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

High-order nonlinearities in disordered media

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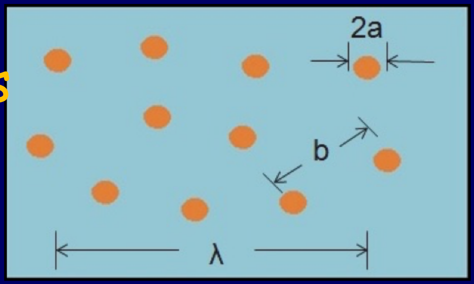
Second lecture

Transverse high-order nonlinear phenomena in composites

Metal-dielectric nanocomposites

Nanoparticles

Dielectric

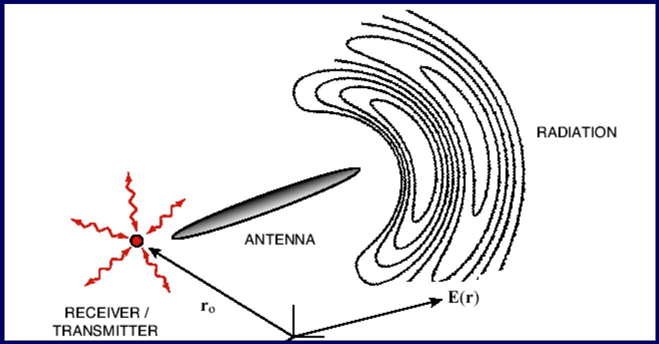


- Glasses (bulk and thin films) with metallic NPs
- Polymers with metallic nanostructures
- Liquid colloids with metallic NPs

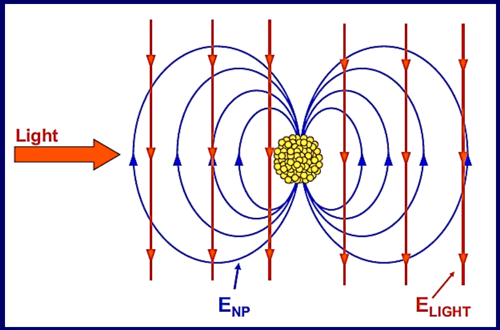
Optical response is controlled through the volume fraction of the NPs

Why metal-dielectric nanocomposites?

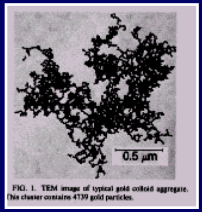
Metallic NPs as optical nanoantennas



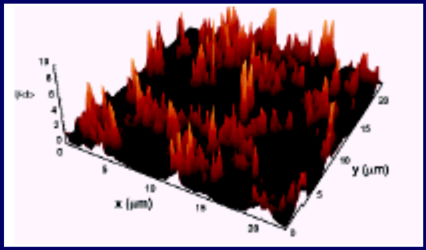
$$E_{local} = \eta \, E_{light}$$

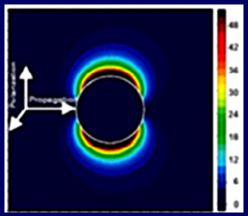


$$\eta = \frac{3\varepsilon_{NP}(\omega)}{\left[\varepsilon_{NP}(\omega) + 2\varepsilon_{h}(\omega)\right]}$$



Fractals and hot-spots



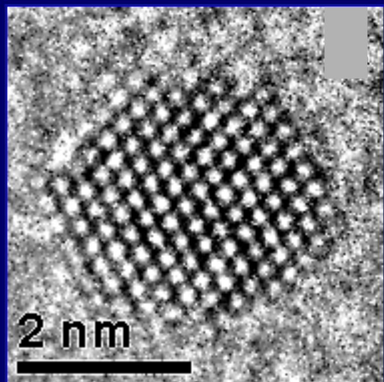


Localized Surface Plasmons

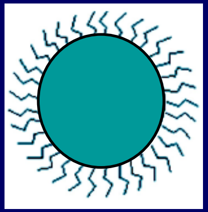
$$\mathrm{Re}\left[\varepsilon(\omega_{sp}) + 2\varepsilon_{m}(\omega_{sp})\right] = 0$$

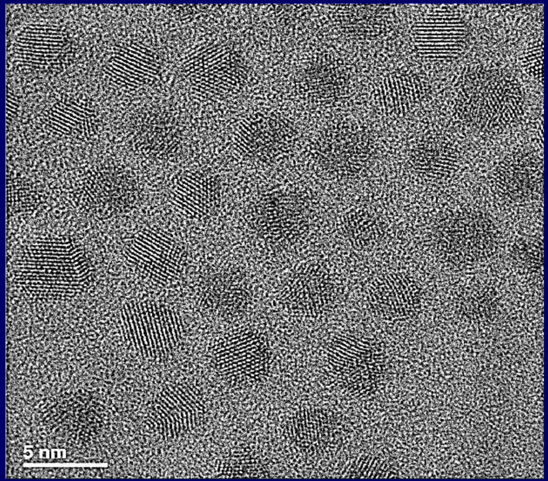
Optical response may be enhanced due to the NPs

Colloids with Ag spherical NPs

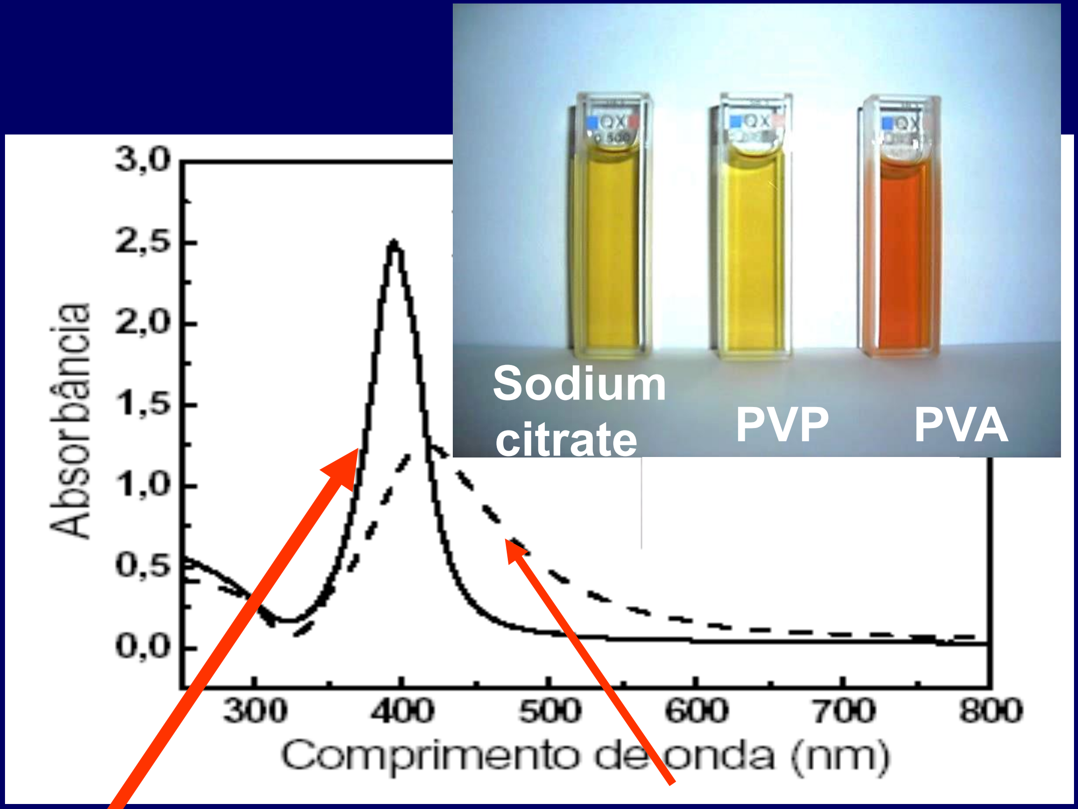


Stabilizing agents to prevent agregation





diameter: 4 nm
≈ 1500 atoms
≈ 30% in the surface

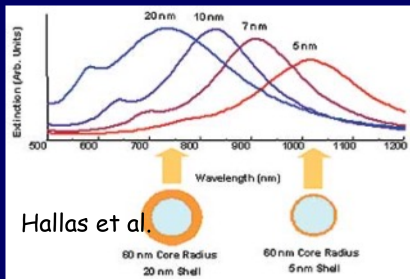


spheres

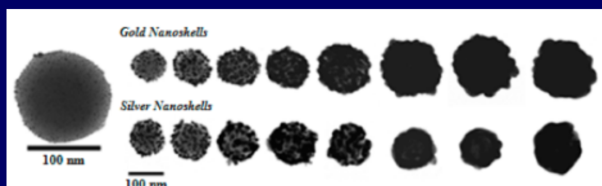
several shapes and sizes

Metallic nanoshells

Plasmon frequency depends on the ratio between the shell thickness and the core radius



