

Random Lasers and Photonic Spin-Glasses: an introduction to theoretical and experimental challenges

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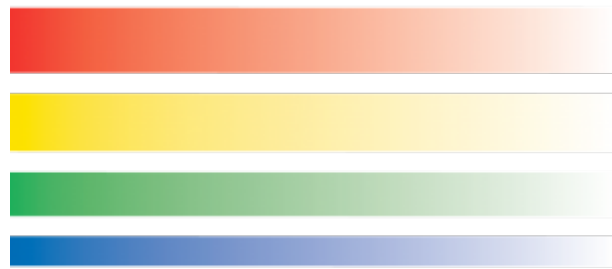
in collaboration with

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Neda Ghofraniha, Miguel Ibanez Berganza, Payal Tyagi - IPCF-CNR

Viola Folli, Marco Leonetti - ISC-CNR

Fabrizio Antenucci, Alessia Marruzzo - Sapienza University.



NETADIS

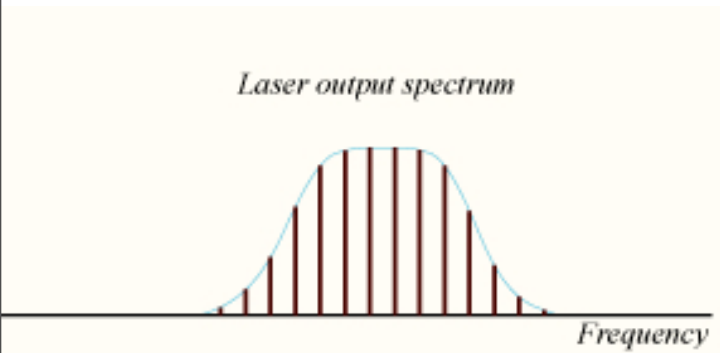
Statistical Physics Approaches
to
Networks Across Disciplines



SAPIENZA
UNIVERSITÀ DI ROMA

Laser

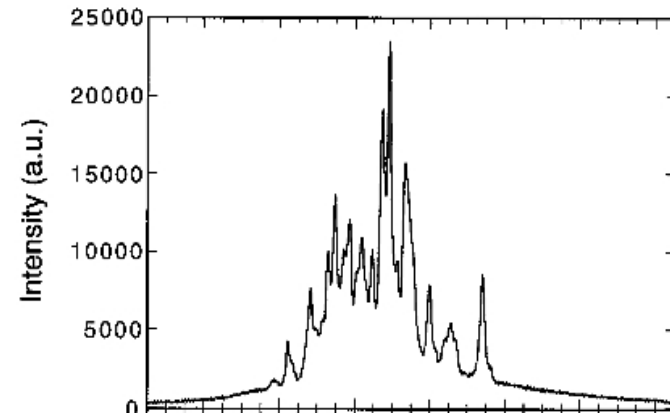
1958:
Infrared and Optical Masers, by
 Arthur L. Schawlow, Charles H. Townes,
 Physical Review



Random Laser

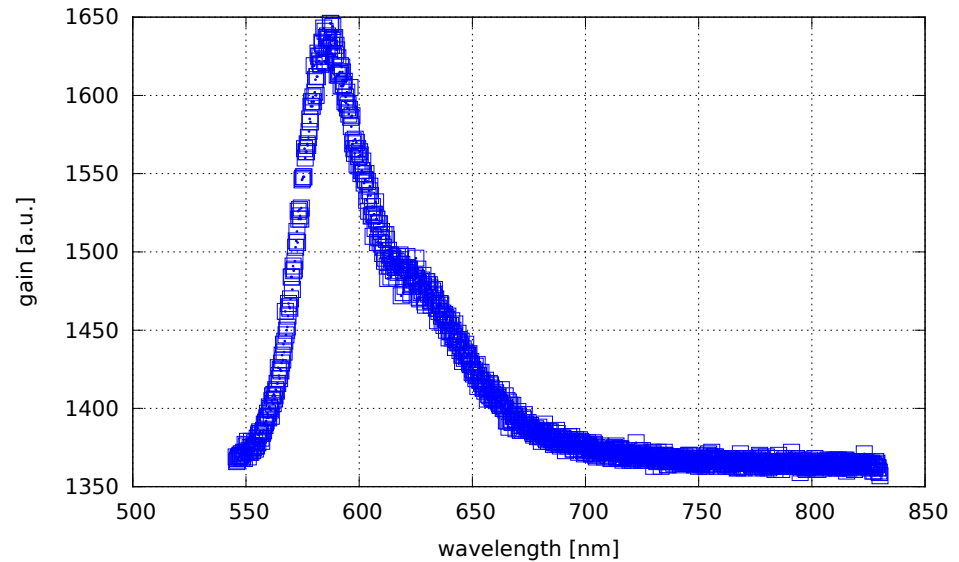
1967: VS Letokhov - JETP Lett. (theory)
 Idea: Stimulated emission without a resonant cavity

1994: Lawandy
Laser action in strongly scattering media (experiments)
 Realization: multiple scattering medium (i.e., set of stochastic resonators), a multimode laser with disorder.



Random Laser

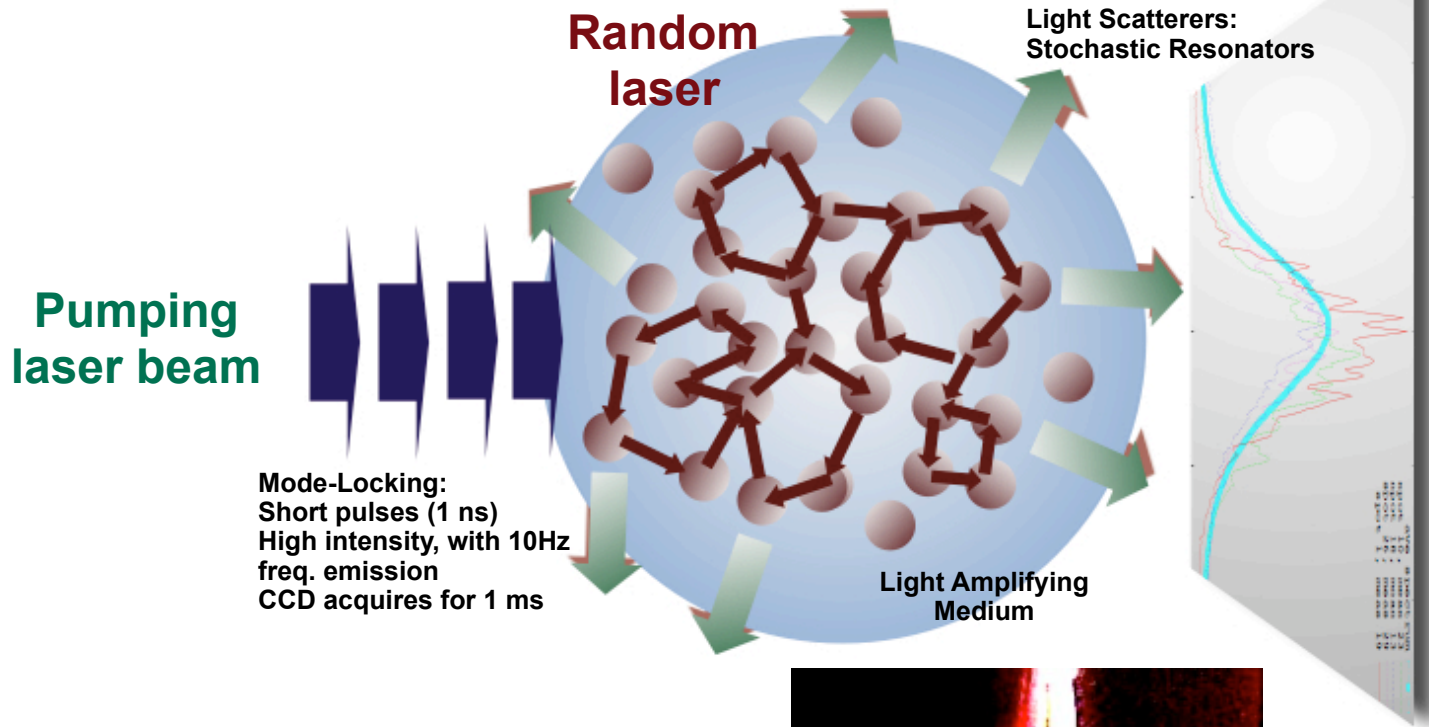
Pumping mode-locked laser (pulses of high intensity)
in an optically active disordered material (powder/precipitate in light amplifying medium)



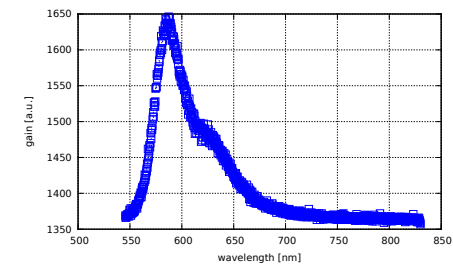
E.g., precipitate of TiO_2 particles (scatterers) in methanol solution
doped by **rhodamine** (light amplifying)

Random Laser

Pumping mode-locked laser (pulses of high intensity)
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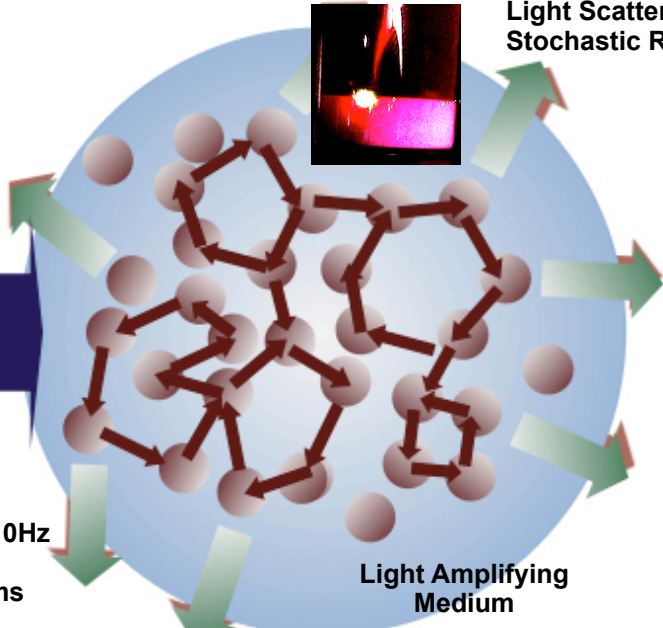
E.g., precipitate of TiO_2 particles (scatterers)
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rhodamine (light amplifying)



Random laser

Pumping laser beam

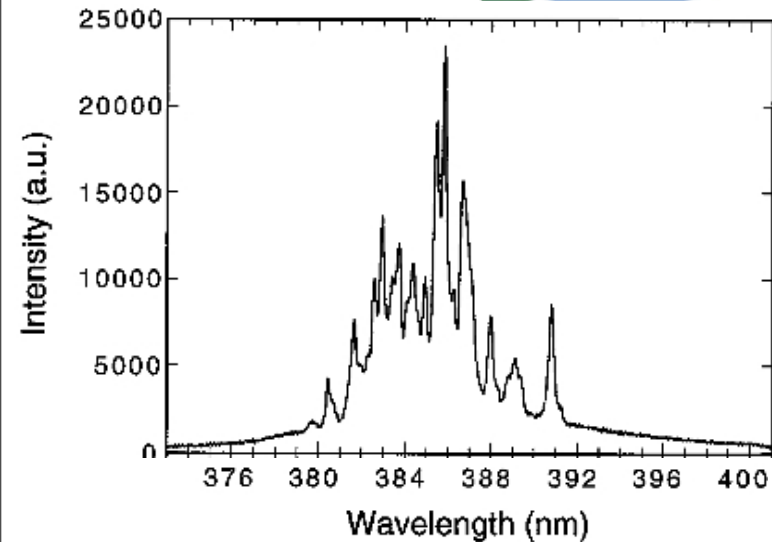
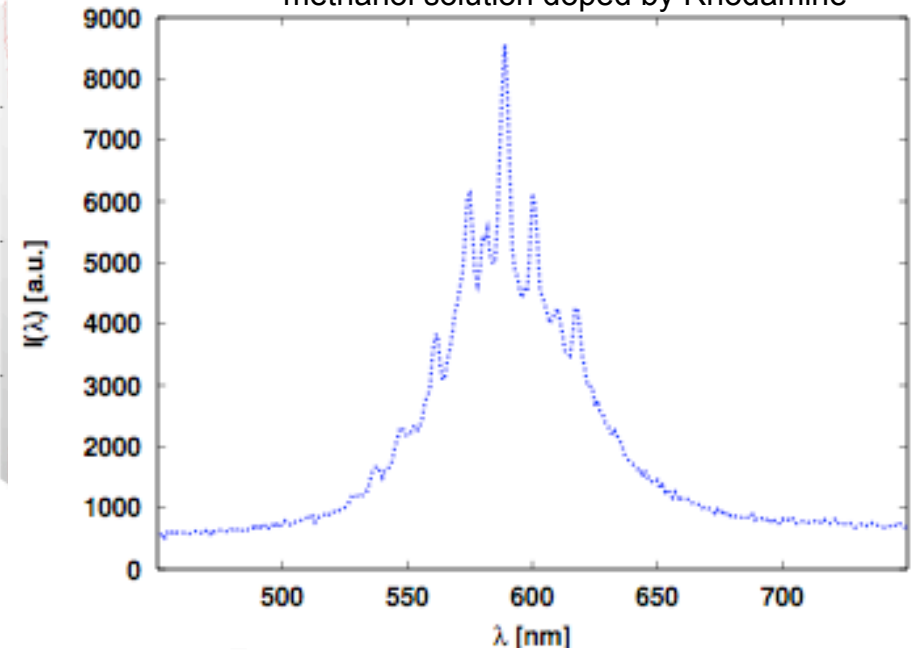
Mode-Locking:
Short pulses (1 ns)
High intensity, with 10Hz
freq. emission
CCD acquires for 1 ms



Light Scatters:
Stochastic Resonators

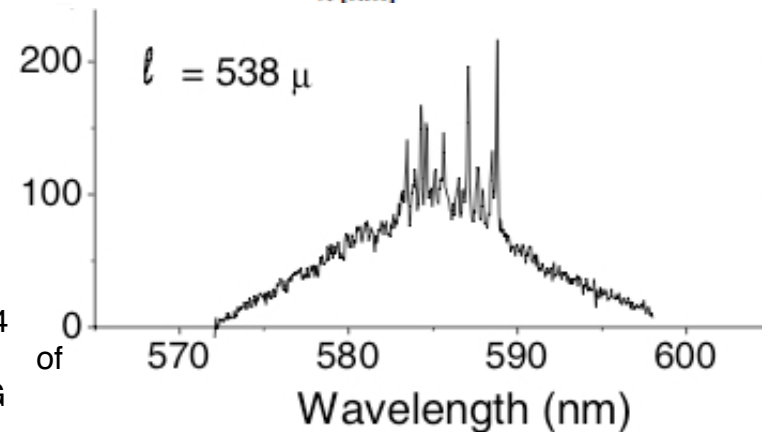
Light Amplifying Medium

M. Leonetti, LL, C. Conti, unpublished,
colloidal dispersion of TiO₂ particles in
methanol solution doped by Rhodamine



H. Cao et al.
PRL 99
ZnO powder

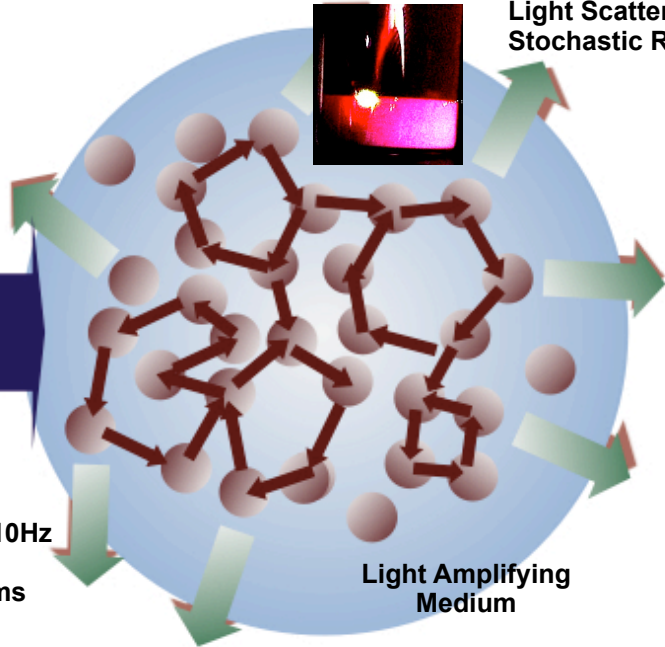
S. Mujumdar
et al. PRL '04
Suspensions
of ZnO in Rh6G



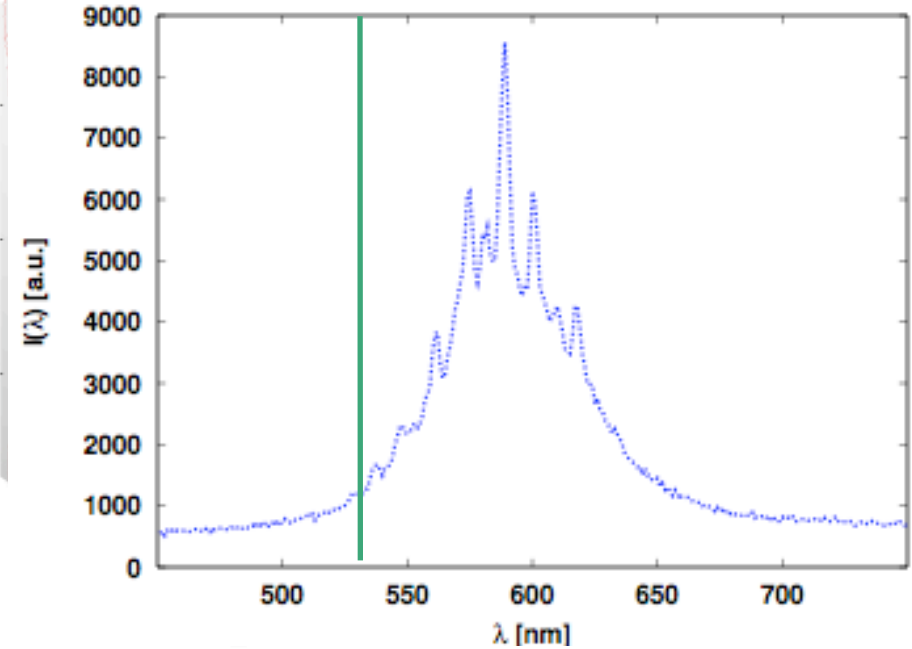
Random laser

Pumping laser beam

Mode-Locking:
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 High intensity, with 10Hz
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M. Leonetti, LL, C. Conti, unpublished

