Introduction to Nonlinear Optics

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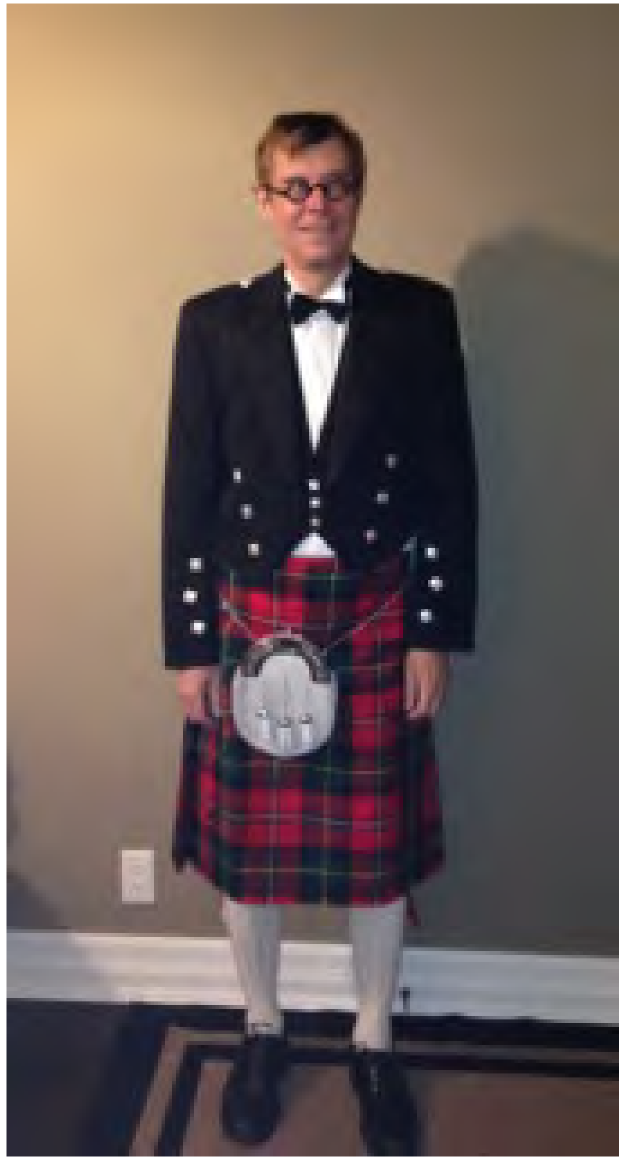
Course 190 - Frontiers in Modern Optics, 30 June - 5 July, 2014.

Outline of Presentation

Part I. Tutorial Introduction to Nonlinear Optics

Part II. Recent Research in Quantum Nonlinear Optics



Why Study Nonlinear Optics?

It is good fundamental physics.

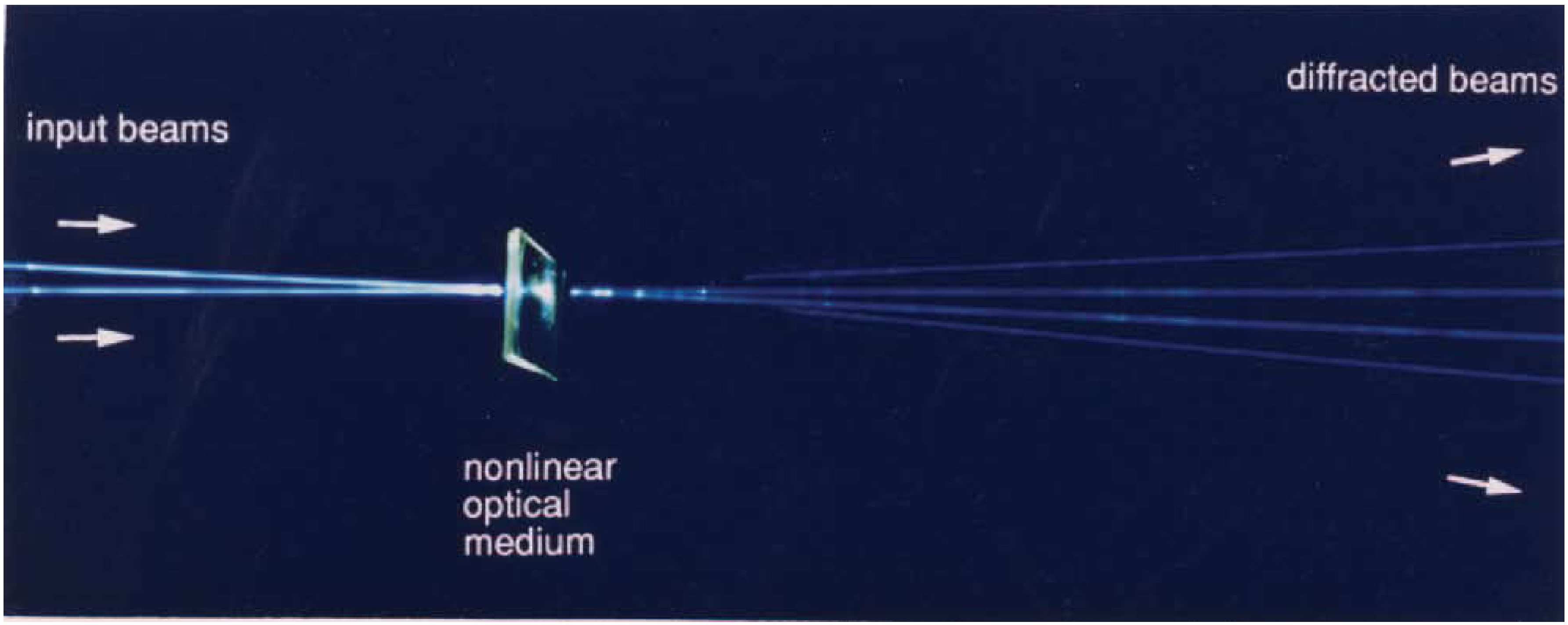
It leads to important applications.

It is a lot of fun.

Demonstrate these features with examples in remainder of talk.

1. What is Nonlinear Optics?

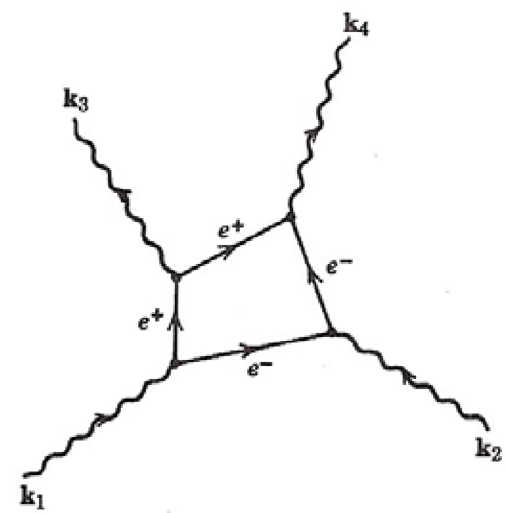
Nonlinear Optics and Light-by-Light Scattering



The elementary process of light-by-light scattering has never been observed in vacuum, but is readily observed using the nonlinear response of material systems.

Nonlinear material is fluorescein-doped boric acid glass (FBAG)

n_2 (FBAG) ≈ 10^{14} n_2(silica)   [But very slow response!]



M. A. Kramer, W. R. Tompkin, and R. W. Boyd, Phys. Rev. A, 34, 2026, 1986.
W. R. Tompkin, M. S. Malcuit, and R. W. Boyd, Applied Optics 29, 3921, 1990.

Nonlinear Optics

THIRD EDITION

Robert W. Boyd

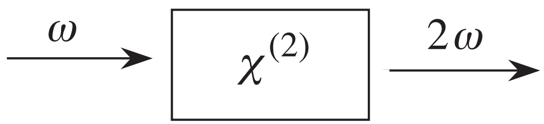
Simple Formulation of the Theory of Nonlinear Optics

$$P = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \ldots$$

Here P is the induced dipole moment per unit volume and E is the field amplitude

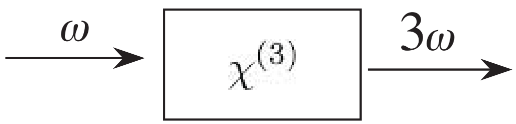
$\chi^{(1)}$ describes linear optics, e.g., how lenses work: 

$\chi^{(2)}$ describes second-order effects, e.g., second-harmonic generation (SHG)

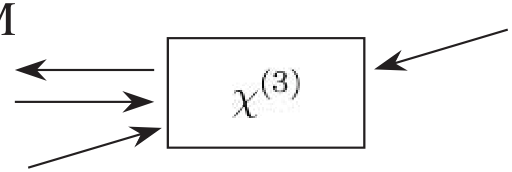


$\chi^{(3)}$ describes third-order effects such as third-harmonic generation, four-wave mixing, and the intensity dependence of the index of refraction.