Langmuir 2013
29, 4366-4372

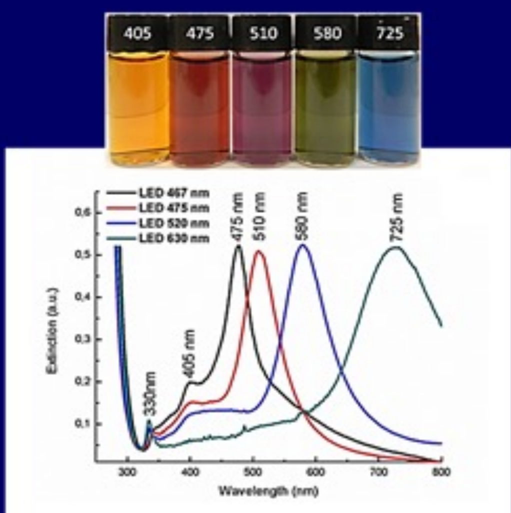


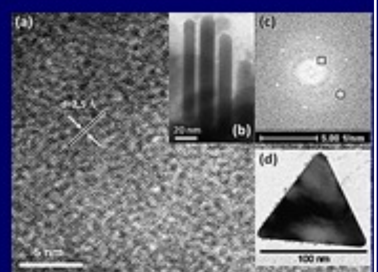
High-order nonlinearity of silica-gold nanoshells in chloroform at 1560 nm

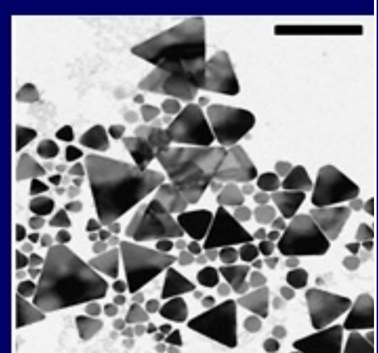
Falcão-Filho et al.
Opt. Express 18 (2010) 21616

Improved synthesis of gold and silver nanoshells
Brito-Silva et al.
Langmuir 29 (2013) 4366

Silver Nanoprisms







Mater. Chem. Phys. 2014 – to appear

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

Saade, de Araújo
Mater. Chem. Phys. 148 (2014) 1184

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

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

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

Effective 3rd. order susceptibility

Local field factor

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

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

Nonlinear response depends strongly on the laser frequency

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

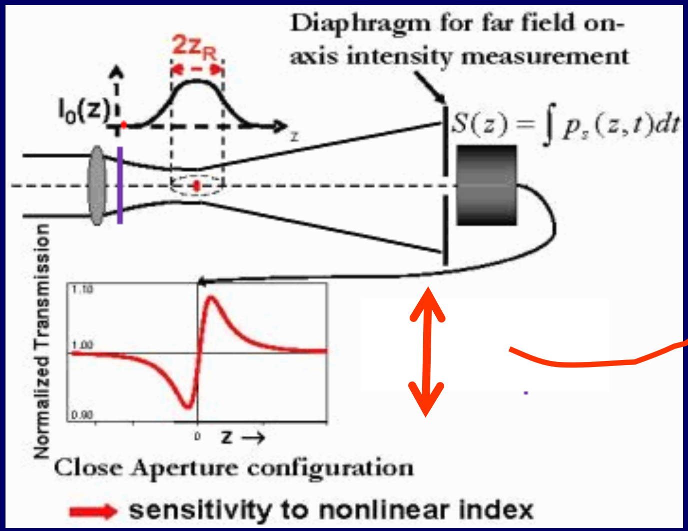
Nonlinear refraction

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

Nonlinear absorption

When high-order nonlinearities are present:

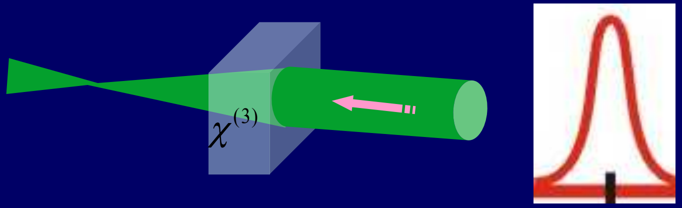
Self - focusing medium



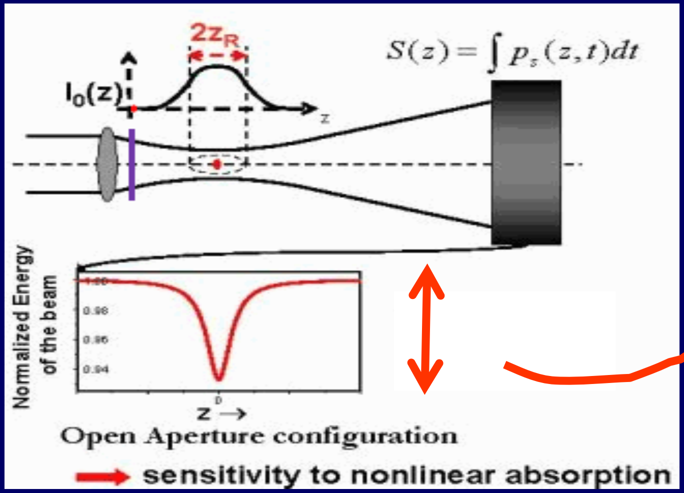
NL refraction

"Closed-aperture" Z scan

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



I (r)



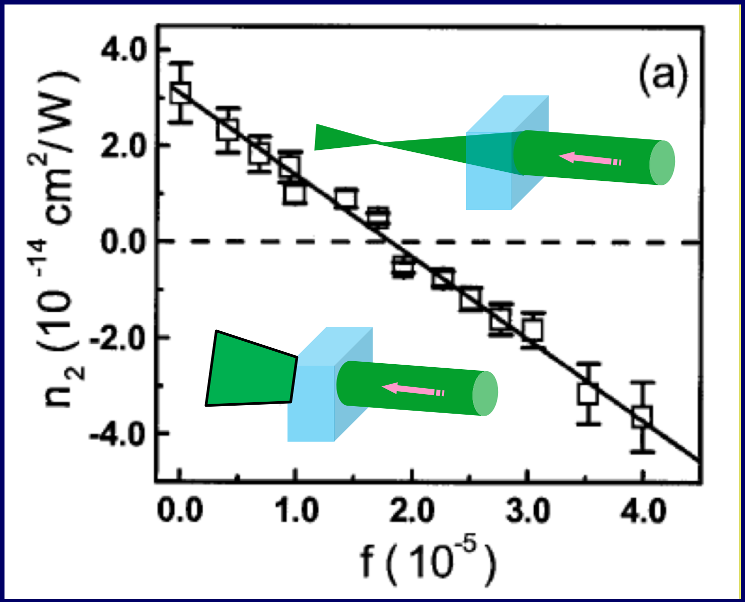
NL absorption

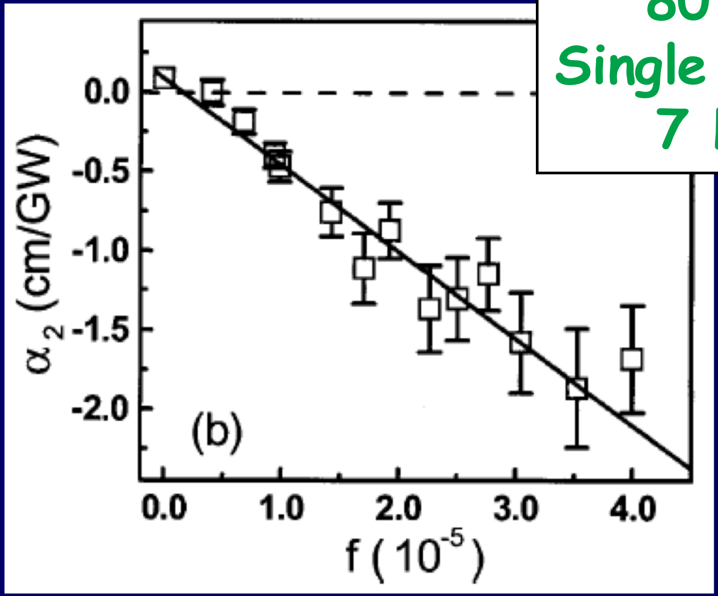
"Open-aperture" Z scan

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

Silver NPs in CS_2

532 nm
80 ps
Single pulses
7 Hz





$$\chi^{(3)}_{eff} \approx f\left(2.3 + i1.0\right)\chi^{(3)}_{NP} + \chi^{(3)}_{host}$$

$$n_2 \propto \left\{f\left(2.3\,\mathrm{Re}\,\chi^{(3)}_{NP} - 1.0\,\mathrm{Im}\,\chi^{(3)}_{NP}\right) + \mathrm{Re}\,\chi^{(3)}_{host}\right\}$$

$$\alpha_2 \propto \left\{f\left(2.3\,\mathrm{Im}\,\chi^{(3)}_{NP} + 1.0\,\mathrm{Re}\,\chi^{(3)}_{NP}\right) + \mathrm{Im}\,\chi^{(3)}_{host}\right\}$$

