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Random Laser Dynamics in Disordered and Semi-ordered Cavities

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Outline

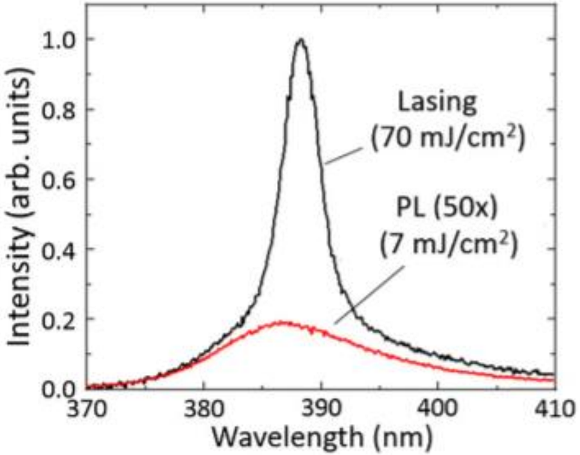
- Instabilities in ZnO random lasers
- Reproducibility with different pump pulse times
- Self-consistent time-dependent laser theory
- Dynamics and stability

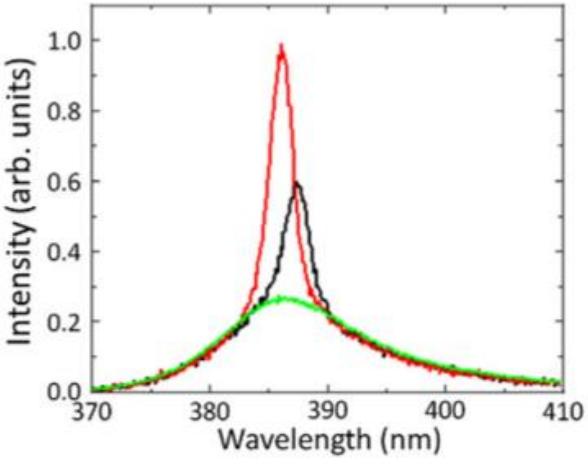
ZnO Random Lasers

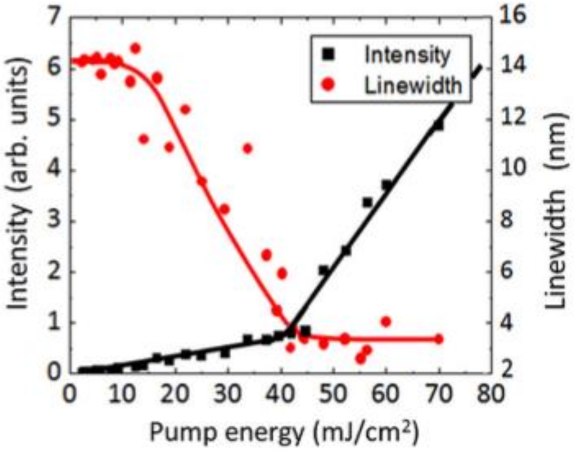
- ZnO: 3.37 eV bandgap, 60 meV exciton binding energy
- Carrier recombination rate $\gamma_a \sim (200\ ps)^{-1}$ [1]
- Emission fluctuations when pumped with 5-10 ns UV pulses [1]
- Emission reproducible with ≤800 ps UV pulses [1]

Motivation: Observed Instabilities

- Pumped with 5 ns 355 nm Nd:YAG laser (10 Hz Q-switched)
- Polydisperse 240 nm ZnO with 30 wt.% MgO (R = 5 nm)







