Fabrication and characterization of electrically pumped disordered ZnO lasers*

Myela Paige[1], B.S., Kannatassen Appavoo[2], PhD, Matthew Y. Sfeir, PhD[2]
[1]University of Maryland Baltimore County, Baltimore, MD, 21250, paigemy1@umbc.edu
[2]Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, NY 11973, msfeir@bnl gov

Abstract*– The development of low-cost, high-efficiency, light amplification processes is crucial for the future of high-performance in photonics. The electrical and optical characteristics of solution processed random lasers are being studied to change optically pumped to electrically pumped devices. Hydrothermally grown ZnO nanospheres in a thin film have exhibited low lasing thresholds and a reduction of void space, but electrical shorts are an ongoing issue. ZnO nanosphere-film about 400 nm in thick comprised of nanospheres (~35nm average size) were spun onto indium-tin-oxide layered glass and atomic layer deposition was used to infill the voids in the solution processed thin film with Al_2O_3 to assist with the reduction of electrical shorts. The diode-like behavior has been exhibited through J-V curves in ITO structures because of holes being injected into the ZnO films. Infilling the ITO device decreases the lasing threshold which allows the device to show electroluminescence and future applications in optoelectronic devices, such as displays.

Keywords— infilling; lasing; photonics; zinc oxide

I. INTRODUCTION

In nanotechnology, devices are comprised of many different materials for many different uses ranging from biomaterials to shoe repellant. The ability for metal oxide nanospheres to be versatile make them attractive because of their functionality and diverse properties. Zinc oxide (ZnO) is a promising semiconductor material being used in many applications: sensors, storage, and optical and electrical devices. ZnO is attractive because of its wide band gap at room temperature. ZnO is particularly interesting because of the versatility of the growing methods. ZnO can be grown as nanowires, a thin film, and nanospheres. [1]

We demonstrated intrinsic (cavity-free) lasing at a low threshold in an ultrathin film of randomly arranged zinc oxide nanospheres.[2] Lasing in solution-processed nanomaterials has gained significant interest because of the potential for low-cost integrated, low-power, high-efficiency integrated photonic devices. Most commercial lasers are well ordered, but ZnO exhibits random lasing where everything is scattered, but still emits at high intensities. There has not been an exhibition of random lasing in a thin film as thin as 400nm. We have fabricated and characterized an electrically pumped random ZnO laser that has the potential to use for sensor and photonics applications. [2] In this work, the fabrication process the indium-tin-oxide (ITO) devices and the optical and electrical results of the lasers will be discussed.

II. METHODS

ZnO is particularly sought after because of its wide band gap and its structural consistency when grown. ZnO is versatile on the nanoscale level and is size and structure dependent based on the morphology and synthesis process. In this work, we focus on ZnO nanostructures because of their robust and highly crystalline structures with a high refractive index with low-thresholds and random lasing. We used hydrothermally grown zinc oxide nanospheres (~35nm in size) dispersed in ethanol 40% by wt. concentration was further diluted in ethanol to 0.35% volume concentration (Figure 1).

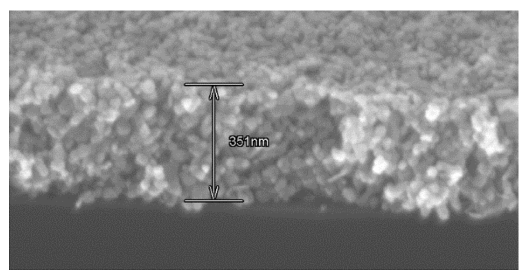

Figure 1. Ultra-thin film of ZnO nanospheres obtained by SEM

To obtain diode-like behavior and electroluminescence from the ZnO nanosphere-film, we need a p-i-n junction. P-i-n junctions are used primarily for the hole and electron injection for increased current and decrease the emission voltage. We aim to obtain a structure for optimized hole injection into the intrinsic layer (i) of the ZnO nanospheres. We created two devices with different base substrates to understand and test the optical and electrical properties of the ZnO and lasing by using (ITO) as the devices' base substrates.

Imaging. Scanning electron microscopy (SEM) was used to observe the random structure of the ZnO nanosphere-film on a 70° cross-section holder. SEM revealed nanosphere-film is

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about ~400nm thick, nanospheres shown to be on average 35 nm in size and somewhat uniform in shape.

