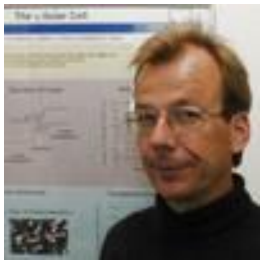
Unstable Emission From Random Lasers

Zachariah Peterson
2019 IEEE Photonics Society Conference
8 July 2019

Acknowledgements



- Rolf Könenkamp
  - Professor, Portland State University

- Robert C. Word
  - Associate Professor, Portland State University

Research supported by Department of Energy
Basic Science Grant #DE-FG02-07ER46406

Outline

- Instabilities in ZnO random lasers
- Reproducibility with different pump pulse times
- Self-consistent time-dependent laser theory
- Stability conditions

Instabilities in ZnO Random Lasers

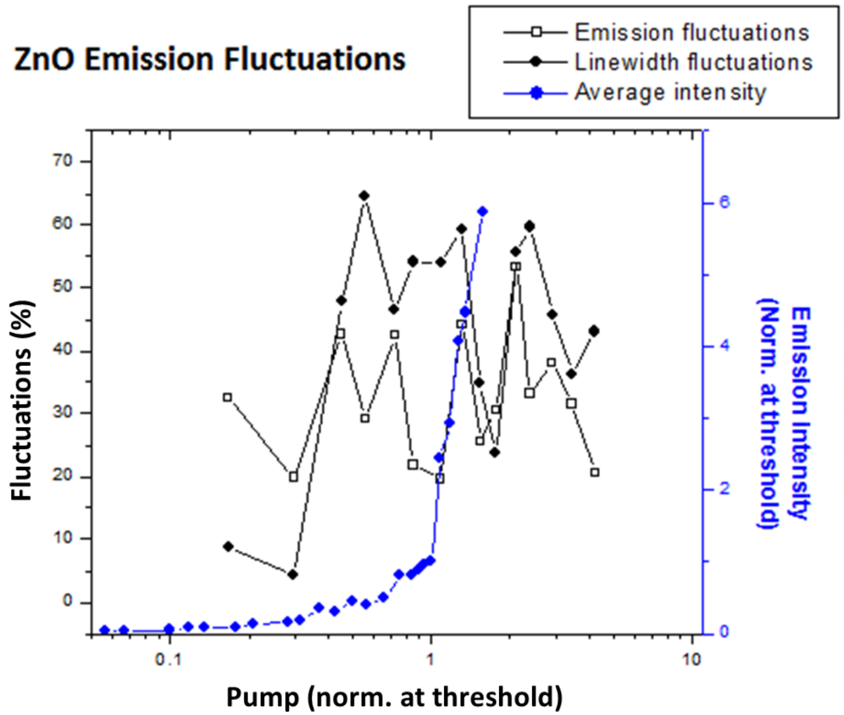
- ZnO: 3.37 eV bandgap, 60 meV exciton binding energy
- Carrier recombination rate $\gamma_a \sim (200\,ps)^{-1}$ [1]
- Emission fluctuations when pumped with 5-10 ns UV pulses [1]
- Emission reproducible with ≤800 ps UV pulses [1]

