Spin-glass photonics: a statistical mechanical theory for lasing in random media

Luca Leuzzi

Inst. Chem. Phys. Processes, IPCF - National Research Council, CNR

@CNR.IT

Fabrizio Antenucci, Andrea Crisanti, Alessia Marruzzo
La Sapienza University, Rome

Miguel Ibanez Berganza, Payal Tyagi
IPCF-CNR, Rome

Claudio Conti, Andrea Puglisi
ISC-CNR, Rome

Viola Folli, Neda Ghofraniha, Marco Leonetti
IPCF-CNR, Rome

SEVENTH FRAMEWORK PROGRAMME
MARIE CURIE ACTIONS
NETADIS Statistical Physics Approaches to Networks Across Disciplines
PRIN
FIRB Fondo per gli Investimenti della Ricerca di Base
FUTURO IN RICERCA

Laser

- 1953-1955: **Charles H. Townes, Nikolay Basov, Aleksandr Prokhorov**: Microwave Amplification by Stimulated Emission of Radiation – MASER. They implemented continuous output, gain media with multienergy level atoms, optical pumping for population inversion.

Nobel Prize in Physics 1964, "for fundamental work in the field of quantum electronics, which has led to the construction of oscillators and amplifiers based on the maser–laser principle"

- 1958: *Infrared and Optical Masers*, Arthur L. Schawlow and Charles H. Townes

Optical Maser = **Laser**

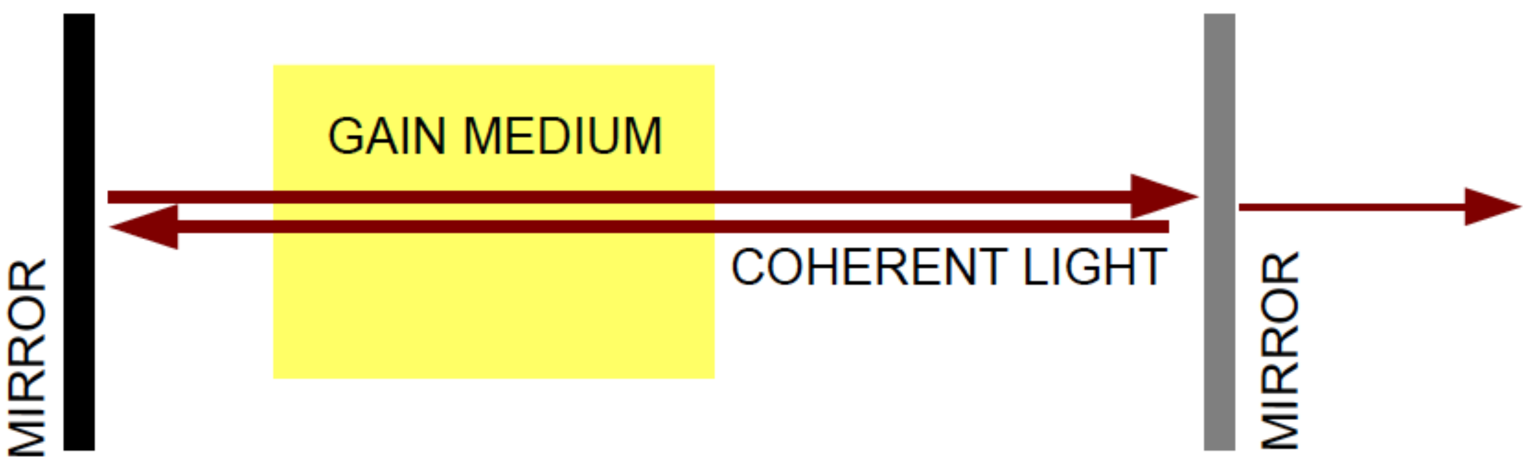
Gordon Gould (1957, 1959). He also invented Xaser, Uaser, …, Raser….

- Laser can be single mode or **multimode**, continuous wave (laser pointer) or **pulsed** (“ps”, “fs”), …

Laser

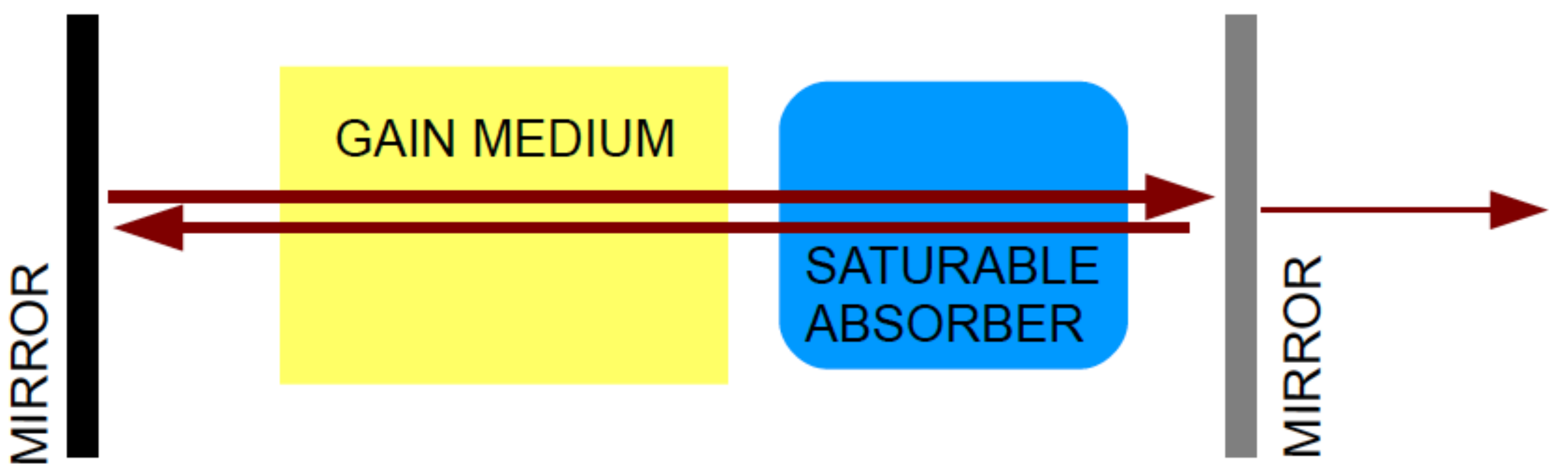
Two essential components

- Cavity — Coherent feedback
- Gain medium — Amplification by Stimulated Emission



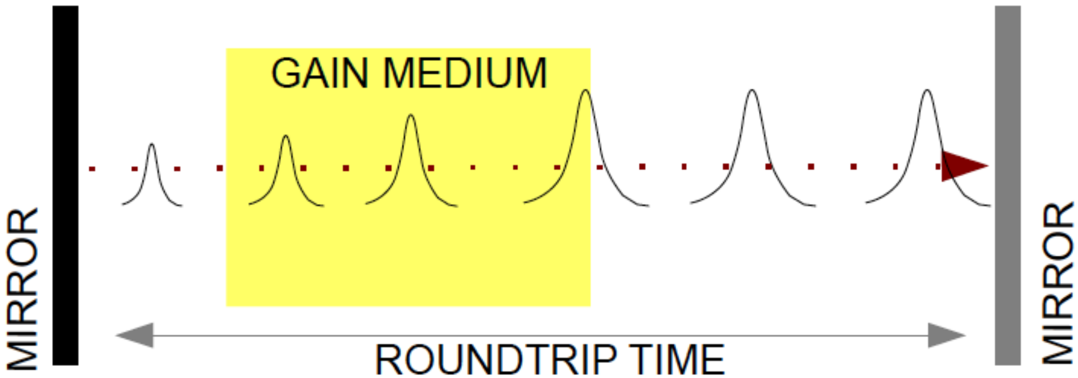
Ultrafast multimode laser

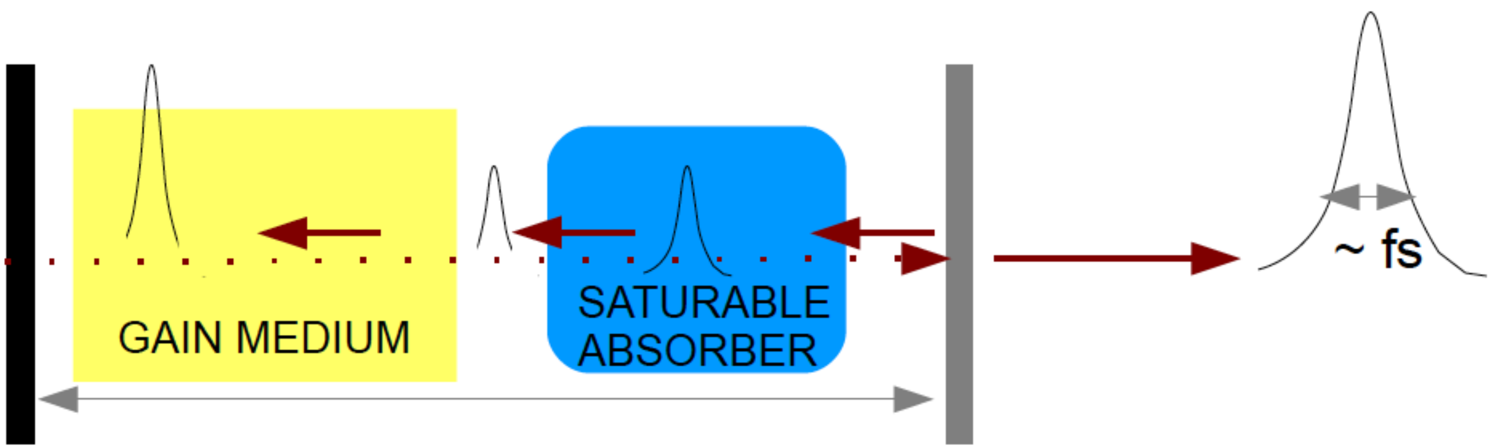
- Cavity — Coherent feedback
- Gain medium — Amplification by S.E.
- Saturable absorber — Passive mode-locking



Ultrafast multimode laser

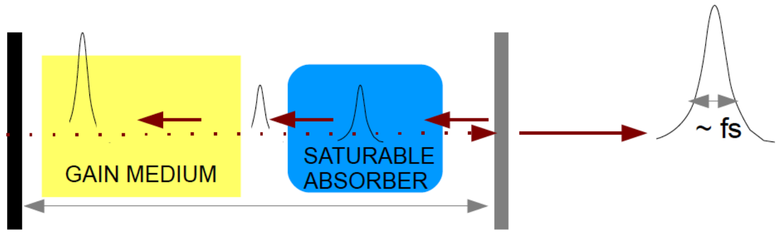
- Cavity — Coherent feedback
- Gain medium — Amplification by S.E.
- Saturable absorber — Passive mode-locking

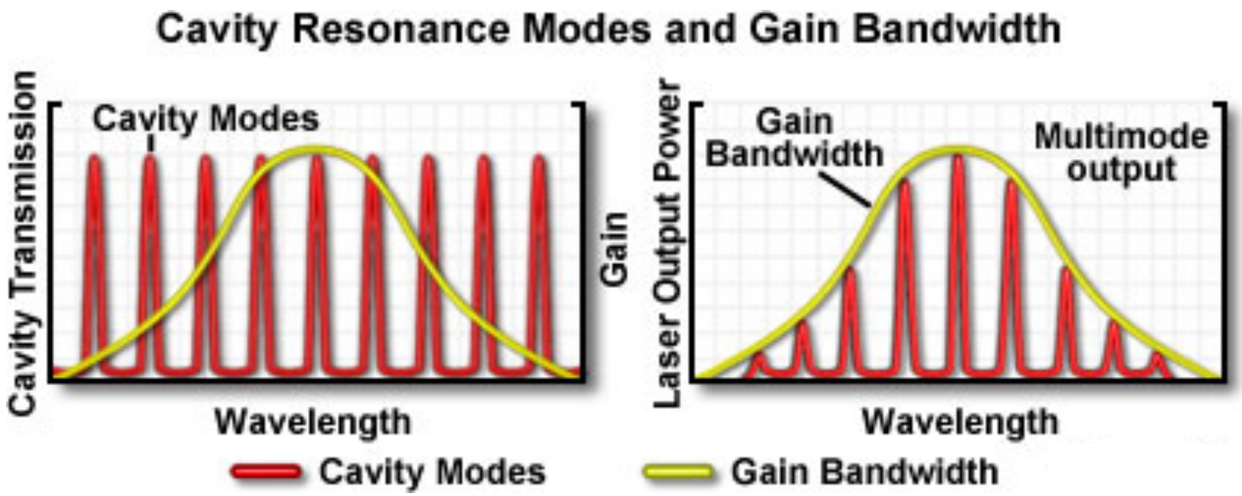




Ultrafast multimode laser

- Cavity — Coherent feedback
- Gain medium — Amplification by S.E.
- Saturable absorber — Passive mode-locking





$\Delta\nu$ Free spectral range

$\delta\nu$ Frequencies spacing

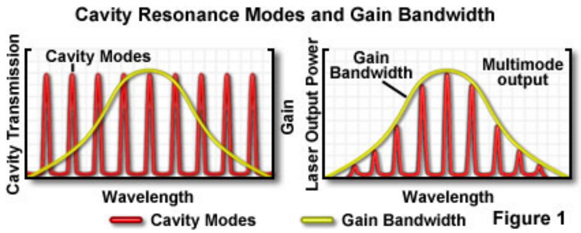
γ Linewidth

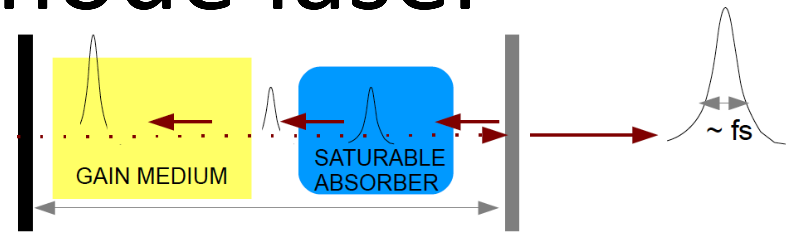
Finesse $f \equiv \delta\nu / \gamma$

Quality factor

$$f \propto Q = 2\pi \frac{\text{STORED ENERGY in the cavity}}{\text{DISSIPATED ENERGY per cycle}}$$

Ultrafast multimode laser





The saturable absorber induces self-starting synchronous oscillations of modes in the cavity: mode-locking -> fast pulses

Mode locking/Phase locking

takes place above the lasing optical power threshold.
It is triggered by a non-linear frequency matching condition occurring in the saturable absorber:

$$\omega_{n_1} - \omega_{n_2} + \omega_{n_3} - \omega_{n_4} = 0.$$

HA Haus, *Mode-Locking of Lasers*, IEEE J. Quantum Electron., 2000

In the lasing regime, the phases of the amplified modes acquire a ~ linear relationship to the frequencies:

$$\phi(\omega) = \phi_0 + \phi' \ \omega + O(\omega^2)$$

$$\phi_{n_1} - \phi_{n_2} + \phi_{n_3} - \phi_{n_4} = 0$$

ALL STANDARD SO FAR

Random laser

A Laser with nonresonant scatterer,
Ambartsumyan, Basov, Kryukov, Lethokov (1966)
"Scatterer-mirror", strong mode interaction due to scattering in different directions: there is coherent feedback but not on a narrow frequency interval -> "nonresonant".

