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Zinc Oxide Random Laser Threshold Enhancement via Addition of Passive Scatterers

Zachariah M. Peterson
Portland State University, zmp@pdx.edu

Rolf Könenkamp
Portland State University, rkoe@pdx.edu

Robert Campbell Word
Portland State University, wordr@pdx.edu

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Zachariah Peterson[1], Rolf Könenkamp[1], Rob Word[1]
1 Department of Physics, Portland State University

Portland State UNIVERSITY

Abstract
Zinc oxide (ZnO) is a wide bandgap n-type semiconductor with a variety of optical and electrical applications and many methods of fabrication. Strong optical scattering and photoluminescence from ZnO nanoparticles and films makes the material an ideal candidate for a random laser. Previous studies have shown both incoherent and coherent random lasing from ZnO films and particles agglomerations. When used as a passive scatterer in a laser dye gain medium, the addition of ZnO has been shown to improve the threshold for lasing. By combining active scattering ZnO with a passive scatterer, MgO, we show here that the lasing threshold is reduced. We also demonstrate strong optical feedback in laser pumped ZnO nanoparticle films. Photoluminescence (PL) results show a clear amplification threshold and the resulting non-linear behavior. We find that shortening the pump pulse time by a factor 6 causes a feedback mechanism transition from Amplified Spontaneous Emission (ASE) to Non-resonant feedback (NRF). The pulse time is still longer than the excitonic lifetime (~200 ps), however the randomness from spontaneous emission is greatly reduced. NRF in our samples can be characterized by a dramatic narrowing of the photoluminescence peak around 387 nm to FWHM of ~3 nm, as well as a high degree of reproducibility in the emitted spectra. A new statistical model for the generation of random laser modes was formulated and it reproduces the experimental results. Further work will focus on studying the transition from non-resonant to resonant feedback in the nanoparticle films.

Random Lasing: Theory

- Maxwell-Bloch equations describe light transport for an N-level lasing system:

$$\frac{\partial^2\vec{E}(\boldsymbol{x},t)}{\partial t^2} = \frac{1}{\varepsilon(\boldsymbol{x})}\nabla^2\vec{E} - \frac{4\pi}{\varepsilon(\boldsymbol{x})}\frac{\partial^2\vec{P}}{\partial t^2} \quad (1)$$

$$\frac{\partial\vec{P}(\boldsymbol{x},t)}{\partial t} = -(i\omega_a + \gamma_p)\vec{P} + \frac{g^2}{i\hbar}\vec{E}\cdot D(\boldsymbol{x},t) \quad (2)$$

$$\frac{\partial D(\boldsymbol{x},t)}{\partial t} = \gamma_a(\alpha D_0 - D) - \frac{2}{i\hbar}\left(\vec{E}(\vec{P}^*) - \vec{P}(\vec{E}^*)\right) \quad (3)$$

- Self-consistent Ab-initio Laser Theory reformulation of the above equations:

$$-k_\mu^2\Phi_\mu(\boldsymbol{x}) = \frac{1}{\varepsilon(\boldsymbol{x})}\nabla^2\Phi_\mu(\boldsymbol{x}) + \left(\frac{1}{\varepsilon(x)}\right)\left(\frac{\gamma_p}{\gamma_p - i(k_\mu - k_a)}\right)\left(\frac{\alpha D_0(x)}{1+\sum_{j=1}^N \Gamma_j|\Phi_j(x)|^2}\right)k_\mu^2\Phi_\mu(\boldsymbol{x}) \quad (4)$$

$$\Gamma(\omega) = \frac{\gamma_p^2}{\gamma_p^2 + (\omega - \frac{2\pi c}{\lambda_a})^2} \quad (5)$$

$$\boldsymbol{T}^\mu_{mn} = \frac{(k_\mu^2/k_a^2)}{(k_\mu^2 - k_m^2)}\left(\frac{i\gamma_p}{\gamma_p - i(k_\mu - k_a)}\right)\int \frac{D_0(\boldsymbol{x}')\bar{\varphi}_m^{\mu*}(\boldsymbol{x}')\varphi_m^\mu(\boldsymbol{x}')}{\varepsilon(\boldsymbol{x})[1+\sum_{j=1}^N \Gamma_j|\Phi_j(\boldsymbol{x})|^2]}dx' \quad (6)$$