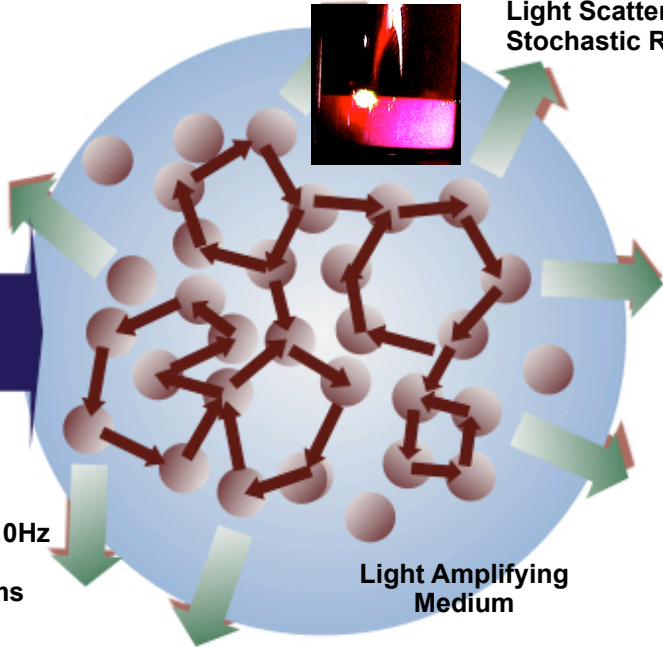


FAQS

Random laser

Pumping laser beam

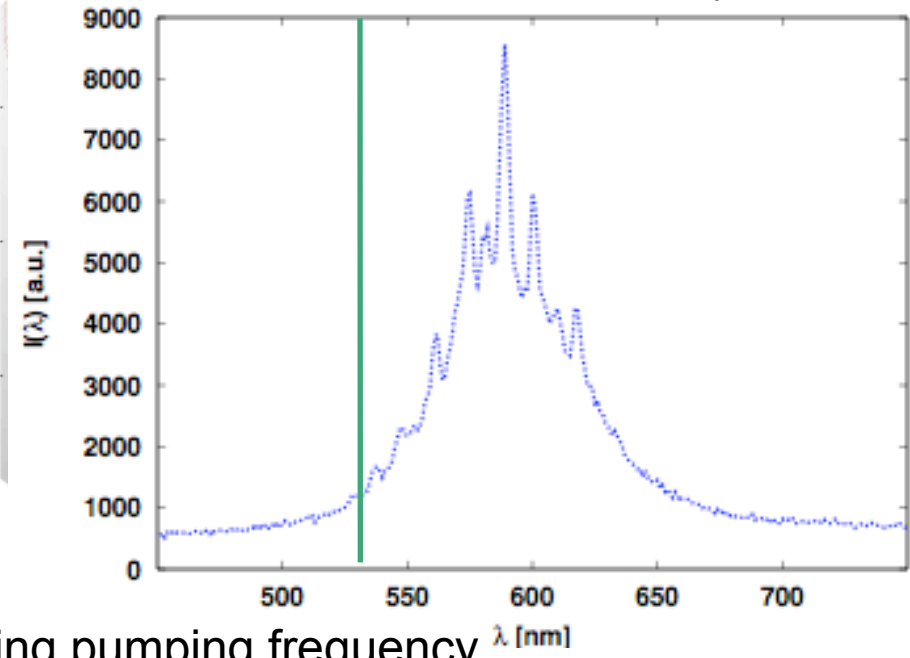
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Light Scatterers:
 Stochastic Resonators

Light Amplifying Medium

M. Leonetti, LL, C. Conti, unpublished

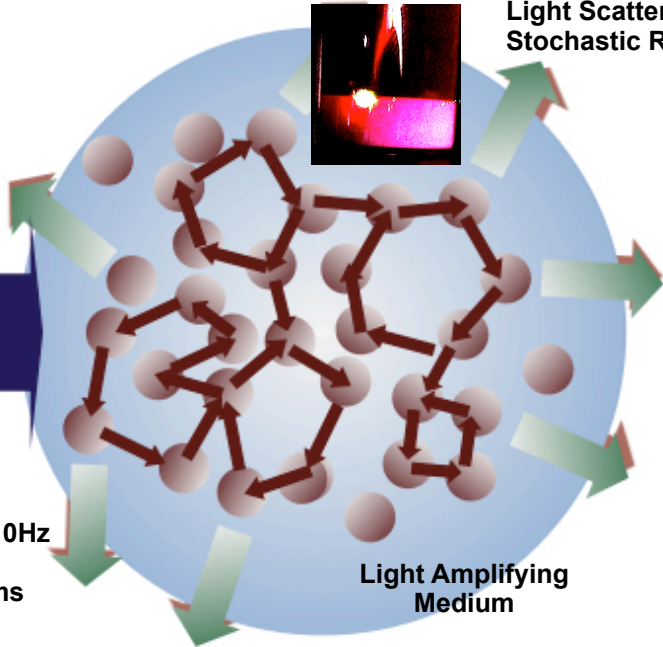


Frequency band of excited modes not including pumping frequency

Random laser

Pumping laser beam

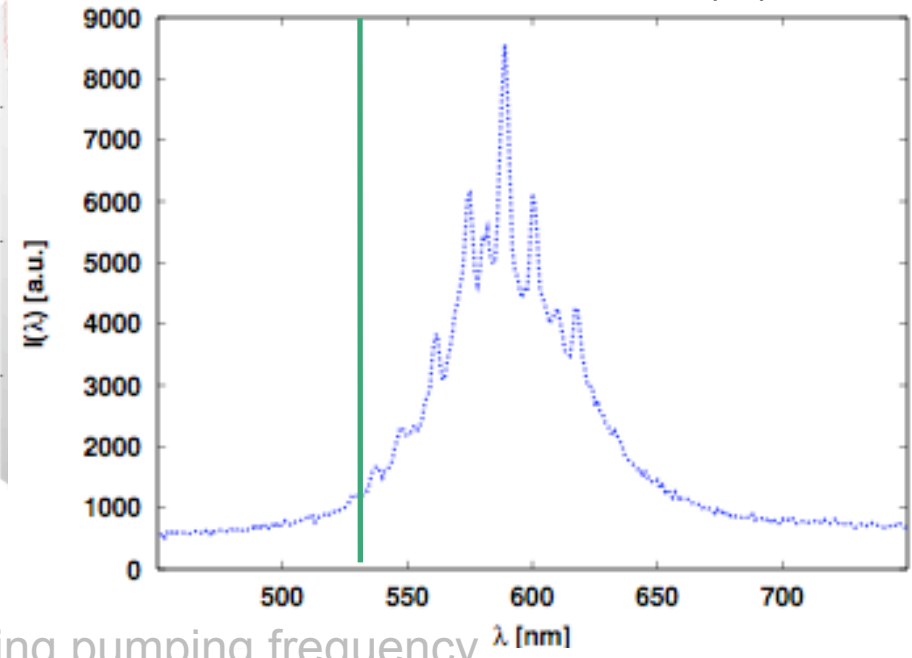
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**Light Scatterers:
 Stochastic Resonators**

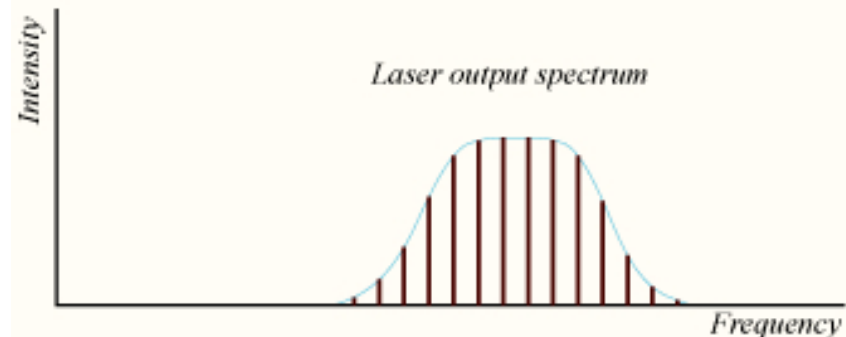
**Light Amplifying
 Medium**

M. Leonetti, LL, C. Conti, in preparation



Frequency band of excited modes not including pumping frequency

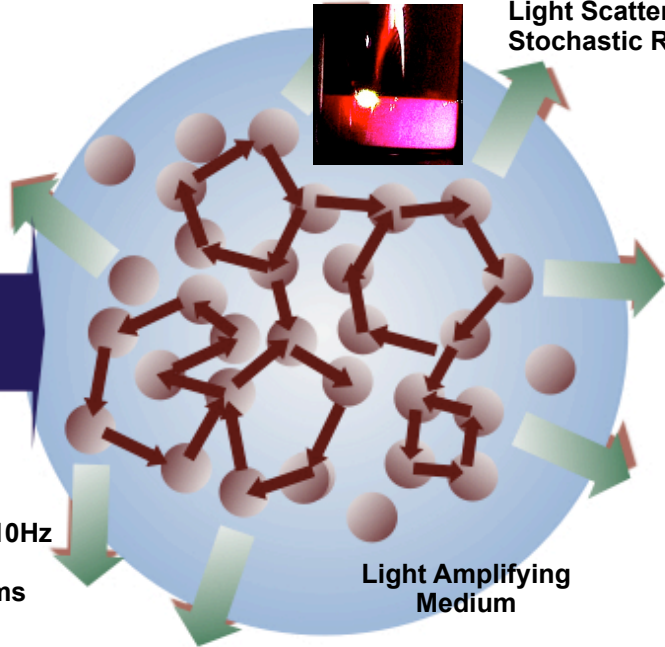
Different from multimode standard laser profile



Random laser

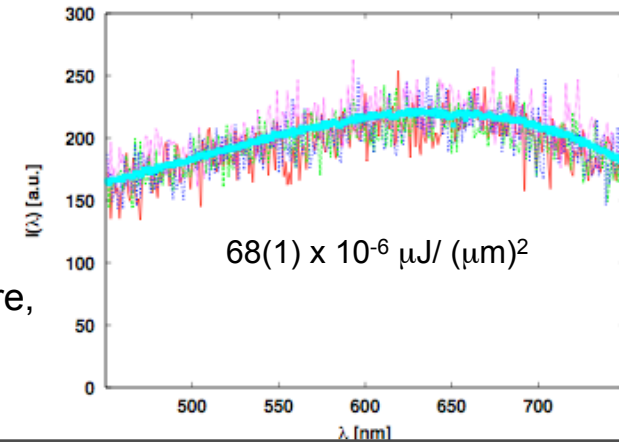
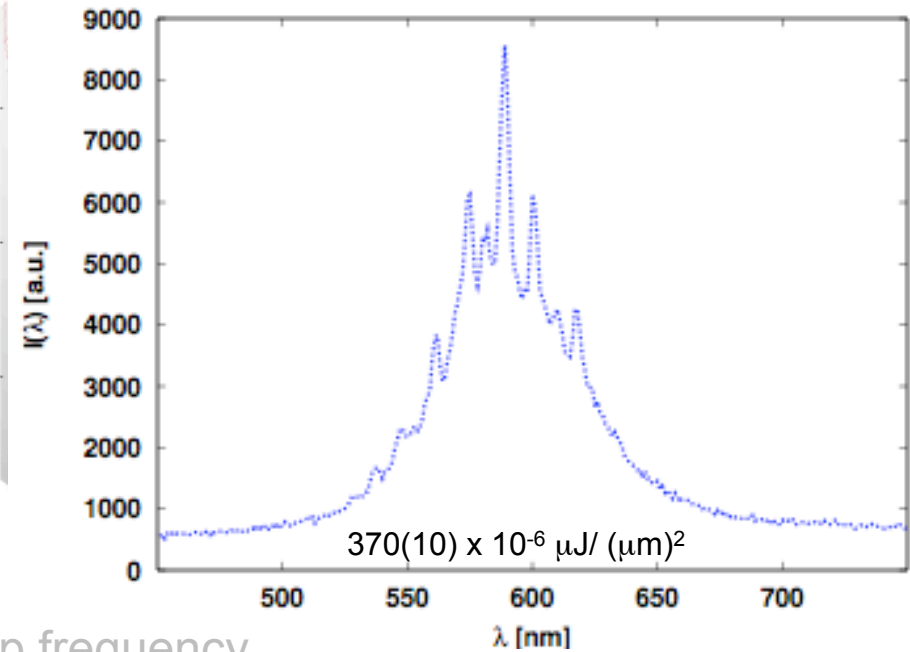
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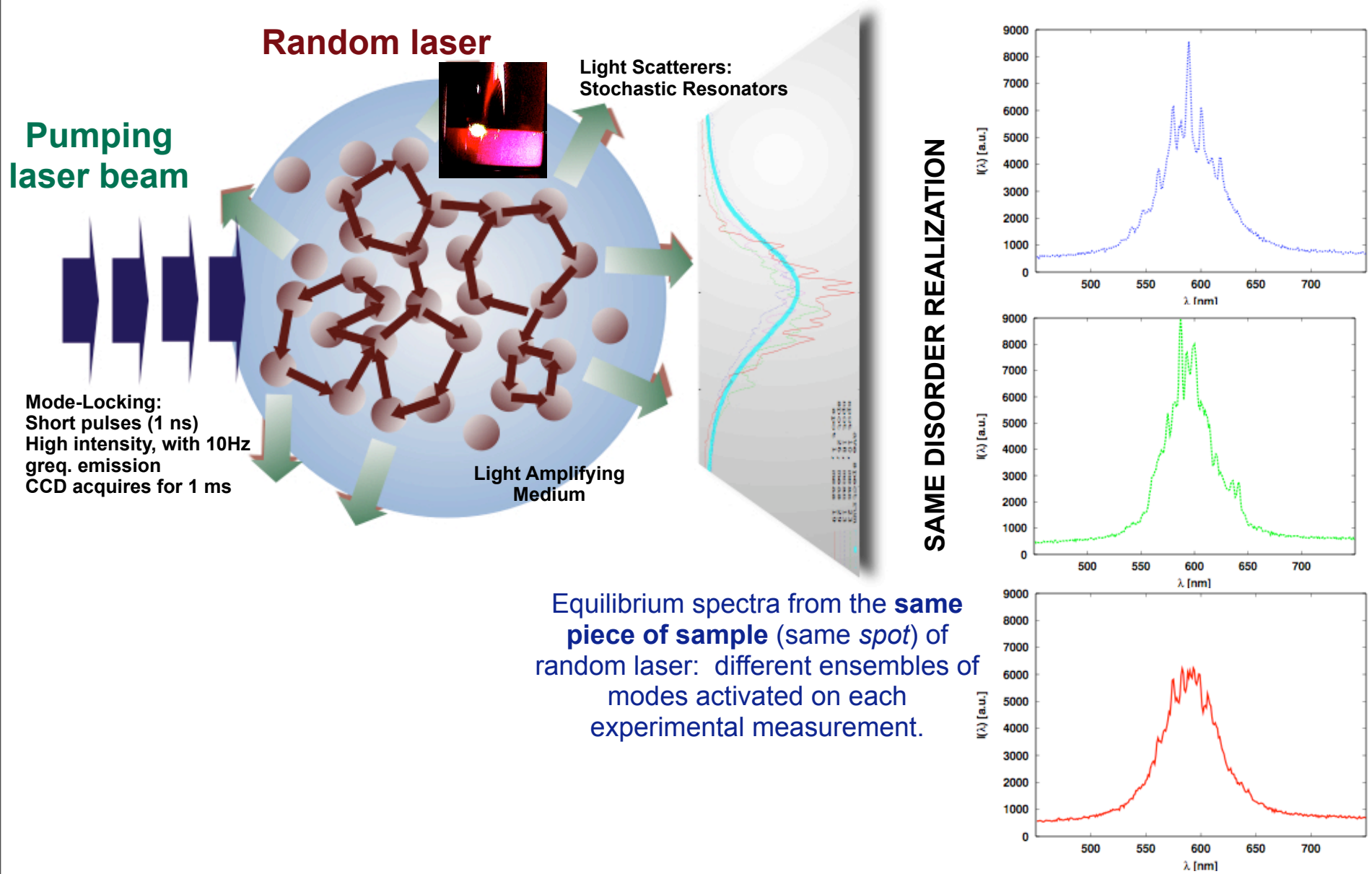
Light Scatters:
Stochastic Resonators

M. Leonetti, LL, C. Conti, unpublished

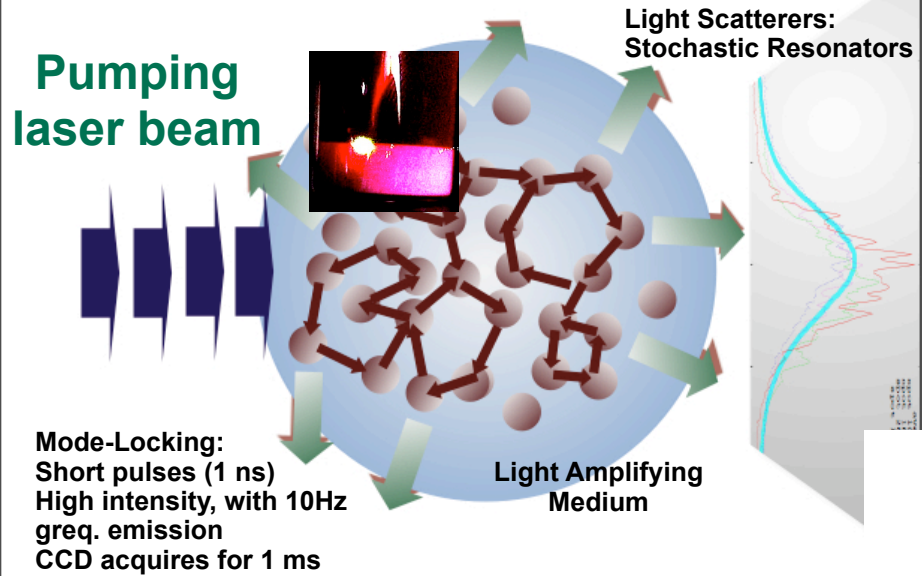


Frequency band of modes not including pump frequency
Different from multimode standard laser profile
Profile typical for high pumping intensity, qualitatively
different from low pumping intensity: **“transition”** from
continuous wave emission to random lasing regime.

This also happens decreasing temperature,
D Wiersma and S Cavalieri Nature 2001



Random laser

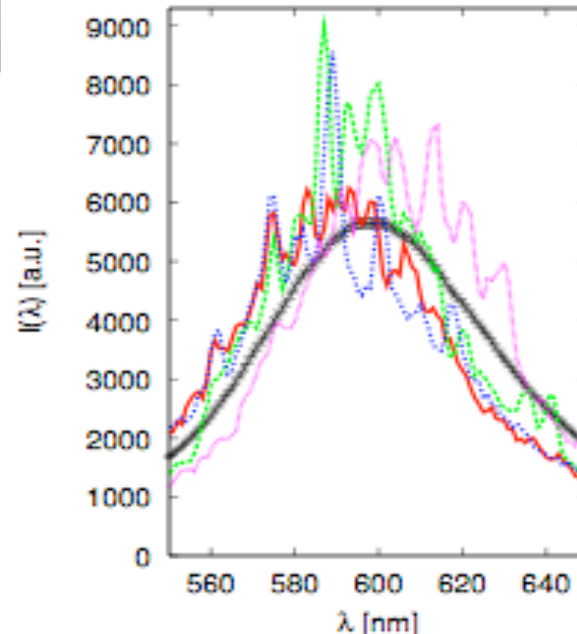


Equilibrium spectra from the **same piece of sample** (same *spot*) of random laser: different ensembles of modes activated on each experimental measurement.

Explanation:

Chaos?

Experimental uncertainty?



