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

Nonlinear Optics

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Recife

Founded in 1535 2 milions of inhabitants

Optics in Recife

Nonlinear spectroscopy of condensed matter systems

Random lasers

Nano optics: Optical microcavities

High-harmonic generation

Ultrafast phenomena

Metrology of optical frequencies

Cold atoms: coherent effects

Quantum information and entanglement

Laser dynamics

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PD U. California 1982

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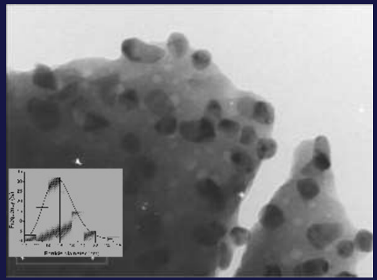
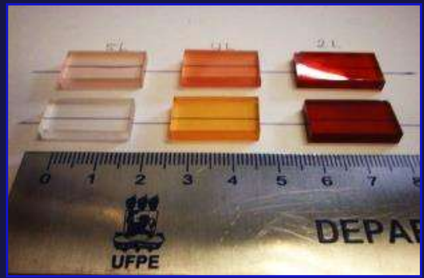
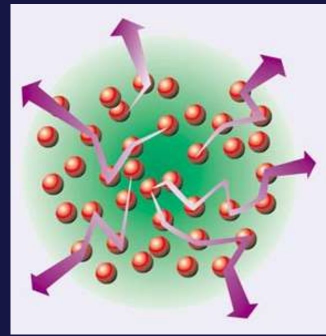
PD UFPE 2019-2022

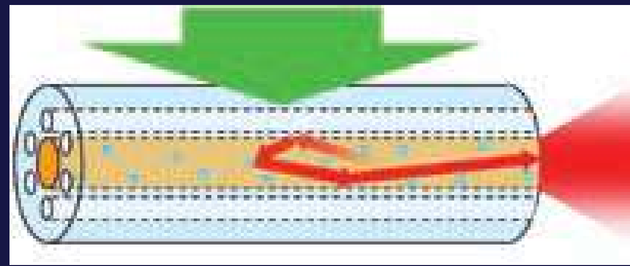
Plasmonics in random media



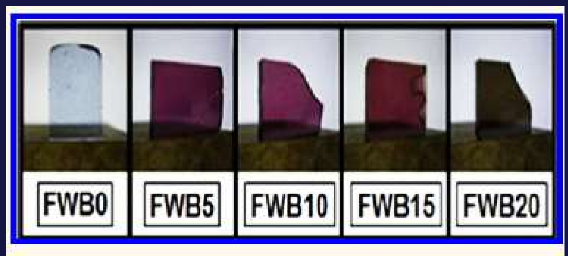


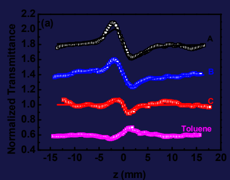
Random lasers



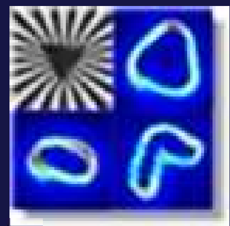


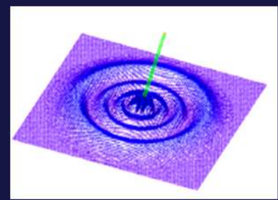
NLO LAB

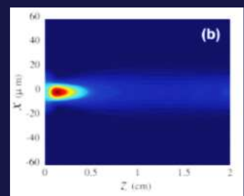




Transition metal oxides
Organic materials
(nanoparticles and films)







NL transverse effects (phase modulation, vortices, solitons...)

J. A. Swieca School 2023

1st. lecture

Introduction to nonlinear optics

2nd lecture

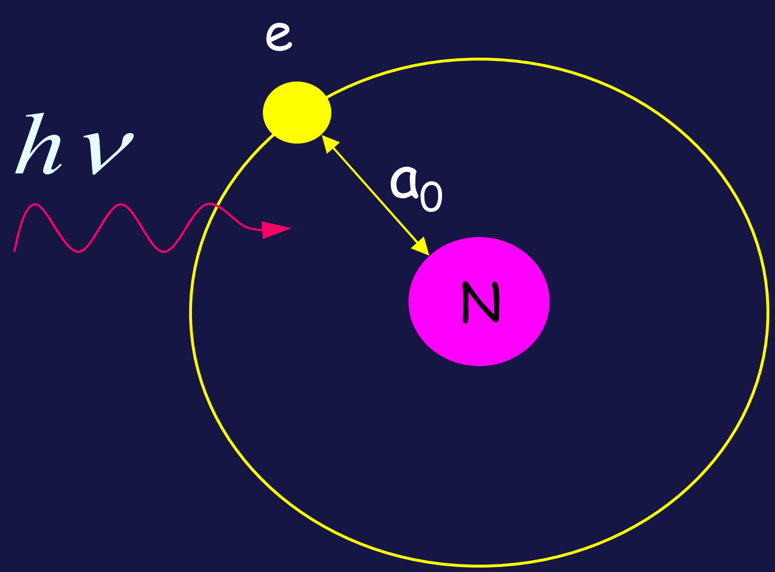
Nonlinear Photonics in Plasmonic media

3rd lecture

Stimulated Emission and Random Lasers

How does optical nonlinearity appear ?

The strength of the electric field of the light wave should be in the range of atomic fields



$$E_{at} = e / a_0^2$$

$$a_0 = \hbar^2 / me^2$$

$$E_{at} \approx 2 \times 10^{-7}\,\text{esu}$$

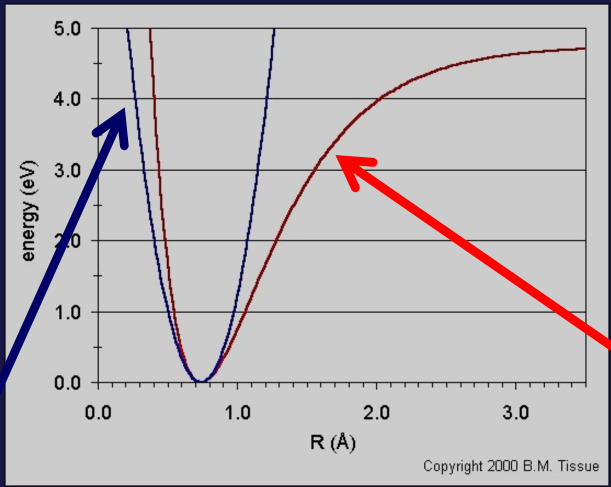
$3 \; x \; 10^8$ V/cm

Atom as an oscillator

Electron potential energy

Small optical intensity

$$\boxed{\omega_0 = \sqrt{\frac{K}{m_0}}}$$



Harmonic oscillator

Large optical intensity

Anharmonic oscillator

$$U(x) = \frac{1}{2}m_0\omega_0^2x^2 + \frac{1}{3}m_0K_ax^3 + \frac{1}{4}\,m_0K_bx^4 + \cdots$$

Restoring Force

$$F(x) = -\frac{dU}{dx} = -m_0\omega_0^2x - m_0K_ax^2 - \,m_0K_bx^3 - \cdots$$

Considering only the second order nonlinearity

$$m_0 \frac{d^2x}{dt^2} + m_0\gamma \frac{dx}{dt} + m_0\omega_0^2 x + m_0 K_a x^2 = -eE(t)$$

$$E(t) = E_0 \cos\omega t = \frac{1}{2} E_0 \left(e^{i\omega t} + e^{-i\omega t}\right)$$

Tentative solution

$$x(t) = \frac{1}{2}\left(X_1 e^{i\omega t} + X_2 e^{i2\omega t} + \text{c.c.}\right) \qquad X_1 \gg X_2$$

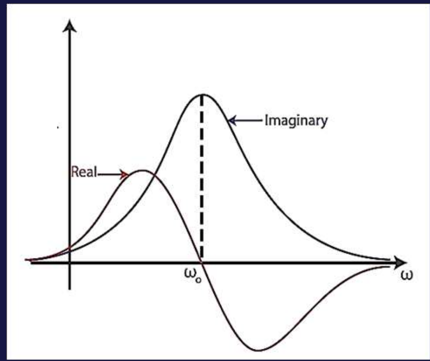
$$X_1 = -\frac{eE_0}{m_0} \frac{1}{(\omega_0^2 - \omega^2) + i\gamma\omega}$$

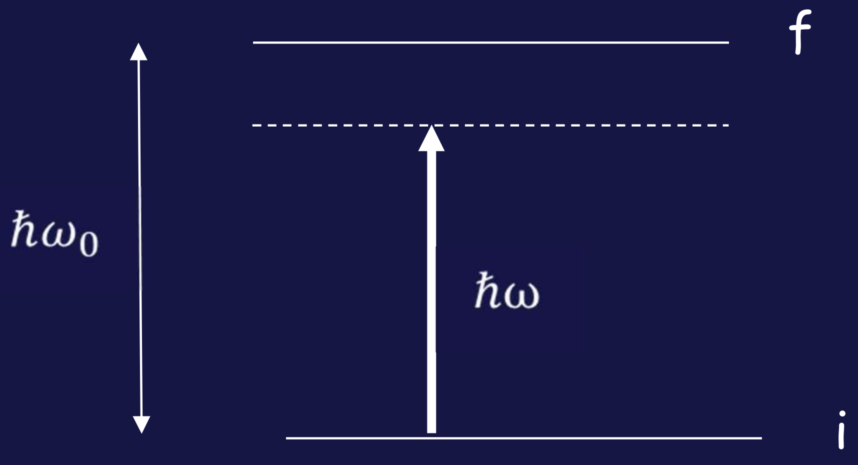
Induced polarization

$$P(\omega, t) = -Nex(\omega, t) = -\frac{Ne}{2}\left(X_1 e^{i\omega t} + \text{c.c.}\right) = \epsilon_0 \chi^{(1)}(\omega) E(t)$$

$$\chi^{(1)}(\omega) = \frac{N e^2}{m_0 \epsilon_0 [(\omega_0^2 - \omega^2) + i\gamma\omega]}$$

$$\chi^{(1)}(\omega,\omega) = \frac{Ne^2}{m\epsilon_0[(\omega_0^2 - \omega^2) + i\gamma\omega]}$$





Representation based on the quantum description

$$n_0 \propto Re\, \chi^{(1)}(\omega,\omega)$$

Linear refrative index

$$\alpha_0 \propto Im\, \chi^{(1)}(\omega,\omega)$$

Linear absorption coefficient

By considering the next term in the perturbative solution

$$P(2\omega,t) = -Nex(2\omega,t) = -\frac{Ne}{2}\left(X_2 e^{i2\omega t} + \text{c.c.}\right) = \epsilon_0 \chi^{(2)}(2\omega) E^2(t)$$

$$\chi^{(2)}(2\omega) = \frac{Ne^3 K}{\epsilon_0 m^2 [(\omega_0^2 - \omega^2) + i\gamma\omega]^2 (\omega_0^2 - 4\omega^2 + i2\omega\gamma)}$$

Second Harmonic Generation

$$P^{(2)} = \epsilon_0\, \chi^{(2)}(2\omega) \left[E \cos \omega t\right]^2$$

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

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

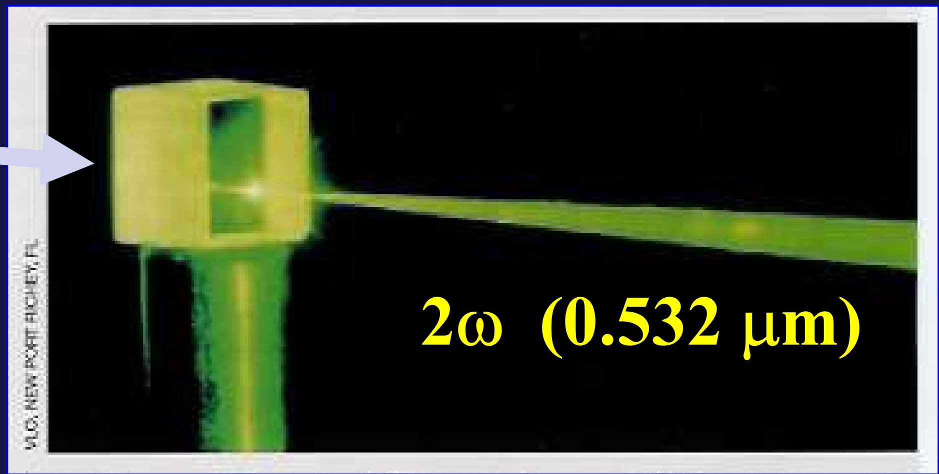
GENERATION OF OPTICAL HARMONICS*

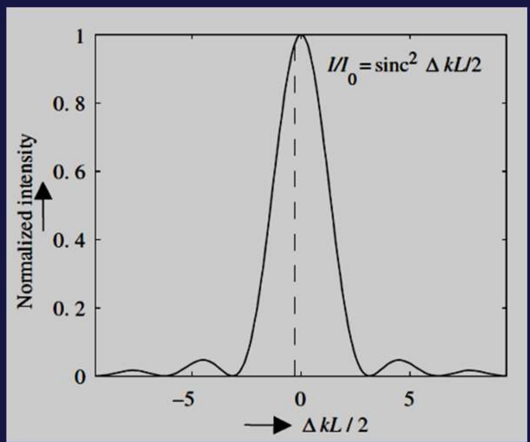
P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

Transparent crystal

Infra-red

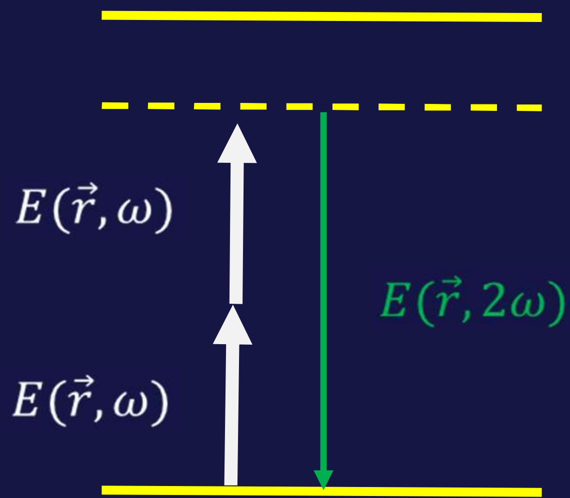
ω (1.064 μm)





Phase matching:

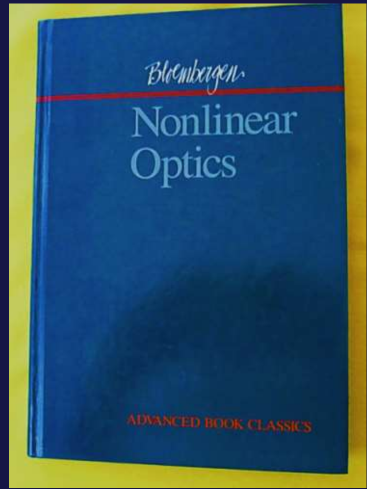
$$k(2\omega) = 2k(\omega)$$



Classical and Quantum formalisms (1962-1964)

N. Bloembergen
(1920-2017)

1981
Physics Nobel Prize



Induced polarization

Nonlinear susceptibilities

$$\vec{P} = \epsilon_0 \{ \overleftrightarrow{\chi}^{(1)} \cdot \vec{E} + \overleftrightarrow{\chi}^{(2)} : \vec{E}\vec{E} + \overleftrightarrow{\chi}^{(3)} \vdots \vec{E}\vec{E}\vec{E} + \cdots \}$$

$\vec{P}$ induces changes in the speed of light in the medium and new frequencies may be generated

Meaning of each term

Polarization vector

$$\overrightarrow{P^{(1)}} = \epsilon_0 \overleftrightarrow{\chi^{(1)}} \cdot \vec{E}$$

$$\overrightarrow{P^{(2)}} = \epsilon_0 \overleftrightarrow{\chi^{(2)}} : \vec{E}\vec{E}$$

$$\overrightarrow{P^{(3)}} = \epsilon_0 \overleftrightarrow{\chi^{(3)}} \vdots \vec{E}\vec{E}\vec{E}$$

Vector components

$$P_i^{(1)} = \epsilon_0 \chi_{ij}^{(1)} E_j$$

$$P_i^{(2)} = \epsilon_0 \chi_{ijk}^{(2)} E_j E_k$$

$$P_i^{(3)} = \epsilon_0 \chi_{ijkl}^{(3)} E_j E_k E_l$$

Analogously for the other terms...