Possible Sources of Instability

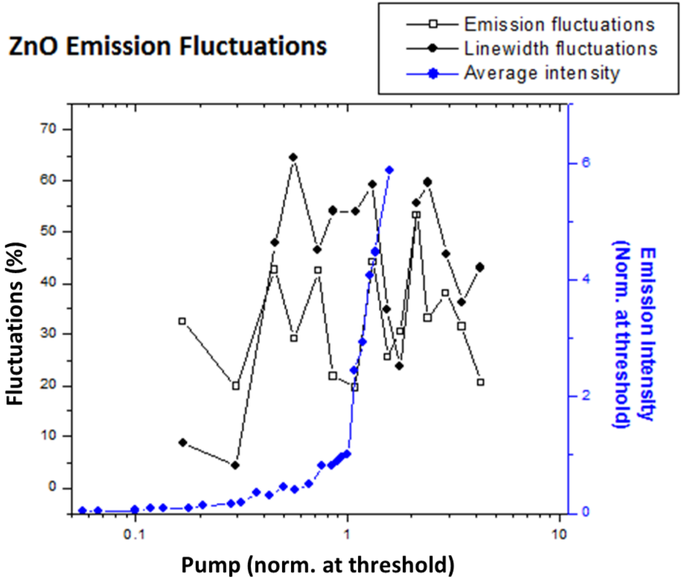
- 1) Fluctuations in pump power near threshold [2]

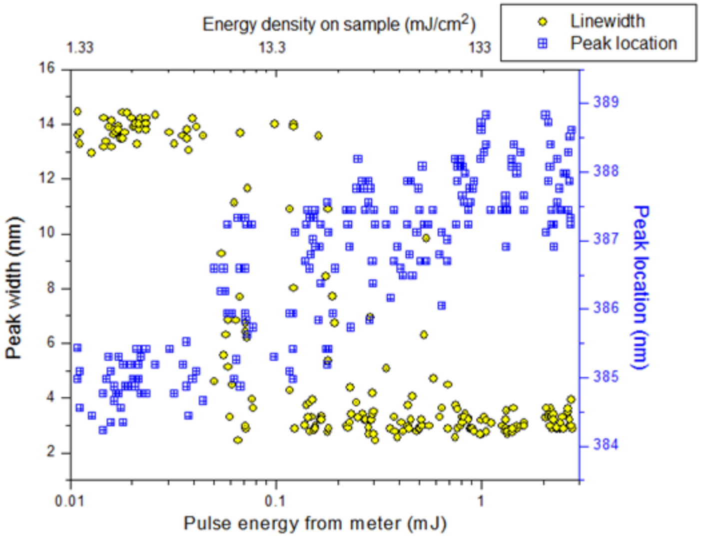
- 2) Slight changes in the microstructure during pumping [3]

- 3) Coupling between emission and population inversion fluctuations [1]

Avg. Pump Power Fluctuations?

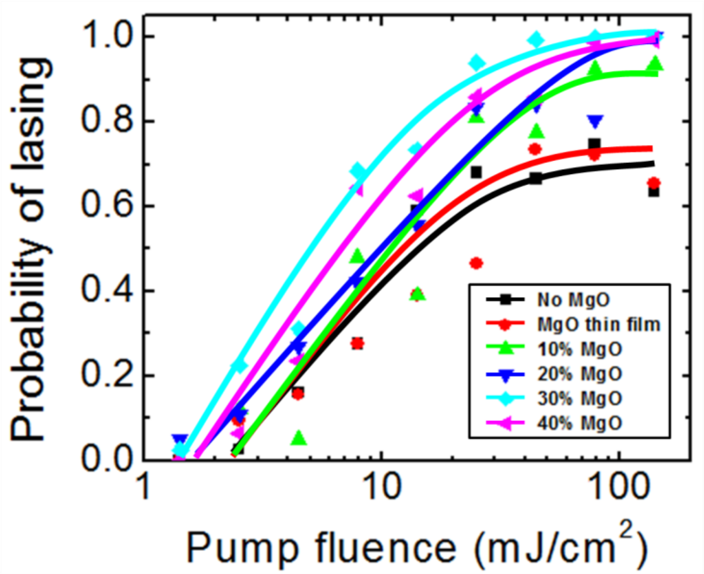
Instability observed far above and below threshold [4]

