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Lasing from ZnO Nanorods on ITO-coated substrates

M.N. Nordin[a], D. Priante[b], M.K. Shakfa[b], and W. Maryam[*a]

[a]School of Physics, Universiti Sains Malaysia, 11800, Penang, Malaysia
[b]Photonics Laboratory, Computer, Electrical, and Mathematical Sciences and Engineering (CEMSE) Division, King Abdullah University of Science & Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

ABSTRACT

Lasing was observed from ZnO nanorods prepared by a simple method of chemical bath deposition (CBD) on ITO-coated glass substrates. The X-ray diffraction pattern showed a dominant peak for (002) plane typical for good crystalline quality of ZnO grown in the z-direction with a wurtzite structure. Continuous-wave photoluminescence (PL) spectra revealed a peak centered at 380 nm corresponding to the band gap of ZnO. Under pulsed optical pumping, lasing was observed above the nominal PL peak, initially for one mode at 384 nm. Two additional modes at 386 nm and 390 nm was observed when the pumping power is further increased. Threshold was achieved at 0.7 µJ which was 10 times smaller than that reported for powder-based random lasers. In addition, gain pinning was also observed for the dominant mode and the additional two modes appeared upon onset of this gain pining behavior.

Keywords: Random lasing, ZnO vertical nanorods, laser mode switching, chemical bath deposition.

1. INTRODUCTION

Conventional lasers require a cavity made of parallel mirrors for lasing to occur. However, lasing may also occur in a high scattering gain medium whereby the so-called 'cavity' is a closed loop path created by the photons [1]. When these photons are in such a gain medium, they stimulate more photons and build up in the closed loop path. Above a certain threshold, lasing is then observed. This kind of lasing, termed as random lasing, has been observed recently in ZnO nanostructures prepared on glass substrates by chemical bath deposition (CBD) method, also referred to as wet chemical method [2]–[4] and other simple methods such as sol gel and vapour phase techniques [5]–[7]. However, to realize electrically pumped random lasers, it is important to prepare the nanorods structure on a conductivity layer for electrical contacts. Hence, in this work, random lasing in ZnO nanorods prepared by CBD on ITO coated glass substrates is presented. As photon backscattering may be geometrical dependent, it is likely that a controlled random laser is realized in this case for nanorods or vertical nanowires due to the closely packed arrangement of nanorods minimizing out of plane scattering. If the photons go through similar scattering paths, it is possible to retain the same number of modes. Typically in a random laser, a random increase in terms of the mode and number of modes is expected.

Annealing ZnO has been shown to reduce random lasing threshold and to increase the chance of lasing [7]. However, upon only sample's annealing at a temperature as low as 600 ºC, lasing was observed with a low threshold compared to structures prepared by other simple growth techniques like sol-gel methods that require annealing above 700 ºC to reach the lasing threshold [8]. As annealing reduces point-defects due to O-vacancy, Zn-vacancy, O-interstitial and Zn-interstitial, near-band-edge luminescence would increase, which in turn may reduce the threshold condition. Previous work on ZnO random lasers on ITO coated substrates were from powdered ZnO prepared by electrophoresis. For these samples, the threshold was as high as 763 kW/cm^2 (equivalent to 76.3 kJ/cm^2) and the number of lasing peaks were more than 10 [9]. To our knowledge, no works on random lasing from ZnO nanorods prepared on ITO coated substrates by CBD have been reported.

In this work, we report on random lasing observed from ZnO nanorods prepared by CBD on ITO coated substrates. Under pulsed optical pumping, at the maximum pump power, three narrow lasing peaks at 384 nm, 386 nm and 390 nm were observed with a full width half maximum (FWHM) between 0.35 nm and 0.52 nm. Lasing was observed when the

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incident pump pulse energy exceeds threshold as low as 0.7 μJ (energy density of 100 mJ/cm^2); which is more than 10 times less than that for powder-based random lasers [10]. The 384 nm mode remains except at higher pumping energy. . The lasing observed here shows stability in the number of modes for a range of pump energy, which is an important characteristic for laser-based optical sensors [11], [12]. In addition, the gain saturation was observed for the mode with the lowest threshold.

2. EXPERIMENTAL METHOD

The ZnO nanorods were grown on ITO-coated substrates by CBD method with a pretreated seed layer of 200 nm thickness. The uniform seed layer was obtained using radio-frequency sputtering technique and annealed at 600 ºC before immersing the substrate in the chemical solution. The as-grown ZnO nanorods samples were also annealed at 600 ºC. The morphology of the ZnO nanorods was characterized by field emission scanning electron microscope (FESEM; Nova Nano SEM 450, FEI, Japan). The crystalline structure of the nanorods was analyzed by high-resolution X-ray diffraction (HRXRD) system with a Cu Kα radiation source of λ= 1.5406 Å. Micro-photoluminescence (μ-PL) was used to study the optical properties and random lasing behavior from the samples. The μ-PL setup utilizes a fourth harmonic from Nd: YAG pulsed laser (20 KHz repetition rate, 550 ps pulse width) with an excitation wavelength of 266 nm for random lasing measurements and a continuous-wave (CW) 325 nm HeCd laser for typical PL measurements. All measurements were performed at room temperature.