THG



FWM

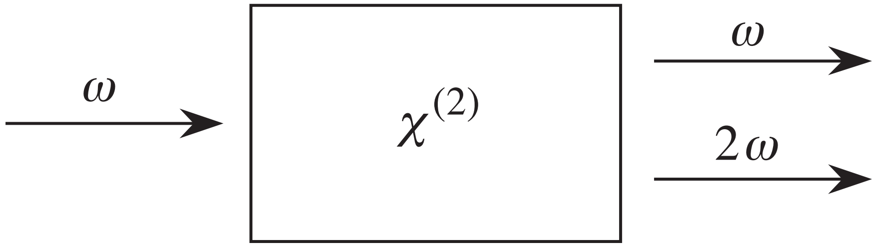


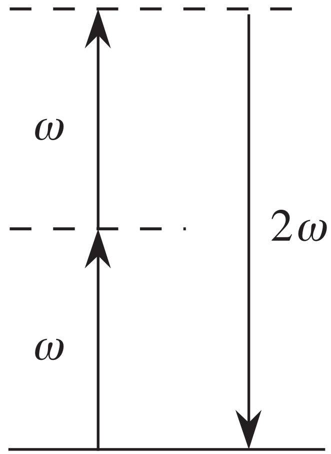
NL index

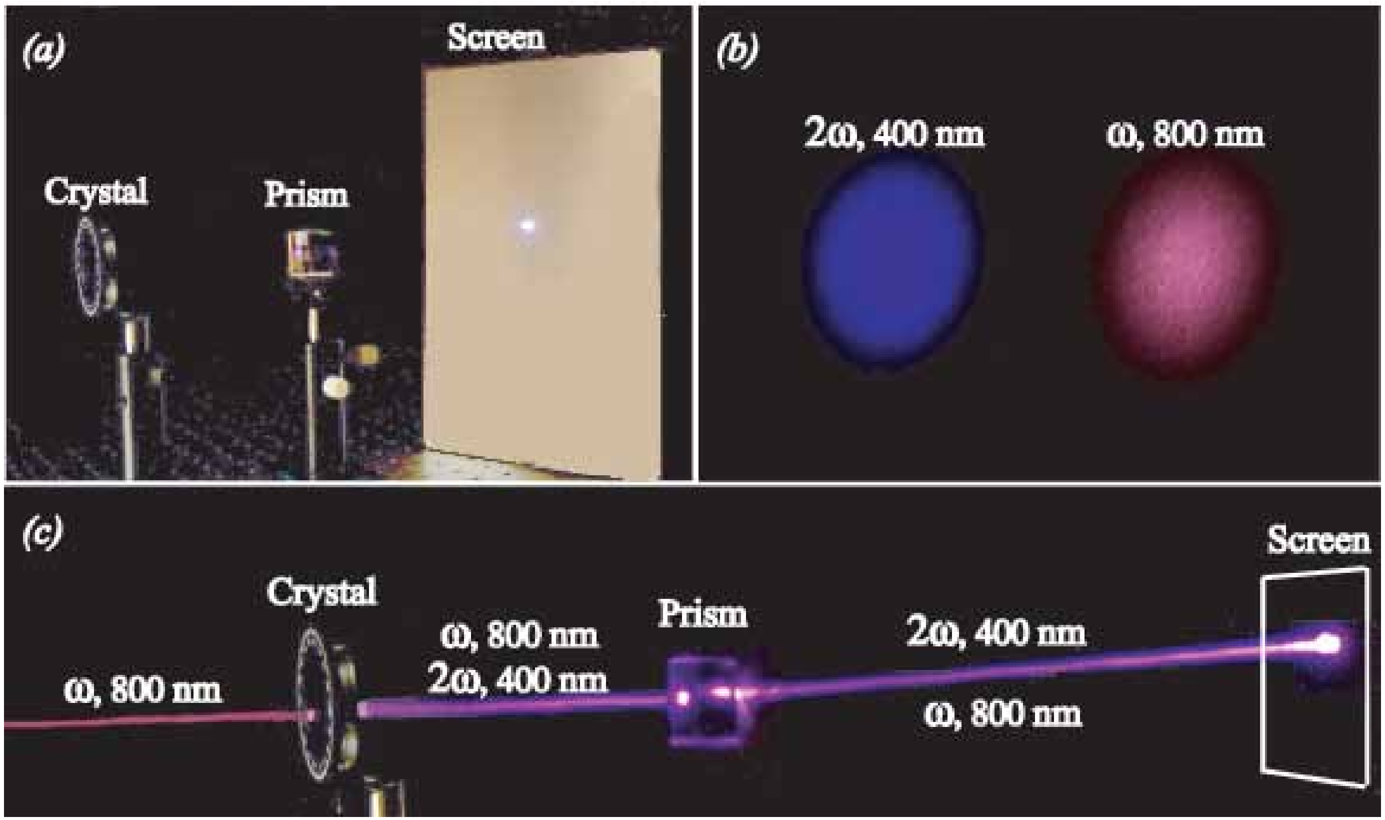
$$n = n_0 + n_2 I \quad \text{where} \quad n_2 = \frac{3}{4n_0^2\epsilon_0 c}\chi^{(3)}$$

Some Fundamental Nonlinear Optical Processes: I

Second-Harmonic Generation



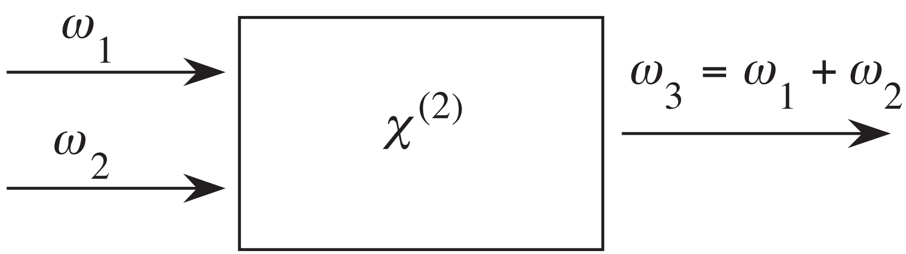


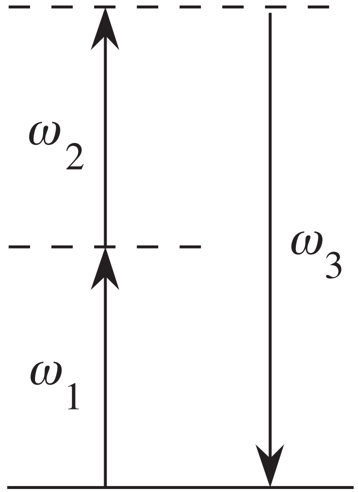


Dolgaleva, Lepeshkin, and Boyd

Some Fundamental Nonlinear Optical Processes: II

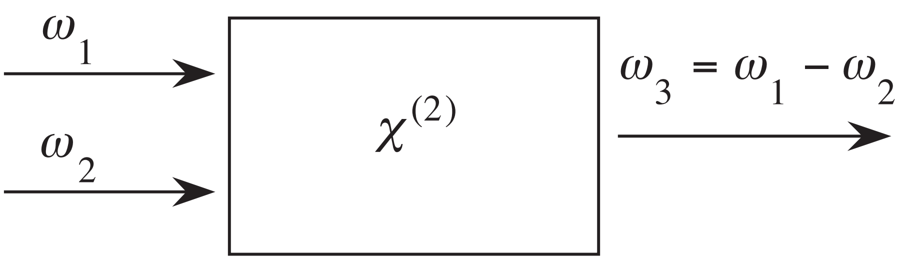
Sum-Frequency Generation

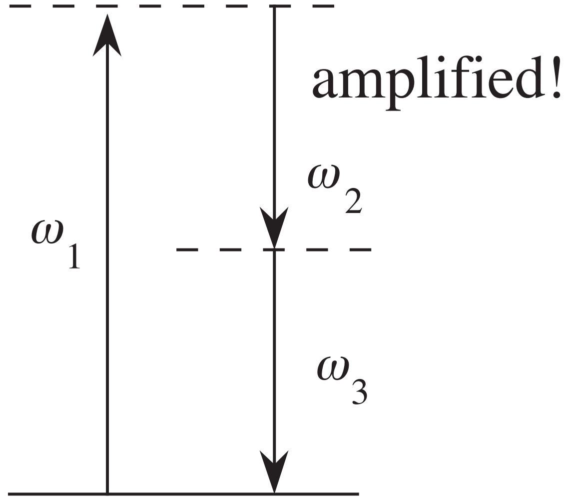




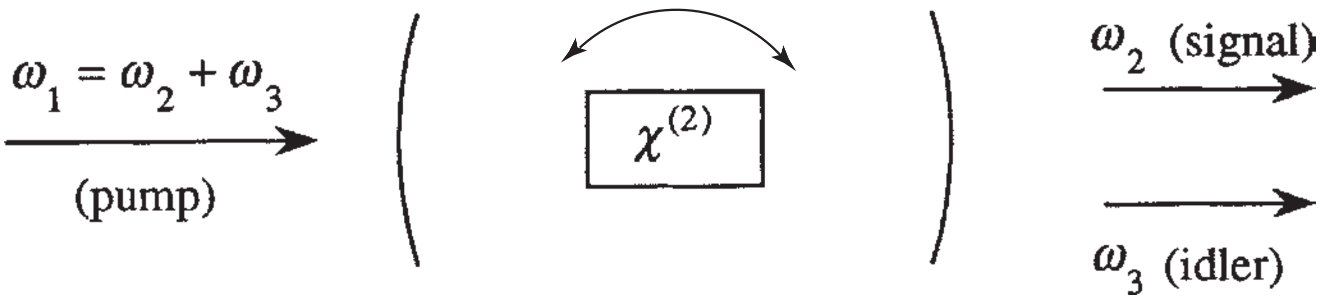
Some Fundamental Nonlinear Optical Processes: III

Difference-Frequency Generation

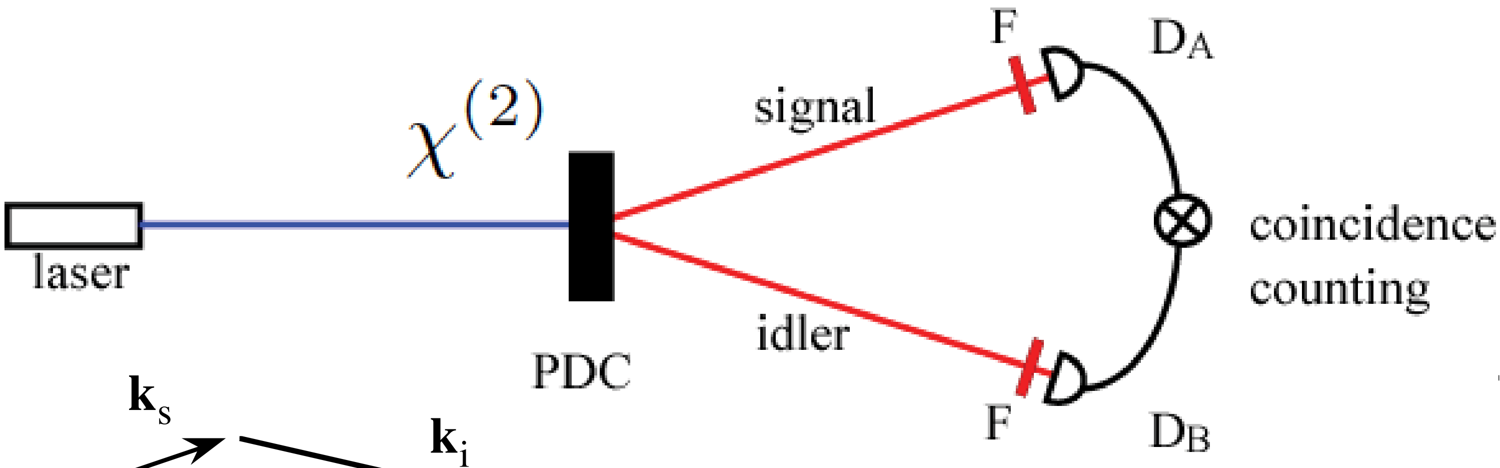


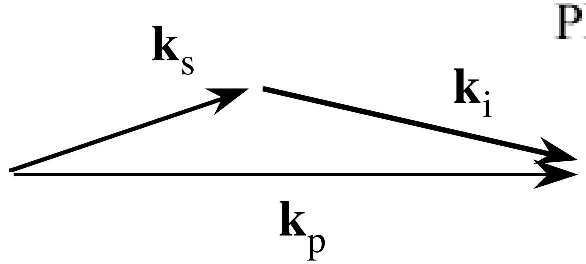


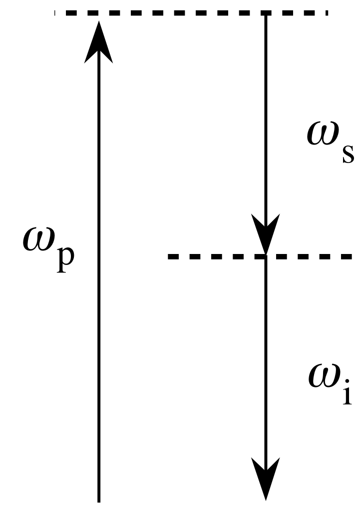
Optical Parametric Oscillation



Parametric Downconversion: A Source of Entangled Photons







The signal and idler photons are entangled in:

(a) polarization

(b) time and energy

(c) position and transverse momentum

(d) angular position and orbital angular momentum

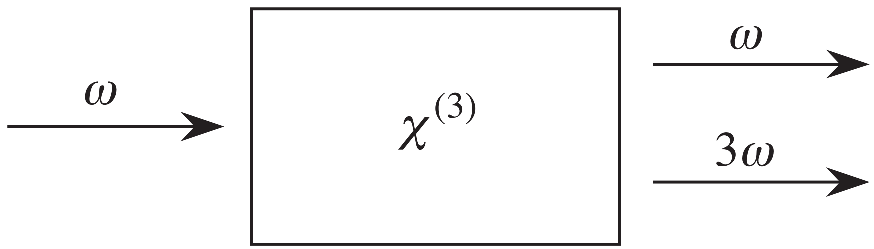
Entanglement is important for:

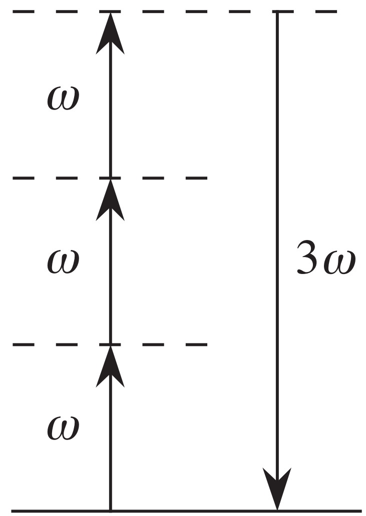
(a) Fundamental tests of QM (e.g., nonlocality)

(a) Quantum technologies (e.g., secure communications)

