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

High-order nonlinearities in disordered media

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Founded in 1535

1.7 milions of inhabitants

1st. High-order optical nonlinearities. Pure and homogeneous systems.

2nd. Transverse high-order nonlinear phenomena in nanocomposites.

3rd. Multiphoton absorption and stimulated emission in random media.

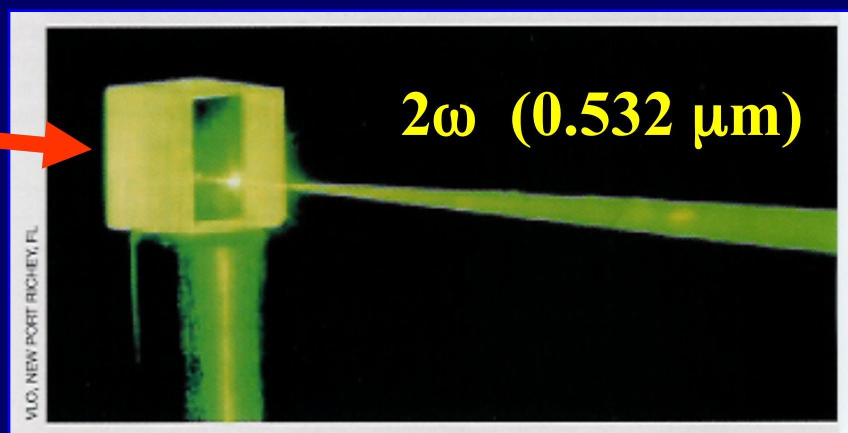
VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

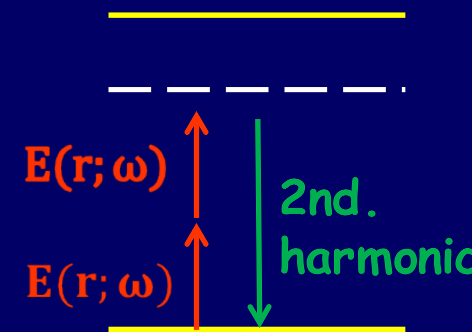
GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

Crystalline quartz

ω (1.064 μm)

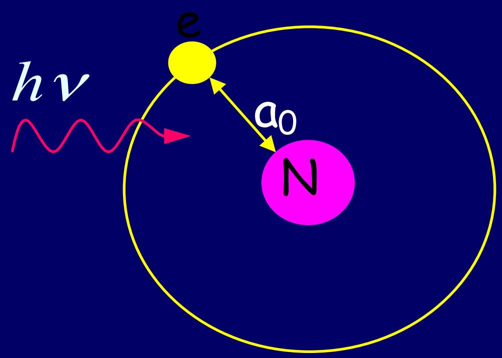




$$k(2\omega) = 2k(\omega)$$

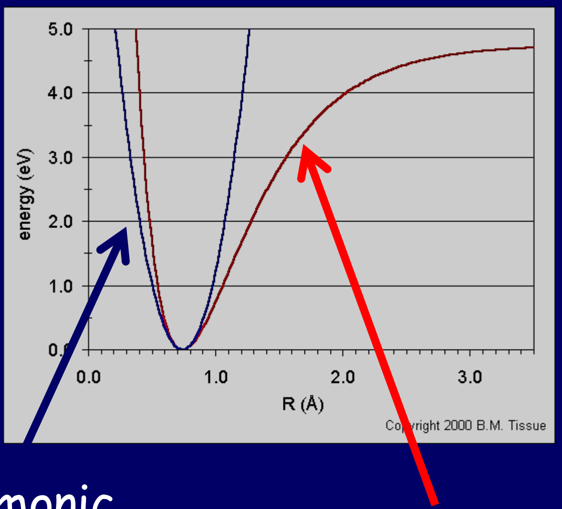
Phase matching

Atom as anharmonic oscillator



$$E_{at} \approx 2\times10^{-7}\,\text{esu}$$

3 x 10^{8} V/m



Harmonic oscillator

Anharmonic oscillator

$$U(x) = \frac{1}{2}m\omega_0^2x^2 + \frac{1}{3}mKx^3 + \frac{1}{4}mK'x^4 + \cdots.$$

$$\omega_0 = \sqrt{\frac{K_0}{m}}$$

Restoring force

$$F(x) = -\frac{dU}{dx} = -m\omega_0^2x - mKx^2 - \cdots$$

$$m\frac{d^2x}{dt^2} + \gamma\frac{dx}{dt} + m\omega_0^2 x + mKx^2 + \cdots = -eE(\omega,t)$$

$$P(\omega,t) = -\mathbb{N}ex(\omega,t)$$

$$P = P^{(1)} + P^{(2)} + P^{(3)} + P^{(4)} + \cdots$$

$$P^{(N)} = \epsilon_0\chi^{(N)}E^N$$

second harmonic generation

$$E(\omega,t) = E\cos\omega t$$

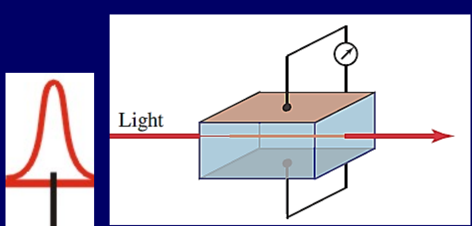
$$P^{(2)} = \varepsilon_0\chi^{(2)}(E\cos\omega t)^2 = \frac{1}{2}\varepsilon_0\chi^{(2)}E^2(1+\cos 2\omega t)$$

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

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

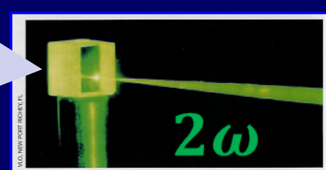
$$P_0^{(2)} + P_{2\omega_1}^{(2)} + P_{2\omega_2}^{(2)} + P_{\omega_1+\omega_2}^{(2)} + P_{\omega_1-\omega_2}^{(2)}$$

Optical retification

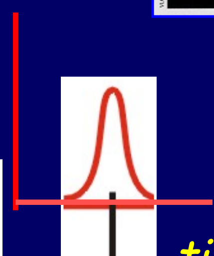


Light pulse

ω

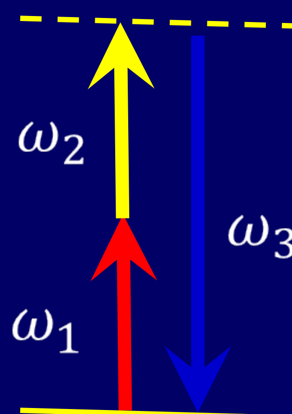


Voltage

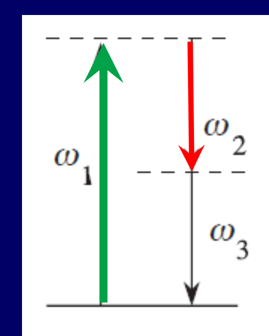


time

SFG



DFG

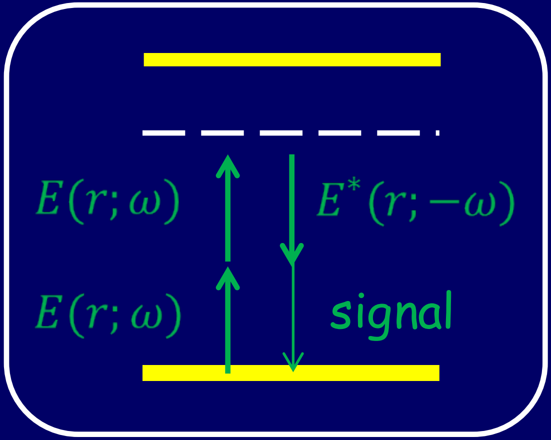


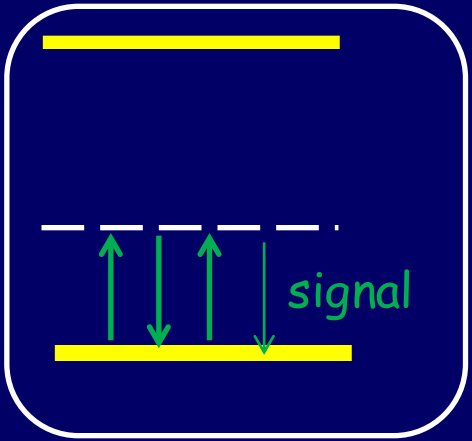
$\chi^{(3)}$: third order polarization

$$P^{(3)}(r;\omega)e^{-i\omega t} = \varepsilon_0\left[\chi^{(3)}(r;\omega,\omega,-\omega,\omega)E(r;\omega)E^*(r;-\omega)E(r;\omega)\right]e^{-i\omega t}$$

Four wave-mixing - 4WM

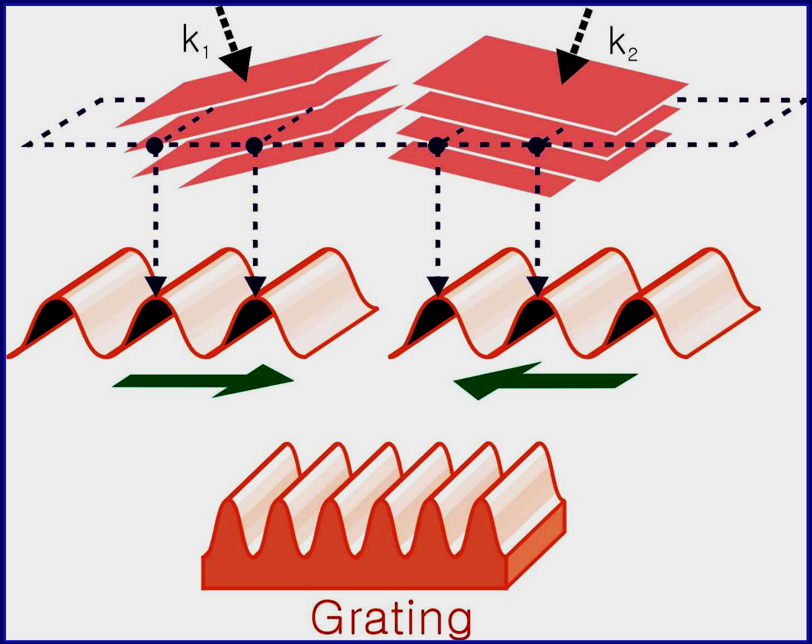
Transparent medium

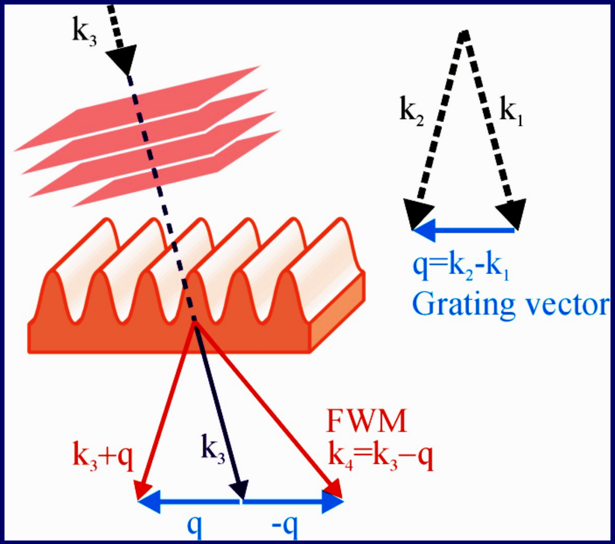




Parametric process. No changes in the population of states.

