



DISORDERED SYSTEMS DAYS AT KING'S COLLEGE LONDON

Random lasers as complex disordered systems:
a spin-glass-like theory for amplified mode-locking waves in random media

King's College London
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+ Experimental collaborations with

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Dario Ballarini, Milena De Giorgi, Luisa De Marco, Giovanni Lerario, Daniele Sanvitto.

- Standard and random lasers
- Statistical physics approach to laser physics
 - Theory for ultrafast mode-locked multimode lasers (order, closed cavity)
 - Theory for random lasers: a mode-locked spin-glass theory (disorder, open cavity)
- The narrow-band solution, phase diagrams, replica symmetry breaking, a new overlap: intensity fluctuation overlap
- Intermezzo: the experimental measurement of the Parisi distribution of overlaps
- In between theory and experiment: a mode-locking model
 - Monte Carlo dynamics simulation with exchange Monte Carlo, GPU parallel computing
- Power distribution among modes in the glassy light regime: condensation vs equipartition at high pumping
- Outlook (work in progress)



- Standard and random lasers

- 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** :-) by Gordon Gould (1957, 1959).
 - Also terms Xaser, Uaser, ..., Raser.... :-/
- Laser can be single mode or **multimode**, continuous wave (laser pointer) or **pulsed** ("ns", "ps", "fs"), ...

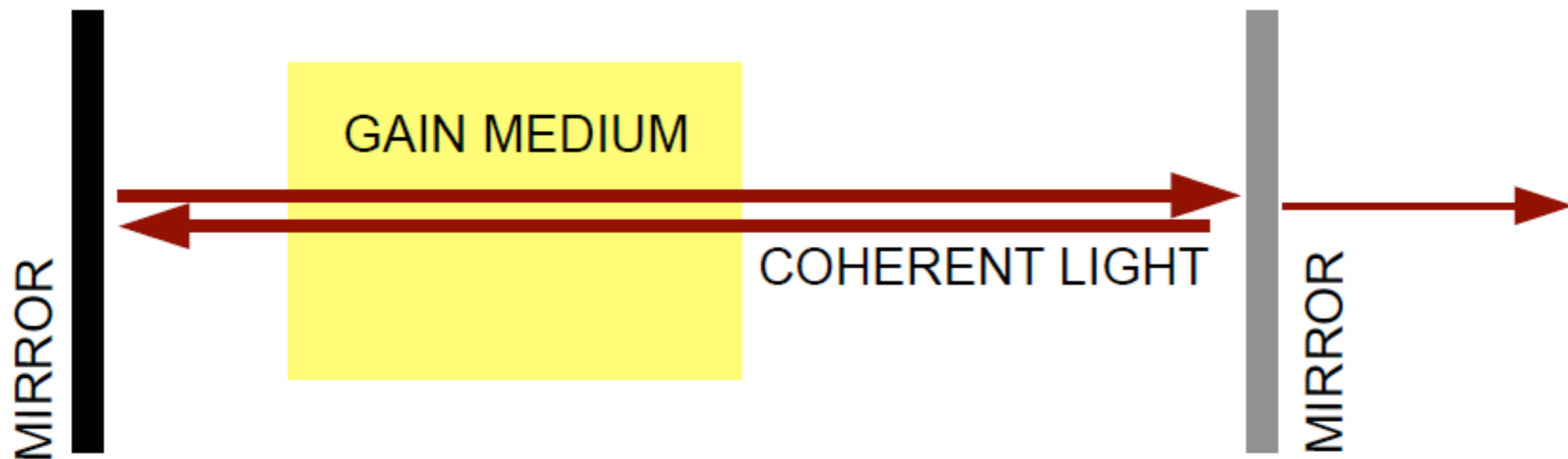


Ultrafast multimode Laser

Two essential components

- Cavity Coherent feedback
- Gain medium Amplification by Stimulated Emission
- Saturable absorber Passive mode-locking

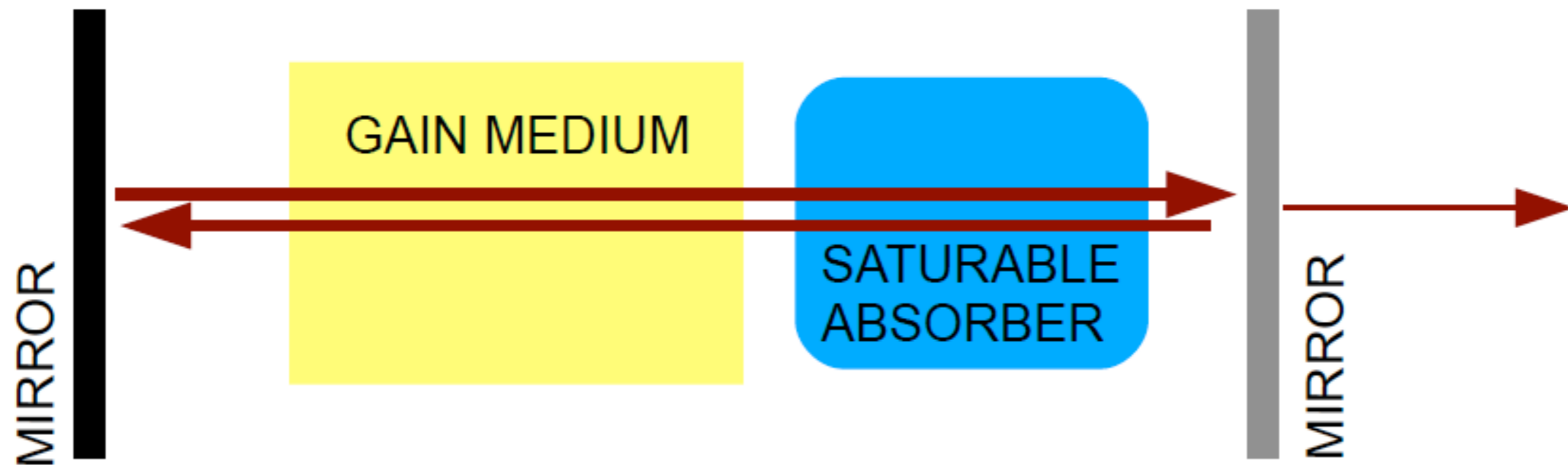
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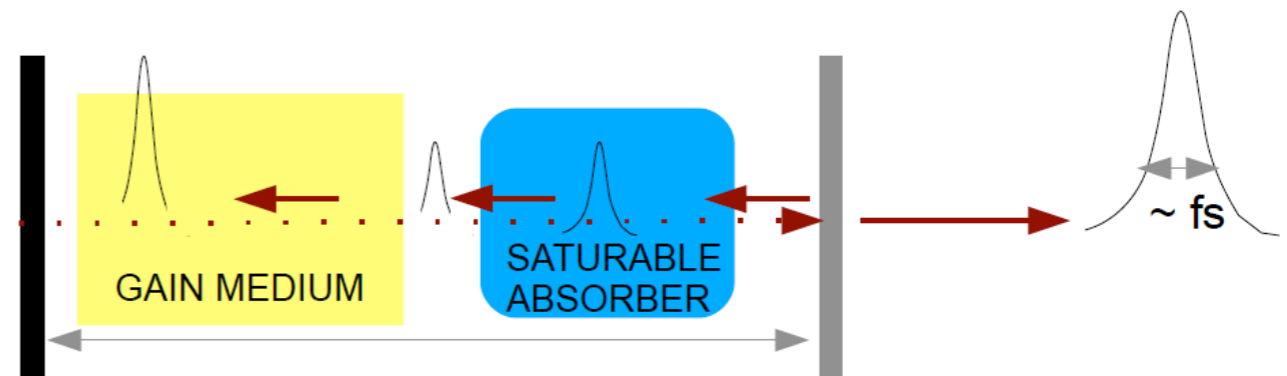
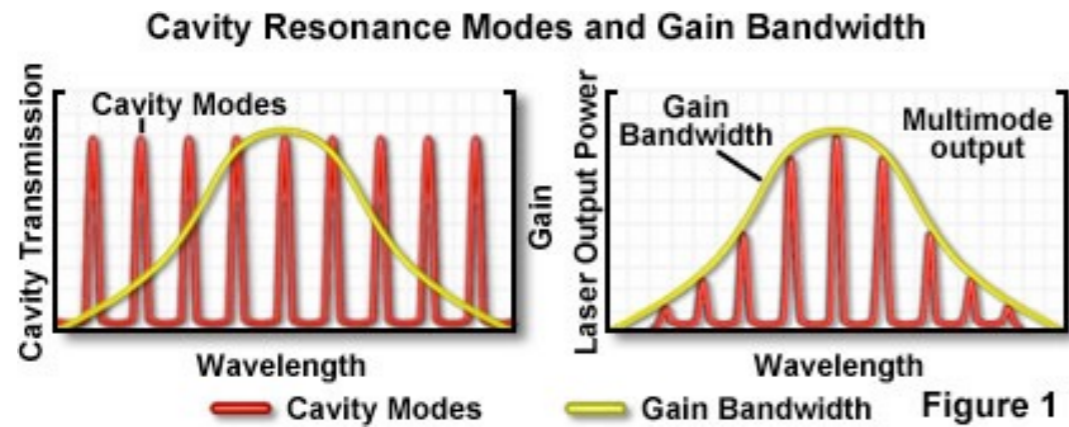
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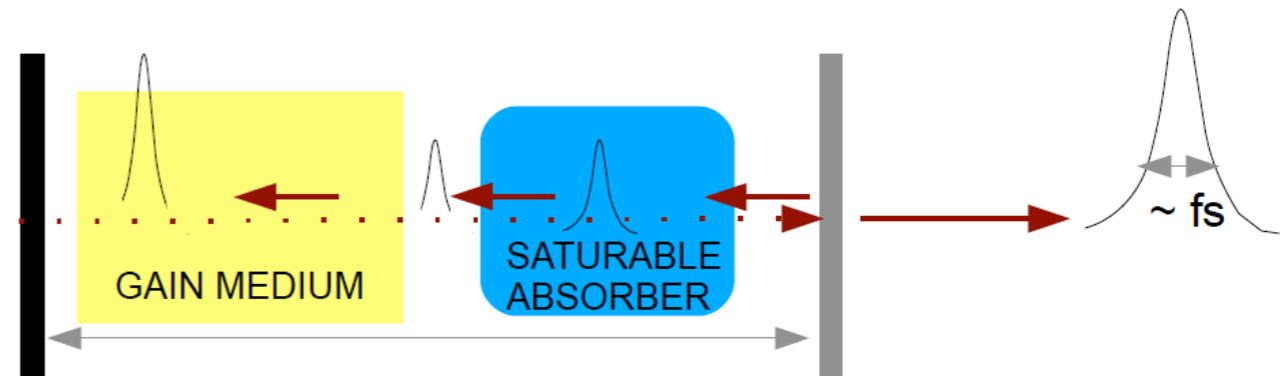
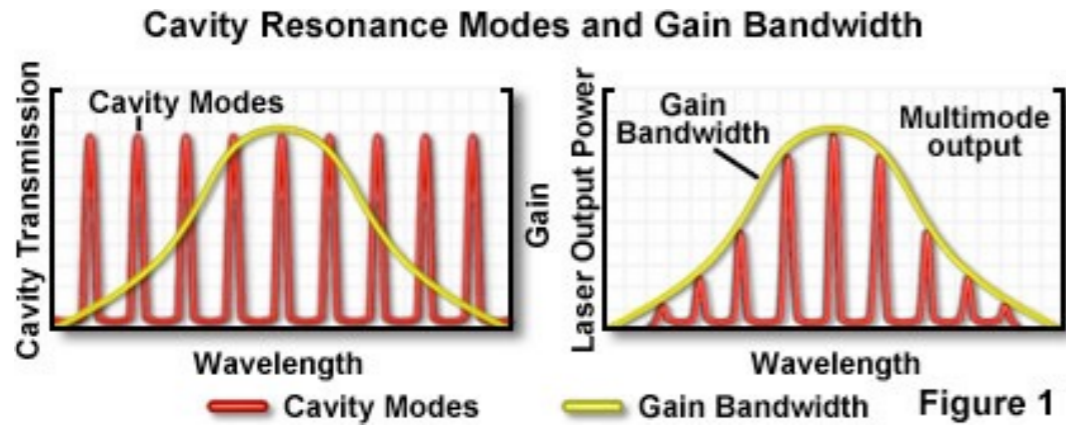
Ultrafast Multimode Laser



The **saturable absorber** induces self-starting **synchronous oscillations** of modes in the cavity: passive **mode-locking** \rightarrow fast pulses.

Related to a non-linear frequency matching condition occurring in the **saturable absorber**:

Ultrafast Multimode Laser

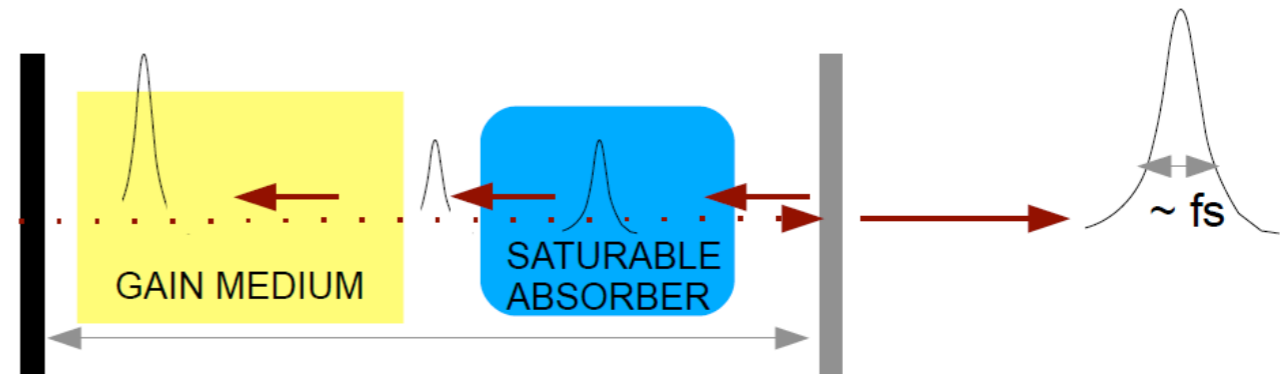
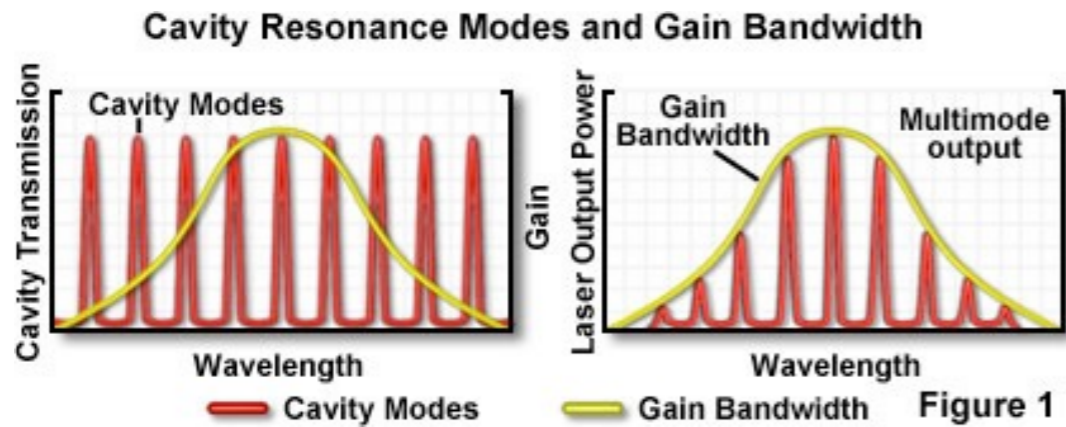


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Ultrafast Multimode Laser



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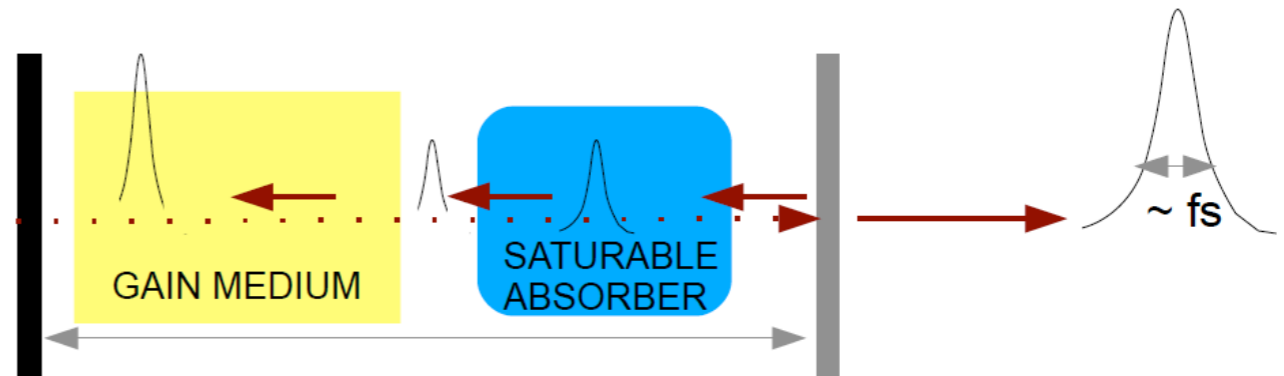
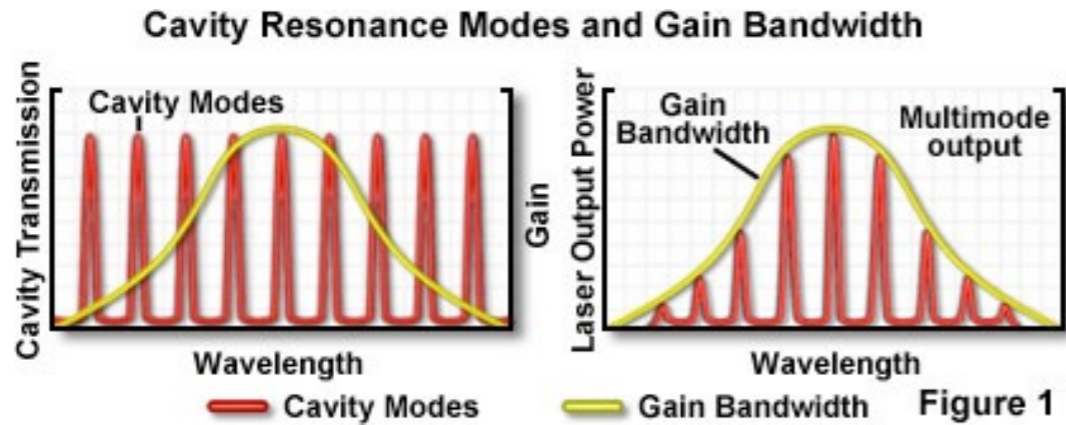
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In the lasing regime, the phases of the amplified modes acquire a linear relationship to the frequencies:

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

Ultrafast Multimode Laser



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Related to a non-linear frequency matching condition occurring in the **saturable absorber**:

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

Mode locking/Phase locking

takes place above the lasing optical power threshold.

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

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

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

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

ALL STANDARD MULTIMODE LASER THEORY SO FAR



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”.

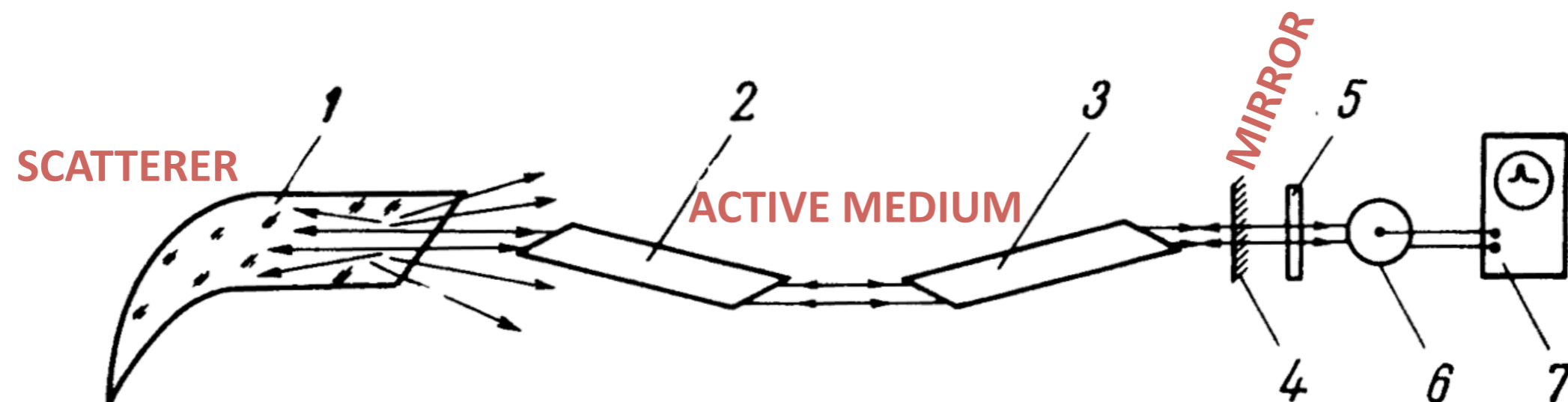


FIG. 1. Experimental setup—laser with scattering feedback.

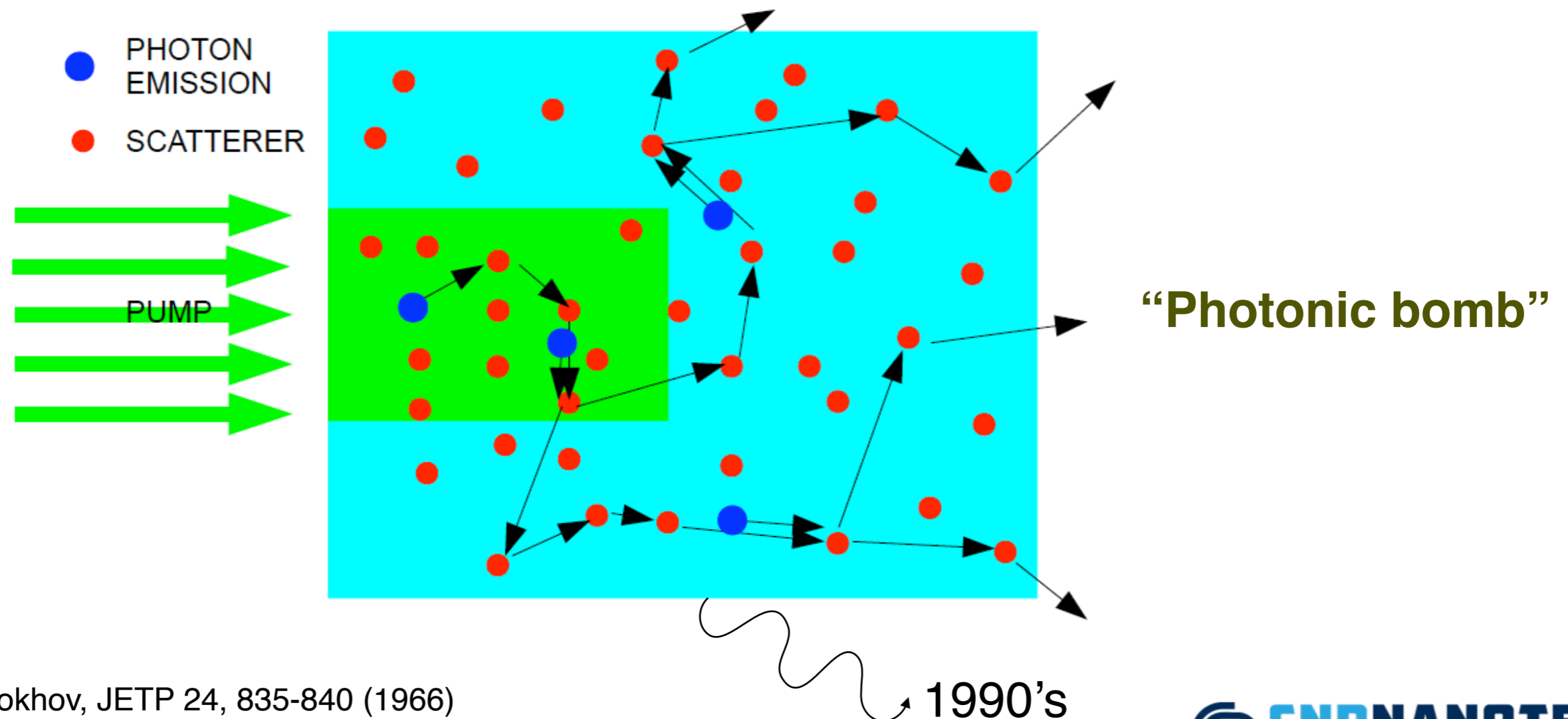
Ambartsumyan, Basov, Kryukov, Lethokov,
IEEE Journal of Quantum Electronics 2, 442 - 446 (1966)
JETP 24, 481-485 (1967)



Generation of Light by a Scattering Medium with Negative Resonance Absorption

Letokhov (1968)

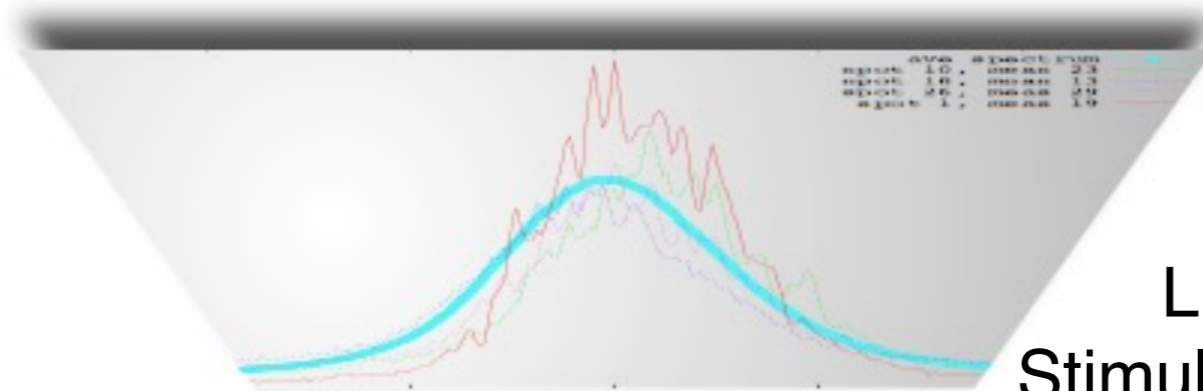
When photon path length is larger than amplification length:
photon multiplication



Letokhov, JETP 24, 835-840 (1966)

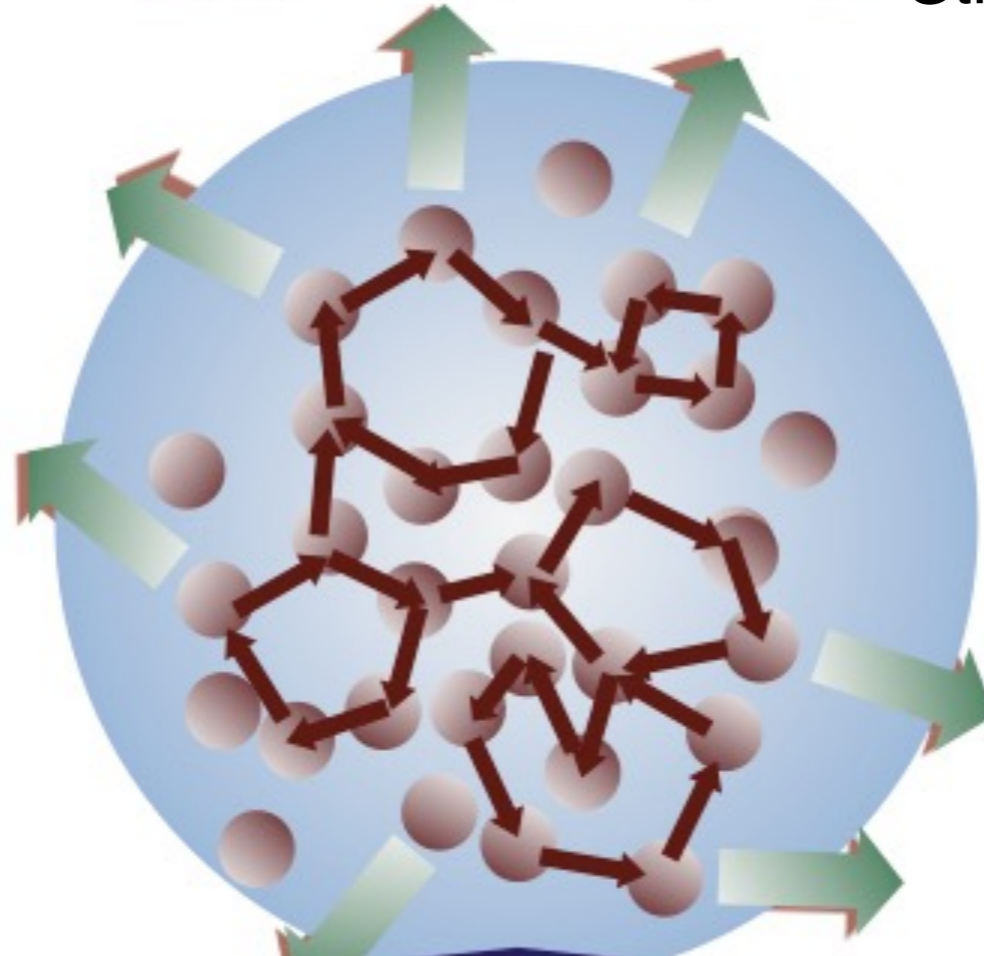


Random laser



Light Amplification by Stimulated Emission Radiation

Multiple scattering of photons



Gain prevails over loss: standing modes

Population inversion by external power pumping

H. Cao, *Waves in Random Media and Complex Media* 13, R1–R39 (2003)

D. S. Wiersma, *Nature Physics* 4, 359 (2008)

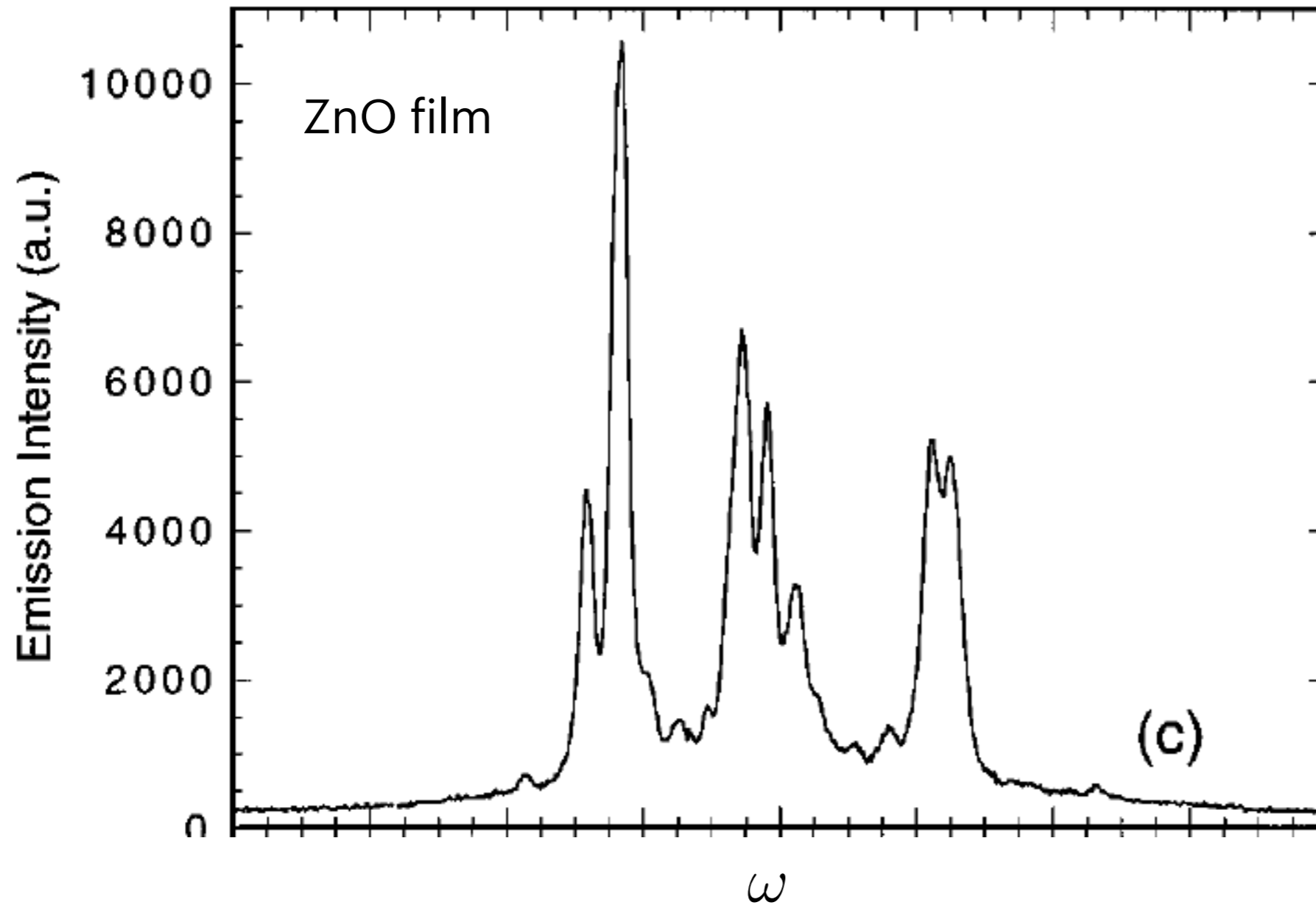
J. Andreasen et al. *Adv. Optics and Photonics* 3, 88–127 (2011)

A. S. Gomes et al. *Progress in Quantum Electronics* 78, 100343 (2021)

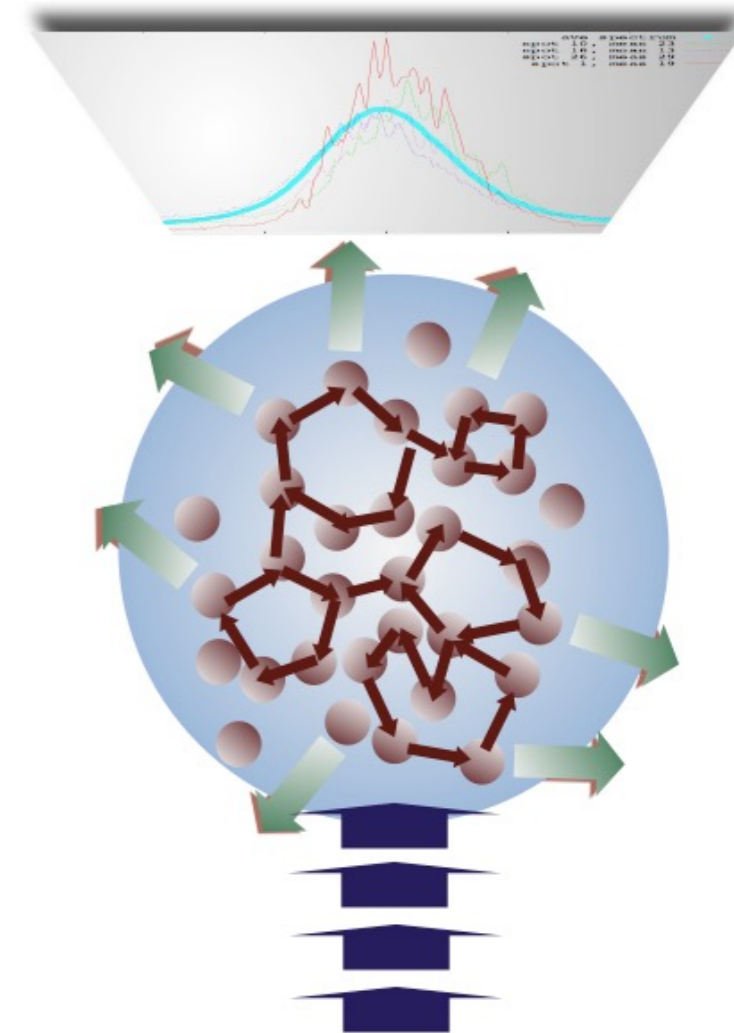


Random laser emission spectrum at high pumping

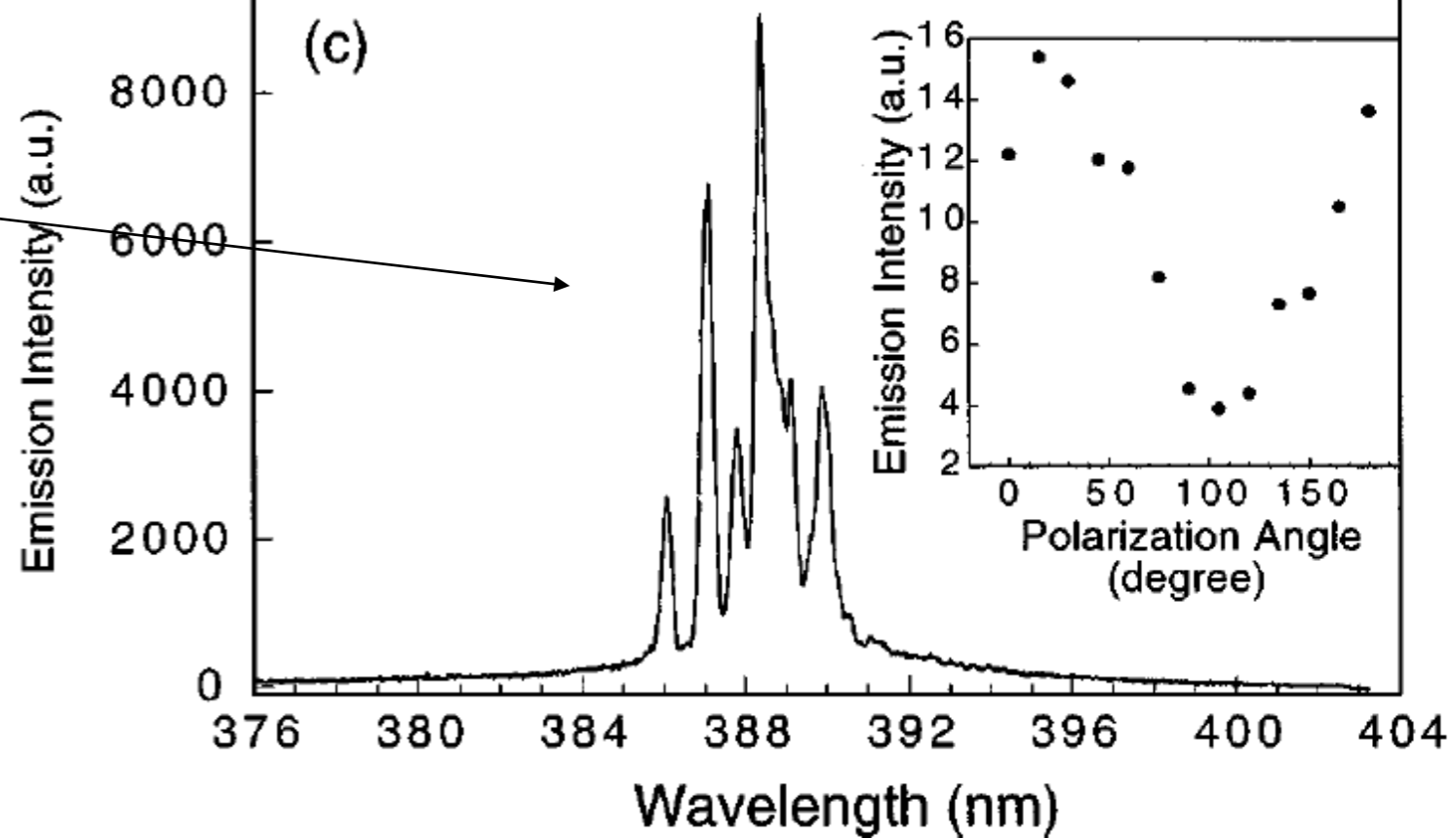
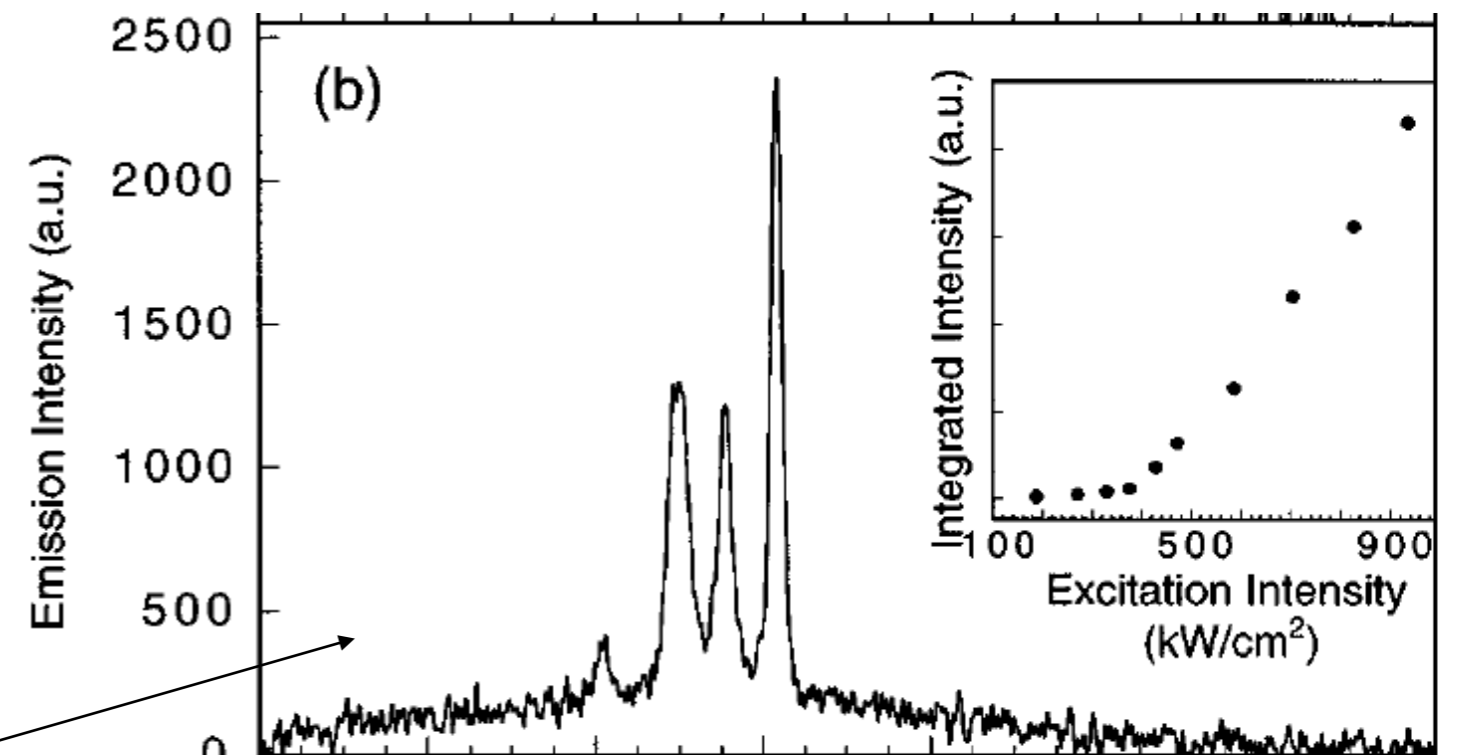
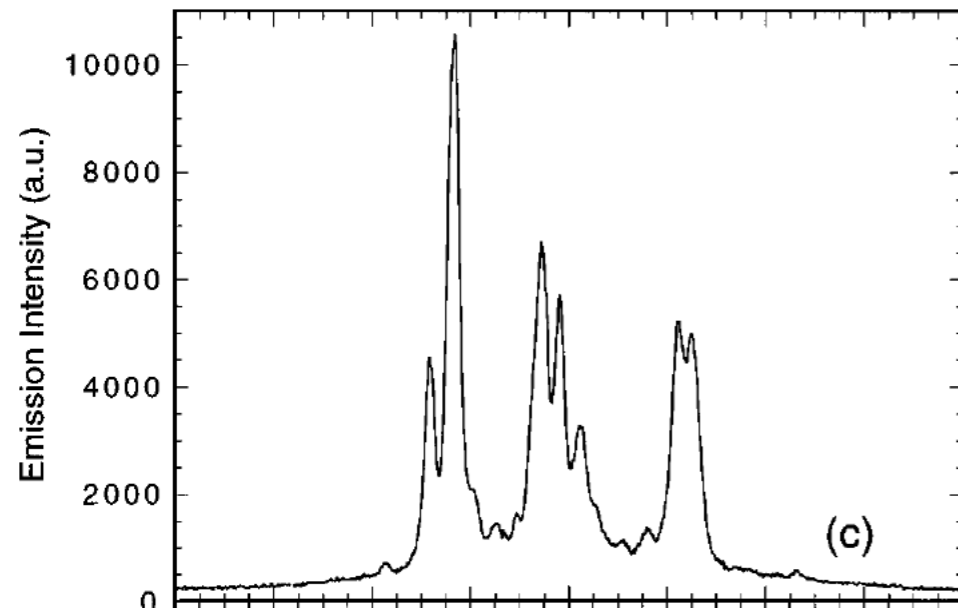
Film of ZnO



H. Cao et al., Appl. Phys. Lett. 73, 3656 (1998)



Random laser emission spectrum at high pumping



Changing the pumping power
resonances move

ZnO film

H. Cao et al.,

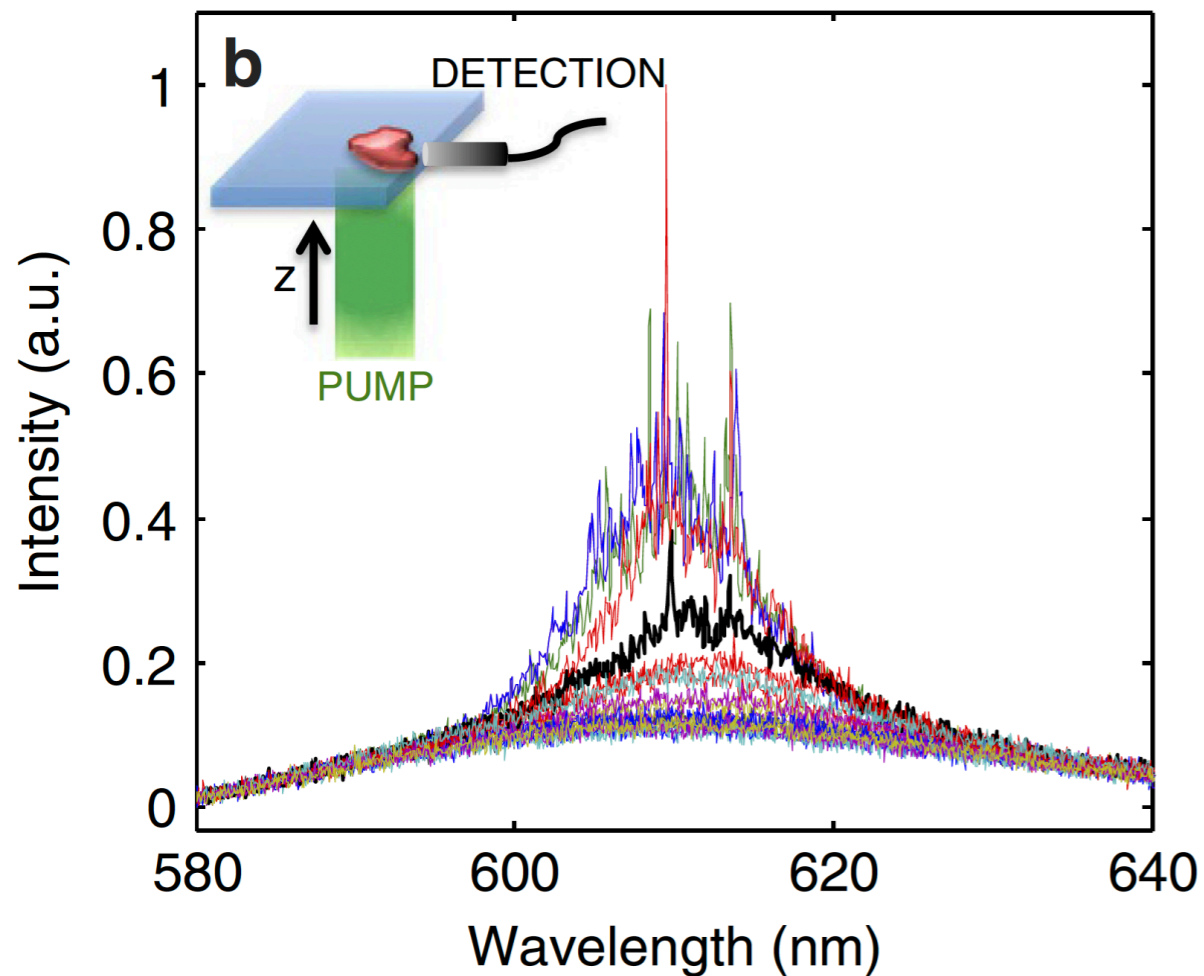
Appl. Phys. Lett. 73, 3656 (1998)



Random laser emission spectrum at high pumping

(In some random lasers) changing the pumping power the resonances move.
What happens at the emission after different shots at the same pumping power
(aka at each realization of the same random laser)?

