UNIVERSIDADE FEDERAL DE PERNAMBUCO

Departamento de Física

INFO Instituto Nacional de Ciência e Tecnologia de Fotônica

XVII J. A. Swieca School - Curitiba (March, 2023)

Nonlinear Optics

2nd lecture

Cid B. de Araújo

cid.araujo@ufpe.br

http://cidbdearaujo.com

mini-course – Nonlinear Optics:

1st lecture

Introduction to nonlinear optics (NLO)

2nd lecture

Nonlinear Nanophotonics in Plasmonic media

3rd lecture

Stimulated Emission and Random Lasers

Research of new materials for NLO

- To increase the efficiency of new frequencies generation
- Reduce the size of photonic devices
- Reduce the optical power levels required for NLO
- Optical processing with low power levels

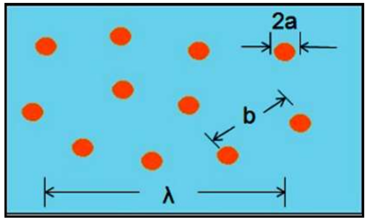
etc, etc, etc.....

Materials: metal NPs, nanoclusters of metal atoms, crystalline nanopowders, glasses doped with NPs, liquid colloids, organic membranes, semiconductor "quantum dots"

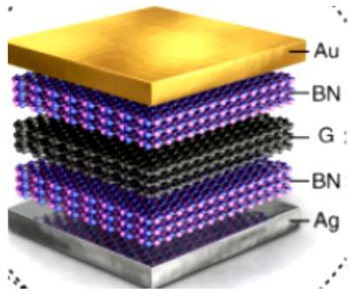
NANOMATERIALS

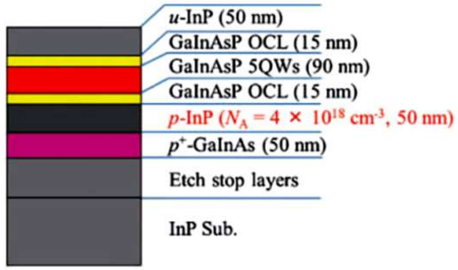
How to increase the NLO response of materials?

Examples

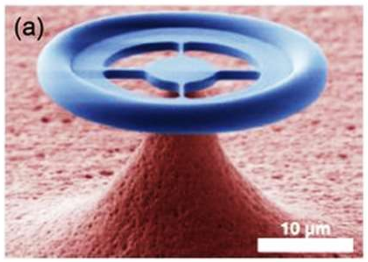


Nanocomposites
Inclusions with larger NL response than the host

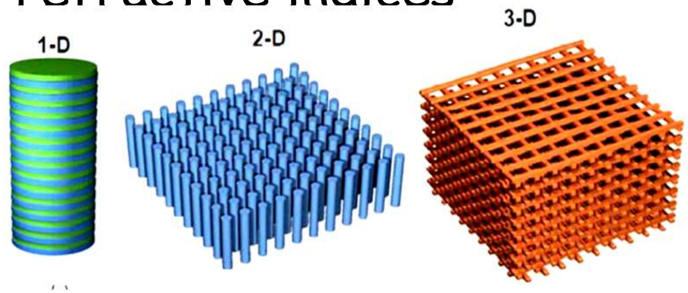




Hetero-structures with components having different refractive indices



Micro-cavities with large quality factor



Photonic crystals

When humans started manipulating objects in the nanoscale?

Roman nanotechnology

Lycurgus cup
<u>400 yrs A.D.</u>

Gold + silver NPs embeded in the glass

Gold-Doped Glass: A Maxwell-Garnett Composite

Developmental Glass, Corning Inc.

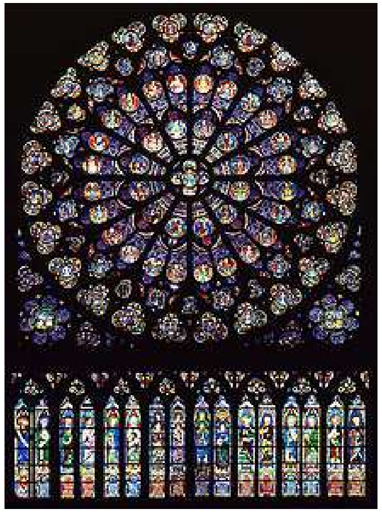
Red Glass Caraffe
Nurenberg, ca. 1700
Huelsmann Museum, Bielefeld

gold volume fraction approximately 10^{-6}
gold particles approximately 10 nm diameter

- Composite materials can possess properties very different from those of their constituents.
- Red color is because the material absorbs very strong in the blue, at the surface plasmon frequency

Slide: R. W. Boyd

Middle age



Cu, Ag, Au nanoparticles

Michael Faraday's Recognition of Ruby Gold: the Birth of Modern Nanotechnology

His 1857 Lecture to the Royal Society in London

David Thompson
DTThompson@aol.com

Figure 1
Faraday's colloidal ruby gold. Reproduced by Courtesy of the Royal Institution of Great Britain

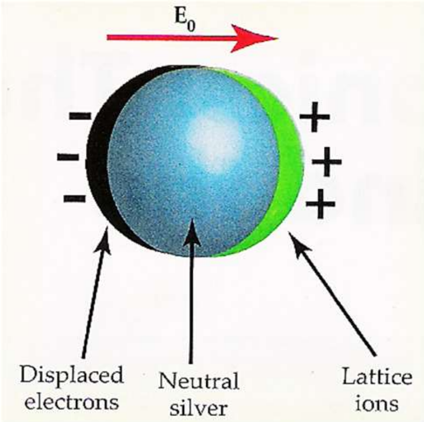
From the paper in Philosophical Transactions entitled 'Experimental relations of gold (and other metals) to light' (1), based on his Bakerian lecture to the Royal Society in London on 5 February 1857, it is clear that Michael Faraday was fascinated by the ruby colour of colloidal gold. The objective of his investigations was to examine the interaction of light with metal particles, but much of this paper focused on various solution such as phosphorus in carbon disulfide in a two phase system. The yellow colour of sodium chloroaurate ($NaAuCl_4$) changes within minutes to the deep ruby colour of colloidal gold. Faraday concluded that the ruby fluid was gold dispersed in the liquid in a very finely divided metallic form not visible in any of the microscopes available in his day. Nearly 100 years later Turkevich et al (2, 4) used electron microscopic

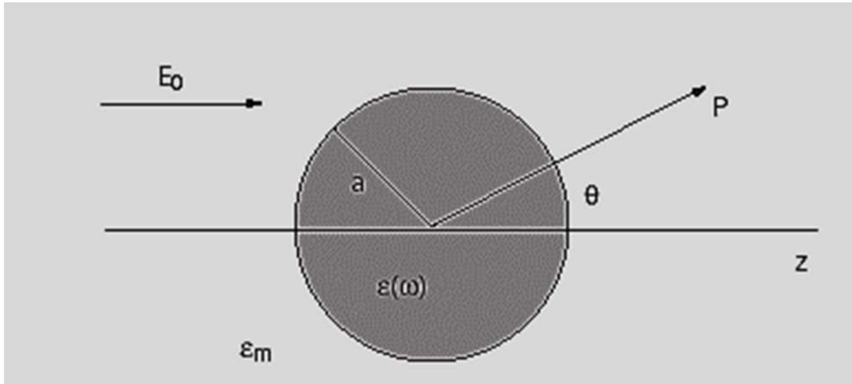
Experimental Relations of Gold (and Other Metals) to Light, M. Faraday, Philos. Trans. R. Soc. London, 147 (1857) 145-181

Metal nanoparticles for NLO

Dimensions smaller than 100 nm

Quase-static approximation





Solve Laplace's equation

$$\Phi(r,\theta) = \sum_{l=0}^{\infty}\left[A_l r^l + B_l r^{-(l+1)}\right] P_l(\cos\theta)$$

$$\Phi_{\text{in}} = -\frac{3\varepsilon_m}{\varepsilon + 2\varepsilon_m} E_0 r \cos\theta$$

$$\Phi_{\text{out}} = -E_0 r \cos\theta + \frac{\varepsilon - \varepsilon_m}{\varepsilon + 2\varepsilon_m} E_0 a^3 \frac{\cos\theta}{r^2}.$$

$$\Phi_{\mathrm{out}} = -E_0 r \cos\theta + \frac{\mathbf{p}\cdot\mathbf{r}}{4\pi\varepsilon_0\varepsilon_m r^3}$$

$$\mathbf{p} = 4\pi\varepsilon_0\varepsilon_m a^3 \frac{\varepsilon - \varepsilon_m}{\varepsilon + 2\varepsilon_m}\mathbf{E}_0.$$

$$\mathbf{p} = \varepsilon_0\varepsilon_m \alpha \mathbf{E}_0$$

$$\alpha = 4\pi a^3 \frac{\varepsilon - \varepsilon_m}{\varepsilon + 2\varepsilon_m}.$$

Polarizability

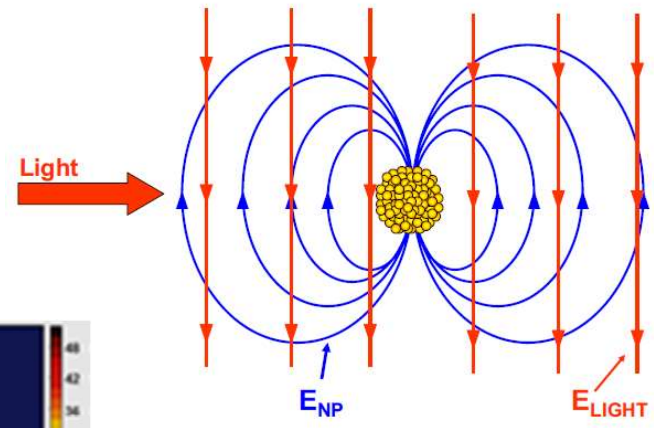
$$\mathbf{E}_{\mathrm{in}} = \frac{3\varepsilon_m}{\varepsilon + 2\varepsilon_m}\mathbf{E}_0$$

$$\mathbf{E}_{\mathrm{out}} = \mathbf{E}_0 + \frac{3\mathbf{n}(\mathbf{n}\cdot\mathbf{p}) - \mathbf{p}}{4\pi\varepsilon_0\varepsilon_m}\frac{1}{r^3}.$$

$$\mathrm{Re}\left[\varepsilon(\omega_{sp}) + 2\varepsilon_m(\omega_{sp})\right] = 0$$

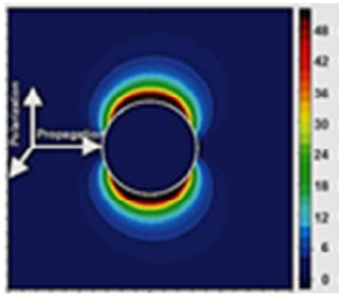
Surface plasmon resonance

Metal NPs as nanoantennae

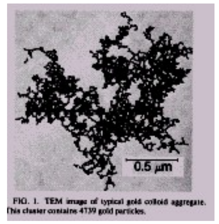


$$E_{local} = \boldsymbol{L} E_{light}$$

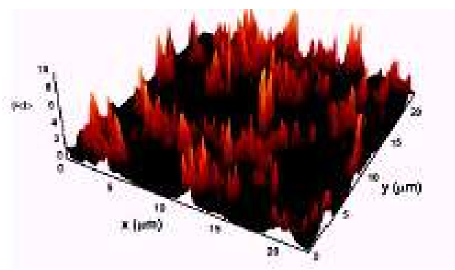
Local field factor



$$\boldsymbol{L} = \frac{3\epsilon_h(\omega)}{[\epsilon_{NP}(\omega) + 2\epsilon_h(\omega)]}$$



Fractals and "hot-spots"



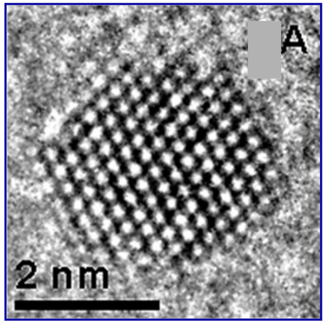
$$Re\ [\epsilon_{NP}(\omega_{SP}) + 2\epsilon_h(\omega_{SP})] = 0$$

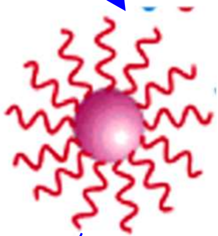
Localized surface plasmons resonance

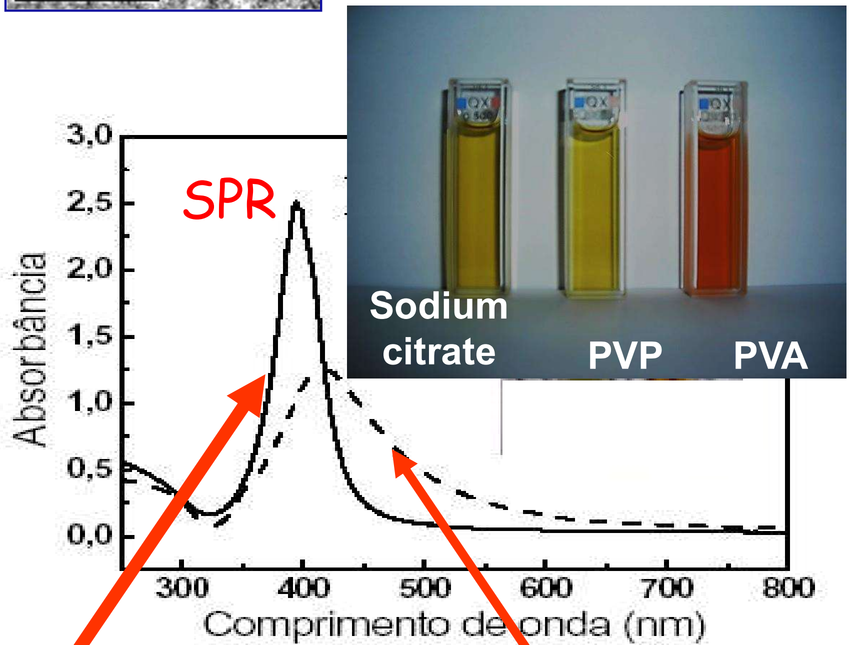
Optical response increase much when $\omega \approx \omega_{SP}$

Colloids with Ag-NPs (spheres)

