XV J. A. Swieca School and SPSAS Nanophotonics– Campinas July, 2016

High-order nonlinearities in disordered media

Cid B. de Araújo

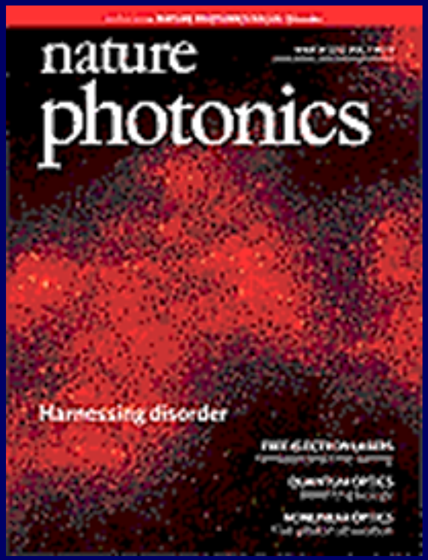
Universidade Federal de Pernambuco, Recife, Brasil

Third lecture:

Multiphoton absorption and stimulated emission in random media.

Disordered Photonics

Disordered structures that strongly scatter light



March 2013

Anderson localization of light
and other transport phenomena

Imaging through scattering media.

Optical nonlinearities.

Random lasers

Measurements of nonlinear refractive index in scattering media

Prathyush Samineni,[1] Zachary Perret,[1] Warren S. Warren,[2] and Martin C. Fischer[1,*]

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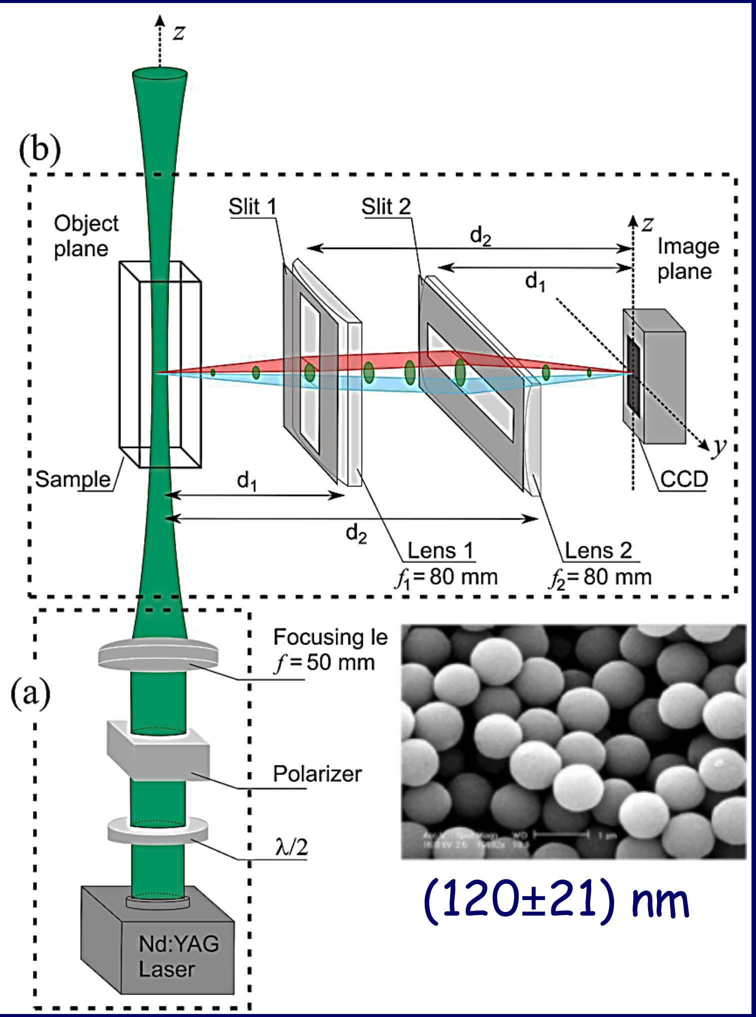
Abstract: We have recently developed a spectral re-shaping technique to simultaneously measure nonlinear refractive index and nonlinear absorption. In this technique, the information about the nonlinearities is encoded in the frequency domain, rather than in the spatial domain as in the conventional Z-scan method. Here we show that frequency encoding is much more robust with respect to scattering. We compare spectral re-shaping and Z-scan measurements in a highly scattering environment and show that reliable spectral re-shaping measurements can be performed even in a regime that precludes standard Z-scans.

Opt. Express 18 (2010) 12727

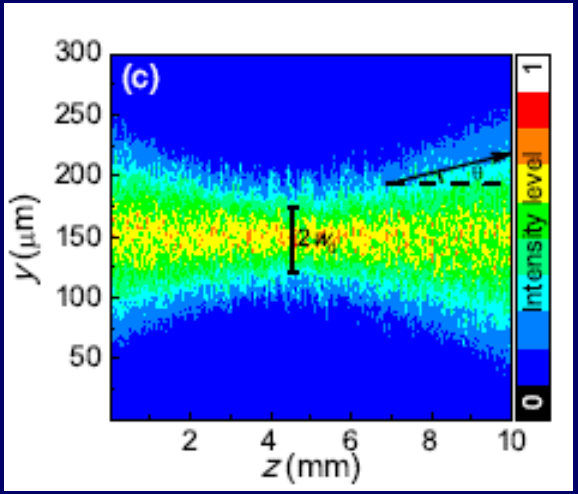
Laser with rep. rate: 20 kHz
Pulses with amplitude modulated at 5 kHz

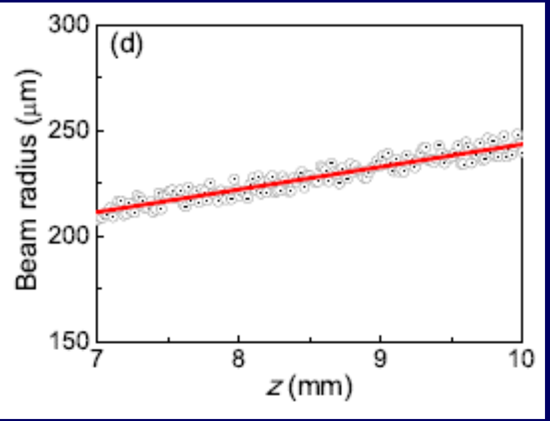
Scattered Light Imaging Method - SLIM

Silica NPs + ethanol +acetone



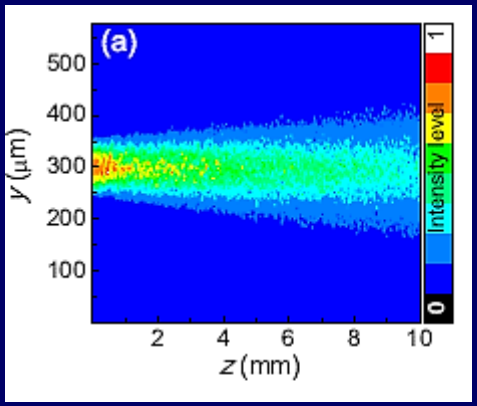
(120±21) nm

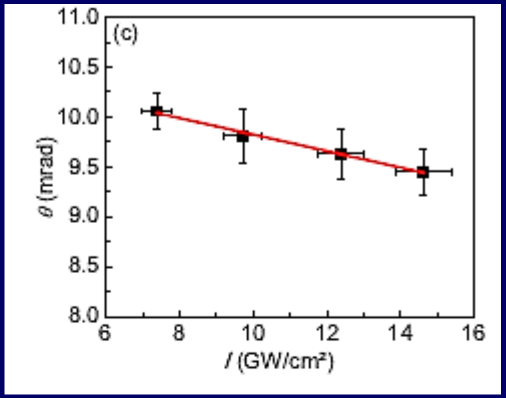


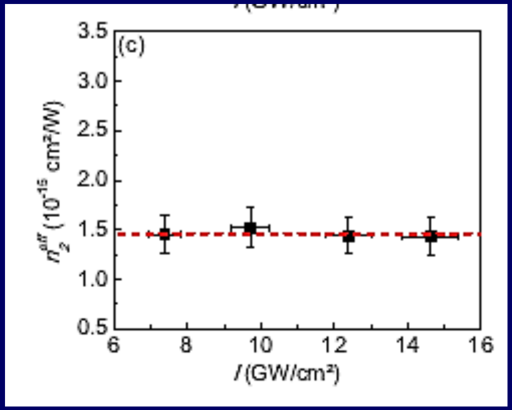


Silica NPs + ethanol +acetone

Avg. diameter = (120±21) nm







$$\frac{1}{f_{eff}} = \frac{a\lambda M^2}{4\pi n_2^{eff} I_0} J$$

$$\frac{\theta_{NL}(z)}{\theta_0(z)} = 1 - \frac{z}{f_{eff}} + \mathcal{O}\left(\frac{z^2}{f_{eff}^2}\right)$$

a=6.4 $J \approx \pi/4$

Table 1. Characteristics of the samples.

Parameters	Sample A	Sample B	Sample C	Sample D
$f(\times 10^{-4})$	0.045	2.25	2.7	4.05
$V_{acetone}$	99%	50%	40%	10%
$n_2^{eff}(\times 10^{-15} cm^2/W)$	2.38	1.58	1.42	0.93

Opt.Express 23 (2015) 19512-19521

Random Lasers

- Introduction – short history + comments on light propagation in scattering media + potential applications
- Examples and applications
  - Stokes and anti-Stokes lasers
  - Multicolor emission from RLs

RLs: lasers without mirrors

No optical cavity

The optical feedback is due to light scattering in a disordered medium, and the interference of the scattered light gives rise to resonant modes at particular frequencies

RL can be implemented in 1D, 2D and 3D geometries depending on the scattering medium: optical waveguide, membrane, or colloids and aggregates/powders

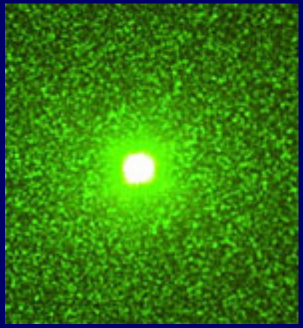
Conventional laser

Narrow linewidth

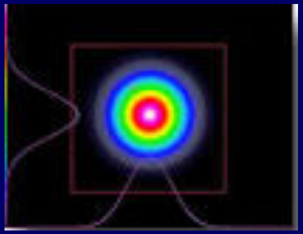
Long coherence time

Large spatial coherence

Directionality



Speckle



Beam profile

Random laser

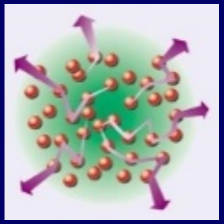
Large linewidth

Short coherence time

Small spatial coherence

No directionality

No speckle



"photon bomb"
or
"laser paint"