Sometimes resonances change a lot

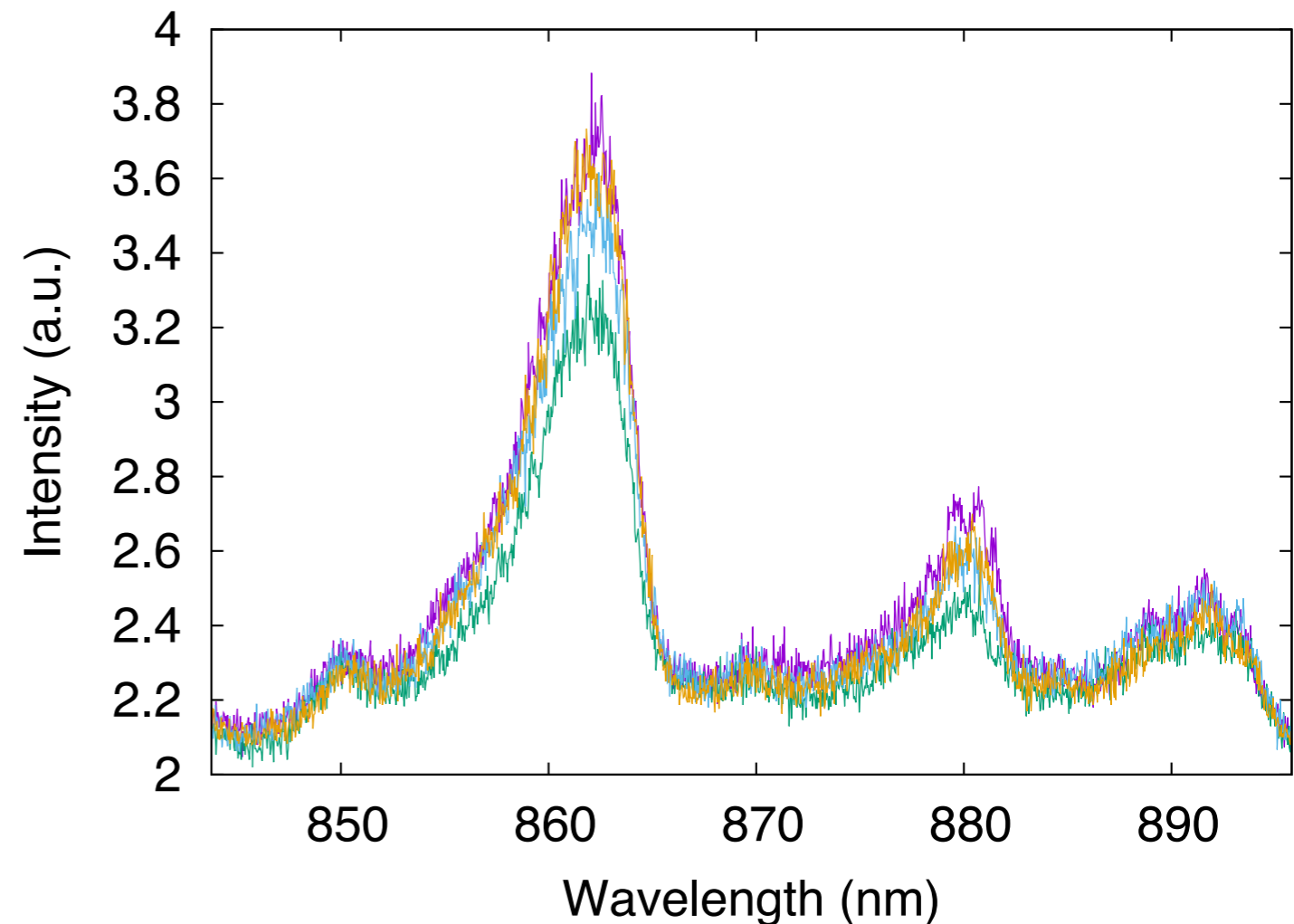


T5OCx
N Ghofraniha et al.,
Nat. Commun. 6, 6058 (2015)



T5OCx = thienyl-S,S-dioxide quinquethiophene

Sometimes they just oscillate a bit

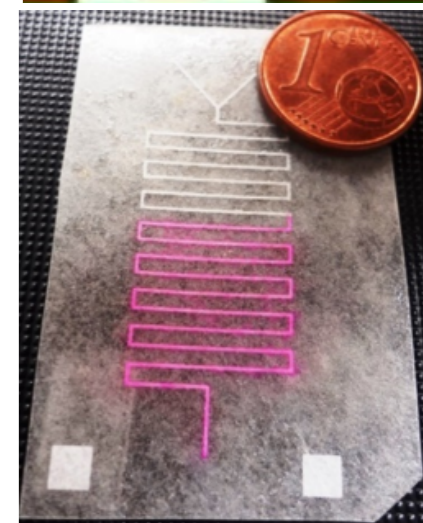
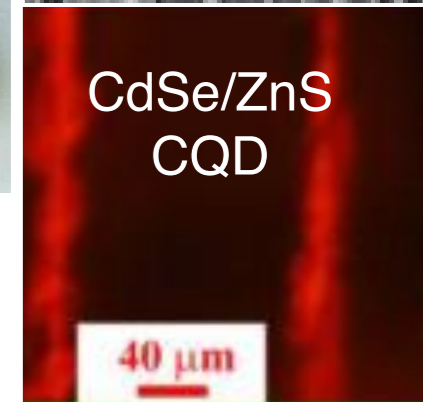
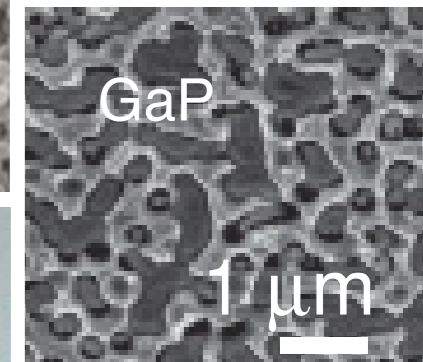
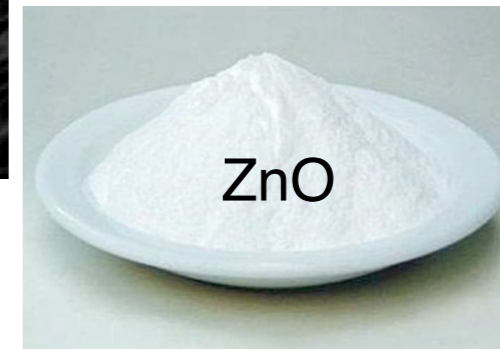
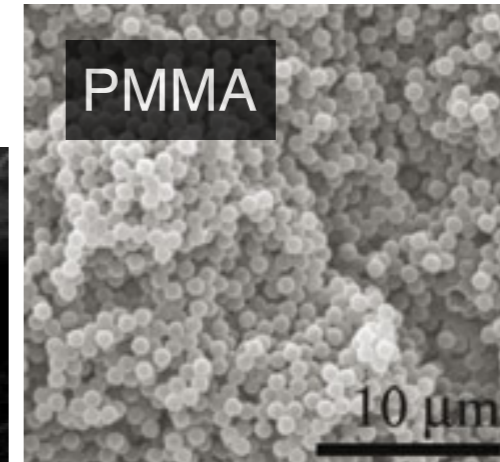
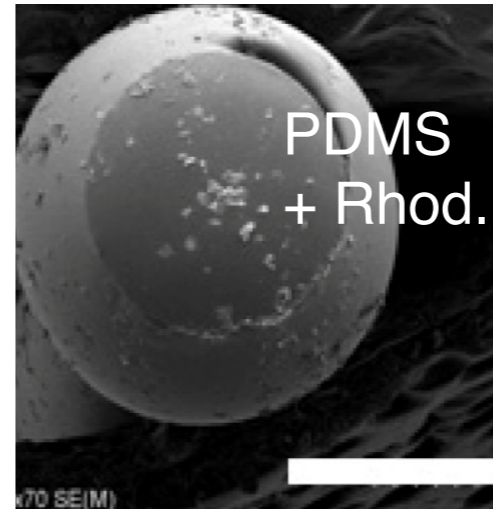


GaAs powder
F Antenucci et al.,
PRL 126, 173901 (2021)

Many random laser materials

Many kinds of random matrices for random lasers:

- Photonics glass [Galisteo-López et al. (2011)]
- **Nanoparticles powders** (TiO₂, ZnO, GaN, GaAs)
- Porous media, [El-Dardiry et al., (2010)]
- Plasmonic waveguides [Zhai et al. (2011)]
- Quantum dots [Chen et al. (2011)]
- Disordered fibers [de Matos et al. (2007); Turitsyn et al. (2010)],
- Polymeric micro channels [Bhaktha et al. (2012)],
- Micro droplets [Tiwari et al. (2012)],
- Granular beads [Folli et al. (2013)],
- Paper [Viola et al. (2013); Ghofraniha et al. (2013)]
- Bio inspired materials [Wang et al. (2014)]
- Organic semiconductors*, used for LEDs and as gain in lasers, among which:
 - Organic conjugated polymers, PPV, MEH-PPV, DOO-PPV, PF [Moses (1992); Hide et al. (1996); Tessler et al. (1996); Holzer et al. (1996); Frolov et al. (1996); Polson and Vardeny (2005); Tulek and Vardeny (2010)];
 - Solutions of laser dyes (rhodamine 6G) with nanoparticles (TiO₂, ZnO, GaN) [Cao et al. (2003); Wu et al. (2006); Polson and Vardeny (2005); Mujumdar et al. (2004)];
 - Organic-inorganic nanocomposites [Anglos et al. (2004)];
 - Organic nanofibers [Quochi et al. (2004, 2006); Andreev et al. (2006)];
 - **Thiophene-based oligomers** [Barbarella et al. (2005, 1999); Anni et al. (2004); Pisignano et al. (2002); Ghofraniha et al. (2013a)].



*Organic random lasers in *Organic lasers*, Viola et al. 2018

Many more since 2018.....



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- Statistical physics approach
to laser physics

Modeling multimode lasing with statistical mechanics

1) Electromagnetic field quantum dynamics can be mapped onto classical stochastic dynamics

Hackenbroich, Viviescas, Haken PRL, PRA 2003,
Antenucci, Crisanti, LL PRA 2015, Antenucci, Springer 2016.

2) Under stationarity conditions the system can be considered as if at equilibrium, coupled to an effective “thermal” *reservoir*

Ordered lasers: Gordon, Fisher PRL 2002, Opt. Commun. 2003;
Gat, Gordon, Fisher, PRE 2004; Weill et al PRL 2005;
Antenucci, Ibanez Berganza, LL PRA, PRB 2015; Marruzzo, LL PRB 2015.

Random lasers (quenched homogeneous amplitudes — phase only):
Angelani et al. PRL, PRB 2006; LL et al. PRL 2009; Conti, LL PRB 2011;

Random lasers: Antenucci, Conti, Crisanti, LL PRL 2015; Antenucci, Crisanti, LL SciRep 2015;
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- Statistical physics approach
to laser physics

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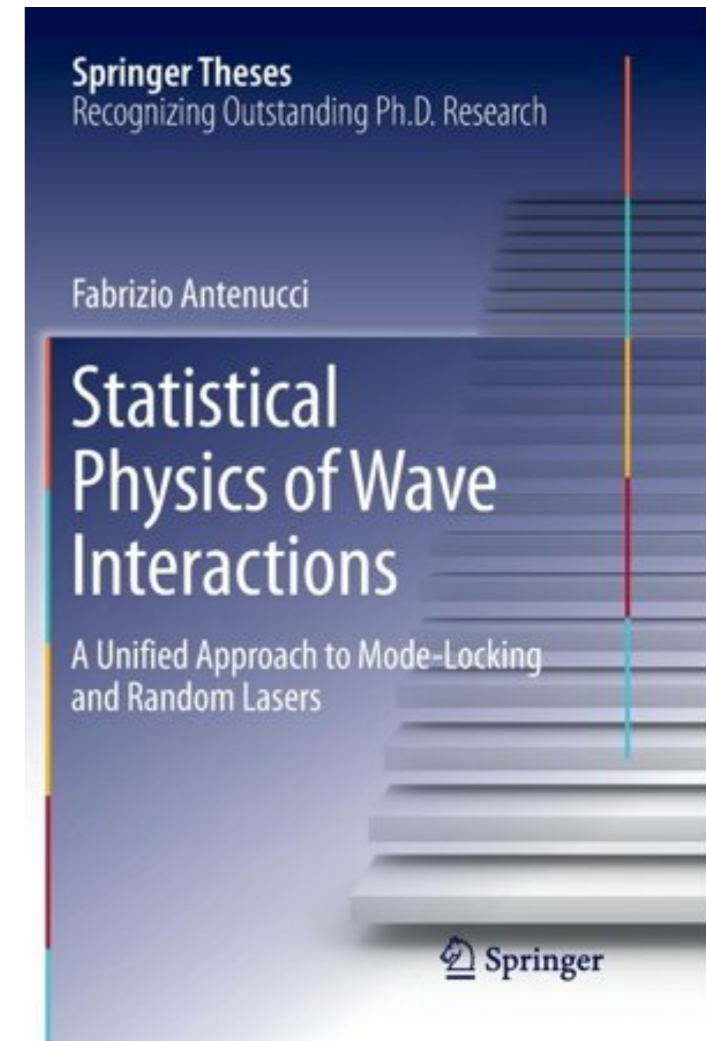
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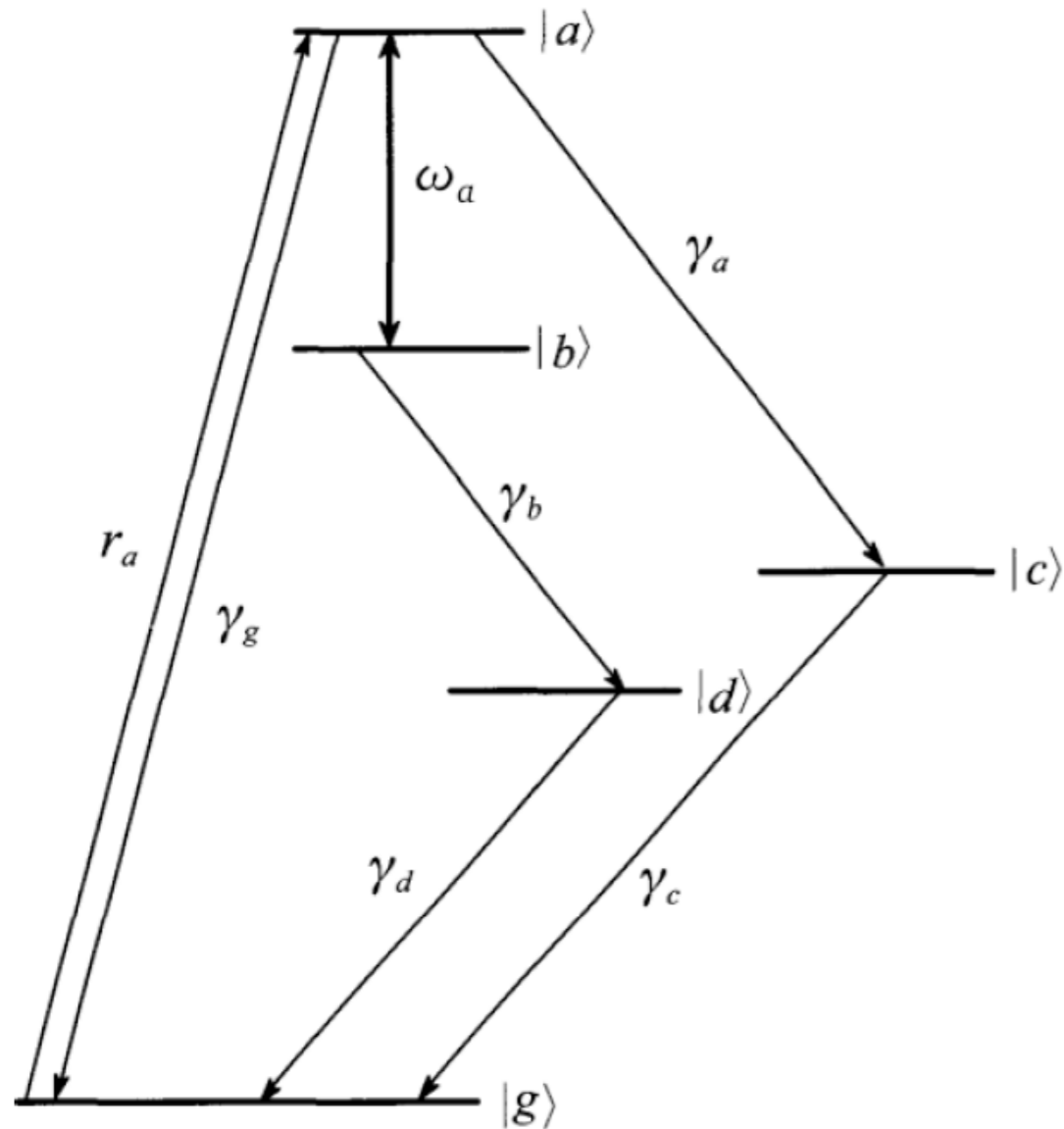
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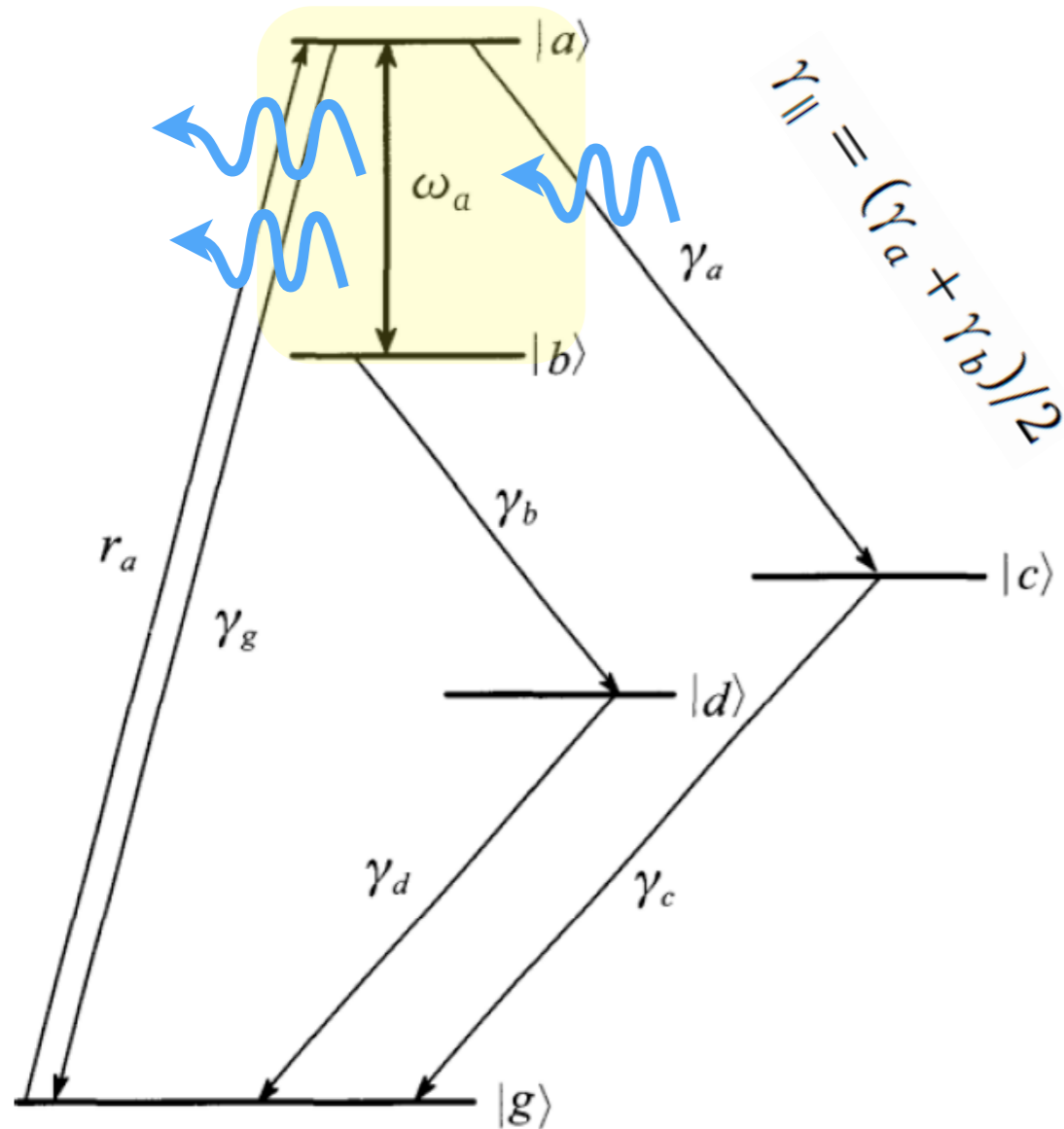
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- Statistical physics approach
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RADIATION

$a_\lambda, a_\lambda^\dagger$
creation, annihilation operators of the e.m. field

MATTER

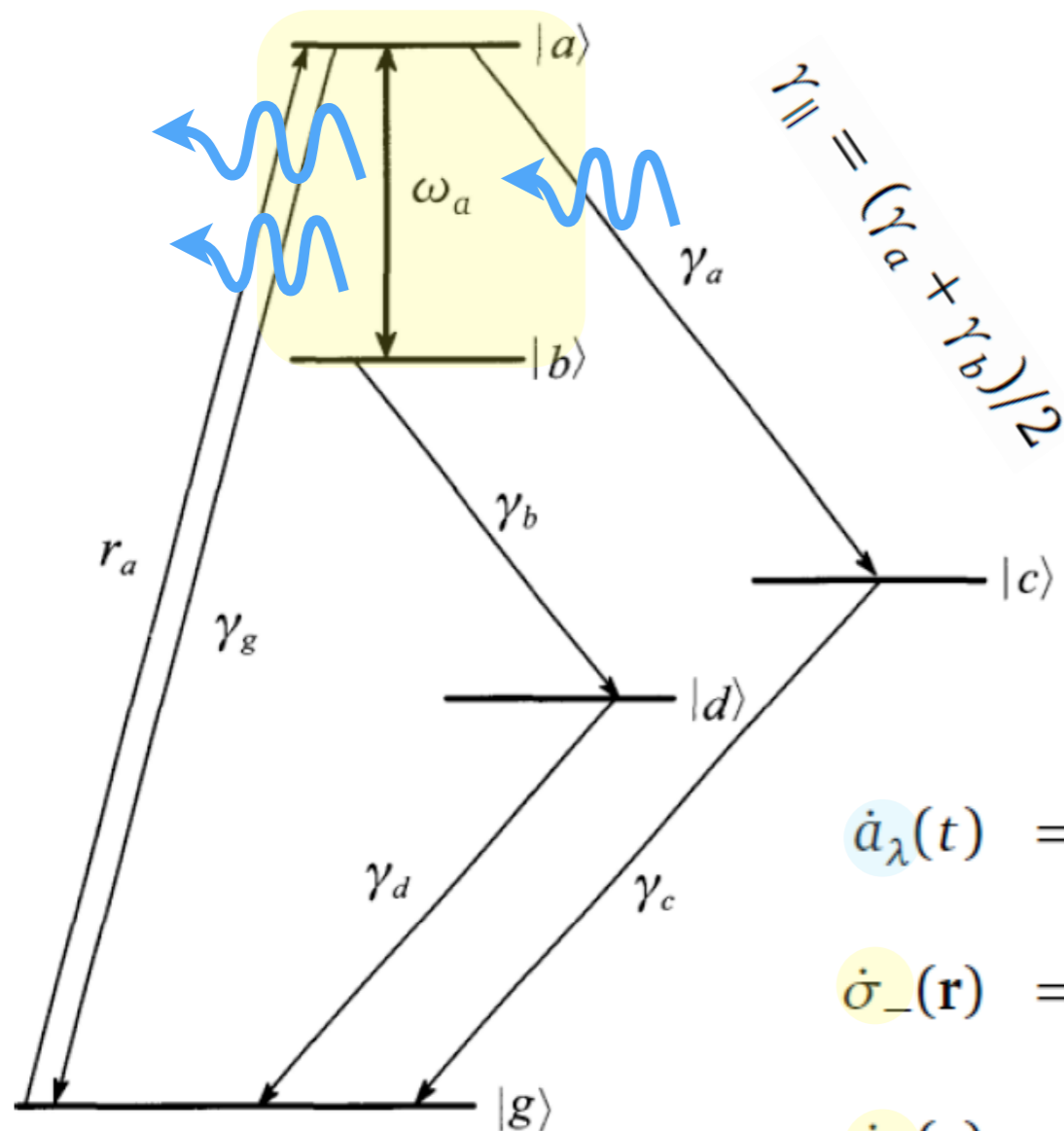
$\sigma_-(\mathbf{r}) \equiv |b\rangle\langle a|$
lowering

$\sigma_+(\mathbf{r}) \equiv |a\rangle\langle b|$
raising

$\sigma_z(\mathbf{r}) \equiv |a\rangle\langle a| - |b\rangle\langle b|$
population inversion

- Statistical physics approach to laser physics

Jaynes-Cummings quantum stochastic dynamics for light-matter interaction



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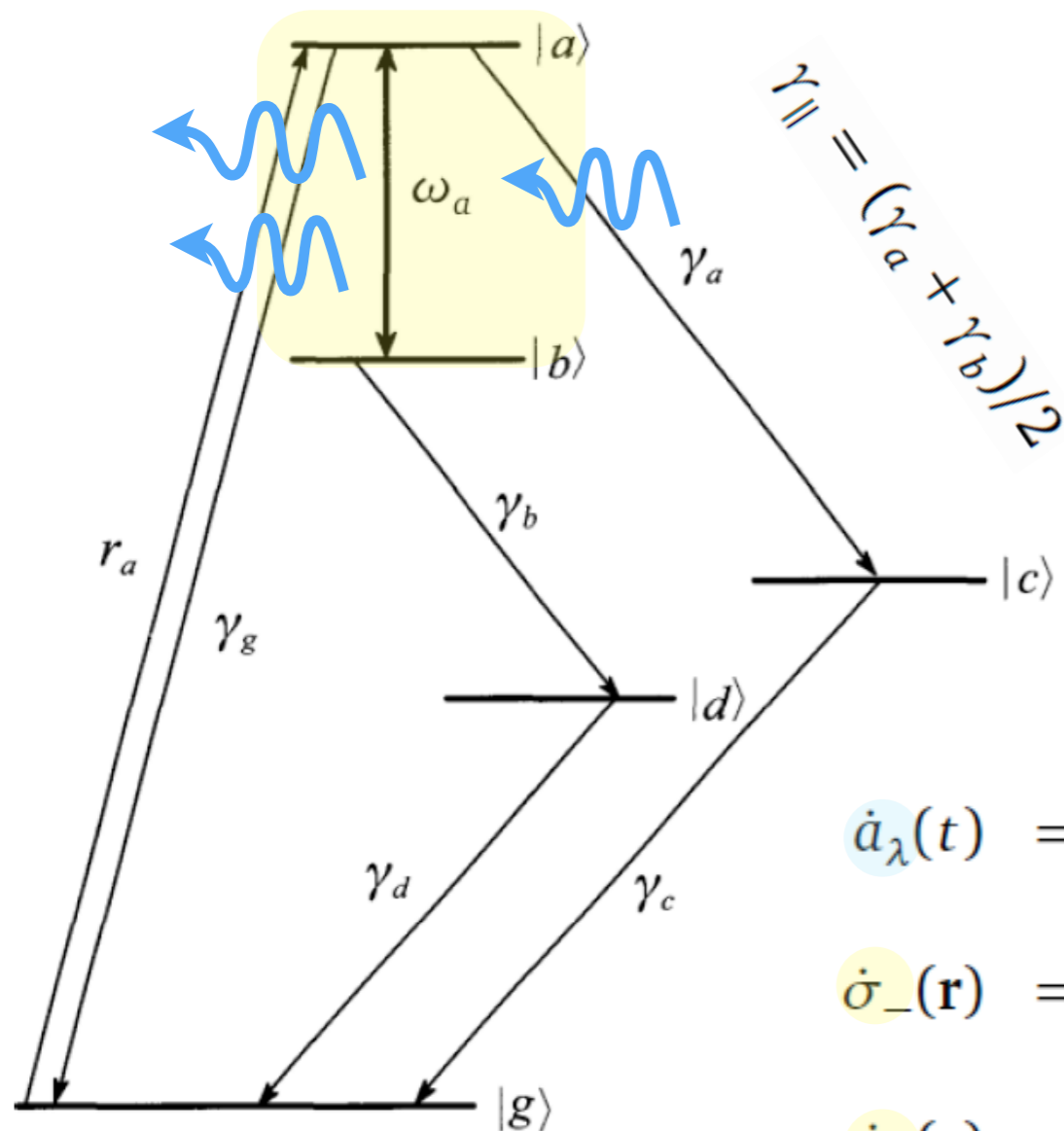
$$\dot{a}_\lambda(t) = -i\omega_\lambda a_\lambda(t) + \int d\mathbf{r} g_\lambda^*(\mathbf{r}) \sigma_-(\mathbf{r}) - \sum_{\lambda'} \Gamma_{\lambda\lambda'} a_{\lambda'}(t) + F_\lambda(t)$$

$$\dot{\sigma}_-(\mathbf{r}) = -(\gamma_\perp + i\omega_a) \sigma_-(\mathbf{r}) + 2 \sum_\lambda g_\lambda(\mathbf{r}) \sigma_z(\mathbf{r}) a_\lambda + F_-(\mathbf{r})$$

$$\dot{\sigma}_z(\mathbf{r}) = \gamma_\parallel (S\rho(\mathbf{r}) - \sigma_z(\mathbf{r})) - \sum_\lambda [g_\lambda^*(\mathbf{r}) a_\lambda^\dagger \sigma_-(\mathbf{r}) + h.c.] + F_z(\mathbf{r})$$

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Downgrading operators to complex numbers we obtain a classical
description of the mode amplitudes stochastic dynamics

- Statistical physics approach
to laser physics

Random laser stochastic differential equations
for the complex amplitudes (the *phasors*)

$$\dot{a}_n(t) = \mathcal{F}_n[\{a\}|\{J\}] + \eta_n(t) \quad n = 1, \dots, N$$

T is related to spontaneous emission, i.e., to the real temperature of the system

Potential solution to the
Fokker-Planck equation

Boltzmann-Gibbs like distribution of the amplitudes' configurations,
at some effective temperature

Ordered lasers:

Gordon, Fisher PRL 89, 103901 (2002); Opt. Commun. 223, 151 (2003); Gat, Gordon, Fisher, PRE 70, 046108 (2004);

Random lasers, quenched homogeneous amplitude approx:

L Angelani et al. PRL 96, 065702 (2006); PRB 74, 104207 (2006), LL et al. PRL 102, 083901 (2009), Conti, LL PRB 83, 134204 (2011)

Random lasers, phasors:

F Antenucci, C Conti, A Crisanti, LL, PRL 114, 043901 (2015); F Antenucci, A Crisanti, LL, PRA 91, 053816 (2015), F Antenucci et al Phil. Mag. 96, 704-731 (2016), F. Antenucci, Springer 2016



- Statistical physics approach
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Random laser stochastic differential equations
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$$\dot{a}_n(t) = \mathcal{F}_n[\{a\}|\{J\}] + \eta_n(t) \quad n = 1, \dots, N$$

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

T is related to spontaneous emission, i.e., to the real temperature of the system

Potential solution to the
Fokker-Planck equation

$$\mathcal{F}_n[\{a\}|\{J\}] \sim -\frac{\partial}{\partial a_n^*} \mathcal{H}[\{a\}|\{J\}]$$

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- Statistical physics approach
to laser physics

What are the phasors $\{a\}$, variables/degrees of freedom of our theory?

They are the complex amplitudes in the
slow amplitude e.m. field expansion in normal modes

$$\mathbf{E}(\mathbf{r}, t) = \sum_k \underbrace{a_k(t)}_{\substack{\text{slow amplitude} \\ \text{complex coefficient}}} \underbrace{\mathbf{E}_k(\mathbf{r})}_{\substack{\text{normal} \\ \text{mode}}} e^{i\omega_k t} + \text{c.c.}$$

$e^{i\omega_k t}$ mode frequency



- Statistical physics approach
to laser physics

Laser stationary regime and equilibrium statistical mechanics

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

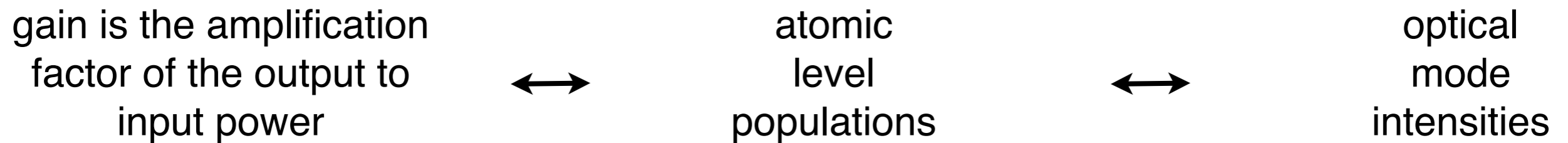
Yet lasers are stable and stationary
Because of **gain saturation**



- Statistical physics approach
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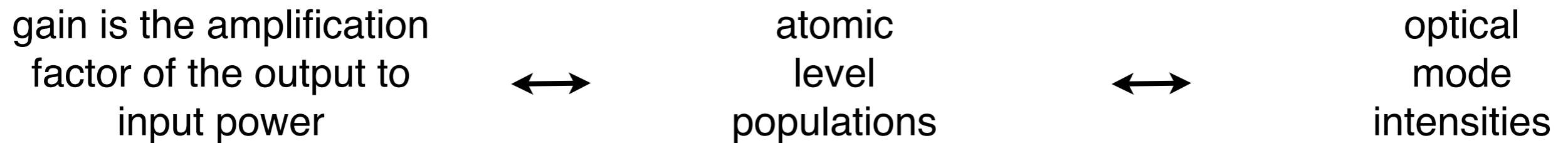
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Laser stationary regime and equilibrium statistical mechanics

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



Yet lasers are stable and stationary
Because of **gain saturation**

as the energy shared by the modes (number of photons emitted) increases the gain is depleted. The energy of modes consequently decreases, then gain increases...

$$g(\mathcal{E}) = \frac{g_0}{1 + \frac{\mathcal{E}}{E_{\text{sat}}}}$$

$$\simeq g_0 - \frac{g_0 \mathcal{E}}{E_{\text{sat}}}$$

$$\mathcal{E} \ll E_{\text{sat}}$$

weak saturation

$$\boxed{\mathcal{E}(t)} = \int_t^{t+\mathcal{T}_{\text{fast}}} d\tau |a(t, \tau)|^2 = \int_t^{t+\mathcal{T}_{\text{fast}}} d\tau \sum_{n,m} e^{i(\omega_n - \omega_m)\tau} a(t, \omega_n) a^*(t, \omega_m)$$

$$\propto \sum_{n,m} \delta(\omega_n - \omega_m) a(t, \omega_n) a^*(t, \omega_m) = \boxed{\sum_n |a(t, \omega_n)|^2}$$

$\mathcal{T}_{\text{fast}}$ roundtrip time, stochastic resonator period

$t \gg \mathcal{T}_{\text{fast}}$



- Statistical physics approach
to laser physics

Laser stationary regime and equilibrium statistical mechanics

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

gain is the amplification
factor of the output to
input power



atomic
level
populations

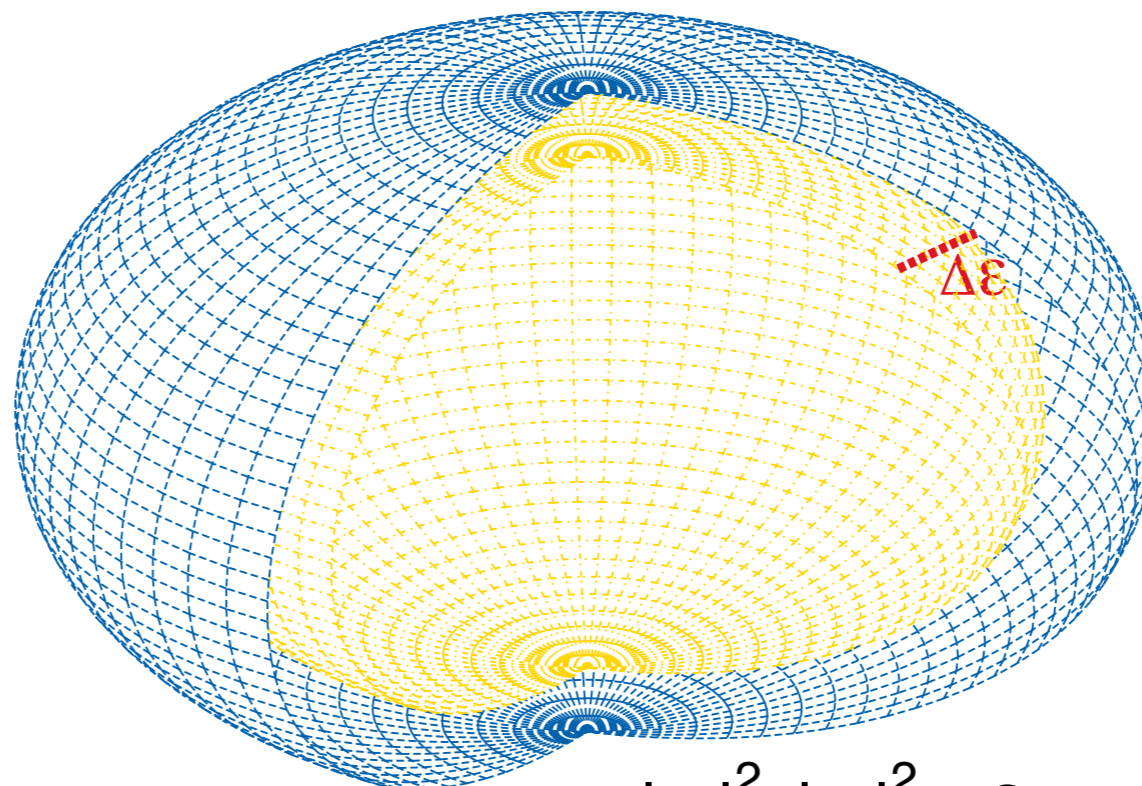


optical
mode
intensities

Yet lasers are stable and stationary
Because of **gain saturation**

$$g(\mathcal{E}) = \frac{g_0}{1 + \frac{\mathcal{E}}{E_{\text{sat}}}}$$

$$\mathcal{E} = \sum_n |a_n(\omega_n)|^2$$



$$|a_1|^2 + |a_2|^2 = \mathcal{E}$$

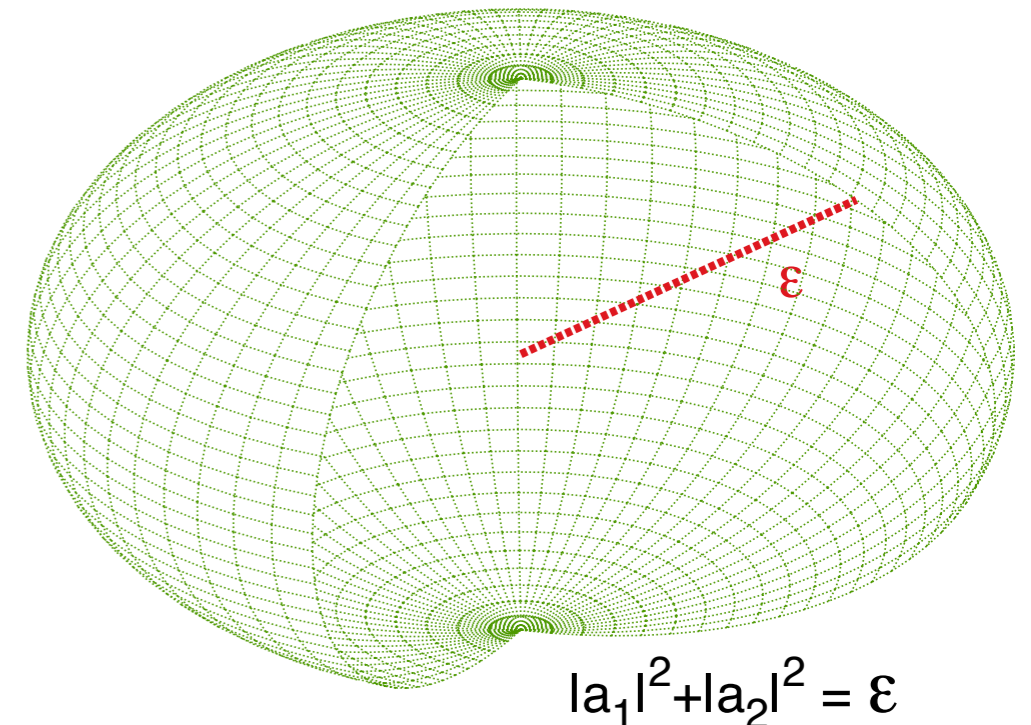
- Statistical physics approach
to laser physics

Laser stationary regime and equilibrium statistical mechanics:
encoding gain saturation

$$\mathcal{E} = \sum_n |a_n(\omega_n)|^2 = N\epsilon$$

As the total optical power of the
amplified modes is kept strictly constant
— quenched dynamics —

$$\dot{\mathcal{E}} = 0$$



Gain saturation yields a global constraint
on the overall intensity of all phasors

- Statistical physics approach
to laser physics

Laser stationary regime and equilibrium statistical mechanics: encoding gain saturation

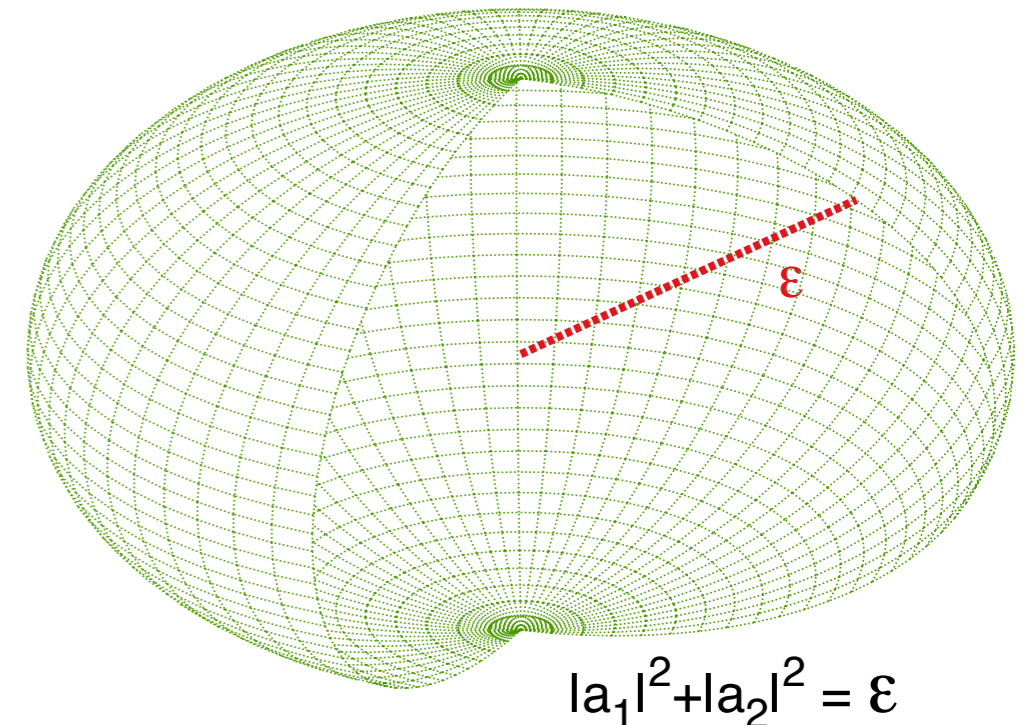
$$\mathcal{E} = \sum_n |a_n(\omega_n)|^2 = N\epsilon$$

As the total optical power of the
amplified modes is kept strictly constant
— quenched dynamics —

$$\dot{\mathcal{E}} = 0$$

the related stationary system can be
considered as if at equilibrium with an effective
“heat-bath” at the “photonic” temperature

$$“T_{\text{laser}}” = \frac{T}{\epsilon^2}$$



We will also use the PUMPING RATE parameter $\mathcal{P} = \epsilon \sqrt{\beta J_0}$

Gain saturation yields a global constraint
on the overall intensity of all phasors



- Statistical physics approach
to laser physics

Random laser stochastic differential equations
for the complex amplitudes (the *phasors*)

$$\dot{a}_n(t) = \mathcal{F}_n[\{a\}|\{J\}] + \eta_n(t) \quad n = 1, \dots, N$$

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

T is related to spontaneous emission, i.e., to the real temperature of the system

Gain saturation yields a global constraint
on the overall intensity of all phasors

$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2 = \text{const}$$

Potential solution to the
Fokker-Planck equation

$$\mathcal{F}_n[\{a\}|\{J\}] \sim -\frac{\partial}{\partial a_n^*} \mathcal{H}[\{a\}|\{J\}]$$

Boltzmann-Gibbs like distribution of
the amplitudes' configurations, with

$$“T_{\text{laser}}” = \frac{T}{\epsilon^2}$$

Ordered lasers: Gordon, Fisher PRL 89, 103901 (2002); Opt. Commun. 223, 151 (2003);
Gat, Gordon, Fisher, PRE 70, 046108 (2004);

Random lasers, quenched homogeneous amplitude approx: L Angelani et al. PRL 96, 065702 (2006);
PRB 74, 104207 (2006), LL et al. PRL 102, 083901 (2009), Conti, LL PRB 83, 134204 (2011)

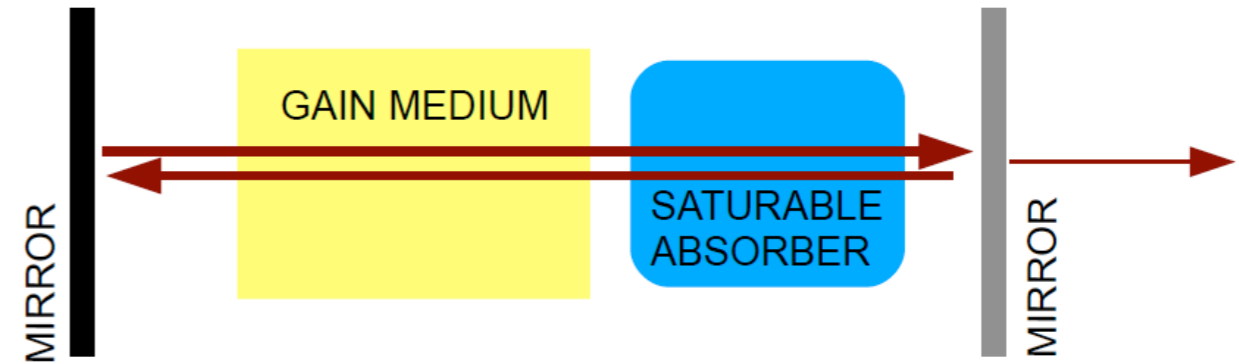
Random lasers, phasors:

F Antenucci, C Conti, A Crisanti, LL, PRL 114, 043901 (2015); F Antenucci, A Crisanti, LL, PRA 91, 053816
(2015), F Antenucci et al Phil. Mag. 96, 704-731 (2016), F. Antenucci, Springer 2016



- Theory for ultrafast mode-locked multimode lasers (order, closed cavity)

Link to Standard Mode-Locking



The equation for the completely closed and ordered limit is known:
Haus standard Mode-Locked laser master equation (1984)

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

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

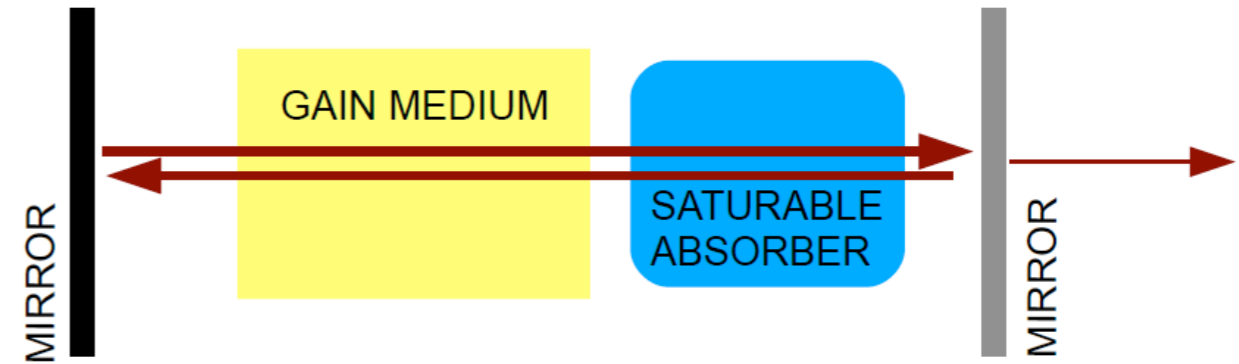
HA Haus, *Mode-Locking of Lasers*, IEEE J. Quantum Electron., 2000

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SAM: SELF-AMPLITUDE MODULATION
COEFFICIENT OF THE SATURABLE ABSORBER

$$\dot{a}_n = \underbrace{(g_m - \ell_m)}_{\text{GAIN}} + \underbrace{iD_m}_{\text{GROUP VELOCITY DISPERSION}} a_n + \underbrace{(\gamma - i\delta)}_{\text{KERR LENS}} \sum_{\omega_j - \omega_k + \omega_l = \omega_n} a_j a_k^* a_l + \underbrace{\eta_n}_{\text{SPONTANEOUS EMISSION}}$$

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

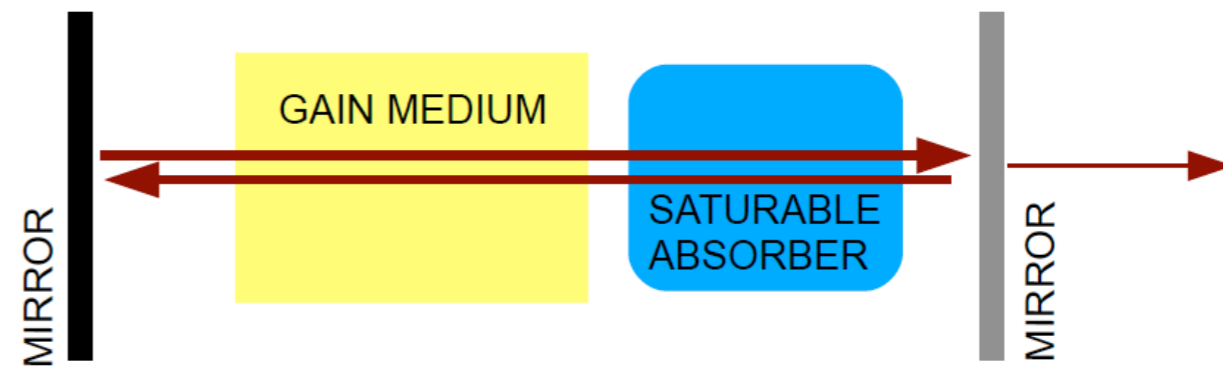
HA Haus, *Mode-Locking of Lasers*, IEEE J. Quantum Electron., 2000

Gordon & Fischer, PRL 2002; Opt. Commun. 2003; Gat, Gordon, Fisher PRE 2004



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SAM: SELF-AMPLITUDE MODULATION
COEFFICIENT OF THE SATURABLE ABSORBER

$$\dot{a}_n = \underbrace{(g_m - \ell_m + iD_m)}_{\text{LOSS}} a_n + \underbrace{(\gamma - i\delta)}_{\text{GAIN}} \sum_{\omega_j - \omega_k + \omega_l = \omega_n} \underbrace{a_j a_k^*}_{\text{GROUP VELOCITY DISPERSION}} a_l + \underbrace{\eta_n}_{\text{KERR LENS}} + \underbrace{\eta_n}_{\text{SPONTANEOUS EMISSION}}$$

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

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

Gordon & Fischer, PRL 2002; Opt. Commun. 2003; Gat, Gordon, Fisher PRE 2004

$$\dot{a}_n = -i \frac{\partial \mathcal{H}}{\partial a_n^*} + \eta_n = iJ_n a_n + iJ_4 \sum_{\omega_j - \omega_k + \omega_l = \omega_n} a_j a_k^* a_l + \eta_n$$

mode-locking and mode-coupling: no locking -> no coupling



- Theory for random lasers: a mode-locked spin-glass theory (disorder, open cavity)

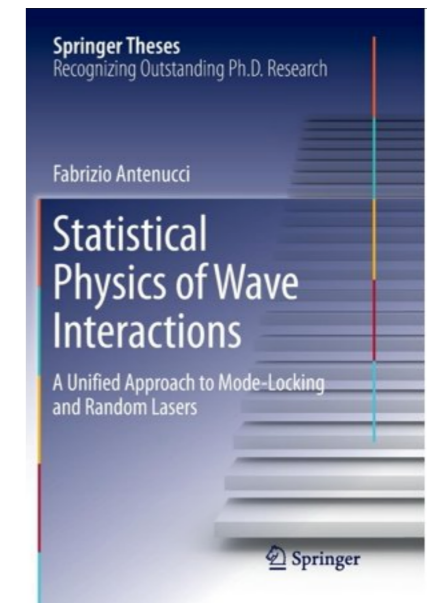
Random laser mode-locked spherical 2+4 phasors' Hamiltonian

Potential, equilibrium-like distribution at inverse temperature $\mathcal{P}^2 = \frac{\epsilon^2}{T} = \frac{1}{T_{\text{photonic}}}$

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

Gain saturation $\leftrightarrow \mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$

Effective interaction



Mode-locking

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

The mode frequencies must satisfy this condition in order for their mutual interaction to be non-zero.