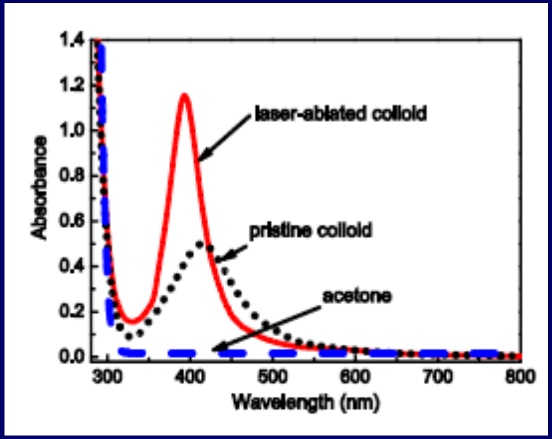
$$\chi^{(3)}_{host} = 2.9\times10^{-20} + i3.5\times10^{-22}\,(m/V)^2$$

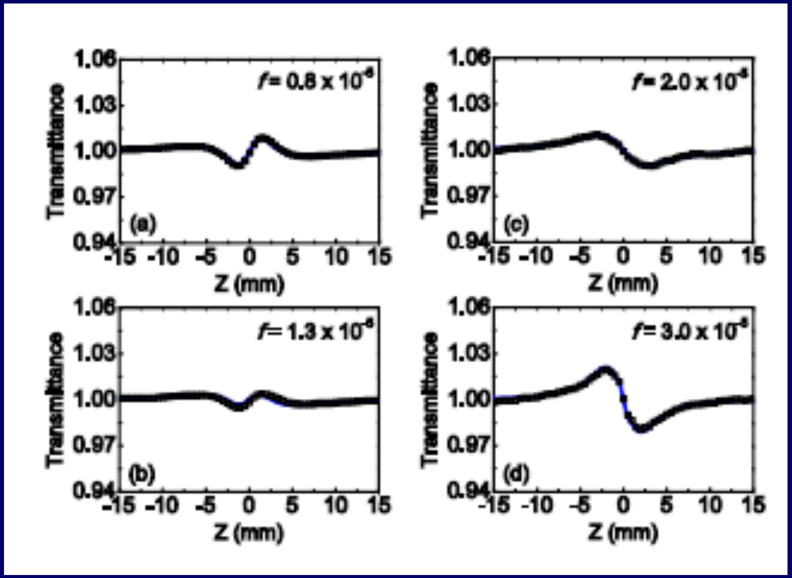
This experiment

$$\chi^{(3)}_{NP} = -(6.3 - i1.9)\times10^{-16}\,(m/V)^2$$

Observation of fifth-order refraction in a metal-colloid

Silver NPs in acetone





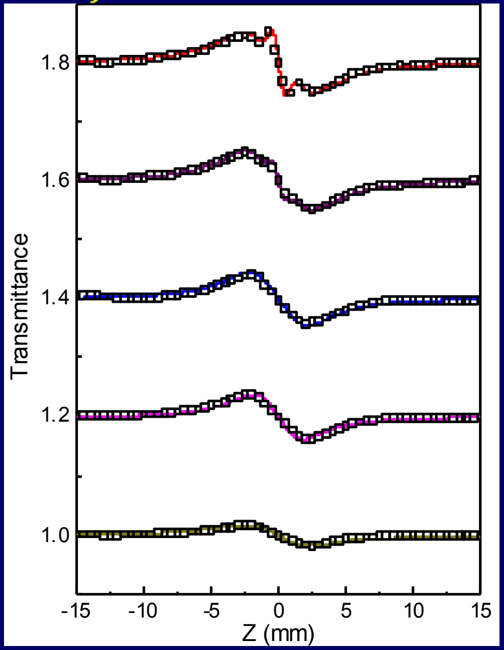
Z-scan

532 nm

Single pulses

5 GW/cm^2

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



9

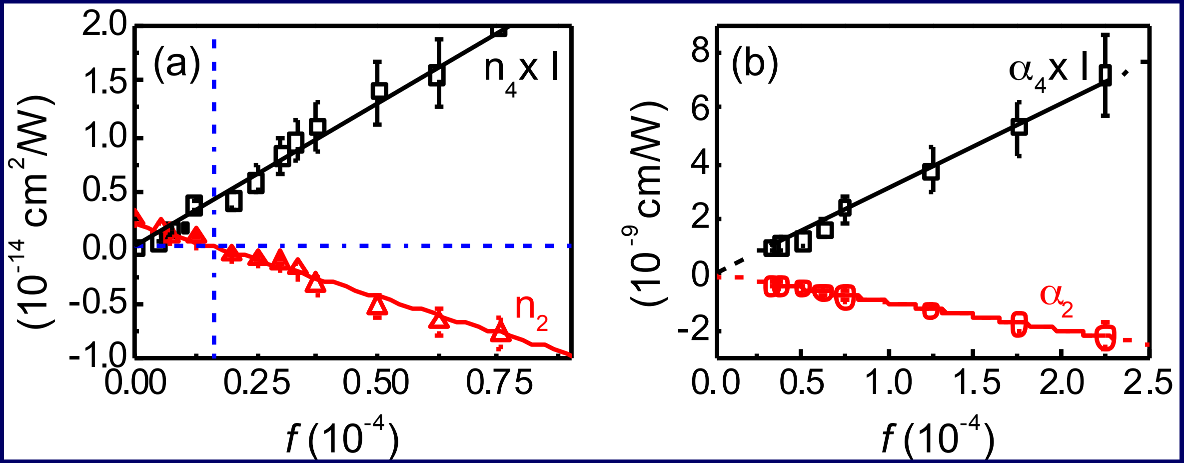
8

6

4

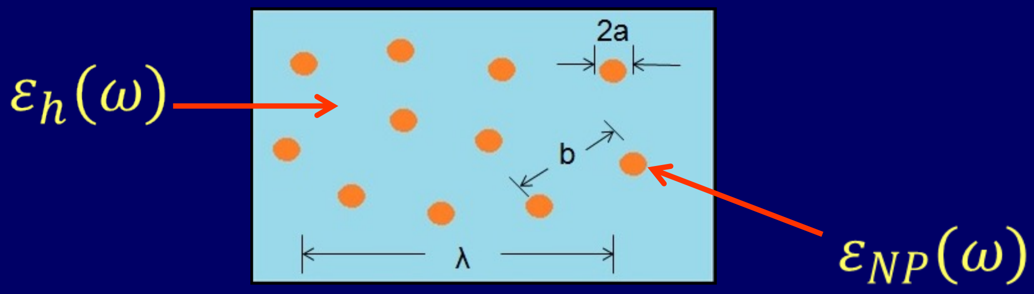
2

GW/cm^2



9 GW/cm^2

Generalized Maxwell-Garnet model



Ag NPs in acetone

$\chi^{(5)}_{host}$ – *negligible*

$a < b < \lambda$

Maxwell-Garnet model

$$\chi^{(3)}_{eff} = f\, L^2 |L|^2 \chi^{(3)}_{np} + \chi^{(3)}_h ,$$

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

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

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

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

Nonlinearity Management

A procedure to obtain exotic metal-dielectric composites

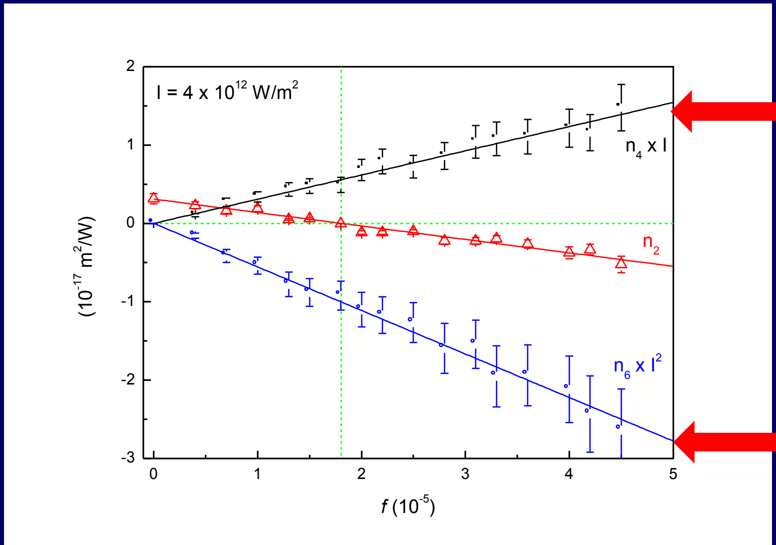
It is possible to supress one specific order of nonlinearity and enhance another one