- Possible feedback mechanisms for scattered light in random media:

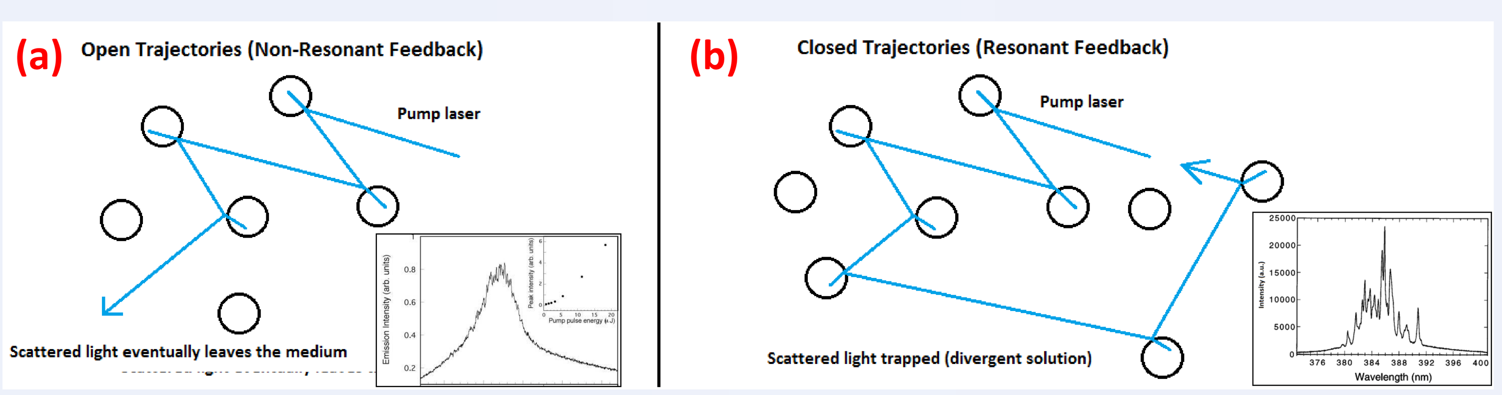

Figure 1. (a) Open scattering trajectories corresponding to amplified spontaneous emission and non-resonant feedback. Inset shows the expected PL spectrum with peak FWHM of ~4 nm. (b) Closed scattering trajectories corresponding to resonant feedback. Inset shows the expected spectrum with multiple thin lasing peaks.

Random Lasing Samples and Optical Apparatus

- ZnO nanoparticles deposited on Si/MgO thin film with MgO nanoparticles added to ZnO (10% increments). Pumped with 5 ns (355 nm) and 800 ps (337 nm) pulses.

