Waveguiding-assisted random lasing in epitaxial ZnO thin film

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Zinc oxide thin films were grown on c-sapphire substrates using pulsed laser deposition. Pump power dependence of surface emission spectra, acquired using a quadrupled 266 nm laser, revealed room temperature stimulated emission (threshold of 900 kW/cm^2). Time dependent spectral analysis plus gain measurements of single-shot, side-emission spectra pumped with a nitrogen laser revealed random lasing indicative of the presence of self-forming laser cavities. It is suggested that random lasing in an epitaxial system rather than a three-dimensional configuration of disordered scattering elements was due to waveguiding in the film. Waveguiding causes light to be amplified within randomly formed closed-loops acting as lasing cavities. © *2010 American Institute of Physics*. [doi:10.1063/1.3527087]

Properties of zinc oxide (ZnO) are the subject of intense research[1] and many optoelectronic applications are emerging. ZnO is interesting as it has intense excitonic ultraviolet stimulated emission, due to high exciton binding energy (60 meV) and a direct wide band-gap of ~3.37 eV.[1]

We studied random laser action, observed in many different forms of ZnO.[2] A random laser has no deliberately formed cavity, and light amplification occurs scattering at dielectric reflectors. With enough scatterers due to optical gain, light can reflect back to its initial position forming closed-loops, and act as self-forming laser cavities. Random lasing was seen in GaAs, ZnO, and TiO_2[3] and reported in powders,[3] in dyes embedded in films,[4] and in highly textured[5] and nonoriented polycrystalline films.[6] Electrical pumping was also achieved.[7] In contrast to these results, we report random lasing in an epitaxial thin film.

In this study, ZnO thin films were grown on c-sapphire (c-Al_2O_3) substrates using pulsed laser deposition (PLD). High resolution x-ray diffraction studies were conducted in a Panalytical X-Pert MRD PRO system using a Cu $K\alpha$ source. The incident beam optics comprised a four-bounce Ge (220) monochromator and a multilayer mirror. In the diffracted beam path, the system was equipped with a three-bounce Ge (220) monochromator. Optical emission was investigated using two geometries: in the first, a frequency-quadrupled neodymium-doped yttrium aluminum garnet laser (266 nm, 5 ns pulse duration, 10 Hz frequency, and 4 mm spot diameter) was employed as a pump with the beam having an angle of 45° to the sample surface. The emission was collected normal to the sample surface using a spectrometer (50 cm focal length) coupled to a Peltier-cooled CCD camera. The second "edge-emission" geometry used a nitrogen pump laser emitting at 337 nm (5 ns pulse duration and 10 Hz frequency) with the incident beam normal to the sample surface and the collection at the edge of the sample (at 90° from the excitation beam, see the inset in Fig. 4).

In the edge-emission configuration, a cylindrical lens with a 35 mm focal length was employed to create a stripe with a width of 30 μm, with a power density of up to 6.6 MW/cm^2. The CCD camera acquisition time was fixed at 100 ms in order to obtain "single-shot spectra" (corresponding to the 10 Hz frequency of the laser.

Gain measurements were taken using the variable stripe-length (VSL) method. In this approach, the laser stripe length is defined using a movable beam-block. Side-emission intensity is then measured as a function of the stripe length. This allows for the excitation volume to be increased while maintaining a constant pump power density.

X-ray diffraction measurements revealed epitaxial growth[8,9] with a FWHM of 0.006° for the (0002) peak high resolution omega scan and an (open-detector) omega rocking curve value of about 0.05°, characteristic of a highly oriented material with very low crystallographic dispersion. The 2θ-ω scan of the (0002) peak gave a c lattice parameter of 5.215 Å. This is slightly higher than the equilibrium value, suggesting the film was under compressive stress in the basal plane. The 2θ-ω scan also showed strong Pendellösung oscillations, characteristic of a high parallelicity of the crystal planes combined with a smooth sample surface over a relatively large area compared with the laser spot size (several mm^2). Fringes spacing gave an estimate of film thickness at 140 nm as compared to a value of 159 nm calculated from angularly resolved photoluminescence (PL) and spectroscopic ellipsometry measurements (not showing).