Stabilizing aggents to prevent aggregation





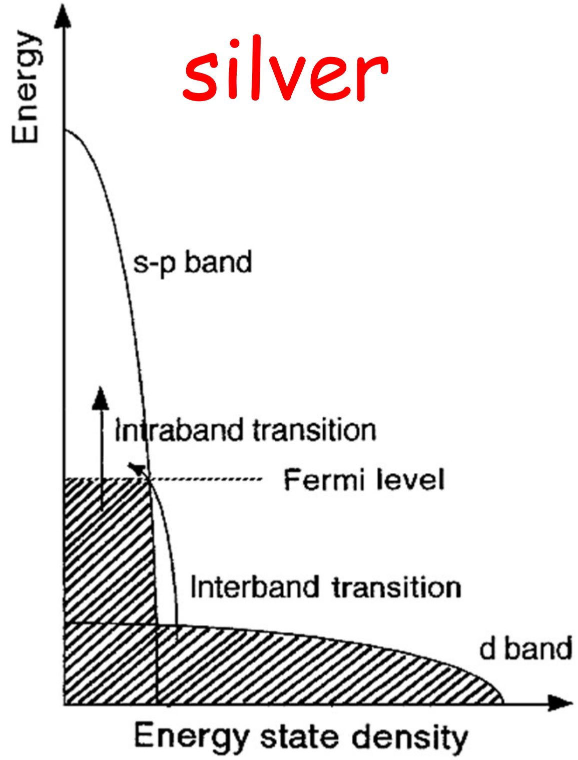


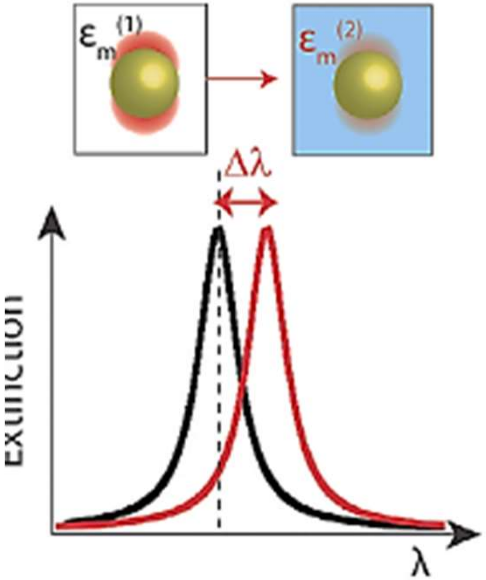
SPR

spheres

Many shapes and sizes

silver



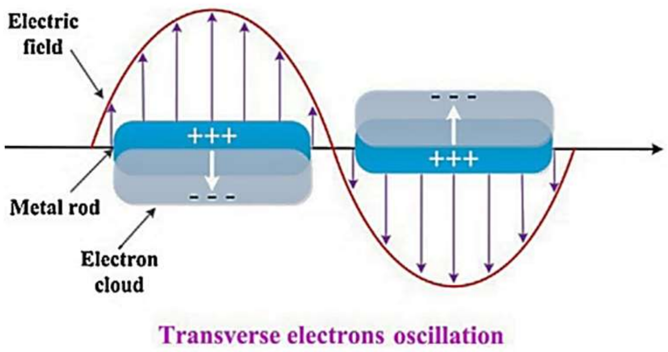


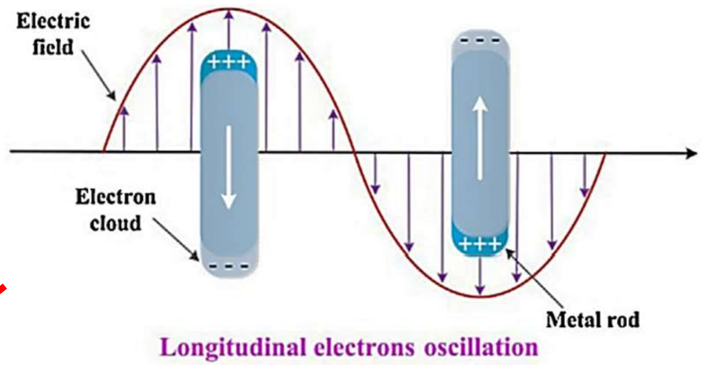
diameter: 4 nm

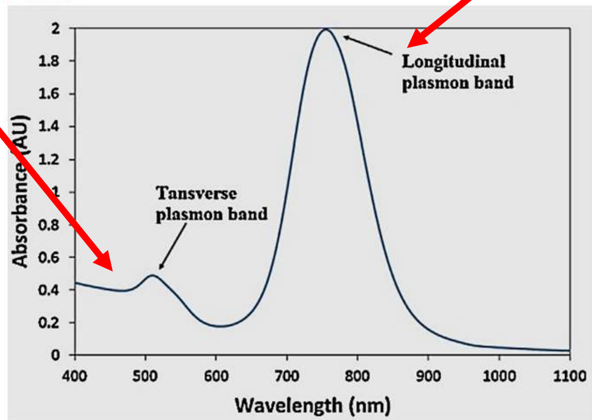
≈ 1500 atoms

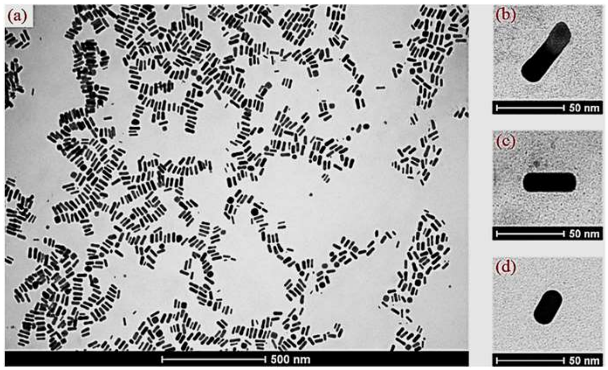
≈ 30% on surface

Gold nanorods



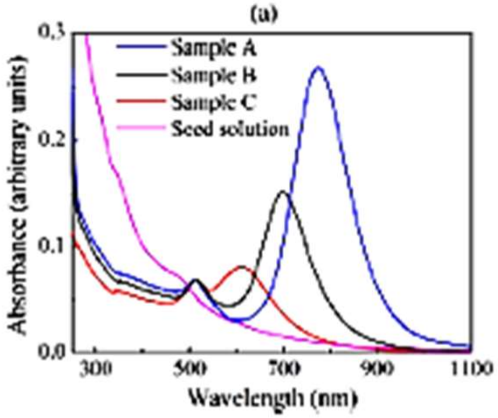


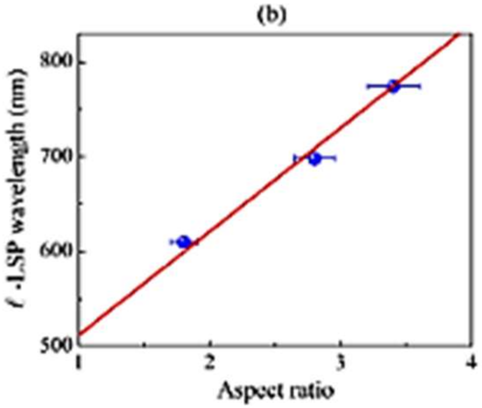




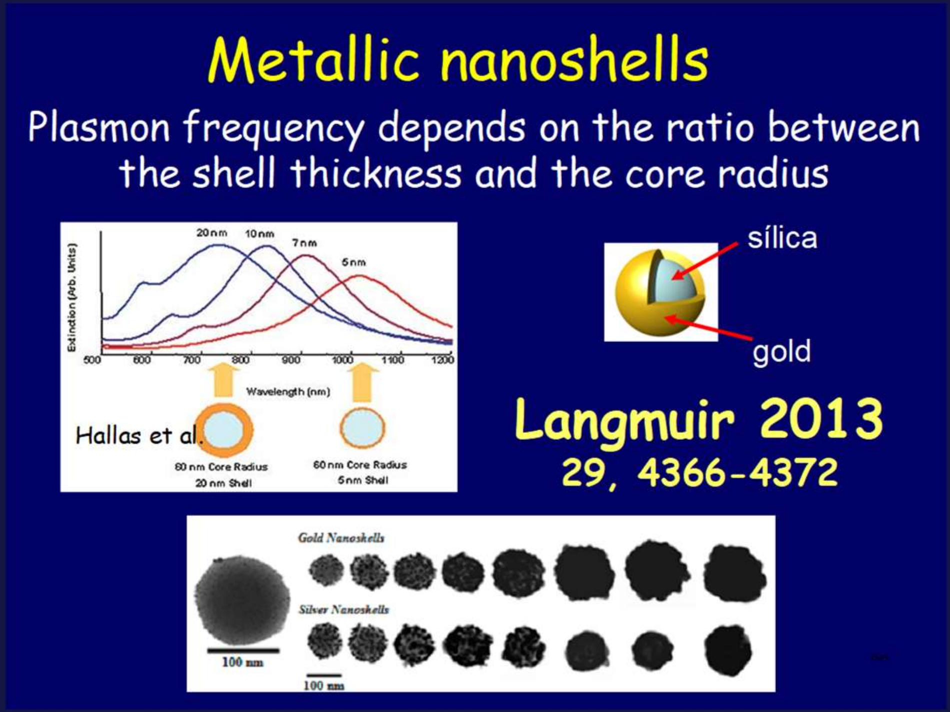
Aspect ratio

A: 3.4
B: 2.8
C: 1.8





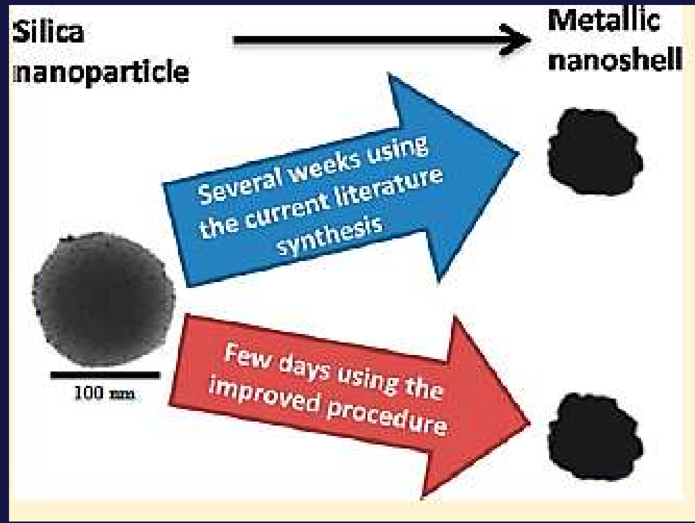
Oliveira et al. J. Phys. Chem. C 123 (2019) 12997-13008

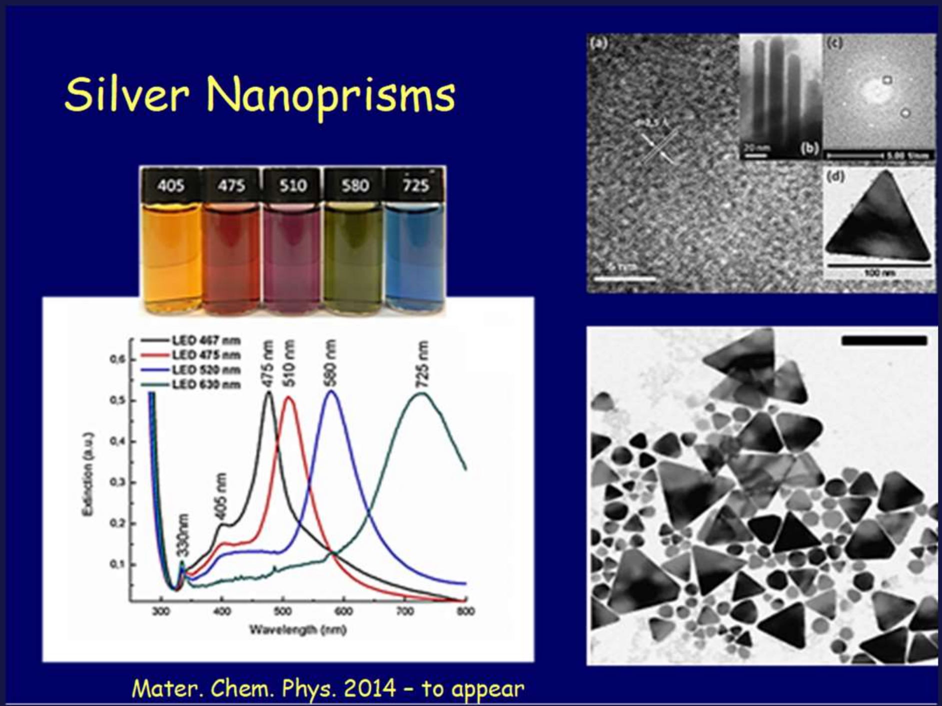


Improved synthesis of gold and silver nanoshells

Brito-Silva et al.

Langmuir 29 (2013) 4366



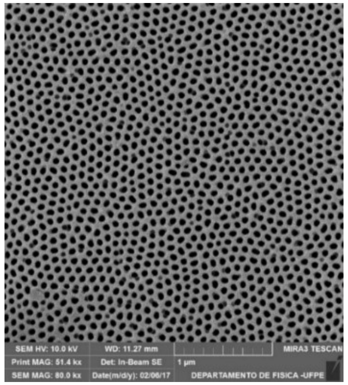


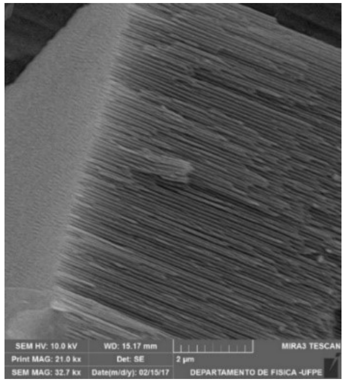
Synthesis of silver nanoprisms:
A photochemical approach
using light emission diodes

Saade, de Araújo

Mater. Chem. Phys. 148 (2014) 1184

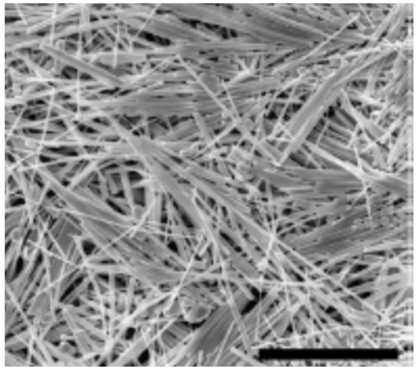
Silver nanowires in alumina membranes electrochemistry





Thesis: Manoel L Silva-Neto UFPE 2021

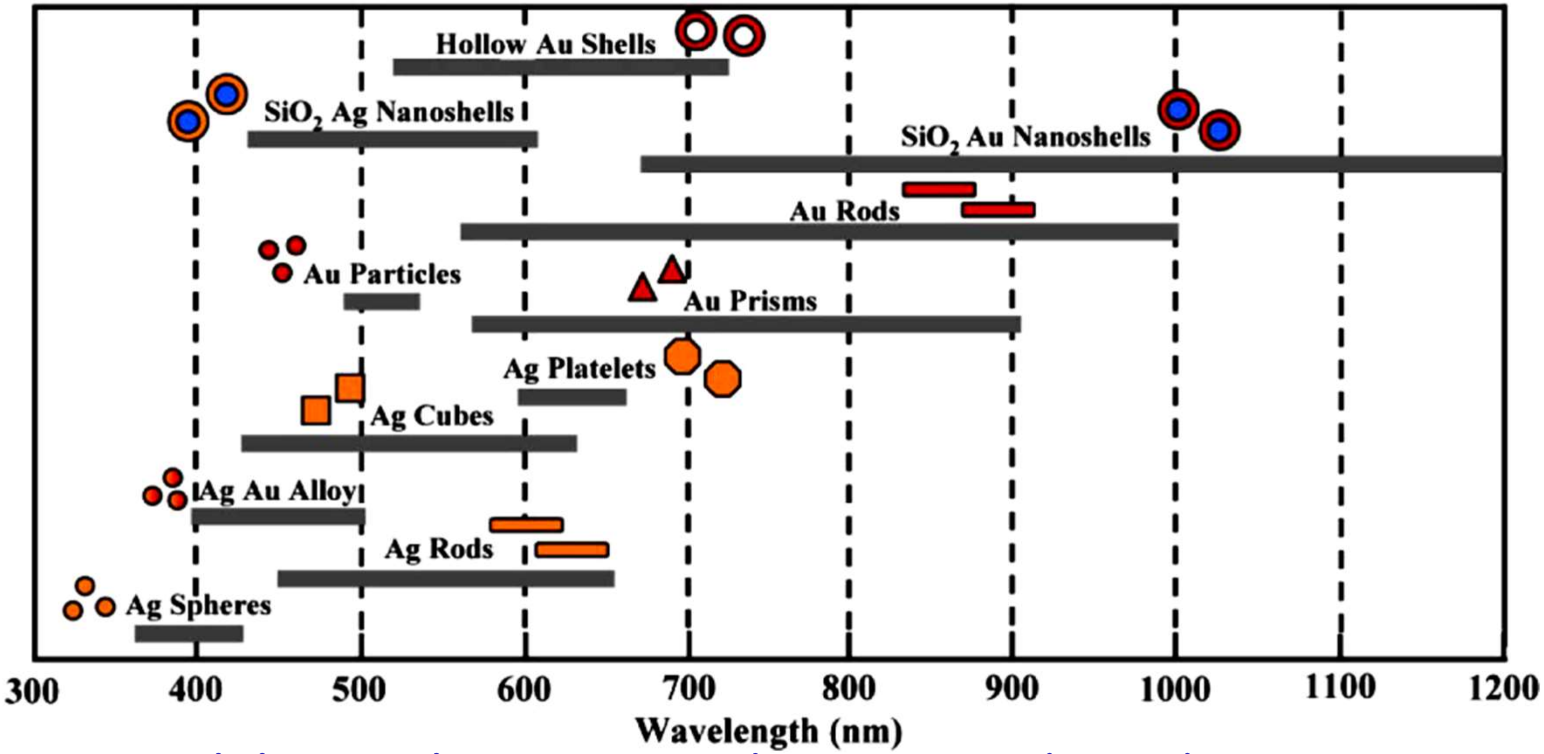
Silver nanowires (chemical synthesis)



PVP-assisted reaction in EG and employing NaCl

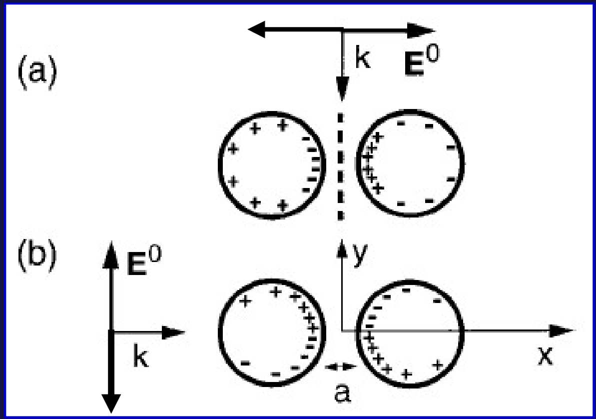
5 microns

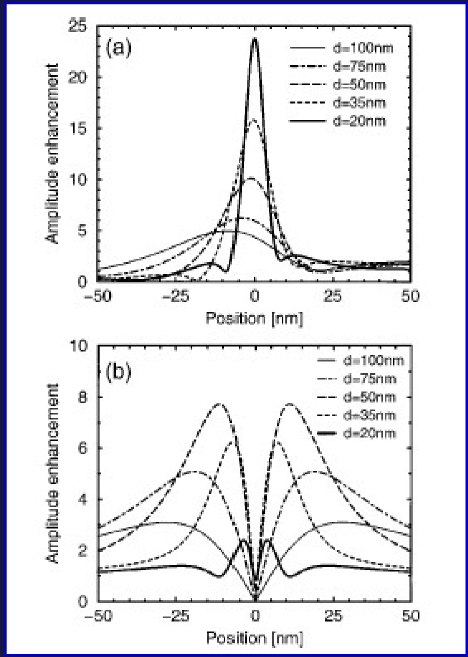
Thesis: Talita Oliveira UFPE 2021



Kalele et al., J. Nanophotonics 1 (2007) 012501

Coupled nanoparticles





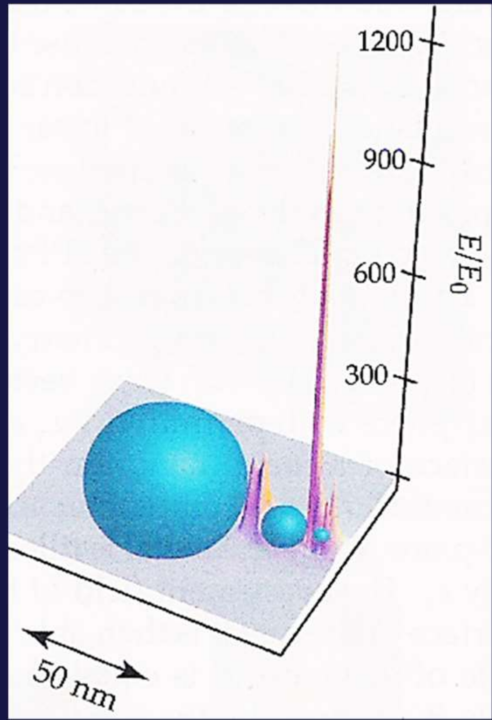
d/a = 5

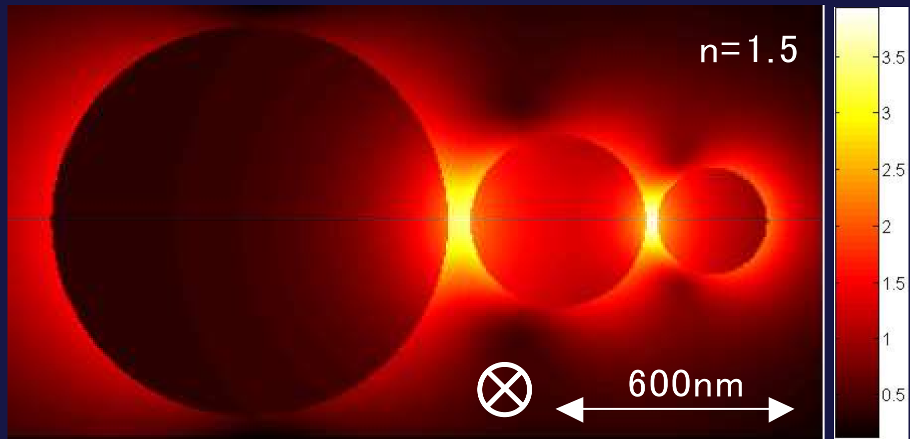
Field increases by 24 for cylinders with diameter d=20 nm.

Intensity enhancement > 500

Kottmann, Martin
Opt. Lett 26 (2001) 1096

Plasmonic nanolens





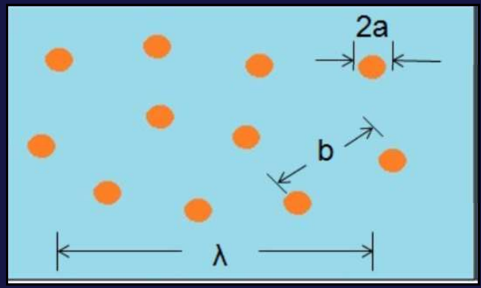
Log E/E_0

Stockman, Physics Today 2011

Metal-dielectric nanocomposites

Nanoparticles

Dielectric



$$\lambda > b > 2a$$

- Glasses (bulk and thin films) with metallic NPs
- Polymers with metallic nanostructures
- Liquid colloids with metallic NPs

Optical response is controlled through the volume fraction of the NPs

Colloids with very small filling fractions

$$10^{-6} < f < 10^{-4}$$

Why metal-dielectric nanocomposites?

Wide range of properties can be chosen to meet the needs of various applications

Control of the optical response through the density and shape of nanoparticles

- Can be doped with large amount of RE ions or transition metals
- Large variety of forms and sizes (ex: fibers and waveguides)
- Nonlinearity can be controlled by changing the NPs concentration

Emission coupling between metal NPs and fluorophores

- SPR does not always enhance the emission properties of the fluorophore.
- Non-radiative decay rate (or quenching) depends on the distance fluorophore - metal particles.
- Emission directionality may be altered

Anger et al. PRL 96 (2006) 113002
Kuhn et al. PRL 97 (2006) 017402

Glasses and glass-ceramics

Kassab, Bordon, Reyna, de Araújo
Nanoparticles-based photonic metal–dielectric composites: A survey of recent results
Optical Materials X 12 (2021) 100098

de Araújo, Kassab, da Silva
Optical properties of glasses and glass-ceramics for optical amplifiers, photovoltaic devices, color displays, optical limiters, and Random Lasers
Optical Materials 131 (2022) 112648

Nonlinear optics of a nanocomposite

$$P_L + P_{NL} = \epsilon_0 \sum_{N=0}^{\infty} \chi_{eff}^{(2N+1)} E^{(2N+1)}$$

Centro-symmetric media

$$\chi_{\text{eff}}^{(\text{even})} = 0$$

Effective 3rd. order susceptibility

$$\chi_{eff}^{(3)} = f\, L^2 \left|L\right|^2 \chi_{np}^{(3)} + \chi_h^{(3)},$$

Local field factor

$$L = 3\varepsilon_h^{(L)} \Big/ \left(\varepsilon_{np}^{(\mathrm{L})} + 2\varepsilon_h^{(L)}\right)$$

Nonlinear response depends strongly on the laser frequency

$$n_2 \propto Re\chi_{eff}^{(3)}$$

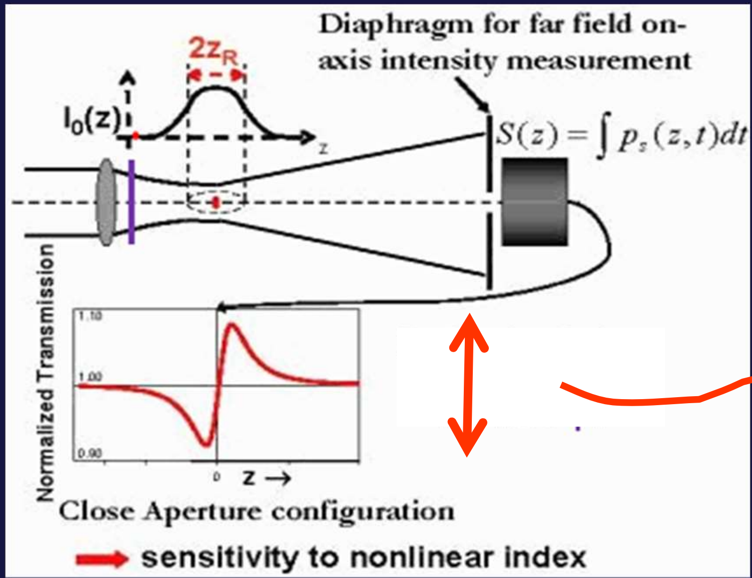
Nonlinear refraction

$$\alpha_2 \propto Im\chi_{eff}^{(3)}$$

Nonlinear absorption

When high-order nonlinearities are present:

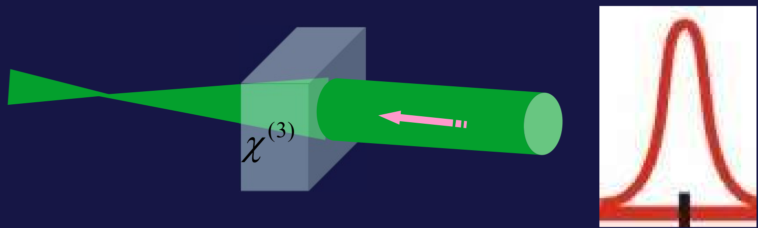
Self - focusing medium



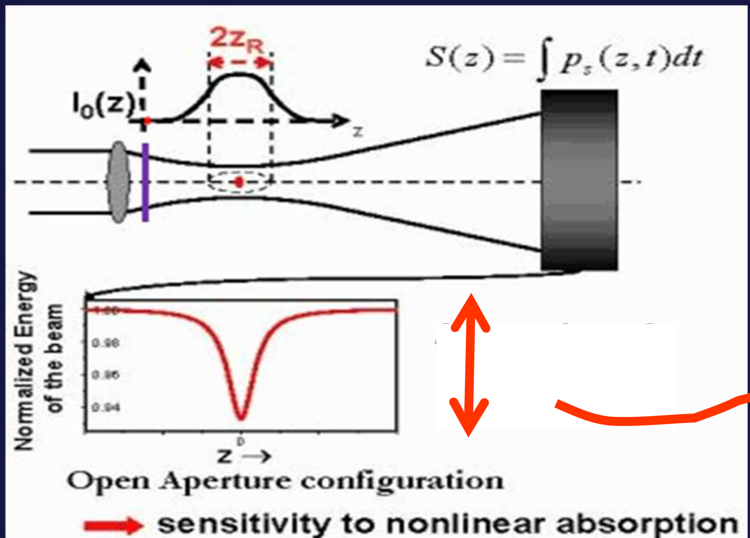
NL refraction

"Closed-aperture" Z scan

$$\Delta T \propto n_2 I + n_4 I^2 + n_6 I^3 + \cdots$$



I (r)