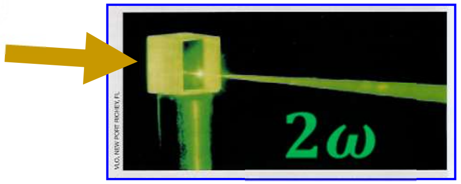
The linear "superposition principle" is not valid

$\chi^{(2)}$ 2nd. order polarization — Three Wave- Mixing

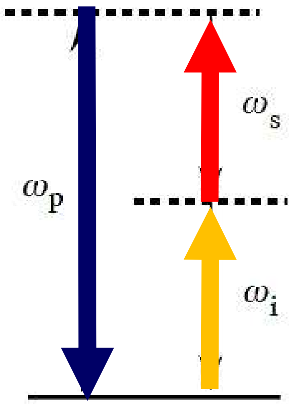
$$P^{(2)} = \varepsilon_0 \chi^{(2)} (E_1 \cos \omega_1 t + E_2 \cos \omega_2 t)^2 =$$

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

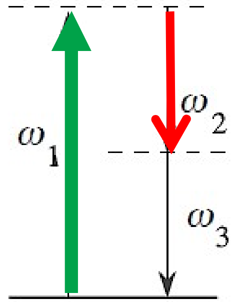
SHG



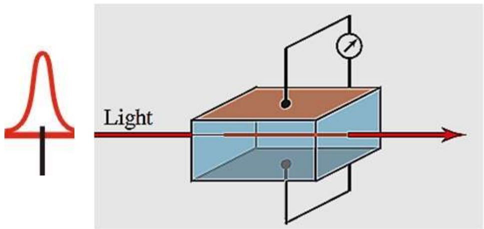
SFG



DFG

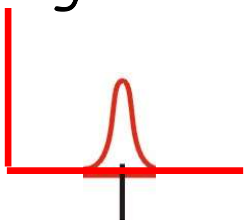


Optical retification



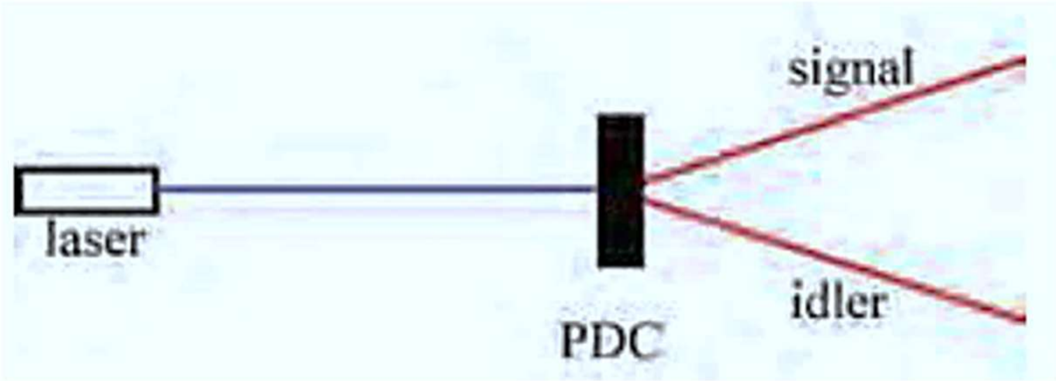
Light pulse

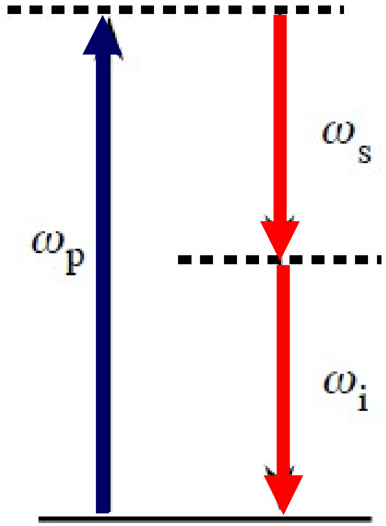
Voltage



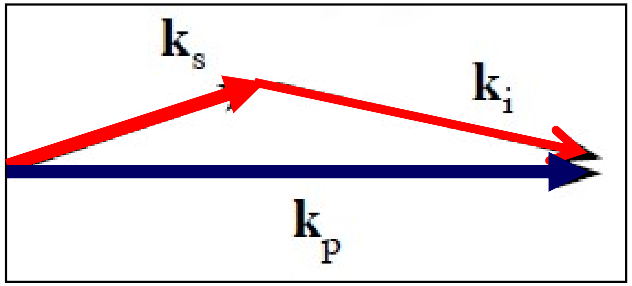
time

$\chi^{(2)}$ Parametric down-conversion





Phase - matching



Signal and idler photons are entangled in polarization, time, energy, position, transverse momentum, angular position and orbital angular momentum

Entanglement is important for fundamental tests of QM and quantum technologies.

Third order nonlinearity

Medium with inversion symmetry $\quad \chi^{(n)} = 0$, n = 2,4, ...

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

$$P^{(1)}(t) + P^{(3)}(t) + \cdots = \epsilon_0 \{ \chi^{(1)} E(t) + \chi^{(3)} E^3(t) + \ \ldots \}$$

$$cos^3 \omega t = \frac{1}{4} \cos 3\omega t + \frac{3}{4} \cos \omega t$$

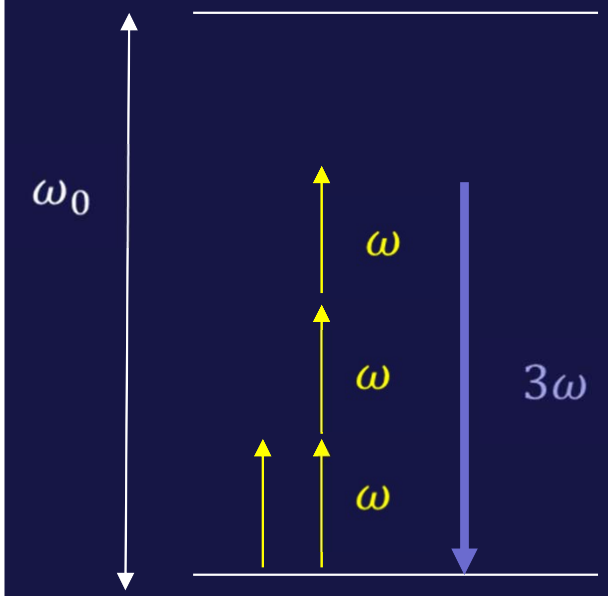
$$P(t) = P^{(1)}(t) + P^{(3)}(t)$$

$$= \epsilon_0 \chi^{(1)} E cos\ \omega t + \frac{1}{4} \epsilon_0 \chi^{(3)} E^3 \cos 3\ \omega t + \frac{3}{4} \epsilon_0 \chi^{(3)} E^3\ cos\ \omega t$$

$$P(t) = \epsilon_0 \left\{ \chi^{(1)} + \frac{3}{4} \chi^{(3)} E^2 \right\} E cos\ \omega t + \frac{1}{4} \epsilon_0 \chi^{(3)} E^3 \cos 3\ \omega t$$

$$P(t) = \epsilon_0 \left\{\chi^{(1)} + \frac{3}{4}\chi^{(3)}E^2\right\} E\cos\omega t + \frac{1}{4}\epsilon_0\chi^{(3)}E^3\cos 3\,\omega t$$

This polarization generates the third harmonic



NL refractive index

$$\boxed{n \propto Re\left\{\chi^{(1)} + \frac{3}{4}\chi^{(3)}E^2\right\}}$$

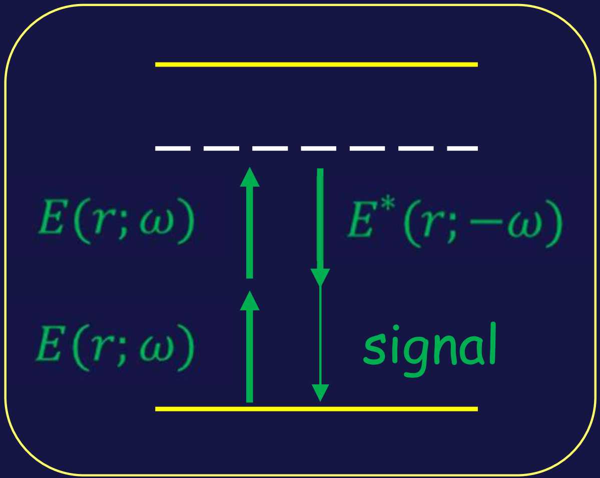
$$n = n_0 + n_2 I \qquad n_2 = \frac{3\,\mathrm{Re}\,\chi^{(3)}}{2n_0^2\epsilon_0 c}$$

$$P^{(3)}(\omega) = \epsilon_0 \{\chi^{(3)}\left(E\, e^{+i\omega t} + c.c.\right)^3$$

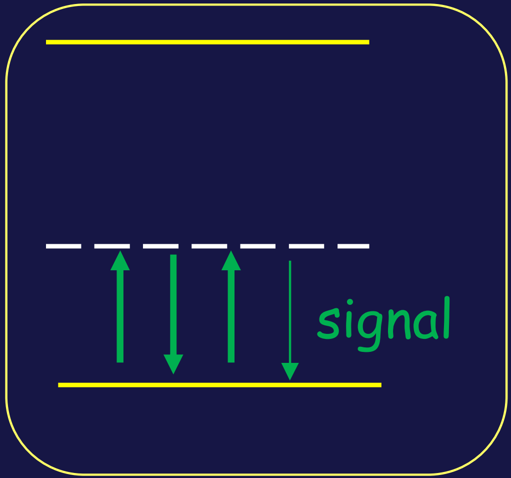
$$P^{(3)}(\omega)e^{-i\omega t} = \epsilon_0\chi^{(3)}(\omega,\omega,\omega,-\omega)E(\omega)E(\omega)E^*(-\omega)\, e^{-i\omega t}$$

Degenerate wave-mixing

Transparent medium

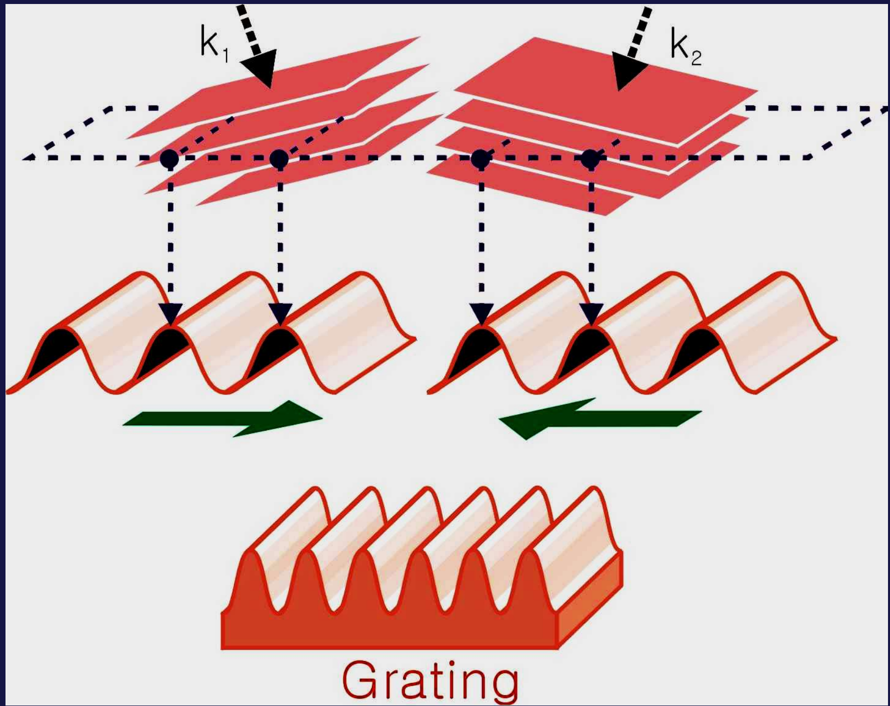


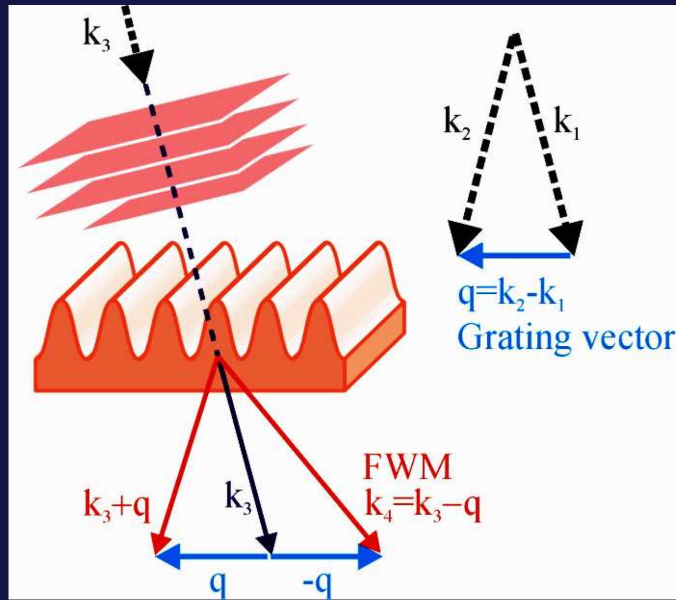
$\chi^{(3)}(\omega,\omega,\omega,-\omega)$