Random Laser Ti:O
rhodamine sol.
in methanol
Pumping Laser Nd:YAG

peak RL: $\lambda \sim 600$ nm

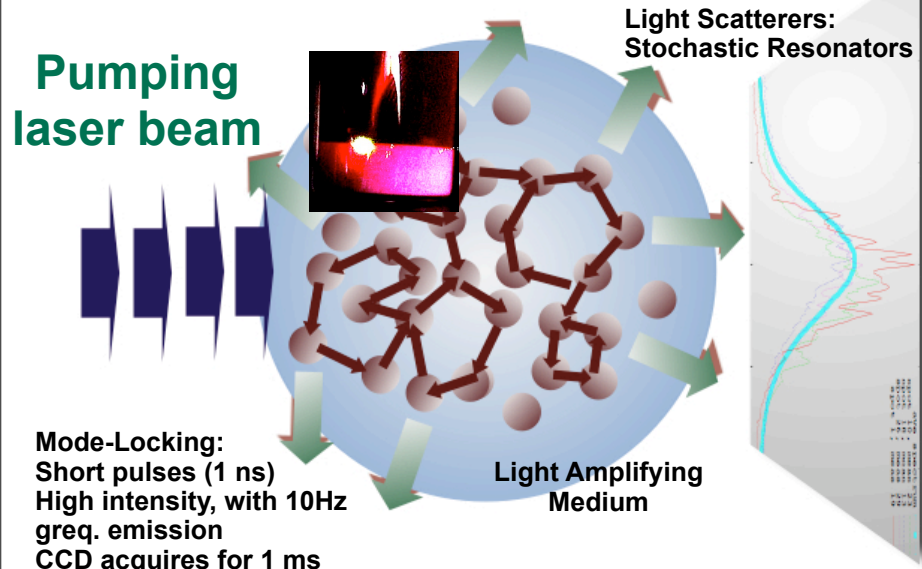
pumping: $\lambda = 532$ nm

bin: $\delta\lambda = 0.03$ nm

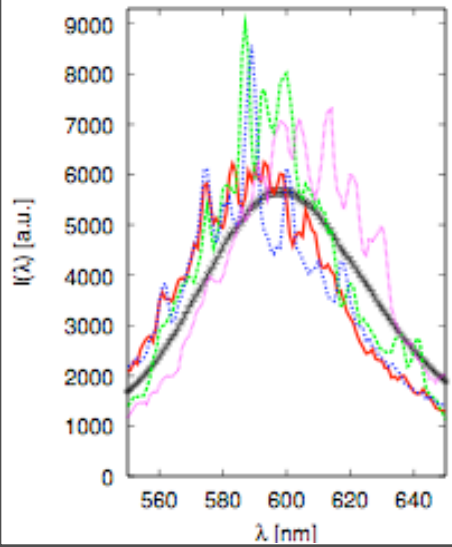
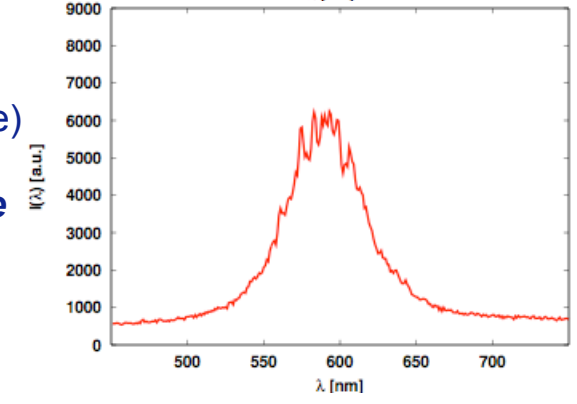
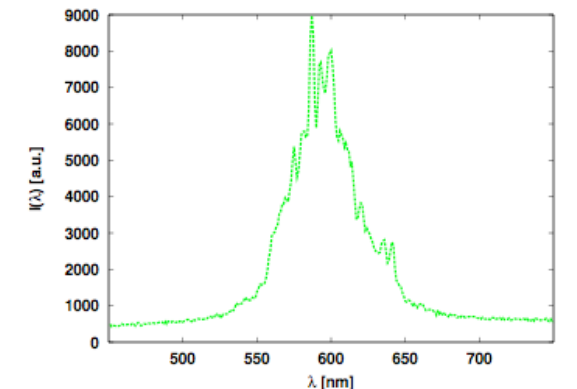
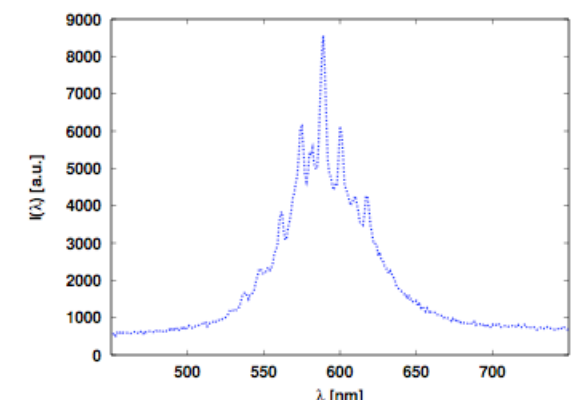
I of pumping beam
 ~ 0.0004 [$\mu\text{J}/\mu\text{m}^2$]

I of RL absorbed by CCD:
 $611(8) \times 10^3$ (countings)

Random laser



SAME DISORDER REALIZATION



Random Laser Ti:O
rhodamine sol.
in methanol

Pumping Laser Nd:YAG

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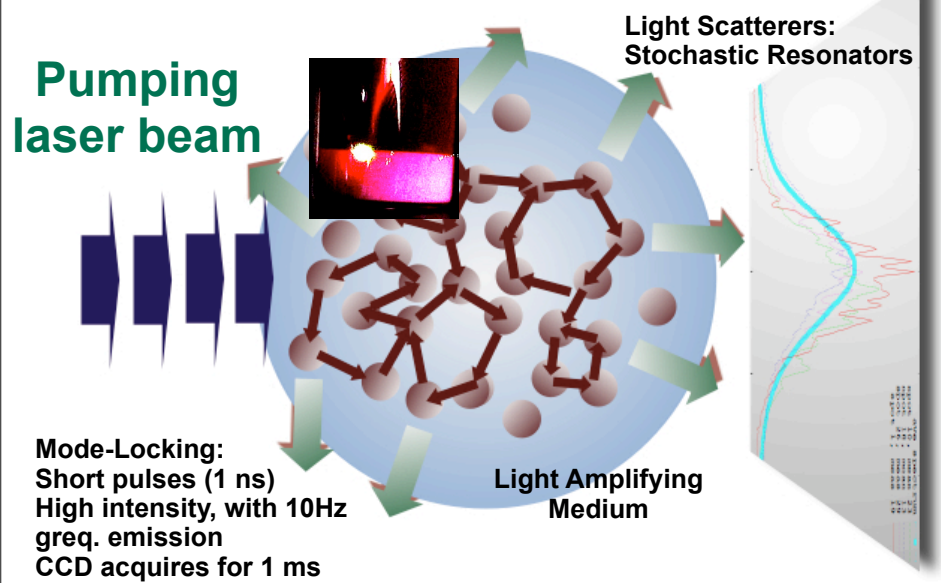
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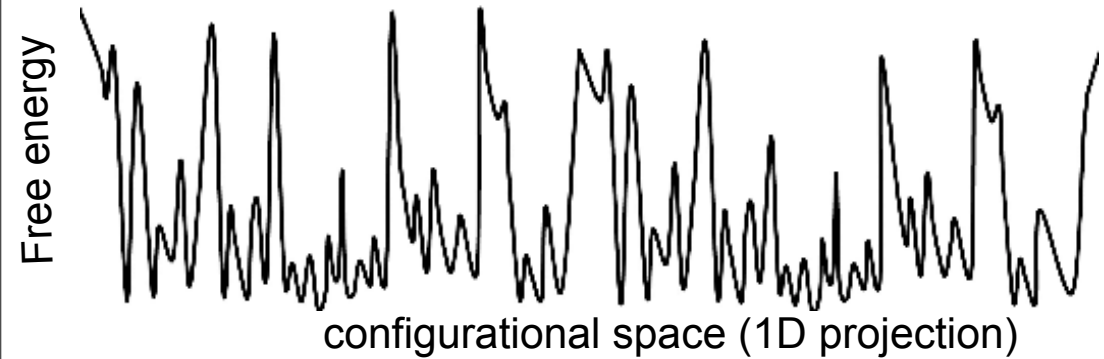
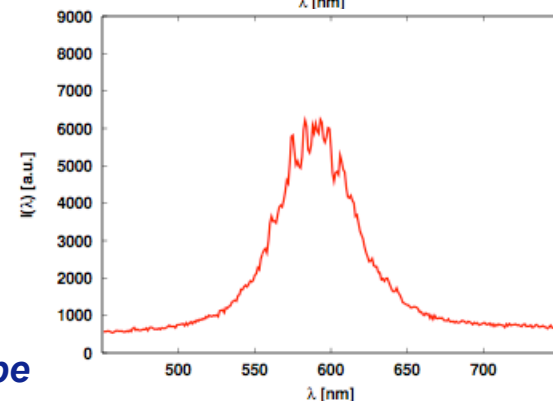
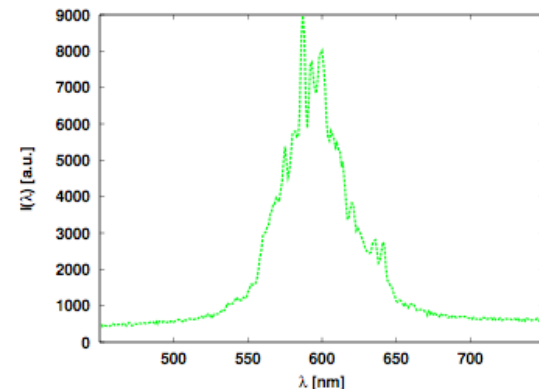
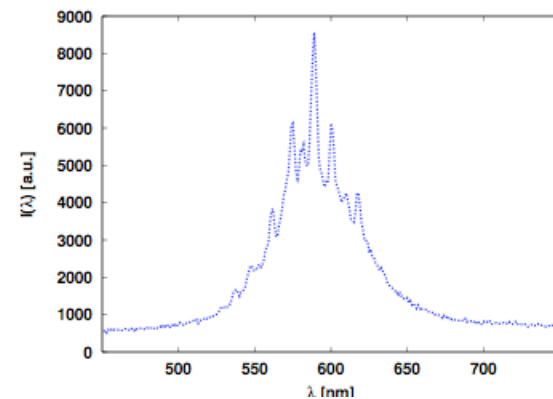
Explanation (proposal):
Mode competition phenomena
(frequency locking at each measure)

Conjecture:
Complex free energy landscape

Random laser



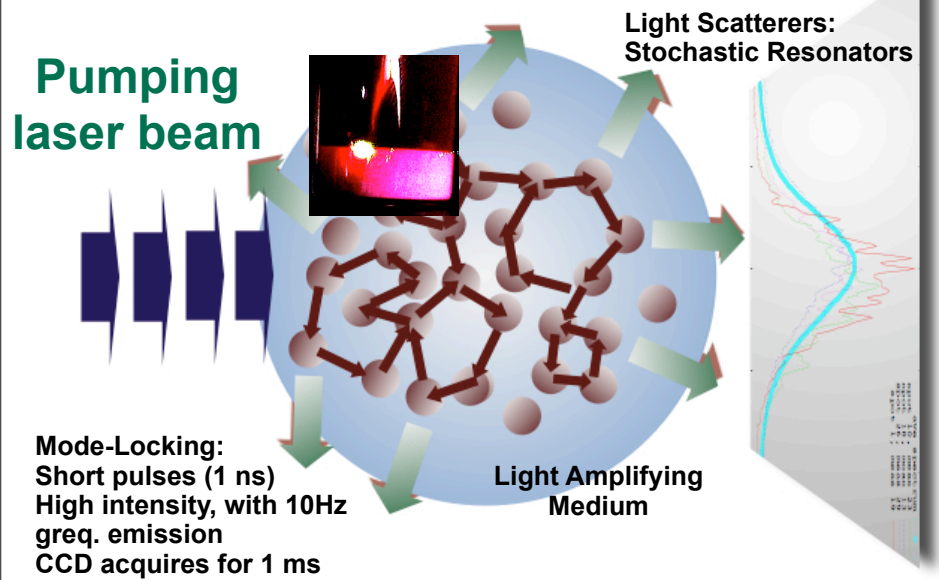
SAME DISORDER REALIZATION



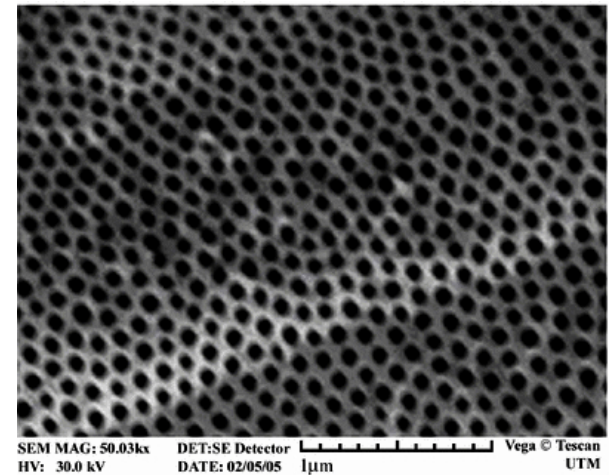
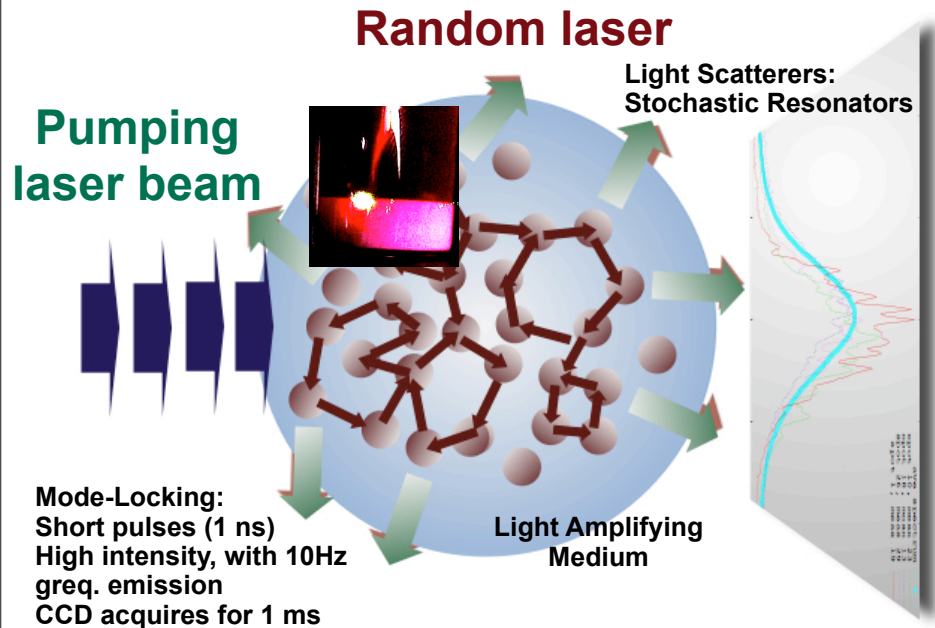
Complex free energy landscape

M. Leonetti, LL, C. Conti, unpublished

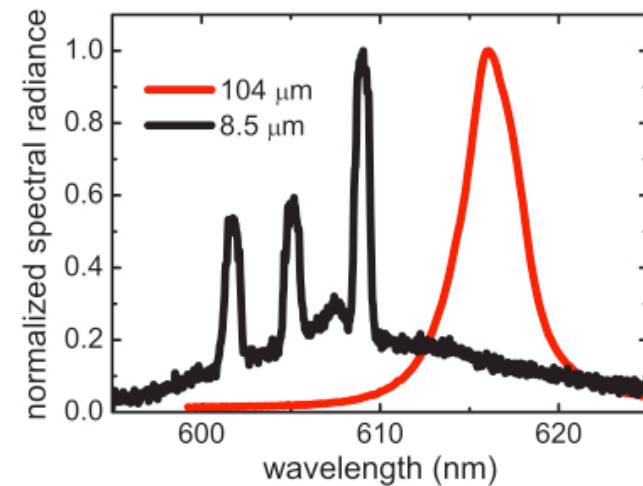
Random laser



This is not general for all random lasers



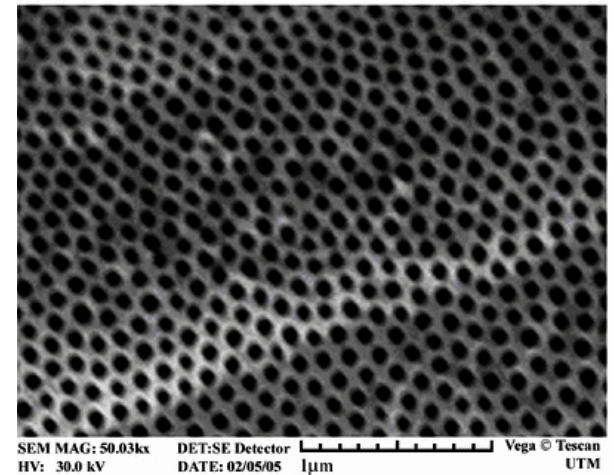
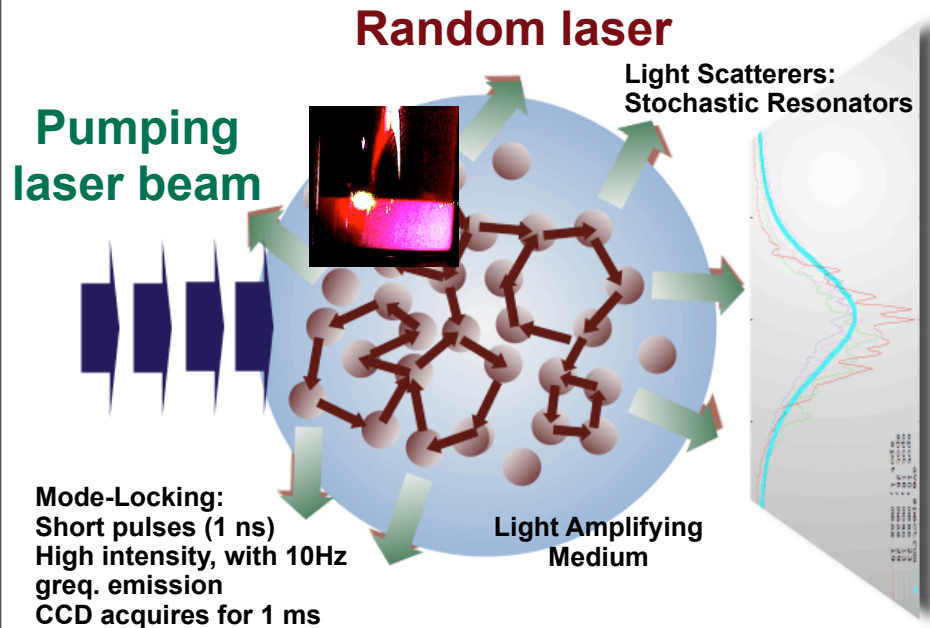
Porous GaP



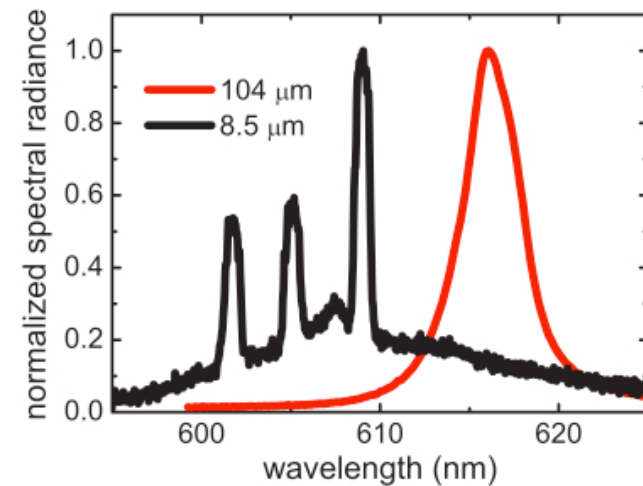
This is not general for all random lasers

There are also random lasing materials [porous semiconducting matrix infiltrated with and embedded in laser dye] where reproducibility of spikes is claimed.

