Experiments on Random Lasers

Claudio Conti

Institute for Complex Systems

National Research Council

People

- Neda Ghofraniha
- Silvia Gentilini
- Marco Leonetti
- Viola Folli
- Giancarlo Ruocco
- Luca Leuzzi
- Luca Angelani
- Francesco Zamponi
- Andrea Puglisi
- Andrea Crisanti
- Ilenia Viola
- F. Di Maria
- G. Barbarella
- G. Gigli
- Luca La Volpe
- Fabrizio Antenucci
- Cefe Lopez
- Cordt Zollfrank

John Templeton Foundation

Outline

- Complexity and spin glasses
- Lasers and Random lasers
- Observation of a Photonic Spin Glass

Complexity

Many years ago…

- My first meeting with complexity

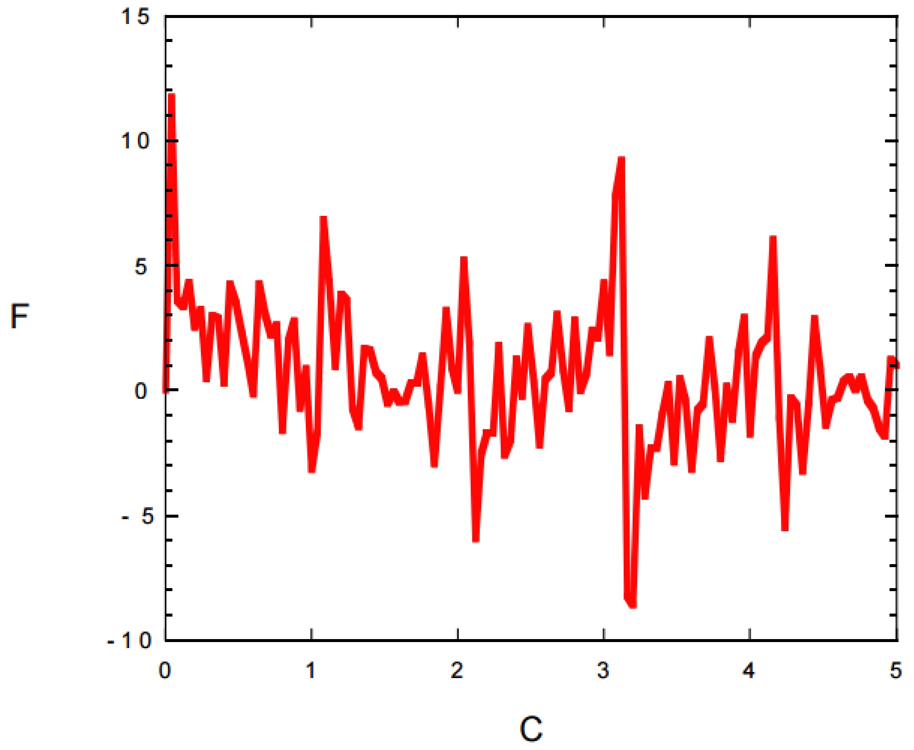
The physical Meaning of Replica Symmetry Breaking

Giorgio Parisi
Dipartimento di Fisica, Sezione INFN and unità INFM,
Università di Roma "La Sapienza",
Piazzale Aldo Moro 2, I-00185 Roma (Italy)
giorgio.parisi@roma1.infn.it

(Dirac medal talk, **arXiv:cond-mat/0205387v1**)

$$H = -\sum_{i,k} J_{i,k}\sigma_i\sigma_k - \sum_i h_i\sigma_i$$

The Landscape



$$w_\alpha \propto \exp(-\beta F_\alpha)$$

Figure 1: An artistic view of the free energy of a complex system as function of the configuration space.

(Parisi, arXiv:cond-mat/0205387v1)

The overlap and the P(q)

$$q[\sigma,\tau] = \frac{1}{N}\sum_{i=1,N}\sigma(i)\tau(i) \ .$$

We define $P_J(q)$ as probability distribution of the overlap q at given J, i.e. the histogram of $q[\sigma,\tau]$ where σ and τ are two equilibrium configurations. Using eq. (2), one finds that

$$P_J(q) = \sum_{\alpha,\gamma} w_\alpha w_\gamma \delta(q - q_{\alpha,\gamma}) \ , \qquad (6)$$

where in a finite volume system the delta functions are smoothed. If there is more than one state, $P_J(q)$ is not a single delta function

$$P_J(q) \neq \delta(q - q_{EA}) \ . \qquad (7)$$

If this happens we say that the replica symmetry is broken: two identical replicas of the same system may stay in a quite different state.

(Parisi, arXiv:cond-mat/0205387v1)

P(q) for different J

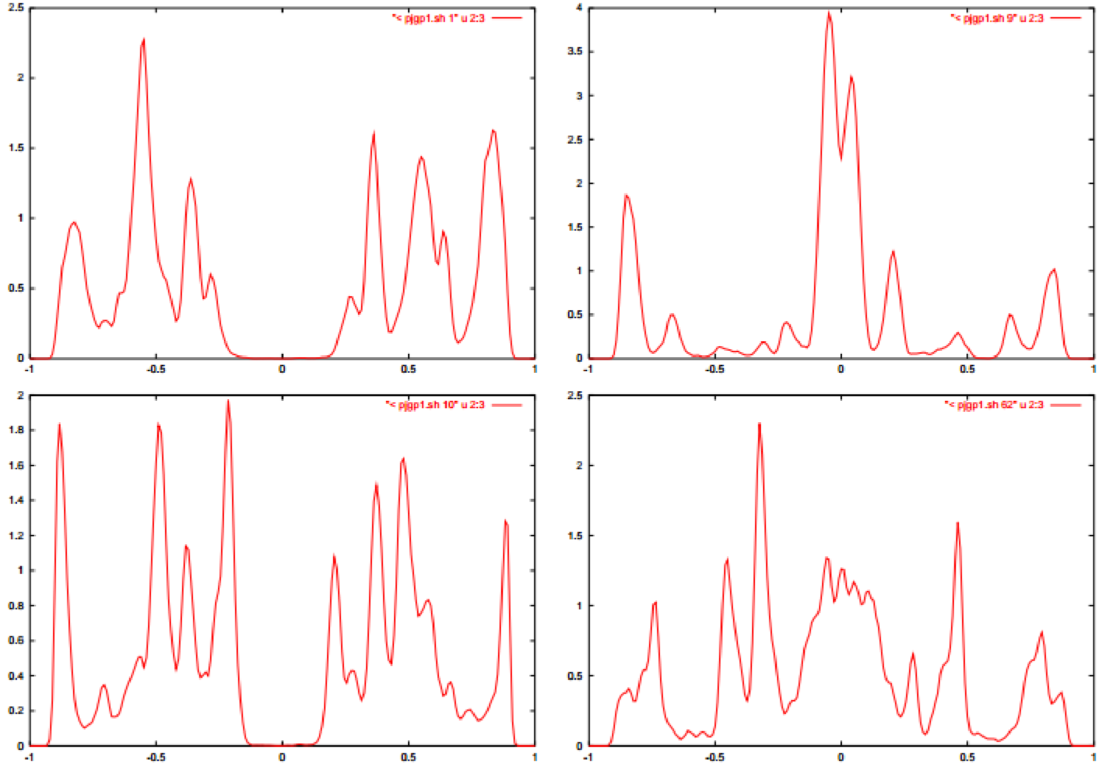


Figure 2: The function $P_J(q)$ for four different samples (i.e different choices of J) for $D = 3$ $L = 16$ (16^3 spins).

(Parisi, arXiv:cond-mat/0205387v1)

Disorder average P(q)

$$P(q) = \overline{P_J(q)} \ .$$

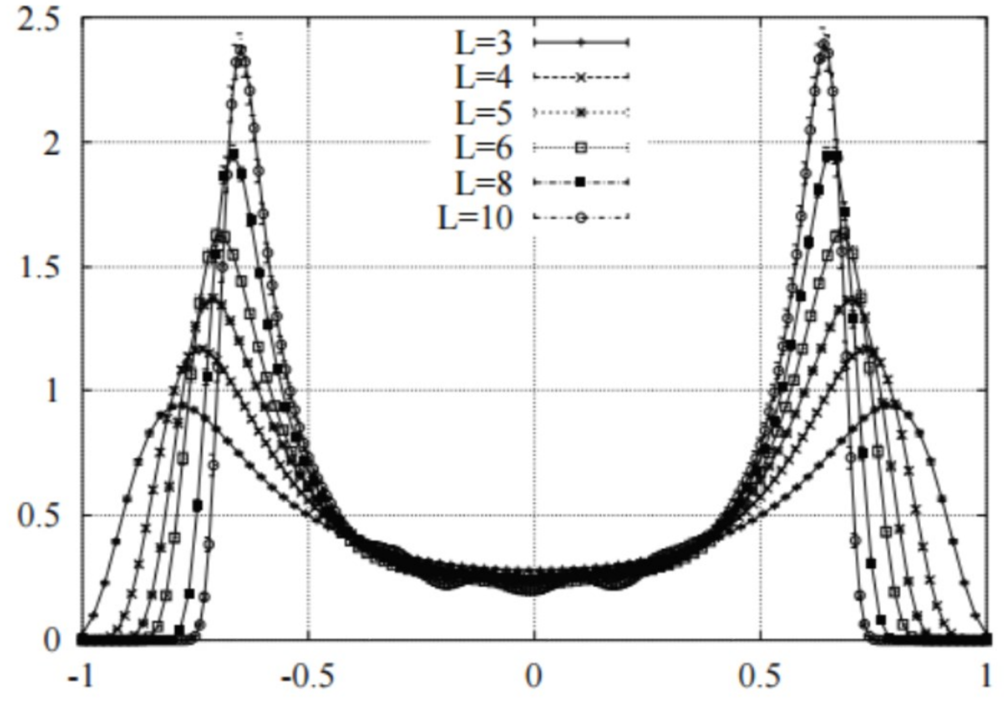
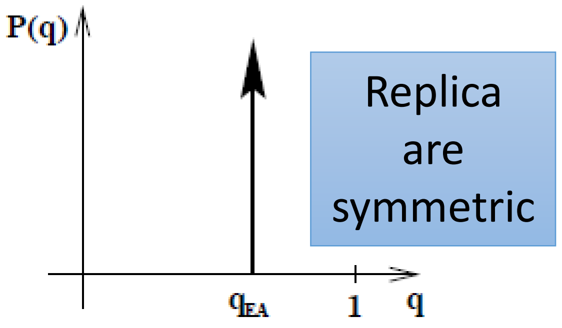
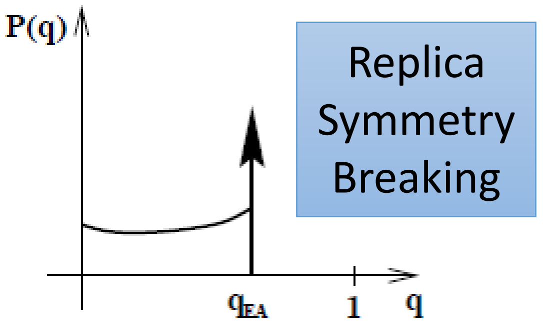


Figure 3: The function $P(q) = \overline{P_J(q)}$ after average over many samples (D=4, L=3...10) .

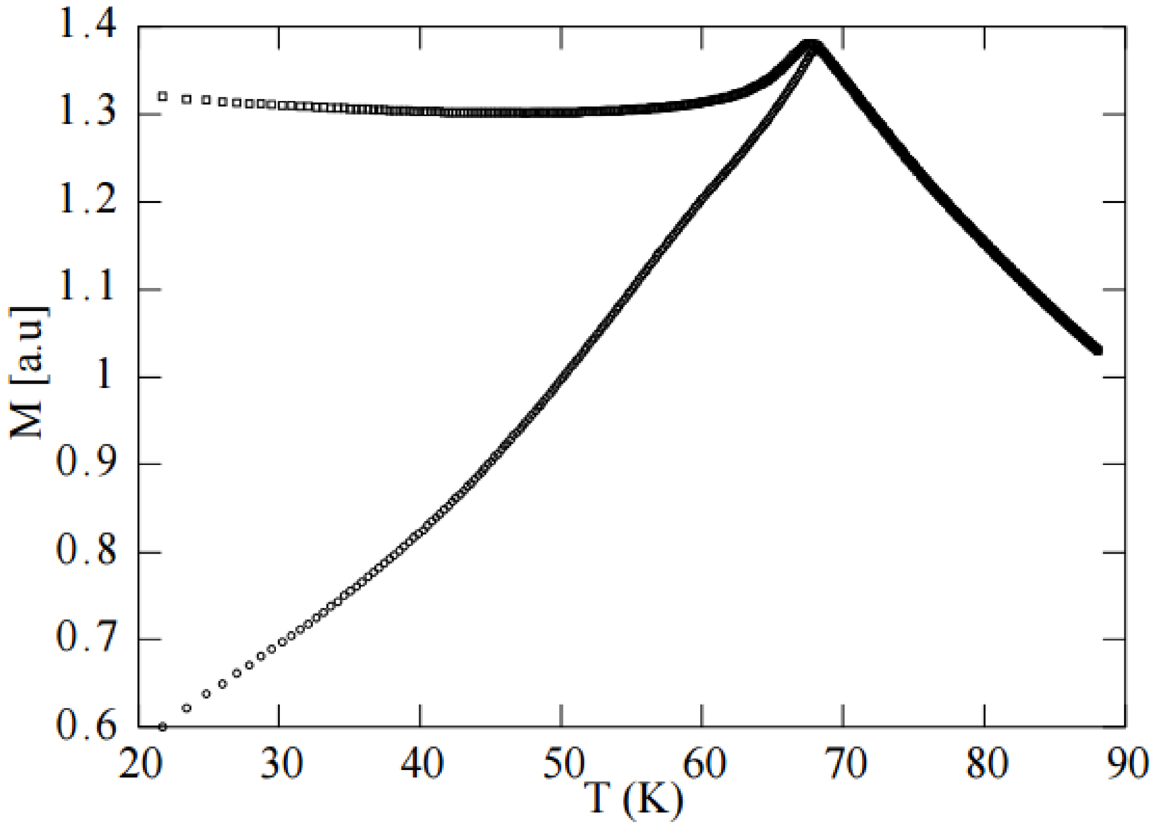




(Parisi, arXiv:cond-mat/0205387v1)

Experimental evidence

Magnetization Vs Temperature



$$\chi_{eq} = \beta \int dq \ P(q)(1-q)$$

(Parisi, arXiv:cond-mat/0205387v1)

Questions

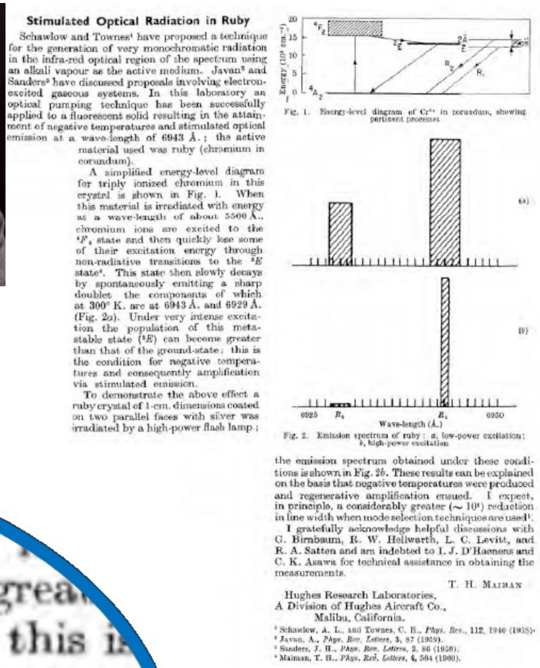
- Photonic spin-glass?
- Direct measurement of P(q) ?
- Replica Symmetry Breaking ?