Some Fundamental Nonlinear Optical Processes: III

Third-Harmonic Generation

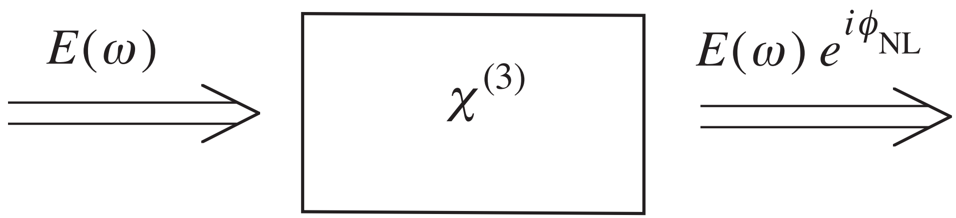




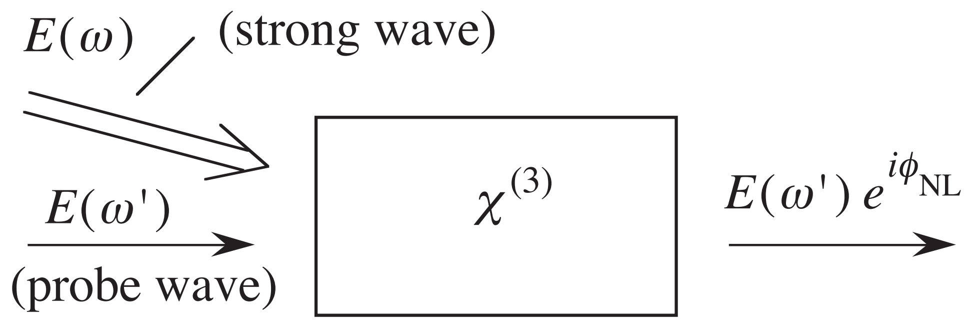
Some Fundamental Nonlinear Optical Processes: IV

Intensity-Dependent Index of Refraction

- Single beam:
  self-phase modulation



- Two beam:
  cross-phase modulation



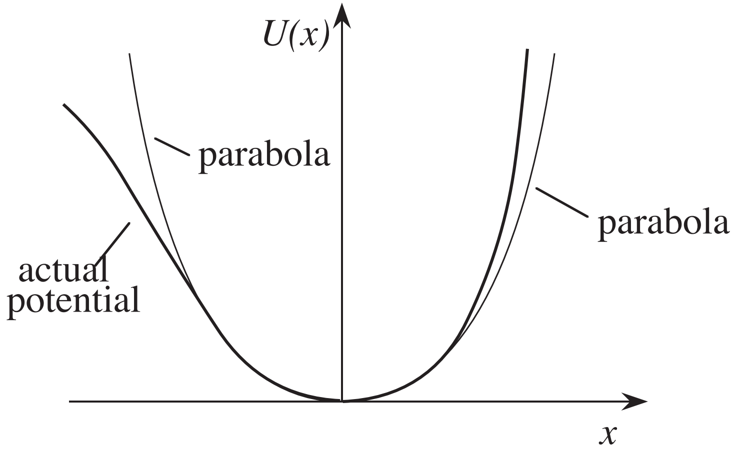
The refractive index is given by

$$n = n_0 + n_2 I \qquad \text{where} \qquad n_2 = \frac{3}{4 n_0^2 \epsilon_0 c} \chi^{(3)}$$

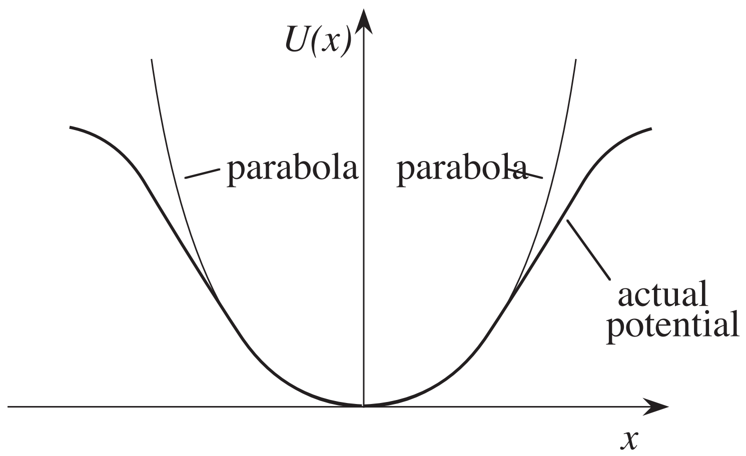
Role of Material Symmetry in Nonlinear Optics

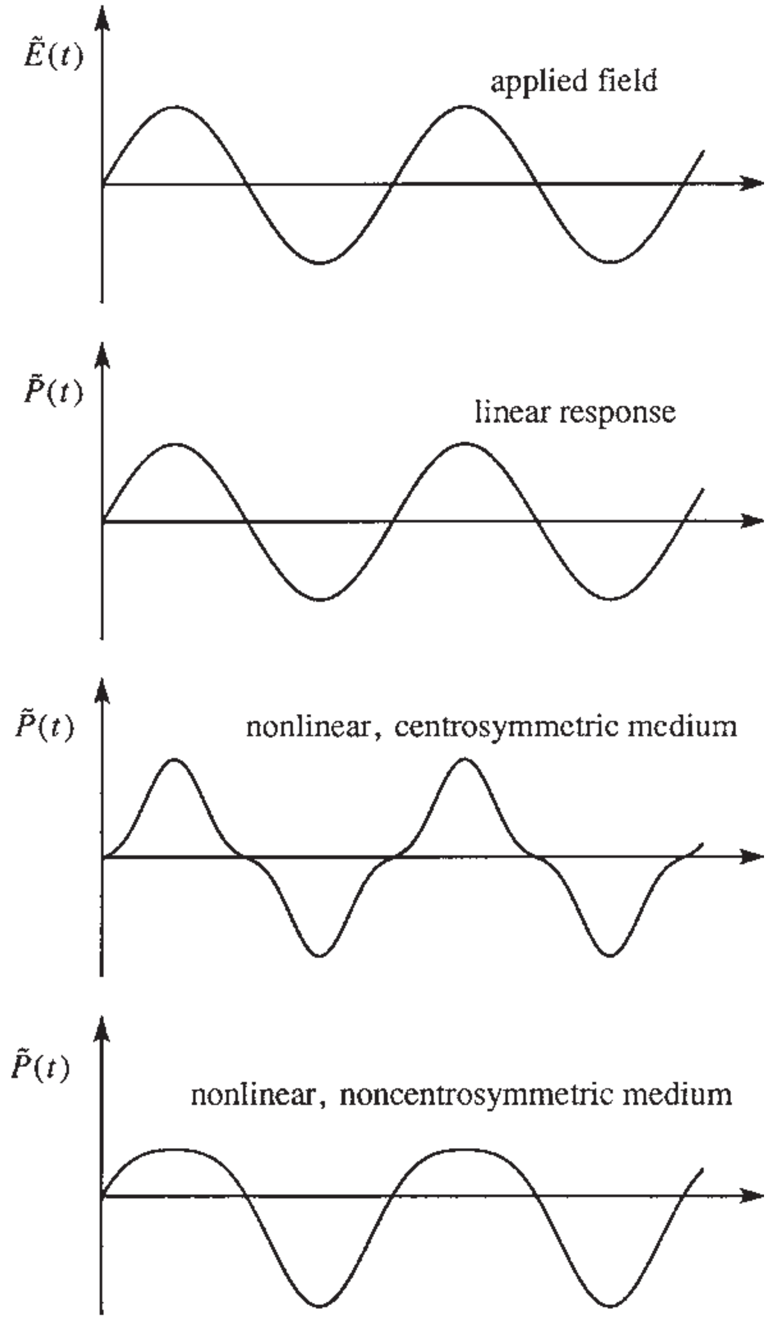
$\chi^{(2)}$ vanishes identically for a material possessing a center of inversion symmetry (a centrosymmetric medium).

non-centrosymmetric medium



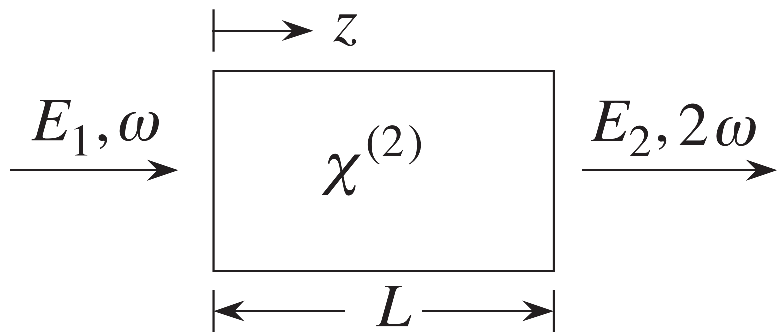
centrosymmetric medium





2. Coupled Wave Equations and Harmonic Generation

Treatment of Second-Harmonic Generation – I



Let

$$\tilde{E}_1(z,t) = E_1(z)e^{-i\omega t} + \text{c.c.} = A_1 e^{i(k_1 z - \omega t)} + \text{c.c.} \tag{1}$$

$$\tilde{E}_2(z,t) = E_2(z)e^{-i2\omega t} + \text{c.c.} = A_2(z) e^{i(k_2 z - 2\omega t)} + \text{c.c.} \tag{2}$$

where $k_1 = n_1\omega/c$ and $k_2 = n_2 2\omega/c$.

We have assumed that the pump wave E_1 at frequency ω is undepleted by the nonlinear interaction. We take A_2 to be a function of z to allow the second harmonic wave to grow with z. We also set

$$\tilde{P}_2(t) = P_2\, e^{-i2\omega t} \quad \text{where} \quad P_2 = \epsilon_0 \chi^{(2)} E_1^2 = \epsilon_0 \chi^{(2)} A_1^2\, e^{i2k_1 z} \tag{3}$$

The generation of the wave at 2ω is governed by the wave equation

$$\nabla^2 \tilde{E}_2 - \frac{n^2}{c^2}\frac{\partial^2 \tilde{E}_2}{\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2 \tilde{P}_2}{\partial t^2}. \tag{4}$$

Treatment of Second-Harmonic Generation – II

Note that the first term in the wave equation is given by

$$\nabla^2 \tilde{E}_2 = \left[\frac{\partial^2 \tilde{A}_2}{\partial z^2} + 2ik_2 \frac{\partial \tilde{A}_2}{\partial z} - k_2^2 A_2\right] e^{i(k_2 z - 2\omega t)} \tag{5}$$

$$\approx \left[2ik_2 \frac{\partial \tilde{A}_2}{\partial z} - k_2^2 A_2\right] e^{i(k_2 z - 2\omega t)} \tag{6}$$

The second form is the *slowly varying amplitude approximation.*

Note also that

$$\frac{\partial^2 \tilde{A}_2}{\partial t^2} = -(2\omega)^2 A_2\, e^{i(k_2 z - 2\omega t)} \quad \frac{\partial^2 \tilde{P}_2}{\partial t^2} = -(2\omega)^2 P_2\, e^{i(2k_1 z - 2\omega t)} \tag{7}$$

By combining the above equations, we obtain

$$2ik_2 \frac{dA_2}{dz} = \frac{-4\omega^2}{c^2} \chi^{(2)} A_1^2 e^{i\Delta k\, z} \quad \text{where} \quad \Delta k = 2k_1 - k_2. \tag{8}$$

The quantity Δk is known as the phase (or wavevector) mismatch factor, and it is crucially important in determining the efficiency of nonlinear optical interactions