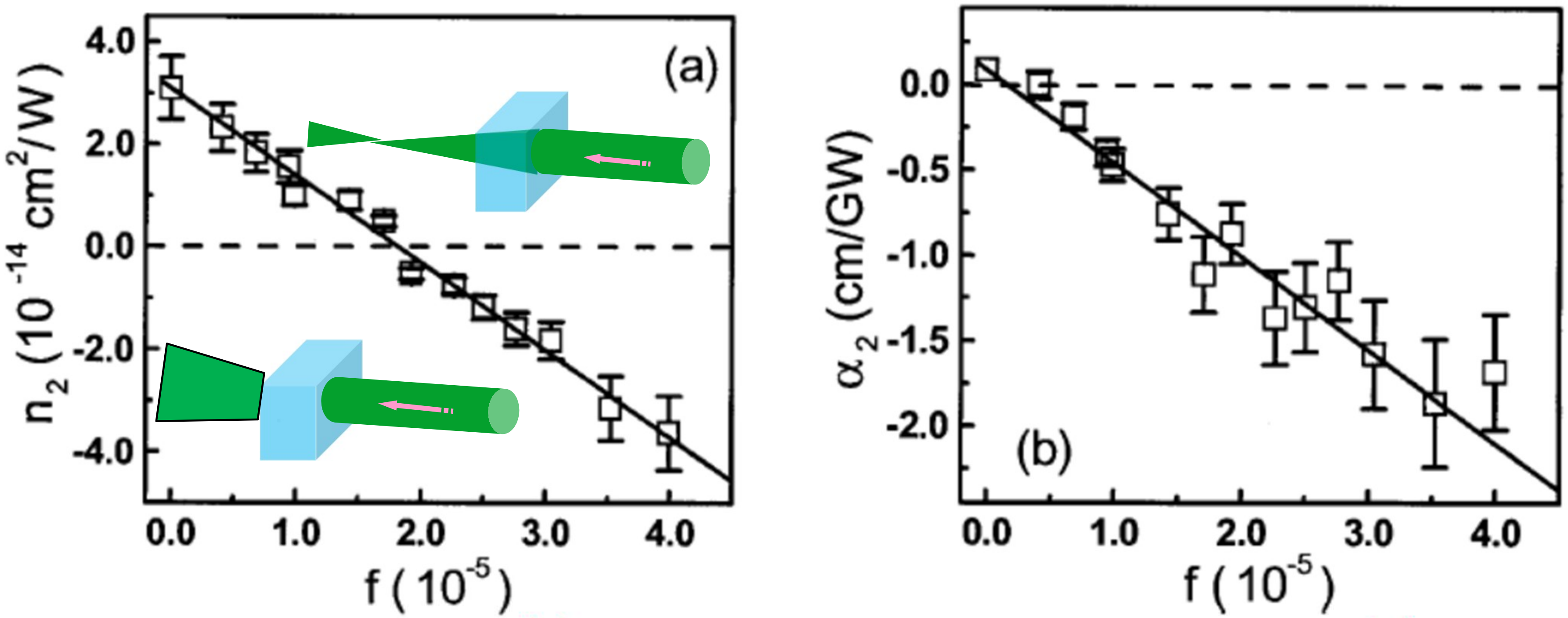
NL absorption

"Open-aperture" Z scan

$$\Delta T \propto \alpha_2 I + \alpha_4 I^2 + \alpha_6 I^3 + \cdots$$

Ag-NPs in CS_2

532 nm; 80 ps; 7 Hz; 1 GW/cm²



$n_2 \propto Re\chi_{eff}^{(3)}$ $\alpha_2 \propto Im\chi_{eff}^{(3)}$

$$\chi_{eff}^{(3)} = \chi_h^{(3)} + fL^2|L|^2\chi_{np}^{(3)} \qquad L = 3\epsilon_h/(\epsilon_{np} + 2\epsilon_h)$$

$$\chi_{np}^{(3)} = -(6.3 - i1.9) \times 10^{-16}(m/V)^2$$

Falcão Filho et al. J. Opt. Soc. Am. B 22 (2005) 2444-2449

Large optical nonlinearity and fast response

Fast response is due to the

induced dipole relaxation

Surface plasmon optical dephasing, T_2

Measured using the "Persistent Hole-Burning Technique"

$$T_2 < 3fs$$

T_2 is influenced by the environment

Almeida et al., Appl. Phys. B 108 (2012) 9-16

Falcão-Filho, de Araújo, Rodrigues, Jr.

High-order nonlinearities of aqueous colloids containing silver nanoparticles

J. Opt. Soc. Am. B 24 (2007) 2948-2956

$\chi^{(3)}, \chi^{(5)}, \chi^{(7)}, \chi^{(9)}$

Reyna, de Araújo

Nonlinearity management of photonic composites and observation of spatial-modulation instability due to quintic nonlinearity

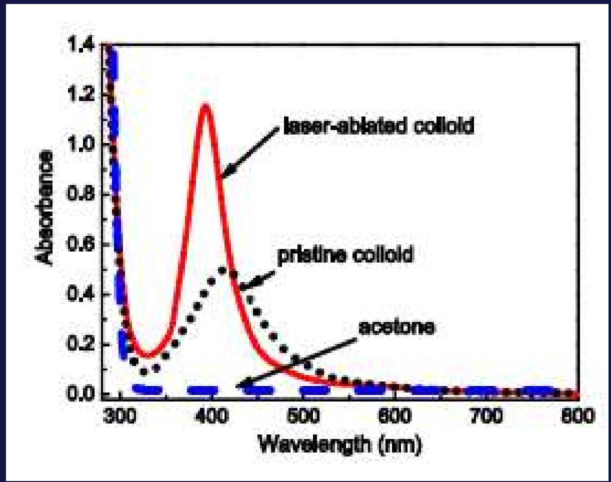
Phys. Rev. A 89 (2014) 063803

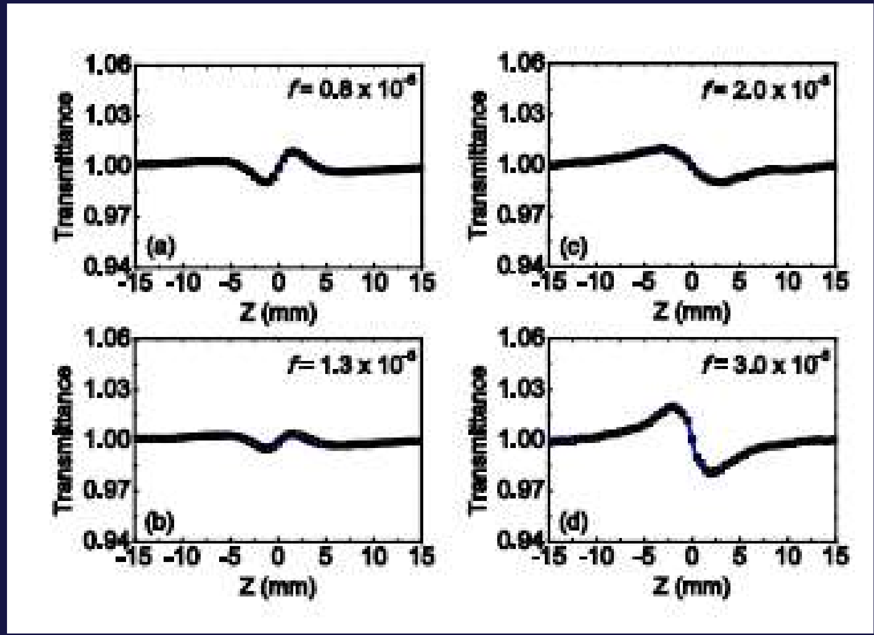
Changing the NPs concentration we may control the NL response of the composites

$\chi_{eff}^{(3)}$ $\chi_{eff}^{(5)}$ $\chi_{eff}^{(7)}$ ... .

Observation of fifth-order refraction in a metal-colloid

Silver NPs in acetone





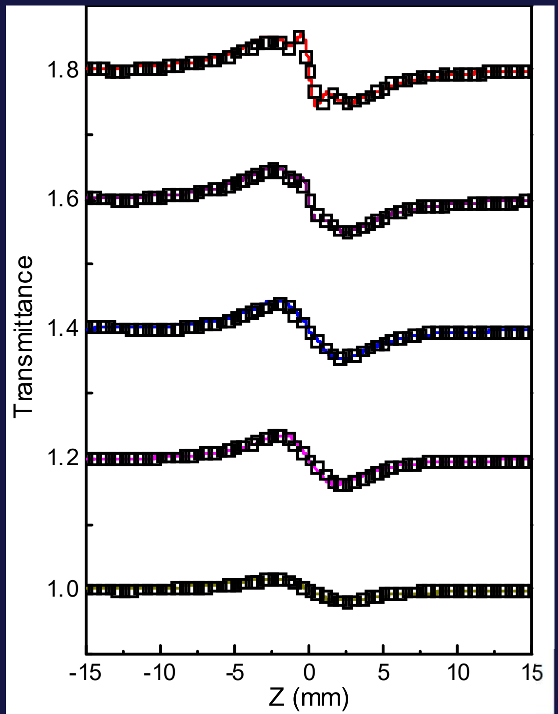
Z-scan

532 nm

Single pulses

5 GW/cm²

$f = 5.0 \ x \ 10^{-5}$



9

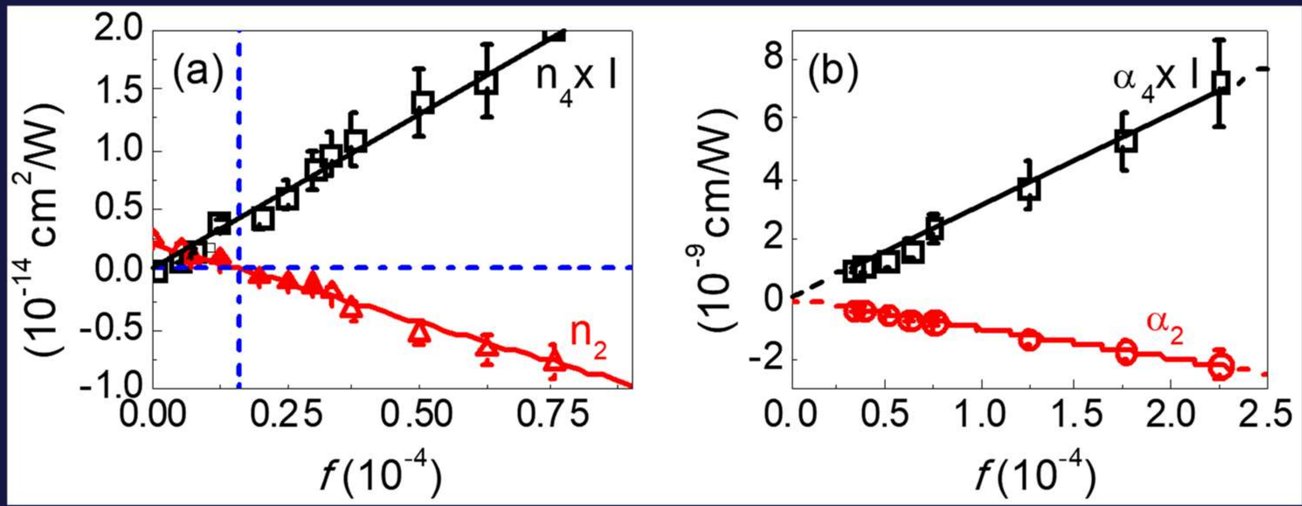
8

6

4

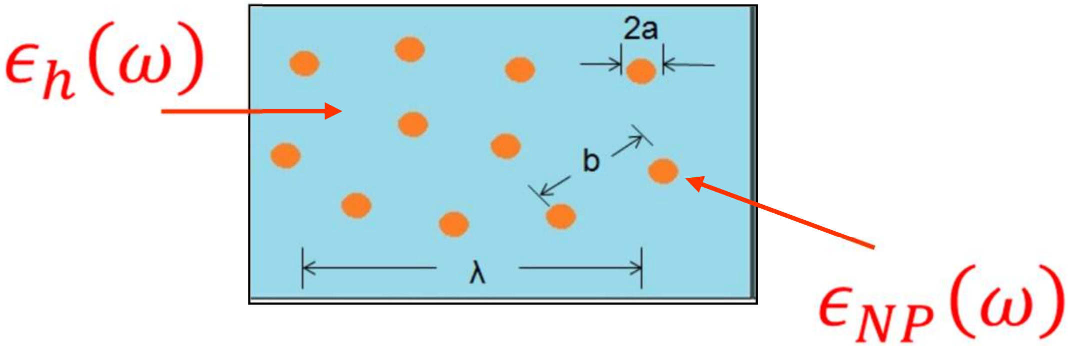
2

GW/cm^2



9 GW/cm²

Generalized Maxwell-Garnet model



Ag NPs in acetone

$\chi^{(5)}_{host}$ - negligible

$$\boxed{\begin{array}{c} a < b < \lambda \\ \text{Maxwell-Garnet model} \end{array}}$$

$$\chi^{(3)}_{eff} = f\, L^2 |L|^2 \chi^{(3)}_{np} + \chi^{(3)}_h, \qquad L = 3\varepsilon_h^{(L)} \big/ \left(\varepsilon_{np}^{(L)} + 2\varepsilon_h^{(L)}\right)$$

$$\chi^{(5)}_{eff} = f\, L^2 |L|^4 \chi^{(5)}_{np} - \frac{6}{10} f\, L^3 |L|^4 \left(\chi^{(3)}_{np}\right)^2 - \frac{3}{10} f\, L |L|^6 \left|\chi^{(3)}_{np}\right|^2,$$

$$\chi^{(7)}_{eff} = f\, L^2 |L|^6 \chi^{(7)}_{np} + \frac{12}{35} f\, L^4 |L|^6 \left(\chi^{(3)}_{np}\right)^3 + \frac{3}{35} f |L|^8 \left[4L^2 \chi^{(3)}_{np} + |L|^2 \left(\chi^{(3)}_{np}\right)^*\right] \left|\chi^{(3)}_{np}\right|^2$$
$$- \frac{4}{7} f\, L |L|^6 \left[2L^2 \chi^{(3)}_{np} + |L|^2 \left(\chi^{(3)}_{np}\right)^*\right] \chi^{(5)}_{np}.$$

Reyna, de Araújo, Optics Express 22 (2014) 22456

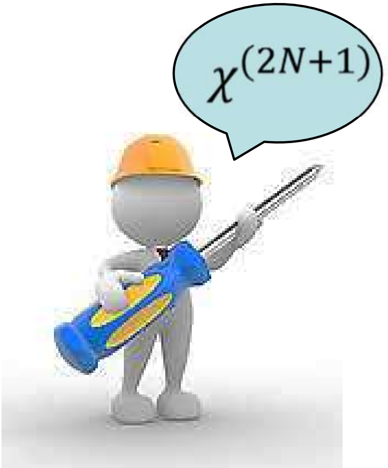
Nonlinearity Management

For nanocomposites it is possible to vary the material's composition and incident light intensity in order to manage its nonlinearity in such way that the material may present HON on demand.

It means that we may, for instance, to adjust the material's parameters to supress one particular NL susceptibility and enhance the others

For example, one may supress the third-order refractive index such that the refraction can be dominate by the fifth-order susceptibility

Nonlinearity Management



A procedure to obtain exotic metal-dielectric composites

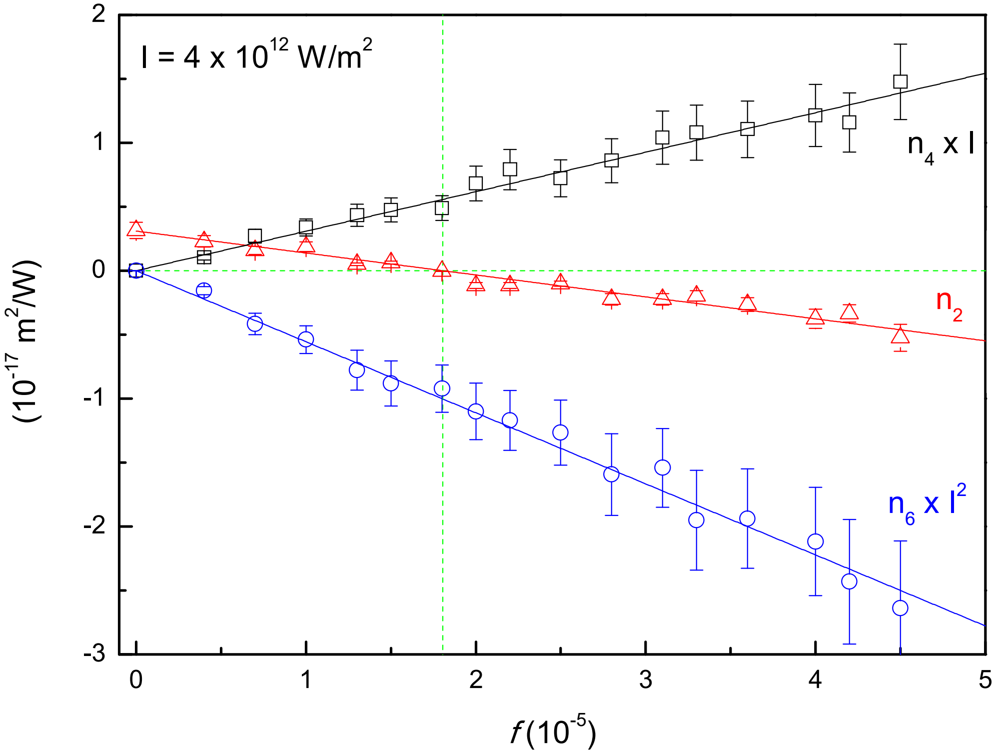
It is possible to supress one specific order of nonlinearity and enhance another one

Example: n_2=0 and n_4≠0

$n_2 \propto Re\, \chi_{eff}^{(3)}$

$n_4 \propto Re\, \chi_{eff}^{(5)}$

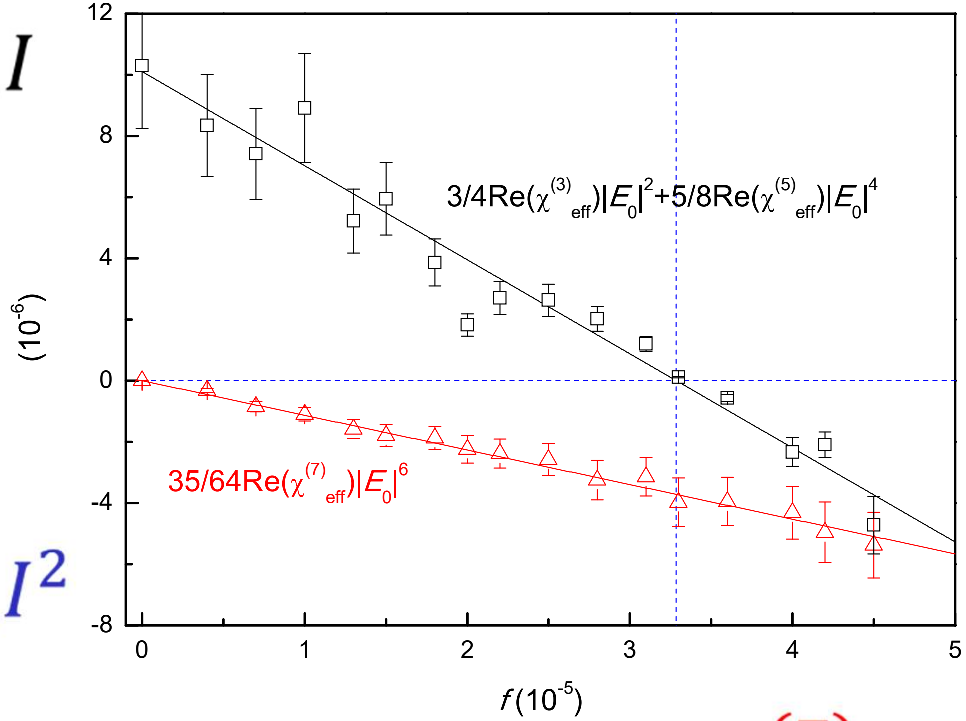
Nonlinearity management: Silver NPs + CS_2