$\chi^{(3)}(\omega,\omega,-\omega,\omega)$

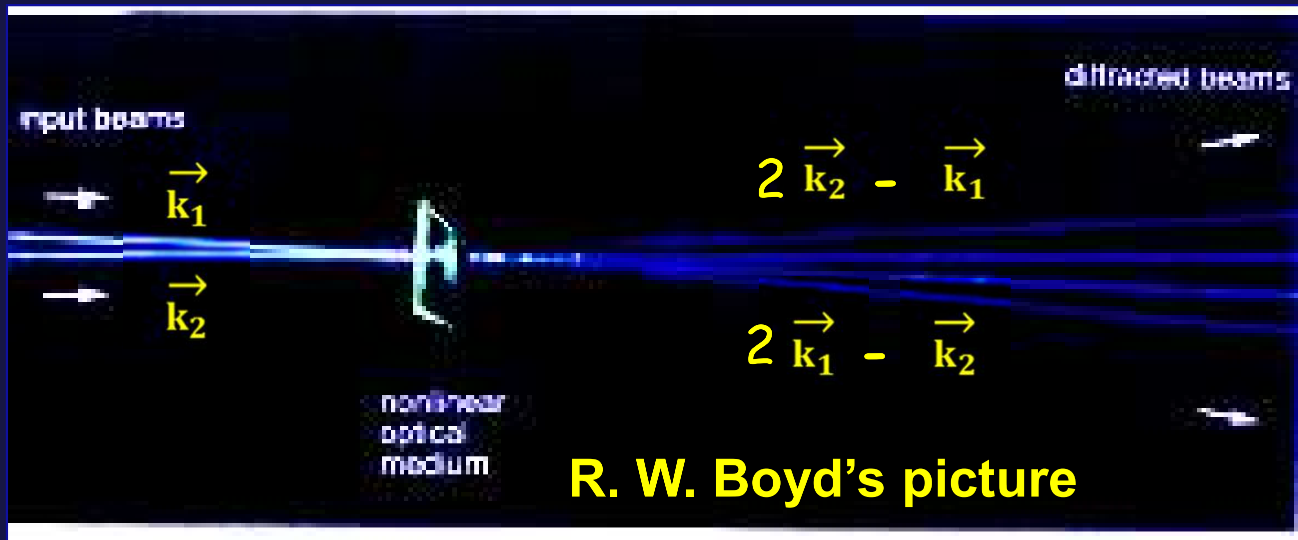
$\chi^{(3)}$: degenerate four-wave-mixing





$$P^{(3)} = \epsilon_0 \chi^{(3)} E^3$$

Only one optical frequency



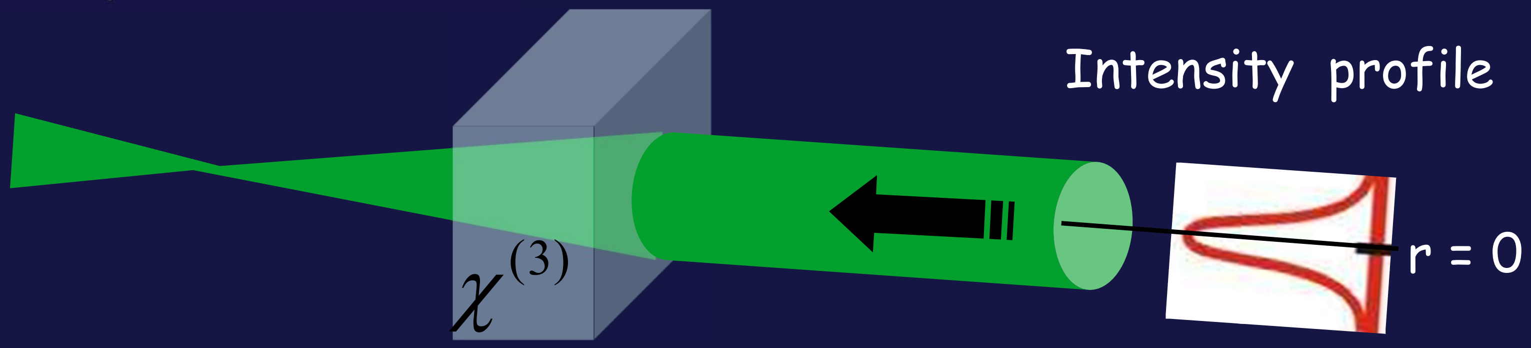
$\chi^{(3)}$: self - focusing

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

A laser beam with Gaussian intensity profile will induce a Gaussian refractive index profile inside the NL sample.

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

$n_2 > 0$



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

Sample behaves as a convergent lens

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

Mechanism	n_2 (cm^2/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}

Electronic polarization

$$\Delta n\,(r=0) = n_2 I_0 \qquad I_0 = 1\,GW/cm^2$$

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

$$\boxed{P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t)}$$

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t}$$

ω_1, ω_2, ω_3, $3\omega_1$, $3\omega_2$, $3\omega_3$, $(\omega_1 - \omega_2 + \omega_3)$, $(-\omega_1 + \omega_2 + \omega_{3,})$, $(2\omega_1 \pm \omega_2)$, $(2\omega_1 \pm \omega_3)$, $(2\omega_2 \pm \omega_1)$, $(2\omega_2 \pm \omega_3)$, $(2\omega_3 \pm \omega_{1,})$, $(2\omega_3 \pm \omega_2)$

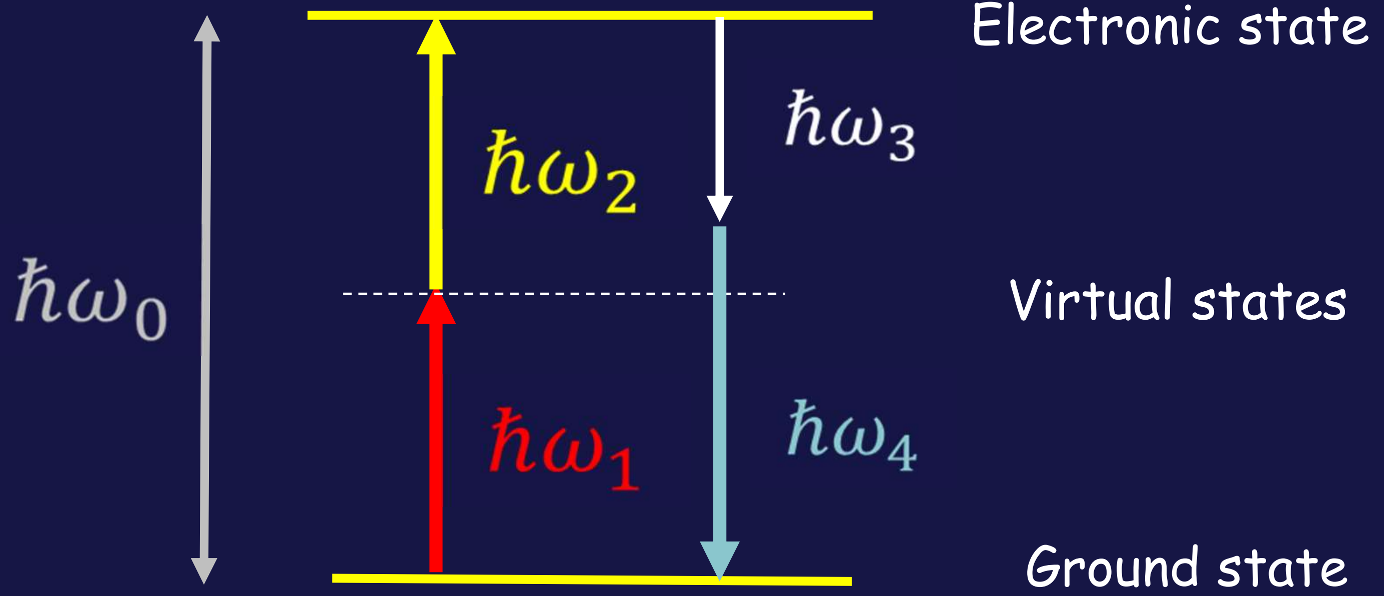
$$P^{(3)}(\omega_4 = 3\omega_1) = \epsilon_0 \chi^{(3)}(3\omega_1, \omega_1, \omega_1, \omega_1) E_1^3$$

$$P^{(3)}(\omega_4 = \omega_1 + \omega_2 - \omega_3) = 6\epsilon_0 \chi^{(3)}(\omega_4, \omega_1, \omega_2, -\omega_3)\, E_1 E_2 E_3^*$$

$$P^{(3)}(t) = \sum_n P^{(3)}(\omega_n) e^{i\omega_n t}$$

Two-photon resonant four wave-mixing

$$P(\omega_4 = \omega_1 + \omega_2 - \omega_3) = 6\epsilon_0 \chi^{(3)}(\omega_4, \omega_1, \omega_2, -\omega_3)\, E_1 E_2 E_3^*$$

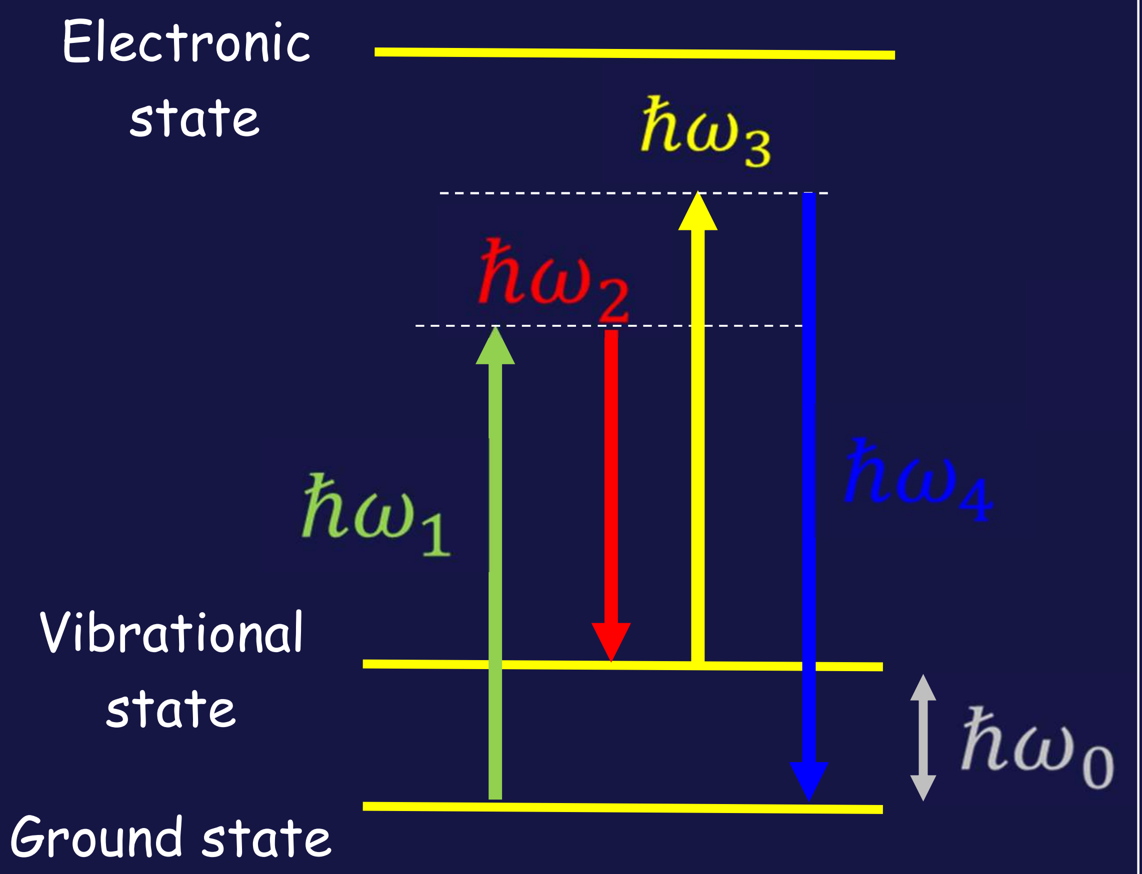


Excited state with same parity than the ground state

2PA cross section

Four wave-mixing with Raman resonance CARS

$$P(\omega_4 = \omega_1 - \omega_2 + \omega_3) = 6\epsilon_0 \chi^{(3)}(\omega_4, \omega_1, -\omega_2, \omega_3)\, E_1 E_2^* E_3$$



$$\boxed{\hbar\omega_1 - \hbar\omega_2 = \hbar\omega_0}$$

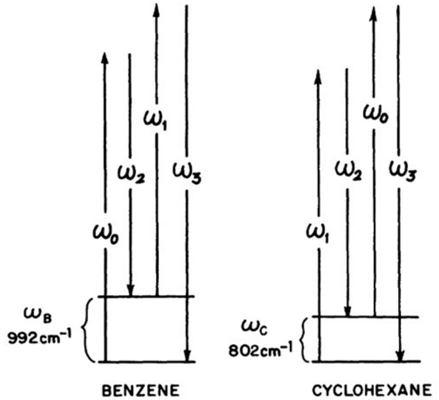
Interaction with non-resonant background

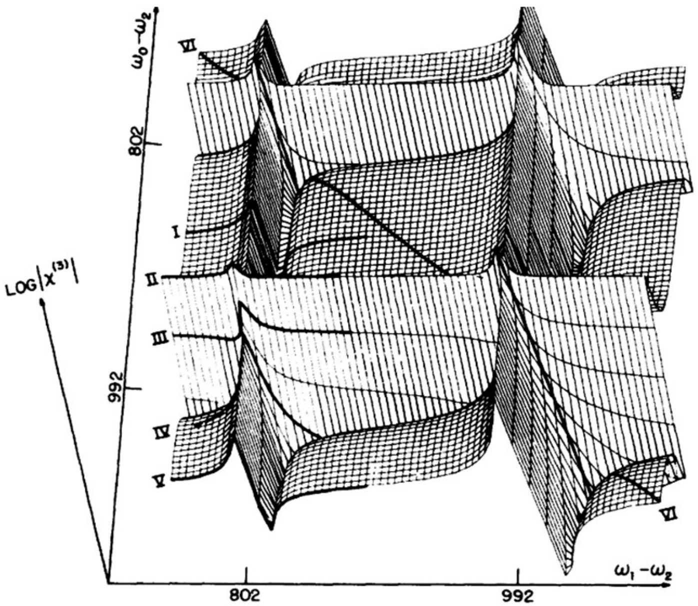
$$P_{NL}^{(3)} = P_{NR}^{(3)} + P_{RES}^{(3)}$$

Y. R. Shen
The Principles of NLO
Wiley, 1984.

M. D. Levenson
Introduction to NL spectroscopy
Academic, 1982

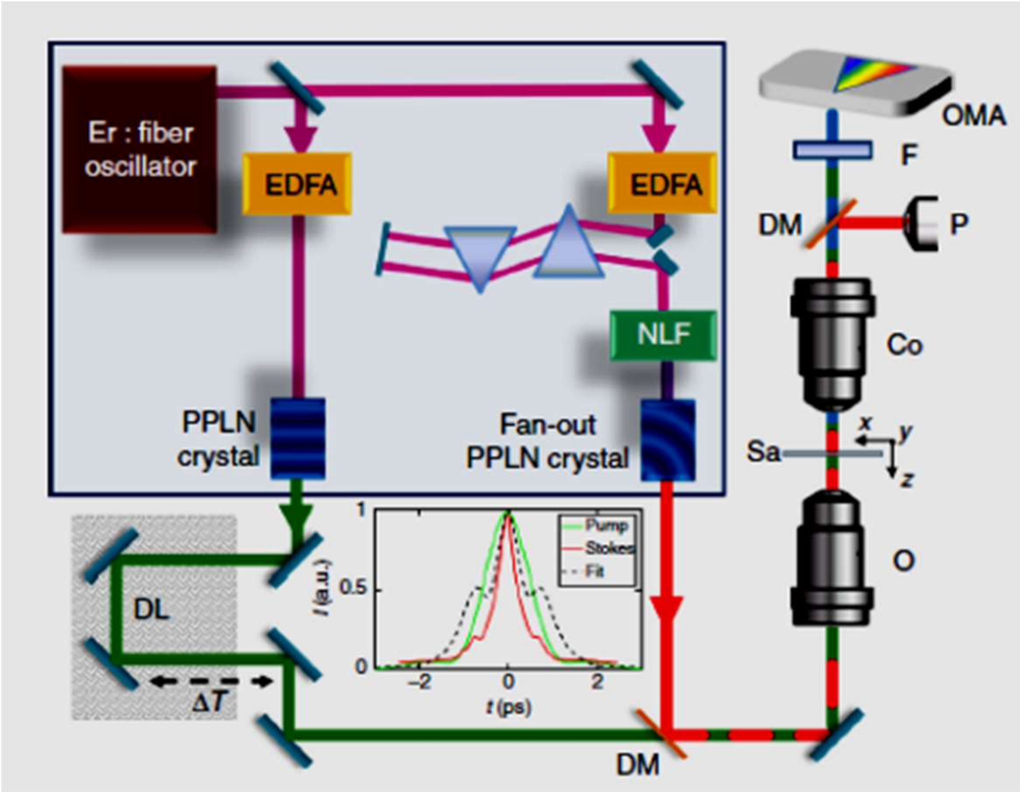
Lotem, Lynch, Bloembergen
Phys. Rev. A 14 (1976) 1748





