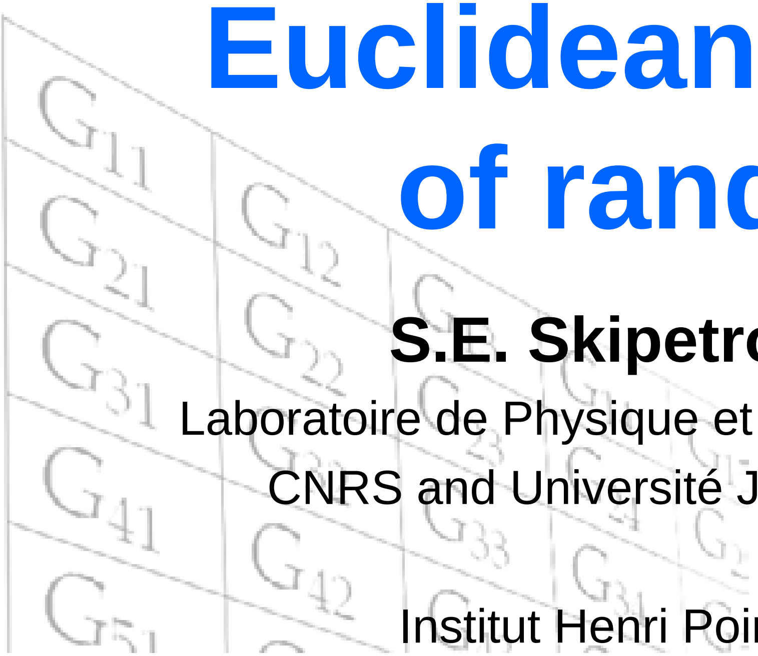
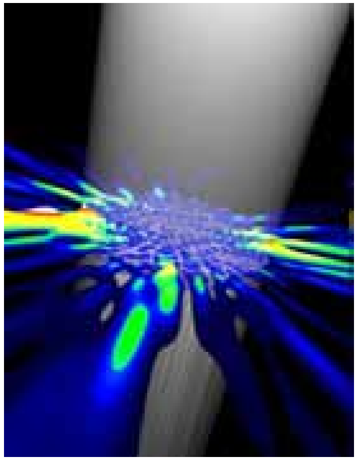


Euclidean matrix theory of random lasing

S.E. Skipetrov and **A. Goetschy**
Laboratoire de Physique et Modélisation des Milieux Condensés
CNRS and Université Joseph Fourier, Grenoble, France

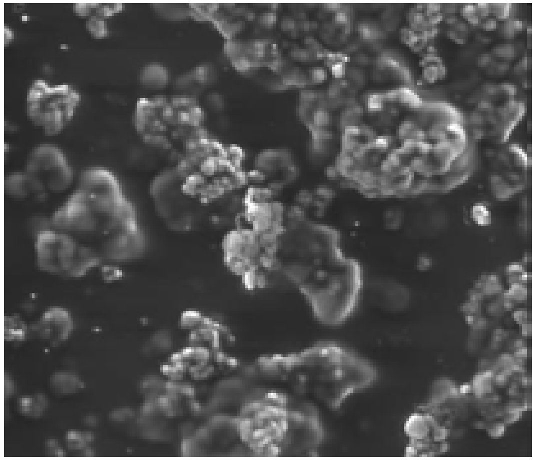
Institut Henri Poincaré, Paris, May 15, 2012

Random laser in pictures

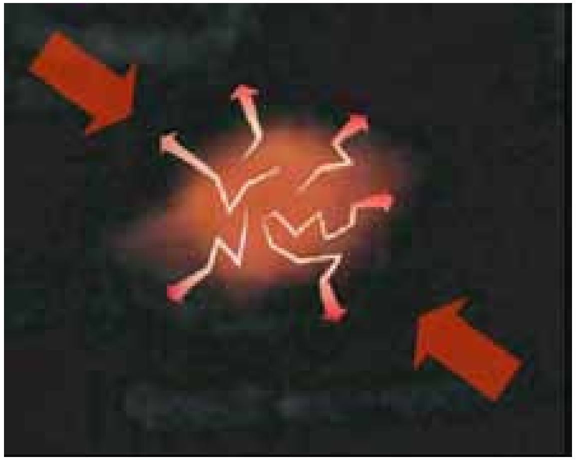


H.E. Tureci et al.

D.S. Wiersma & A. Lagendijk

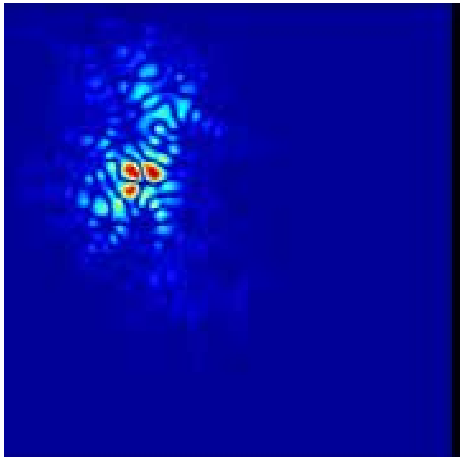


R. Polson et al.



W. Guerin et al.

M.A. Noginov

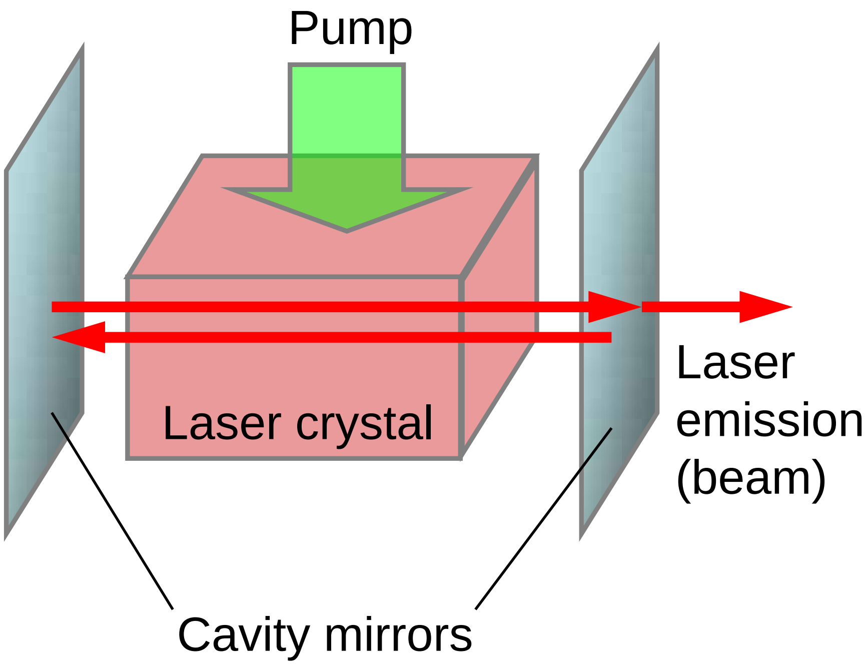


P. Sebbah & Ch. Vanneste

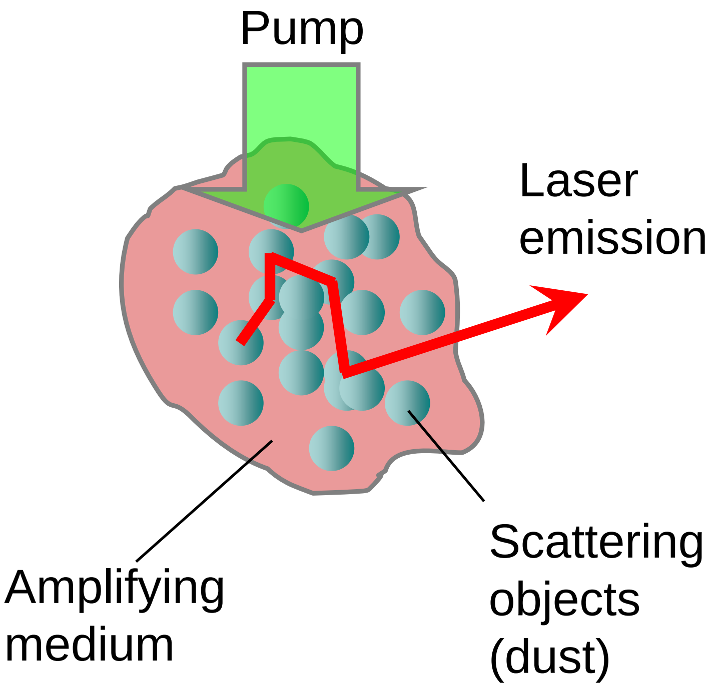
“Making lasers from dust”

(The title of an article by D.S. Wiersma in Photonics Spectra, February 2007)

“Normal” laser



Random laser



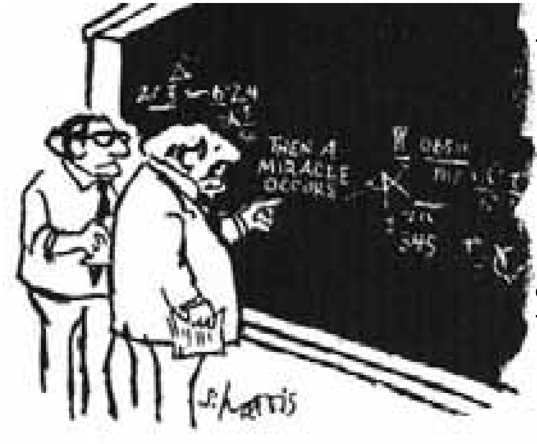
Theory of random laser

Diffusion theory

Idea: Replace intensity I in laser equations by its average value $\langle I \rangle$ that obeys diffusion equation

Advantage: Allows for many analytical results (threshold, dynamics beyond the threshold, etc.)

Drawback: Replacement $I \rightarrow \langle I \rangle$ not justified



"I think you should be more explicit here in step two."

http://revelation4-11.blogspot.com/

Anomalous states

Idea: Study untypical states (modes) that might be most adapted for lasing

Advantage: Analytical approach beyond the diffusion theory

Drawback: No guarantee of finding *the* best states, results specific for a particular system, not always realizable

Numerical methods

Idea: Study lasing for a given realization of disorder, then average (if computer time allows)

Advantage: Numerically exact, easy to adapt to a specific system

Drawback: Results are difficult to interpret and to generalize, too time consuming in 3D

Theory of random laser

Diffusion theory

Idea: Replace intensity *I* in laser equations by its average value <*I* > that obeys diff