Why lasers?

- Lasers are known for being thermodynamic systems
  - Black body radiation
  - Negative temperature

- «Random lasers» are disordered systems

Maiman, 1960



Stimulated Optical Radiation in Ruby

Schawlow and Townes[1] have proposed a technique for the generation of very monochromatic radiation in the infra-red optical region of the spectrum using an alkali vapour as the active medium. Javan[2] and Sanders[3] have discussed proposals involving electron-excited gaseous systems. In this laboratory an optical pumping technique has been successfully applied to a fluorescent solid resulting in the attainment of negative temperatures and stimulated optical emission at a wave-length of 6943 Å.; the active material used was ruby (chromium in corundum).

A simplified energy-level diagram for triply ionized chromium in this crystal is shown in Fig. 1. When this material is irradiated with energy at a wave-length of about 5500 Å., chromium ions are excited to the $^4F_2$ state and then quickly lose some of their excitation energy through non-radiative transitions to the ^2E state[4]. This state then slowly decays by spontaneously emitting a sharp doublet the components of which at 300° K. are at 6943 Å. and 6929 Å. (Fig. 2a). Under very intense excitation the population of this metastable state (^2E) can become greater than that of the ground-state; this is the condition for negative temperatures and consequently amplification via stimulated emission.

To demonstrate the above effect a ruby crystal of 1-cm. dimensions coated on two parallel faces with silver was irradiated by a high-power flash lamp;

Fig. 1. Energy-level diagram of Cr^{3+} in corundum, showing pertinent processes

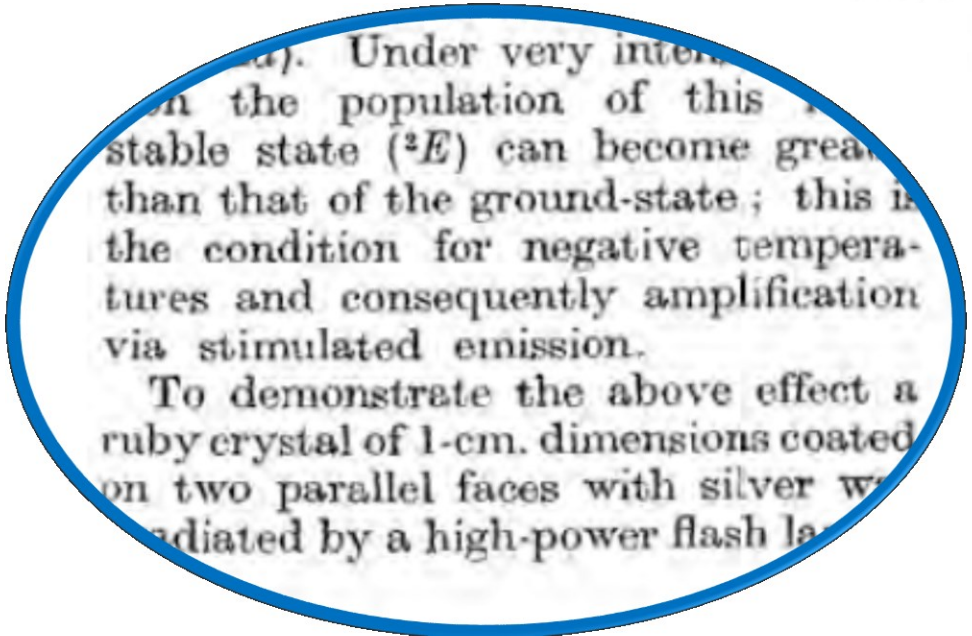
Fig. 2. Emission spectrum of ruby: a, low-power excitation; b, high-power excitation

the emission spectrum obtained under these conditions is shown in Fig. 2b. These results can be explained on the basis that negative temperatures were produced and regenerative amplification ensued. I expect, in principle, a considerably greater (~ 10^4) reduction in line width when mode selection techniques are used[1].

I gratefully acknowledge helpful discussions with G. Birnbaum, R. W. Hellwarth, L. C. Levitt, and R. A. Satten and am indebted to I. J. D'Haenens and C. K. Asawa for technical assistance in obtaining the measurements.

T. H. Maiman
Hughes Research Laboratories,
A Division of Hughes Aircraft Co.,
Malibu, California.

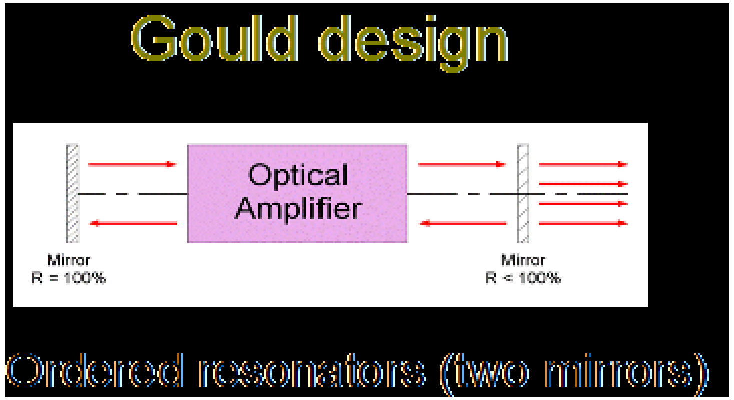
[1] Schawlow, A. L., and Townes, C. H., *Phys. Rev.*, 112, 1940 (1958).
[2] Javan, A., *Phys. Rev. Letters*, 3, 87 (1959).
[3] Sanders, J. H., *Phys. Rev. Letters*, 3, 86 (1959).
[4] Maiman, T. H., *Phys. Rev. Letters*, 4, 564 (1960).

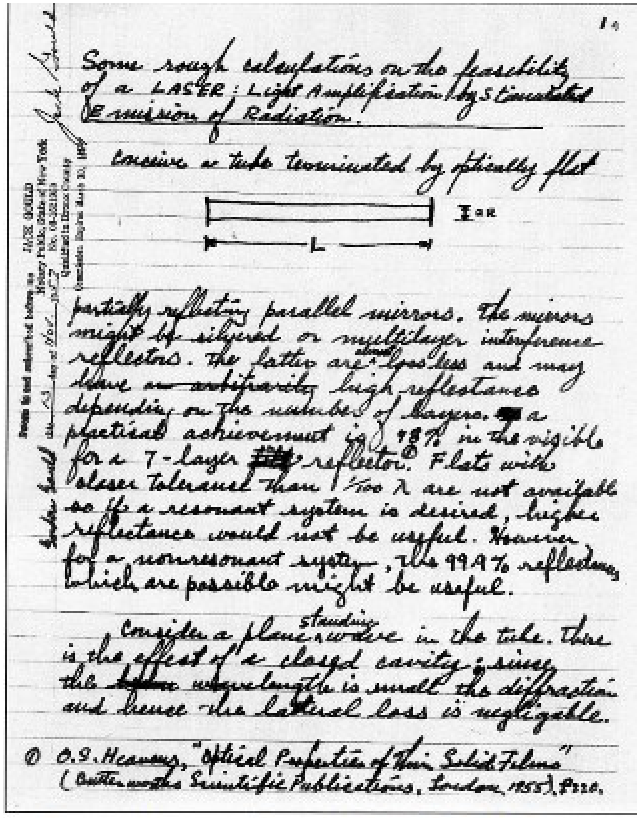


Lasers

- Light amplification by stimulated emission of radiation

- The original laser model:
  - Gould, Prokhorov, Schawlow, Townes
  - Invented the open cavity design (1957)




Some rough calculations on the feasibility of a LASER: Light Amplification by Stimulated Emission of Radiation.

Conceive a tube terminated by optically flat partially reflecting parallel mirrors. The mirrors might be silvered or multilayer interference reflectors. The latter are almost loss-less and may have an arbitrarily high reflectance depending on the number of layers. A practical achievement is 98% in the visible for a 7-layer reflector. Flats with closer tolerance than 1/100 λ are not available so if a resonant system is desired, higher reflectance would not be useful. However for a nonresonant system, the 99.9% reflectance which are possible might be useful.

Consider a plane standing wave in the tube. There is the effect of a closed cavity; since the wavelength is small the diffraction and hence the lateral loss is negligible.

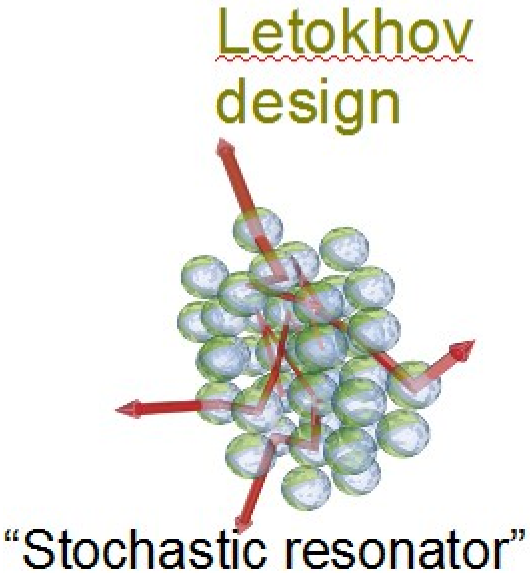
① O.S. Heavens, "Optical Properties of Thin Solid Films" (Butterworths Scientific Publications, London, 1955), p.220.

Gordon Gould 1957
(source:wikipedia)

Random lasers

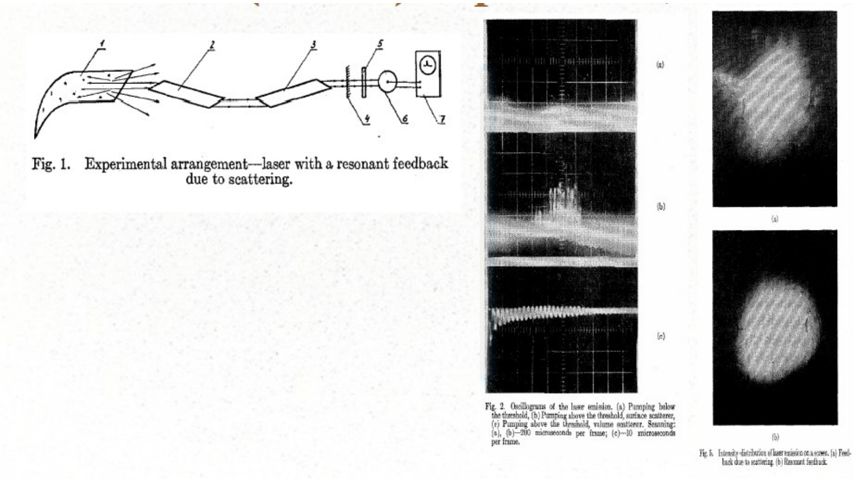
- A new design for lasers, dating back to 1966
- The photonic bomb