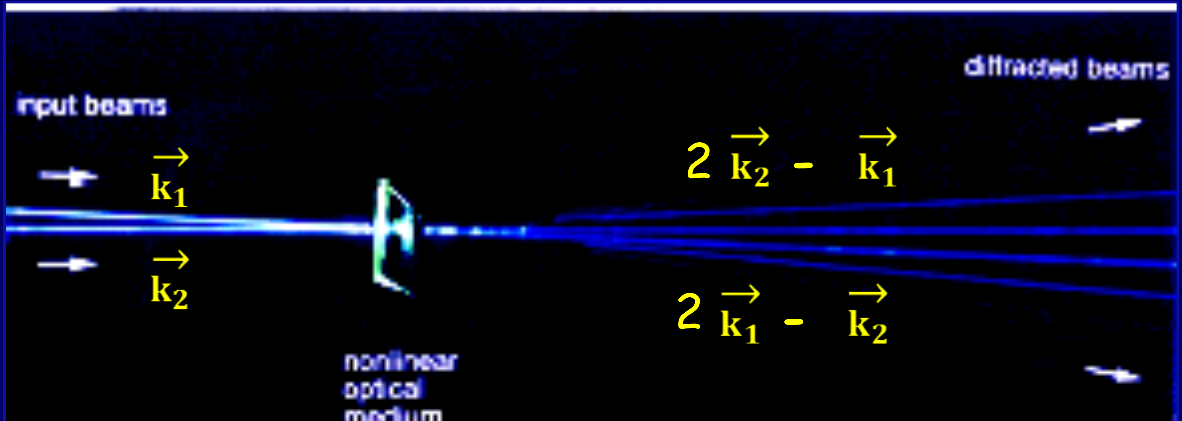
$\chi^{(3)}$: degenerate 4WM





$$P^{(3)} = \varepsilon_o \chi^{(3)} E^3$$

Only one frequency



Boyd, Escola Swieca (Belo Horizonte, 2004))

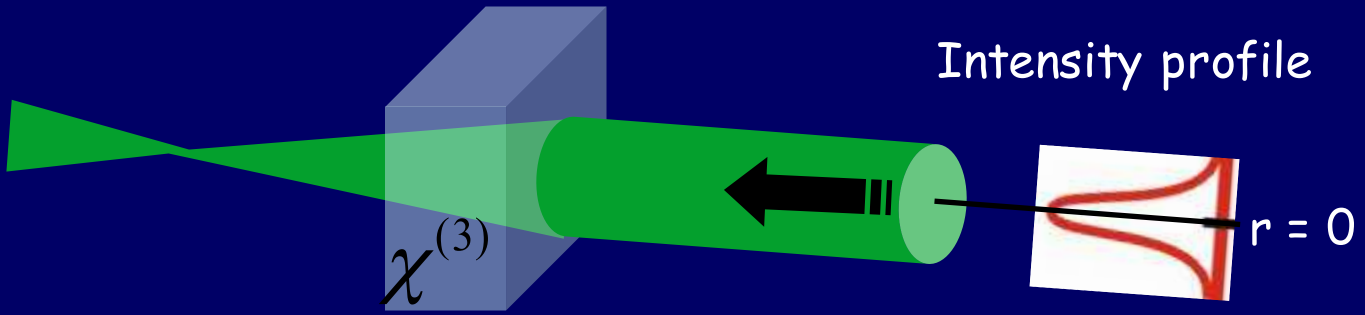
$\chi^{(3)}$: self-focusing

$n_2 \propto \mathrm{Re}\,\chi(3)$

$$E(\omega)\exp\left(\frac{-r^2}{\Delta^2}\right)e^{-i\omega t}$$

$$n = n_0 + n_2 I_0 \exp\{-r^2 / w^2\}$$

$n_2 > 0$



Sample behaves like a biconvex lens

$n_2 \propto \mathrm{Re}\,\chi(3)$

Mechanism	n_2 (cm²/W)	$\chi^{(3)}_{1111}$(esu)	Response time (sec)
Electronic Polarization	10^{-16}-10^{-13}	10^{-14}-10^{-11}	10^{-15}
Molecular Orientation	10^{-14}	10^{-12}	10^{-12}
Electrostriction	10^{-14}	10^{-12}	10^{-9}
Saturated Absorption	10^{-10}	10^{-8}	10^{-8}
Thermal effects	10^{-6}	10^{-4}	10^{-3}

$\Delta n(r=0) = n_2 I_0$

$I_0 = 1$ GW/cm²

$\Delta n = 10^{-7} - 10^{-4}$

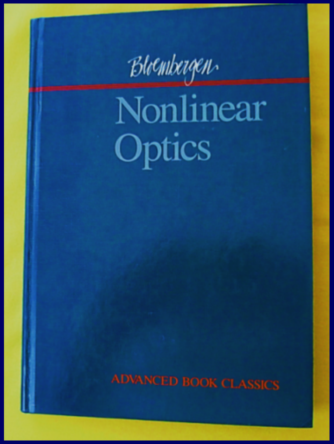
Electronic polarization

General theoretical approach

PHYSICAL REVIEW VOLUME 127, NUMBER 6 SEPTEMBER 15, 1962

Interactions between Light Waves in a Nonlinear Dielectric*

J. A. Armstrong, N. Bloembergen, J. Ducuing,† and P. S. Pershan
Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts



When there is inversion symmetry:

$$\chi^{(j)} \equiv 0$$

j = even

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

$$n_N \propto Re\, \chi^{(2N+1)}$$

Nonlinear refractive index

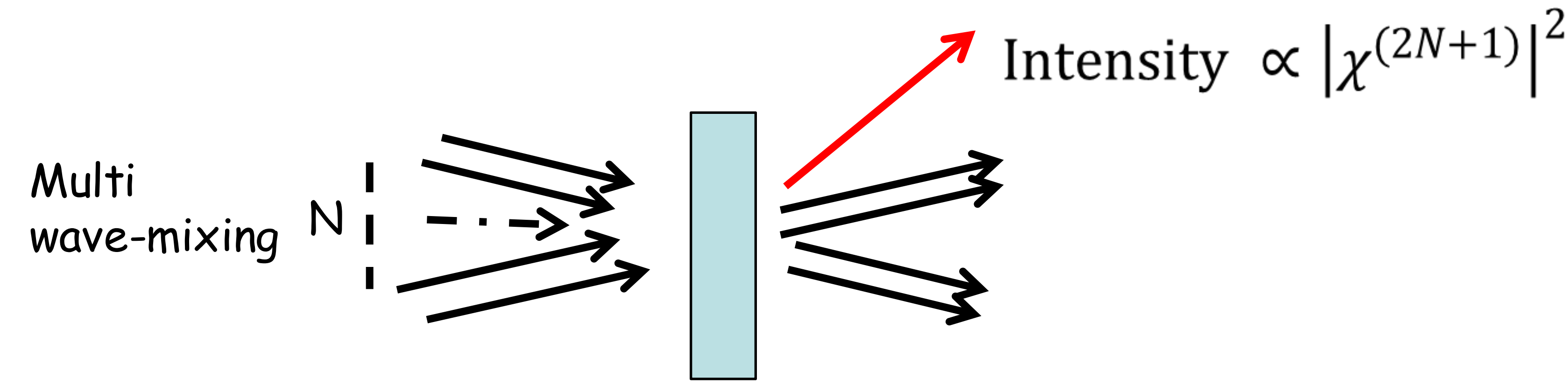
$$\alpha_N \propto Im\, \chi^{(2N+1)}$$

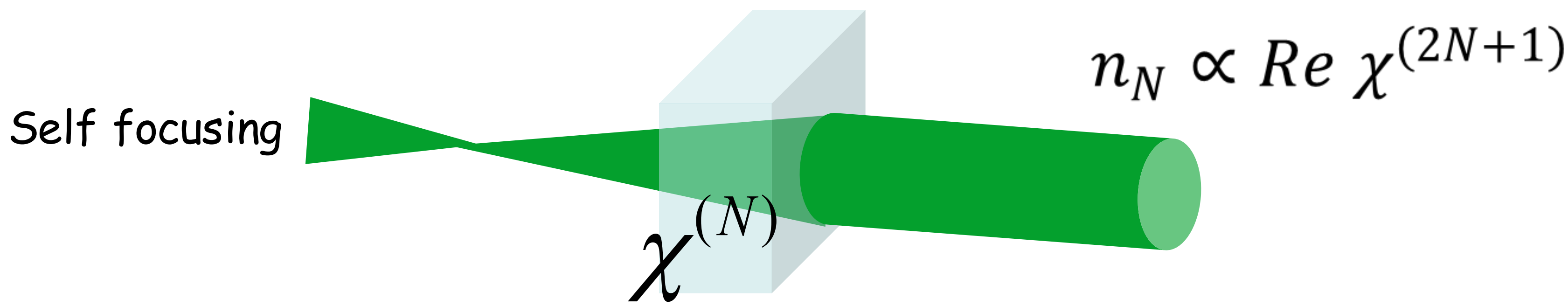
Nonlinear absorption coefficient

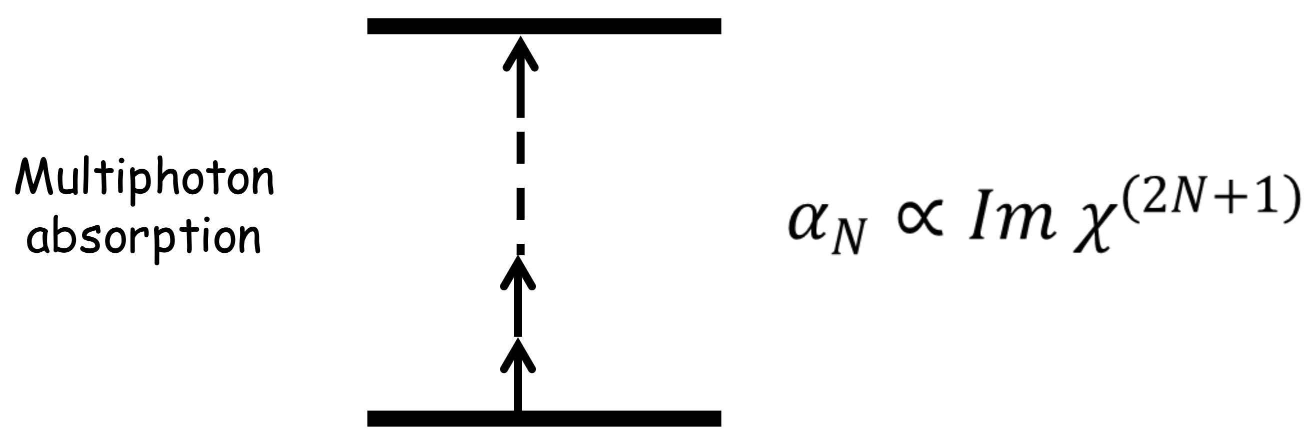
linear + nonlinear

$$n = n_0 + n_2 I + n_4 I^2 + n_6 I^3 + \cdots$$

$$\alpha = \alpha_0 + \alpha_2 I + \alpha_4 I^2 + \alpha_6 I^3 + \cdots$$


Multi
wave-mixing
N
Intensity $\propto |\chi^{(2N+1)}|^2$


Self focusing
$\chi^{(N)}$
$n_N \propto Re\ \chi^{(2N+1)}$


Multiphoton
absorption
$\alpha_N \propto Im\ \chi^{(2N+1)}$

Light propagation inside a NL medium

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

$$\vec{P}_L(\vec{r},t) = \varepsilon_0 \chi_L \vec{E}(\vec{r},t)$$

$$\vec{P}_{NL}(\vec{r},t) = \varepsilon_0 \chi_{NL} \vec{E}(\vec{r},t)$$

$\mathrm{Re}\,\chi^{(2N+1)}$ → NL refractive index

$\mathrm{Im}\,\chi^{(2N+1)}$ → NL absorption coefficient

N = 1, 2, 3...

Solitons

solutions of Maxwell's equation with NL polarization terms

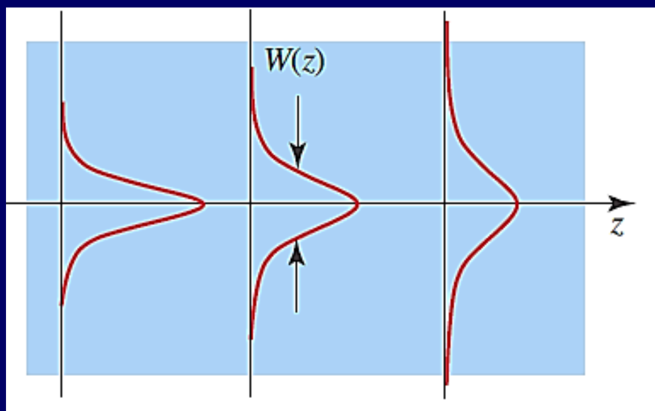
"Nonlinear Schrödinger equation" - NLSE -

Wave with special shape such that it propagates without change their shape.

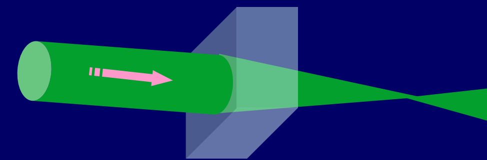
When one soliton interacts with another soliton they do not change their shape but, the phases of the electric fields change.

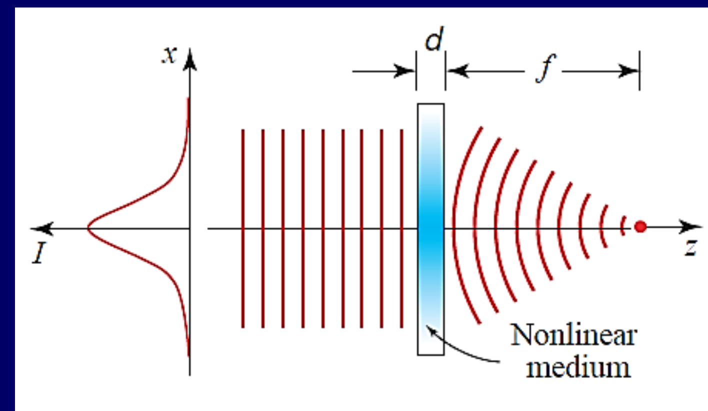
Temporal and spatial solitons

Diffraction

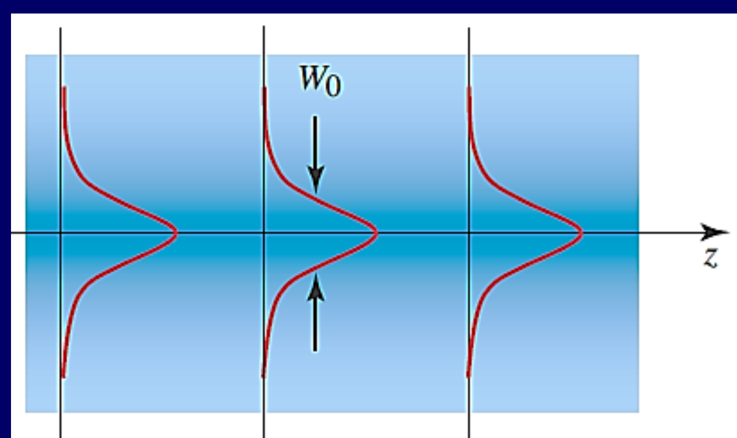


Self-focusing





Bright Spatial Soliton



Nonlinear Schrödinger Equation: (1+1)D

Scalar theory

$$E(x,z,t) = A_m a(x,z) e^{i(k_0 n z - \omega t)}$$

$$\frac{1}{2k_0 n_0}\frac{\partial^2 a}{\partial x^2} + i\frac{\partial a}{\partial z} + \frac{k_0 n_0 n_2}{2\eta_0}|a|^2 a = 0$$

Dispersion x self-focusing

$$\left|\frac{\partial^2 a(x,z)}{\partial z^2}\right| << \left|k_0 \cdot \frac{\partial a(x,z)}{\partial z}\right|$$

$$\eta_0 = \sqrt{\frac{\mu_0}{\varepsilon_0}}$$

$$a(x,z) = \mathrm{sech}(x / X_0)\exp(i\,z / 2L_d)$$

Soliton solution

(1+1)D

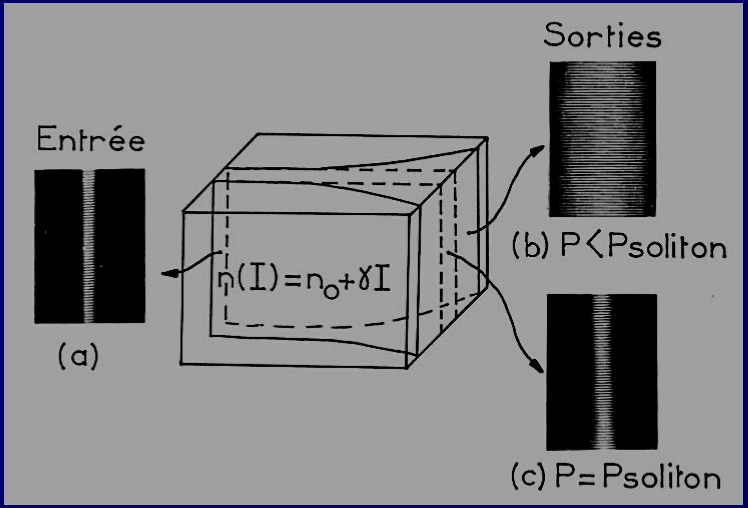
$$L_d = X_0^2 k_0 n_0$$

$$X_0 = \frac{1}{k_0 n_0}\sqrt{\frac{2\eta_0}{|n_2||A_m|}}$$

(1+1)D spatial solitons in CS_2

$n_2 > 0$

Beam focused by cylindrical lenses



Barthélémy, Maneuf, Froehly, Opt. Commun. 55 (1985) 201.

Planar waveguides

Maneuf, Desailly, Froehly, Opt. Commun. 65 (1988) 193.

Picosecond lasers - 532 nm

(2+1)D Soliton

$$E(x,y,z,t) = A\, a(x,y,z)\exp[\mathrm{i}(k_0 n z - \omega t)]$$

NLSE

$$\frac{1}{2k_0 n_0}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)a + i\frac{\partial a}{\partial z} + \frac{k_0 n_0 n_2}{2\eta_0}|a|^2 a = 0$$

(2+1)D soliton is unstable in a pure Kerr medium

Catastrophic self-focusing

High-order nonlinearity

Modified NLS equation

$\chi^{(5)}$

$$\frac{1}{2k_0 n_0}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)a + i\frac{\partial a}{\partial z} + \frac{k_0 n_0 n_2}{2\eta_0}|a|^2 a + \frac{k_0 n_0^2 n_4}{4\eta_0^2}|a|^4 a - \frac{\alpha_3}{2}\left(\frac{n_0}{2\eta_0}\right)^2 |a|^4 a = 0$$

These terms may stabilize the (2+1)D spatial soliton

This problem was known
since the years 60 '
but no homogeneous and isotropic
material was identified such
that the propagation of
Spatial Solitons
would be possible for long distances