Advantag
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Drawback
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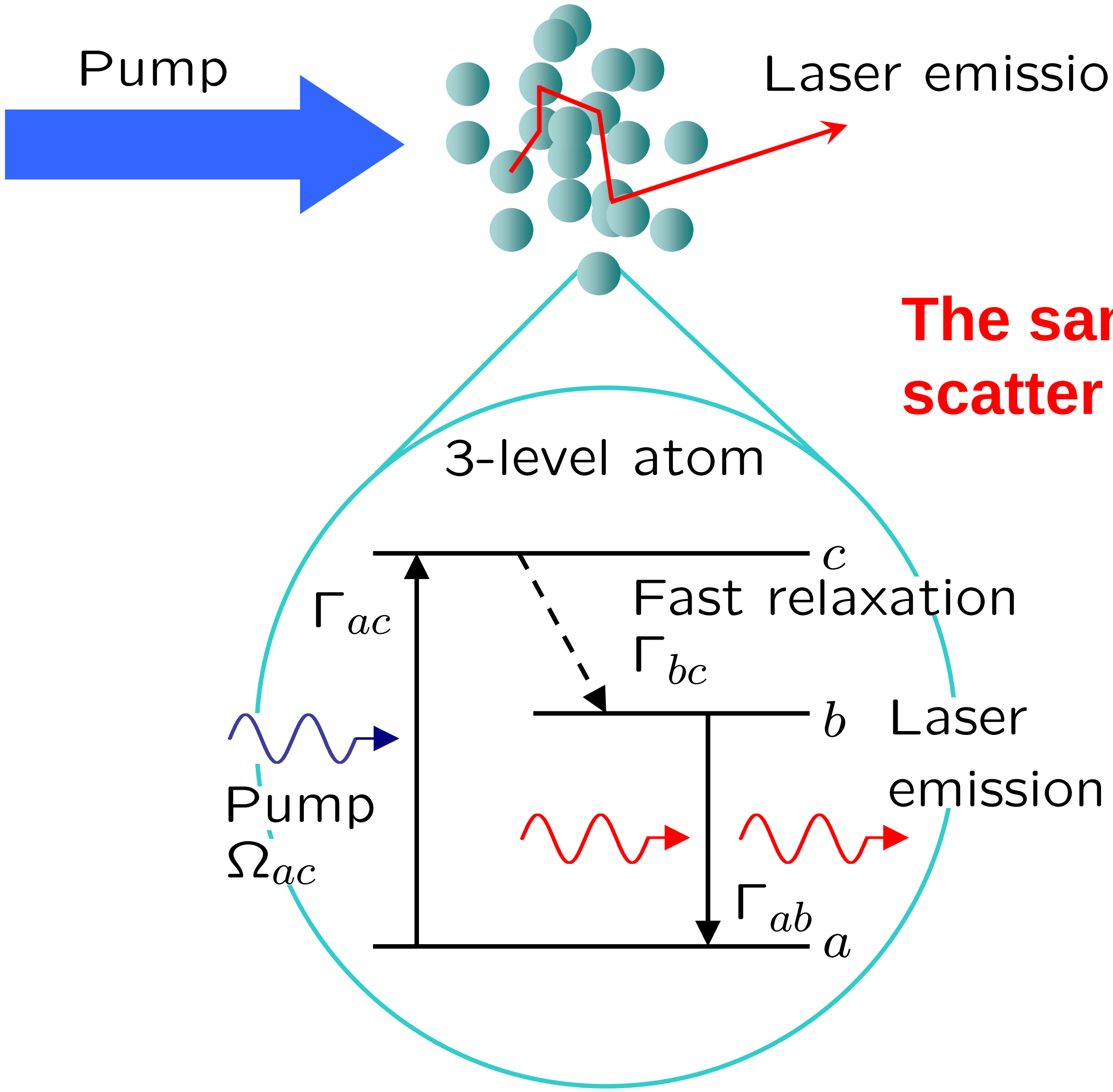
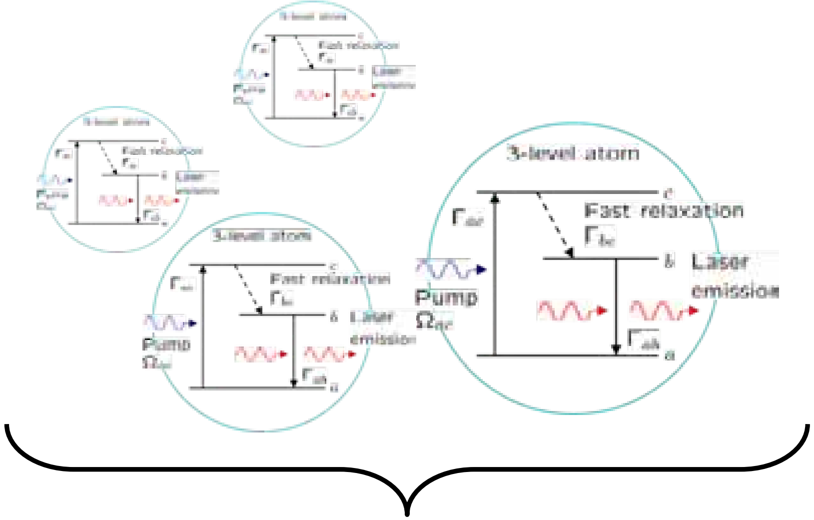
Need for a simple model that allows for an “exact” solution

The simplest random laser



The same atoms scatter and amplify light !

Random lasing in a cloud of 3-level atoms



N atoms

Dipole approximation for the field-atom interaction

↓

Heisenberg equations for field and atomic operators

↓

Heisenberg-Langevin equations for atomic operators

$\Pi = |b\rangle\langle b| - |a\rangle\langle a|$, $S^+ = |b\rangle\langle a|$:

$$\frac{dS_i^+}{dt} = \left[\mathrm{i}\omega_0 - \frac{\Gamma}{2}(1+W_i)\right] S_i^+ + \mathrm{i}\frac{\Gamma}{2}\Pi_i \sum_{j\neq i}^{N} G_{ij}^* S_j^+ + F_i^+(\mathbf{r}_i, t)$$

$$\frac{d\Pi_i}{dt} = -\Gamma\left[(1+W_i)\,\Pi_i + (1-W_i)\right] - 2\Gamma\,\mathrm{Im}\left[S_i^+ \sum_{j\neq i}^{N} G_{ij} S_j^-\right]$$

$$+ \quad F_i^{\Pi}(\mathbf{r}_i, t)$$

Pump

Green's matrix $G_{ij} = \dfrac{e^{\mathrm{i}k_0|\mathbf{r}_i - \mathbf{r}_j|}}{k_0|\mathbf{r}_i - \mathbf{r}_j|}$

(scalar model)

[Similar approach: Savels, Mosk and Lagendijk (2005)]

Random lasing in a cloud of 3-level atoms

- Laser threshold:

$$\boxed{\lambda_n = \frac{1}{t}}$$

The dimensionless scattering matrix of a single atom:

$$t(\omega) = \frac{1}{2} \times \frac{W-1}{W+1} \times \frac{1}{\omega - \omega_0 + \frac{\mathrm{i}}{2}(W+1)}$$

An eigenvalue of the matrix $\widehat{G}$: $\widehat{G}\vec{\Psi}_n = \lambda_n \vec{\Psi}_n$

The true scattering matrix is $\tilde{t} = (4\pi/k_0)t$ [see Savels, Mosk and Lagendijk (2005)]

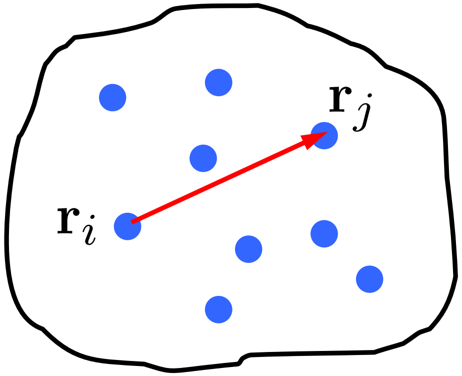
- Rate equations beyond threshold:

$$\frac{dI_n}{dt} = -2\kappa_n I_n + \sum_m \gamma_{nm} I_n I_m$$

$$\gamma_{nm} = 4\Gamma \frac{W-1}{(W+1)^3} \times \mathrm{Im}\left[\lambda_n \frac{\sum_{i=1}^N |\Psi_n^i|^2 (\Psi_m^i)^2}{\sum_{i=1}^N |\Psi_n^i|^2}\right]$$

$$\kappa_n = \frac{\Gamma}{2} \times \frac{W-1}{W+1}\left[\frac{(W+1)^2}{W-1} - \mathrm{Im}\lambda_n\right]$$

Euclidean random matrices



$i, j = 1, \dots N$

$N \times N$ matrix $G_{ij} = G(\mathbf{r}_i, \mathbf{r}_j)$

Eigenvalue problem:

$$\widehat{G}\vec{\Psi}_n = \lambda_n \vec{\Psi}_n$$

λ_n — eigenvalues

$\vec{\Psi}_n = \{\Psi_n^i\}$ — eigenvectors

$n = 1, \dots, N$

Euclidean matrix $G_{ij} = \frac{e^{ik_0|\mathbf{r}_i - \mathbf{r}_j|}}{k_0|\mathbf{r}_i - \mathbf{r}_j|}$ determines the behavior of our random laser

The notion of Euclidean random matrices was first introduced by M. Mézard, G. Parisi and A. Zee (1999)

Eigenvalue density of Euclidean matrices: main idea

Hermitian matrices: $p(\lambda) = -\frac{1}{\pi}\mathrm{Im}\mathcal{G}(z=\lambda+\mathrm{i}\epsilon)$

Green's function (resolvent):

$$\mathcal{G}(z) = \frac{1}{N}\left\langle \mathrm{Tr}\frac{1}{z-\widehat{G}}\right\rangle = \frac{1}{N}\left\langle \mathrm{Tr}\left(\frac{1}{z}+\frac{1}{z}\widehat{G}\frac{1}{z}+\dots\right)\right\rangle = \frac{1}{z-\Sigma(z)}$$

Non-Hermitian matrices: $\widehat{G}_{N\times N} \xrightarrow{\quad} \widehat{G}_2 = \begin{pmatrix}\widehat{G} & 0\\ 0 & \widehat{G}^+\end{pmatrix}_{2N\times 2N}$

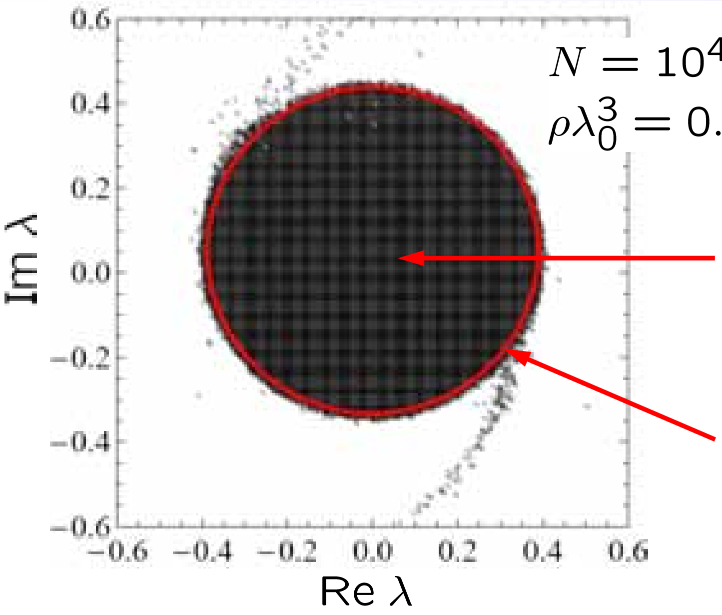
$$\widehat{\mathcal{G}} = \left\langle \frac{1}{\hat{z}_\epsilon - \widehat{G}_2}\right\rangle = \begin{pmatrix}\widehat{\mathcal{G}}_{qq} & \widehat{\mathcal{G}}_{q\bar{q}}\\ \widehat{\mathcal{G}}_{\bar{q}q} & \widehat{\mathcal{G}}_{\bar{q}\bar{q}}\end{pmatrix} \quad \text{where} \quad \hat{z}_\epsilon = \begin{pmatrix} z & \mathrm{i}\epsilon\\ \mathrm{i}\epsilon & \bar{z}\end{pmatrix}$$

Green's function (resolvent):

$$g(z,\bar{z}) = \frac{1}{N}\left\langle \mathrm{Tr}\frac{\bar{z}-\widehat{G}^+}{(z-\widehat{G})(\bar{z}-\widehat{G}^+)+\epsilon^2}\right\rangle = \frac{1}{N}\mathrm{Tr}\widehat{\mathcal{G}}_{qq}$$

$$\boxed{p(\lambda) = \frac{1}{\pi}\frac{\partial}{\partial\bar{z}}g(z,\bar{z})\Big|_{z=\lambda}}$$

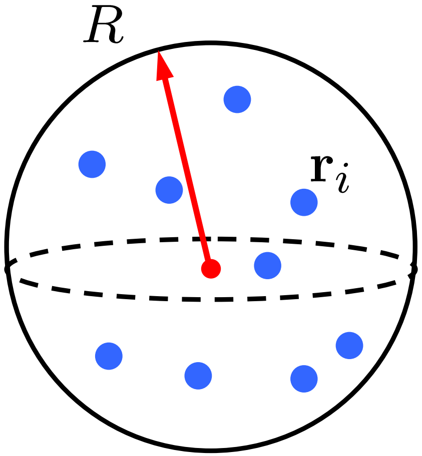
Eigenvalue density of the matrix $\hat{G}$: main results



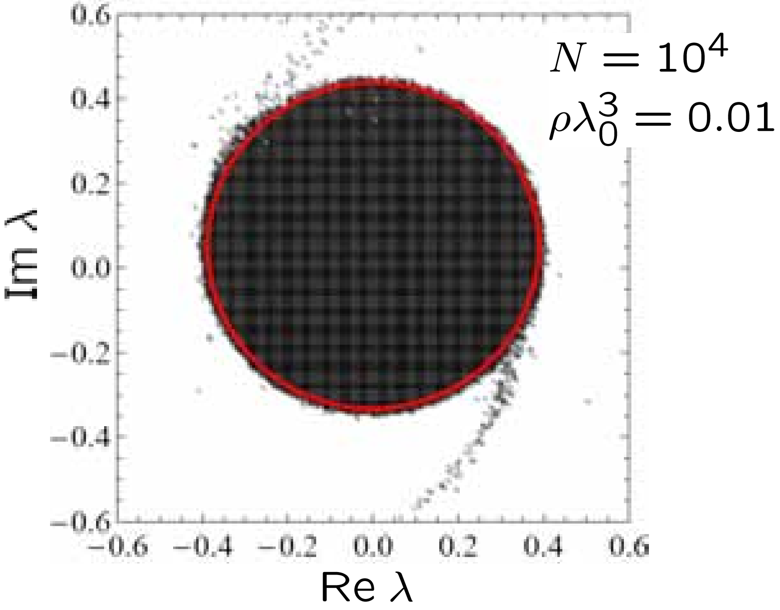
Density of eigenvalues of $\widehat{G}$ (numerical)