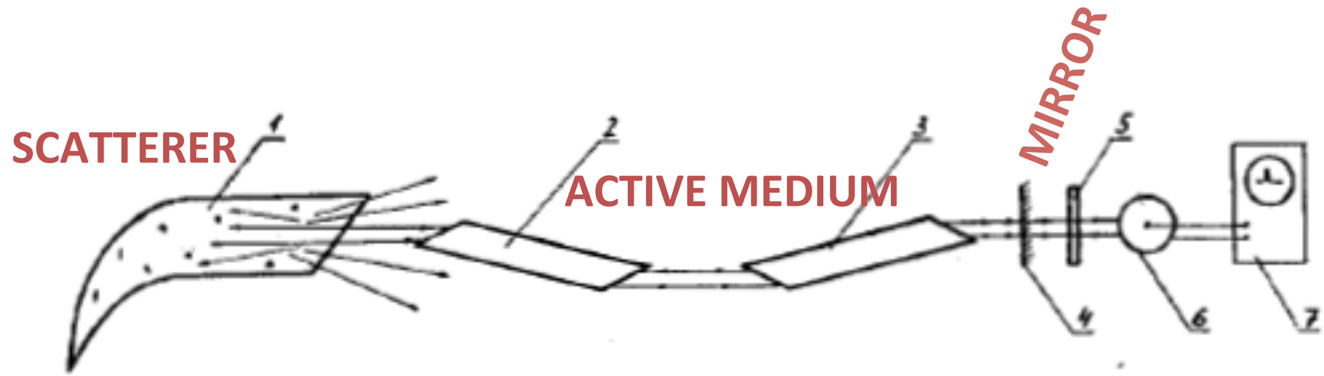
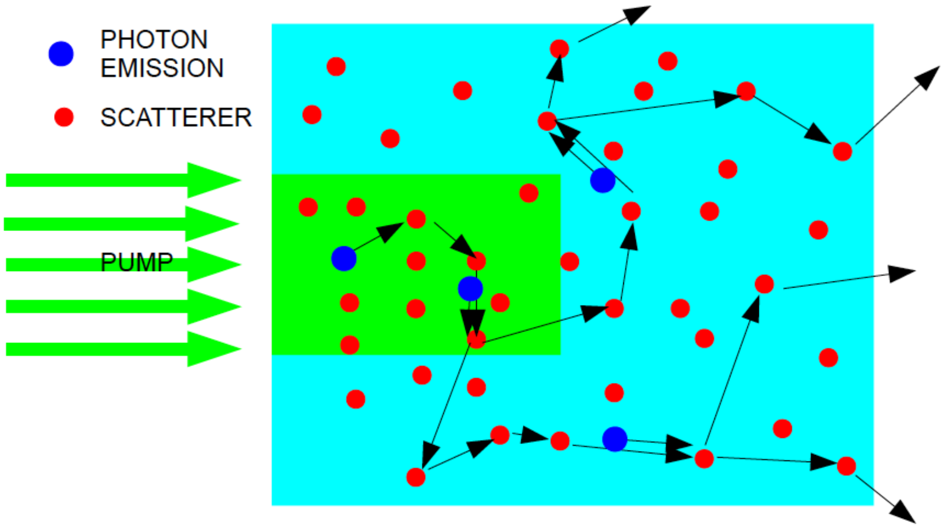


Fig. 1. Experimental arrangement—laser with a resonant feedback due to scattering.

Random laser

Generation of Light by a Scattering Medium with Negative Resonance Absorption, Letokhov (1968)

When photon path length is larger than amplification length: photon multiplication



Photonic bomb

26 years of silence....

Random laser

26 years of silence….

Laser action in strongly scattering media
Lawandy et al. 1994
A multiple scattering medium (i.e., a set of *stochastic resonators*) leads to a multimode **random laser** (non-resonant)

Hui Cao et. al 1998
Resonant RL

ZnO powder

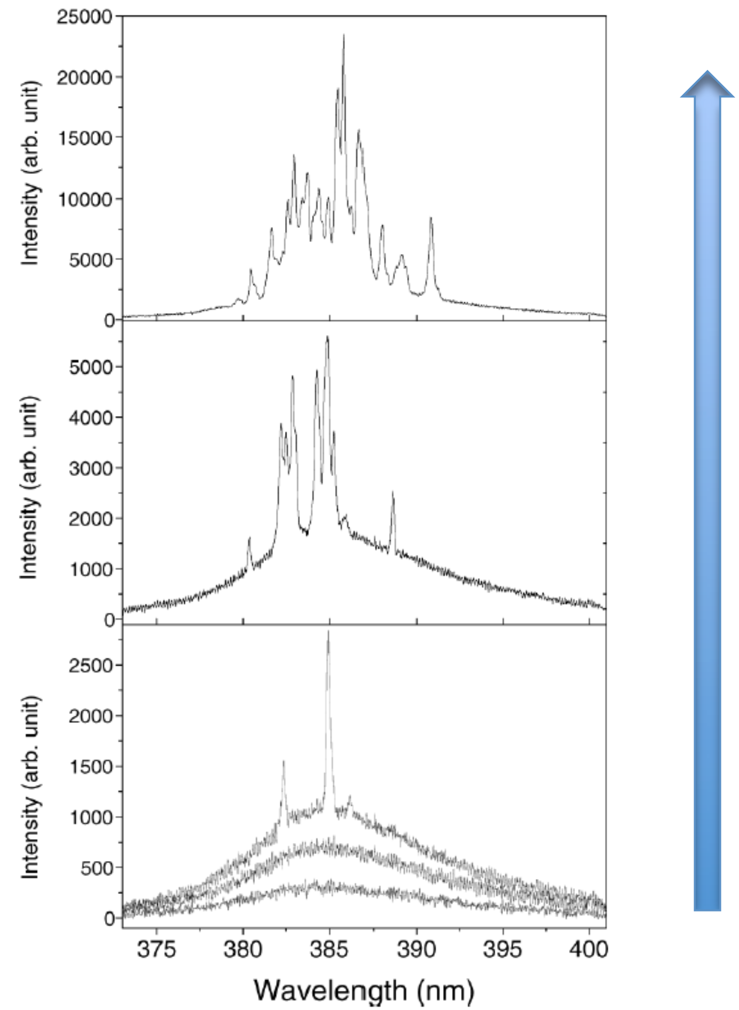
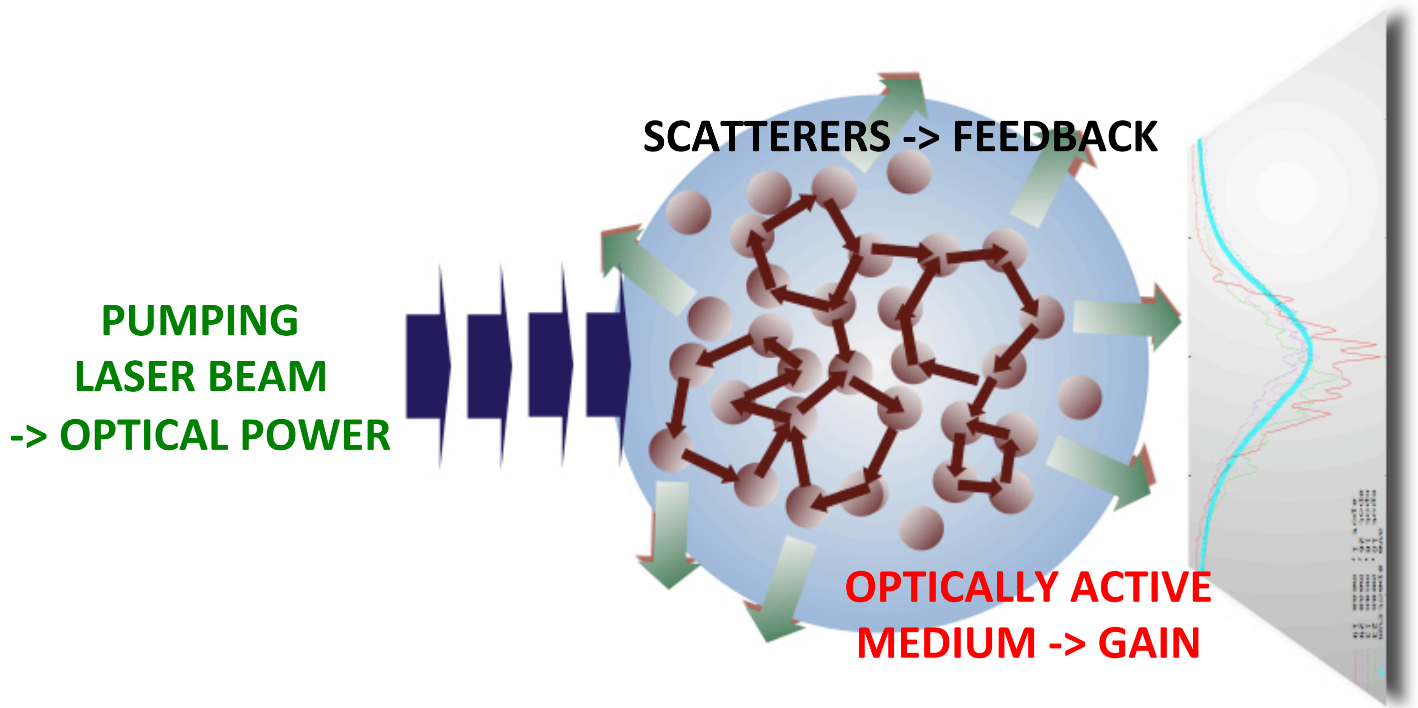


Figure 1. Spectra of emission from ZnO powder when the excitation intensity is (from bottom to top) 400, 562, 763, 875 and 1387 kW cm^{-2}.

Random laser



- Cavity-less stimulated amplified emission: after multiple scattering photons return to a coherence region visited before -> stochastic resonator.
- The frequency of a lasing mode is determined by the interference with backscattered photons.
- Multimode lasing: many localized modes are established. Is there mode-locking?

Random laserS

Many kinds of : TiO_2 or ZnO powder, TiO_2 or ZnO+Rhodamine-methanol solution, DDO-PPV film, porous GaP, oligomer T5OCx,…

Grouped in

- strongly and weakly scattering (Mujumdar et al. 2004, Fallert 2009)
  - S.ly Scat: $kl \leq 1$. Modes are strongly localized in space - linear extension O(1 μm). Small spatial overlap among different modes.
  - W.ly Scat: $kl >> 1$. Modes extension can be as large as the sample, strong leaks at the boundary. Large spatial overlap among modes.

- resonant and non-resonant feedback (Cao 2002)
  - Resonant: modes frequencies are well resolved in spectra, linewidth $\gamma << \delta\nu$. High *finesse,* high Q-factor.
  - Non-resonant: modes frequencies are strongly overlapping, $\gamma \approx \delta\nu$. Low *f*, low Q.

Random laserS

- One might group them also according to their spectral reproducibility under identical experimental conditions (sample structure, pumping): *shot-to-shot*
  – Fairly reproducible spectra (small fluctuations between spectra in different experiments, but same peaks)
  – Always different spectra (large fluctuations in intensity, different peaks activated)

Mode-locking RL's?

Mode-Locking Random Laser

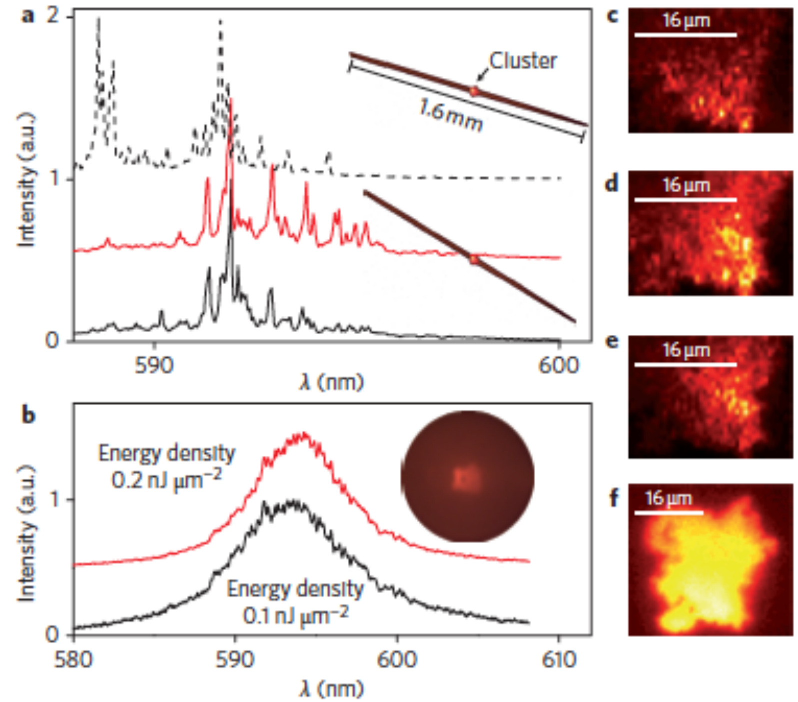
Directional pumping protocol enabling the selection of the number of activated modes: modes oscillates synchronously, also in absence of a saturable absorber (and of a cavity!).

As modes interact (i.e., spatially overlap) their emissions are correlated: mode-locked.

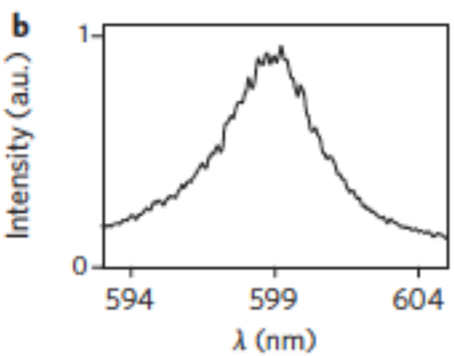
As number modes increase: resonant to (apparently) non-resonant RL spectra.

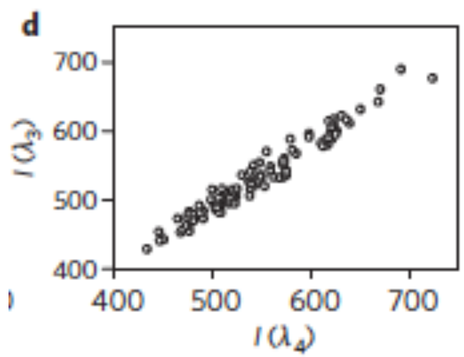
In a “non-resonant” RL so many resonances are there making the spectrum smooth and apparently spike-less.

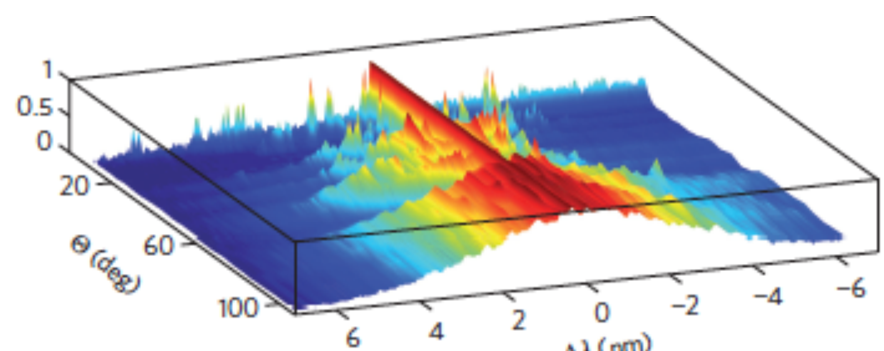
Small fluctuations are strongly correlated.