Treatment of Second-Harmonic Generation – III

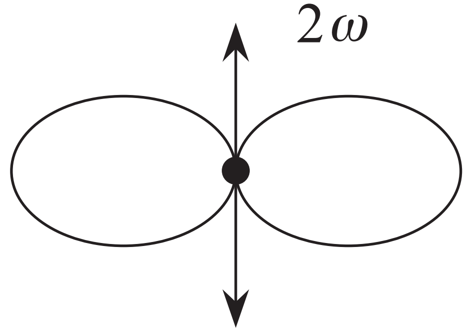
For the case $\Delta k = 0$, Eq. (8) becomes

$$2ik_2 \frac{dA_2}{dz} = \frac{-4\omega^2}{c^2} \chi^{(2)} A_1^2 \tag{9}$$

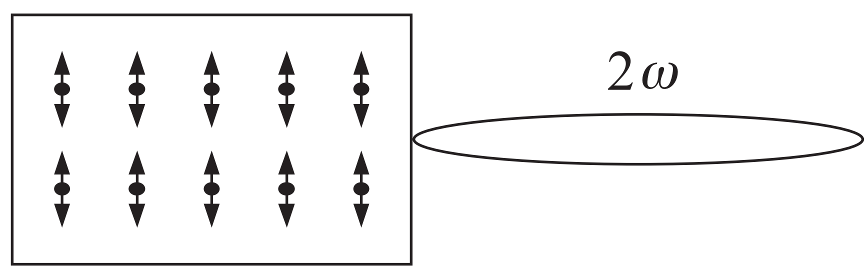
with solution evaluated at $z = L$ of

$$A_2(L) = \frac{2i\omega}{n_2 c} \chi^{(2)} A_1^2 L \quad \text{or} \quad |A_2(L)|^2 = \frac{4\omega^2}{n_2^2 c^2} [\chi^{(2)}]^2 |A_1|^4 L^2. \tag{10}$$

Note that the SHG intensity scales as the square of the input intensity and also as the square of the length L of the crystal.



dipole emitter



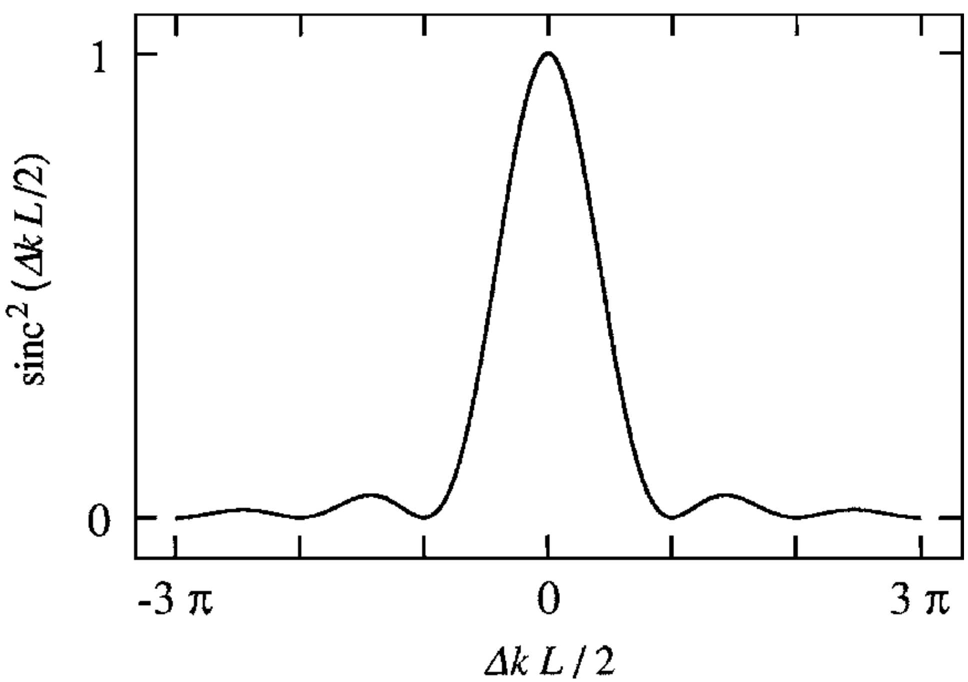
phased array of dipoles

Treatment of Second-Harmonic Generation – IV

For the general case of $\Delta k \neq 0$, Eq. (8) can still be solved to yield

$$|A_2(L)|^2 = \frac{4\omega^2}{n_2^2 c^2} \, [\chi^{(2)}]^2 \, |A_1|^4 L^2 \, \mathrm{sinc}^2(\Delta k \, L/2) \qquad (11)$$

Note that $\Delta k \, L$ must be kept much smaller than π radians in order for efficient SHG to occur.



Nonlinear Optical Microscopy

An important application of harmonic generation is nonlinear microscopy. . .

Microscopy based on second-harmonic generation in the configuration of a confocal microscope and excited by femtosecond laser pulses was introduced by Curley et al. (1992). Also, harmonic-generation microscopy can be used to form images of transparent (phase) objects, because the phase matching condition of nonlinear optics depends sensitively on the refractive index variation within the sample being imaged (Muller et al., 1998).

Boyd, NLO, Subsection 2.7.1

Caution!

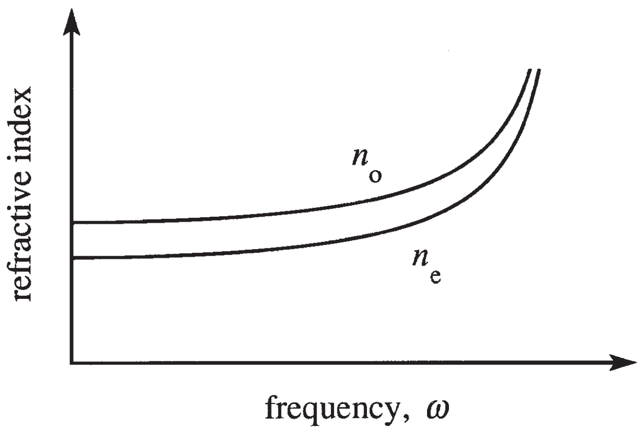
Curley et al., not Curly et al.

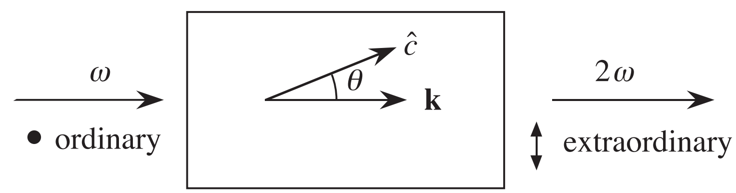
How to Achieve Phase Matching: Birefringence Phase Matching

The phase matching condition $\Delta k = 0$ requires that

$$\frac{n_1\omega_1}{c} + \frac{n_2\omega_2}{c} = \frac{n_3\omega_1}{c} \quad \text{where} \quad \omega_1 + \omega_2 = \omega_3$$

These conditions are incompatible in an isotropic dispersive material. However, for a birefringent material phase matching can be achieved.

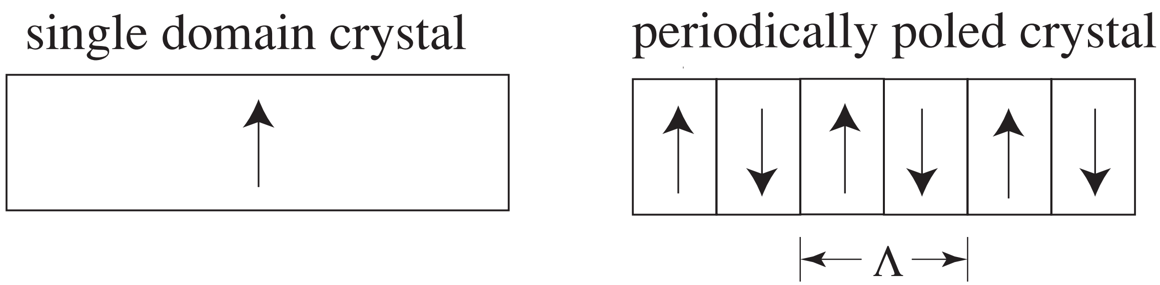




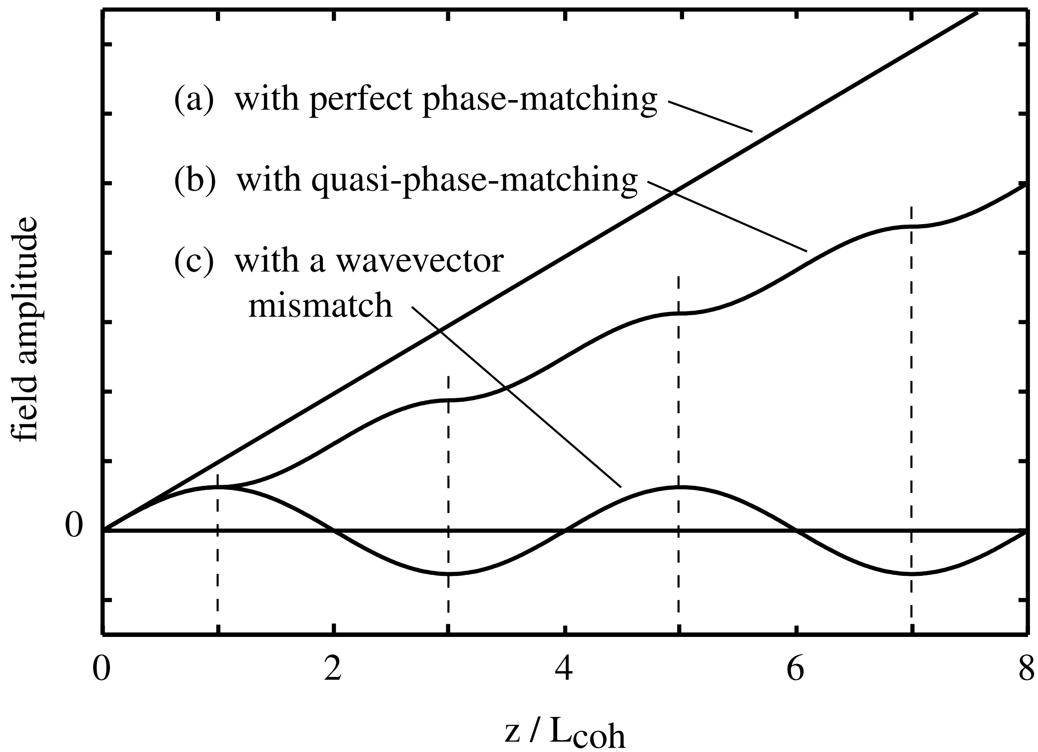
Midwinter and Warner showed that there are two ways to achieve phase matching:

	Positive uniaxial ($n_e > n_0$)	Negative uniaxial ($n_e < n_0$)
Type I	$n_3^0\omega_3 = n_1^e\omega_1 + n_2^e\omega_2$	$n_3^e\omega_3 = n_1^o\omega_1 + n_2^o\omega_2$
Type II	$n_3^0\omega_3 = n_1^o\omega_1 + n_2^e\omega_2$	$n_3^e\omega_3 = n_1^e\omega_1 + n_2^o\omega_2$

How to Achieve Phase Matching: Quasi Phase Matching



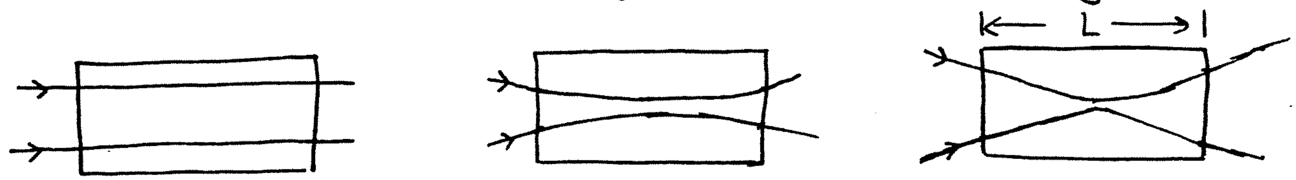
Sign of $\chi^{(2)}$ is periodically inverted to prevent reverse power flow.



Nonlinear Optics with Focused Laser Beams

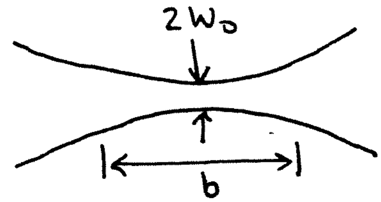
(Real experiments don't use infinite plane waves)

Focus beam to increase laser intensity.
What is optimum degree of focusing?