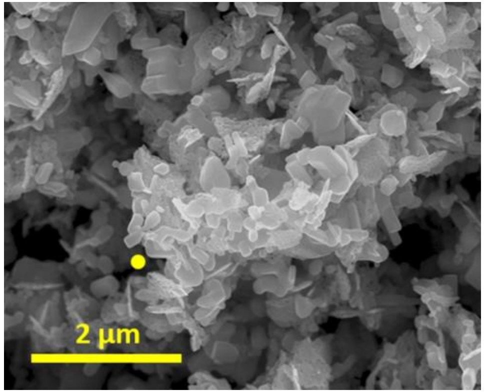


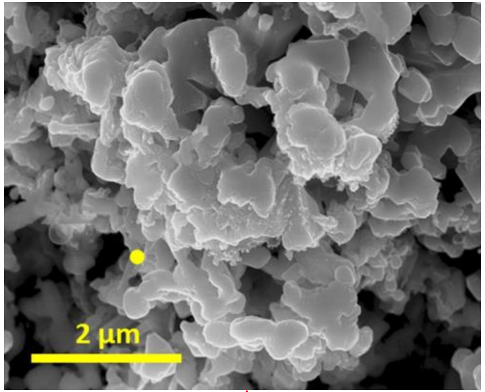


Many modes lasing randomly, **Eliminates #1**

Accumulated Damage?



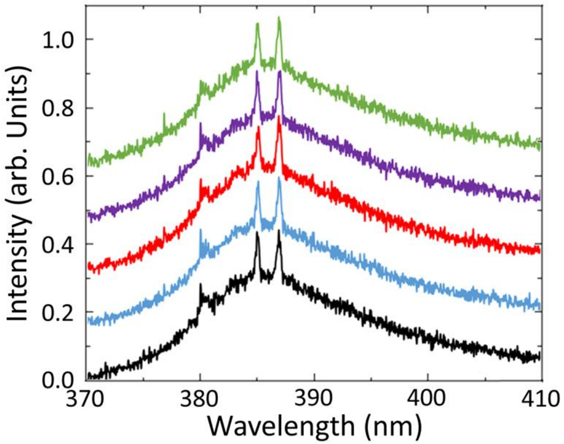


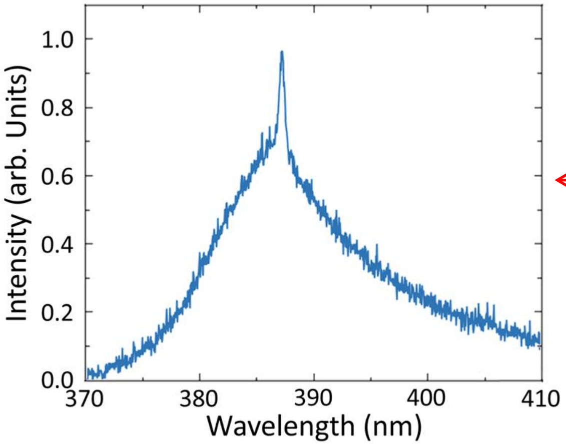


Damage accumulates over ~7500 pulses, reproducible emission at 800 ps or faster [6]

Eliminates #2

Emission Control





ZnO/MgO with PS spheres can exhibit single mode emission [*]

- Pumped with 800 ps 337 nm N_2 laser, Threshold ~7 mJ/cm^2
- Polydisperse 240 nm ZnO with 860 nm PS spheres (1-5 mm^{-2})

*Peterson, Z., Word, R. C., Könenkamp, R. Coherence Enhancement in ZnO Random Lasers with Spherical Defects. *In preparation*.