$n_4 I$

n_2

$n_6 I^2$

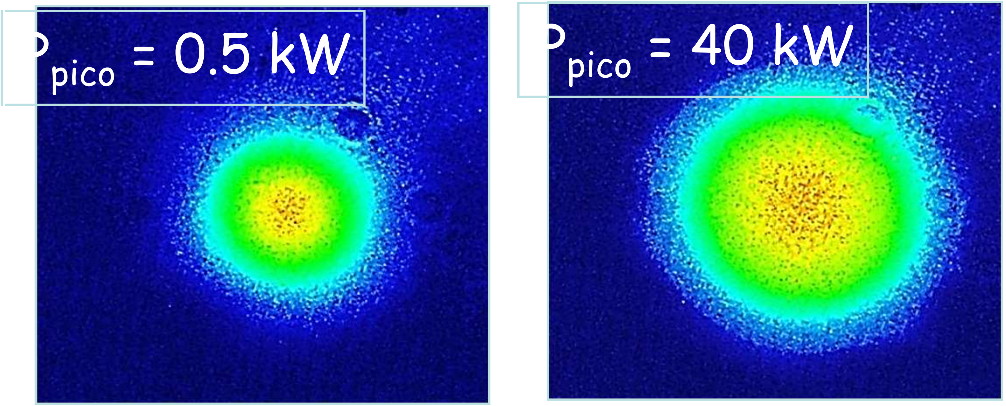


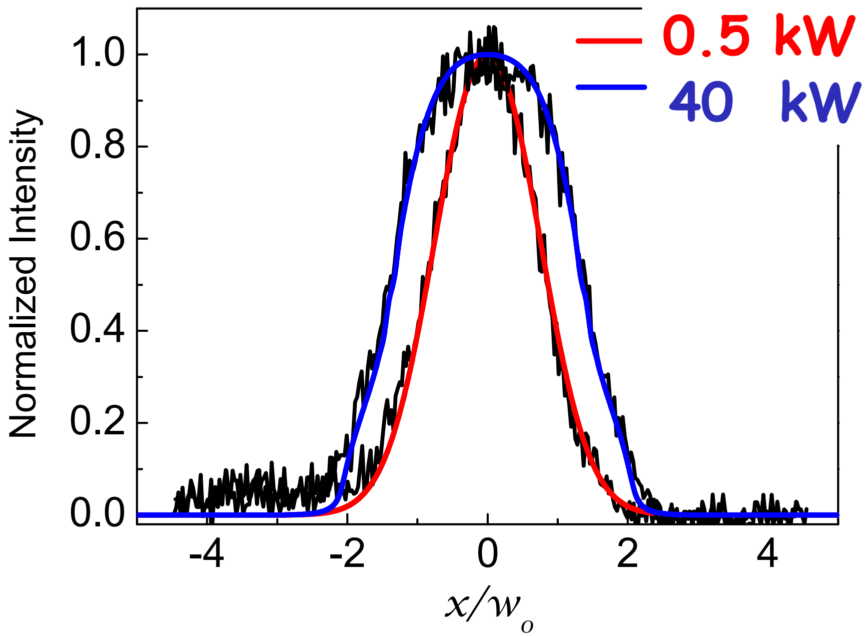
Effective $\chi_{eff}^{(7)}$

4×10^8 W/cm^2 1×10^8 W/cm^2

Self defocusing due to $\chi^{(7)}$

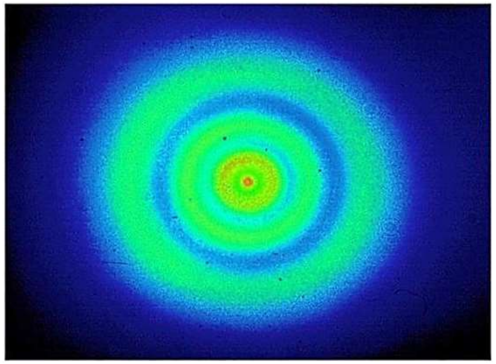
$f = 3.3 \times 10^{-5}$

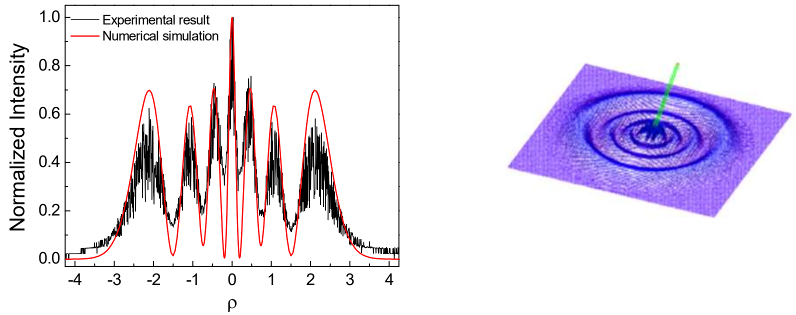




Input plane out of the lens focus

70 GW/cm²





Reyna, de Araújo, Phys. Rev. A 89 (2014) 063803

Cross-phase modulation with two counter - propagating beams

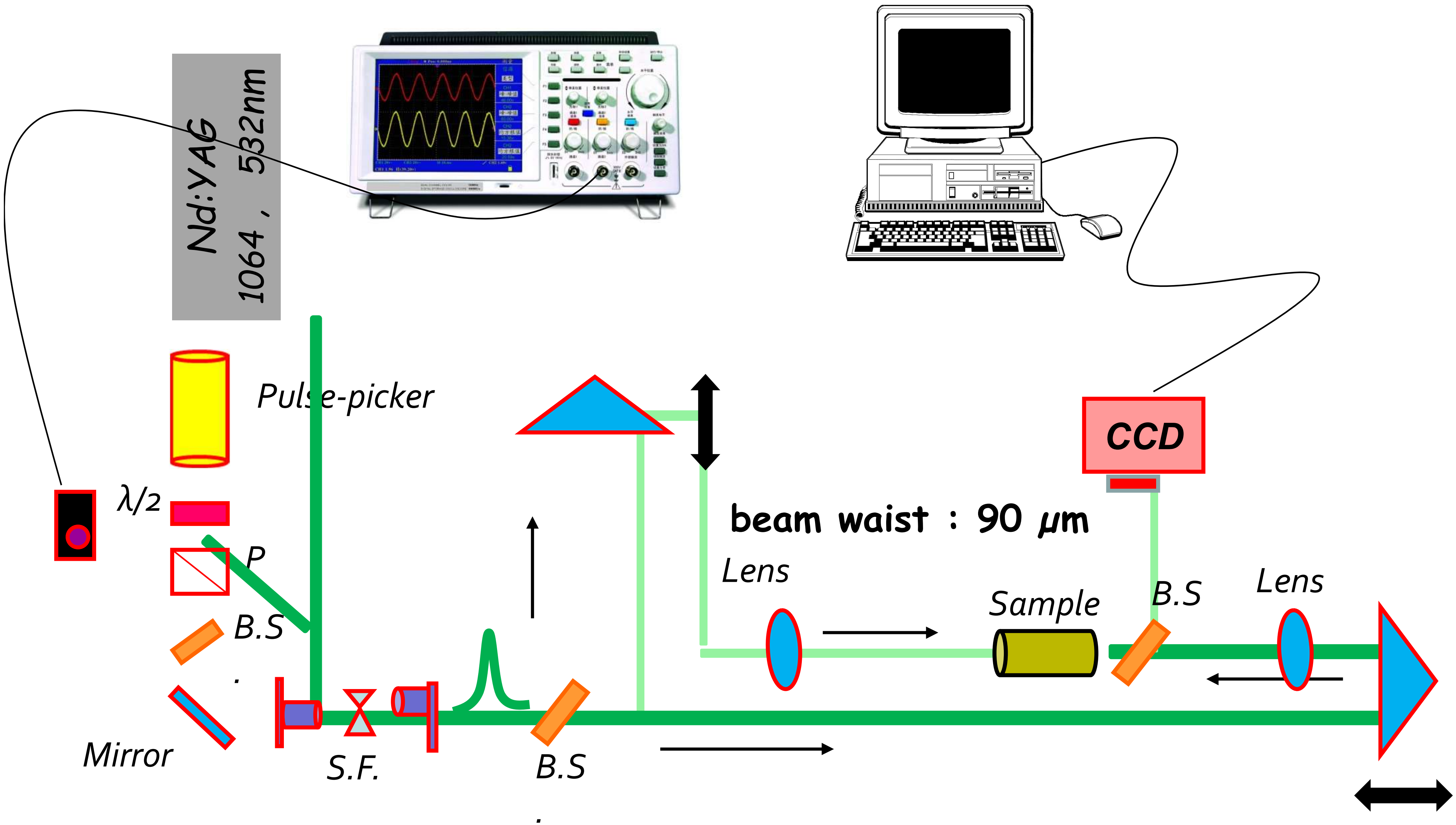
$$-\frac{\partial A_1}{\partial z}-\frac{i}{2k}\left(\frac{\partial^2 A_1}{\partial x^2}+\frac{\partial^2 A_1}{\partial y^2}\right)=\frac{ikn_2}{n_0}\left(|A_1|^2+2|A_2|^2\right)A_1+\frac{ikn_4}{n_0}\left(|A_1|^4+6|A_1|^2|A_2|^2+3|A_2|^4\right)A_1,$$

$$\frac{\partial A_2}{\partial z}-\frac{i}{2k}\left(\frac{\partial^2 A_2}{\partial x^2}+\frac{\partial^2 A_2}{\partial y^2}\right)=\frac{ikn_2}{n_0}\left(|A_2|^2+2|A_1|^2\right)A_2+\frac{ikn_4}{n_0}\left(|A_2|^4+6|A_1|^2|A_2|^2+3|A_1|^4\right)A_2,$$

Third-order

Fifth-order

Spatial Cross-Phase Modulation



NPs: 9 nm, L=5 cm, I_{pump}=2 GW/cm^2 , I_{probe} = 0.1 I_{pump}

Counter-propagating beams

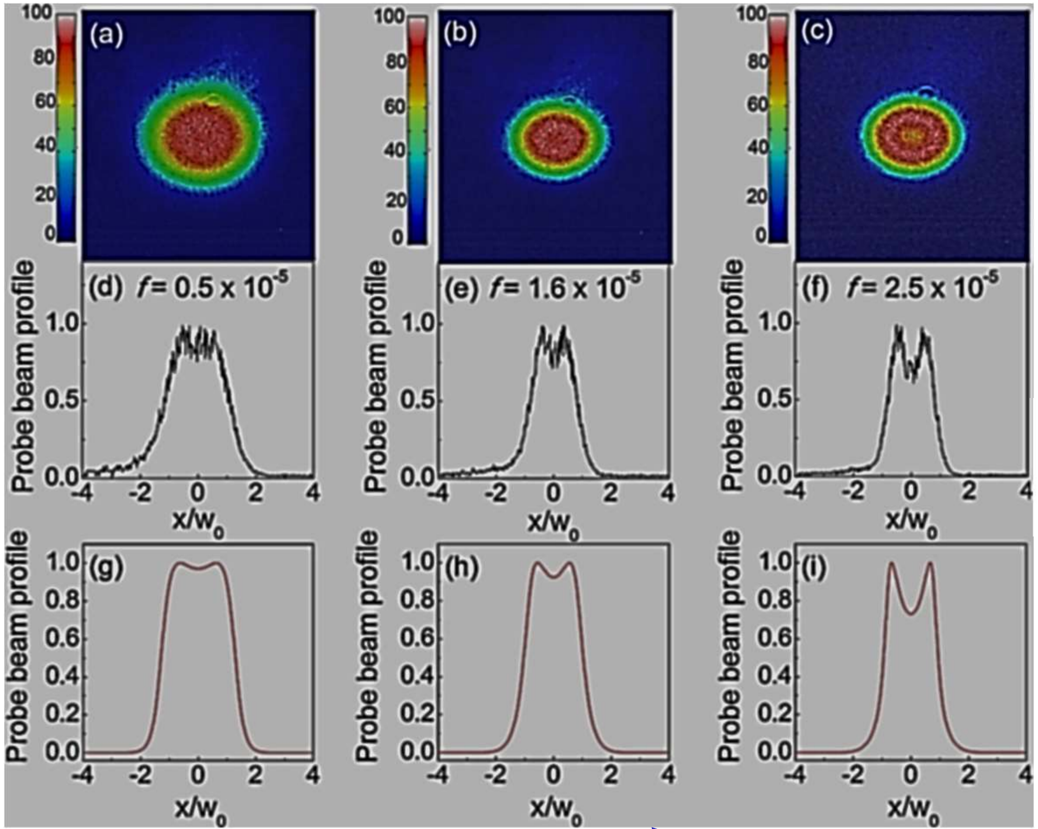
Ag NPs + acetone

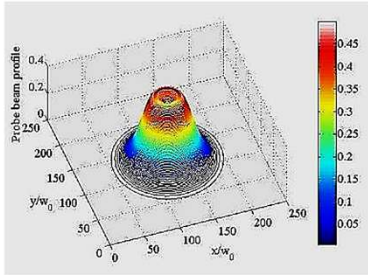
First observation of Spatial Modulational Instability due to $\chi_{eff}^{(5)}$

Probe beam profile

Profiles from the images

Theory





$Re\ \chi_{eff}^{(3)} = 0$

$Re\ \chi_{eff}^{(5)} > 0$

$n_2 = 0$ $\quad n_4 = +3.2 \times 10^{-25}\ cm^4/W^2$

Reyna, de Araújo, Phys. Rev. A 89 (2014) 063803

Cross-phase modulation

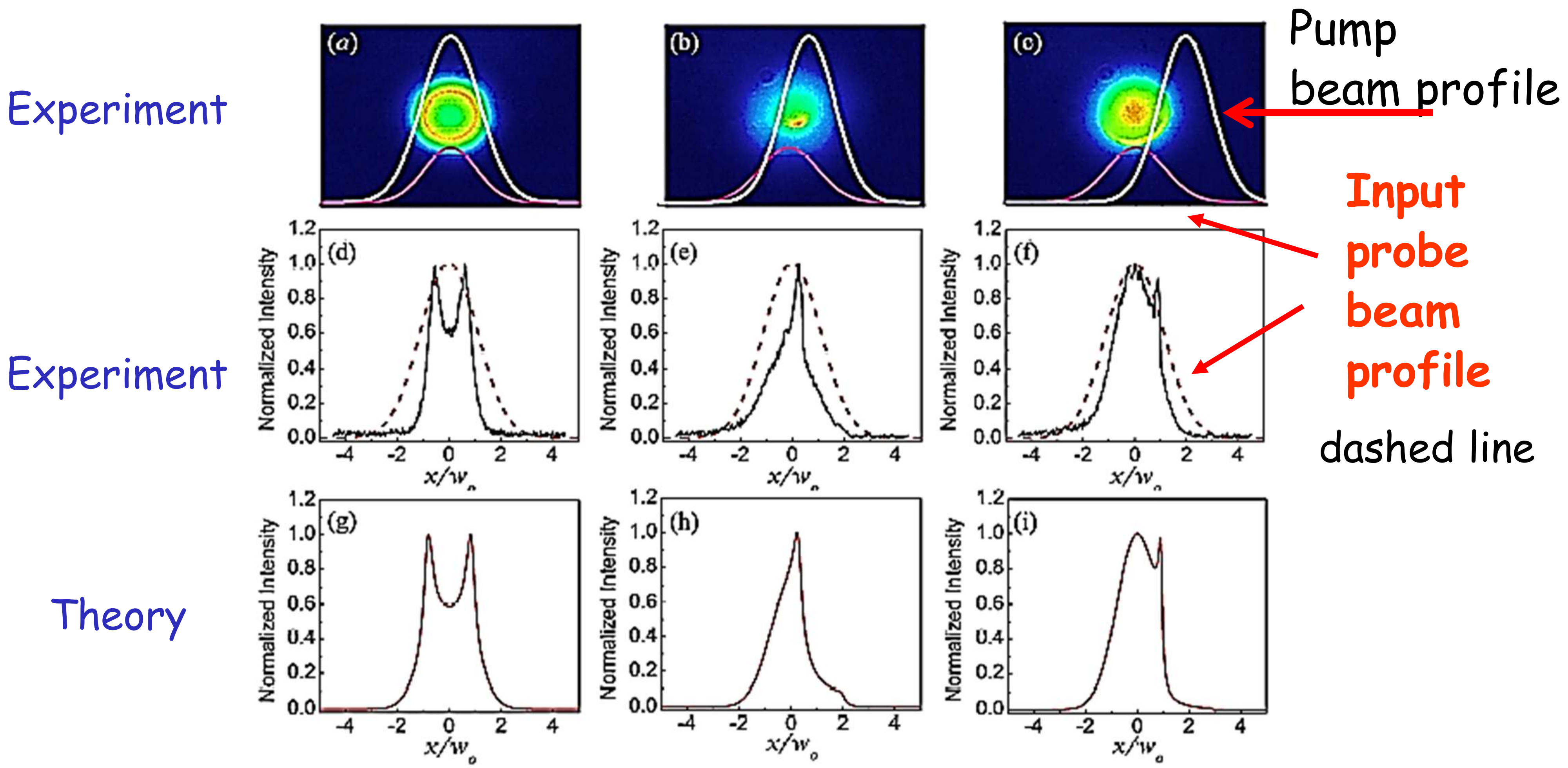
Co-propagating beams

$$-2ik\frac{\partial E_1}{\partial z}+\Delta E_1=-\frac{\omega^2}{c^2}\Big[3\chi_{eff}^{(3)}\left(|E_1|^2+2|E_2|^2\right)E_1$$
$$+10\chi_{eff}^{(5)}\left(|E_1|^4+6|E_1|^2|E_2|^2+3|E_2|^4\right)E_1$$
$$+35\chi_{eff}^{(7)}\left(|E_1|^6+18|E_1|^2|E_2|^4+12|E_1|^4|E_2|^2+4|E_2|^6\right)E_1\Big]$$

$$2ik\frac{\partial E_2}{\partial z}+\Delta E_2=-\frac{\omega^2}{c^2}\Big[3\chi_{eff}^{(3)}\left(2|E_1|^2+|E_2|^2\right)E_2$$
$$+10\chi_{eff}^{(5)}\left(3|E_1|^4+6|E_1|^2|E_2|^2+|E_2|^4\right)E_2$$
$$+35\chi_{eff}^{(7)}\left(4|E_1|^6+12|E_1|^2|E_2|^4+18|E_1|^4|E_2|^2+|E_2|^6\right)E_2\Big].$$

Reyna, de Araújo - Optics Express 22 (2014) 22456-22469

Induced focusing due to the seventh-order susceptibility



$$n_2 = 0;\ n_4 = 0;\ n_6 < 0$$

Analagous to $\chi^{(3)}$: Hickmann, Gomes, de Araújo, PRL 68 (1992) 3547

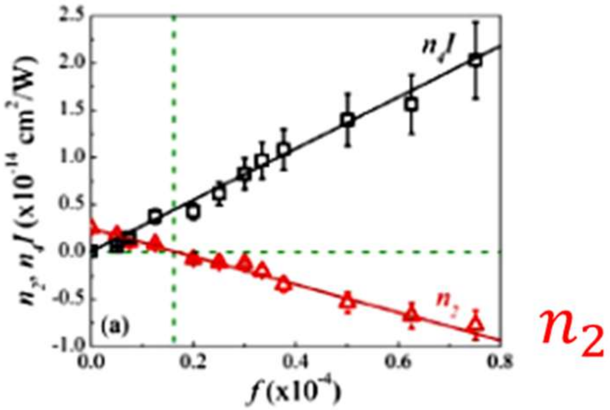
“Nonlinearity management”