A. Virga, ….G. Cerullo… et al
Coherent anti-Stokes Raman spectroscopy of single and multi-layer graphene
Nature Commun. 10 (2019) 3658

$$P_{NL}^{(3)} = P_{NR}^{(3)} + P_{RES}^{(3)}$$



Optical susceptibilities depend on the geometrical arrangements of atoms

Susceptibility tensors ⇔ structural symmetry

$$\chi_{ij}^{(1)}(\omega;\ \omega)$$

9 elements

Isotropic media: scalar

$$\chi_{ijk}^{(2)}(\omega;\ \omega_1, \omega_2)$$

27 elements

All elements are identically null in systems with inversion symmetry

$$\chi_{ijkl}^{(3)}(\omega;\ \omega_1, \omega_2, \omega_2)$$

81 elements

In isotropic media: 21 nonzero elements (only 3 are independent)

Higher order susceptibilities: more elements

Quantum approach

$$\langle P \rangle = NTr(\rho\mu) \qquad \frac{\partial \rho}{\partial t} = -\frac{i}{\hbar}[H,\rho]$$

$$H = H_o + H_{\text{int}} + H_{random}$$

$$H_{\text{int}} = -\vec{\mu}\cdot\vec{\mathrm{E}} \cancel{- \frac{e}{2}\overleftrightarrow{Q}\,\nabla\vec{E} + \vec{M}\cdot\vec{H}} = \sum_i H_{\text{int}}(\omega_i)$$

Electric dipole coupling

Electric quadrupole coupling

Magnetic dipole coupling

$$H_{\text{int}} \propto \mathrm{E}_i \exp(-i\omega_i t)$$

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar}\left[H_0 + H_{int}, \rho\right] + \left(\frac{\partial \rho}{\partial t}\right)_{relax}$$

$$\left(\frac{\partial \rho}{\partial t}\right)_{relax} = -\frac{i}{\hbar}\left[H_{random}, \rho\right] \quad \Rightarrow \quad \left(\frac{\partial \rho_{nm}}{\partial t}\right)_{relax} = -\Gamma_{nm}\rho_m$$

Diagonal elements ⇔ ***atomic population***

Off-diagonal elements ⇔ ***quantum coherences***

$$\rho = \rho^{(0)} + \rho^{(1)} + \rho^{(2)} + \cdots \qquad \rho^{(n)} = \sum_j \rho^{(n)}(\omega_j)$$

After solving system of coupled equations

$$\boxed{\rho^{(n)} \Rightarrow \langle P^{(n)} \rangle \Rightarrow \chi^{(n)}}$$

Rabi frequency has to be smaller than the frequency detuning

When there is inversion symmetry:

$$\chi^{(j)} \equiv 0$$
j = *even*

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

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

Nonlinear refractive index

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

Nonlinear absorption coefficient

linear + nonlinear

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

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

High-order nonlinearities (HON)

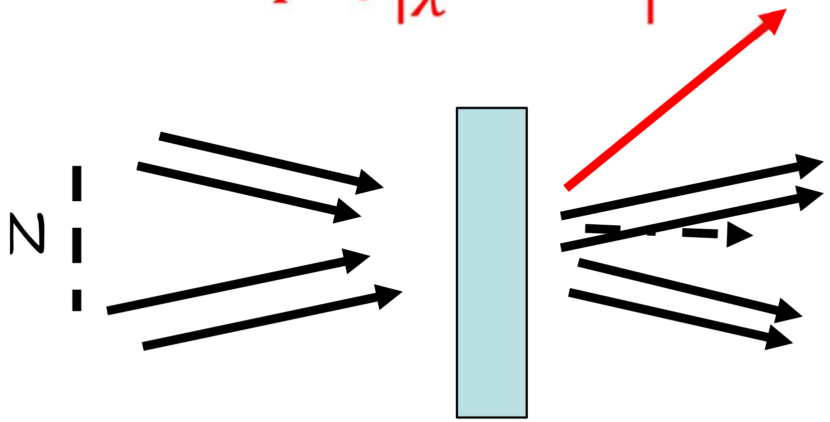
Normally HON are weaker than low-order effects

- Generation of high-order harmonics
- Multiphoton excitation processes
- Multi-wave mixing
- Stability of multidimension solitons

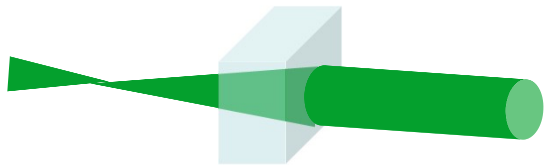
2nd lecture

Multi wave-mixing

$I \propto \left|\chi^{(2N+1)}\right|^2$

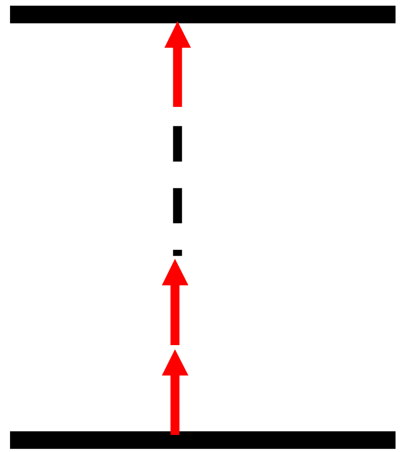


Self focusing



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

Multiphoton absorption



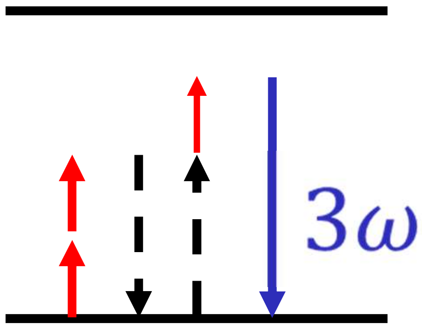
$\alpha_N \propto Im\,\chi^{(2N+1)}$

Cascade processes

HON may be due to repeated low-order susceptibilities

Macroscopic cascading: involves propagation effects

$\omega + \omega$ creates 2ω, then $(2\omega + \omega)$ creates 3ω



$$\chi^{(2)} \cdot \chi^{(2)} \equiv \chi^{(3)}_{eff}$$

Microscopic cascading:

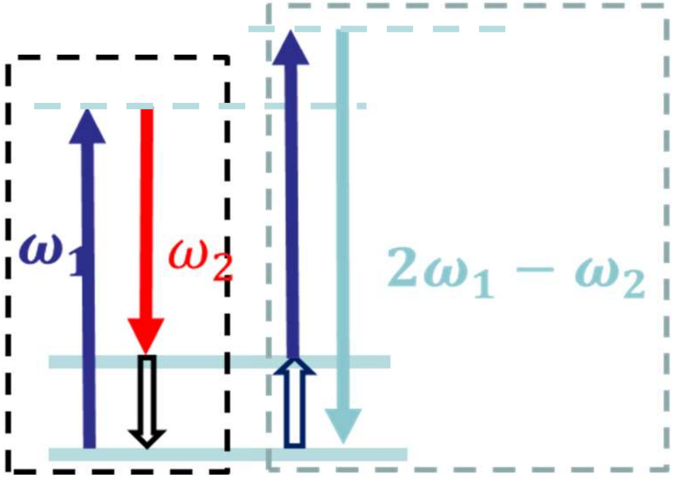
two neighbor atoms interact through local field effects to create a HON process

Medium without inversion symmetry

Yablonovitch, Flytzanis, Bloembergen
Phys. Rev. Lett. 29 (1972) 865

$$\chi^{(2)} \neq 0$$

Infrared transition (polariton)

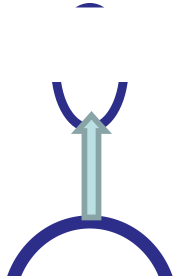


Local OR non-local

$$E(\omega_1 - \omega_2) \propto \chi^{(2)} E(\omega_1) E^*(\omega_2)$$

$$E(2\omega_1 - \omega_2) \propto \chi^{(2)} E(\omega_1 - \omega_2) E(\omega_1) =$$
$$\propto \chi^{(2)} \{\chi^{(2)} E(\omega_1)\ E^*(\omega_2)\}\ E(\omega_1)$$

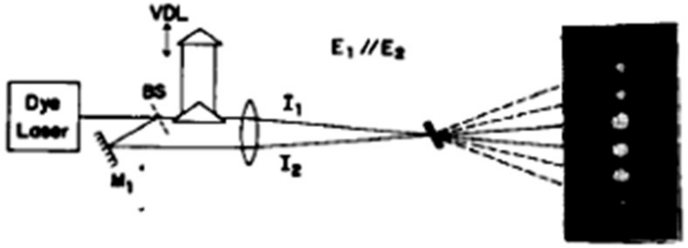
Effective susceptibility $\quad \boldsymbol{\chi_{eff}^{(3)} = A\chi^{(2)}.\chi^{(2)}}$



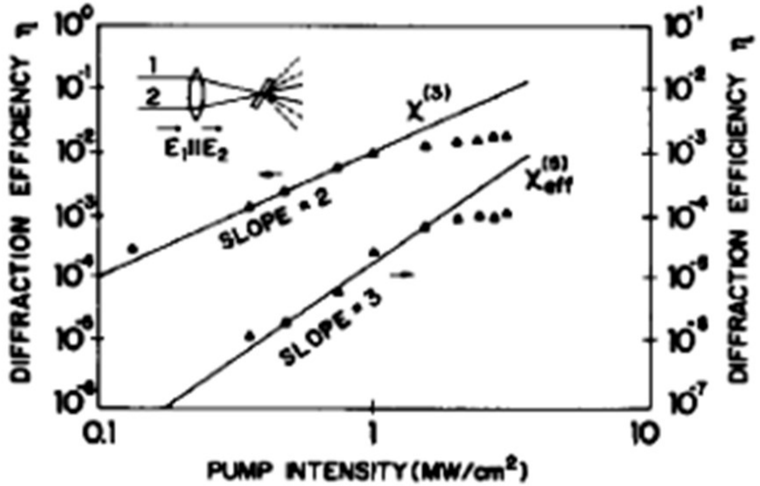
Glass with nanocrystals of CdS_xSe_{1-x}

Acioli et al. IEEE QE 26 (1990) 1277

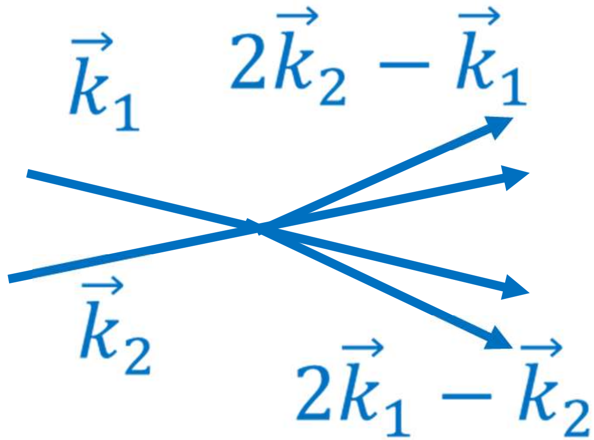
$\hbar\omega_{laser} \approx E_{gap}$



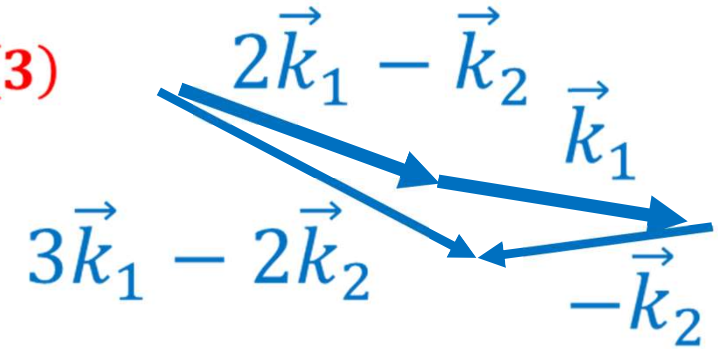
Self-diffraction



$\chi^{(3)}$

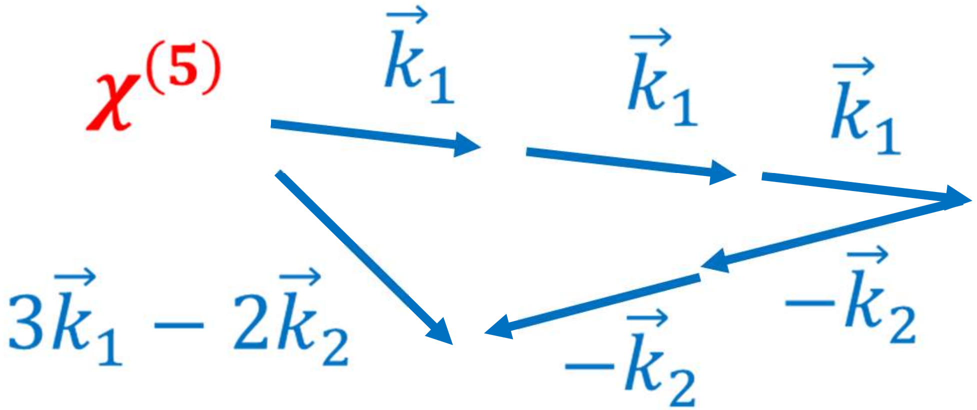


$\chi^{(3)}$



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

$\chi^{(5)}$



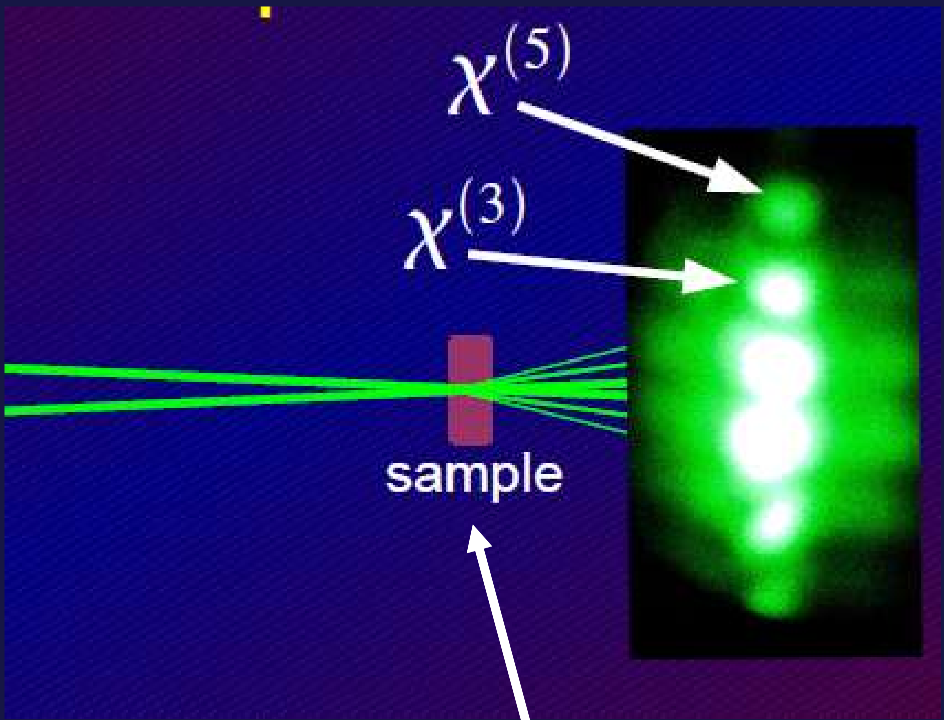
$$\boxed{\chi^{(5)}_{eff} = A\chi^{(5)} + B\chi^{(3)} . \chi^{(3)}}$$

PRL **103,** 113902 (2009) PHYSICAL REVIEW LETTERS week ending 11 SEPTEMBER 2009

Observation of a Microscopic Cascaded Contribution to the Fifth-Order Nonlinear Susceptibility

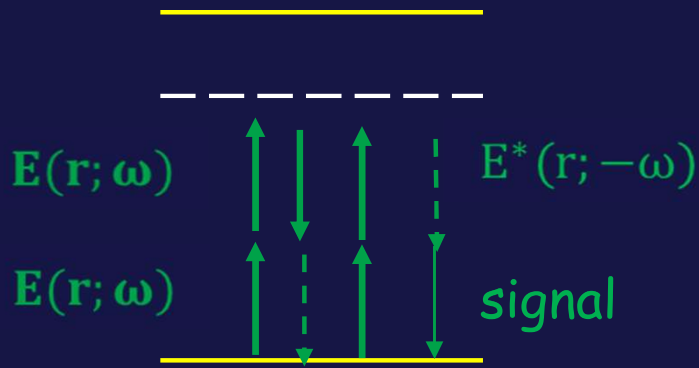
Ksenia Dolgaleva,* Heedeuk Shin, and Robert W. Boyd

$$\chi^{(5)}_{eff} \propto A\chi^{(3)}:\chi^{(3)} + B\chi^{(5)}$$



Cascade

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



There are other possibilities

Mixture of CS_2 and Fulerene (C_{60})

Microscopic cascading by local field effects

Third-order hyperpolarizability

$$\chi^{(3)} = N\gamma_{at}^{(3)}|L|^2 L^2,$$

Lorentz local field factor

$$L = \frac{\varepsilon^{(1)} + 2}{3}$$

Direct contribution from the fifth-order hyperpolarizability

$$\chi^{(5)} = N\gamma_{at}^{(5)}|L|^4 L^2 + \frac{24\pi}{10} N^2 (\gamma_{at}^{(3)})^2 |L|^4 L^3 + \frac{12\pi}{10} N^2 |\gamma_{at}^{(3)}|^2 |L|^6 L.$$

Contributions by the third-order hyperpolatizability

Optical solitons

solutions of Maxwell's equation with NL terms

Wave with special shape such that propagates without change its shape.