Old history

Ambartsumyan, Basov, Kryukov, Lethokov
Laser with nonresonant feedback
JETP Lett. 1966

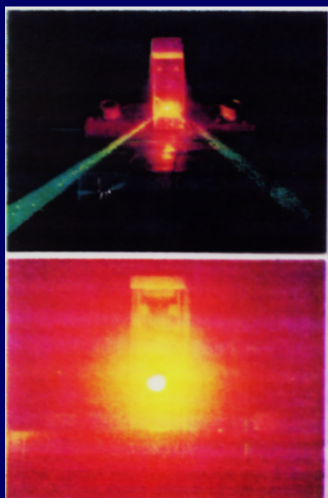
Lethokov: proposal of scattering with "negative absorption" to describe emissions from astrophysical molecular clouds
Sov. Phys. JETP 1967

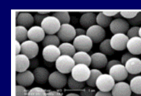
Lawandy, Balachandran, Gomes, Sauvain
Laser action in strong scattering media
Nature 1994

Wiersma
Random Lasers?
Nature 1995

Random laser

Lawandy et al. Nature 1994





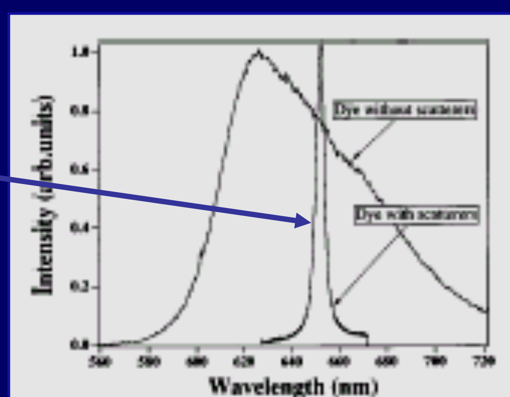
Photons follow random pathways due to reflections by TiO_2 NPs

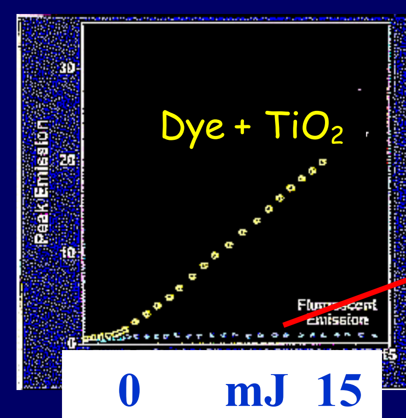
Rh: 2 x 10^{-3} M 10^{11} particles / cm^3

Mean free path: 120 μ m @ 532 nm

140 μ m @ 650 nm

Intensity ÷ 100

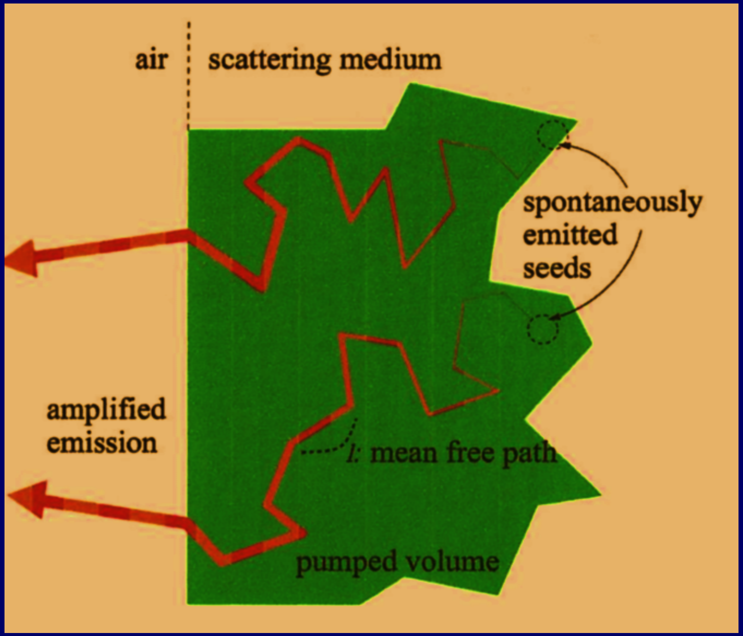




Only dye

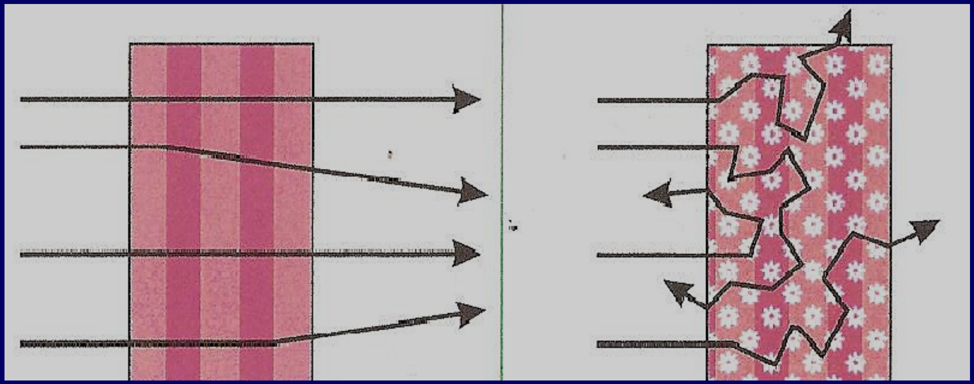
Multiple light scattering is essential

Light follows a random pathway through the excited volume



mean free path larger than the sample volume

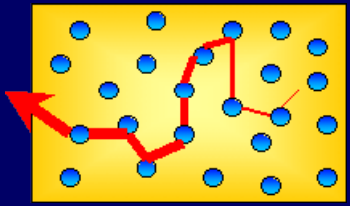
mean free path smaller than the sample dimension



Balistic propagation

Diffusion

Incoherent feedback



Coherent feedback

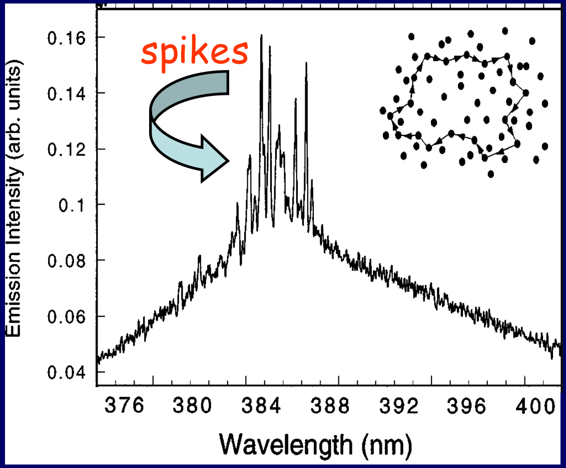
"Optical cavities" $n\lambda/2=L$

First observation of RL with coherent feedback

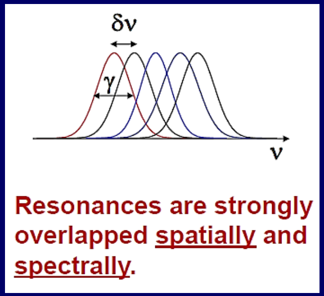
H. Cao et al. PRL 82 (1999) 2278

ZnO powder – grains diameter: 100 nm

Nd: YAG
355 nm



Constructive interference



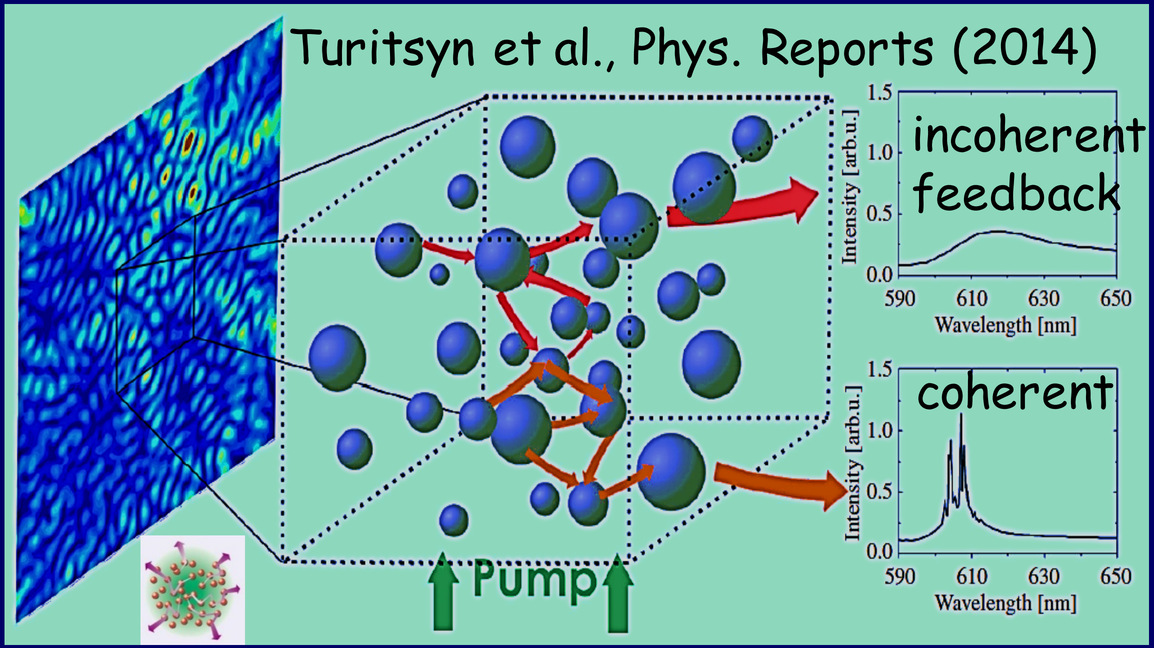
$L = n \ \lambda/2$

analogous of ring cavity of conventional lasers

The direction of each mode is different

Co-existence of strongly and weakly localized random modes. Fallert et al. Nature Photon. (2009)

The mode-locking transition of random lasers. Leonetti et al. Nature Photon. (2011)

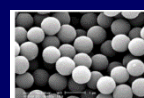


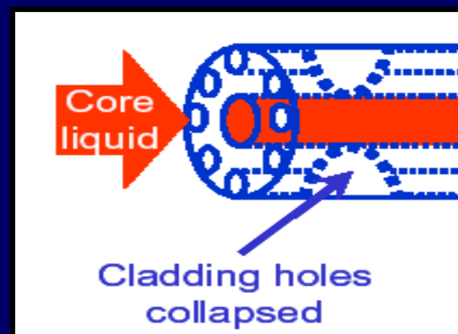
Directionality without mirrors?