(too loose) (just right) (too tight)

Trade off between peak intensity $P/\pi w_0^2$ and length of interaction region $\sim b$.



$$b = \frac{2\pi w_0^2}{\lambda}$$

For SHG, Boyd and Kleinman (1968) showed that maximum power conversion occurs for

$$b = L/2.84$$

$$\Delta k = 3.2/L$$

← because of Gouy phase shift!

and is given by

$$\frac{P(2\omega)}{P(\omega)} = 1.068 \left[\frac{128\pi^2 \omega_1^3 d_{eff}^2 L}{c^4 n_1 n_2}\right] P(\omega)$$

Additional Studies of Wave Propagation Effects

3. Mechanisms of Optical Nonlinearity

Typical Values of the Nonlinear Refractive Index

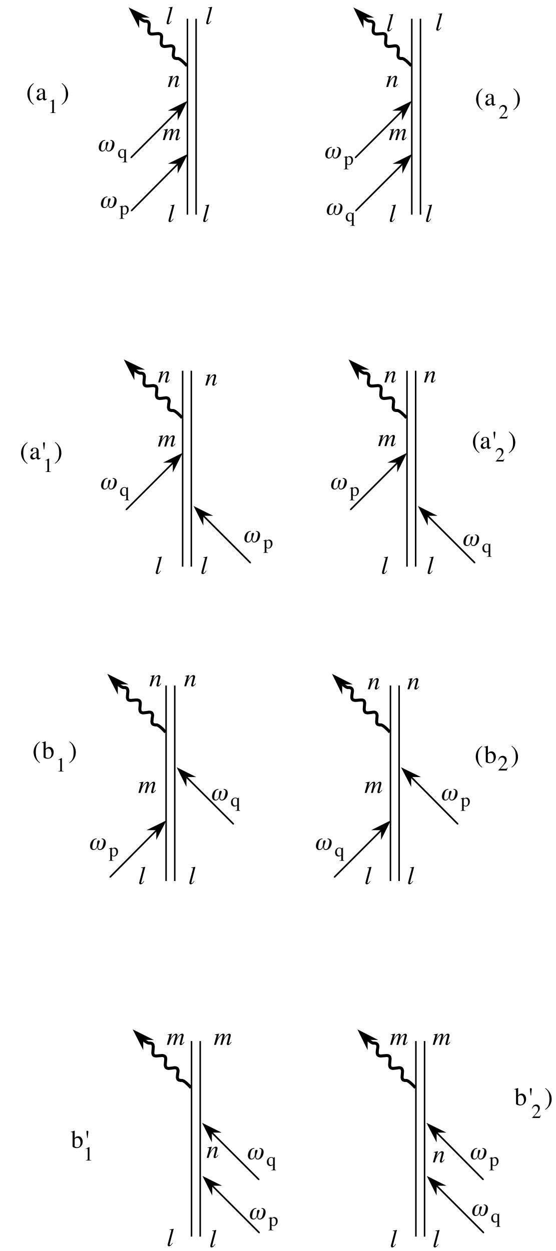
Mechanism	n_2 [a] (cm^2/W)	$\chi^{(3)}_{1111}$ (m^2/V^2)	Response time (sec)
Electronic polarization	10^{-16}	10^{-22}	10^{-15}
Molecular orientation	10^{-14}	10^{-20}	10^{-12}
Electrostriction	10^{-14}	10^{-20}	10^{-9}
Saturated atomic absorption	10^{-10}	10^{-16}	10^{-8}
Thermal effects	10^{-6}	10^{-12}	10^{-3}
Photorefractive effect[b]	(large)	(large)	(intensity-dependent)

[a] For linearly polarized light.

[b] The photorefractive effect often leads to a very strong nonlinear response. This response usually cannot be described in terms of a $\chi^{(3)}$ (or an n_2) nonlinear susceptibility, because the nonlinear polarization does not depend on the applied field strength in the same manner as the other mechanisms listed.

Quantum Mechanical Origin of the Nonlinear Optical Suesceptibility

$$
\begin{aligned}
&\chi^{(2)}_{ijk}(\omega_p+\omega_q, \omega_q, \omega_p) \\
&= \frac{N}{2\,\epsilon_0 \hbar^2} \sum_{lmn} \rho^{(0)}_{ll} \Bigg\{ \frac{\mu^i_{ln}\mu^j_{nm}\mu^k_{ml}}{[(\omega_{nl}-\omega_p-\omega_q)-i\gamma_{nl}][(\omega_{ml}-\omega_p)-i\gamma_{ml}]} && (\mathrm{a}_1) \\
&+ \frac{\mu^i_{ln}\mu^k_{nm}\mu^j_{ml}}{[(\omega_{nl}-\omega_p-\omega_q)-i\gamma_{nl}][(\omega_{ml}-\omega_q)-i\gamma_{ml}]} && (\mathrm{a}_2) \\
&+ \frac{\mu^k_{ln}\mu^i_{nm}\mu^j_{ml}}{[(\omega_{mn}-\omega_p-\omega_q)-i\gamma_{mn}][(\omega_{nl}+\omega_p)+i\gamma_{nl}]} && (\mathrm{a}'_1) \\
&+ \frac{\mu^j_{ln}\mu^i_{nm}\mu^k_{ml}}{[(\omega_{mn}-\omega_p-\omega_q)-i\gamma_{mn}][(\omega_{nl}+\omega_q)+i\gamma_{nl}]} && (\mathrm{a}'_2) \\
&+ \frac{\mu^j_{ln}\mu^i_{nm}\mu^k_{ml}}{[(\omega_{nm}+\omega_p+\omega_q)+i\gamma_{nm}][(\omega_{ml}-\omega_p)-i\gamma_{ml}]} && (\mathrm{b}_1) \\
&+ \frac{\mu^k_{ln}\mu^i_{nm}\mu^j_{ml}}{[(\omega_{nm}+\omega_p+\omega_q)+i\gamma_{nm}][(\omega_{ml}-\omega_q)-i\gamma_{ml}]} && (\mathrm{b}_2) \\
&+ \frac{\mu^k_{ln}\mu^j_{nm}\mu^i_{ml}}{[(\omega_{ml}+\omega_p+\omega_q)+i\gamma_{ml}][(\omega_{nl}+\omega_p)+i\gamma_{nl}]} && (\mathrm{b}'_1) \\
&+ \frac{\mu^j_{ln}\mu^k_{nm}\mu^i_{ml}}{[(\omega_{ml}+\omega_p+\omega_q)+i\gamma_{ml}][(\omega_{nl}+\omega_q)+i\gamma_{nl}]} \Bigg\} && (\mathrm{b}'_2)
\end{aligned}
$$



Nonresonant Electronic Nonlinearities

Estimate size:

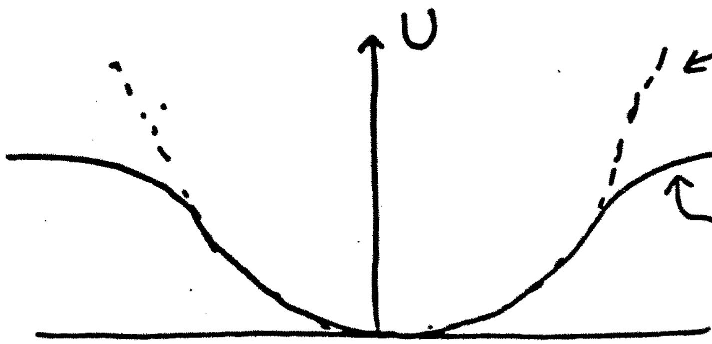
$$P = \chi^{(1)} E + \chi^{(3)} E^3$$

nonlinear term becomes comparable to linear term when

$$E \sim E_{at} \equiv \frac{e}{a_0^2}$$ ← Bohr radius

$$\Rightarrow \quad \chi^{(3)} \approx \frac{\chi^{(1)}}{E_{at}^2} \approx \frac{1}{E_{at}^2} = 10^{-16} \text{ esu}$$

Must generalize the Lorentz model of atom to allow a nonlinearity in restoring force.

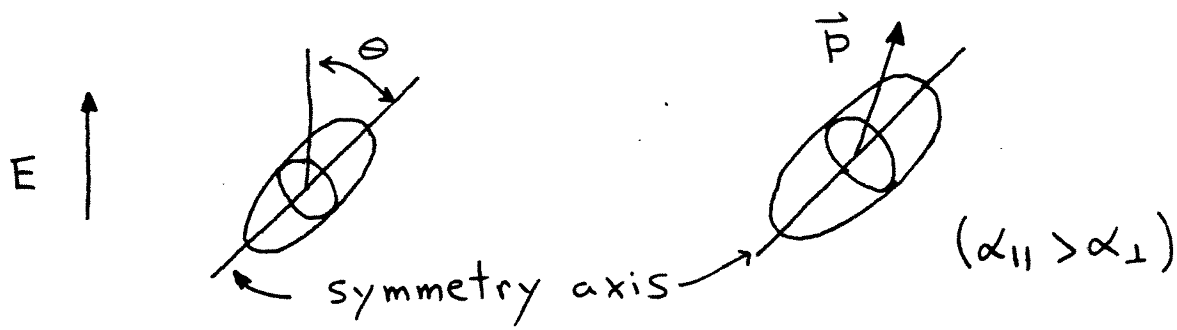


Molecular Orientation Effect

Dominant nonlinear effect in most organic liquids.

Due to optical-field induced alignment of anisotropic molecules.

Picosecond response time



$\alpha_{\parallel}$ = polarizability parallel to symmetry axis

$\alpha_{\perp}$ = polarizability perpendicular to symmetry axis

The induced dipole moment

$$\vec{p} = \overleftrightarrow{\alpha} \cdot \vec{E}$$

is <u>not</u> parallel to the applied electric field.

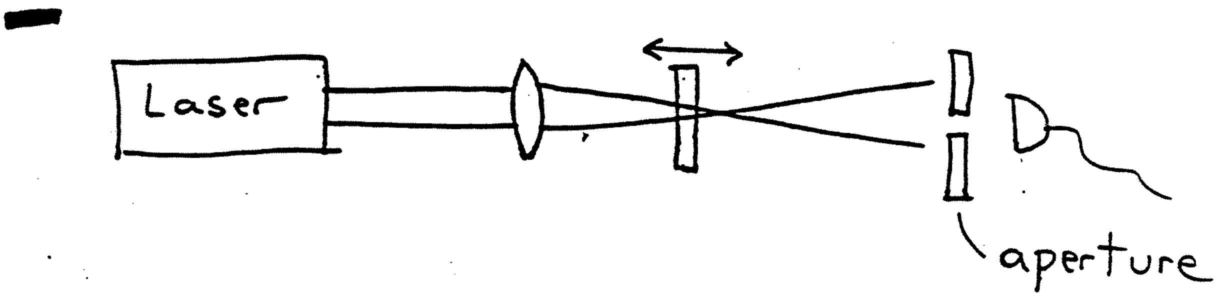
The molecule hence experiences a torque

$$\vec{\tau} = \vec{p} \times \vec{E}$$

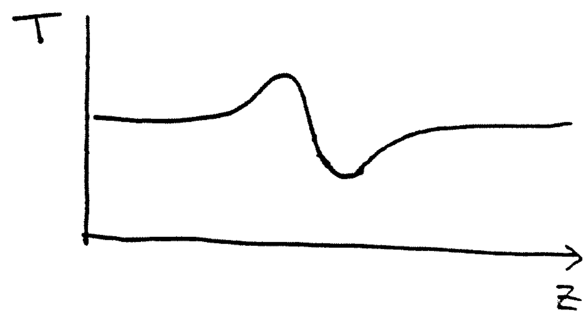
which tends to align the molecule with field.