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

Temporal solitons

Spatial solitons

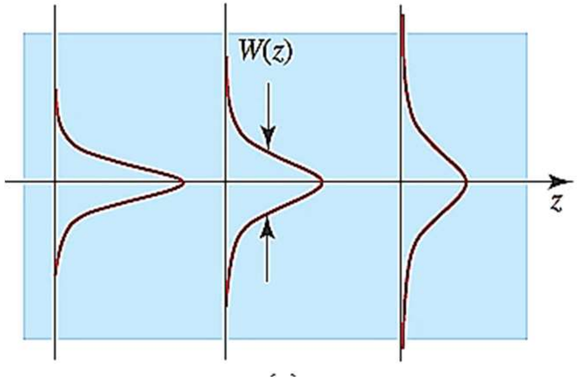
Temporal + spatial solitons ("light bullets")

Spatial Optical Solitons

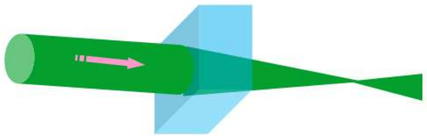
Solutions of Maxwell equations including nonlinear polarization

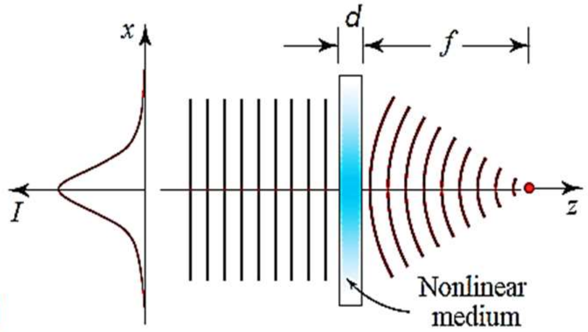
Example: NL Schrodinger equation

Diffraction

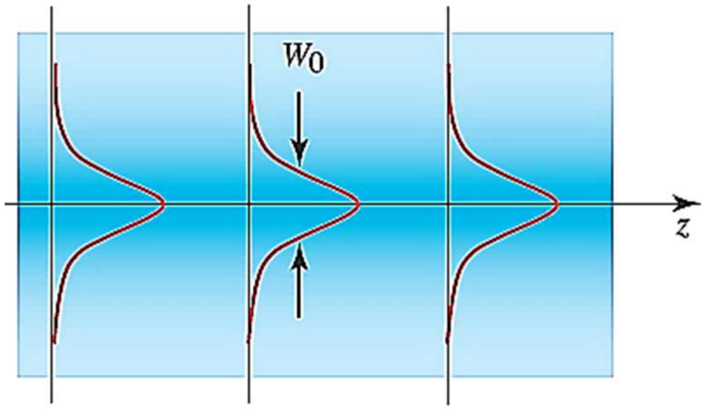


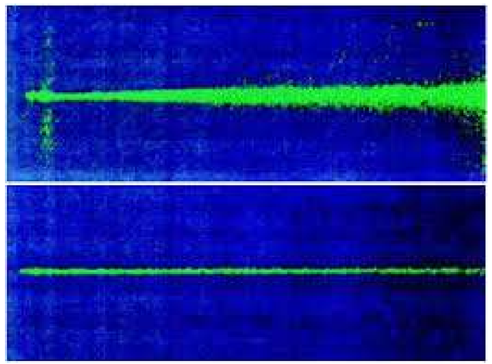
Self-focusing material





Bright spatial soliton





Small intensity

Large intensity

Soliton

Light propagation inside a NL medium

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

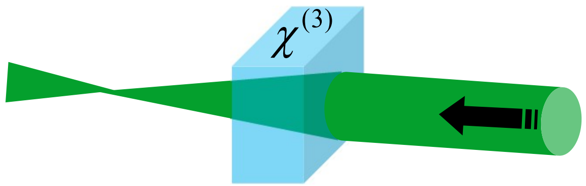
$$\vec{E}(\vec{r},t) = \vec{A}(\vec{r}) \exp[i(kz - \omega t)]$$

third-order nonlinearity

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

diffraction

Self-focusing



Nonlinear Schrödinger Equation: (1+1)D

Scalar theory

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

Inside a waveguide

Diffraction x self-focusing

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

$$\left|\frac{\partial^2 a(x,z)}{\partial z^2}\right| << \left|k_0 \cdot \frac{\partial a(x,z)}{\partial z}\right| \qquad \eta_0 = \sqrt{\frac{\mu_0}{\varepsilon_0}}$$

$$\boxed{a(x,z) = \mathrm{sech}\left(\frac{x}{X_0}\right)\exp(iz/2L_d)}$$

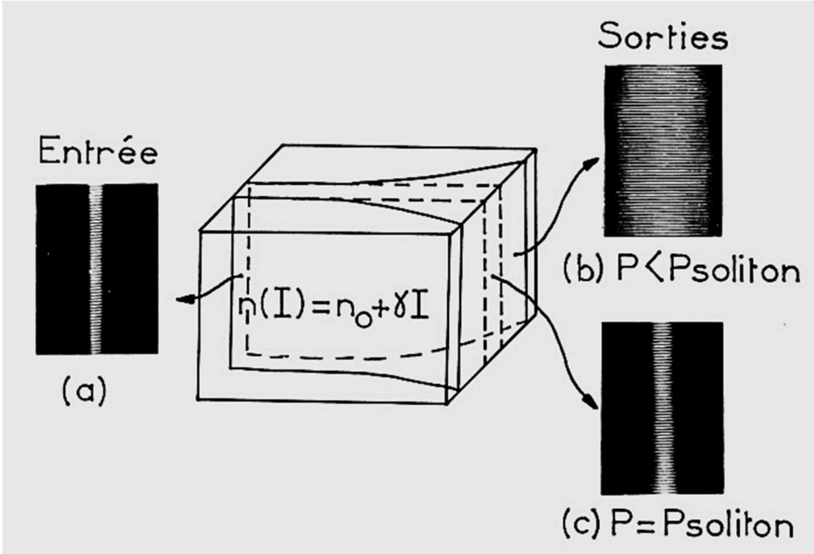
$$L_d = X_0^2 k_0 n_0 \qquad X_0 = \frac{1}{k_0 n_0}\sqrt{\frac{2\eta_0}{|n_2||A_m|}}$$

(1+1)D spatial solitons in CS_2

$n_2 > 0$

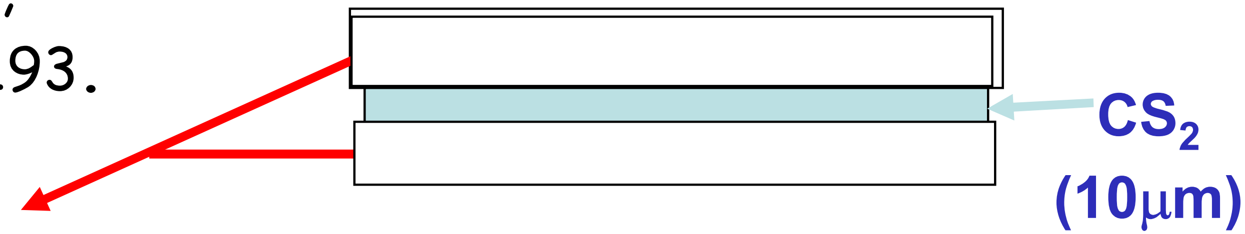
Beam focused by cylindrical lenses

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

$$E(x,y,z,t) = C\,a(x,y,z,t)\exp[i\,(k_0 nz - \omega t)]$$

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

$n_2 > 0$

neglecting 2PA

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

Catastrophic self-focusing

High-order nonlinearity

$\chi^{(5)}$

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

Stable solution if $n_2 > 0$ and $n_4 < 0$

This problem was known
since the years 60s but
no homogeneous and isotropic
material was identified up until recently.

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

Important: contributions of third and fifth order of opposite signs

Carbon disulfide: CS_2

VOLUME 74, NUMBER 15 PHYSICAL REVIEW LETTERS 10 APRIL 1995

Fifth Order Optical Response of Liquid CS_2 Observed by Ultrafast Nonresonant Six-Wave Mixing

Keisuke Tominaga and Keitaro Yoshihara

Results in the femtosecond regime:

***n_2*: Couris *et al.*, Chem. Phys. Lett. 369, 318 (2003).**

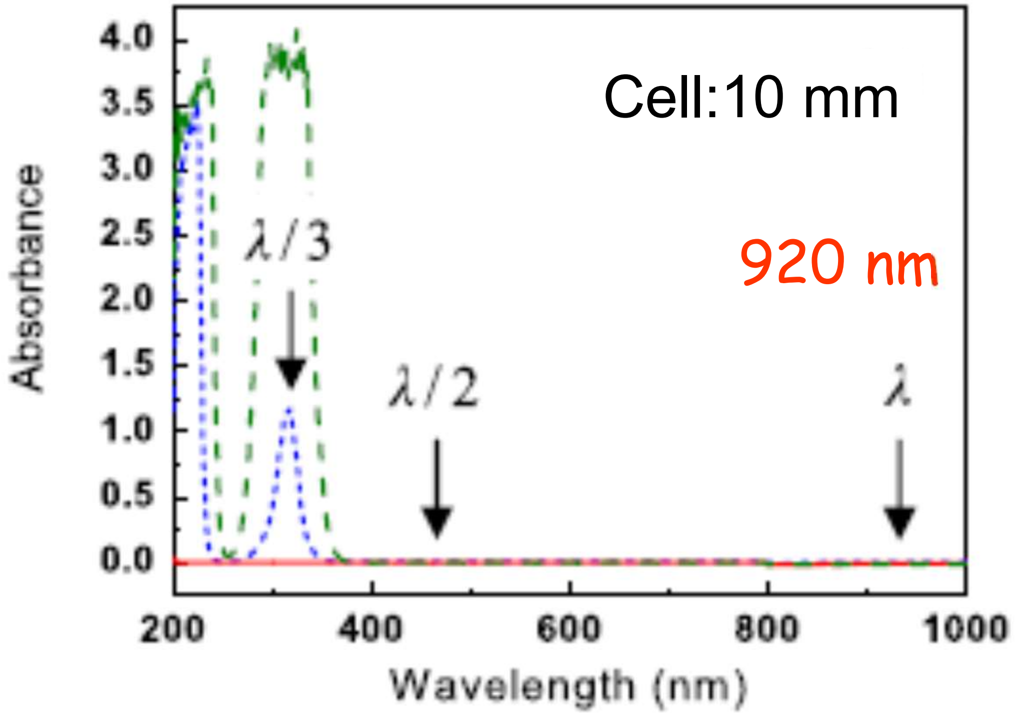
***n_4*: Kong *et al.*, J. Phys. B: At. Mol. Phys. 42, 065401 (2009).**

$n_2 = +3.1 \times 10^{-19}\ m^2/W$ $\quad$ $n_4 = -2.0 \times 10^{-35}\ m^4/W^2$

Focusing nonlinearity $\quad$ Defocusing nonlinearity

Absorbance spectrum of CS_2 diluted in ethanol

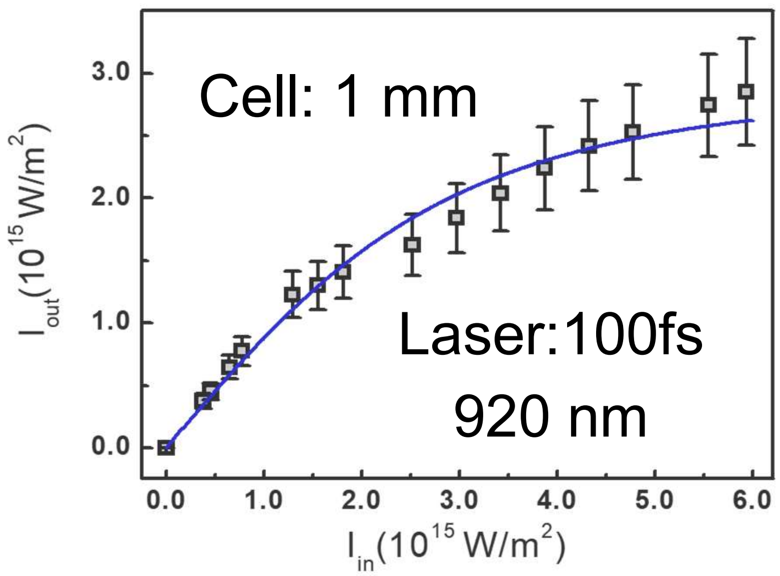
$n_2 > 0 \quad n_4 < 0$



Optical limiting experiment

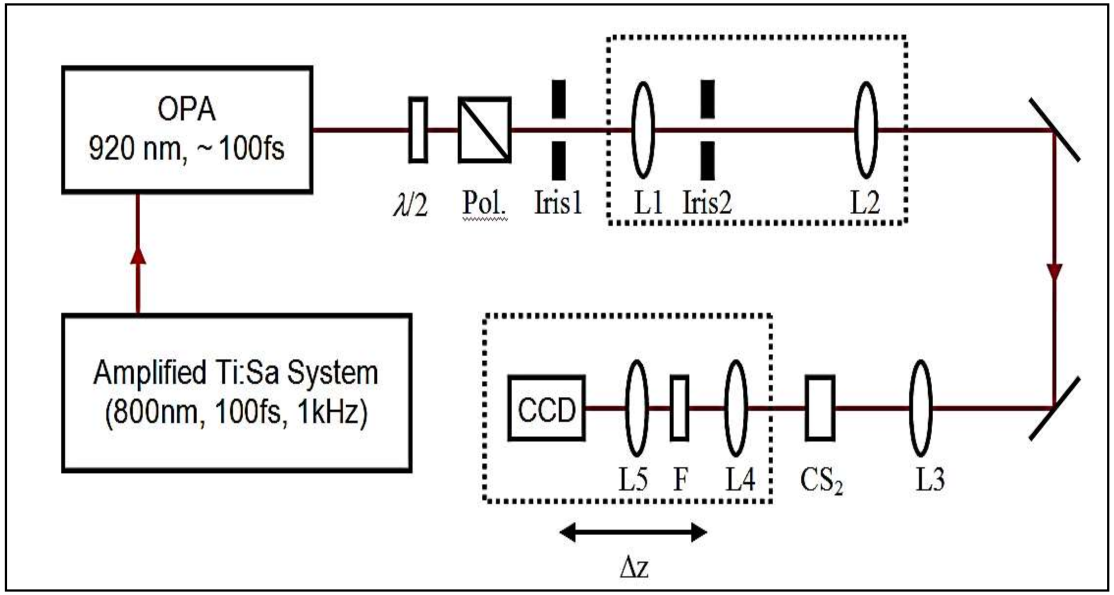
$$I_L = (1-R)^2 \frac{I_0}{\sqrt{1 + 2I_0^2(1-R)^2\alpha_4 L}}$$