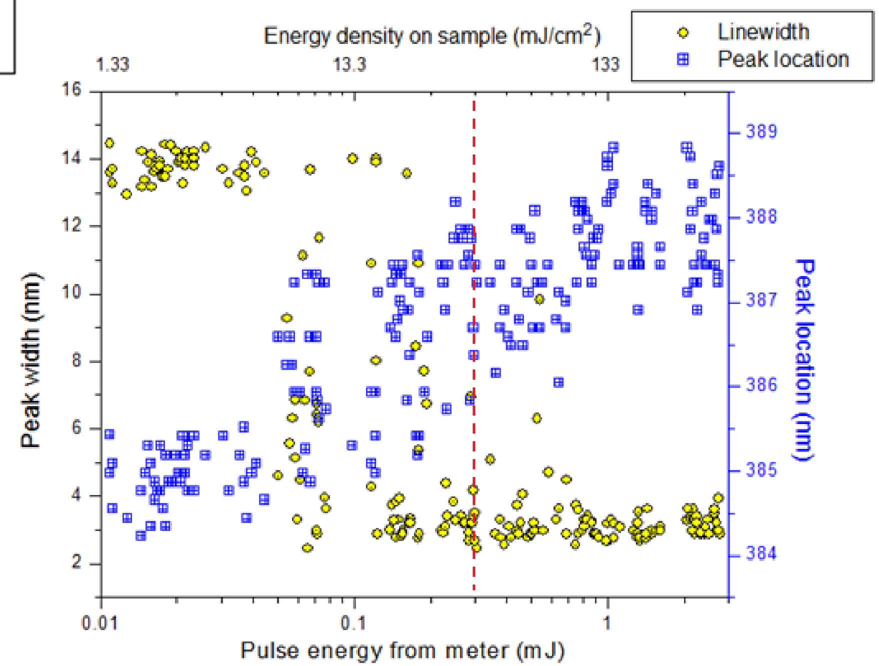
Instabilities in ZnO Random Lasers

- Supposed 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]

Pump power fluctuations

- Instability observed far above and below threshold [4]

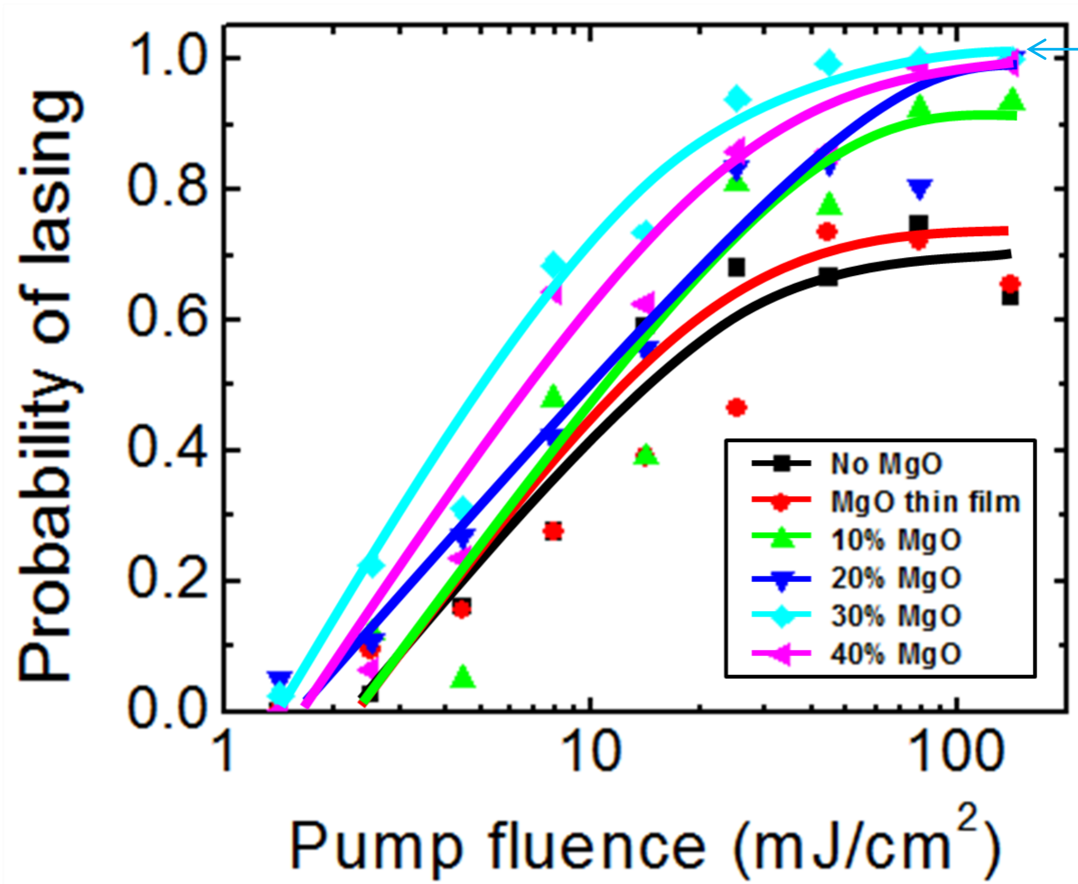




- Many modes lasing randonly
- Eliminates possibility #1

Instability: ZnO/MgO samples

- Statistics from 200 single pumping shots (5 ns pulses) [5]