For surface emission PL spectra are shown in Fig. 1. Figure 1(a) presents the evolution of the luminescence as a function of the pump power for an accumulation of 50 single-shot spectra at various power densities. The inset shows a plot of integrated emission intensity as a function of pump power. A stimulated emission threshold of 900 kW/cm^2 was identified and is a typical value for an epitaxial ZnO thin film.[10] For a pump power of 400 kW/cm^2, there is spontaneous emission is absorbed at 378 nm due to near band edge recombination. Above the stimulated emission threshold, however, the main peak is over ~394 nm. This is most likely due to electron-hole plasma recombination.[11] With increasing pump power, peak broadening and red-shifting are observed. This is probably attributable to band-gap renormalization.[12] Figure 1(b) pre-

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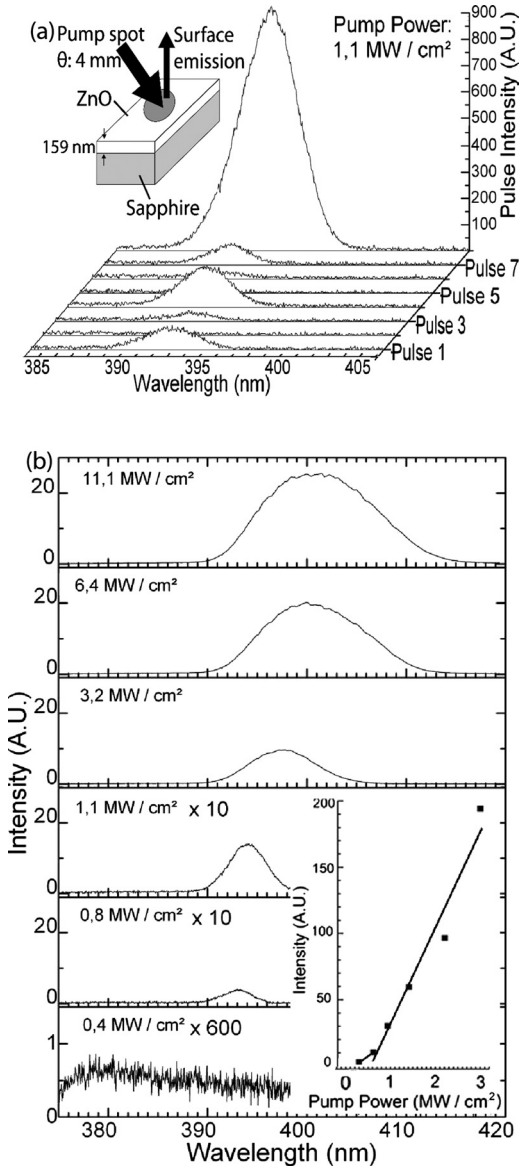

FIG. 1. (a) Single-shot surface emission PL spectra. Inset: experimental configuration used. (b) Accumulated spectra for 50 surface emission pulses at different pump powers. Inset: integrated emission intensities at different pump powers. The threshold is at 0.9 MW/cm^2.

sents a series of single-shot spectra, for which the power density was just above the stimulated emission threshold (1.1 MW/cm^2). The PL intensity was observed to fluctuate significantly between spectra on a random basis over time.

Figure 2(a) shows groups of five single-shot side-emission spectra acquired at various pump powers. For a pump power of 3.7 MW/cm^2 (above threshold), individual spectra show multiple emission peaks, with each having significantly lower line width than observed for the surface emission spectra (at similar pump powers). This is consistent with the presence of laser cavities in the sample, which select precisely defined wavelengths. Moreover, the various emission peaks were observed to toggle on and off randomly over time (as was also observed for the surface emission), which is coherent with varying lasing modes being excited by each pulse. Thus it was concluded that random lasing was occurring in the film, with different self-forming laser cavities being selected at each pump pulse. Figure 2(b) shows 50 spectra juxtaposed for different pump powers (ranging from