VS Letokhov



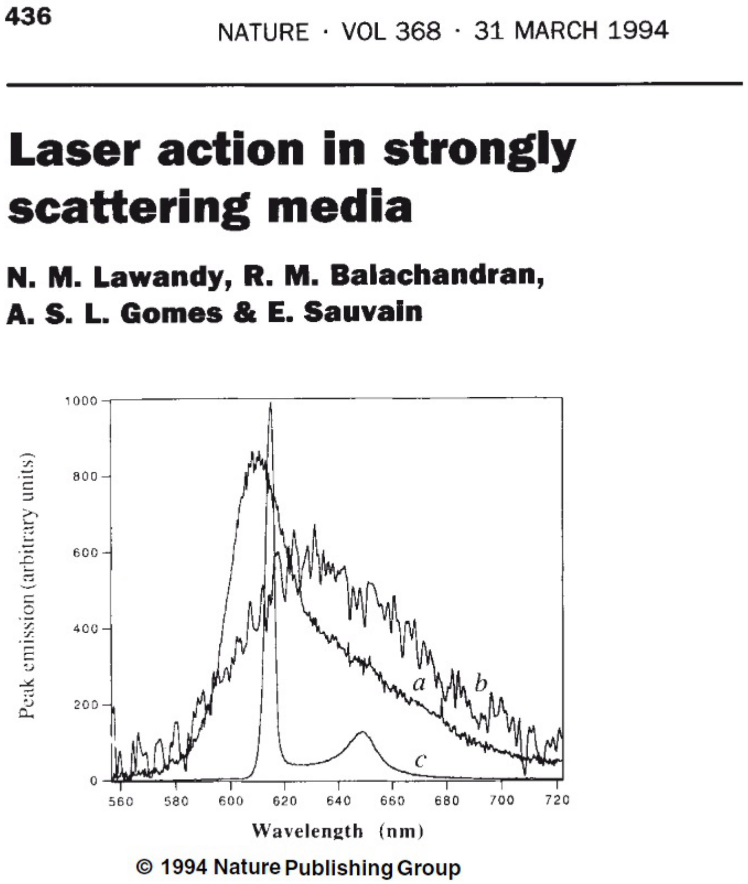
Several different random lasers

The first one



Ambartsumyam, Basov, Kryukov, Lethokov (ABKL) experiment, 1966

The most famous



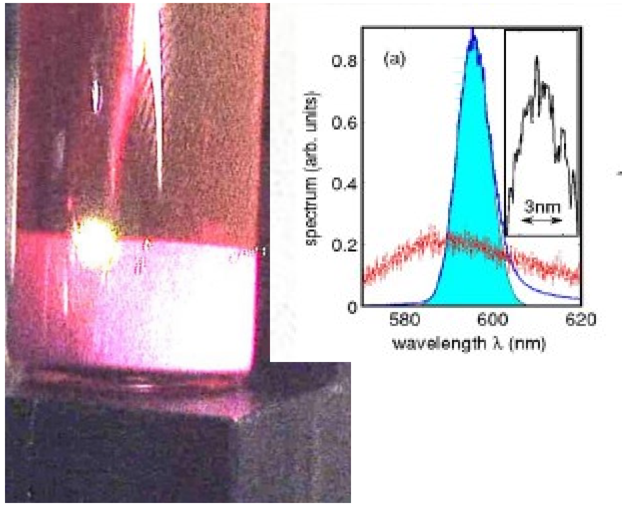
436 NATURE · VOL 368 · 31 MARCH 1994

Laser action in strongly scattering media

N. M. Lawandy, R. M. Balachandran, A. S. L. Gomes & E. Sauvain

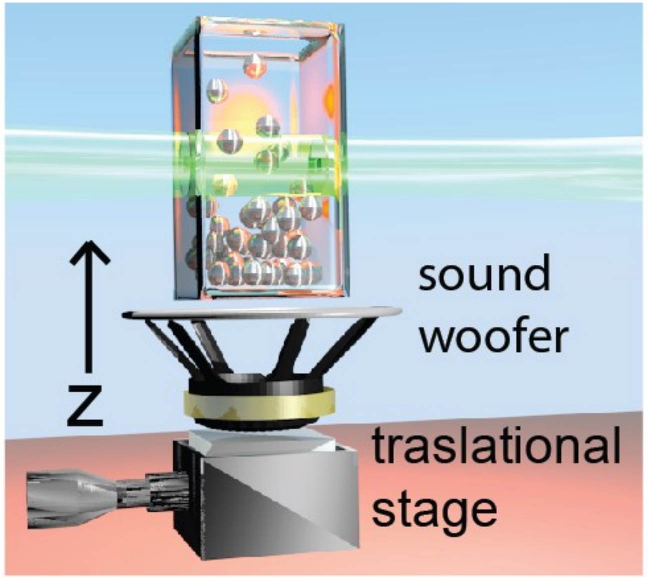
© 1994 Nature Publishing Group

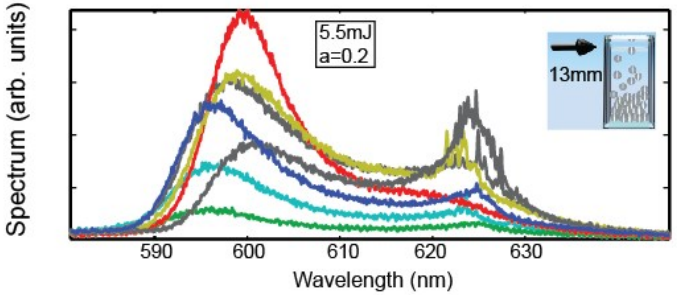
Our first one

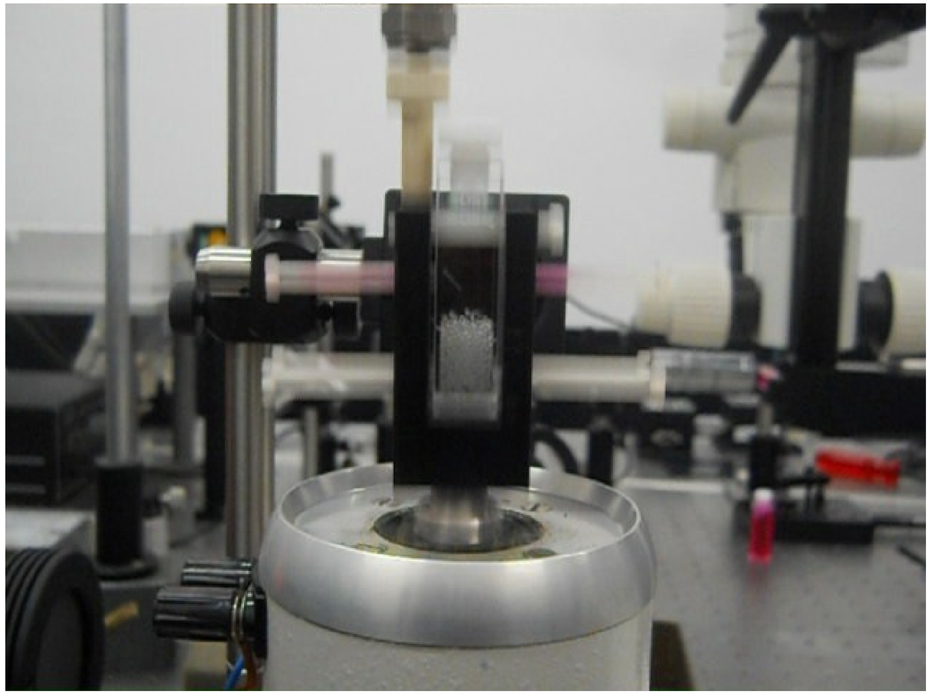


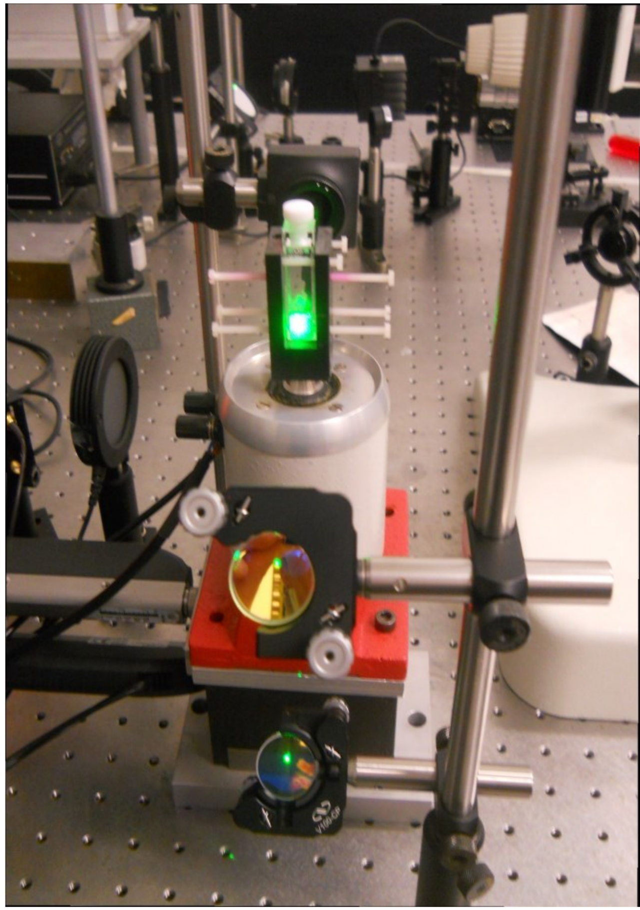
CC et al,
PRL 101, 143901 (2008)

