBR,[7] the ideal reflectivity between ZnO and Si interface at around 380 nm is only around 20%, and most of the light penetrates into the silicon and is absorbed by the substrate. Since the reflectivity of the DBR structure in the present device reaches 95% at 380–390 nm, most of light will be reflected back to the ZnO nanowires/thin film structure, leading to more light scattering among the nanowires. The threshold of a laser device is strongly related to the reflectivity and it can be reduced by increasing the reflective of the reflectors.[18,19] For both the devices with and without DBR, the reflectivity of the top ITO and ZnO interface is the same. The much lesser loss from the DBR results in a lower threshold current and a higher output power.

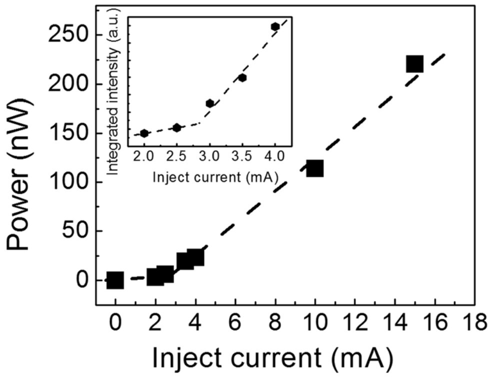

FIG. 5. Output power as a function of drive current of the laser device. The inset is the integrated intensity of the EL spectra versus drive current. Both measurements show the threshold at around 3 mA.

In conclusion, nitrogen doped p-type ZnO nanowires were grown on undoped n-type ZnO thin film on a 10-period DBR structure of alternative layers made from SiO_2 and SiN_x. The nitrogen doped nanowires were studied by XPS and low-temperature PL measurements. The p-n junction was confirmed by I-V and photocurrent characteristics. The device showed strong random lasing emissions. The threshold current as low as 3 mA was achieved and the output power was also increased to be around 220 nW owing to the high reflectivity of the DBR.

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