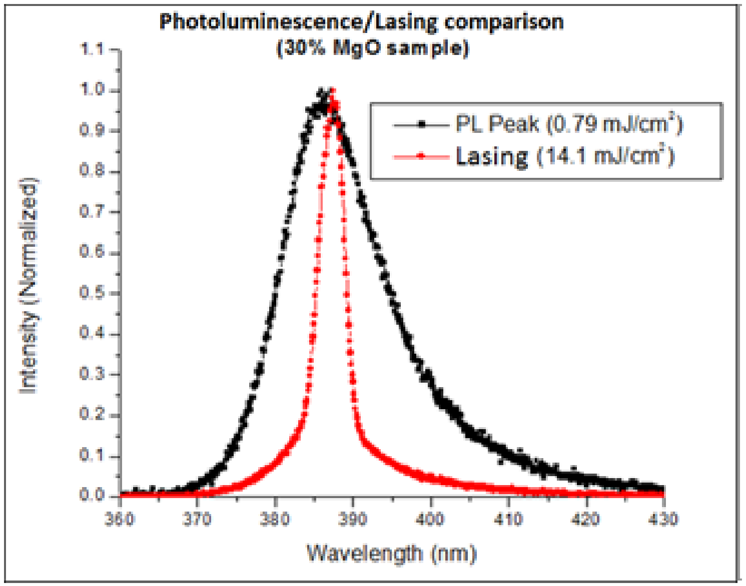


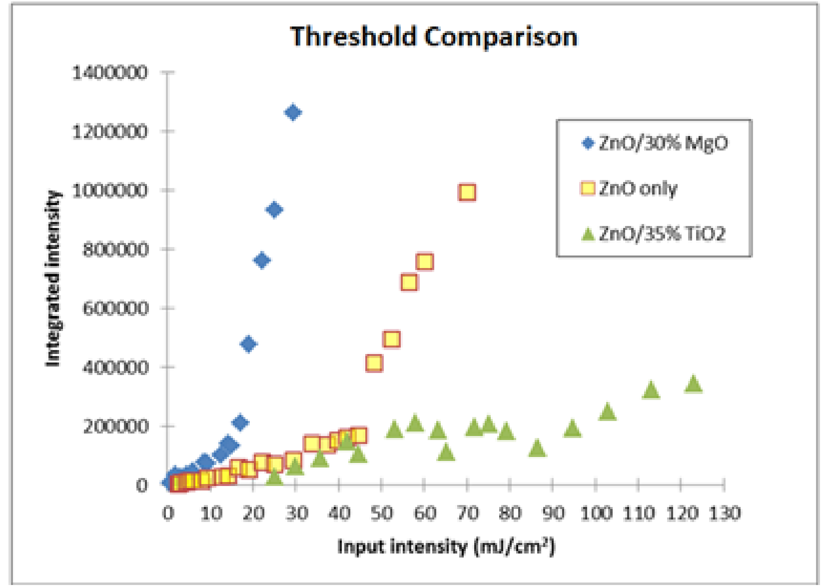
30 wt.% MgO sample has 100% probability for lasing.

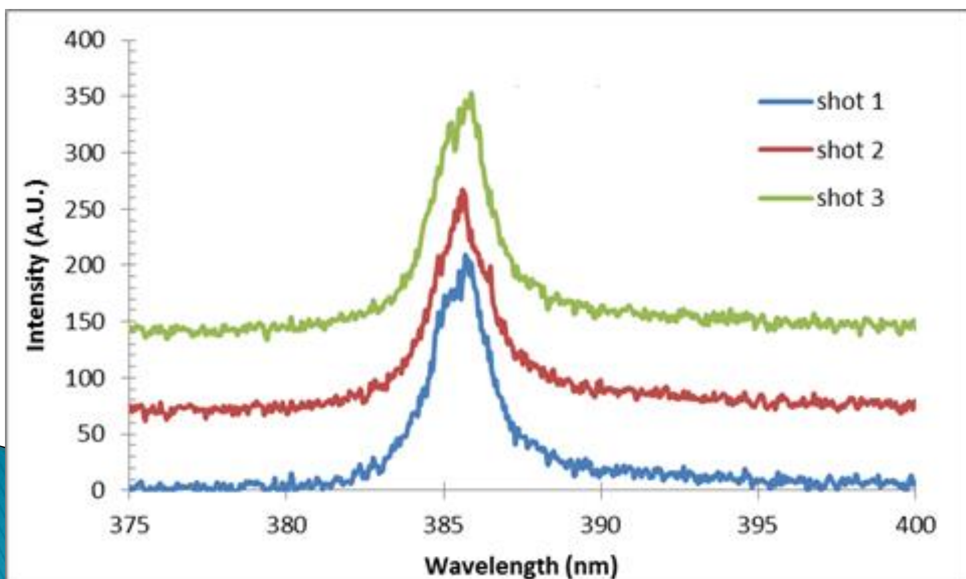
Intensity is still irreproducible!

