FI255 - Tópicos de Óptica e Fotônica II

Óptica Não-Linear

11ª. aula

Prof. Cid B. de Araújo
UNICAMP - 25 de maio de 2018

Roteiro

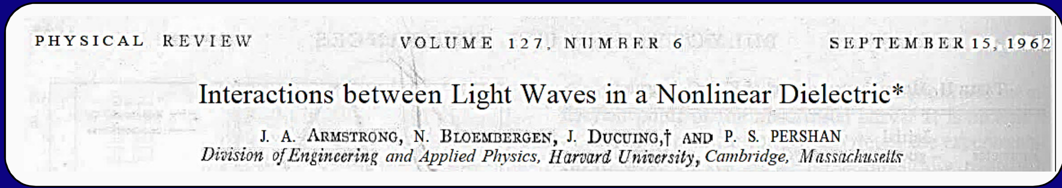
1. Técnicas não lineares para caracterização de materiais-Review

2. Representação gráfica dos processos não lineares-Review

3. Não linearidades de alta ordem

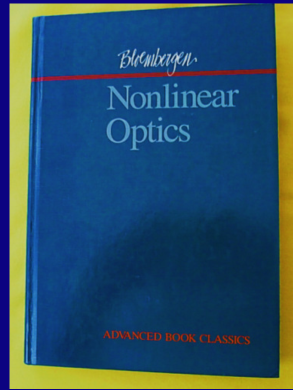
4. Cascatas de não linearidades - how to eliminate their influence

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

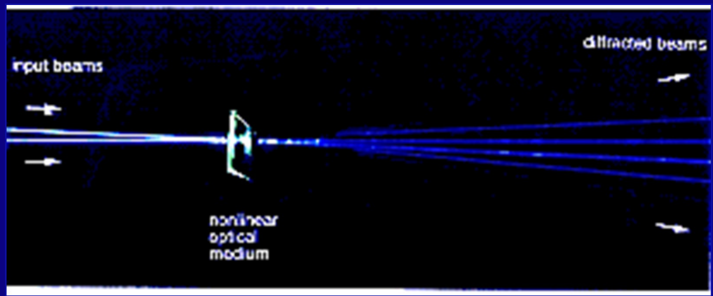
$$n = n_0 + n_2 I + n_4 I^2 + n_6 I^3 + \cdots$$

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

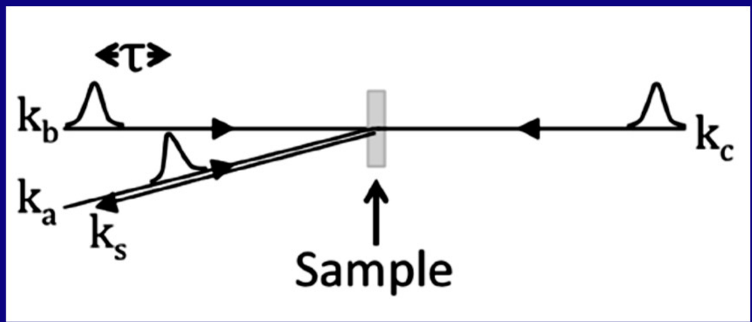
9ª. aula

Degenerate four wave-mixing

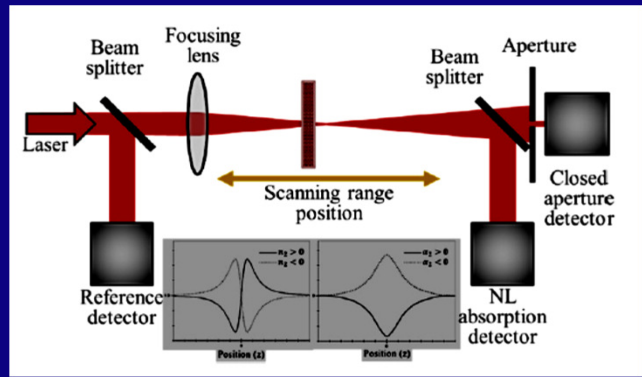
$|\chi^{(3)}|^2$



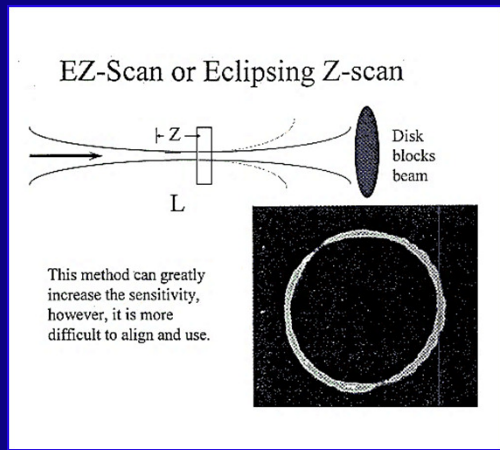
Phase conjugation



Z-scan



EZ-scan



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

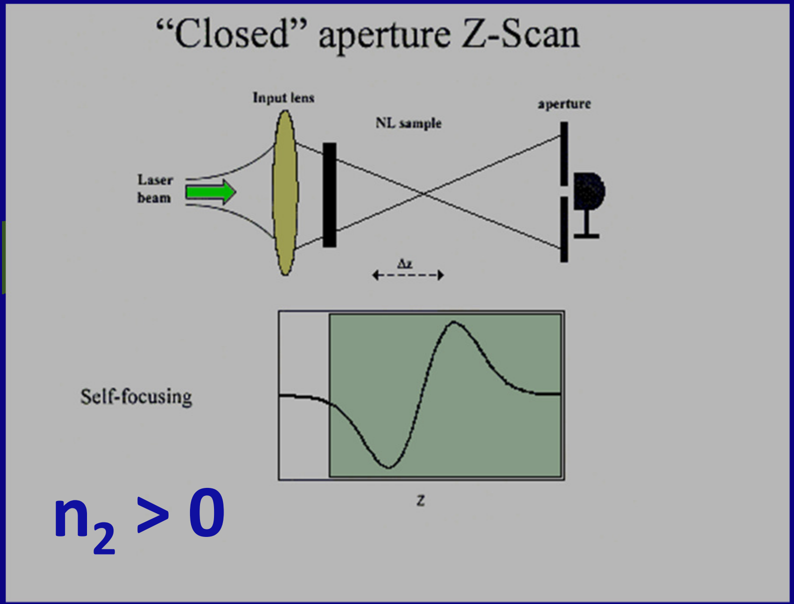
$Im\ \chi^{(3)}$

Nonlinear refraction

$$n = n_0 + n_2\, I(r)$$

$$I(r) = I_0 \exp\left[-r^2/w^2\right]$$

$n_2 > 0$ self focusing
$n_2 < 0$ self defocusing



Sample ≡ lens

$L \ll z_0$ $\quad L \ll z_0/\Delta\Phi_0$

$$T(z, \Delta\Phi_0) \cong 1 - \frac{4 \cdot \Delta\Phi_0 \cdot x}{(x^2+9)(x^2+1)};$$

$x = z/z_0$

$\Delta\Phi_0 = (n_2 I_0)k\, L_{eff}$

$L_{eff} = (1 - e^{-\alpha L})/\alpha$

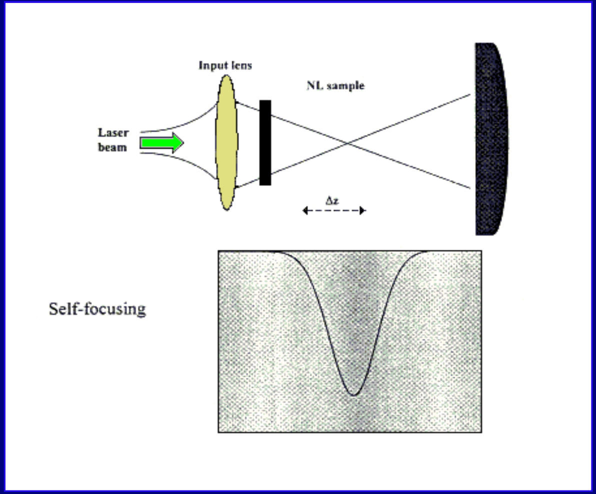
Sheik-Bahae et al.
IEEE J. Quantum Electron. 1990