Z-scan

Ag-NPs (<10 nm) + CS_2

$\chi_{eff}^{(3)}$, $\chi_{eff}^{(5)}$, $\chi_{eff}^{(7)}$

Reyna, de Araújo. Opt. Express 22 (2014) 22456-22469

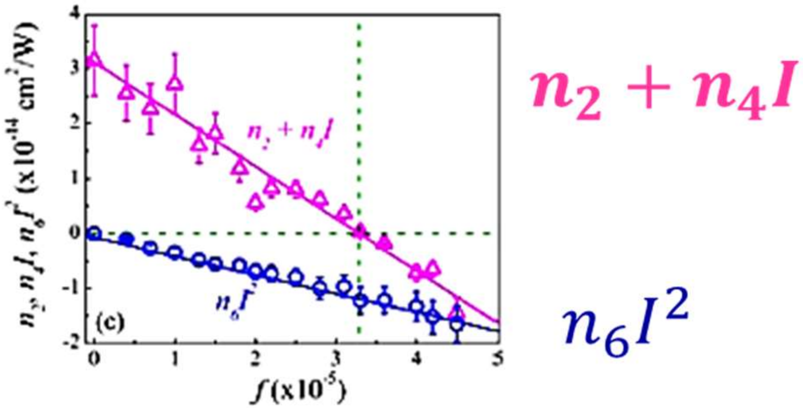


f = 1.6 x 10^{-5}

4 x 10^8 W/cm^2

$$Re\chi^{(3)} = 0$$

$$Re\chi^{(5)} \neq 0$$



f = 3.3 x 10^{-5}

1 x 10^8 W/cm^2

$$Re\left(\chi^{(3)} + I\,x\,\chi^{(5)}\right) = 0$$

$$Re\chi^{(7)} \neq 0$$

Non linear propagation of light

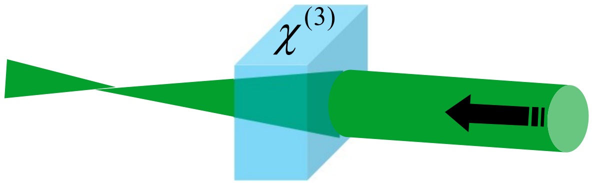
$$\nabla^2 \vec{E} - \frac{1}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}_L}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2}$$

$$\vec{E}(\vec{r},t) = \vec{A}(\vec{r}) \exp[i(kz - \omega t)]$$

$$i\frac{\partial \vec{A}}{\partial z} = -\frac{1}{2k}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\vec{A} - \frac{n_2}{n_0} k |\vec{A}|^2 \vec{A}$$

difração

auto-focalização

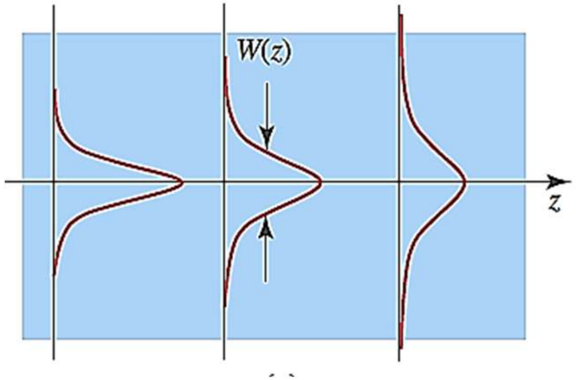


Optical Spatial Solitons

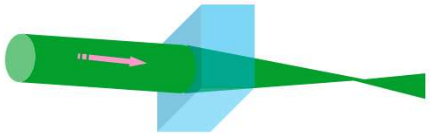
Solutions of the Maxwells equation including nonlinear terms

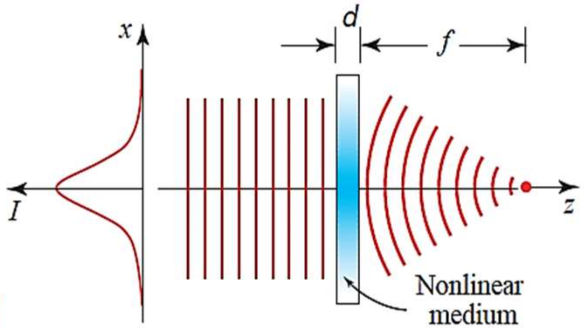
NL Schrödinger Equation

Difraction



Self-focusing material

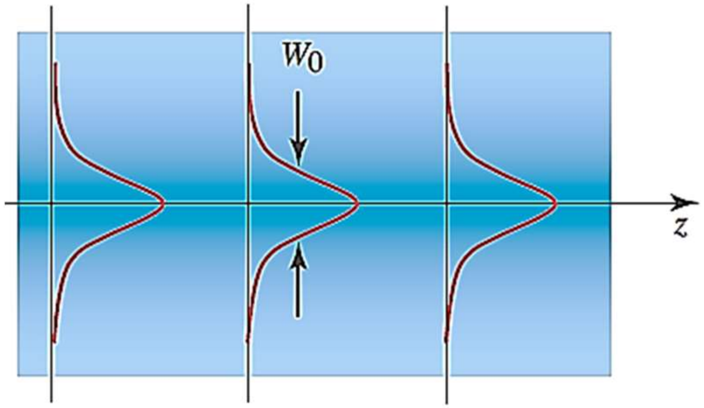


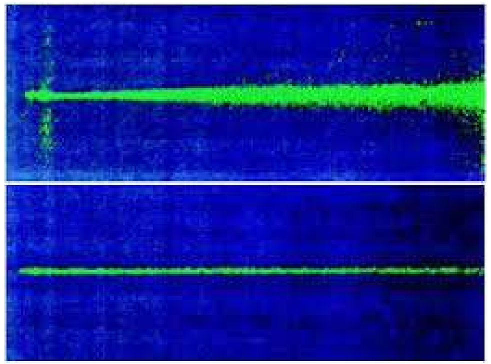


$n_2 > 0$

$$n = n_0 + n_2 I + \ \ldots$$

Bright Spatial soliton





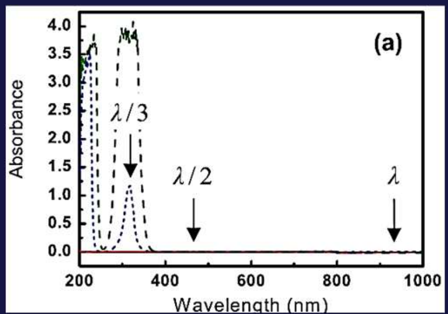
Low intensity

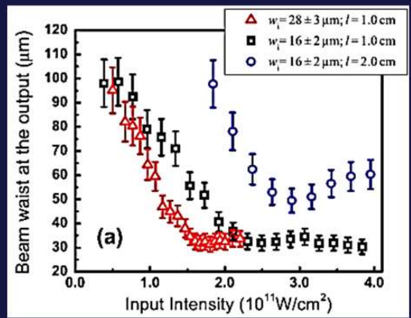
High intensity Soliton

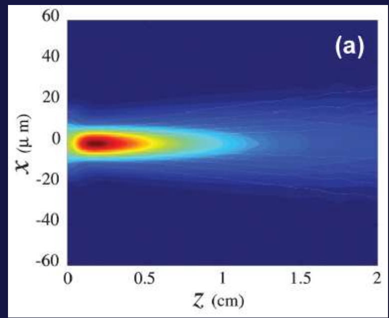
1st. lecture

First demonstration of (2+1)D soliton propagating in a homogeneous medium with local nonlinearity

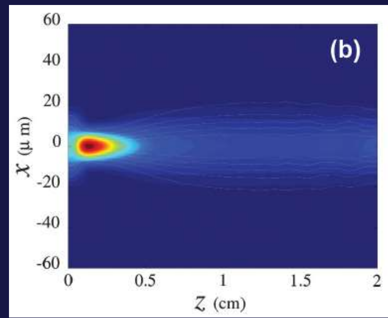
Falcão-Filho, de Araújo, Boudebs, Leblond, Skarka
Robust two-dimensional spatial solitons in liquid carbon disulfide
Phys. Rev. Lett. 110 (2013) 013901.







Low intensity



soliton

Very important: contributions of third and fifth order of opposite signs

CS_2: stable (2+1)D soliton

$$Re\,\chi^{(3)} > 0 \qquad\qquad Re\,\chi^{(5)} < 0$$

Is it possible to observe a stable (2+1)D soliton in a system with:

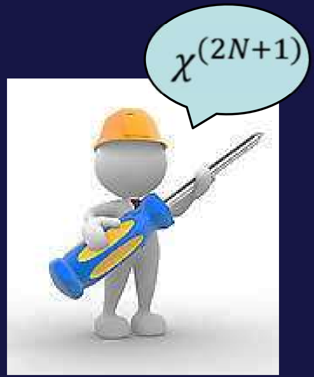
$$Re\,\chi^{(3)} = 0 \;, \qquad Re\,\chi^{(5)} > 0 \;, \qquad Re\,\chi^{(7)} < 0 \qquad ?$$

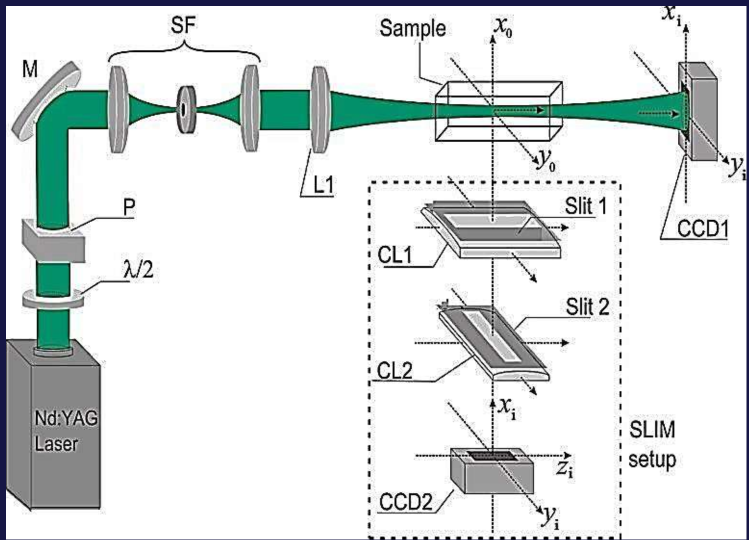
$$2ik\frac{\partial E}{\partial z} + \Delta E = -\frac{\omega^2}{c^2}\left[3\chi_{eff}^{(3)}|E|^2 E + 10\chi_{eff}^{(5)}|E|^4 E + 35\chi_{eff}^{(7)}|E|^6 E\right]$$

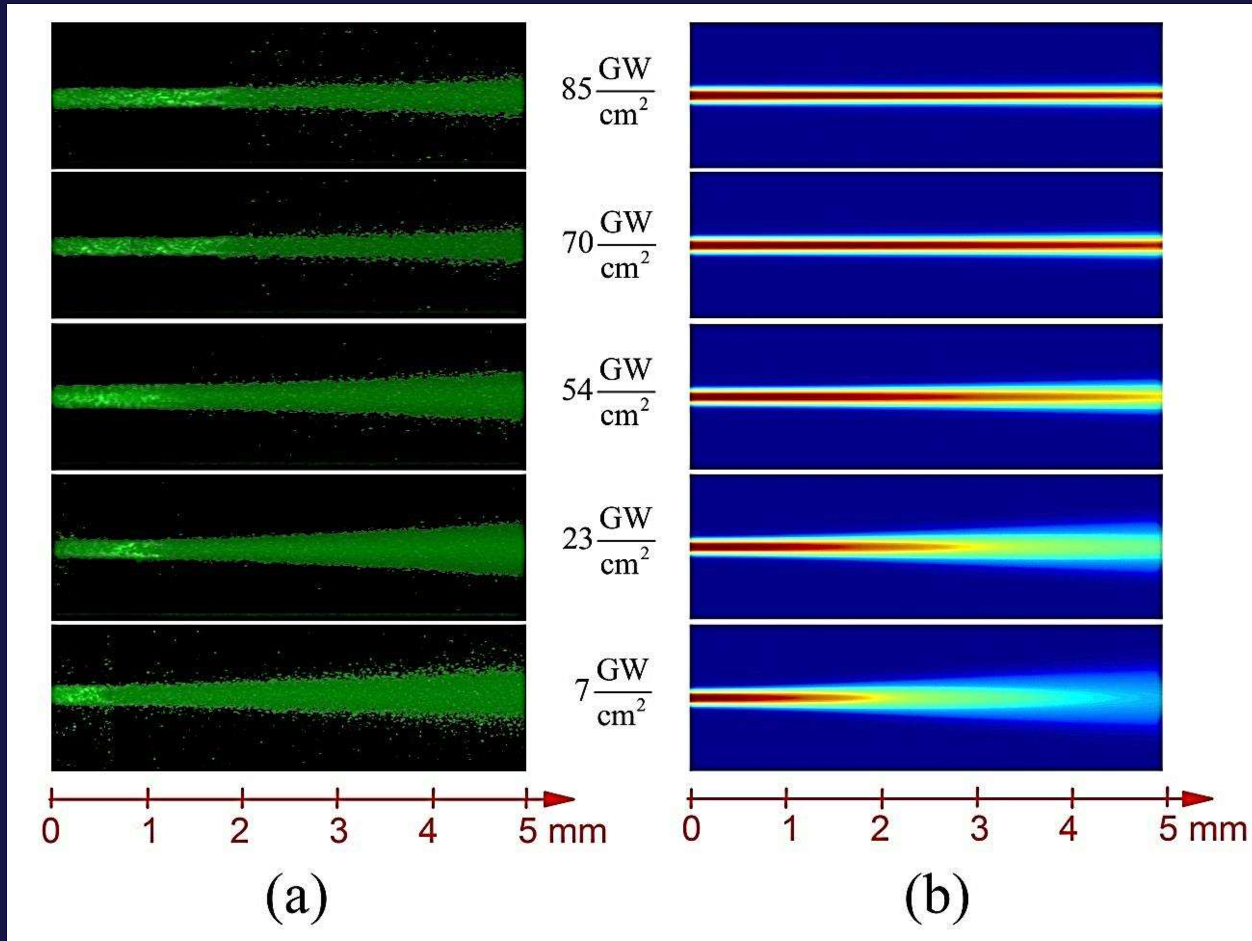
First observation of 2D Spatial-Solitons in a quintic-septimal medium

Silver NPs in acetone

$$n_2 = 0; \quad n_4 > 0; \quad n_6 < 0$$



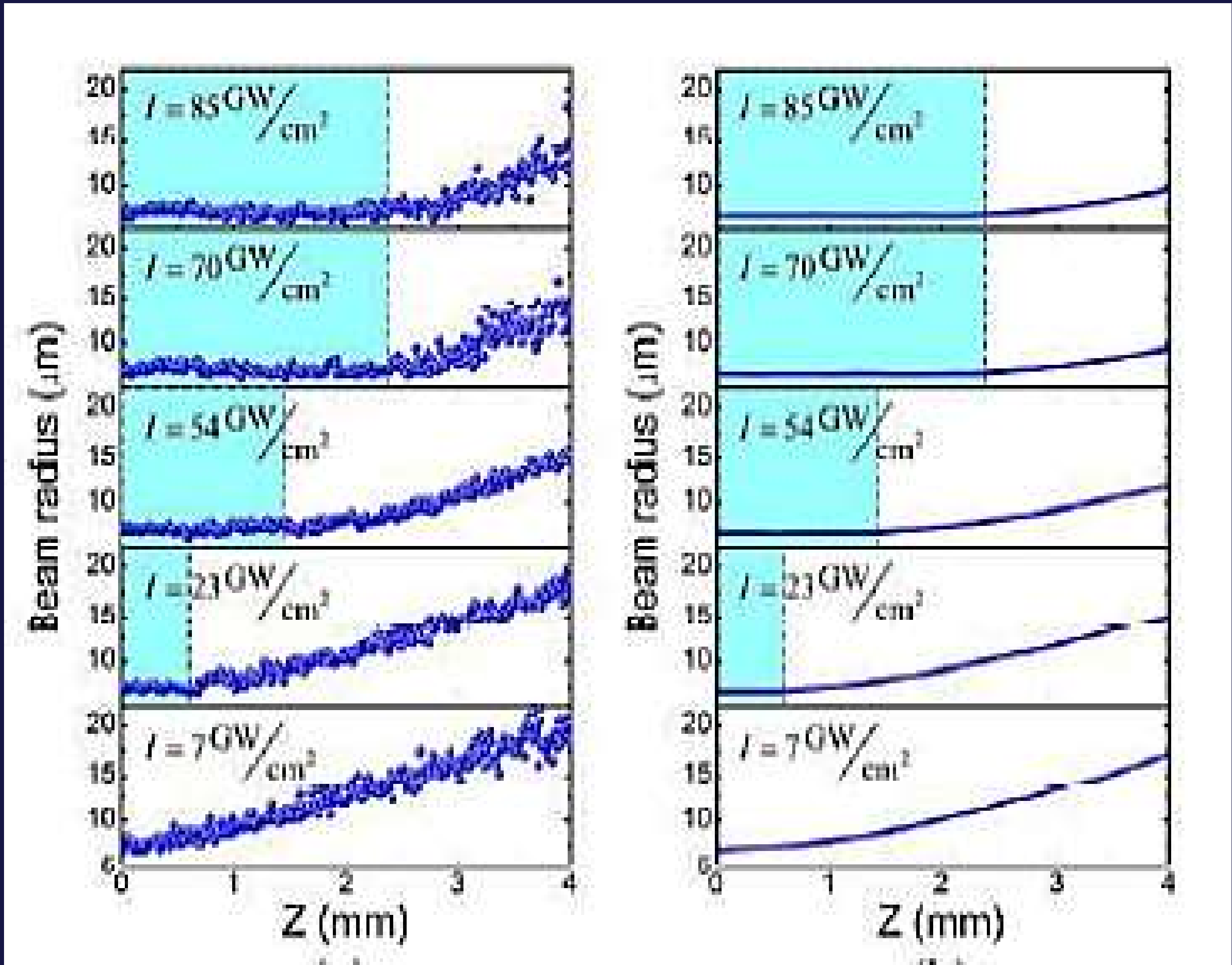




experiment theory

Reyna, Jorge, de Araújo Phys. Rev. A 90 (2014) 063835
de Araújo et al. J. Lumin. 169 (2016) 492-496

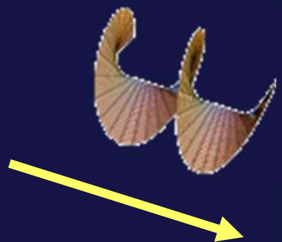
$$2ik\frac{\partial E}{\partial z}+\Delta E=-\frac{\omega^2}{c^2}\left[3\chi_{eff}^{(3)}|E|^2E+10\chi_{eff}^{(5)}|E|^4E+35\chi_{eff}^{(7)}|E|^6E\right]$$



experiment theory

Optical vortices

- Beams with phase singularity
- Zero field in the center of the vortex
- Helical wavefront
- Phase



$$\phi(t, z, \theta) = kz + \omega t + m\theta$$

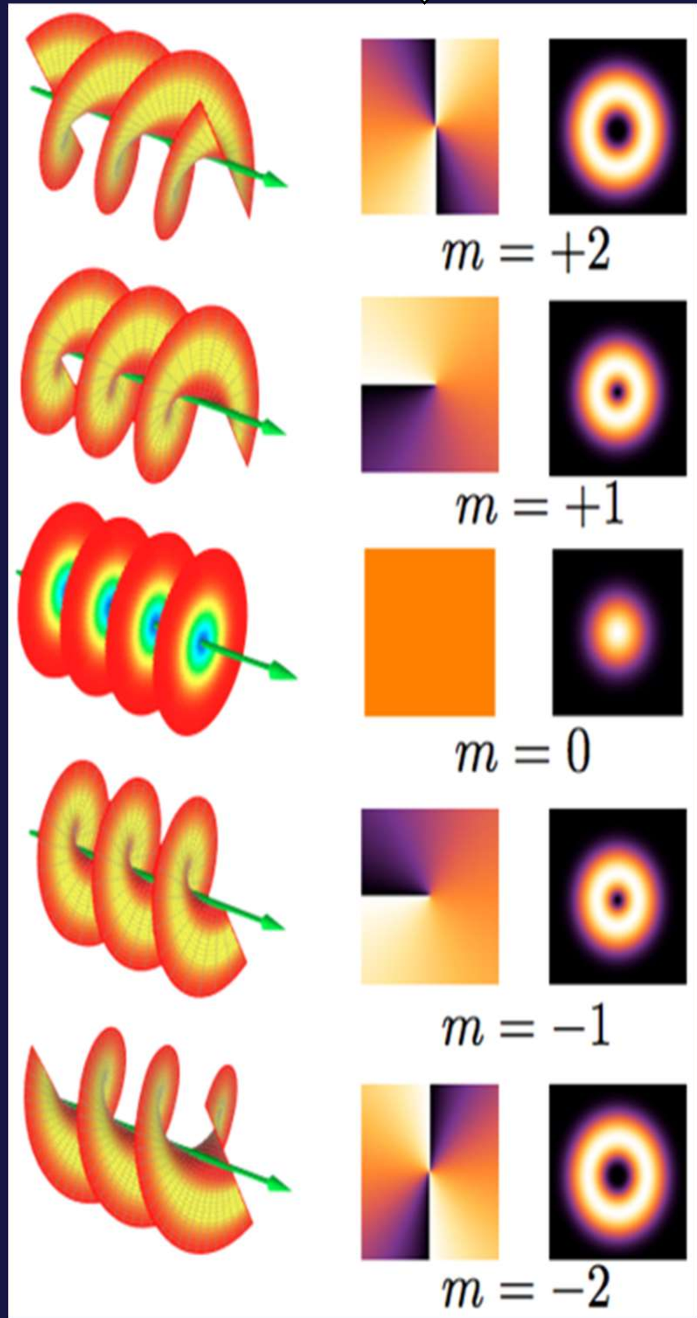
$m = 0$ Plane wave

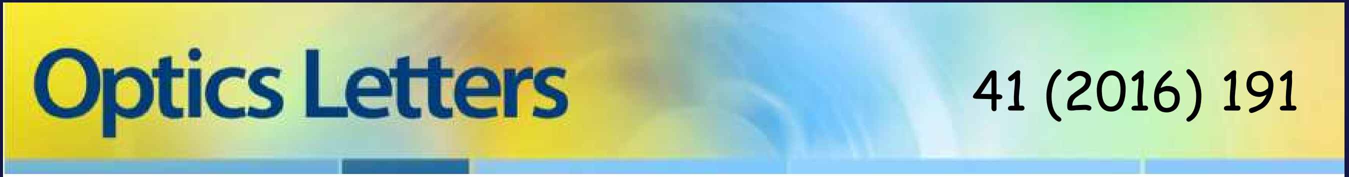
$m \neq 0$ Wave with topological phase

m is the “topological charge”