Example: n_2=0 and n_4≠0

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

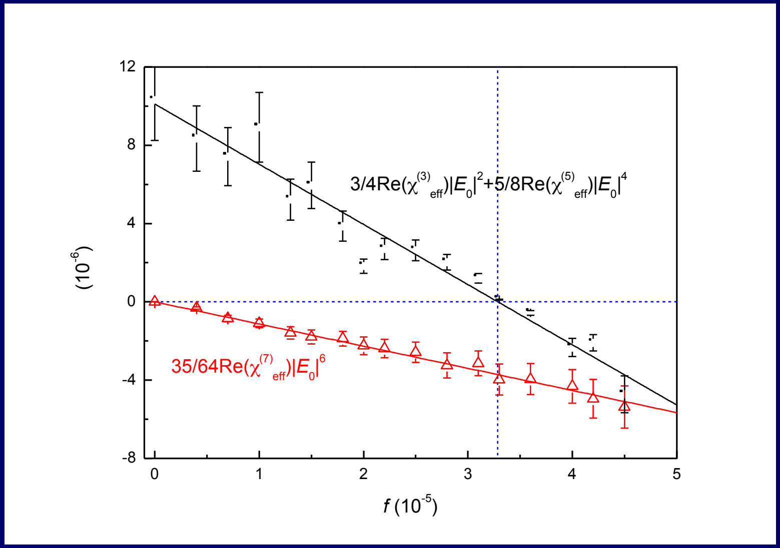
$n_4 \propto Re\chi^{(5)}_{eff}$

Nonlinearity management: Silver NPs + CS_2



$n_4 I$

$n_6 I^2$



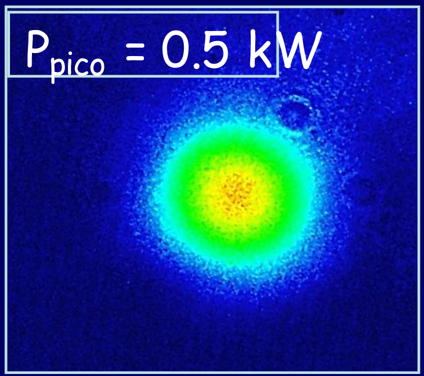
4×10^8 W/cm^2

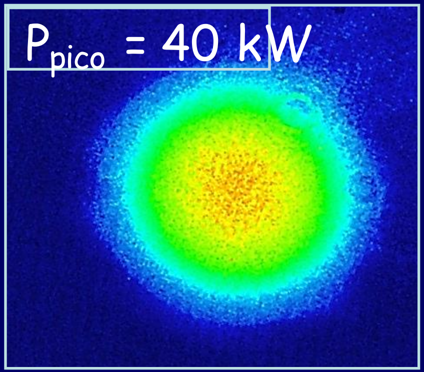
1×10^8 W/cm^2

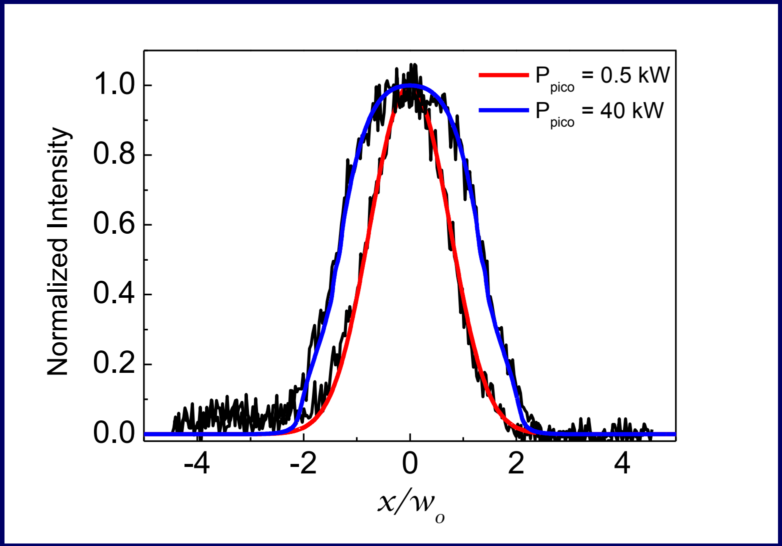
Effective $\chi^{(7)}$

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

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

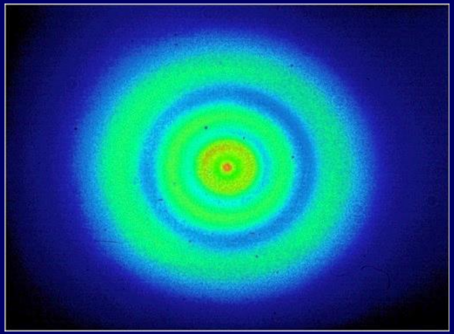


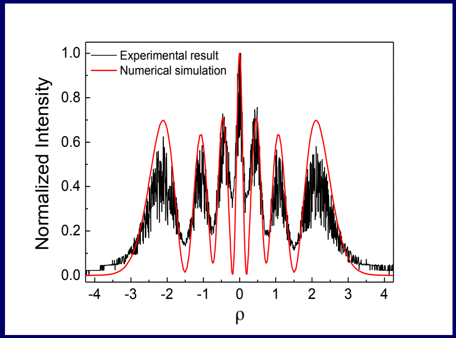


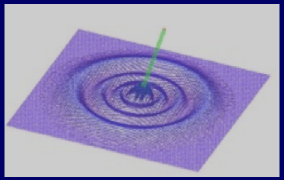


Input plane out of the lens focus

$I = 70\ GW/cm^2$







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

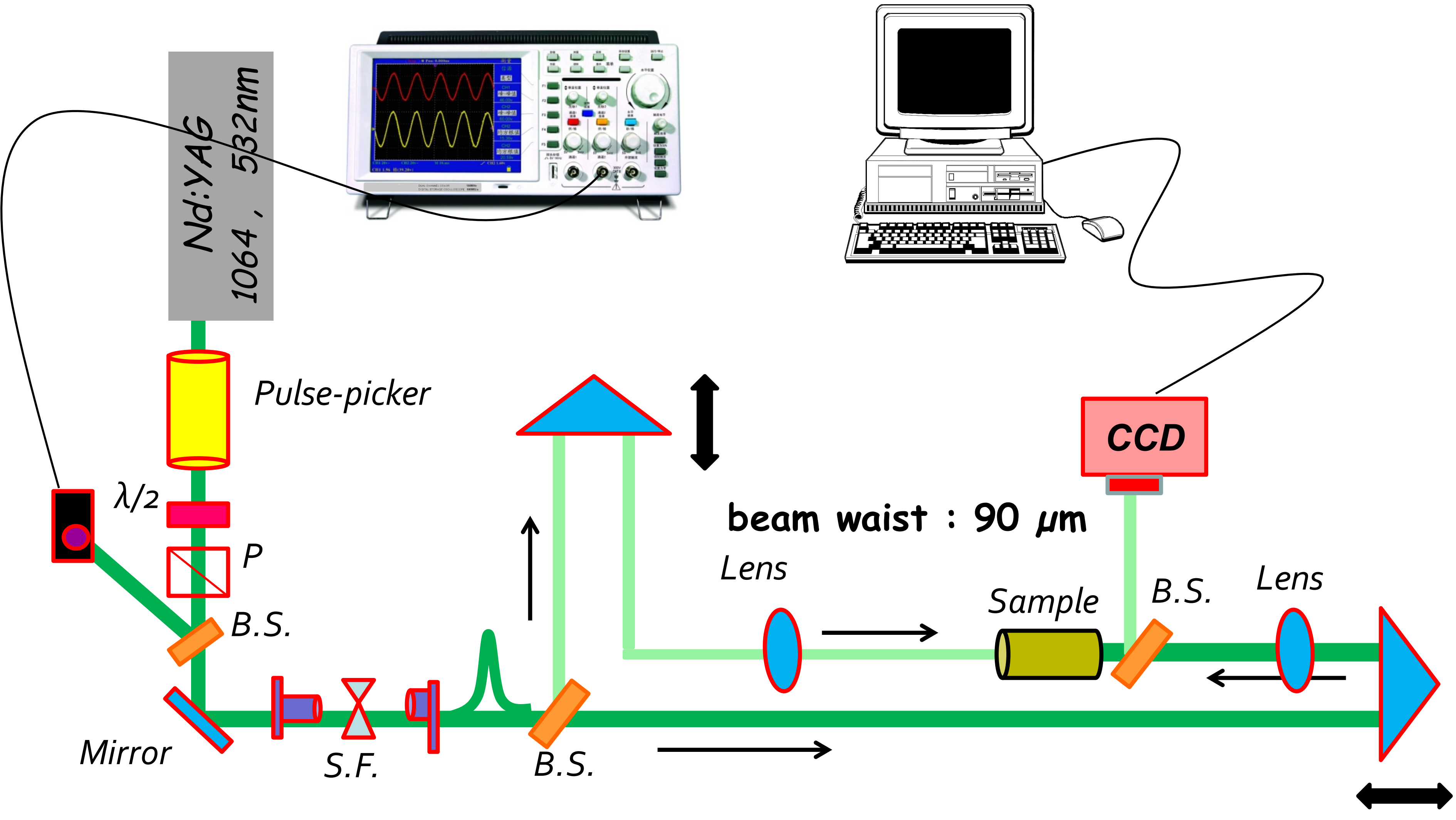
Cross-phase modulation with two counter – propagating beams

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

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

Third-order ↑ (n_2) Fifth-order ↑ (n_4)