M. Leonetti, C. Conti, C. Lopez, 2011







TiO_2 in rhodamine sol.

Random laser

- Basic questions:
  - What are the lasing modes?
  - **How do modes interact?**
  - How is the laser pulse? How coherent is a random laser?
  - Use? Control? Applications? Security? Health? Environment?

Modeling multimode lasing with statistical mechanics

Electromagnetic field dynamics mapping onto stochastic dynamics:

identification of a (classical) Hamiltonian and of a “thermal” *reservoire*.

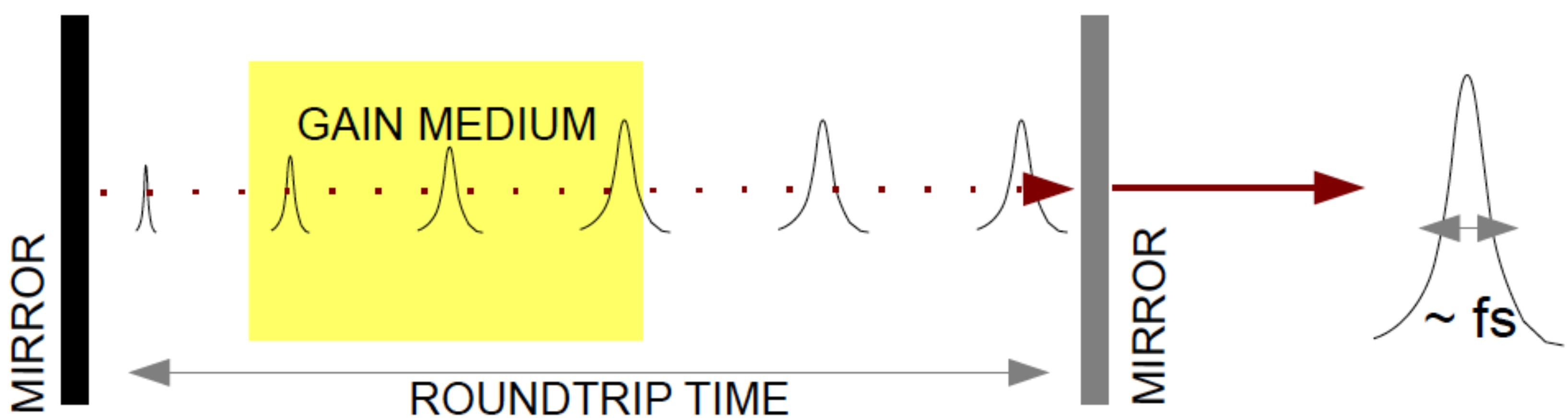
Different ways (somewhat complementary) to derive the Hamiltonian

- Comparing to a generalized Master equation + stochastic noise (Gordon-Fisher 2002) and adding disorder
- Deriving a quantum theory of localized and radiative modes and degrading the stochastic Langevin equation from operators to complex numbers (Hackenbroich-Viviescas, 2003)
- Solving classical Maxwell equations in presence of nonlinear polarization, long time perturbation (Angelani et al., 2006)
- **Computing classical electromagnetic energy averaged over short times (Conti-Leuzzi, 2011)**

Fast and slow time-scales

Each approach involves separation of time-scales

- Time-width of the light pulse.
  Its Fourier transform yields the contribution to the intensity spectra due to a single emission. The shortest the pulse, the more the frequencies, i.e. the wider the free spectral range Δν of the gain profile.
- Time interval between two pulses.
- Round-trip time: the time a light pulse employs to perform an optical cycle in the cavity.



Light in a dielectric (open) cavity

Electromagnetic energy in a dielectric

$$\mathcal{E}_{\mathrm{em}} = \int \boldsymbol{E}^*(\boldsymbol{r},t)\cdot D(\boldsymbol{r},t)\; dV$$

$$\boldsymbol{D}(\boldsymbol{r}) = \epsilon_0\epsilon_r(\boldsymbol{r})\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\mathrm{nl}}(\boldsymbol{r})$$

$$n^2(\boldsymbol{r}) = \epsilon_r(\boldsymbol{r})$$

$$\begin{aligned}\boldsymbol{D}(\boldsymbol{r}) &= \epsilon_0[1+\chi^{(1)}(\boldsymbol{r})]\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\mathrm{nl}}(\boldsymbol{r}) \\ &= \epsilon_0\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\mathrm{lin}}(\boldsymbol{r}) + \boldsymbol{P}_{\mathrm{nl}}(\boldsymbol{r}) \\ &= \epsilon_0 n^2(\boldsymbol{r})\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\mathrm{nl}}(\boldsymbol{r})\end{aligned}$$

Light in a dielectric (open) cavity

Electromagnetic energy in a dielectric

$$\mathcal{E}_{\rm em} = \int \boldsymbol{E}^*(\boldsymbol{r},t)\cdot D(\boldsymbol{r},t)\ dV$$

$$\boldsymbol{D}(\boldsymbol{r}) = \epsilon_0\epsilon_r(\boldsymbol{r})\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\rm nl}(\boldsymbol{r})$$

$$n^2(\boldsymbol{r}) = \epsilon_r(\boldsymbol{r})$$

$$\begin{aligned}\boldsymbol{D}(\boldsymbol{r}) &= \epsilon_0[1+\chi^{(1)}(\boldsymbol{r})]\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\rm nl}(\boldsymbol{r})\\ &= \epsilon_0\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\rm lin}(\boldsymbol{r}) + \boldsymbol{P}_{\rm nl}(\boldsymbol{r})\\ &= \epsilon_0 n^2(\boldsymbol{r})\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\rm nl}(\boldsymbol{r})\end{aligned}$$

Maxwell equations

$$\begin{aligned}\boldsymbol{\nabla}\wedge\boldsymbol{H} &= \partial_t\boldsymbol{D}\\ \boldsymbol{\nabla}\wedge\boldsymbol{E} &= -\mu_0\partial_t\boldsymbol{H}\end{aligned}$$

$$\boldsymbol{P}_{\rm nl} = 0$$

$$\begin{aligned}\boldsymbol{E}(\boldsymbol{r},t) &= \sum_n \boldsymbol{E}_n(\boldsymbol{r})e^{-\imath\omega_n t} + \text{ c.c.}\\ \boldsymbol{H}(\boldsymbol{r},t) &= \sum_n \boldsymbol{H}_n(\boldsymbol{r})e^{-\imath\omega_n t} + \text{ c.c.}\end{aligned}$$

E.m. field expansion in normal modes

Light in a dielectric (open) cavity

Electromagnetic energy in a dielectric

$$\mathcal{E}_{\rm em} = \int \boldsymbol{E}^*(\boldsymbol{r},t)\cdot D(\boldsymbol{r},t)\; dV$$

$$\boldsymbol{D}(\boldsymbol{r}) = \epsilon_0\epsilon_r(\boldsymbol{r})\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\rm nl}(\boldsymbol{r})$$

$$n^2(\boldsymbol{r}) = \epsilon_r(\boldsymbol{r})$$

$$\begin{aligned}\boldsymbol{D}(\boldsymbol{r}) &= \epsilon_0[1+\chi^{(1)}(\boldsymbol{r})]\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\rm nl}(\boldsymbol{r})\\ &= \epsilon_0\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\rm lin}(\boldsymbol{r}) + \boldsymbol{P}_{\rm nl}(\boldsymbol{r})\\ &= \epsilon_0 n^2(\boldsymbol{r})\boldsymbol{E}(\boldsymbol{r}) + \boldsymbol{P}_{\rm nl}(\boldsymbol{r})\end{aligned}$$

Maxwell equations

$$\begin{aligned}\boldsymbol{\nabla}\wedge\boldsymbol{H} &= \partial_t\boldsymbol{D}\\ \boldsymbol{\nabla}\wedge\boldsymbol{E} &= -\mu_0\partial_t\boldsymbol{H}\end{aligned}$$

$$\boldsymbol{P}_{\rm nl} = 0$$

$$\begin{aligned}\boldsymbol{E}(\boldsymbol{r},t) &= \sum_n \boldsymbol{E}_n(\boldsymbol{r})e^{-\imath\omega_n t} + \text{ c.c.}\\ \boldsymbol{H}(\boldsymbol{r},t) &= \sum_n \boldsymbol{H}_n(\boldsymbol{r})e^{-\imath\omega_n t} + \text{ c.c.}\end{aligned}$$

E.m. field expansion in normal modes

$$\boldsymbol{P}_{\rm nl} \neq 0$$

$$\begin{aligned}\boldsymbol{E}(\boldsymbol{r},t) &= \sum_n a_n(t)\boldsymbol{E}_n(\boldsymbol{r})e^{-\imath\omega_n t} + \text{ c.c.}\\ \boldsymbol{H}(\boldsymbol{r},t) &= \sum_n a_n(t)\boldsymbol{H}_n(\boldsymbol{r})e^{-\imath\omega_n t} + \text{ c.c.}\end{aligned}$$

$$\boldsymbol{P}_{\rm nl}(\boldsymbol{r},t) = \sum_n \boldsymbol{p}_n(\boldsymbol{r},t)e^{-\imath\omega_n t}$$

Laser. Stat. Mech.

$$\boldsymbol{P}_{\rm nl}(\boldsymbol{r},t) = \sum_n \boldsymbol{p}_n(\boldsymbol{r},t) e^{-\imath\omega_n t}$$

Sargent, Scully, Lamb , *Laser Physics,* 1974

$$p_n^\delta(\boldsymbol{r},t) = \sum_{\substack{\omega_j-\omega_k+\omega_l=\omega_n \\ \alpha,\beta,\gamma,\delta=x,y,z}} \chi^{(3)}_{\alpha\beta\gamma\delta}(\omega_j,\omega_k,\omega_l,\omega_j-\omega_k+\omega_l;\boldsymbol{r})\ E_j^\alpha(\boldsymbol{r})E_k^\beta(\boldsymbol{r})E_l^\gamma(\boldsymbol{r})\ a_j(t)a_k^*(t)a_l(t)$$

It is the frequency matching condition leading to mode-locking in standard lasers

$\boldsymbol{P}_{\rm nl} = 0$ Constant amplitudes

$\boldsymbol{P}_{\rm nl} \gtrsim 0$ Slowly varying amplitudes

Hamiltonian for the dynamics of the slow complex amplitudes:

$$\mathcal{E}_{\rm em}(t) \rightarrow \mathcal{H} = \langle \int \boldsymbol{E}^*(\boldsymbol{r},t)\cdot\boldsymbol{D}(\boldsymbol{r},t)\ dV\rangle_{\rm fast}$$

Laser Hamiltonian

$$\mathcal{E}_{\rm em}(t) \;=\; \epsilon_0 \int \boldsymbol{E}^*(\boldsymbol{r},t)\cdot\boldsymbol{E}(\boldsymbol{r},t)\; dV + \epsilon_0 \int E^{\alpha\;*}(\boldsymbol{r},t)\chi^{(1)}_{\alpha\beta}(\boldsymbol{r})E^{\beta}(\boldsymbol{r},t)\; dV + \int \boldsymbol{E}^*(\boldsymbol{r},t)\cdot\boldsymbol{P}_{\rm nl}(\boldsymbol{r},t)\; dV$$

In terms of superposition of normal modes $\boldsymbol{E}(\boldsymbol{r},t) \;=\; \sum_n a_n(t)\boldsymbol{E}_n(\boldsymbol{r})e^{-\imath\omega_n t} + \text{ c.c.}$

$$\begin{aligned}\mathcal{E}_{\rm em}(t) \;=&\; \epsilon_0 \sum_{nm} a_n^*(t)a_m(t)e^{\imath(\omega_n-\omega_m)t}\int E_n^{\alpha}(\boldsymbol{r})\left[\delta_{\alpha\beta}+\chi^{(1)}_{\alpha\beta}(\boldsymbol{r})\right]E_m^{\beta}(\boldsymbol{r})\; dV \\ &+ \sum_{\omega_j-\omega_k+\omega_l-\omega_n=0} a_j(t)a_k^*(t)a_l(t)a_n^*(t)e^{\imath(\omega_j-\omega_k+\omega_l-\omega_n)t}\int E_j^{\alpha}(\boldsymbol{r})E_k^{\beta}(\boldsymbol{r})E_l^{\gamma}(\boldsymbol{r})E_n^{\delta}(\boldsymbol{r})\chi^{(3)}_{\alpha\beta\gamma\delta}(\omega_j,\omega_k,\omega_l,\omega_n;\boldsymbol{r})\; dV\end{aligned}$$

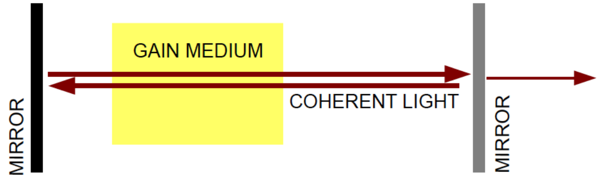
Hamiltonian

$$\mathcal{E}_{\rm em}(t)\to\mathcal{H} = \langle\int \boldsymbol{E}^*(\boldsymbol{r},t)\cdot\boldsymbol{D}(\boldsymbol{r},t)\; dV\rangle_{\rm fast}$$

$$\begin{aligned}\mathcal{H} \;=&\; \sum_n |a_n(t)|^2\int\left[|E_n(\boldsymbol{r})|^2 + E_n^{\alpha}(\boldsymbol{r})\chi^{(1)}_{\alpha\beta}(\boldsymbol{r})E_n^{\beta}(\boldsymbol{r})\right]\; dV \\ &+ \sum_{n\neq m} a_n^*(t)a_m(t)\langle e^{\imath(\omega_n-\omega_m)t}\rangle_{\rm fast}\int E_n^{\alpha}(\boldsymbol{r})\left[\delta_{\alpha\beta}+\chi^{(1)}_{\alpha\beta}(\boldsymbol{r})\right]E_m^{\beta}(\boldsymbol{r})\; dV \\ &+ \sum_{\omega_j-\omega_k+\omega_l-\omega_n=0} a_j(t)a_k^*(t)a_l(t)a_n^*(t)\int \boldsymbol{E}_j^{\alpha}(\boldsymbol{r})E_k^{\beta}(\boldsymbol{r})E_l^{\gamma}(\boldsymbol{r})E_n^{\delta}(\boldsymbol{r})\;\chi^{(3)}_{\alpha\beta\gamma\delta}(\omega_j,\omega_k,\omega_l\omega_n;\boldsymbol{r})\; dV\end{aligned}$$

Laser. Stat. Mech.

