*Supporting Information*

# Excitonic Lasing in Solution-Processed Subwavelength Nanosphere Assemblies

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**Scanning Electron Microscopy images of the disordered ZnO nanosphere film.**

In Figure S1, we show a cross-sectional SEM image that shows the extended lateral dimension of the film represented in Figure 1a of the manuscript.

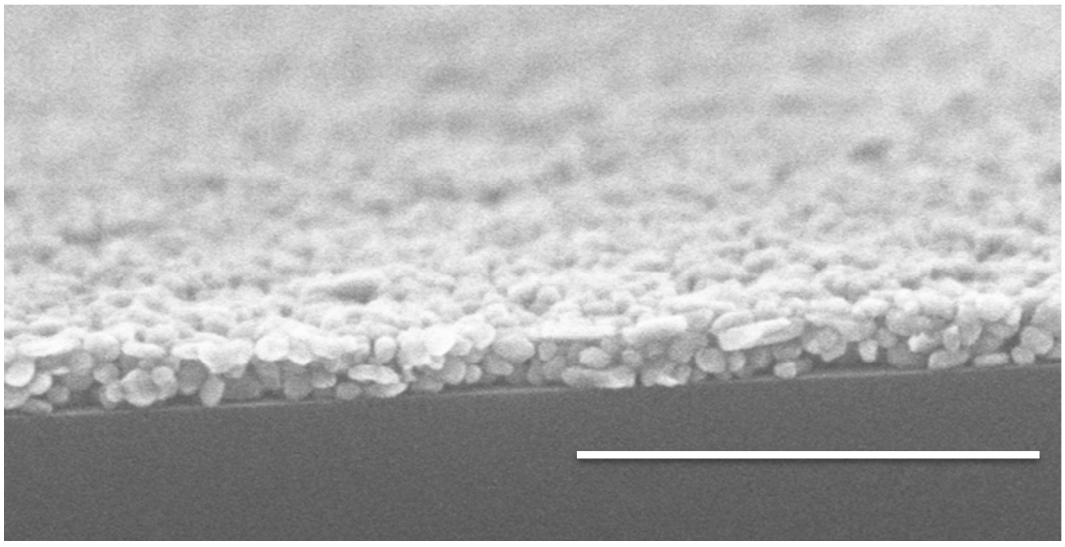

**Figure S1.** Cross-sectional SEM images showing the thickness and disorder of a ZnO nanosphere films that exhibits random lasing. The scale bar represents 500 nm.

**Excitation Spot Size Measurements**

Lasing behavior was observed for all pump spot sizes used in this measurement. By modifying the experimental geometry to measure the samples in transmission mode, we were able to vary the spot size from ~ 2 μm to 1 mm. As can be seen in Figure S2, line narrowing and threshold behavior is apparent for all pump sizes. Below threshold, the luminescence spectra are identical. Above threshold, variations in the lasing spectrum are observed for different spot sizes.