Shaken granular lasers





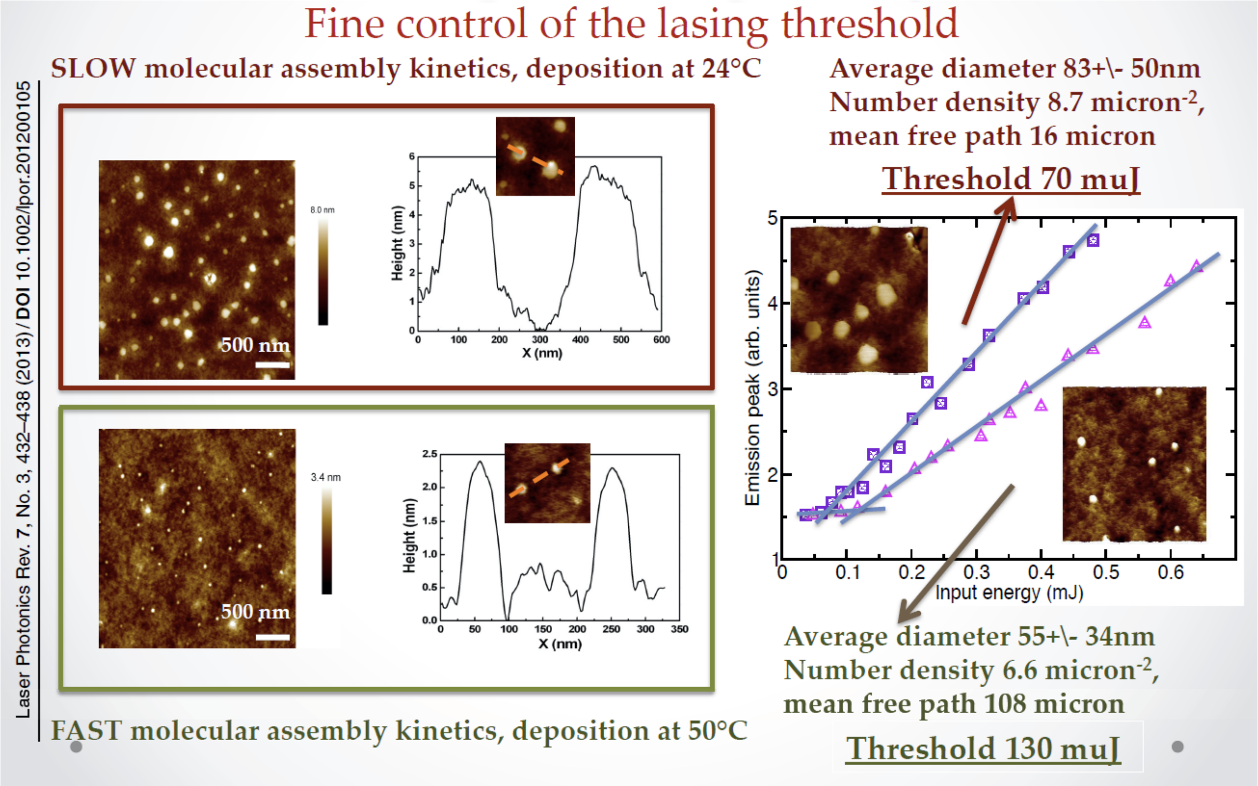


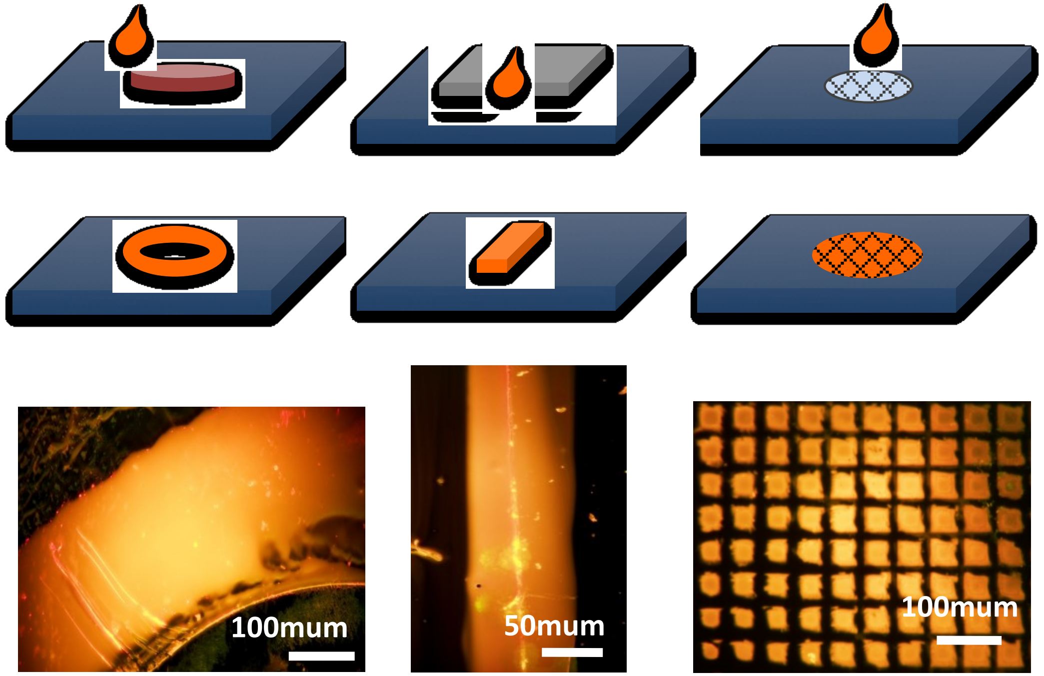


PRL 2012,
Scientific Reports 2013

Laser with soft matter

- Colloidal dye

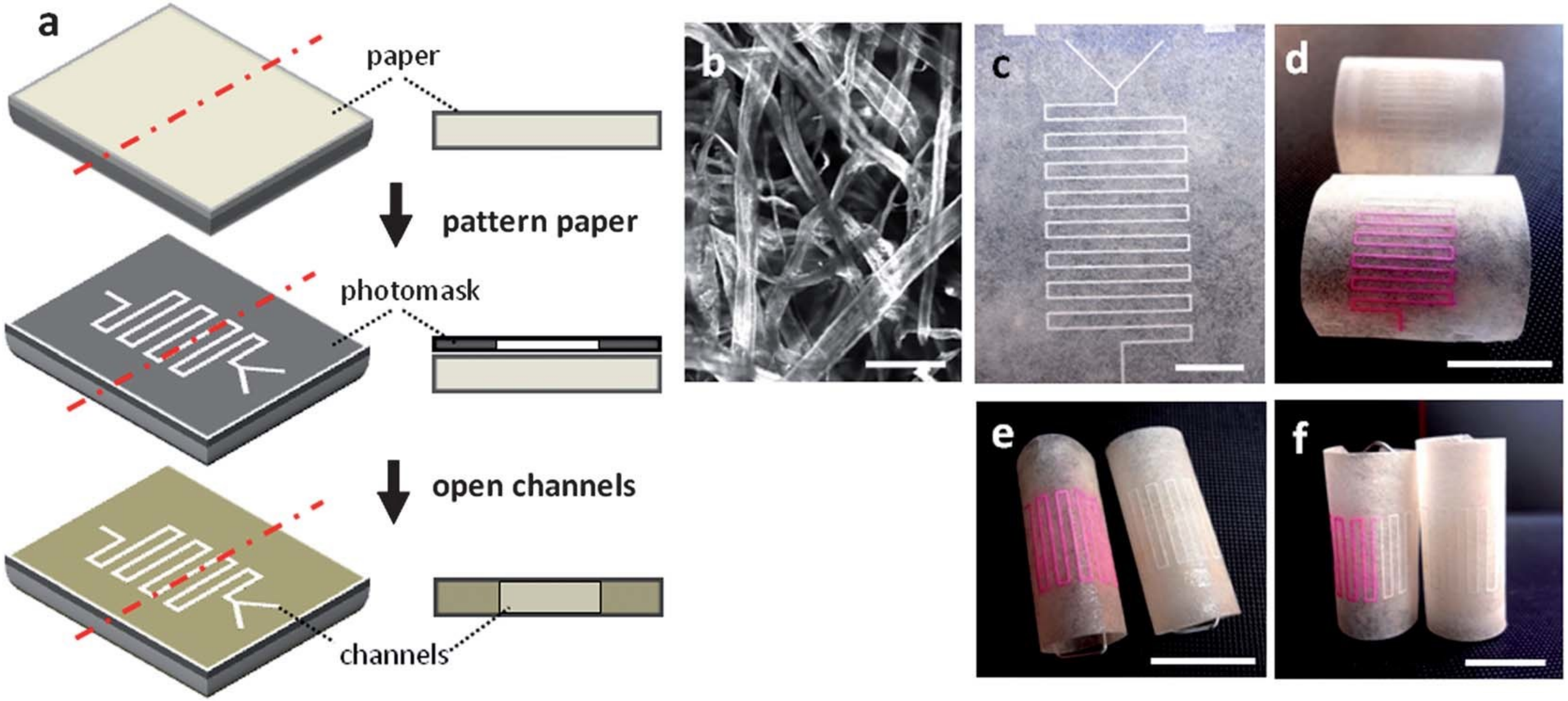




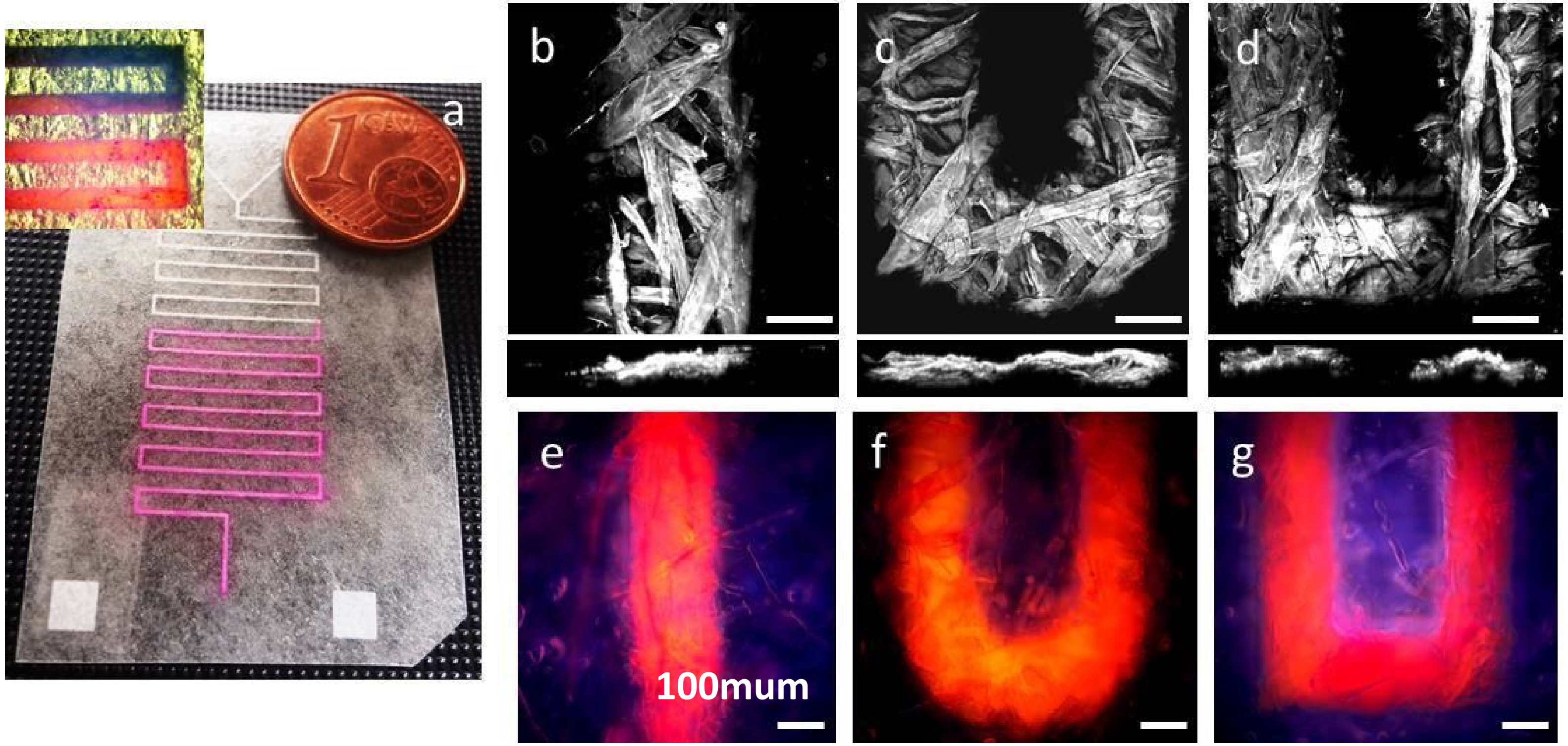
Laser Photonics Rev. **7,** No. 3, 432–438 (2013) / **DOI** 10.1002/lpor.201200105

Experimental results
Paper-based microchannels

Fabrication by I. Viola, A. Zacheo, V. Arima and G. Gigli from Nano-CNR in Lecce

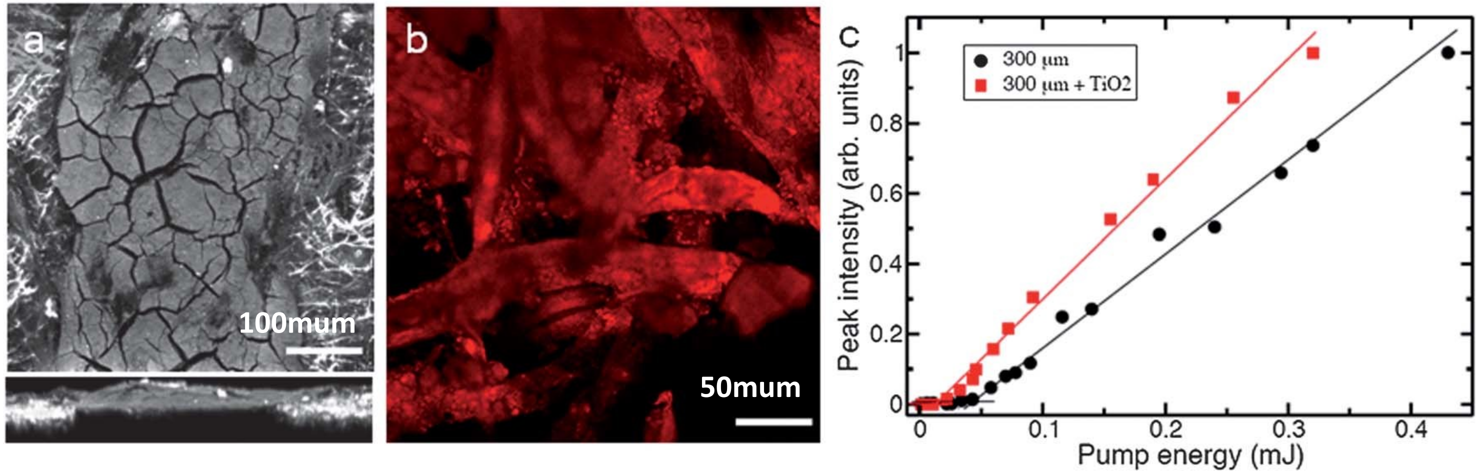


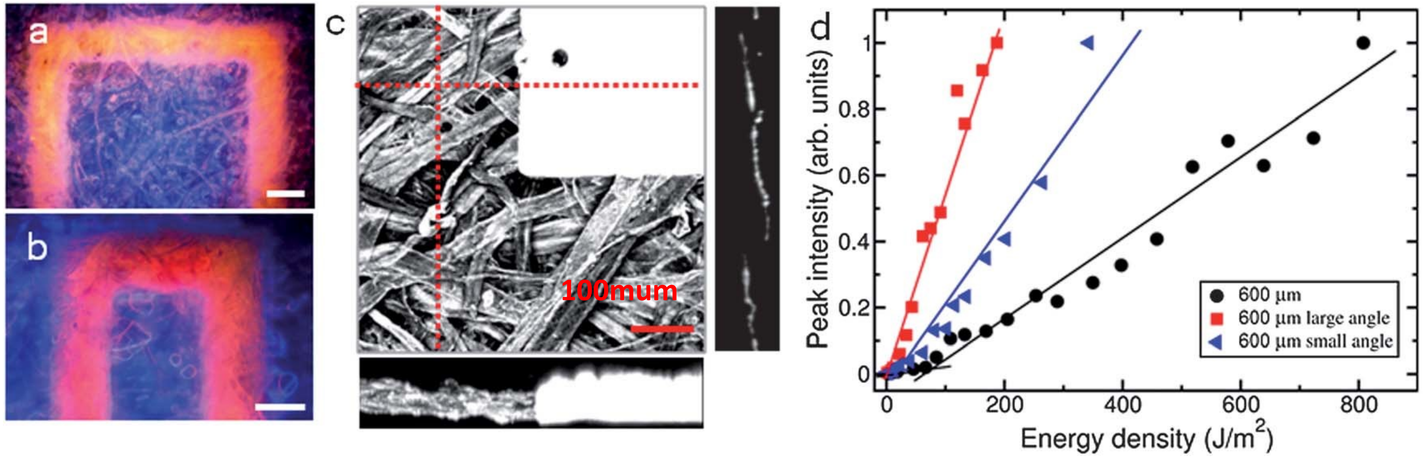
Experimental results
Paper-based microchannels



The lasing circuits are realized by using, firstly standard soft-lithography techniques to define the channel geometry, then imbibing with a lasing dye (RhB) through the channel inlet and, finally, fulfilling the channels by capillary processes.