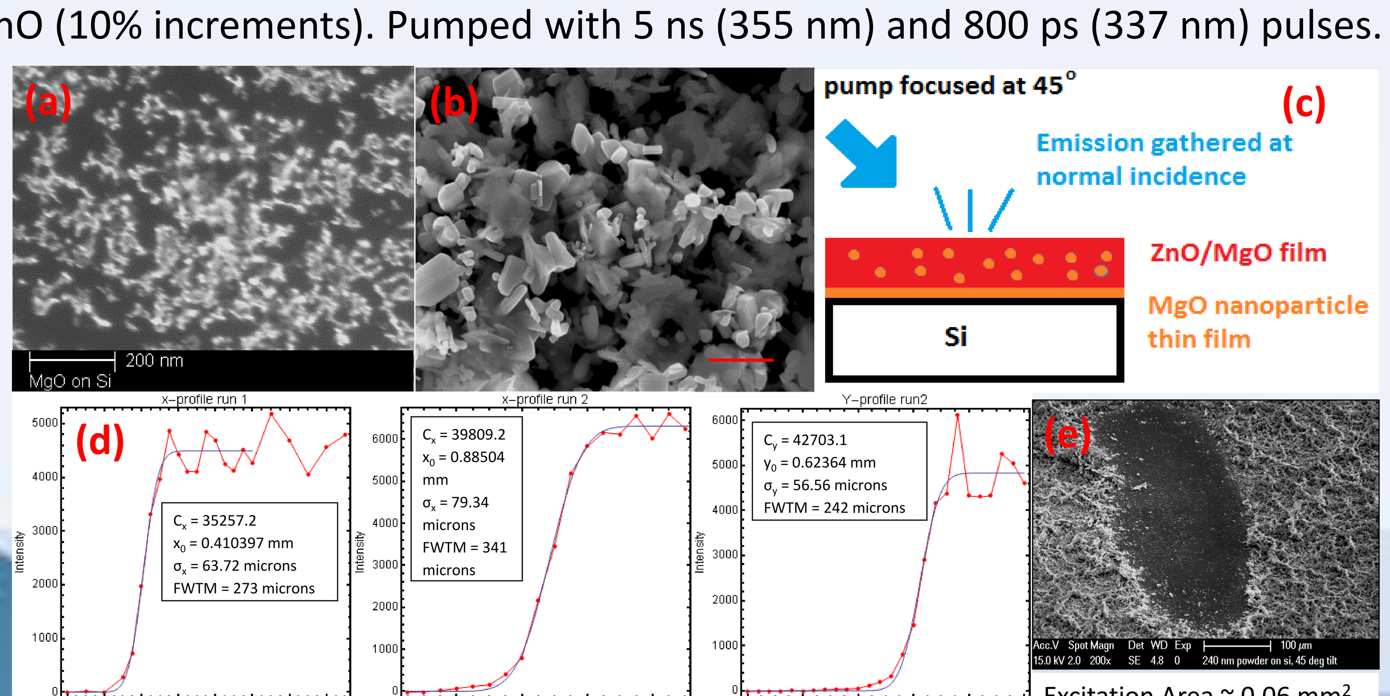

Figure 2. (a) MgO nanoparticles on Si. (b) ZnO nanoparticles on Si. The inset scale bar is 1 micron. (c) Diagram showing the sample and pump light configuration. (d) Data for spot size measurements and (e) SEM of laser ablated sample for comparison.

Optical feedback under nanosecond pumping

- Data for ZnO/MgO samples (single shot at 5 ns). 30% MgO initially seems optimum.