CLOSED CAVITY: in a closed cavity with regular mirrors, in which each light pulse takes a roundtip time T_R to complete an “optical cycle” between two mirros



$$\langle e^{\imath(\omega_n-\omega_m)t}\rangle_{\text{opt.cyc.}} = \frac{1}{T_R}\int_0^{T_R} dt\ e^{\imath(\omega_n-\omega_m)t} = \frac{1}{T_R}\int_0^{T_R} dt\ e^{\frac{2\pi}{T_R}\imath(n-m)t} = 0 \quad ; \quad n \neq m$$

We are back to the mode-locking standard laser:

no off-diagonal linear interaction.

OPEN CAVITY:

(i) Mirror cavities with leakages: there will be radiative modes, whose frequencies take values over a continuous dominion, thus the integral above can be a non-zero complex number.

(ii) Mirror-less lasers in random media, with inhomogeneous optical susceptibility profiles: also the discrete lasing frequencies will not be all equispaced. Furthermore, the “optical cycle” and the “roundtrip time” are not defined. Their random analogues depend on the scatterers structure.

Linear non-diagonal contribution to the Hamiltonian.

Eigenmodes basis in open cavity

OPEN CAVITY:

(i) Mirror cavities with leakages: there will be radiative modes, whose frequencies take values over a continuous dominion, thus the integral above can be a non-zero complex number.

(ii) Mirror-less lasers in random media, with inhomogeneous optical susceptibility profiles: also the discrete lasing frequencies will not be all equispaced. Furthermore, the “optical cycle” and the “roundtrip time” are not defined. Their random analogues depend on the scatterers structure.

$$\boldsymbol{E}(\boldsymbol{r},t) \;=\; \sum_n a_n(t)\boldsymbol{E}_n(\boldsymbol{r})e^{-\imath\omega_n t} + \;\text{c.c.}$$

What is a complete basis in an open system?

Fox-Li modes, quasi-bound states, *constant flux modes*, ….

Strong Interactions in Multimode Random Lasers
Hakan E. Türeci, Li Ge, Stefan Rotter, A. Douglas Stone 2008

Laser. Stat. Mech.

OPEN CAVITY : "Input-output" quantum field theory

Viviescas and Hackenbroich 2003

A Hamiltonian can be written in term of creation-annihilation operators for both inner and outer modes

Separation of localized/inner modes inside the stimulated region and radiative/outer modes

By projection onto two complementary subspaces, completing the whole space.

Complete basis divided in

localized eigenmodes with discrete frequencies $\boldsymbol{u}_k(\boldsymbol{r})$ ω_k

radiative eigenmodes whose frequencies are distributed on a continuum $\boldsymbol{v}_{\mathsf{A}}(\omega, \boldsymbol{r})$

In terms of creation-annihilation operators for inner (a) and outer (b) modes:

$$\mathcal{H}_{\text{open}} = \sum_n a_n^\dagger a_n + \sum_{\mathsf{A}} \int d\omega\ b_{\mathsf{A}}^\dagger(\omega) b_{\mathsf{A}}(\omega) + \sum_n \sum_{\mathsf{A}} \int d\omega\ \left[\mathcal{W}_{n\mathsf{A}}\ a_n^\dagger\ b_{\mathsf{A}}(\omega) + \text{ h.c.}\right]$$

Eventually for the internal modes one obtains an effective non-diagonal interaction

$$J_{\vec{n}_2}^{\text{rad}} = \pi \sum_{\mathsf{A}} \mathcal{W}_{n_1 \mathsf{A}} \mathcal{W}_{\mathsf{A} n_2}^\dagger$$

$$\mathcal{W}_{k\mathsf{A}}(\omega) \equiv \frac{c^2}{2\sqrt{\omega_k \omega}} \int_{\text{boundary}} d\mathcal{S}\ \boldsymbol{u}_k^*(\boldsymbol{r}) \cdot \{\hat{\boldsymbol{n}} \wedge [\nabla \wedge \boldsymbol{v}_{\mathsf{A}}(\omega, \boldsymbol{r})]\}$$

Laser. Stat. Mech.

$$\mathcal{H} = -\sum_{n_1,n_2} J_{n_1 n_2} a_{n_1} a^*_{n_2} - \sum_{\omega_{n_1}-\omega_{n_2}+\omega_{n_3}-\omega_{n_4}=0} J_{\vec{n}_4} a_{n_1} a^*_{n_2} a_{n_3} a^*_{n_4} + \text{ c.c.}$$

$$J_n \equiv \epsilon_0 \left[1 + \int E_n^{\alpha_1}(\boldsymbol{r}) \chi^{(1)}_{\vec{\alpha}_2}(\boldsymbol{r}) E_n^{\alpha_2}(\boldsymbol{r})\right] dV$$

$$J^{\text{inh}}_{n_1 \neq n_2} \equiv \epsilon_0 \langle e^{\imath(\omega_{n_1}-\omega_{n_2})t}\rangle_{\text{fast}} \int E_{n_1}^{\alpha_1}(\boldsymbol{r}) \chi^{(1)}_{\vec{\alpha}_2}(\boldsymbol{r}) E_{n_2}^{\alpha_2}(\boldsymbol{r})\ dV$$

$$J^{\text{rad}}_{\vec{n}_2} = \pi \sum_{\mathsf{A}} \mathcal{W}_{n_1 \mathsf{A}} \mathcal{W}^\dagger_{\mathsf{A} n_2}$$ (operators -> c-numbers)

$$J_{\vec{n}_2} = J^{\text{inh}}_{\vec{n}_2} + J^{\text{rad}}_{\vec{n}_2}$$

E.M.F. SPATIAL OVERLAP

⟵⟶

AMPS INTERACTION

$$J_{\vec{n}_4} \equiv \int \boldsymbol{E}_{n_1}^{\alpha_1}(\boldsymbol{r}) E_{n_2}^{\alpha_2}(\boldsymbol{r}) E_{n_3}^{\alpha_3}(\boldsymbol{r}) E_{n_4}^{\alpha_4}(\boldsymbol{r})\ \chi^{(3)}_{\vec{\alpha}_4}(\{\omega_{\vec{n}_4}\}; \boldsymbol{r})\ dV$$

All coupling coefficients are in general complex numbers

Can you have other kind of nonlinearity: $\chi^{(2)}$, $\chi^{(4)}$,… ? SURE!

$\chi^{(3)}$ is just a (motivated) working choice

Langevin equation for light-mode complex amplitudes

The dynamics of the amplified localized modes created by stimulated emission in presence of spontaneuous emission is given by the Langevin equation

$$\dot{a}_n = -\imath \frac{\partial \mathcal{H}}{\partial a_n^*} + \eta_n(t);$$

$$\langle \eta_n(t) \rangle = 0; \qquad \langle \eta_n(t) \eta_m(s) \rangle = 2T\delta(t-s)\gamma_{nm} \simeq 2T\delta(t-s)\delta_{nm}$$

N:B.: the more the cavity is open the less accurate is the diagonal approximation for the noise

Standard Mode-Locking Laser

The completely closed and ordered limit:

Haus standard ML laser master equation

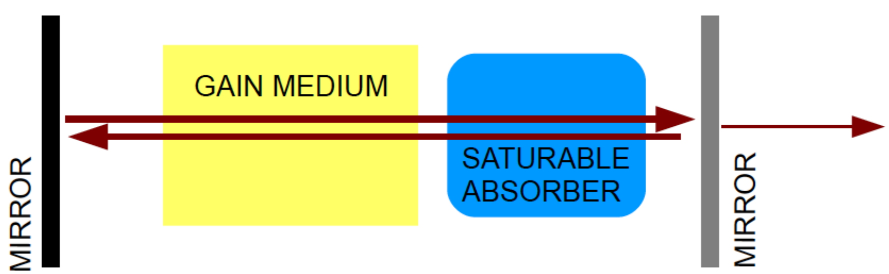
HA Haus, *Waves and Fields in Optoelectonics, 1984*

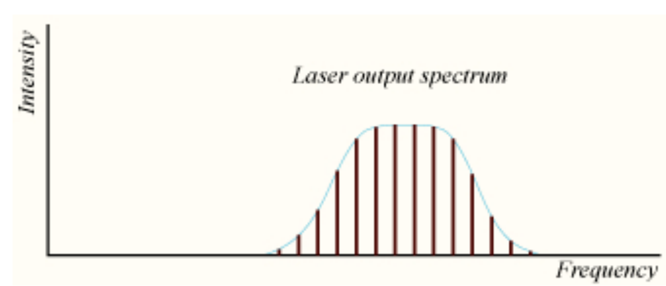
HA Haus, *Mode-Locking of Lasers*, IEEE J. Quantum Electron., 2000

$$\dot{a}_n = (g_m - \ell_m + \imath D_m)a_n + (\gamma - \imath\delta) \sum_{\omega_j - \omega_k + \omega_l = \omega_n} a_j a_k^* a_l + \eta_n$$

GAIN **LOSS** **GROUP VELOCITY DISPERSION** **SAM** **KERR LENS** **SPONTANEOUS EMISSION**

SAM: SELF-AMPLITUDE MODULATION COEFFICIENT OF THE SATURABLE ABSORBER





Standard ML Laser

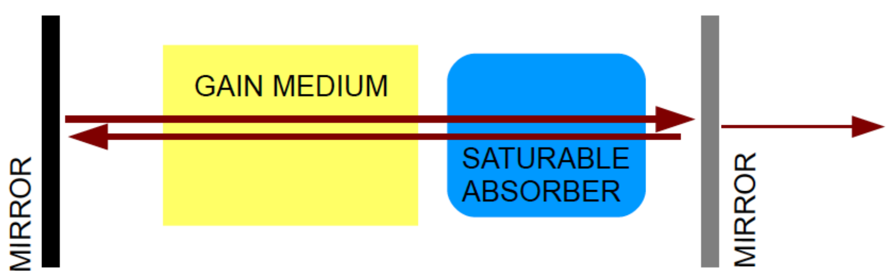
$$\dot{a}_n = (g_m - \ell_m + \imath D_m)a_n + (\gamma - \imath\delta) \sum_{\omega_j - \omega_k + \omega_l = \omega_n} a_j a_k^* a_l + \eta_n$$

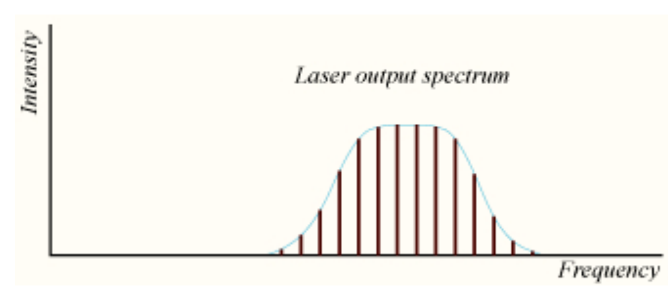
GAIN **LOSS** **GROUP VELOCITY DISPERSION** **SAM** **KERR LENS** **SPONTANEOUS EMISSION**

In the strong cavity limit with space-homogeneous gain (linear susceptibility) we compare with our Hamiltonian:

$$\dot{a}_n = -\imath \frac{\partial \mathcal{H}}{\partial a_n^*} + \eta_n = \imath J_n a_n + \imath J_4 \sum_{\omega_j - \omega_k + \omega_l = \omega_n} a_j a_k^* a_l + \eta_n$$

Physical meaning of the coupling parameters by comparison





Laser stationary regime and equilibrium stat. mech.

Lasers are not at equilibrium: energy is pumped to mantain the population inversion and the stimulated emission and in open cavities energy is lost by radiation.

As the optical power pumped into the system is kept strictly constant

$$\mathcal{E}_P = \epsilon N = \sum_{m=1}^{N} |a_m|^2$$

The so obtained stationary system can be considered as in equilibrium with an effective "heat-bath" at "temperature"

$$\text{``}T_{\text{laser}}\text{''} = \frac{T}{\epsilon^2}$$

PUMPING RATE

$$\mathcal{P} = \epsilon\sqrt{\beta J_0}$$

Notice that in Random Lasers the effect of lowering the T is shown to be experimentally equivalent to raise the optical power.

D.S. Wiersma and S. Cavalieri (2002), *Temperature-controlled random laser action in liquid crystal infiltrated systems.*

Mean-field RL replica theory

$$\mathcal{H} = -\Re\left[\frac{1}{2} \sum_{n_1,n_2}^{1,N} J_{\vec{n}_2} a_{n_1} a_n^* \quad + \frac{1}{4!} \sum_{\omega_{n_1}+\omega_{n_3}=\omega_{n_2}+\omega_{n_4}}^{n_k=1,N} J_{\vec{n}_4} a_{n_1} a_{n_2}^* a_{n_3} a_{n_4}^* \right]$$

arXiv:1406.7826v1
F. Antenucci et al.

NARROWBAND APPROXIMATION $\omega_n \simeq \omega_0,$

implies

$$J_n = g(\omega_n) \simeq g(\omega_0) = g_0,$$

$$\sum_{n_1,n_2} J_{\vec{n}_2} a_{n_1} a_{n_2}^* = g_0 \mathcal{E} + \sum_{n_1 \neq n_2} J_{\vec{n}_2} a_{n_1} a_{n_2}^*.$$

Given a system of fixed scatterers the couplings are quenched disordered.
We consider them as Gaussian distributed

$$P(J_{\vec{n}_p}) = \sqrt{\frac{N^{p-1}}{2\pi J_p^2}} \exp\left\{ -\frac{N^{p-1}}{2J_p^2} \left[J_{\vec{n}_p} - \frac{J_0^{(p)}}{N^{p-1}} \right]^2 \right\}$$

p=2,4

Mean-field RL replica theory

$$\mathcal{H} = -\Re\left[\frac{1}{2}\sum_{n_1,n_2}^{1,N} J_{\vec{n}_2} a_{n_1} a_n^* \quad + \frac{1}{4!}\sum_{\omega_{n_1}+\omega_{n_3}=\omega_{n_2}+\omega_{n_4}}^{n_k=1,N} J_{\vec{n}_4} a_{n_1} a_{n_2}^* a_{n_3} a_{n4}^*\right]$$

$$\omega_n \simeq \omega_0, \qquad \sum_{n_1,n_2} J_{\vec{n}_2} a_{n_1} a_{n_2} = g_0\mathcal{E} + \sum_{n_1 \neq n_2} J_{\vec{n}_2} a_{n_1} a_{n_2}.$$

arXiv:1406.7826v1
F. Antenucci et al.

$$P(J_{\vec{n}_p}) = \sqrt{\frac{N^{p-1}}{2\pi J_p^2}} \exp\left\{-\frac{N^{p-1}}{2J_p^2}\left[J_{\vec{n}_p} - \frac{J_0^{(p)}}{N^{p-1}}\right]^2\right\}$$

A bit of parameter definitions

"nonlinearity" or "closeness" degrees

$$J_0^{(4)} = \alpha_0 J_0; \;\; \alpha_0 = \left[\frac{J_0^{(2)}}{J_0^{(4)}} + 1\right]^{-1}; \;\; J_0 = J_0^{(2)} + J_0^{(4)}$$

$$J_4 = \alpha J; \qquad \alpha = \left[\frac{J_2}{J_4} + 1\right]^{-1}; \quad J = J_2 + J_4$$

Degree of randomness

$$R_J = J/J_0$$

Pumping rate

$$\mathcal{P} = \epsilon\sqrt{\beta J_0}$$

Mean-field RL replica theory: thermodynamic phases, order parameters, phase diagrams