ITO Device Structure. Ideally we would want a laser that is transparent for visible emission. The ITO is a transparent layer of conductive material spun onto one side of glass substrate. The glass is conductive because of the ITO, which is an n-type semiconductor. We sputtered ~100nm of ZnO onto the ITO to help make the ZnO more n-type. We spin coated the nanospheres and used low temperature ALD to infill the ITO device. Using PVD, we deposited ~70nm of p-type 60-40 gold/palladium alloy 2mm diameter electrode on top of the ZnO nanosphere- films (Figure 2).

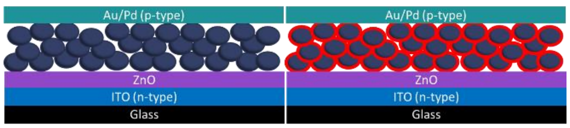

Figure 2. Schematic of ITO control (left) and ALD (right) device

Electrical Characterization. We used bias sweeping for current measurements on each type of device. We measured the current using a bias sweep from -1V to 4V.

Optical Characterization. Photoluminescence (PL) and electroluminescence (EL) were performed on the infilled ITO sample by electrically pumping the sample through the electrodes, where the ITO was ground. Steady –state emission was used to the effect of ALD on the emission intensity.

III. Results

Current-density (J-V) graph (Figure 3) shows the ITO ALD device had a turn on voltage of about 3.2V. This is reasonable because the band gap for ZnO is about 3.4eV.[1] The control device did not exhibit diode-like [1]behavior and we attribute that to the fact the devices were shorted while depositing the electrodes.

The ALD device emitted light around the edge of the visible light spectrum and UV light spectrum (~400nm). The normalized comparison of EL and PL line (Figure 4) in shows that the infilling ITO did not create defects in the device.

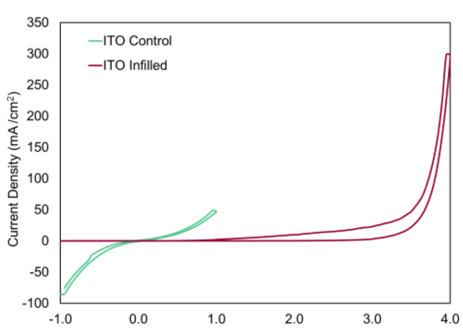

Figure 3. Current density as a function of bias voltage graph for ITO devices

The peak intensity wavelength of the EL measurement is slightly higher than the PL peak intensity wavelength because of possible increase of the work function of the ZnO from electrical stimulation.

Effects of ALD. Steady-state emission has revealed the lasing threshold's dependence in the device. ALD reduces the lasing threshold infilled sample due to enhanced scattering compared to the ITO control sample. The infilling also shows the prevention of shorting due to Al_2O_3 creating a barrier between the p-type contact and the nanospheres.

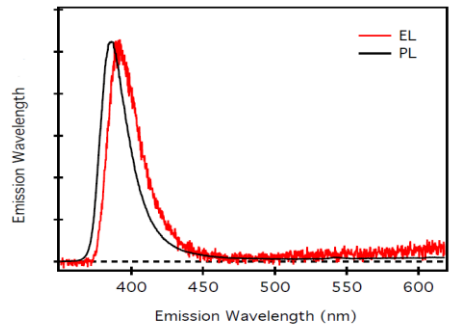

Figure 4. PL vs EL graph of emission intensity as a function of emission wavelength

IV. Conclusion and Outlook

The work revealed the distinct differences in electrical measurements and did show how dependent the lasing threshold is on the device structure. The ALD acted as the barrier in the ITO infilled device and the device optically and electrically lased possibly due to possibly the Al_2O_3 barrier that hindered the rate at which we injected electrons in to the ZnO region.

There are promising applications for electrically pumped ZnO lasers in high-performance optoelectronic devices. The potential of these applications are based on the amount of light is emitted through electroluminescence and the optimization of the laser size. In the future, we plan on changing the geometry of the device, contact materials, and testing ranges to obtain a highly efficient, low-cost, random ZnO laser.

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References

[1] M. Vaseem, A. Umar, and Y. Hahn. *Metal Oxide Nanostructures and Their Applications*, American Scientific Publishers, 2010. pp. 1–36

[2] K. Appavoo, X. Liu, V. Menon, and M. Sfeir. "Excitonic lasing in solution-processed subwavelength nanosphere assemblies." *Nano Letters*. Vol 16, no. 3, pp 2004-2010. March 2016

[1] http://maeresearch.ucsd.edu/mckittrick/index_files/Page945.htm