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

PRL 110, 013901 (2013) PHYSICAL REVIEW LETTERS week ending 4 JANUARY 2013

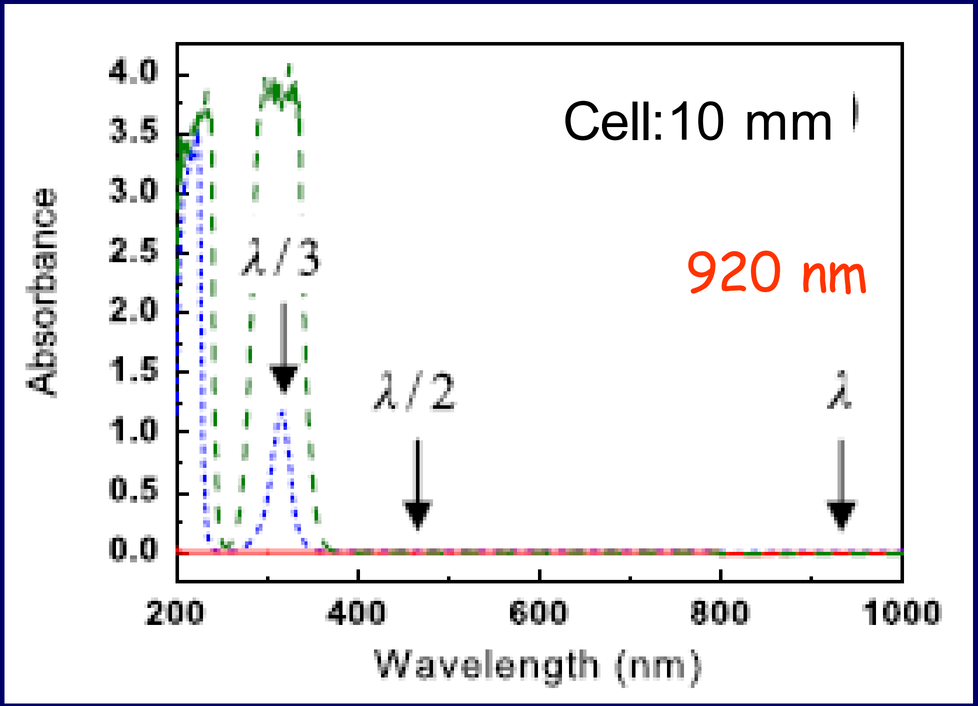
Robust Two-Dimensional Spatial Solitons in Liquid Carbon Disulfide

Edilson L. Falcão-Filho* and Cid B. de Araújo
Departamento de Física, Universidade Federal de Pernambuco, 50670-901 Recife, Pernambuco, Brazil

Georges Boudebs, Hervé Leblond, and Vladimir Skarka
LUNAM Université, Université d'Angers, Laboratoire de Photonique d'Angers, EA 4464, 49045 Angers, France

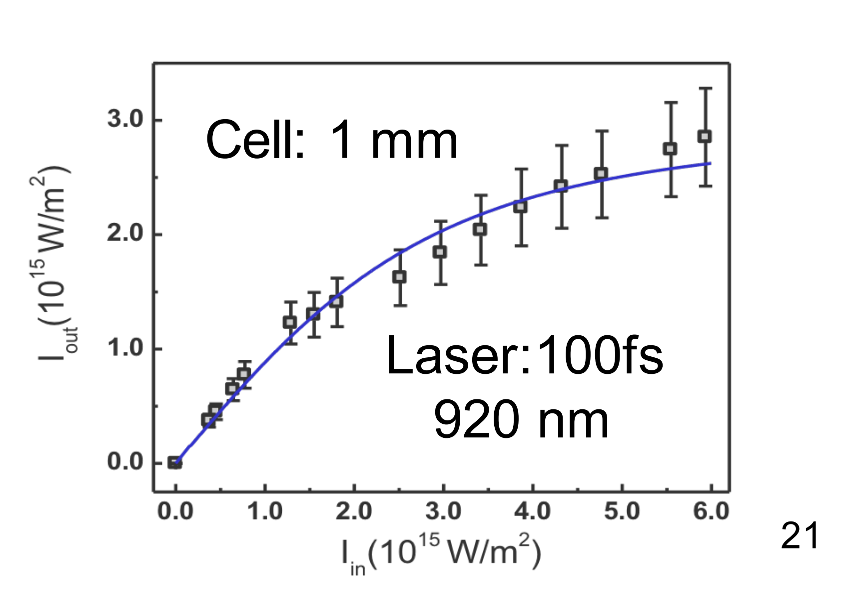
Very important: contributions of third and fifth order of opposite signs

Absorbance spectrum of CS_2 diluted in ethanol



$n_2 > 0 \qquad n_4 < 0$

Three photon absorption of pure CS_2

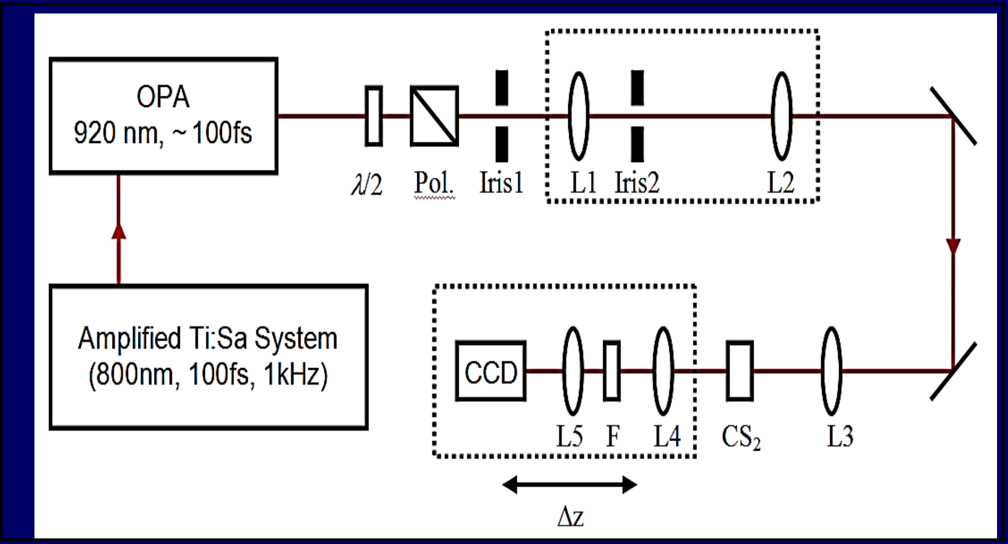


Optical limiting experiment

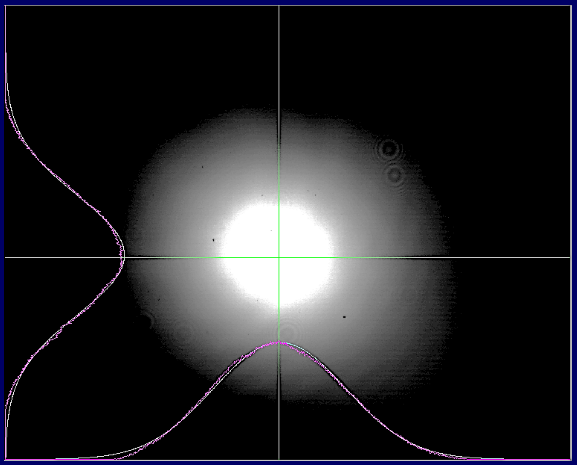
$$I_L = (1-R)^2 \frac{I_0}{\sqrt{1+2I_0^2(1-R)^2\alpha_3 L}}$$