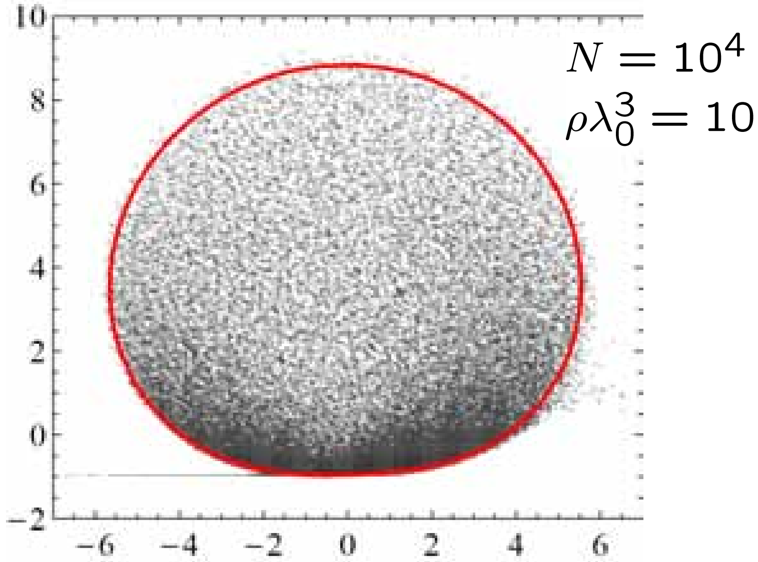
Borderline of the domain of existence of eigenvalues of $\widehat{G}$ (analytical)

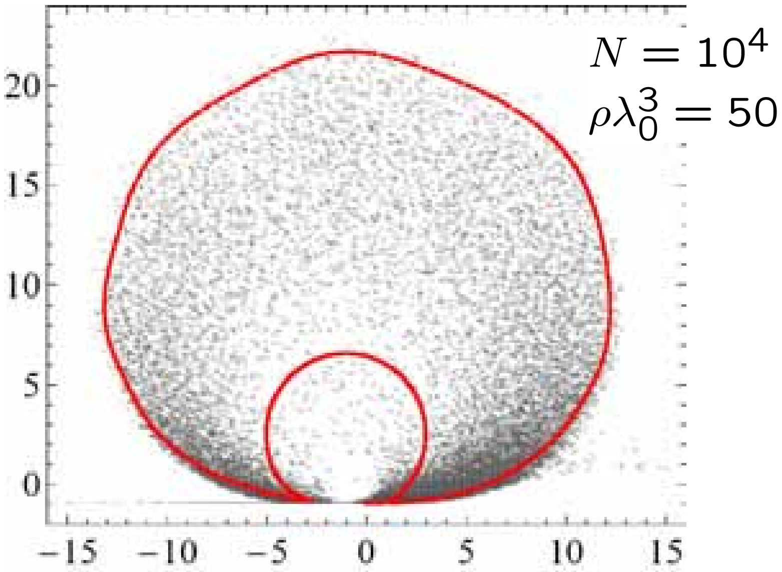
$N \gg 1$ atoms in a sphere:

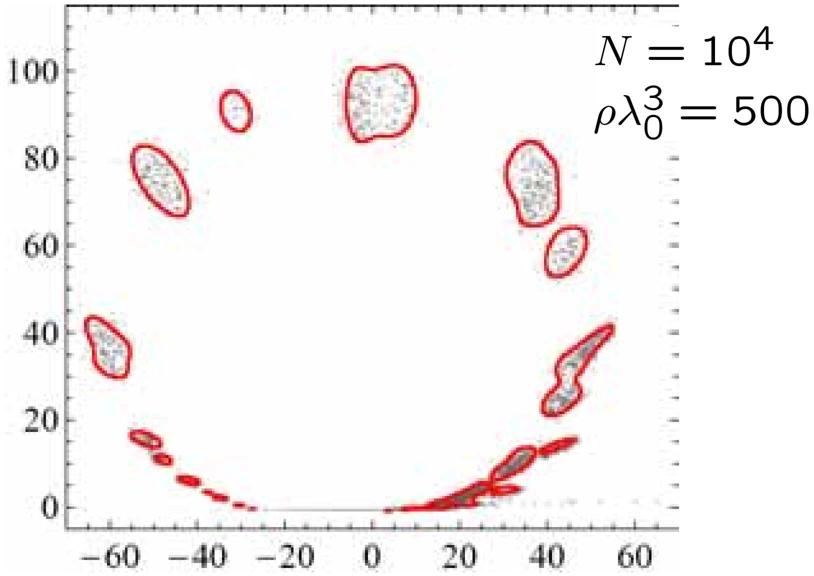


Eigenvalue density of the matrix $\hat{G}$: main results

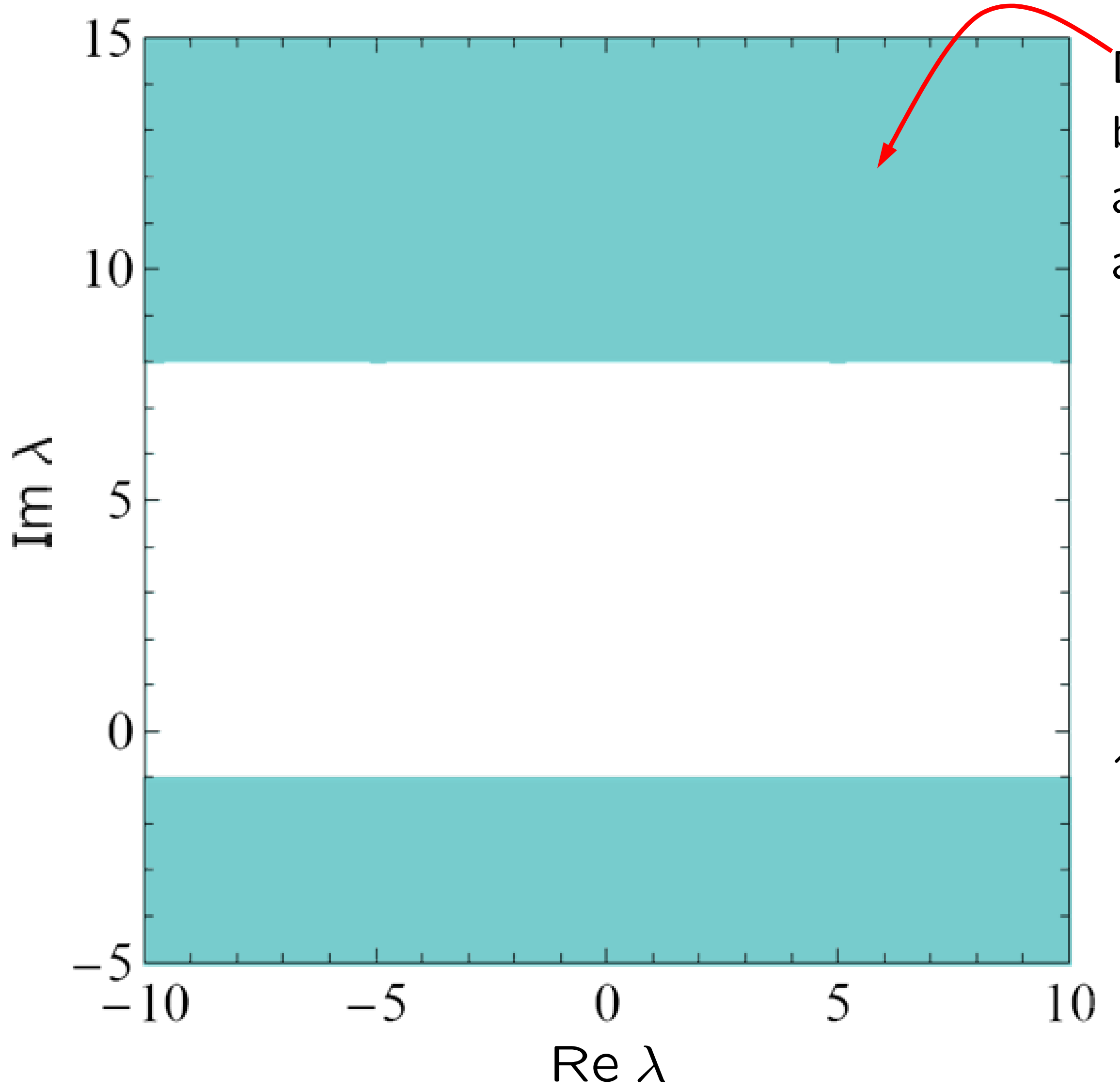








Random lasing in a cloud of 3-level atoms



Domain spanned by $1/t(\omega)$ when ω and the pump W are varied

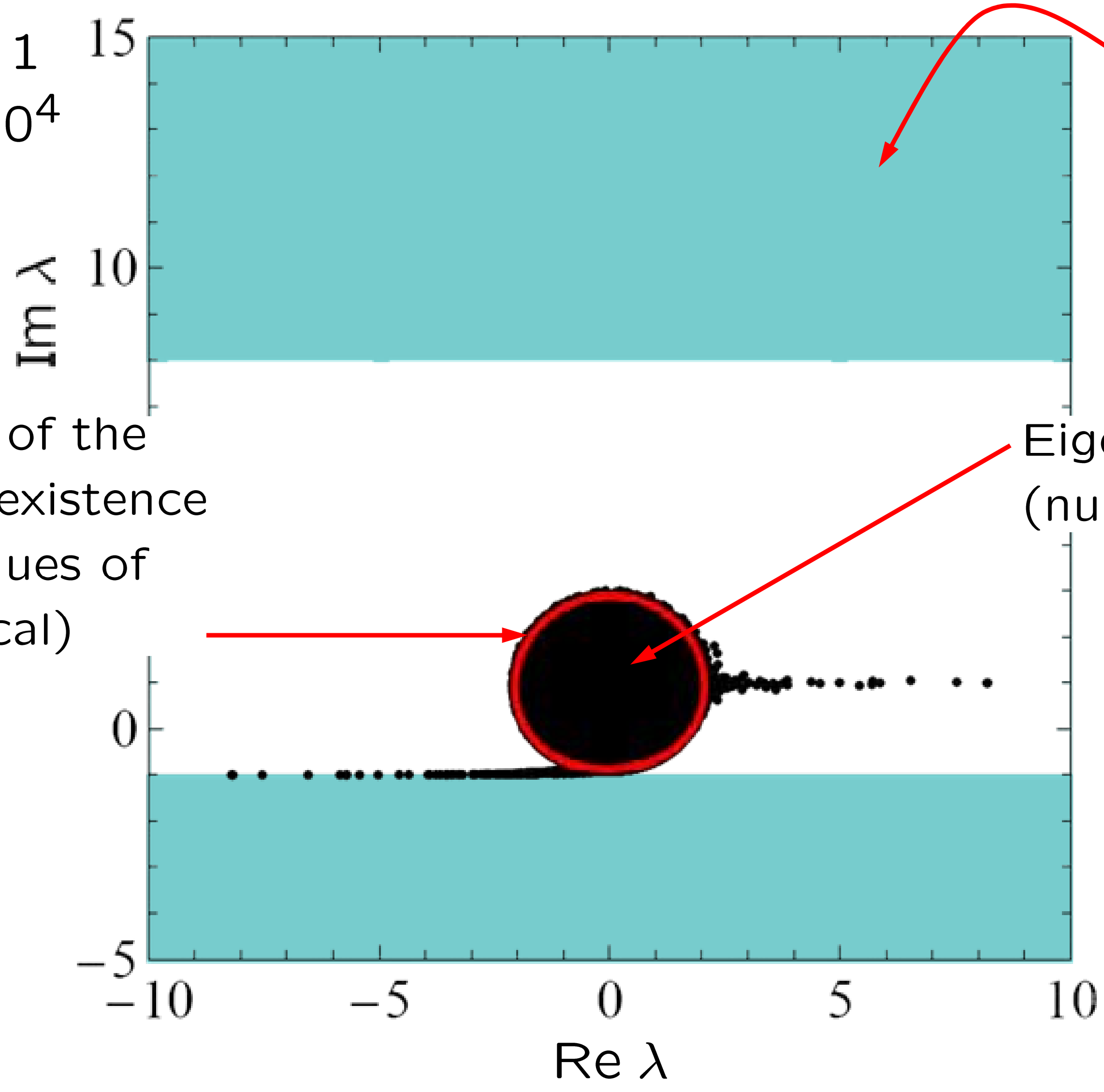
Laser threshold:

$$\lambda_n = \frac{1}{t}$$

Random lasing in a cloud of 3-level atoms

$\rho\lambda_0^3 = 1$

$N = 10^4$



Domain spanned by $1/t(\omega)$ when ω and the pump W are varied

Borderline of the domain of existence of eigenvalues of $\widehat{G}$ (analytical)

Eigenvalues of $\widehat{G}$ (numerical)

Laser threshold:

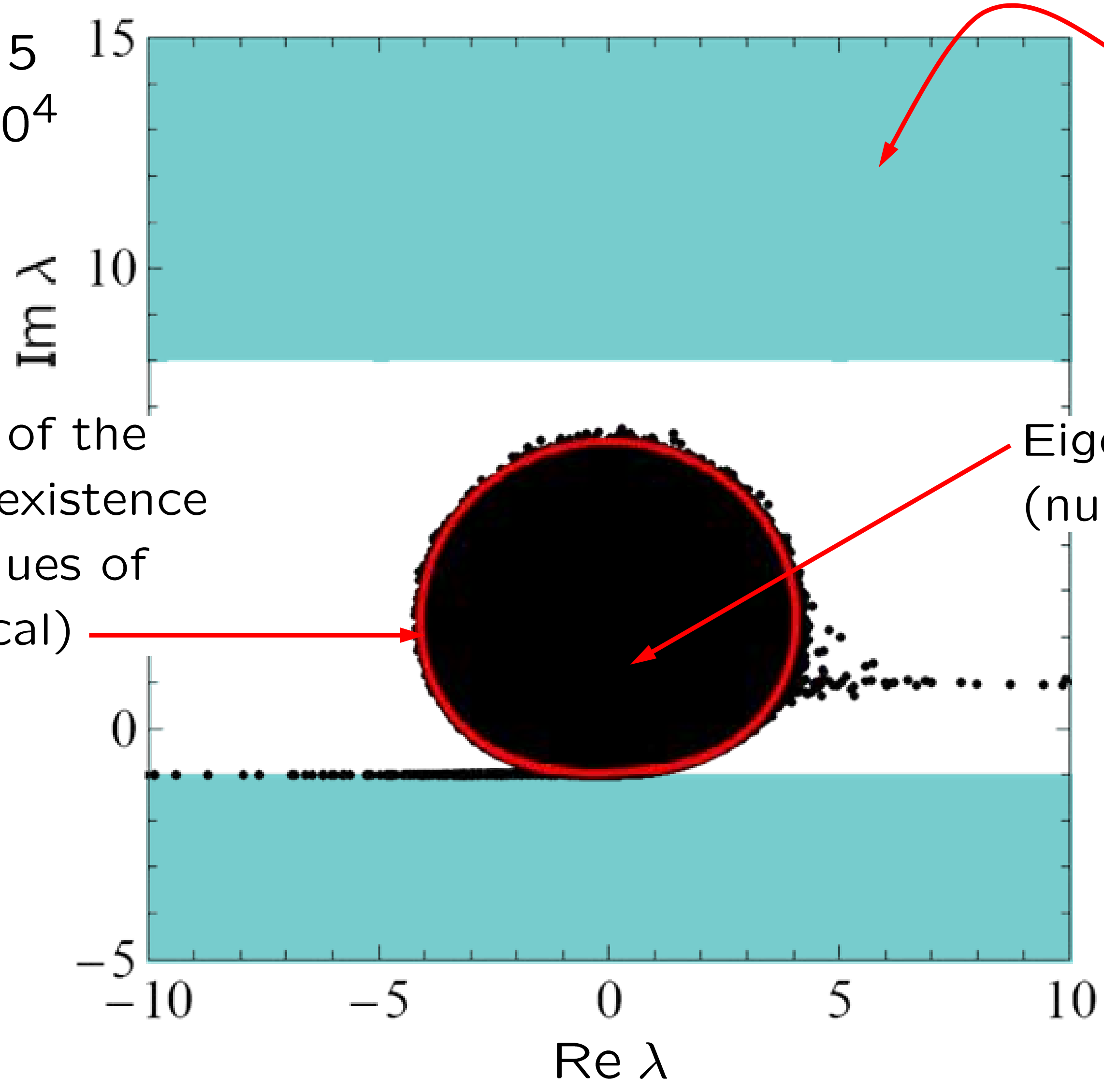
$$\lambda_n = \frac{1}{t}$$

Well below threshold...

Random lasing in a cloud of 3-level atoms

$\rho\lambda_0^3 = 5$

$N = 10^4$



Domain spanned by $1/t(\omega)$ when ω and the pump W are varied

Borderline of the domain of existence of eigenvalues of $\widehat{G}$ (analytical)

Eigenvalues of $\widehat{G}$ (numerical)

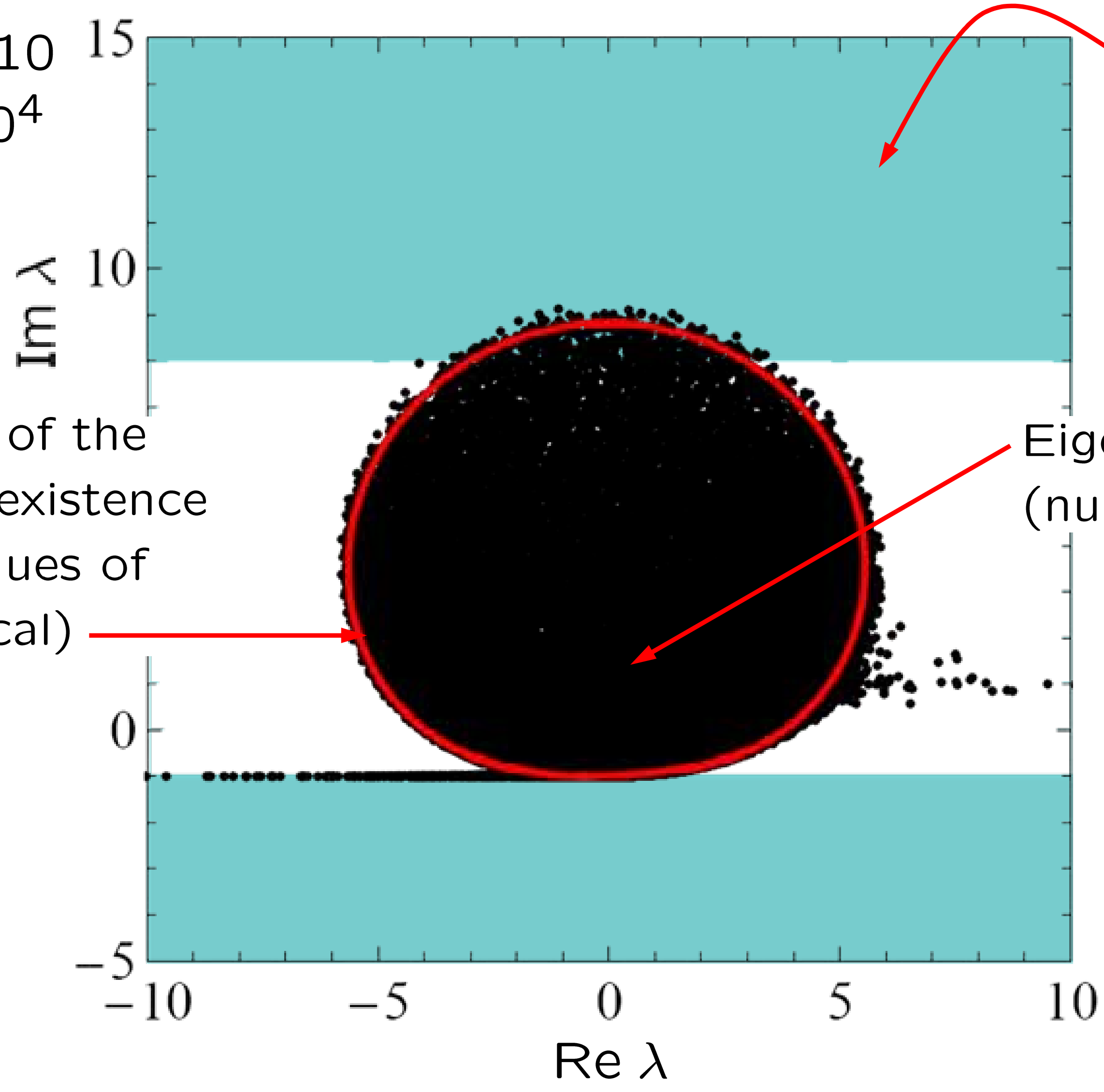
Laser threshold:

$$\lambda_n = \frac{1}{t}$$

Approaching threshold...

Random lasing in a cloud of 3-level atoms

$\rho\lambda_0^3 = 10$
$N = 10^4$



Domain spanned by $1/t(\omega)$ when ω and the pump W are varied

Borderline of the domain of existence of eigenvalues of $\widehat{G}$ (analytical)

Eigenvalues of $\widehat{G}$ (numerical)

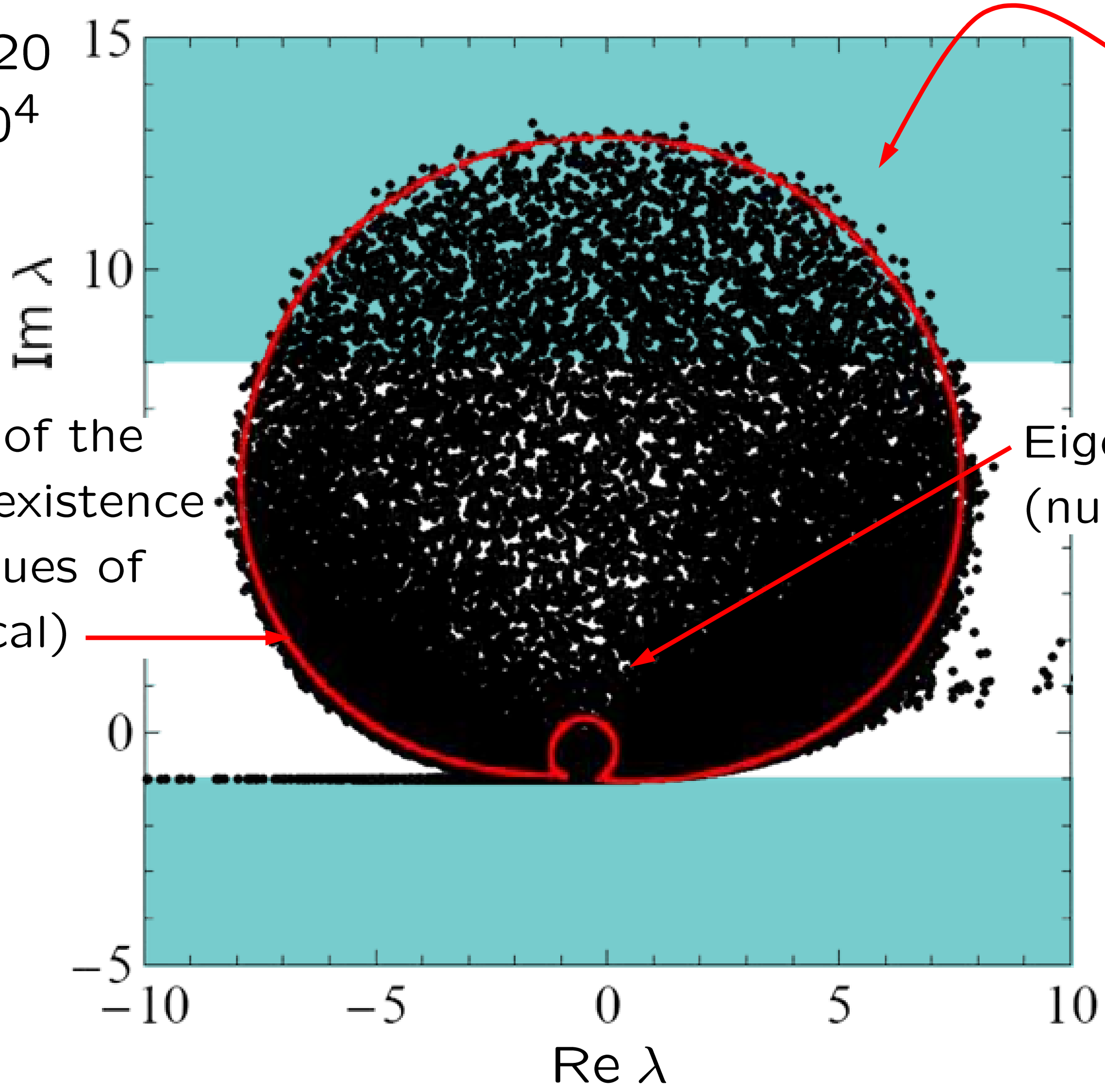
Laser threshold:

$$\lambda_n = \frac{1}{t}$$

Threshold reached!

Random lasing in a cloud of 3-level atoms

$\rho\lambda_0^3 = 20$
$N = 10^4$



Domain spanned by $1/t(\omega)$ when ω and the pump W are varied

Borderline of the domain of existence of eigenvalues of $\widehat{G}$ (analytical)

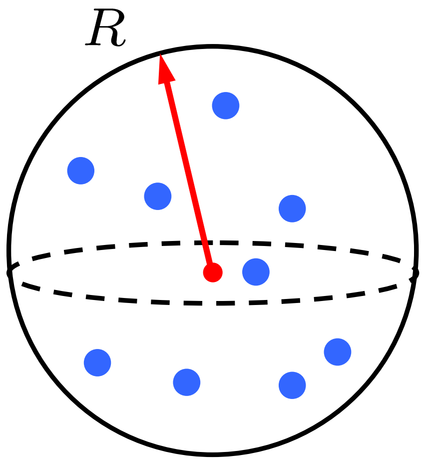
Eigenvalues of $\widehat{G}$ (numerical)

Laser threshold:

$$\lambda_n = \frac{1}{t}$$

Far above threshold...

And what about the “standard” diffusion theory?