El-Dardiry *et al.* Phys. Rev. A 2010



Porous GaP



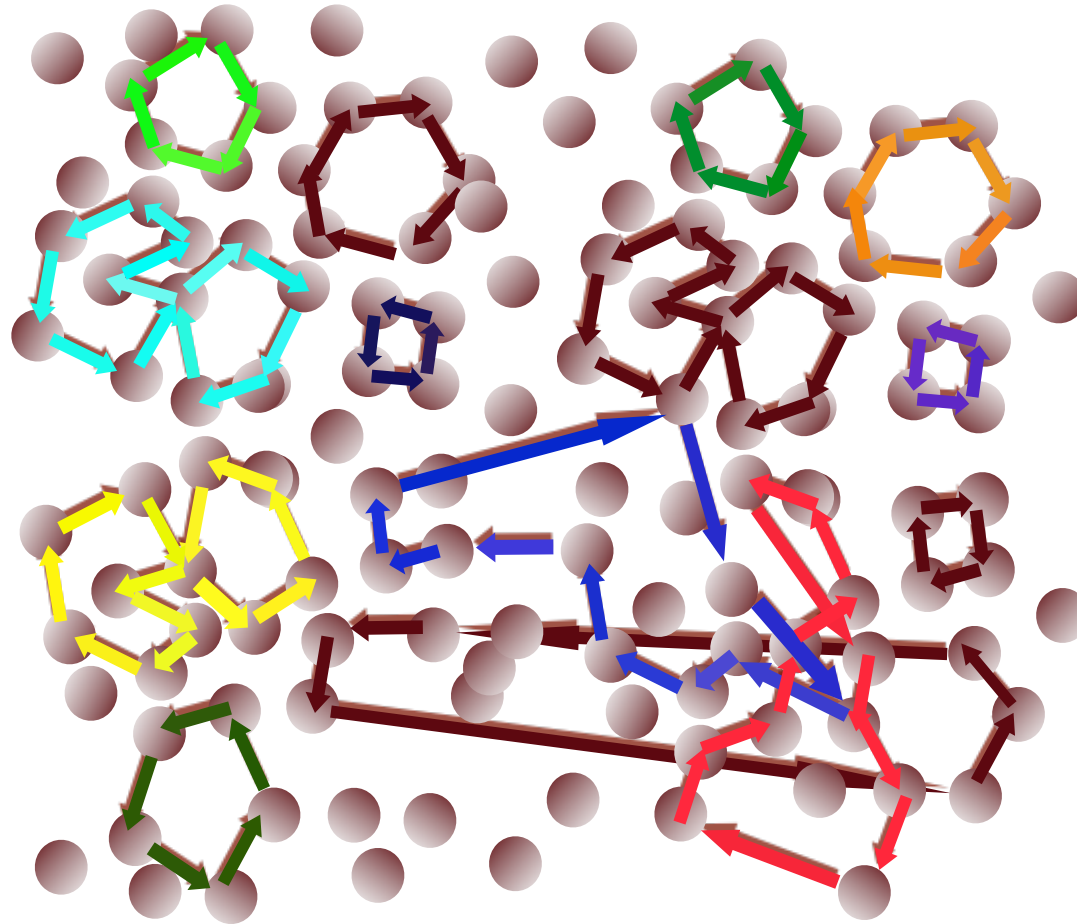
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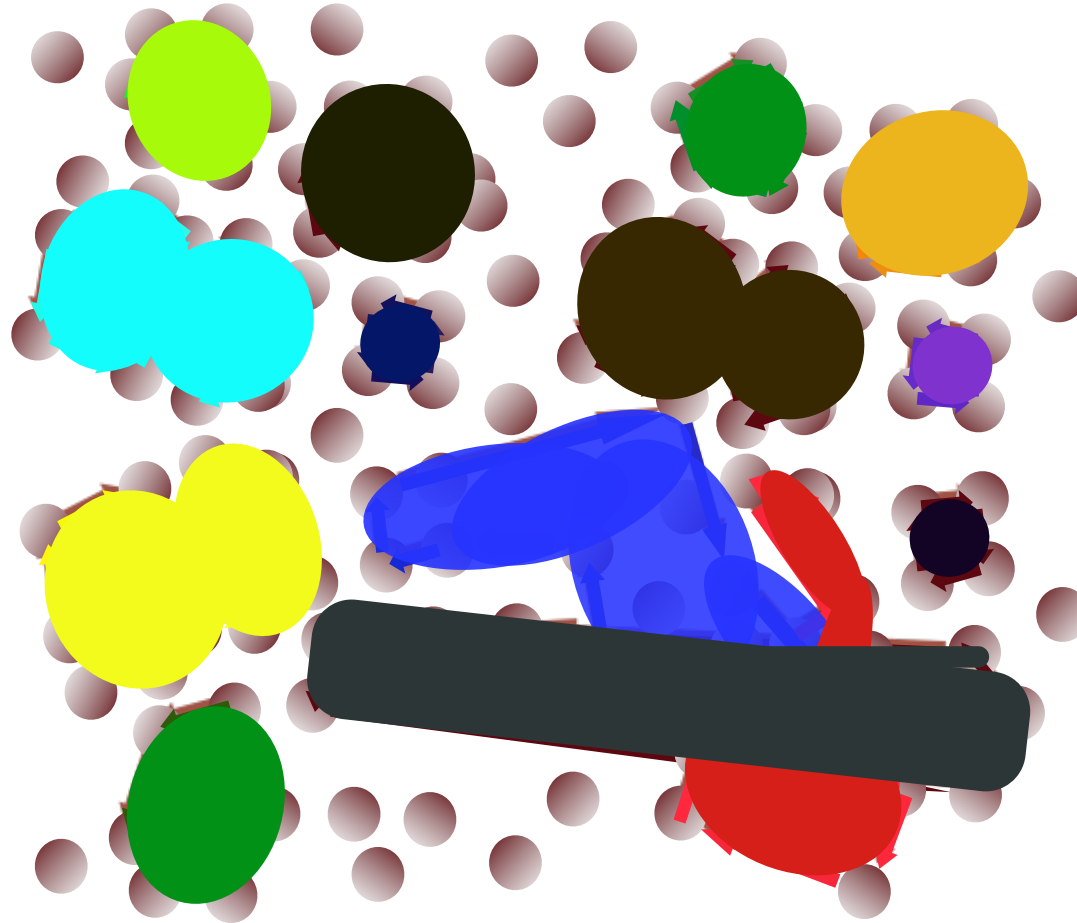
El-Dardiry *et al.* Phys. Rev. A 2010

cavity-less laser?

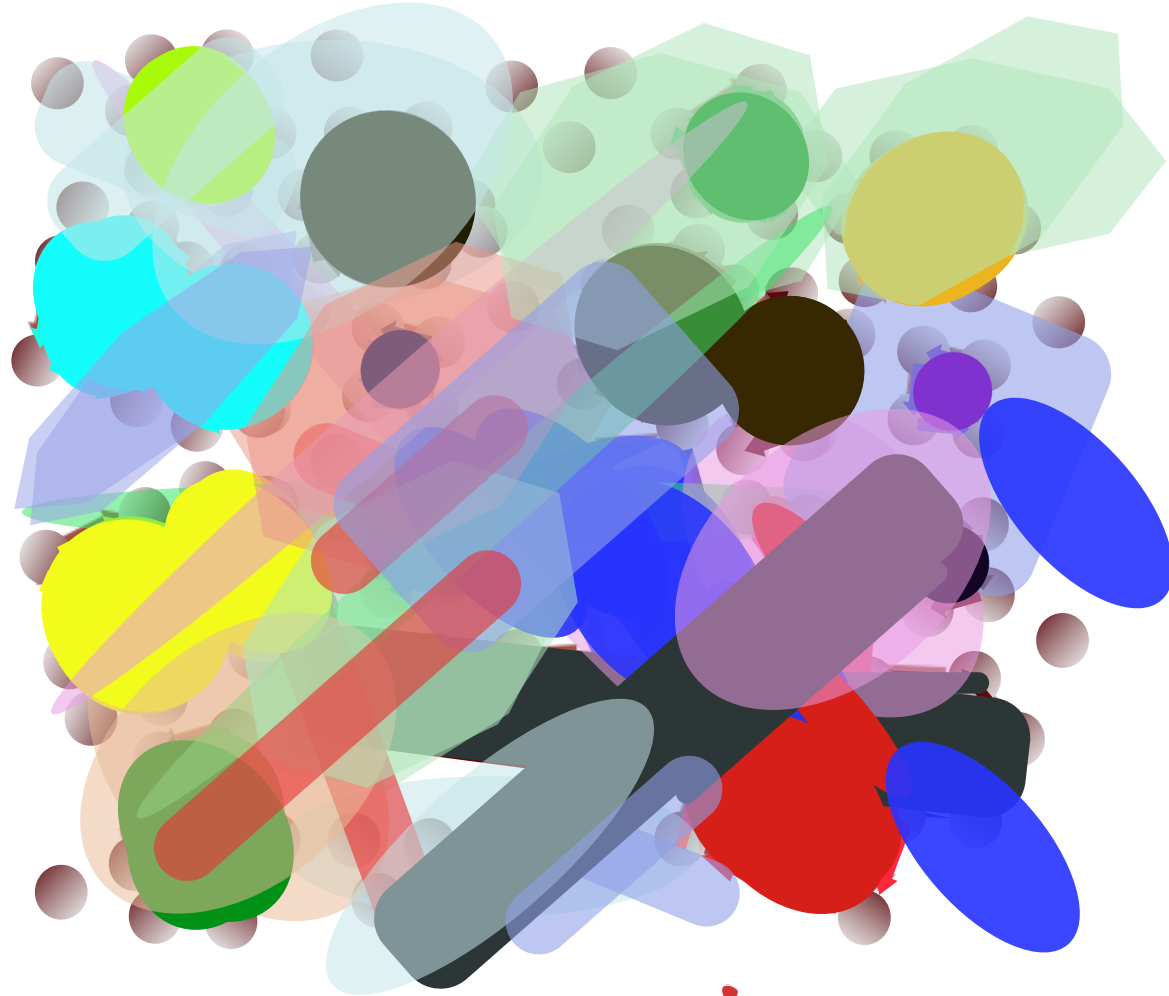
Modeling random laser modes in space:
 the electromagnetic field of each localized light mode of
 frequency ω is non-zero in a given region of space r .



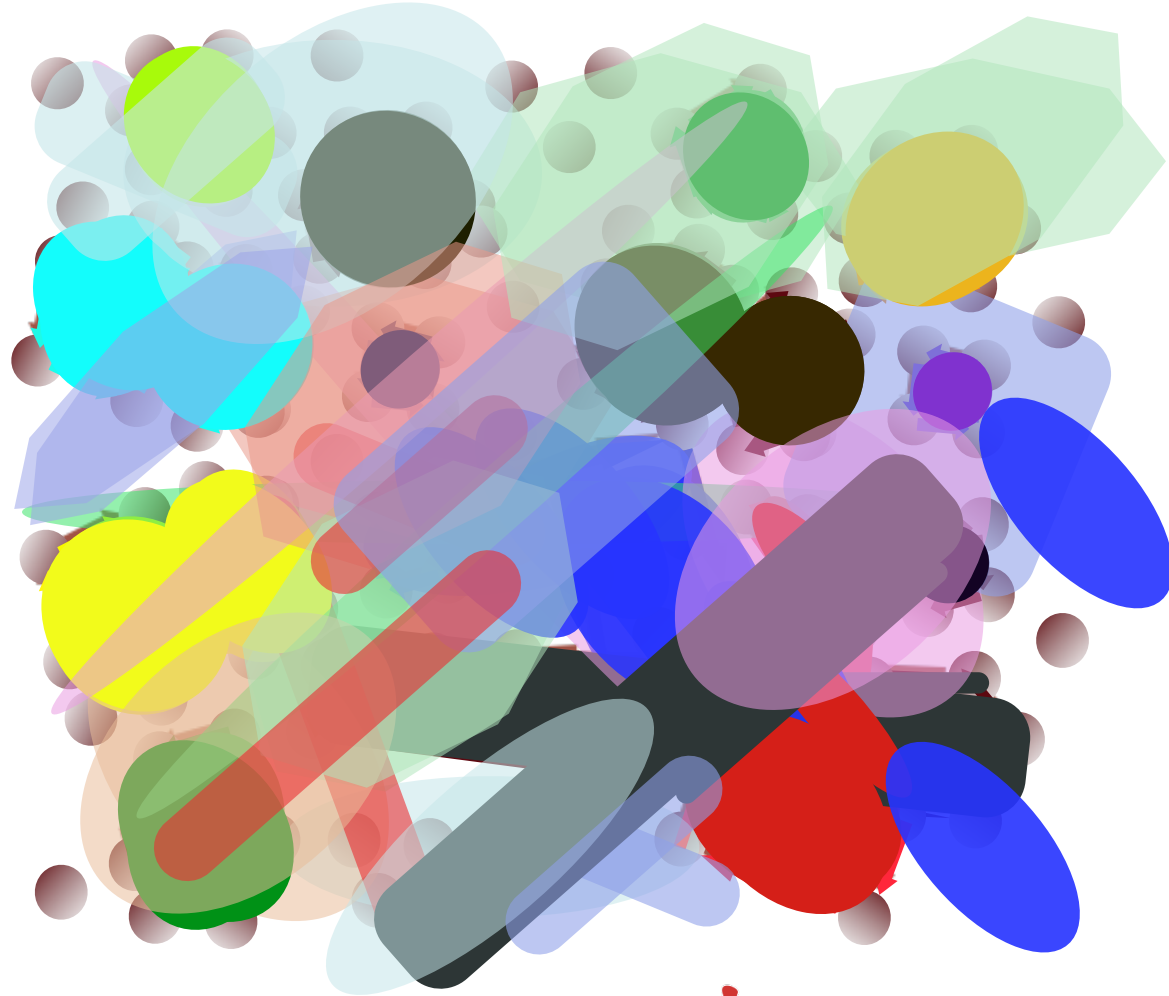
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*A bit less
pictorial...?*

Electromagnetic Cavity of refractive index profile $n(\mathbf{r})$
 non linear polarization $\mathbf{P}_{NL}(\mathbf{r})$
 displacement vector $\mathbf{D}(\mathbf{r}) = \epsilon_0 n^2(\mathbf{r})\mathbf{E}(\mathbf{r}) + \mathbf{P}_{NL}(\mathbf{r})$

Maxwell equations in presence of nonlinear polarization in an electromagnetic cavity

$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} = \epsilon_0 n^2(\mathbf{r}) \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}_{NL}}{\partial t}$$

$$\nabla \times \mathbf{E} = -\mu_0 \partial_t \mathbf{H}$$

K Sakoda, *Optical Properties of Photonic Crystals*, 2001
 HA Haus, *Waves and Fields in Optoelectronics*, 1984
 L Angelani et al. PRB 06

Solution to the equations is a superposition of modes:

$$\mathbf{E} = \text{Re} \left[\sum_n a_n(t) \mathbf{E}_n(\mathbf{r}) \exp(-i\omega_n t) \right]$$

$$\mathbf{H} = \text{Re} \left[\sum_n a_n(t) \mathbf{H}_n(\mathbf{r}) \exp(-i\omega_n t) \right]$$

Complex amplitudes such that total energy stored in the EM cavity (closed):

$$\mathcal{E} = \sum_k \mathcal{E}_k = \sum_k |a_k|^2$$

Electromagnetic Cavity of refractive index profile $n(\mathbf{r})$

non linear polarization \mathbf{P}_{NL} and displacement vector $\mathbf{D}(\mathbf{r}) = \epsilon_0 n^2(\mathbf{r})\mathbf{E}(\mathbf{r}) + \mathbf{P}_{NL}(\mathbf{r})$

$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} = \epsilon_0 n^2(\mathbf{r}) \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}_{NL}}{\partial t}$$

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$$\mathcal{E} = \sum_k \mathcal{E}_k = \sum_k |a_k|^2$$

$$\mathbf{H} = \text{Re} \left[\sum_n a_n(t) \mathbf{H}_n(\mathbf{r}) \exp(-i\omega_n t) \right]$$

If $\mathbf{P}_{NL}=0$, amplitudes are constant the whole time dependence is in the oscillation and $\mathbf{E}_n, \mathbf{H}_n$ are the eigenvalues of the system

$$\mathcal{L} \mathcal{F}_n^{(0)} = \omega_n \mathcal{M} \mathcal{F}_n^{(0)}$$

$$\mathcal{F}_n^{(k)} = \begin{pmatrix} \mathbf{E}_n^{(k)} \\ \mathbf{H}_n^{(k)} \end{pmatrix}$$

$$\mathcal{L} = \begin{pmatrix} 0 & i\nabla \times \\ -i\nabla \times & 0 \end{pmatrix}$$

$$\mathcal{M} = \begin{pmatrix} \epsilon_0 n^2(\mathbf{r}) & 0 \\ 0 & \mu_0 \end{pmatrix}$$

Electromagnetic Cavity of refractive index profile $n(\mathbf{r})$

non linear polarization \mathbf{P}_{NL} and displacement vector $\mathbf{D}(\mathbf{r}) = \epsilon_0 n^2(\mathbf{r})\mathbf{E}(\mathbf{r}) + \mathbf{P}_{NL}(\mathbf{r})$

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$$\mathbf{H} = \text{Re} \left[\sum_n a_n(t) \mathbf{H}_n(\mathbf{r}) \exp(-i\omega_n t) \right]$$

If \mathbf{P}_{NL} is not zero, amplitudes depend on time and solution has the general form above
To obtain the time evolution of the amplitudes we expand around weak polarization, i.e.,
slowly varying amplitudes: multiscale approach

$$a(t) = a(t_1, t_2, \dots, t_n)$$

$$t_n = \eta^n t$$

$$t_0 = t$$

$$\partial_t = \sum_k \frac{\partial t_k}{\partial t} \partial_{t_k} = \sum_k \eta^k \partial_{t_k} \simeq \partial_{t_0} + \eta \partial_{t_1} + \dots$$

If \mathbf{P}_{NL} is not zero, amplitudes depend on time and solution has the general form above
 To obtain the time evolution of the amplitudes we expand around weak polarization, i.e.,
 slowly varying amplitudes: multiscale approach

$$\mathbf{E} \simeq \text{Re} \left\{ \sum_n \left[\mathbf{E}_n^{(0)} + \eta \mathbf{E}_n^{(1)} + \dots \right] \exp(-i\omega_n t) \right\}$$

$$\mathbf{H} \simeq \text{Re} \left\{ \sum_n \left[\mathbf{H}_n^{(0)} + \eta \mathbf{H}_n^{(1)} + \dots \right] \exp(-i\omega_n t) \right\}$$

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$$\mathbf{P}_{NL} = \text{Re} \left[\sum_n \mathbf{P}_n(t_1, t_2, \dots) \exp(-i\omega_n t) \right]$$

$$t_n = \eta^n t$$

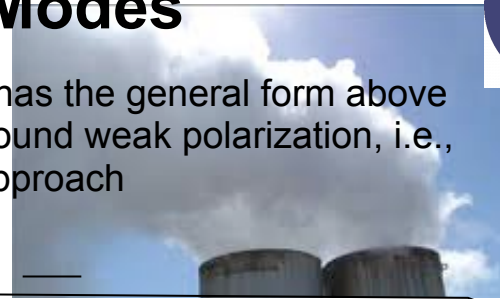
$$\simeq \text{Re} \left[\sum_n \left(\mathbf{P}_n^{(0)} + \eta \mathbf{P}_n^{(1)} + \dots \right) \exp(-i\omega_n t) \right]$$

$$\mathbf{J} = \partial_t \mathbf{P}_{NL} = \text{Re} \left[\sum_n \mathbf{J}_n(t_1, t_2, \dots) \exp(-i\omega_n t) \right]$$

$$\simeq \text{Re} \left[\sum_n \left(\mathbf{J}_n^{(0)} + \eta \mathbf{J}_n^{(1)} + \dots \right) \exp(-i\omega_n t) \right]$$

$$\mathbf{J}_n^{(0)} = -i\omega_n \mathbf{P}_n^{(0)}$$

If \mathbf{P}_{NL} is not zero, amplitudes depend on time and solution has the general form above
 To obtain the time evolution of the amplitudes we expand around weak polarization, i.e., slowly varying amplitudes: multi-scale approach



$$\mathbf{J} = \partial_t \mathbf{P}_{NL} = \text{Re} \left\{ \sum_n \left[\mathbf{E}_n^{(0)} + \eta \mathbf{E}_n^{(1)} + \dots \right] \exp(-i\omega_n t) \right\}$$