3. RESULTS AND DISCUSSION

Figure 1(a) shows FESEM image of the surface of the grown ZnO nanorods. No uniform pattern in shape and size has been observed. However, the FESEM image shows a good population of nanorods grown vertically from the substrate surface. The nanorods varied in size which may be attributed to multifaceted orientation of the seed layer, resulting in coalesced formation of the nanorods [13]. Figure 1(b) shows an XRD pattern obtained from the sample. One can see a dominant peak at 34.4º which refers to ZnO in the (002) plane. This indicates the preferential growth of the nanorods along the c-axis direction perpendicular to the surface of sample. This is in line with the FESEM image shown in Figure 1 (a). Figure 1(c) shows the PL spectrum of the sample recorded at room temperature using CW laser source. The spectrum shows a strong dominant near-band-edge emission in the ultraviolet (UV) region, namely from the recombination of free excitons. The peak observed is centered at 378 nm, which is typical for ZnO thin films. On the visible region, no defects-related peaks was detected. This indicates that the sample has low defect- and site vacancy densities.

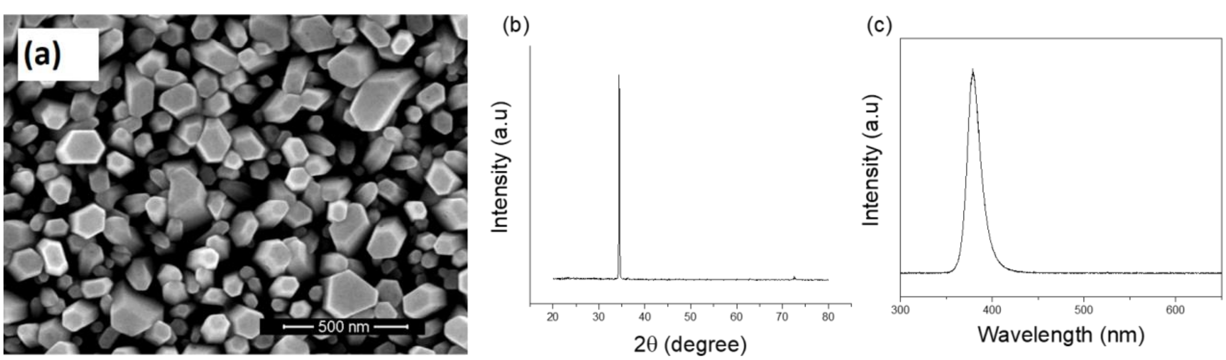

Figure 1: (a) SEM image of ZnO nanorods prepared on an ITO-coated substrate view from top. (b) XRD spectra showing the dominant (002) peak at 34.4 degrees, typical for ZnO grown in the c-direction. (c) CW-PL spectrum of nanorods with a single UV emission band centered at 378 nm.

The sample was optically excited with a laser spot size of 8 µm perpendicular to the sample surface. The emitted light from the sample was then collected normal to the surface. Figure 2 shows sharp narrow peaks that arise from the broad spontaneous UV emission band due to preferential amplification at frequencies close to the maximum of the gain spectrum. The narrow line-widths provide an evidence of stimulated emission from the nanorods array. Threshold occurred at 100 mJ/cm^2 with one lasing peak emerging at 384 nm. Single mode lasing was observed until the pump energy density reached 258.8 mJ/cm^2 whereby the number of peaks multiplied into three. These modes were observed at 384 nm, 386 nm, and 390 nm and maintained with incising the pump power. The low threshold of the observed lasing emission can be attributed to the high reflectivity of the optical reflectors formed by the nanorods. In addition, the closely packed population form vertical walls minimizes out of the plane scattering. Therefore, enhancing long paths for the photons can then benefit from high gain over the entire spot size [1], [14]