Ensemble of N randomly distributed point-like scatterers with a scattering matrix t

Scattering mean free path: $\ell = \frac{4\pi}{\rho|\tilde{t}|^2}$

Extinction length: $\ell_{\text{ex}} = \frac{k_0}{\rho \, \text{Im}\tilde{t}}$

Absorption/amplification length: $\frac{1}{\ell_a} = \frac{1}{\ell_{\text{ex}}} - \frac{1}{\ell}$

Macroscopic absorption/amplification length:
$L_a^2 = \frac{\ell\ell_a}{3}$

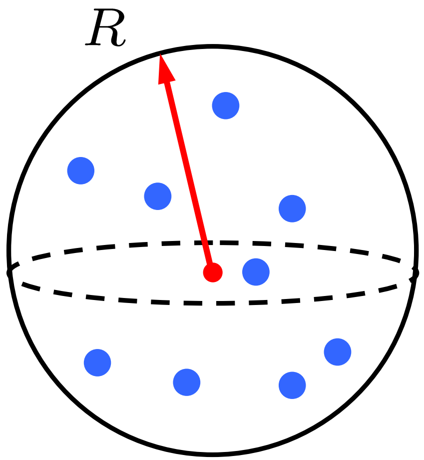
Diffusion equation for the average intensity:

$$\nabla^2 \langle I(\mathbf{r},\mathbf{r}')\rangle - \frac{1}{L_a^2}\langle I(\mathbf{r},\mathbf{r}')\rangle = \delta(\mathbf{r}-\mathbf{r}')$$

Boundary conditions in a sphere:

$$\langle I(\mathbf{r},\mathbf{r}')\rangle = 0 \text{ for } r = \frac{2}{3}\ell\frac{1}{1+\frac{2}{3}\frac{\ell}{R}}$$

And what about the “standard” diffusion theory?



Ensemble of N randomly distributed point-like scatterers with a scattering matrix t

The laser thereshold is reached when $\langle I(\mathbf{r},\mathbf{r}')\rangle$ diverges:

$$\boxed{1 = \frac{3b_0^2}{(2\pi)^2}|t|^2\left(|t|^2 - \mathrm{Im}t\right)\left(1 + \frac{1}{1+\frac{3}{4}b_0|t|^2}\right)^2}$$

$b_0 = 2R/\ell$ — optical thickness at resonance without pump

Link between random matrix and scattering theories

Resolvent of the random matrix theory:

$$\hat{\mathcal{G}} = \left\langle \frac{1}{\hat{z}_\epsilon - \hat{G}_2} \right\rangle = \begin{pmatrix} \hat{\mathcal{G}}_{qq} & \hat{\mathcal{G}}_{q\bar{q}} \\ \hat{\mathcal{G}}_{\bar{q}q} & \hat{\mathcal{G}}_{\bar{q}\bar{q}} \end{pmatrix}_{2N\times 2N} \qquad \hat{g} = \frac{1}{N}\mathrm{Tr}_{\mathrm{block}}\hat{\mathcal{G}} = \begin{pmatrix} g & g_2 \\ g_2 & g \end{pmatrix}$$

$$g_2 = \frac{-\mathrm{i}\epsilon}{N}\,\mathrm{Tr}\left\langle \frac{1}{[t-\hat{G}][\bar{t}-\hat{G}^{+}]+\epsilon^2} \right\rangle\Bigg|_{\epsilon\,\to\,0^+}$$

correlation of left and right eigenvectors

yields $p(\lambda)$

Green's function of Helmholtz equation: $\hat{\mathcal{G}} = \dfrac{\hat{G}}{1-t\hat{G}} = \dfrac{1}{\hat{G}^{-1}-t}$

Average intensity: $I_{ij} = \langle \mathcal{G}_{ij}\bar{\mathcal{G}}_{ij} \rangle$

Sum over i and average over j:

$$I \;=\; \frac{1}{N}\sum_{i,j=1}^{N} I_{ij} = \frac{1}{N}\mathrm{Tr}\left\langle \frac{1}{[t-\hat{G}^{-1}][\bar{t}-(\hat{G}^{-1})^{+}]} \right\rangle$$

Link between random matrix and scattering theories

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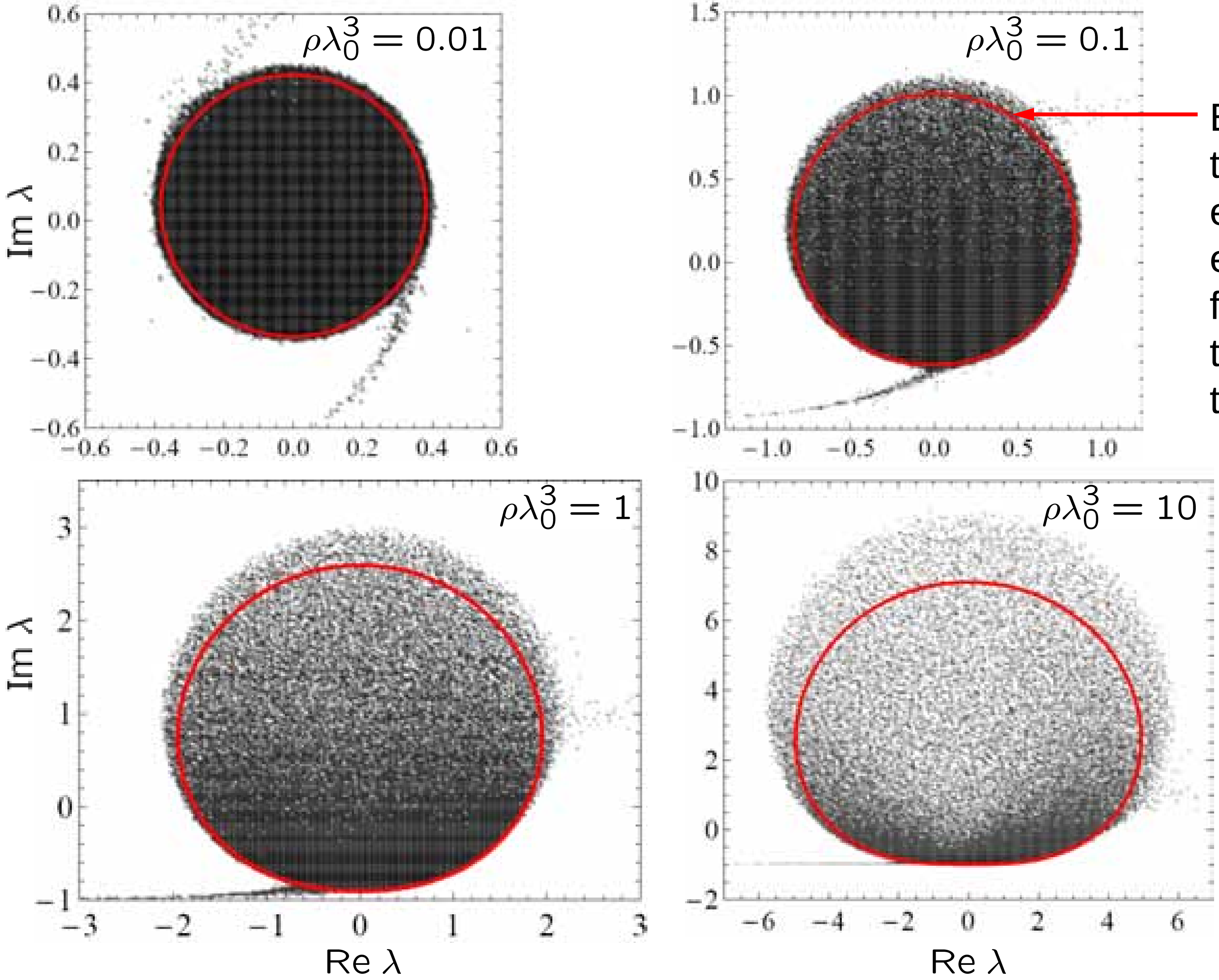
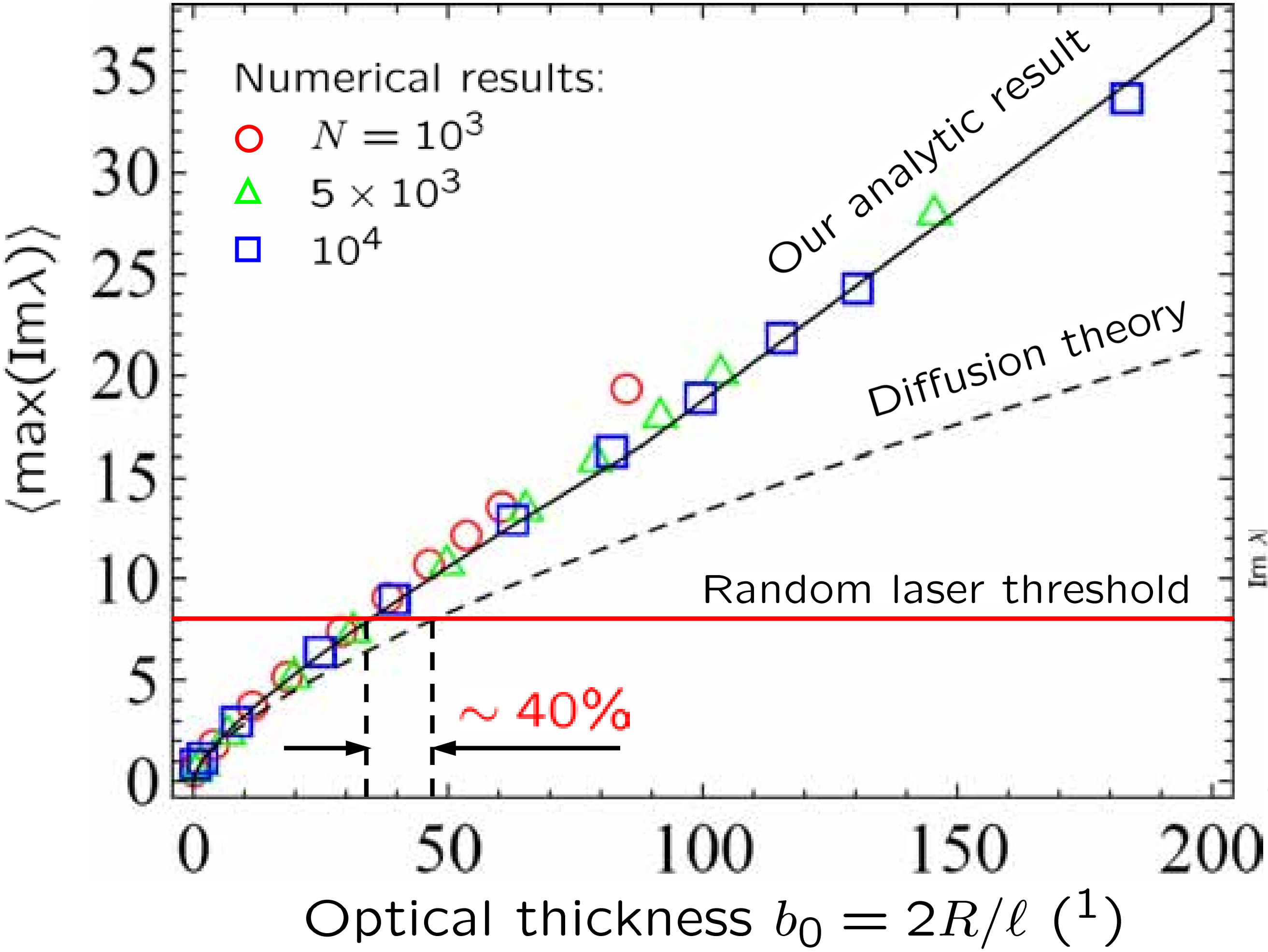
Eigenvalue domain from the diffusion theory

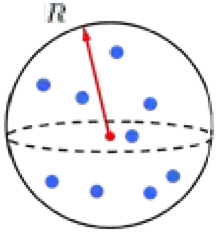


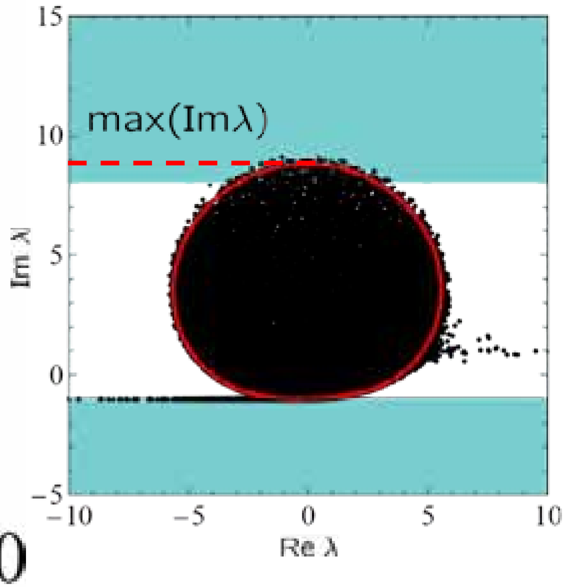
Borderline of the domain of existence of eigenvalues found from the diffusion theory