Experimental results
Paper-based microchannels

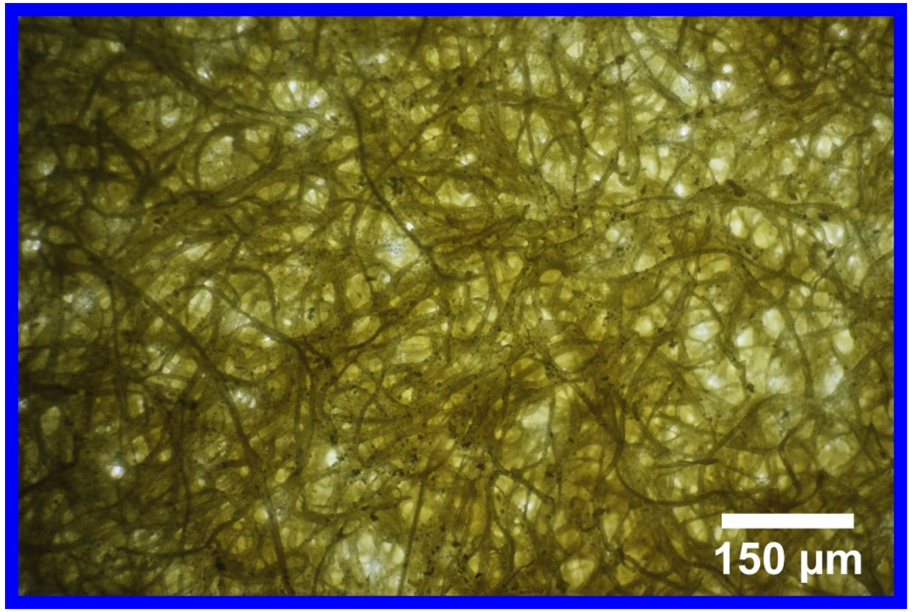


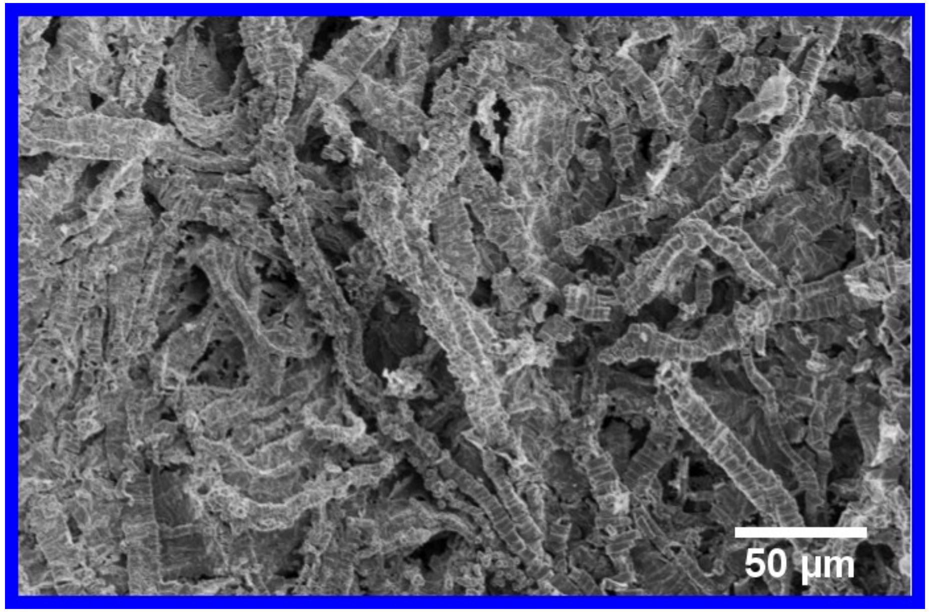


J. Mater. Chem. C, 2013, **1**, 8128–8133

Experimental results
Bio-templated titanium

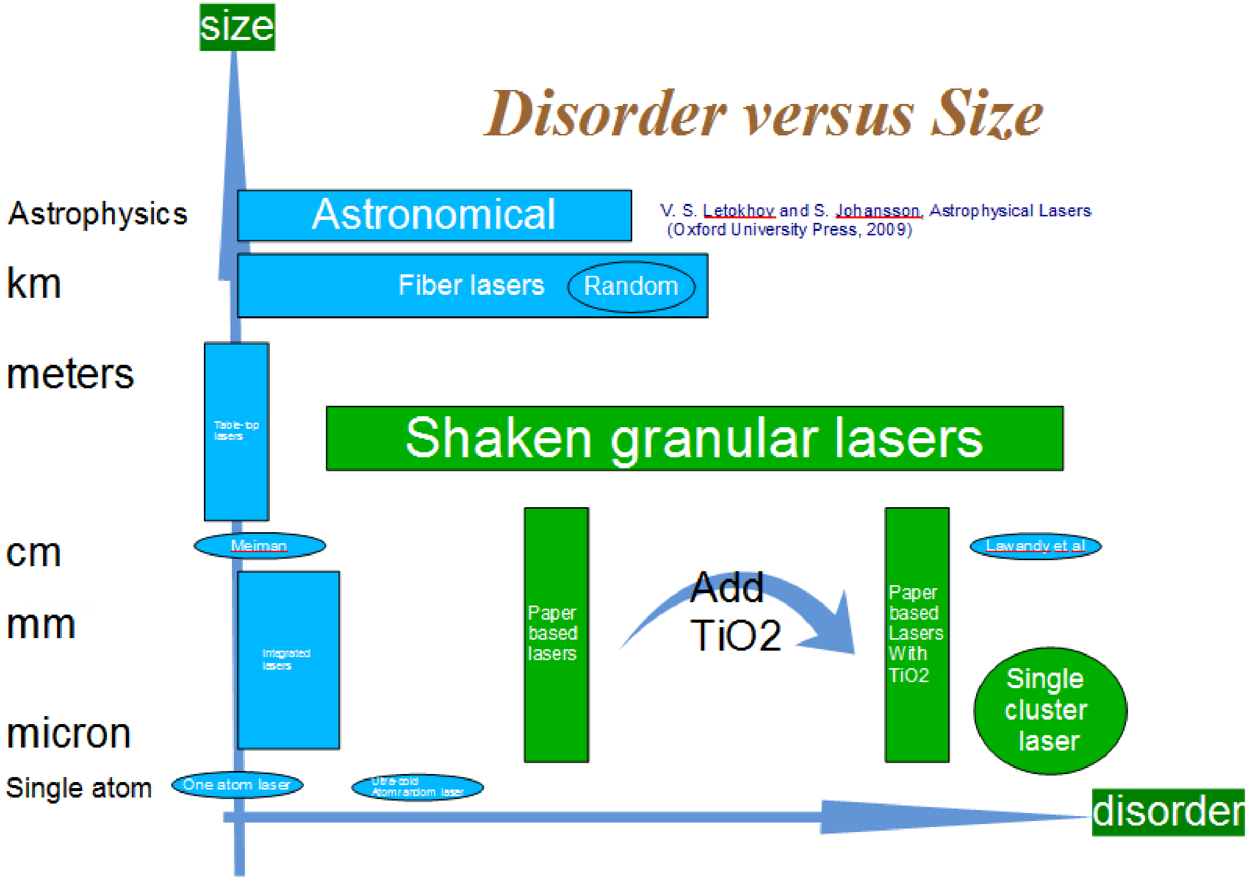
TiO_2 from cellulose templates:
Titanium with the structure of paper



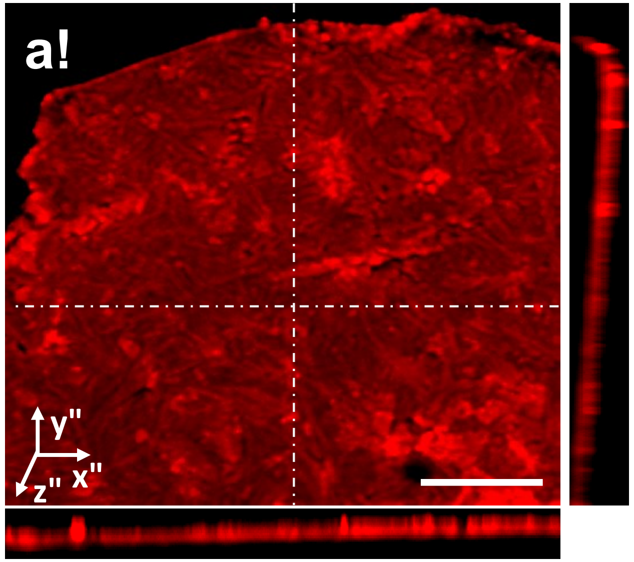


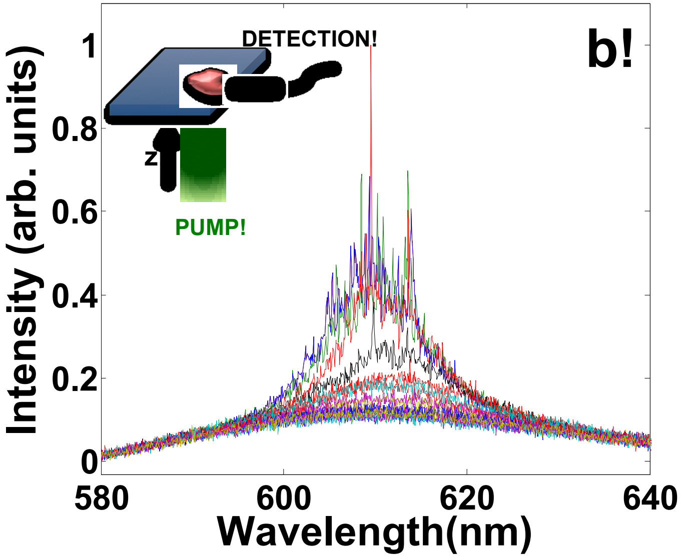
http://www.professoren.tum.de/en/zollfrank-cordt/

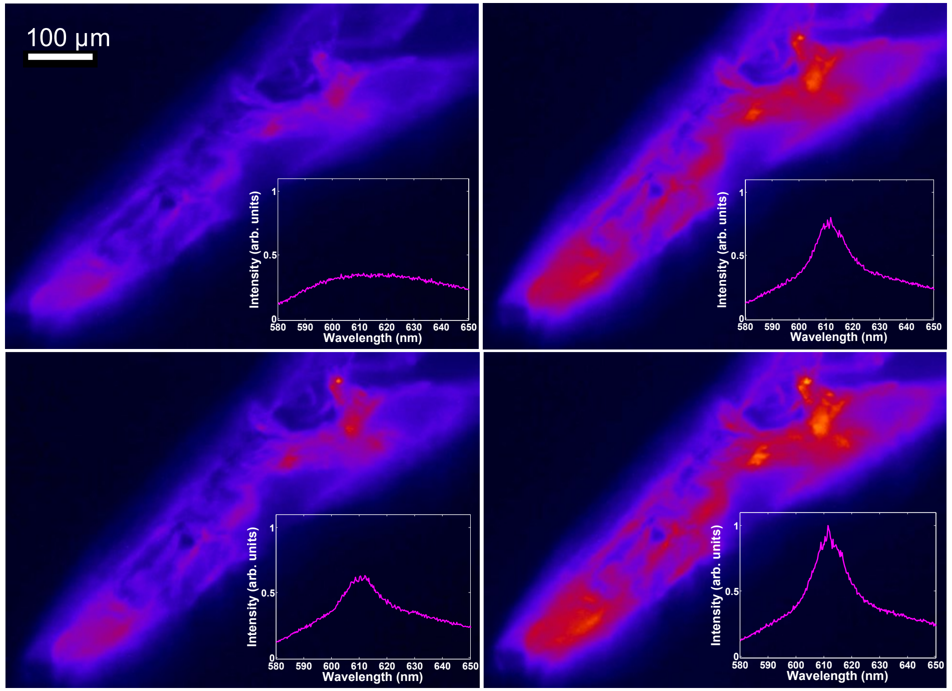
Disorder Vs Size



First hints about a possible RSB







The Hamiltonian for Random Lasers

$$\mathcal{H} = -\Re\left[\sum_{j<k} G^{(2)}_{jk}\, a_j a_k^* + \sum_{\omega_j+\omega_k=\omega_l+\omega_m} G^{(4)}_{jklm}\, a_j a_k a_l^* a_m^*\right]$$

Glassy behavior of light

L. Angelani, C. Conti, G. Ruocco, F. Zamponi

(Submitted on 17 Nov 2005)

We study the nonlinear dynamics of a multi-mode random laser using the methods of statistical physics of disordered systems. A replica-symmetry breaking phase transition is predicted as a function of the pump intensity. We thus show that light propagating in a random non-linear medium displays glassy behavior, i.e. the photon gas has a multitude of metastable states and a non vanishing complexity, corresponding to mode-locking processes in random lasers. The present work reveals the existence of new physical phenomena, and demonstrates how nonlinear optics and random lasers can be a benchmark for the modern theory of complex systems and glasses.

Comments:	5 pages, 1 figure
Subjects:	**Disordered Systems and Neural Networks (cond-mat.dis-nn)**; Statistical Mechanics (cond-mat.stat-mech); Optics (physics.optics)
Journal reference:	Phys. Rev. Lett. 96, 065702 (2006)
DOI:	10.1103/PhysRevLett.96.065702
Cite as:	**arXiv:cond-mat/0511427 [cond-mat.dis-nn]**
	(or **arXiv:cond-mat/0511427v1 [cond-mat.dis-nn]** for this version)

Problems

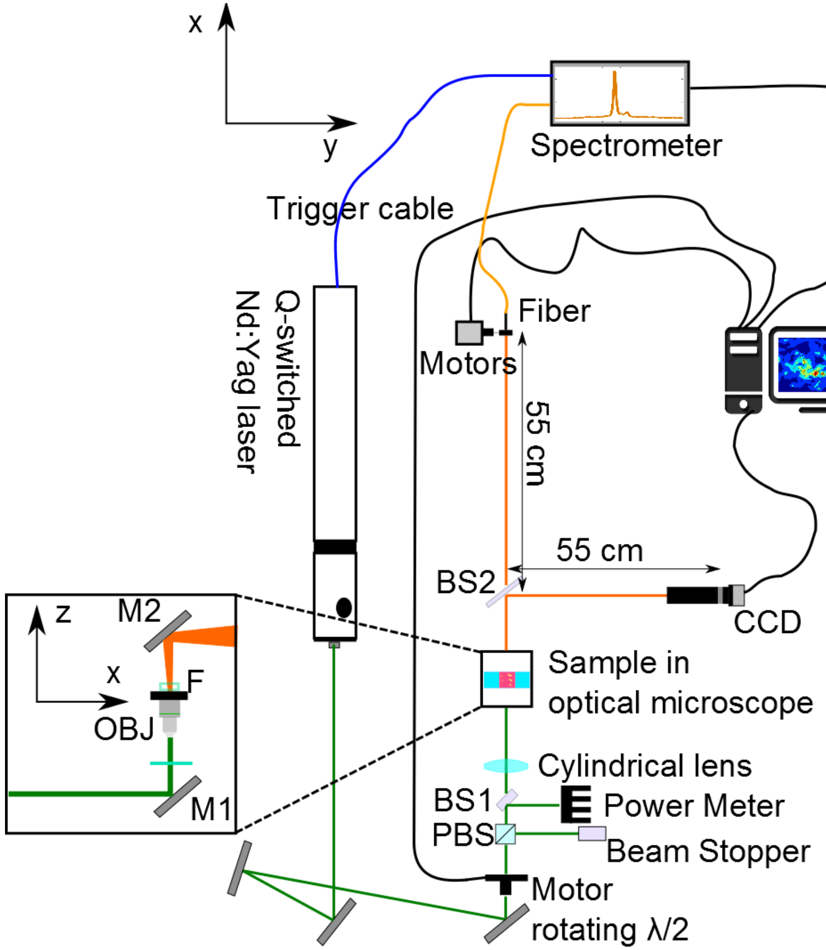
- Is it true that random lasers have modes?
- Do random laser modes interact?