Spatial Cross-Phase Modulation



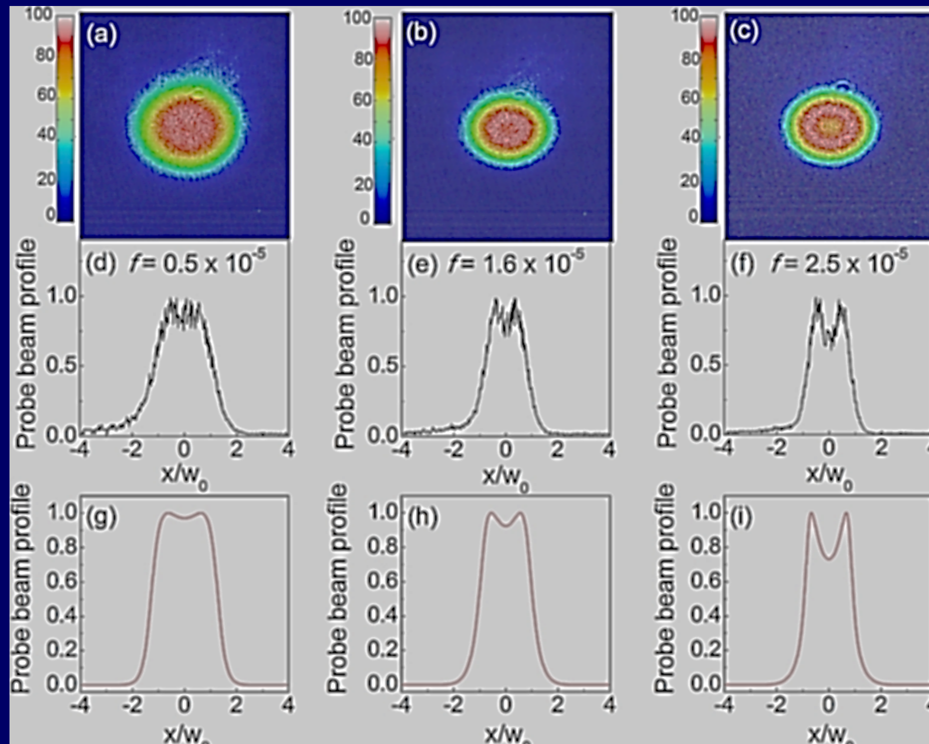
NPs: 9 nm, L=5 cm, I_{pump}=2 GW/cm^2 , I_{probe} = 0.1 I_{pump}

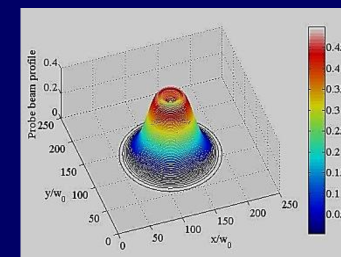
Counter-propagating beams
First observation of Spatial Modulational Instability due to $\chi^{(5)}_{eff}$

Probe beam profile

Profiles from the images

Theory





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

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

Ag NPs + acetone

$n_2 = 0$

$n_4 = +3.2\times10^{-25}\, cm^4 / W^2$

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

Cross-phase modulation

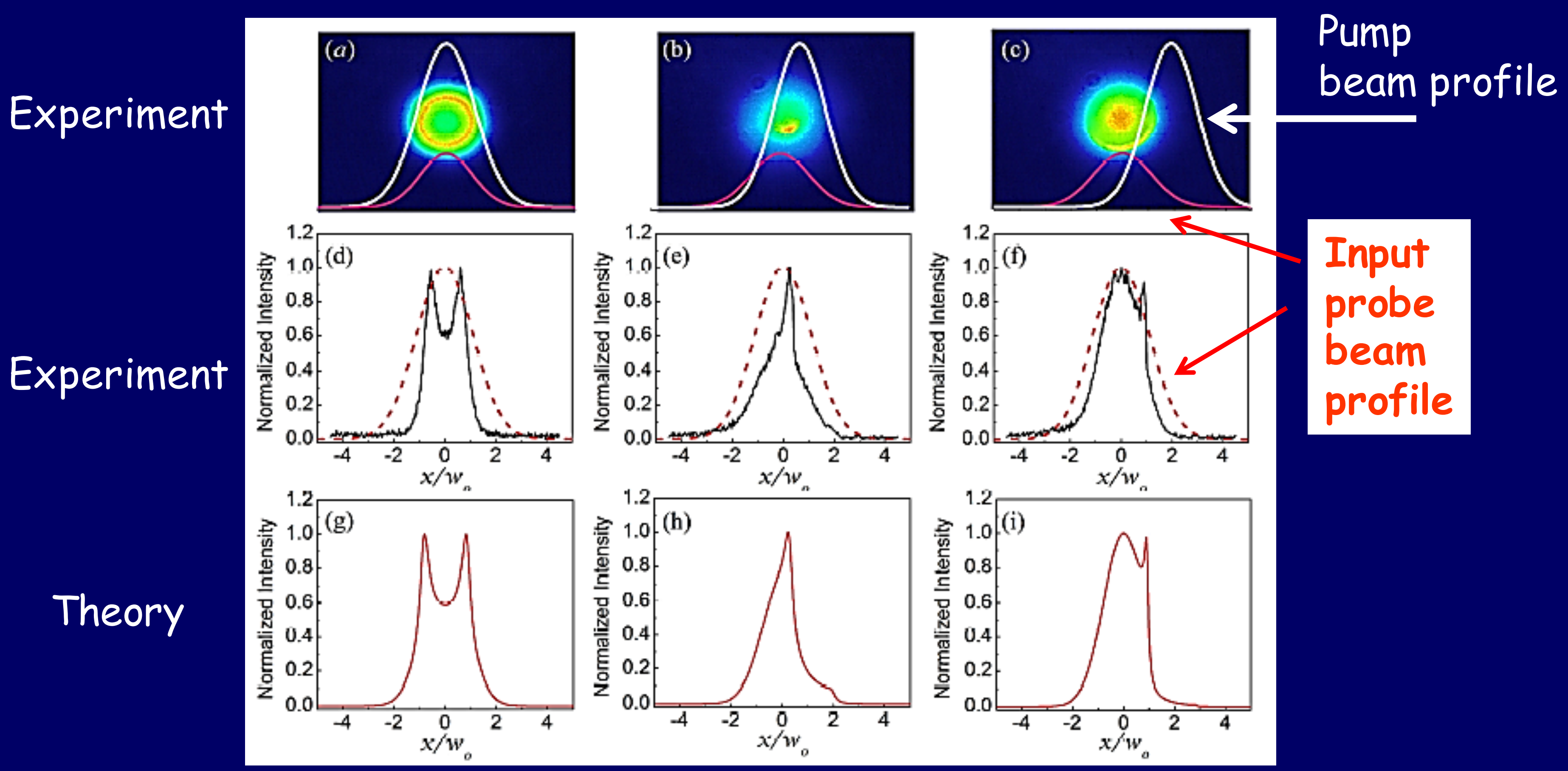
Co-propagating beams

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

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

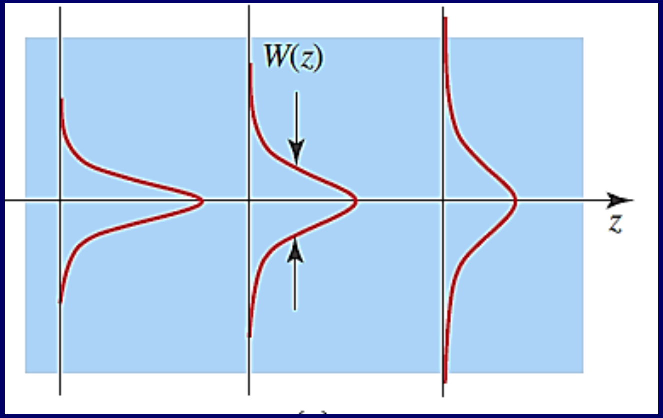
Reyna, de Araújo - Optics Express 22 (2014) 22456

Induced focusing due to the seventh-order susceptibility

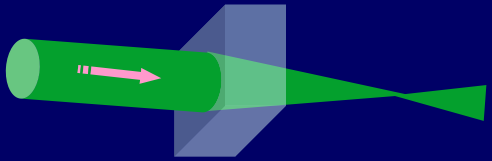


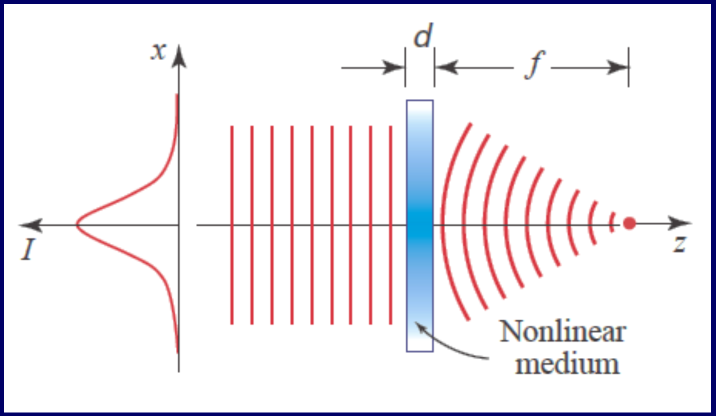
$$n_2 = 0; \quad n_4 = 0; \quad n_6 < 0$$