$N = 10^4$

Minimal optical thickness to lase







(^{1}) optical thickness at resonance,
in the absence of pump

Random laser beyond threshold

$$\widehat{G}\vec{\Psi}_n = \lambda_n \vec{\Psi}_n$$

Field: $\vec{\Omega} = \sum_n a_n(t)\vec{\Psi}_n e^{-\mathrm{i}\omega_n t}$

Intensity of n-th mode: $I_n(t) \propto |a_n(t)|^2$

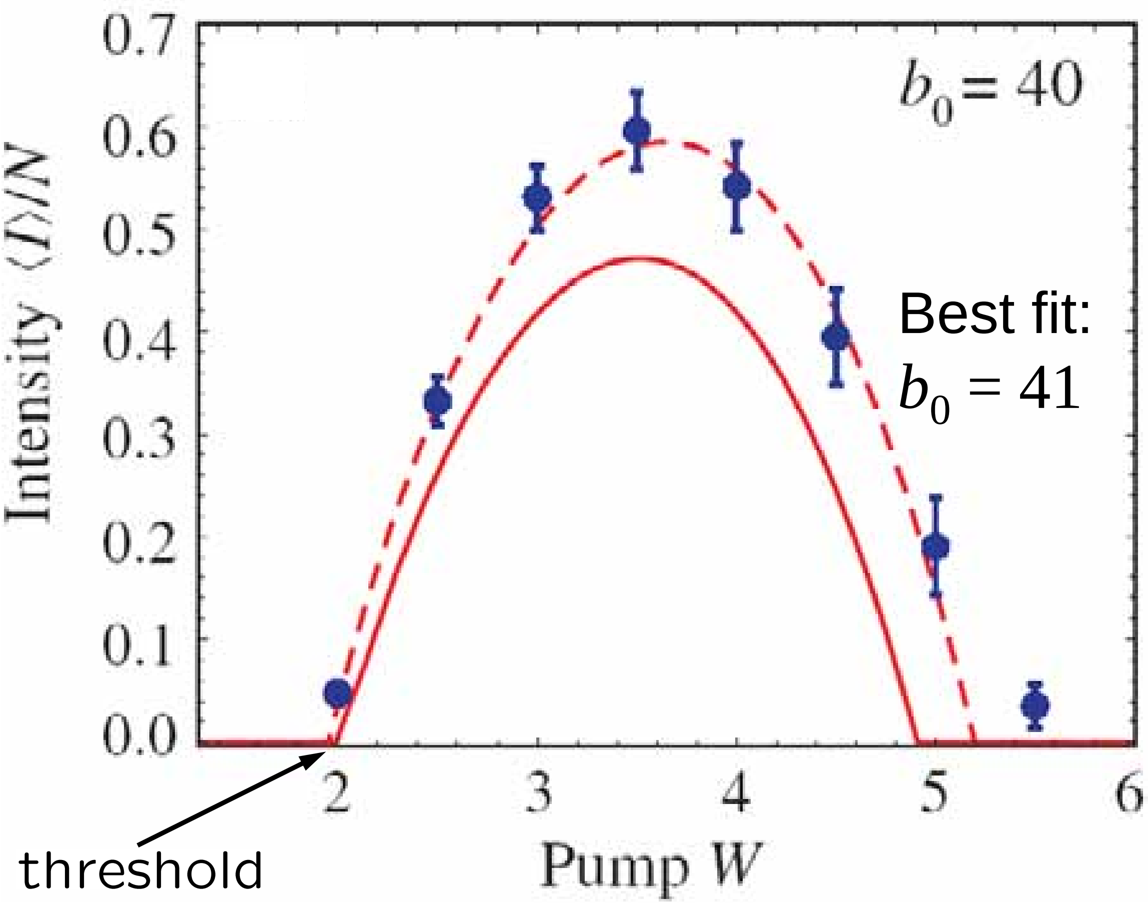
$$\frac{dI_n}{dt} = -2\kappa_n I_n + \sum_m \gamma_{nm} I_n I_m$$

$$\gamma_{nm} = 4\Gamma \frac{W-1}{(W+1)^3} \times \mathrm{Im}\left[\lambda_n \frac{\sum_{i=1}^N |\Psi_n^i|^2 (\Psi_m^i)^2}{\sum_{i=1}^N |\Psi_n^i|^2}\right]$$

$$\kappa_n = \frac{\Gamma}{2} \times \frac{W-1}{W+1}\left[\frac{(W+1)^2}{W-1} - \mathrm{Im}\lambda_n\right]$$

Total intensity: $I = \sum_n I_n$

Random laser beyond threshold



Best fit:
$b_0 = 41$

- ● numerical solution of coupled equations
- ─── analytical result
- - - - analytical result for a larger optical thickness

$$\text{Number of lasing modes} \propto \sqrt{\text{Number of modes beyond threshold}}$$

A universal result

Laser threshold:

$$\boxed{\lambda_n = \frac{1}{t}}$$

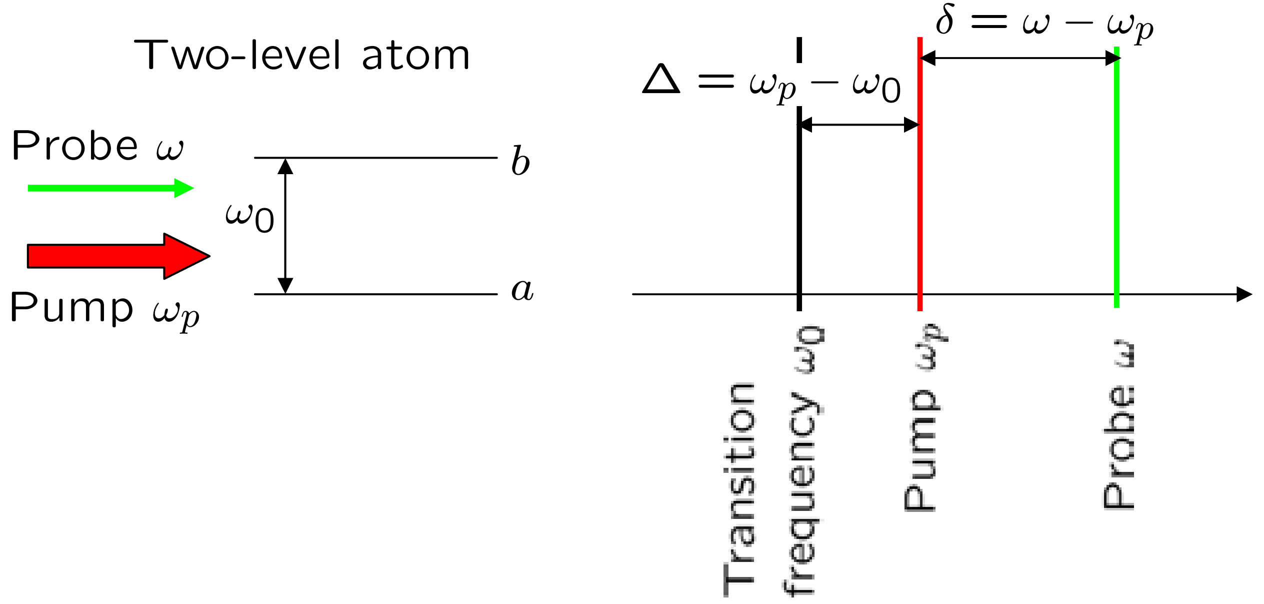
The dimensionless scattering matrix of a single scatterer (atom)

An eigenvalue of the matrix $\widehat{G}$:
$\widehat{G}\vec{\Psi}_n = \lambda_n \vec{\Psi}_n$

Recipe to find the laser threshold:

- Draw the domain of existence of complex eigenvalues λ of $\widehat{G}$ on the complex plane $z = \lambda$
- Draw the region spanned by $z = 1/t$ on the same complex plane
- Find the values of parameters for which the two above 2D regions touch

An even simpler laser: 2-level atoms



Dimensionless scattering matrix:

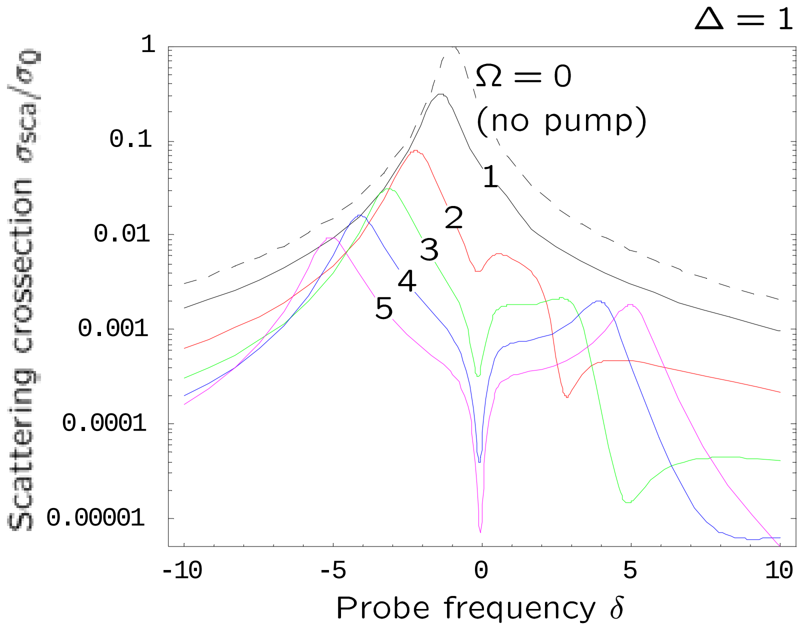
$$
\begin{aligned}
t(\Delta, \delta, \Omega) &= -\frac{1}{2} \frac{1+4\Delta^2}{1+4\Delta^2+2\Omega^2} \\
&\times \frac{(\delta+i)(\delta-\Delta+\frac{i}{2}) - \frac{1}{2}\Omega^2 \frac{\delta}{\Delta-\frac{i}{2}}}{(\delta+i)(\delta-\Delta+\frac{i}{2})(\delta+\Delta+\frac{i}{2}) - \Omega^2(\delta+\frac{i}{2})}
\end{aligned}
$$

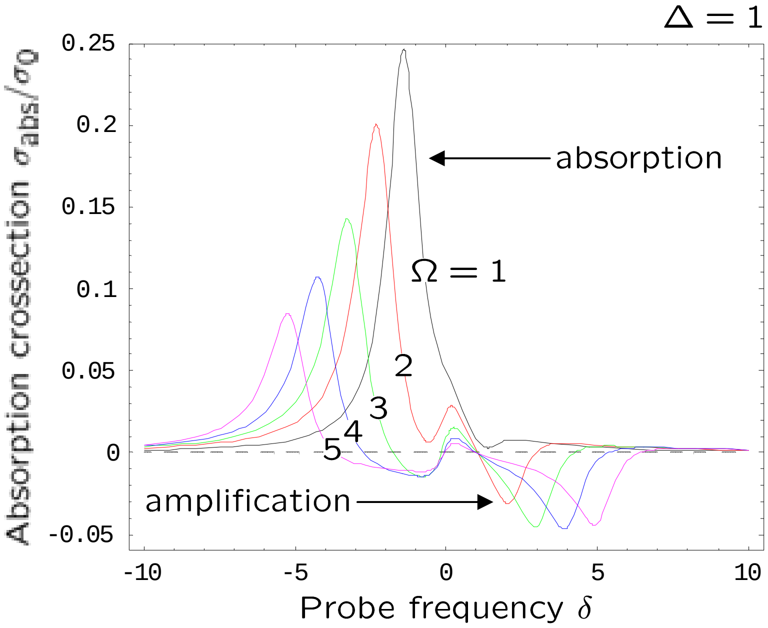
Δ, δ, Ω

in units of Γ

[B.R. Mollow, Phys. Rev. A 5, 2217 (1972)]