- Theory for random lasers: a mode-locked spin-glass theory (disorder, open cavity)

Random laser mode-locked spherical 2+4 phasors' Hamiltonian

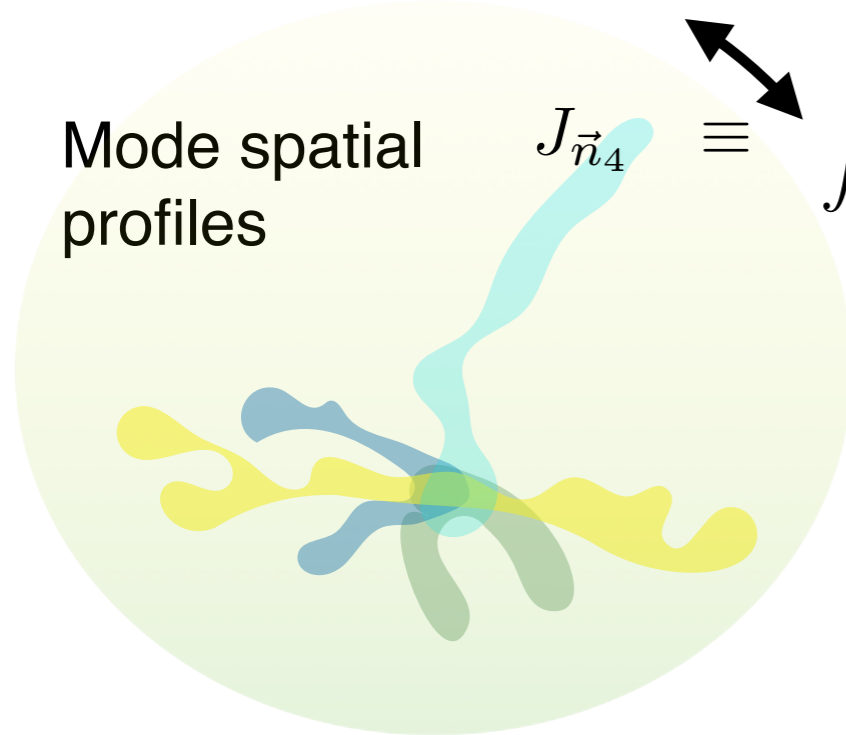
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Gain saturation $\leftrightarrow \mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$

Effective interaction

Mode spatial profiles



$$J_{\vec{n}_4} \equiv \int d\mathbf{r} E_{n_1}^{\alpha_1}(\mathbf{r}) E_{n_2}^{\alpha_2}(\mathbf{r}) E_{n_3}^{\alpha_3}(\mathbf{r}) E_{n_4}^{\alpha_4}(\mathbf{r}) \chi_{\vec{\alpha}_4}^{(3)}(\omega_{n_1}, \omega_{n_2}, \omega_{n_3}, \omega_{n_4}; \mathbf{r})$$

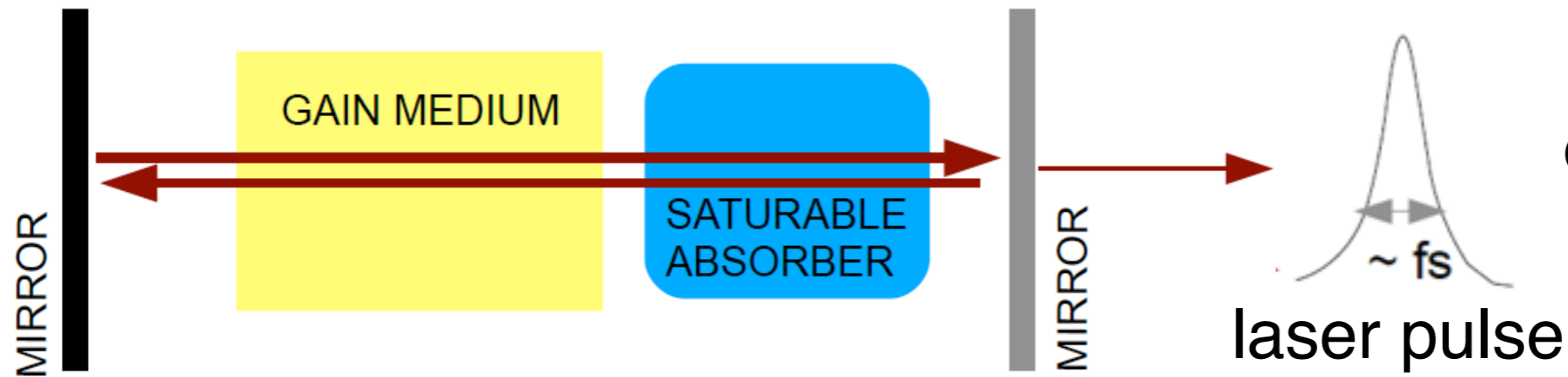
The “interaction” among phasors’ expresses the overlap of the spatial profiles of the e.m. modes, modulated by the nonlinear (random) optical susceptibility.
If the modes do not overlap they do not compete for the energy pumped into the system.

Mode-locking

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

The mode frequencies must satisfy this condition in order for their mutual interaction to be non-zero.

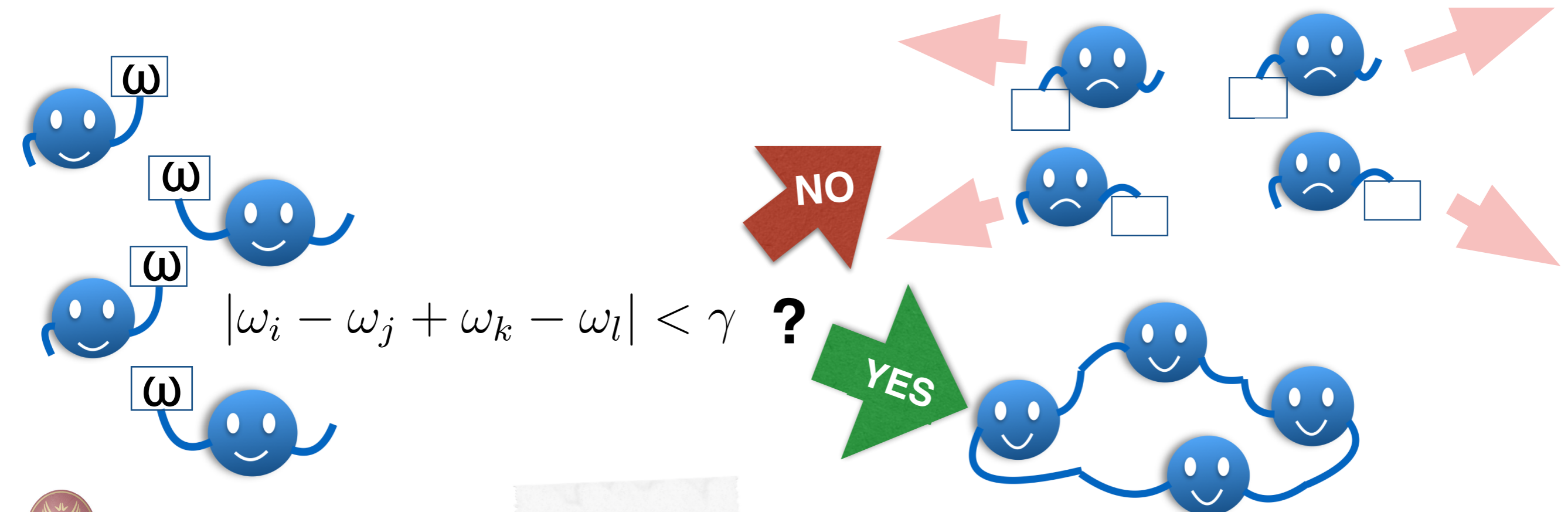
- Theory for ultrafast mode-locked multimode lasers (order, closed cavity)



many modes of different frequencies compose the pulse

these **modes** are *locked*, for any set of modes (i,j,k,l)

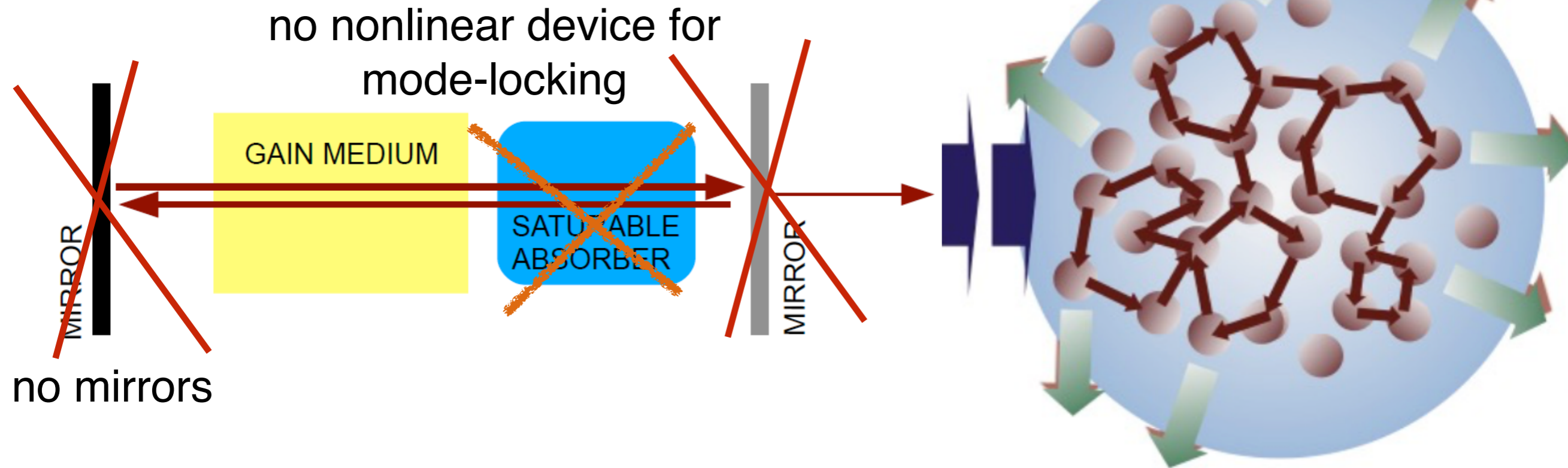
the **frequency matching condition** holds: $|\omega_i - \omega_j + \omega_k - \omega_l| < \gamma$



mode-locking and mode-coupling: no locking -> no coupling

MODE LOCKING

Mode-locking in random lasers



multiple scattering in place of mirror reflection

mode space overlap and heterogeneous non-linear optical response induces the mode-coupling

*no ad hoc device:
self-starting mode-locking in random lasers*

F. Antenucci, G. Lerario, B. Silva Fernandez, L. De Marco, M. De Giorgi, D. Ballarini, D. Sanvitto, and LL,
Phys. Rev. Lett. 126, 173901 (2021).



- Standard and random lasers
- Statistical physics approach to laser physics
 - Theory for ultrafast mode-locked multimode lasers (order, closed cavity)
 - Theory for random lasers: a mode-locked spin-glass theory (disorder, open cavity)
- **The narrow-band solution, phase diagrams, replica symmetry breaking, a new overlap: intensity fluctuation overlap**
- Intermezzo: the experimental measurement of the Parisi distribution of overlaps
- In between theory and experiment: a mode-locking model
 - Monte Carlo dynamics simulation with exchange Monte Carlo, GPU parallel computing
- Power distribution among modes in the glassy light regime: condensation vs equipartition at high pumping
- Outlook (work in progress)



Random laser mode-locked spherical 2+4 phasors' Hamiltonian *Narrow-band*, aka fully connected

$$\mathcal{P}^2 = \frac{\epsilon^2}{T} = \frac{1}{T_{\text{photonic}}}$$

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

$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

It is exactly solvable in the narrowband approximation

$$\omega_n \simeq \omega_0 \quad \forall n$$

the mode locking relation

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

is always satisfied and the interaction network is fully connected

At high pumping (low T)
Replica Symmetry Breaking phases
(one step, full, 1+full)



Narrow-band random laser spherical 2+4 phasors' Hamiltonian

$$\mathcal{H} = -\frac{1}{2} \sum_{jk}^{1,N} J_{jk} a_j a_k^* - \frac{1}{4!} \sum_{j<k<l<m}^{1,N} J_{jklm} a_j a_k^* a_l a_m^* + \text{c.c.},$$
$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

The narrow-band solution:

- order parameters,
- phase diagram,
- a new overlap: intensity fluctuation overlap,
- replica symmetry breaking



Narrow-band random laser

spherical 2+4 phasors' Hamiltonian

$$\mathcal{H} = -\frac{1}{2} \sum_{jk}^{1,N} J_{jk} a_j a_k^* - \frac{1}{4!} \sum_{j < k < l < m}^{1,N} J_{jklm} a_j a_k^* a_l a_m^* + \text{c.c.},$$
$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

ORDER PARAMETERS

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

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

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



Narrow-band random laser

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ORDER PARAMETERS

It is real (or pure imaginary)

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INTENSITY COHERENCE m



Narrow-band random laser spherical 2+4 phasors' Hamiltonian

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It can be chosen real without loss of generality

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INTENSITY COHERENCE m

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

**REPLICA
SYMMETRY
BREAKING**



Narrow-band random laser

spherical 2+4 phasors' Hamiltonian

$$\mathcal{H} = -\frac{1}{2} \sum_{jk}^{1,N} J_{jk} a_j a_k^* - \frac{1}{4!} \sum_{j<k<l<m}^{1,N} J_{jklm} a_j a_k^* a_l a_m^* + \text{c.c.},$$

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It can be chosen real without loss of generality

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

The imaginary part can be set to 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



Narrow-band random laser spherical 2+4 phasors' Hamiltonian

$$\mathcal{H} = -\frac{1}{2} \sum_{jk}^{1,N} J_{jk} a_j a_k^* - \frac{1}{4!} \sum_{j < k < l < m}^{1,N} J_{jklm} a_j a_k^* a_l a_m^* + \text{c.c.},$$

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INTENSITY COHERENCE

m

PHASE COHERENCE

r_d

COMPLEX AMPLITUDES OVERLAP

$q_{\alpha\beta}$



Narrow-band random laser

spherical 2+4 phasors' Hamiltonian

$$\mathcal{H} = -\frac{1}{2} \sum_{jk}^{1,N} J_{jk} a_j a_k^* - \frac{1}{4!} \sum_{j < k < l < m}^{1,N} J_{jklm} a_j a_k^* a_l a_m^* + \text{c.c.},$$
$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

PHASE DIAGRAMS

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

PUMPING RATE (INVERSE TEMPERATURE)

$$R_J \equiv \frac{J}{J_0}$$

DISORDER DEGREE (msd / mean)

$$J_4^2 = \alpha^2 J^2, \quad J_2^2 = (1 - \alpha)^2 J^2,$$

"CLOSEDNESS"



Narrow-band random laser

spherical 2+4 phasors' Hamiltonian

$$\mathcal{H} = -\frac{1}{2} \sum_{jk}^{1,N} J_{jk} a_j a_k^* - \frac{1}{4!} \sum_{j < k < l < m}^{1,N} J_{jklm} a_j a_k^* a_l a_m^* + \text{c.c.},$$

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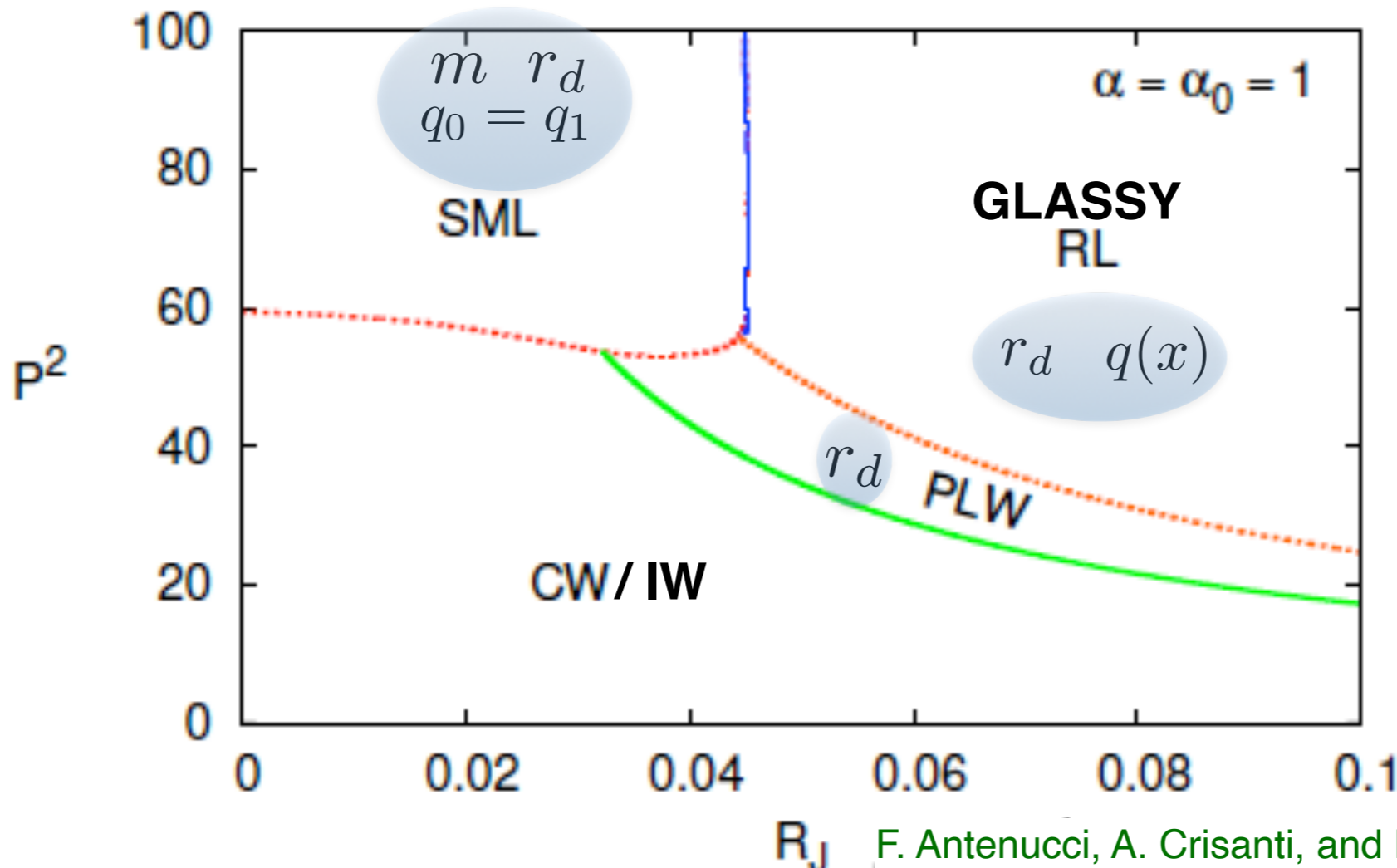
PHASE DIAGRAMS

CLOSED CAVITY + ANY DEGREE OF DISORDER

SML: Standard (and random) ML laser
 CW/IW: Continuous/Incoherent Wave regime
 PLW: Phase-Locked Wave regime
 GRL: Gassy Random Laser

SML \leftrightarrow (random) Ferromagnet
 CW/IW \leftrightarrow Paramagnet
 PLW \leftrightarrow ----
 GRL \leftrightarrow **Glassy phase**

$$R_J \equiv \frac{J}{J_0}$$



$$J_4^2 = \alpha^2 J^2, \quad J_2^2 = (1 - \alpha)^2 J^2,$$



Narrow-band random laser

spherical 2+4 phasors' Hamiltonian

$$\mathcal{H} = -\frac{1}{2} \sum_{jk}^{1,N} J_{jk} a_j a_k^* - \frac{1}{4!} \sum_{j < k < l < m}^{1,N} J_{jklm} a_j a_k^* a_l a_m^* + \text{c.c.},$$

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PHASE DIAGRAMS

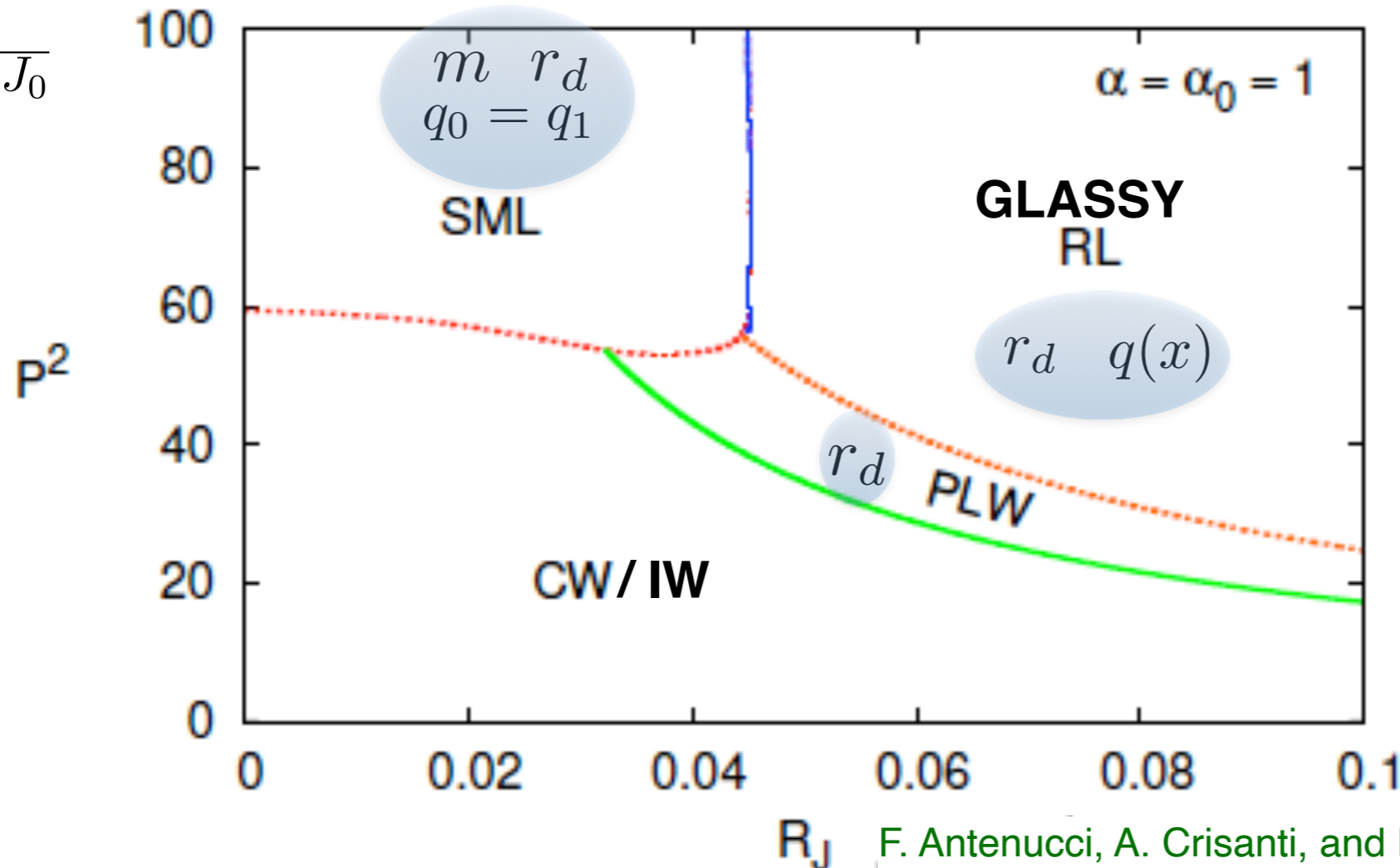
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 PLW ↔ ----
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$$\mathcal{P} = \epsilon \sqrt{\beta J_0}$$

$$R_J \equiv \frac{J}{J_0}$$



$$J_4^2 = \alpha^2 J^2, \quad J_2^2 = (1 - \alpha)^2 J^2,$$



Narrow-band random laser

spherical 2+4 phasors' Hamiltonian

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$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

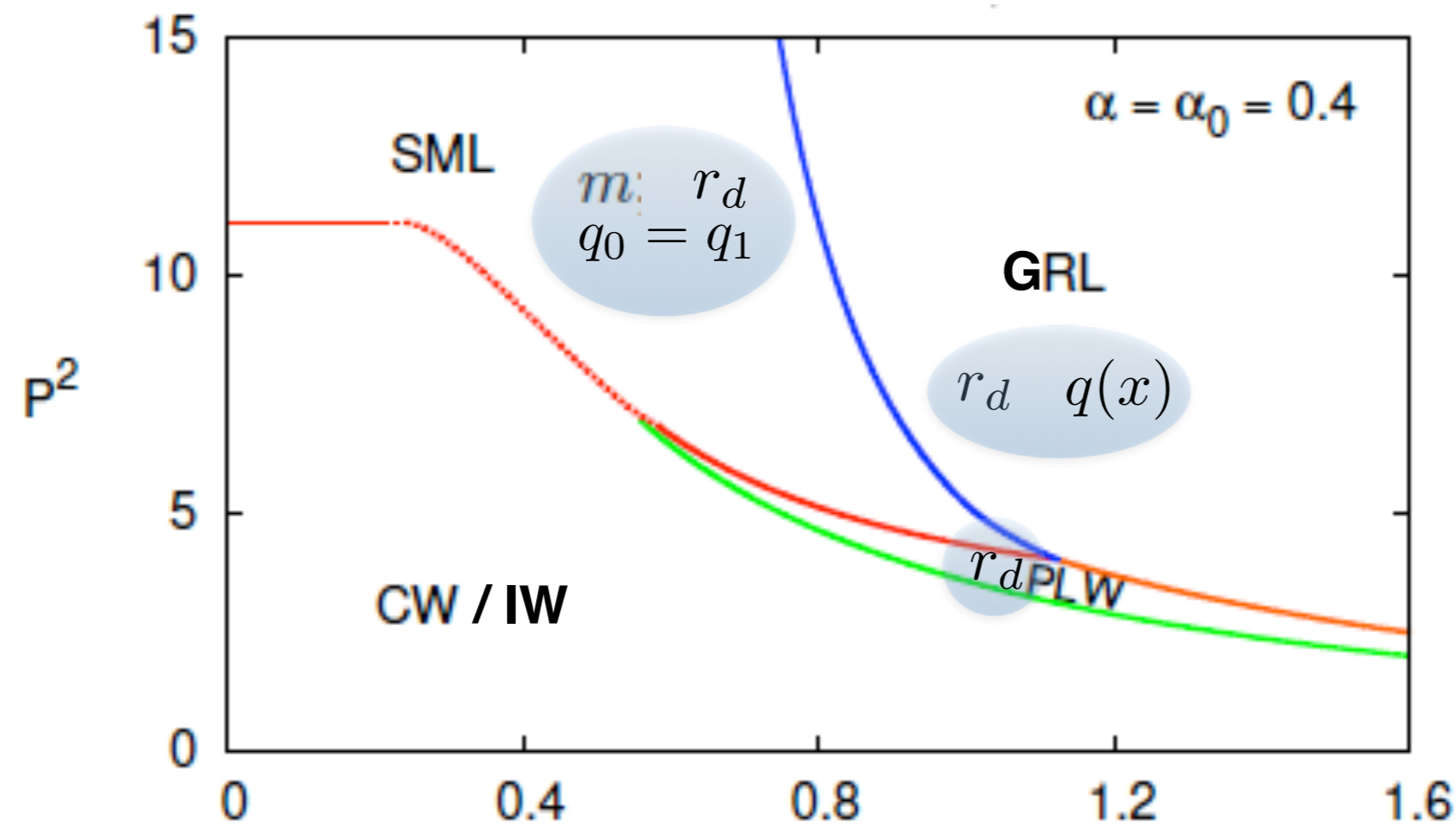
PHASE DIAGRAMS

OPEN CAVITY + ANY DEGREE OF DISORDER

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 CW/IW: Continuous/Incoherent Wave regime
 PLW: Phase-Locked Wave regime
 GRL: Gassy Random Laser

SML ↔ (random) Ferromagnet
 CW/IW ↔ Paramagnet
 PLW ↔ ----
 GRL ↔ **Glassy phase**

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



$$J_4^2 = \alpha^2 J^2, \quad J_2^2 = (1 - \alpha)^2 J^2,$$



$$R_J = J/J_0$$

R_J F. Antenucci, A. Crisanti, and LL,
PRA 91, 053816 (2015)



Narrow-band random laser spherical 2+4 phasors' Hamiltonian

An extra (useful) parameter: IFO Intensity Fluctuation Overlap

Since our 'spins' are not *locally* bounded we can define an
intensity fluctuation overlap between replicas

$$Q_{\alpha\beta} = \frac{1}{N} \sum_{k=1}^N \left(|a_k^{(\alpha)}|^2 |a_k^{(\beta)}|^2 - \langle |a_k|^2 \rangle^2 \right)$$

This is the *standard* Parisi overlap in this model: $q_{\alpha\beta} = \frac{1}{N} \sum_k a_k^\alpha a_k^{*\beta}$

In the *fully connected* mean-field mixed spherical phasor model one proves

$$Q_{ab} = q_{ab}^2 - \frac{|m|^4}{4}$$

*There is a one-to-one correspondence between elements of the
standard (Parisi) overlap and IFO matrices
In principle IFO can be experimentally measured*

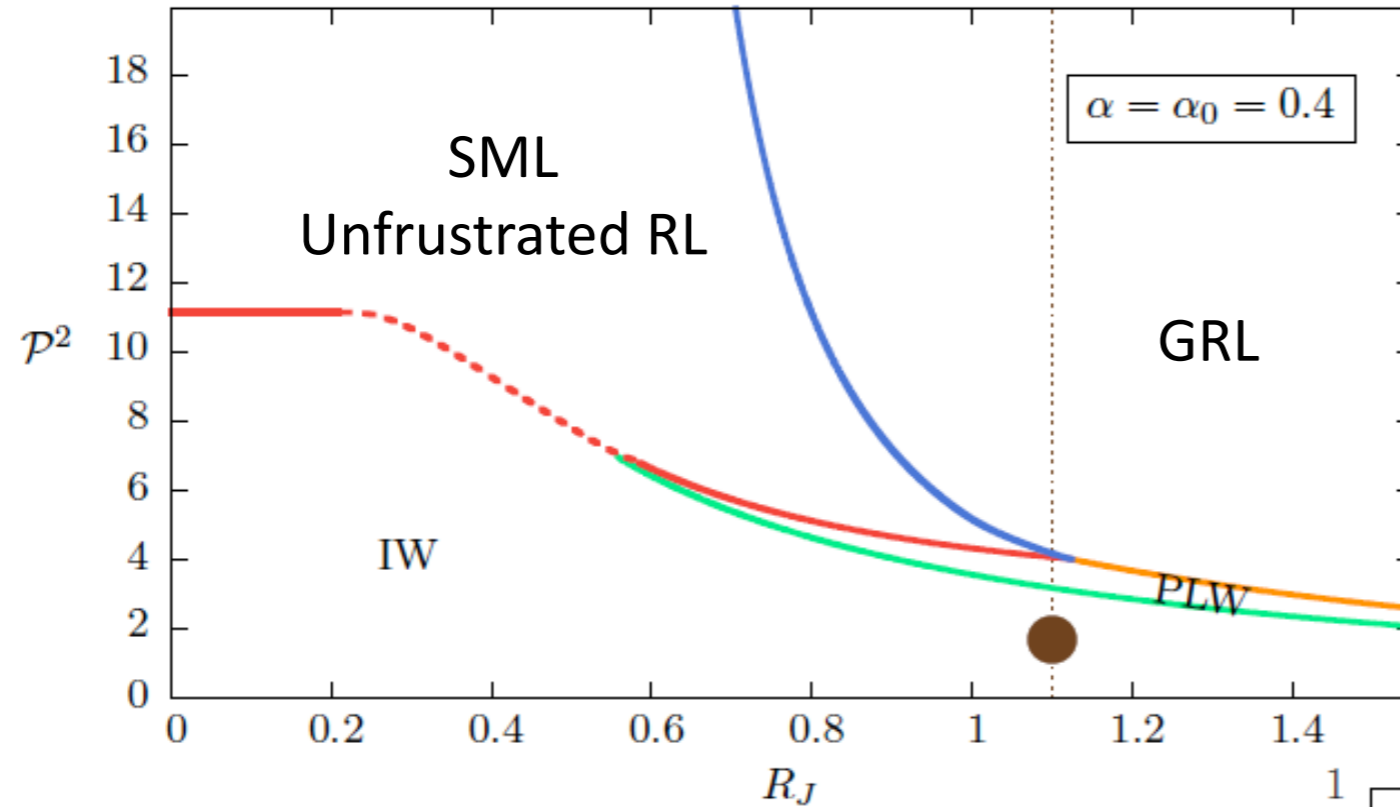


Narrow-band random laser spherical 2+4 phasors' Hamiltonian

$$\mathcal{H}[\mathbf{a}] = -\frac{1}{2} \sum_{n_1 n_2}^{1,N} J_{n_1 n_2} a_{n_1} a_{n_2}^* - \frac{1}{4!} \sum_{n_1 n_2 n_3 n_4}^{1,N} J_{n_1 n_2 n_3 n_4} a_{n_1} a_{n_2}^* a_{n_3} a_{n_4}^*$$

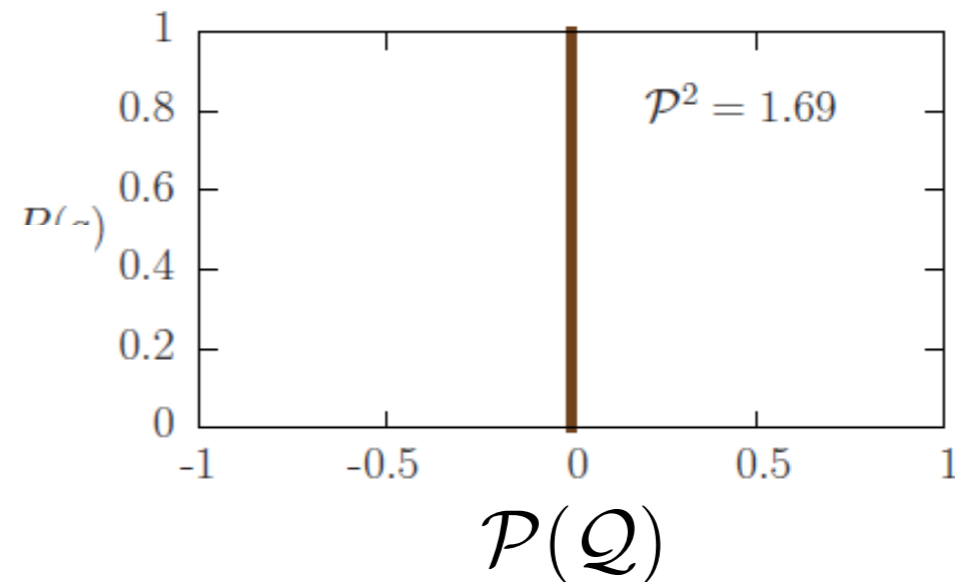
PHASE DIAGRAM

MFT: Intensity Fluctuations Overlap



REPLICA SYMMETRY BREAKING

IFO DISTRIBUTION
Replica Symmetry

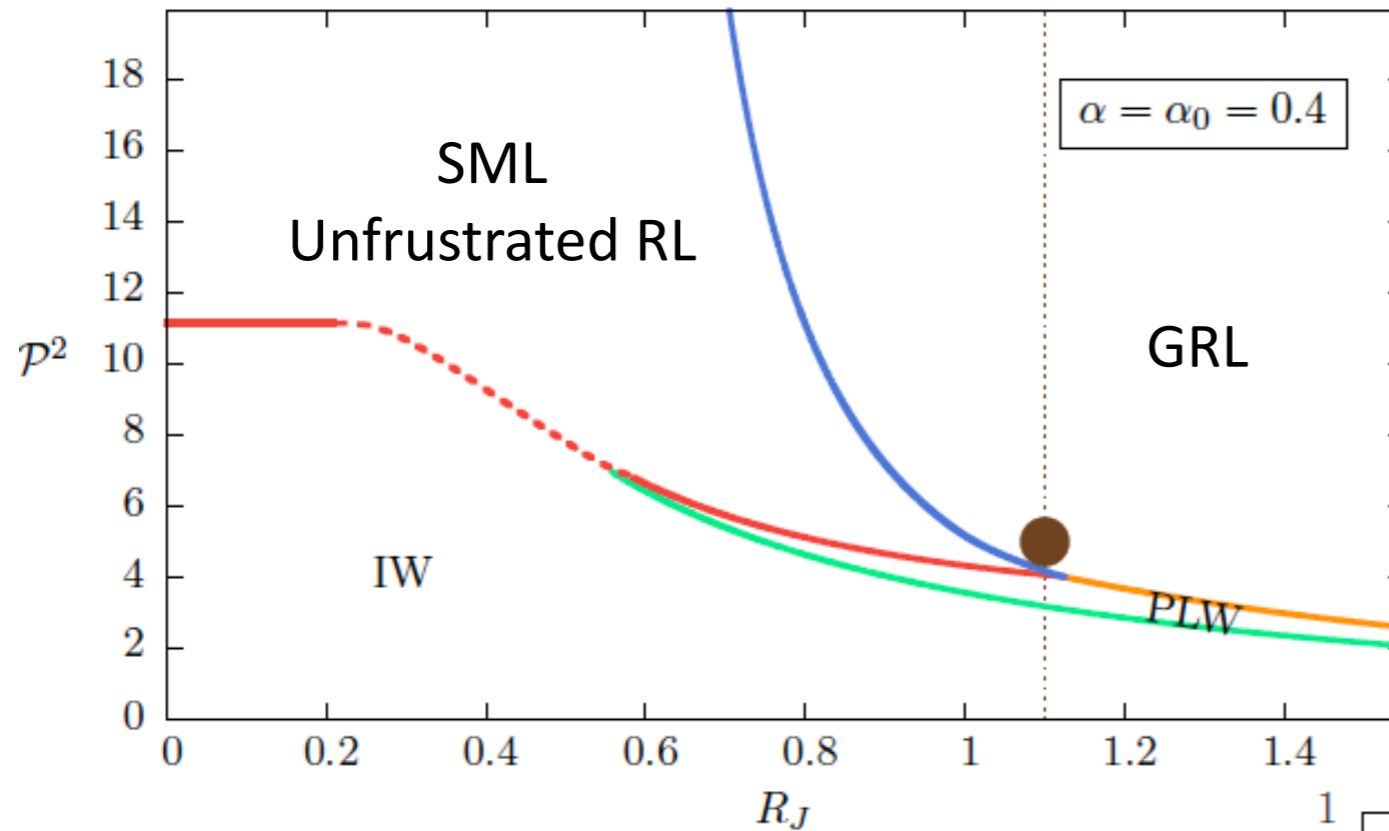


Narrow-band random laser spherical 2+4 phasors' Hamiltonian

$$\mathcal{H}[a] = -\frac{1}{2} \sum_{n_1 n_2}^{1,N} J_{n_1 n_2} a_{n_1} a_{n_2}^* - \frac{1}{4!} \sum_{n_1 n_2 n_3 n_4}^{1,N} J_{n_1 n_2 n_3 n_4} a_{n_1} a_{n_2}^* a_{n_3} a_{n_4}^*$$

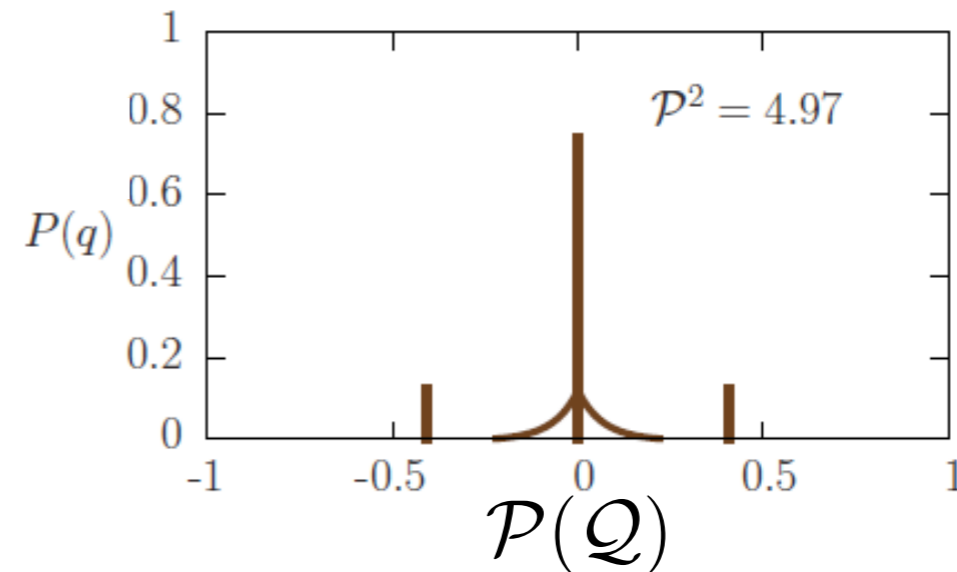
PHASE DIAGRAM

MFT: Intensity Fluctuations Overlap



REPLICA SYMMETRY BREAKING

IFO DISTRIBUTION
1+Full Replica
Symmetry Breaking

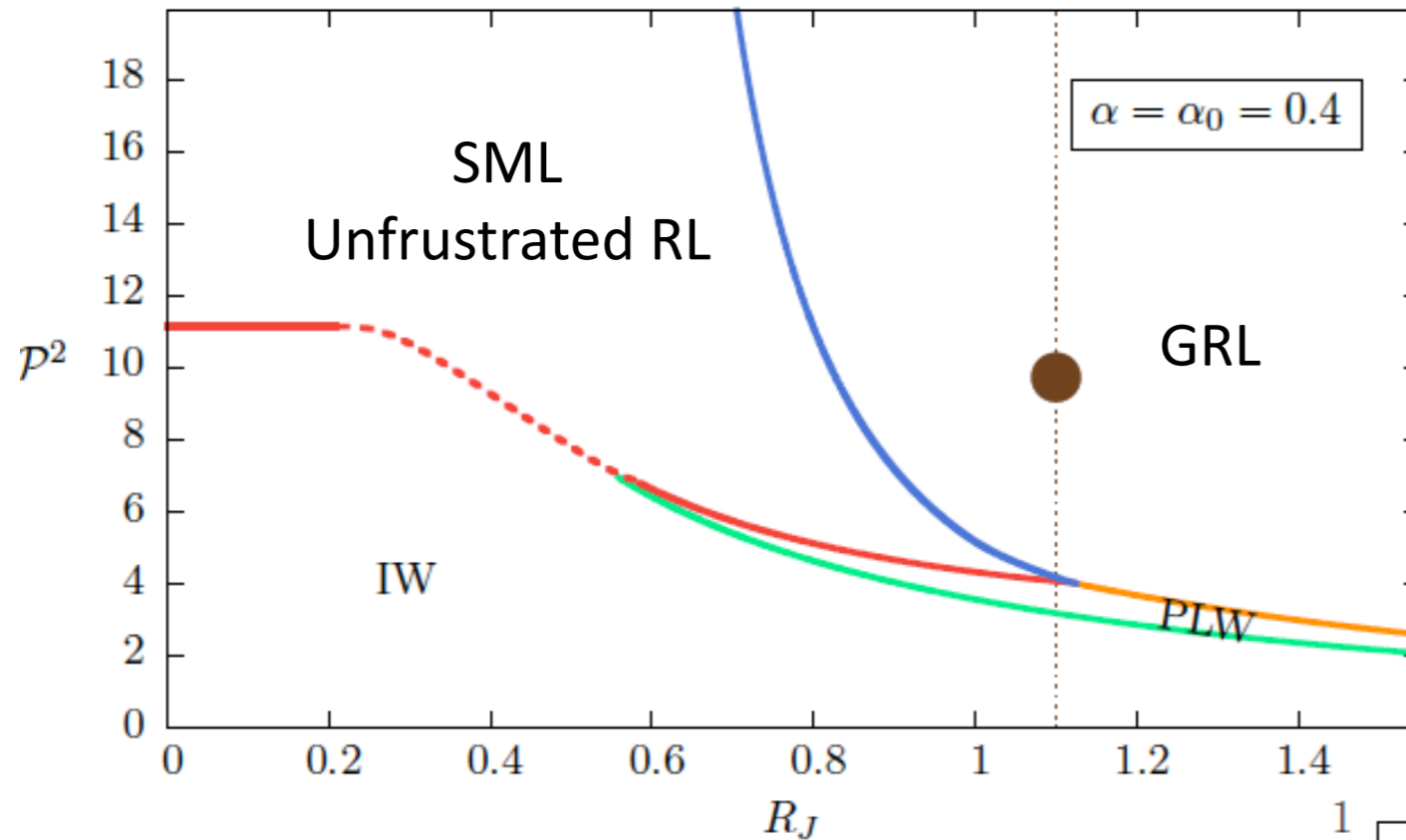


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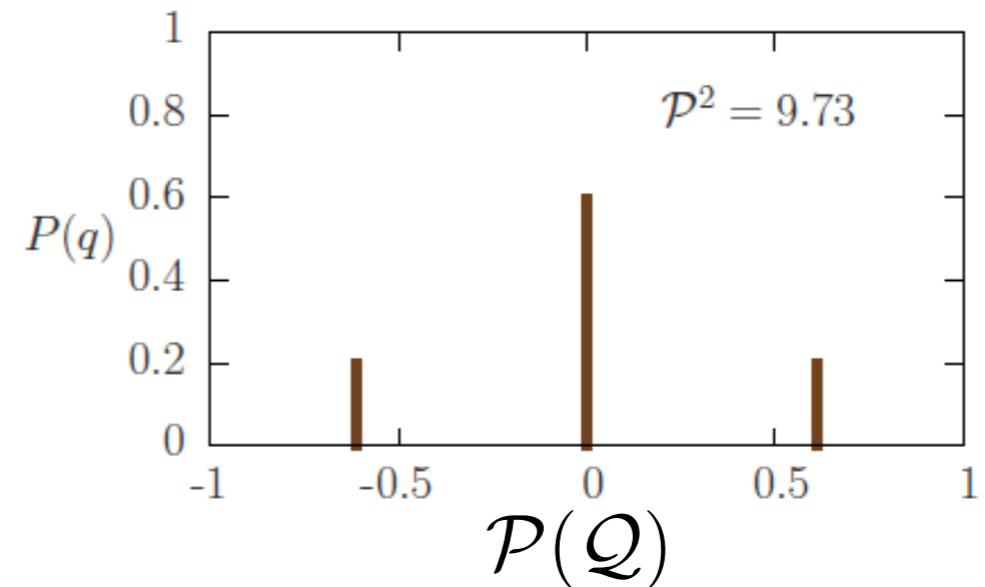
PHASE DIAGRAM