PRL 99, 153903 (2007) PHYSICAL REVIEW LETTERS week ending 12 OCTOBER 2007

Random Fiber Laser

Christiano J. S. de Matos,[1,*] Leonardo de S. Menezes,[2] Antônio M. Brito-Silva,[3] M. A. Martinez Gámez,[4] Anderson S. L. Gomes,[2] and Cid B. de Araújo[2]

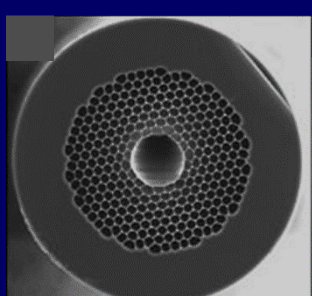


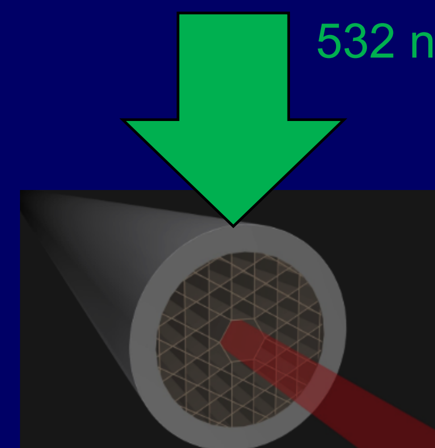


100 times
more efficient
than conventional RL

Rh 6G + TiO_2 NPs

Hollow fiber
10 μm – 3 μm





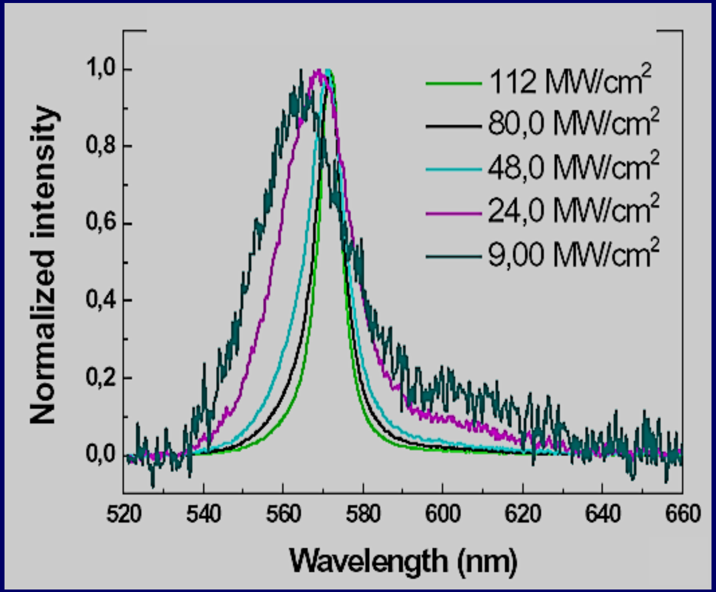
<u>Feedback</u>

Transverse: total internal reflection

Axial : multiple scattering

Random Fiber Laser

1D laser - Directional emission



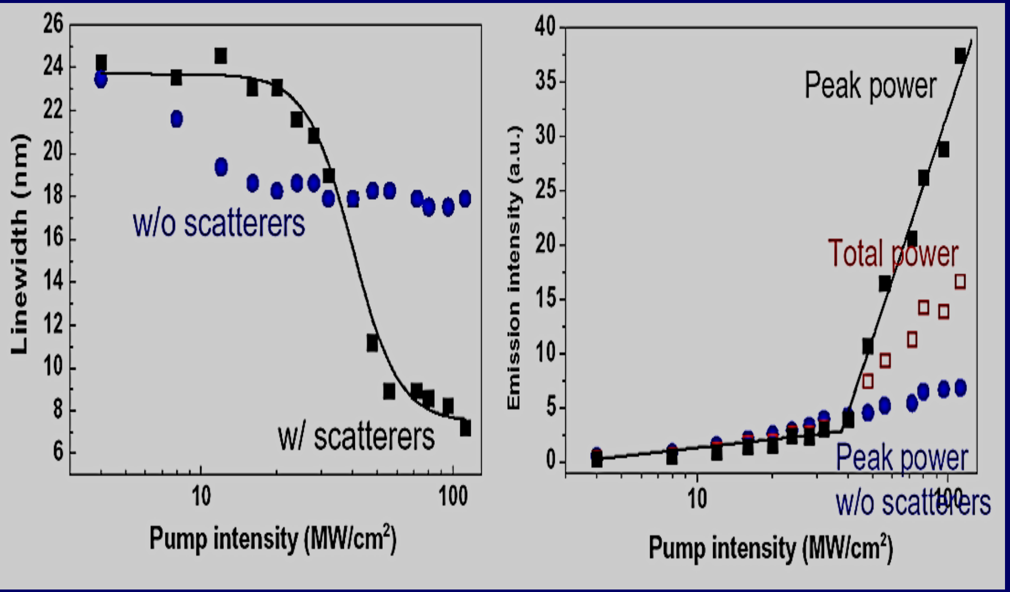


Figure of merit = $(I_{thr} \cdot \rho_{dye} \cdot \rho_{scatt})^{-1}$

At least 100 times improvement with the Fiber Random Laser

de Matos, Menezes, Brito-Silva, Gamez, Gomes, de Araújo.
Phys. Rev. Lett. 99 (2007) 153903

Smirnov et al. - Modeling of spectral and statistical properties of a random distributed fiber laser Opt. Express 21 (2013) 21236

Turitsyn et al. - Random distributed feedback fibre lasers. Phys. Reports 542 (2014) 133

Tang et al. - A random Q-switchd fiber laser Sci. Reports 5 (2015) 9338

Churkin et al. - Wave kinetics of random fiber lasers Nature Commun. 6 (2015) 6214

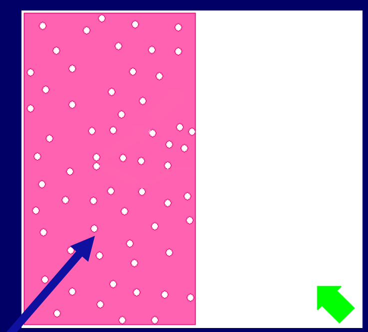
Churkin et al. - Recent advances in fundamental and applications of random fiber lasers Adv. Opt. Photon. 7 (2015) 516

Dependence of RL emission on silver nanoparticle density in PMMA films containing rhodamine 6G

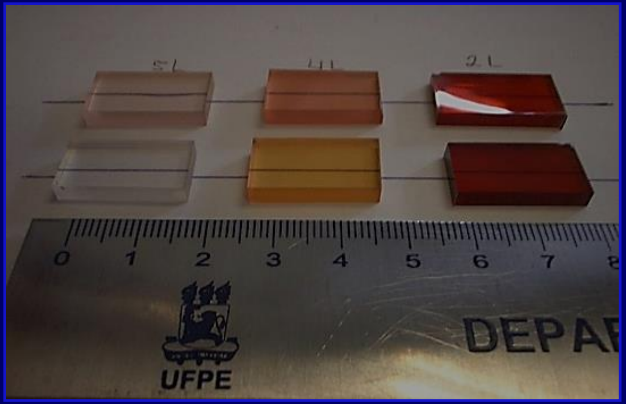
Polyester or PMMA with Ag NPs and Rh 6G

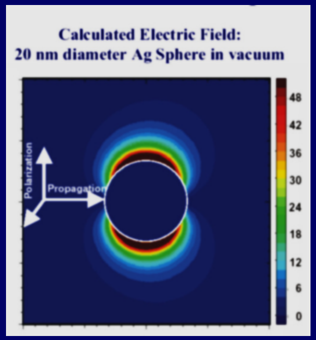
Brito-Silva et al. Polymer Eng. & Science 2350 (2010)

Rh6G



Nanoparticles



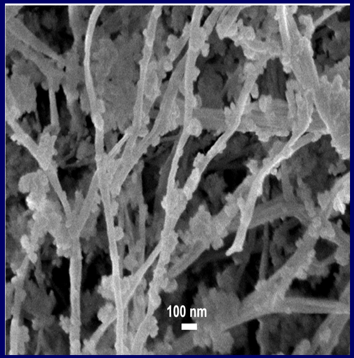


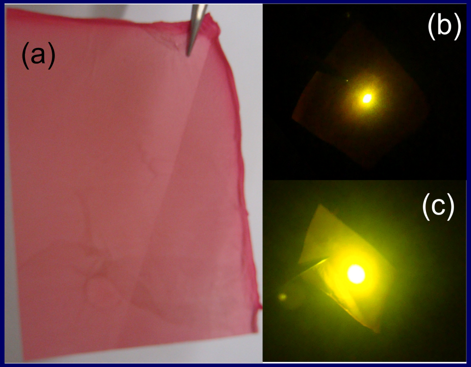
Dominguez, Maltez, Reis, Melo, de Araújo, Gomes
J. Opt. Soc. Am. B 28 (2011) 1118

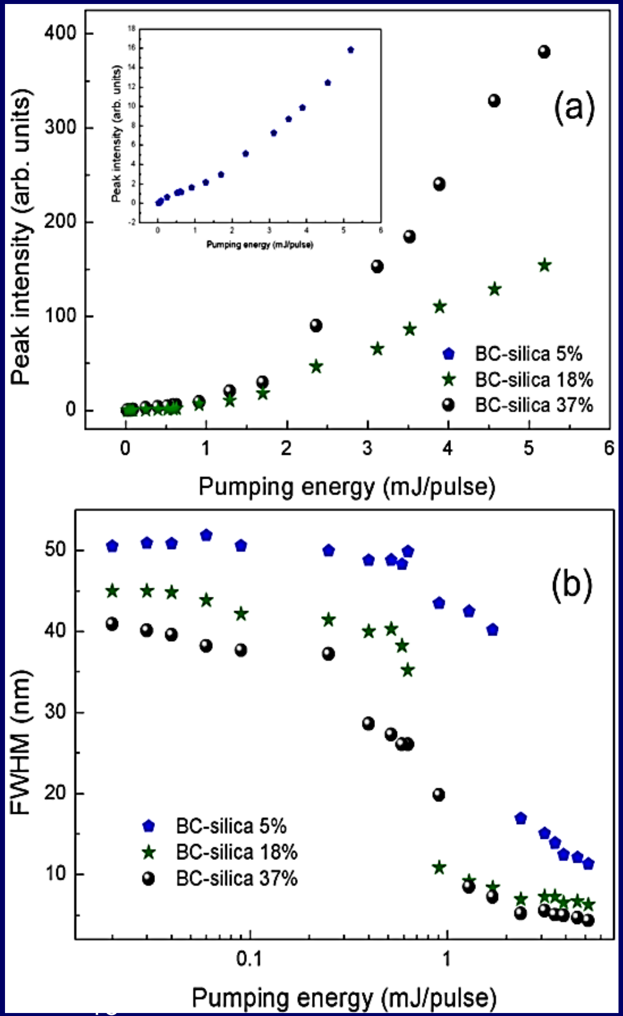
Random laser action from flexible biocellulose-based device

biocompatible - produced by the bacteria *Gluconacetobacter xylinus*

3D network composed of microfibrils having nanometric diameters.