Scattering and amplification by a pumped 2-level atom



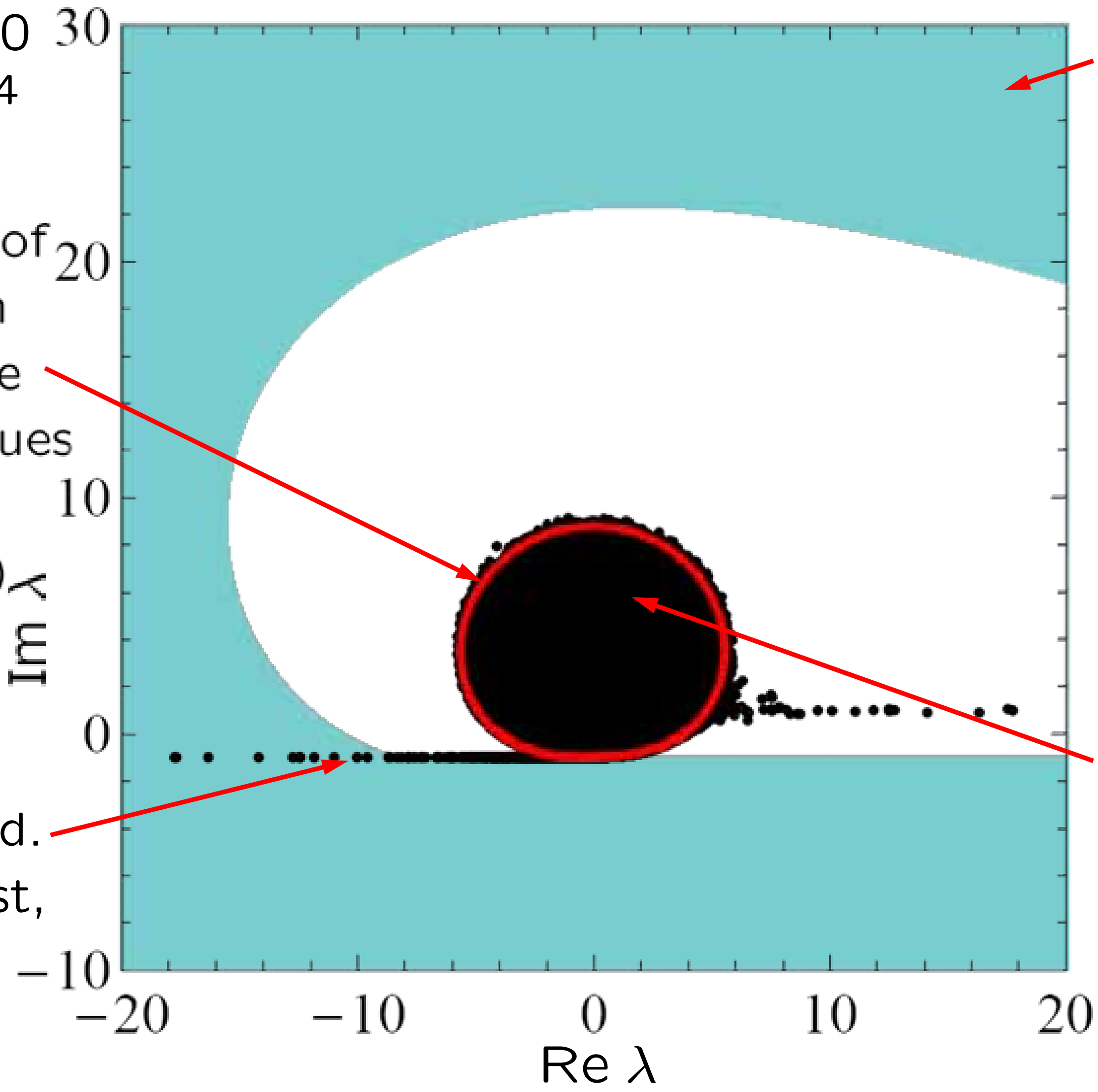


Mollow random laser at a fixed pump frequency

$\rho\lambda_0^3 = 10$

$N = 10^4$

$\Delta = 1$



Borderline of the domain of existence of eigenvalues of $\widehat{G}$ (analytical)

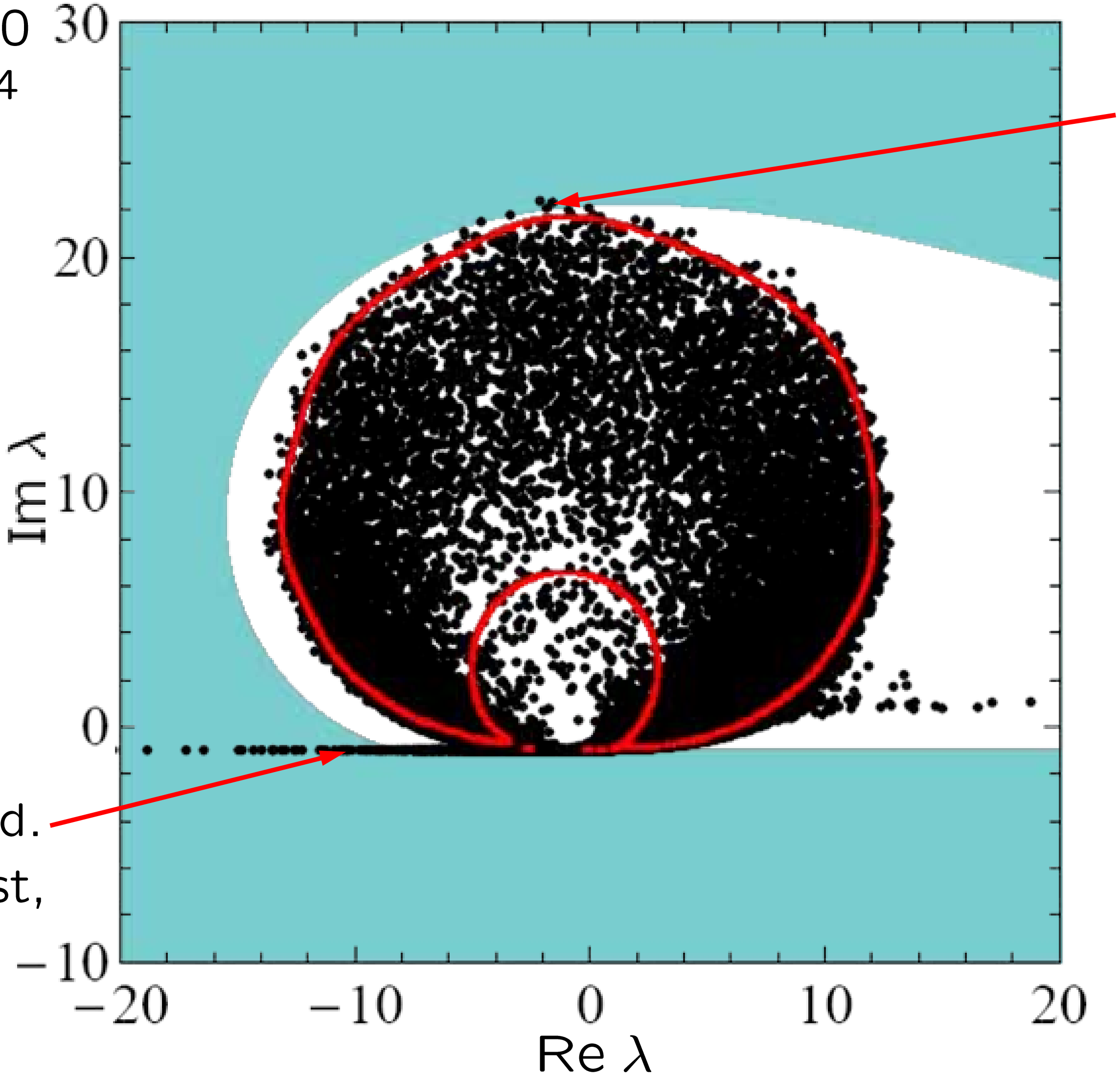
"Laser" of the 1st kind. Always exist, even for 2 atoms

Domain spanned by $1/t$ when δ and the pump Ω are varied

Eigenvalues of $\widehat{G}$ (numerical)

Mollow random laser at a fixed pump frequency

$\rho\lambda_0^3 = 50$
$N = 10^4$
$\Delta = 1$



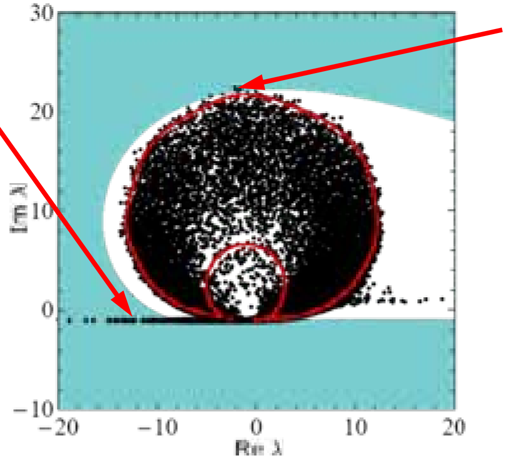
Laser of the 2nd kind. Requires sufficiently large optical thickness

"Laser" of the 1st kind. Always exist, even for 2 atoms

Two types of emission mechanisms

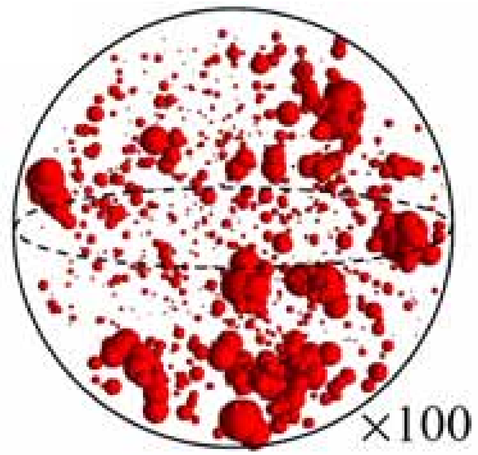
“Laser” of the 1st kind

- Emission by pairs of very closely situated atoms
- Not a true collective effect
- Does not require many atoms: also exists for 2 atoms
- Typical mode localized on 2 atoms:

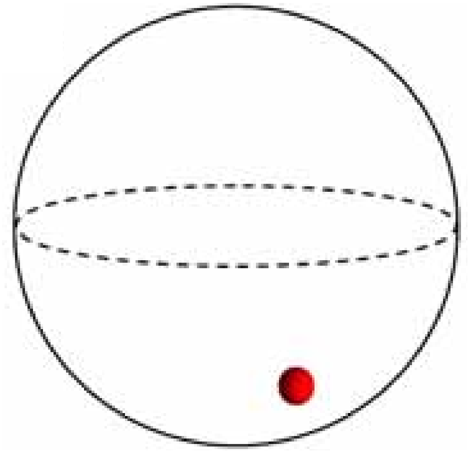


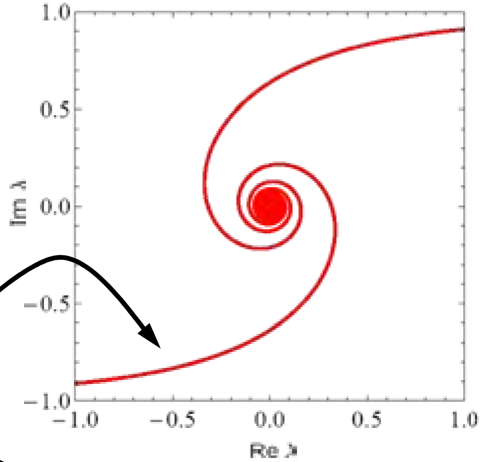
Laser of the 2nd kind

- Emission by all atoms
- A true collective effect
- Requires multiple scattering by many atoms
- Typical mode delocalized:



This is a true random laser



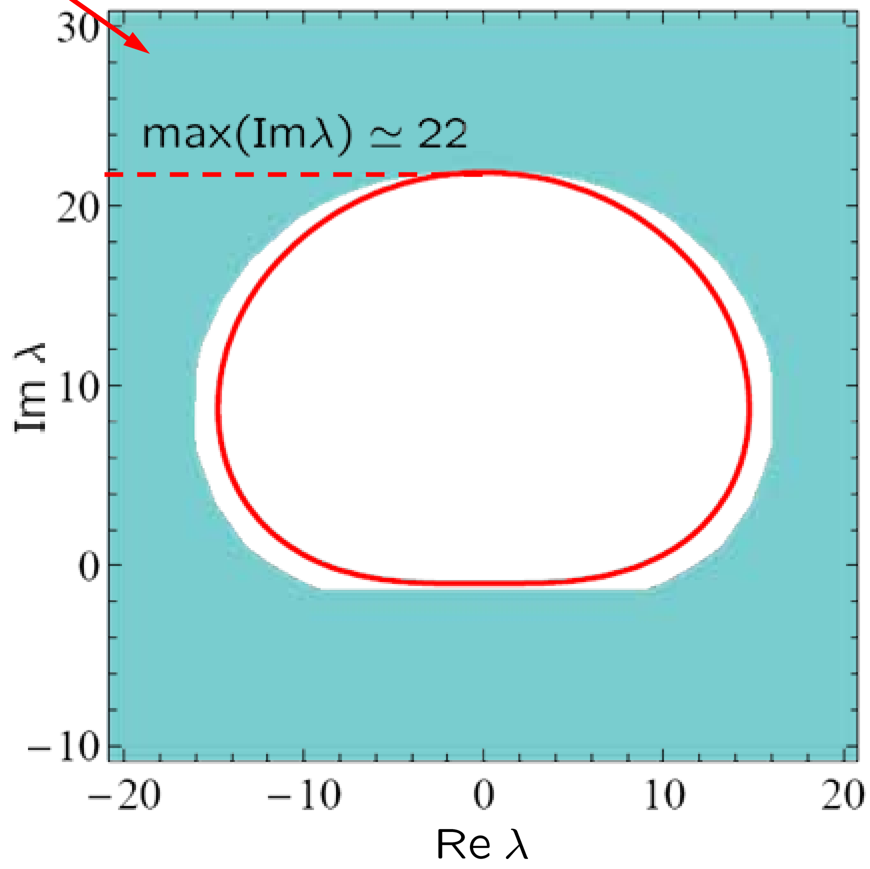


Long-lived sub-radiant states

Is the Mollow random laser realizable ?