~70% fluctuations in emission intensity

Reproducibility With 800 ps Pulses



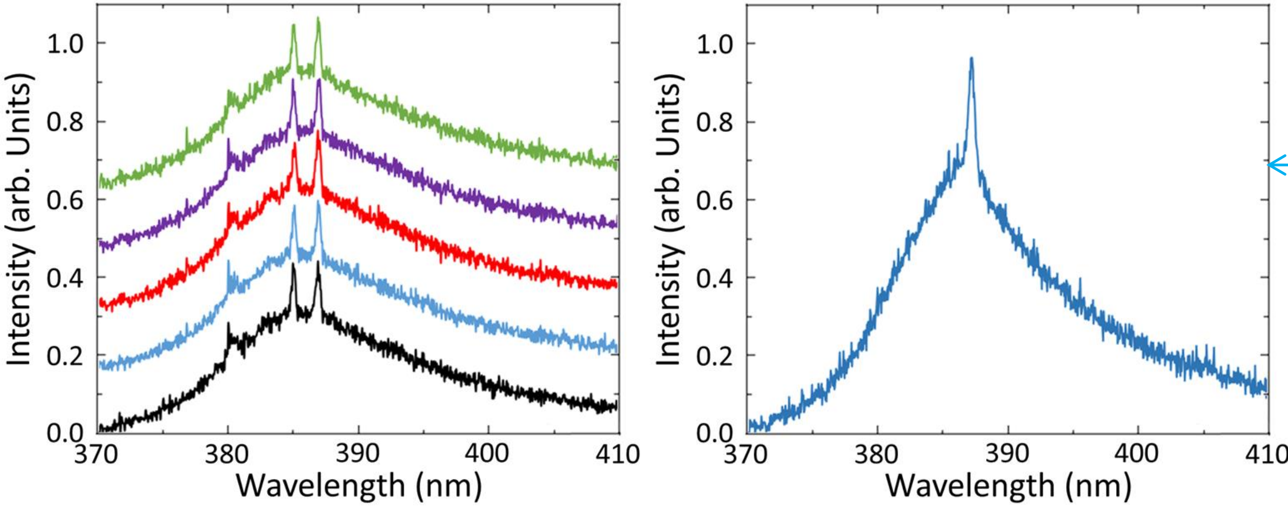




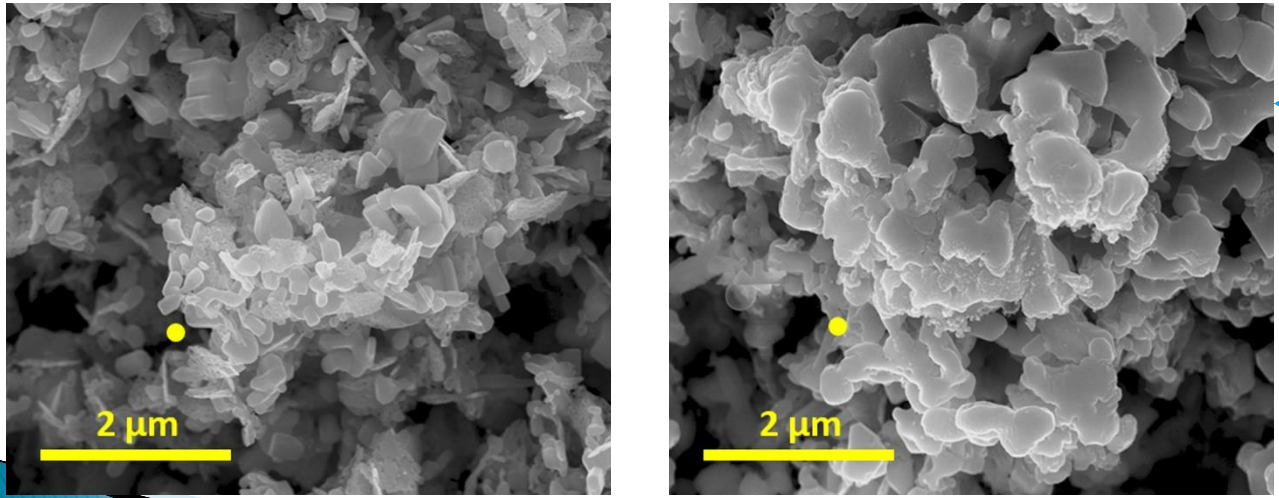
ZnO/30% MgO, 9 mJ/cm^2, 800 ps pulses, reproducible emission [6]