phas
e

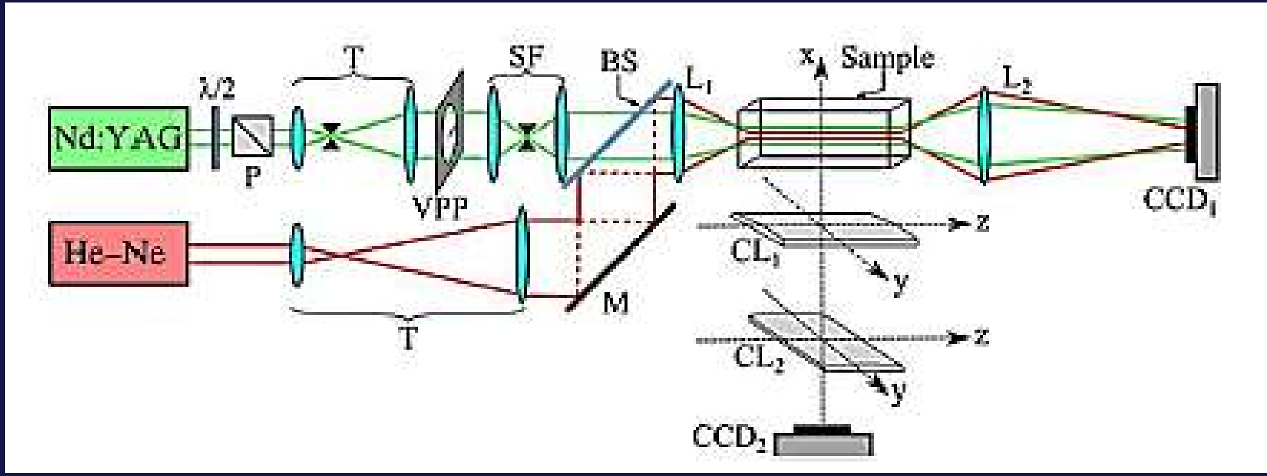
intensity

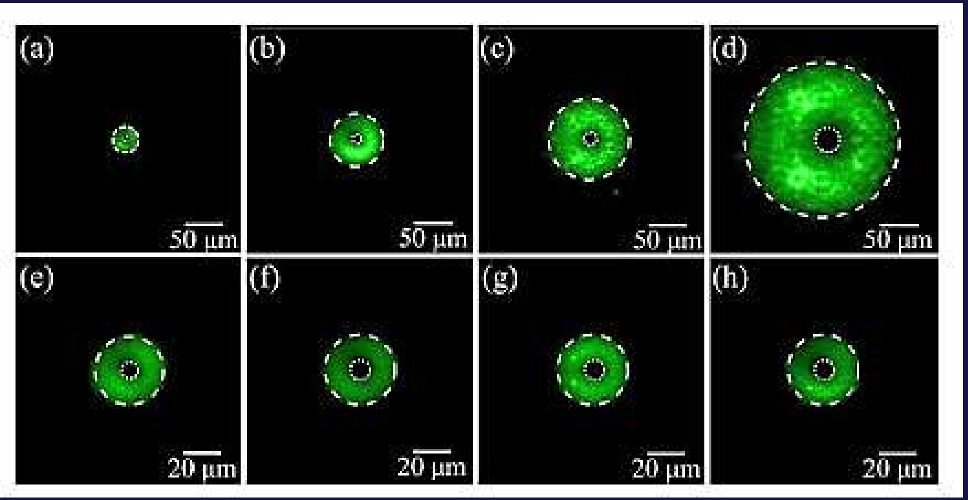



Optics Letters 41 (2016) 191

Guiding and confinement of light induced by optical vortex solitons in a cubic–quintic medium

Albert S. Reyna* and Cid B. de Araújo



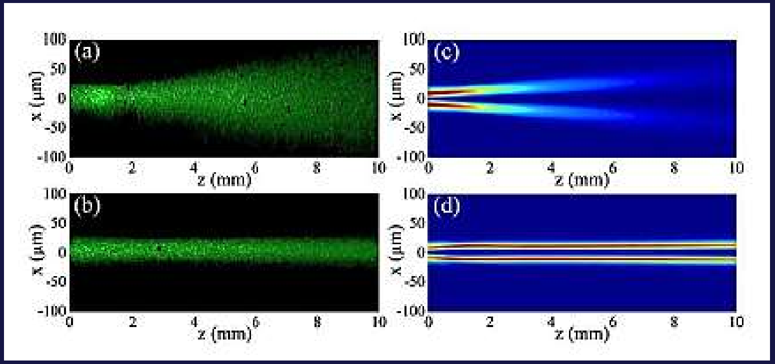


Z= 0 3 5 10 mm

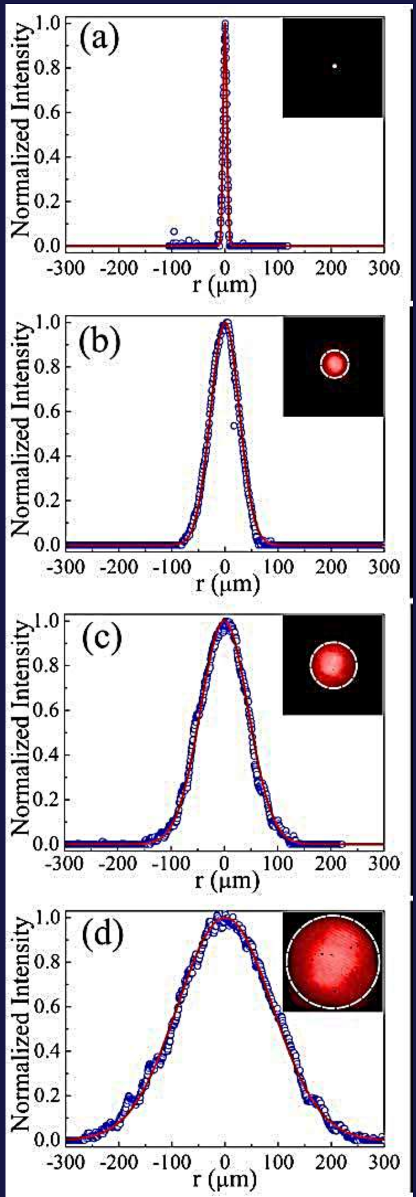
0.1

3.0

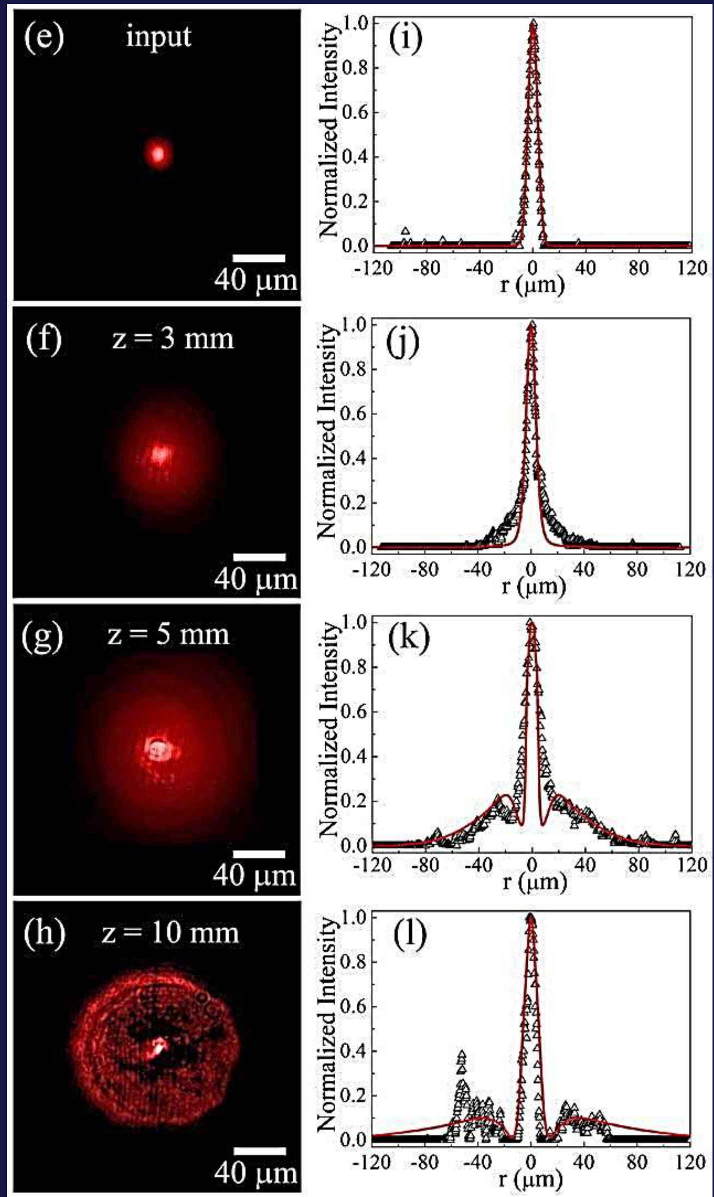
GW/cm^2



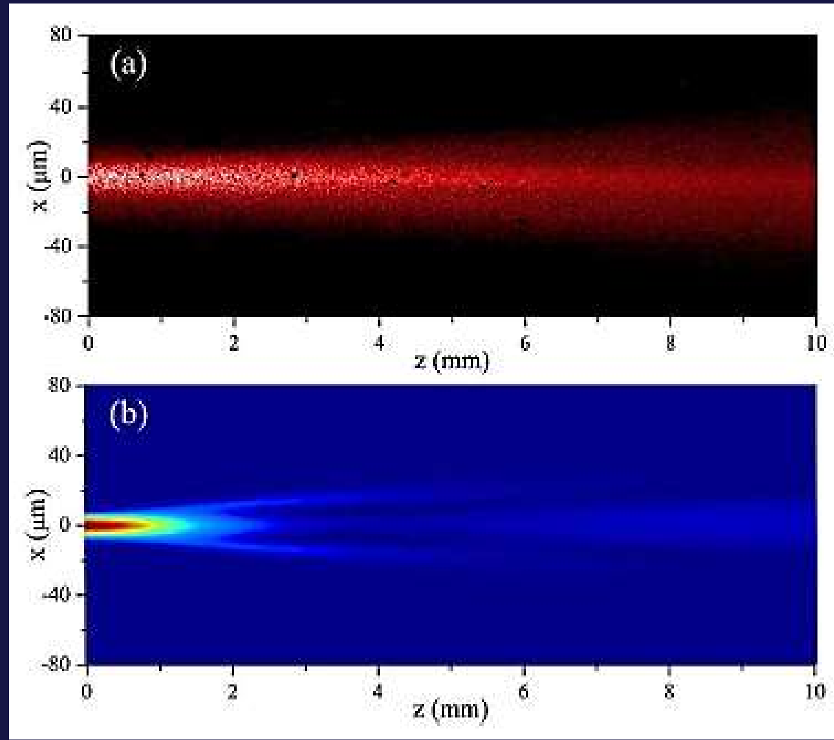
10 mm - 25 Z_R



HeNe



Guided HeNe



I_{OVS} = 3.0 GW/cm^2

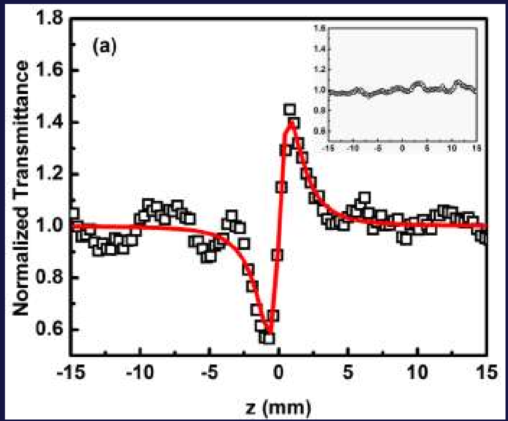
I_{HeNe} = 0.1 GW/cm^2

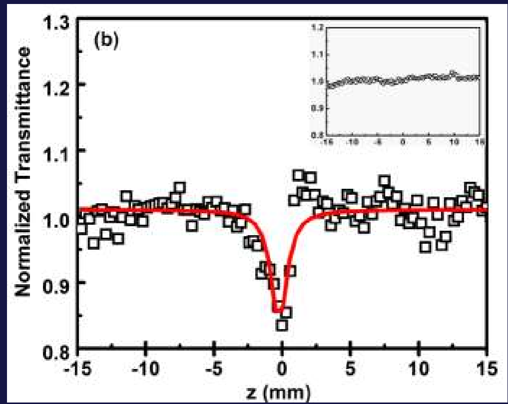
How to address the long standing problem of discovering a very good material for all-optical switching?

We need a material with large NL refraction and low NL absorption

In general large NL refraction presents large NL absorption

PbO-GeO$_2$ films with gold NPs for all-optical switching





RF sputtering

800 nm 150 fs

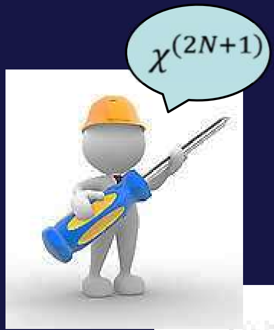
Figure-of-merit enhanced by two orders of magnitude

Germanate film	$n_2/\lambda\alpha_2$
As grown	8.3×10^{-4}
With Au NPs	$>2.1 \times 10^{-1}$

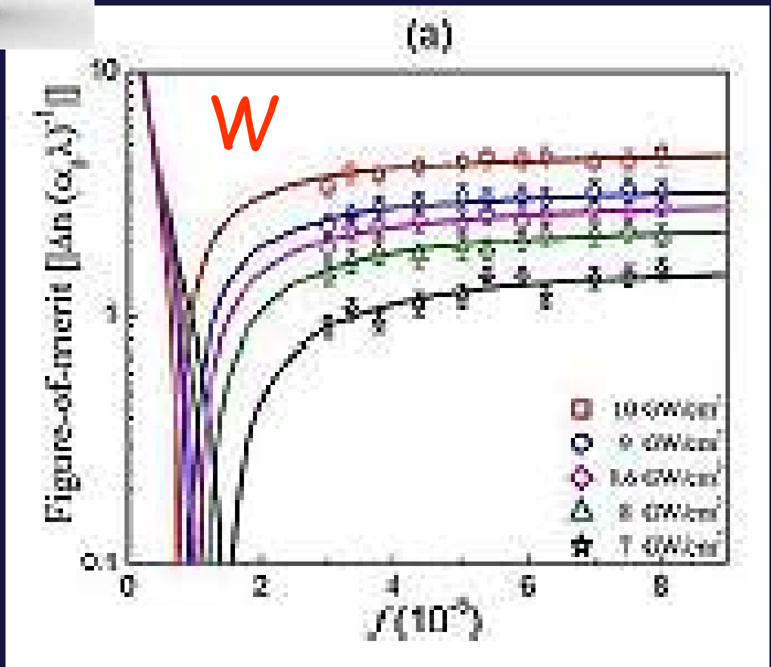
de Araújo et al. J. Luminescence 133 (2013) 180

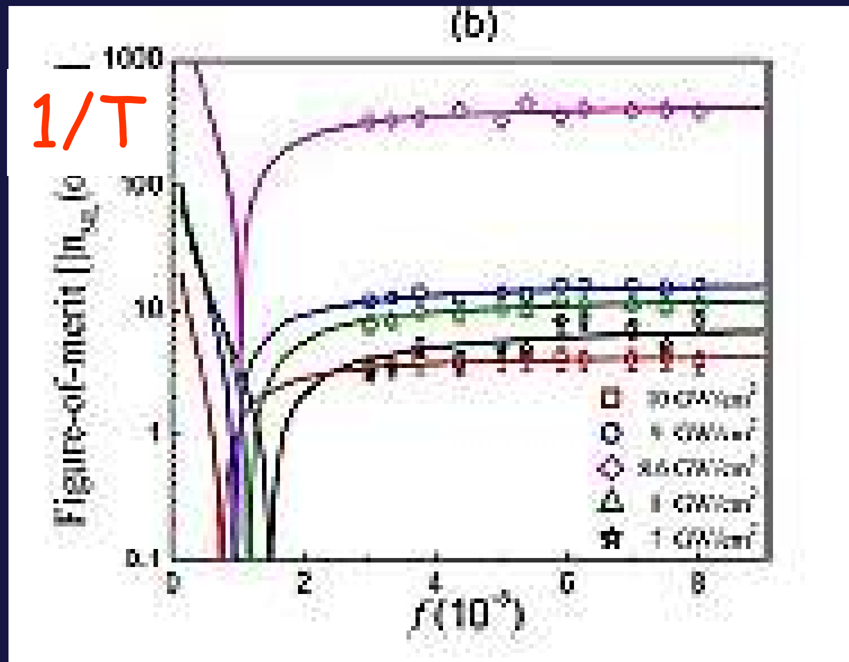
Optimization procedure for the design of all-optical switches based on metal-dielectric nanocomposites

$$\Delta n = n_2 I + n_4 I^2 + n_6 I^3 + \cdots = n_{NL} I \qquad \alpha_{NL} = \alpha_2 + \alpha_4 I + \alpha_6 I^2 + \cdots$$



$$\boxed{W = \frac{\Delta n}{\lambda \alpha_0} > 1} \qquad \boxed{T = \frac{\lambda\, \alpha_{NL}}{n_{NL}} < 1}$$



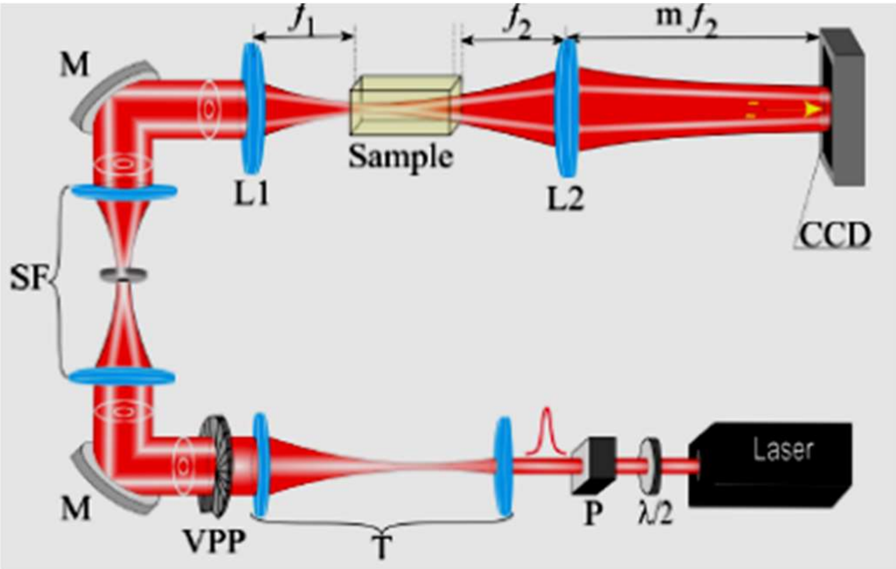


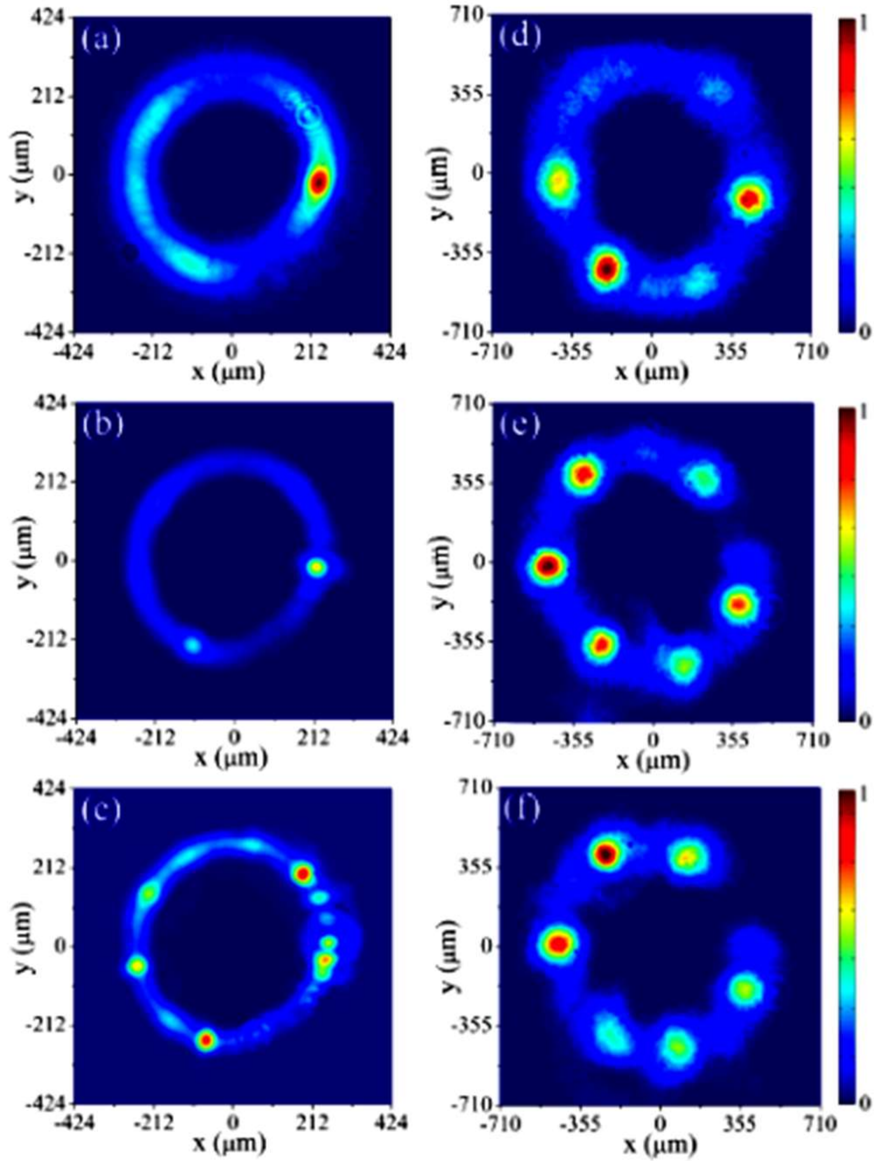
Reyna, de Araújo, Opt. Express 23 (2015) 7659-7666

These results show that it is possible to have an efficient all-optical switch if a nanocomposite is made
according to the nonlinearity management procedure presented

Challange for
materials scientists

Soliton clusters





Reyna et al.