Domain spanned by $1/t$ when δ, Δ and the pump Ω are varied

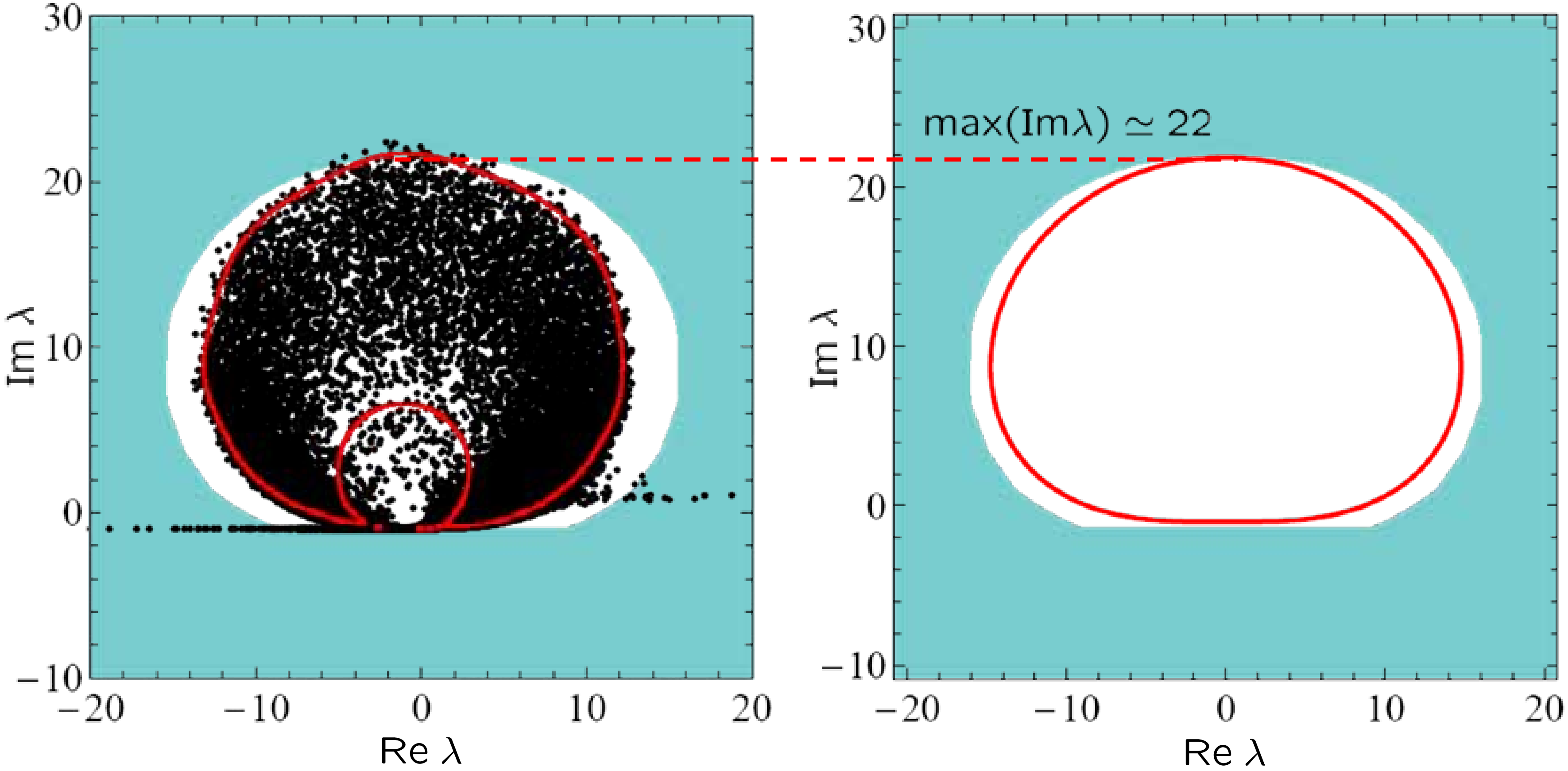
Diffusion theory



Minimal optical thickness needed to realize random lasing:

$$\boxed{b_0 \simeq 200}$$

Is the Mollow random laser realizable ?



Our Euclidean random matrix theory

Diffusion theory

Minimal optical thickness needed to realize random lasing:

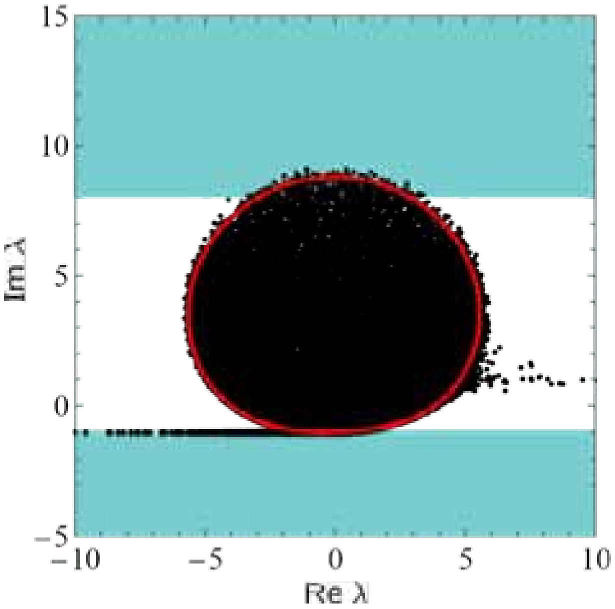
$\boxed{b_0 \simeq 110}$ $\boxed{b_0 \simeq 200}$

Why does the diffusion theory not apply?

3-level atoms

Threshold reached for

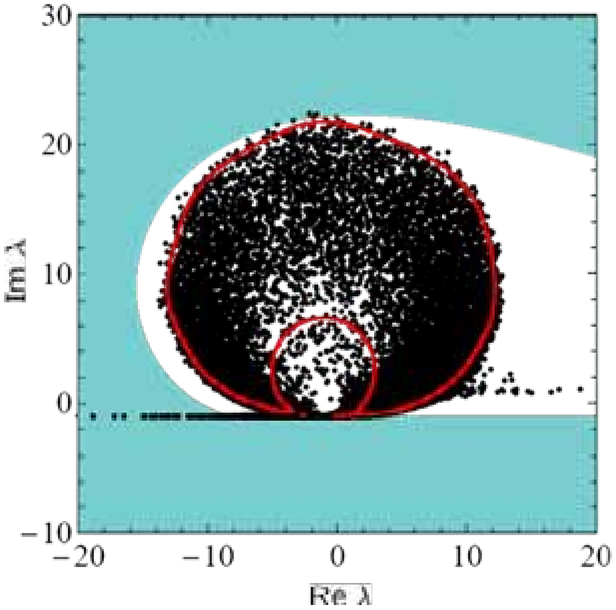
$b_0 \simeq 35,\ 1/t \simeq 8\mathrm{i}$



2-level atoms

Threshold reached for

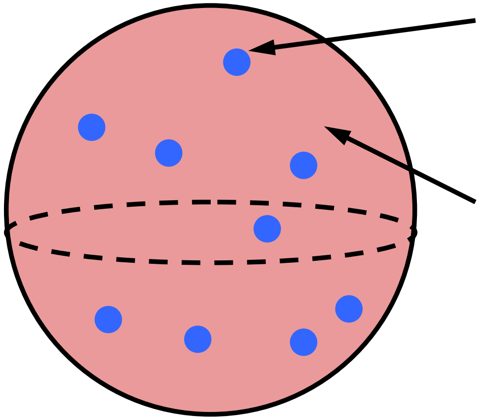
$b_0 \simeq 110,\ 1/t \simeq 22\mathrm{i}$



The actual optical thickness at threshold

$$b = b_0 |t|^2 < 1$$

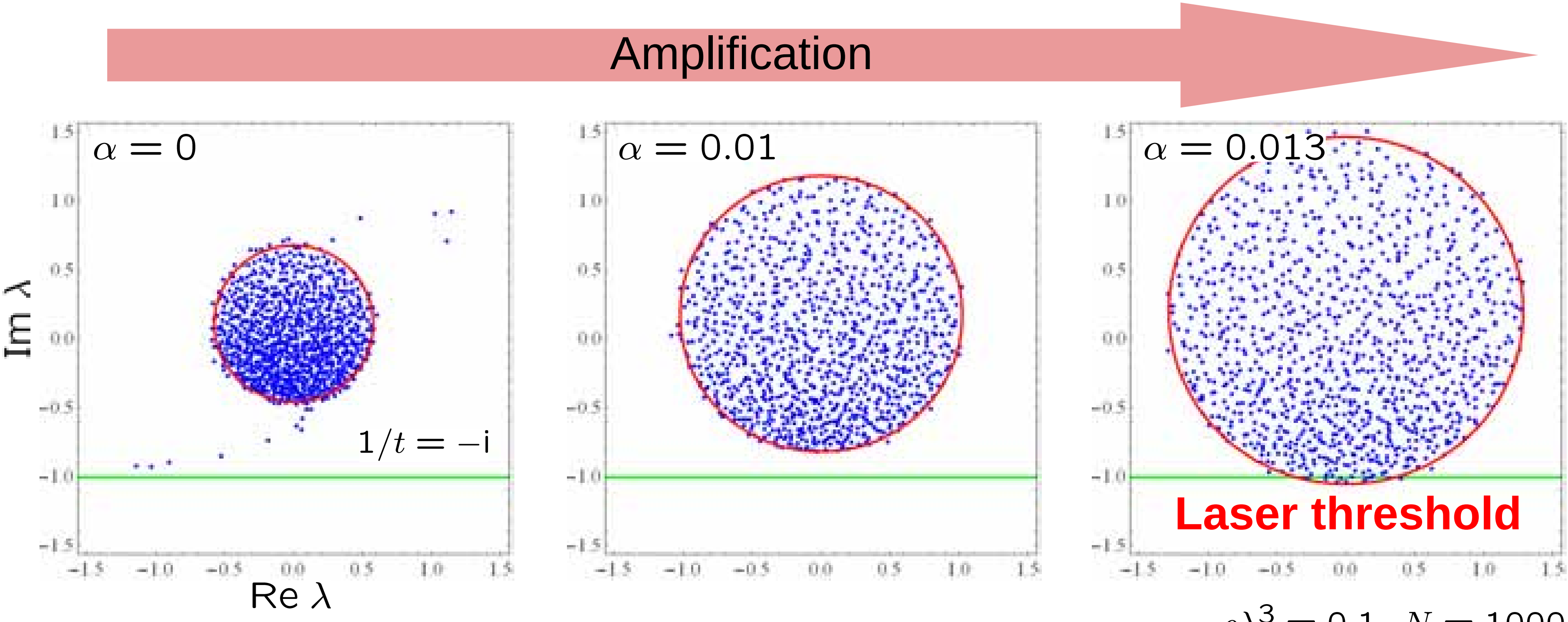
Application to a more “conventional” random laser



t-matrix:
$1/t = -\mathrm{i}$
(optical theorem)

Green’s matrix

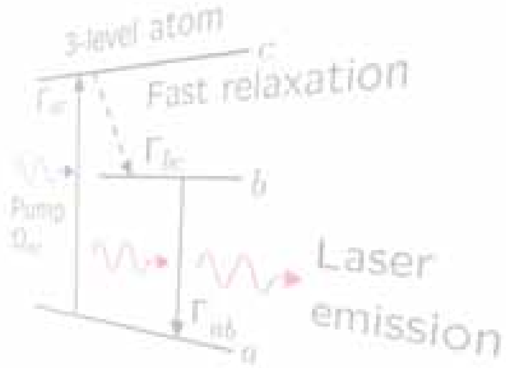
$$G_{ij} = \frac{e^{(\mathrm{i}+\alpha)k_0|\mathbf{r}_i-\mathbf{r}_j|}}{k_0|\mathbf{r}_i-\mathbf{r}_j|}$$



$\rho\lambda_0^3 = 0.1,\ N = 1000$

Conclusions

- Finding the domain of existence of eigenvalues of the Green’s matrix is mathematically equivalent to finding the threshold of a random laser
- In a cloud of 3-level atoms, random lasing should be realizable starting from optical thickness b_0 ~ 35 (instead of 50 in diffusion theory)
- In a cloud of 2-level atoms, Mollow random laser should be realizable starting from optical thickness b_0 ~ 110 (instead of 200 in diffusion theory)
- Euclidean matrix approach applies to passive scatterers in an amplifying matrix as well
- Much remains to be done: lasing beyond threshold, quantum statistics, etc.



Find details in our papers:

- *Physical Review E* **84**, 011150 (2011)
- *Europhysics Letters* **96**, 34005 (2011)

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Announcements

CNRS research group

Méso Im@ge GDR 3219

is organizing
a special session
“Wave propagation in disordered media”
(JMC13, Montpellier, August 27-31, 2012)

... and a workshop
“Recent developments in wave propagation
and imaging in complex media”
(IHP, Paris, November 7-9, 2012)

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