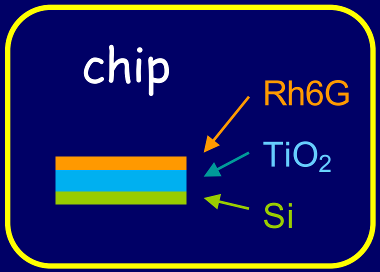


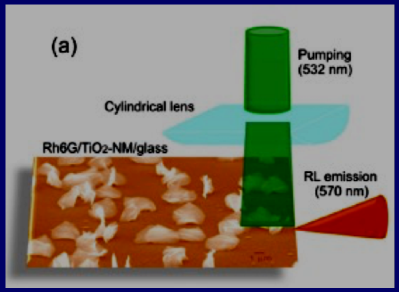


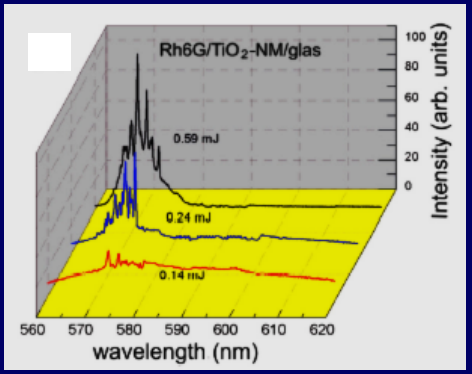


Santos, Dominguez, Schiavon, Barud, de Melo, Ribeiro, Gomes, de Araújo. J. Appl. Phys. 115 (2014) 083108

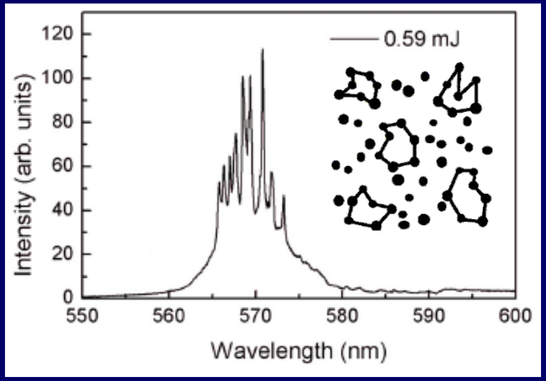
Microchip RL based on a disordered TiO_2 nanomembranes arrangement







MOCVD technique



532 nm

5 ns, 5 Hz

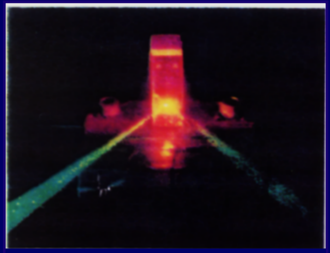
Coherent feedback

Dominguez, Lacroute, Chaumont, Sacilotti, de Araújo, Gomes. Optics Express 20 (2012) 17381

Random Lasers

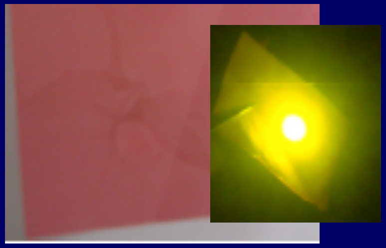
Can be implemented in 1D, 2D and 3D geometries in various scattering media: waveguides, membranes, colloids andpowders

Colloids

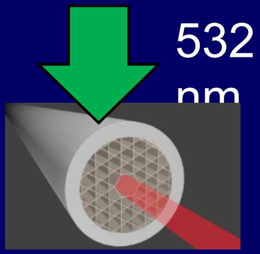


Powder

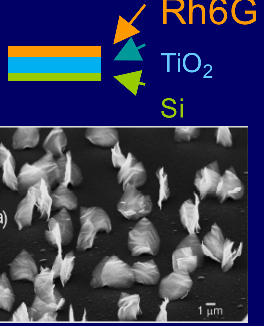
Membrane



Microstructued Fibers



Chips



IOP PUBLISHING — JOURNAL OF OPTICS
J. Opt. 12 (2010) 024010 (4pp) doi:10.1088/2040-8978/12/2/024010

Cancerous tissue mapping from random lasing emission spectra

R C Polson and Z V Vardeny

Department of Physics and Astronomy, University of Utah, Salt Lake City, UT 84112, USA

Received 16 May 2009, accepted for publication 2 September 2009
Published 11 January 2010
Online at stacks.iop.org/JOpt/12/024010

Abstract
Random lasing emission spectra have been collected from both healthy and cancerous tissues. The two types of tissue with optical gain have different light scattering properties as obtained from an average power Fourier transform of their random lasing emission spectra. The difference in the power Fourier transform leads to a contrast between cancerous and benign tissues, which is utilized for tissue mapping of healthy and cancerous regions of patients.

Keywords: random lasing, cancerous tissue mapping, data processing, Fourier transform

New Astronomy Reviews 51, 443 (2007)

Astrophysical lasers and nonlinear optical effects in space

S. Johansson [a,*], V.S. Letokhov [a,b]

Detection of nanoscale structural changes in bone using random lasers

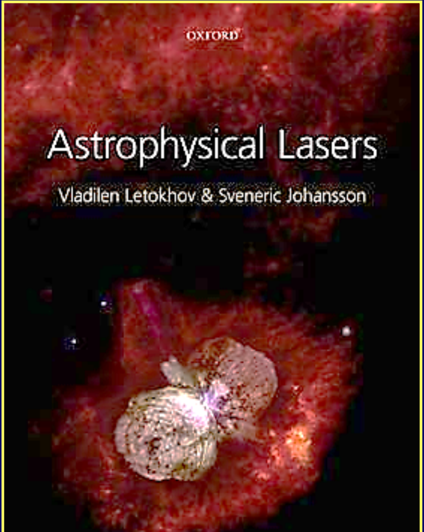
Qinghai Song,[1] Zhengbin Xu,[1] Seung Ho Choi,[1] Xuanhao Sun,[1] Shumin Xiao,[2] Ozan Akkus[1], and Young L. Kim[1,*]

[1]Weldon School of Biomedical Engineering, Purdue University, West Lafayette, IN 47907
[2]School of Electrical and Computer Engineering and Birck Nanotechnology Center, Purdue University, West Lafayette, IN 47907
*youngkim@purdue.edu

Abstract: We demonstrate that the unique characteristics of random lasing in bone can be used to assess nanoscale structural alterations as a mechanical or structural biosensor, given that bone is a partially disordered biological nanostructure. In this proof-of-concept study, we conduct photoluminescence experiments on cortical bone specimens that are loaded in tension under mechanical testing. The ultra-high sensitivity, the large detection area, and the simple detection scheme of random lasers allow us to detect prefailure damage in bone at very small strains before any microscale damage occurs. Random laser-based biosensors could potentially open a new possibility for highly sensitive detection of nanoscale structural and mechanical alterations prior to overt microscale changes in hard tissue and biomaterials.

Biomedical Optics Express 1 (2010) 1401

A random laser *could* happen naturally in space



2008

nature physics — LETTERS
PUBLISHED ONLINE: 5 MAY 2013 | DOI: 10.1038/NPHYS2614

A cold-atom random laser

Q. Baudouin, N. Mercadier†, V. Guarrera†, W. Guerin and R. Kaiser*

Prior 1994 – Theoretical Proposal and Experimental demonstrations with MICRON SIZED Nd powders (solid state)

1994 – 250nm TiO_2/Dye colloid (Nature milestone paper) +

1995/7 – Bio-tissue; polymeric gain media; solid state

1998/2000 – ZnO Nanopowders; opal photonic crystals;

2001/3 – pi-conjugated polymers; temperature control; liquid crystals; plasmonic effect for directionality

2004/6 – Human Tissues+dye; organic/inorganic; silver nanoparticles

2007/9 – Random Fiber Lasers; DNA; mid-infrared; one & two photon GaAs;

2010/14 – colloidal QD; plasmonically controlled core-shell; UV UC in Nd; Mode-locking; cicada wings; Cold Atom; TiO_2 nanomembranes; 3-photons in ZnO and colloids

nature photonics

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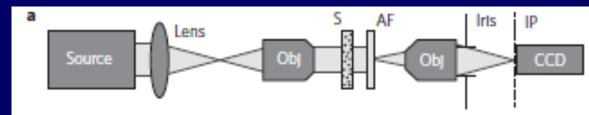
NATURE PHOTONICS | LETTER

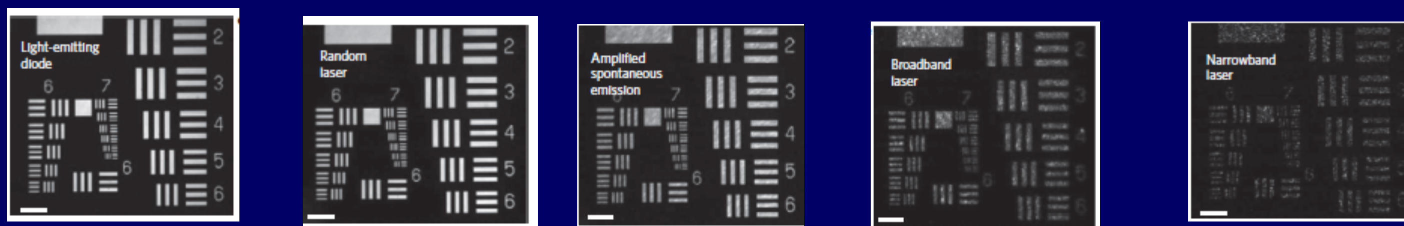
Speckle-free laser imaging using random laser illumination

Brandon Redding, Michael A. Choma & Hui Cao

Many imaging applications require increasingly bright illumination sources, motivating the replacement of conventional thermal light sources with light emitting diodes (LEDs), superluminescent diodes (SLDs) and lasers. Despite their brightness, lasers and SLDs are poorly suited for full-field imaging applications because their high spatial coherence leads to coherent artifacts known as speckle that corrupt image formation[1, 2]. We recently demonstrated that random lasers can be engineered to provide low spatial coherence[3]. Here, we exploit the low spatial coherence of specifically-designed random lasers to perform speckle-free full-field imaging in the setting of significant optical scattering. We quantitatively demonstrate that images generated with random laser illumination exhibit higher resolution than images generated with spatially coherent illumination. By providing intense laser illumination without the drawback of coherent artifacts, random lasers are well suited for a host of full-field imaging applications from full-field microscopy[4] to digital light projector systems[5].