$\alpha_4 = 5.8 \times 10^{-29}\ m^3/W^2$

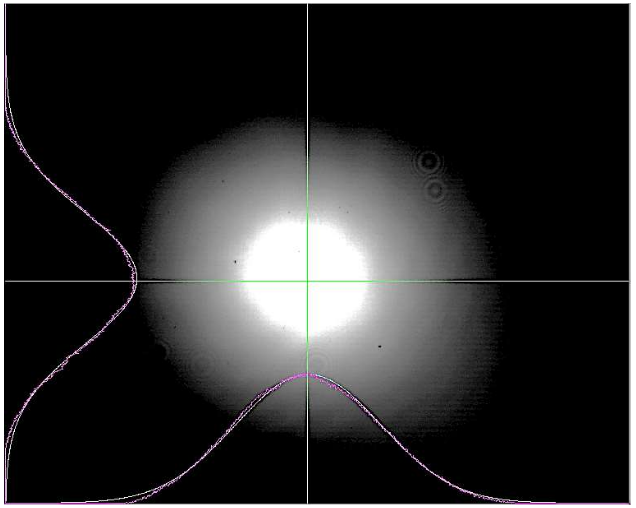


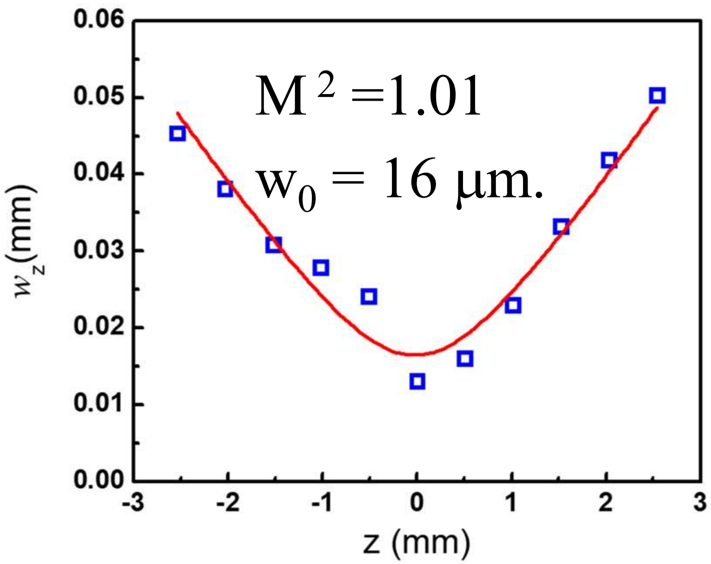
Three photon absorption of pure CS_2

experimental setup

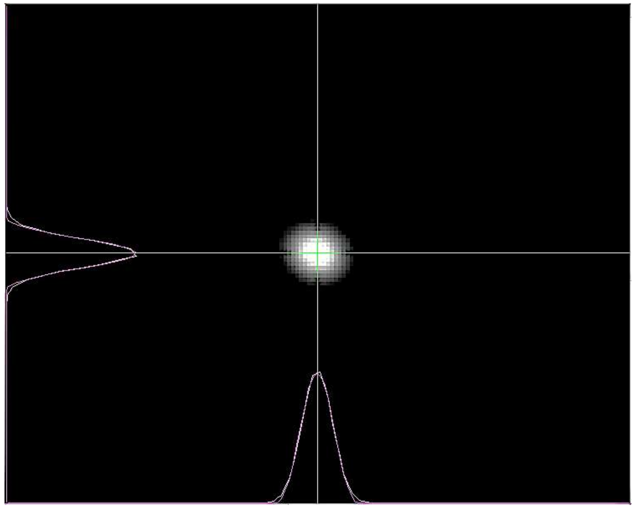


Beam waist on L3: $w = 2.0$ mm





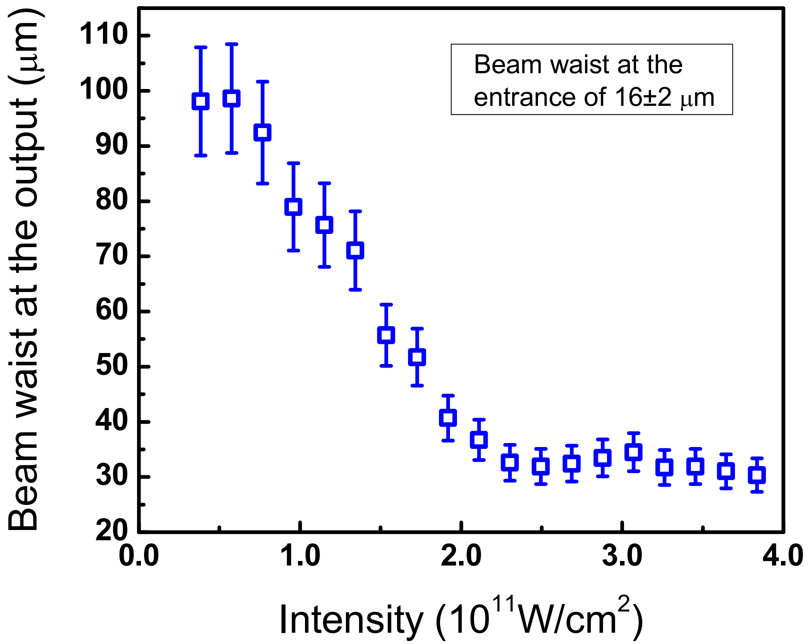
Focal region: $w_0 = 16\ \mu m$

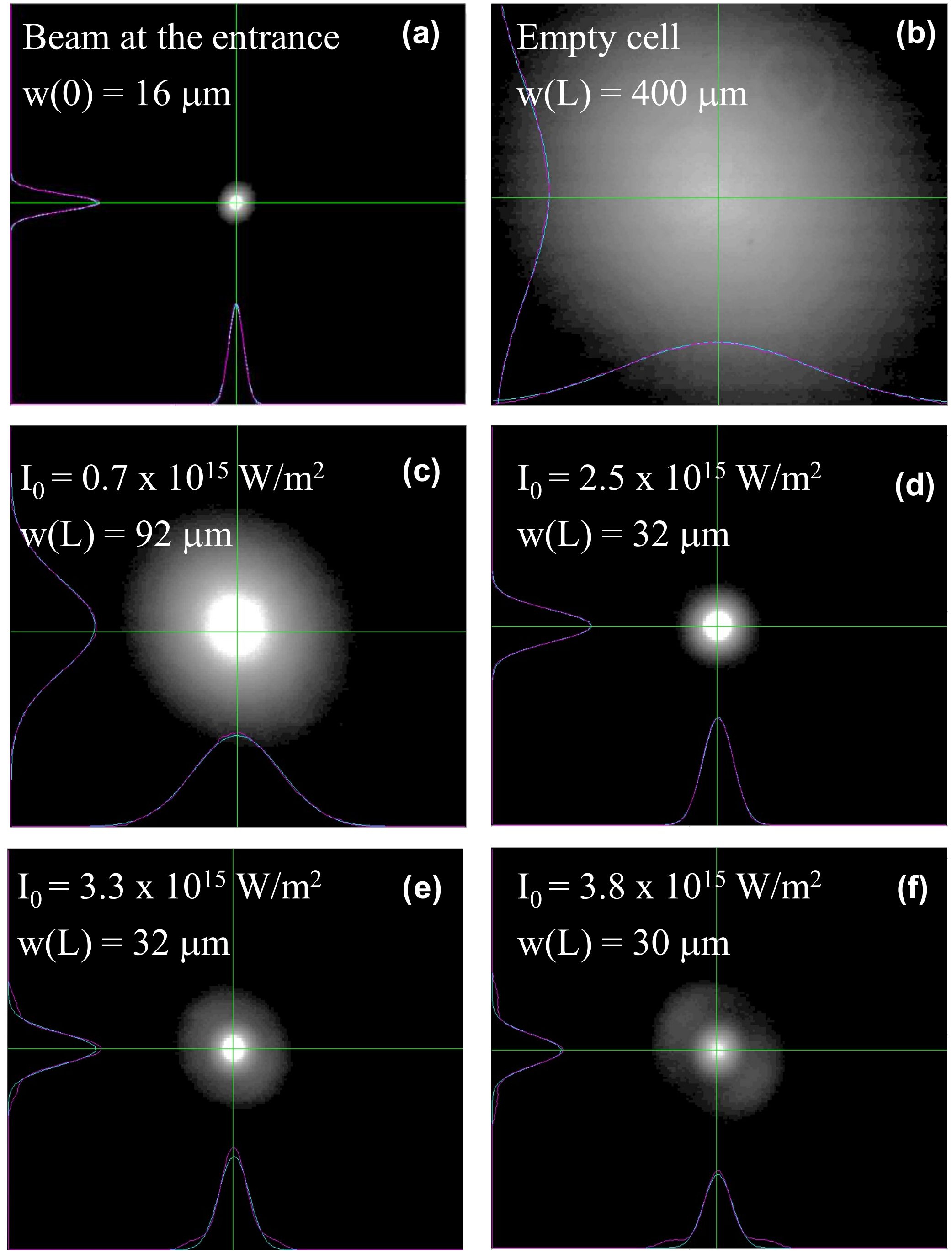


45

cell length: 1.0 cm

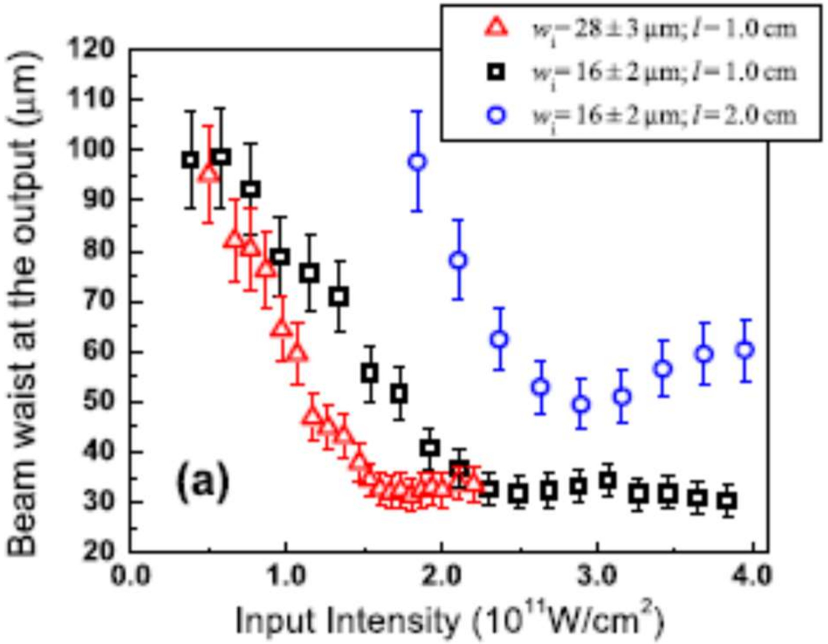
Beam focused on the entrance face



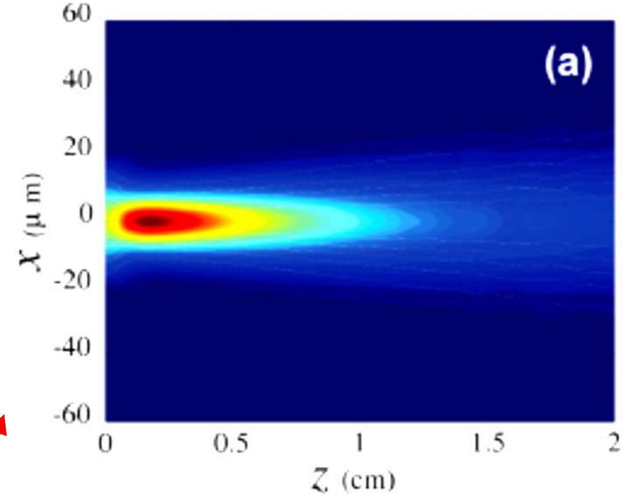


1 cm → $10z_0$

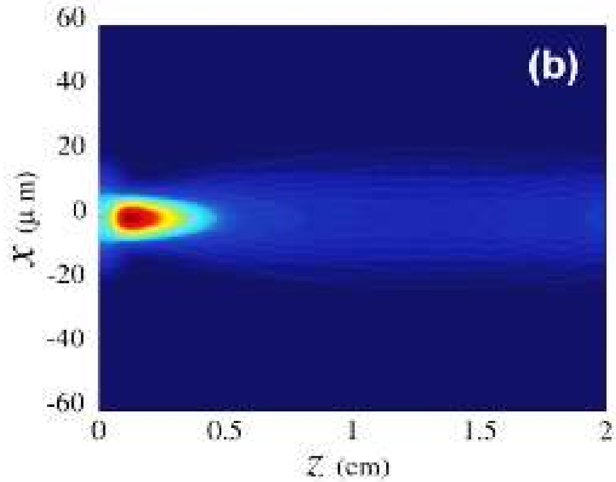
Beam waist versus laser intensity



Cells: 1 and 2 mm



0.8×10^{11} W/cm^2



NLS equation

1.6×10^{11} W/cm^2

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 agreement with the experimental data.