Similar # of modes

Reproducibility With 800 ps Pulses



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



Damage accumulates over ~7500 pulses, reproducible emission [6]

Eliminates #2

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

Unanswered Questions

- *How does the pulse time affect the transition from stability to instability?*
  - Clearly related to the pump pulse time [1]

- Requires a self-consistent time-dependent theory for random lasers

- Steady-state Ab-initio Laser Theory (SALT) [7] has some drawbacks:
  - Only considers steady pump sources (DC voltage, CW lasers, etc.)

  - Cannot describe transient behavior (relaxation oscillations, transition to steady state) or predict instability

Self-consistent Time-dependent Laser Theory

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

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

$$\frac{\partial \vec{P}}{\partial t} = -\left(ik_a + \gamma_p\right)\vec{P} + \frac{g^2}{i\hbar}\vec{E}D \quad (1b)$$

$$\frac{\partial D}{\partial t} = \gamma_a\left(D_0(\vec{\boldsymbol{r}}, t) - D\right) - \frac{2}{i\hbar}\left(\vec{E}\cdot\vec{P}^* - \vec{E}^*\cdot\vec{P}\right) \quad (1c)$$

- Define a series solution ansatz for E and P:

$$E = \sum_{\mu} \Phi_{\mu}(\vec{\boldsymbol{r}}, t)\, e^{-ik_{\mu}t} \quad (2a)$$

$$P = \sum_{\mu} P_{\mu}(\vec{\boldsymbol{r}}, t)\, e^{-ik_{\mu}t} \quad (2b)$$

*Full treatment found in [8]

Self-consistent Time-dependent Laser Theory

- Define two normalization scales for E and D:

$$E_c = \frac{\hbar\sqrt{\gamma_p \gamma_a}}{2g} \qquad\qquad D_c = \frac{\hbar\gamma_p}{4\pi g^2}$$

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

$$\frac{\partial D}{\partial t} = \gamma_a D_0(\vec{\boldsymbol{r}}, t) - \gamma_a D\left(1 + \sum_{\mu} \Gamma_\mu \left|\Phi_\mu\right|^2\right) \tag{3}$$

- **Inversion is not stationary!** Contrast this with SALT. [7]

- Normalized wave equation for mode μ:

$$\nabla^2\Phi_\mu + \varepsilon(\vec{\boldsymbol{r}})\left(2ik_\mu \frac{\partial \Phi_\mu}{\partial t} + k_\mu^2 \Phi_\mu\right) = \frac{i\gamma_p}{\left(\gamma_p + i\left(k_a - k_\mu\right)\right)}\left(2ik_\mu \frac{\partial}{\partial t}\left(D\Phi_\mu\right) + k_\mu^2 D\Phi_\mu\right) \tag{4}$$

Self-consistent Time-dependent Laser Theory

- Define an orthonormal set of basis states for each mode with time-dependent amplitudes:

$$\Phi_\mu(\vec{\boldsymbol{r}},t) = \sum_{\mu} a_m^{\mu}(t)\varphi_m(\vec{\boldsymbol{r}}) \qquad (5a)$$