Speckle-free laser imaging using random laser illumination
Cao et al. Nature Photon. 6 (2012) 355





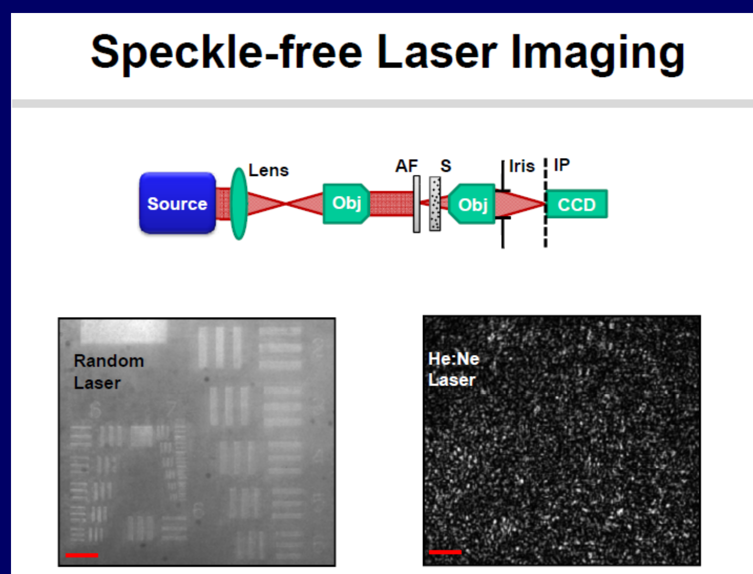
LED

RL

ASE

Broadband laser

Narrowband laser



Some review articles

Wiersma - Review article: *The physics and applications of random lasers*. Nature Phys. 4 (2008) 359

Turitsyn et al. - *Random distributed feedback fibre lasers*. Phys. Reports 542 (2014) 133

Churkin et al. - Recent advances in fundamental and applications of random fiber lasers Adv. Opt. Photon. 7 (2015) 516

Sebbah et al. - *Breakthrough in Photonics 2014: Random Lasers.* IEEE Photonics Journal 7 (2015) 0700207

Luan, Gu, Gomes, Yong, Wen, Prasad. *Lasing in nanocomposites random media.* Nano Today 10 (2015) 168

Multiphoton absorption in Random Lasers

RL wavelength is shorter than the excitation wavelength

Anti-Stokes RLs

$\chi^{(3)}$ and $\chi^{(5)}$ effects

Direct three-photon excitation of upconversion random laser emission in a weakly scattering organic colloidal system

Anderson S. L. Gomes,[1] Mariana T. Carvalho,[1] Christian T. Dominguez,[1,3] Cid B. de Araújo,[1] and Paras N. Prasad[2]

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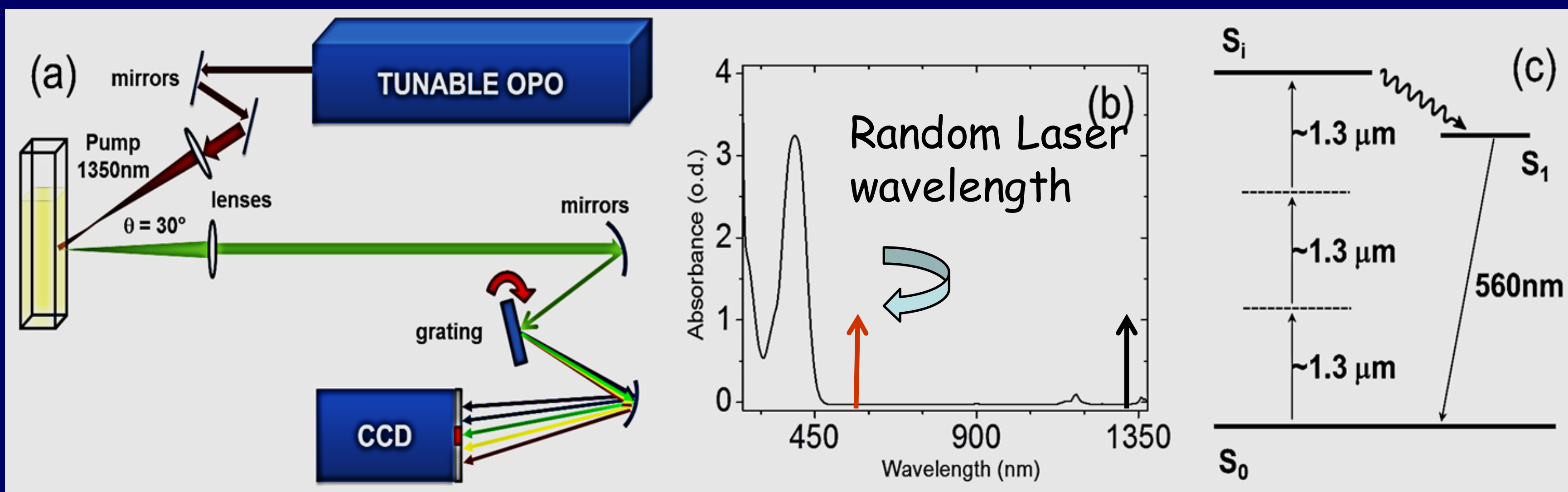
[2]*Institute for Lasers, Photonics and Biophotonics - University at Buffalo, The State University of New York, Buffalo, New York 14260-3000, USA*
[3]*Laboratório de Óptica Biomédica e Imagem, Universidade Federal de Pernambuco 50740-530, Recife, PE, Brazil*

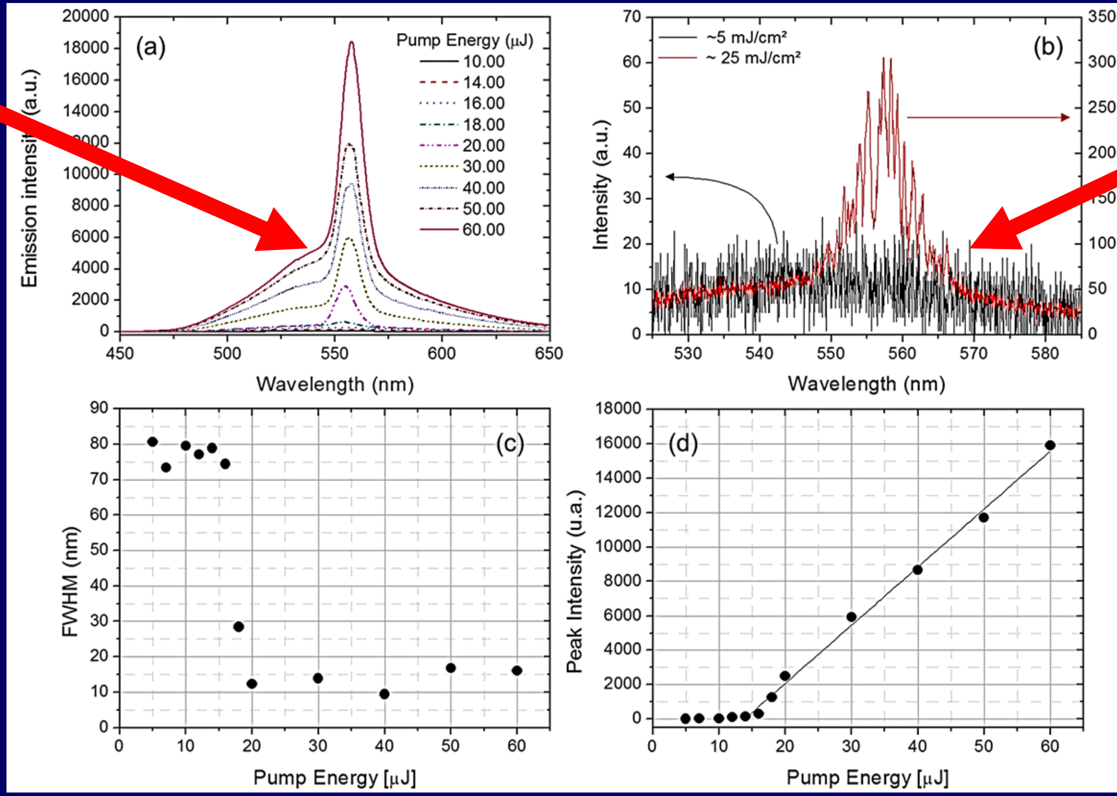
Opt. Express 22 (2014) 14305



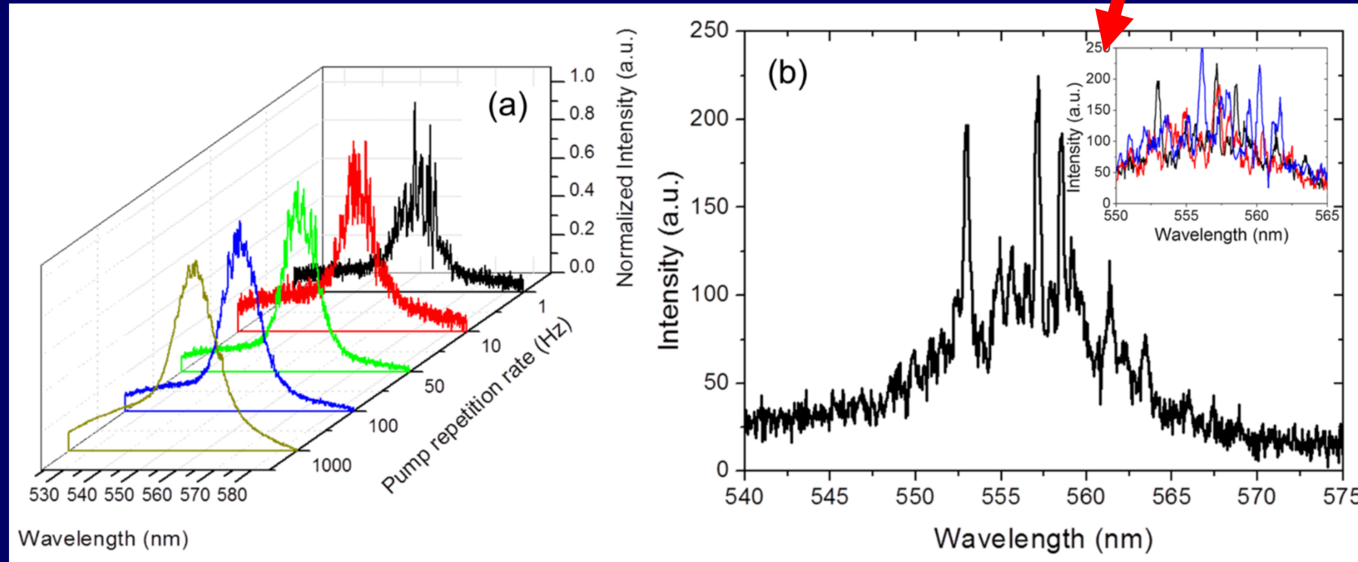
Dye: APSS in DMSO
TiO_2 NPs dia:250 nm $2 \times 10^9/cm^3$