Z-scan technique
Nonlinear absorption

when $\alpha_2 I_0 L_{eff} << 1$

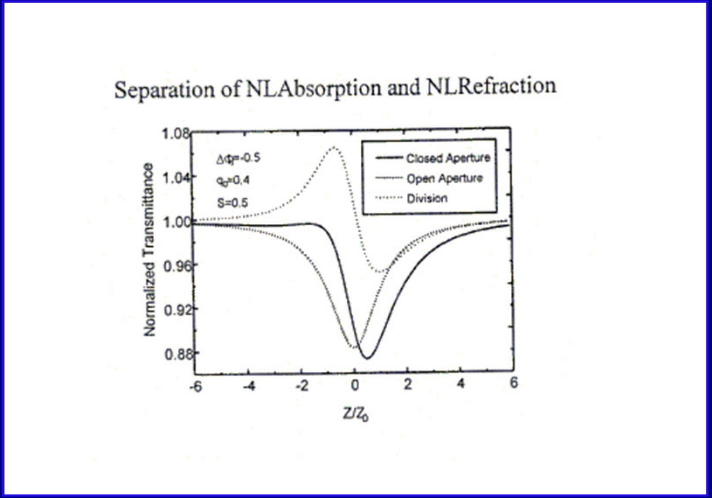
$$\Delta T(z) \approx -\frac{\alpha_2 \cdot I \cdot L_{eff}}{2\sqrt{2}} \frac{1}{1 + z^2 / Z_0^2}$$