Diffraction

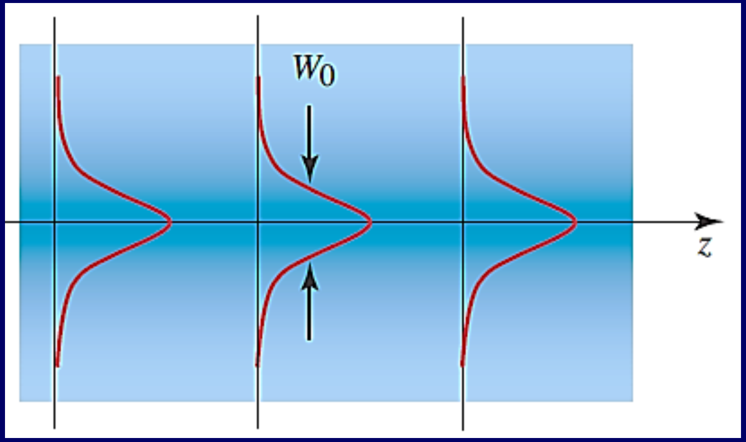


Self-focusing



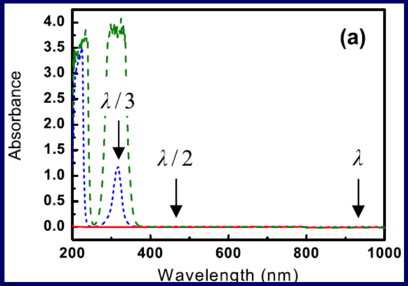


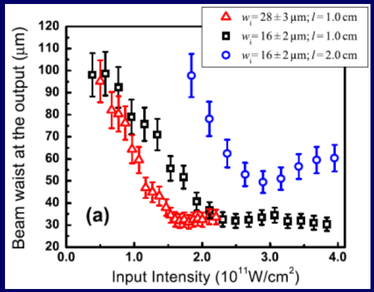
Bright Spatial Soliton

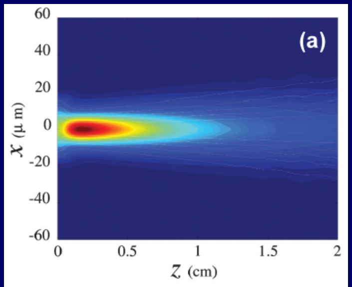


First demonstration of (2+1)D soliton propagating in a homogeneous medium with local nonlinearity

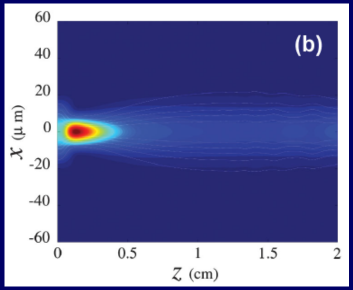
Falcão-Filho, de Araújo, Boudebs, Leblond, Skarka
Robust two-dimensional spatial solitons in liquid carbon disulfide
Phys. Rev. Lett. 110 (2013) 013901.







Low intensity



soliton

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

CS_2: stable (2+1)D soliton

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

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

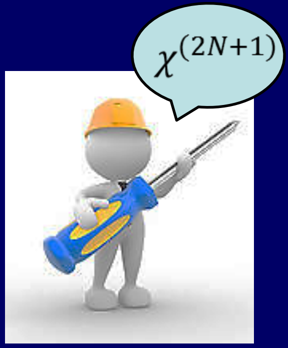
$Re\ \chi^{(3)} = 0$, $Re\ \chi^{(5)} > 0$, $Re\ \chi^{(7)} < 0$?

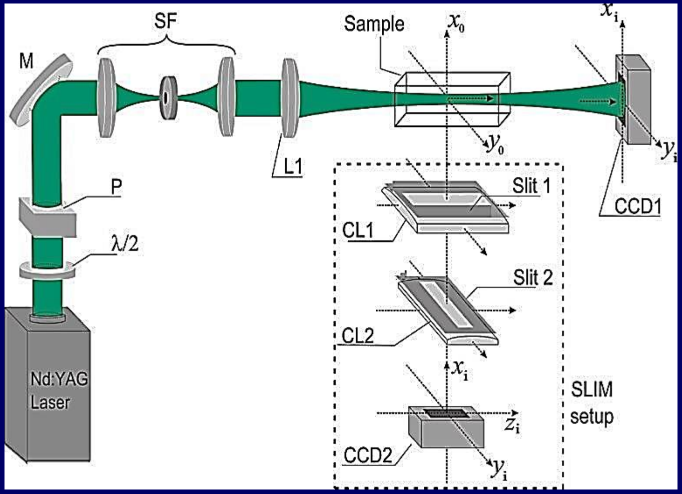
$$2ik\frac{\partial E}{\partial z} + \Delta E = -\frac{\omega^2}{c^2}\left[3\chi_{eff}^{(3)}|E|^2 E + 10\chi_{eff}^{(5)}|E|^4 E + 35\chi_{eff}^{(7)}|E|^6 E\right]$$

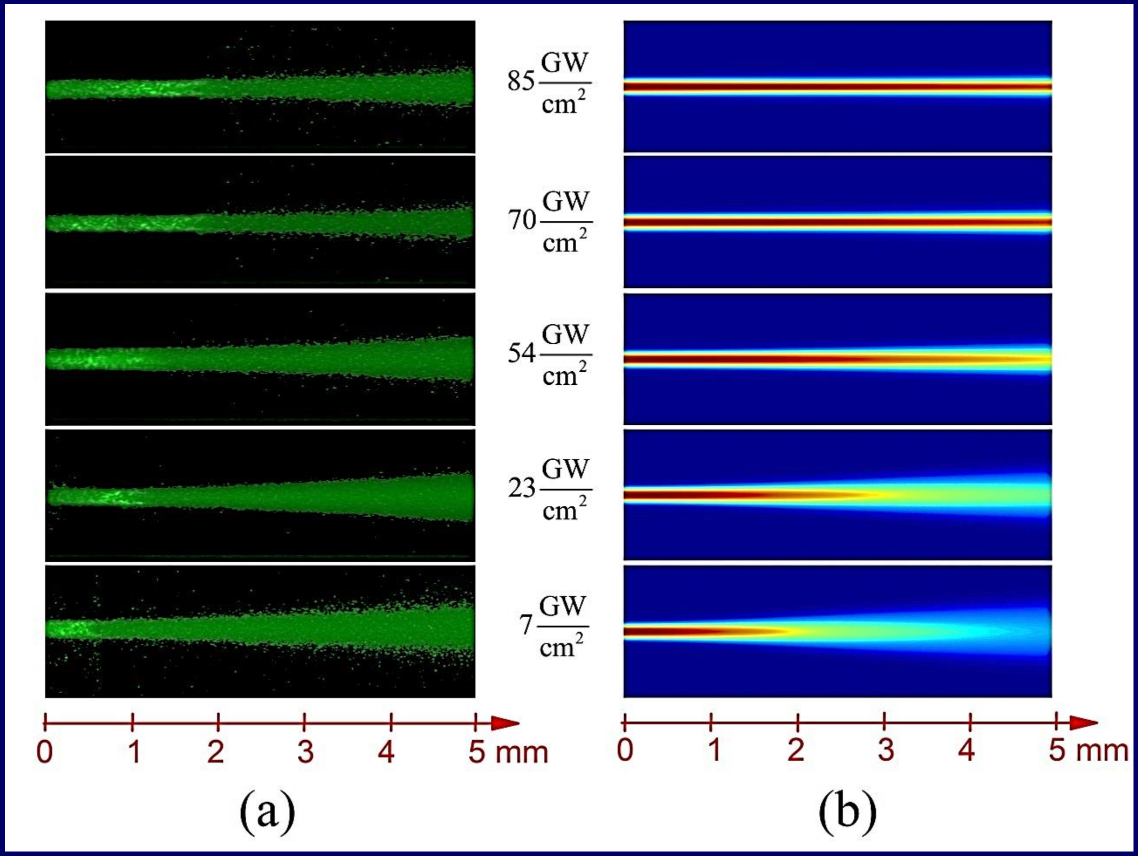
First observation of 2D Spatial-Solitons in a quintic-septimal medium