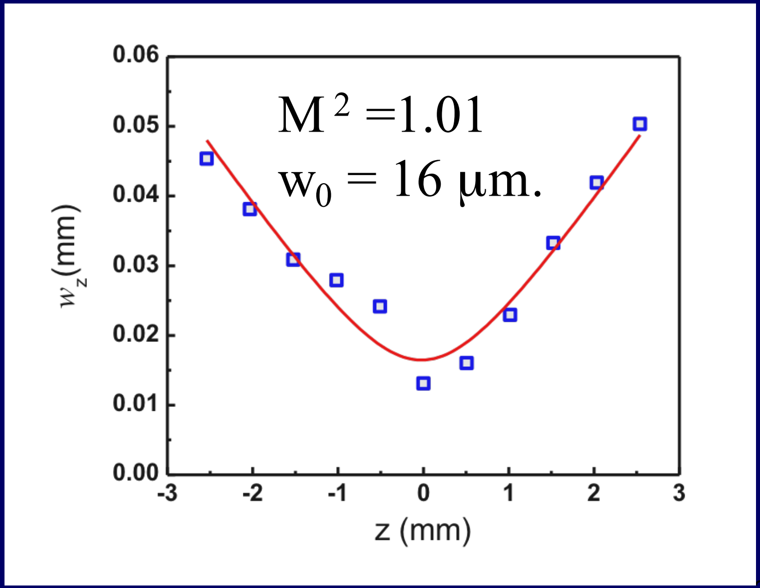
$a_3 = 5.8 \times 10^{-29}\ m^3/W^2$

experimental setup

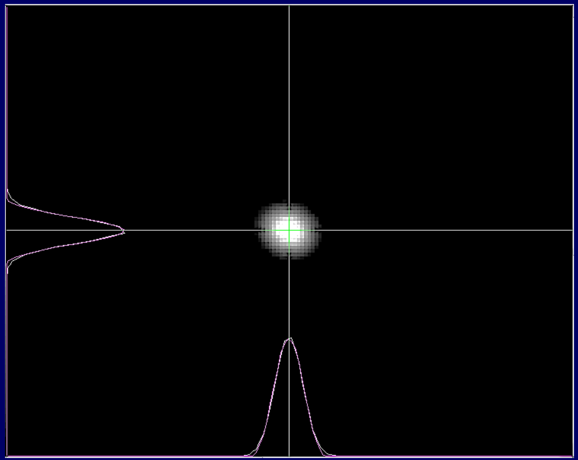


Beam waist on L3: $w = 2.0$ mm



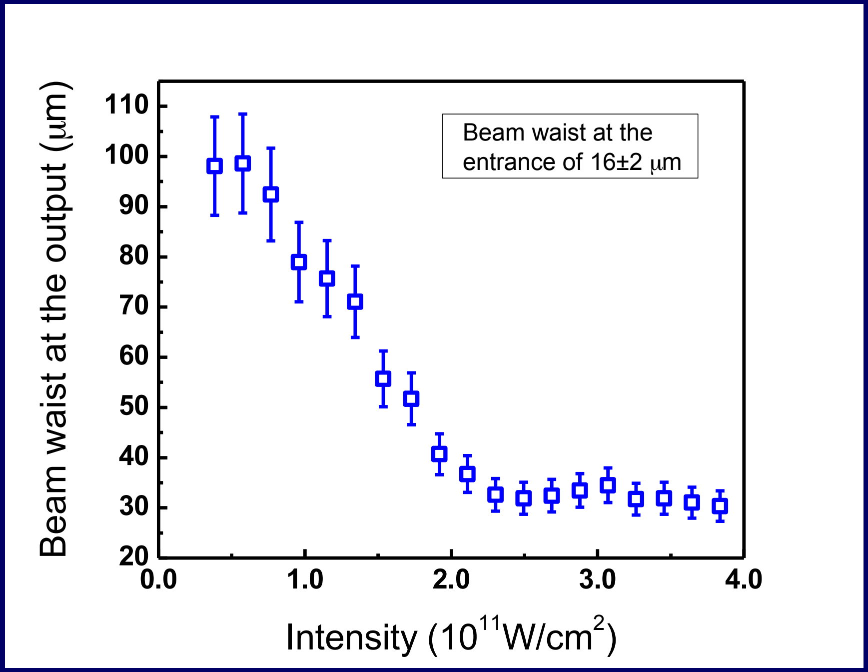


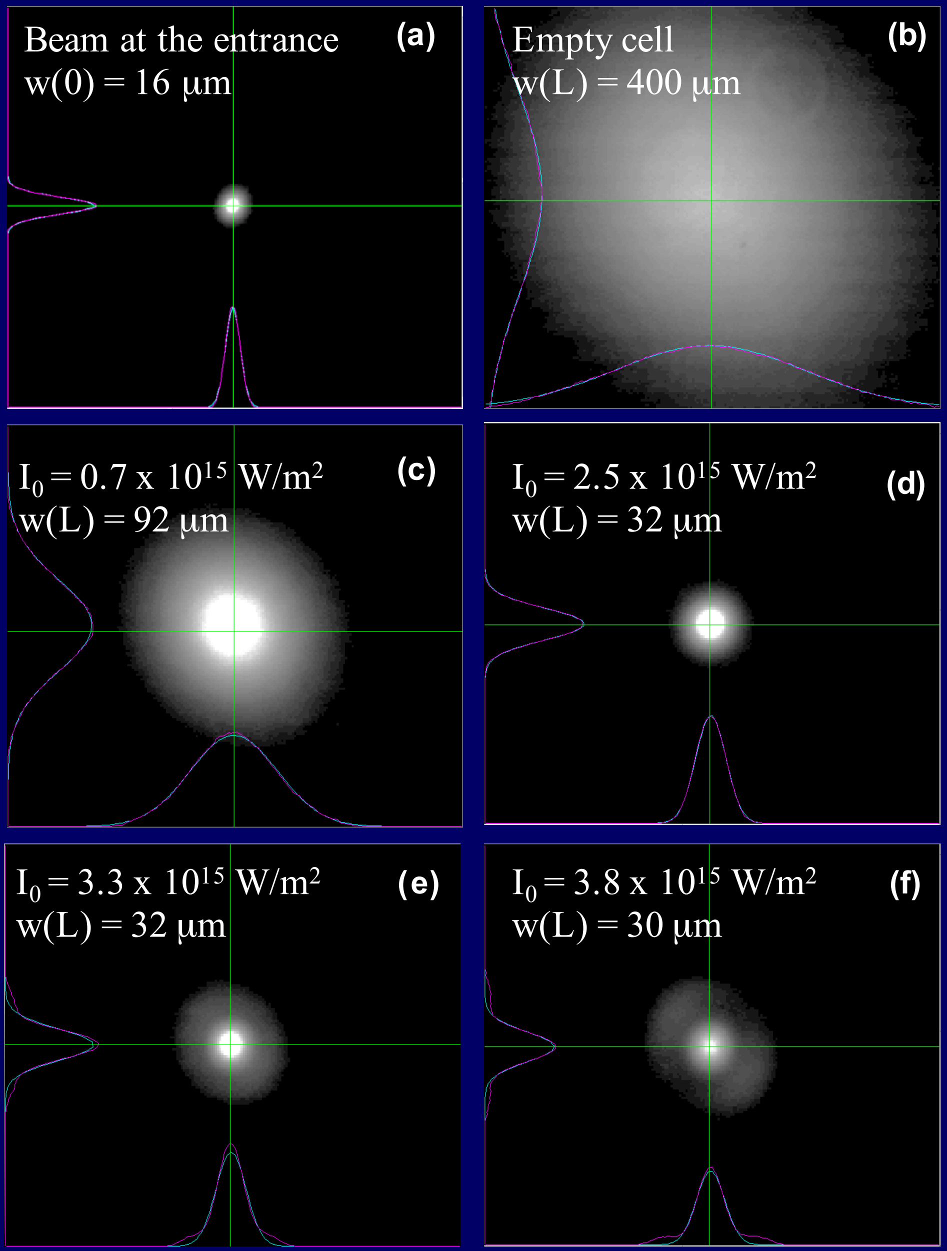
Focal region: $w_0 = 16\ \mu m$



cell length: 1.0 cm

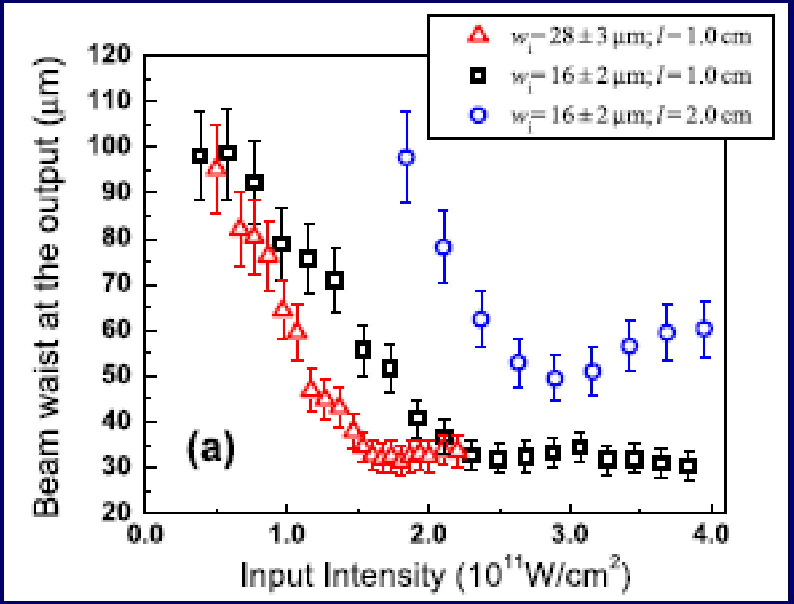
Beam focused on the entrance face



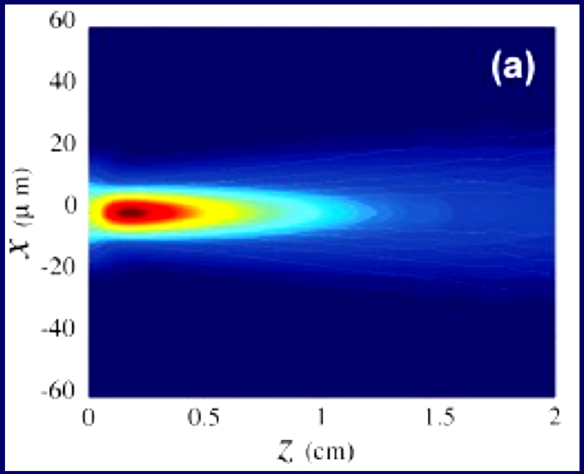


1 cm → $10z_0$

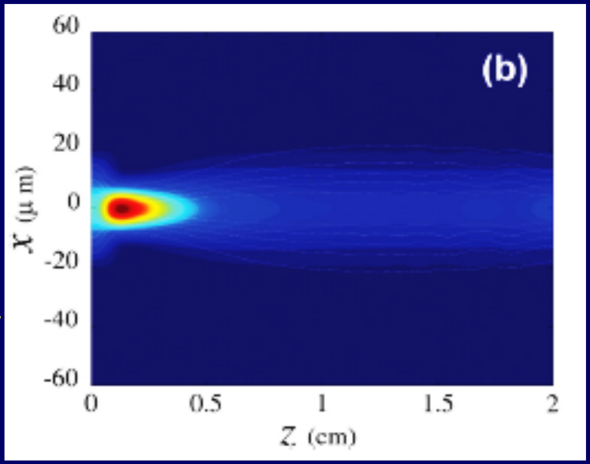
Beam waist versus laser intensity



Cells: 1 and 2 mm



0.8×10^{11} W/cm^2



NLS equation

1.6×10^{11} W/cm^2

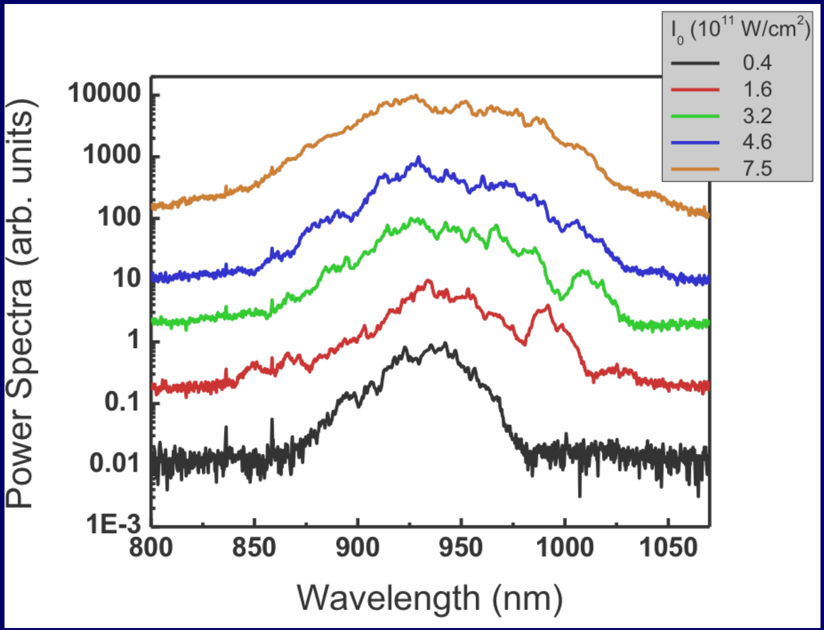
Spectral broadening

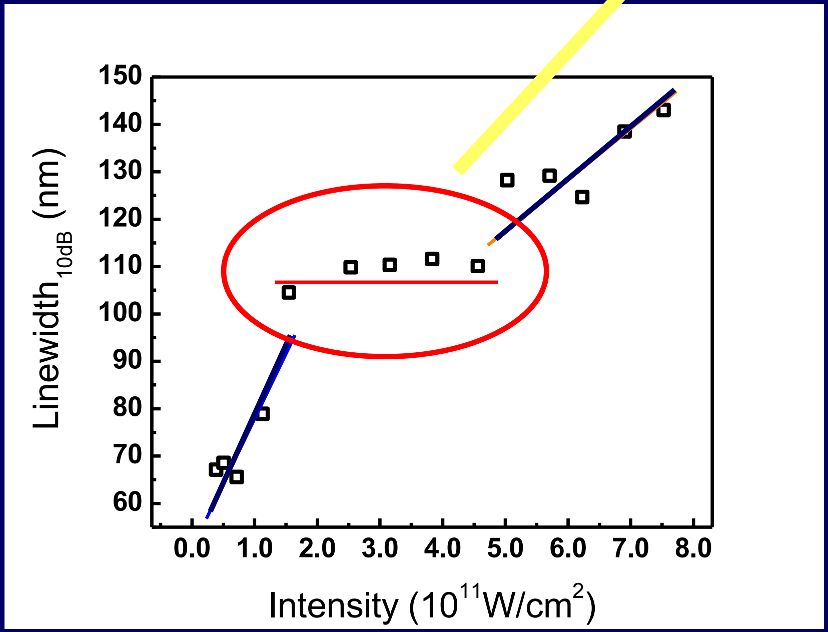
$$\phi(t) = \omega_0 t - \frac{2\pi}{\lambda_0} \cdot n(I) L$$