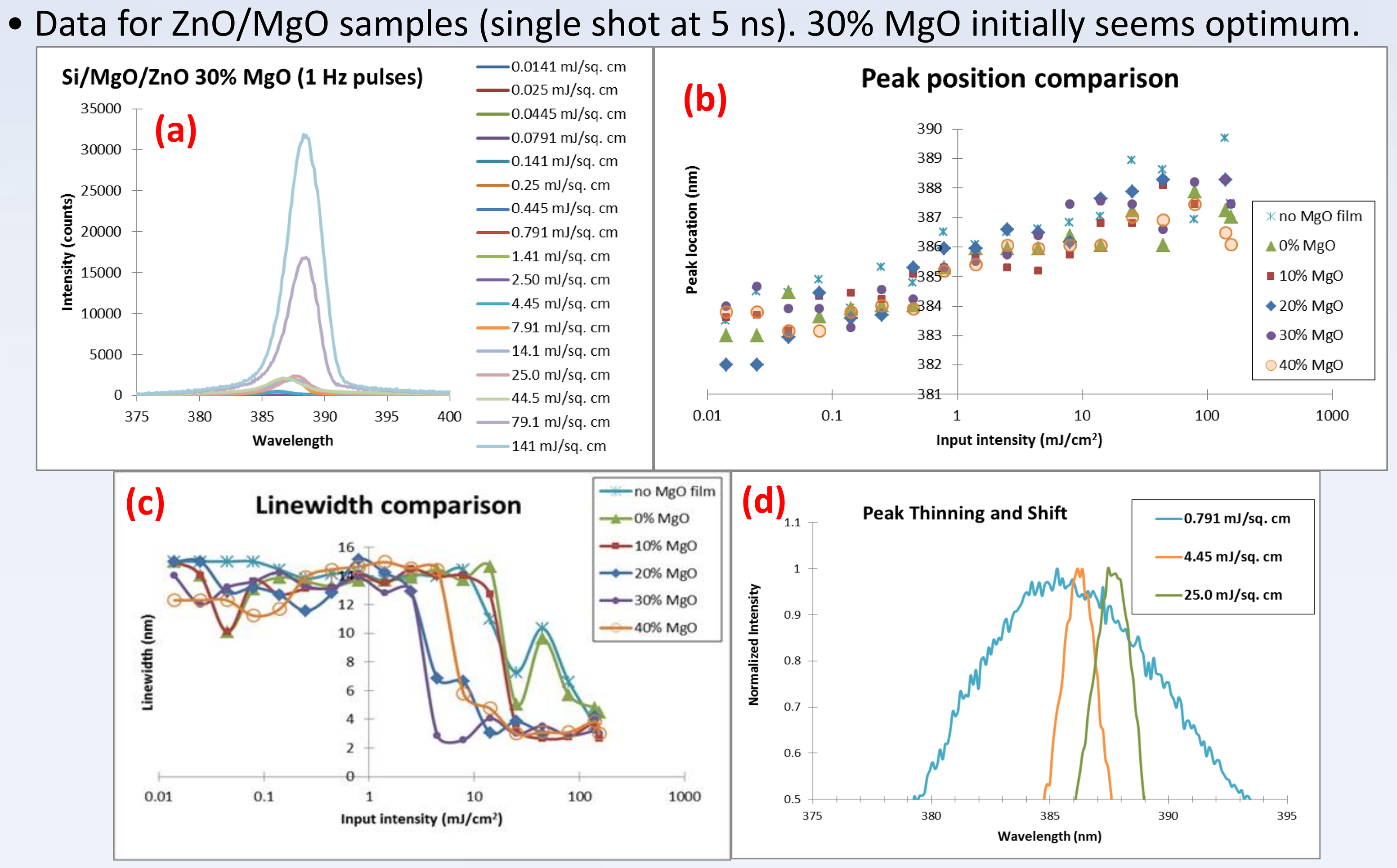

Figure 3. (a) Random laser spectra under single shot pumping. (b) Peak redshift data vs. pumping intensity. (c) Linewidth data from all samples. (d) Normalized spectra comparing photoluminescence spectrum with two lasing spectra above threshold.

- Data showing spectral randomness due to intense ASE.