Open aperture



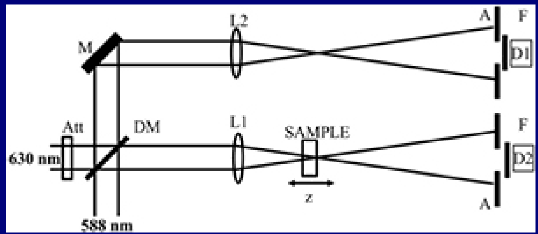
Closed aperture

NL refraction + NL absorption

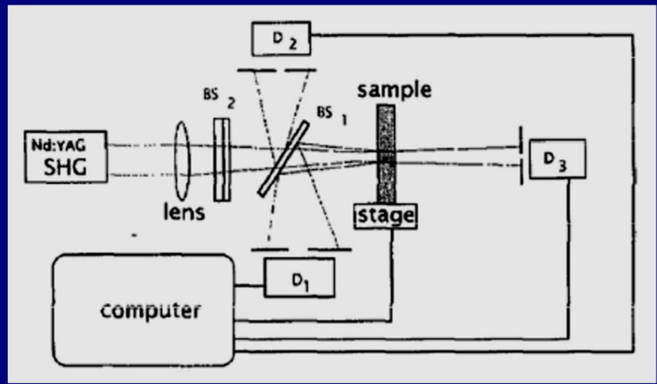


9ª. aula

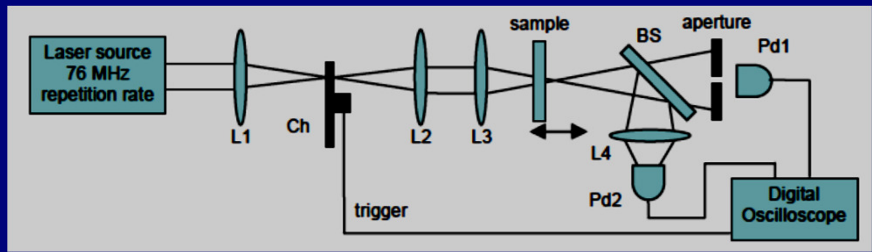
Two-color Z-scan



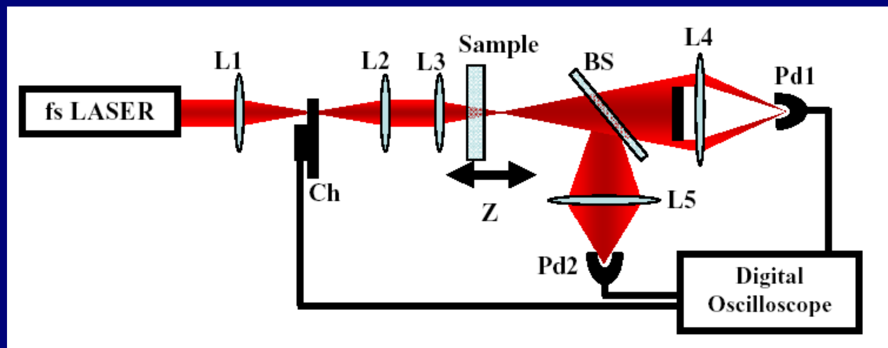
RZ-scan



TM Z-scan



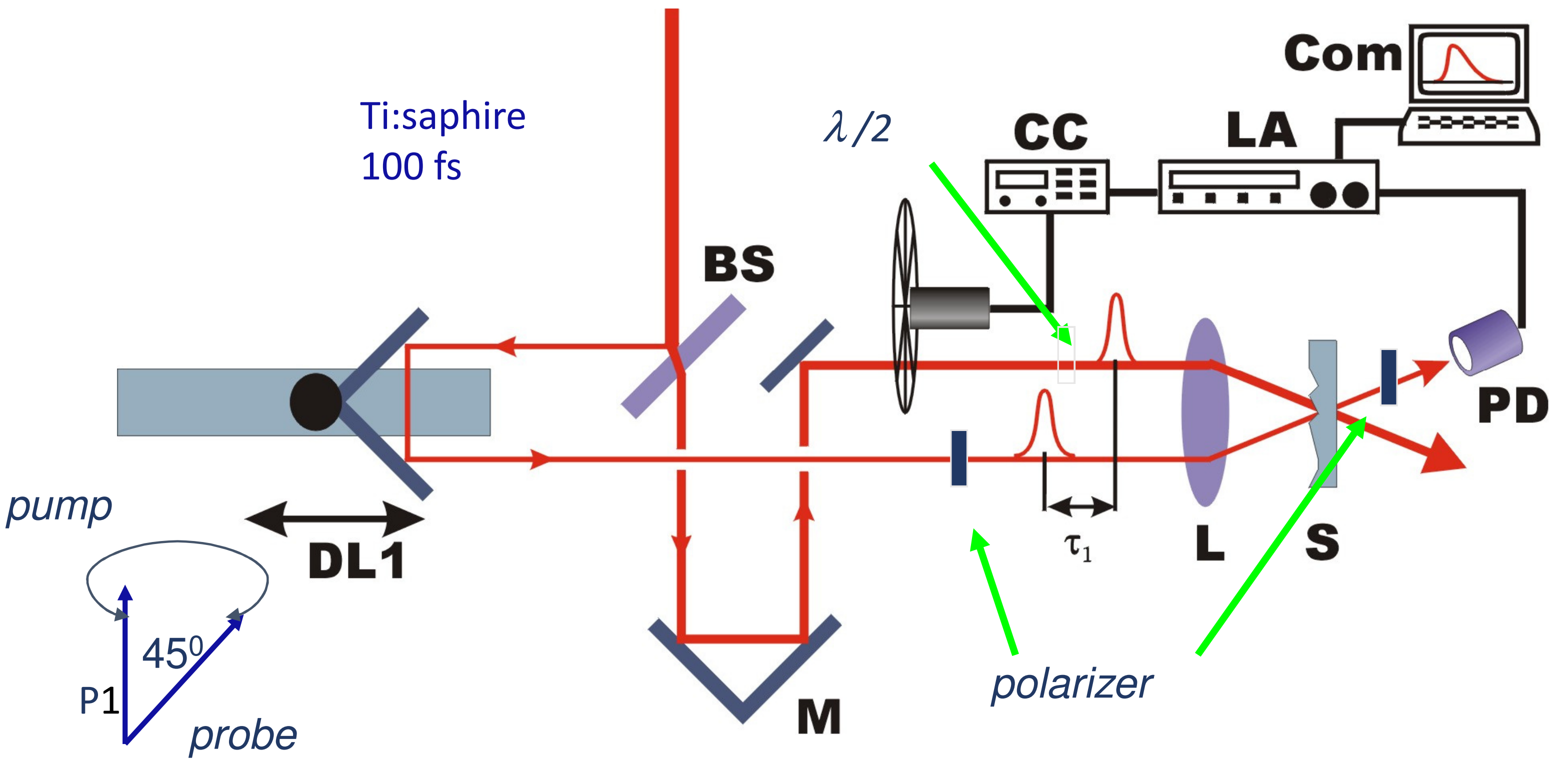
TM- EZ scan



Kerr shutter setup
Dynamics of the nonlinearity

$\chi^{(3)}_{ijkl}(t)$

Pump beam induces birrefringence



Kerr shutter

Isotropic medium

$$\vec{E}_{exc} = E_{exc}\,\hat{x}$$

$$\vec{E}_{prova} = \sqrt{2}\,E_{prova}\,(\hat{x}+\hat{y})/2$$

$$P_x^{(3)} = \chi^{(3)}_{xxxx}|E_{exc}|^2 E_{prova,\,x}$$

$$P_y^{(3)} = \chi^{(3)}_{yxxy}|E_{exc}|^2 E_{prova,\,y}$$

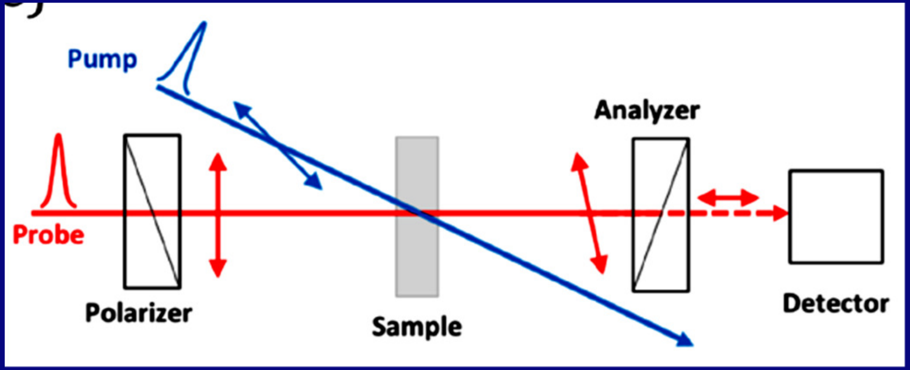
$$\Delta n_x = \frac{2\pi}{n_0}\chi^{(3)}_{xxxx}|E_{exc}|^2$$

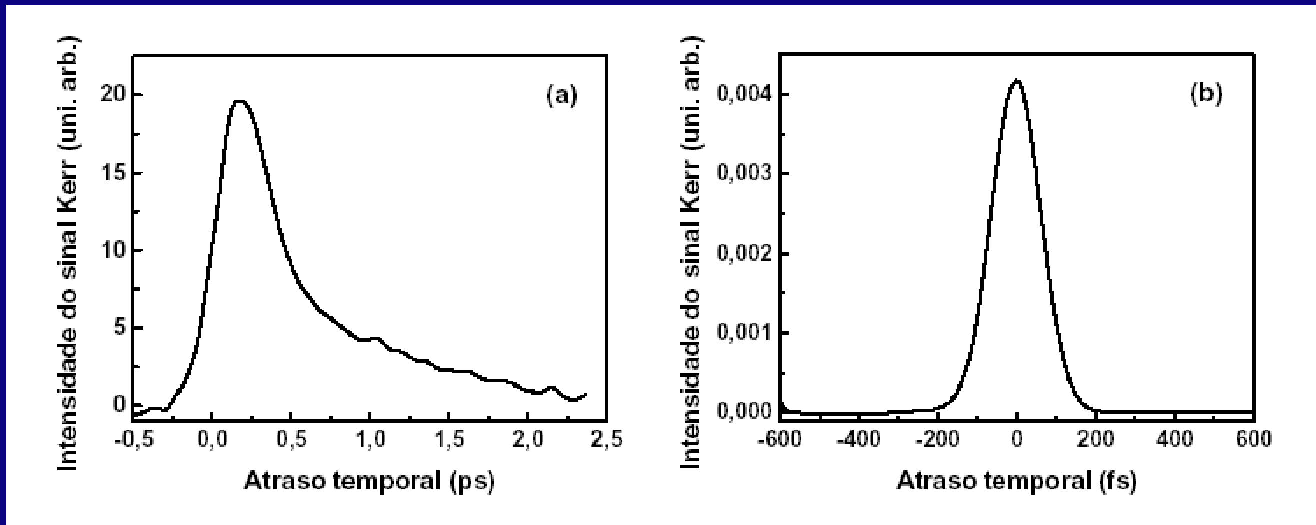
$$\Delta n_y = \frac{2\pi}{n_0}\chi^{(3)}_{yxxy}|E_{exc}|^2$$

$$\Delta n_{NL} = \Delta n_x - \Delta n_y = \frac{4\pi}{3n_0}\chi^{(3)}_{xxxx}|E_{exc}|^2$$

$$\vec{E}_{prova}(L) = \frac{\sqrt{2}}{2}E_{prova}\left[\hat{x}+\hat{y}\exp(-i\,\Delta\phi_{NL})\right]$$

$$\Delta\phi_{NL} = k\,L\,\Delta n_{NL}$$



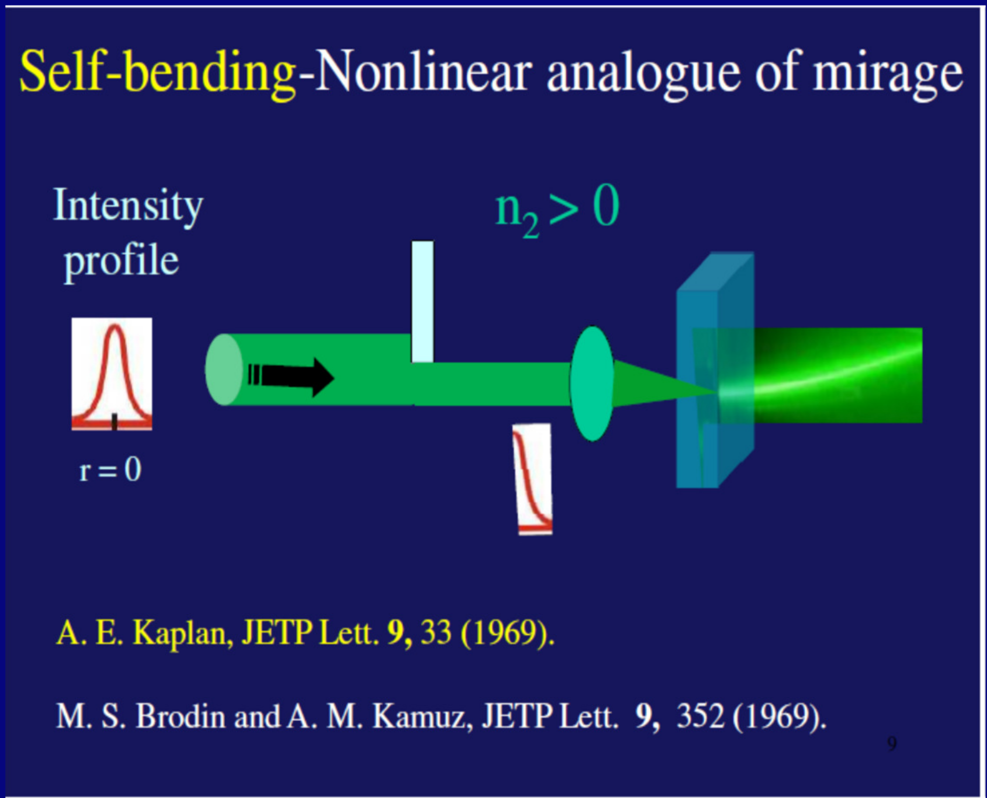


Carbon disulfide CS_2

Silica (Fused SiO_2)