- where

$$\nabla^2\varphi_m(\vec{\boldsymbol{r}}) + \varepsilon(\vec{\boldsymbol{r}})k_m^2\varphi_m(\vec{\boldsymbol{r}}) = 0 \qquad (\vec{\boldsymbol{r}} \in C) \qquad (5b)$$

$$\nabla^2\varphi_m(\vec{\boldsymbol{r}}) + \varepsilon_0 k_\mu^2\varphi_m(\vec{\boldsymbol{r}}) = 0 \qquad (\vec{\boldsymbol{r}} \notin C) \qquad (5c)$$

$$\int \varepsilon(\vec{\boldsymbol{r}})\varphi_n(\vec{\boldsymbol{r}})\varphi_m^*(\vec{\boldsymbol{r}})\, dV = \delta_{nm} \qquad (\vec{\boldsymbol{r}} \in C) \qquad (5d)$$

subject to: $(\nabla\varphi_m)\cdot\hat{n}\ \Big|_S = ik_\mu\varphi_m\ \Big|_S$ and $\lim_{r\to\infty}\varphi_m \propto \frac{e^{ik_\mu r}}{r^{\frac{d-1}{2}}}$

- Eqs. (3)–(5) fit within standard SALT treatment [7]
- Allows time-varying pumps $D_0(\vec{\boldsymbol{r}},t')$ to be considered
- Allows transient behavior to be considered

Self-consistent Time-dependent Laser Theory

- Solution for the population inversion: [8]

$$D(\vec{\boldsymbol{r}},t) = \gamma_a e^{-\gamma_a \int_0^t (1+F(\vec{\boldsymbol{r}},t'))dt'} \int_0^t D_0(\vec{\boldsymbol{r}},t')\, e^{\gamma_a \int_0^{t'} (1+F(\vec{\boldsymbol{r}},s))ds} dt' \qquad (5)$$

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

- Define $\langle g(\vec{\boldsymbol{r}})\rangle_{nm} = \int g(\vec{\boldsymbol{r}})\varphi_n^* \varphi_m dV$

- Solution for the basis state amplitudes: [8]

$$\frac{d}{dt}[a] = \frac{k_\mu}{2}[\varepsilon]^{-1}[M][a] \qquad (6a)$$

- where $M_{nm} = \Gamma_\mu \langle D\rangle_{nm} - \langle \varepsilon_I(\vec{\boldsymbol{r}})\rangle_{nm}$, $[a] = \left[a_1^\mu(t) \ldots a_m^\mu(t)\right]^T$ (6b), (6c)

Stability in SCTDLT

- Split (4) into real and imaginary parts, multiply by Φ_μ^*, and add complex conjugate of the result:

$$\frac{\partial}{\partial t}\left|\Phi_\mu\right|^2 = \frac{k_\mu}{2\varepsilon_R}\left|\Phi_\mu\right|^2\left(\Gamma_\mu D - \varepsilon_I\right) \tag{7}$$

- Taking $F(\vec{\boldsymbol{r}},t) = \sum_\mu \Gamma_\mu \left|\Phi_\mu(\vec{\boldsymbol{r}},t)\right|^2 = \sum_\mu F_\mu$ in (3) yields:

$$\frac{\partial D}{\partial t} = \gamma_a D_0(\vec{\boldsymbol{r}},t) - \gamma_a D\left(1 + F(\vec{\boldsymbol{r}},t)\right) \tag{8a}$$

$$\frac{\partial F_\mu}{\partial t} = \frac{k_\mu}{2\varepsilon_R} F_\mu\left(\Gamma_\mu D - \varepsilon_I\right) \tag{8b}$$

- Family of 1[st] order coupled nonlinear differential equations with $\vec{\boldsymbol{r}}$ as a parameter → stability analysis for fluctuations.

Stability in SCTDLT

- Define fluctuations $D \rightarrow D + \Delta D$ and $F_\mu \rightarrow F_\mu + \Delta F_\mu$ in (8a) and (8b): [9]

$$\frac{\partial \Delta D}{\partial t} = -\gamma_a \Delta D \left(1 + \sum F_\mu\right) - \gamma_a D \sum \Delta F_\mu \tag{9a}$$

$$\frac{\partial \Delta F_\mu}{\partial t} = \frac{k_\mu}{2\varepsilon_R}\left(\Delta F_\mu (\Gamma_\mu D - \varepsilon_I) + \Delta D \Gamma_\mu F_\mu\right) \tag{9b}$$

- For the moment, assume 1 mode. Write (10) as a matrix:

$$\frac{\partial}{\partial t}\begin{bmatrix}\Delta F_\mu \\ \Delta D\end{bmatrix} = \begin{bmatrix} k_\mu (2\varepsilon_R)^{-1}\left(\Gamma_\mu D - \varepsilon_I\right) & k_\mu (2\varepsilon_R)^{-1}\Gamma_\mu F_\mu \\ -\gamma_a D & -\gamma_a\left(1 + F_\mu\right) \end{bmatrix}\begin{bmatrix}\Delta F_\mu \\ \Delta D\end{bmatrix} \tag{10}$$

- Can treat stability with pulsed (around some arbitrary time t_0) and steady pumps (around critical points).