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$$\mathbf{H} \simeq \text{Re} \left\{ \sum_n \left[\mathbf{H}_n^{(0)} + \eta \mathbf{H}_n^{(1)} + \dots \right] \exp(-i\omega_n t) \right\}$$

$$\mathbf{P}_{NL} = \text{Re} \left[\sum_n \mathbf{P}_n(t_1, t_2, \dots) \exp(-i\omega_n t) \right]$$

$$\simeq \text{Re} \left[\sum_n \left(\mathbf{P}_n^{(0)} + \eta \mathbf{P}_n^{(1)} + \dots \right) \exp(-i\omega_n t) \right]$$

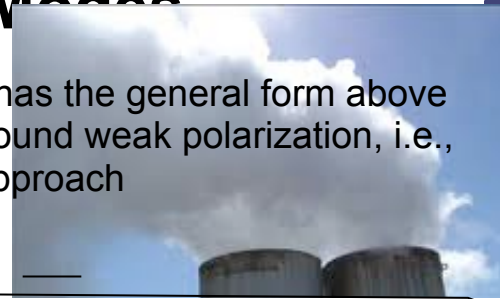
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$$\nabla \times \mathbf{E} = -\mu_0 \partial_t \mathbf{H}$$

$$t_n = \eta^n t$$

If **PNL** is not zero, amplitudes depend on time and solution has the general form above
 To obtain the time evolution of the amplitudes we expand around weak polarization, i.e., slowly varying amplitudes: multi-scale approach



$$\mathbf{J} = \partial_t \mathbf{P}_{NL} = \text{Re} \left\{ \sum_n \left[\mathbf{E}_n^{(0)} + \eta \mathbf{E}_n^{(1)} + \dots \right] \exp(-i\omega_n t) \right\}$$

$$\mathbf{H} \simeq \text{Re} \left\{ \sum_n \left[\mathbf{H}_n^{(0)} + \eta \mathbf{H}_n^{(1)} + \dots \right] \exp(-i\omega_n t) \right\}$$

$$\mathbf{P}_{NL} = \text{Re} \left[\sum_n \mathbf{P}_n(t_1, t_2, \dots) \exp(-i\omega_n t) \right]$$

$$\simeq \text{Re} \left[\sum_n \left(\mathbf{P}_n^{(0)} + \eta \mathbf{P}_n^{(1)} + \dots \right) \exp(-i\omega_n t) \right]$$

$$\mathbf{J}_n^{(0)} = -i\omega_n \mathbf{P}_n^{(0)}$$

$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} = \epsilon_0 n^2(\mathbf{r}) \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}_{NL}}{\partial t}$$

$$\nabla \times \mathbf{E} = -\mu_0 \partial_t \mathbf{H}$$

$$\mathcal{L} = \begin{pmatrix} 0 & i\nabla \times \\ -i\nabla \times & 0 \end{pmatrix}$$

$$\mathcal{M} = \begin{pmatrix} \epsilon_0 n^2(\mathbf{r}) & 0 \\ 0 & \mu_0 \end{pmatrix}$$

$$\mathcal{F}_n^{(k)} = \begin{pmatrix} \mathbf{E}_n^{(k)} \\ \mathbf{H}_n^{(k)} \end{pmatrix}$$

$$\mathcal{L} \mathcal{F}_n^{(1)} - \omega_n \mathcal{M} \mathcal{F}_n^{(1)} = \mathcal{B}_n$$

$$\mathcal{B}_n = \begin{pmatrix} i\epsilon_0 n^2(\mathbf{r}) \frac{da_n}{dt_1} \mathbf{E}_n^{(0)} + i\mathbf{J}_s^{(0)} \\ i\mu_0 \frac{da_n}{dt_1} \mathbf{H}_n^{(0)} \end{pmatrix}$$

$$t_n = \eta^n t$$

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$$\mathcal{L} = \begin{pmatrix} 0 & i\nabla \times \\ -i\nabla \times & 0 \end{pmatrix} \quad \mathcal{M} = \begin{pmatrix} \epsilon_0 n^2(\mathbf{r}) & 0 \\ 0 & \mu_0 \end{pmatrix} \quad \mathcal{F}_n^{(k)} = \begin{pmatrix} \mathbf{E}_n^{(k)} \\ \mathbf{H}_n^{(k)} \end{pmatrix}$$

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Fredholm theorem: orthogonality with the kernel (0) solution:

$$\mathcal{F}_n : \quad (\mathcal{F}_n, \mathcal{B}_n) = \int_V \mathcal{F}_n^* \cdot \mathcal{B}_n dV = 0$$

$$\mathbf{J}_n^{(0)} = -i\omega_n \mathbf{P}_n^{(0)}$$

yields

$$\frac{da_n(t)}{dt} = i \frac{\omega_n}{4} \int_V \mathbf{E}_n^*(\mathbf{r}) \cdot \mathbf{P}_n(\mathbf{r}) dV$$

Master equation

$$\frac{da_m(t)}{dt} = \frac{i\sqrt{\omega_m}}{4} \int_V \mathbf{E}_m^*(\mathbf{r}) \cdot \mathbf{P}_m(\mathbf{r}) d^3\mathbf{r}$$

$\alpha = x, y, z$
 $m = 1, \dots, N$

$$\mathbf{P}_{NL}(\mathbf{r}) = \text{Re} \left[\sum_m \sqrt{\omega_m} a_m(t) \mathbf{P}_m(\mathbf{r}) e^{-i\omega_m t} \right]$$

$$P_m^\alpha = \sum_{\omega_l} \chi_{\alpha\beta}^{(1)}(\omega_l, \omega_m; \mathbf{r}) E_l^\beta(\mathbf{r}) \sqrt{\omega_l} a_l$$

$$+ \sum_{\omega_j + \omega_k = \omega_l + \omega_m} \chi_{\alpha\beta\gamma\delta}^{(3)}(\omega_m; \omega_k, \omega_l, -\omega_j; \mathbf{r}) E_j^\beta(\mathbf{r}) E_k^\gamma(\mathbf{r}) E_l^\delta(\mathbf{r}) \sqrt{\omega_j \omega_k \omega_l} a_j^* a_k a_l$$

Optical response

Master equation

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Optical response

Langevin equation

$$\frac{da_m(t)}{dt} = \sum_l \gamma_{lm} a_l + \sum_{[jkl]} g_{jklm} a_j^* a_k a_l + \Gamma_m(t)$$

$$\langle \Gamma_j(t) \Gamma_k^*(t') \rangle = 2k_B T \delta_{jk} \delta(t - t')$$

Spontaneous emission

Master equation

$$\frac{da_m(t)}{dt} = \frac{i\sqrt{\omega_m}}{4} \int_V \mathbf{E}_m^*(\mathbf{r}) \cdot \mathbf{P}_m(\mathbf{r}) d^3\mathbf{r}$$

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$$\omega_j - \omega_k = \omega_l - \omega_m$$

Mode-locking condition

$$\langle \Gamma_j(t) \Gamma_k^*(t') \rangle = 2k_B T \delta_{jk} \delta(t - t')$$

Spontaneous emission

$$\omega_j - \omega_k - \omega_l + \omega_m < \delta_\omega \quad \text{spectral line width}$$

Master equation

$$\frac{da_m(t)}{dt} = \frac{i\sqrt{\omega_m}}{4} \int_V \mathbf{E}_m^*(\mathbf{r}) \cdot \mathbf{P}_m(\mathbf{r}) d^3\mathbf{r}$$

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Optical response

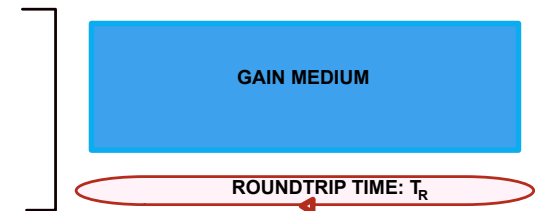
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Multimode mode-locking laser

- Once cavity is known, fields localization is known.
- Optical susceptibility can be computed in gas lasers (classical em field approx.) with two and three levels (and more..).
- To derive the optical susceptibility in solid state lasers is more complicated: em field must be quantized. No known (by myself!) results for multimode lasing.



Master equation

$$\frac{da_m(t)}{dt} = \frac{i\sqrt{\omega_m}}{4} \int_V \mathbf{E}_m^*(\mathbf{r}) \cdot \mathbf{P}_m(\mathbf{r}) d^3\mathbf{r}$$

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Optical response

$$\frac{da_m(t)}{dt} = \sum_l \gamma_{lm} a_l + \sum_{[jkl]} g_{jklm} a_j^* a_k a_l + \Gamma_m(t)$$

Spontaneous emission

Langevin equation

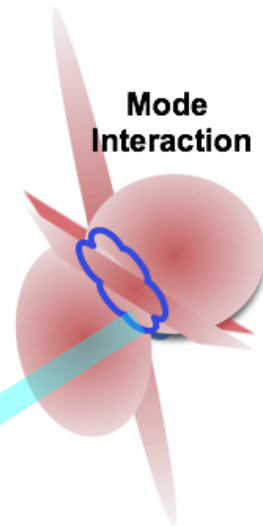
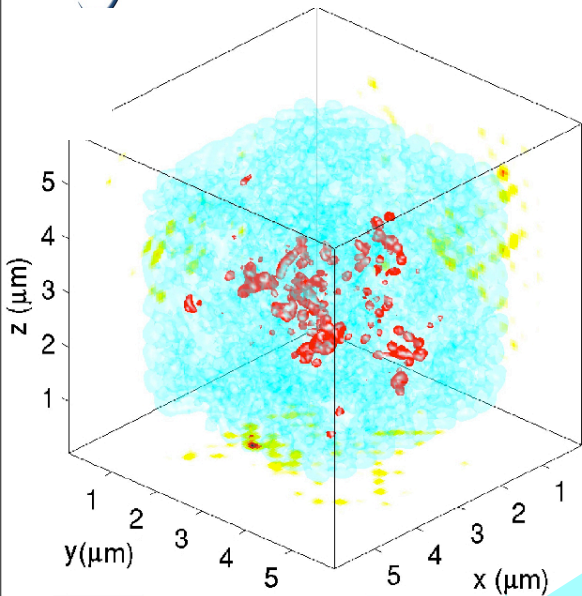
$$\langle \Gamma_j(t) \Gamma_k^*(t') \rangle = 2k_B T \delta_{jk} \delta(t - t')$$

This generically holds for light modes interacting in a non-linearly polarized medium

What are the specific features of the modes of a Random Laser?

LOCALIZED MODES

S. Gentilini *et al.*, Opt. Lett **34**, 130 (2009).



RANDOMNESS

Cavity-less light amplification: modes are amplified by scattering through randomly placed dielectric particles. This implies **random spatial distribution of modes**, and induces **random susceptibility**.

Mode spatial overlaps modulated by non-linear susceptibility yield **quenched interactions** with (so far) **unknown probability distributions**

**DISORDERED
DISTRIBUTED
INTERACTION
COEFFICIENT**

$$g_{jklm} = i \frac{\sqrt{\omega_j \omega_k \omega_l \omega_m}}{2} \int_V \chi_{\alpha, \beta, \gamma, \delta}^{(3)}(\omega_j, \omega_k, \omega_l, \omega_m) E_m^\alpha(\mathbf{r}) E_j^\beta(\mathbf{r}) E_k^\gamma(\mathbf{r}) E_l^\delta(\mathbf{r}) d^3 \mathbf{r}$$

NONLINEAR RANDOM SUSCEPTIBILITY
α COMPONENT OF EM FIELD IN MODE m

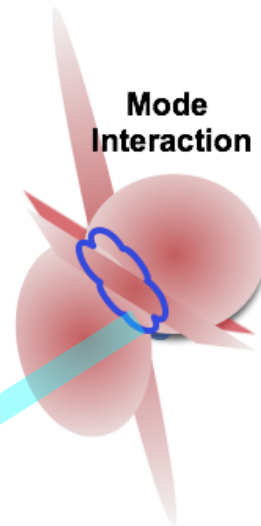
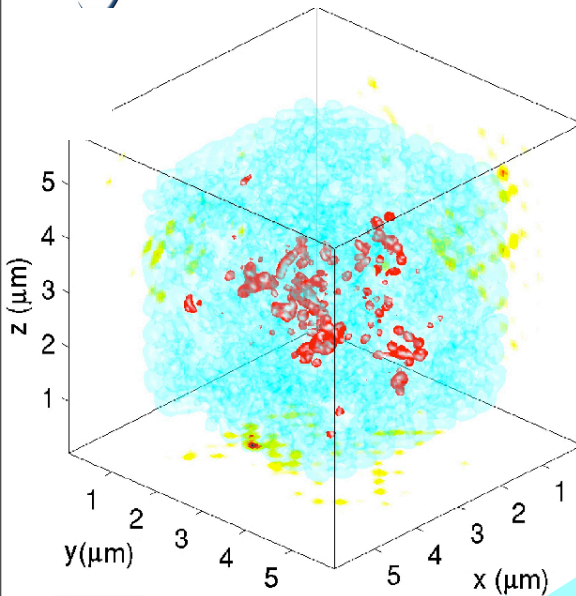
$$\frac{da_m(t)}{dt} = \sum_l \gamma_{lm} a_l + \sum_{[jkl]} g_{jklm} a_j^* a_k a_l + \Gamma_m(t)$$

NON-LINEARITY + RANDOMNESS

$$\langle \Gamma_j(t) \Gamma_k^*(t') \rangle = 2k_B T \delta_{jk} \delta(t - t')$$

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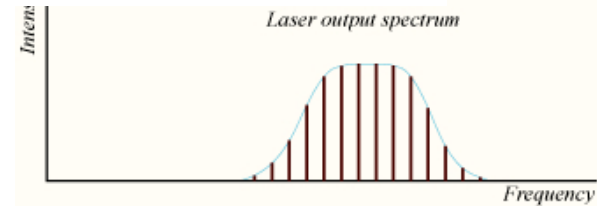
NON-LINEARITY + RANDOMNESS

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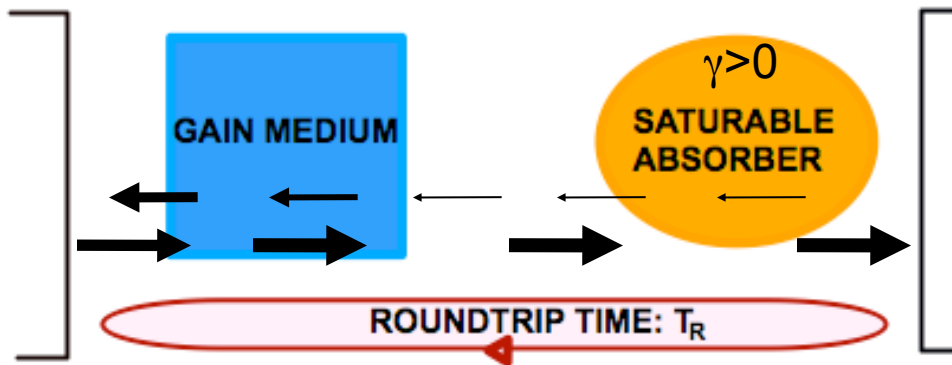
Physical meaning of coefficients in master equation?

$$\frac{da_m(t)}{dt} = \sum_l \gamma_{lm} a_l + \sum_{[jkl]} g_{jklm} a_j^* a_k a_l + \Gamma_m(t)$$

In absence of disorder the Master equation is the one of mode-locking lasers



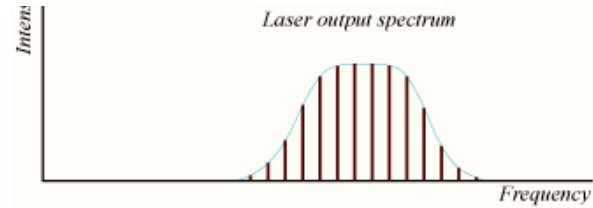
$$\frac{\partial a_m}{\partial t} \equiv \dot{a}_m = [g_m - \ell_m + iD_m] a_m + (\gamma - i\delta) \sum_{\omega_j + \omega_k - \omega_l = \omega_m} a_j^* a_k a_l + \Gamma_m(t)$$



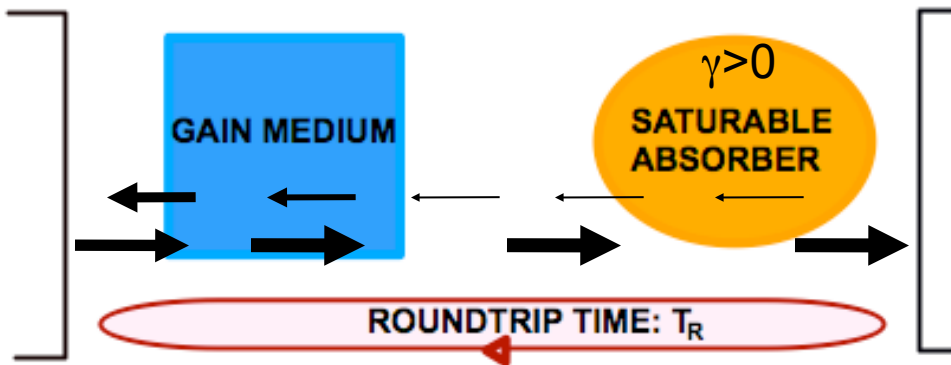
HA Haus, *Waves and Fields in Optoelectronics*, 1984

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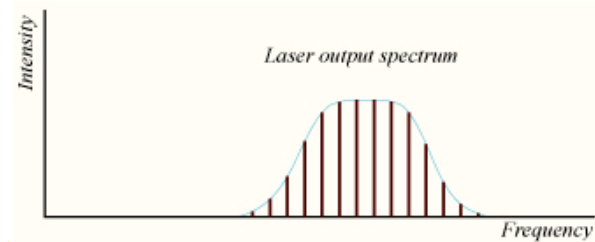
$$\frac{\partial a_m}{\partial t} \equiv \dot{a}_m = \overset{\text{GAIN}}{[g_m - \ell_m + iD_m]} a_m + \overset{\text{LOSS}}{(\gamma - i\delta)} \sum_{\omega_j + \omega_k - \omega_l = \omega_m} \overset{\text{GROUP VELOCITY DISPERSION}}{a_j^* a_k a_l} + \overset{\text{SATURABLE ABSORBER}}{\Gamma_m(t)} \overset{\text{KERR LENS EFFECT}}{+ \Gamma_m(t)} \overset{\text{SPONTANEOUS EMISSION}}{+ \Gamma_m(t)}$$



HA Haus, *Waves and Fields in Optoelectronics*, 1984

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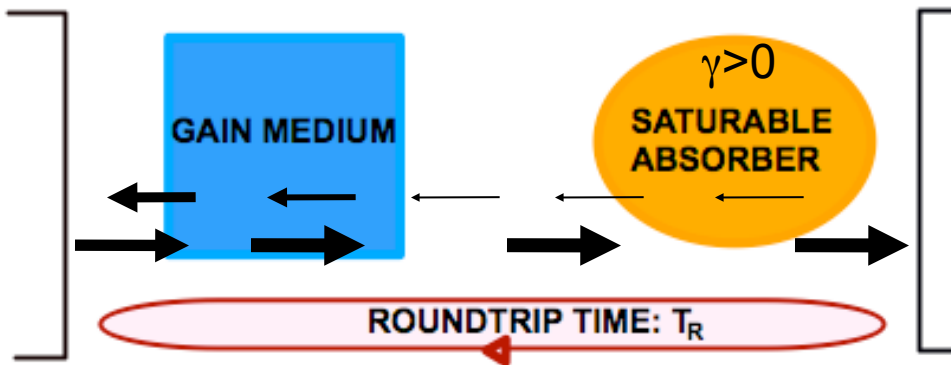
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SPONTANEOUS EMISSION

HA Haus, *Waves and Fields in Optoelectronics*, 1984



$$\gamma \sim \text{Re} [g]$$

From Langevin to Hamilton

- $$\frac{da_m(t)}{dt} = \sum_l \gamma_{lm} a_l + \sum_{[jkl]} g_{jklm} a_j^* a_k a_l + \Gamma_m(t) \longrightarrow \dot{a}_j = -\frac{\partial \mathcal{H}}{\partial a_j^*} + \Gamma_j$$

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From instantaneous EM energy of the non-linear (closed) cavity to Hamiltonian

- $$\mathcal{E} = \frac{1}{2} \int [\mathbf{D}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r}) + \mu_0 |\mathbf{H}(\mathbf{r})|^2] dV = \sum_m \mathcal{N}_m \omega_m |a_m(t)|^2 + \frac{1}{2} \int \mathbf{E}(\mathbf{r}) \cdot \mathbf{P}_{NL}(\mathbf{r}) dV$$

$$\longrightarrow \mathcal{H} = -\frac{1}{2} \left\langle \int \mathbf{E}(\mathbf{r}) \cdot \mathbf{P}_{NL}(\mathbf{r}) dV \right\rangle_{\text{optical cycle}}$$

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Hamiltonian description:

$$\mathcal{H}[\{a_j\}] = -\text{Re} \left[\sum_{jk} \gamma_{jk} a_j a_k^* + \sum_{[jklm]} g_{jklm} a_j a_k^* a_l a_m^* \right]$$

Localized mode interaction mediated by radiating modes

Non-linear localized mode-coupling expressed by 4-body interaction between amplitudes

with a global energy constraint

$$\mathcal{E} = \sum_k \omega_k |a_k|^2$$

Mean-Field model with Gaussian distribution of couplings:

2+4 spherical spin-glass [A Crisanti & LL, PRL 04, PRB 06, PRB 07, NPB 13]

Topology? Light modes Network?