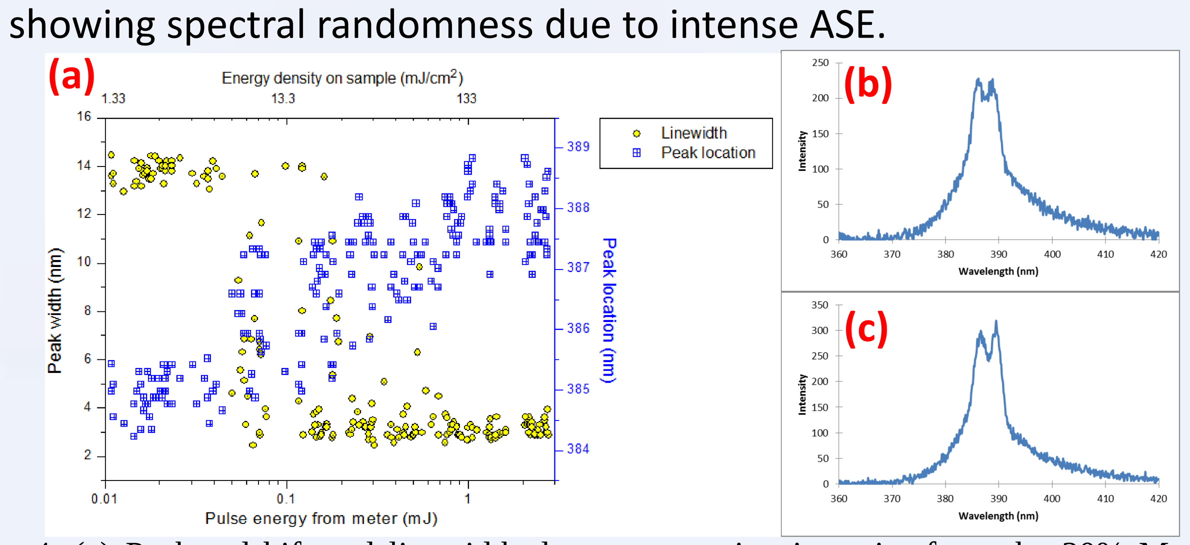

Figure 4. (a) Peak redshift and linewidth data vs. pumping intensity from the 30% MgO sample under single pump pulses. (b) and (c) show spectra with the rare occurrence of more than one visible peak.

- Replacing MgO with equal volume of TiO_2 (35% by weight) shows opposite result.

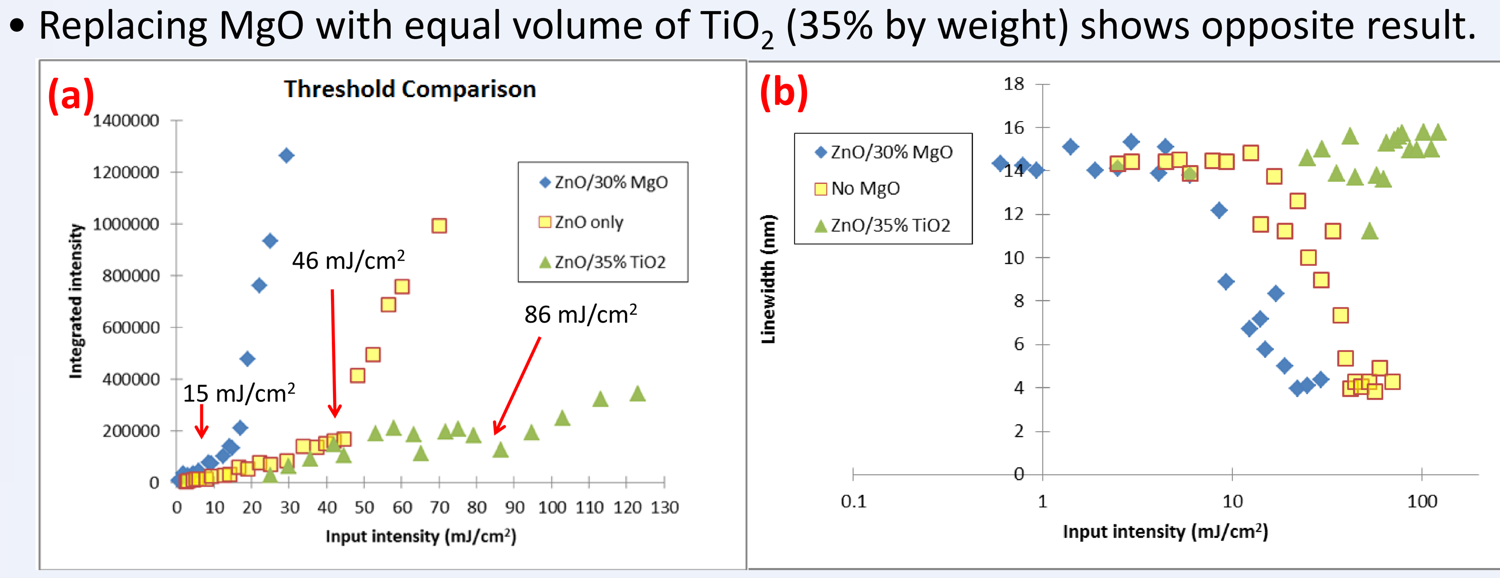

Figure 5. (a) Threshold measurements from averaged spectra (150 pumping shot average). (b) Corresponding linewidth (FWHM) data showing threshold improvement with addition of MgO.