MFT: Intensity Fluctuations Overlap



**REPLICA SYMMETRY
BREAKING**

IFO DISTRIBUTION
1-step Replica
Symmetry Breaking

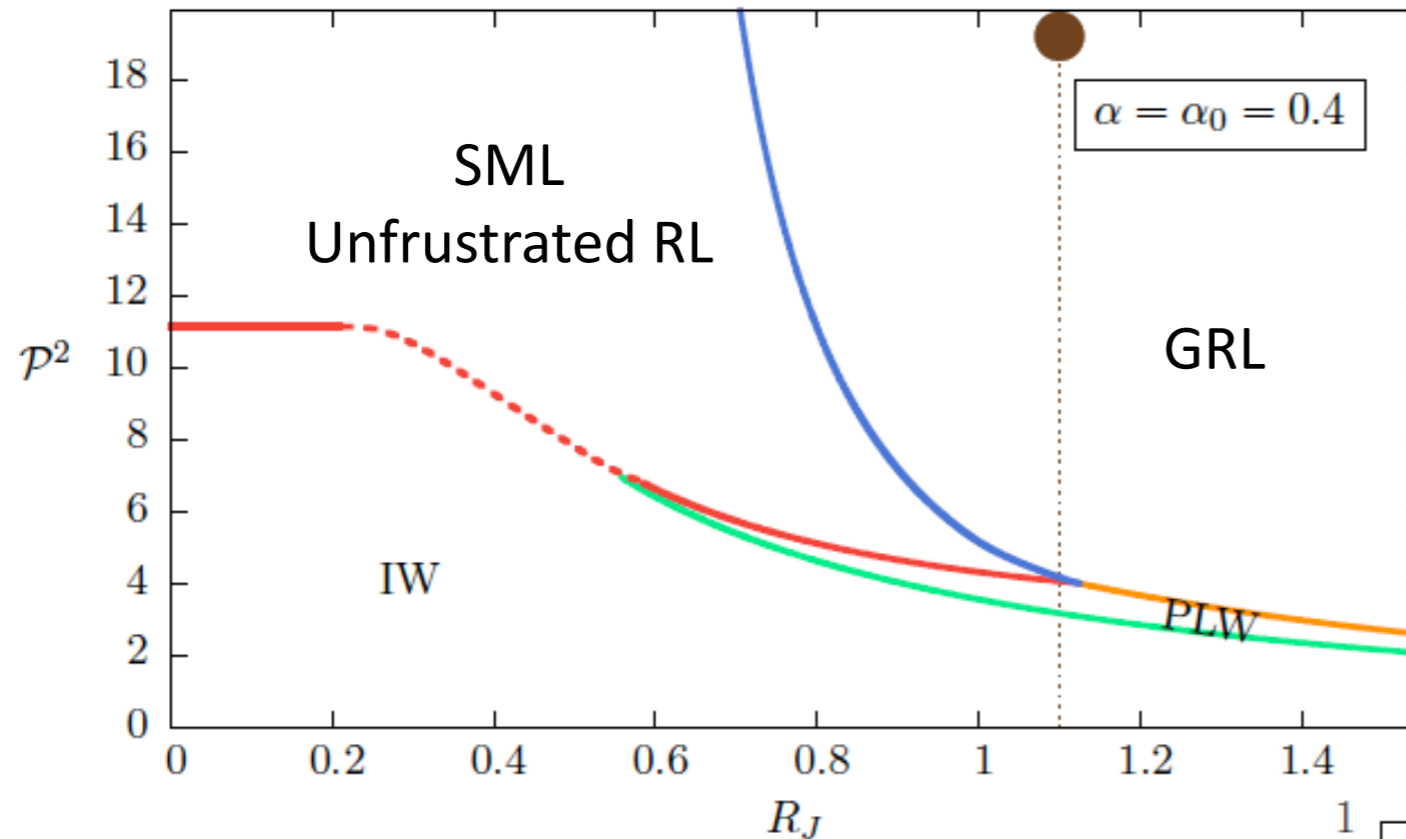


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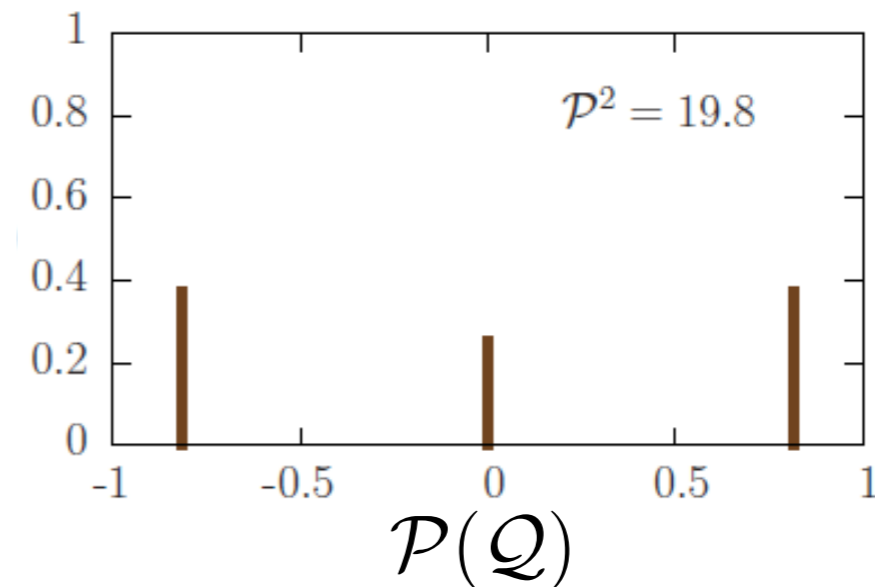
PHASE DIAGRAM

MFT: Intensity Fluctuations Overlap



REPLICA SYMMETRY BREAKING

IFO DISTRIBUTION
1-step Replica
Symmetry Breaking



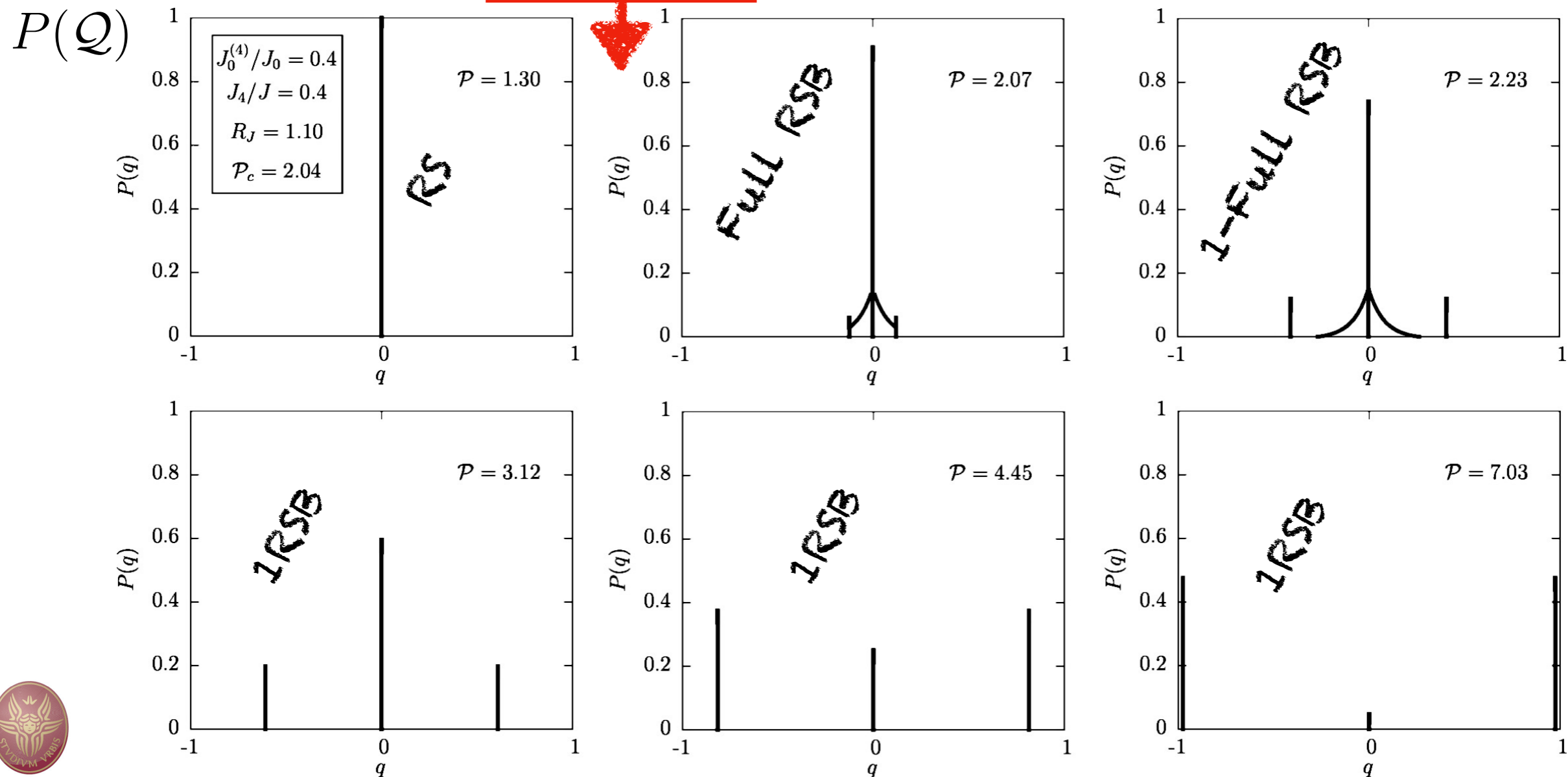
Narrow-band random laser spherical 2+4 phasors' Hamiltonian

REPLICA SYMMETRY BREAKING

Intensity Fluctuation Overlap
probability distribution in the
fully connected 2+4 spherical phasor model

F. Antenucci, A. Crisanti, LL, SciRep 2015

RSB transition



REPLICA SYMMETRY BREAKING

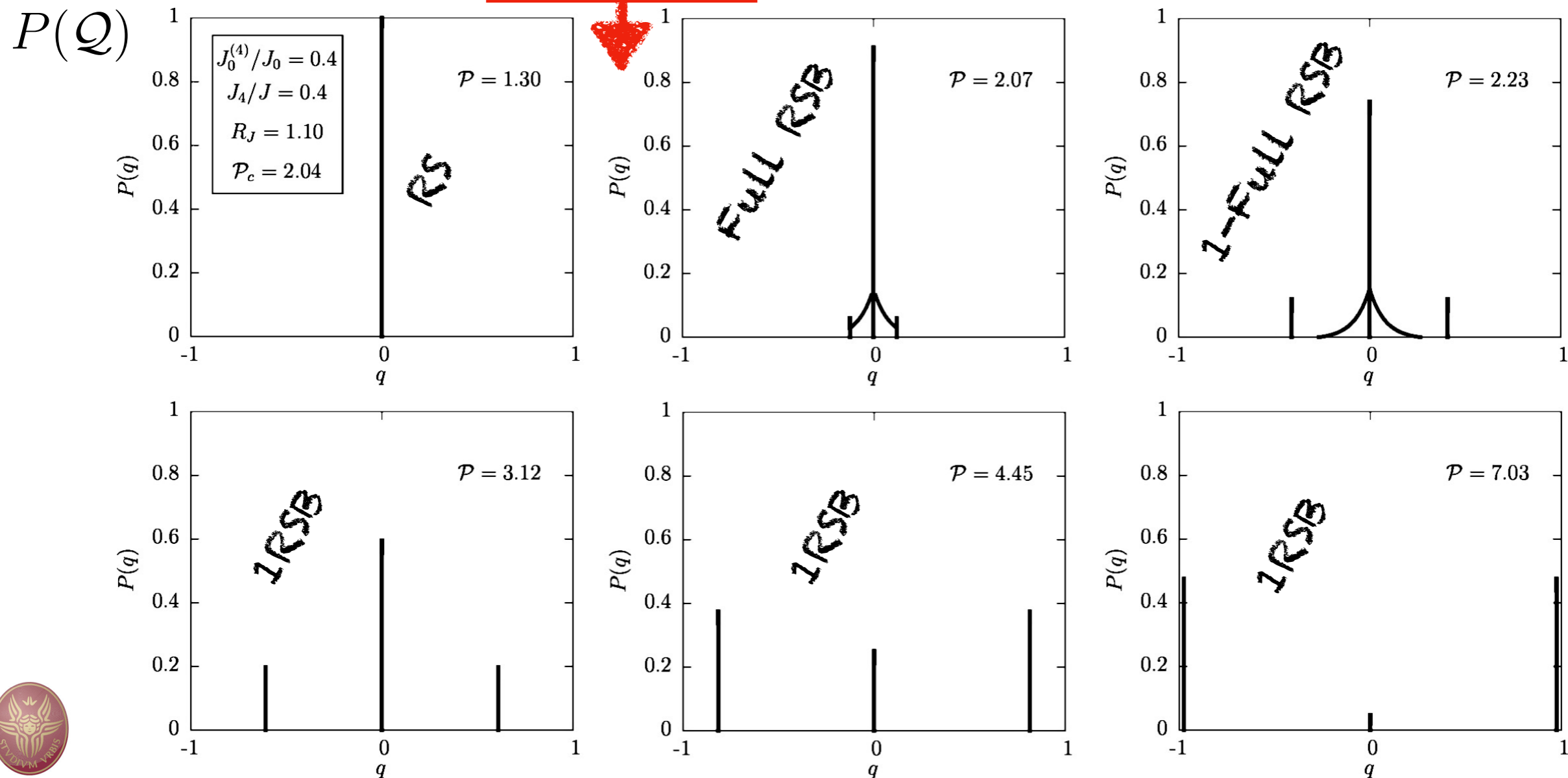
Intensity Fluctuation Overlap
probability distribution in the
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Mean-field $Q_{ab} = q_{ab}^2$
fully connected theory

F. Antenucci, A. Crisanti, LL, SciRep 2015

Element-element relation: any
Replica Symmetry Breaking is
detectable and measurable.

RSB transition

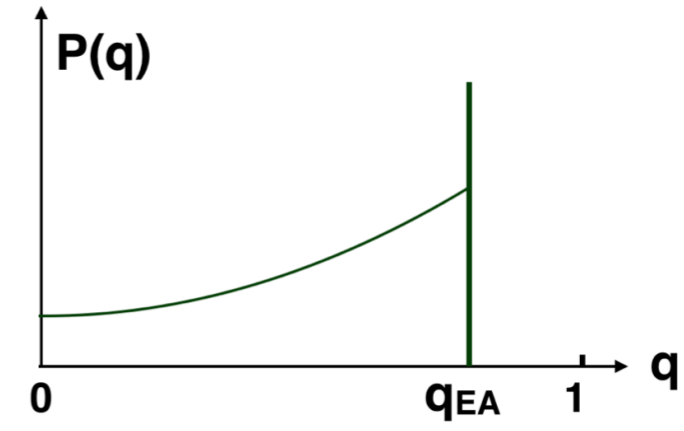


- Standard and random lasers
- Statistical physics approach to laser physics
 - Theory for ultrafast mode-locked multimode lasers (order, closed cavity)
 - Theory for random lasers: a mode-locked spin-glass theory (disorder, open cavity)
- The narrow-band solution, phase diagrams, replica symmetry breaking, a new overlap: intensity fluctuation overlap
- **Intermezzo: the experimental measurement of the Parisi distribution of overlaps**
- In between theory and experiment: a mode-locking model
 - Monte Carlo dynamics simulation with exchange Monte Carlo, GPU parallel computing
- Power distribution among modes in the glassy light regime: condensation vs equipartition at high pumping
- Outlook (work in progress)



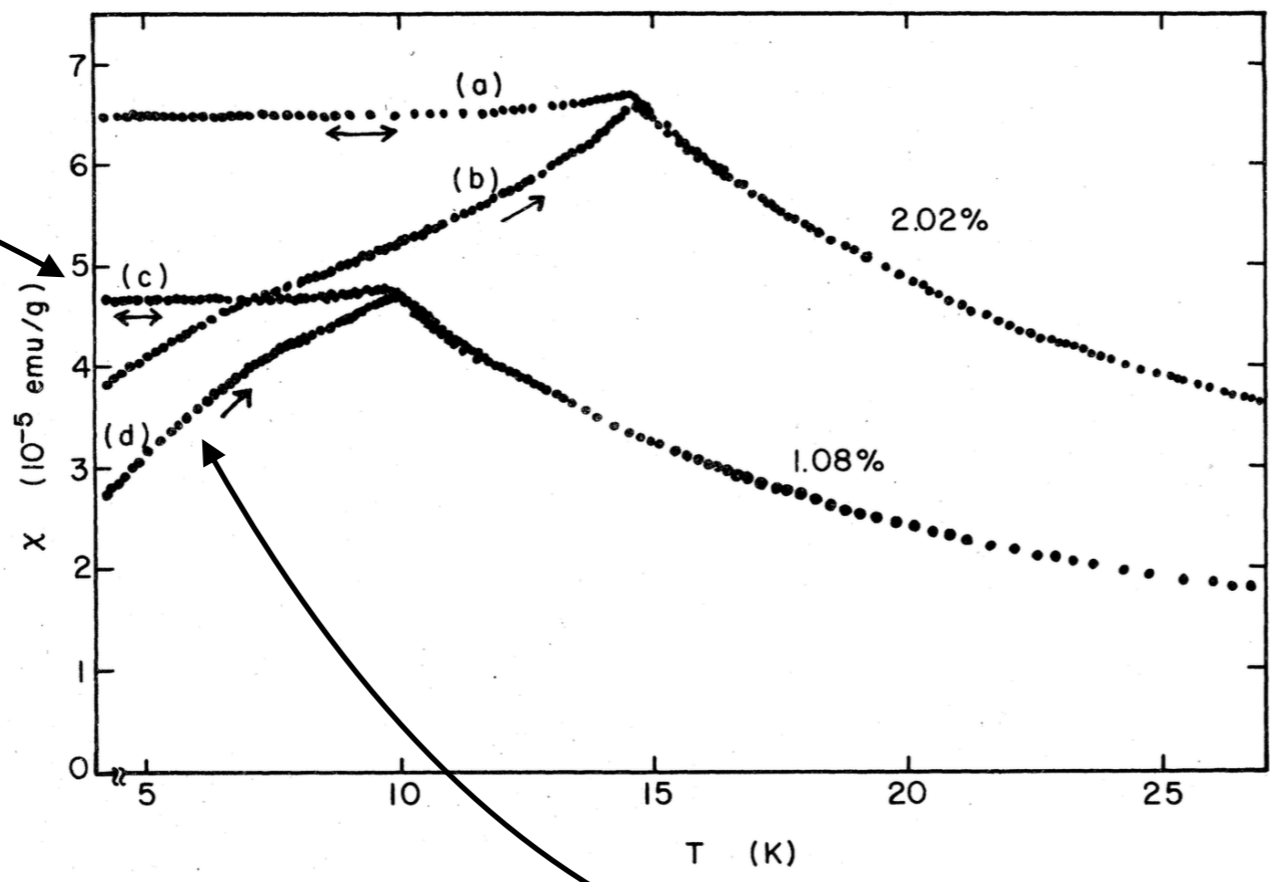
- Intermezzo: the experimental measurement of the Parisi distribution of overlaps

The RSB order parameter is the distribution of the overlap values



What is accessible to experiments?

Static susceptibility after cooling in zero magnetic field or in a field h



Field Cooled Susceptibility

Zero Field Cooled Susceptibility

$$\chi_{FC} = \beta (1 - \langle q \rangle)$$

$$\chi_{ZFC} = \beta (1 - q_{EA})$$

$$\langle q \rangle = \int_0^1 dq q P(q)$$

$$q_{EA} = \frac{1}{N} \sum_i \overline{m_i^2} = \max_q P(q)$$



- Intermezzo: the experimental measurement of the Parisi distribution of overlaps

What hinders the whole $P(q)$ measure?

We need microscopic configurations at equilibrium

$$q_{ab} = \frac{1}{N} \sum_{i=1}^N \sigma_i^{(a)} \sigma_i^{(b)}$$

configuration of replica a
at equilibrium

$$\frac{1}{Z} \sum_{\{\sigma^{(a)}\}} \exp \left\{ -\beta \mathcal{H}_J[\{\sigma^{(a)}\}] \right\}$$

$$q_{\alpha_1 \alpha_2} \equiv \frac{1}{N} \sum_{i=1}^N m_i^{(\alpha_1)} m_i^{(\alpha_2)}$$

pure state α
with clustering property

$P(q)$

Microscopic atomic spin configurations in spin glasses
are hard to be measured

Equilibrium is hardly/never attained
in experiments on spin-glasses



- Intermezzo: the experimental measurement of the Parisi distribution of overlaps

Statics-dynamics equivalence

$$X(q) = \lim_{\substack{t, t' \rightarrow \infty \\ C(t, t') = q}} \frac{\partial \chi(t, t')}{\partial t'} \bigg/ \frac{\partial C(t, t')}{\partial t'} \quad \text{Fluctuation-Dissipation Ratio}$$

Cumulative distribution of overlap $\tilde{X}(q) = \int_0^q dq' \tilde{P}(q')$



- Intermezzo: the experimental measurement of the Parisi distribution of overlaps

Statics-dynamics equivalence

$$X(q) = \lim_{\substack{t, t' \rightarrow \infty \\ C(t, t') = q}} \frac{\partial \chi(t, t')}{\partial t'} \bigg/ \frac{\partial C(t, t')}{\partial t'}$$

Fluctuation-Dissipation Ratio

Cumulative distribution of overlap

$$\tilde{X}(q) = \int_0^q dq' \tilde{P}(q')$$

If the dynamic FDR can be measured in experiments
and if the system is **stochastically stable***

$$X(q) = \tilde{X}(q)$$

and we can recover the Parisi distribution

*A reshuffling of pure states takes place in the (limit) procedure of the construction of $\tilde{P}(q)$ in presence of ergodicity breaking. If such reshuffling only lift the degeneracy due to symmetries of the system, then $\tilde{P}(q)$ is the right distribution of the overlap.

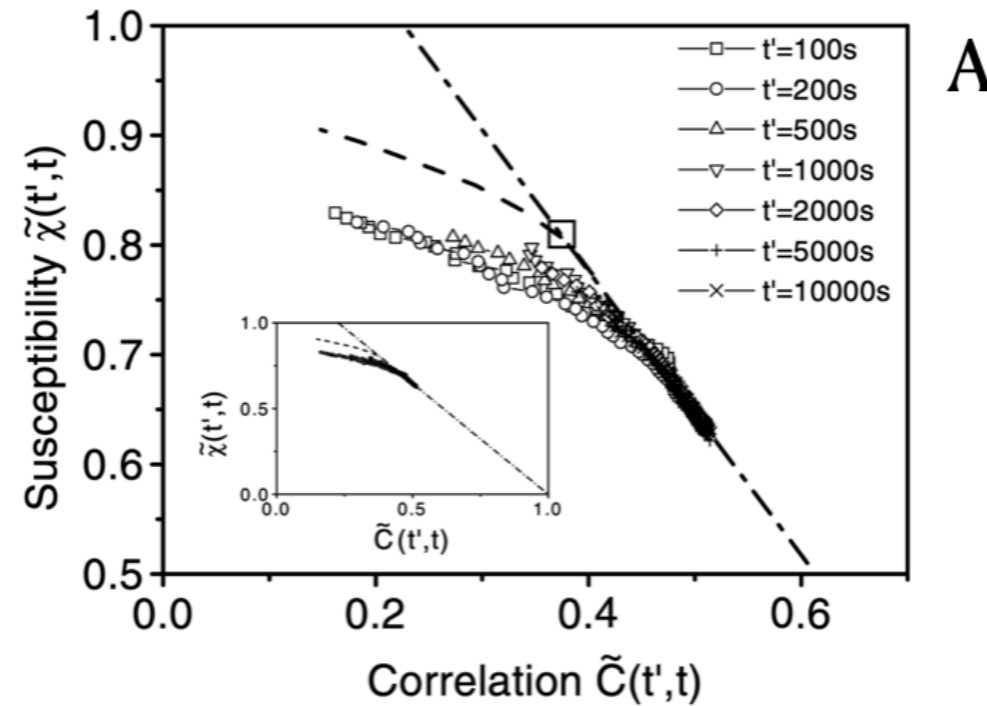


- Intermezzo: the experimental measurement of the Parisi distribution of overlaps

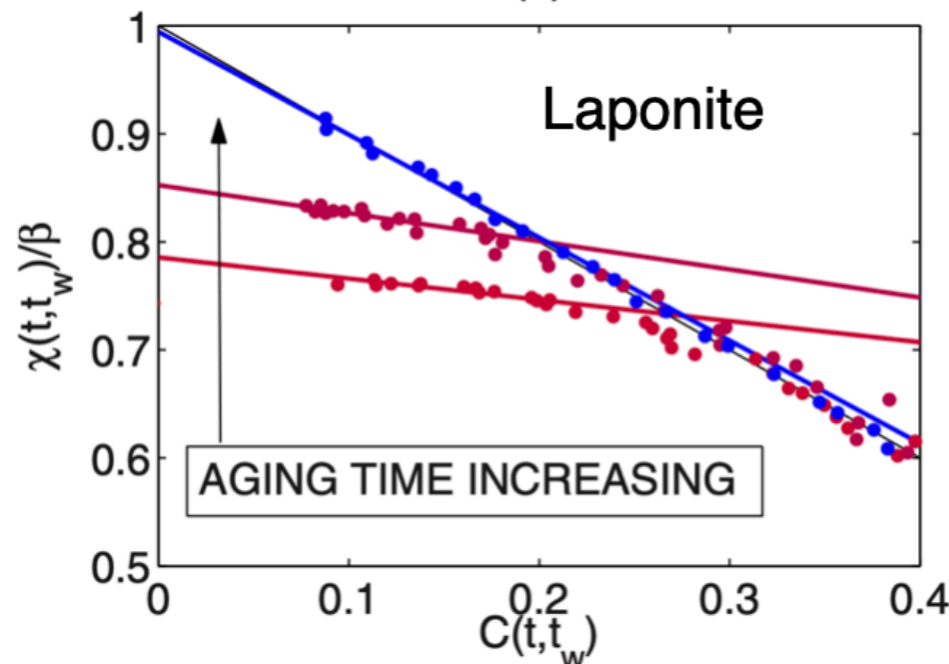
Statics-dynamics equivalence

Cd Cr_{1.7} In_{0.3} S₄
spin-glass

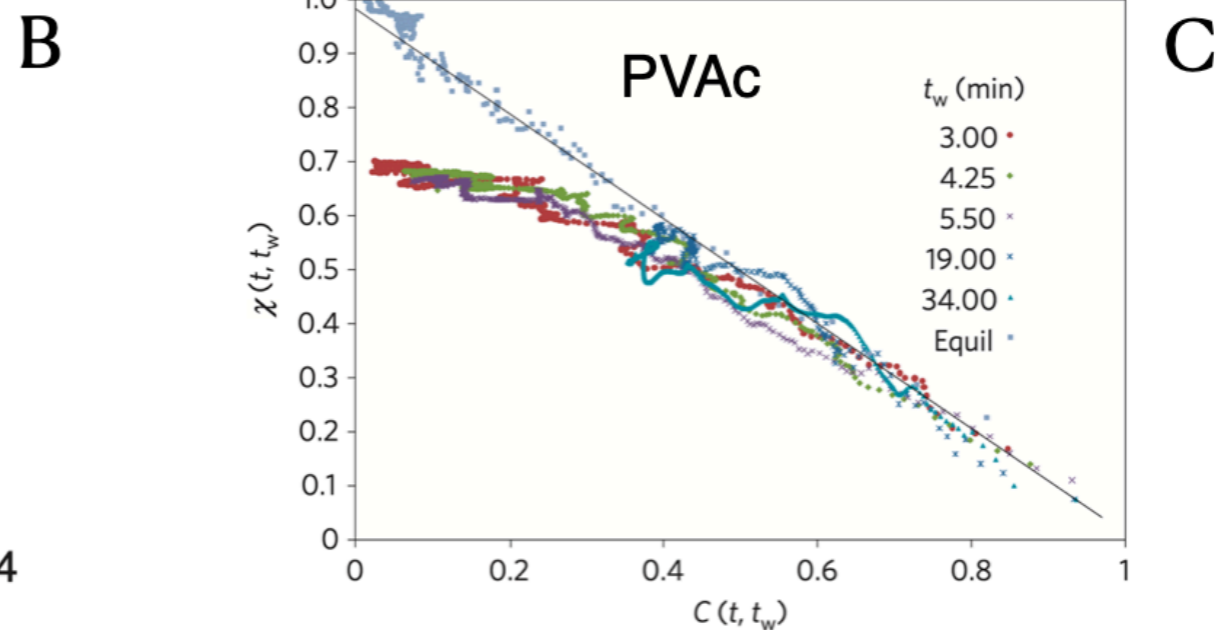
Herisson, Ocio, PRL 88, 257202 (2002)



FDR measure is harder than expected in real glassy systems



Maggi et al, PRB 81, 104201 (2010)



Ourkis, Israeloff, Nature Phys 6, 135 (2010)



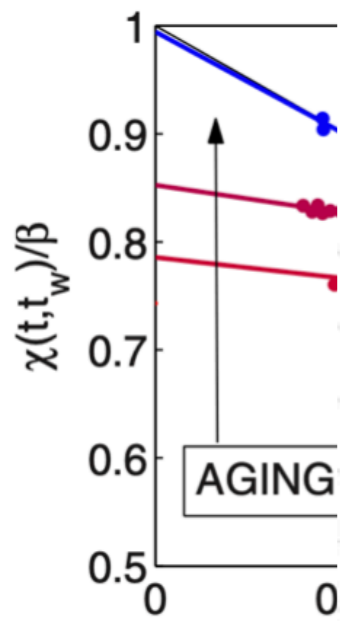
- Int of

Cd Cr spir

Herisson, Ocio, PRL 88, 257

CHAPTER 16

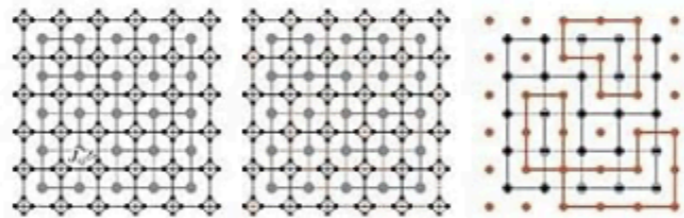
arXiv:2209.03781



Maggi et al, PRB 8

Spin Glass Theory and Far Beyond

Replica Symmetry Breaking After 40 Years



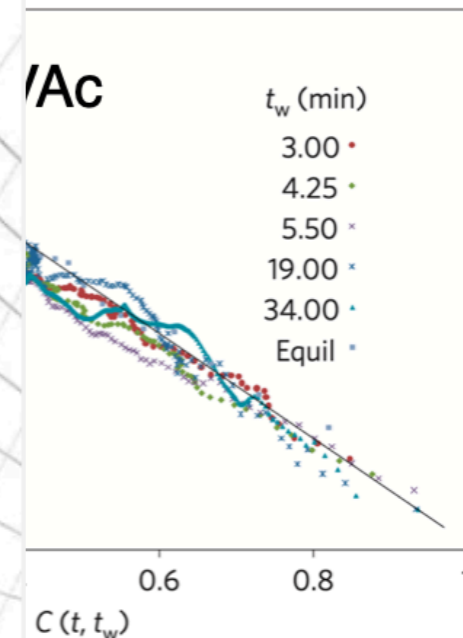
Editors
 Patrick Charbonneau
 Enzo Marinari
 Marc Mézard
 Giorgio Parisi
 Federico Ricci-Tersenghi
 Gabriele Sicuro
 Francesco Zamponi



ent

FDR measure is harder than expected in real glassy systems

No $P(q)$ measure from dynamic stochastic stability approach so far.



Israeloff, Nature Phys 6, 135 (2010)



• Intermezzo: the experimental measurement of the Parisi distribution of overlaps

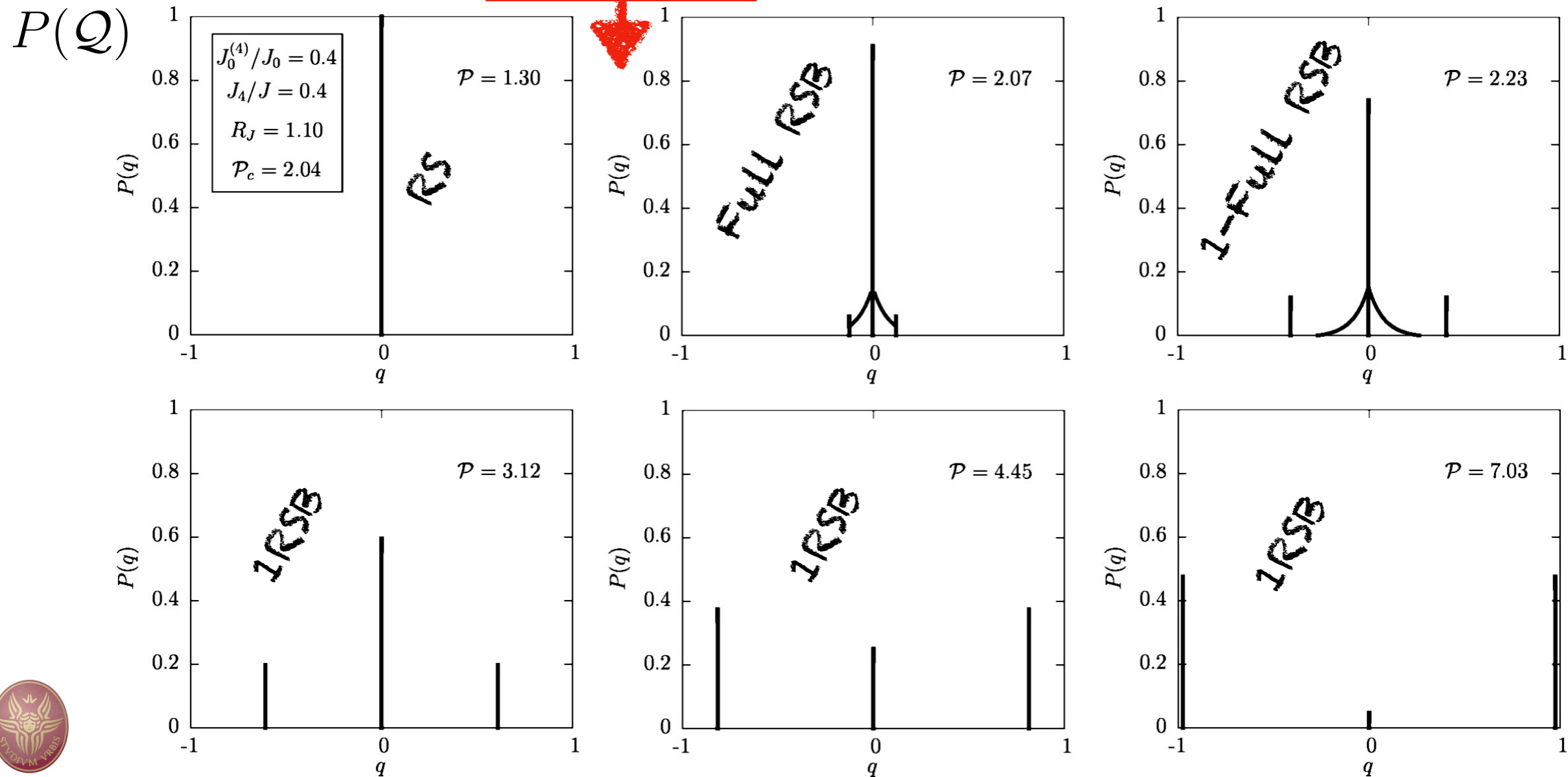
Intensity Fluctuation Overlap probability distribution in the Fully connected 2+4 spherical phasor model

$$Q_{\alpha\beta} = \frac{1}{N} \sum_{k=1}^N \left(|a_k^{(\alpha)}|^2 - \langle |a_k|^2 \rangle \right) \left(|a_k^{(\beta)}|^2 - \langle |a_k|^2 \rangle \right)$$

Mean-field $Q_{ab} = q_{ab}^2$ - fully connected theory

F. Antenucci, A. Crisanti, LL, SciRep 2015

RSB transition



- Intermezzo: the experimental measurement of the Parisi distribution of overlaps

Intensity Fluctuation Overlap - IFO
Fully connected 2+4 spherical phasor model

$$Q_{\alpha\beta} = \frac{1}{N} \sum_{k=1}^N \left(|a_k^{(\alpha)}|^2 - \langle |a_k|^2 \rangle \right) \left(|a_k^{(\beta)}|^2 - \langle |a_k|^2 \rangle \right)$$

Experimentally the same sample of random optical medium can be repeatedly illuminated by an external pumping laser under exactly the same conditions if the compound is solid (scatterers do not move between different random laser dynamics) and this implies that the mode-coupling random realization will be the same in different random laser dynamic stories.

Real replicas are feasible and in this cases, in principle

The IFO can be measured in experiments

Why not directly the phasor overlap?

Phases are unknown

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

Also instantaneous intensities are not measurable

$$I_k(\omega, t) = |a_k(\omega, t)|^2$$

We can only measure intensities on data acquisition intervals, i. e., averaged over the whole dynamics.

$$I_k(\omega) = \frac{1}{\mathcal{T}} \sum_{t=t_0}^{t_0+\mathcal{T}} |a_k(\omega, t)|^2 \simeq \langle |a_k(\omega)|^2 \rangle$$



- Intermezzo: the experimental measurement of the Parisi distribution of overlaps

Intensity Fluctuation Overlap - IFO
Fully connected 2+4 spherical phasor model

$$Q_{\alpha\beta} = \frac{1}{N} \sum_{k=1}^N \left(|a_k^{(\alpha)}|^2 - \langle |a_k|^2 \rangle \right) \left(|a_k^{(\beta)}|^2 - \langle |a_k|^2 \rangle \right)$$

If real replicas are feasible IFO's can be measured in experiments

The experimental intensity fluctuation of the shot (replica) α with respect to the average spectrum is

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

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

and their overlap is

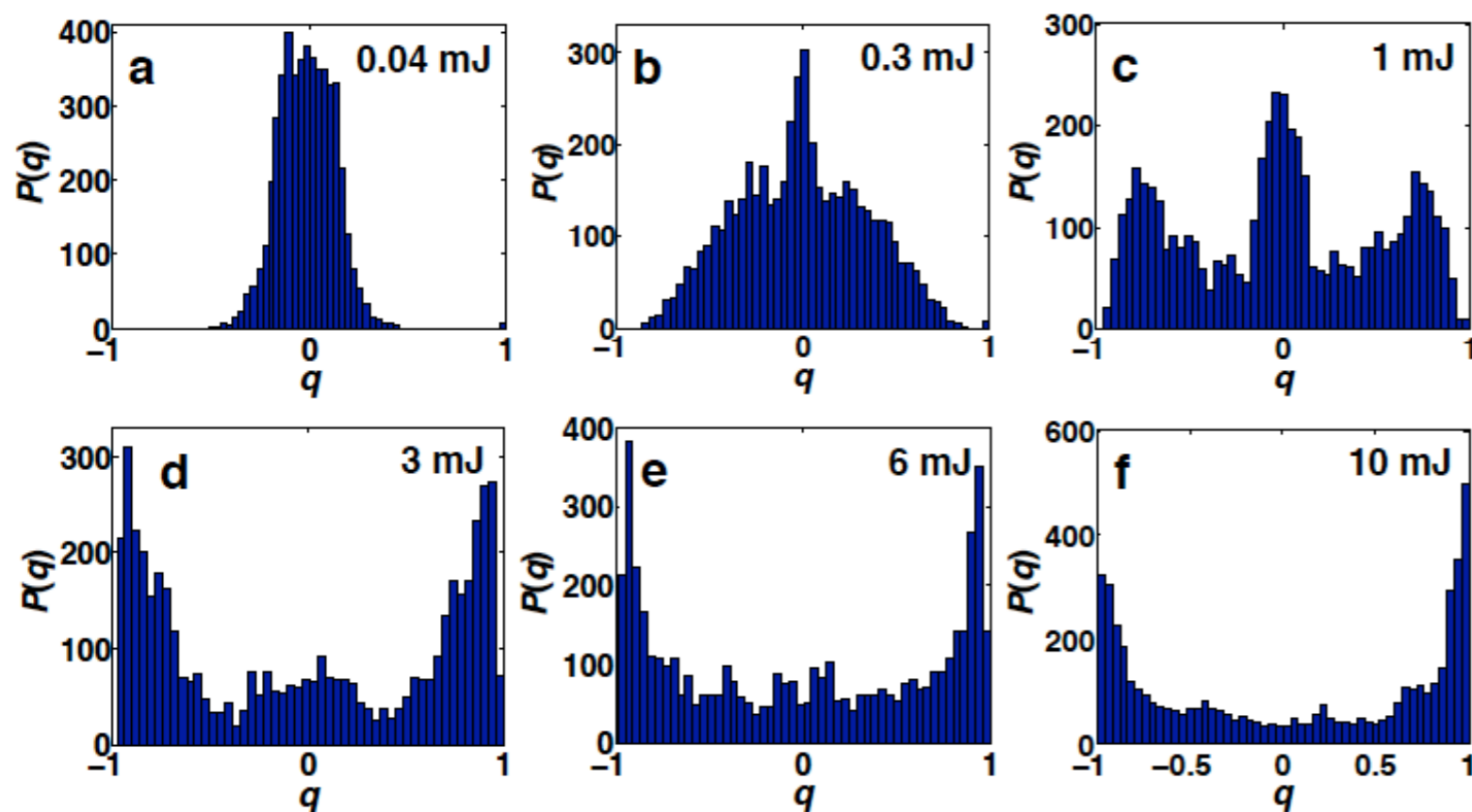
$$Q_{\alpha\beta} \equiv \frac{1}{N} \sum_{k=1}^N \Delta_k^\alpha \Delta_k^\beta$$

These overlaps can be measured over many shots at fixed external pumping power and the procedure can be repeated at different external pumping powers across the lasing transition to see how their distribution behaves



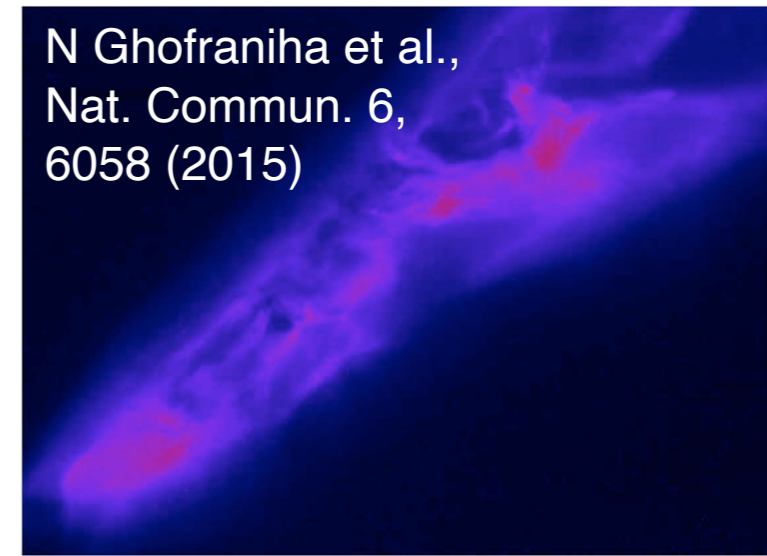
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Replica Symmetry Breaking is experimentally detected in IFO in the T50Cx Random Laser

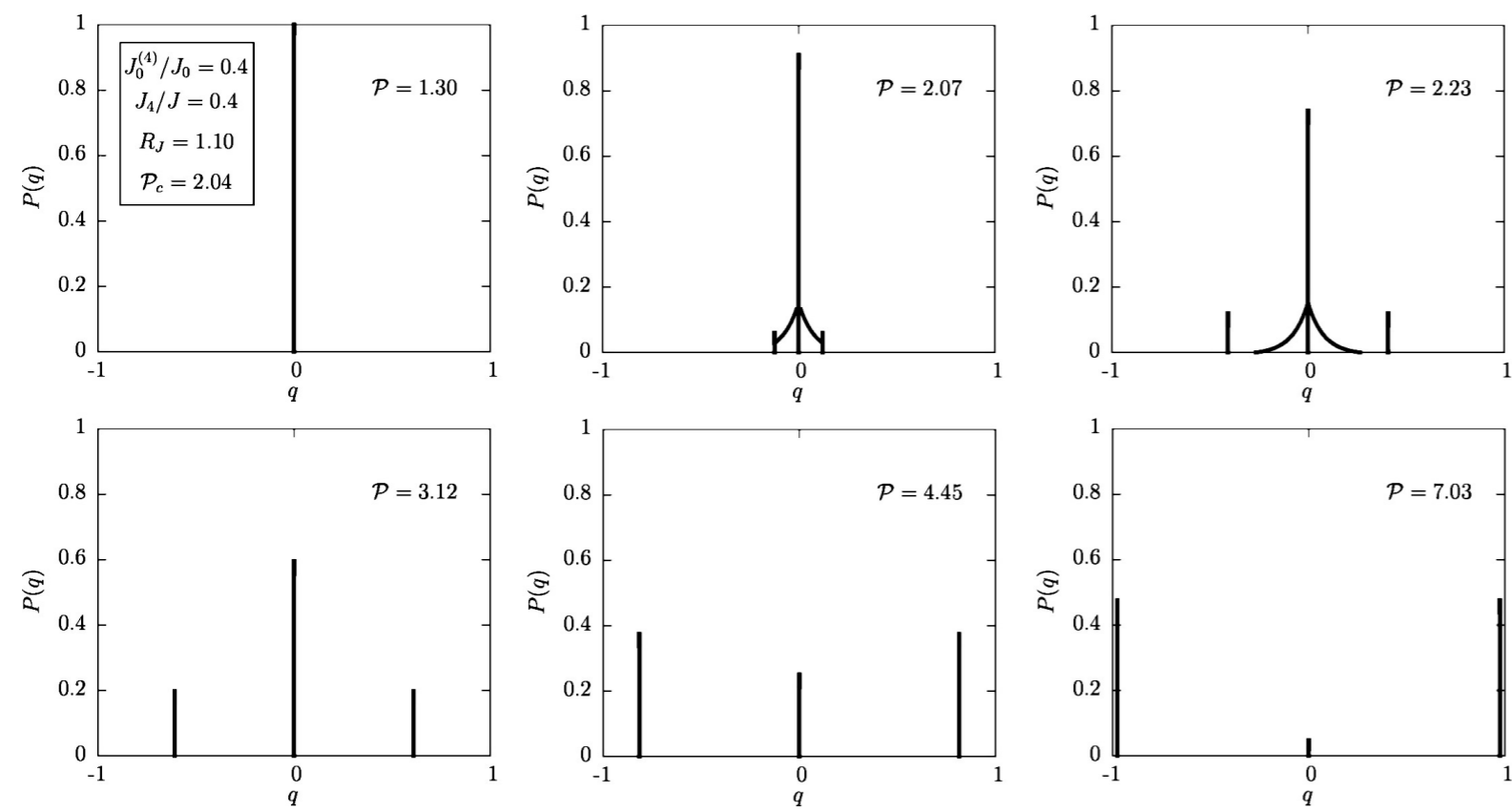


T50Cx grains

N Ghofraniha et al.,
Nat. Commun. 6,
6058 (2015)



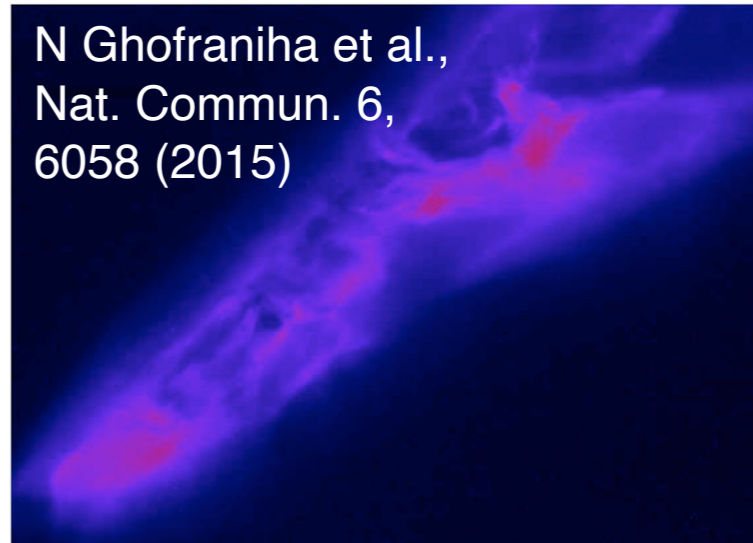
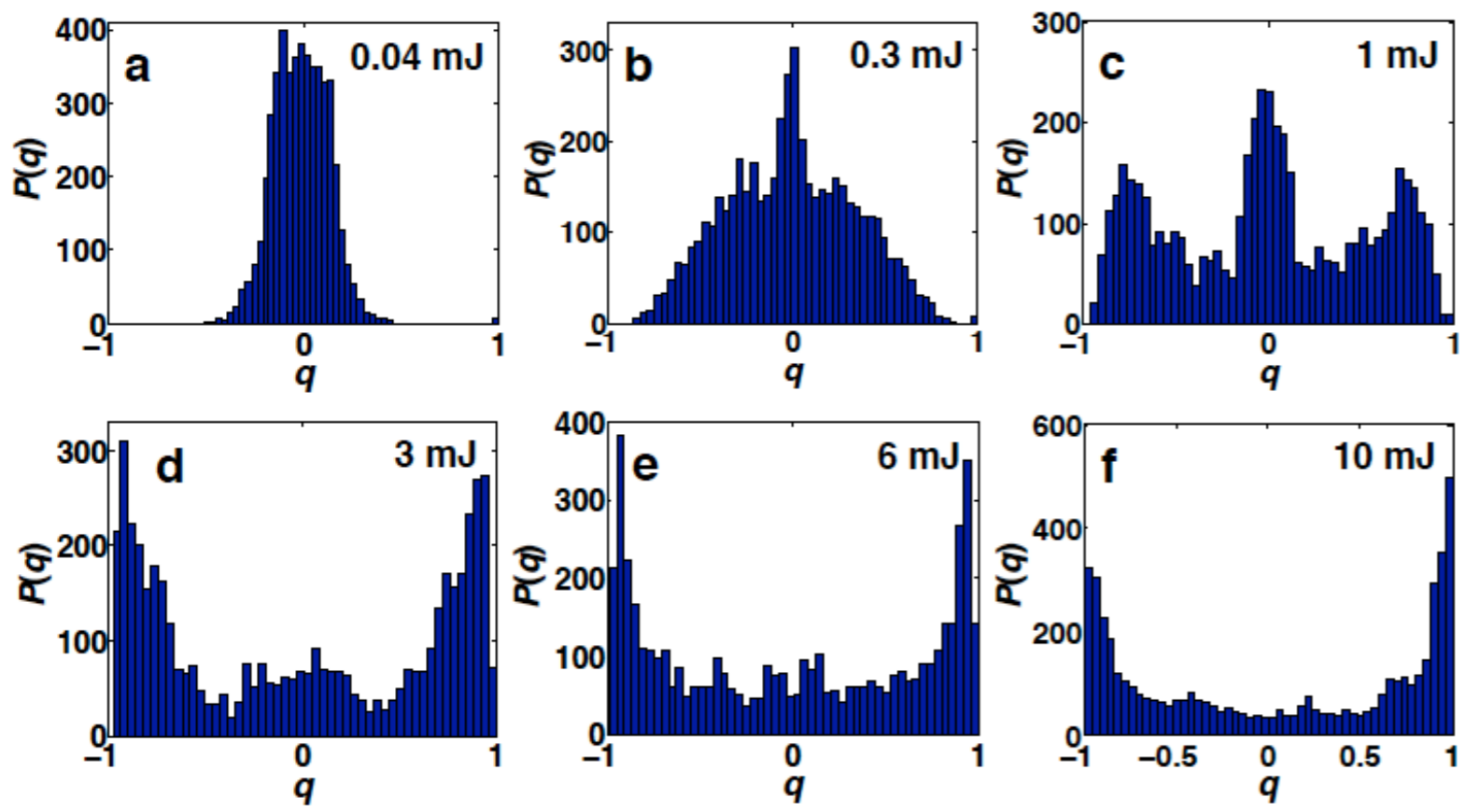
• Intermezzo: the experimental measurement of the Parisi distribution of overlaps



Mean-field fully connected theory $Q_{ab} = q_{ab}^2$

F. Antenucci, A. Crisanti, LL, SciRep 2015

Element-element relation: any Replica Symmetry Breaking is detectable and it is experimentally detected in IFO in the T5OCx Random Laser
first direct measurement



N Ghofraniha et al., Nat. Commun. 6, 6058 (2015)

T5OCx grains

- In between theory and experiment

Differences between mean-field theory on complete graph and experiments

Fully connected RSB theory

- In the narrowband approximation **mode-locking does not play any role** and the **interaction graph is complete**
- In the *thermodynamic limit of infinite* number of modes
- under constant energy and **effective equilibrium** assumptions and using an equilibrium ensemble of *instantaneous* modes

$$I_k(t) = |a_k(t)|^2$$

In experiments:

- **mode-locking** is expected to occur and the interaction **graph is unknown**, as well as the magnitude of the couplings
- the number of modes is **finite** and also their resolution
- **equilibration is not under control** and we do not have access to instantaneous resonances but only to the total intensity acquired

$$\langle I_k(t) \rangle = \frac{1}{\mathcal{T}} \int_{t_0}^{t_0 + \mathcal{T}} dt I_k(t)$$



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- In between theory and experiment:
a mode-locking model

We include mode frequencies and frequency matching for a more realistic model

We also simplify some (momentarily less essential) features:

- * only 4-“spins” (no losses, flat gain profile)
- * comb-like distributed frequencies (easier combinatorics)

spherical 4-phasor
mode-locked
random laser

$$\mathcal{H}[\mathbf{a}] = - \sum_{\mathbf{k} | \text{FMC}(\mathbf{k})} J_{k_1 k_2 k_3 k_4} \bar{a}_{k_1} a_{k_2} \bar{a}_{k_3} a_{k_4} + \text{c.c.}$$

$$\text{FMC}(\mathbf{k}) : |\omega_{k_1} - \omega_{k_2} + \omega_{k_3} - \omega_{k_4}| \lesssim \gamma$$

$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

Random couplings

$$\mathcal{P}(J_{k_1 \dots k_p}) = \frac{1}{\sqrt{2\pi\sigma_p^2}} \exp \left\{ -\frac{J_{k_1 \dots k_p}^2}{2\sigma_p^2} \right\}$$

$$\sigma_p^2 = \overline{J_{k_1 \dots k_p}^2} \propto \frac{1}{N^{p-2}}$$

$$p = 4 \quad \sigma_4^2 = \overline{J_{k_1 k_2 k_3 k_4}^2} \propto \frac{1}{N^2}$$

Equispaced comb-like frequencies

$$\omega_k = \omega_0 + \delta\omega k \quad \longrightarrow \quad |k_1 - k_2 + k_3 - k_4| = 0 \quad , k_i = 1, \dots, N$$

G. Gradenigo, F. Antenucci, LL, Phys Rev Res 2020

J. Niedda, G. Gradenigo, LL and G. Parisi, SciPost Phys. 2023

J. Niedda, G. Gradenigo, LL, JSTAT 2023



- In between theory and experiment: a mode-locking model

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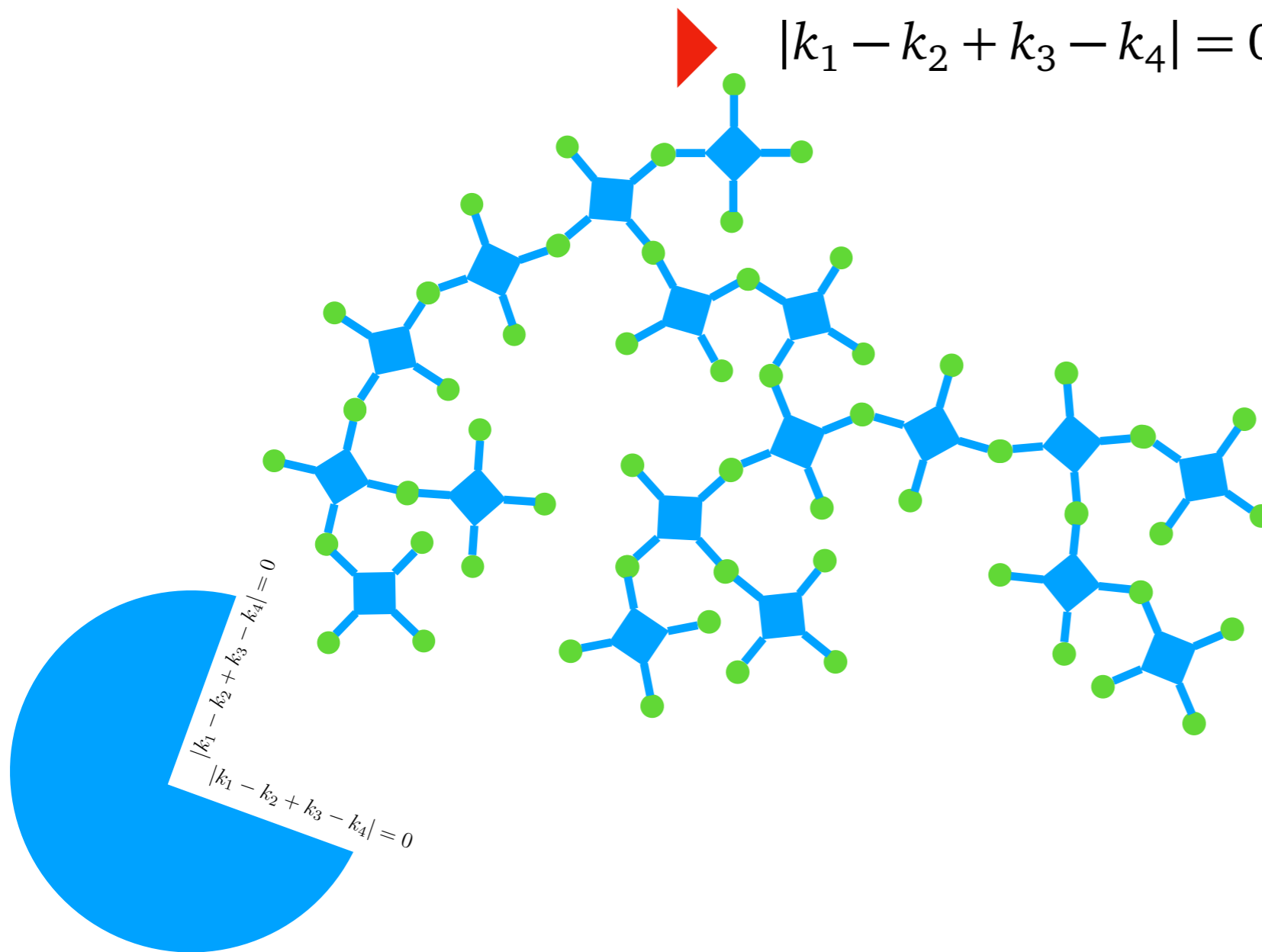
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Effect of frequency matching condition on interaction graph connectivity

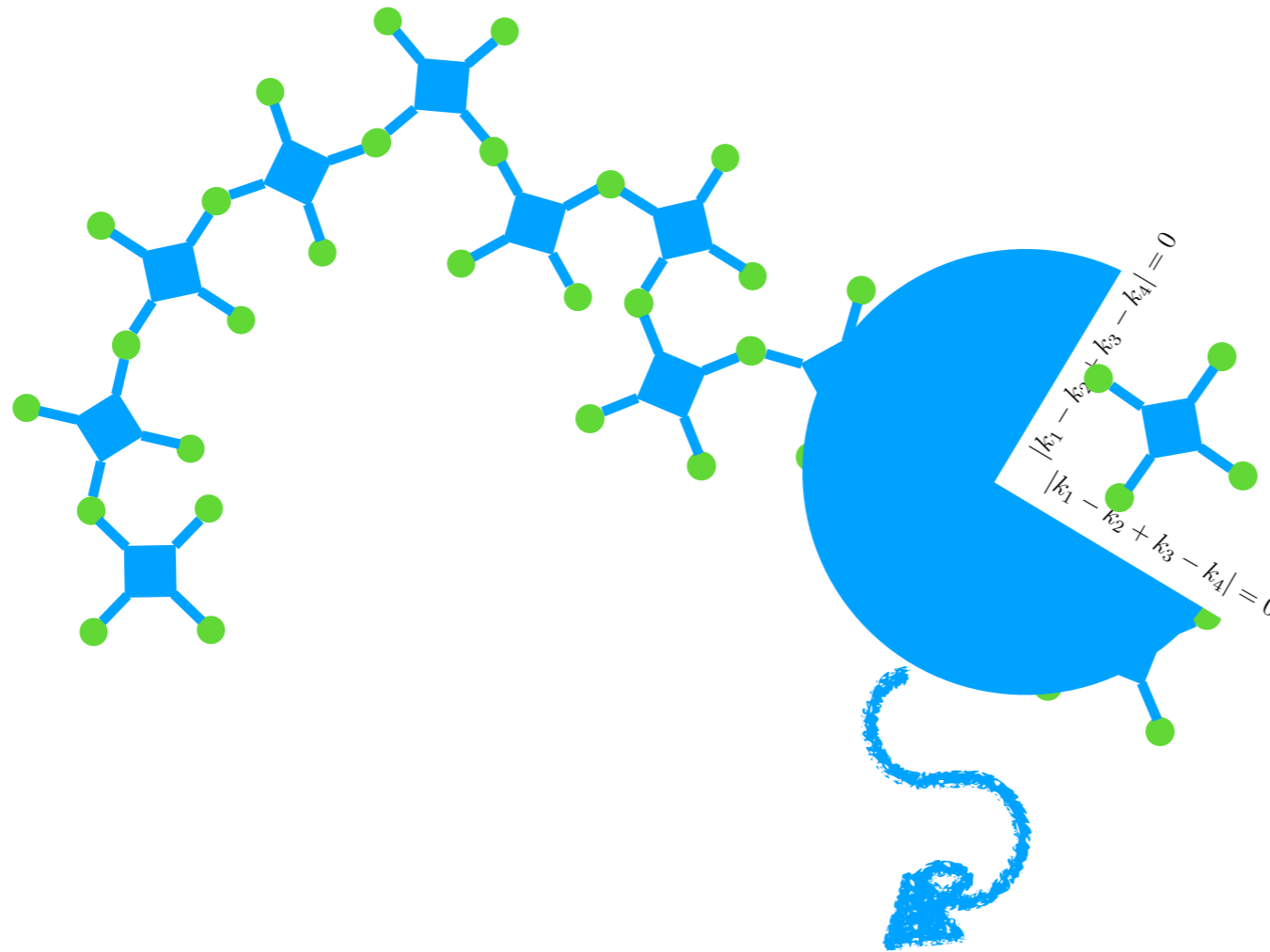


spherical 4-phasor
mode-locked
random laser



Effect of frequency matching condition on interaction graph connectivity

▶ $|k_1 - k_2 + k_3 - k_4| = 0$



spherical 4-phasor
mode-locked
random laser

$$\binom{N}{4} \times \left(\frac{2}{3N} + \frac{1}{3N^3} \right) = \mathcal{O}(N^3)$$

Complete factor graph

Frequency matching condition pruning
~~decimation~~



- * Exchange Monte Carlo for equilibrium study
- * Parallel computation of contribution to single mode energy update

Energy of a configuration of N phasors

$$\mathcal{H}[\mathbf{a}] = - \sum_{\mathbf{k} | \text{FMC}(\mathbf{k})} J_{k_1 k_2 k_3 k_4} \bar{a}_{k_1} a_{k_2} \bar{a}_{k_3} a_{k_4} + \text{c.c.} = \underbrace{J_{1234} \bar{a}_1 a_2 \bar{a}_3 a_4 + J_{1235} \bar{a}_1 a_2 \bar{a}_3 a_5 + J_{1239} \bar{a}_1 a_2 \bar{a}_3 a_9 + \dots}$$

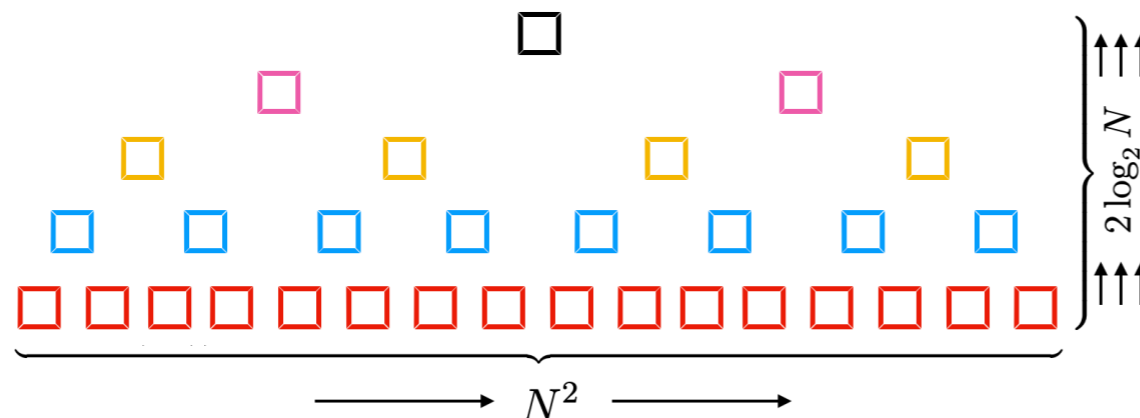
is the sum of $\binom{N}{4} \times \left(\frac{2}{3N} + \frac{1}{3N^3} \right) = \mathcal{O}(N^3)$ terms

Complete factor graph Frequency matching condition pruning

Single mode energy update

$$a_k \rightarrow a'_k \Rightarrow \Delta E \equiv \mathcal{H}[\mathbf{a}'] - \mathcal{H}[\mathbf{a}] \text{ is the sum of } \mathcal{O}(N^2) \text{ terms}$$

Each term in the sum computed in parallel on GPU kernels



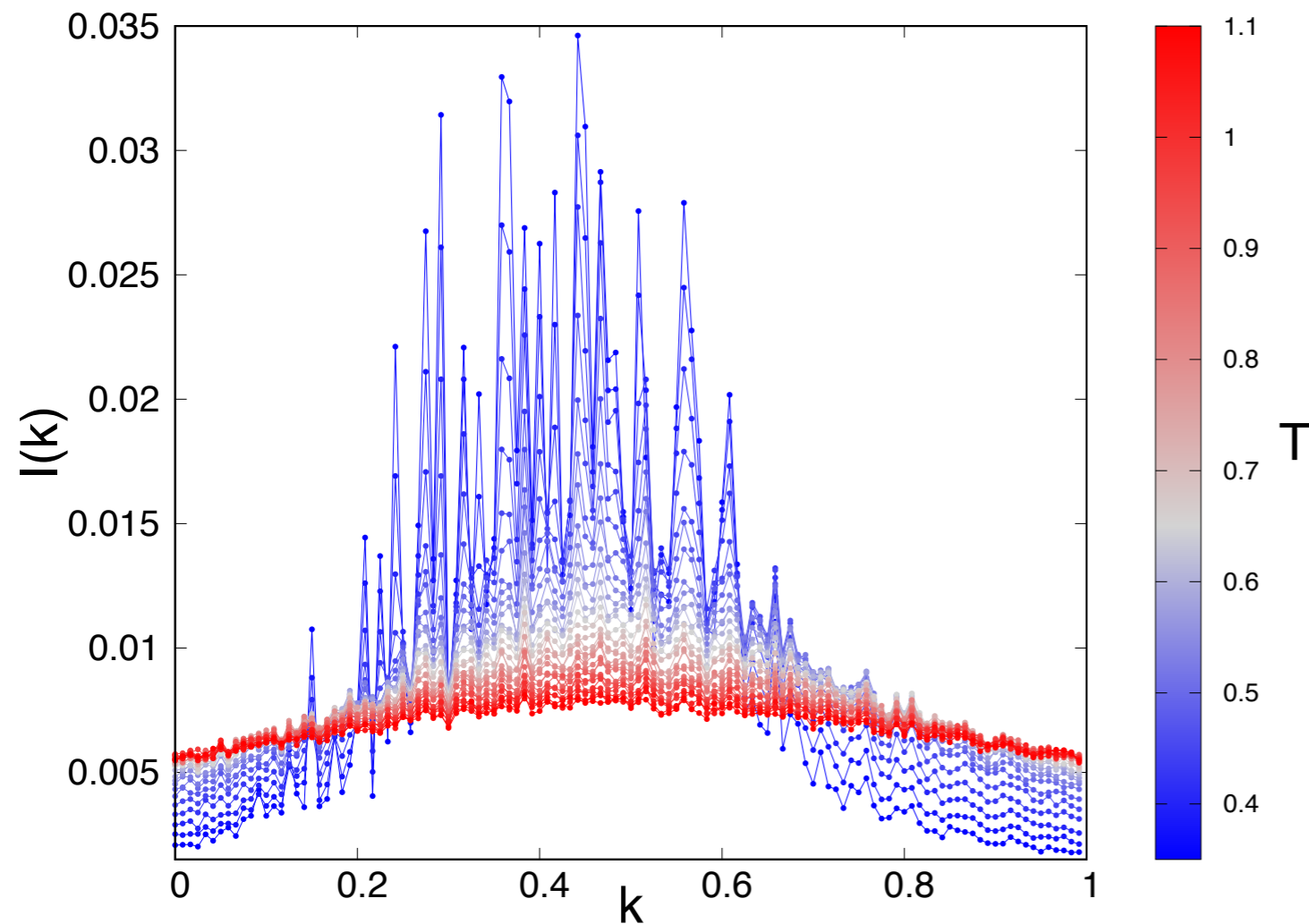
The time required to carry out
the sum scales as $\log_2 N$



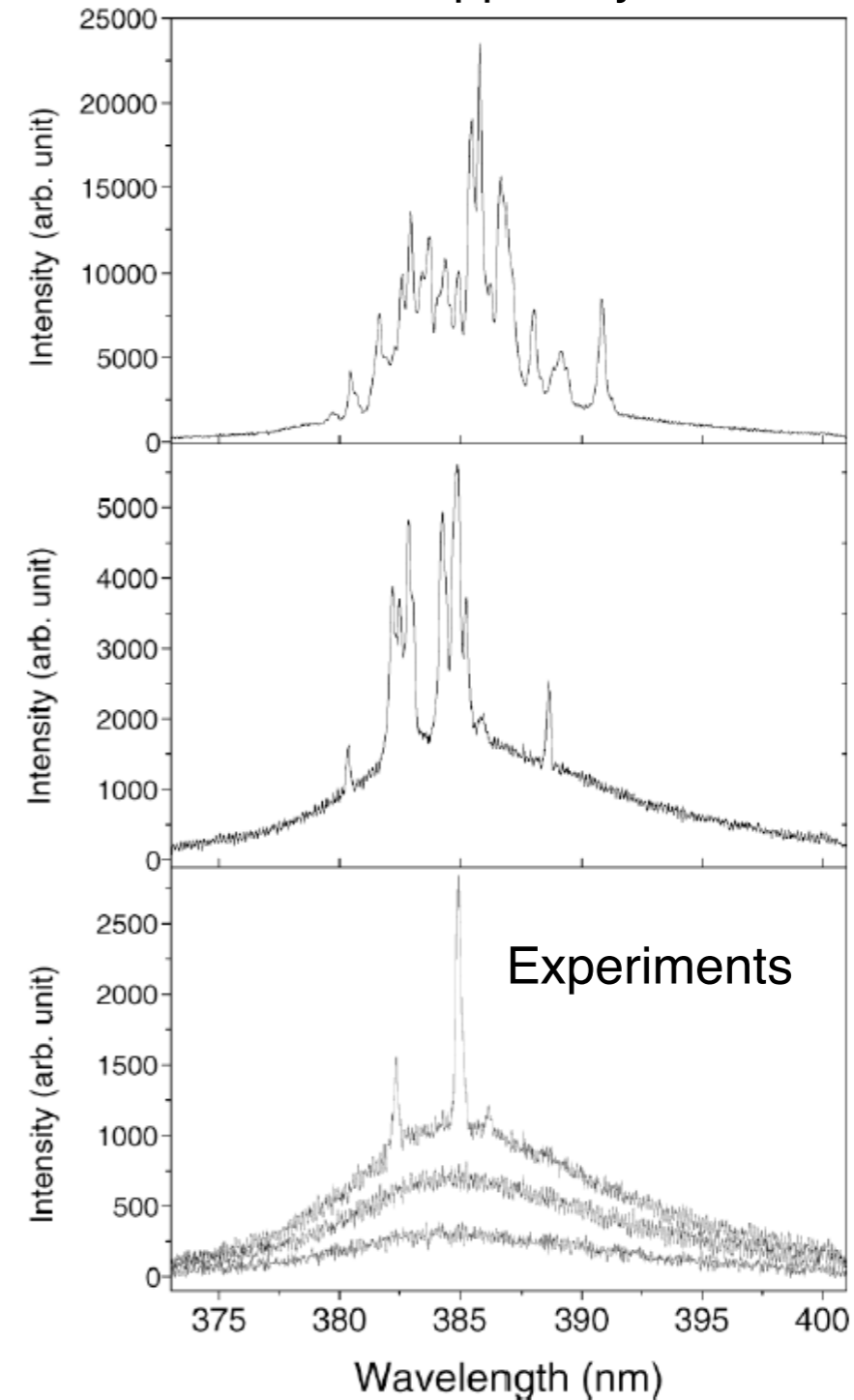
Monte Carlo simulations of the spherical 4-phasor mode-locked random laser

Spectral emission

Spectral narrowing and sharp resonances as pumping increases



Hui Cao et. al , Appl. Phys. Lett. 1998



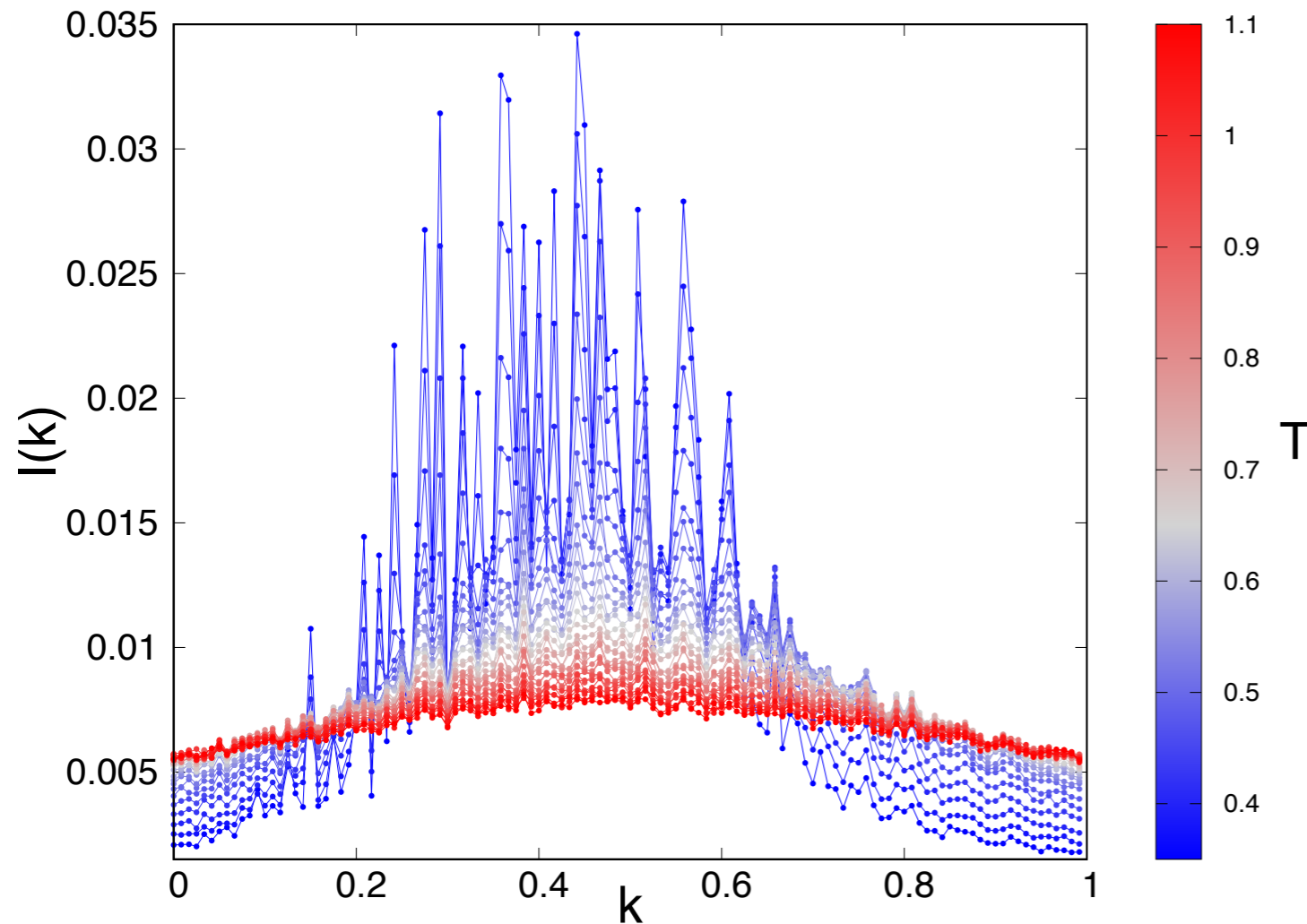
$$\mathcal{P}^2 = \frac{\epsilon^2}{T} = \frac{1}{T_{\text{photonic}}} \quad |k_1 - k_2 + k_3 - k_4| = 0$$



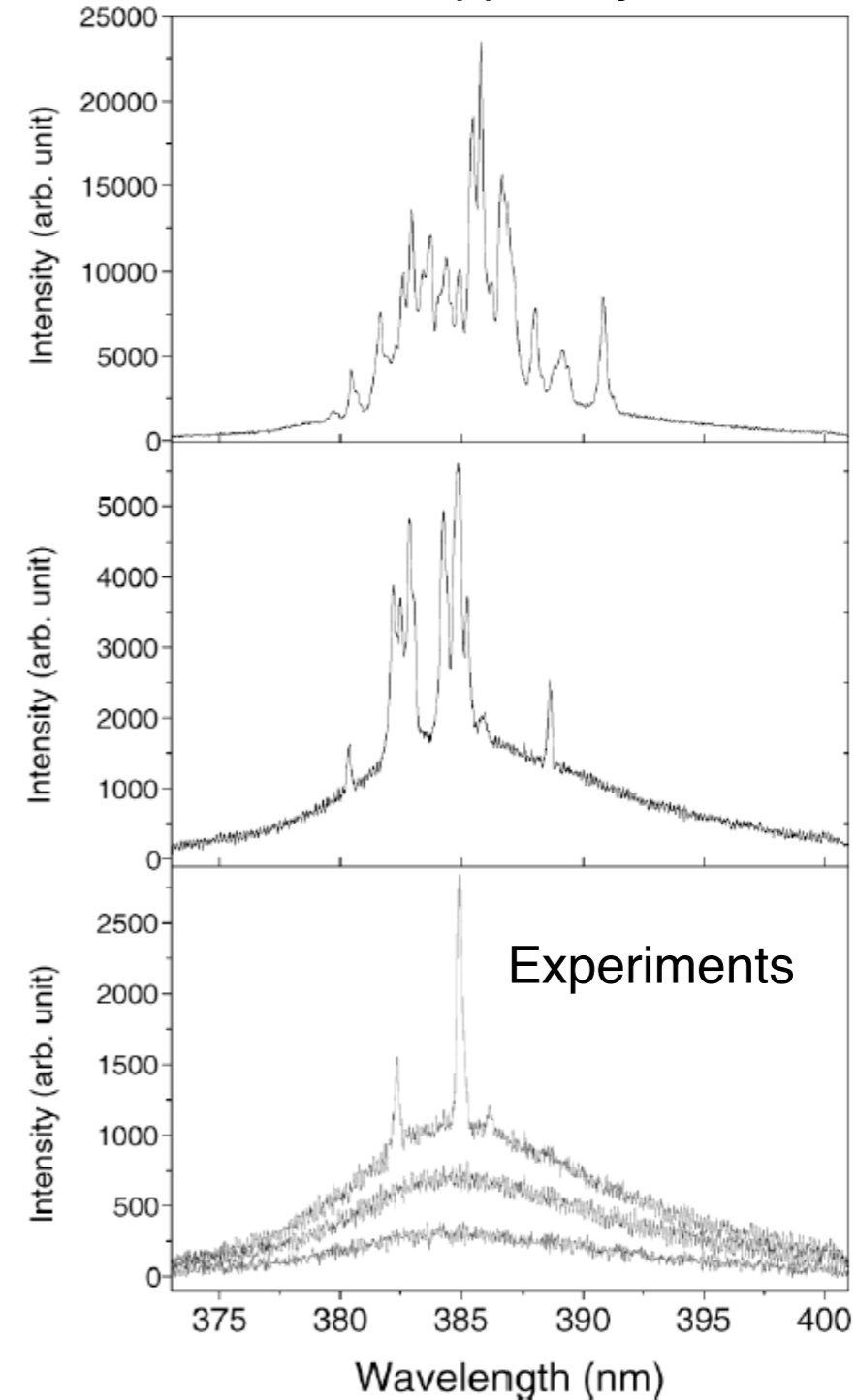
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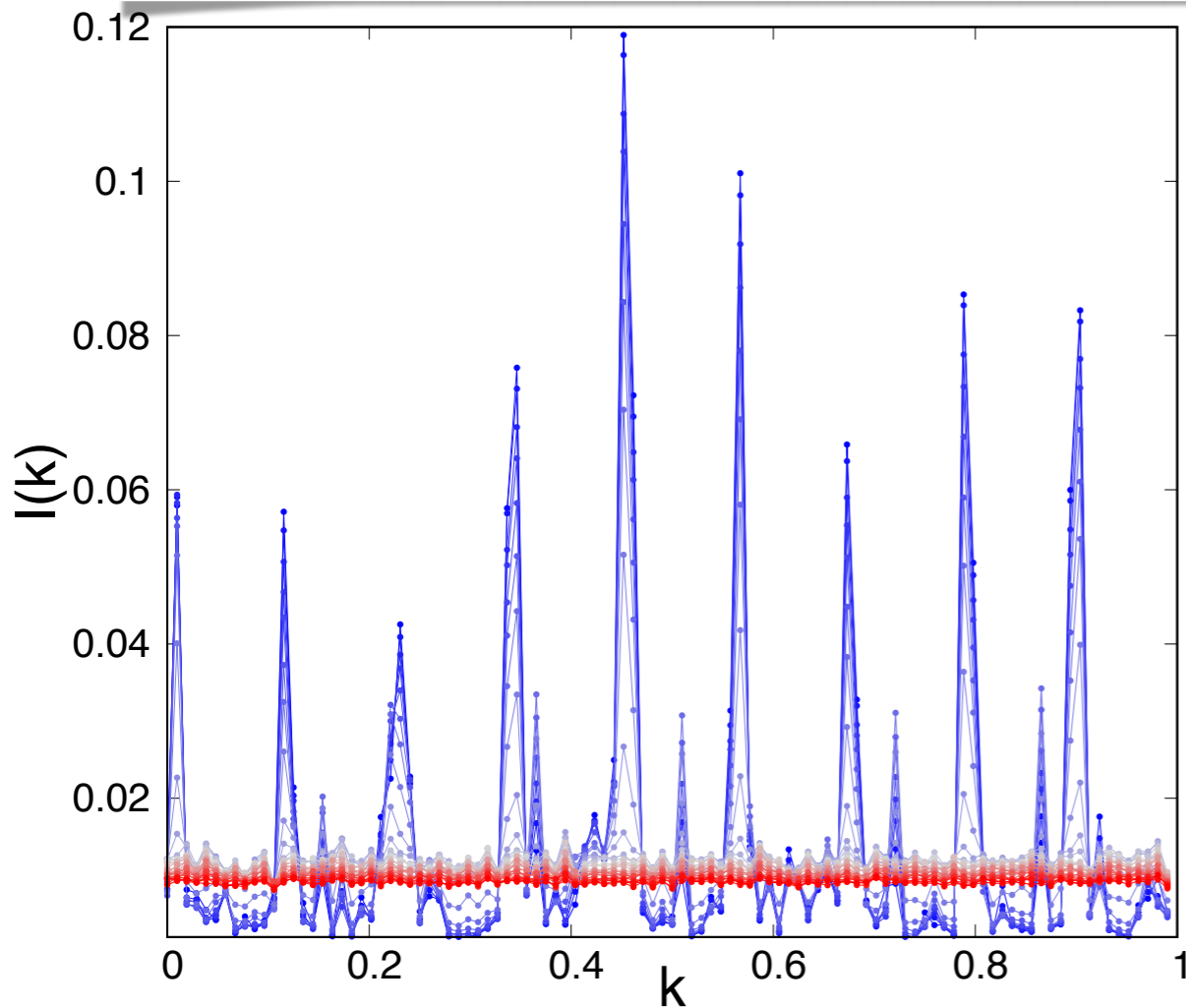
Strong finite size effects (large sizes of continuous variables on densely connected interaction graphs take long times to simulate)



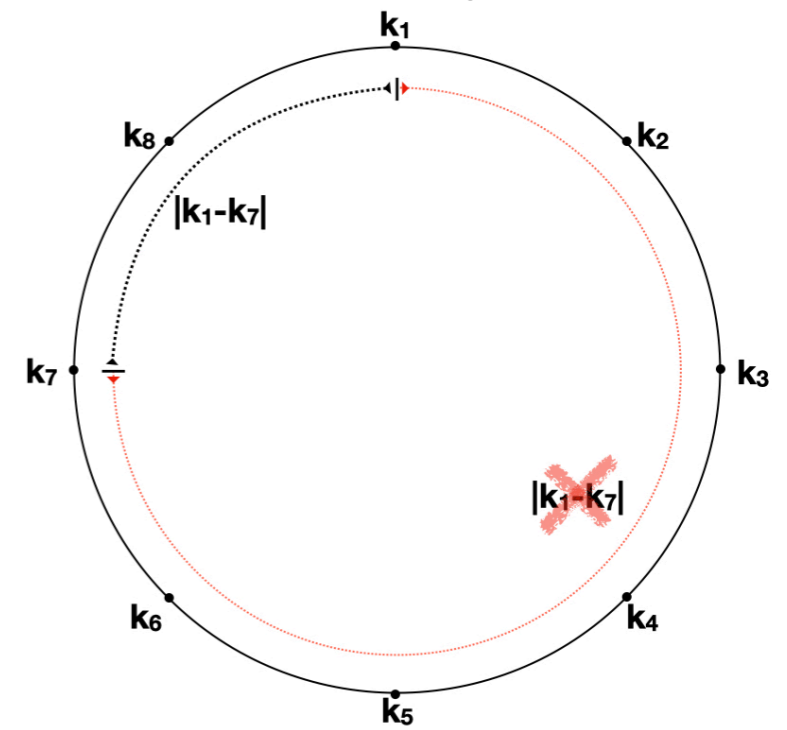
Figure 1. Spectra of emission from ZnO powder when the excitation intensity is (from bottom to top) 400, 562, 763, 875 and 1387 kW cm⁻².

Monte Carlo simulations of the spherical 4-phasor mode-locked random laser

A trick to reduce large FSE is to impose periodic boundary conditions *on the mode frequencies*



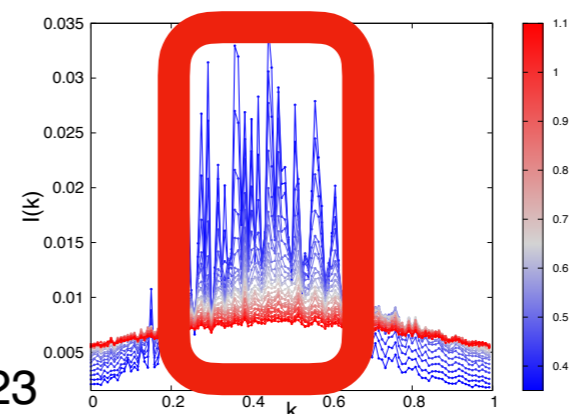
Frequency periodic boundary conditions



$$|k_1 - k_2 + k_3 - k_4| = 0$$

$$|k_a - k_b| = \begin{cases} |k_a - k_b| & \text{if } |k_a - k_b| \leq \lfloor \frac{N}{2} \rfloor \\ N - |k_a - k_b| & \text{if } |k_a - k_b| \geq \lfloor \frac{N}{2} \rfloor \end{cases}$$

As if only central modes count



$$\mathcal{P}^2 = \frac{\epsilon^2}{T} = \frac{1}{T_{\text{photonic}}}$$



Monte Carlo simulations of the spherical 4-phasor mode-locked random laser

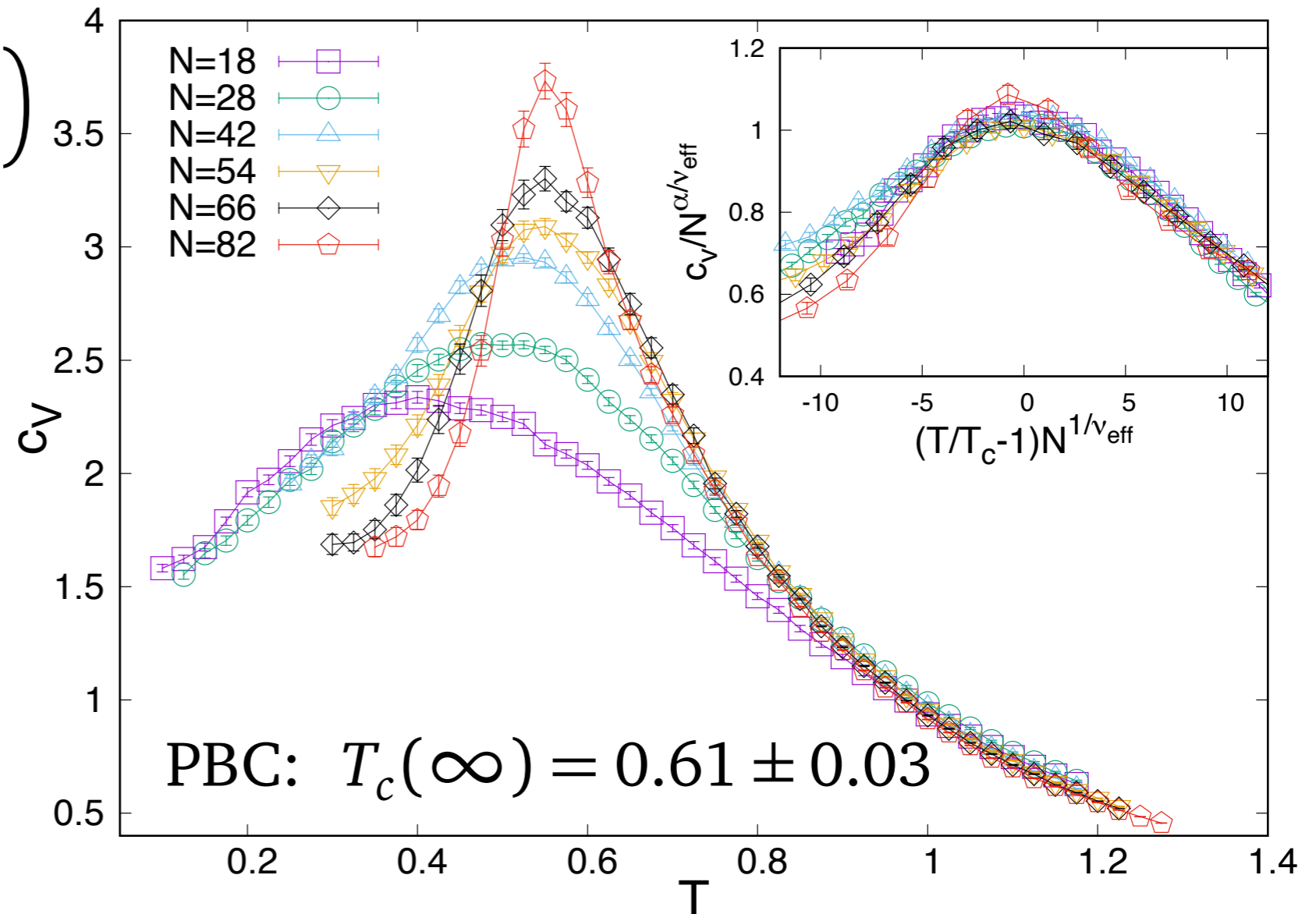
Phase transition and universality class

$$c_{V_N}(T) = N^{\frac{\alpha}{\nu_{\text{eff}}}} \hat{f}_{c_{V_N}} \left(N^{\frac{1}{\nu_{\text{eff}}}} t_N \right)$$

$$\nu_{\text{eff}} = 2\beta + \gamma$$

$1 \leq \nu_{\text{eff}} \leq 2$
for a mean-field model

PBC: Periodic boundary conditions on
frequency to reduce finite size effects



Strong finite size effects, yet **compatible with a mean-field theory**

PBC: $\alpha = 0.27 \pm 0.05$,

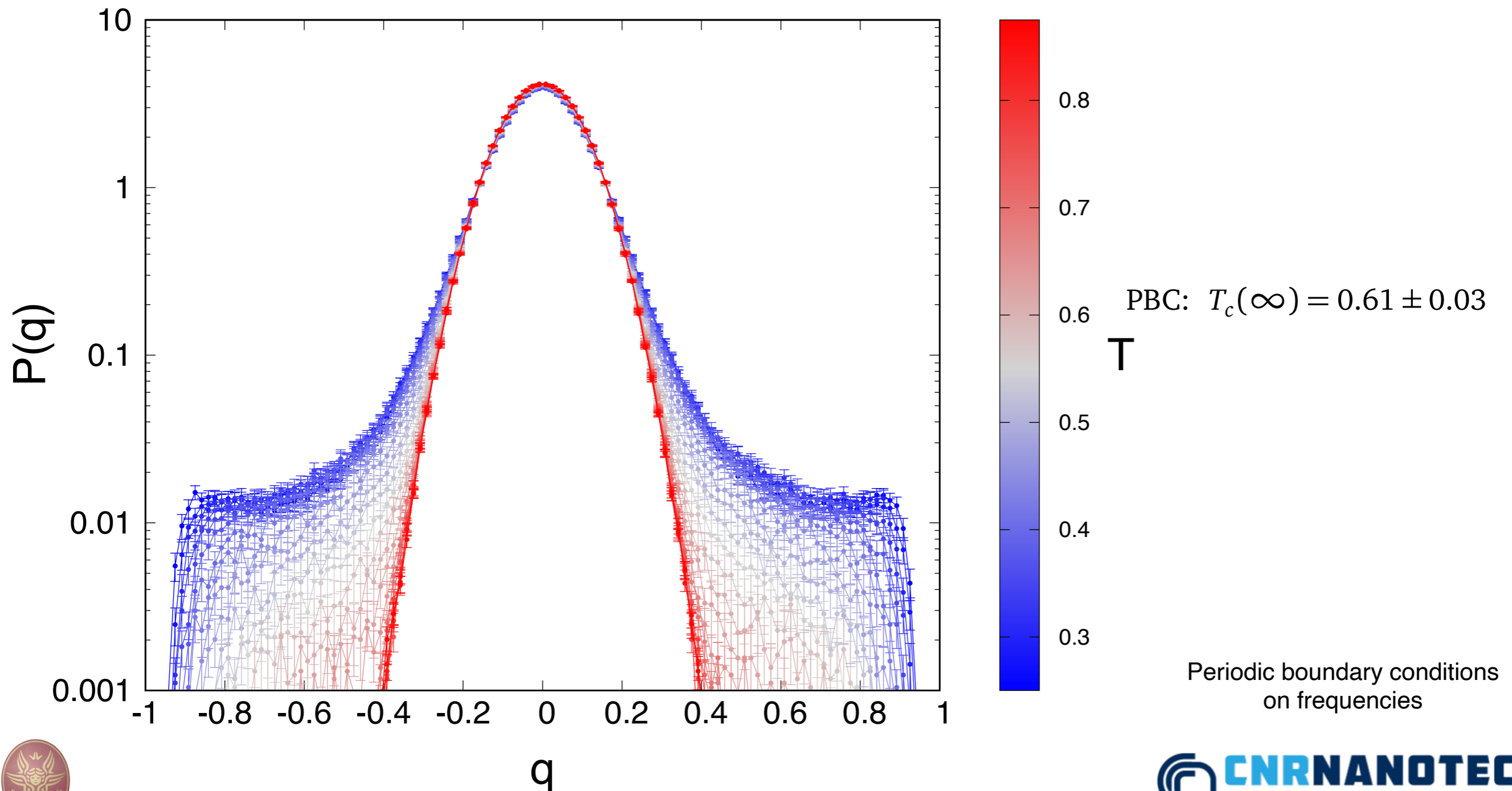
$1/\nu_{\text{eff}} = 0.86 \pm 0.14$



Monte Carlo simulations of the spherical 4-phasor mode-locked random laser

Replica symmetry breaking

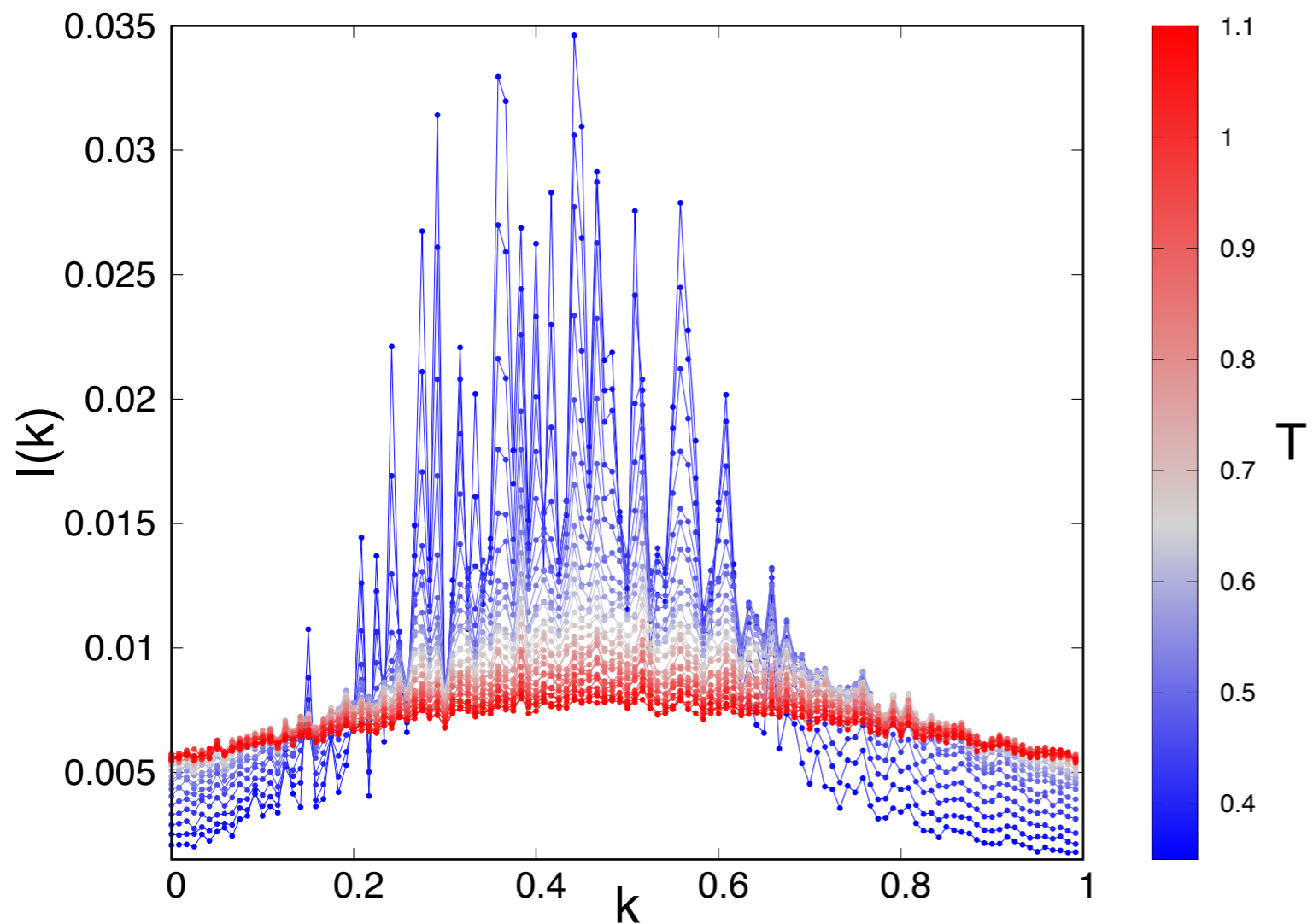
non-trivial distribution below the transition point:
non-gaussian tails and side peaks at low T



- Standard and random lasers
- Statistical physics approach to laser physics
 - Theory for ultrafast mode-locked multimode lasers (order, closed cavity)
 - Theory for random lasers: a mode-locked spin-glass theory (disorder, open cavity)
- The narrow-band solution, phase diagrams, replica symmetry breaking, a new overlap: intensity fluctuation overlap
- Intermezzo: the experimental measurement of the Parisi distribution of overlaps
- In between theory and experiment: a mode-locking model
 - Monte Carlo dynamics simulation with exchange Monte Carlo, GPU parallel computing
- Power distribution among modes in the glassy light regime: condensation vs equipartition at high pumping
- Outlook (work in progress)



How is the power distributed among modes?