$$I(t) = I_0 \exp\left(-\frac{t^2}{\tau^2}\right)$$

$$\omega(t) = \frac{d\phi(t)}{dt}$$

Intensity clamping





Summary – bright spatial solitons

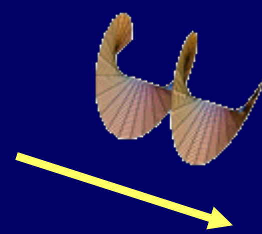
- (2+1)D spatial soliton propagation over more than $10z_0$ in CS_2 due to simultaneous contribution of the third- and the fifth-order susceptibilities.

- Intensity clamping effect which corroborates the soliton stability

- Computer simulations with the NLSE. Results in greement with the experimental data.

Optical vortices

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

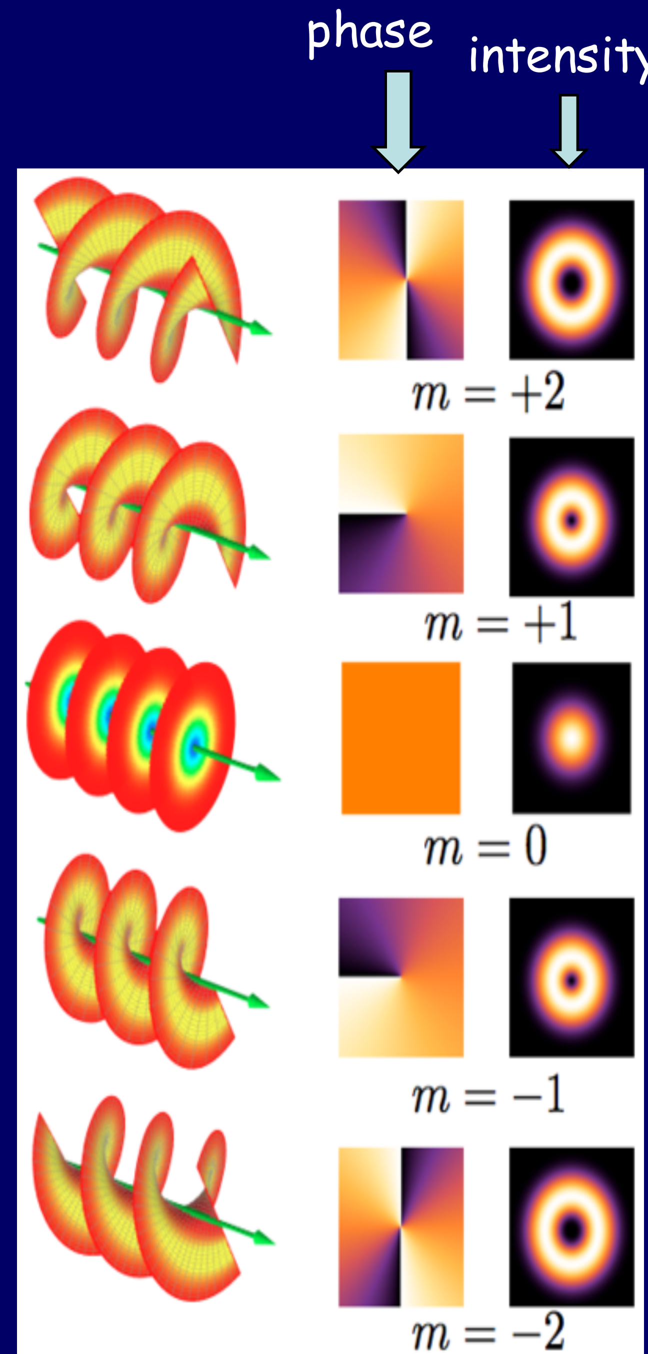


$$\phi(t, z, \theta) = kz + \omega t + m\theta$$

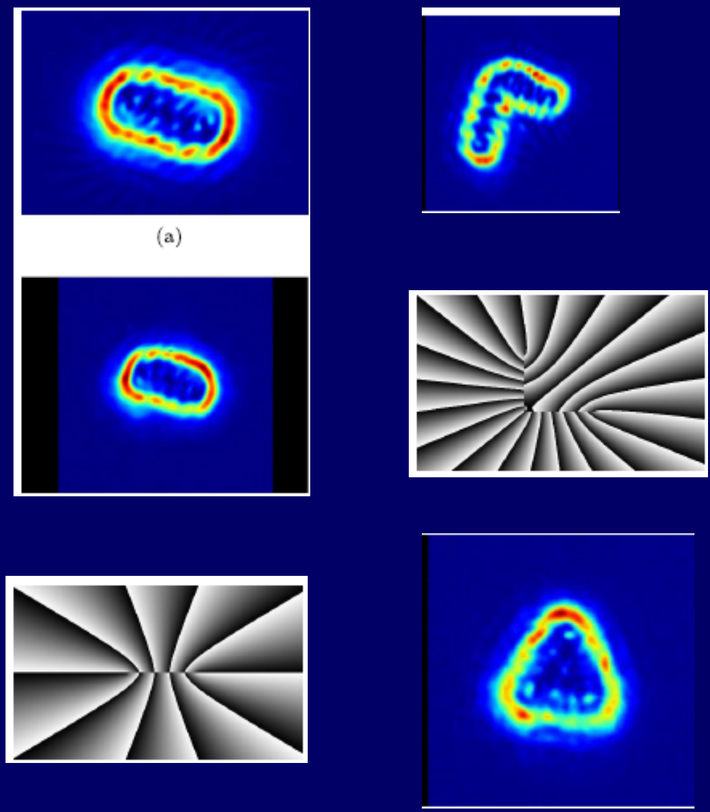
$m = 0$ Plane wave

$m \neq 0$ Wave with topological phase

m is the "topological charge"

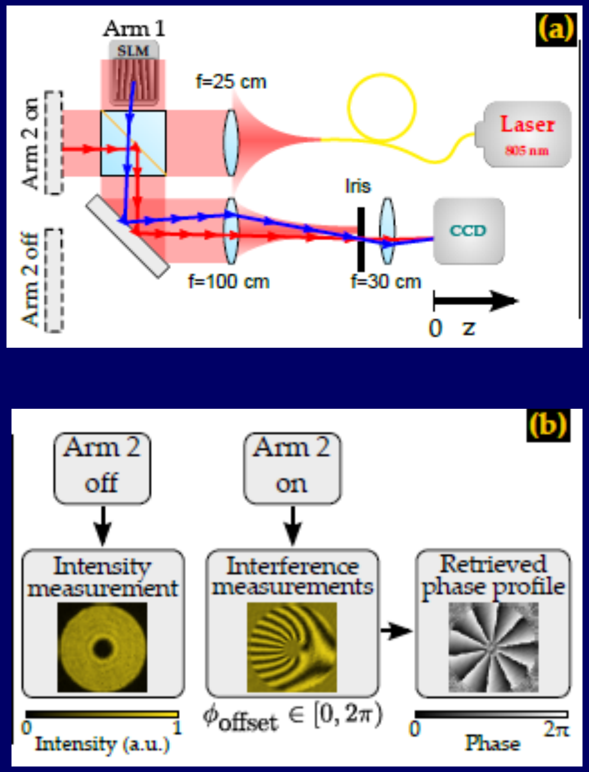


Shaping optical beams with topological charges



Amaral, Falcão-Filho, de Araújo
Opt. Lett. 38 (2013) 1579

Characterization of topological charge and orbital angular momentum of shaped optical vortices



Amaral, Falcão-Filho, de Araújo
Opt. Express 22 (2013) 30315

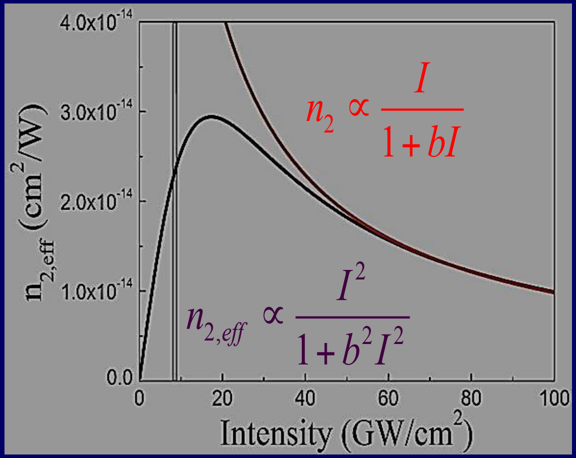
Optical Vortex Soliton - OVS

Stable propagation in a self-defocusing medium
Unstable propagation in self-focusing

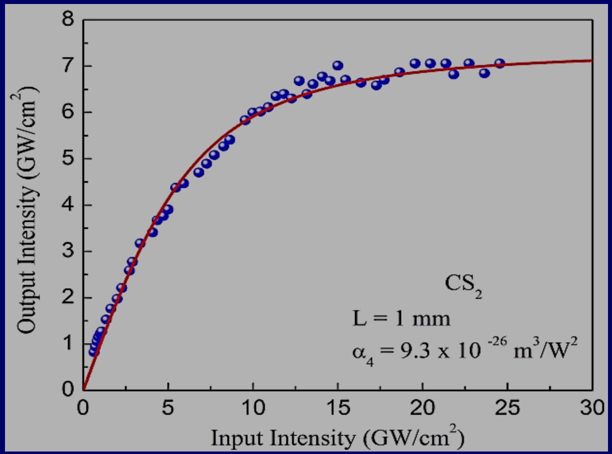
Conditions on the NLSE to observe stable OVS in a self-focusing medium?