Hamiltonian description:

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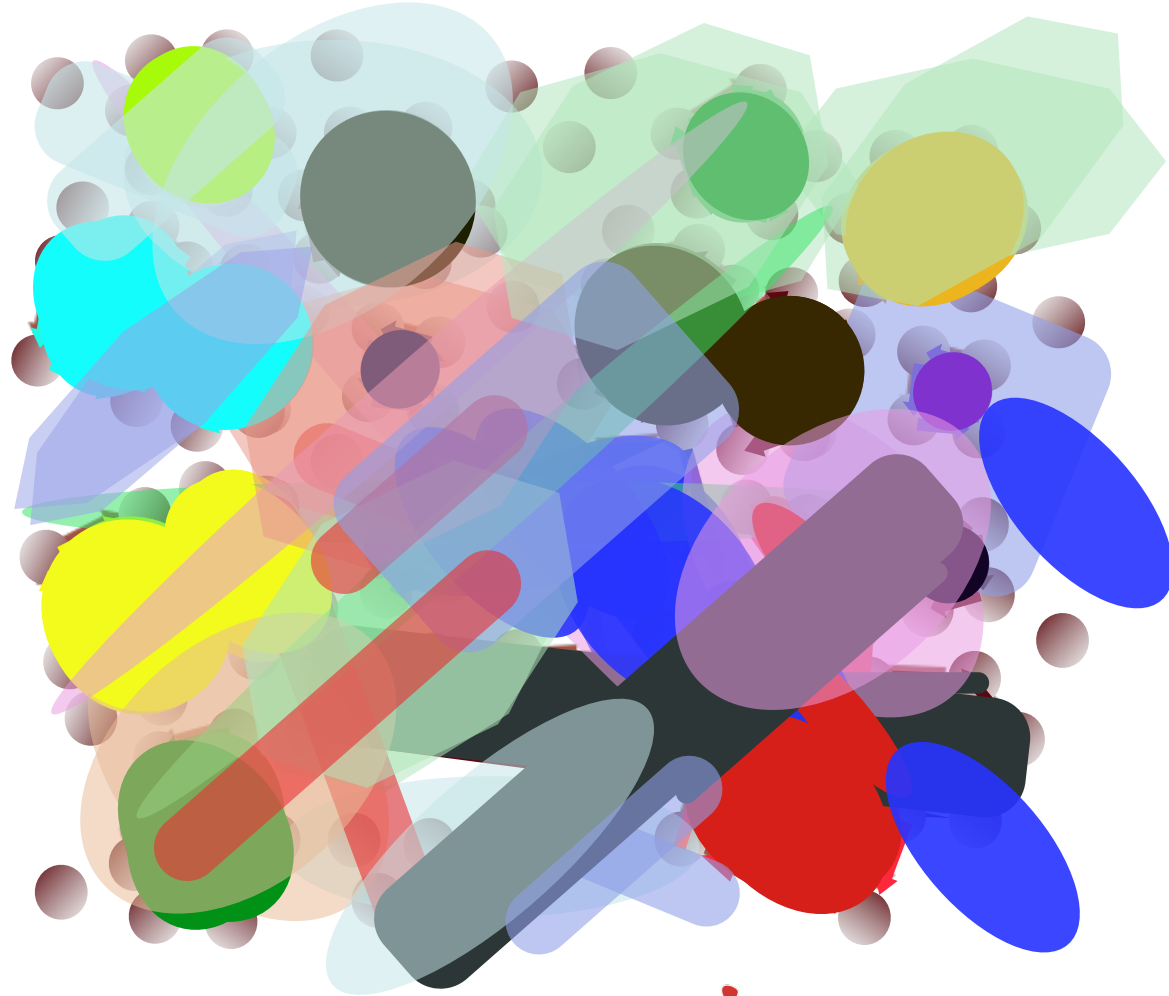
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Modeling random laser modes in space:
the electromagnetic field of each localized light mode of
frequency ω is non-zero in a given region of space \mathbf{r} .



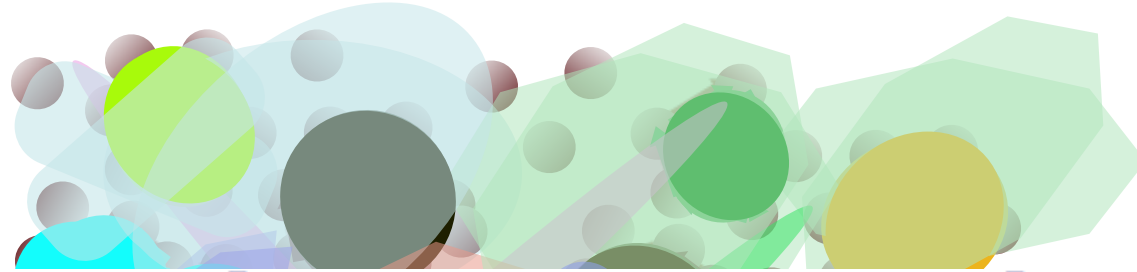
Modes interaction depends on their spatial overlap (4-uple).



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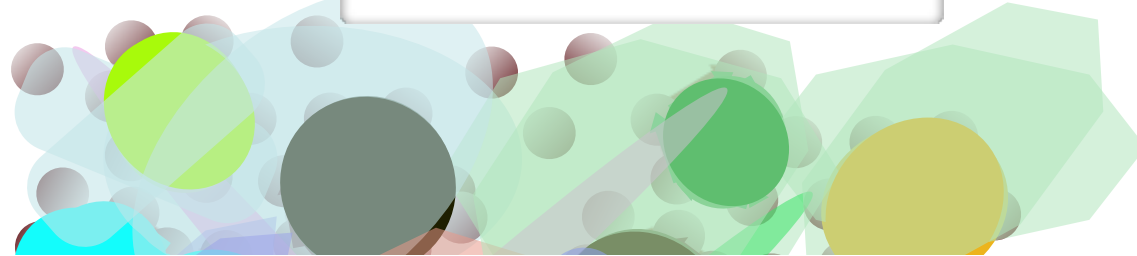


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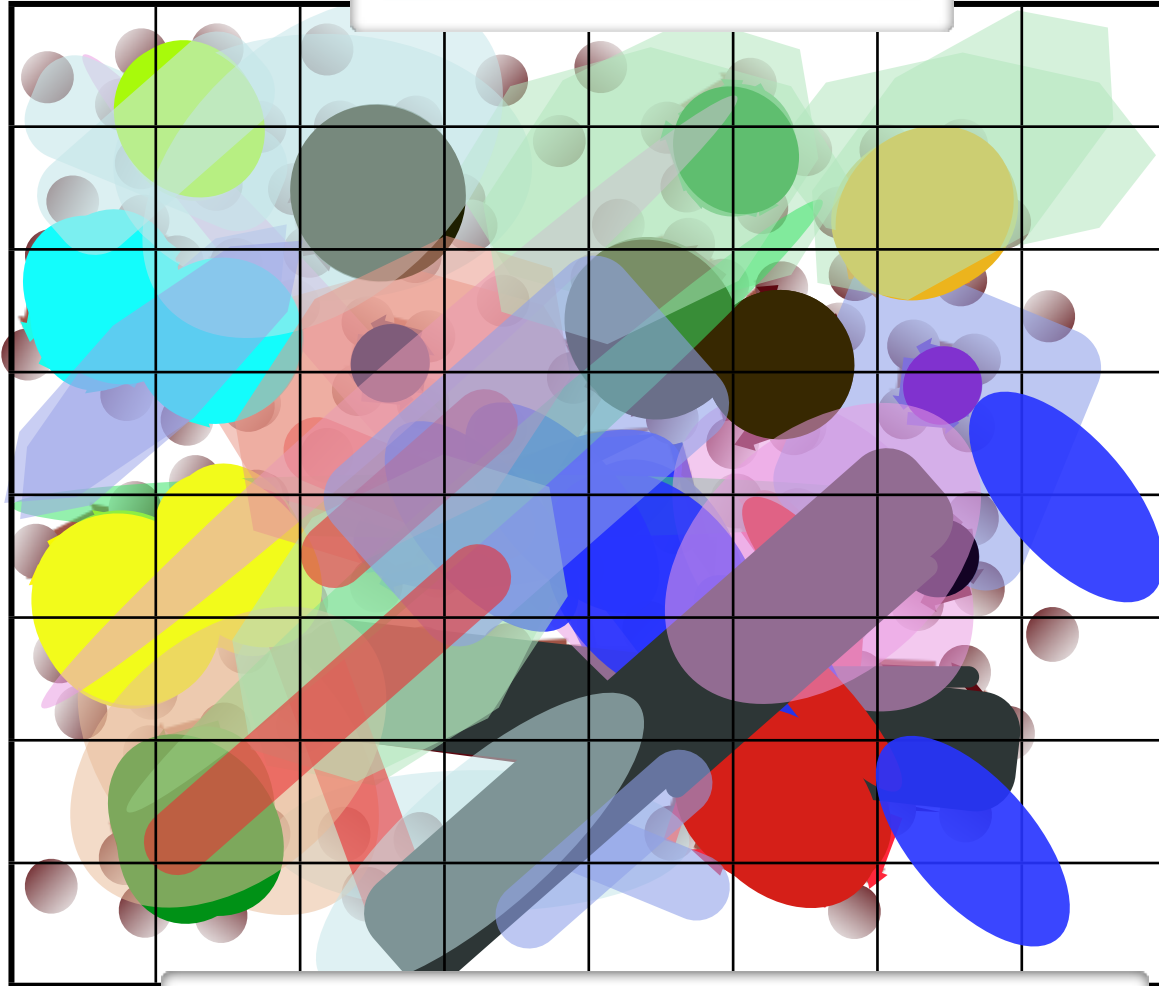
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Modeling random laser modes in space:
 localized modes can be **coarse-grained** as nodes of a graph with links to other nodes if their spatial overlap is non-zero and the **mode locking condition** is satisfied.

$$\omega_j - \omega_k = \omega_l - \omega_m$$

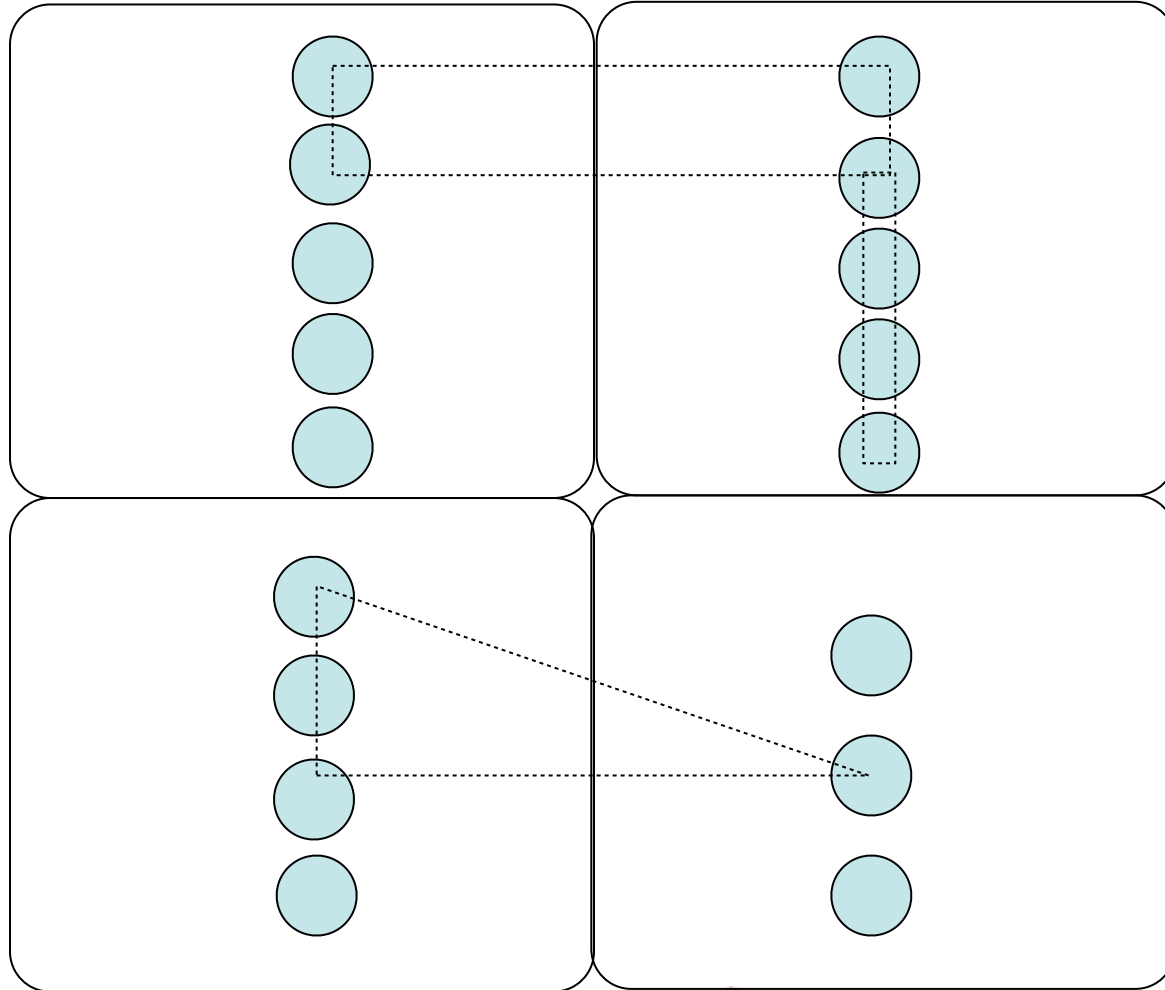


$$g_{jklm} \propto \int_V \chi_{\alpha,\beta,\gamma,\delta}^{(3)}(\omega_j, \omega_k, \omega_l, \omega_m) E_m^\alpha(\mathbf{r}) E_j^\beta(\mathbf{r}) E_k^\gamma(\mathbf{r}) E_l^\delta(\mathbf{r}) d^3\mathbf{r}$$

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Modeling random laser modes in space:
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$$\mathcal{H}\{a_j\} = -\text{Re} \left[\sum_{jk} \gamma_{jk} a_j a_k^* + \sum_{[jklm]} g_{jklm} a_j a_k^* a_l a_m^* \right]$$



M modes
 in each
 cell

Modeling random laser modes in space:
localized modes can be coarse-grained as nodes of a graph with links to other nodes if their spatial overlap is non-zero and the mode-locking condition is satisfied.

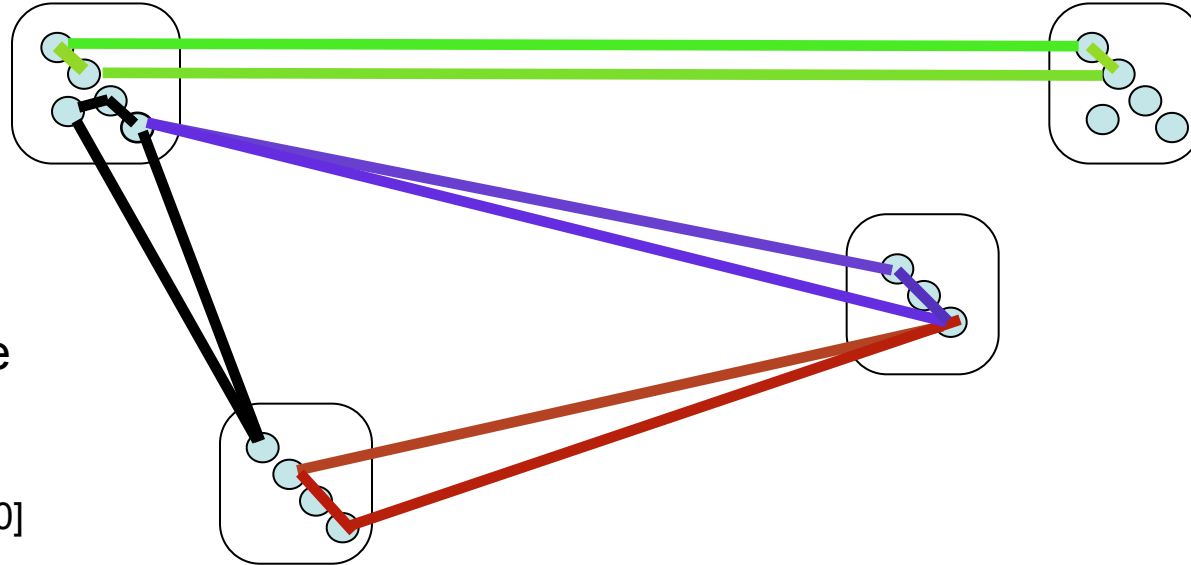
M modes in each cell.

A cell is a node in a network.

4-mode but 2-node interaction

$\sim(M,p)$ model

[Caltagirone et al. PRB10]



Once all non-zero “4-modes” couplings have been selected:
network of cells/nodes, each one containing M modes.