Optical vortices

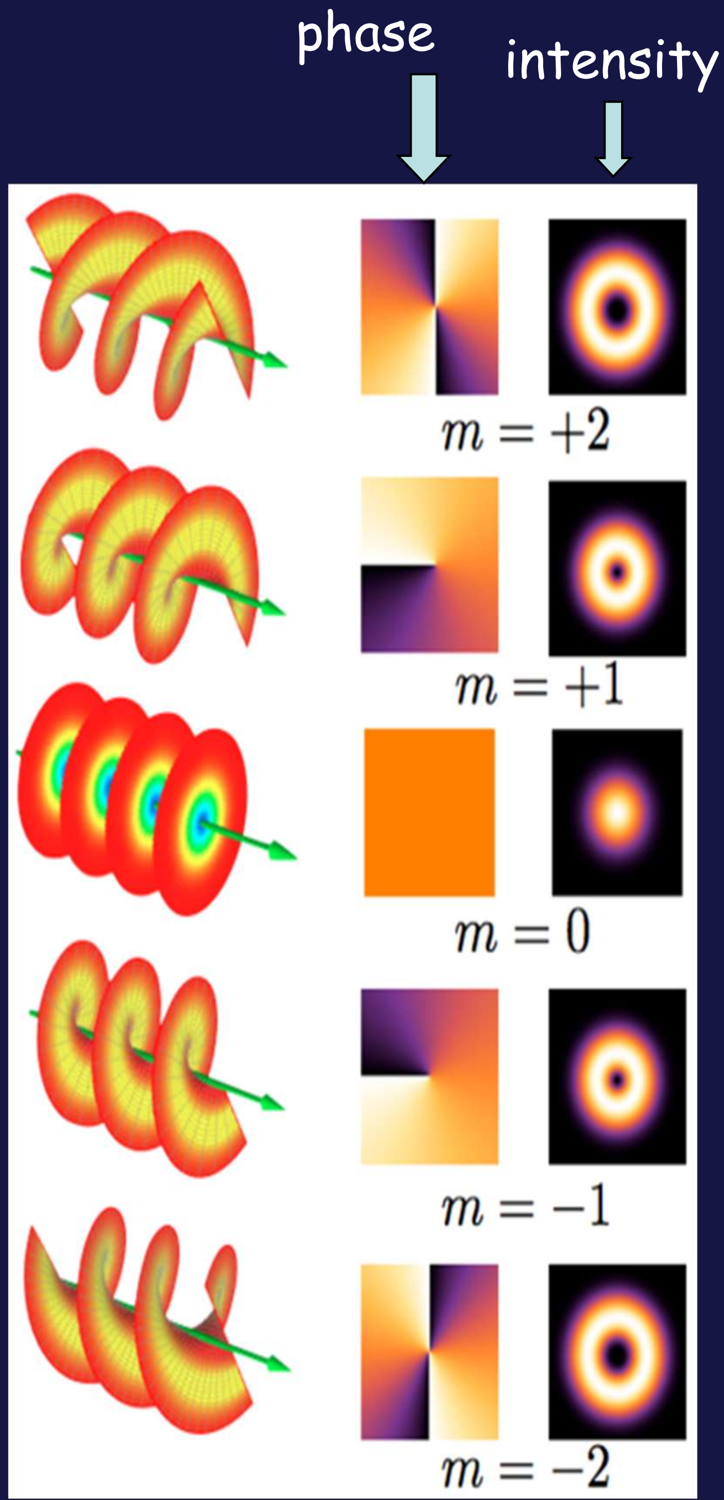
- Beams with phase singularity
- Zero field in the center of the vortex
- Helical wavefront
- Phase

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

$m = 0$ Plane wave

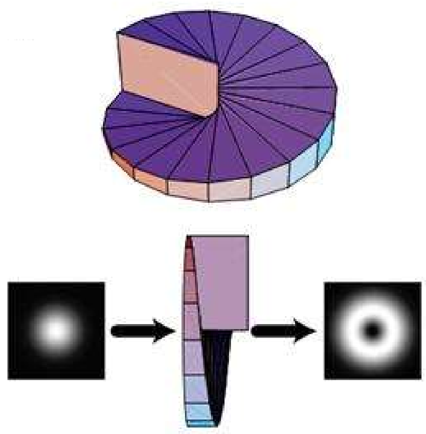
$m \neq 0$ Wave with topological phase

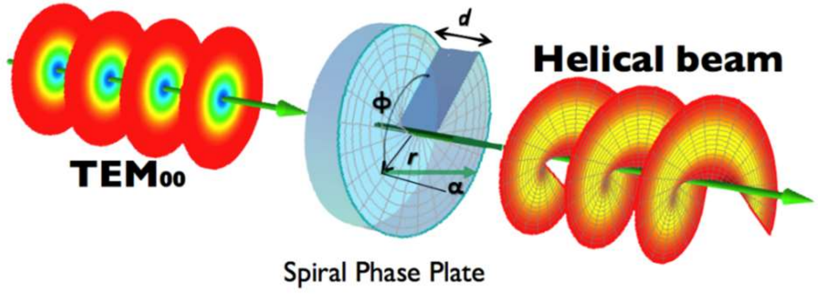
m is the "topological charge"



Optical Vortex beam carries an orbital angular momentum of $m\hbar$ per photon

Vortex phase plate





Spatial light modulator

Images from the internet

Optical Vortex Soliton - OVS

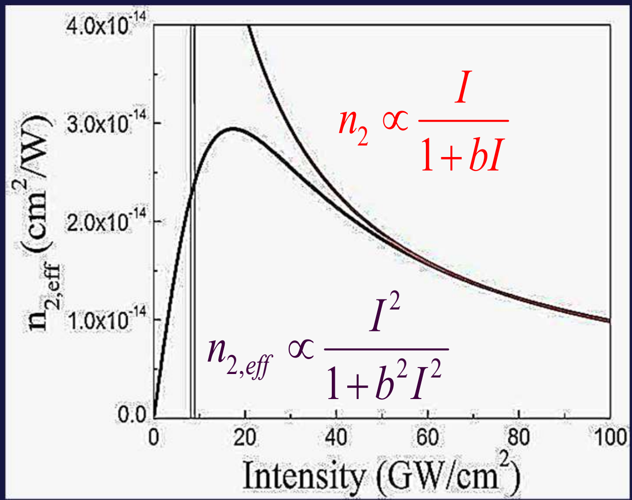
Stable propagation in a self-defocusing medium

Unstable propagation in self-focusing media

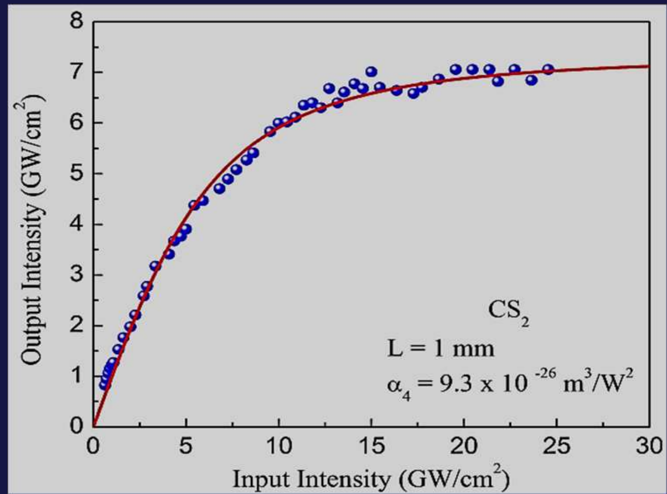
Conditions on the NLSE to observe stable OVS in a self-focusing medium?

Saturable NL refractive index and NL absorption

CS_2
532 nm
psec



Effective NL refractive index



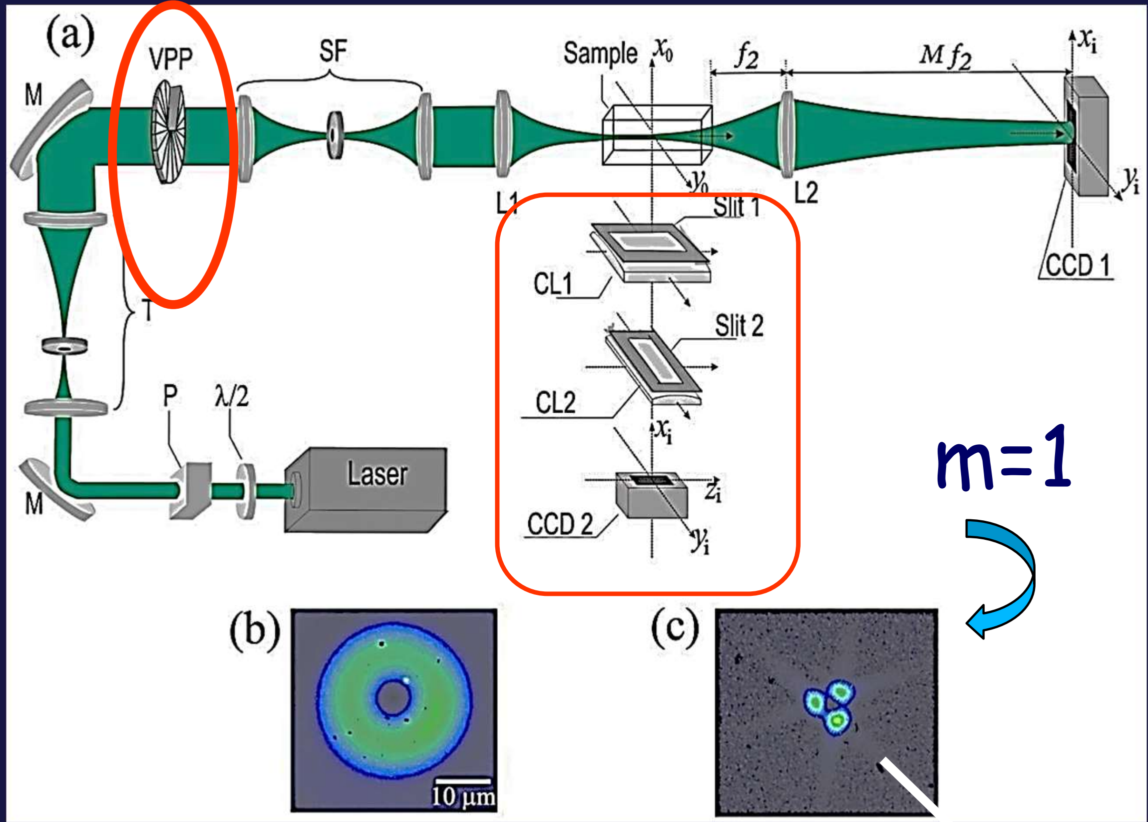
NL transmittance

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]

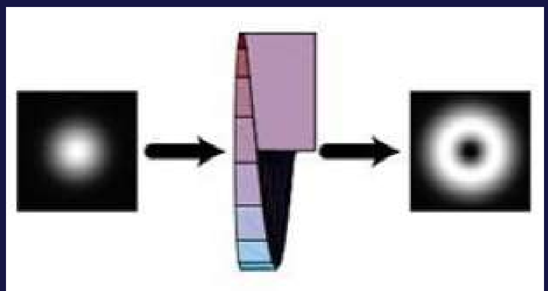
Propagation of OVS in CS_2



m=1

532 nm
80 ps
10 Hz

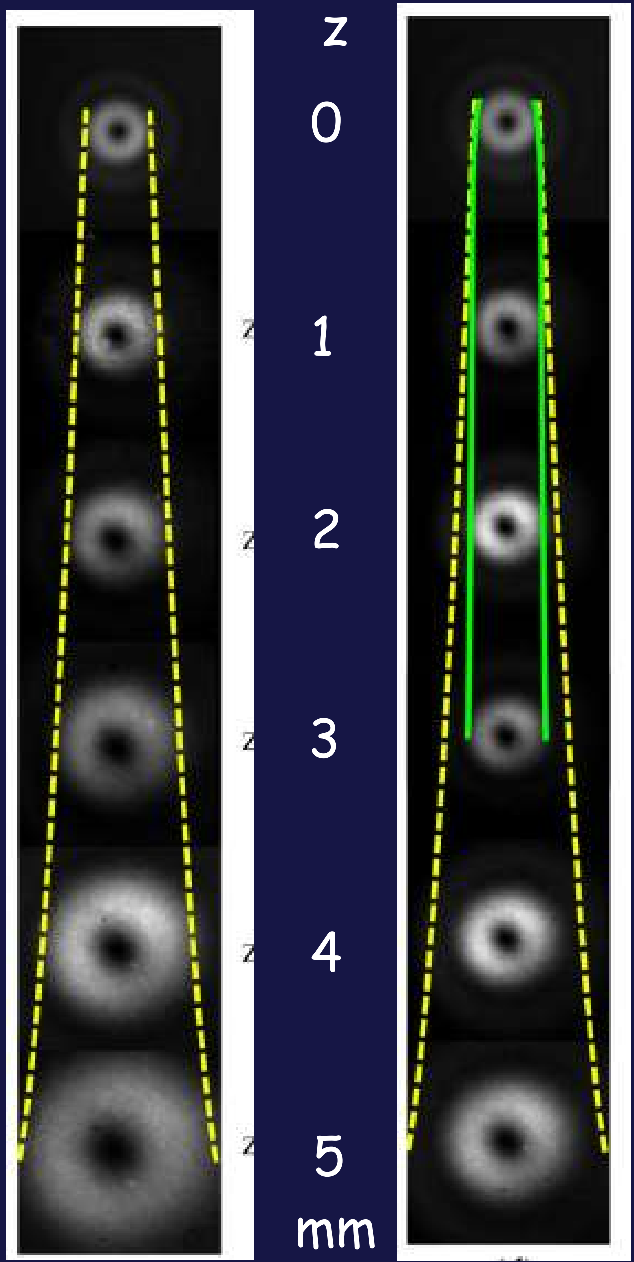
VPP- vortex phase plate



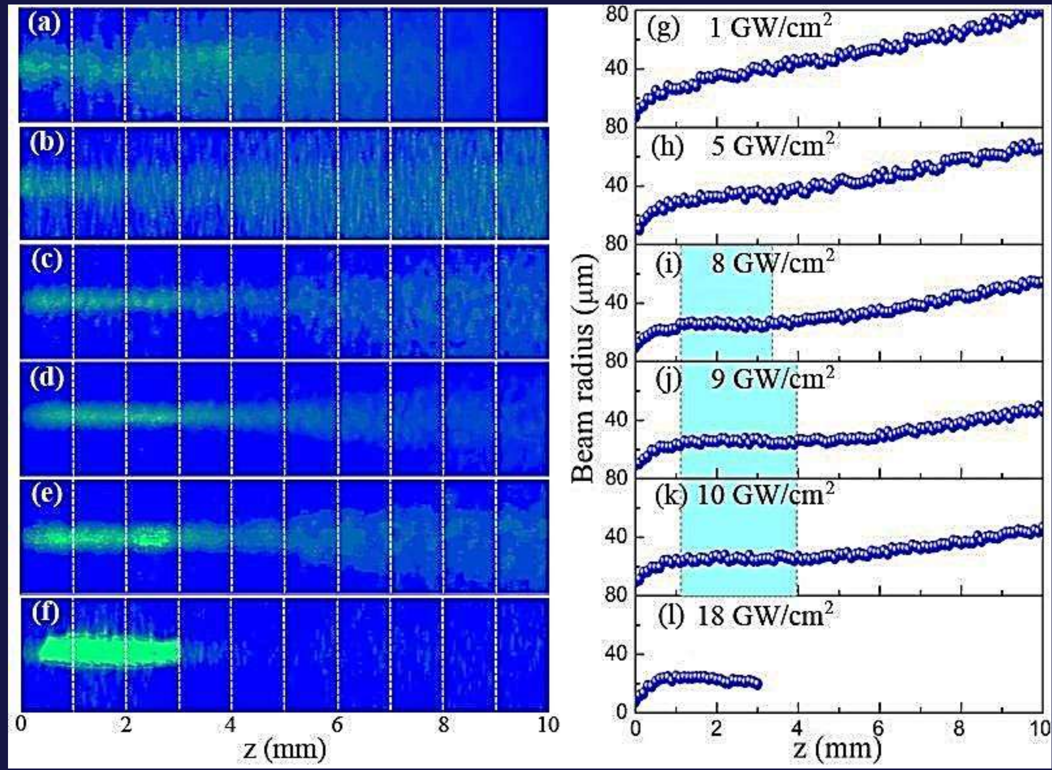
Optical Vortex beam carries an orbital angular momentum of $m\hbar$ per photon