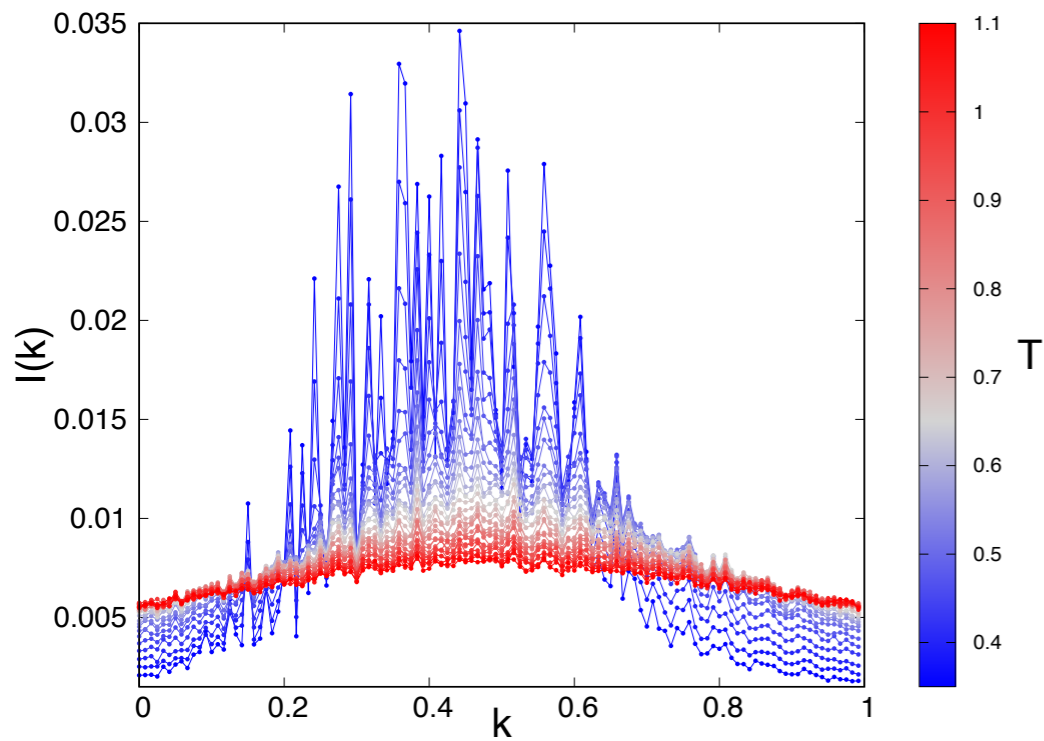


$$I_k = |a_k|^2$$

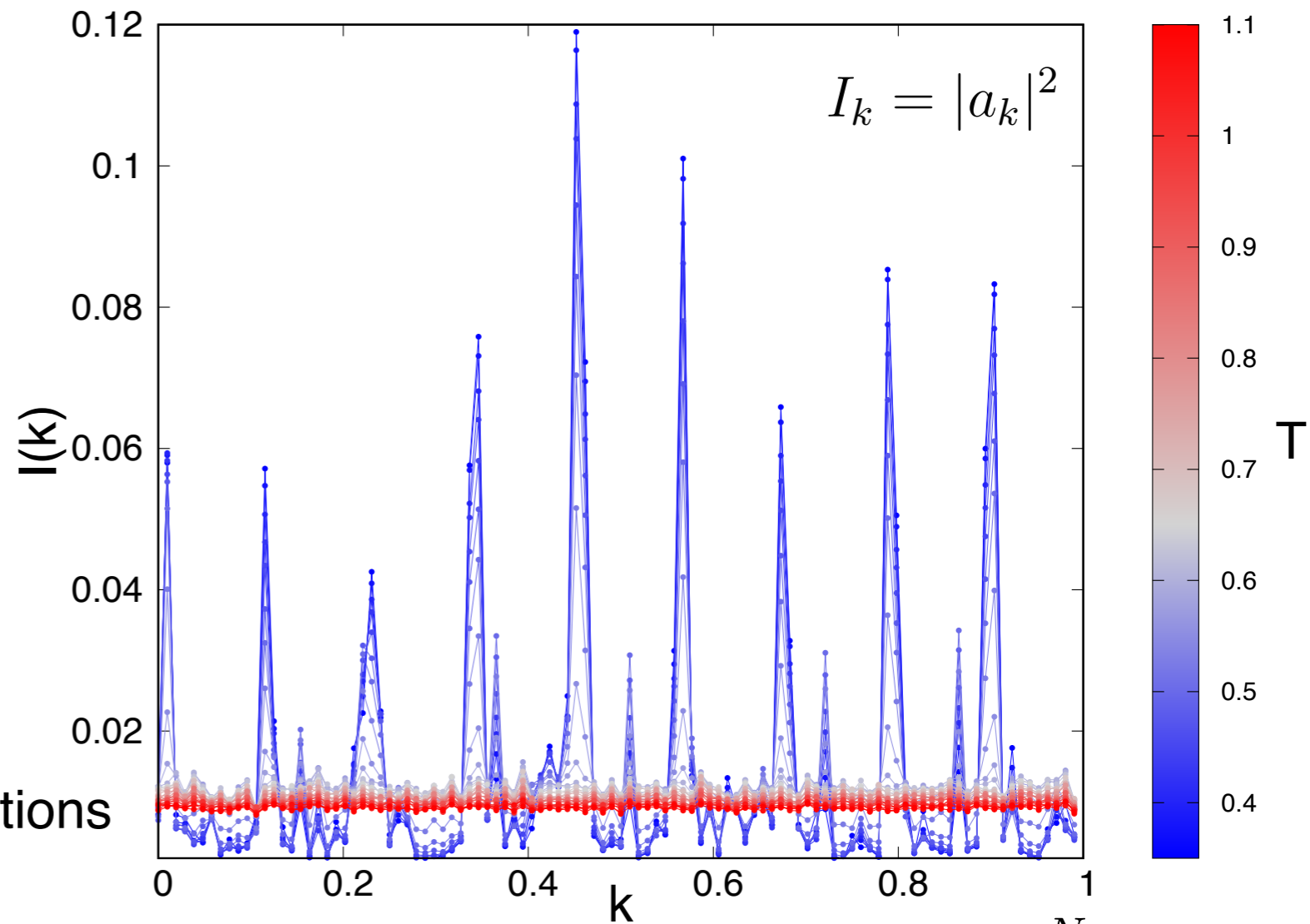
$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$



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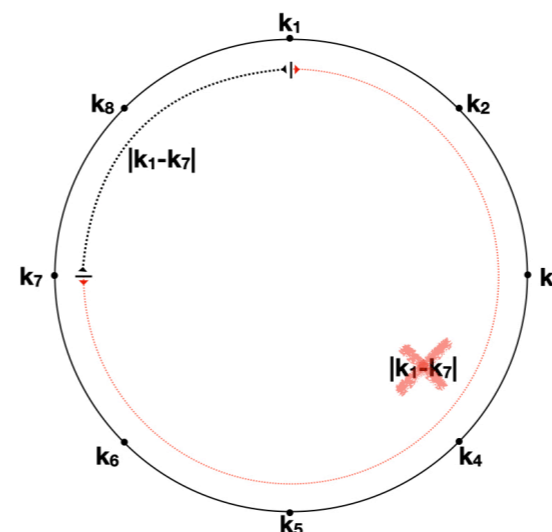


Frequency Periodic Boundary Conditions



$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

PBC reduce finite size effects



$$|k_1 - k_2 + k_3 - k_4| = 0$$

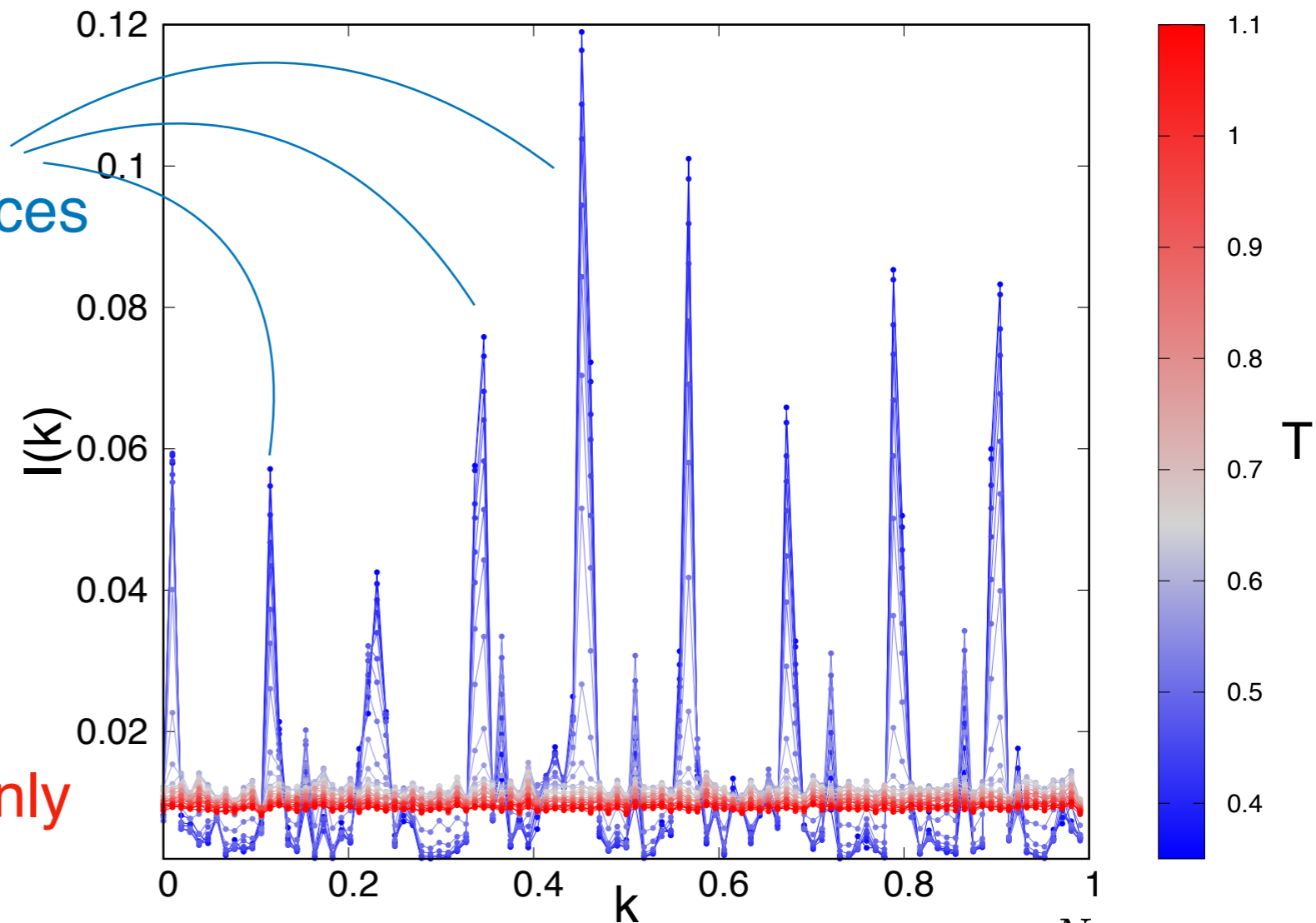
$$|k_a - k_b| = \begin{cases} |k_a - k_b| & \text{if } |k_a - k_b| \leq \lfloor \frac{N}{2} \rfloor \\ N - |k_a - k_b| & \text{if } |k_a - k_b| \geq \lfloor \frac{N}{2} \rfloor \end{cases}$$



How is the power distributed among modes?

High P/low T: intensity is
concentrated on a few resonances

High T/low P: intensity is evenly
distributed among modes



$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

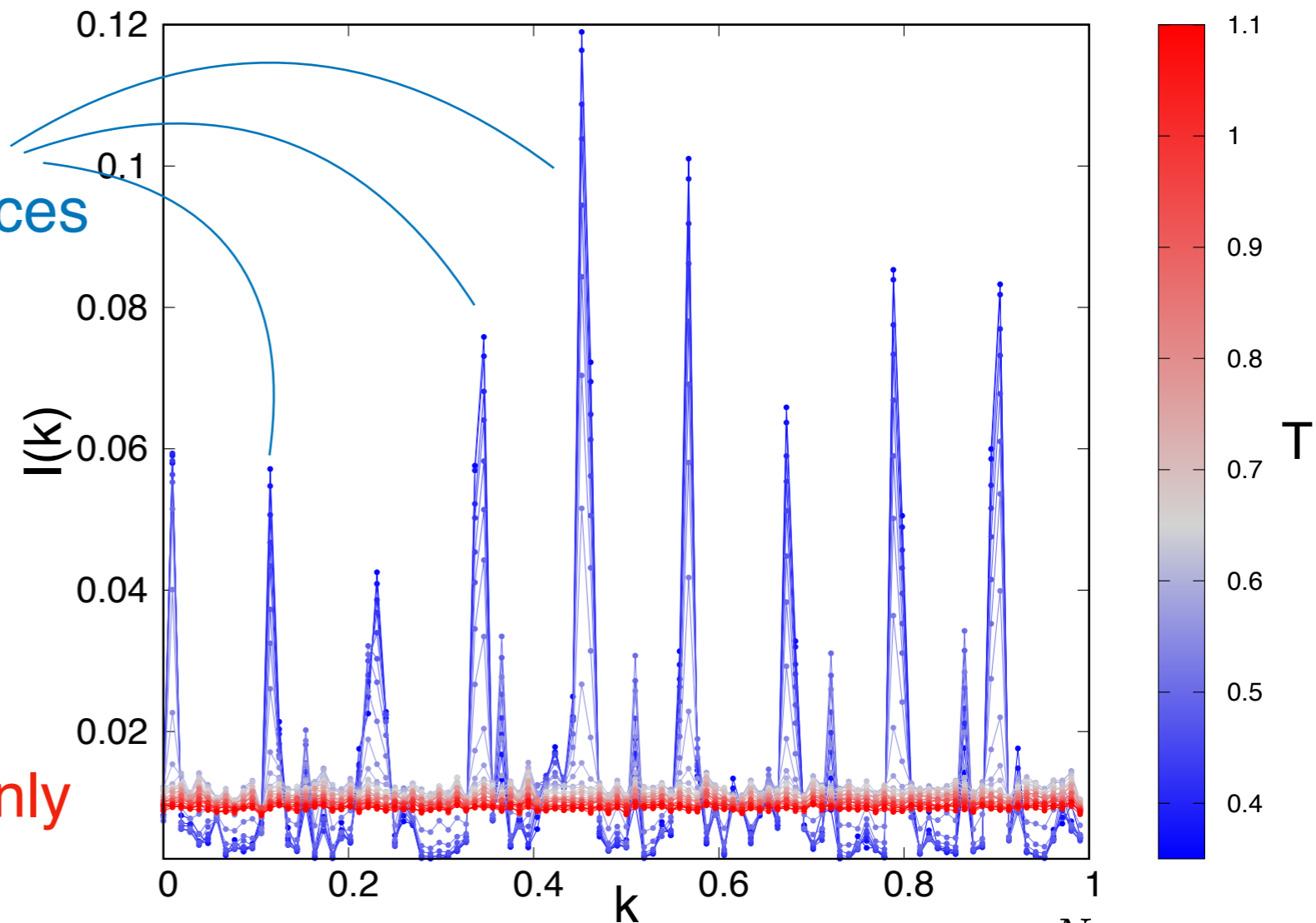


How is the power distributed among modes?

High P/low T: intensity is concentrated on a few resonances

Are "a few" very few?
Do they take all the intensity distributed in the system?

High T/low P: intensity is evenly distributed among modes



$$\mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

$$\sum_{k=1}^N \frac{|a_k|^2}{\epsilon} = N$$

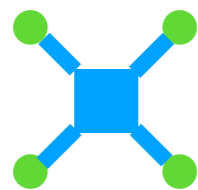
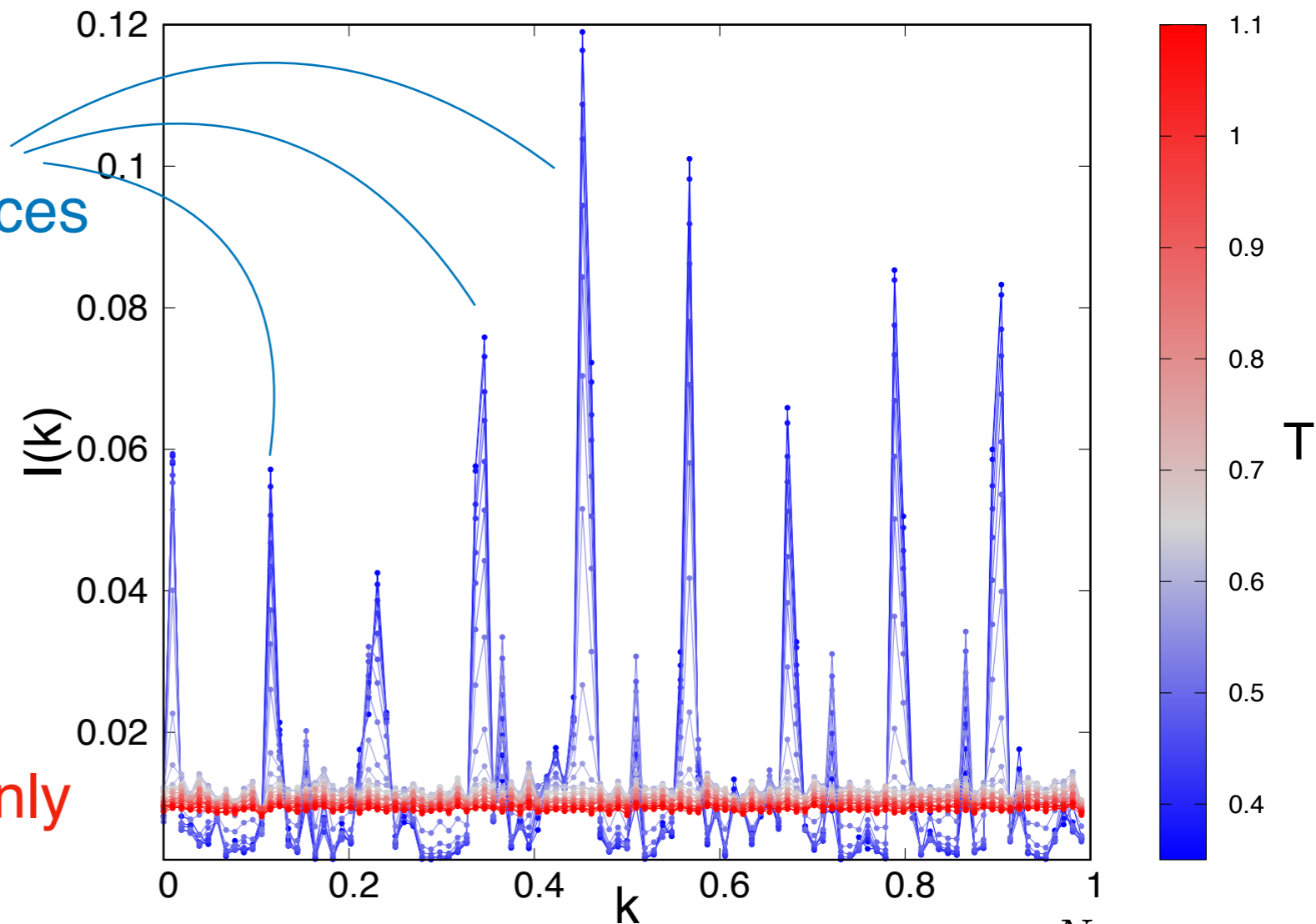


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$\{\mathbf{a}\}$:
CONDENSED

$$|a_k| \in \square \propto \sqrt{N}, \quad |a_k| \notin \square = 0$$

?

$$\sum_{k=1}^N \frac{|a_k|^2}{\epsilon} = N$$

$\{\mathbf{a}\}$:
UNIFORM

$$|a_k| \simeq 1 \quad \forall k = 1, \dots, N$$



How is the power distributed among modes?

Participation ratio:
$$Y_2 = \left\langle \frac{\sum_{k=1}^N I_k^2}{(\sum_{k=1}^N I_k)^2} \right\rangle = \frac{1}{N^2} \left\langle \sum_{k=1}^N I_k^2 \right\rangle \quad I_k = |a_k|^2$$

vs T

$$\lim_{N \rightarrow \infty} Y_2 = \text{const}$$

condensation

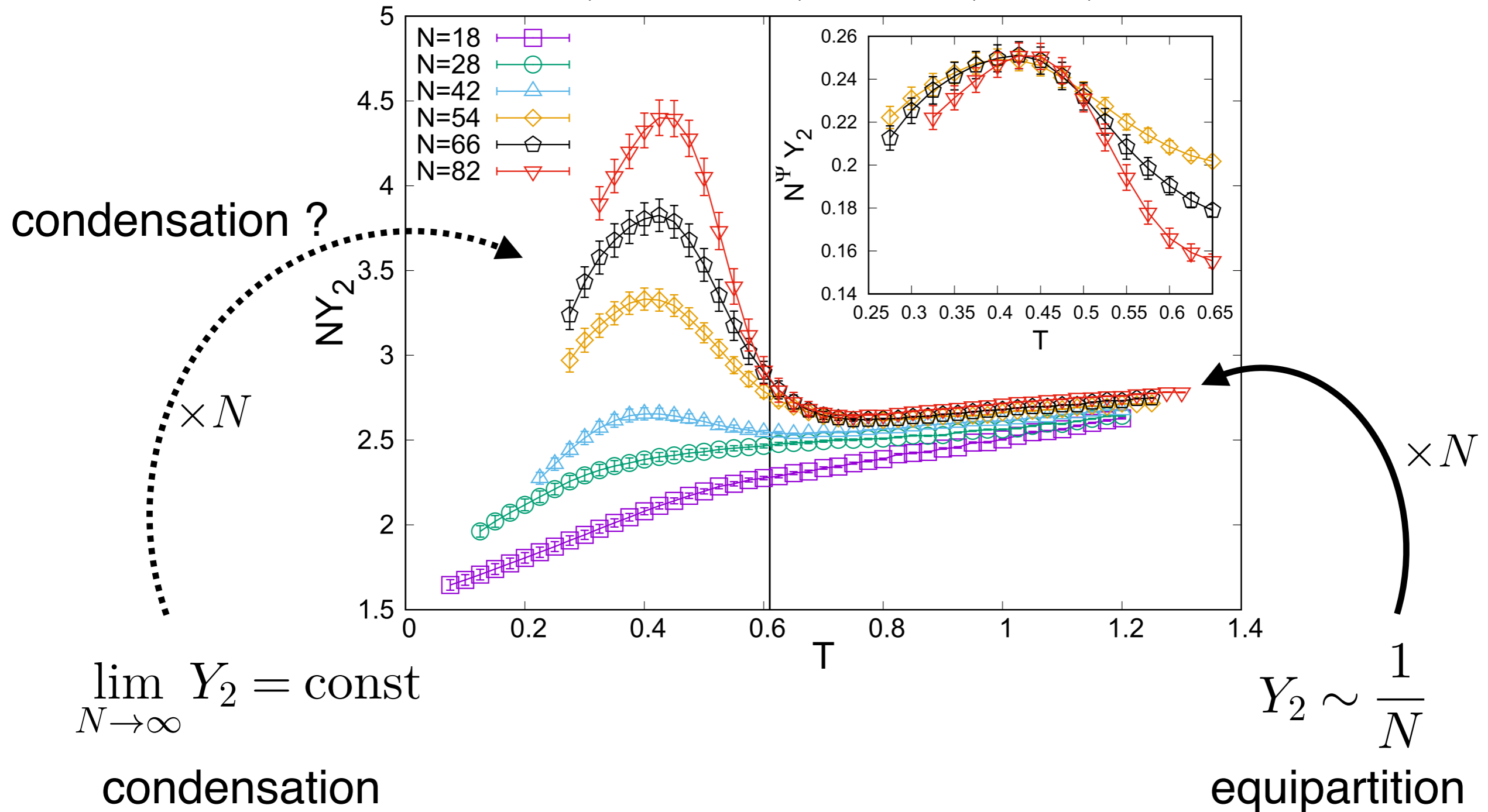
$$Y_2 \sim \frac{1}{N}$$

equipartition



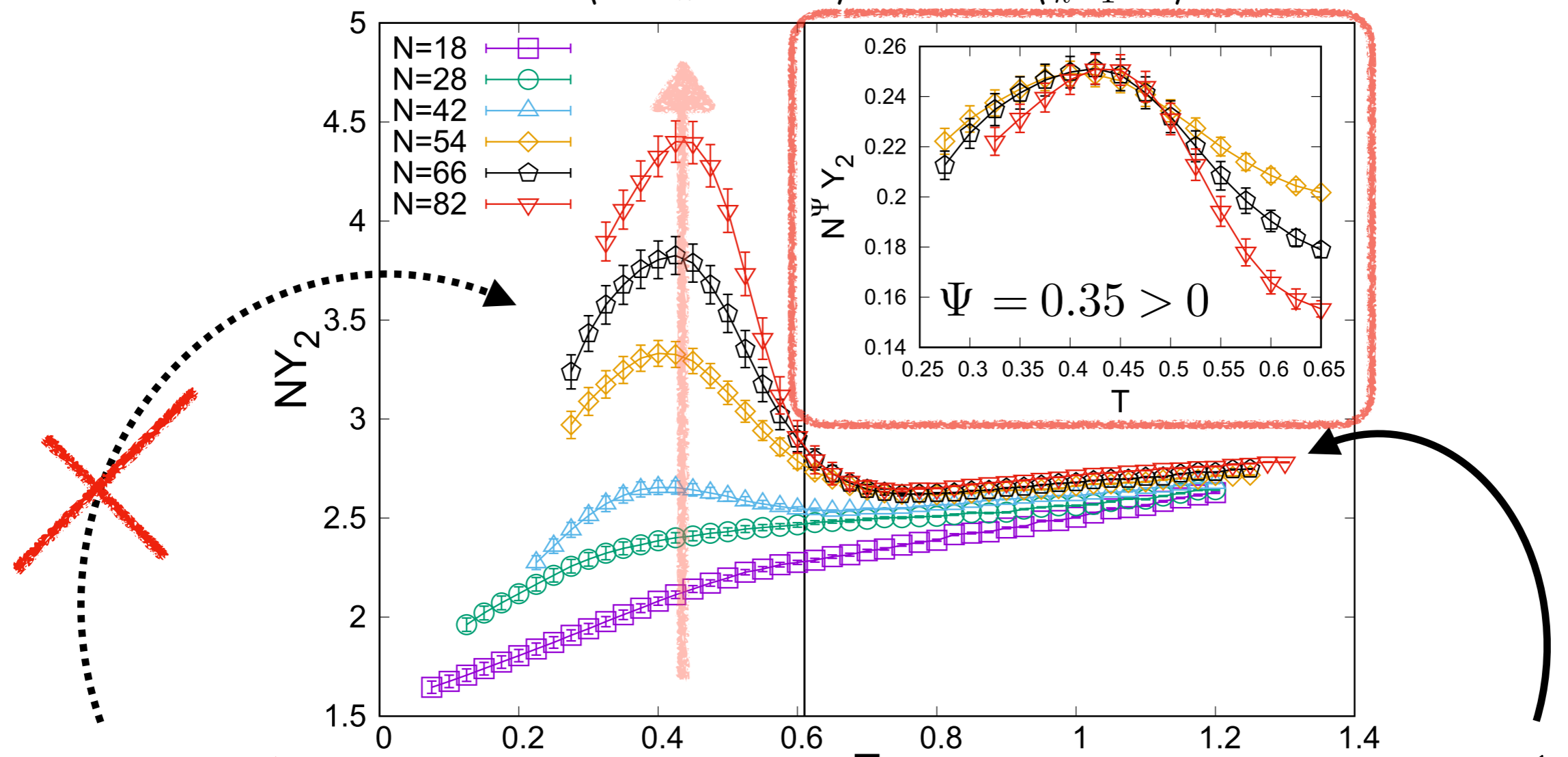
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Power pseudo-condensation

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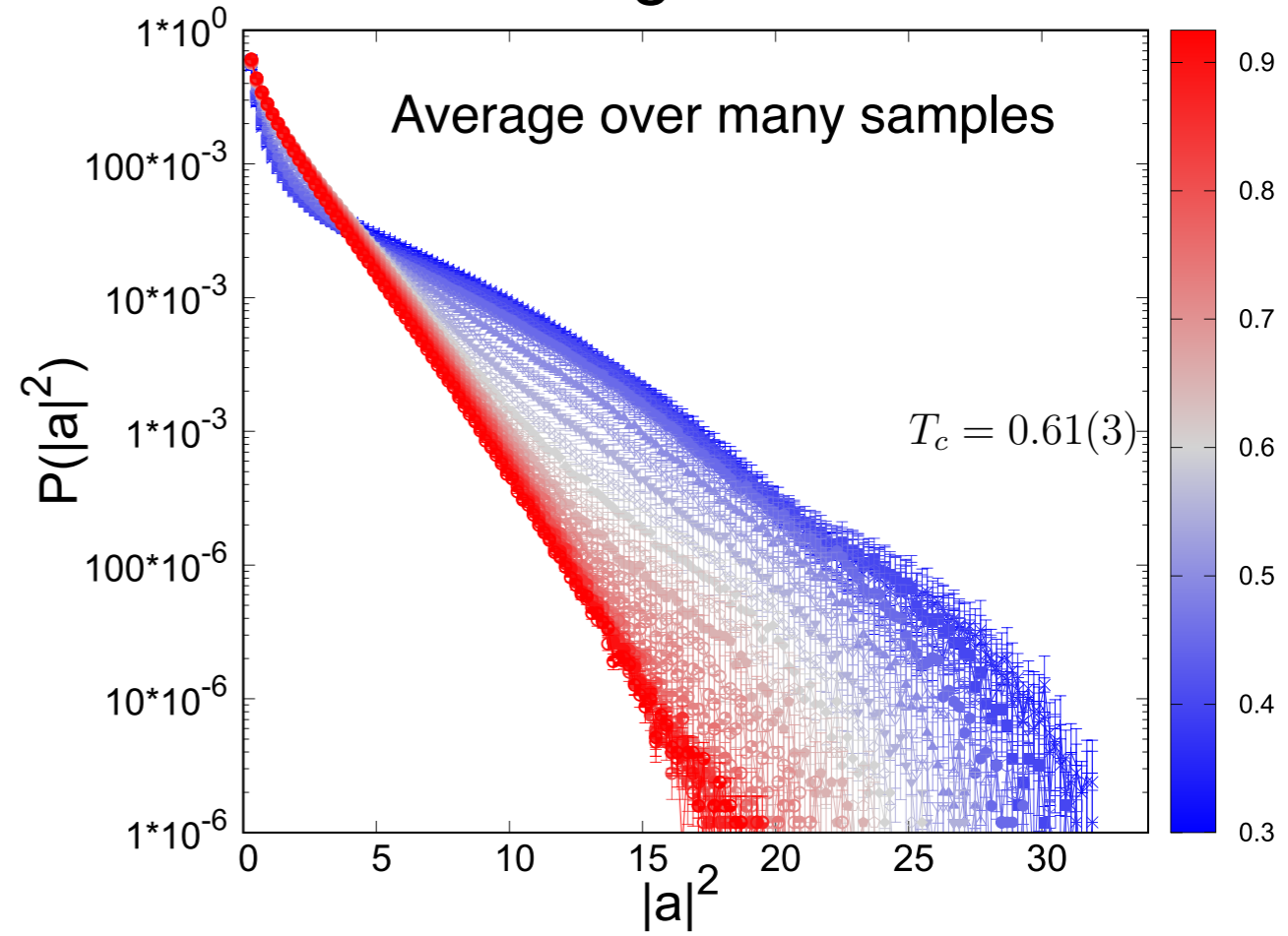
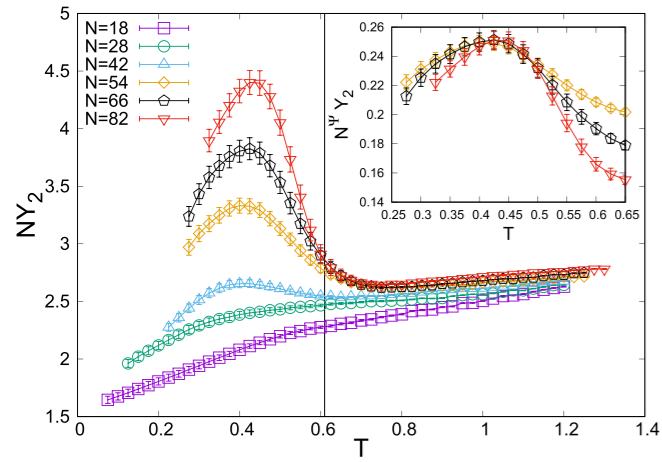
~~$\lim_{N \rightarrow \infty} Y_2 = \text{const}$~~
~~condensation~~

no condensation at low T
(but no equipartition either):
pseudo-condensation

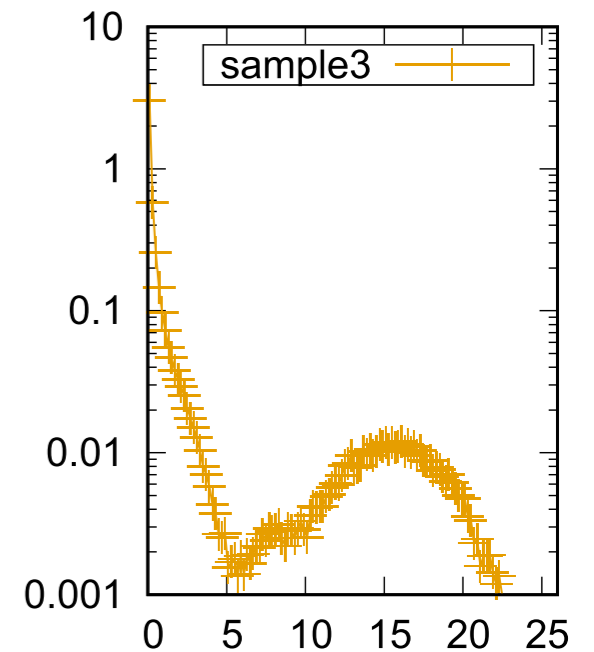
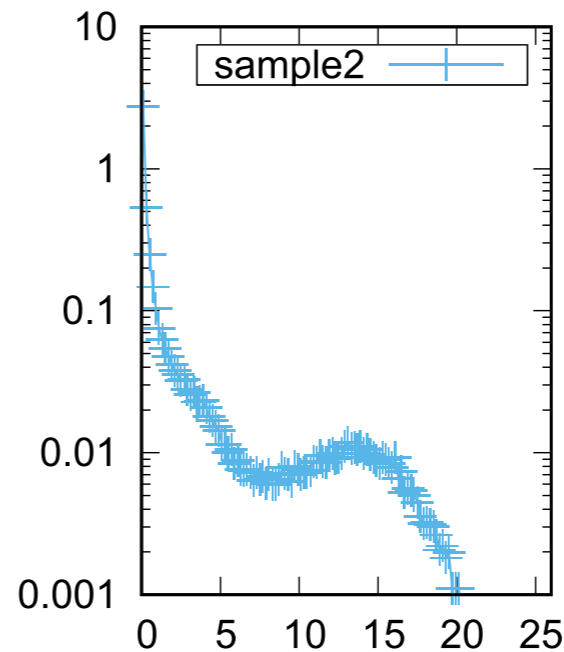
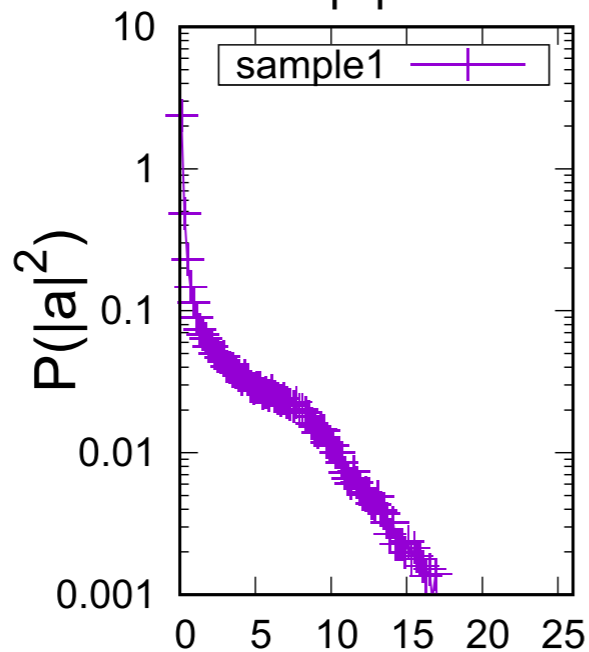
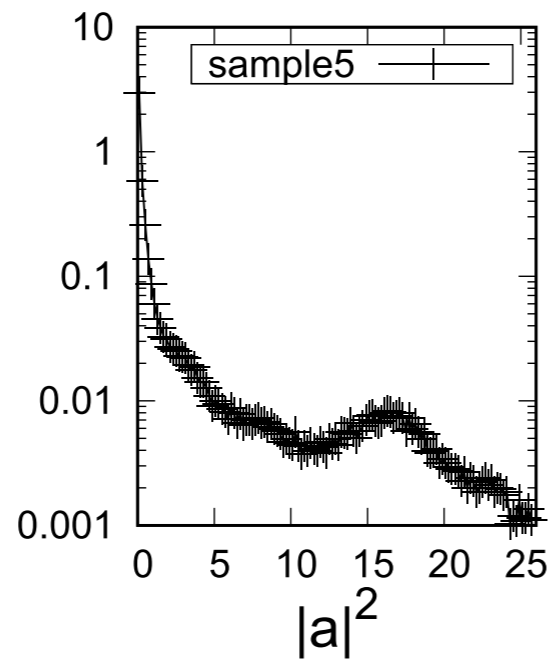
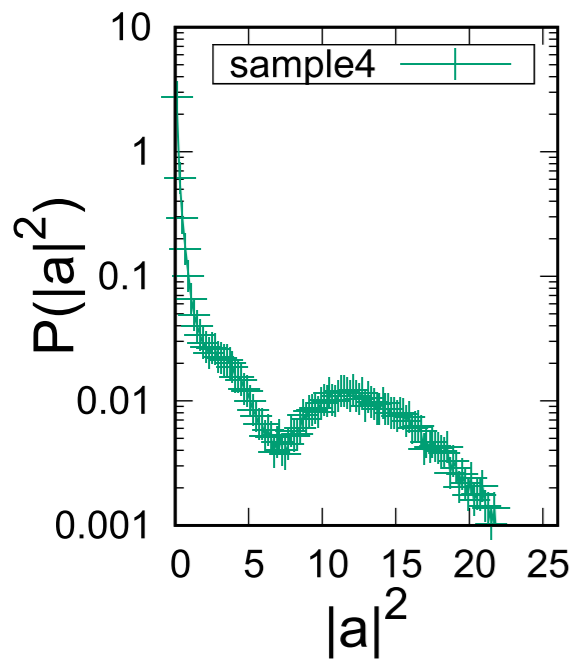
$Y_2 \sim \frac{1}{N}$
equipartition



Intensity distribution in the pseudo-condensation regime



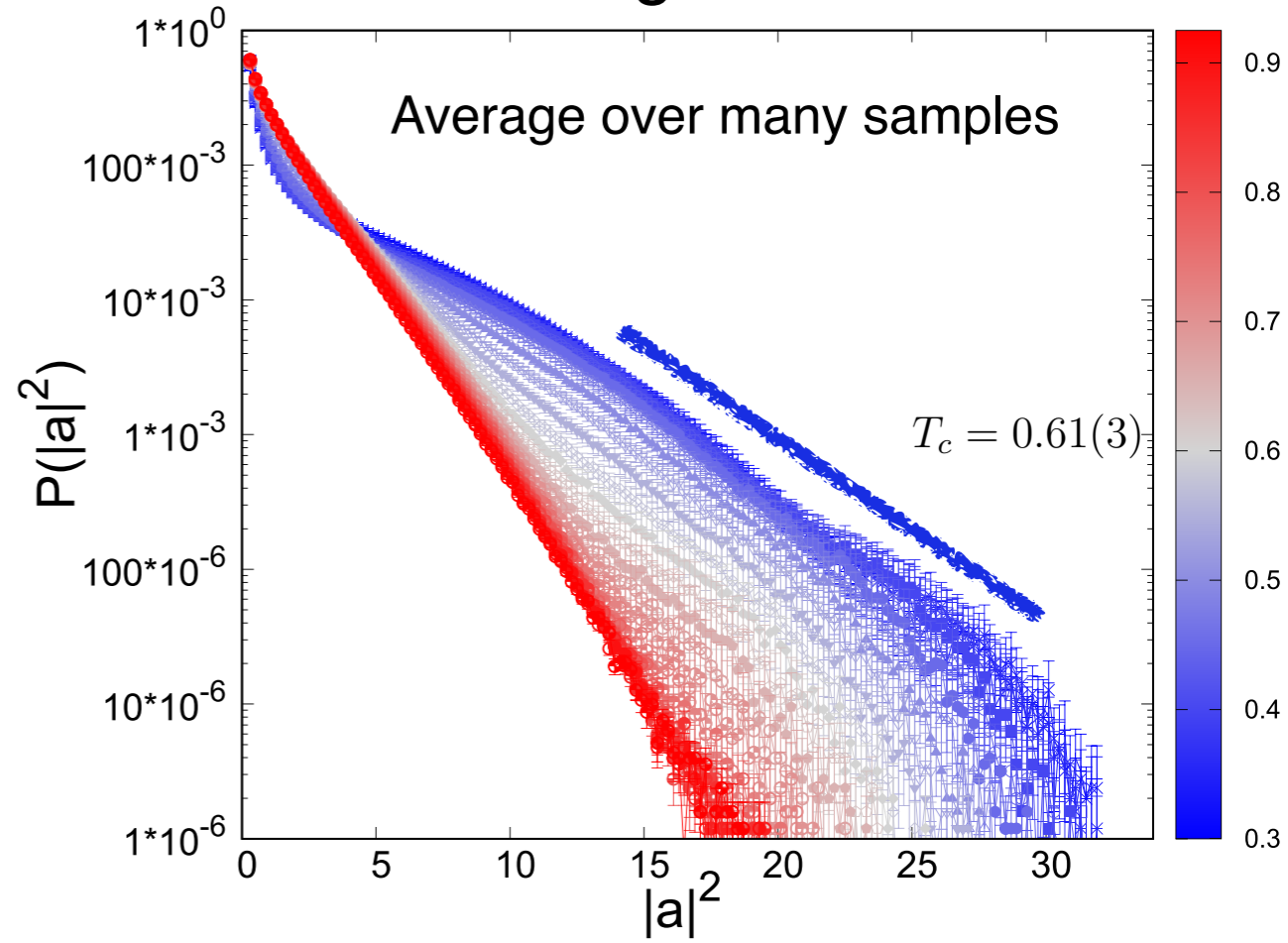
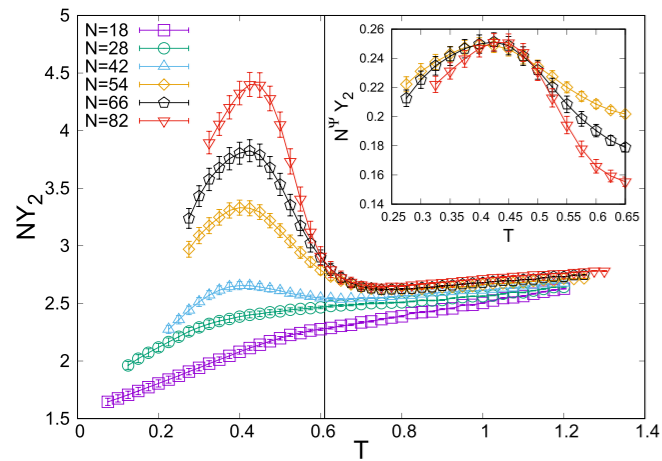
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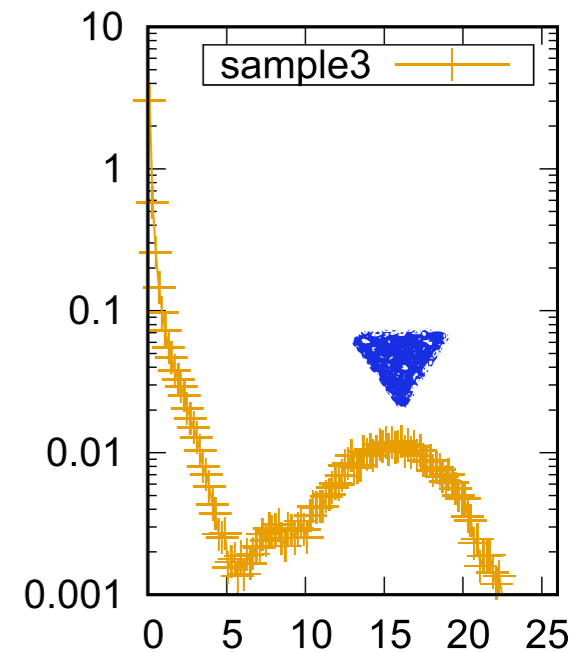
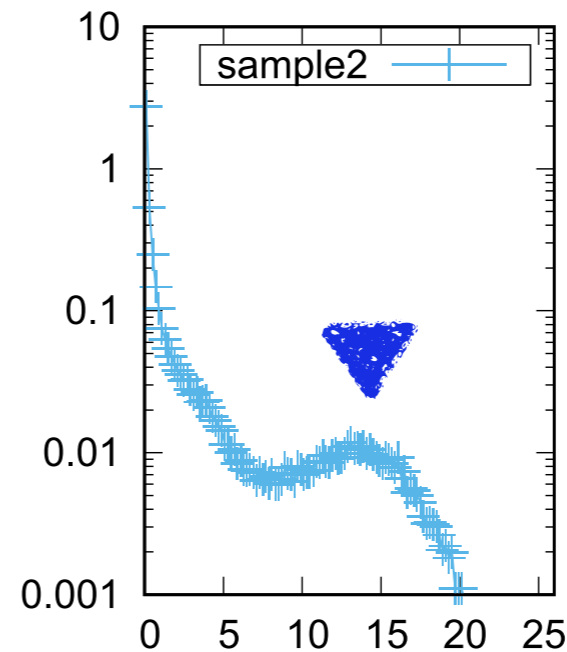
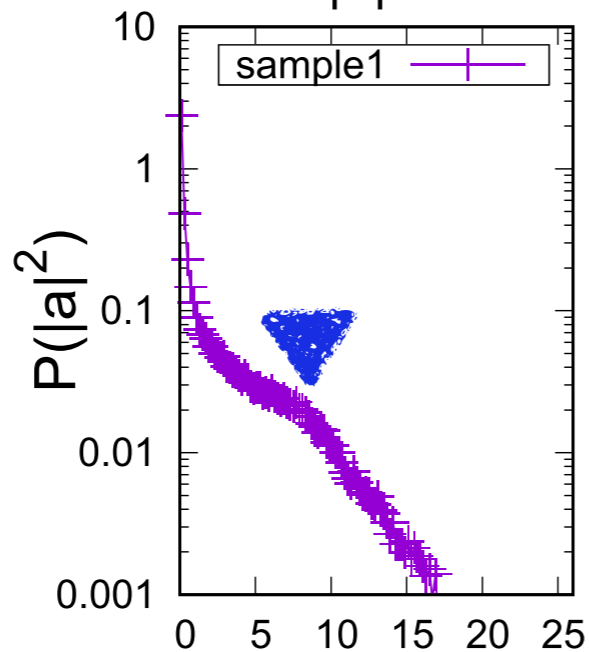
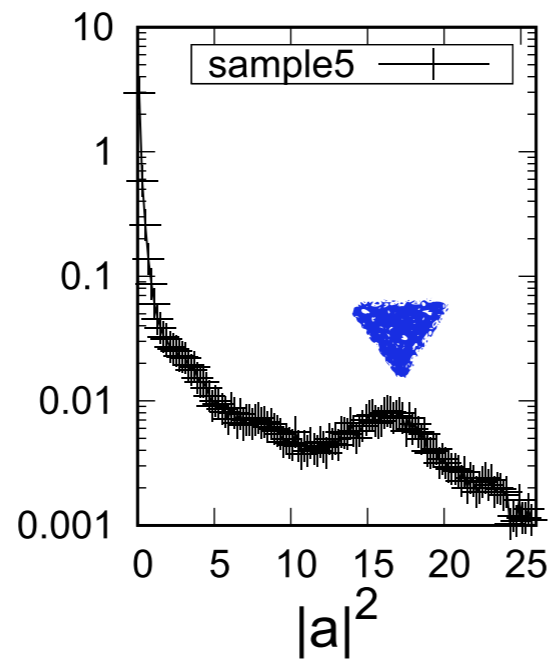
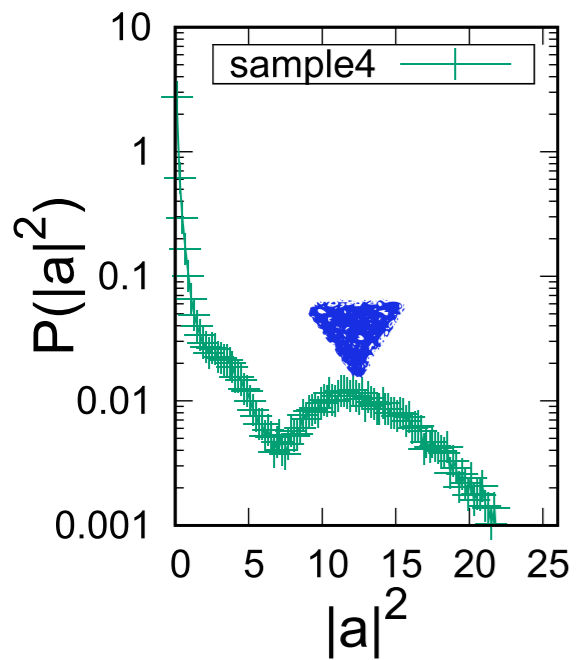
Single samples at $T=0.3$ (lowest)



Intensity distribution in the pseudo-condensation regime



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Single samples at $T=0.3$ (lowest)



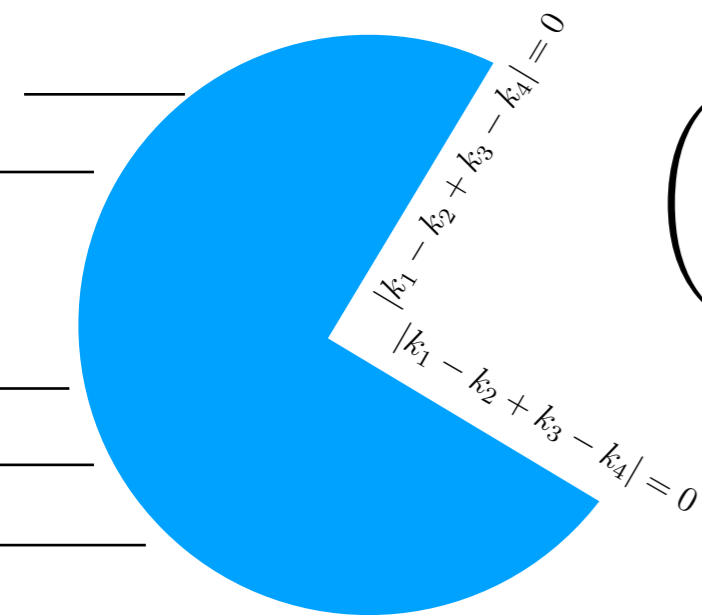
Why pseudo-condensation ?

There is a connection to connectivity

$$\mathcal{H}[\mathbf{a}] = - \sum_{\mathbf{k} | \text{FMC}(\mathbf{k})} J_{k_1 k_2 k_3 k_4} \bar{a}_{k_1} a_{k_2} \bar{a}_{k_3} a_{k_4} + \text{c.c.} \quad \mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

$$\sigma_4^2 = \overline{J_{k_1 k_2 k_3 k_4}^2} \propto \frac{1}{N^2}$$

Applying the Frequency Matching Condition the interaction graph is diluted by a factor $1/N$



$$\binom{N}{4} = \mathcal{O}(N^4)$$



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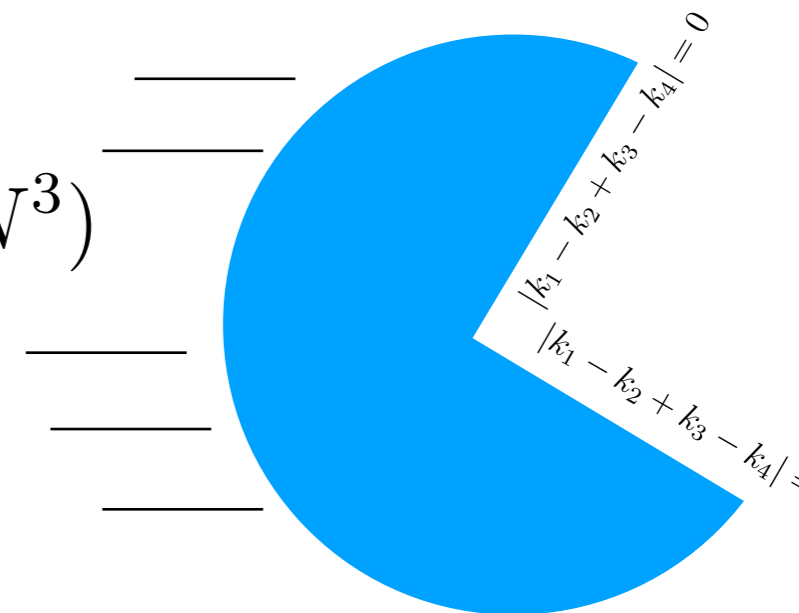
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Applying the Frequency Matching Condition the interaction graph is diluted by a factor 1/N

The scaling of the number of elements in the Hamiltonian is

$$\binom{N}{4} \times \left(\frac{2}{3N} + \frac{1}{3N^3} \right) = \mathcal{O}(N^3)$$

Complete factor graph Frequency matching condition pruning



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EXTENSIVE ENERGY

$$\min_a \mathcal{H}[\mathbf{a}] = \mathcal{O}(N)$$

UNIFORM

This holds if

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UNIFORM

$$|a_k| \simeq 1 \quad \forall k = 1, \dots, N$$



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UNIFORM

But what if condensation occurs
(on a single quadruplet)?