Order parameters

$$m_\alpha = \frac{1}{N}\sum_k a_k^\alpha$$

It is either real or pure imaginary

$$q_{\alpha\beta} = \frac{1}{N}\sum_k a_k^\alpha a_k^{*\beta}$$

It is real

$$s_{\alpha\beta} = \frac{1}{N}\sum_k a_k^\alpha a_k^\beta$$

The imaginary part is zero,
the off-diagonal real part is equal to $q_{\alpha\beta}$
The diagonal real part is r_d

INTENSITY COHERENCE m

PHASE COHERENCE r_d

COMPLEX AMPLITUDES OVERLAP $q_{\alpha\beta}$

RSB PARAMETER x (1RSB)

REPLICA SYMMETRY BREAKING

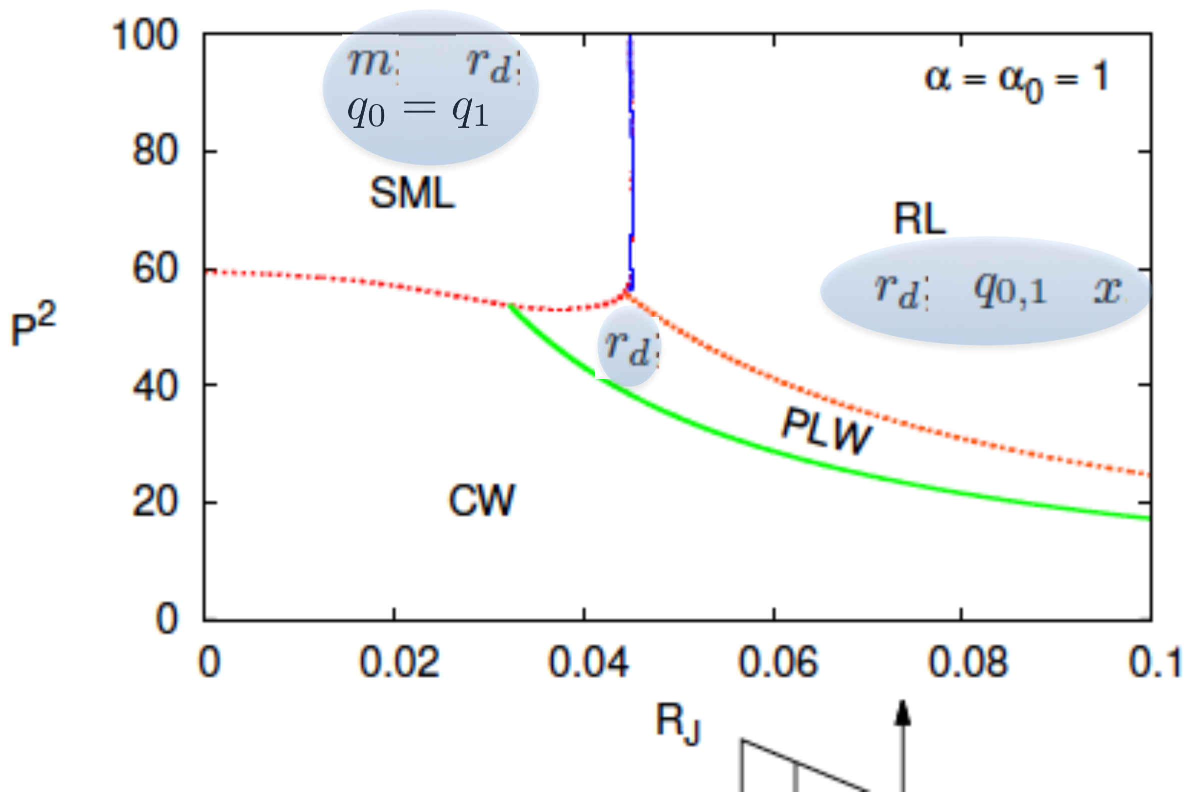
JJ Ruiz-Lorenzo, F. Guerra
This morning

Mean-field RL replica theory: thermodynamic phases, order parameters, phase diagrams

PHASE DIAGRAM: **CLOSED CAVITY** + ANY DEGREE OF DISORDER

SML: Standard Mode Locking laser
CW: Continuous Wave regime
PLW: Phase-Locked Wave regime
RL: Random Laser

SML: Ferromagnetic light
CW: Paramagnetic light
PLW: ??
RL: **Glassy light**



$$\mathcal{P} = \epsilon\sqrt{\beta J_0}$$

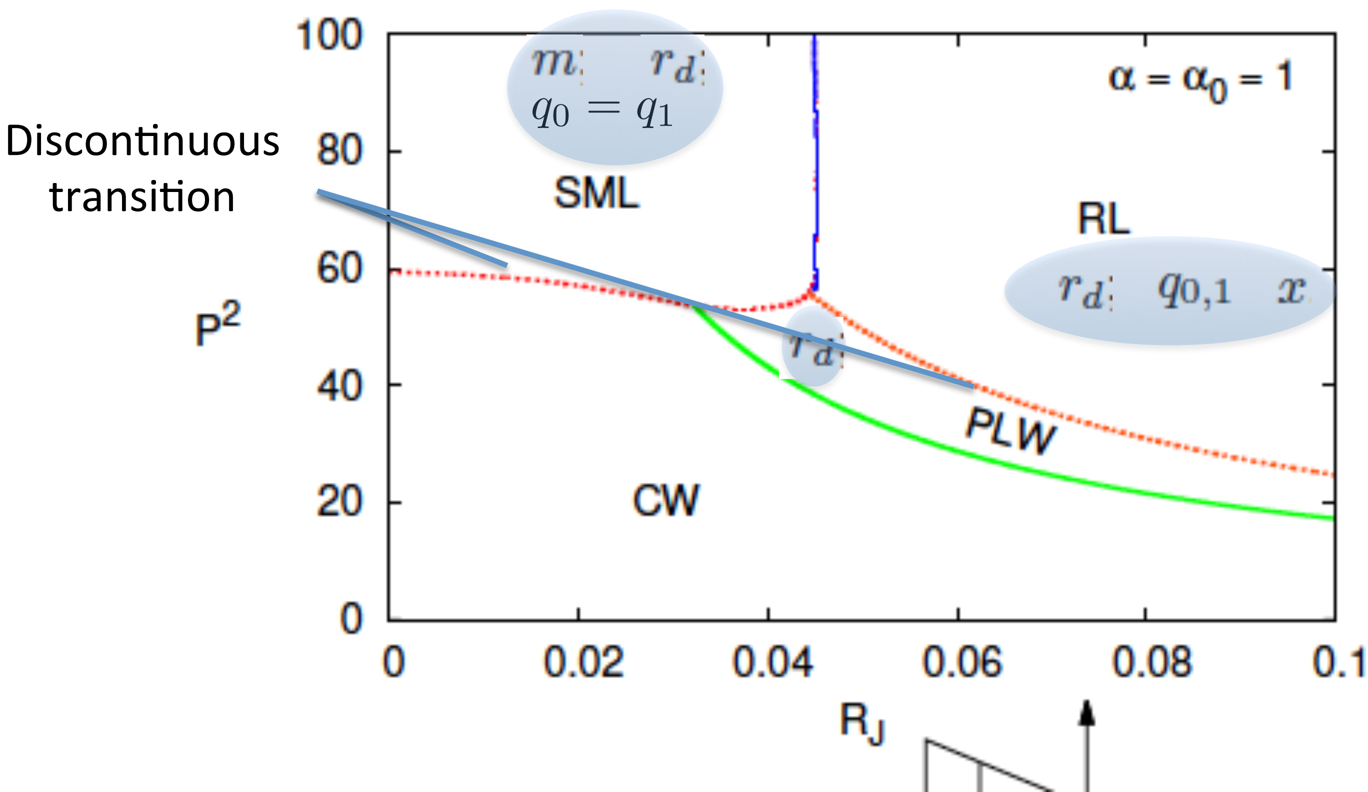
$$R_J = J/J_0$$

Mean-field RL replica theory: thermodynamic phases, order parameters, phase diagrams

PHASE DIAGRAM: **CLOSED CAVITY** + ANY DEGREE OF DISORDER

SML: Standard Mode Locking laser
CW: Continuous Wave regime
PLW: Phase-Locked Wave regime
RL: Random Laser

SML: Ferromagnetic light
CW: Paramagnetic light
PLW: ??
RL: **Glassy light**



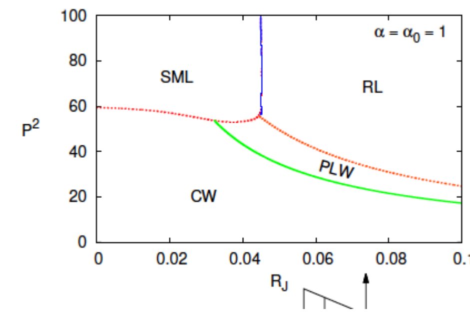
$$\mathcal{P} = \epsilon\sqrt{\beta J_0}$$

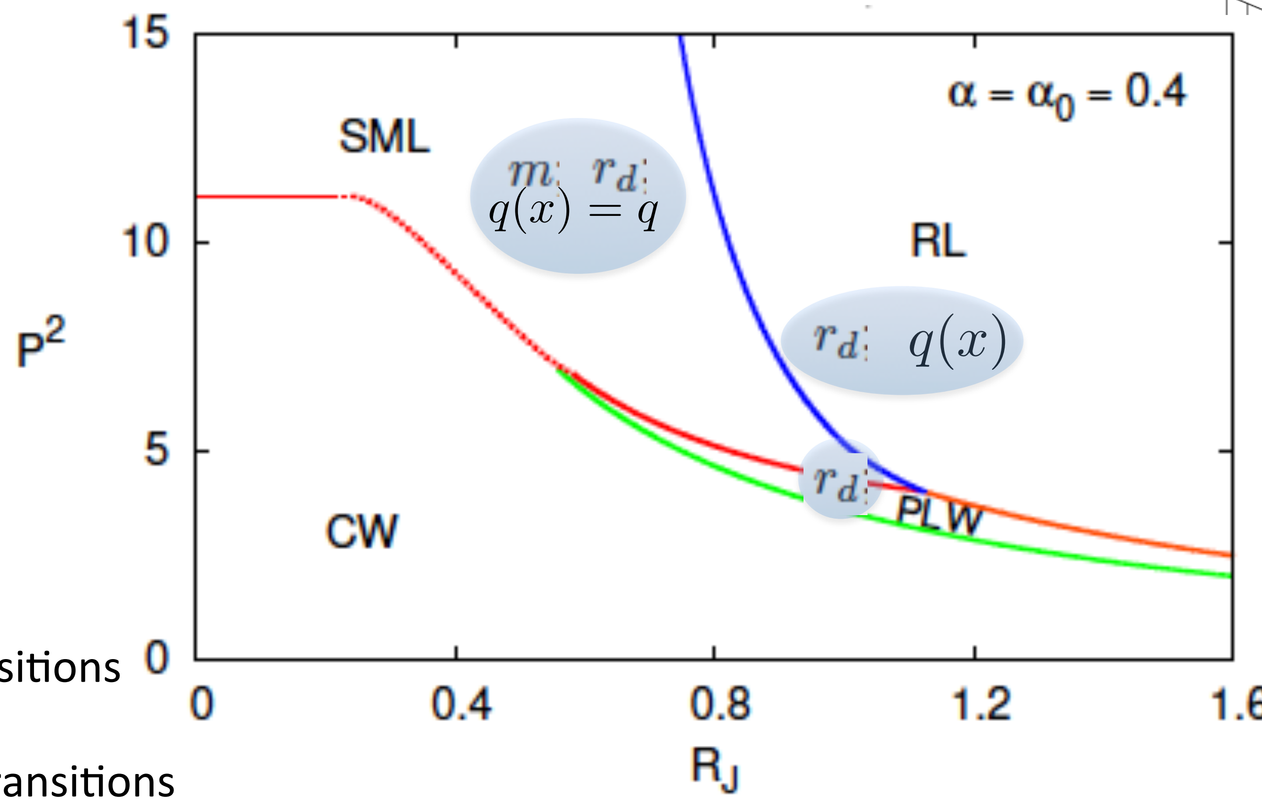
$$R_J = J/J_0$$

Mean-field RL replica theory: thermodynamic phases, order parameters, phase diagrams

PHASE DIAGRAM: **OPEN CAVITY**

SML: Standard Mode Locking laser
CW: Continuous Wave regime
PLW: Phase-Locked Wave regime
RL: Random Laser



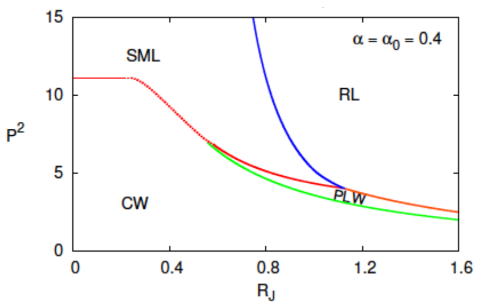


$$\mathcal{P} = \epsilon\sqrt{\beta J_0}$$

$$R_J = J/J_0$$

Solid lines:
Continuous transitions
Dashed lines:
Discontinuous transitions

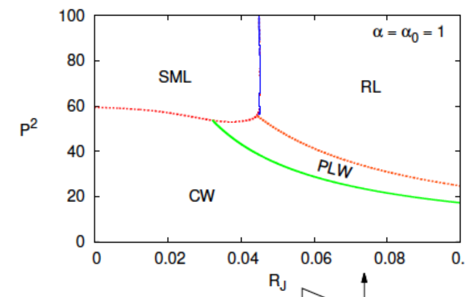
Mean-field laser replica theory: thermodynamic phases, order parameters, phase diagrams



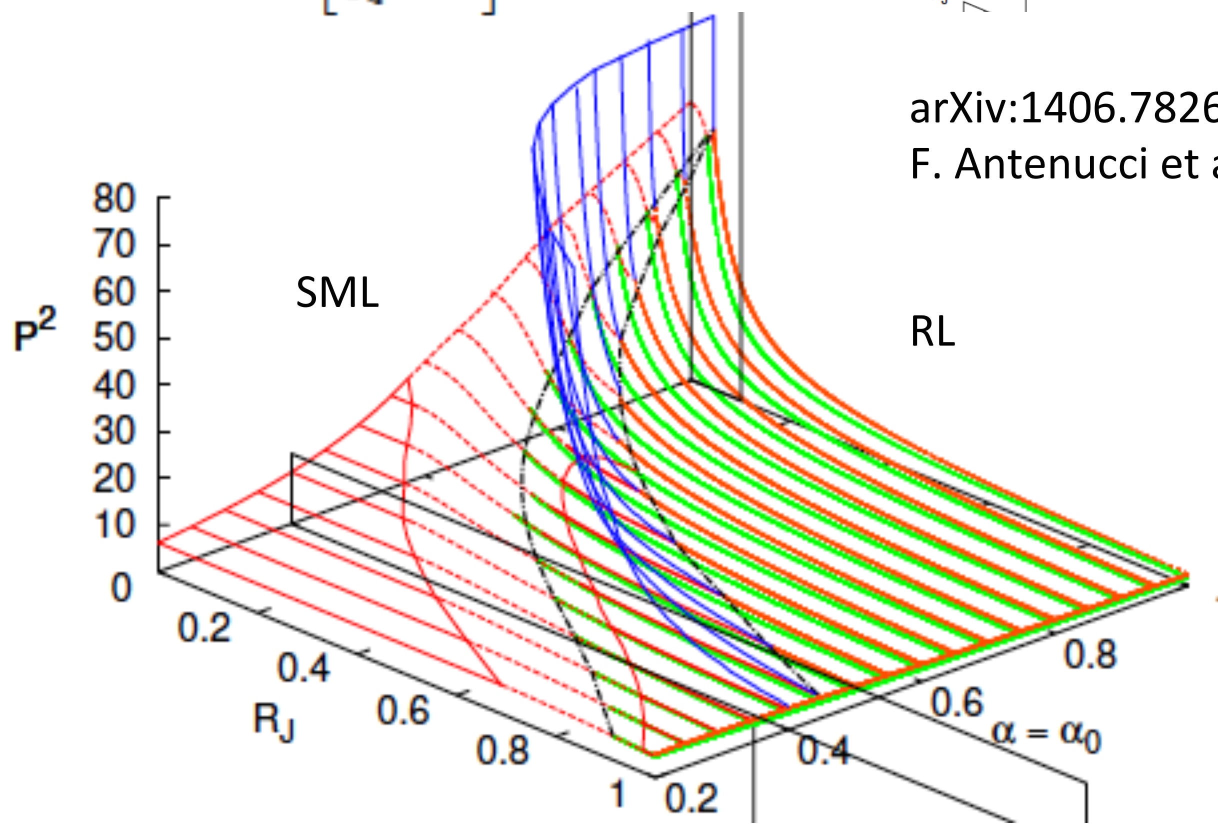
$$\mathcal{P} = \epsilon\sqrt{\beta J_0}$$

$$R_J = J/J_0$$

$$\alpha = \left[\frac{J_2}{J_4} + 1\right]^{-1}$$



arXiv:1406.7826v1
F. Antenucci et al.