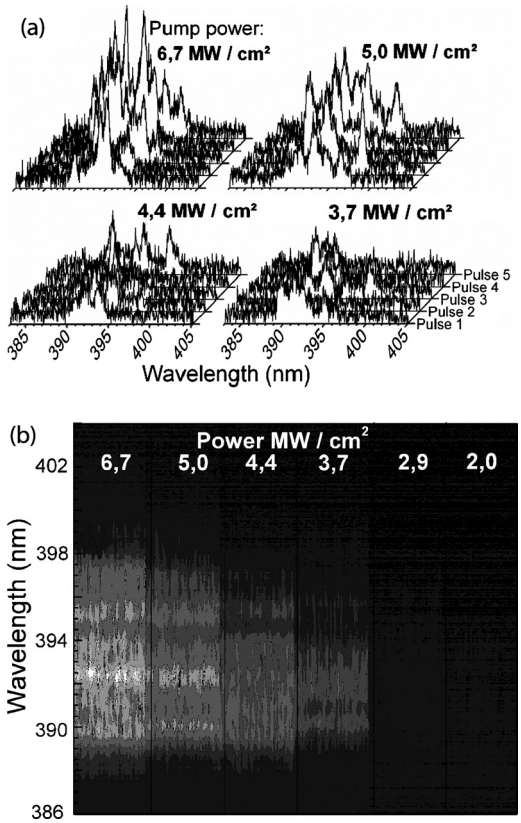

FIG. 2. (a) Several single-shot side-emission spectra at various pump powers. (b) Three-dimensional contour plot for single-shot side-emission spectra at different pump powers. The *y*-axis represents the wavelength while the *x*-axis represents the pump power from the right (2 MW/cm^2) to the left (6.7 MW/cm^2). Each section of pump power contains 50 spectra side by side. The *z*-axis represents the intensity, which increases from low intensity (black) to high intensity (white), while going through intermediate intensity (grey). Note the recurrence of certain peak positions at higher powers.

2 to 6.7 MW/cm^2) as a contour plot. We see that although the peaks in the single-shot spectra occur randomly, they appear at well-defined positions, which recur over time. Figure 3 shows the result of accumulating 50 side-emission

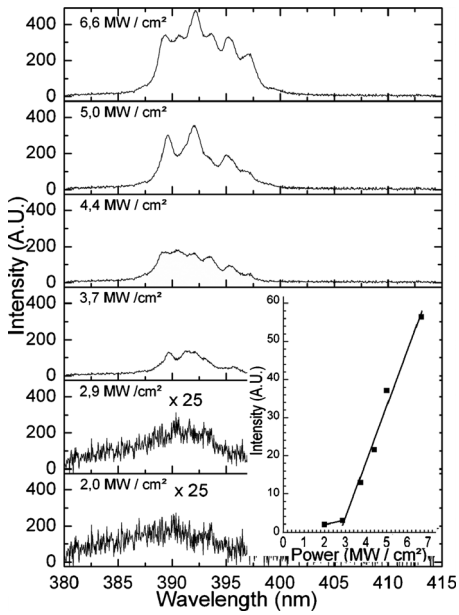

FIG. 3. Averaged spectra for 50 side-emission pulses at different pump powers. Inset: integrated emission intensities as a function of pump power. The stimulated emission threshold is 2.9 MW/cm^2.

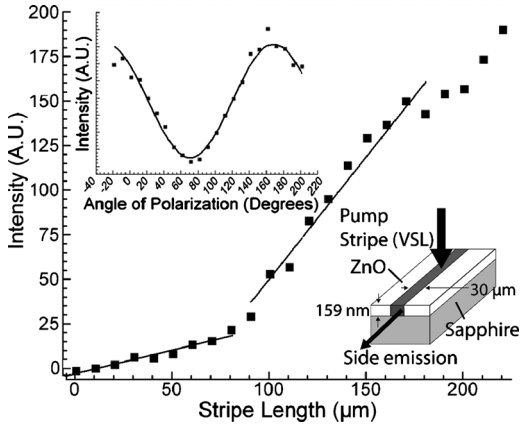

FIG. 4. Side-emission VSL gain measurements (bottom inset). Top inset: emission intensity vs angle of polarization. The maximum corresponds to the TE mode.