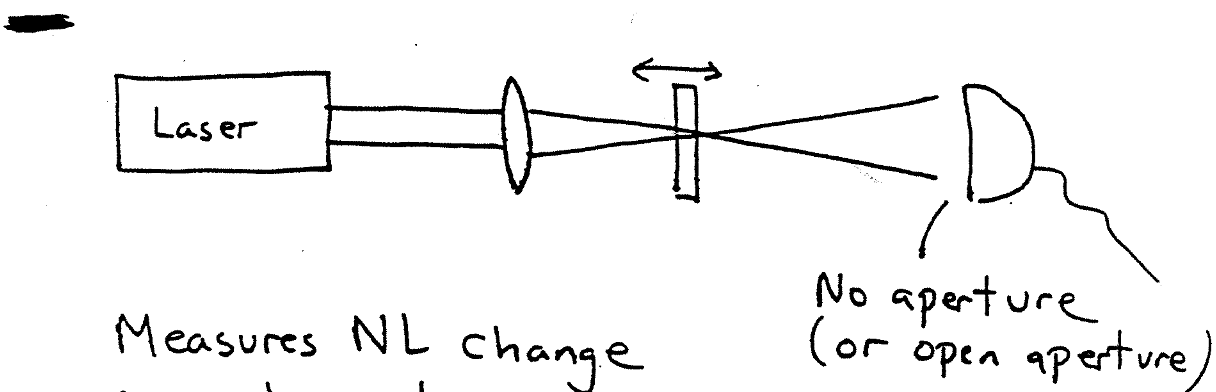
$$\chi^{(1)} = N\left(\frac{1}{3}\alpha_{\parallel} + \frac{2}{3}\alpha_{\perp}\right) \qquad \chi^{(3)} = \frac{2N}{45}\frac{(\alpha_{\parallel} - \alpha_{\perp})^2}{kT}$$

Z-Scan Measurement of $\chi^{(3)}$

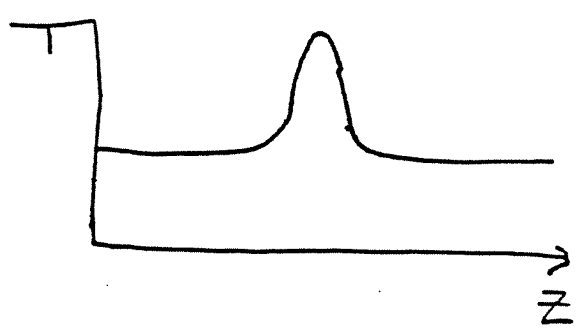
- 

  Measures NL change in refraction ($\mathrm{Re}\,\chi^{(3)}$)

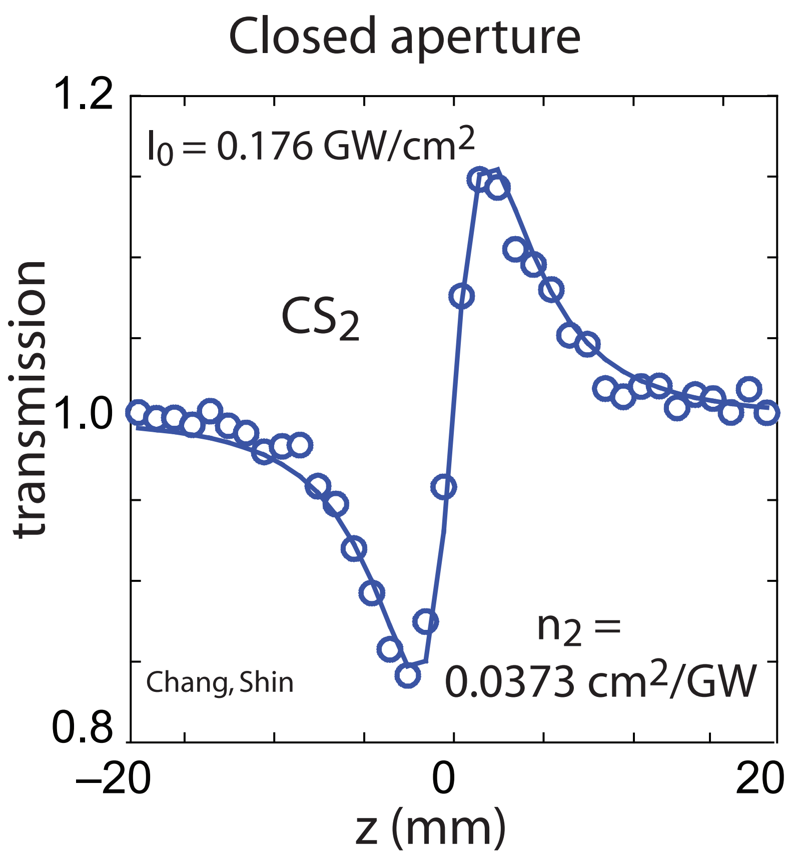
  

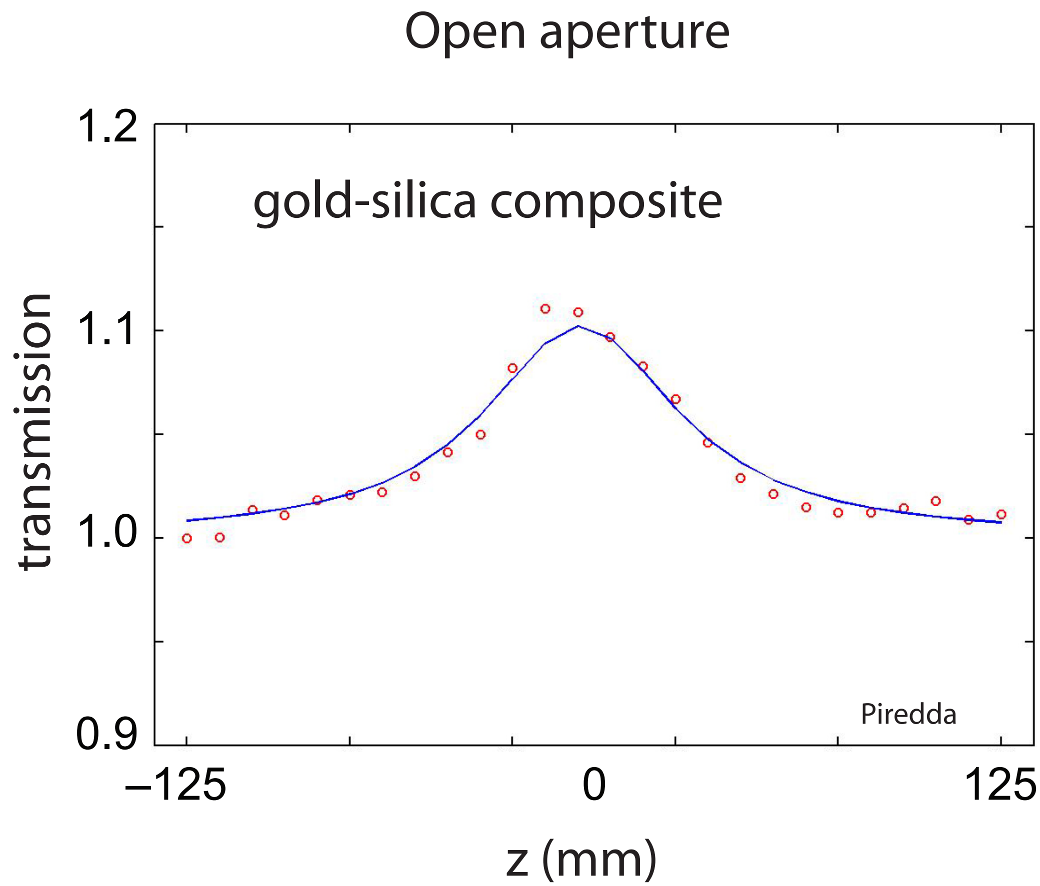
- 

  Measures NL change in absorption. ($\mathrm{Im}\,\chi^{(3)}$)

Some Actual Z-Scan Data





For closed aperture z-scan

$$\Delta T_{pv} = 0.406\ \Phi^{NL}$$

where

$$\Phi^{NL} = n_2\,(\omega/c)\ I_0\ L$$

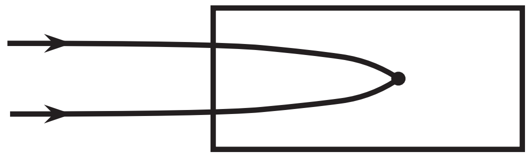
M. Sheik-Bahae et al., IEEE J. Quantum Electron. 26 760 (1990).

6. Self-Action Effects in Nonlinear Optics

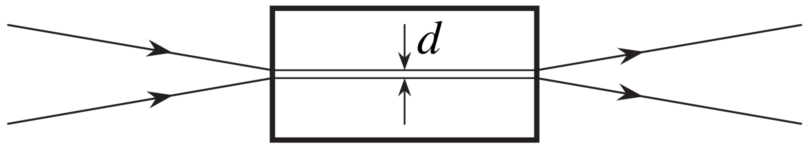
Self-Action Effects in Nonlinear Optics

Self-action effects: light beam modifies its own propagation

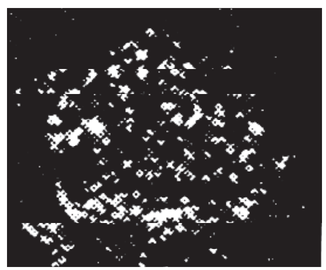
- self focusing



- self trapping

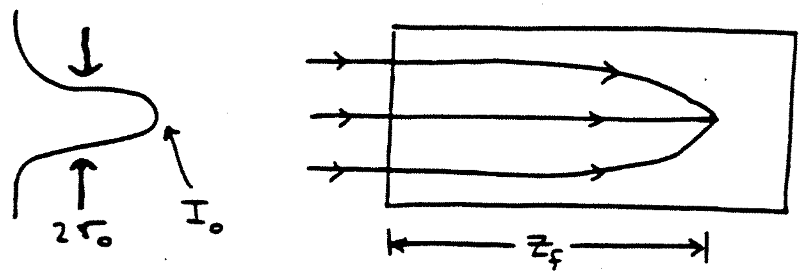


- small-scale filamentation

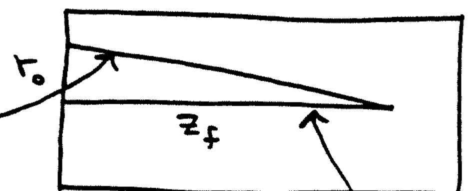


Self - Focusing

Assume that $P >> P_{cr}$ and estimate the propagation distance z_f to the self-focus.