$\{\mathbf{a}\} : |a_k| \in \square \propto \sqrt{N} \quad , \quad |a_k| \notin \square = 0$
CONDENSED

$\mathcal{H}[\mathbf{a}] = -J_{1234} \bar{a}_1 a_2 \bar{a}_3 a_4 = \frac{1}{\mathcal{O}(N)} \mathcal{O}(N^2) = \mathcal{O}(N)$
CONDENSED



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CONDENSED

There is no
advantage
in condensation,
nor in equipartition.
They coexists:
pseudo-condensation



Pseudo-condensation is a special case

The behaviour of the 4-phasor mode-locking model is borderline

$$\mathcal{H}[\mathbf{a}] = - \sum_{\mathbf{k} | \text{FMC}(\mathbf{k})} J_{k_1 k_2 k_3 k_4} \bar{a}_{k_1} a_{k_2} \bar{a}_{k_3} a_{k_4} + \text{c.c.} \quad \mathcal{E} = \epsilon N = \sum_{n=1}^N |a_n|^2$$

In the narrow
band case

$$\sigma_4^2 = \overline{J_{k_1 k_2 k_3 k_4}^2} \propto \frac{1}{N^3}$$

$$\mathcal{H}[\mathbf{a}] = -J_{1234} \bar{a}_1 a_2 \bar{a}_3 a_4 = \frac{1}{\mathcal{O}(N^{3/2})} \mathcal{O}(N^2) = \mathcal{O}(N^{1/2}) \ll \mathcal{O}(N)$$

Starting with a dilute
 $\mathcal{O}(N^3)$ bipartite ER
graph and pruning
with FMC

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CONDENSED

Equipartition holds

Starting with a dilute
 $\mathcal{O}(N^3)$ bipartite ER
graph and pruning
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$$\sigma_4^2 = \overline{J_{k_1 k_2 k_3 k_4}^2} \propto \frac{1}{N}$$

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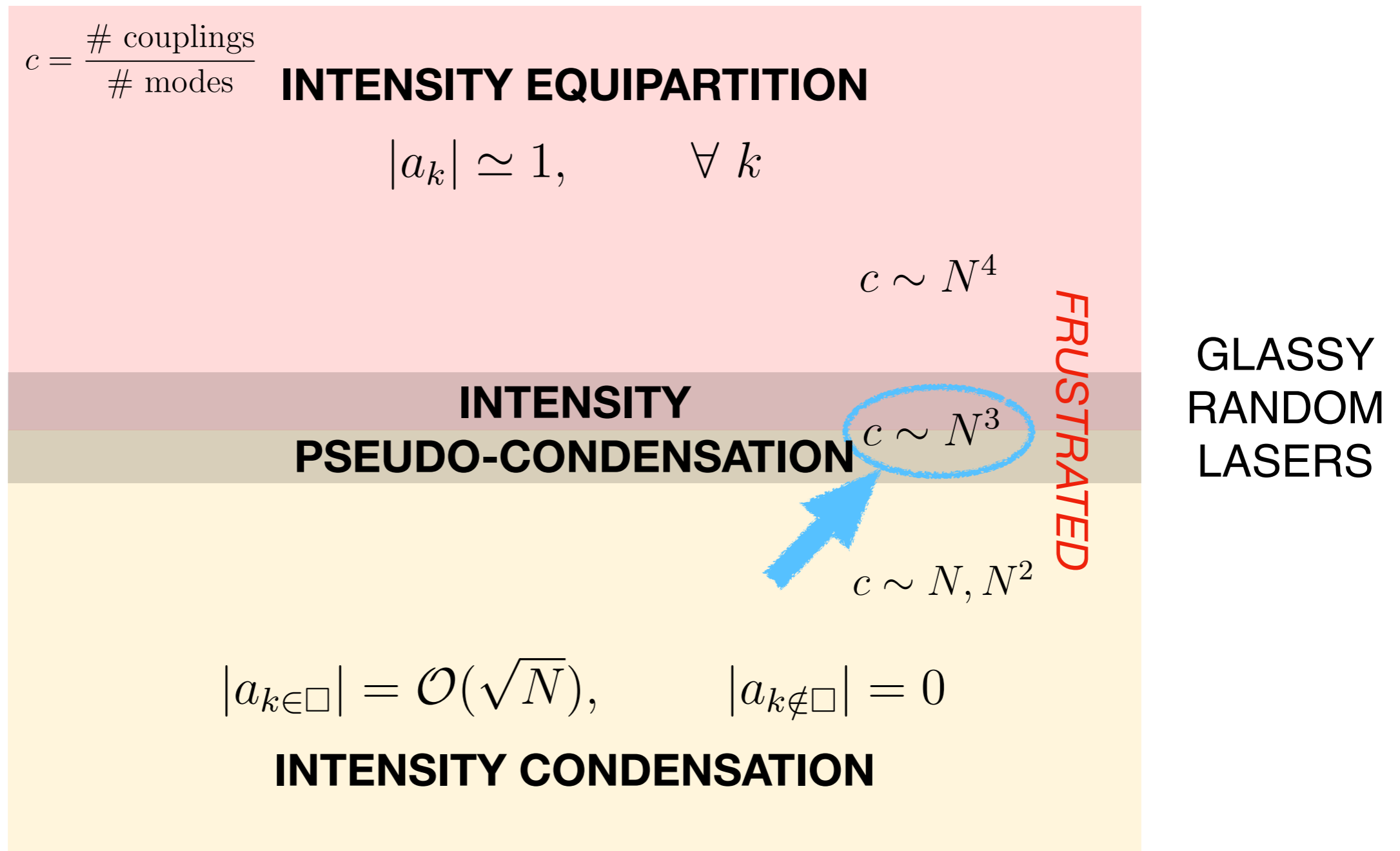
CONDENSED

Power condensation occurs

Ensemble equivalence breaks down!



Why pseudo-condensation ?

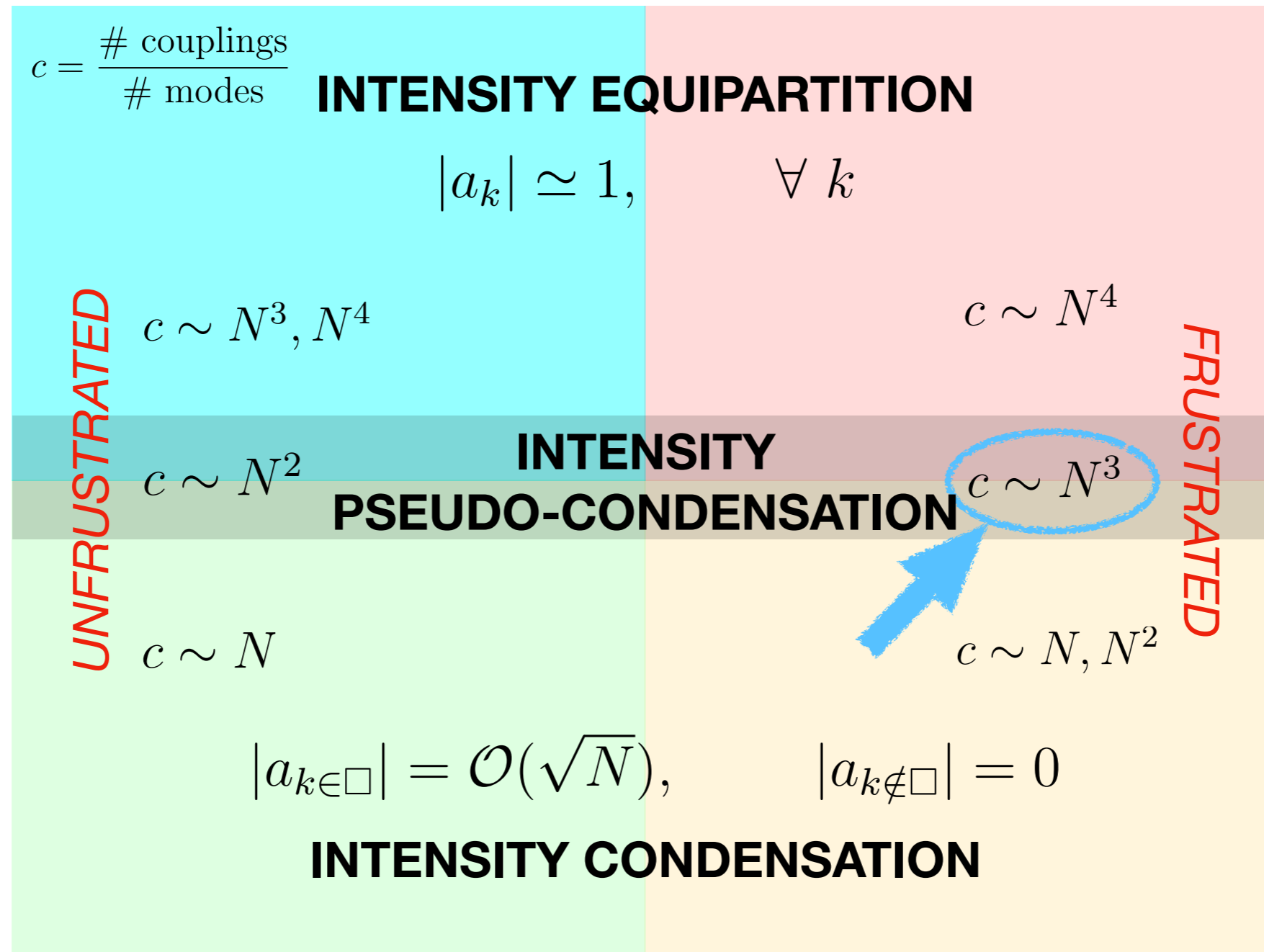


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Why pseudo-condensation ?

ORDERED OR
NON-GLASSY
RANDOM
LASERS



GLASSY
RANDOM
LASERS

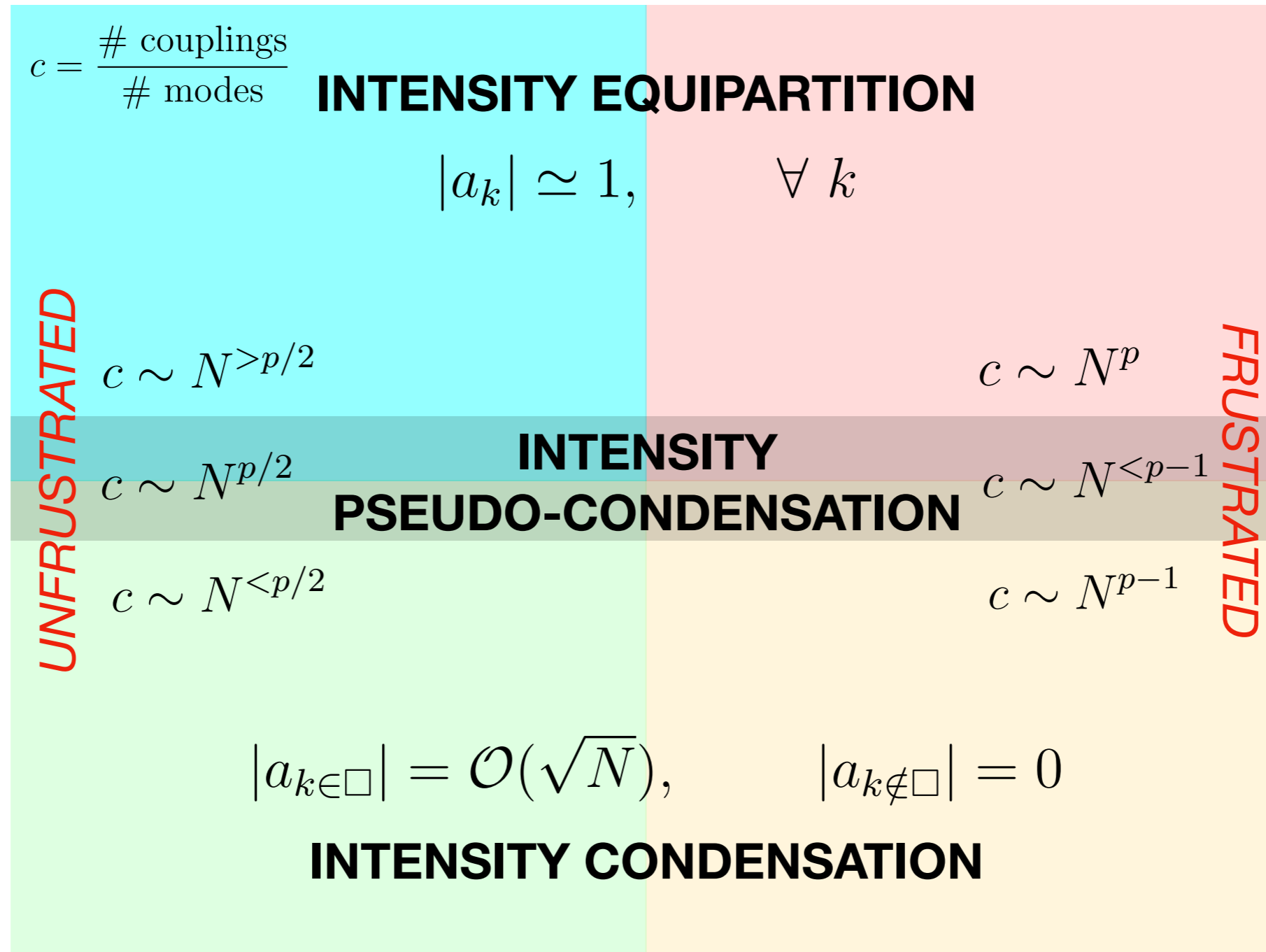
F Antenucci, M Ibanez Berganza, LL,
PRB 92, 014204 (2015), PRA 91, 043811 (2015)

J. Niedda, G. Gradenigo, LL,
JSTAT 053302 (2023)



Why pseudo-condensation ?

For any p-interacting number of modes, any spherical p-spin model



Conclusions

- Random lasers can be effectively modelled by statistical mechanics of disordered systems (mode-locking, spectral properties, lasing threshold,...)
- **Some random lasers display glassy (multi-equilibria) features and allow for directly measuring the RSB order parameter distribution**
- Monte Carlo dynamics of the leading model, the spherical 4-phasor mode-locked model, yield evidence for
 - Mean-field universality class for the lasing critical point
 - RSB at high pumping/low temperature
 - and pseudo-condensation of the overall intensity (connection with connectivity of the interaction network)



- In progress:

- * Monte Carlo simulations at and off-equilibrium with continuous band of frequencies, real material gain profiles, losses (M Benedetti, G Trinca-Cintioli, J Niedda)
- * Interpolation between analytic and experimental IFO, out of equilibrium effects (G Trinca Cintioli, J Niedda)
- * Analytic theory for spin-glasses on mode-locked graphs — merit factor revisited (J Niedda, G Parisi)
- * Mode-locked random lasers on sparse graphs avoiding condensation (M Benedetti)
- * Measurements campaign on different random laser compounds (solids, viscous liquids, organic, inorganic, 2D, 3D, varying mode extensions, ..) probed by different pumping lasers (ns, ps, fs pulses) to tune disorder, understand the relationship between mode extension and coupling network connectivity, deepen the reliability of photonics measure of the Parisi $P(q)$ (CNR-NANOTEC, D Sanvitto, L De Marco, I Viola, M De Giorgi, ...)



Post-doc positions opening soon on the MUR-funded project
Statics and Dynamics of Spin Glass and Disordered Systems:
Theory and Numerics to Understand Experiments.



Random lasers as complex disordered systems

Luca Leuzzi

Lucabib

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FUNDING AGENCIES



Eigenmode basis in open cavity

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

OPEN CAVITY:

- (i) Mirror cavities with leakages: there will be also radiative modes, whose frequencies take values in a continuous dominion. Different modes can have the same frequency.
- (ii) Mirror-less lasers in random media, with inhomogeneous optical susceptibility profiles. Discrete lasing frequencies will not be all equispaced and may overlap. Furthermore, the “optical cycle” and the “roundtrip time” are not defined. Their random analogues depend on the scatterers structure.

Non-diagonal linear contribution to the Hamiltonian.

What is a complete basis in an open system?

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

Strong Interactions in Multimode Random Lasers

Türeci, Ge, Rotter, Douglas Stone 2008

Feschbach projection onto radiative and localized mode subspaces

Hackenbroich, Viviescas, Haken 2003



Random graph ML laser

$$\mathcal{H} = - \sum_{n=1}^N g_n^{(0)} |a_n|^2 - J \sum_{\omega_{n_1} - \omega_{n_2} + \omega_{n_3} - \omega_{n_4} = 0} a_{n_1} a_{n_2}^* a_{n_3} a_{n_4}^* + \text{c.c.} \quad \rightarrow \quad \mathcal{H} = - \sum_{jklm}^{\text{ML}} J_{jklm} \cos(\phi_j - \phi_k + \phi_l - \phi_m)$$

quenched amplitude or amplitude equipartition $|a| \sim 1$: 4-XY phase model

A. Marruzzo and LL, PRB 2015

$$\text{ML: } |\omega_j - \omega_k + \omega_l - \omega_m| \leq \gamma$$



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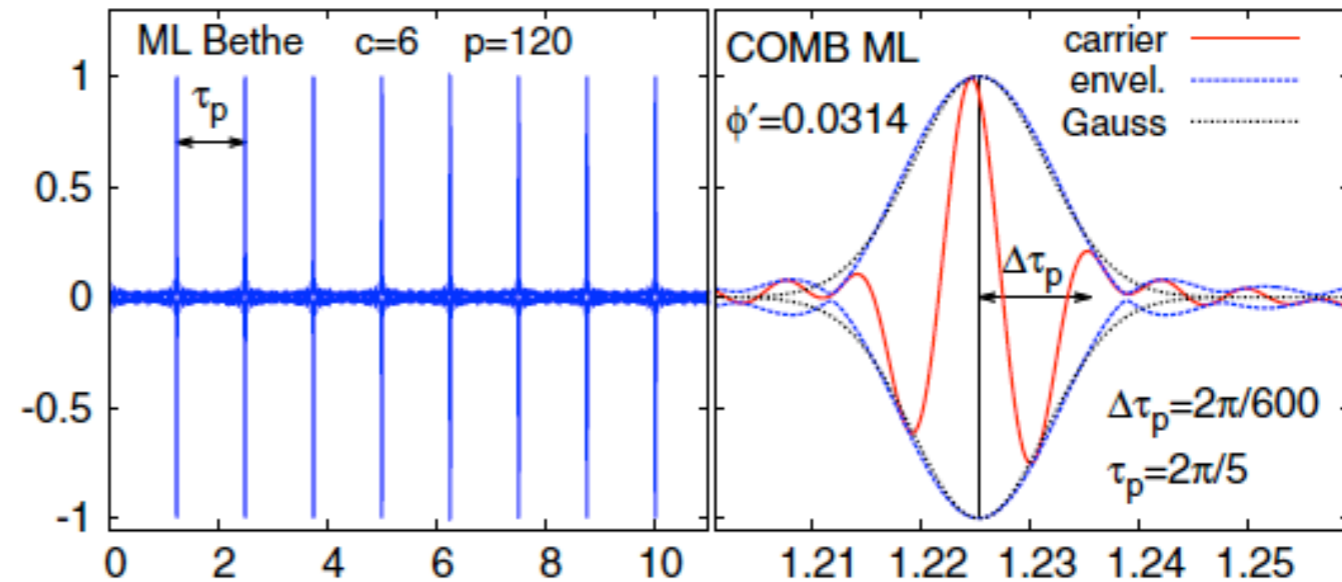
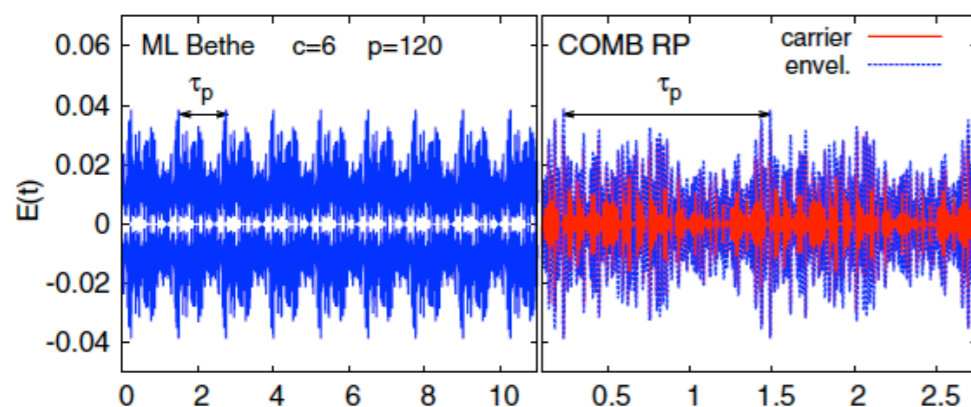
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High power mode-locking

$$\phi(\omega) \simeq \phi(\omega_0) + \phi' \times (\omega - \omega_0)$$



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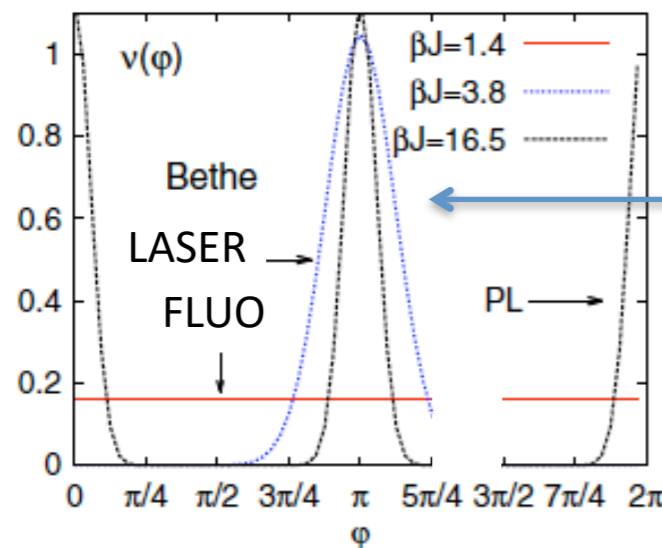


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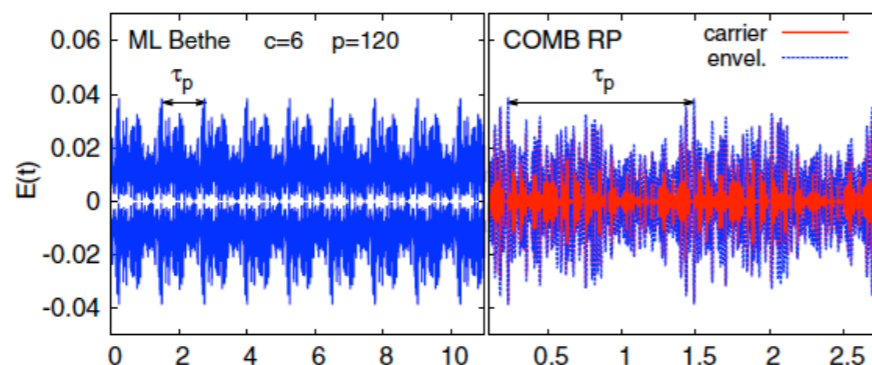
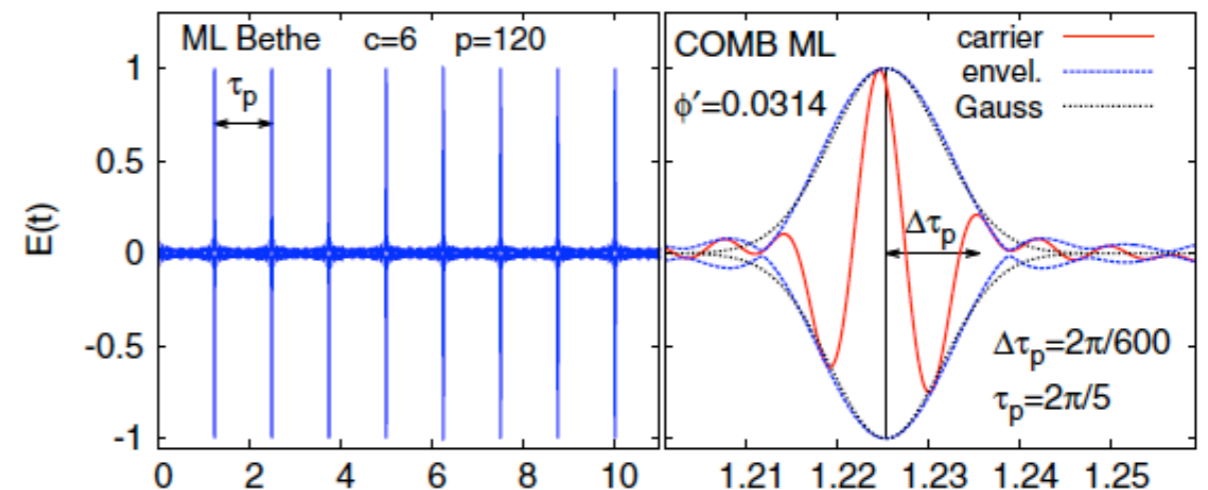


The optical regime is characterized by the phase *marginal*: the distribution of the phase values $v(\phi)$

$$\phi(\omega) \simeq \phi(\omega_0) + \phi' \times (\omega - \omega_0)$$



High power mode-locking

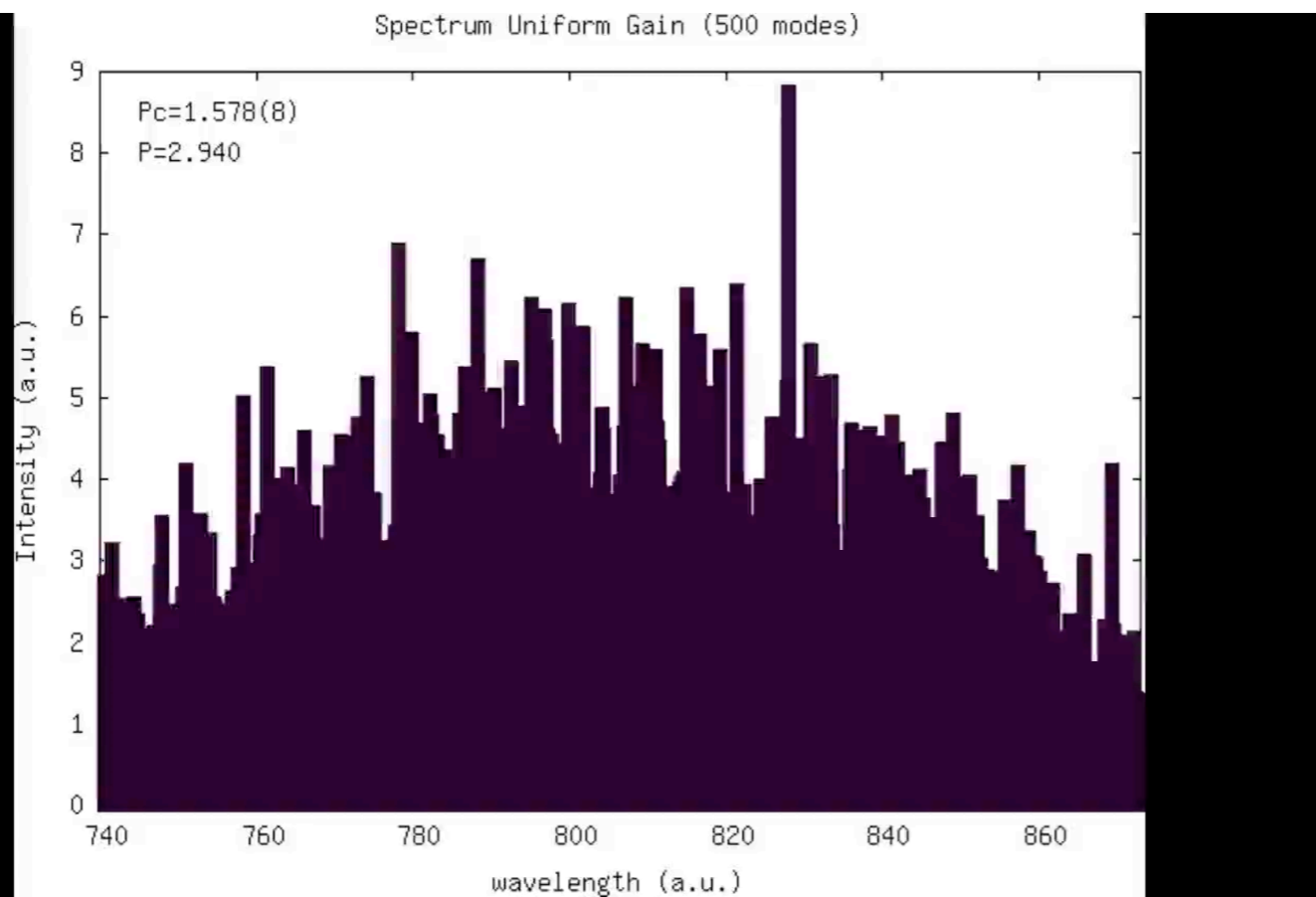


Loopy random graph ML lasers

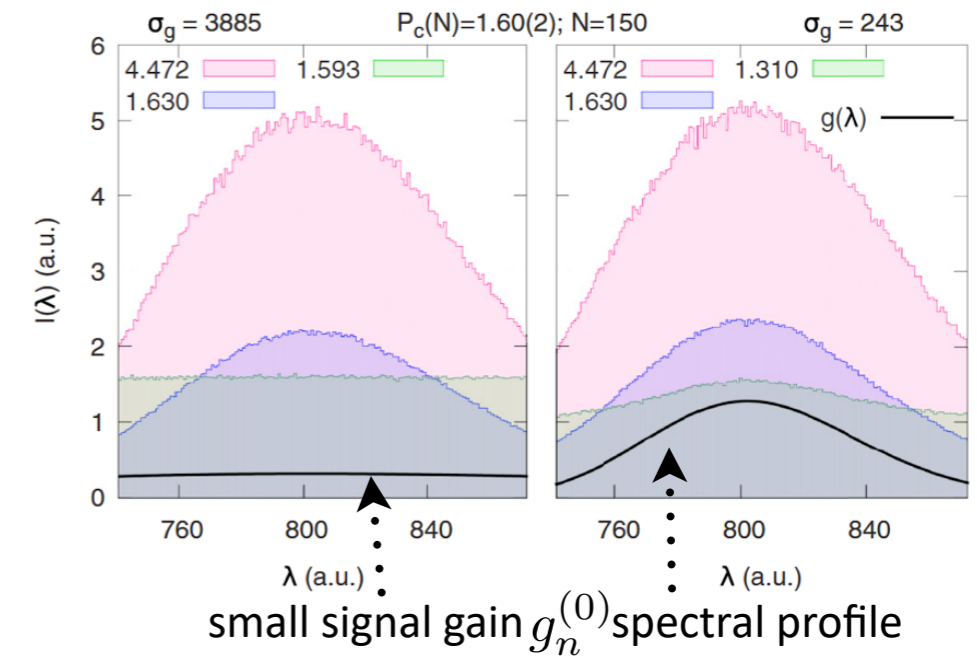
Beyond mean-field approximations we study mode-locking systems with non-trivial gain profile and interaction networks: Monte Carlo simulations.

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Single pulse spectra



Integrated spectra



F. Antenucci, M. Ibañez Berganza,
LL, PRA 2015, PRB 2015

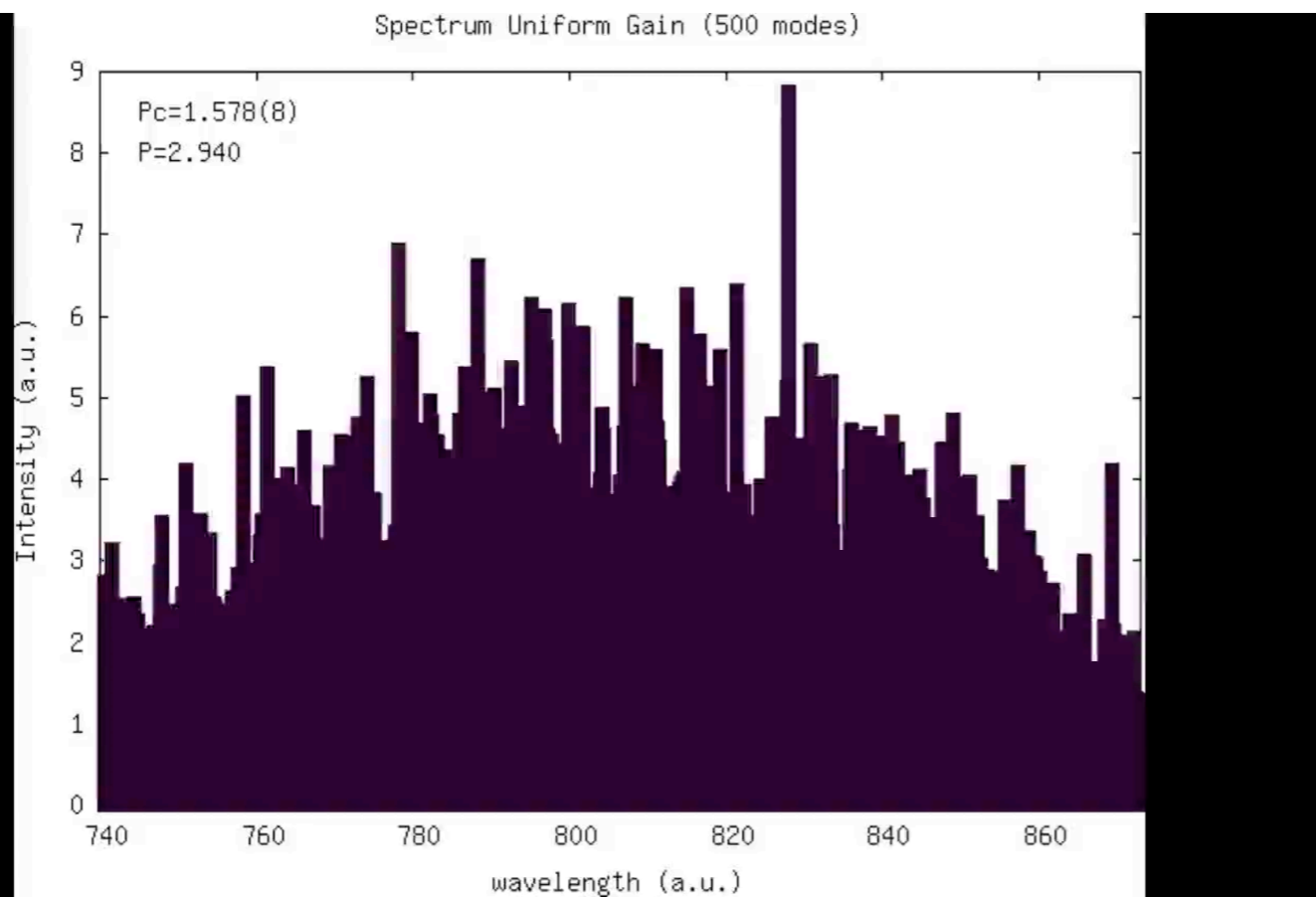


Loopy random graph ML lasers

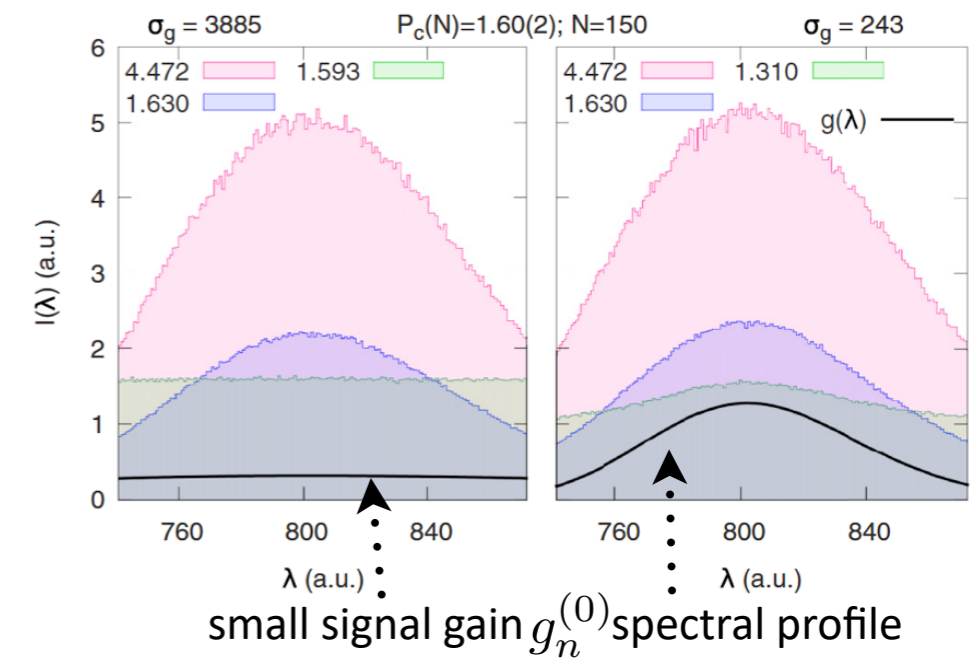
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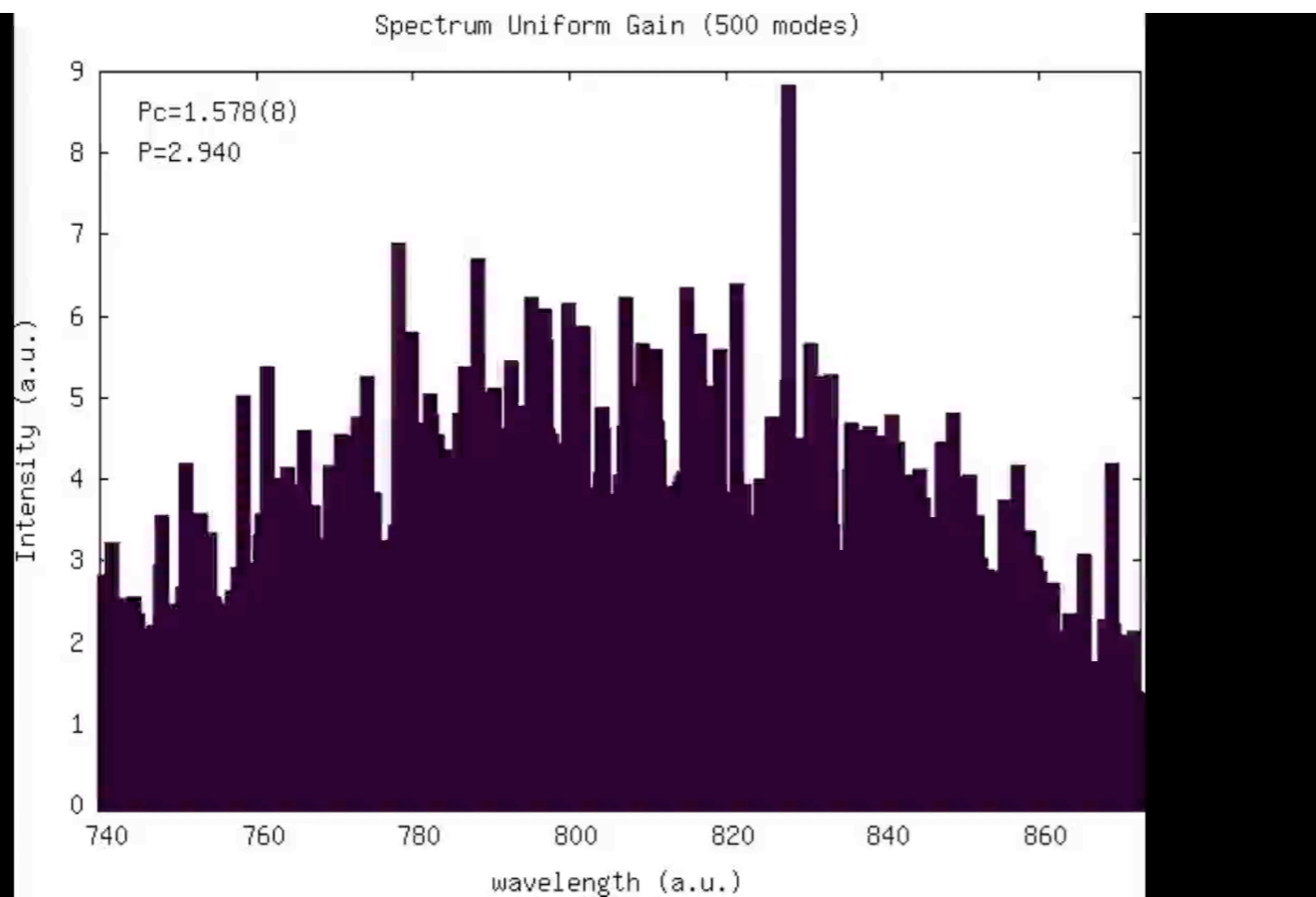


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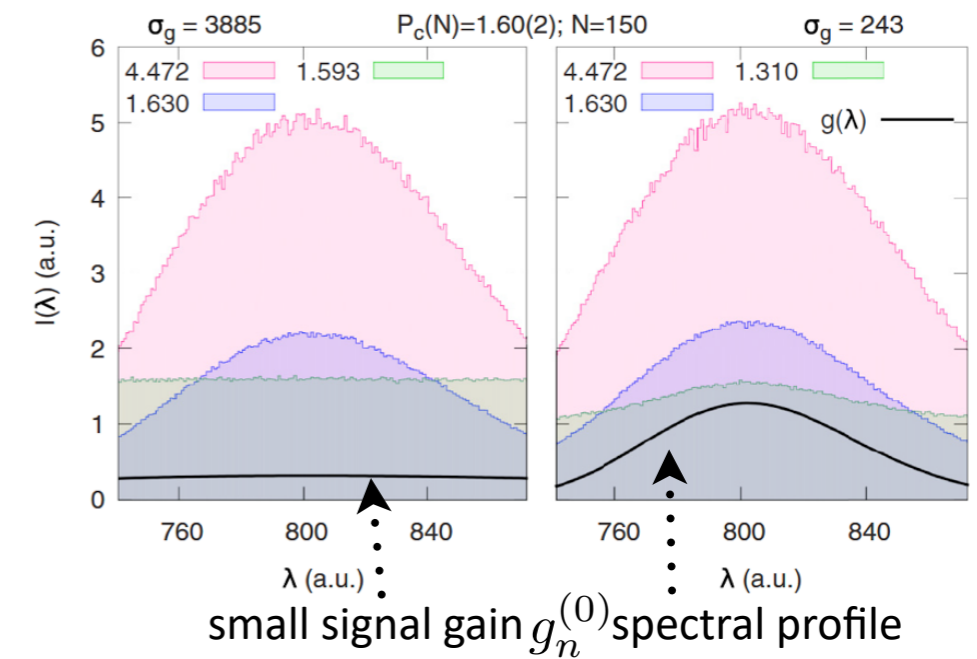
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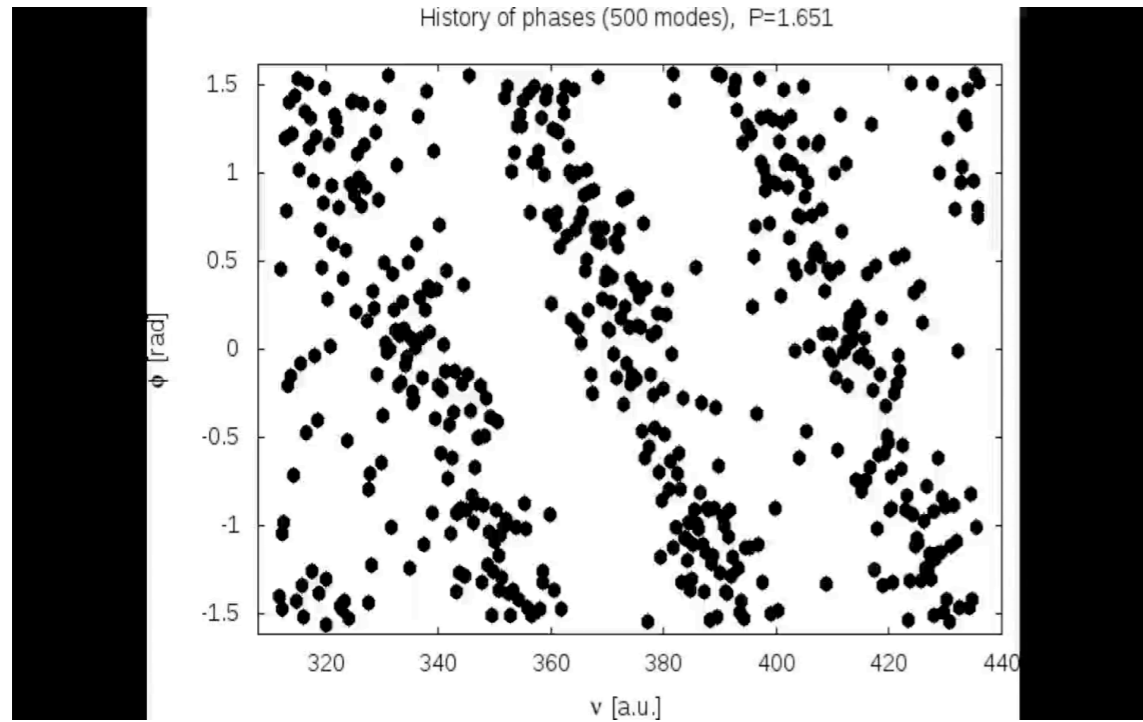


F. Antenucci, M. Ibañez Berganza,
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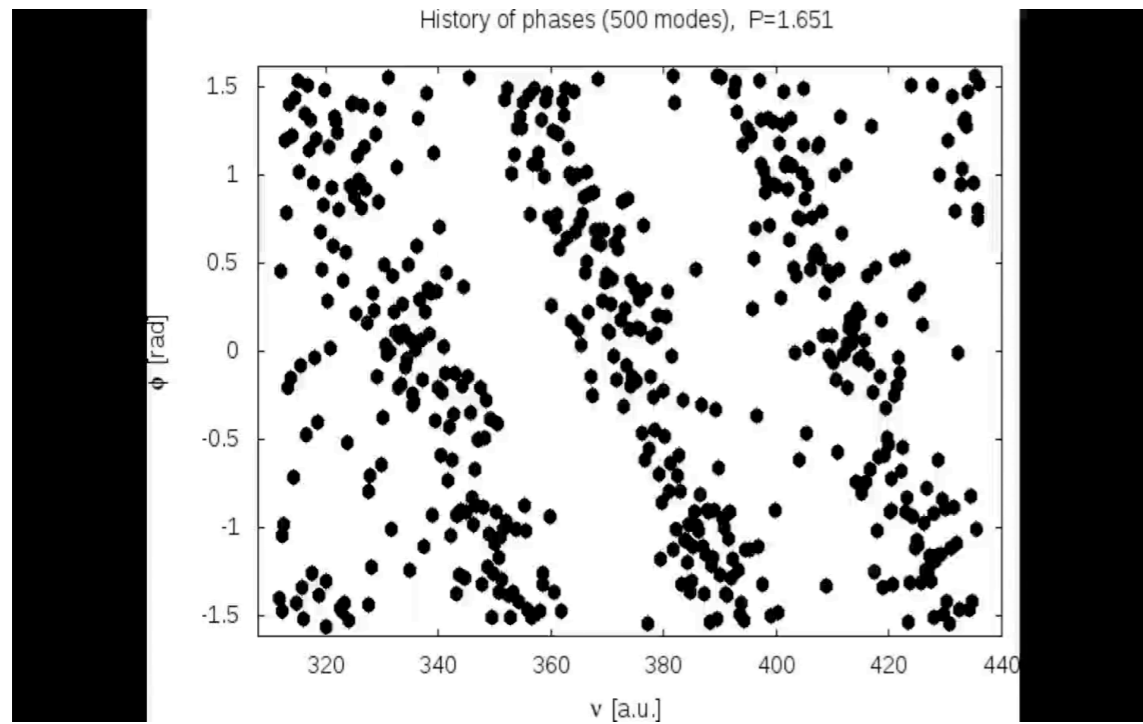
F. Antenucci, M. Ibañez Berganza, LL, PRA 2015, PRB 2015