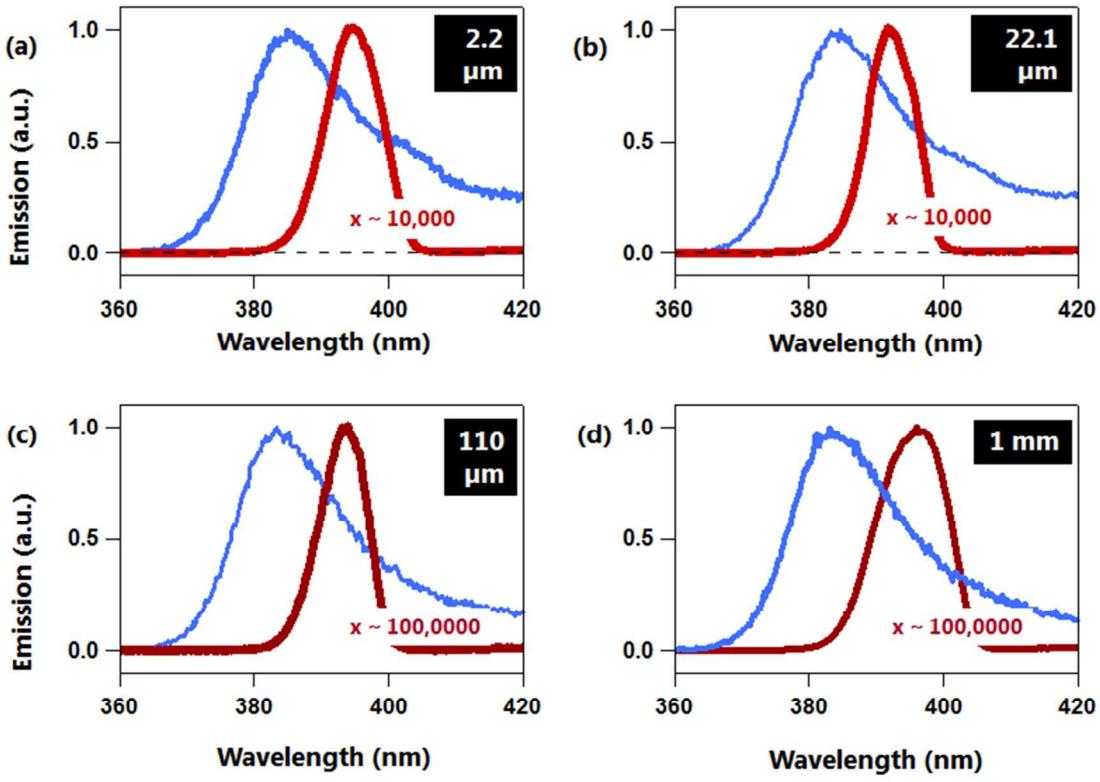


**Figure S2.** Below (blue) and above (red) threshold pumping for a pump spot size of (a) 2.2 μm, (b) 22.1 μm, (c) 110 μm, and (d) 1 mm.

**Ultrafast Optical Kerr Gating for Time-Resolved Emission Studies.**

To record temporally the broadband emission features of our samples, we use a home-built ultrafast emission spectrometer as schematically shown in Figure S3 and described in details elsewhere.[1] Notably, the modular design of our emission spectrometer can either probe weak emission features[2] with a temporal resolution of ~530 fs when benzene is used as the Kerr medium or alternatively, intense emission can also be recorded with a temporal accuracy of ~220 fs when using quartz. The time-resolution was determined by performing cross-correlation at 380 nm, that is near the band-edge emission of the zinc oxide (ZnO), using pulses derived from the optical parametric amplifier. The gate pulse has a temporal duration of ~100 fs.