Emission Control

- Annealing → Up to 3x threshold reduction
- Adding wide bandgap passive scatterers (MgO, BeO, etc.) → Up to 3x threshold reduction
- Shorter pulse width → Reproducibility
- High-quality nanostructures → Mode selection, threshold reduction

Self-consistent Time-dependent Laser Theory

- Begin with Maxwell-Bloch equations for N-level medium (1 radiative transition) in Gaussian units: [7][*]

$$\left(\frac{1}{\varepsilon(\vec{\boldsymbol{r}})}\nabla^2 - \frac{\partial^2}{\partial t^2}\right)\vec{E} = \frac{4\pi}{\varepsilon(\vec{\boldsymbol{r}})}\frac{\partial^2 \vec{P}}{\partial t^2}$$

$$\frac{\partial \vec{P}}{\partial t} = -\left(ik_a + \gamma_p\right)\vec{P} + \frac{g^2}{i\hbar}\vec{E}D$$

$$\frac{\partial D}{\partial t} = \gamma_a\left(D_0(\vec{\boldsymbol{r}}, t) - D\right) - \frac{2}{i\hbar}\left(\vec{E}\cdot\vec{P}^* - \vec{E}^*\cdot\vec{P}\right)$$

- Define a series solution ansatz for E and P:

$$E = \sum_{\mu} \Phi_\mu(\vec{\boldsymbol{r}}, t)\, e^{-ik_\mu t} \qquad P = \sum_{\mu} P_\mu(\vec{\boldsymbol{r}}, t)\, e^{-ik_\mu t}$$

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Dynamic Treatment

- Under the rotating wave approximation, $\dot{D}$ reduces to:

$$\frac{\partial D}{\partial t} = \gamma_a D_0(\vec{\boldsymbol{r}}, t) - \gamma_a D\left(1 + \sum_{\mu} \Gamma_\mu \left|\Phi_\mu\right|^2\right)$$

- **Inversion is not stationary!** Contrast this with SALT. [7]
- Normalized wave equation for mode μ:

$$\nabla^2\Phi_\mu + \varepsilon(\vec{\boldsymbol{r}})\left(2ik_\mu \frac{\partial \Phi_\mu}{\partial t} + k_\mu^2 \Phi_\mu\right) = \frac{i\gamma_p}{\left(\gamma_p + i\left(k_a - k_\mu\right)\right)}\left(2ik_\mu \frac{\partial}{\partial t}\left(D\Phi_\mu\right) + k_\mu^2 D\Phi_\mu\right)$$

Dynamic Treatment

- Split into real and imaginary parts, multiply by Φ_μ^*, add complex conjugate of the result:

$$\frac{\partial}{\partial t}\left|\Phi_\mu\right|^2 = \frac{k_\mu}{2\varepsilon_R}\left|\Phi_\mu\right|^2\left(\Gamma_\mu D - \varepsilon_I\right)$$

- Taking $F(\vec{\boldsymbol{r}},t) = \sum_\mu \Gamma_\mu \left|\Phi_\mu(\vec{\boldsymbol{r}},t)\right|^2 = \sum_\mu F_\mu$ yields:

$$\frac{\partial D}{\partial t} = \gamma_a D_0(\vec{\boldsymbol{r}},t) - \gamma_a D(1 + F(\vec{\boldsymbol{r}},t))$$

$$\frac{\partial F_\mu}{\partial t} = \frac{k_\mu}{2\varepsilon_R} F_\mu\left(\Gamma_\mu D - \varepsilon_I\right)$$

- Family of 1st order coupled nonlinear differential equations with $\vec{\boldsymbol{r}}$ as a parameter

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Stability and Transient Analysis

- Define fluctuations $D \rightarrow D + \Delta D$ and $F_\mu \rightarrow F_\mu + \Delta F_\mu$: [9]

$$\frac{\partial \Delta D}{\partial t} = -\gamma_a \Delta D \left(1 + \sum F_\mu\right) - \gamma_a D \sum \Delta F_\mu \qquad \frac{\partial}{\partial t}\Delta F_\mu = \frac{k_\mu}{2\varepsilon_R}\left(\Delta F_\mu(\Gamma_\mu D - \varepsilon_I) + \Delta D \Gamma_\mu F_\mu\right)$$