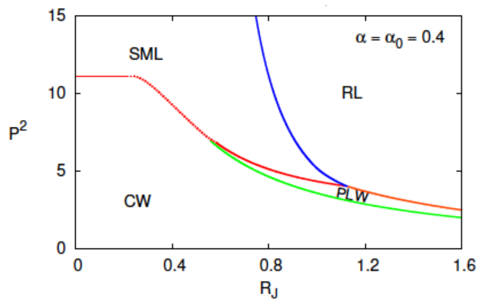


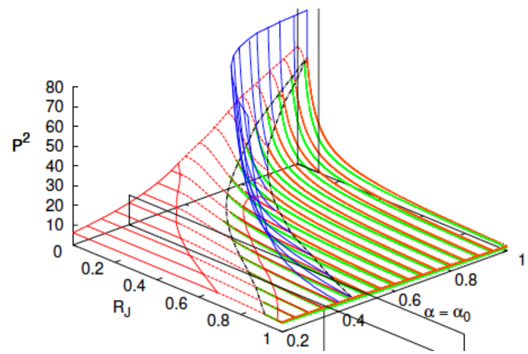
SML: Standard Mode Locking laser
CW: Continuous Wave regime
PLW: Phase-Locked Wave regime
RL: Random Laser

Solid lines:
Continuous transitions
Dashes, dotted lines:
Discontinuous transitions

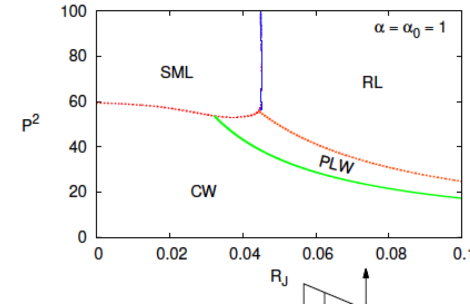
Glassy light phase

OPEN





CLOSED



arXiv:1406.7826v1

The GLASSY LIGHT phase above the pumping threshold depends on the degree of openness of the cavity.

$$\alpha_{\rm nl} = 0.6297.$$

SML: Standard Mode Locking laser
CW: Continuous Wave regime
PLW: Phase-Locked Wave regime
RL: Random Laser

If α is larger, the **nonlinear** contribution dominates,
The transition is **discontinuous** in the order parameters and there is also a dynamic transition. The glassy light phase is characterized by a stable 1**-step RSB** solution.

Solid lines:
Continuous transitions
Dashes, dotted lines:
Discontinuous transitions

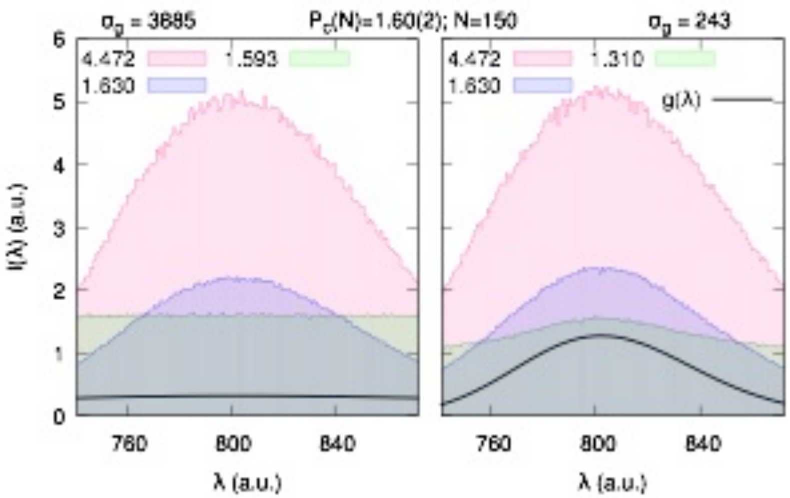
If α is smaller, the **linear** contribution dominates,
The transition is **continuous** in the order parameters. The glassy light phase is characterized by an **∞ -step RSB** solution.

Mean-field to real world contact

- Realistic standard ML laser as a statistical mechanical problem: gain with finite bandwidth
- Standard ML <-> RL? Is it feasible in real photonic systems to tune the degree of disorder?
- Threshold and nature of RL as a glassy light phase: can they be experimentally detected?

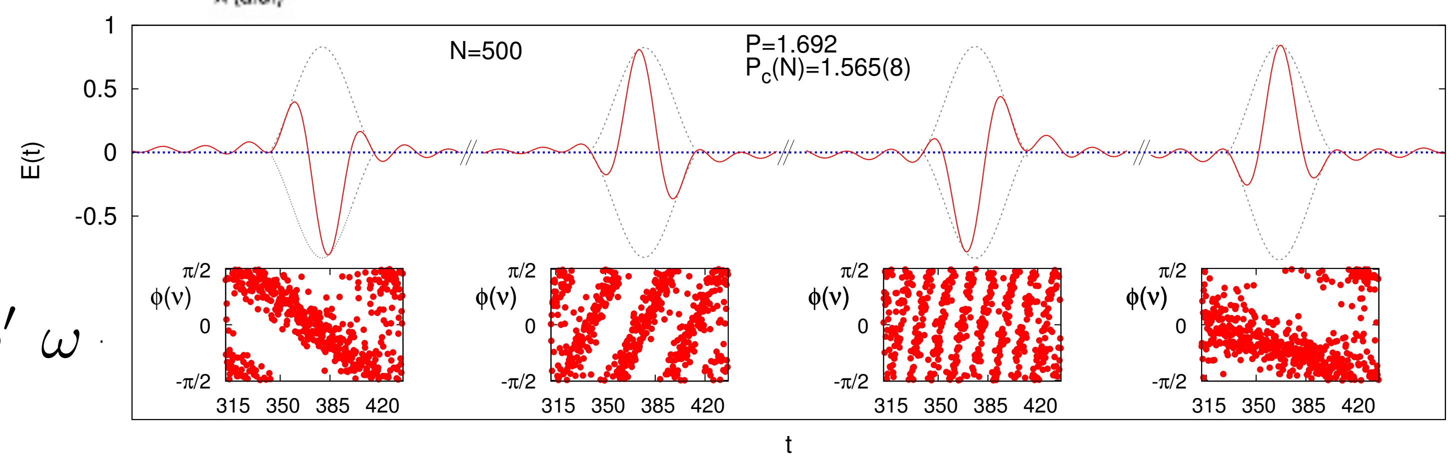
Standard ML (ordered limit)

We can go beyond narrow-band (and mean-field) approximation and study mode-locking systems with non-trivial gain profile by means of Monte Carlo simulations.



$$\mathcal{H} = -\sum_{k=1}^{N} g_k |a_k|^2 - J \sum_{\vec{k}_4}^{\mathrm{ML}} a_{k_1} a^*_{k_2} a_{k_3} a^*_{k_4}$$

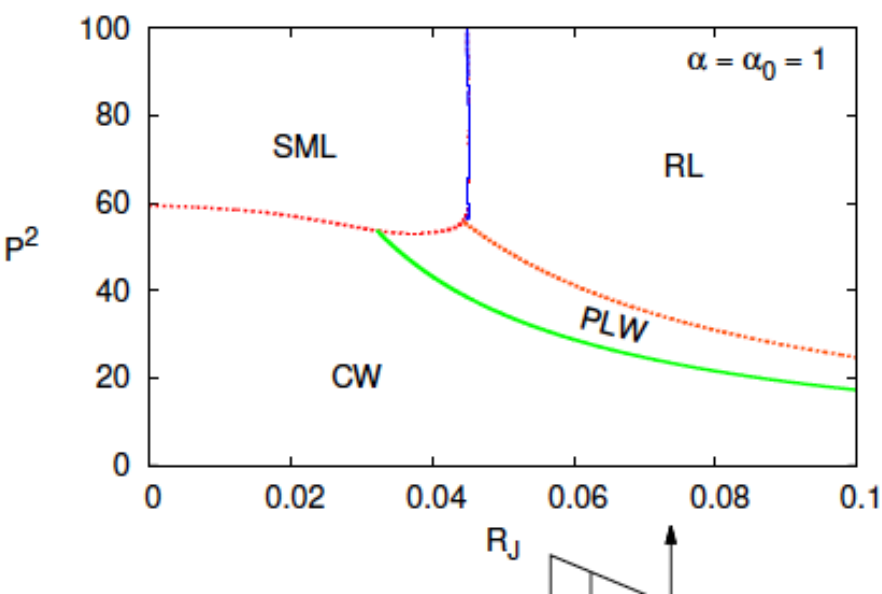
POSTER: Fabrizio Antenucci, Miguel Ibañez Berganza, LL, next room arXiv:1409.6345v1

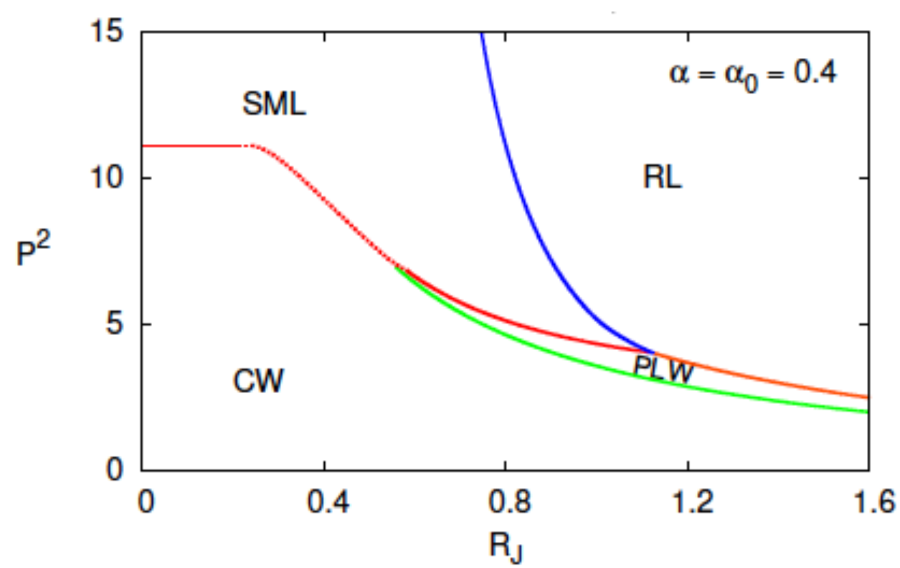


$$\phi(\omega) = \phi_0 + \phi' \, \omega$$

Order/disorder: glassy light – standard ML phase transition

- For a limited amount of disorder the standard ML structure holds. Up to a tolerance threshold.
- This threshold has never been detected in experiments so far since optically active systems (with or without cavity) display a fixed amount of disorder.
- How can we tune the degree of disorder in experiments?
  Taking photonic crystals and hammering them….?
  High Q factor but very expensive!





Glassy light – standard ML phase transition

Else using granular beads…?

Granular matter is unexpensive. Start with an array of millimetric beads and disorder them progressively by tapping. But… is it a laser?

Granular random laser

Is it a laser?

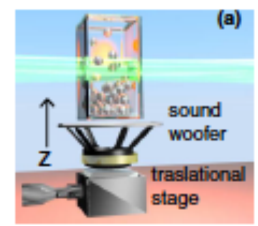
Yes! Metallic and glassy beads in Rhodamine B both display lasing as the pumped energy is high enough: ***granular random laser***

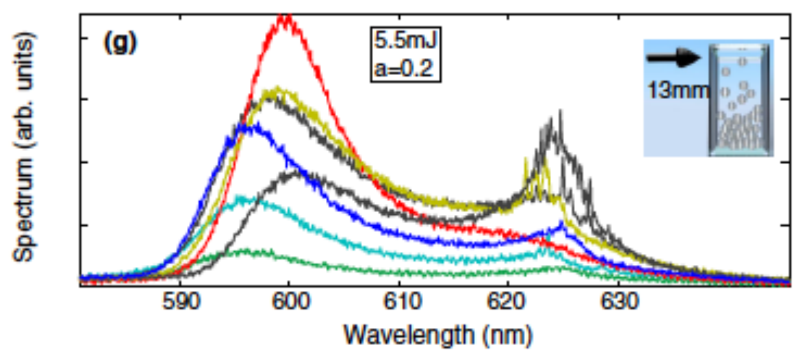
. 108, 248002 (2012) PHYSICAL REVIEW LETTERS week en 15 JUNE

Shaken Granular Lasers

Viola Folli,[1,2] Andrea Puglisi,[1,2] Luca Leuzzi,[3,2] and Claudio Conti[2,1]
[1]*ISC-CNR, Unit of University Sapienza, Piazzale Aldo Moro 5, 00185, Rome, Italy*
[2]*Department of Physics, University Sapienza, P.le Aldo Moro 5, 00185, Rome, Italy*
[3]*IPCF-CNR, Unit Kerberos of University Sapienza, Piazzale Aldo Moro 5, 00185, Rome, Italy*
(Received 3 February 2012; published 14 June 2012)

Granular materials have been studied for decades, driven by industrial and technological applications. These very simple systems, composed of agglomerations of mesoscopic particles, are characterized, in specific regimes, by a large number of metastable states and an extreme sensitivity (e.g., in sound transmission) to the arrangement of grains; they are not substantially affected by thermal phenomena, but can be controlled by mechanical solicitations. Laser emission from shaken granular matter is so far unexplored. Here we provide experimental evidence that laser emission can be affected and controlled by the status of the motion of the granular material; we also find that competitive random lasers can be observed. We hence demonstrate the potentialities of gravity-affected moving disordered materials for optical applications, and open the road to a variety of novel interdisciplinary investigations, involving modern statistical mechanics and disordered photonics.