Hickmann et al.
PRL 105 (2010) 053904

Optical Vortex Solitons in CS_2



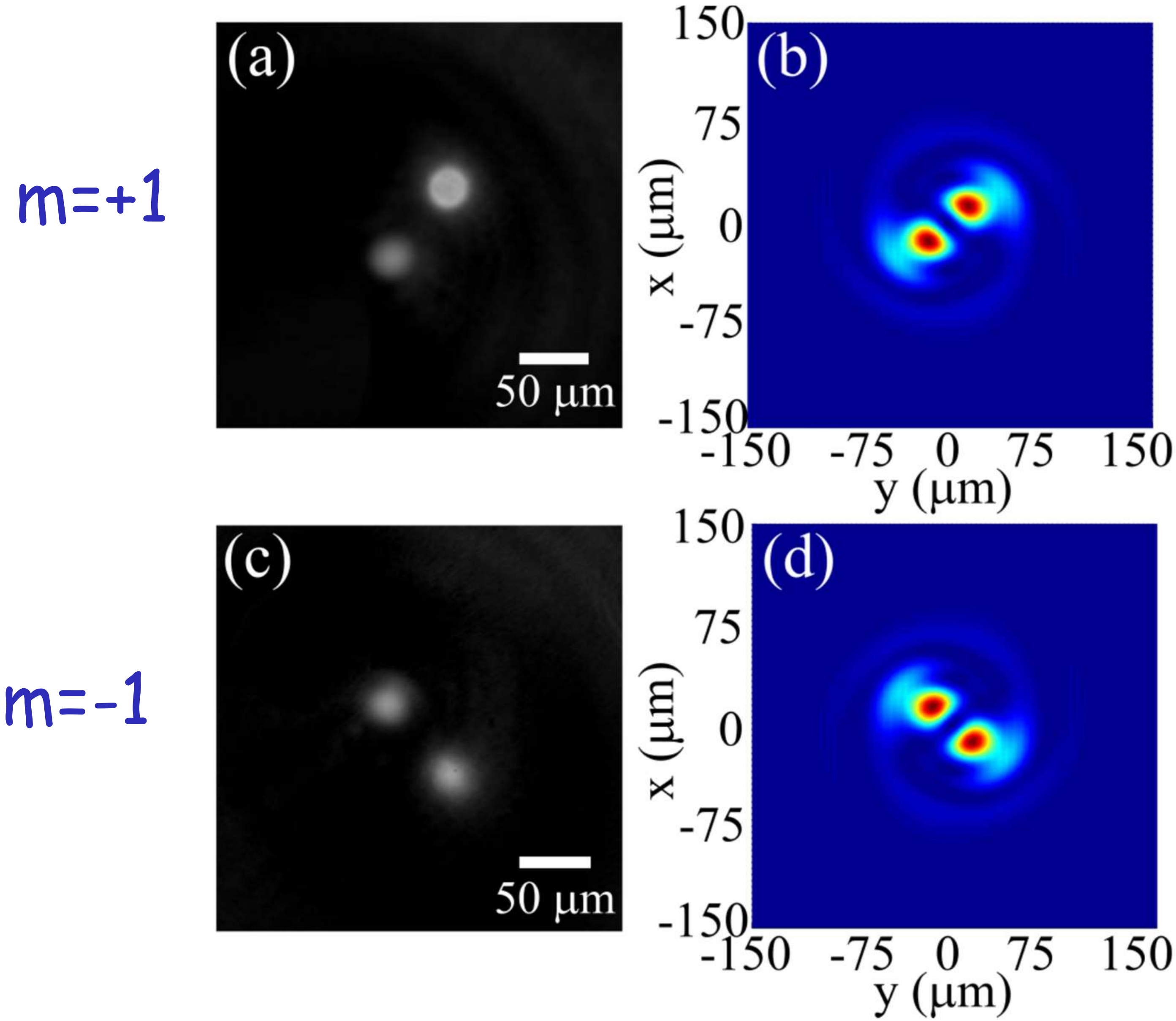
1.0 GW/ cm^2 9.0 GW/ cm^2



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

First observation of an OVS in a homogenous medium with local nonlinearity

Image at the output of cell after fragmentation

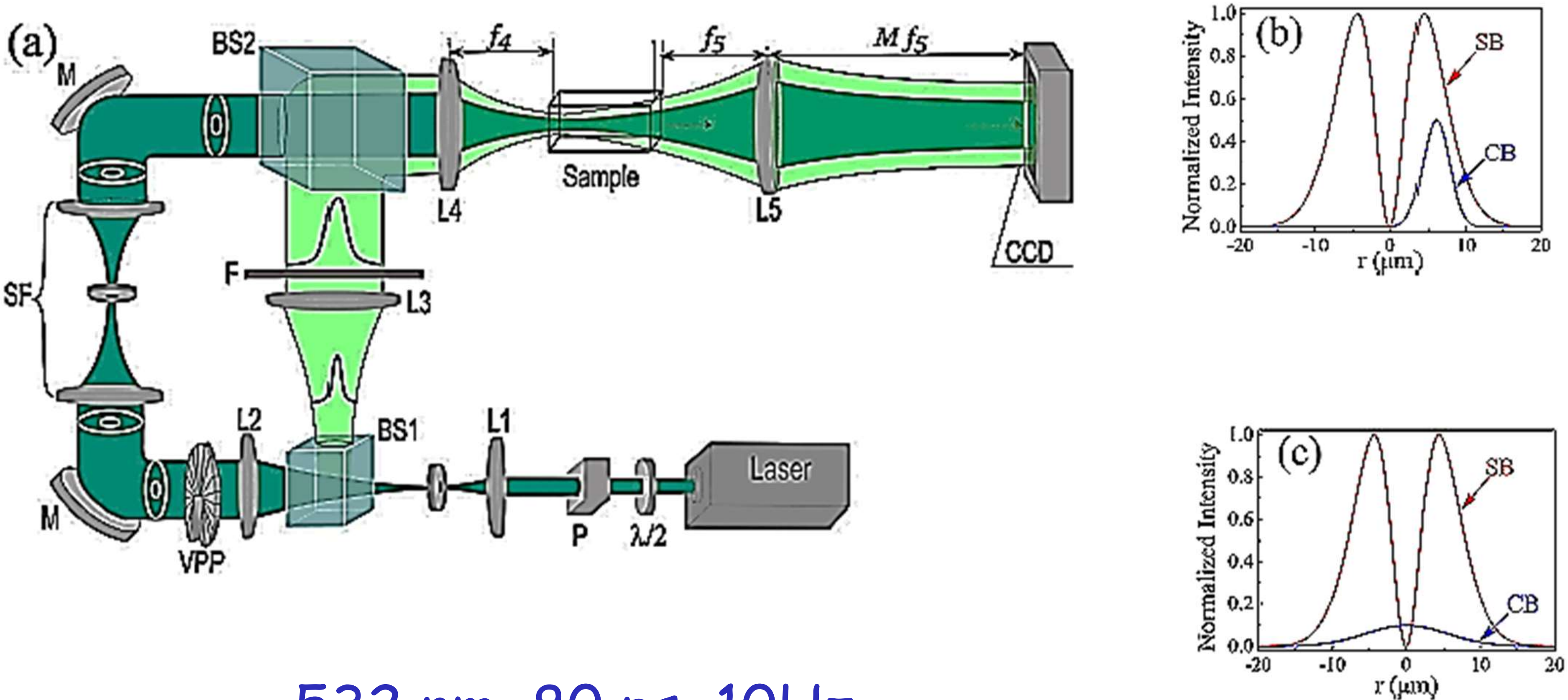


Experiment: 18 GW/cm^2 Simulation 15 GW/cm^2

PHYSICAL REVIEW A 93, 013843 (2016)

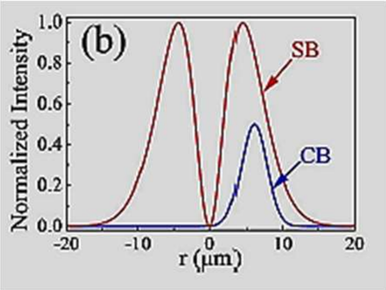
Taming the emerging beams after the split of optical vortex solitons in a saturable medium

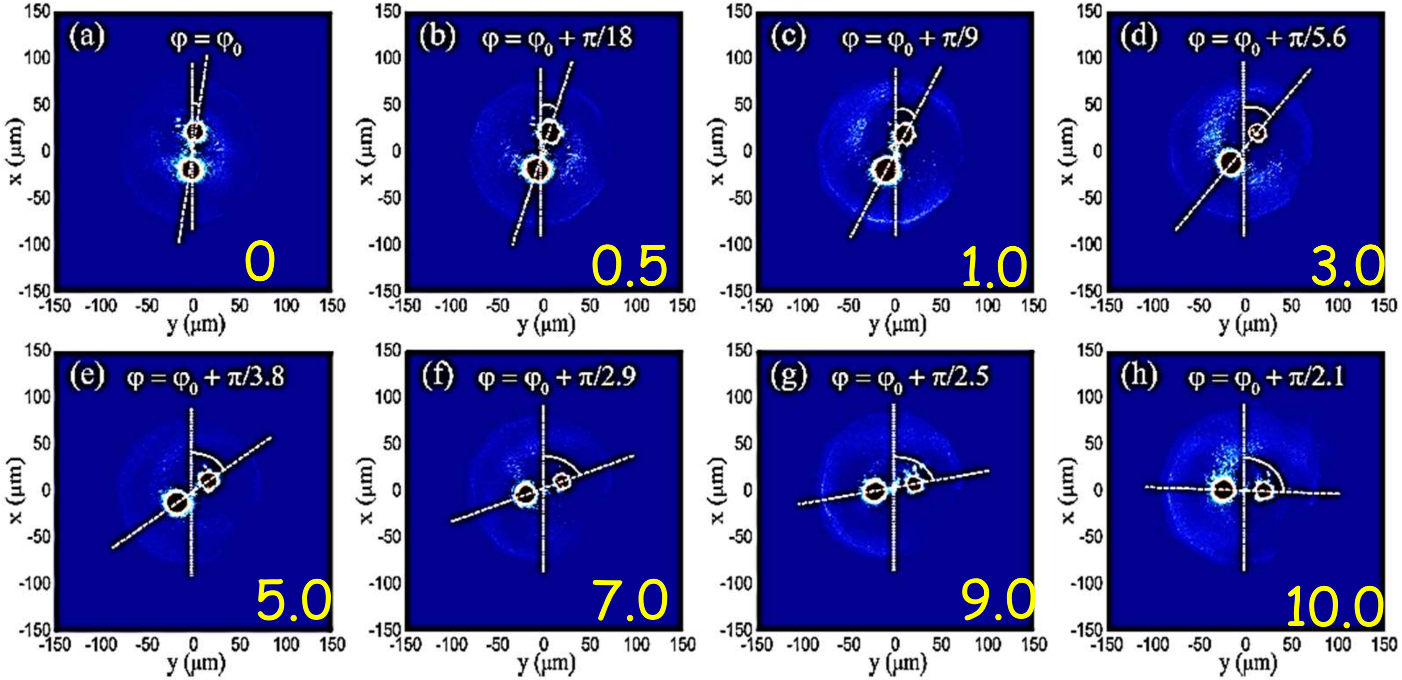
Albert S. Reyna* and Cid B. de Araújo



532 nm, 80 ps, 10Hz

cell length: 10 mm

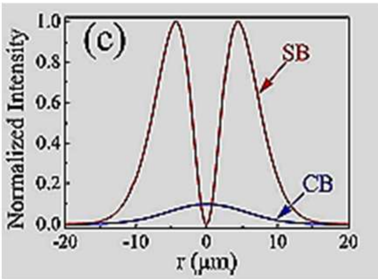


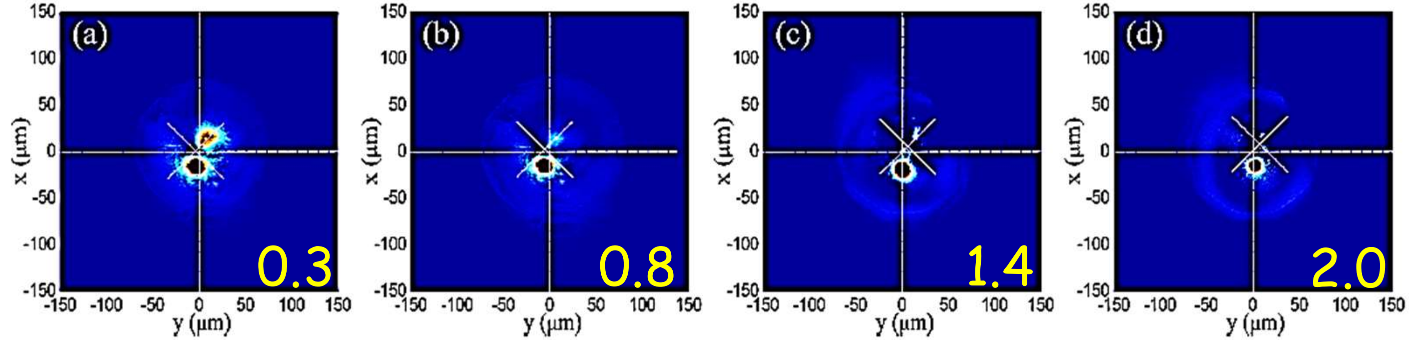


Vortex beam
18 GW/cm^2

Control beam
Gaussian
GW/cm^2

Control beam less intense than the signal beam





Control beam
0.3-2.0 GW/cm^2

Control of energy transfer

Final comments

Exploitation of high-order electronic nonlinearities allows::

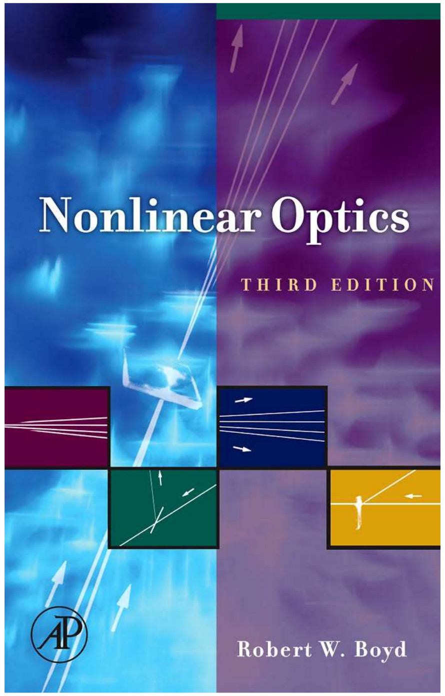
observation of (2+1)D bright spatial solitons in a homogeneous NL medium with electronic nonlinearity.

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

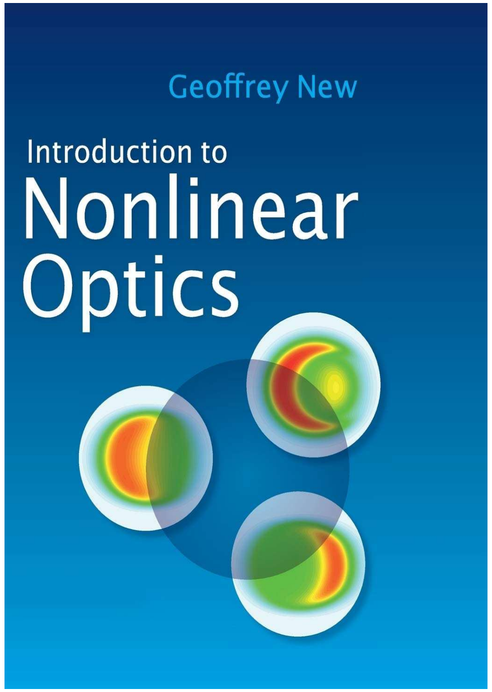
controlling the instability of an OVS by using a control beam with smaller intensity than the OVS.

Optical systems offer a simple ground for studies of <u>fundamental effects</u> in physics of nonlinear waves and different types of nonlinear phenomena

In the next two lectures I will present examples to illustrate these statements in Plasmonic systems and Random Lasers



R. W. Boyd



G. New

Christodoulides, Khoo, Salamo, Stegeman, van Stryland
Nonlinear refraction and absorption: mechanisms and magnitudes,
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High-order optical nonlinearities in plasmonic nanocomposites – a review
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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]

2nd lecture - tomorrow

Nonlinear Photonics in Plasmonic media

Thank you for your attention

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