Stability in SCTDLT: Bifurcation with Steady Pump

- Critical points in (10):

$$F_{\mu,c} = 0 \; \forall \, \mu, \, D_c = D_0 \qquad (11a)$$

$$F_{\mu,c} = \frac{\Gamma_\mu D_0}{\varepsilon_I} - 1 \; \forall \, \mu, \, D_c = \frac{\varepsilon_I}{\Gamma_\mu} \qquad (11b)$$

- Stability determined using Poincaré–Bendixson theorem or the eigenvalues of (10). For (11a):

$$\det \begin{bmatrix} k_\mu (2\varepsilon_R)^{-1} \left(\Gamma_\mu D_0 - \varepsilon_I \right) - \lambda & 0 \\ -\gamma_a D_0 & -\gamma_a - \lambda \end{bmatrix} = 0$$

- Transition to instability:
  - Stable growth for $\varepsilon_I < \Gamma_\mu D_0 < 2\varepsilon_R \gamma_a k_\mu^{-1} + \varepsilon_I$
  - Hopf bifurcation at $\Gamma_\mu D_0 = 2\varepsilon_R \gamma_a k_\mu^{-1} + \varepsilon_I$
  - Unstable growth for $\Gamma_\mu D_0 - \varepsilon_I > 2\varepsilon_R \gamma_a k_\mu^{-1}$

Stability in SCTDLT: Relaxation oscillations

- For critical points in (12b) (non-zero photon density):

$$\frac{\partial}{\partial t}\begin{bmatrix}\Delta F_\mu \\ \Delta D\end{bmatrix} = \begin{bmatrix} 0 & k_\mu (2\varepsilon_R)^{-1}\Gamma_\mu \left(\frac{\Gamma_\mu D_0}{\varepsilon_I} - 1\right) \\ -\gamma_a \frac{\varepsilon_I}{\Gamma_\mu} & -\gamma_a \frac{\Gamma_\mu D_0}{\varepsilon_I} \end{bmatrix}\begin{bmatrix}\Delta F_\mu \\ \Delta D\end{bmatrix} \qquad (12)$$

- Equation of motion for ΔD:

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

- Underdamped: $\Gamma_\mu D_0(1 + 2c\varepsilon_I) - c\Gamma_\mu^2 D_0^2 < c\varepsilon_I^2$, where $c = \frac{k_\mu^2 \gamma_a \varepsilon_I}{2\varepsilon_R^2}$

$$P_\mu = \frac{k_\mu}{2\pi}\int \left(\Gamma_\mu[D(\vec{\boldsymbol{r}},t) + \Delta D(\vec{\boldsymbol{r}},t)] - \varepsilon_I(\vec{\boldsymbol{r}})\right)\left|\Phi_\mu + \Delta\Phi_\mu\right|^2 dS - \frac{1}{2\pi}\frac{\partial}{\partial t}\int \varepsilon_R(\vec{\boldsymbol{r}})\left|\Phi_\mu + \Delta\Phi_\mu\right|^2 dS$$

where $\Gamma_\mu(\vec{\boldsymbol{r}}), D(\vec{\boldsymbol{r}},t) \in \boldsymbol{G}$ and $\Phi_\mu(\vec{\boldsymbol{r}},t), \varepsilon_I(\vec{\boldsymbol{r}}) \in \boldsymbol{C}$

Stability in SCTDLT: Time-dependent Pumping

- Stable limit cycles for the system in (12) do not exist:

- For time-dependent pumping, we have a non-autonomous system with no equilibria.

- Example: Define $z = \frac{t}{\tau}$, where τ is the pulse time. $D_0 \approx \frac{At}{\tau} = Az$.