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

Silver NPs in acetone





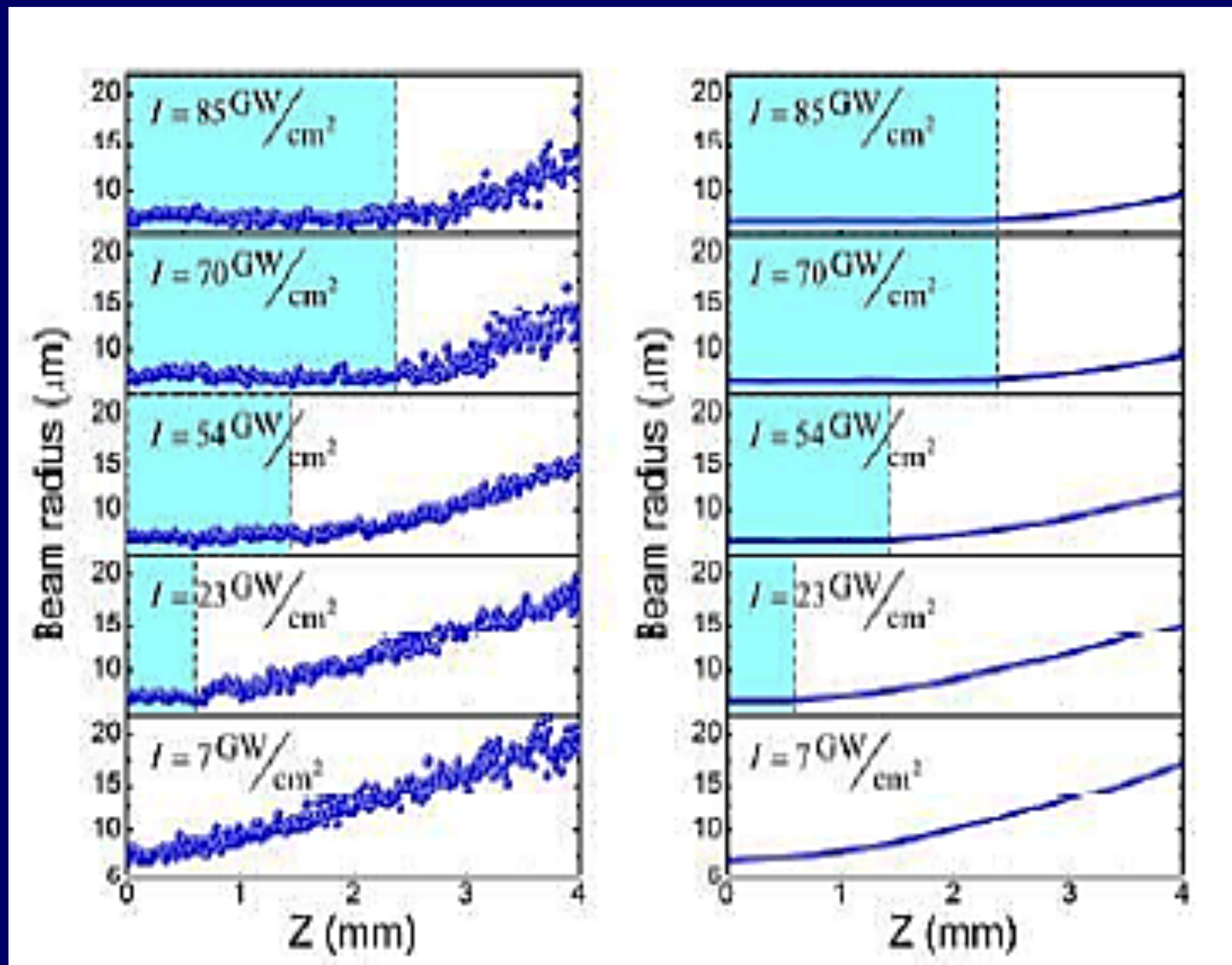


experiment theory

Reyna, Jorge, de Araújo, Phys. Rev. A 90 (2014) 063835

de Araújo et al. , J. Lumin. 169 (2016) 492-496

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

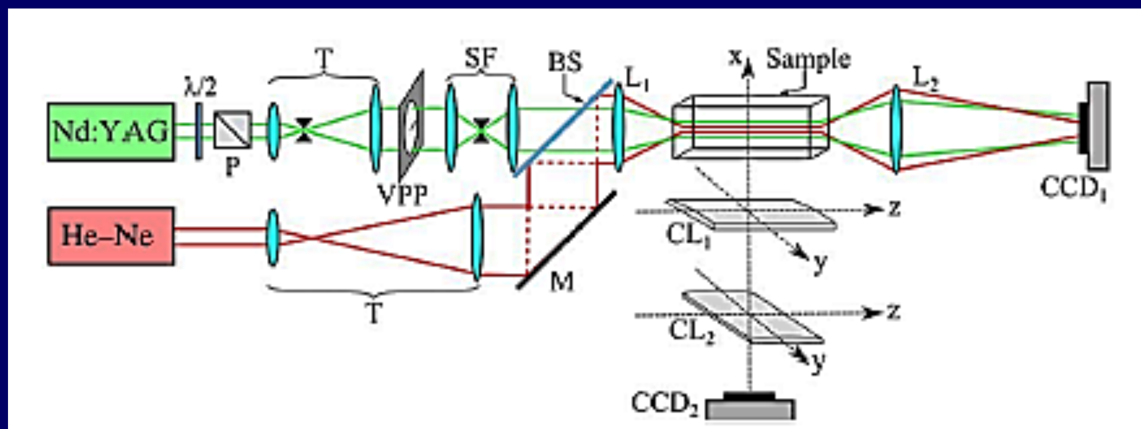


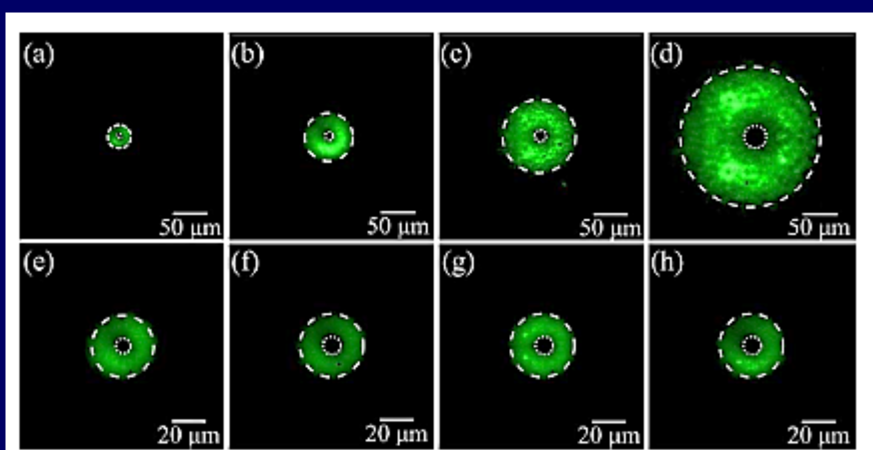
experiment theory

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



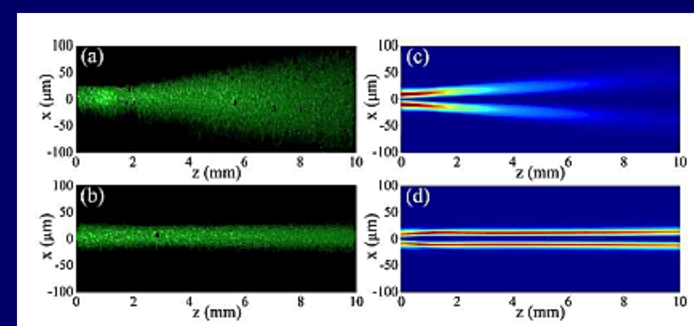


Z= 0 3 5 10 mm

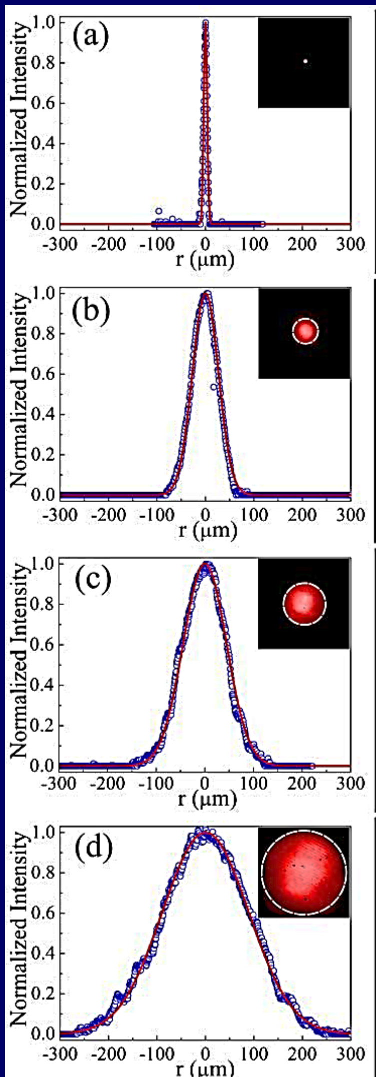
0.1

3.0

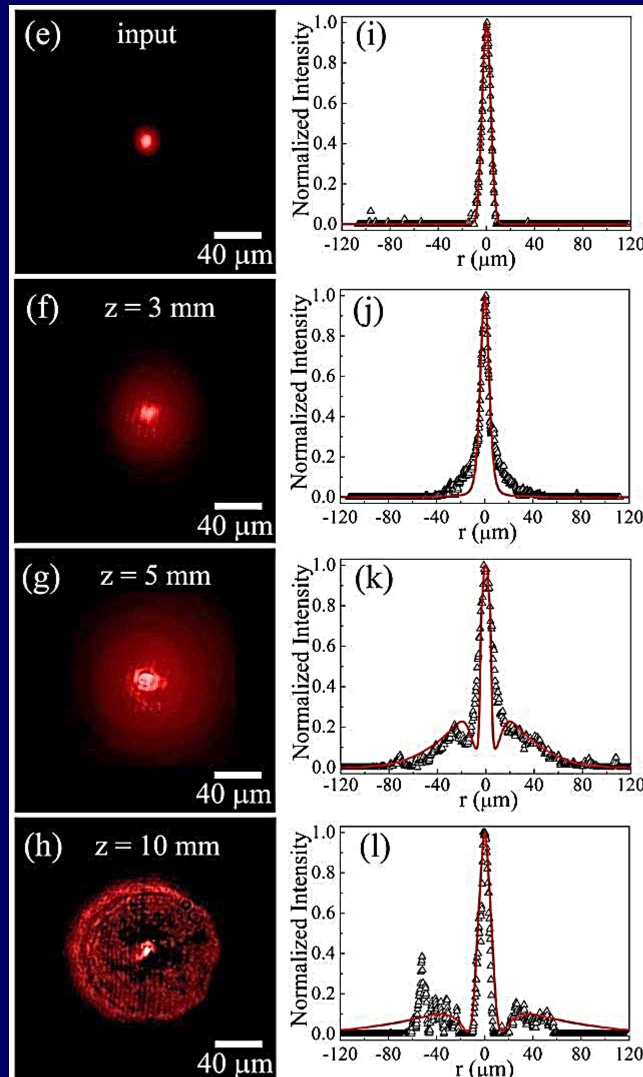
GW/cm^2



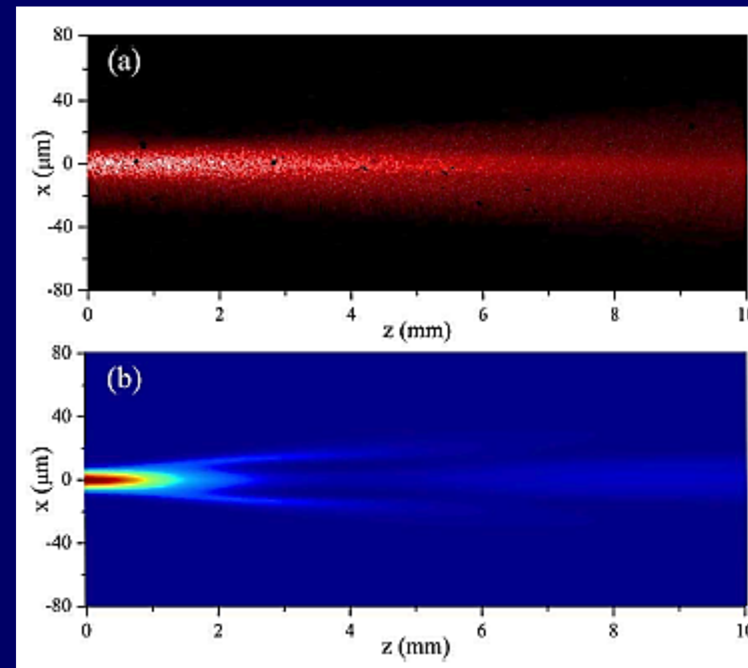
10 mm - 25 Z_R



HeNe



Guided HeNe



$I_{OVS} = 3.0\ GW/cm^2$

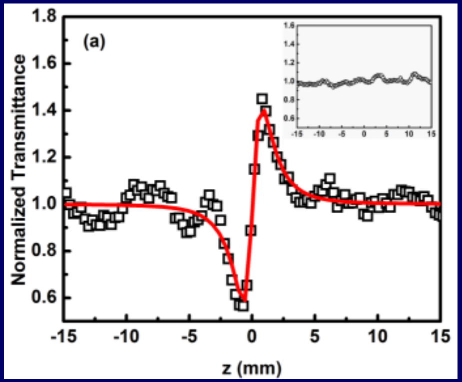
$I_{HeNe} = 0.1\ GW/cm^2$

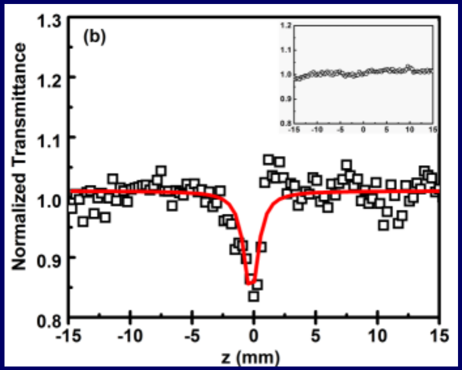
How to address the long standing problem of discovering a very good material for all-optical switching?