Electronic <50 fs
Reorientational > 2ps

Electronic polarization
Faster than the laser pulse duration

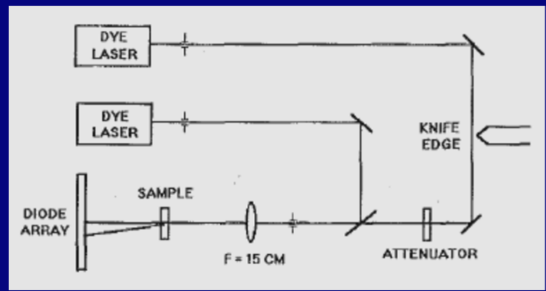


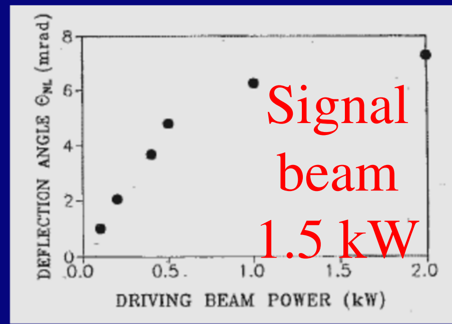
Cross-bending

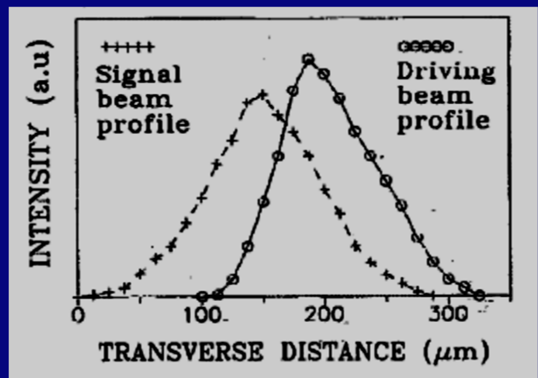
Appl. Phys. Lett. 63 (1993) 3553

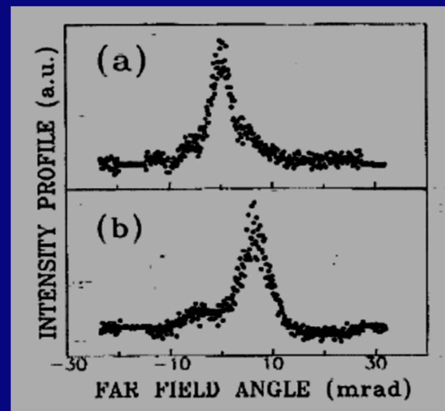
Light-controlled beam deflector in semiconductor doped glasses

H. Ma and Cid B. de Araújo
Departamento de Física, Universidade Federal de Pernambuco, 50732-910 Recife, PE, Brazil







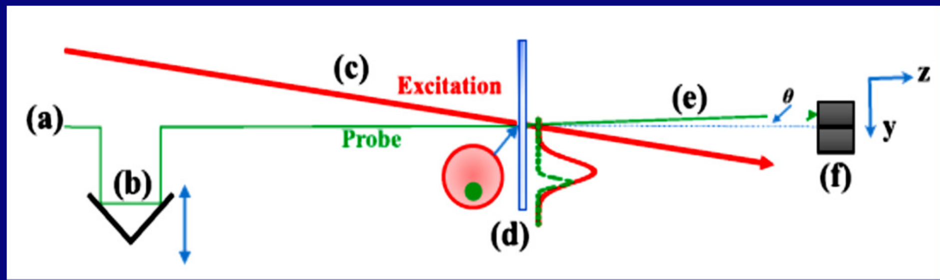


Driving beam peak power: 1.5 kW

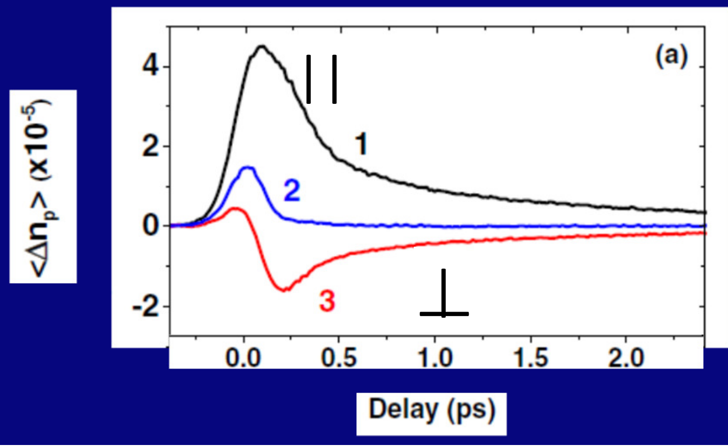
Opt. Lett. 38 (2013) 3518

Beam deflection measurement of time and polarization resolved ultrafast nonlinear refraction

Manuel R. Ferdinandus,[1] Honghua Hu,[1] Matthew Reichert,[1] David J. Hagan,[1,2] and Eric W. Van Stryland[1,2,*]



$$\Delta n_p(x,y,t) = \Delta n_p(t)\exp\left(\frac{-2(x^2+y^2)}{w_e^2}\right)$$



For more details and references for several NL techniques see for example:

IOP Publishing
Rep. Prog. Phys. 79 (2016) 036401 (30pp)

Review
Techniques for nonlinear optical characterization of materials: a review

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

Four wave-mixing
Phase conjugation
Kerr gate

Scattered Light
Imaging Method –SLIM

Pump-and-probe

Beam deflection
techniques

Single beam Z-scan

Two-color Z-scan

Reflection Z-scan
Eclipsing Z-scan
Thermally managed
TM-Z scan

TM-EZ scan

Hartmann - Schack Z-scan

White-light continuum Z-scan

Intensity scan (I-scan)