- Equivalent autonomous system:

$$\frac{\partial}{\partial t}\begin{bmatrix}\Delta F_\mu \\ \Delta D \\ \Delta z\end{bmatrix} = \begin{bmatrix} k_\mu(2\varepsilon_R)^{-1}(\Gamma_\mu D - \varepsilon_I) & k_\mu(2\varepsilon_R)^{-1}\Gamma_\mu F_\mu & 0 \\ -\gamma_a D & -\gamma_a(1+F_\mu) & \gamma_a A \\ 0 & 0 & 0 \end{bmatrix}\begin{bmatrix}\Delta F_\mu \\ \Delta D \\ \Delta z\end{bmatrix} \quad (13)$$

Stability in SCTDLT: Time-dependent Pumping

- Solve (13) as a frozen coefficient system.
- For $D_0 \sim A(x) f(t)$, where $f(t)$ is linear, transition to instability unrelated to pump time for a single mode.
- N-mode non-autonomous system, let $z = \frac{t}{\tau}$:

$$\frac{\partial}{\partial t}\begin{bmatrix}\Delta F_1 \\ \vdots \\ \Delta F_N \\ \Delta D \\ \Delta z\end{bmatrix} = \begin{bmatrix} k_1(2\varepsilon_R)^{-1}(\Gamma_1 D - \varepsilon_I) & 0 & \cdots & 0 & k_1(2\varepsilon_R)^{-1}\Gamma_1 F_1 & 0 \\ \vdots & & & & \vdots & \vdots \\ k_N(2\varepsilon_R)^{-1}(\Gamma_N D - \varepsilon_I) & 0 & \cdots & 0 & k_N(2\varepsilon_R)^{-1}\Gamma_N F_N & 0 \\ -\gamma_a D & \cdots & -\gamma_a D & -\gamma_a\left(1 + \sum F_\mu\right) & \gamma_a A(x) f'(z) \\ 0 & \cdots & & 0 & & \end{bmatrix}\begin{bmatrix}\Delta F_1 \\ \vdots \\ \Delta F_N \\ \Delta D \\ \Delta z\end{bmatrix} \quad (14)$$

Summary

- Mode reduction may not always suppress instabilities
- Eq. (14): for N lasing modes, N+2 possibly distinct, possibly complex eigenvalues
  - Stability conditions vary in each mode
  - Transition to instability affected by spatiotemporal modal overlap
- Theory predicts relaxation oscillations
  - Calculated for steady pump source
  - Limit cycles do not exist for non-zero photon density
  - Different areas of the random laser exhibit mixed damping

Further Research

- Numerically simulate stable/unstable behavior directly from stochastic processes that govern noise in random lasers

- Directly derive photon statistics (Gaussian → Levy)

- Solution algorithm for instabilities in multi-mode lasing (overlapping modes in space and time, Eq. (14)).

Thank you!

References

[1] V. M. Markushev, et al., "Analysis of ZnO random laser spectra under nano-second pumping," *Proc. SPIE6731*, 67311M (2007).

[2] Zhu, G., et. al. "Experimental study of instability in a random laser with immobile scatterers," *Phys. Rev. A*, **85**, 043801 (2012).

[3] Sun, Y., et. al. "Excitonic gain and stimulated ultraviolet emission in nanocrystalline zinc-oxide powder," *Appl. Phys. Lett.*, **77**(15) (2000).

[4] Peterson, Z., et al. "Morphology stability and threshold enhancement in zinc oxide random lasers," *Microscopy and Microanalysis*, Portland, OR (July 2015).

[5] Peterson, Z., et al., "Zinc oxide random lasers: threshold enhancement and operational stability," *Bulletin of the American Physical Society,* **60**(6) (2015).

[6] Peterson, Z., et al. Structural instability and dynamic emission fluctuations in zinc oxide random lasers. *Journal of Applied Physics*, **124**, 063104 (2018).

[7] Ge, L., Chong, Y. D., and Stone, A. D. Steady-state Ab-initio Laser Theory: Generalizations and Analytic Results. *Phys. Rev. A*, **82**, 063824 (2010).

[8] Peterson, Z. Self-consistent theory of random lasing in the time-domain. *Optica Applicata*, **48**(4) (2018).

[9] N. M. Lawandy, "Relaxation Oscillations and Stability," *IEEE J. Quant. Elec.*, QE-18(12) (December 1982).