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

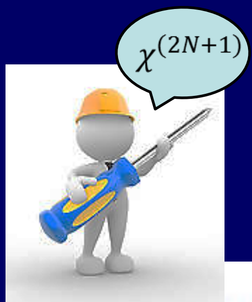
Germanate film	$n_2/\lambda\alpha_2$
As grown	8.3×10^{-4}
With Au NPs	$>2.1 \times 10^{-1}$

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

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

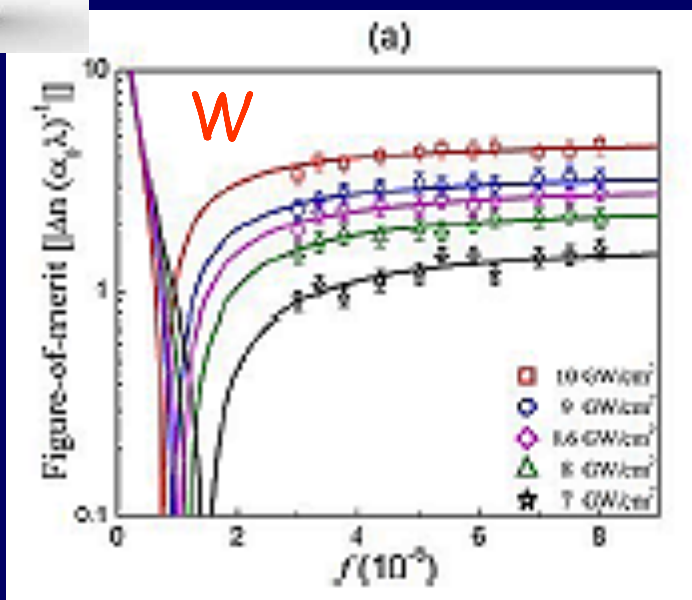
$$\Delta n = n_2 I + n_4 I^2 + n_6 I^3 + \cdots = n_{NL} I$$

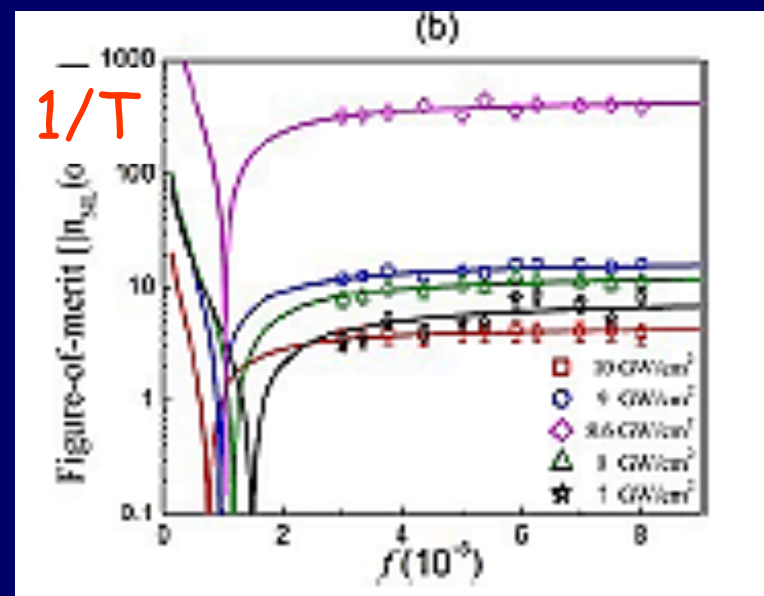
$$\alpha_{NL} = \alpha_2 + \alpha_4 I + \alpha_6 I^2 + \cdots$$



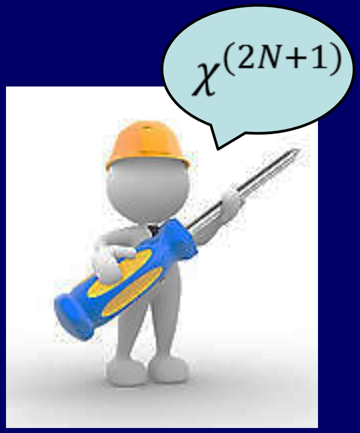
$$W = \frac{\Delta n}{\lambda \alpha_0} > 1$$

$$T = \frac{\lambda\, \alpha_{NL}}{n_{NL}} < 1$$





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



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

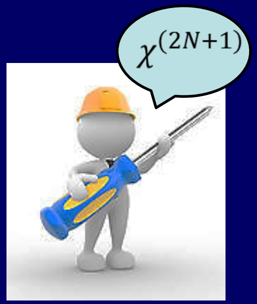
Challange for
materials scientists

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.

Thank you for your attention

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