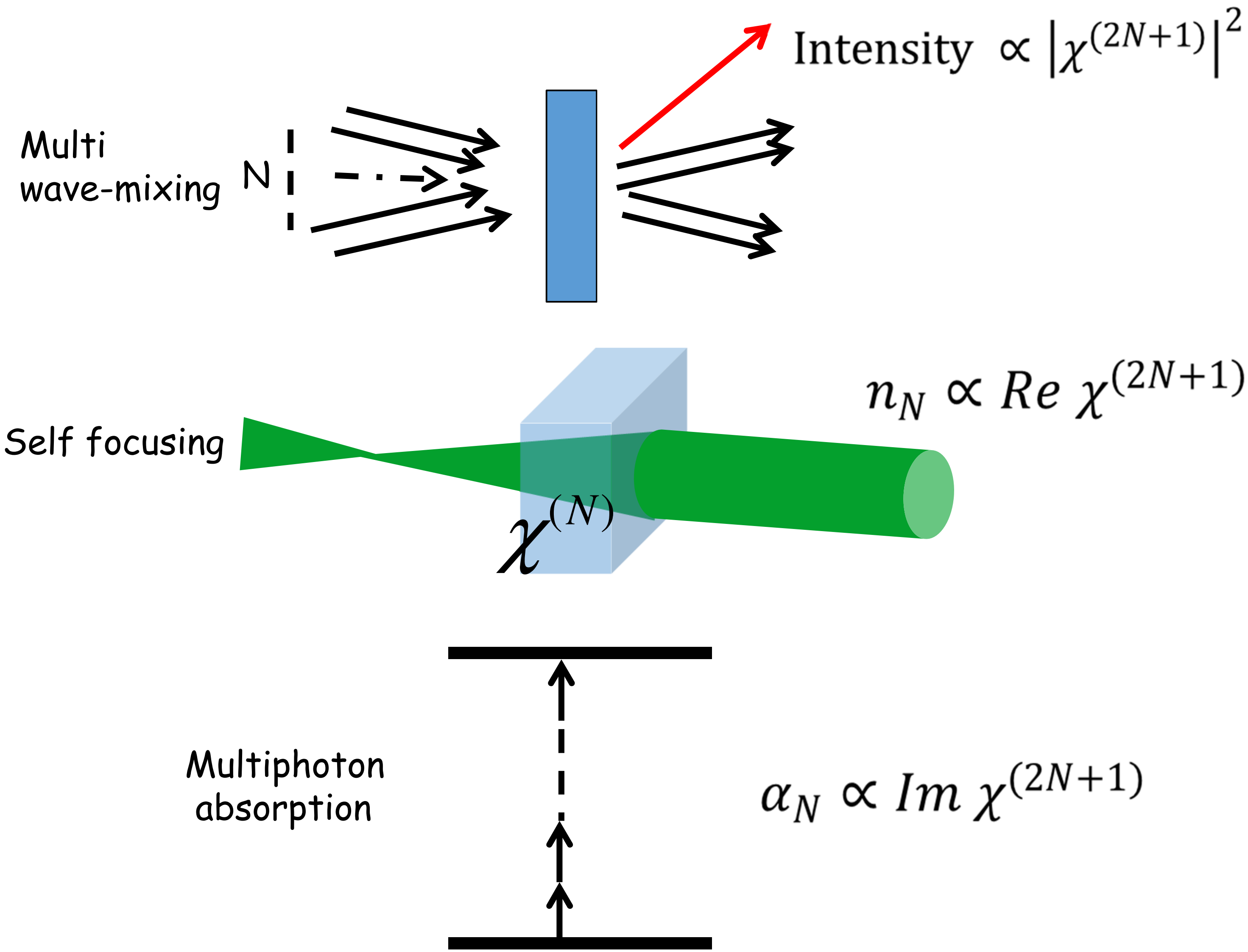
High-order nonlinearities

- Generation of higher harmonics
- Multiphoton excitation processes
- Multi-wave mixing

Studies of multiphoton ionization: 60´s; Many studies in the 70´s, 80´s.

Multiphoton dissociation and ionization processes applied to isotope separation (70´s and 80´s).

More recently:
Solitons, filamentation, extreme events, generation of VUV and soft X-rays – 400th harmonic, proposals for attosecond X-ray pulse generation ....



Multi
wave-mixing
N
Intensity $\propto \left|\chi^{(2N+1)}\right|^2$
Self focusing
$\chi^{(N)}$
$n_N \propto Re\ \chi^{(2N+1)}$
Multiphoton
absorption
$\alpha_N \propto Im\ \chi^{(2N+1)}$

Normally high-order nonlinearities (HON) are weaker than low-order effects

But HON may be due to repeated low-order susceptibilities

cascading processes

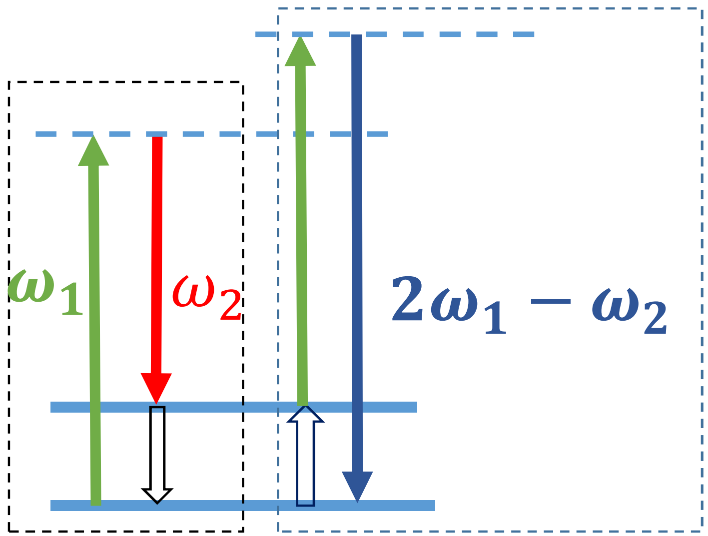
Macroscopic cascading: involves propagation effects

Example: $\omega + \omega$ creates 2ω then $2\omega + \omega$ creates 3ω

Microscopic cascading:

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

Meio sem centro de inversão $\chi^{(2)} \neq 0$

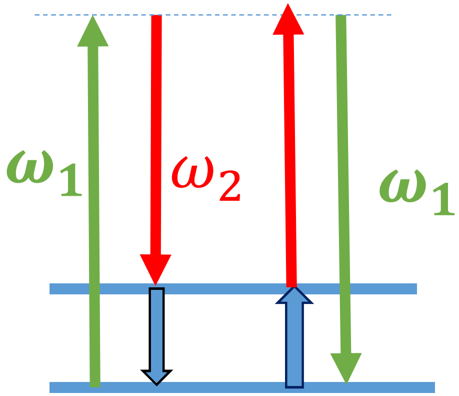


$$E(\omega_1 - \omega_2) \propto \chi^{(2)}(\omega_1 - \omega_2, \omega_1, -\omega_2)E(\omega_1)E^*(\omega_2)$$

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

$$\mathbf{E}(2\omega_1 - \omega_2) \propto \underbrace{\chi^{(2)}(2\omega_1 - \omega_2, \omega_1 - \omega_2, \omega_1)\chi^{(2)}(\omega_1 - \omega_2, \omega_1, -\omega_2)}_{\chi_{eff}^{(3)}}E(\omega_1)E(\omega_1)E^*(\omega_2)$$

Outra possibilidade



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

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

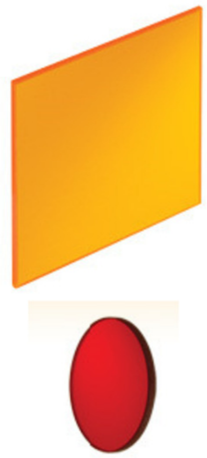
$$\boldsymbol{E}(\omega_1) \propto \boldsymbol{\chi}^{(2)}(\omega_1, \omega_1 - \omega_2, \omega_2)\boldsymbol{E}(\omega_1 - \omega_2)\boldsymbol{E}(\omega_2)$$

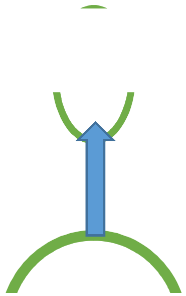
$$\boldsymbol{E}(\omega_1) \propto \boldsymbol{\chi}^{(2)}(\omega_1, \omega_1 - \omega_2, \omega_2)\boldsymbol{\chi}^{(2)}(\omega_1 - \omega_2, \omega_1, -\omega_2)\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)\boldsymbol{E}^*(\omega_2)$$