Selection “tools”:

$$\omega_j - \omega_k = \omega_l - \omega_m$$



$$g_{jklm} \propto \int_V \chi_{\alpha,\beta,\gamma,\delta}^{(3)}(\omega_j, \omega_k, \omega_l, \omega_m) E_m^\alpha(\mathbf{r}) E_j^\beta(\mathbf{r}) E_k^\gamma(\mathbf{r}) E_l^\delta(\mathbf{r}) d^3\mathbf{r}$$

Hamiltonian description:

$$\mathcal{H}[\{a_j\}] = -\text{Re} \left[\sum_{jk} \gamma_{jk} a_j a_k^* + \sum_{[jklm]} g_{jklm} a_j a_k^* a_l a_m^* \right]$$

Localized mode interaction mediated by radiating modes

Non-linear localized mode-coupling expressed by 4-body interaction between amplitudes

with a global energy constraint

$$\mathcal{E} = \sum_k |a_k|^2$$

Statistical Mechanics allows to model different kinds of random lasers characterized by:

- **degree of disorder** (ranging from almost standard lasers with an irreducible noise to completely random lasers),
- **extension of modes** localization,
- **geometry and dimension**, **interaction range**
- **pumping** intensity and mode-locked pulse length,
- role of **radiating modes** in modulating localized modes linear/'two-body' interaction (assumption of pure self-interaction),
- characteristic **times of magnitude and phase** of complex mode amplitudes (assumption of quenched amplitudes)

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Statistical Mechanics allows to model different kinds of random lasers characterized by:

• **degree of disorder**



**COUPLING DISTRIBUTION
PARAMETER VALUES (mean, variance)**

• **extension of modes** localization,
• **geometry and dim., interaction range**



**NETWORK/GRAPH
STRUCTURE**

• **pumping** intensity



TEMPERATURE

• role of **radiating modes** in modulating localized modes 'two-body' interaction



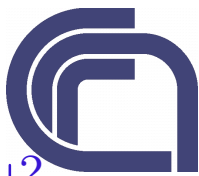
$$\sum_{jk} \gamma_{jk} a_j a_k^* \simeq \sum_j \gamma_j |a_j|^2$$

• characteristic **times of magnitude and phase** of complex mode amplitudes



$$a_j = A_j e^{i\phi_j}$$

SLOW / FAST



$$\mathcal{H}[\{a_j\}] = -\text{Re} \left[\sum_{jk} \gamma_{jk} a_j a_k^* + \sum_{[jklm]} g_{jklm} a_j a_k^* a_l a_m^* \right]$$

$$\mathcal{E} = \sum_k |a_k|^2$$

$$\mathcal{H}[\{a_j\}] \simeq -\sum_j \gamma_j |a_j|^2 - \text{Re} \left[\sum_{[jklm]} g_{jklm} a_j a_k^* a_l a_m^* \right]$$

$$\omega_j - \omega_k = \omega_m - \omega_l$$

Negligible non-local interaction between localized modes via radiation modes (closed cavity)

$$\mathcal{H}[\{A_j; \phi_j\}] \simeq -\sum_j \gamma_j A_j^2 - \sum_{[jklm]} A_j A_k A_l A_m \text{Re} \left[g_{jklm} e^{i(\phi_j - \phi_k + \phi_l - \phi_m)} \right]$$

Decoupled amplitude magnitude and phase



$$\mathcal{H}[\{a_j\}] = -\text{Re} \left[\sum_{jk} \gamma_{jk} a_j a_k^* + \sum_{[jklm]} g_{jklm} a_j a_k^* a_l a_m^* \right] \quad \mathcal{E} = \sum_k |a_k|^2$$

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Quenched approximation for amplitudes

$$\mathcal{H}[\{\phi_j\}] \simeq - \sum_{[j,k,l,m]} J_{jklm} \cos(\phi_j - \phi_k + \phi_l - \phi_m)$$

$$J_{jklm} \propto A_j A_k A_l A_m g_{jklm}$$



$$\mathcal{H}[\{a_j\}] = -\text{Re} \left[\sum_{jk} \gamma_{jk} a_j a_k^* + \sum_{[jklm]} g_{jklm} a_j a_k^* a_l a_m^* \right] \quad \mathcal{E} = \sum_k |a_k|^2$$

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Inferring g 's yields information about **light modes localization**

$$g_{jklm} \propto \int_V \chi_{\alpha,\beta,\gamma,\delta}^{(3)}(\omega_j, \omega_k, \omega_l, \omega_m) E_m^\alpha(\mathbf{r}) E_j^\beta(\mathbf{r}) E_k^\gamma(\mathbf{r}) E_l^\delta(\mathbf{r}) d^3 \mathbf{r}$$

mode coupling
in position (nodes)
and frequency

$$j \begin{cases} \omega_j \\ \mathbf{r}_j \end{cases}$$

**Can we
measure g 's?**

$$\mathcal{H}[\{a_j\}] = -\text{Re} \left[\sum_{jk} \gamma_{jk} a_j a_k^* + \sum_{[jklm]} g_{jklm} a_j a_k^* a_l a_m^* \right] \quad \mathcal{E} = \sum_k |a_k|^2$$

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Inferring g's yields information about light modes localization

$$C_{jklm}^{(4)} = \langle \cos(\phi_j - \phi_k + \phi_l - \phi_m) \rangle \quad j \begin{cases} \omega_j \\ \mathbf{r}_j \end{cases}$$

$$C_{jklm}^{(4)} = \langle \text{Re} [a_j a_k^* a_l a_m^*] \rangle \quad C_{jklm}^{(2)} = \langle \text{Re} [a_j a_k^*] \rangle$$

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Inferring g's yields information about light modes localization

Fitting graphical problem techniques can be applied/generalized, though:

Theory

- variables are continuous: XY (phases) or spherical (amplitudes) “spins”;
- quenched disorder is there;
- four point correlations have to be considered, besides two point.

$$C_{jklm}^{(4)} = \langle \cos(\phi_j - \phi_k + \phi_l - \phi_m) \rangle$$

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Experiments

- Intensities (mode magnitudes) are available versus frequency.
How many modes? From 10 to 10^5 , though refinement is finite in spectra (e.g., .3 nm) -> 100:1000 distinct frequencies can be usually appreciated;
- phases (vs. frequency) have not been measured yet: set up is ready, probing standard lasers and seeking high intensity random lasers (rhodamine+TiO₂ is not “energetic” enough).

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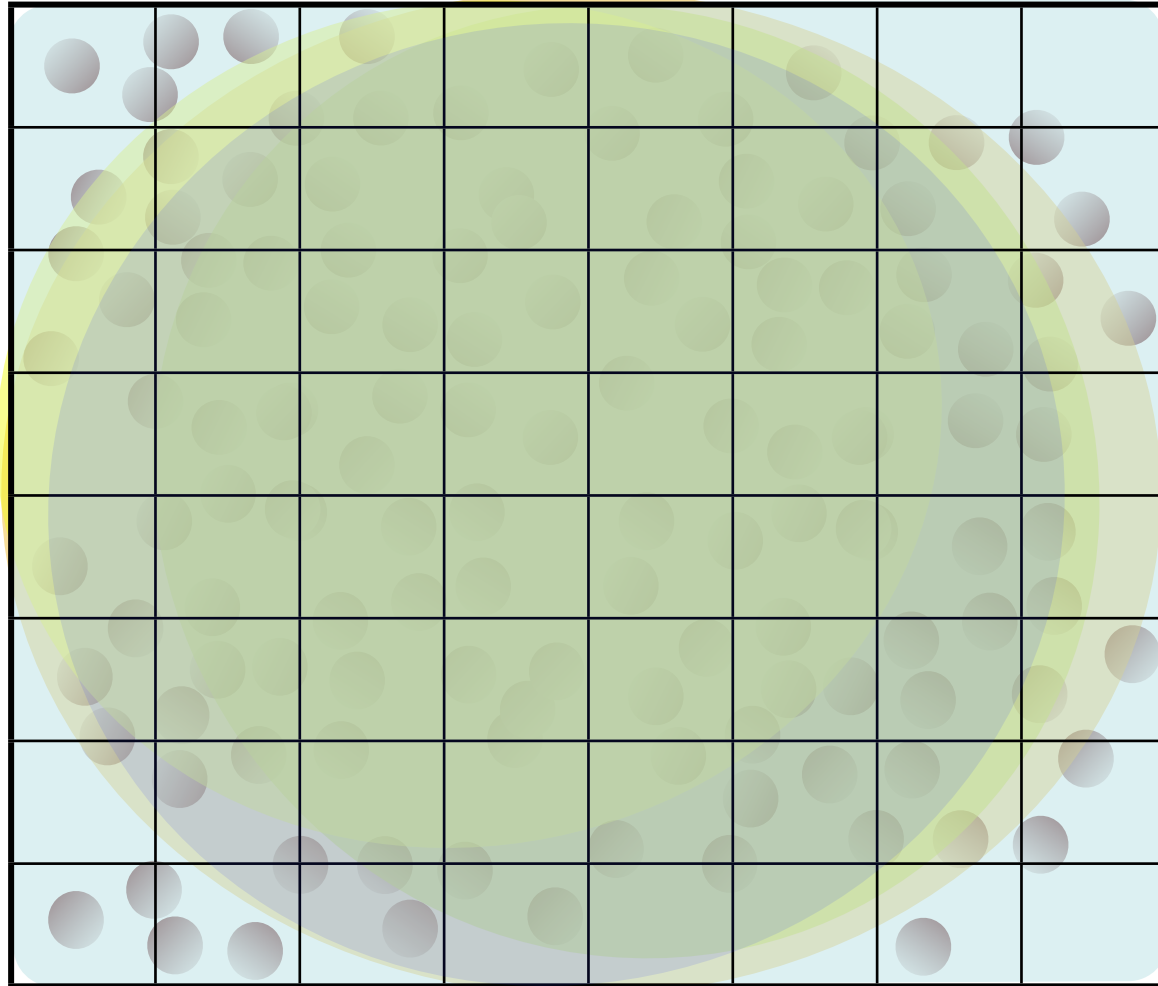
“Warm up” with:

- ordered multimode mode-locking laser;
- linearly interacting waves (not lasers): $C_{jk}^{(2)} = \langle \text{Re} [a_j a_k^*] \rangle$

Mean-field model for slow amplitudes



In some systems modes can be localized
but non-zero almost everywhere in the optically active medium

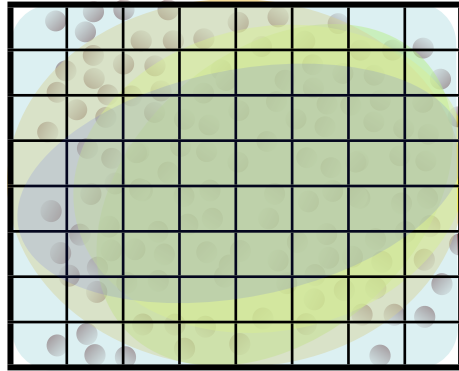


$$\mathcal{H}[\{A_j; \phi_j\}] \simeq - \sum_j \gamma_j A_j^2 - \sum_{[jklm]} A_j A_k A_l A_m \text{Re} \left[g_{jklm} e^{i(\phi_j - \phi_k + \phi_l - \phi_m)} \right]$$

Mean-field model for slow amplitudes



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Mode-locking condition

$$\omega_j - \omega_k = \omega_m - \omega_l$$

implies dilution:

~ Erdos-Renyi random graph [Tyagi]

Phase model

$$\mathcal{H}[\{A_j; \phi_j\}] \simeq - \sum_j \gamma_j A_j^2 - \sum_{[jklm]} A_j A_k A_l A_m \operatorname{Re} \left[g_{jklm} e^{i(\phi_j - \phi_k + \phi_l - \phi_m)} \right]$$

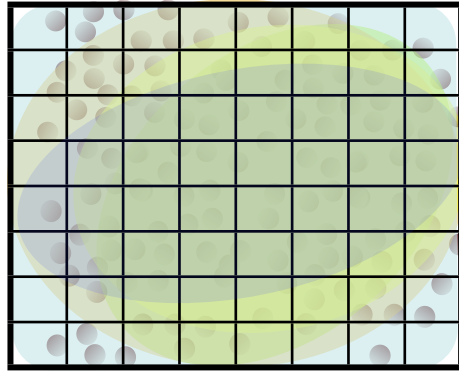
$$\mathcal{H}[\{A_j; \phi_j\}] \simeq -\mathcal{P}^2 \sum_{[jklm]} \operatorname{Re} \left[J_{jklm} e^{i(\phi_j - \phi_k + \phi_l - \phi_m)} \right]; \quad \mathcal{P} \propto \langle A^2 \rangle$$

$$J_{jklm} \equiv g_{jklm} A_j A_k A_l A_m / \mathcal{P}^2$$

Mean-field model for slow amplitudes



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Mode-locking condition

$$\omega_j - \omega_k = \omega_m - \omega_l$$

implies dilution: Erdos-Renyi/
Bethe lattice [Tyagi]

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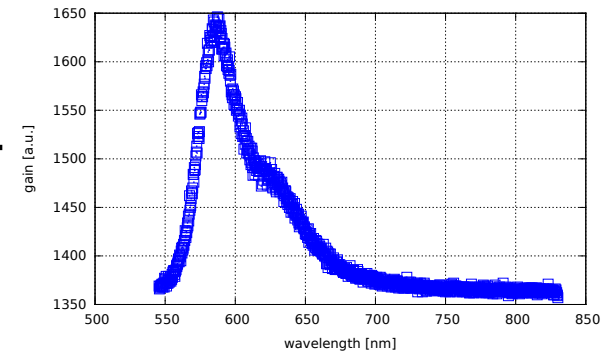
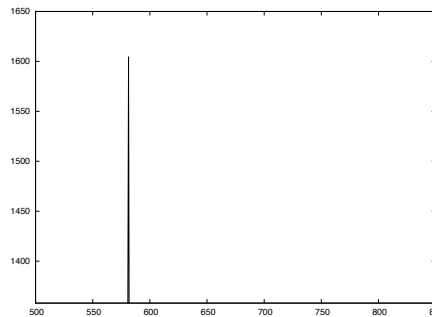
$$\mathcal{H}[\{A_j; \phi_j\}] \simeq -\mathcal{P}^2 \sum_{[jklm]} \operatorname{Re} [J_{jklm} e^{i(\phi_j - \phi_k + \phi_l - \phi_m)}]; \quad \mathcal{P} \propto \langle A^2 \rangle$$

$$J_{jklm} \equiv g_{jklm} A_j A_k A_l A_m / \mathcal{P}^2$$

Fully connected Mean-field approximation

Mode-locking always satisfied

$$\omega_j - \omega_k = \omega_m - \omega_l$$



$$\mathcal{H}[\{\phi_j\}] \simeq -\mathcal{P}^2 \sum_{j < k; l < m; k < l} J_{jklm} \cos(\phi_j - \phi_k + \phi_l - \phi_m)$$

$$\mathcal{H}[\{\phi_j\}] \simeq -\mathcal{P}^2 \sum_{j < k; l < m; k < l} J_{jklm} \cos(\phi_j - \phi_k + \phi_l - \phi_m)$$

Mean-field approximation:

- all 4-plets of phases interact with each other with small couplings (vanishing in the thermodynamic limit $N \rightarrow \infty$) - **GEOMETRY**
 - bandwidth narrows and the spectral distribution of angular frequencies is peaked around a value: $\omega_j \sim \omega_0$ for all modes $j=1, \dots, N$ - **MODE LOCKING**
- condition $\omega_j - \omega_k = \omega_l - \omega_m$ is always satisfied

Gaussian independent identically distributed interaction couplings

$$\langle J_{jklm} \rangle = J_0 / N^3$$

$$\langle (J_{jklm} - \langle J_{jklm} \rangle)^2 \rangle = \sigma_J / N^3$$

$$\mathcal{P}^2 = J_0 \frac{\langle A^2 \rangle^2}{k_B T_{\text{bath}}}$$

PARTITION FUNCTION:

$$Z_J = \int \prod_{j=1}^N d\phi_j e^{-\beta \mathcal{P}^2 \mathcal{H}[\{\phi_k\}]} = \int \prod_{j=1}^N d\phi_j e^{-\tilde{\beta} \mathcal{H}[\{\phi_k\}]}; \quad \tilde{\beta} = \mathcal{P}^2 \beta$$

Statistical mechanical properties, thermodynamic phases, order parameters,

$$\mathcal{H}[\{\phi_j\}] \simeq - \sum_{i < j; l < m; i < l} J_{ijklm} \cos(\phi_i - \phi_k + \phi_l - \phi_m)$$

Mean-field approximation

the role of inverse temperature is played by the square of the average stored energy per mode: "pumping rate"

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$$\tilde{\beta} = \mathcal{P}^2/k_B T$$

$$\mathcal{P} \propto \langle \mathcal{E} \rangle = \langle |a_j|^2 \rangle$$

$$R_J \equiv \sigma_J/J_0$$

$$\beta \Phi = -\frac{1}{N} \langle \log Z_J \rangle_J = \boxed{-\frac{1}{N} \lim_{n \rightarrow 0} \frac{\langle Z_J^n \rangle - 1}{n}}$$

Replica trick

In mean-field replica calculation sites interaction is eliminated and replicas interaction is introduced through the **overlap** order parameters

$$q_{ab} = \langle e^{i(\phi_a - \phi_b)} \rangle$$

$$r_{ab} = \langle e^{i(\phi_a + \phi_b)} \rangle$$

plus standard o.p.'s ("magnetizations")

$$m_a = \langle e^{i\phi_a} \rangle$$

$$\tilde{r}_a = \langle e^{2i\phi_a} \rangle$$

$$\beta\Phi = -\frac{1}{N} \langle \log Z_J \rangle_J = -\frac{1}{N} \lim_{n \rightarrow 0} \frac{\langle Z_J^n \rangle - 1}{n}$$

$$\langle Z \rangle \simeq \int \mathcal{D}\phi \int \prod_{jklm} dJ_{jklm} e^{-J_{jklm}^2} e^{\mathcal{H}[\phi]} \simeq \int \mathcal{D}\phi \int \prod_{jklm} dJ_{jklm} e^{-J_{jklm}^2} e^{J_{jklm} \cos(\dots\phi\dots)}$$

$$\begin{aligned} \langle Z^n \rangle &\simeq \prod_{a=1}^n \left[\int \mathcal{D}\phi^{(a)} \int \prod_{jklm} dJ_{jklm} e^{-J_{jklm}^2} e^{\mathcal{H}[\phi^{(a)}]} \right] \\ &\simeq \int \prod_{a=1}^n \mathcal{D}\phi^{(a)} \int \prod_{jklm} dJ_{jklm} e^{-J_{jklm}^2} e^{J_{jklm} \sum_{a=1}^n \cos(\dots\phi^{(a)}\dots)} \end{aligned}$$

All replicas enter in the same way: symmetry

Solving the thermodynamics replica symmetry is *spontaneously* broken: RSB theory

$$\mathcal{H}[\{\phi_j\}] \simeq - \sum_{i < j; l < m; i < l} J_{ijklm} \cos(\phi_i - \phi_k + \phi_l - \phi_m)$$

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Mean-field approximation

$$\tilde{\beta} = \mathcal{P}^2/k_B T$$

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Replica trick

It is “correct”, e.g., thermodynamically stable / self-consistent in the “**one-step Replica Symmetry Breaking**” scheme of computation

$$P(X) = m \delta(X - X_0) + (1 - m)\delta(X - X_1)$$

G.Parisi, 1979, 1980

“REPLICATED” FREE ENERGY

$$\begin{aligned} \bar{\beta}\Phi = & -\frac{\bar{\beta}R_J}{8} |\tilde{m}|^4 - \frac{\bar{\beta}^2}{32} \left[1 - (1 - m) (|q_1|^4 + |r_1|^4) - m (|q_0|^4 + |r_0|^4) + |r_d|^2 \right] \\ & - \text{Re} \left[\frac{1-m}{2} (\bar{\lambda}_1 q_1 + \bar{\mu}_1 r_1) + \frac{m}{2} (\bar{\lambda}_0 q_0 + \bar{\mu}_0 r_0) - \bar{\mu}_d r_d - \bar{\nu} \tilde{m} \right] \\ & + \frac{\lambda_1^R}{2} - \frac{1}{m} \int \mathcal{D}[0] \log \int \mathcal{D}[1] \left[\int_0^{2\pi} d\phi \exp \mathcal{L}(\phi; \mathbf{0}, \mathbf{1}) \right]^m \end{aligned}$$

$$\mathcal{H}[\{\phi_j\}] \simeq - \sum_{i < j; l < m; i < l} J_{ijklm} \cos(\phi_i - \phi_k + \phi_l - \phi_m)$$

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Mean-field approximation
Gaussian independent identically distributed interaction couplings

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LL et al PRL 09
C Conti and LL, PRB 11

$$\begin{aligned} \mathcal{L}(\phi; \mathbf{0}, \mathbf{1}) \equiv & \text{Re} \left\{ e^{i\phi} \left[\bar{\zeta}_1 \sqrt{\Delta\lambda^R - |\Delta\mu|} + \bar{\zeta}_0 \sqrt{\lambda_0^R - |\mu_0|} + x_1 \sqrt{2\Delta\bar{\mu}} + x_0 \sqrt{2\bar{\mu}_0} + \bar{\nu} \right] + e^{2i\phi} \left(\bar{\mu}_d - \frac{\bar{\mu}_1}{2} \right) \right\} \\ \Delta\lambda = \lambda_1 - \lambda_0 \quad \Delta\mu = \mu_1 - \mu_0 \quad \mathbf{0} = \{x_0, \zeta_0^R, \zeta_0^I\} \quad \mathbf{1} = \{x_1, \zeta_1^R, \zeta_1^I\} \end{aligned}$$