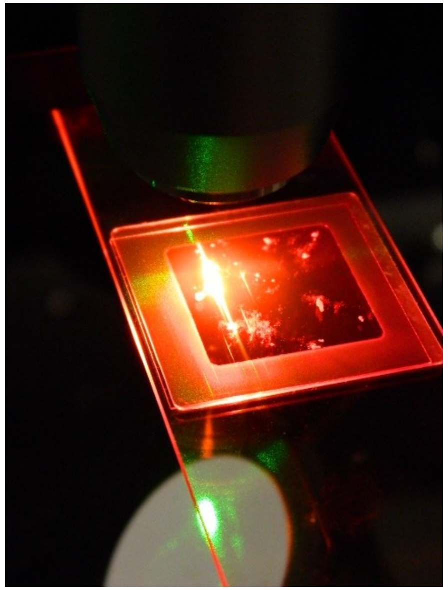
- What can we measure ?

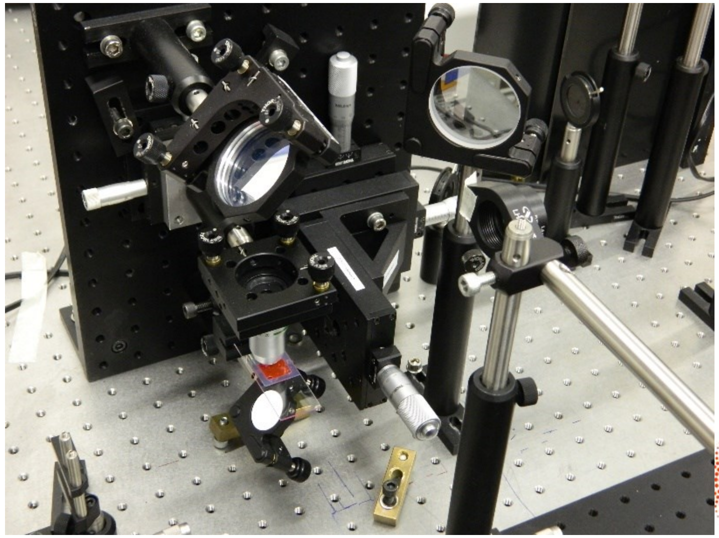
Modes in random lasers?

Experimental results
Random laser from paper cellulose templated
Titanium di-oxide

Experiments for the detection of the spatial extension of the RL modes







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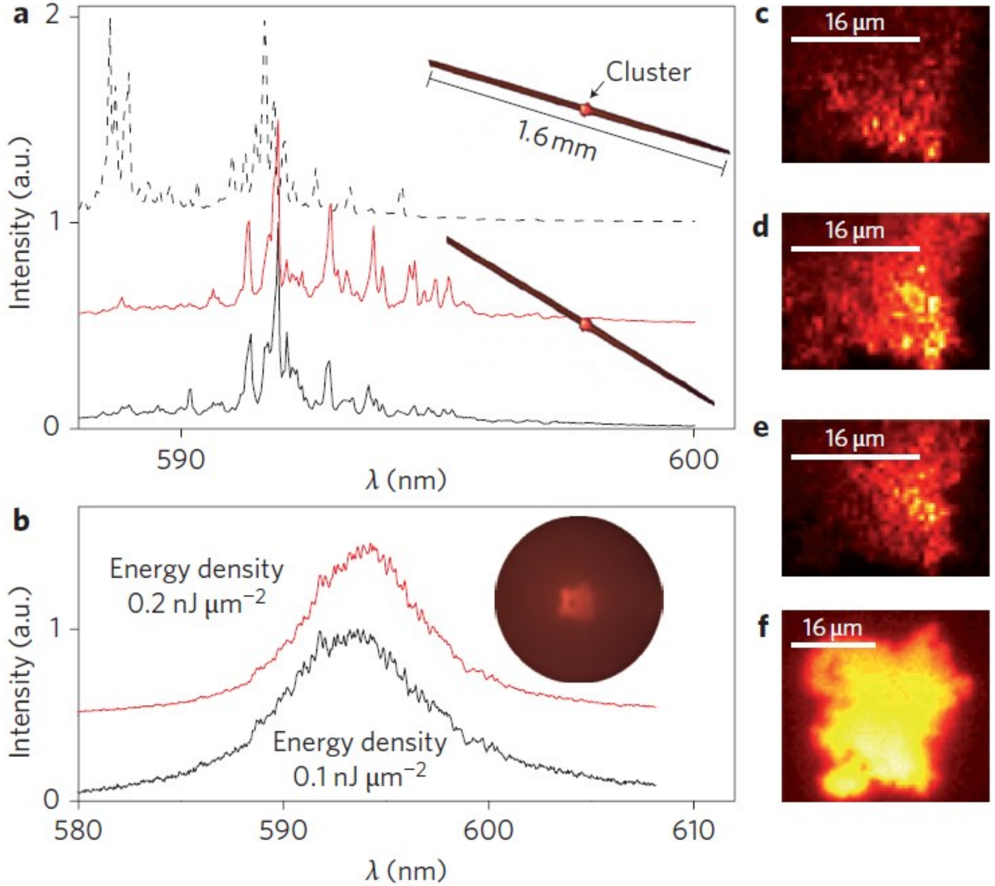
Modes in random lasers



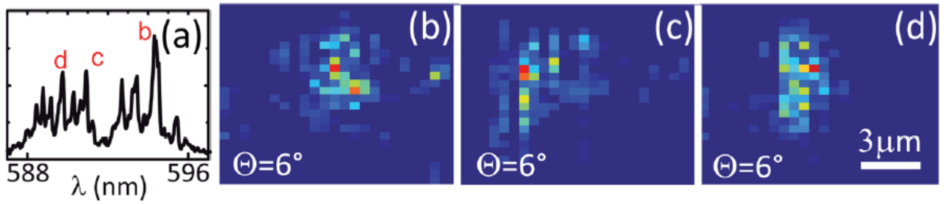


FIG. 3. (a) Spectrum from the cluster C3 obtained by pumping with $\Theta = 6°$; (b)-(d) spatial distribution of intensity for the three modes indicated in (a).

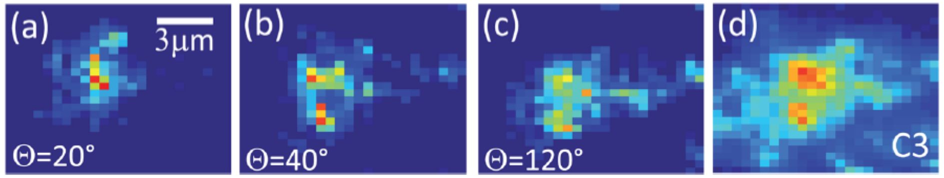
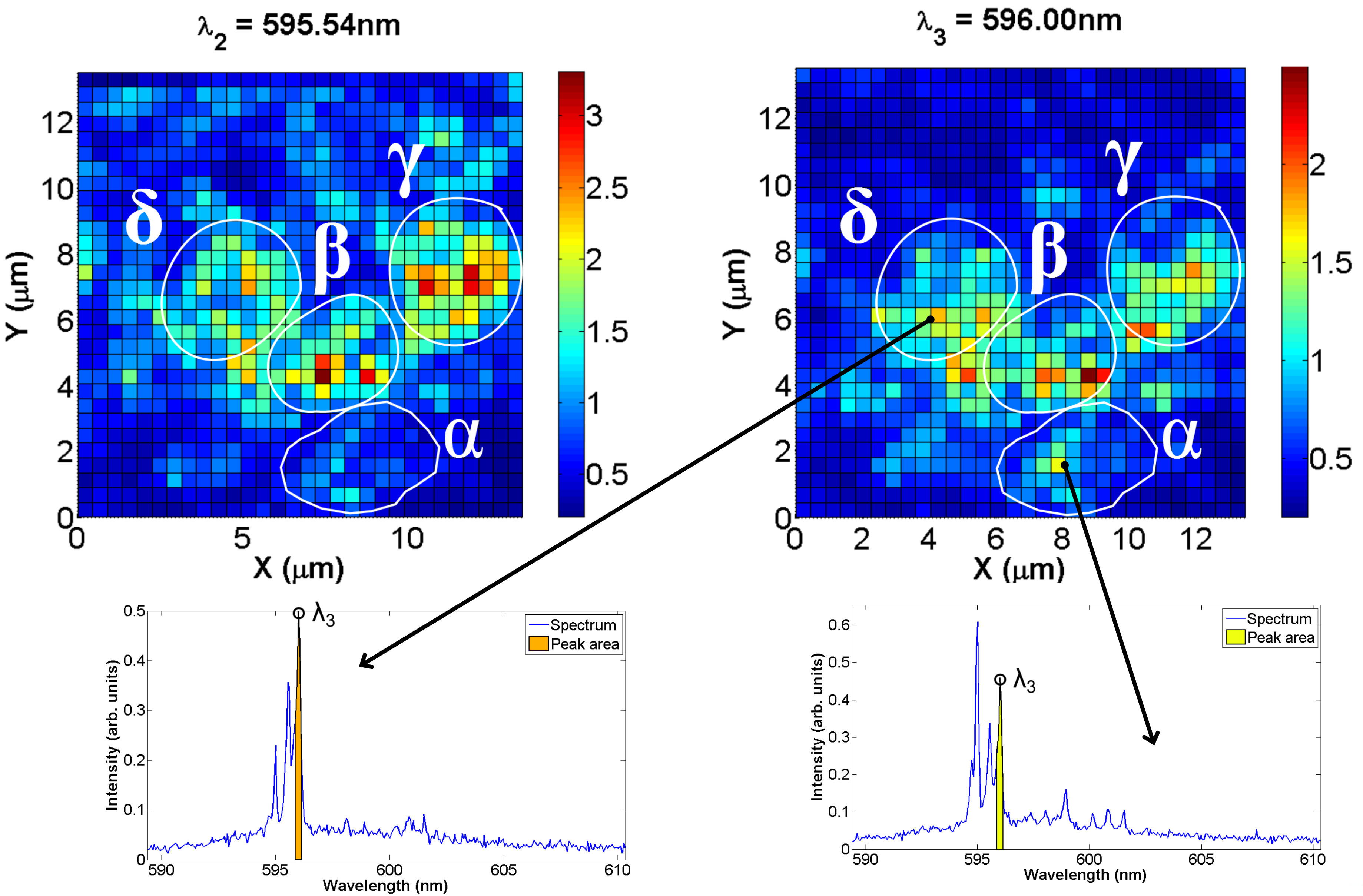


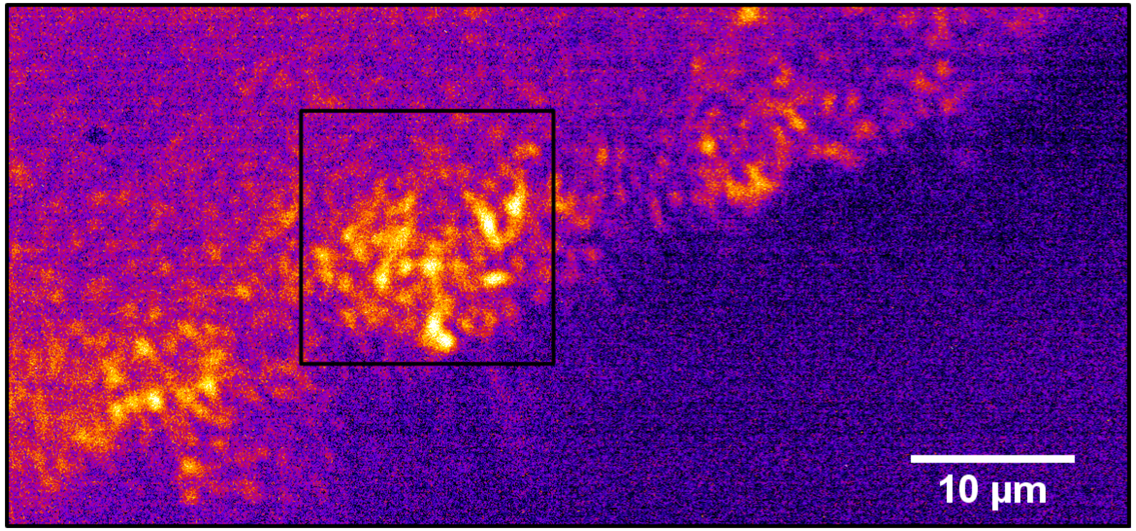
FIG. 4. (a)-(c) Represent the spatial intensity distribution for the most intense mode of cluster C3 when pumped with $\Theta = 20°$, $\Theta = 40°$, and $\Theta = 120°$, respectively, while panel (d) reports the spatial distribution of the intensity (all wavelengths summed) below lasing threshold providing the shape of the cluster.