Saturable NL refractive index and NL absorption

CS_2
532 nm
psec



Effective NL refractive index



NL transmittance

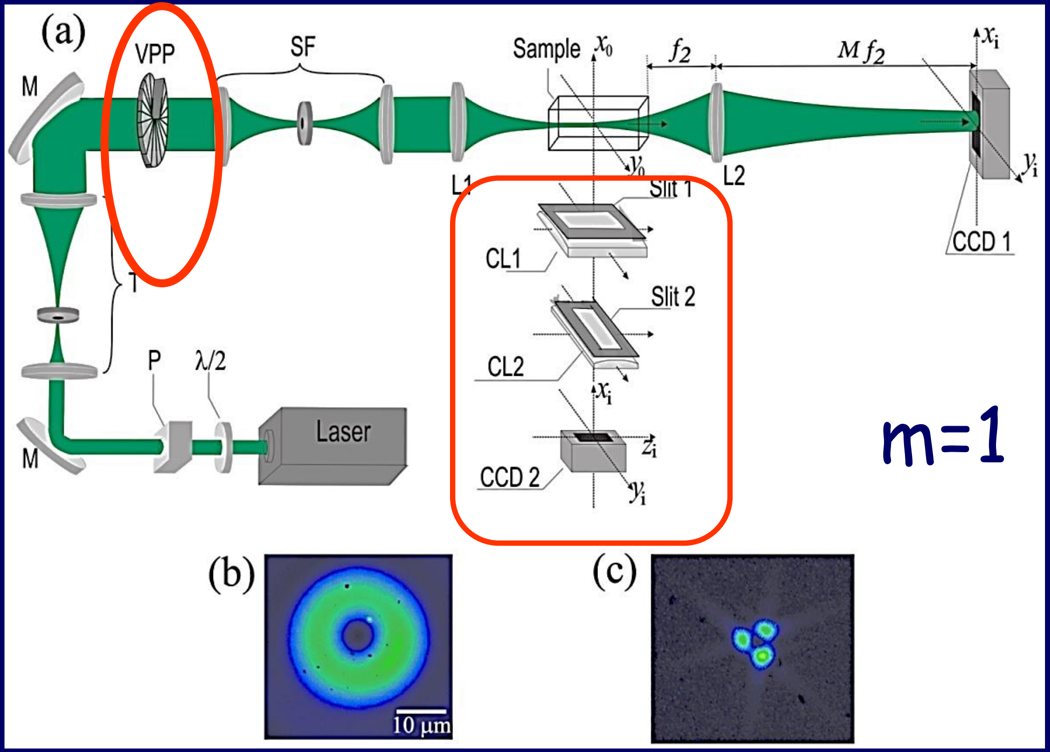
PHYSICAL REVIEW A 93, 013840 (2016)

Robust self-trapping of vortex beams in a saturable optical medium

Albert S. Reyna,[1,*] Georges Boudebs,[2] Boris A. Malomed,[1,†] and Cid B. de Araújo[1]

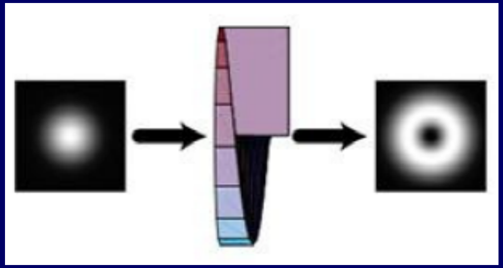
Propagation of OVS in CS_2

532 nm
80 ps
10 Hz



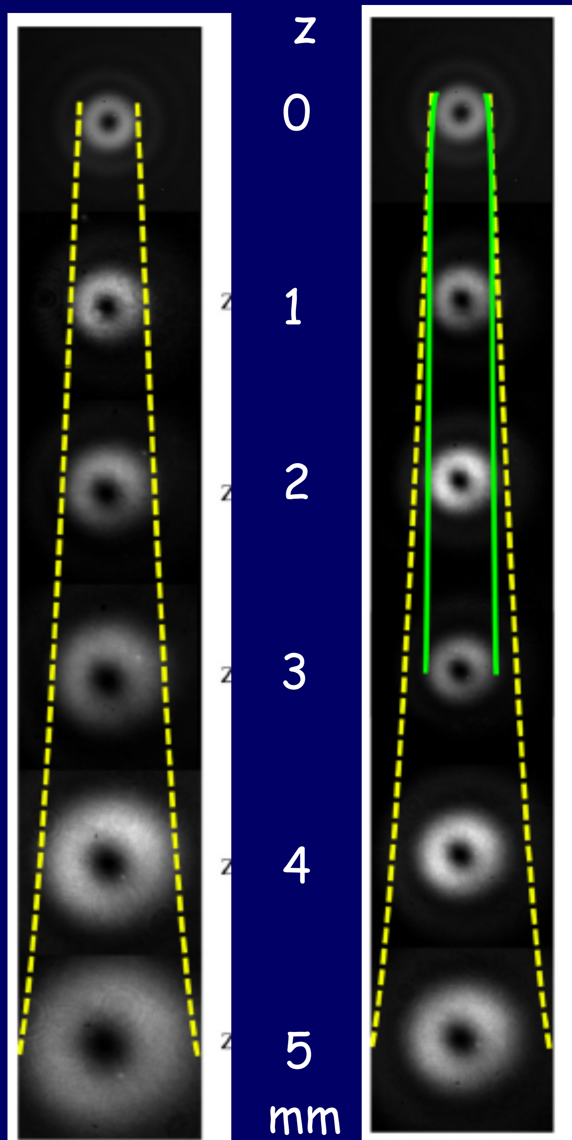
m=1

VPP- vortex phase plate

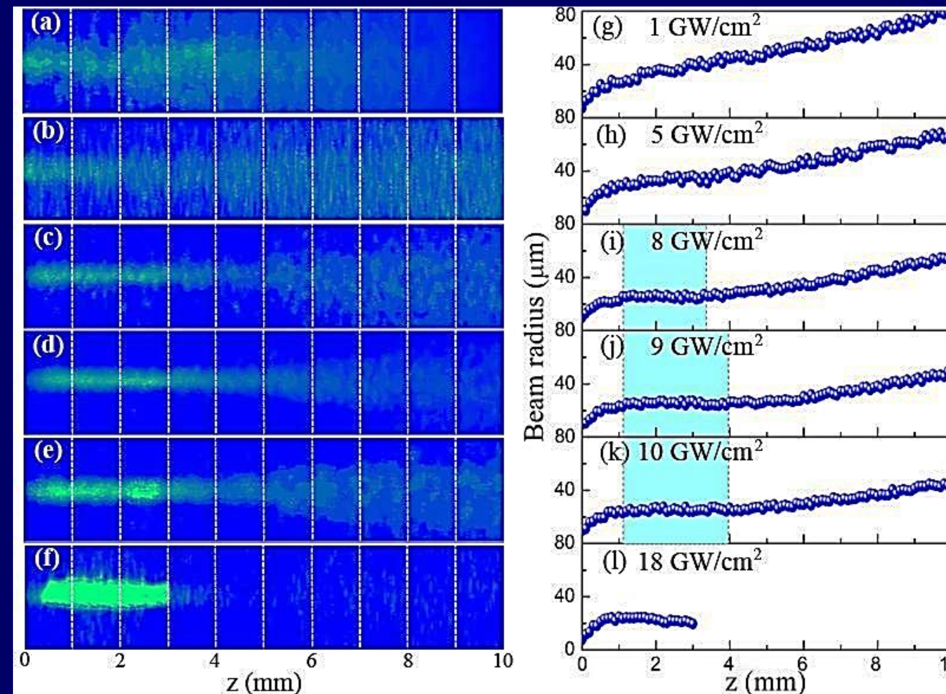


Optical Vortex beam carries an orbital angular momentum of *mħ* per photon

Optical Vortex Solitons in CS_2



1.0 GW/ cm² 9.0 GW/ cm²



$$i\frac{\partial E}{\partial z} = -\frac{1}{2n_0 k}\nabla_\perp E - \left[\frac{k a I^2}{1+b^2 I^2} + i\frac{\gamma I^2}{2}\right]E$$

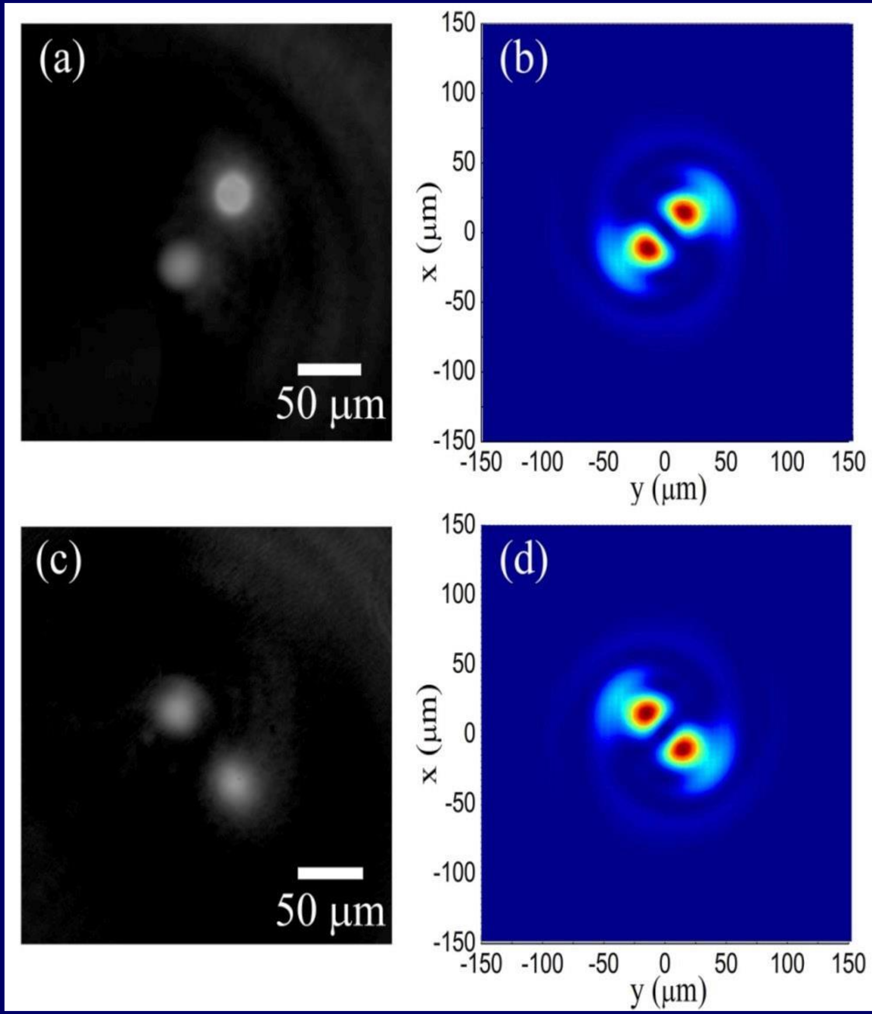
First observation of an OVS in a medium with local nonlinearity