SELF-CONSISTENCY EQUATIONS

$$\begin{aligned} \lambda_{0,1} = \frac{\bar{\beta}^2}{4} (q_{0,1})^3 \quad ; \quad \mu_{0,1} = \frac{\bar{\beta}^2}{4} |r_{0,1}|^2 r_{0,1} \\ \mu_d = \frac{\bar{\beta}^2}{8} |r_d|^2 r_d \quad ; \quad \nu = \frac{\bar{\beta}R_J}{2} |\tilde{m}|^2 \tilde{m} \end{aligned}$$

$$\begin{aligned} q_1 = \langle \langle c_{\mathcal{L}}^2 \rangle_m \rangle_0 + \langle \langle s_{\mathcal{L}}^2 \rangle_m \rangle_0 \\ q_0 = \langle \langle c_{\mathcal{L}}^2 \rangle_m \rangle_0 + \langle \langle s_{\mathcal{L}}^2 \rangle_m \rangle_0 \\ r_1 = \langle \langle c_{\mathcal{L}}^2 \rangle_m \rangle_0 - \langle \langle s_{\mathcal{L}}^2 \rangle_m \rangle_0 + 2i \langle \langle c_{\mathcal{L}} s_{\mathcal{L}} \rangle_m \rangle_0 \\ r_0 = \langle \langle c_{\mathcal{L}}^2 \rangle_m \rangle_0 - \langle \langle s_{\mathcal{L}}^2 \rangle_m \rangle_0 + 2i \langle \langle c_{\mathcal{L}} \rangle_m \rangle_0 \langle \langle s_{\mathcal{L}} \rangle_m \rangle_0 \\ r_d = \langle \langle e^{2i\phi} \rangle_{\mathcal{L}} \rangle_m \rangle_0; \quad \tilde{m} = \langle \langle e^{i\phi} \rangle_{\mathcal{L}} \rangle_m \rangle_0 \end{aligned}$$

$$\mathcal{D}[\mathbf{a}] = \frac{1}{(2\pi)^{3/2}} \exp \left[-\frac{(x_a^2 + (\zeta_a^R)^2 + (\zeta_a^I)^2)}{2} \right]$$

$$\langle \langle \dots \rangle \rangle_m \equiv \frac{\int \mathcal{D}[\mathbf{1}] (\dots) \left[\int_0^{2\pi} d\phi e^{\mathcal{L}(\phi; \mathbf{0}, \mathbf{1})} \right]^m}{\int \mathcal{D}[\mathbf{1}] \left[\int_0^{2\pi} d\phi e^{\mathcal{L}(\phi; \mathbf{0}, \mathbf{1})} \right]^m}$$

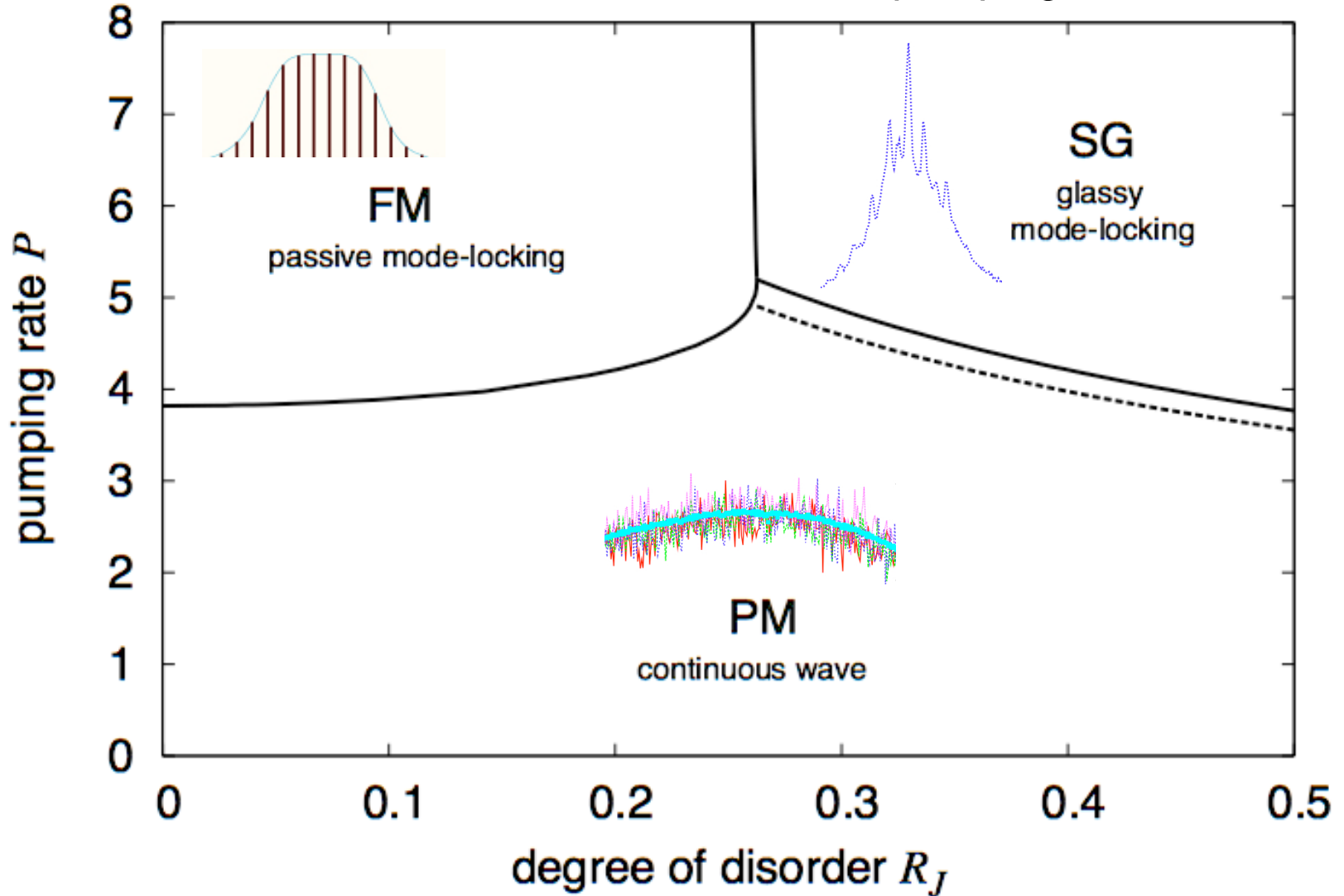
$$\langle \langle \dots \rangle \rangle_0 \equiv \int \mathcal{D}[\mathbf{0}] (\dots)$$

N.B.:
RSB parameter
 m still
undetermined

Mean-field approximation

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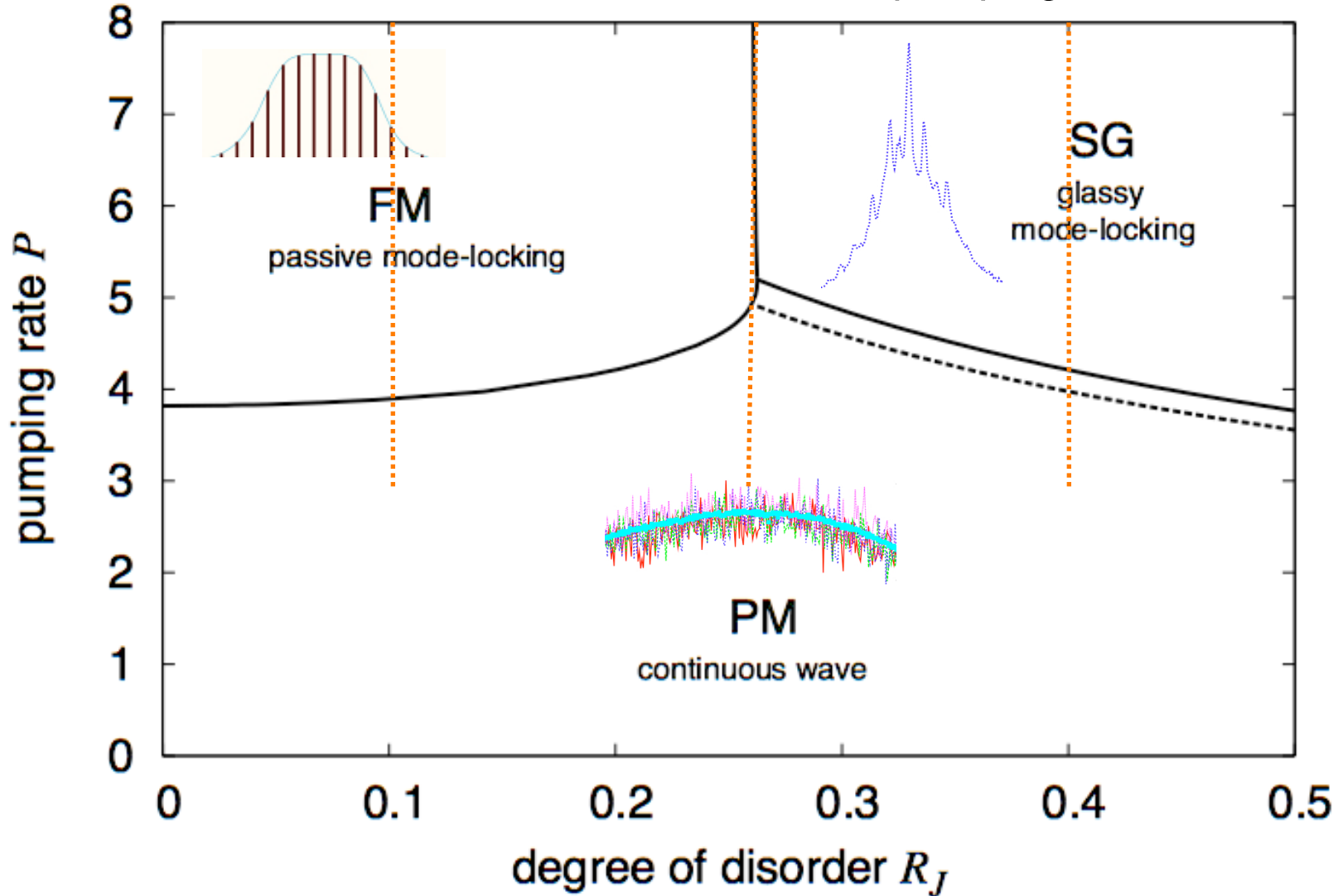
Photonics: pumping vs disorder



Mean-field approximation

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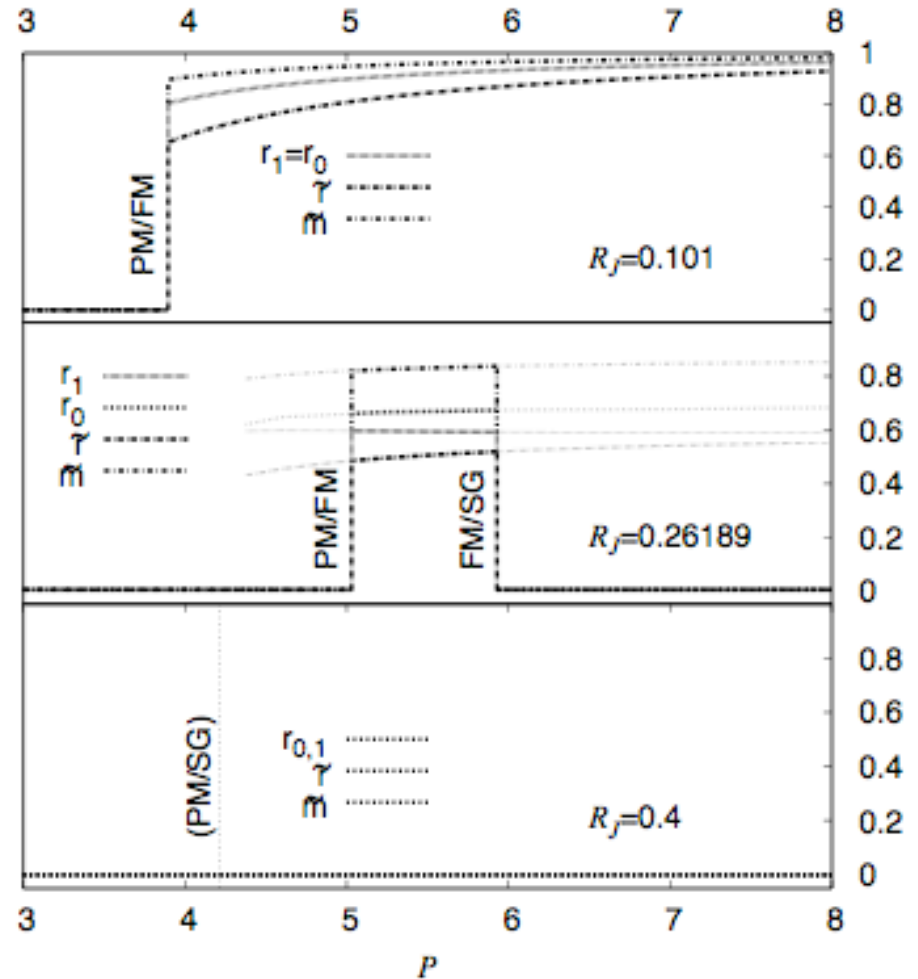
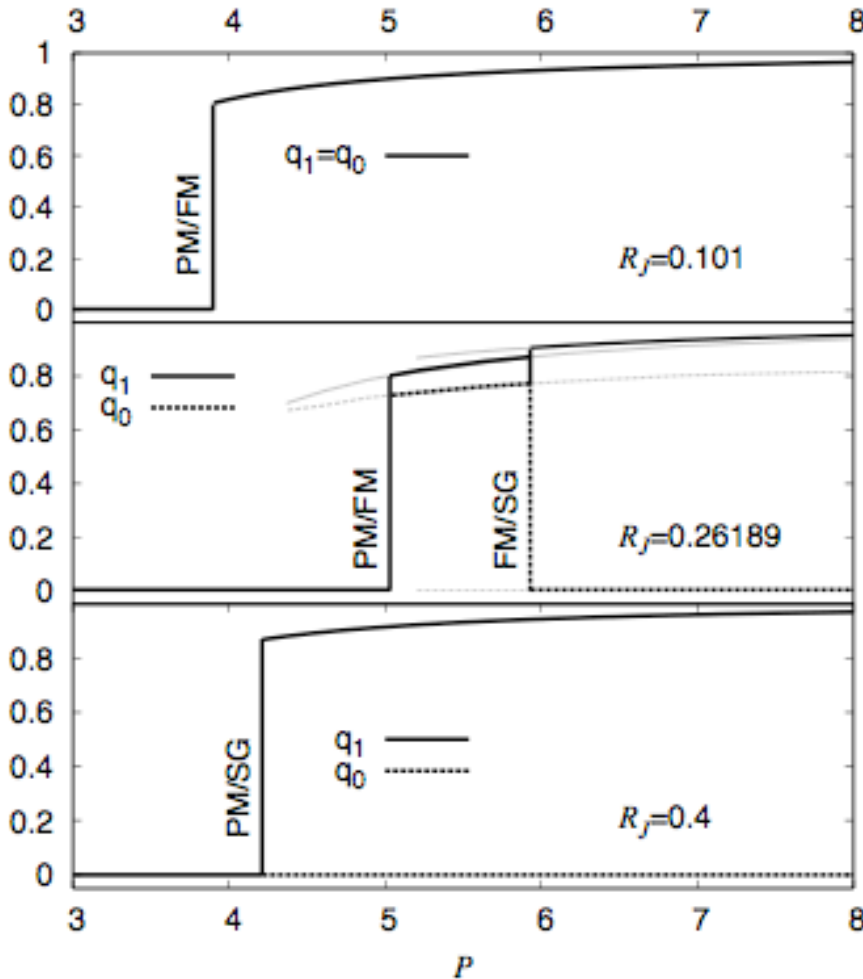
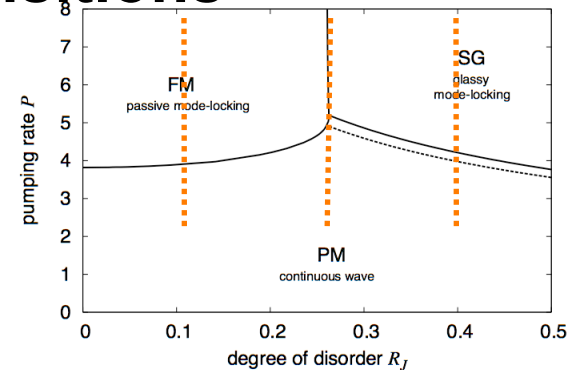
Photonics: pumping vs disorder



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Discontinuous order parameters



Outlook and Perspectives

So far....

- * Experimental spectral evidence compatible with the conjecture that **random laser** thermodynamics/dynamics is ruled by **complex free energy landscape**.
- * **Physical replicas** realization: in experiments the quenched disorder can be kept constant for different measurements.
- * RL behavior can be modeled by **Hamiltonian models with quenched disorder** and effects of tuning disorder strength can be predicted.
[control of disorder is fundamental in the physics and engineering of **nano-structured lasers** (and “cavity-less” lasers)].

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In progress....

- * “photonic spin-glasses” can model and reproduce: (i) **fluorescence/random laser transition** [Angelani et al. PRL06], (ii) **ordered/random laser transition** [LL et al PRL09, Conti & LL PRB11] (e.g., granulars [Folli et al. PRL12], nano crystal lasers,..) (iii) **random laser spectra**.

* Inference of non-linear couplings yield information about **modes localization** and optical response in random (and non-random) lasers. Graphical problems techniques.

- * Construction of **quantitative models** (real distribution of disorder, total energy profile with pumping, diluted interactions, finite dimensional structure, ...).
- * Applications to other wave problems in nonlinear random media (BEC in temperature, optical propagation at $T=0$) [Conti & LL PRB11].

Outlook and Perspectives

So far....

- * Experimental spectral evidence compatible with the conjecture that **random laser** thermodynamics/dynamics is ruled by **complex free energy landscape**.
- * **Physical replicas** realization: in experiments the quenched disorder can be kept constant for different measurements [Leonetti].
- * RL behavior can be modeled by **Hamiltonian models with quenched disorder** and effects of tuning disorder strength can be predicted [LL et al PRL09].
- [control of disorder is fundamental in the physics and engineering of **nano-structured lasers** (and “cavity-less” lasers)].

In progress....

- * “photonic spin-glasses” can model and reproduce: (i) **fluorescence/random laser transition** , (ii) **ordered/random laser transition** [LL et al PRL09, Conti & LL PRB11] (e.g., in granulars Folli et al. PRL12) (iii) **random laser spectra** [Antenucci, Tyagi].
- * Inference of non-linear couplings yield information about **modes localization** and optical response in random (and non-random) lasers. Graphical problems techniques [Tyagi].
- * Non-linear susceptibility computation for multimode gas and solid state lasers, small disorder effects [Marruzzo].
- * Construction of **quantitative models** (real distribution of disorder, total energy profile with pumping, diluted interactions, finite dimensional structure, ...) [Antenucci, Ibanez, Tyagi].
- * Applications to other wave problems in nonlinear random media (BEC in temperature, optical propagation at $T=0$) [Conti & LL PRB11].

Experimentally...

- * Experiments to measure **phases** rather than intensities [Ghofraniha] and directly access the dynamic variables would allow for correlation measure and theory test and inference.
- * Experiments in which the **degree of disorder is tuned changing the compactness of granular stochastic resonators** in random lasing materials [Folli et al. PRL12].