Mode locking

$$\phi(\omega) \simeq \phi(\omega_0) + \phi' \times (\omega - \omega_0)$$

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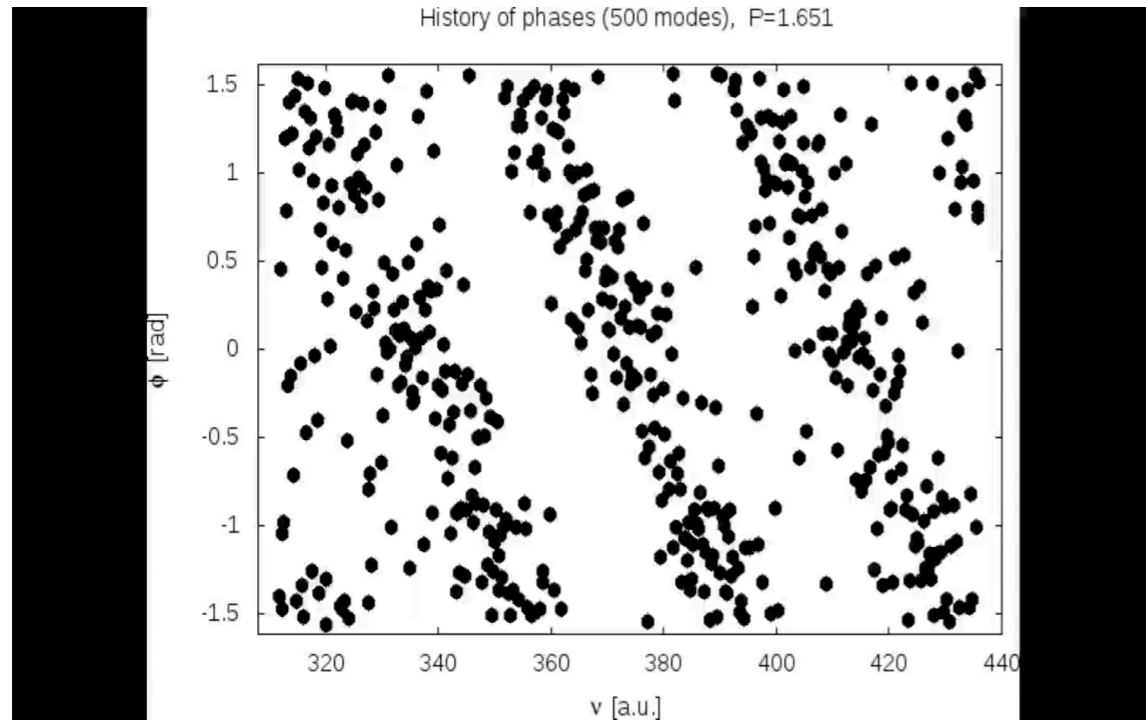
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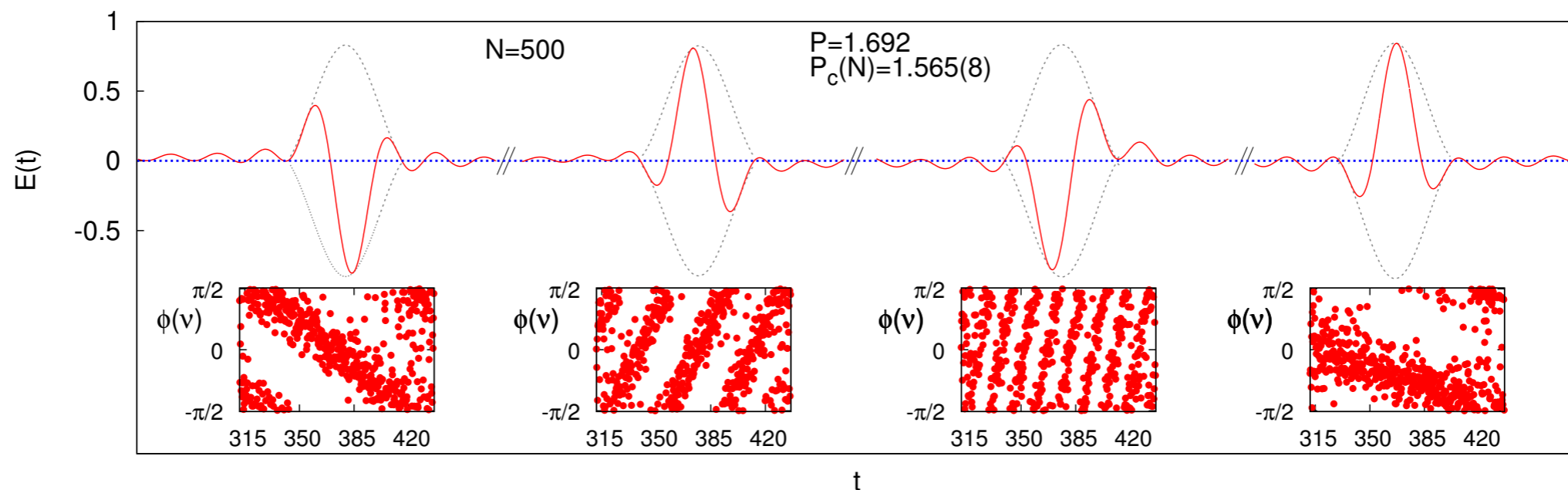


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F. Antenucci, M. Ibañez Berganza, LL, PRA 2015, PRB 2015

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Demonstration of Self-Starting Nonlinear Mode Locking in Random Lasers

Fabrizio Antenucci^{1,4}, Giovanni Lerario,² Blanca Silva Fernández,² Luisa De Marco,² Milena De Giorgi,²
Dario Ballarini,² Daniele Sanvitto,^{2,*} and Luca Leuzzi^{1,3,†}

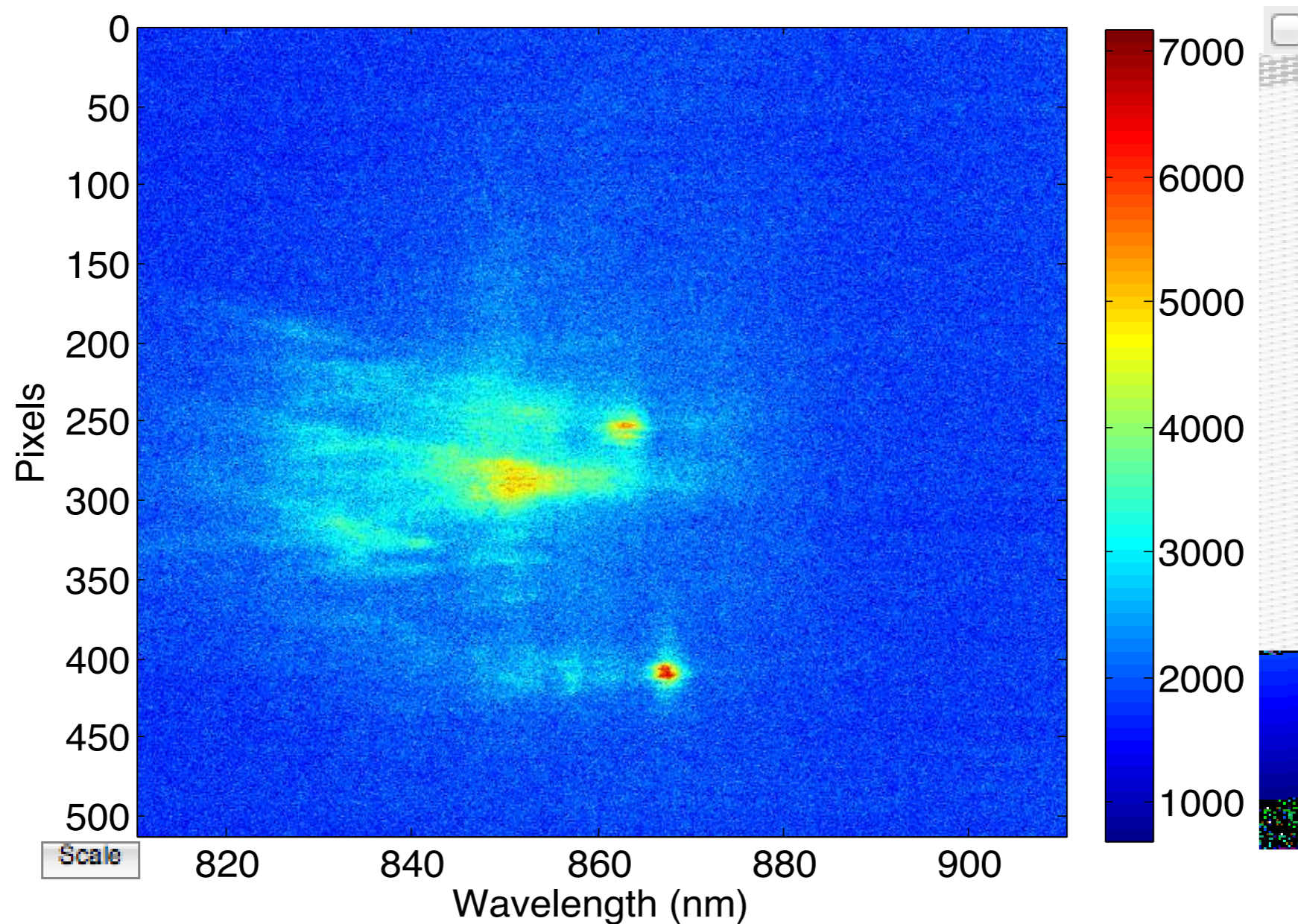
¹CNR-NANOTEC, Institute of Nanotechnology, Soft and Living Matter Laboratory, Piazzale Aldo Moro 5, I-00185 Rome, Italy

²CNR-NANOTEC, Institute of Nanotechnology, Via Monteroni, I-73100 Lecce, Italy

³Dipartimento di Fisica, Università Sapienza, Piazzale Aldo Moro 5, I-00185 Rome, Italy

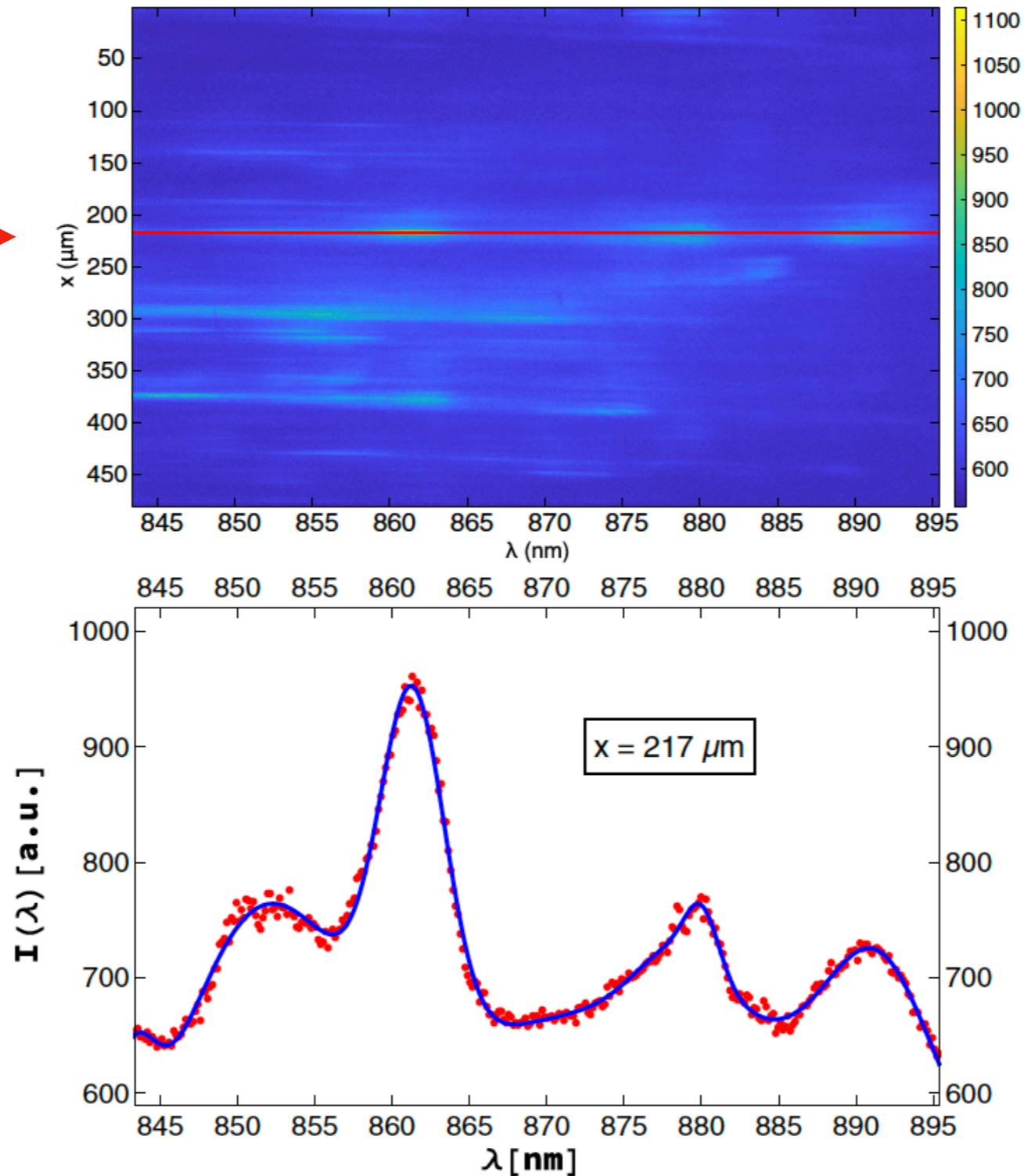
⁴Saddle Point Science Ltd, 71 OAKS Avenue, Worcester Park KT4 8XE, United Kingdom

The key point is to have contemporarily the spectrum and the position of the resonance



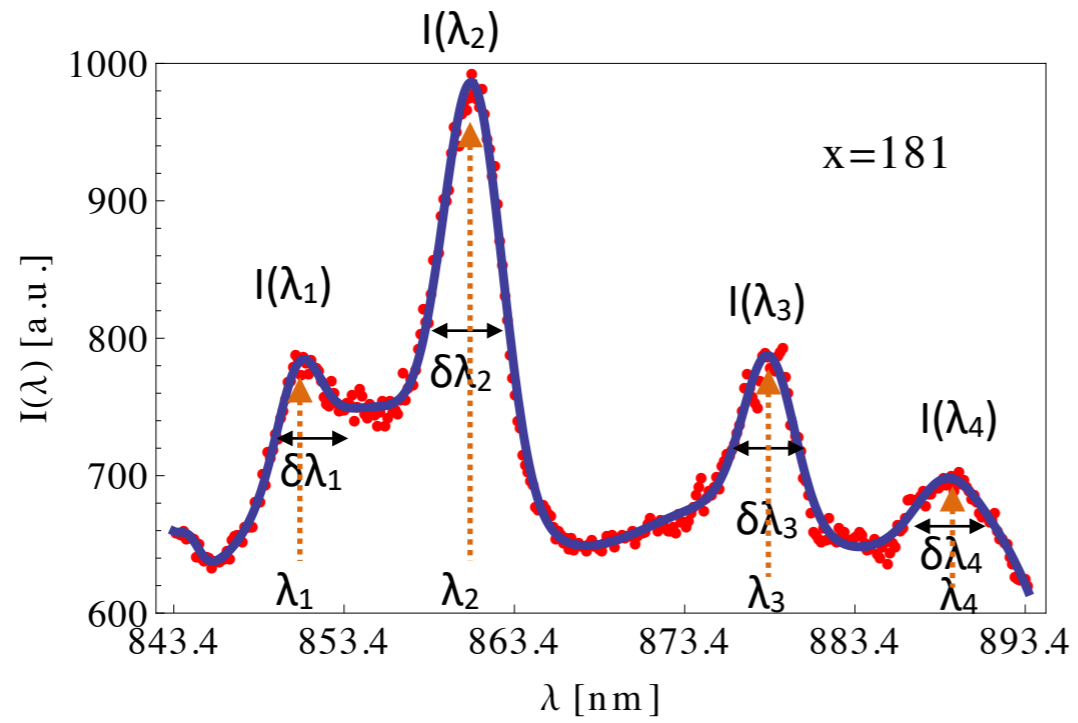
Self-starting mode-locking in random lasers

Modes identification



Multiple Gaussian
interpolation of
thousands of spectra
+
Akaike inference
criterion to limit
overfitting





Self-starting mode-locking in random lasers

Modes 4-point correlation

$$C_4(\omega_j, \omega_k, \omega_l, \omega_m) = \langle I_j I_k I_l I_m \rangle_c = C_4^{(0)} - C_4^{(1)} + 2C_4^{(2)} - 6C_4^{(3)}$$

$$C_4^{(0)} = \langle I_j I_k I_l I_m \rangle$$

$$C_4^{(1)} = \langle I_j I_k I_l \rangle \langle I_m \rangle + \langle I_j I_k I_m \rangle \langle I_l \rangle + \langle I_j I_m I_l \rangle \langle I_k \rangle + \langle I_m I_k I_l \rangle \langle I_j \rangle \\ + \langle I_j I_k \rangle \langle I_l I_m \rangle + \langle I_j I_l \rangle \langle I_k I_m \rangle + \langle I_j I_m \rangle \langle I_k I_l \rangle$$

$$C_4^{(2)} = \langle I_j I_k \rangle \langle I_l \rangle \langle I_m \rangle + \langle I_j I_l \rangle \langle I_k \rangle \langle I_m \rangle + \langle I_j I_m \rangle \langle I_k \rangle \langle I_l \rangle \\ + \langle I_k I_l \rangle \langle I_j \rangle \langle I_m \rangle + \langle I_k I_m \rangle \langle I_j \rangle \langle I_l \rangle + \langle I_l I_m \rangle \langle I_j \rangle \langle I_k \rangle$$

$$C_4^{(3)} = -6 \langle I_j \rangle \langle I_k \rangle \langle I_l \rangle \langle I_m \rangle$$

$$c_4(\omega_j, \omega_k, \omega_l, \omega_m) \equiv \frac{C_4(\omega_j, \omega_k, \omega_l, \omega_m)}{\sigma_j(\omega_j) \sigma_k(\omega_k) \sigma_l(\omega_l) \sigma_m(\omega_m)}$$

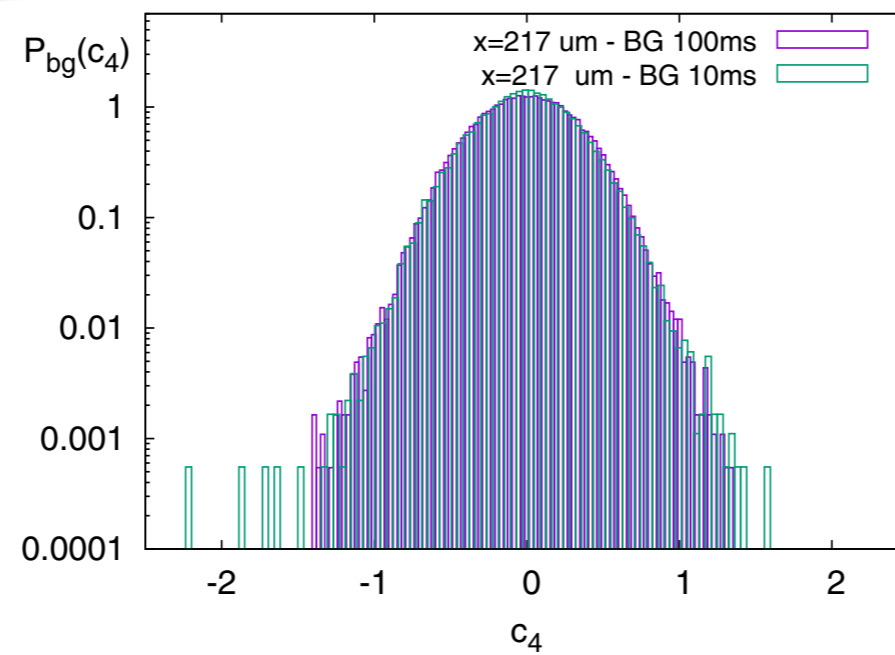
$$\sigma_j(\omega_j) = \sqrt{\langle (I_j - \langle I_j \rangle)^2 \rangle}$$



Self-starting mode-locking in random lasers

Self-starting
mode-locking in
random lasers

4-point correlation
distributions

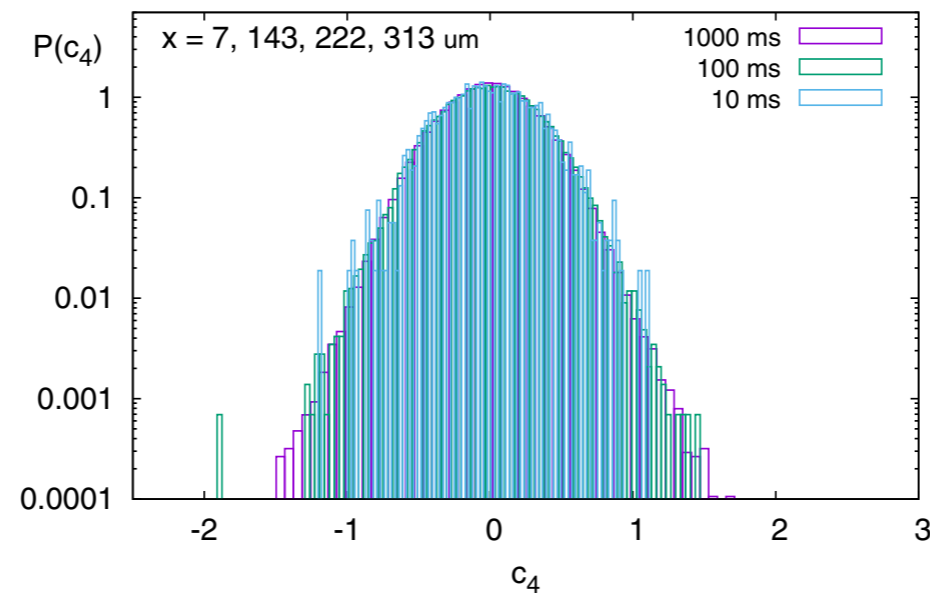


4-point correlations
among resonances in
different spectra



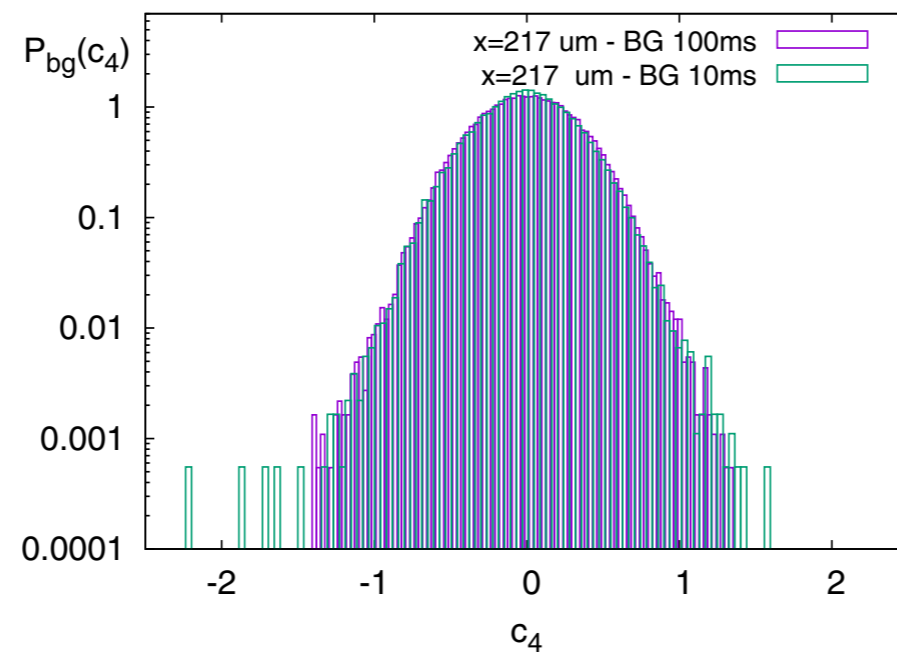
Self-starting mode-locking in random lasers

Self-starting mode-locking in random lasers



4-point correlations among resonances in the same spectrum at far away distances

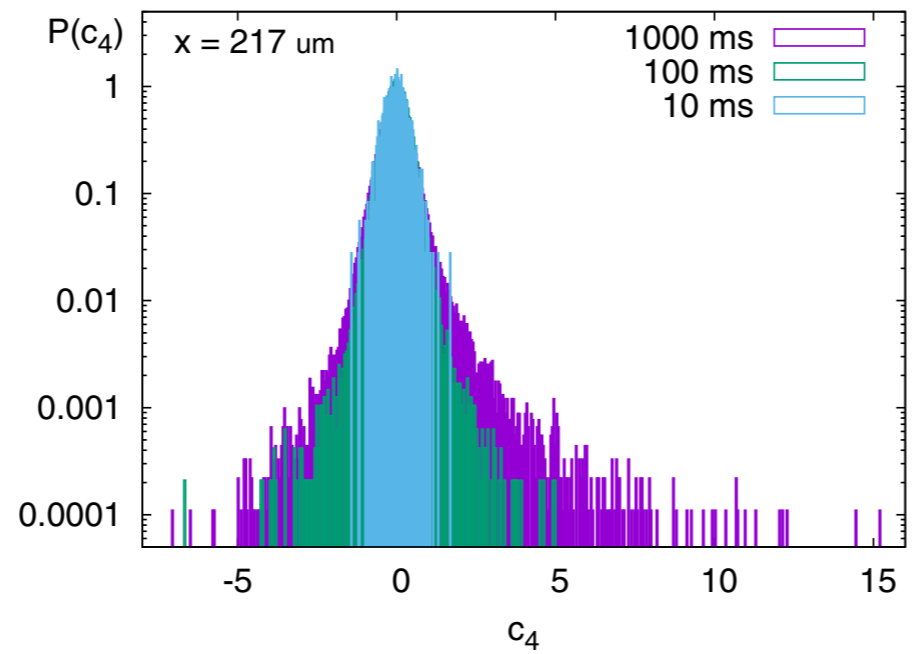
4-point correlation distributions



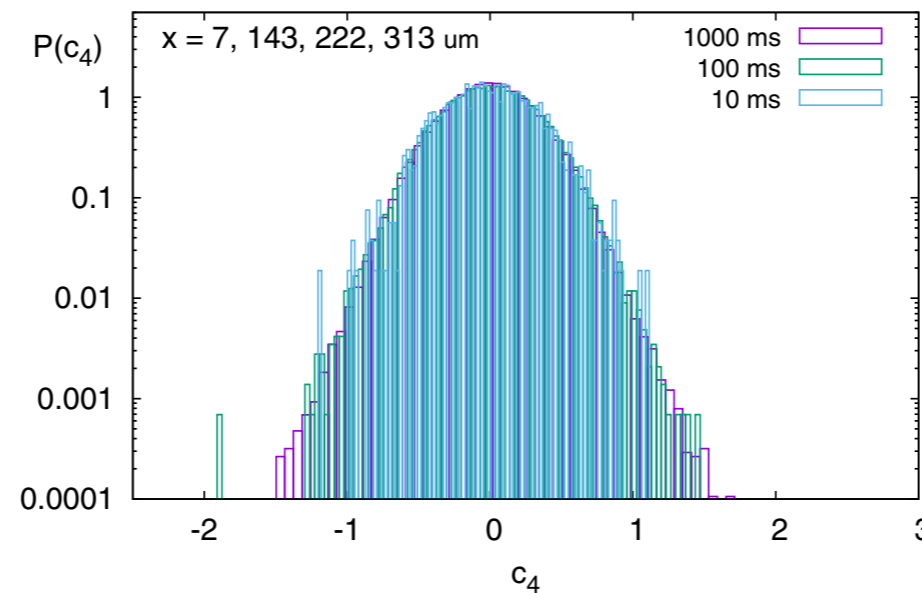
4-point correlations among resonances in different spectra



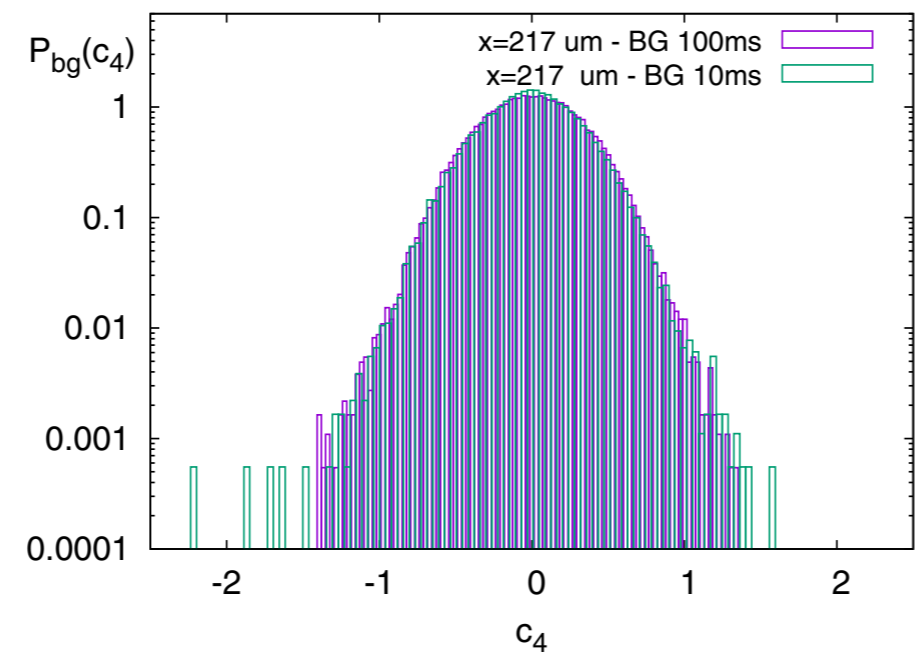
Self-starting mode-locking in random lasers



4-point correlations among resonances in the same spectrum at the same position



4-point correlations among resonances in the same spectrum at far away distances



4-point correlations among resonances in different spectra

4-point correlation distributions



Self-starting mode-locking in random lasers

Correlation vs the mode locking condition on the matching of mode frequencies

$$|\omega_{k_1} - \omega_{k_2} + \omega_{k_3} - \omega_{k_4}| < \gamma; \quad \gamma \equiv \sum_{j=1}^4 \gamma_{k_j}$$

We introduce the control parameter

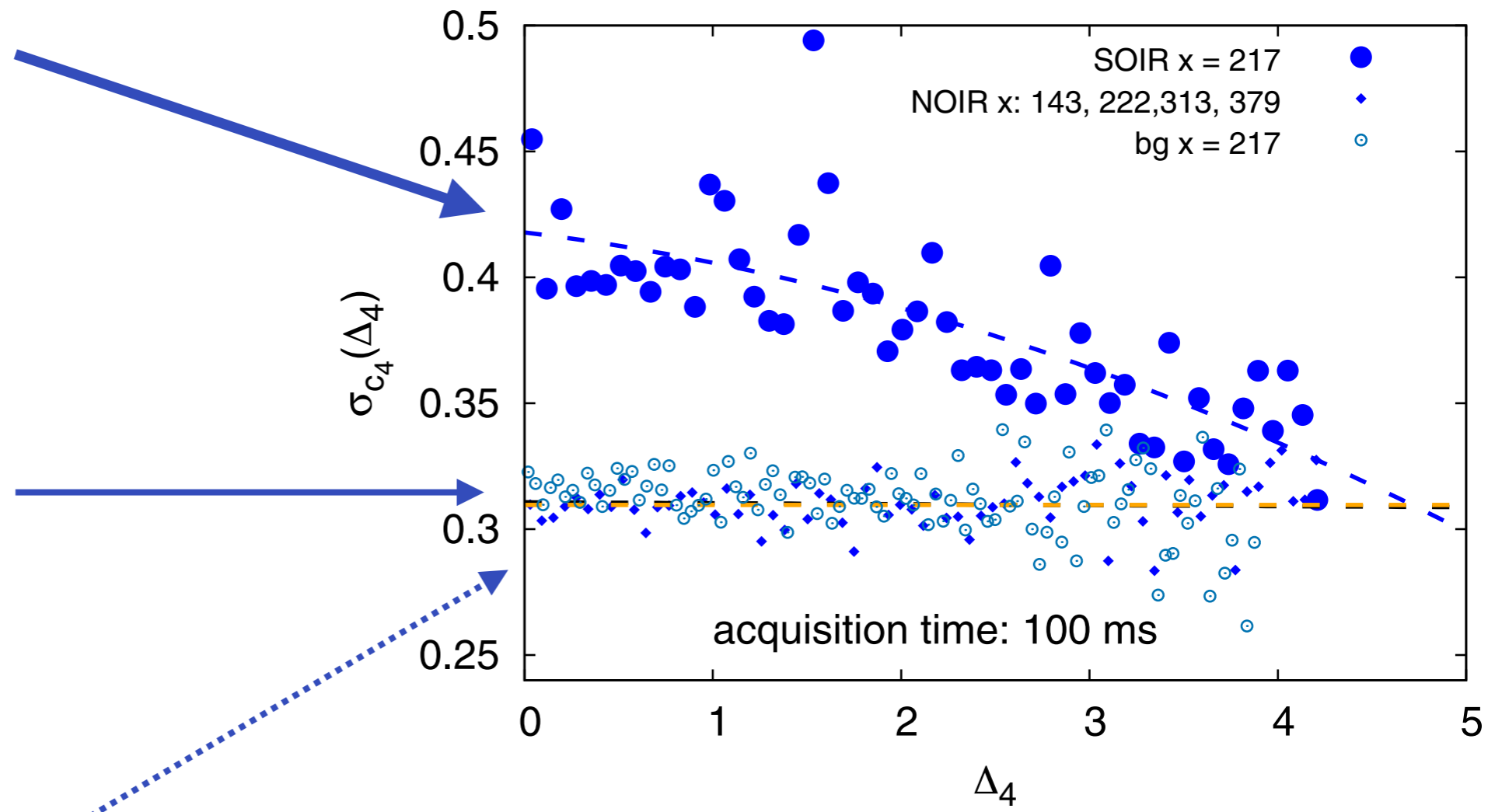
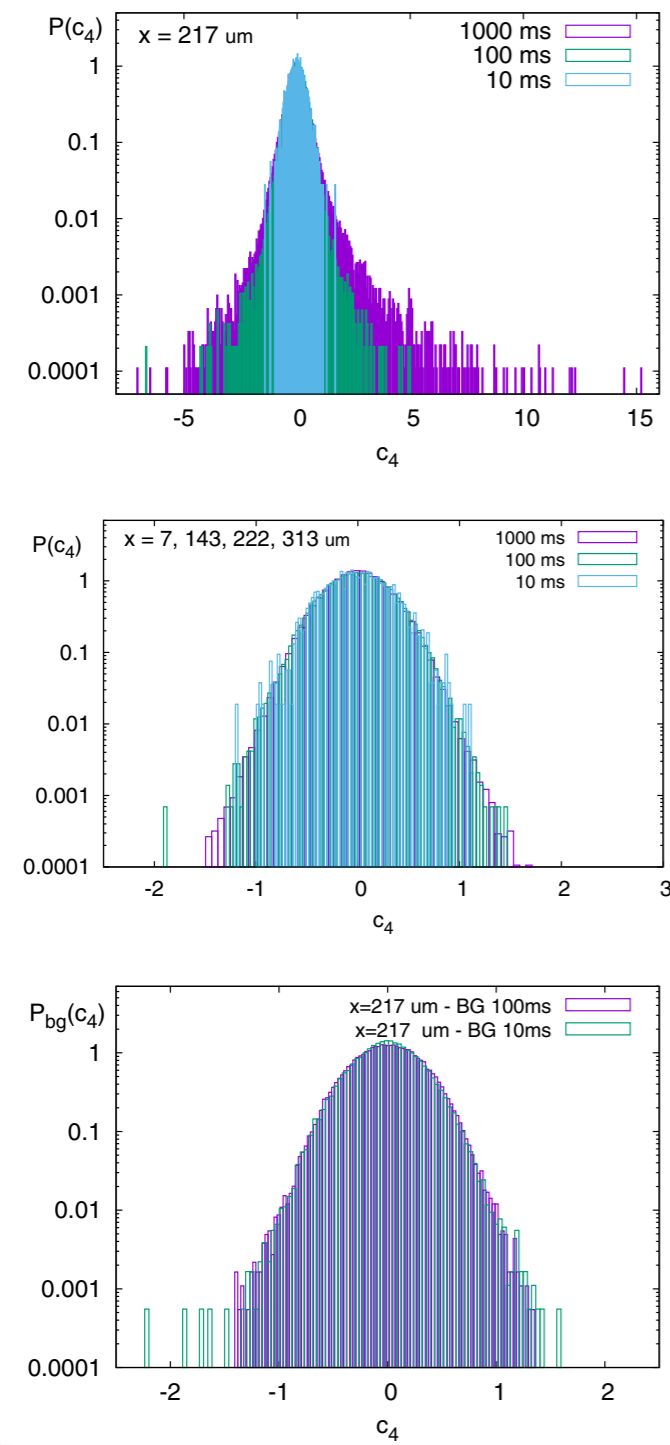
$$\Delta_4 \equiv \frac{|\omega_1 - \omega_2 + \omega_3 - \omega_4|}{\gamma_1 + \gamma_2 + \gamma_3 + \gamma_4}$$

The smaller, the more locked are the modes



Self-starting mode-locking in random lasers

Variance correlation vs the matching of mode frequencies

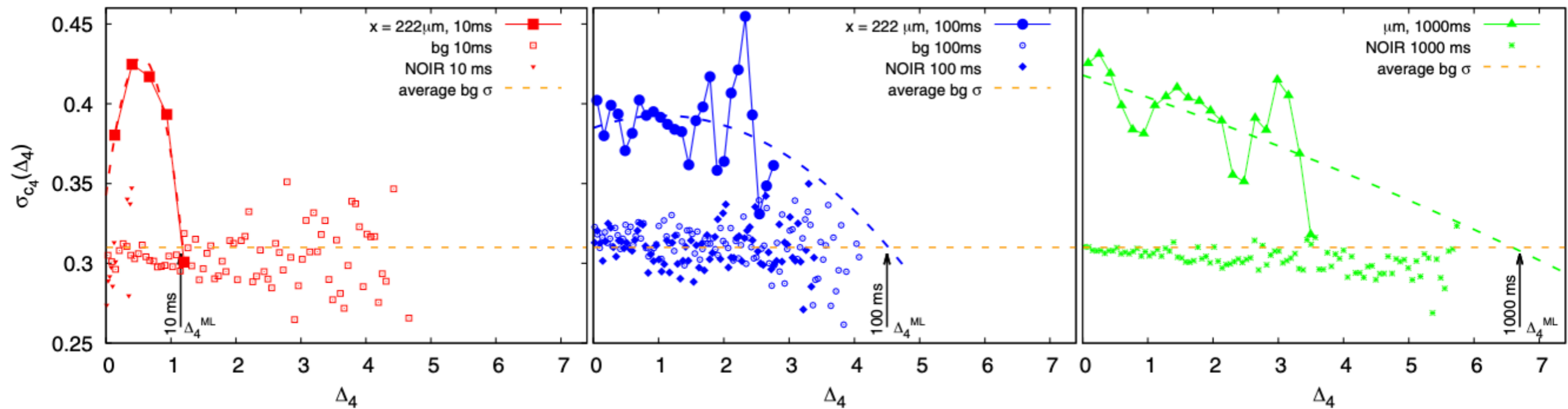


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Self-starting mode-locking in random lasers

Variance correlation vs the matching of mode frequencies



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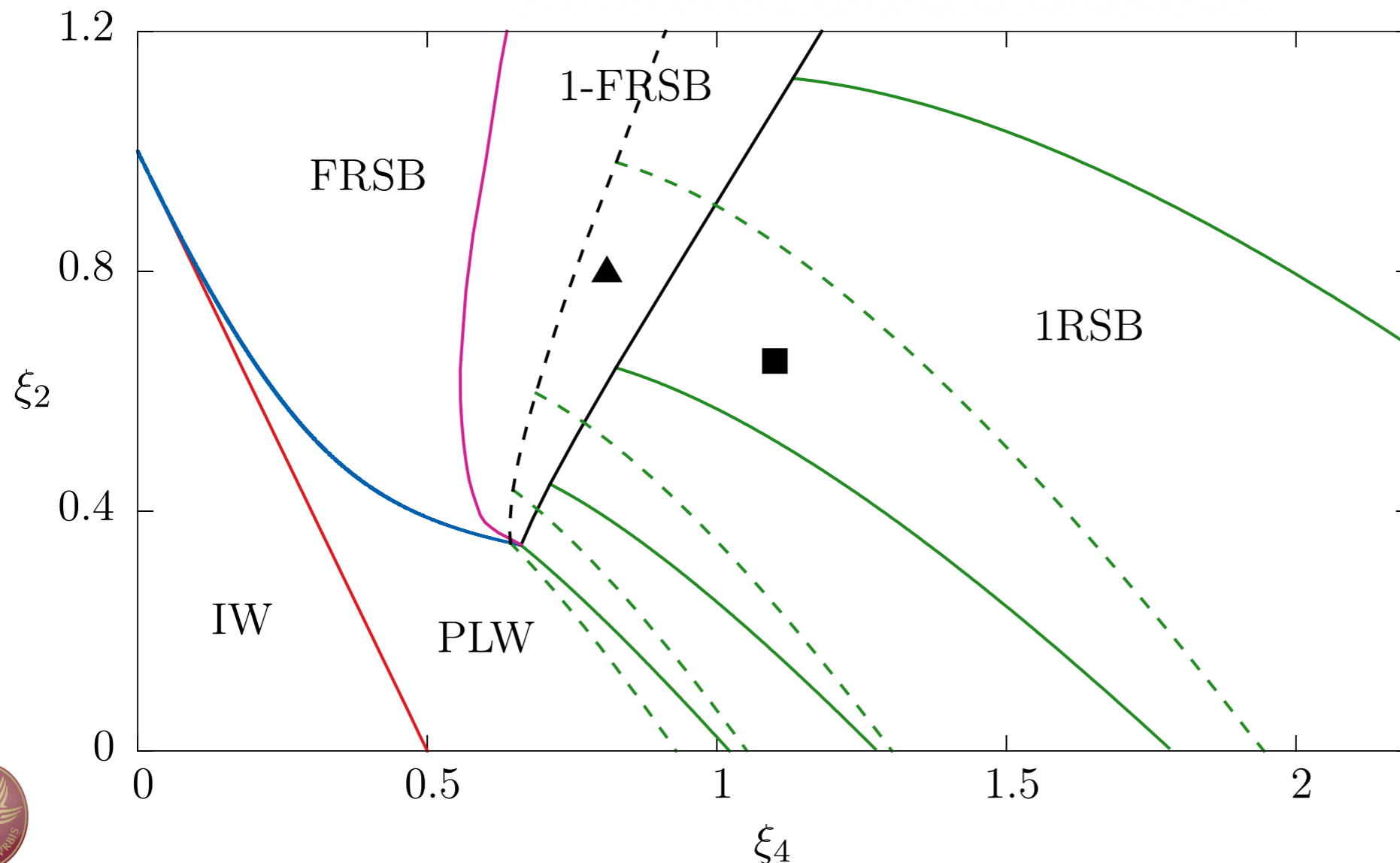


Fully connected 2+4 spherical phasor model

$$\mathcal{H}[\mathbf{a}] = -\frac{1}{2} \sum_{n_1 n_2}^{1,N} J_{n_1 n_2} a_{n_1} a_{n_2}^* - \frac{1}{4!} \sum_{n_1 n_2 n_3 n_4}^{1,N} J_{n_1 n_2 n_3 n_4} a_{n_1} a_{n_2}^* a_{n_3} a_{n_4}^*$$

Phase diagram in “mode-coupling”-like parameters

$$\xi_2 = \frac{\epsilon^2}{4} \beta^2 J_2^2, \quad \xi_4 = \frac{\epsilon^4}{6} \beta^2 J_4^2,$$



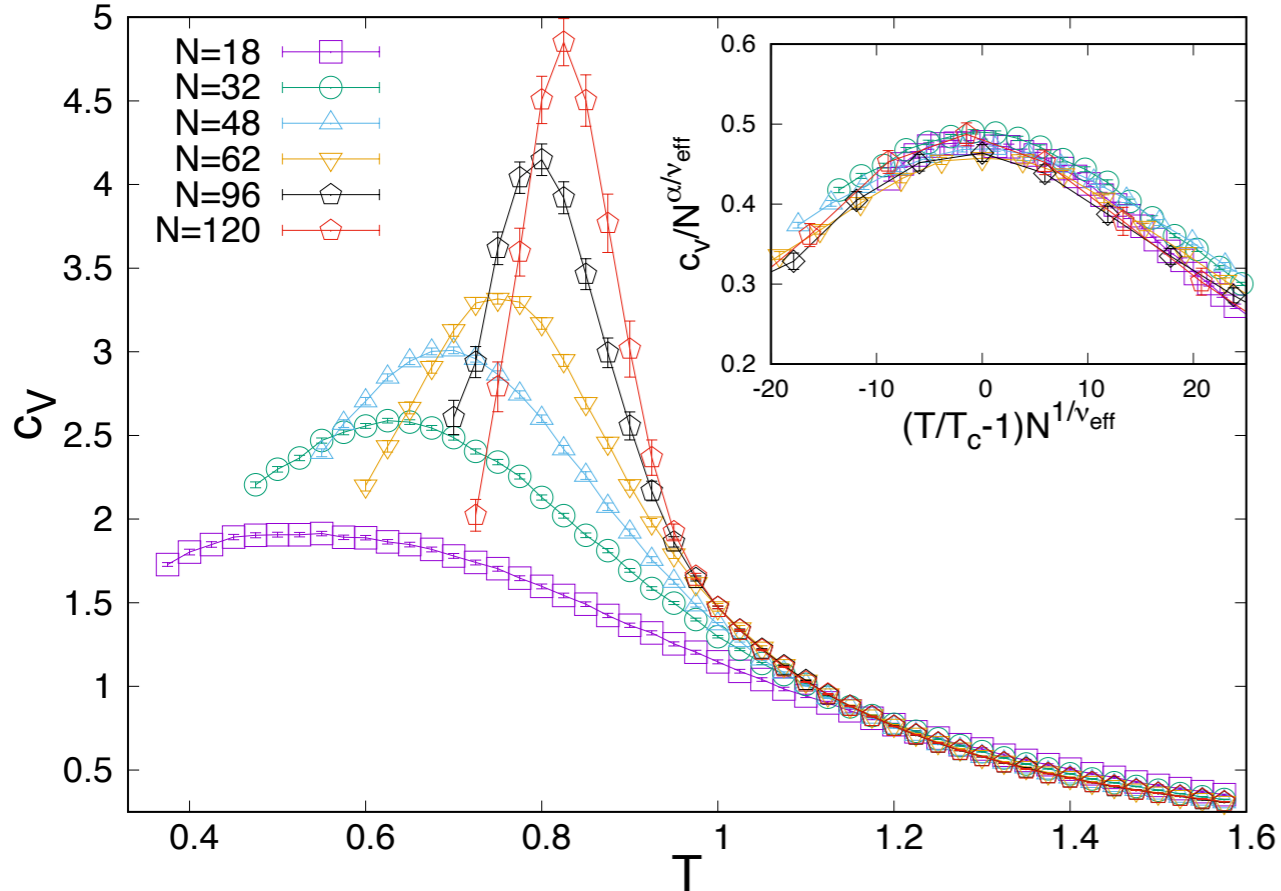
Universality class

$$c_{V_N}(T) = N^{\frac{\alpha}{\nu_{\text{eff}}}} \hat{f}_{c_{V_N}} \left(N^{\frac{1}{\nu_{\text{eff}}}} t_N \right)$$

$$\nu_{\text{eff}} = 2\beta + \gamma$$

$1 \leq \nu_{\text{eff}} \leq 2$ for a mean-field model

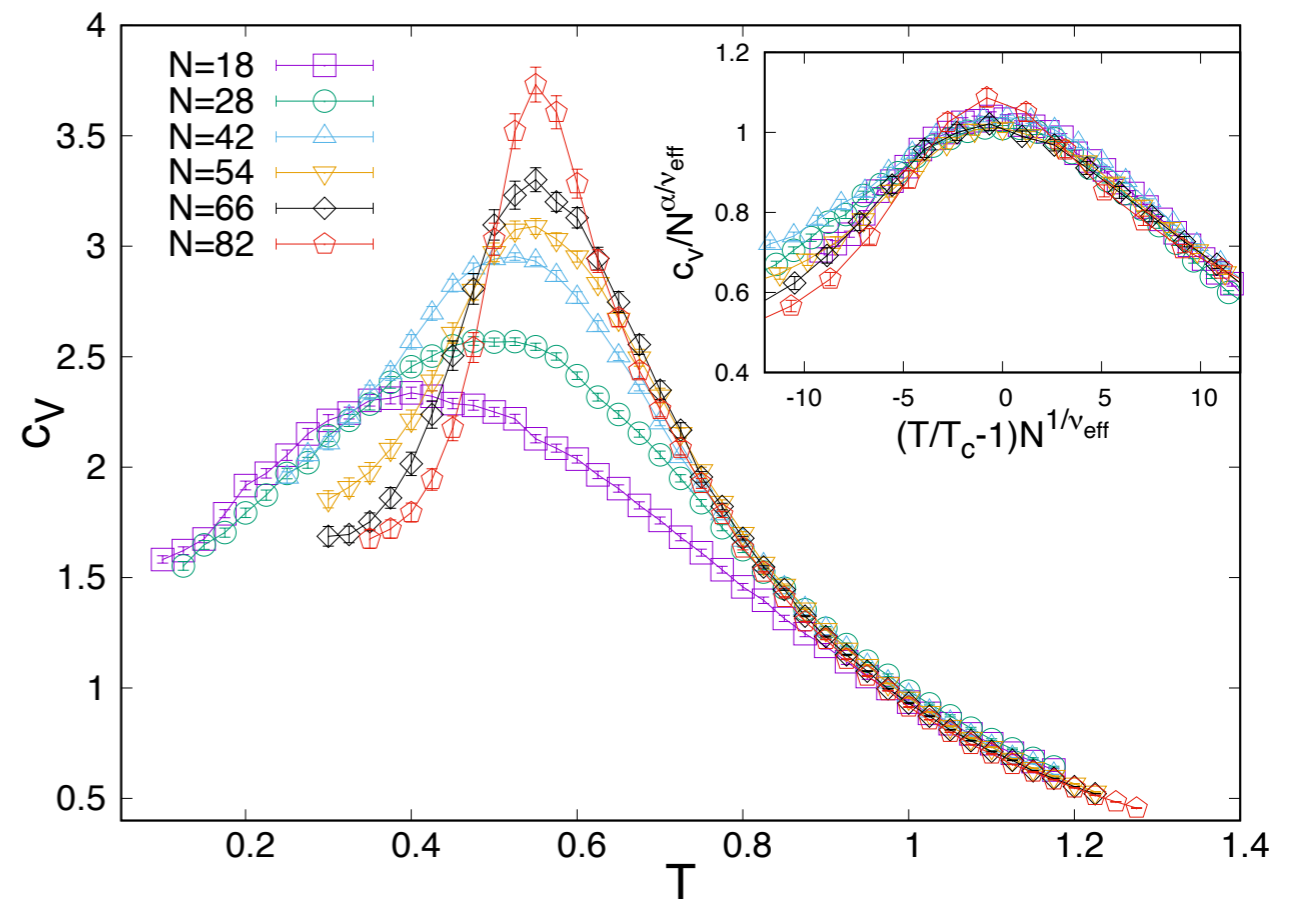
Free boundary conditions on frequencies



FBC: $T_c(\infty) = 0.86 \pm 0.03$

$\alpha = 0.48 \pm 0.05$ $1/\nu_{\text{eff}} = 1.1 \pm 0.1$

Periodic boundary conditions on frequencies



PBC: $T_c(\infty) = 0.61 \pm 0.03$

PBC: $\alpha = 0.27 \pm 0.05$, $1/\nu_{\text{eff}} = 0.86 \pm 0.14$