1 kHz

Single shot



Spectral profile versus repetition rate



Nanoscale

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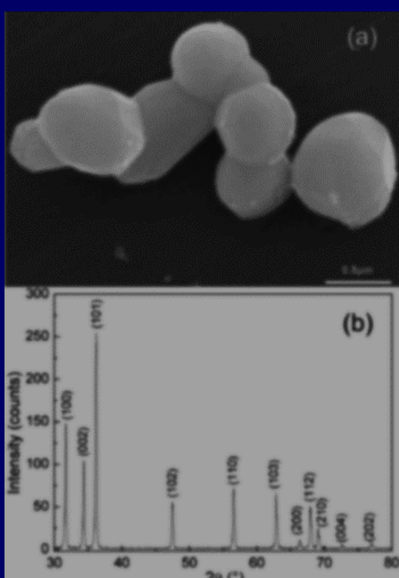
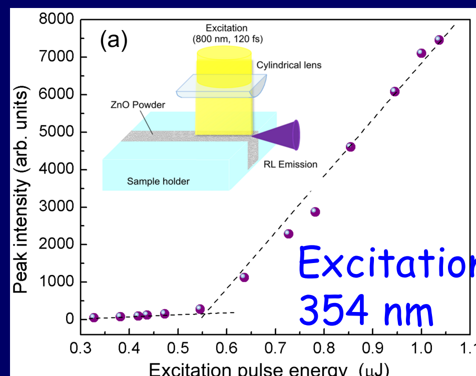
Cite this: *Nanoscale*, 2015, **7**, 317

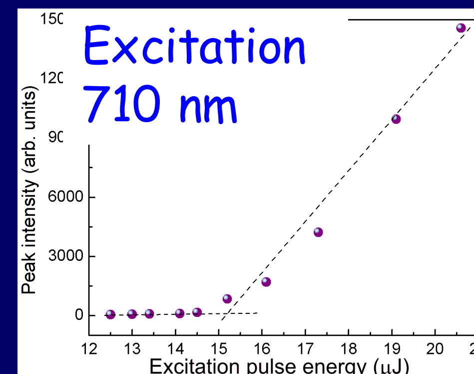
Multi-photon excited coherent random laser emission in ZnO powders

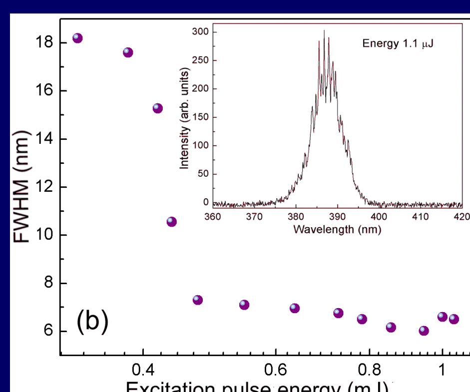
Christian Tolentino Dominguez,*[a,c] Maria de A. Gomes,[b] Zélia S. Macedo,[b] Cid B. de Araújo[c] and Anderson S. L. Gomes*[c]

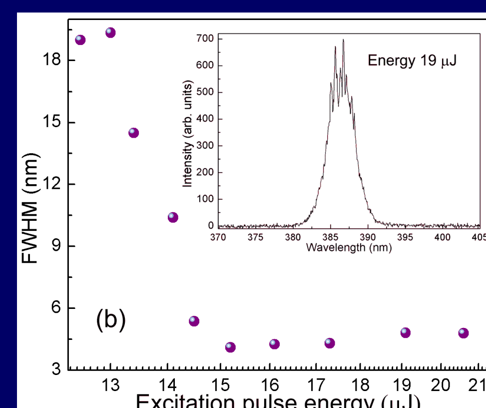
ZnO grains









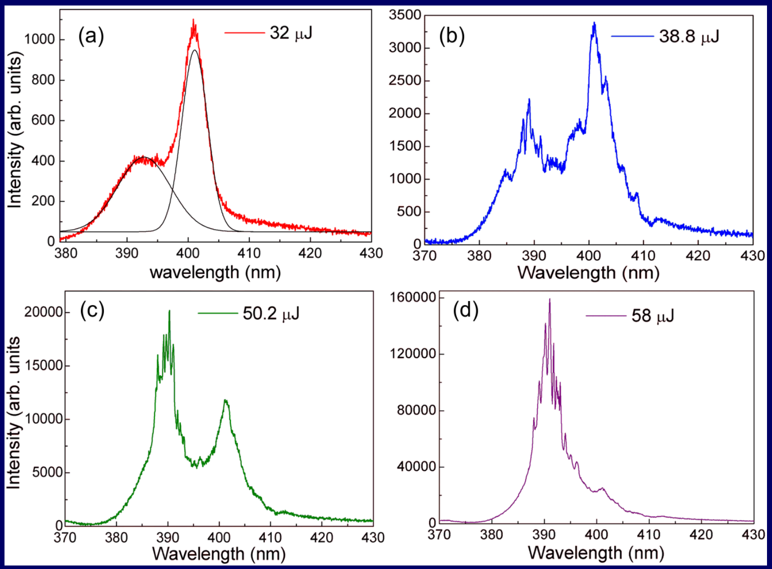


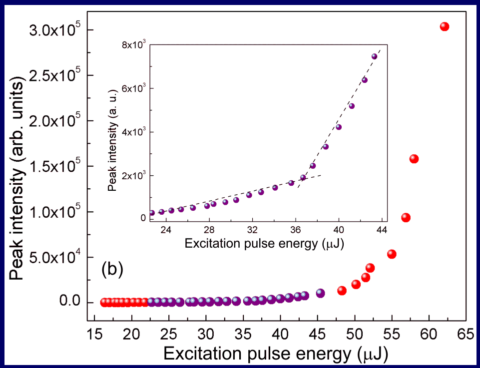
One-photon excitation

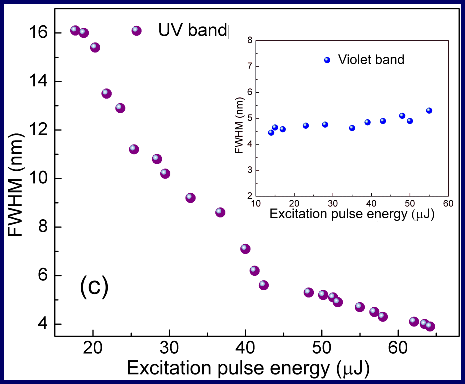
Two-photon excitation

Three – photon excitation

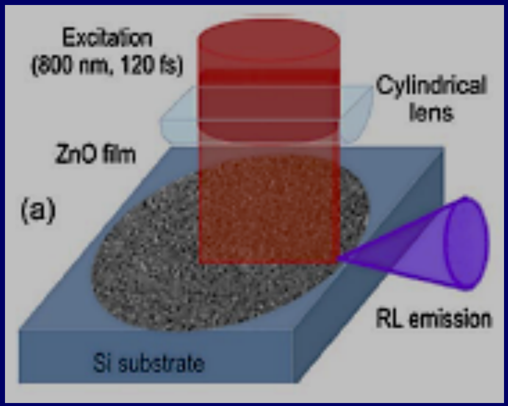
Excitation: 802 nm







Also ZnO films



Dominguez et al.
J. Opt. Soc. Am. B 31 (2014) 1975

Second-order parametric effects in Random Lasers

$$P = \varepsilon_o \chi^{(1)} E + \varepsilon_o [\chi^{(2)} E^2 + \chi^{(3)} E^3 + \chi^{(4)} E^4 ...]$$

linear

nonlinear

$\chi^{(2)}$ Second order polarization

$$P^{(2)} = \varepsilon_0 \chi^{(2)} \left(E_1 \cos \omega_1 t + E_2 \cos \omega_2 t \right)^2 =$$

$$P_0^{(2)} + P_{2\omega_1}^{(2)} + P_{2\omega_2}^{(2)} + P_{\omega_1+\omega_2}^{(2)} + P_{\omega_1-\omega_2}^{(2)}$$

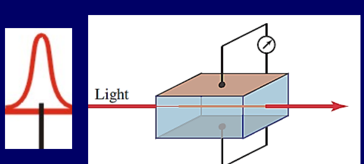
Optical retification

SHG

ω

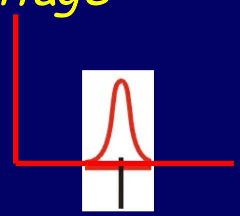
SFG

DFG

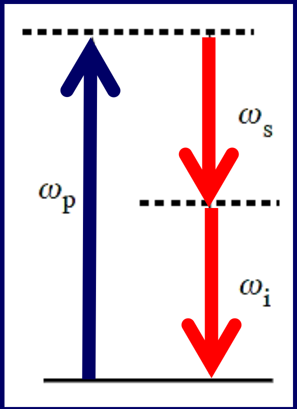


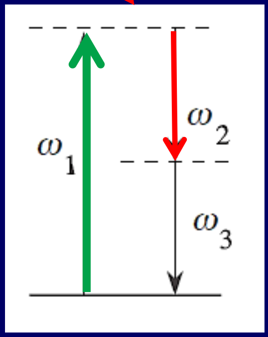
Light pulse

Voltage



time





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2015

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Cite this: J. Mater. Chem. C, 2015, 3, 11689

Structural and luminescence properties of Nd^{3+}/Yb^{3+} codoped $Al_4B_2O_9$ nanocrystalline powders†

Lauro J. Q. Maia,*[a] Fausto M. Faria Filho,[a] Vladimir Jerez,[bc] André L. Moura[cd] and Cid B. de Araújo[c]