- If we have a high-Q nanostructure in single mode case:

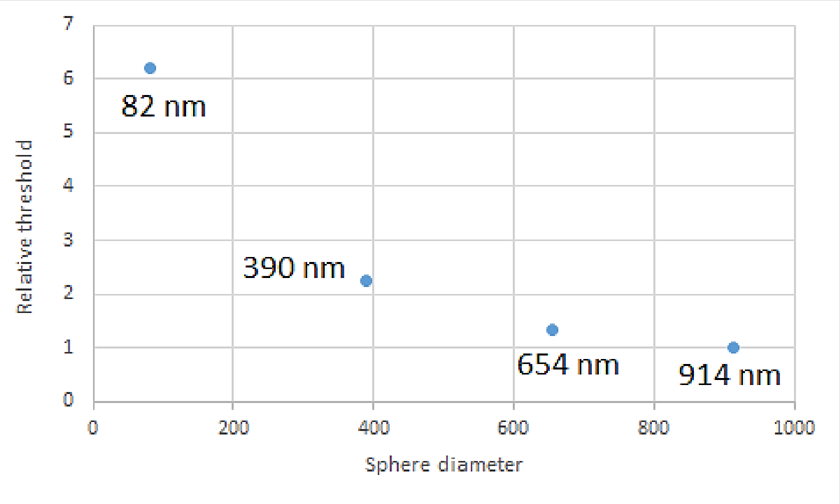
$$\frac{\partial}{\partial t}\begin{bmatrix}\Delta F_\mu \\ \Delta D\end{bmatrix} = \begin{bmatrix} k_\mu (2\varepsilon_R)^{-1}(\Gamma_\mu D - \varepsilon_I) & k_\mu (2\varepsilon_R)^{-1}\Gamma_\mu F_\mu \\ -\gamma_a D & -\gamma_a(1 + F_\mu) \end{bmatrix}\begin{bmatrix}\Delta F_\mu \\ \Delta D\end{bmatrix}$$

- Stability for pulsed (around an arbitrary t_0) and steady pumps (around critical points).

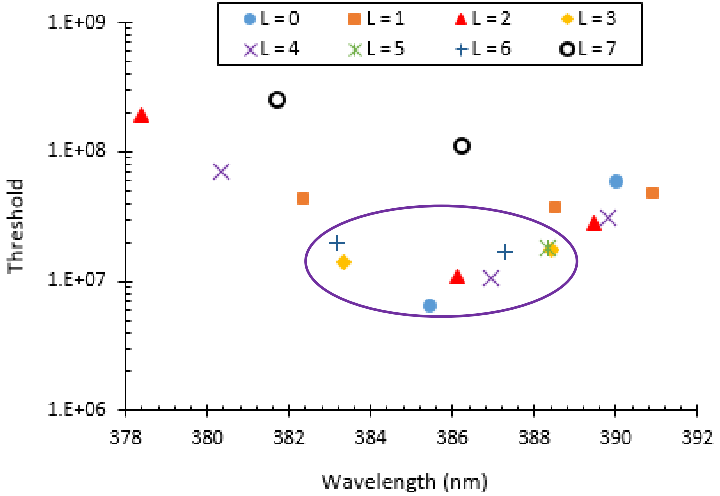
Stability and Transient Analysis w/ Steady Pump

- Relaxation oscillations: $\Delta\ddot{D} + \gamma_a \frac{\Gamma_\mu D_0(\vec{\boldsymbol{r}})}{\varepsilon_I}\Delta\dot{D} + k_\mu(2\varepsilon_R)^{-1}\gamma_a\left(\Gamma_\mu D_0(\vec{\boldsymbol{r}}) - \varepsilon_I\right)\Delta D = 0$