Nature Photonics, **5**, **10**: 615—617 (2011)

Mode 2&3:



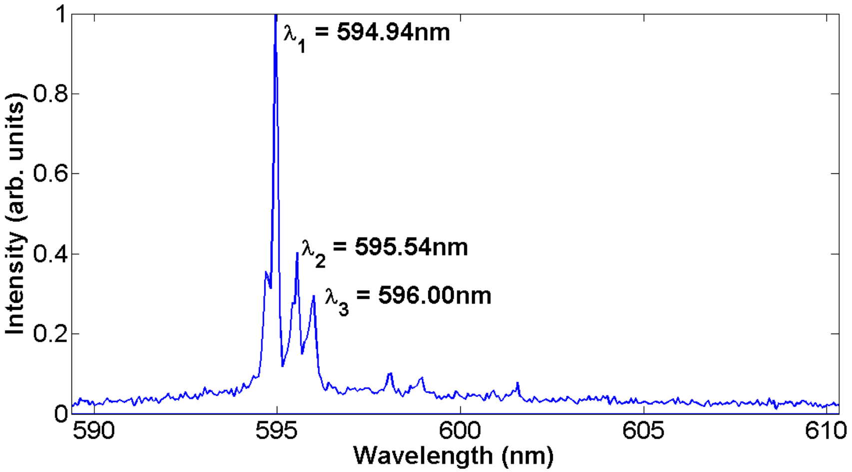
λ2 = 595.54nm
λ3 = 596.00nm
δ
β
γ
α
Y (μm)
X (μm)
Spectrum
Peak area
λ3
Intensity (arb. units)
Wavelength (nm)

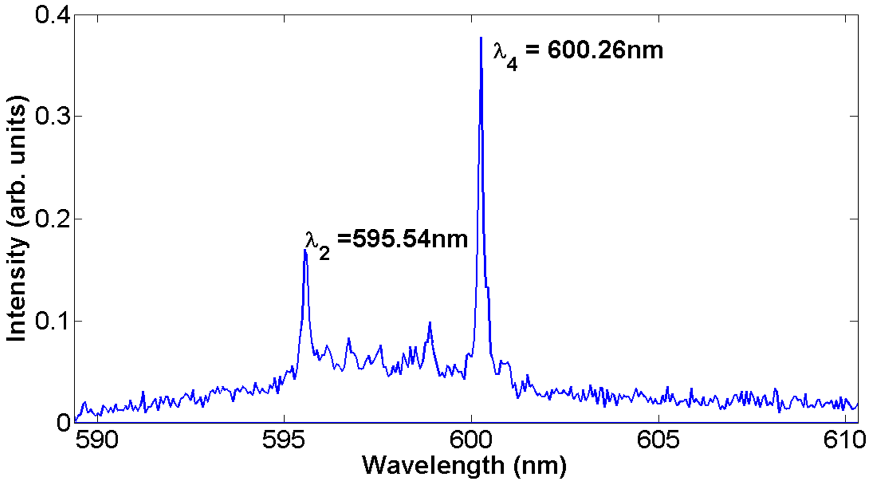


The fiber moves and scans the emission from the sample with a resolution < 1 micron

Scan over 13.5X13.5 micron
Resolution 0.45 micron
Pump energy 10 muJ

Four modes (peaks) with FWHM = 0.2nm





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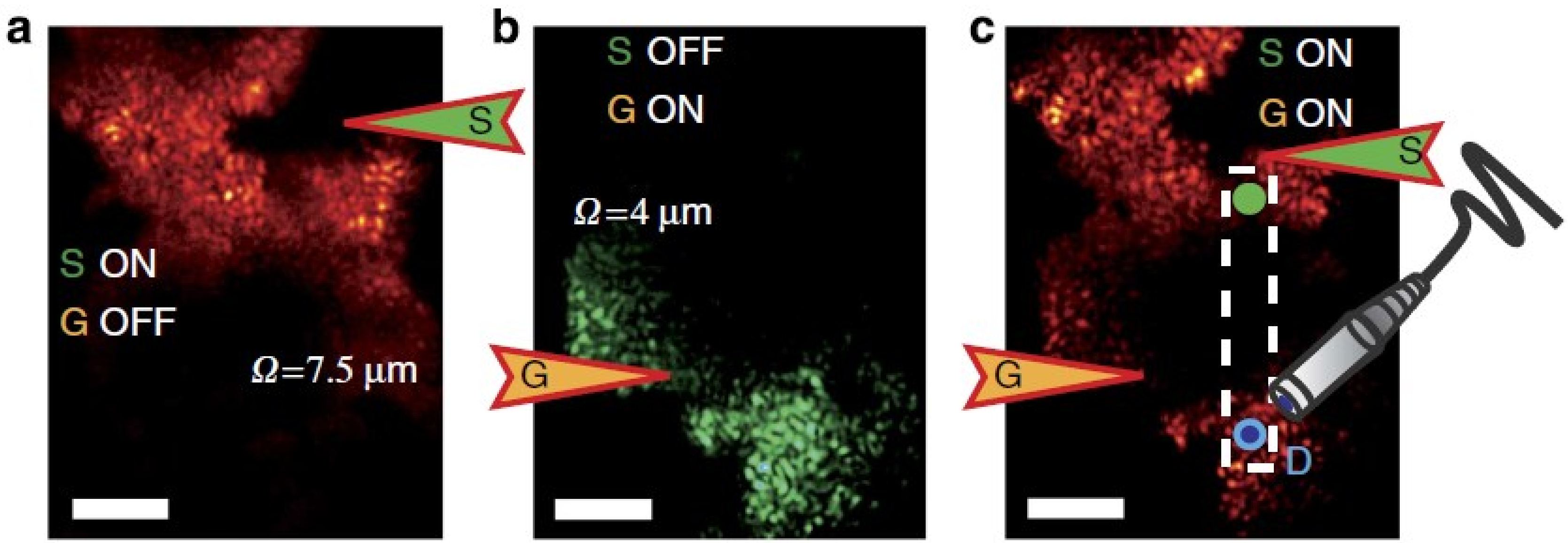
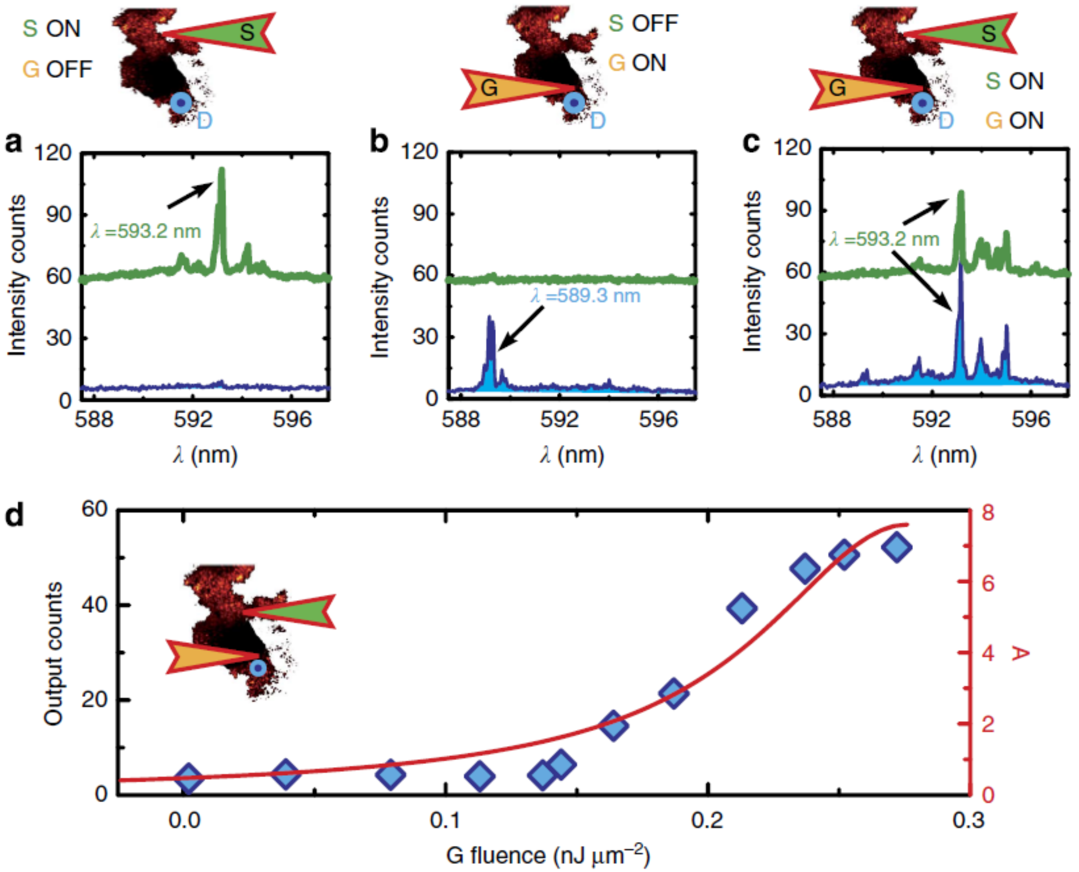
Do random laser modes interact?

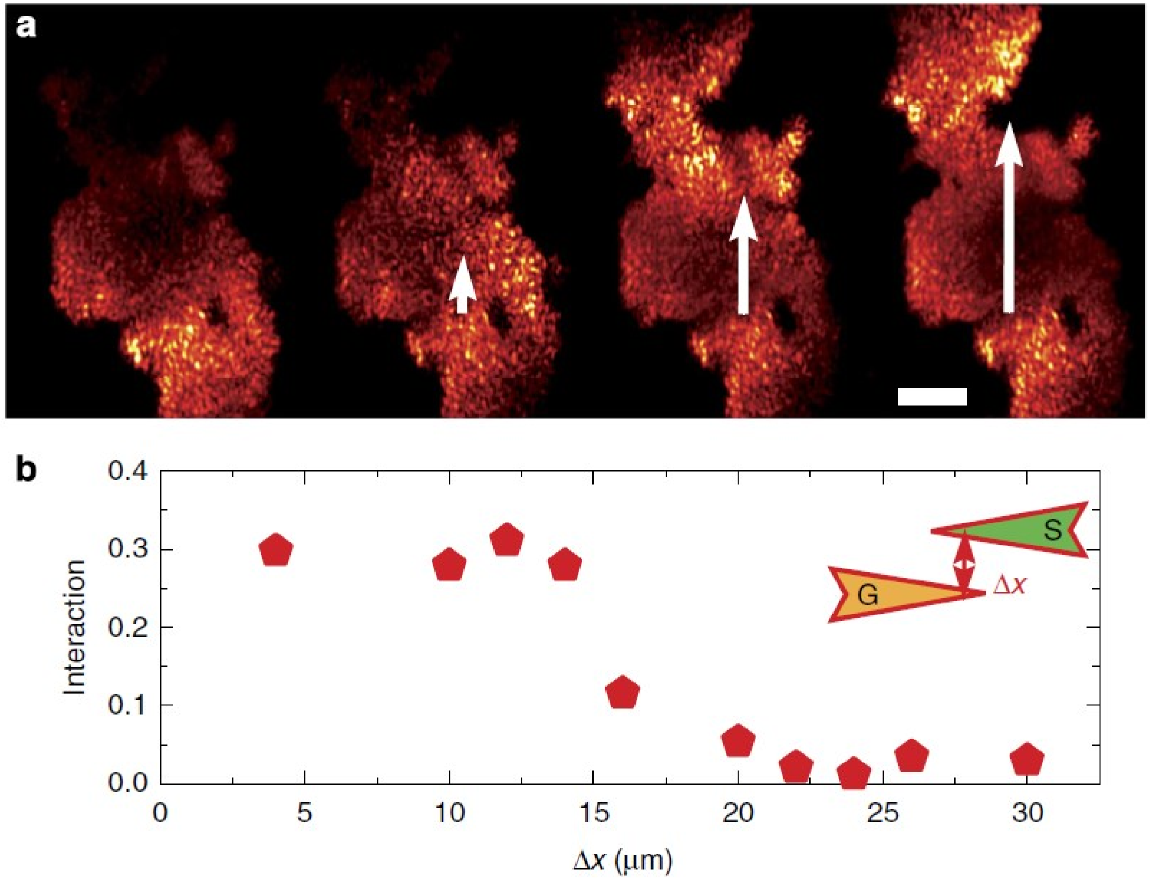
Experiment to measure RL interaction



Nat Commun, **4**: 1740 (2013)

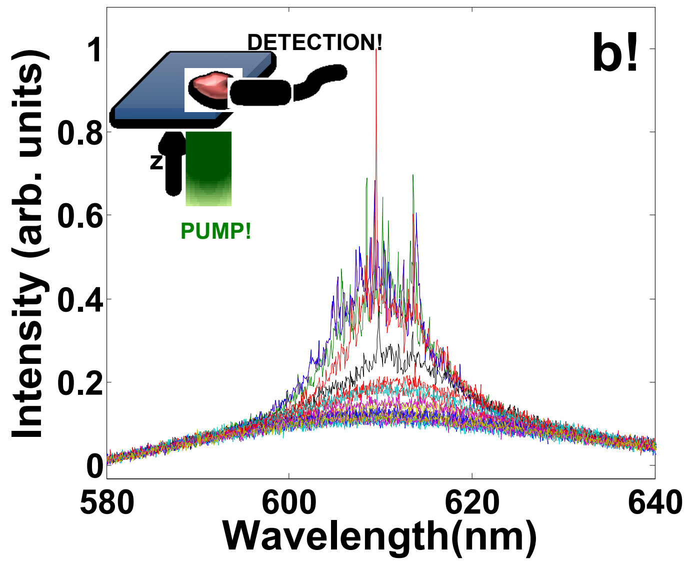

S ON
G OFF
S
D
S OFF
G ON
G
S ON
G ON
a
b
c
d
Intensity counts
λ =593.2 nm
λ =589.3 nm
λ (nm)
120
90
60
30
0
588
592
596
Output counts
G fluence (nJ μm^{-2})
A
0.0
0.1
0.2
0.3
60
40
20
8
6
4
2

Which is the interaction length?



Measure of the Overlap?