$$\chi^{(3)}_{eff} \propto \left[\chi^{(2)}\right]^2$$

Semiconductor doped glasses

Nanocrystals of CdS_xSe_{1-x}





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

Rep. Prog. Phys. **79** (2016) 036401 Rev

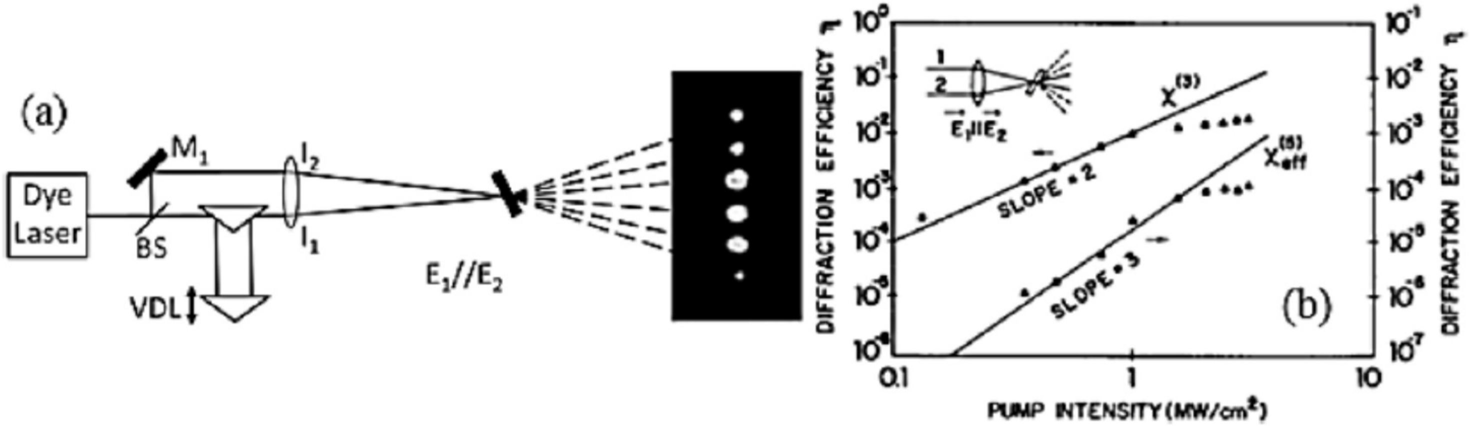
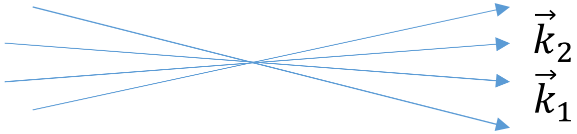
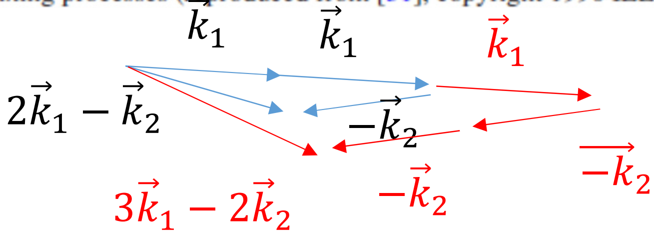
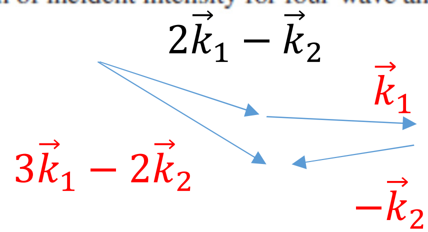


Figure 2. (a) Typical experimental setup for studying forward DFWM. The photographic image shows the two transmitted beams in the center and self-diffracted orders on each side; (b) measured diffraction efficiency as a function of incident intensity for four-wave and effective six-wave mixing processes (reproduced from [31], copyright 1990 IEEE).





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



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

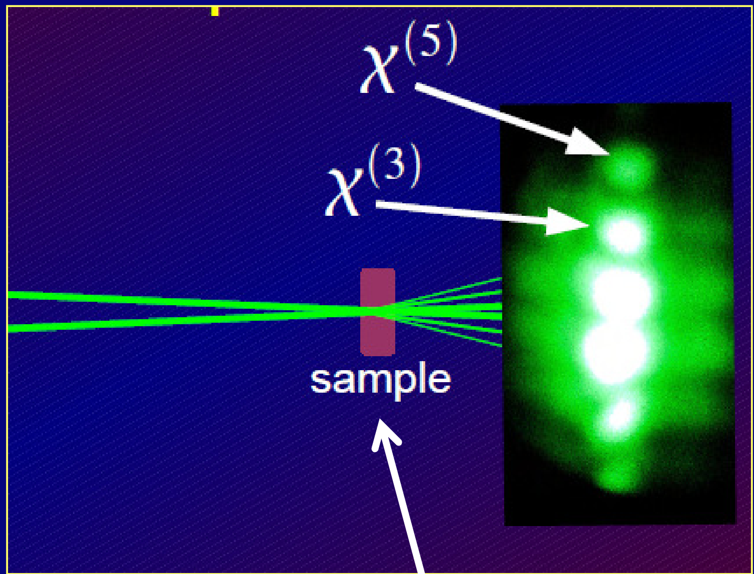
$$\chi^{(5)}_{eff} = A\chi^{(3)}.\chi^{(3)} + B\chi^{(5)}$$

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

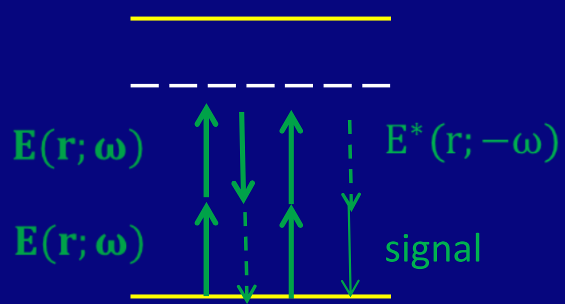
Observation of a Microscopic Cascaded Contribution to the Fifth-Order Nonlinear Susceptibility

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

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



Cascade - $\chi^{(3)}:\chi^{(3)}$



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

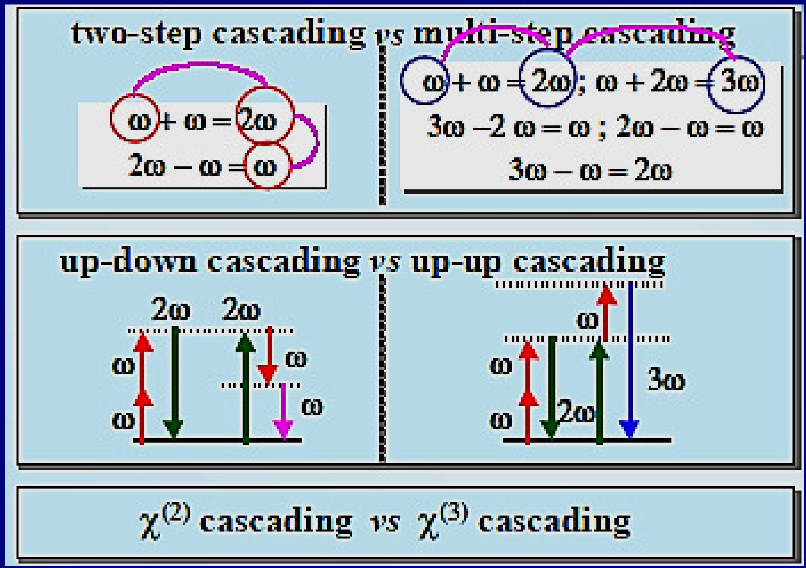
$$L = \frac{\varepsilon^{(1)} + 2}{3}$$

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 hyperpolarizability



Under certain conditions, the cascaded contribution can be as large as the direct contribution

Cascaded NL optical processes play important role

- in frequency upconversion
- in optical communications: for routing, switching, information interchange
- for studying fundamental constants of the materials
- for mode-locking and pulse compression

However it is possible determination of pure NL susceptibilities using phase-matching configurations as described in the following papers

Volume 51, number 2 OPTICS COMMUNICATIONS 15 August 1984

DIRECT OBSERVATION OF HIGH-ORDER OPTICAL SUSCEPTIBILITIES VIA ANGULARLY-RESOLVED MULTIWAVE MIXING

R.K. RAJ, Q.F. GAO *, D. BLOCH and M. DUCLOY

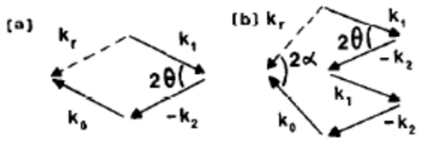


Fig. 1. Wavevector configuration for (a) backward four-wave mixing ($k_0 = -k_1$; $k_r = -k_2$), (b) phase-matched six-wave mixing in which two photons 1 are absorbed and two photons 2 emitted.