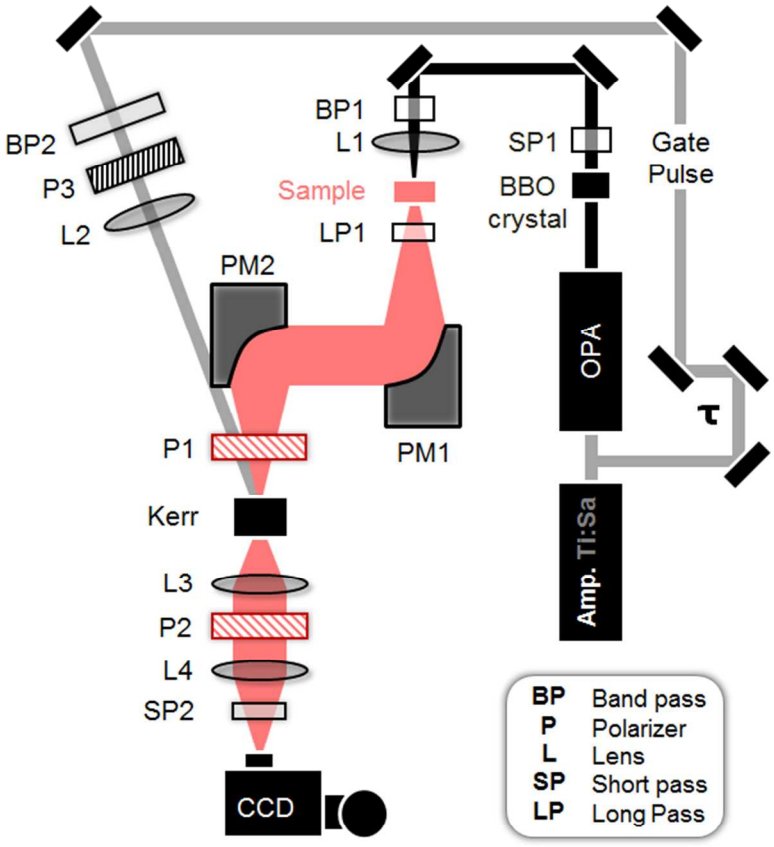


**Figure S3.** Ultrafast broadband optical Kerr spectrometer. A schematic representation of the experimental setup. For details, see text.

**Position and Temporal Variations of the Lasing Spectra**

Because of the random nature of the lasing process, we observe slight changes in the lasing intensity and spectrum as a function of sample position (S4a) and time (S4b). A 2 μm spot size was used for these measurements.

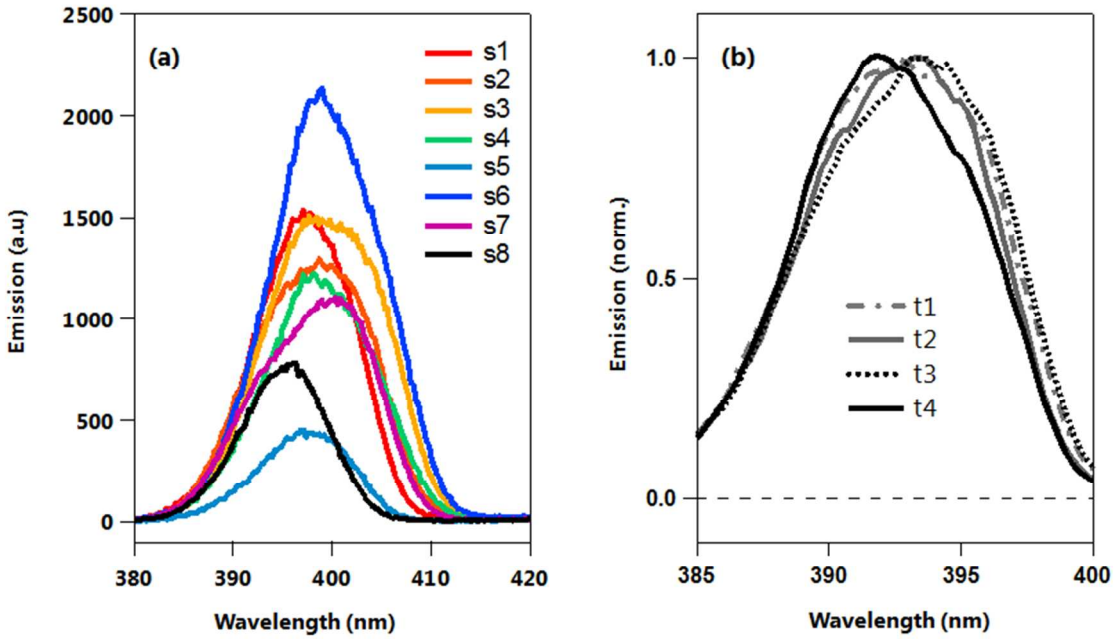


**Figure S4.** (a) Different sample spots show differences in the lasing intensity and spectrum at the same pump fluence (~ 500 μJ/cm$^2$). (b) Sample spot s8 measured four consecutive times with 100 ms integrations (~ 100 shots). Small spectral differences can be clearly seen.