Granular random laser (digression)

Yes! Metallic and glassy beads in Rhodamine B both display lasing as the pumped energy is high enough: ***granular random laser***

Shaking these granular lasers under various tapping accelerations and frequencies we find out that by optical investigation one can get relevant information about the inner structure of the granular material and its dynamics. Making it a random laser is a new probing technique for a granular system … *unfortunately* …

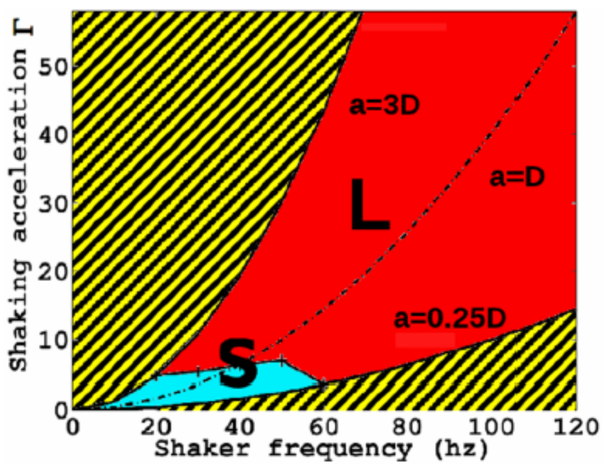


Figure 7 | Phase diagram of glass bead with $D = 1.0$ mm with Rhodamine (B). The region in red (dark gray) corresponds to the liquid-like (G) phase, related to the laser emission spectra oscillations, the region in blue (light gray) indicates the solid-like (S) phase. The region filled with

SCIENTIFIC REPORTS

OPEN

SUBJECT AREAS:
PHYSICS
OPTICS AND PHOTONICS
PHOTONIC DEVICES
MATERIALS SCIENCE

Time-resolved dynamics of granular matter by random laser emission

Viola Folli[1,2], Neda Ghofraniha[3], Andrea Puglisi[1,2], Luca Leuzzi[3,1] & Claudio Conti[1,2]

[1]Department of Physics, University Sapienza, Piazzale Aldo Moro, 5, 00185, Rome (IT), [2]ISC-CNR, UOS Roma Sapienza, Piazzale Aldo Moro 5, 00185, Rome (IT), [3]IPCF-CNR, UOS Roma Kerberos, Univ. Sapienza, Piazzale Aldo Moro 5, 00185, Rome (IT).

Received
17 April 2013

Accepted
4 July 2013

Published
22 July 2013

Because of the huge commercial importance of granular systems, the second-most used material in industry after water, intersecting the industry in multiple trades, like pharmacy and agriculture, fundamental research on grain-like materials has received an increasing amount of attention in the last decades. In photonics, the applications of granular materials have been only marginally investigated. We report the first phase-diagram of a granular as obtained by laser emission. The dynamics of vertically-oscillated granular in a liquid solution in a three-dimensional container is investigated by employing its random laser emission. The granular motion is function of the frequency and amplitude of the mechanical solicitation, we show how the laser emission allows to distinguish two phases in the granular and analyze its spectral distribution. This constitutes a fundamental step in the field of granulars and gives a clear evidence of the possible control on light-matter interaction achievable in grain-like system.

Strong disorder: glassy light

In real systems one can reproduce the same experiment several times on exactly the same realization of disorder and under the same "thermodynamic" conditions, i.e., constant heat-bath temperature and constant energy pumping: *real replicas*.

Each shot yields a replicated dynamics.

Dynamics variables $$a_k(\omega; t) = A_k(\omega; t) e^{\imath \phi_k(\omega; t)}$$

Strong disorder: glassy light

In real systems one can reproduce the same experiment several times on exactly the same realization of disorder and under the same "thermodynamic" conditions, i.e., constant heat-bath temperature and constant energy pumping: *real replicas*.

Each shot yields a replicated dynamics.

So far, we do not have access to complex amplitude configurations in experiments, only to intensity spectra

$$a_k(\omega;t) = A_k(\omega;t)e^{\imath\phi_k(\omega;t)}$$

?

Averaged over the whole dynamics

$$I_k(\omega) = \langle |a_k(\omega;t)|^2 \rangle_t$$

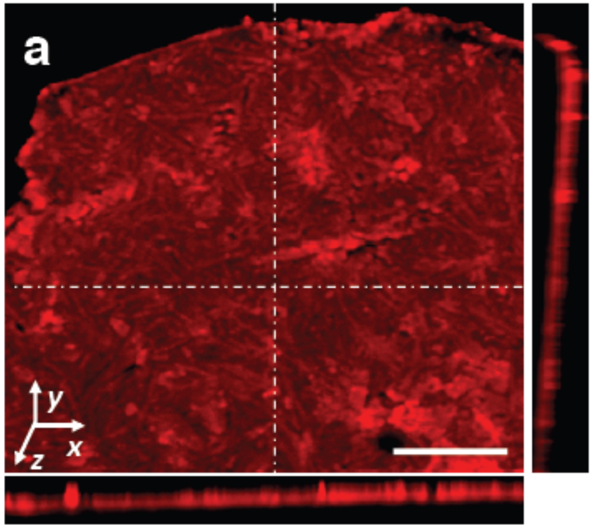
Strong disorder: glassy light

Real replicas. Each shot yields a replicated dynamics.

So far, we do not have access to complex amplitude configurations in experiments, only to intensity spectra.

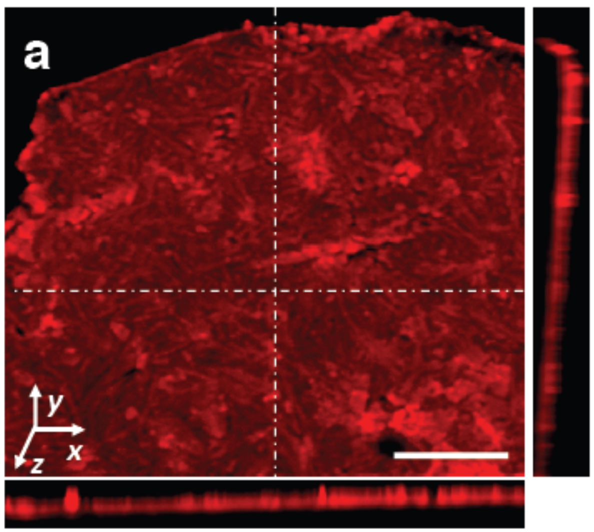
$$a_k(\omega;t) = A_k(\omega;t)e^{\imath\phi_k(\omega;t)} \; ?$$

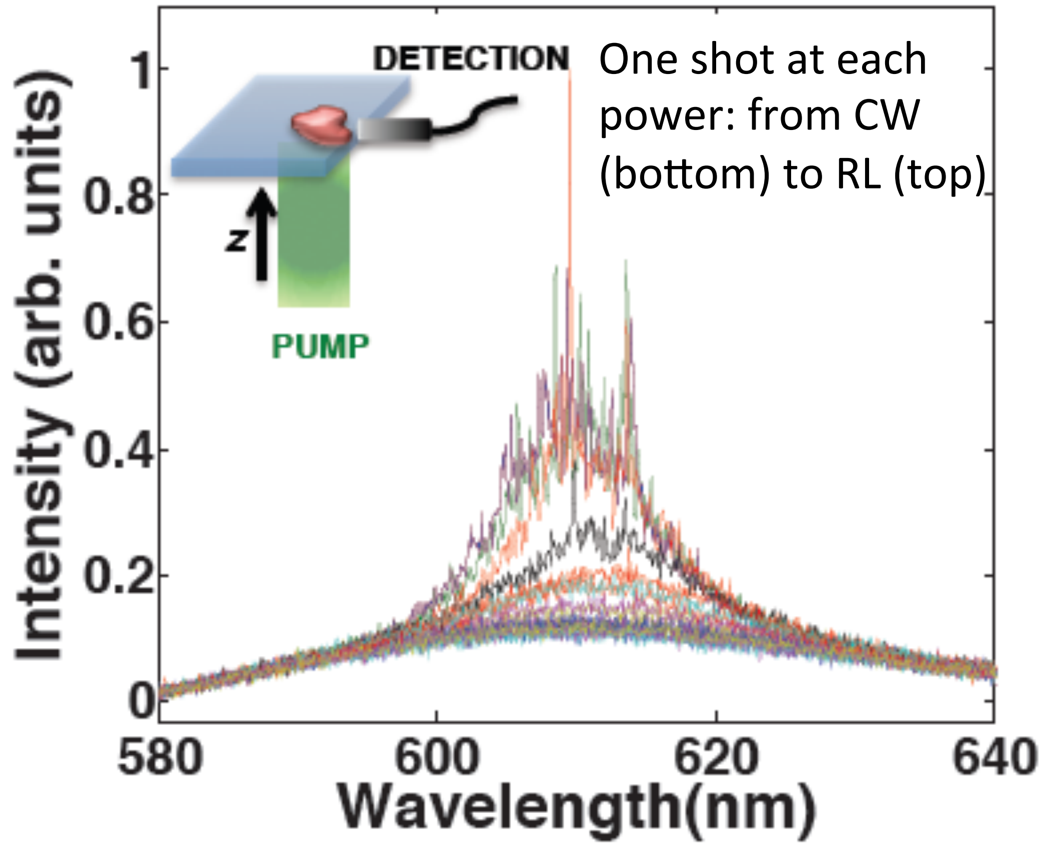
$$I_k(\omega) = \langle |a_k(\omega;t)|^2\rangle_t$$



In some random lasers intensity spectra vary very much from replica to replica: large shot-to-shot fluctuations. We look at the fluctuations of each replicated spectrum with respect to its average spectrum in a "functionalized thiophene based oligomer commonly named T5COx" and compute their normalized overlaps.

Strong disorder: glassy light

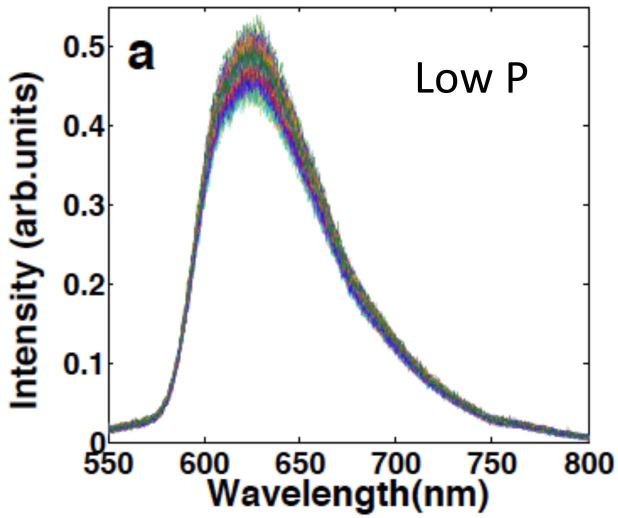


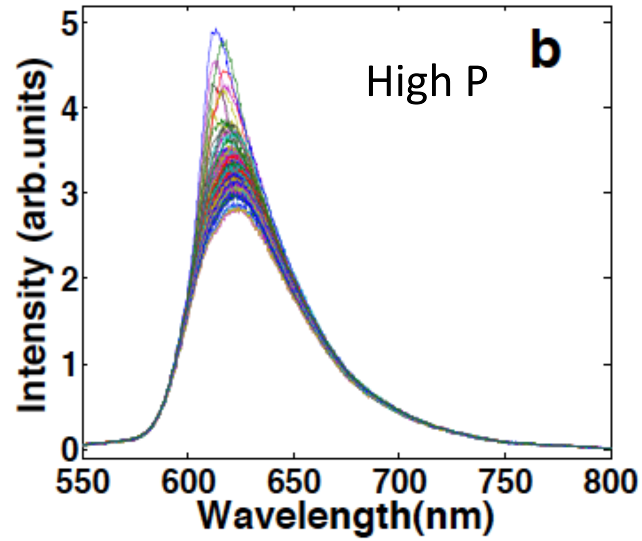


Resonant random laser
with large shot-to-shot spectral fluctuations: T5COx

arXiv:1407.5428v2 Neda Ghofraniha et al.

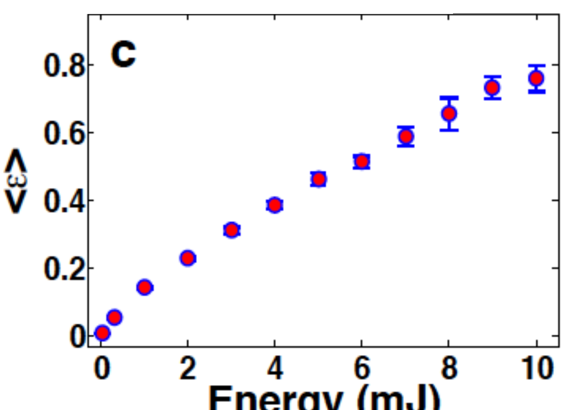
Lasing threshold and fluctuations



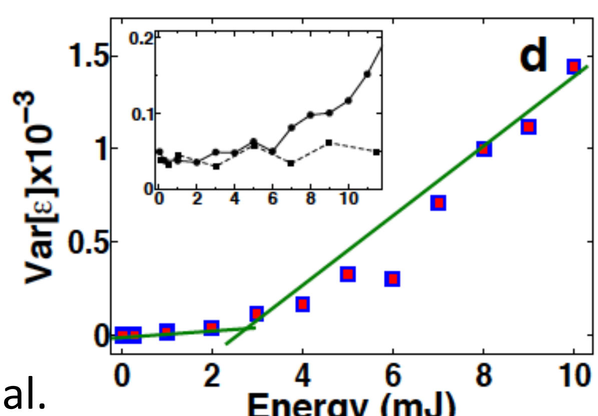


$$\epsilon_\alpha = \frac{1}{N}\sum_{k=1}^{N} I_\alpha(k)$$

$$\mathrm{Var}[\epsilon] = \frac{1}{\mathrm{N_s}}\sum_{\alpha=1}^{\mathrm{N_s}} (\epsilon_\alpha - < \epsilon_\alpha >)^2$$



arXiv:1407.5428v2
Neda Ghofraniha et al.