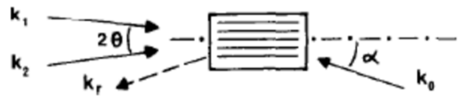


Fig. 3. Grating viewpoint of resonant multiwave mixing. The nth-order Bragg condition is given by $\sin\alpha = n \sin\theta$.

$\chi^{(5)}$: $\quad sen\, \alpha = 2\, sen\, \theta$

$\chi^{(2n+1)}$: $\quad sen\, \alpha = n\, sen\, \theta$

Optics Communications 100 (1993) 193–196

Cascade contributions in the high-order optical nonlinearity measurement

H. Ma, A.S.L. Gomes and Cid B. de Araujo

Phase-matching multi-wave mixing

Unambigous measurement of high-order susceptibilities

Rep. Prog. Phys. **79** (2016) 036401 Review

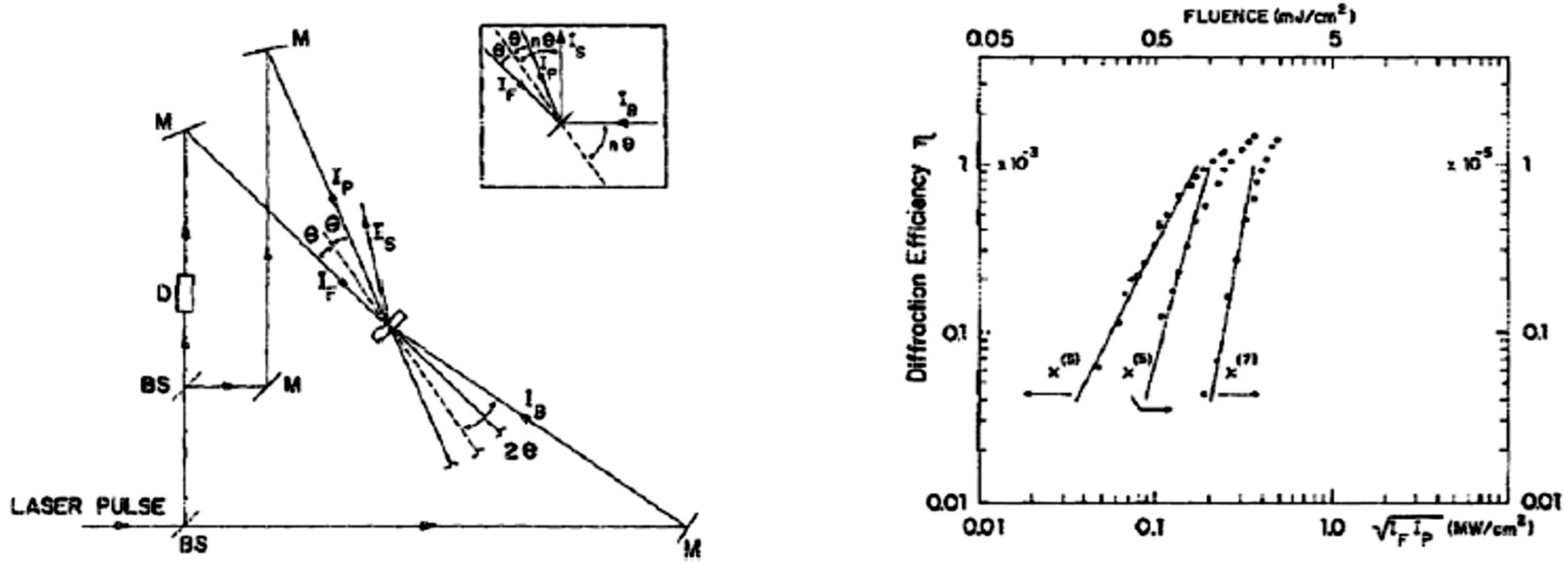
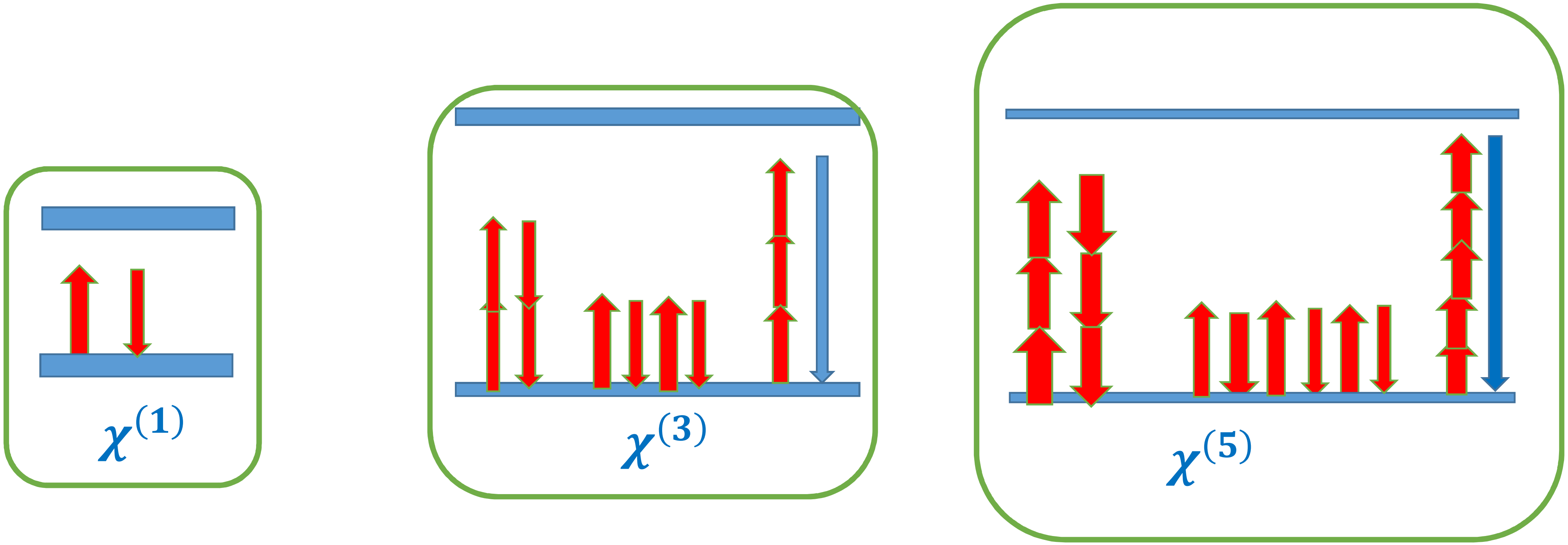


Figure 3. (a) Experimental scheme for unambiguous measurement of phase-matched multi wave-mixing; (b) measured diffraction efficiencies for $|\chi^{(3)}|$, $|\chi^{(5)}|$ and $|\chi^{(7)}|$ in SDG (reproduced from [50], copyright 1988 AIP).