- Steady state: $F_{\mu,c} = \frac{\Gamma_\mu D_0}{\varepsilon_I} - 1 \;\forall\, \mu,\; D_c = \frac{\varepsilon_I}{\Gamma_\mu}$



860 nm spheres, Hankel eigenmodes



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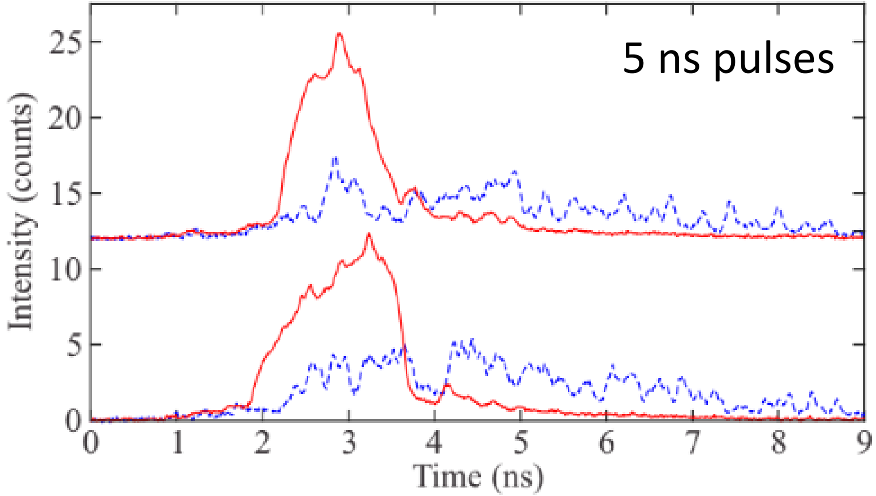
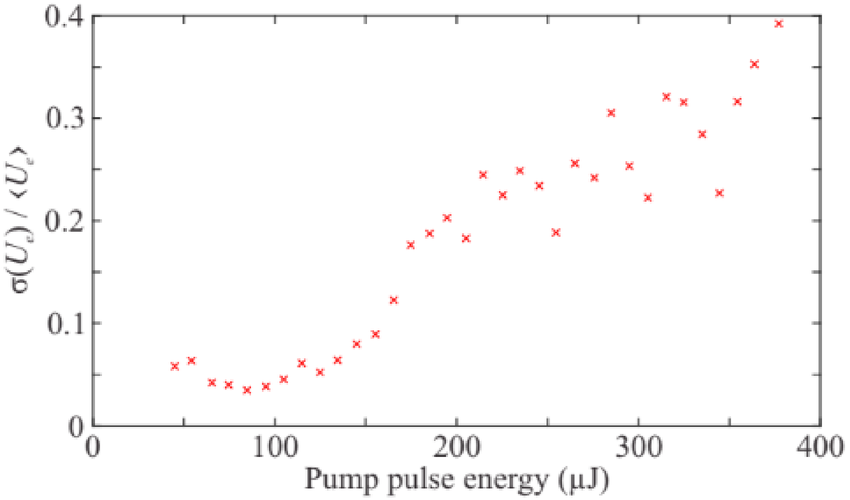
Further research

- Threshold reduction on steady pumping?
- Investigating stochastic behavior
  - Temporally modulated pump → unpredictable large fluctuations*





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*Bittner, Stefan, Sebastian Knitter, Seng Fatt Liew, and Hui Cao. "Random-laser dynamics with temporally modulated pump." *Physical Review A* 99, no. 1 (2019): 013812.

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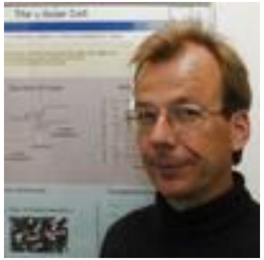

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