Observation and analysis of creation, decay, and regeneration of annular soliton clusters in a lossy cubic-quintic optical medium,

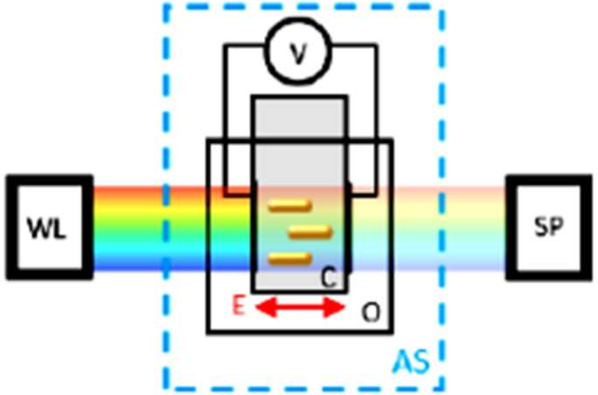
Phys. Rev. A 102 (2020) 033523

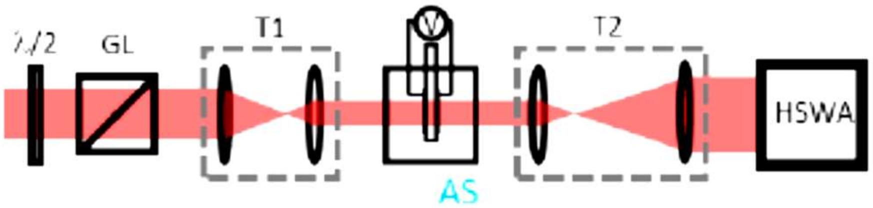
Research Article Vol. 26, No. 16 | 6 Aug 2018 | OPTICS EXPRESS 20298

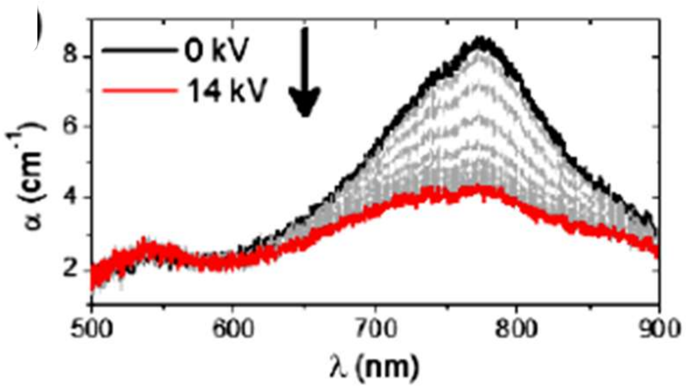
Optics EXPRESS

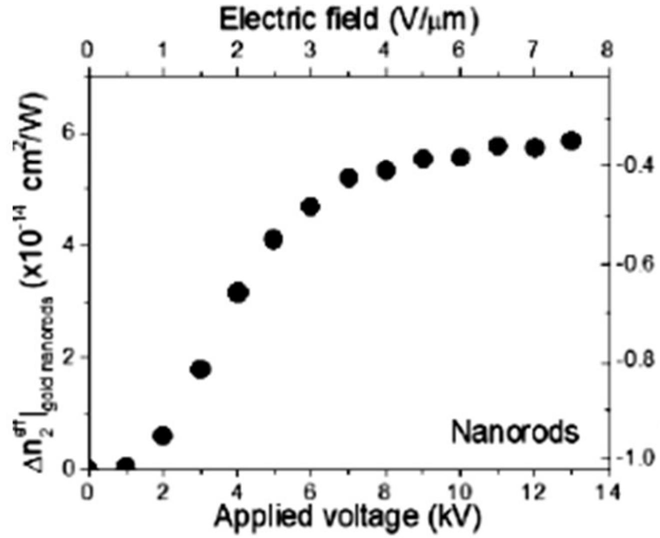
Nonlinear refractive index of electric field aligned gold nanorods suspended in index matching oil measured with a Hartmann-Shack wavefront aberrometer

M. Maldonado et al.





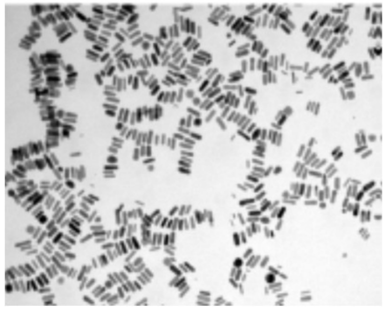


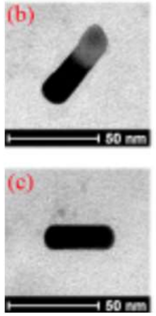


THE JOURNAL OF PHYSICAL CHEMISTRY C

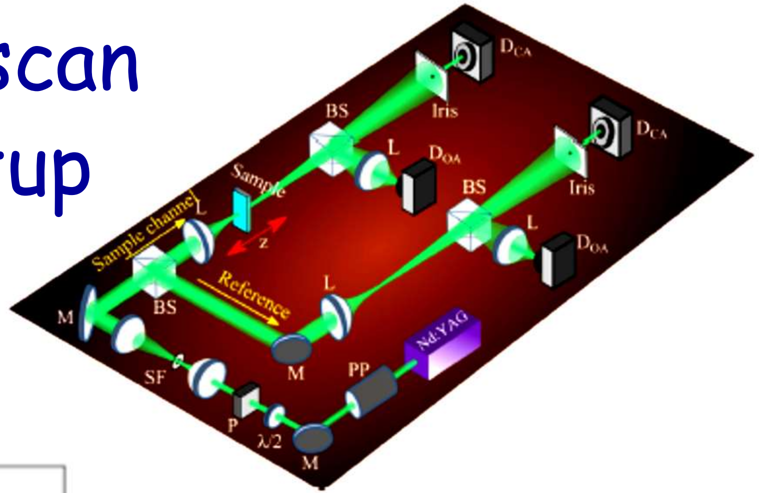
Light Scattering, Absorption, and Refraction due to High-Order Optical Nonlinearities in Colloidal Gold Nanorods

Oliveira et al. J. Phys. Chem. C 2019, 123, 12997–13008



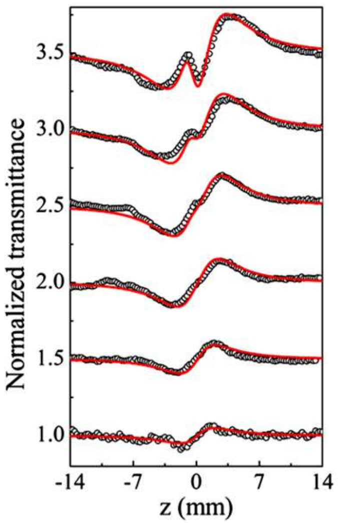


Z-scan setup

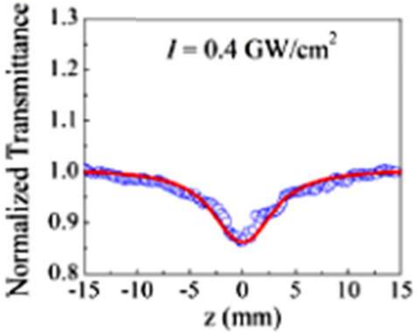


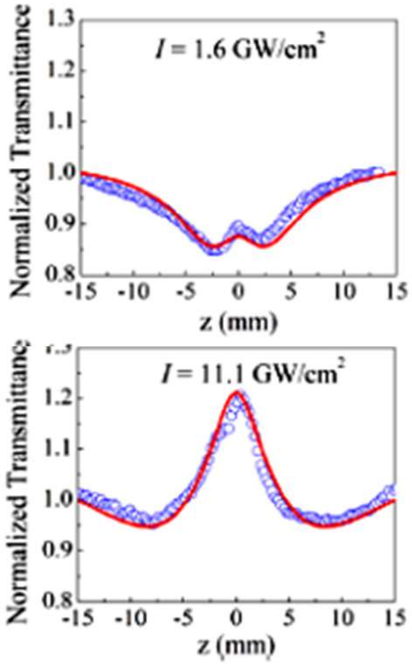
$\chi^{(3)}$ $\chi^{(5)}$

GWcm^{-2}



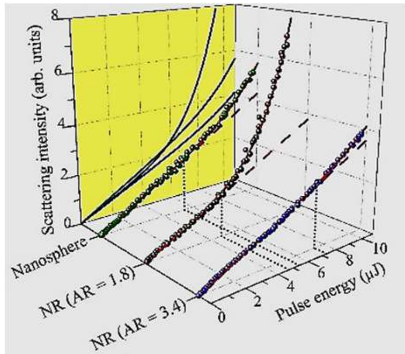
4.0
3.2
2.4
1.6
0.8
0.4





$f = 1.0x10^{-4}$

RA: 3.4

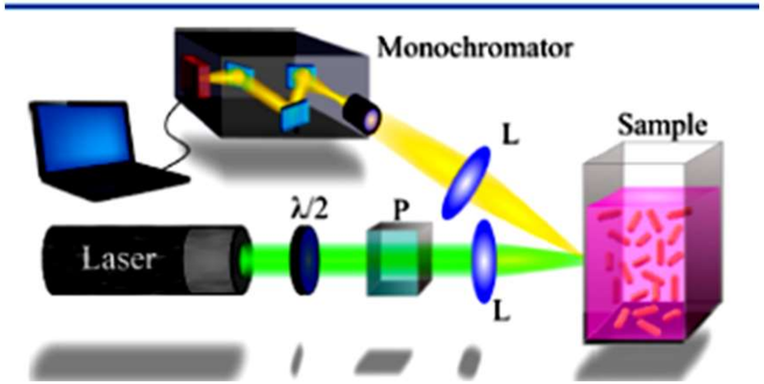


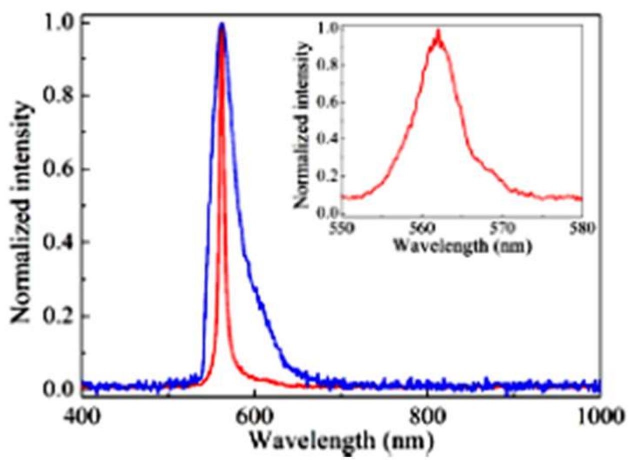
NL scattering

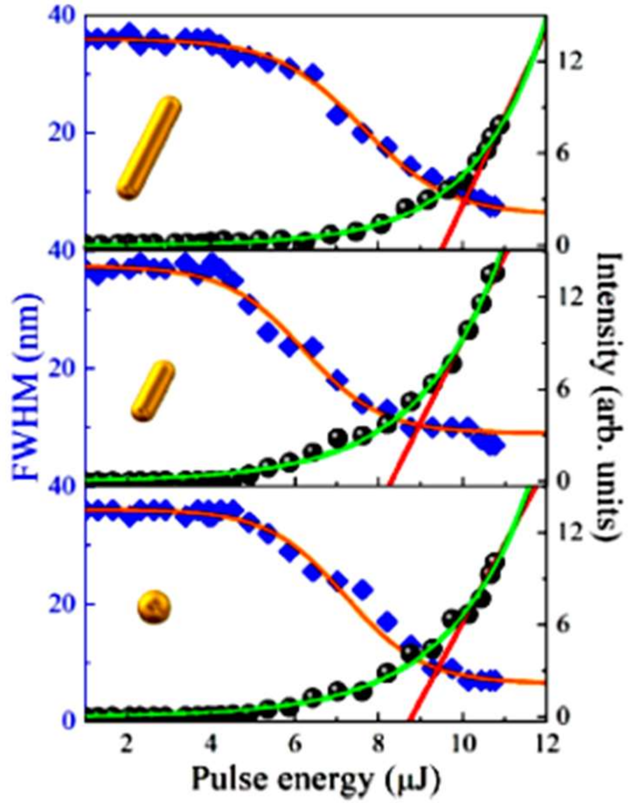
THE JOURNAL OF PHYSICAL CHEMISTRY C

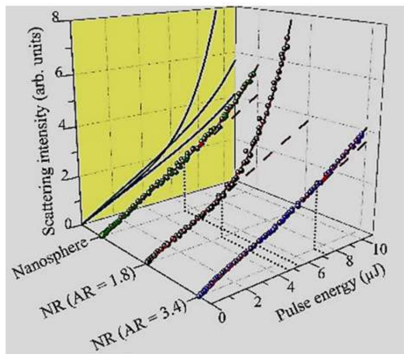
Influence of the Fifth-Order Nonlinearity of Gold Nanorods on the Performance of Random Lasers

Oliveira et al. *J. Phys. Chem. C* 2020, 124, 10705−10709





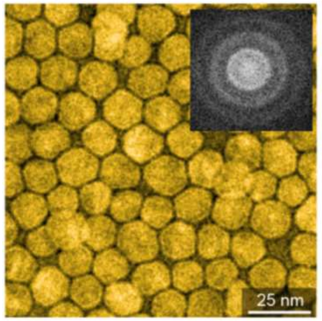


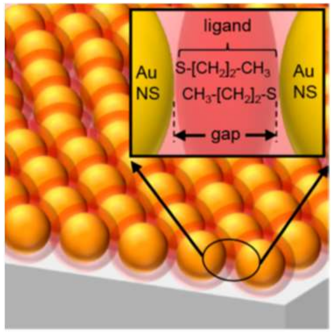


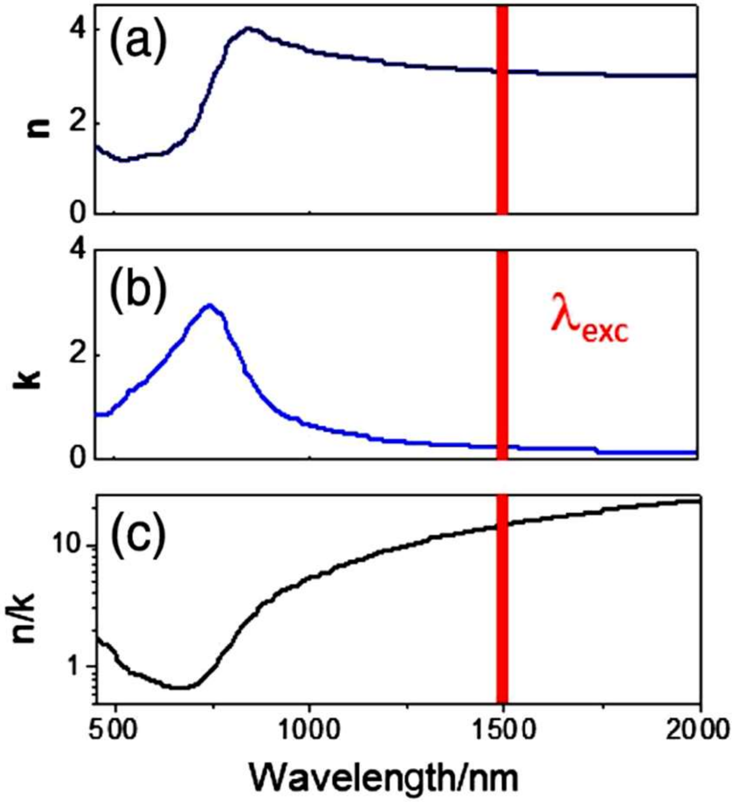
Nonlinear scattering: 5th. order

Gold metasurface

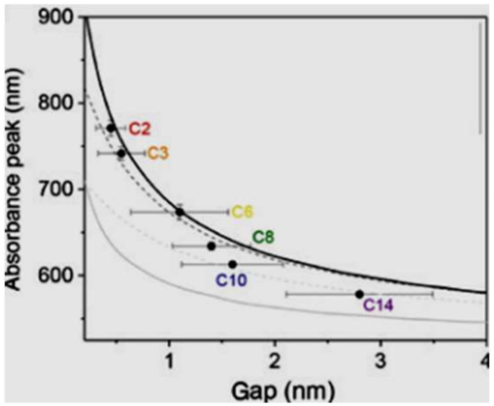
samples: Jake Fontana
NRL USA







spheres : 15 nm diam.
gap: **0.6 nm**



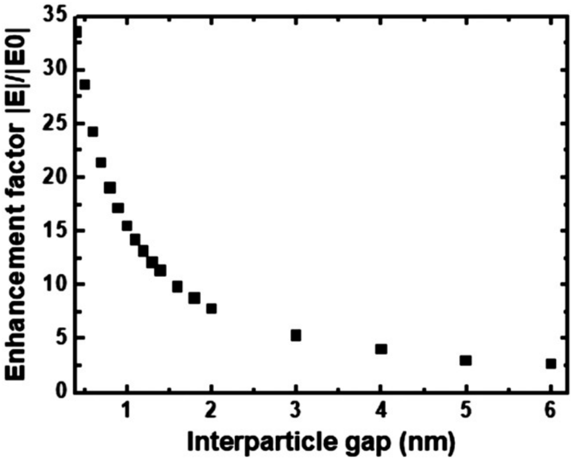
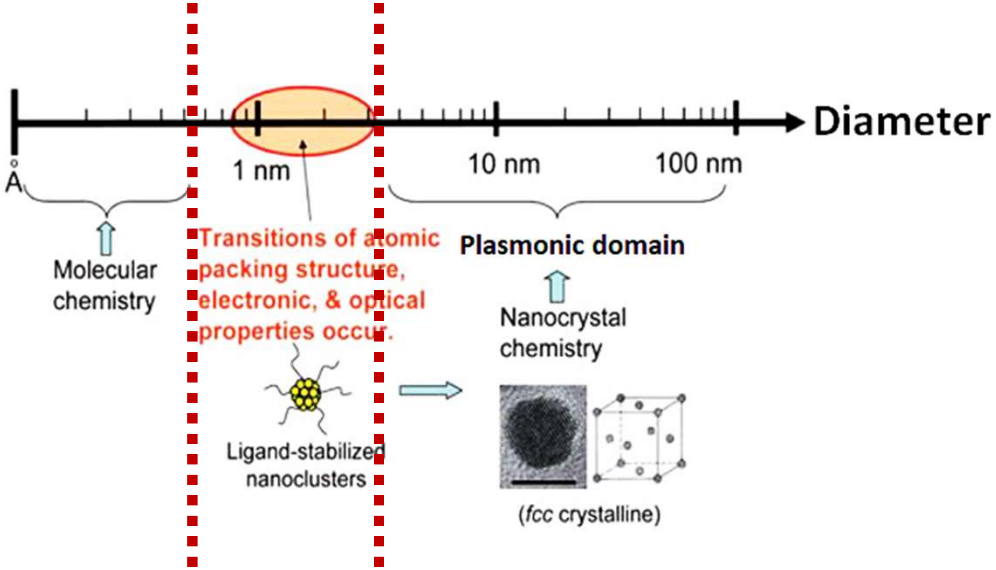


Table 2. Comparison between the NL Optical Coefficients of the Gold MS at 800 nm and 1500 nm, Corrected for Linear Absorption[a]

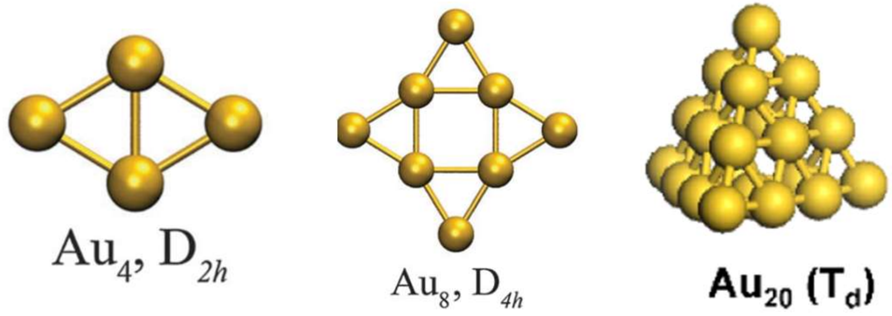
λ (nm)	n_2 ($cm^2 W^{-1}$)	α_2 (cmW^{-1})	$Re\chi^{(3)}$ ($cm^2 V^{-2}$)	$Im\chi^{(3)}$ ($cm^2 V^{-2}$)
800	-7.90×10^{-9}	-0.90×10^{-4}	-3.37×10^{-10}	-1.90×10^{-10}
1500	-1.05×10^{-10}	3.00×10^{-6}	-2.73×10^{-12}	8.08×10^{-13}