Exemples of pure optical processes



$$P^{(1)} = \epsilon_0 \chi^{(1)} E(\omega)$$

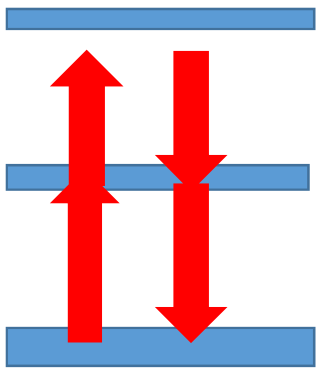
$$P^{(3)} = \epsilon_0 \chi^{(3)} [E(\omega)]^2 E^*(\omega)$$

$$P^{(5)} = \epsilon_0 \chi^{(5)} [E(\omega)]^3 [E^*(\omega)]^2$$

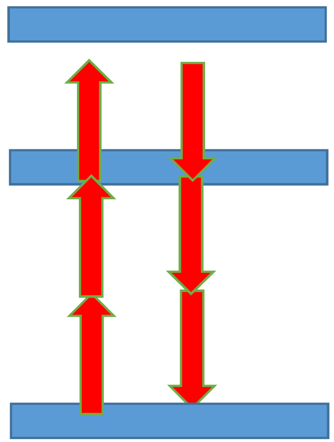
$$P^{(3)} = \epsilon_0 \chi^{(3)} [E(\omega)]^3$$

$$P^{(5)} = \epsilon_0 \chi^{(5)} [E(\omega)]^5$$

Examples of more cascade processes



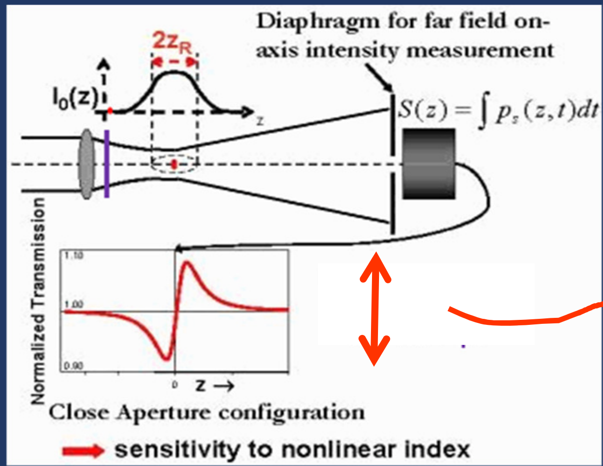
$$\chi^{(1)} \cdot \chi^{(1)} \equiv \chi^{(3)}$$



$$\chi^{(3)} \cdot \chi^{(1)} \equiv \chi^{(5)}$$

When high-order nonlinearities are present:

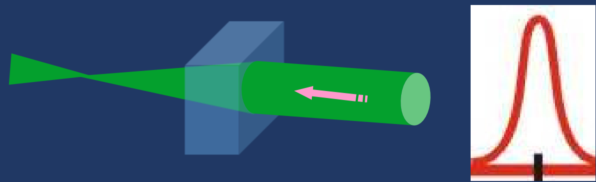
Self – focusing medium



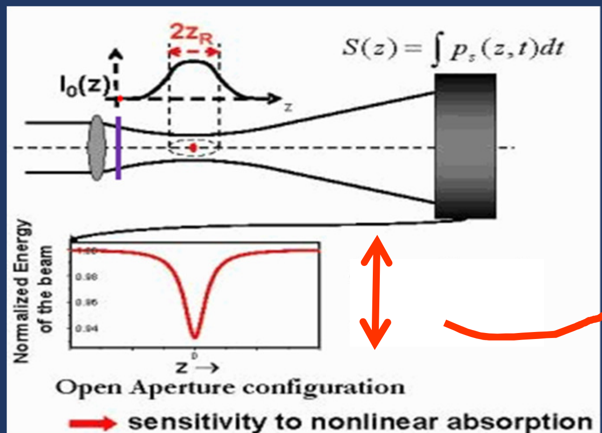
NL refraction

"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

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

Particularly 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.

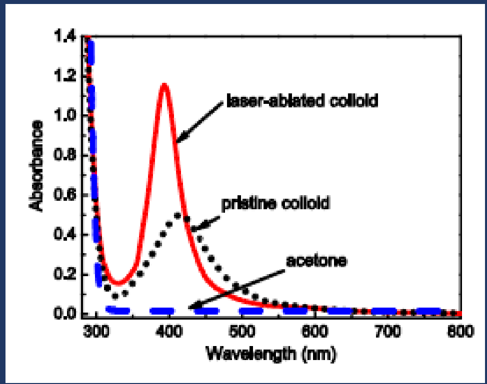
It means that we may, for instance, to adjust the material’s parameters to supress one particular NL susceptibility and enhance the others.

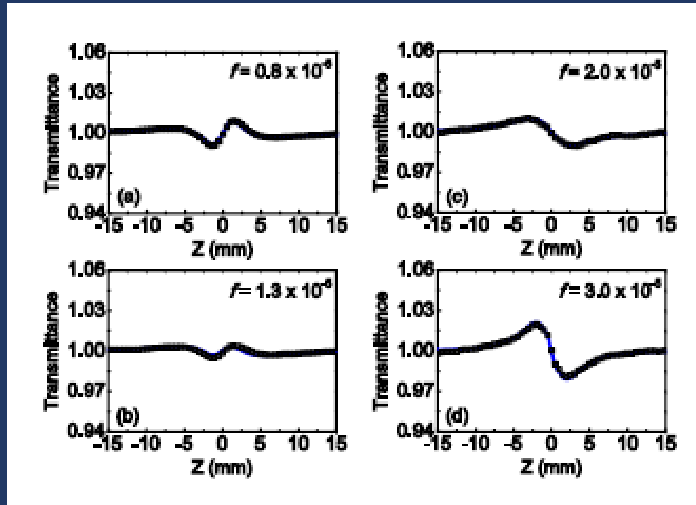
For example, one may supress the third-order refractive index such that the refraction can be dominate by the fifth-order susceptibility

This possibility is illustrated in the next slide where n_2 is canceled but n_4 is not nulled. This nonlinearity management procedure will be the subject presented in the next class.

Observation of fifth-order refraction in a colloid with supressed third-order refraction

Silver NPs in acetone

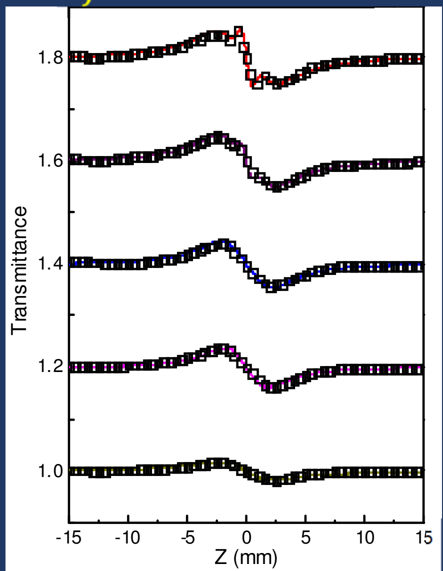




Z-scan

532 nm
Single pulses
5 GW/cm^2

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



9

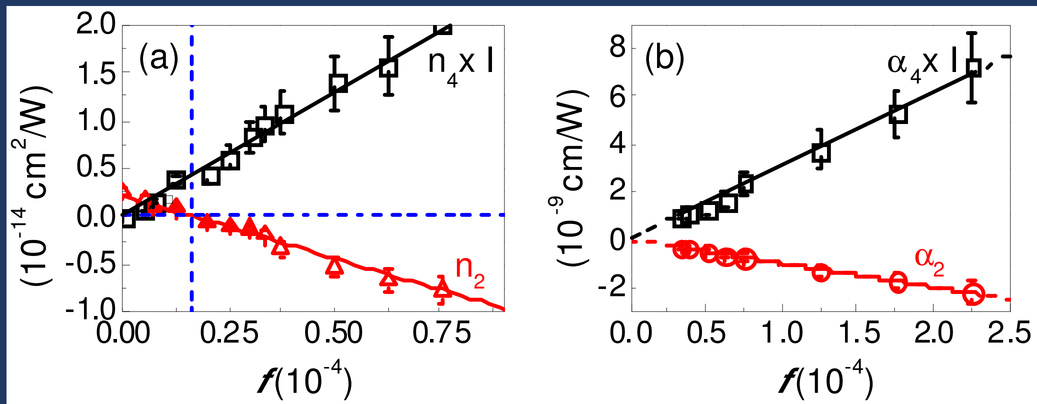
8

6

4

2

GW/cm^2



9 GW/cm^2