- MgO addition also improves feedback from ASE. NRF may also be present.

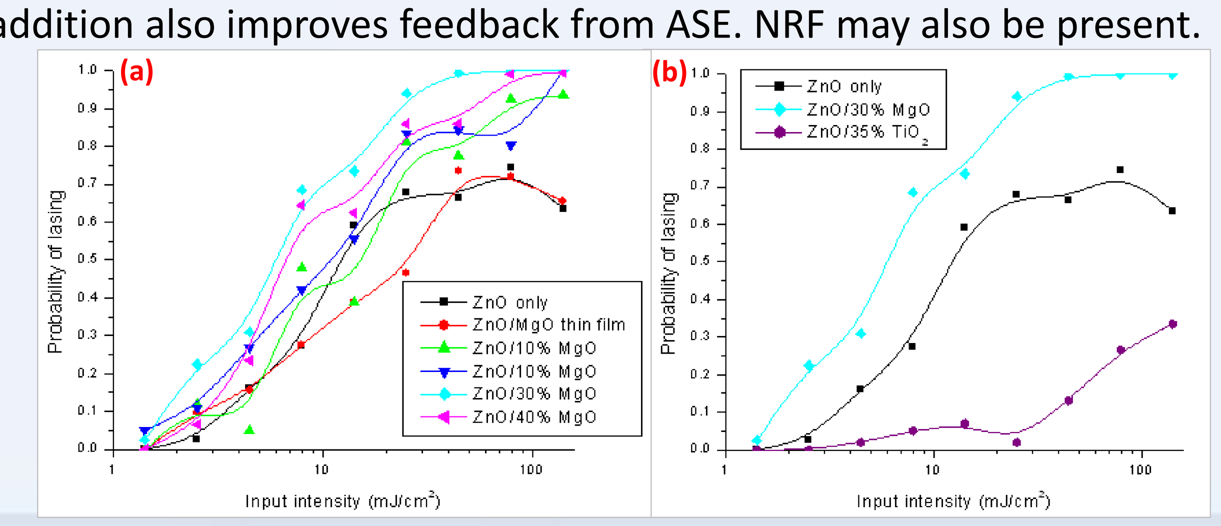

Figure 6. (a) Probability that pumping pulse produces visible optical feedback (200 shots). (b) Results showing comparison between non-absorbing MgO and absorbing TiO_2.

- MgO causes clear improvement in lasing threshold due to decreased absorption coefficient α at the pump (355 nm) and emission (~386 nm) wavelengths.

Material	α (cm^{-1})
ZnO	~5*10^3
MgO	0
TiO_2	~2.5*10^4

Non-resonant feedback under picosecond pumping

- Emitted spectra become reproducible when pumped with picosecond lasing. Amplified spontaneous emission is greatly reduced. Data for single shots (800 ps):