**Evidence of discrete lasing modes, with narrower bandwidth than steady-state measurement:**

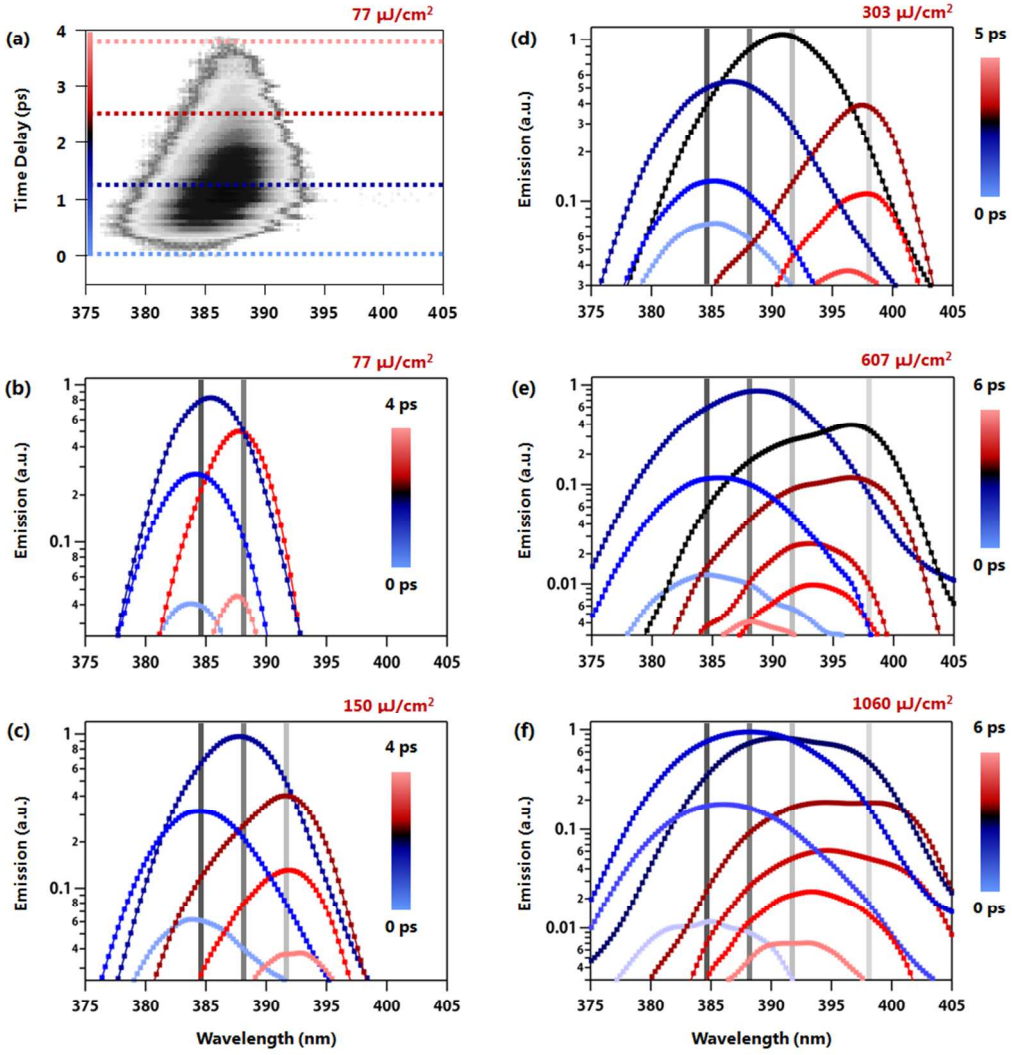


**Figure S5. Temporal dynamics of discrete lasing modes.** (a) A 2D dimensional transient emission (log intensity, greyscale) scan shows the evolution of the lasing modes at 77 μJ/cm$^2$, right above the threshold for lasing, as a function of time and wavelength along with (b) spectral slices at different time points that show the distinct temporal characteristics of individual lasing modes. (c-f) At higher pump fluences, the mode density increases and the spectral dynamics become increasingly complex. Vertical lines are guides to the eyes and represent the various emission peak positions.