Menezes et al. JOSA B 36 (2019) 1485-1491

Gomes et al. Nanophotonics 9 (2020) 725-740

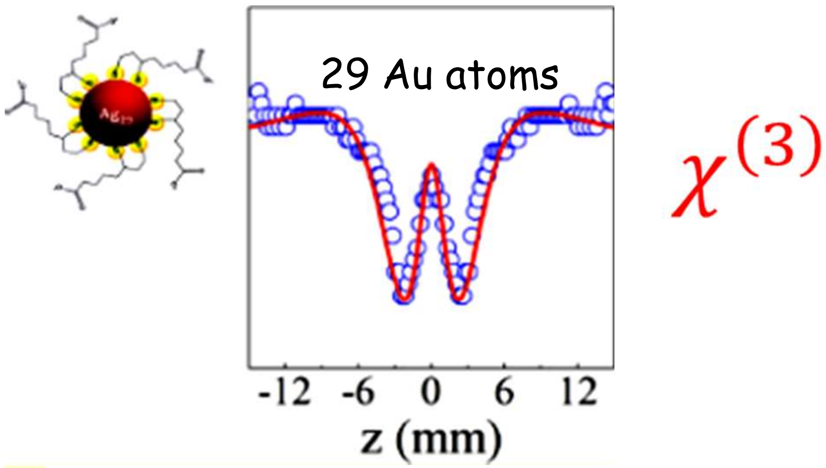


Gold nanoclusters
- Vacuum -



Geometry changes when adding ligands

Au_{29} $(DHLA)_{12}$

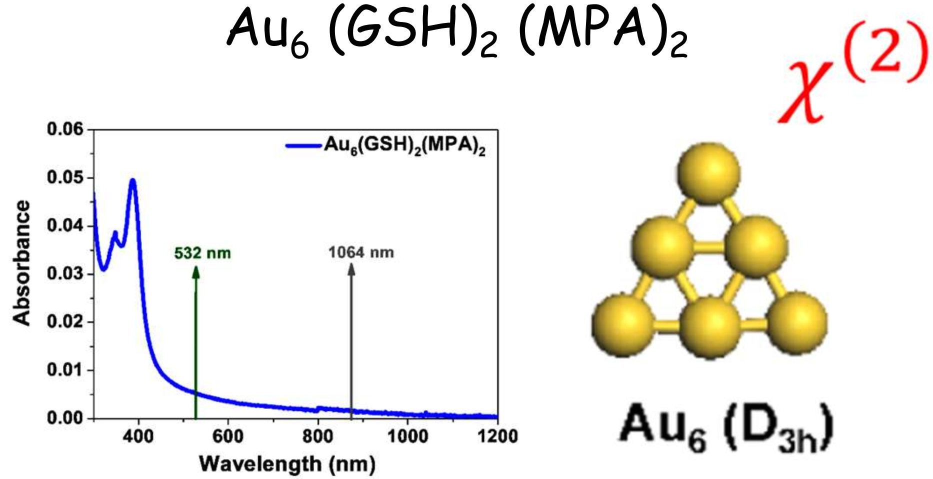


NL refraction and absorption

Saturation of 2PA

J. Phys. Chem. C 122 (2018) 18682

Au_6 $(GSH)_2$ $(MPA)_2$

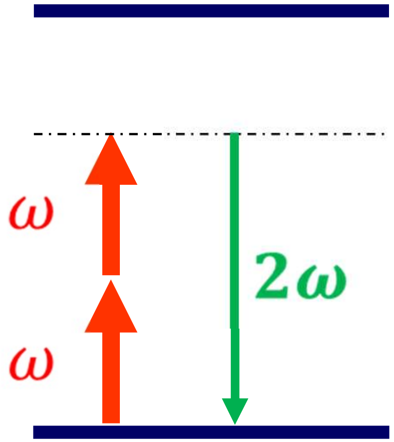


Second harmonic generation by clusters of six gold atoms

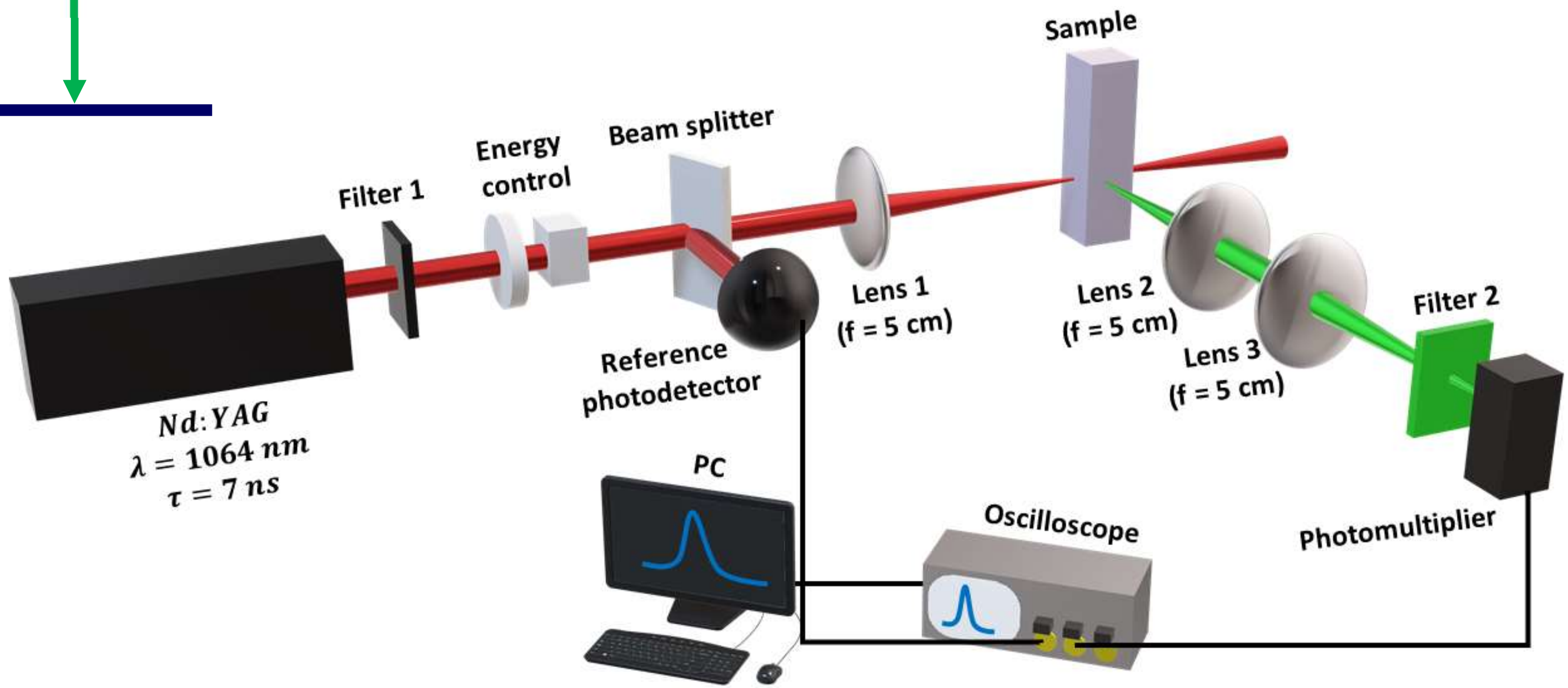
J. Phys. Chem. C 124 (2020) 15440-15447

Incoherent second harmonic generation

hyper Rayleigh scattering



$$P^{(2)} = \epsilon_0 \chi^{(2)} (E \cos \omega t)^2 = \frac{1}{2} \epsilon_0 \chi^{(2)} E^2 (1 + \cos \mathbf{2\omega} t)$$

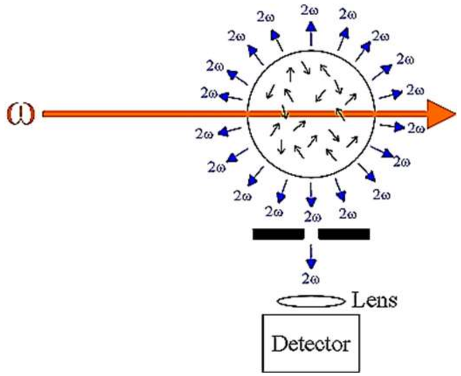


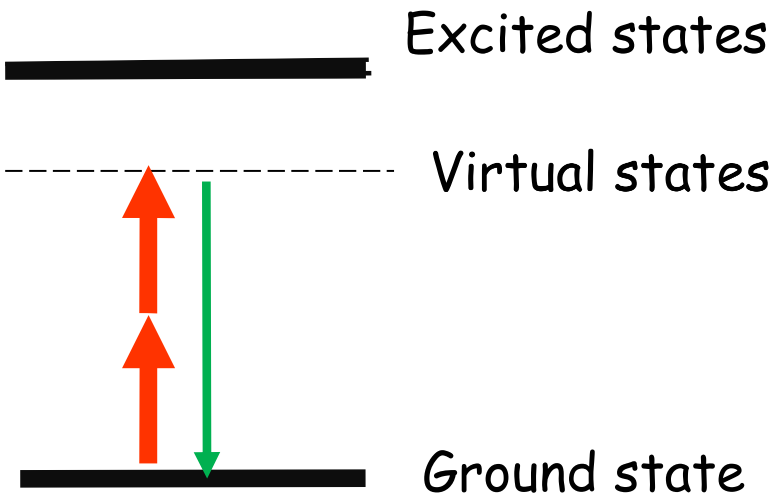
Barbosa-Silva et al. J. Phys. Chem. C 124 (2020) 15440-15447

Second order nonlinearity

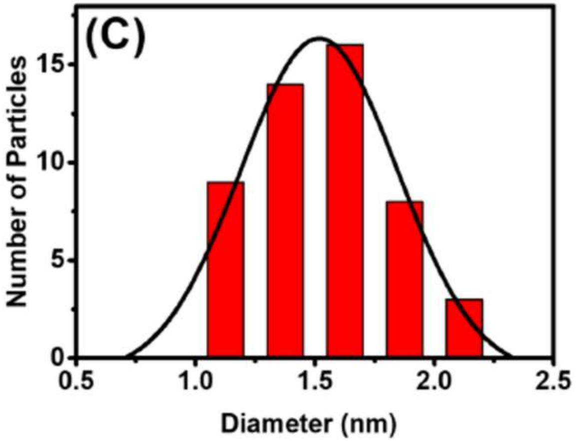
hyper-Rayleigh scattering

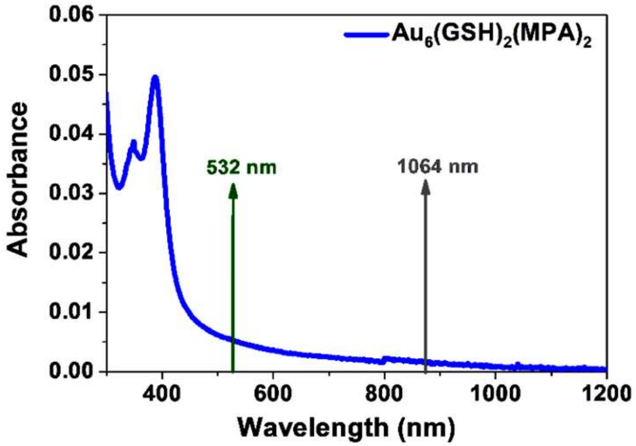
$$\chi^{(2)}$$

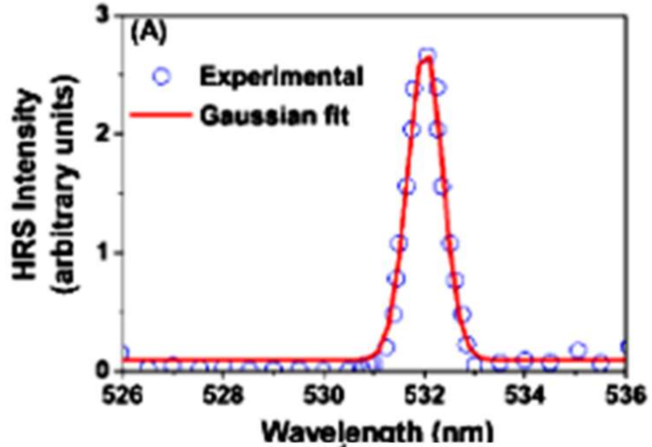




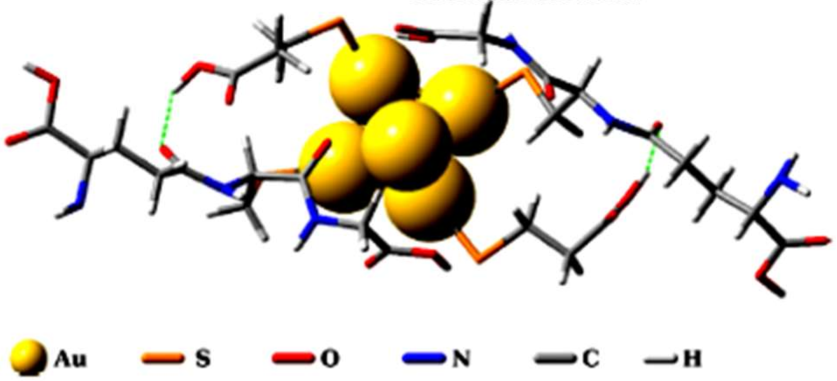
$Au_6\,(GSH)_2\,(MPA)_2$ A. Patra - Índia







Gold nanocluster	Wavelength (nm)	$10^{30}\,\beta(2\omega)$ (esu)	$10^{30}\,\beta(2\omega)$ *per gold atom* (esu)	Reference
Au_6	1064	760	127	This work
Au_{15}	802	509	34	12
Au_{25}	802	128	5.1	12

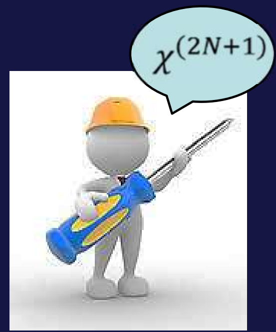


Summary

Metal composites present large NL susceptibility which depends on the shape and volume fraction of NPs

Metallic NPs can be nucleated inside different media allowing enhancement of:

- luminescence properties (Stokes and anti-Stokes)
- optical gain/amplification in waveguides
- random lasers, DFB lasers
- all-optical switching, etc.



Nonlinearity Management

The control of NPs volume fraction allows supression and/or enhancement of nonlinear optical contributions

Robust two-dimensional spatial solitons in liquid carbon disulfide
Phys. Rev. Lett. 110 (2013) 013901.

Two-dimensional solitons in a quintic-septimal medium.
Phys. Rev. A 90 (2014) 063835.

Nonlinearity management of photonic composites and observation of spatial-modulation instability due to quintic nonlinearity.
Phys. Rev. A 89 (2014) 063803.

Spatial phase modulation due to quintic and septimal nonlinearities in metal colloids. Opt. Express 22 (2014) 22456.

An optimization procedure for the design of all-optical switches based on metal-dielectric nanocomposites. Opt. Express 23 (2015) 7659 .

Robust self-trapping of optical vortex beams in a saturable optical medium. Phys. Rev. A 93 (2016) 013840.

Taming the emerging beams after the split of optical vortex solitons in a saturable. Phys. Rev. A 93 (2016) 013843.

Guiding and confinement of light induced by optical vortex solitons in a cubic-quintic medium. Opt. Lett. 41 (2016) 191.

Observation and analysis of soliton clusters
Phys. Rev. A 102 (2020) 033523

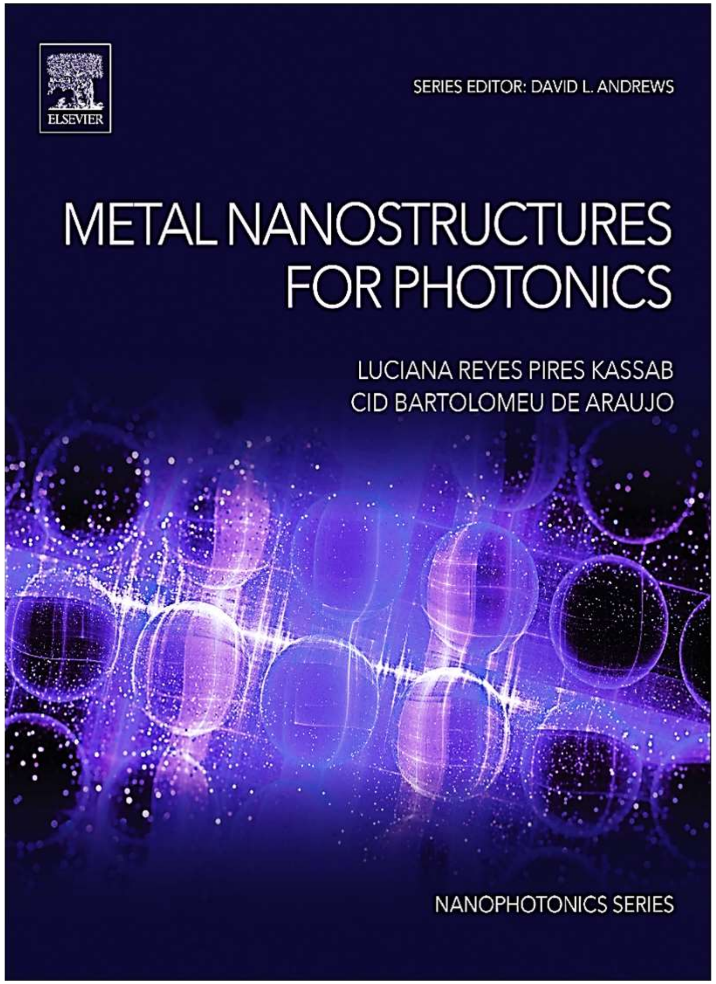
Control of the nonlinear refractive index by applied voltage
Optics Express 26 (2018) 20298

Investigation of high order nonlinearities of colloidal gold nanorods
J. Phys. Chem. C 123 (2019) 12997-13008
J. Phys. Chem. C 124 (2020) 10705-10709

Nonlinear optical behavior of gold metasurface
Nanophotonics 9 (2020) 725-740

NL refraction, NL absorption, Saturated 2PA of silver nanocluster with 29 atoms
J. Phys. Chem. C 122 (2018) 18682

First hyperpolarizability of gold clusters with six atoms
J. Phys. Chem. C 124 (2020) 15440-15447



ELSEVIER (2019)

Section I (Influence of metallic nanoparticles on luminescence of ions in solids)

Section II (Near and far-field optical phenomena associated and/or influenced by metallic nanoparticles)

Section III (Photonic materials and devices with improved performance influenced by metallic nanoparticles)

Reyna, de Araújo
High-order optical nonlinearities in plasmonic nanocomposites – a review
Adv. Opt. Photon. **9** (2017) 720-774

Reyna, de Araújo. Beyond third-order optical nonlinearities in liquid suspensions of metal-nanoparticles and metal-nanoclusters.

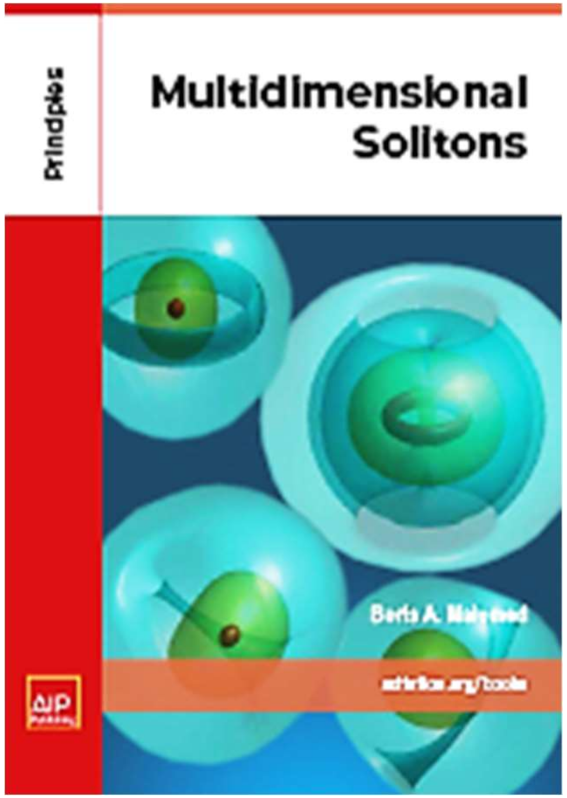
J. Opt. 24 (2022) 104006.

Review Article

Frontiers in multidimensional self-trapping of nonlinear fields and matter

Y. V. Kartashov, G. E. Astrakharchik, B. A. Malomed & L. Torner

Nature Reviews Physics **volume 1**, pages185–197 (2019)



Author: Boris A. Malomed

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