SCIENTIFIC REPORTS

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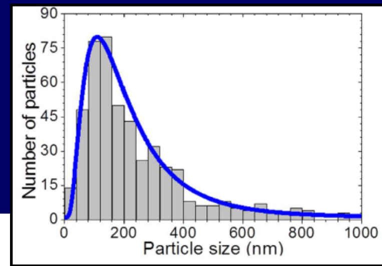
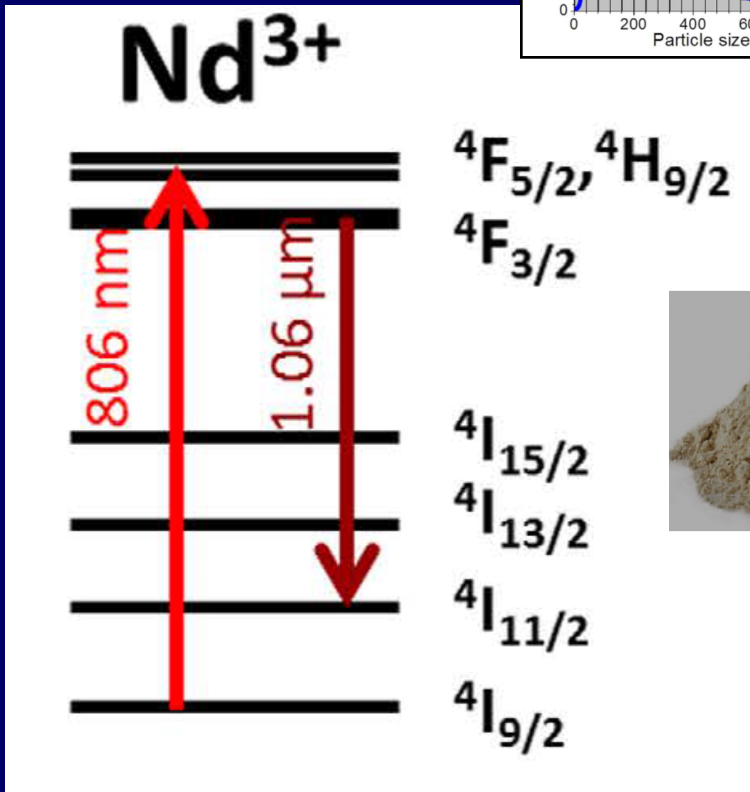
Multi-wavelength emission through self-induced second-order wave-mixing processes from a Nd^{3+} doped crystalline powder random laser

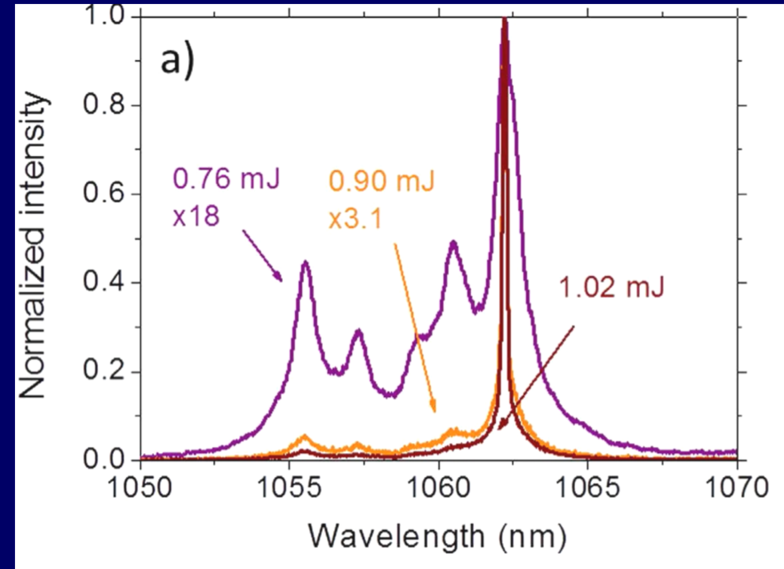
André L. Moura[1,2], Vladimir Jerez[2,3], Lauro J. Q. Maia[4], Anderson S. L. Gomes[2] & Cid B. de Araújo[2]

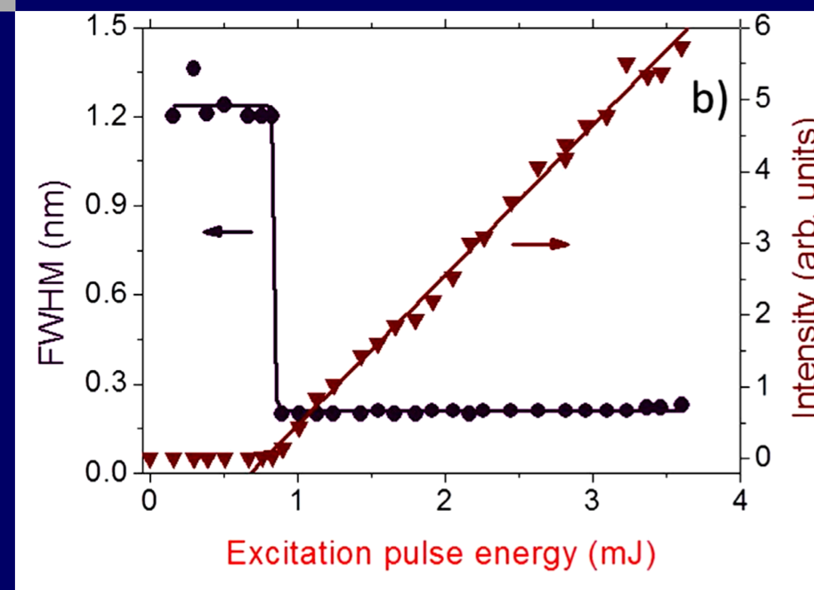
Sci. Rep. 5, 13816 (2015)

$\chi^{(2)}$ Nd^{3+}: $YAl_3(BO_3)_4$





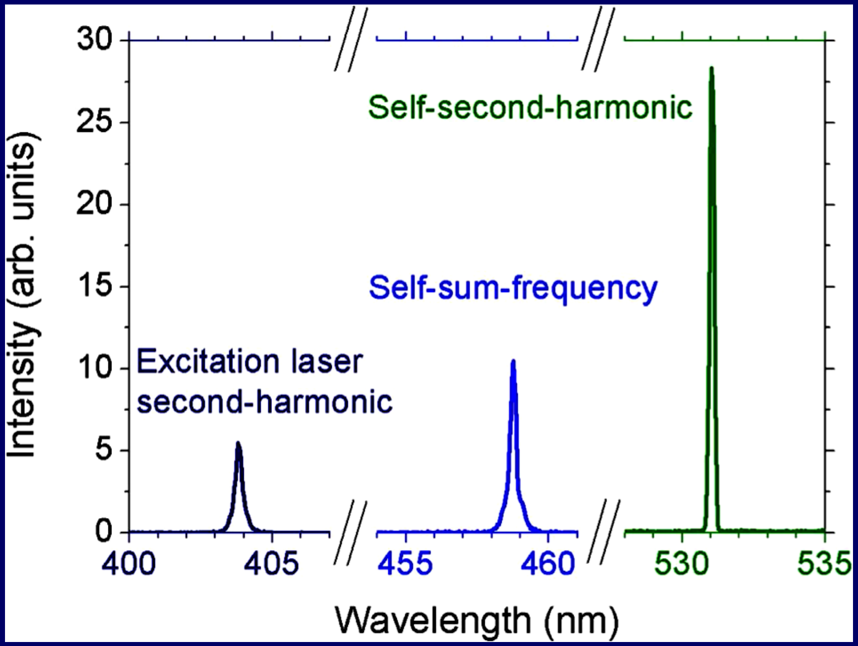


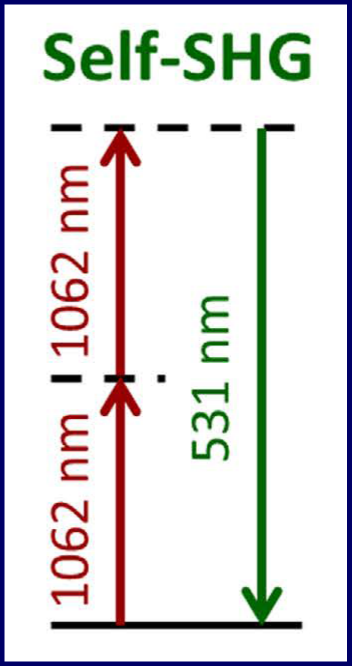


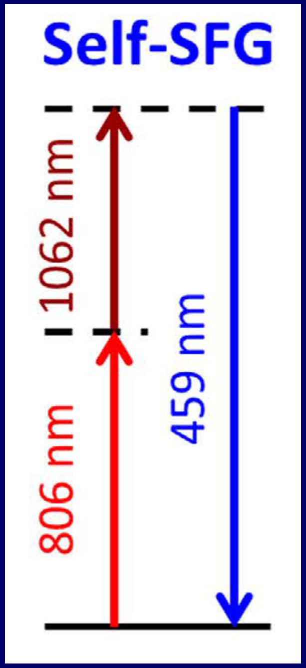
Stimulated emission + multiple light scattering + second harmonic generation + sum frequency generation