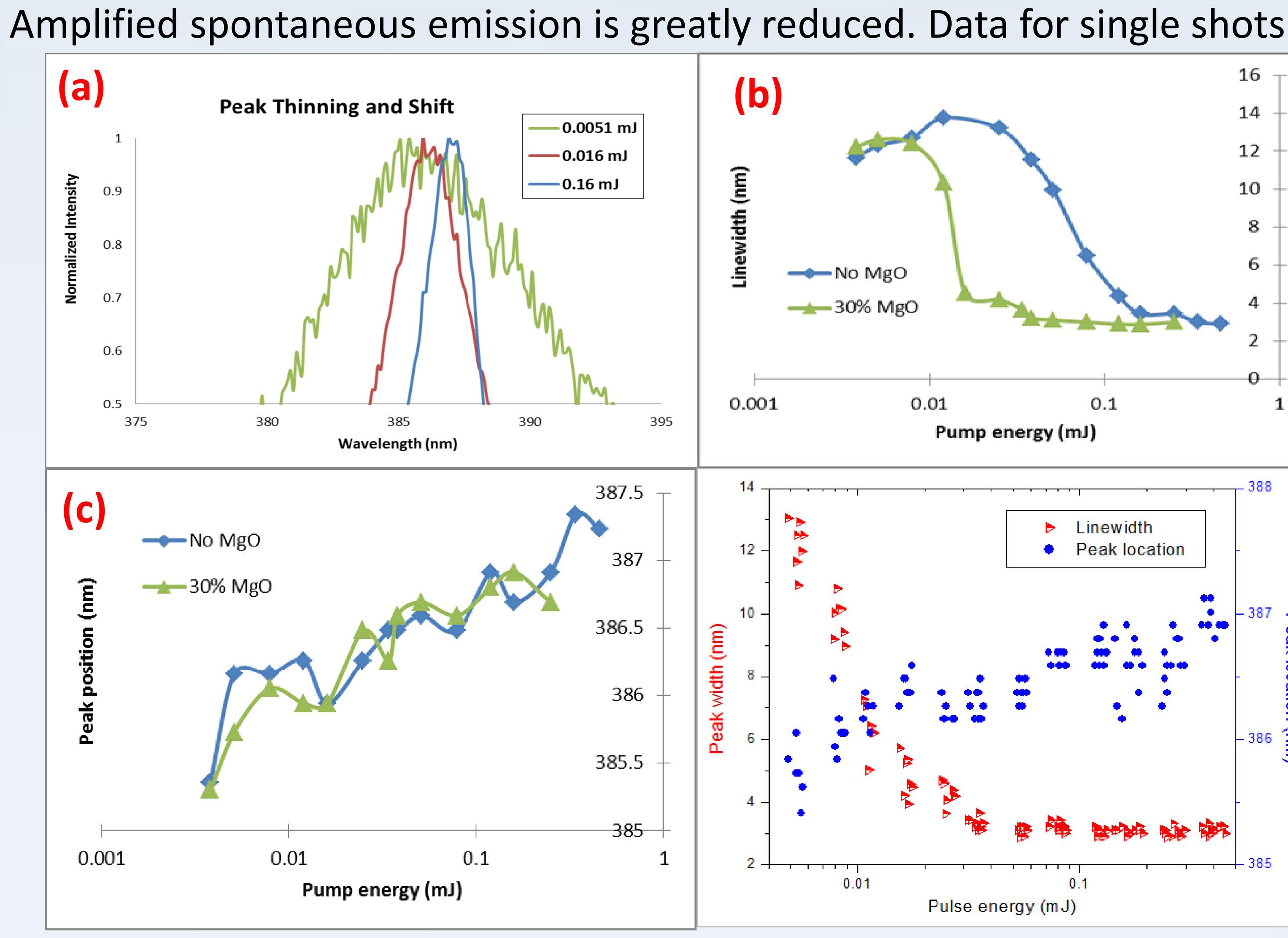

Figure 7. (a) Random laser spectra under single shot picosecond pumping. (b) and (c) show linewidth and peak position data from ZnO only and ZnO/30% MgO samples. (d) Peak shift and linewidth data vs. pumping intensity from 30% MgO sample under single pump pulses.

SALT results and statistical modelling

- 1-D SALT model shows threshold behavior in media with volume averaged refractive indices.
- Statistical model developed using gain curve (Eq. 5) as probability distribution for lasing mode peak location with Poisson distribution on mode number and prescribed mode width. Model results reproduce experimental results.