Use Fermat's principle (approximate ray trajectories as straight lines).



$$OPL = \left[\sqrt{z_f^2 + r_0^2}\right]\left[n_0 + \tfrac{1}{2} n_2 I_0\right]$$

$$OPL = \left[z_f\right]\left[n_0 + n_2 I_0\right]$$

Equate two optical path lengths:

$$z_f = r_0 \sqrt{\frac{n_0}{n_2 I_0}}$$

This result can be expressed equivalently as

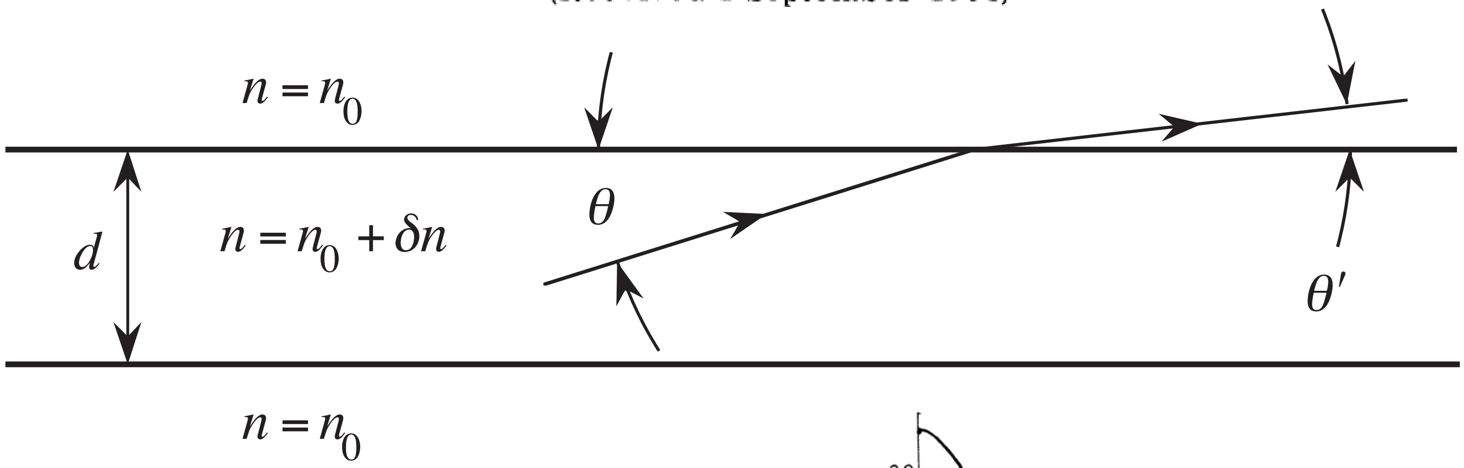
$$z_f = r_0^2 \sqrt{\frac{\pi n_0}{n_2 P}} = \frac{2\sqrt{2}\, n_0 r_0^2}{0.61\ \lambda} \frac{1}{\sqrt{P/P_{cr}}}$$

Prediction of Self Trapping

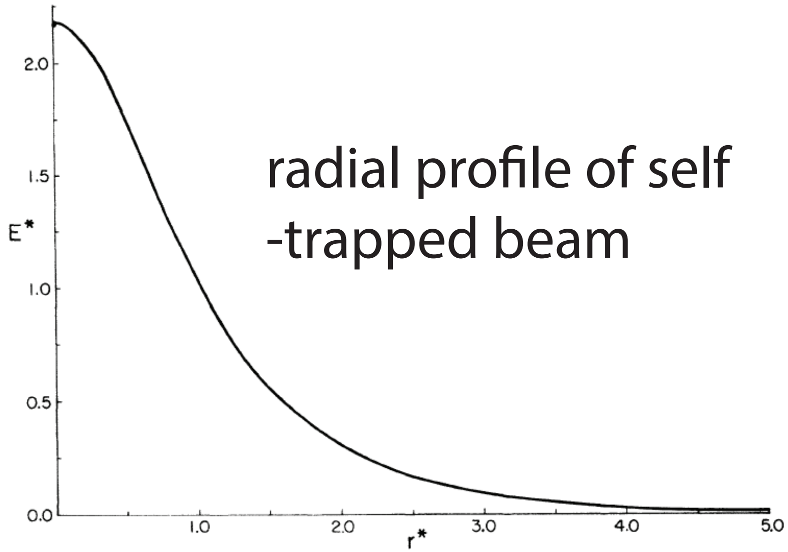
VOLUME 13, NUMBER 15 PHYSICAL REVIEW LETTERS 12 OCTOBER 1964

SELF-TRAPPING OF OPTICAL BEAMS*

R. Y. Chiao, E. Garmire, and C. H. Townes
Massachusetts Institute of Technology, Cambridge, Massachusetts
(Received 1 September 1964)



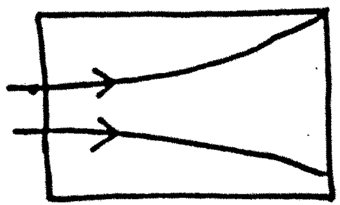
$$P_{\rm cr} = \frac{\pi(0.61)^2\lambda_0^2}{8n_0n_2}$$



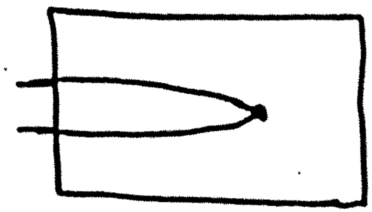
radial profile of self
-trapped beam

Properties and Applications of Spatial Solitons

- A spatial soliton propagates with a uniform transverse dimension because of a perfect balance between diffraction and self focusing.



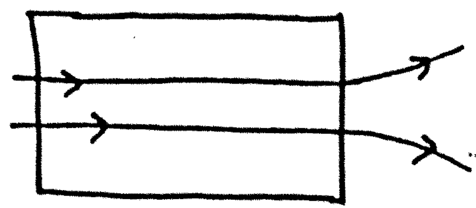
diffraction



self-focusing

$n = n_0 + n_2 I$

$n_2 > 0$



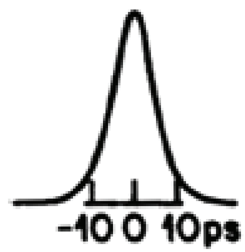
spatial soliton

Optical Solitons

Field distributions that propagate without change of form

Temporal solitons (nonlinearity balances gvd)

$$\frac{\partial \tilde{A}_s}{\partial z} + \frac{1}{2} i k_2 \frac{\partial^2 \tilde{A}_s}{\partial \tau^2} = i\gamma |\tilde{A}_s|^2 \tilde{A}_s.$$

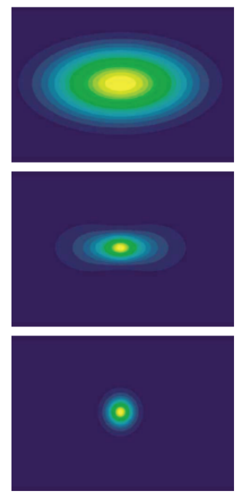


1973: Hasegawa & Tappert

1980: Mollenauer, Stolen, Gordon

Spatial solitons (nonlinearity balances diffraction)

$$2ik_0 \frac{\partial A}{\partial z} + \frac{\partial^2 A}{\partial x^2} = -3\chi^{(3)} \frac{\omega^2}{c^2} |A|^2 A$$



1964: Garmire, Chiao, Townes

1974: Ashkin and Bjorkholm (Na)

1985: Barthelemy, Froehly (CS2)

1991: Aitchison et al. (planar glass waveguide

1992: Segev, (photorefractive)

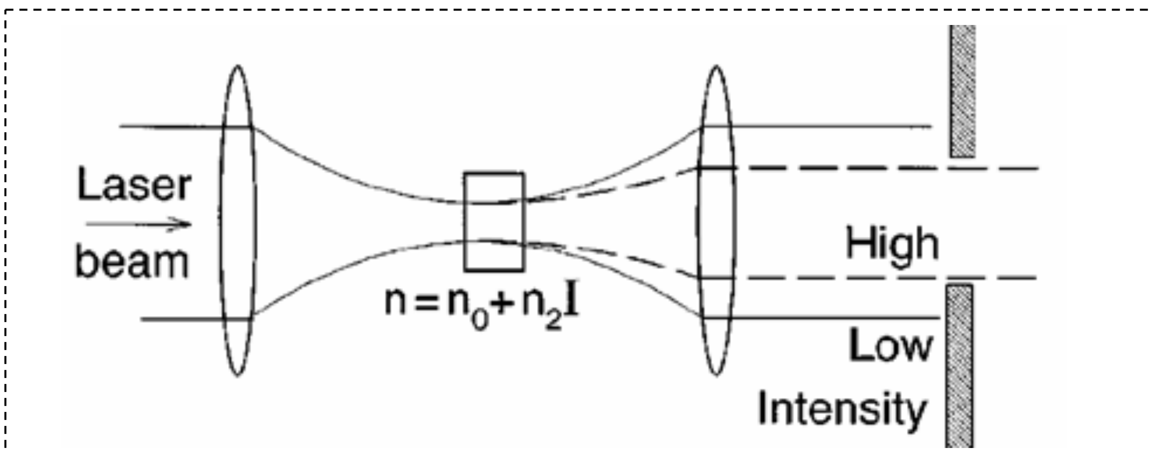
Solitons and self-focussing in Ti:Sapphire

42 OPTICS LETTERS / Vol. 16, No. 1 / January 1, 1991

60-fsec pulse generation from a self-mode-locked Ti:sapphire laser

D. E. Spence, P. N. Kean, and W. Sibbett

J. F. Allen Physics Research Laboratories, Department of Physics and Astronomy, University of St. Andrews, North Haugh, St. Andrews, Fife, KY16 9SS, Scotland



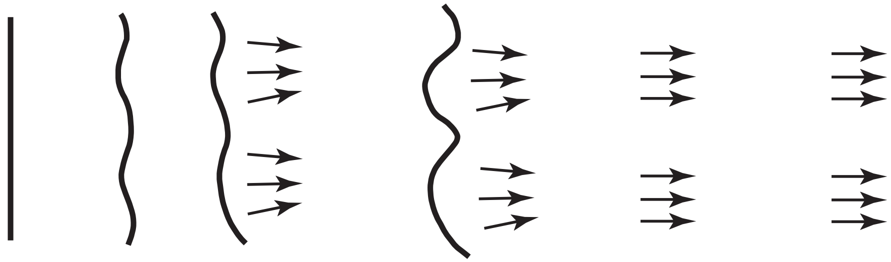
Diffraction-management controls the spatial self-focussing

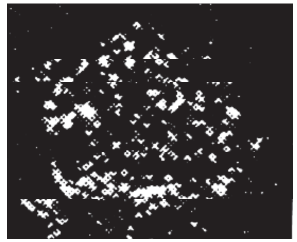
Dispersion-management controls the temporal self-focussing

Beam Breakup by Small-Scale Filamentation

Predicted by Bespalov and Talanov (1966)

Exponential growth of wavefront imperfections by four-wave mixing processes





Sidemode amplitude grows as

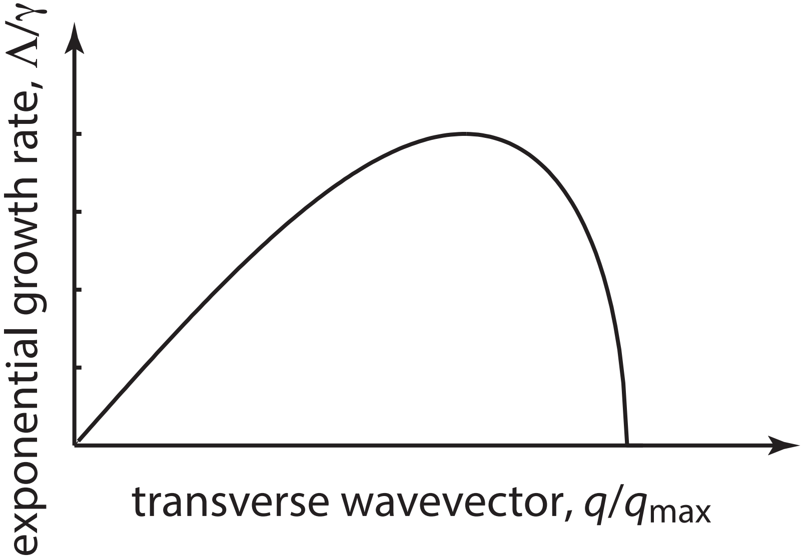
$$A_1(z) = A_1(0)e^{i\gamma z}e^{\Lambda z}$$