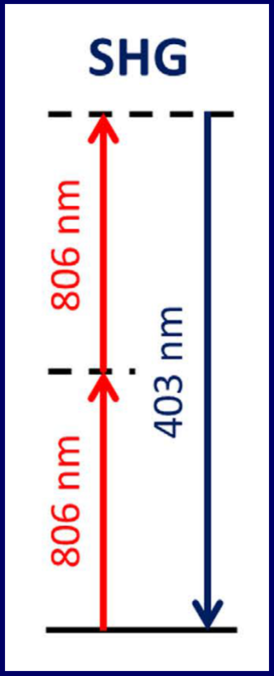
Self-frequency conversion

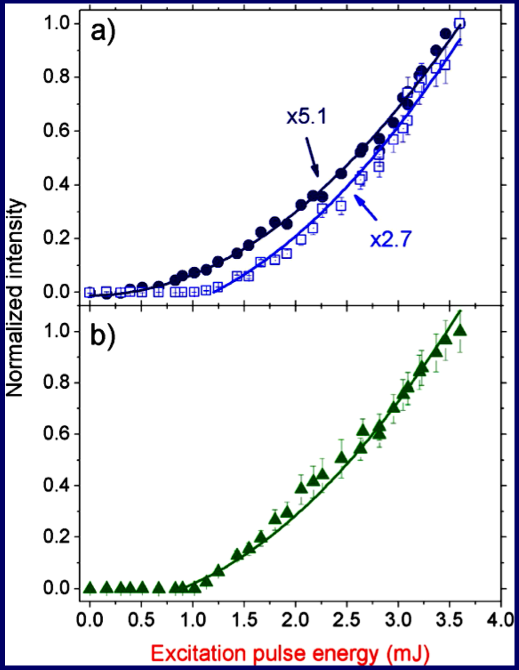
$Nd_{0.04}Y_{0.96}Al_3(BO_3)_4$









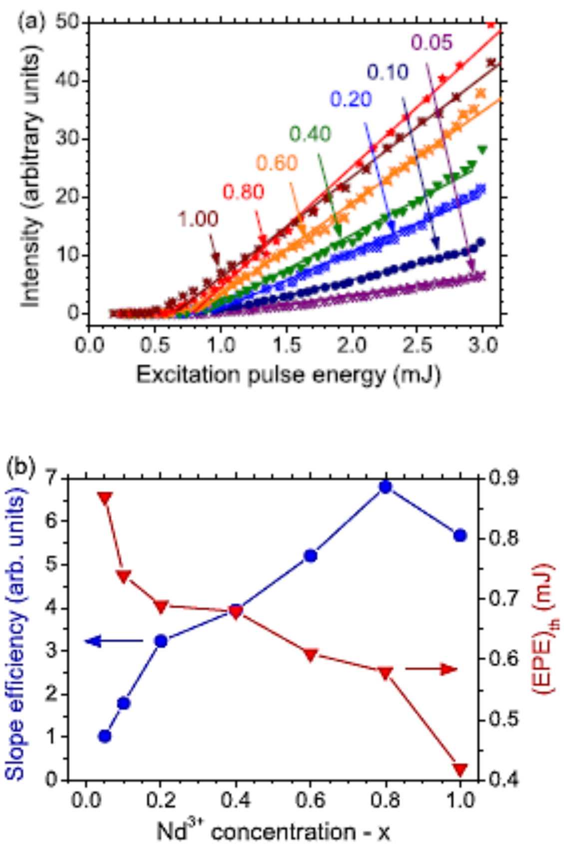


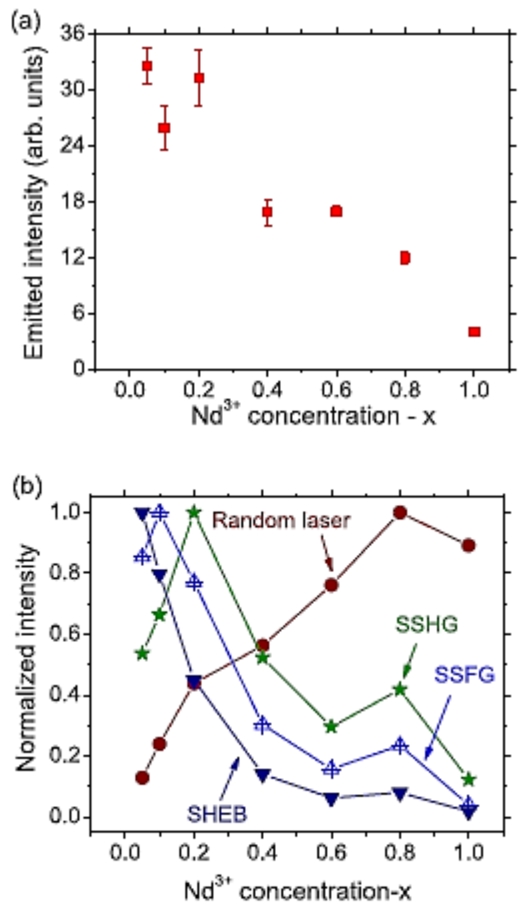
Optical Materials 54 (2016) 262-268

Interplay between random laser performance and self-frequency conversions in $Nd_xY_{1.00-x}Al_3(BO_3)_4$ nanocrystals powders

CrossMark

Sandra J.M. Carreño [a], André L. Moura [a,b,*], Pablo I.R. Pincheira [a], Zanine V. Fabris [c], Lauro J.Q. Maia [c], Anderson S.L. Gomes [a], Cid B. de Araújo [a]





SCIENTIFIC REPORTS

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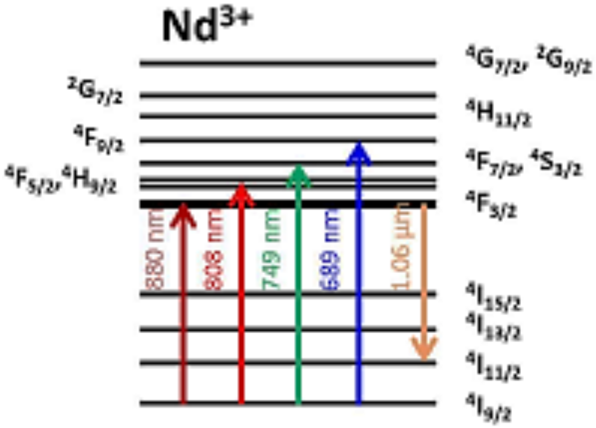
Tunable ultraviolet and blue light generation from Nd:YAB random laser bolstered by second-order nonlinear processes

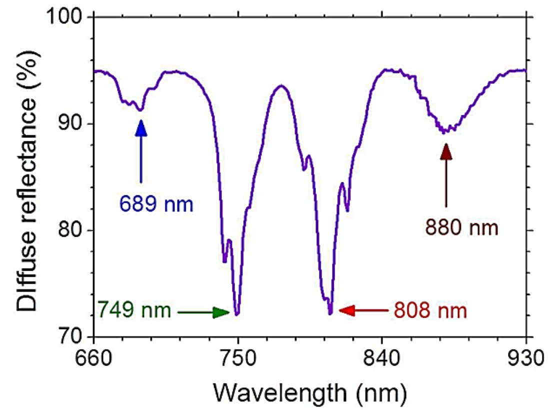
Received: 18 March 2016
Accepted: 11 May 2016
Published: 01 June 2016

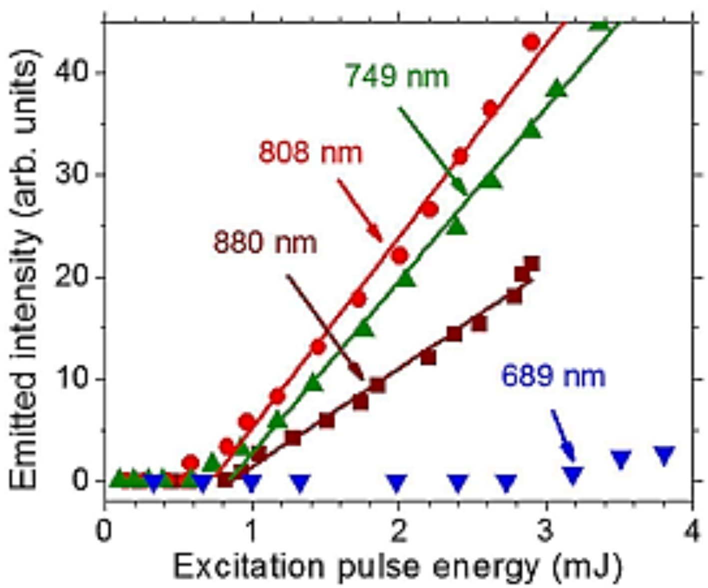
André L. Moura[1], Sandra J. M. Carreño[2], Pablo I. R. Pincheira[2], Zanine V. Fabris[3], Lauro J. Q. Maia[3], Anderson S. L. Gomes[2] & Cid B. de Araújo[2]

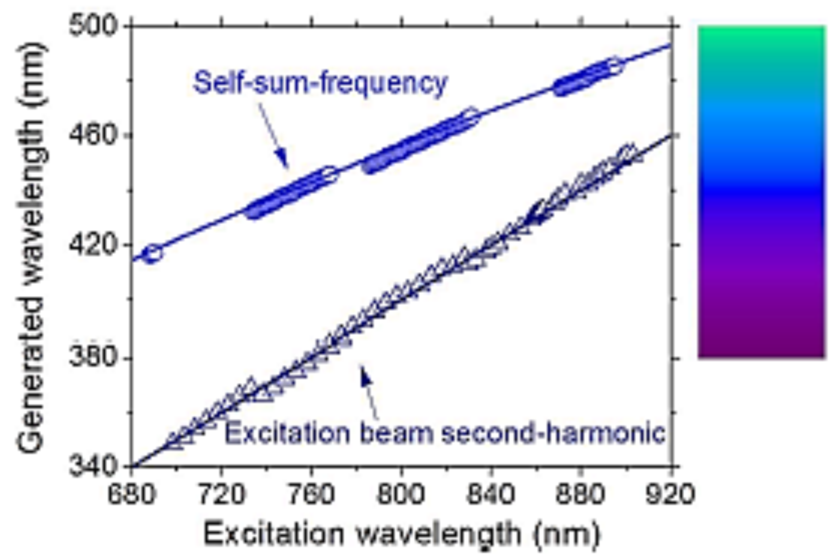
Sci. Reports 6 (2016) 27107

$Nd_{0.10}Y_{0.90}Al_3(BO_3)_4$









Glassy Behavior of Light

L. Angelani,[1] C. Conti,[2,3] G. Ruocco,[3,4] and F. Zamponi[3,4]

[1]*Research Center SMC INFM-CNR, c/o Università di Roma "La Sapienza," I-00185, Roma, Italy*
[2]*Centro Studi e Ricerche "Enrico Fermi," Via Panisperna 89/A, I-00184, Roma, Italy*
[3]*Research Center Soft INFM-CNR, c/o Università di Roma "La Sapienza," I-00185, Roma, Italy*
[4]*Dipartimento di Fisica, Università di Roma "La Sapienza," I-00185, Roma, Italy*

(Received 22 September 2005; published 16 February 2006)

We study the nonlinear dynamics of a multimode 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 nonlinear medium displays glassy behavior; i.e., the photon gas has a multitude of metastable states and a nonvanishing 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.

nature COMMUNICATIONS

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,

Spin-glass theory is one of the leading paradigms of complex physics and describes condensed matter, neural networks and biological systems, ultracold atoms, random photonics and many other research fields. According to this theory, identical systems under identical conditions may reach different states. This effect is known as replica symmetry breaking and is revealed by the shape of the probability distribution function of an order parameter named the Parisi overlap. However, a direct experimental evidence in any field of research is still missing. 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.

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Sci. Reports 6, 27987 (2016)

SCIENTIFIC REPORTS

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Accepted: 26 May 2016
Published: 13 June 2016

Observation of Lévy distribution and replica symmetry breaking in random lasers from a single set of measurements

Anderson S. L. Gomes[1], Ernesto P. Raposo[2], André L. Moura[1,3], Serge I. Fewo[4], Pablo I. R. Pincheira[1], Vladimir Jerez[5], Lauro J. Q. Maia[6] & Cid B. de Araújo[1]

Letter Vol. 41, No. 15 / August 1 2016 / *Optics Letters* 3459

Optics Letters

Observation of photonic paramagnetic to spin-glass transition in a specially designed TiO_2 particle-based dye-colloidal random laser

Pablo I. R. Pincheira,[1] Andréa F. Silva,[2] Serge I. Fewo,[3] Sandra J. M. Carreño,[1] André L. Moura,[1,4,*] Ernesto P. Raposo,[5] Anderson S. L. Gomes,[1] and Cid B. de Araújo[1]

From the basic point of view: disordered media are complex nonlinear systems

Excelent plataform to study

mesoscopic transport, laser physics,

nonlinear optics, quantum optics,

statistical physics, quantum chaos,

nonlinear dynamics, atomic physics,...

Thank you for your attention

Our work has been supported by the Brazilian agencies

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