- The spectra are determined by the amplitude and the phases of the mode
- The spectra fluctuate
- The phases determine the fluctuations
- We measure the overlap of the fluctuations of the spectra



$$\Delta_\alpha(k) = I_\alpha(k) - \bar{I}(k)$$

$$q_{\alpha\beta} = \frac{\sum_{k=1}^{N} \Delta_\alpha(k)\Delta_\beta(k)}{\sqrt{\sum_{k=1}^{N} \Delta_\alpha^2(k)}\sqrt{\sum_{k=1}^{N} \Delta_\beta^2(k)}}.$$

No RSB

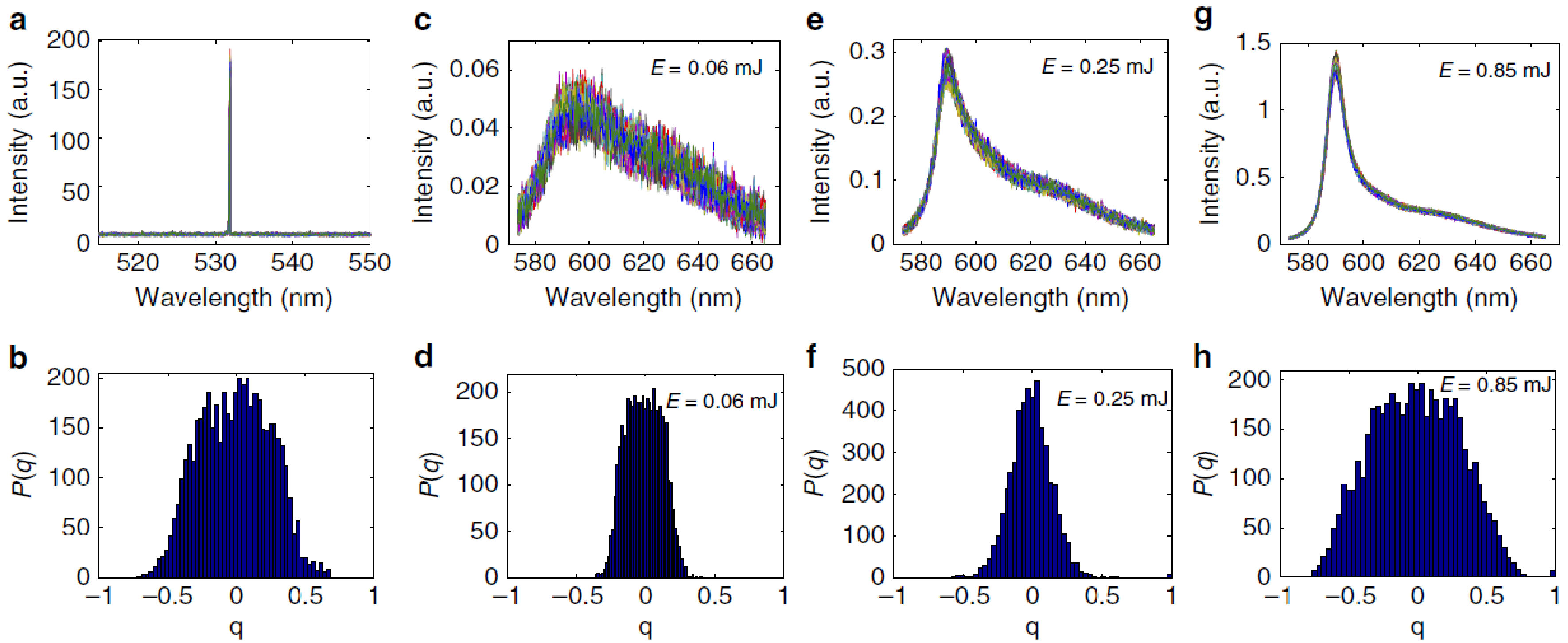
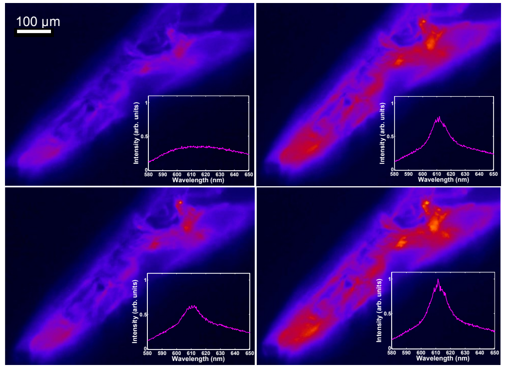
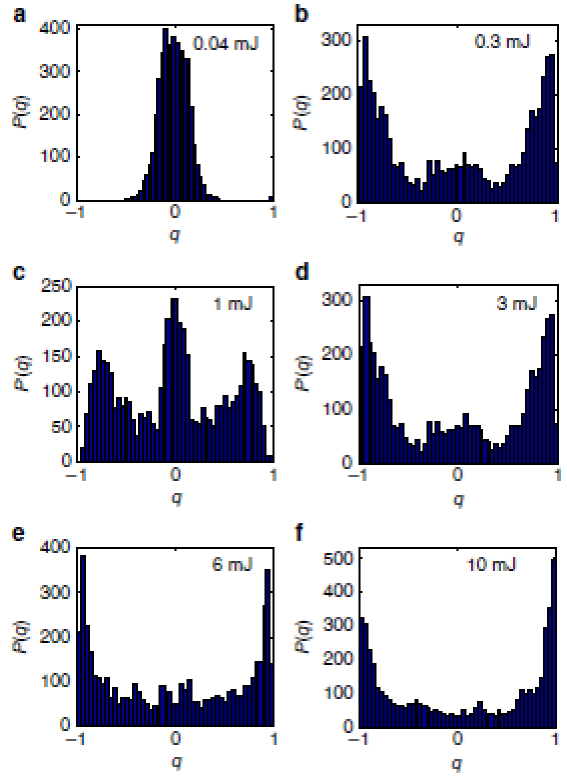


Figure 5 | Distribution function of the overlap in standard and random laser without replica symmetry breaking. (**a**,**b**) Emission spectra (**a**) and $P(q)$ (**b**) of a Q-switched pulsed Nd-Yag standard ordered laser. The analysis is done on 100 shots. (**c**–**h**) Emission spectra (**c**,**e**,**g**) and correspondent $P(q)$ (**d**,**f**,**h**) of a liquid dispersion of titanium dioxide in rhodamine B-ethylene glycol solution at three different pump energy through the threshold.

RSB





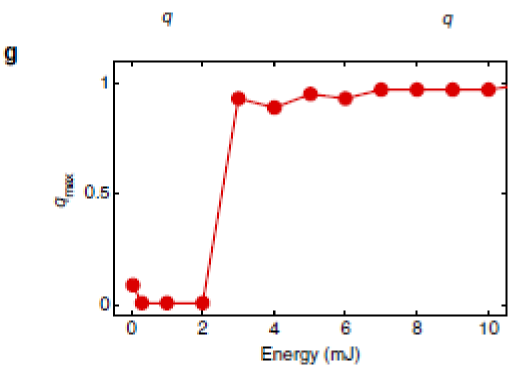


Figure 4 | Distribution function of the overlap showing replica symmetry breaking by increasing pump energy. (**a-f**) Distribution of the overlap q at different pump energy. (**g**) q_{max} corresponding to the position of the maximum of $P(|q|)$ versus pumping.

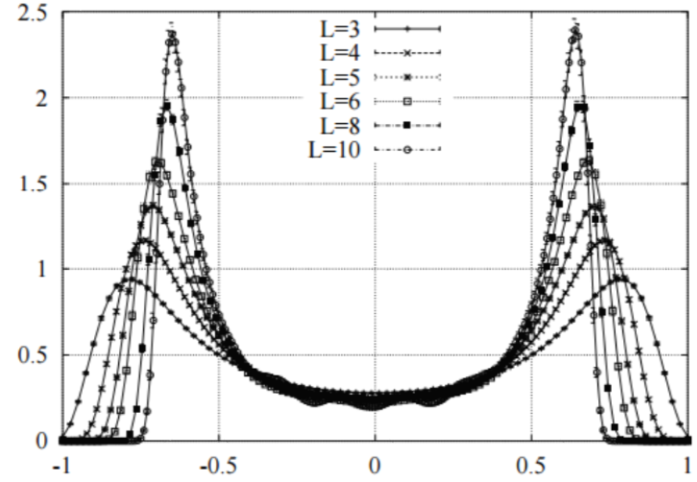


Figure 3: The function $P(q) = \overline{P_J(q)}$ after average over many samples (D=4, L=3…10) .

nature COMMUNICATIONS

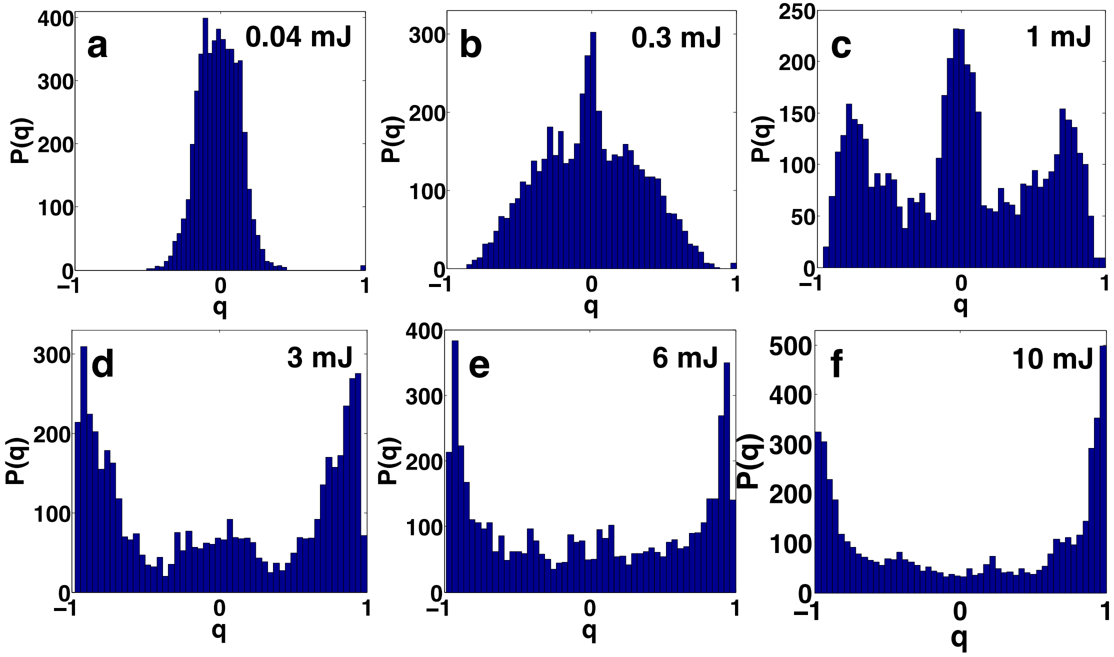
ARTICLE

Received 18 Dec 2013 | Accepted 8 Dec 2014 | Published 14 Jan 2015

DOI: 10.1038/ncomms7058

Experimental evidence of replica symmetry breaking in random lasers

N. Ghofraniha[1,2,3], I. Viola[2,4], F. Di Maria[5,6], G. Barbarella[5], G. Gigli[4,7], L. Leuzzi[1,2] & C. Conti[2,3]



Replica symmetry breaking: identical systems under identical conditions may reach different states. This effect is revealed by the shape of the probability distribution function of an order parameter named the Parisi overlap. Here we investigate pulse-to-pulse fluctuations in random lasers, we introduce and measure the analogue of the Parisi overlap in independent experimental realizations of the same disordered sample, and we find that the distribution function yields evidence of a transition to a glassy light phase compatible with a replica symmetry breaking.

Open issues

- Direct measurement of phases and amplitude of the modes?
- Direct measurement of the couplings J?
- Accurate measurement of the P(q) to assess a Full RSB
- ············ many publications over the years

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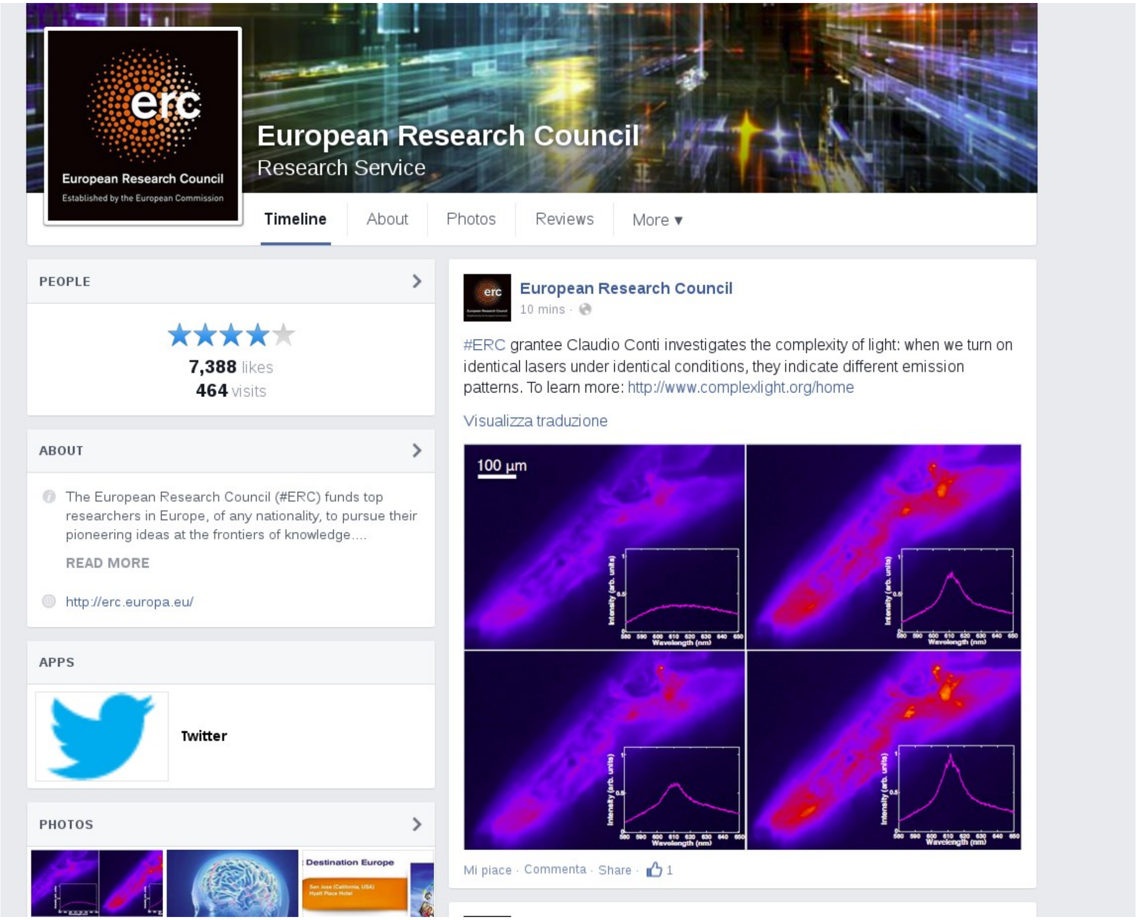

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