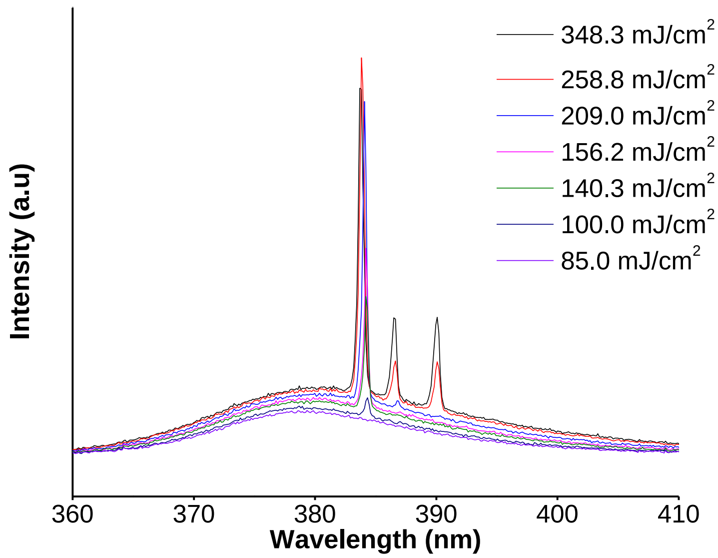

Figure 2: Lasing spectra above the spontaneous PL peak emission under different excitation energies. Threshold is observed at an energy density of 100 mJ/cm^2 with one lasing mode at 384 nm. Above 209 mJ/cm^2, a second mode appeared followed by a third mode at 258.8 mJ/cm^2.

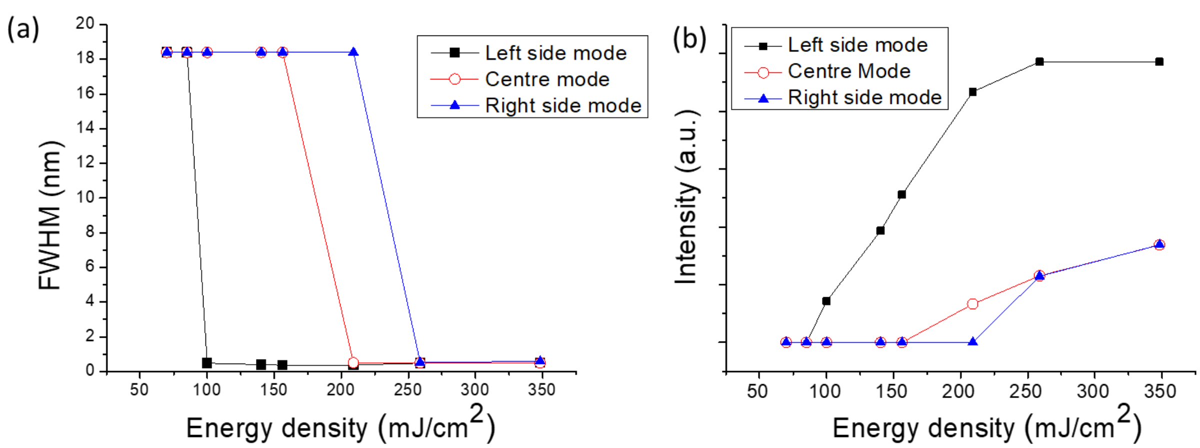

Figure 3: (a) FWHM of the lasing modes. (b) Peak intensity of each lasing peak as a function of the pumping energy density showing spontaneous emission region followed by stimulated emission and for the left side mode.

Figure 3 (a) shows the FWHM of the lasing peaks against the pump energy density. Left-side mode, center mode, and right-side mode are the peaks at 384, 386, and 390 nm, respectively. Lasing was observed initially for the left-side mode and emission from other modes were only observed at higher pump energies indicating the different threshold for each lasing mode. The Q-factor ($\lambda/\Delta\lambda$) is between 800 and 1100 for each mode which is considerably higher than that for previously reported ZnO-based random lasers [15], [16]. An increase in Q-factor of a random laser cavity has been shown to coincide with lasing that lasts longer and stronger laser emission [17]. Figure 3 (b) shows the intensity of the lasing modes with increasing pump energy density. Most intriguing, the saturation of lasing intensity for the left mode was observed above a pump energy density of 209 mJ/cm^2. Such a behavior has been observed in optically pumped GaN Febry Perot lasers, termed as gain-pinning behavior [18]. Interestingly, once the left mode achieved gain saturation, other modes start to overcome threshold and emerge. To our knowledge, this behavior has not reported for ZnO-based random lasing. This can open the possibility of switching to different modes by changing the pump power for advanced sensor applications [11].

4. CONCLUSION

In conclusion, lasing from ZnO nanorod array prepared by CBD on ITO-coated glass substrates were observed. Threshold was achieved at 100 mJ/cm^2 followed by a gain-pinning behavior above 209 mJ/cm^2. When the gain-pinning was achieved, two additional modes appeared. This suggests a relationship between gain-pinning and threshold behavior for other additional modes. Results also suggest a great potential for using this method of CBD to synthesize random-laser materials for future integrated nanophotonic devices.

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