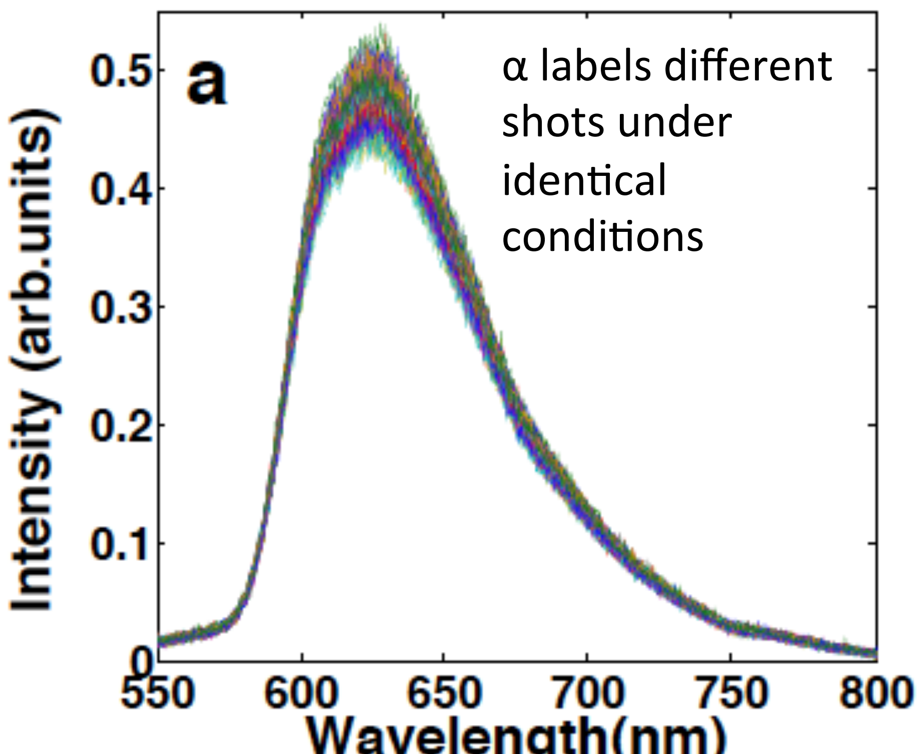


Intensity fluctuation overlap

$$I_k(\omega) = \langle |a_k(\omega;t)|^2 \rangle_t$$

$$\mathcal{Q}_{\alpha\beta} \equiv \frac{1}{N}\sum_{k=1}^{N} \Delta_k^{\alpha} \Delta_k^{\beta}$$

$$\Delta_k^{\alpha} \equiv \frac{I_k^{\alpha} - \bar{I}_k}{\sqrt{\sum_{k=1}^{N} \left(I_k^{\alpha} - \bar{I}_k\right)^2}}$$



α labels different shots under identical conditions

$$\bar{I}_k \equiv \frac{1}{N_R}\sum_{\alpha=1}^{N_R} I_k^{\alpha}$$

<-k labels intensity bins

arXiv:1407.5428v2
N. Ghofraniha et al.

Glassy light: intensity fluctuation overlap

Is this Q_{ab} related to the complex amplitude overlap q_{ab}?

$$\mathcal{Q}_{\alpha\beta} \equiv \frac{1}{N}\sum_{k=1}^{N} \Delta_k^\alpha \Delta_k^\beta \quad \overset{?}{\leftrightarrow} \quad q_{\alpha\beta} = \frac{1}{N}\sum_k a_k^\alpha a_k^{*\beta}$$

$$\Delta_k^\alpha \equiv \frac{I_k^\alpha - \bar{I}_k}{\sqrt{\sum_{k=1}^N \left(I_k^\alpha - \bar{I}_k\right)^2}} \simeq \mathcal{N}\left(I_k^\alpha - \bar{I}_k\right)$$

$$I_k(\omega) = \langle |a_k(\omega;t)|^2\rangle_t$$

Assuming that the normalization fluctuations are negligible, we can compute it the narrow-band mean-field model:

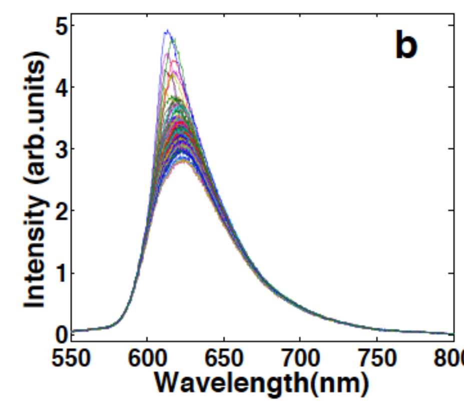
$$\mathcal{Q}_{ab} \equiv \frac{1}{N}\sum_k \left(|a_k^\alpha|^2 |a_k^\beta|^2 - \langle |a_k|^2\rangle^2\right)$$
$$\to \langle |a_k^\alpha|^2 |a_k^\beta|^2\rangle - \langle |a_k^\alpha|^2\rangle \langle |a_k^\beta|^2\rangle$$

In reality

(i) band is not narrow (many frequencies);

(ii) the acquired data correspond to (at least) several nanoseconds of emissions and have a finite wavelength refinement of 0.3nm: different modes of similar frequency appear in the same bin of the intensity spectrum;

(iii) Fluctuation normalization might be non-negligible.



Glassy light: intensity fluctuation overlap

$$\mathcal{Q}_{\alpha\beta} \equiv \frac{1}{N}\sum_{k=1}^{N} \Delta_k^\alpha \Delta_k^\beta \qquad \to \langle |a_k^\alpha|^2 |a_k^\beta|^2\rangle - \langle |a_k^\alpha|^2\rangle\langle |a_k^\beta|^2\rangle$$

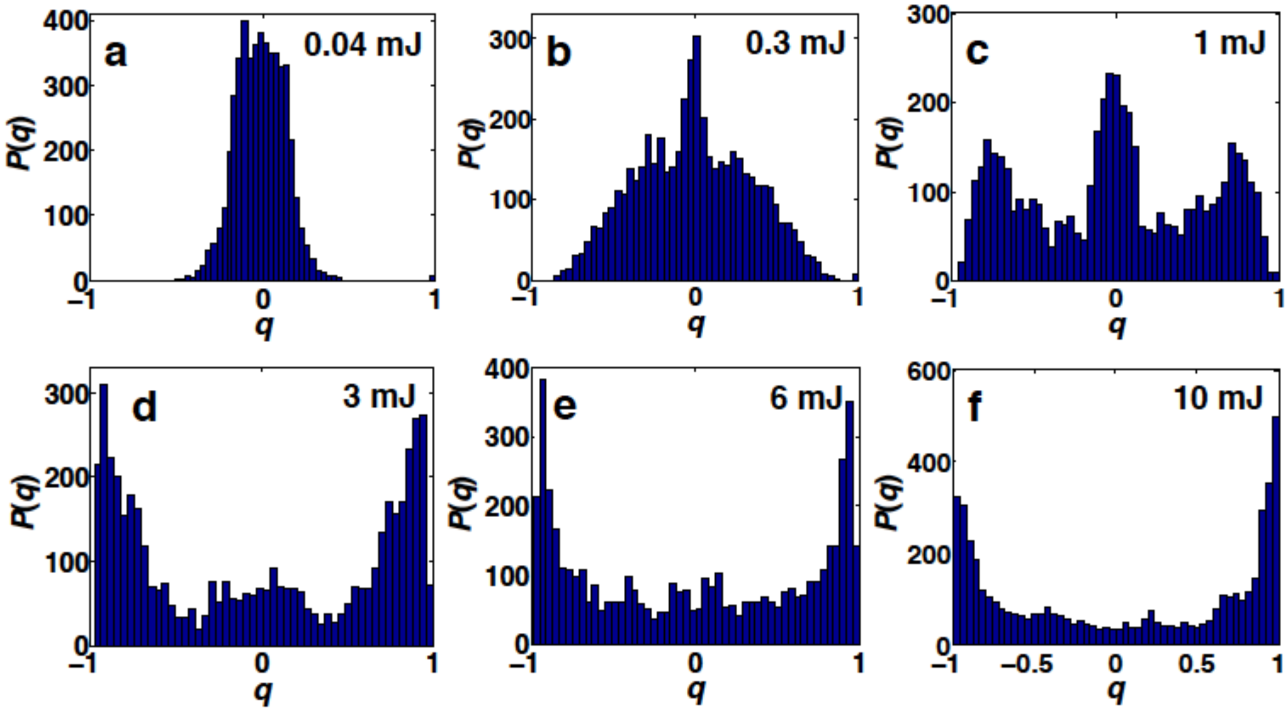
$$\mathcal{Q}_{\alpha\beta} = 8q_{\alpha\beta}^2 \qquad m = 0$$

Element-element relationship: any RSB on $q_{\alpha\beta}$ is detectable

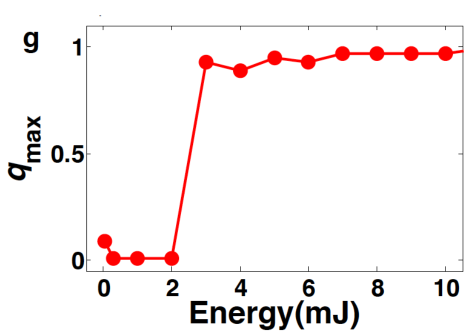
$$\mathcal{Q}_{\alpha\beta} = 2h(m)\left(\epsilon + 2r_d + 4\sum_{\gamma\neq\alpha} q_{\gamma\alpha}\right)^2 \left(2q_{\alpha\beta} - m^2\right) + 2\left(2q_{\alpha\beta} - m^2\right)^2$$

$$h(m) = 4k_2 m + 16k_4 m^3$$

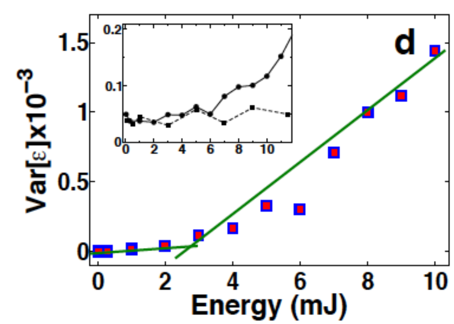
Experimental distribution of $\mathcal{Q}_{\alpha\beta}$



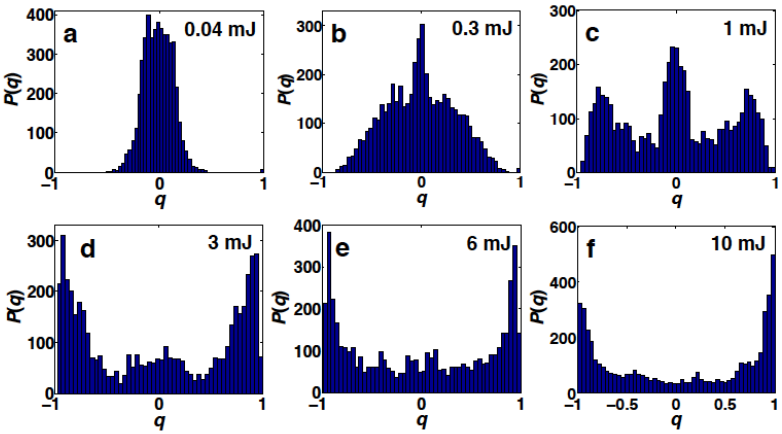
T5COx



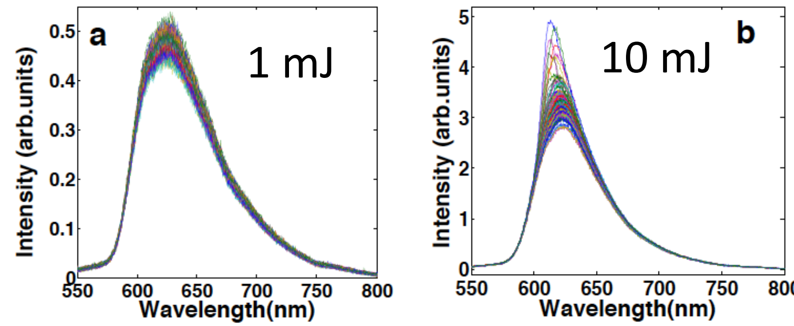
arXiv:1407.5428v2
N. Ghofraniha et al.



Experimental distribution of $\mathcal{Q}_{\alpha\beta}$



T5COx

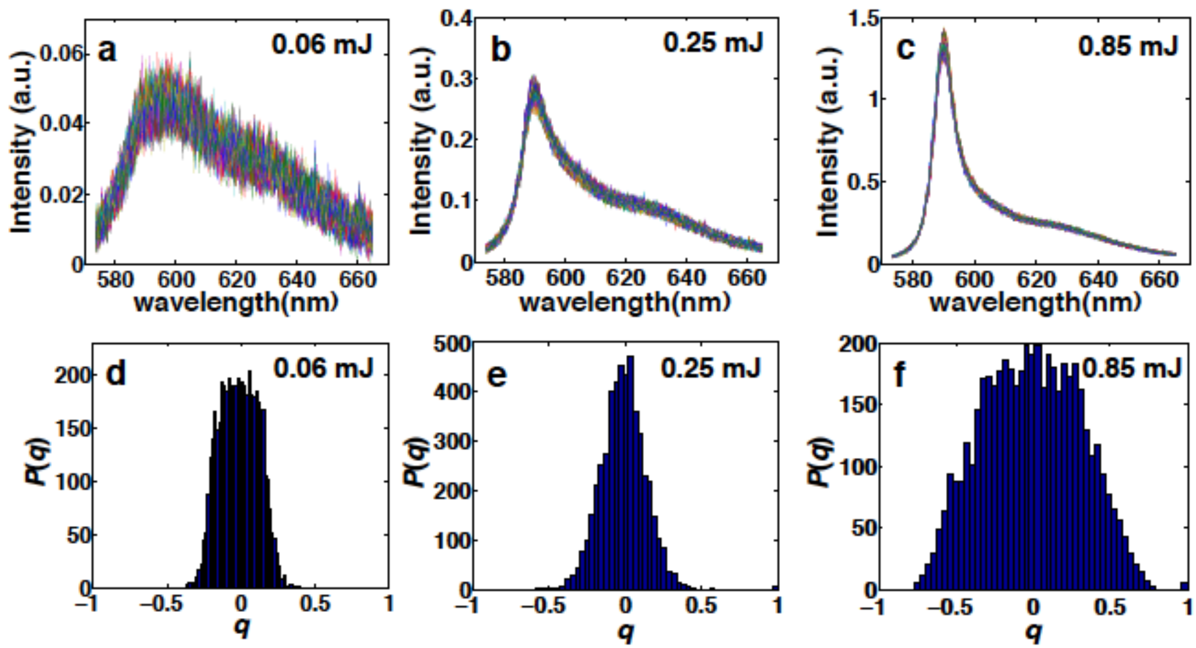


P_{th}≈2-3 mJ

arXiv:1407.5428v2
N. Ghofraniha et al.

TiO2 +
Rhodamine sol.

P_{th}≈0.25 mJ



Conclusions and perspectives

- Mean-field theory for standard and random mode-locking laser transition (new phase: “phase-locked paramagnet”)
- Realistic ML laser model (see Poster by Antenucci and Ibañez)
- Granular random laser: a probe for granular structure and dynamics
- Random laser as a glassy light phase: RSB-like

Stat Mech Laser perspectives

- Random laser beyond mean-field:

Monte Carlo simulations on GPU's for non-trivial gain profile, open cavity, correlated and uncorrelated quenched disorder-> RL pulses, phase delay, dynamics of spectra, direct reproduction of experimentally measured parameters.

- Probe of the $P(Q_{int\ flu})$ for different random laser compounds and in standard mode-locking laser.
- *Quenched amplitude approximation* (XY and clock model) study on diluted bipartite graphs (*mode-locking* vs Erdos-Renyi graphs).
- Inference problems in photonics: couplings reconstruction -> modes localizations