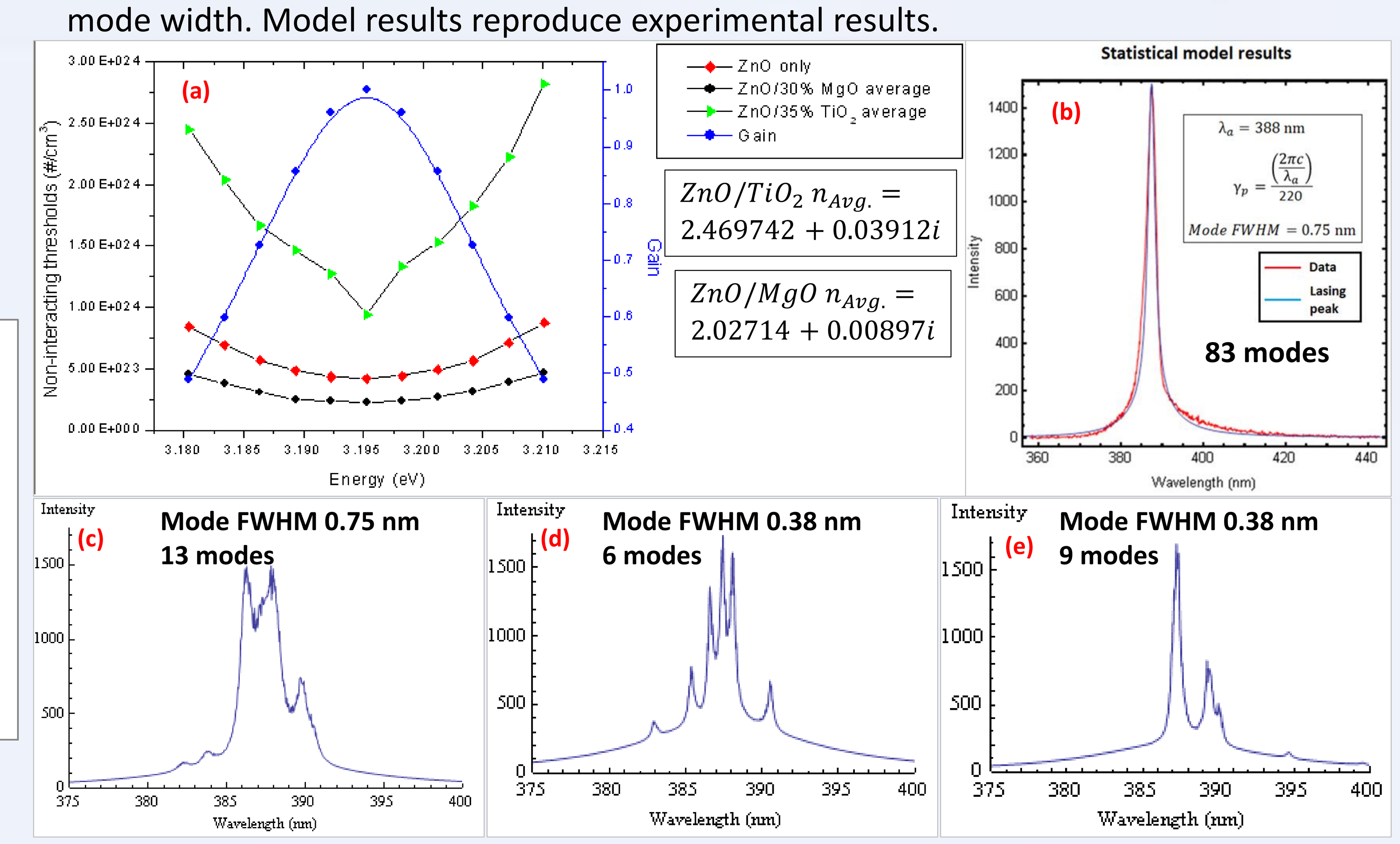

Figure 8. (a) SALT simulation results showing threshold reduction. (b) Statistical model results with Poisson mean = 75. (c)-(e) show generated spectra with Poisson mean = 10.

Conclusions

- Addition of MgO lowers threshold by reducing absorption at each scattering event.
- During pump pulse buildup to maximum energy, spontaneous emission can occur. Longer pulse times allow for more spontaneous emission events.
- Statistical model suggests our spectra contain such a large number of spectrally overlapping modes that they cannot be individually resolved.
- Peak emission wavelength may shift while emitting (EHP recombination).
- More work is needed to achieve coherent feedback.

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