Azimuthal symmetry breaking

$I = 18\,GW/cm^2$ $I = 15\,GW/cm^2$

$m = +1$

$m = -1$



Considering 3PA

Stable OVS

$8\,GW/cm^2 \le I < 10\,GW/cm^2$

splitting

$13\,GW/cm^2 \le I$

No 3PA

$7.4\,GW/cm^2 \le I \le 7.6\,GW/cm^2$

$8\,GW/cm^2 \le I$

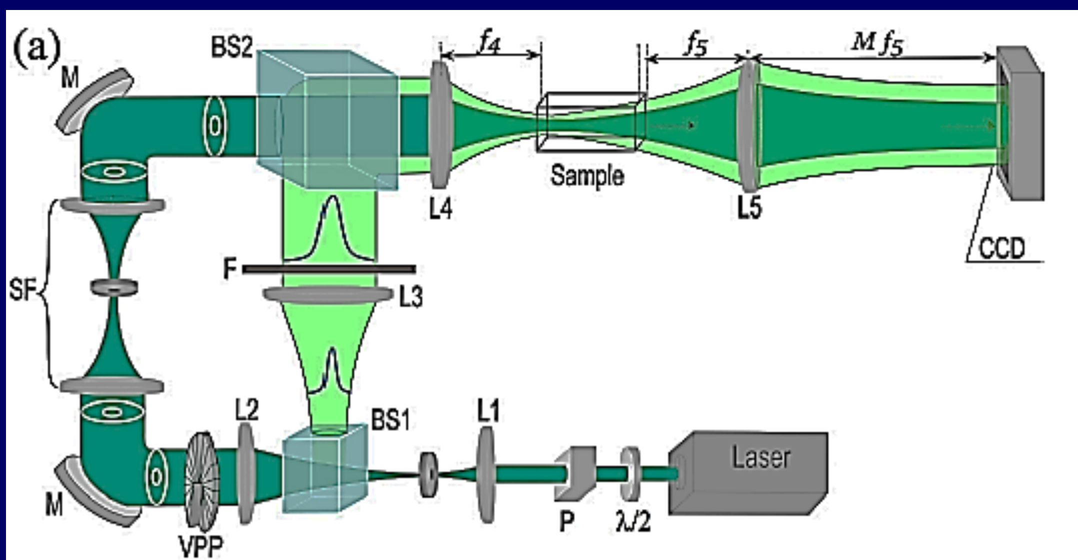
FILM

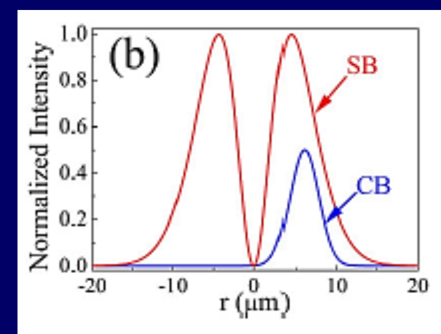
topological charge m=+1 final.mp4

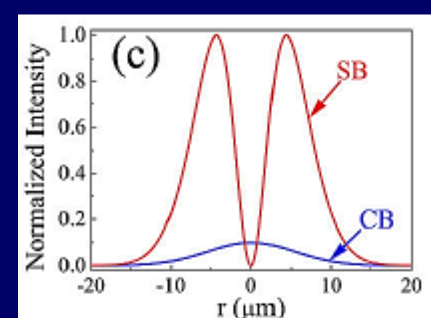
PHYSICAL REVIEW A **93**, 013843 (2016)

Taming the emerging beams after the split of optical vortex solitons in a saturable medium

Albert S. Reyna* and Cid B. de Araújo

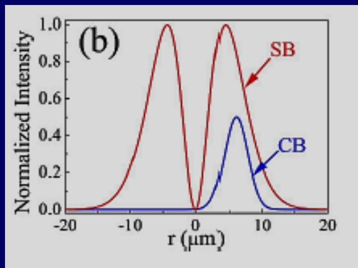


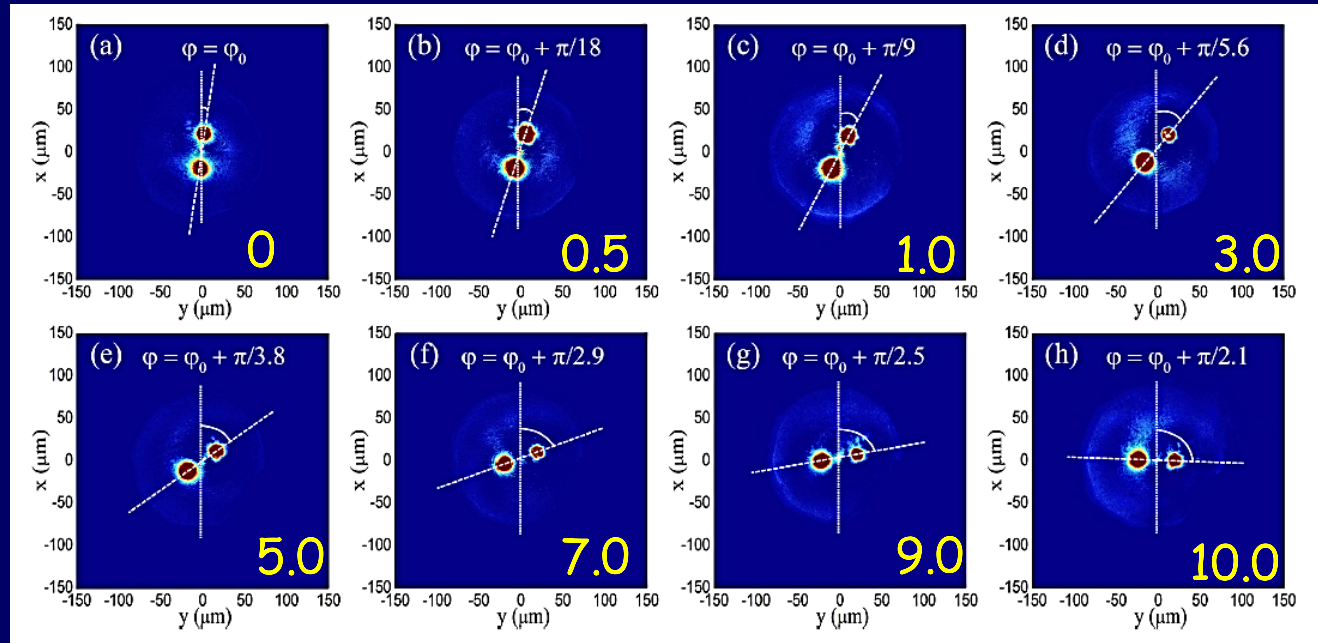




532 nm, 80 ps, 10Hz

cell length: 10 mm

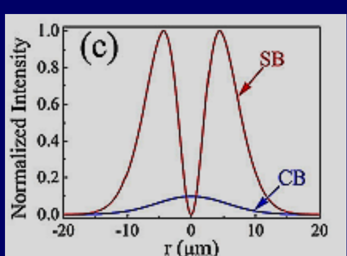


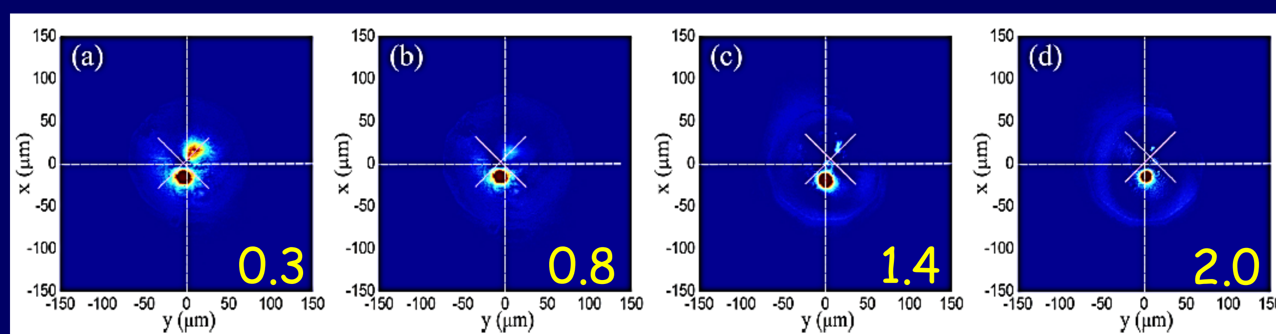


VB intensity
18 GW/cm^2

Control beam
GW/cm^2

Control beam less intense than the signal beam





Control of energy transfer

Exploitation of high-order electronic nonlinearities allows::

- observation of (2+1)D bright spatial solitons in a homogeneous NL medium with electronic nonlinearity.

- observation of stable propagation of optical vortex solitons (OVS) in a medium with saturable refractive index and presenting NL absorption.

- controlling the instability of an OVS by using a control beam with smaller intensity than the OVS.

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Measurements of the third- and fifth-order optical nonlinearities of water at 532 and 1064 nm using the D4σ method

Côme Schnebelin,[1] Christophe Cassagne,[1] Cid B. de Araújo,[2] and Georges Boudebs[1,*]
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Investigations on the nonlinear optical response and losses of toluene at 532 and 1064 nm in the picosecond regime

Georges Boudebs[1] · Hongzhen Wang[1] · Christophe Cassagne[1] · Hervé Leblond Cid B. de Araújo

IOP Publishing Reports on Progress in Physics
Rep. Prog. Phys. **79** (2016) 036401 (30pp) doi:10.1088/0034-4885/79/3/036401

Techniques for nonlinear optical characterization of materials: a review

Cid B de Araújo[1], Anderson S L Gomes[1] and Georges Boudebs[2]

Thank you for your attention

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FACEPE CAPES CNPq