**Calculations of the void as a function of nanostructure size**

Assuming that the nanostructures in the thin films are spherical and arranged in an hexagonal closed packed lattice, with a 74 percent packing ratio, we calculate (i) the number of nanostructures found in a volume defined as the cubic diameter of the nanostructures and (ii) the space between the two spheres. Figure S6 shows the result. For example, decreasing the nanospheres diameter from 200 nm to 35 nm results in a decrease of the void size by about 90 percent.

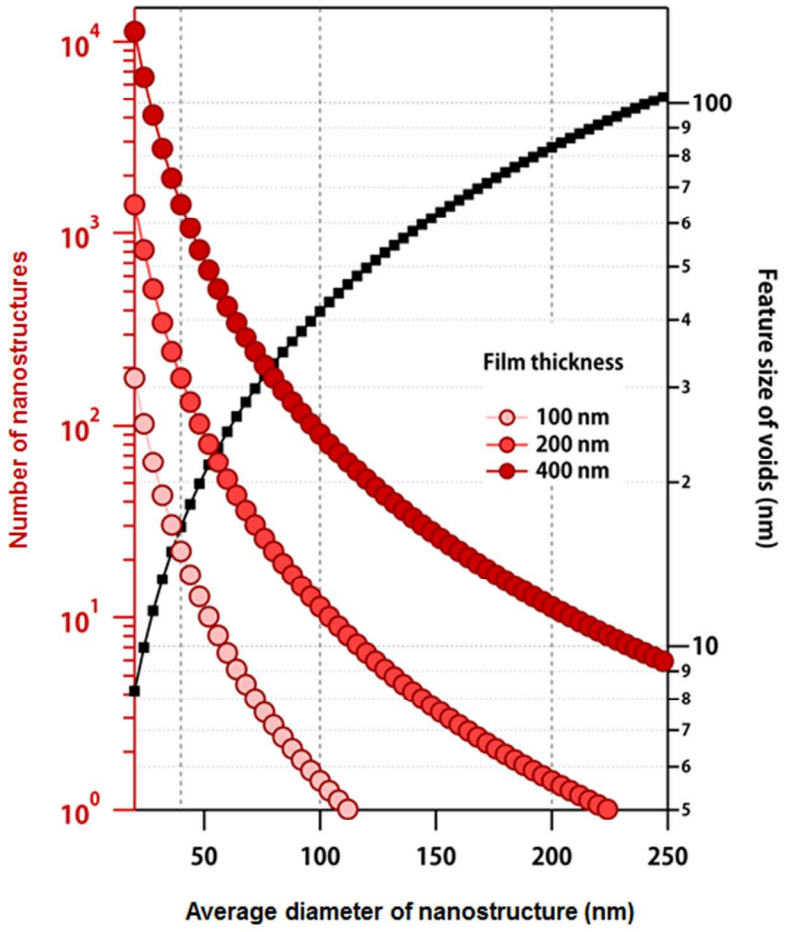


**Figure S6. Number of nanostructures and feature size of the void as a function of film thickness and diameter.** The simulations are carried out for 3 film thicknesses of 100 nm, 200 nm and 400 nm. The black line represents the feature size of the voids for a given nanosphere size (right scale).

**References**


1. Appavoo, K.; Sfeir, M. Y. *Rev. Sci. Instrum.* **2014,** 85, (5), 055114.

2. Appavoo, K.; Liu, M.; Black, C. T.; Sfeir, M. Y. *Nano Lett.* **2015,** 15, (2), 1076-1082.