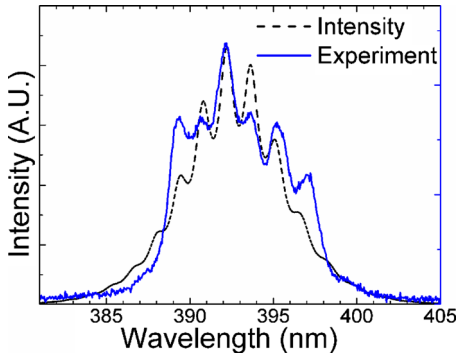

FIG. 5. (Color online) Simulated Fabry–Pérot oscillator (cavity length of 12.03 μm and reflectivity of 1%) with Gaussian gain (maximum: 471 cm^{-1}) fitted against averaged PL spectra at 6.6 MW/cm^2 pump intensity.

spectra at various pump powers. The inset shows the dependence of the spectrally integrated emission intensity as a function of the pump power. A stimulated emission threshold of 2.9 MW/cm^2 deduced. We note that spontaneous emission, observed subthreshold, appeared at a higher wavelength (i.e., 390 nm) than for the normal collection case. This may be due to waveguiding in the film, which would favor TE over TM polarization mode at lower wavelengths. At higher pump powers, the output begins to saturate. Moreover, the stimulated emission thresholds are slightly different for the surface and edge emission. This may be the result of using a stripe laser (with a length of 4 mm and a depth of 200 μm, i.e., more than the film thickness) combined with a cylindrical lens for the side configuration.

Using VSL measurements (Fig. 4), lasing threshold was found to depend not only on the laser pump power but also on the excitation volume, as was concluded in studies on random lasers elsewhere.[13] As the excitation volume increases beyond a threshold of 482 μm^3 (a stripe with a length of 90 μm and a depth of 159 nm), laser action takes place. Peak separation was not observed to vary as a function of stripe length, however, which distinguishes these results from those typically observed (e.g., Tang *et al.*[14]) suggesting our findings should not be interpreted in the same way. The peaks had widths between about 0.3 and 2 nm for single-shot spectra and a gain, g, of 160 cm^{-1} (cf. Fig. 4), which is coherent with the literature for comparable samples.[15]

Light emitted was polarized, with all peaks exhibiting a maximum corresponding to the TE mode (see the inset of Fig. 4). This indicates that the ZnO thin film may act as a waveguide, as observed before,[14] and is also consistent with the red-shift observed for the spontaneous emission in side-collection.

Having an epitaxial thin film poses questions on the origin of the random lasing. Figure 5 shows a simulation for a simple Fabry–Pérot cavity spectrum fitted to these results (assuming Gaussian gain and including cavity wavelength dispersion caused by the change of refractive index in ZnO). Fitting parameters included a maximum gain amplitude, g_{max}, of 471 cm^{-1}, 1% reflectivity, and a cavity length of 12.03 μm. We stress that g_{max} is a nonadjustable parameter as a gain of 160cm^{-1} was measured by VSL (see Fig. 4) (i.e., the Gaussian fit for the gain was made for a FWHM of 160 cm^{-1}, resulting in g_{max} of 471 cm^{-1}). This simulation fits the experimental results fairly well meaning we have scattering due to reflection at defects in the film. We point out the deduced "cavity length" (12.03 μm) does not necessarily imply that there are actual cavities of this dimension in the sample, but rather that the typical path of the photon before arriving back at its original position is of the order of 12 μm. While such a one-dimensional Fabry–Pérot model can be considered as oversimplified, the probability for a photon to return to its emitted point can be deduced from the reflectivity to be 0.01. As far as the scattering process is concerned, it has been reported that space charge effect at grain boundaries increases the refractive index of the medium locally and, therefore, act as a mirror for the light.[16] This hypothesis needs confirmation using high resolution transmission electron microscopy observations.

In conclusion, room temperature excitation dependence of optical gain spectra was investigated for thin films of ZnO epitaxially grown on c-sapphire substrates by PLD. Time dependent analysis of single-shot side-emission spectra using a VLS approach revealed room temperature random lasing from self-forming cavities with a threshold of 2.9 MW/cm^2. It was proposed that waveguiding in the film enhanced the random lasing. The origin of the scattering, however, remains uncertain. Such an experimental approach could be used as a tool to characterize thin film quality via random lasing observations.

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