$$\Lambda^2 = \frac{q^2}{2k}\left(2\gamma - \frac{q^2}{2k}\right),$$

$$\gamma = n_2(\omega / c)I_0, \quad q = |\mathbf{k}_\perp|$$

$$q_{\max} = \sqrt{2k\gamma}$$

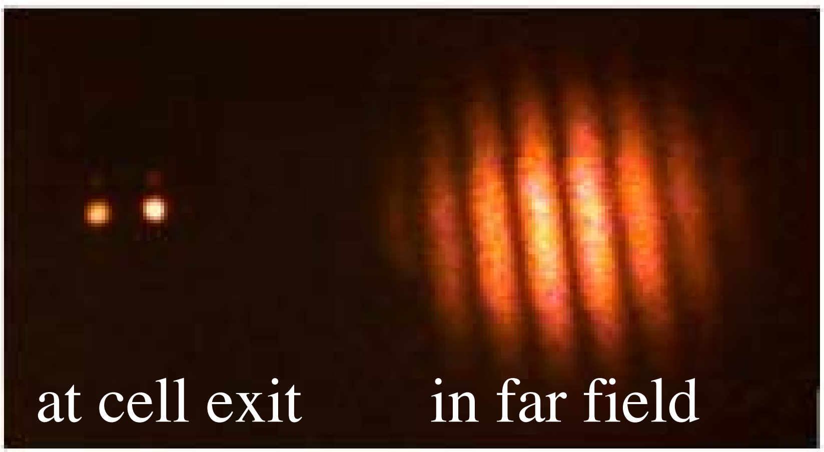
$$\theta_{\max} = \frac{q_{\max}}{k}$$



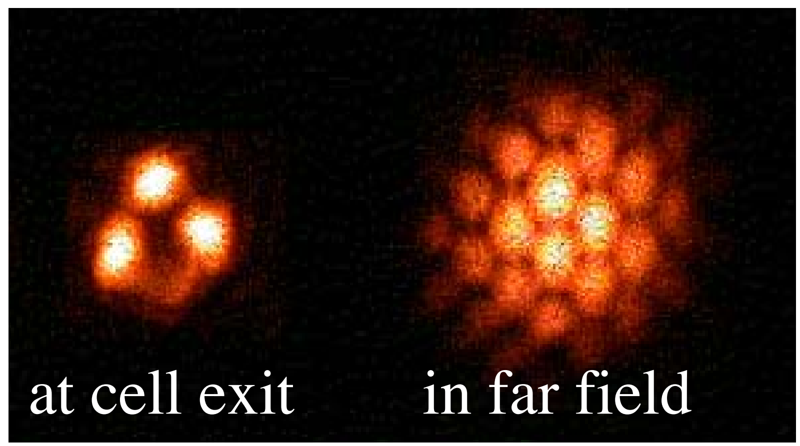
Honeycomb Pattern Formation

Output from cell with a single gaussian input beam

At medium input power



At high input power



Quantum statistics?

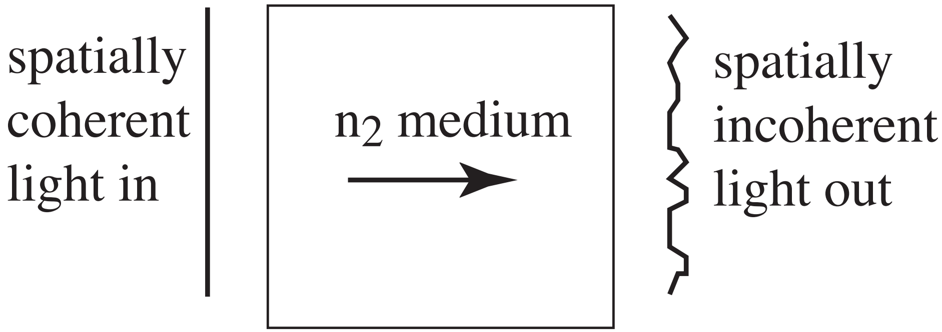
Input power 100 to150 mW
Input beam diameter 0.22 mm

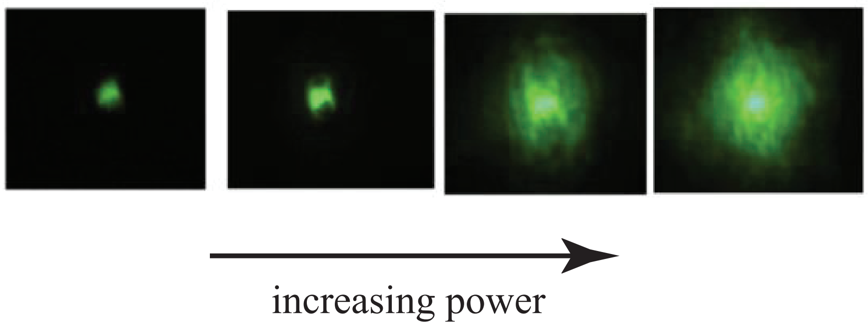
Sodium vapor cell T = 220^o C
Wavelength = 588 nm
Bennink et al., PRL 88, 113901 2002.

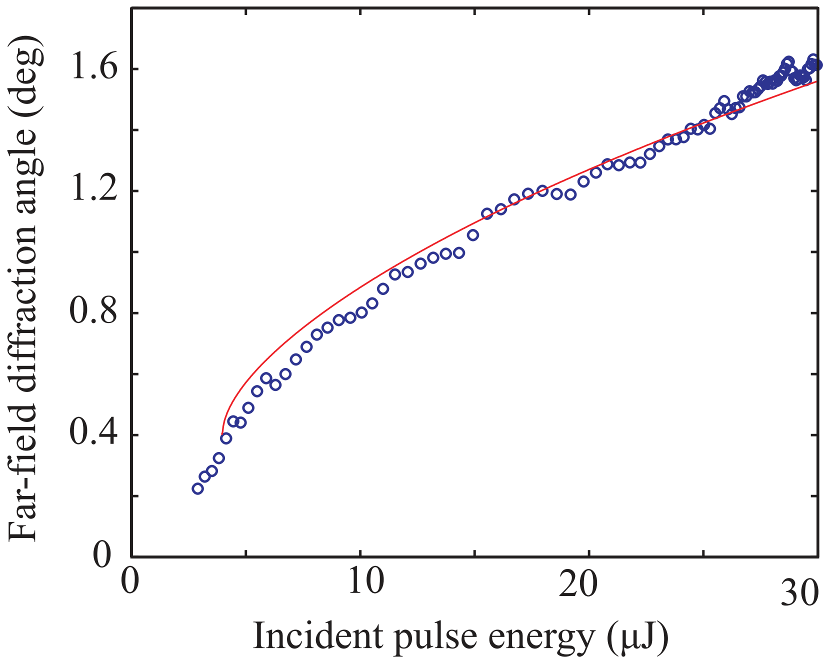
Optical Radiance Limiter Based on Spatial Coherence Control

Controlled small-scale filamentation used to modify spatial degree of coherence

Alternative to standard appropches to optical power limiting





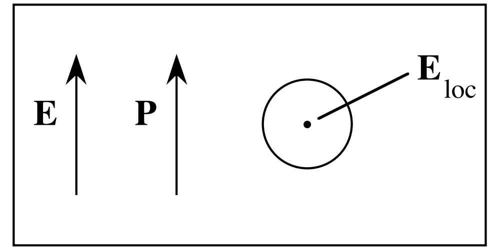


Schweinsberg et al., Phys. Rev. A 84, 053837 (2011).

4. Local-Field Effects in Nonlinear Optics

Local Field Effects in Nonlinear Optics – I

Recall the Lorentz-Lorenz Law
(linear optics)



$$\chi^{(1)} = \frac{N\alpha}{1 - \frac{4}{3}\pi N\alpha} \quad \text{or} \quad \frac{\epsilon^{(1)} - 1}{\epsilon^{(1)} + 2} = \frac{4}{3}\pi N\alpha.$$

This result follows from the assumption that the field that acts on a representative atom is not the macroscopic Maxwell field but rather the Lorentz local field given by

$$E_{\text{loc}} = E + \tfrac{4}{3}\pi P \qquad \text{where} \qquad P = \chi^{(1)} E$$

We can rewrite this result as

$$E_{\text{loc}} = LE \quad \text{where} \quad L = \frac{\epsilon^{(1)} + 2}{3} \quad \text{is the local field factor.}$$

The Lorentz Red Shift

$$\frac{\varepsilon - 1}{\varepsilon + 2} = \frac{4\pi}{3} N \alpha$$

$$\alpha = \frac{c f r_e \lambda_0 e / 4\pi}{\omega_0 - \omega - i\gamma}$$

$\Rightarrow$

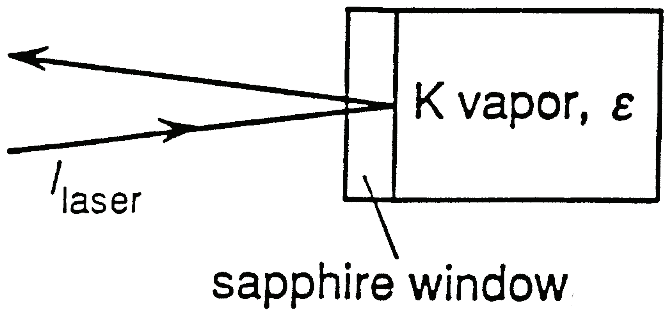
$$\varepsilon = 1 + \frac{N f r_e \lambda_0 c}{\omega_0 + \Delta\omega_L - \omega - i\gamma}$$

$$\Delta\omega_L = -\frac{1}{3} N f r_e \lambda_0 c$$

↑ Lorentz red shift

See, for instance, H. A. Lorentz, Theory of Electrons, Dover, NY (1952).

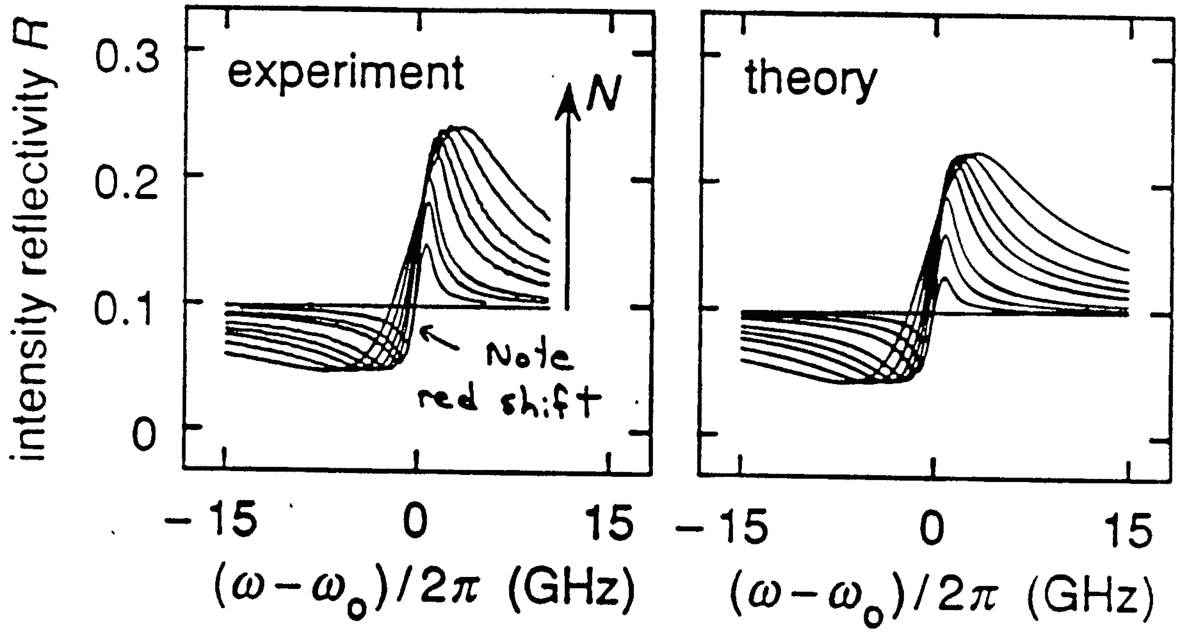
Observation of the Lorentz Red Shift



$N \sim 10^{17}\,\mathrm{cm}^{-3}$

$\Gamma_{Dop} \sim 1\,\mathrm{GHz}$

$|\varepsilon - 1| \sim 1$ and $|L| \sim 4/3$



$4^2S_{1/2} \leftrightarrow 4^2P_{1/2}$

Maki, Malcuit, Sipe, and Boyd, Phys. Rev. Lett. 68, 972 (1991).

Local Field Effects in Nonlinear Optics – II

For the case of nonlinear optics, Bloembergen (1962, 1965) showed that, for instance,

$$\chi^{(3)}(\omega = \omega + \omega - \omega) = N\gamma^{(3)}|L(\omega)|^2[L(\omega)]^2.$$

where $\gamma^{(3)}$ is the second hyperpolarizability and where

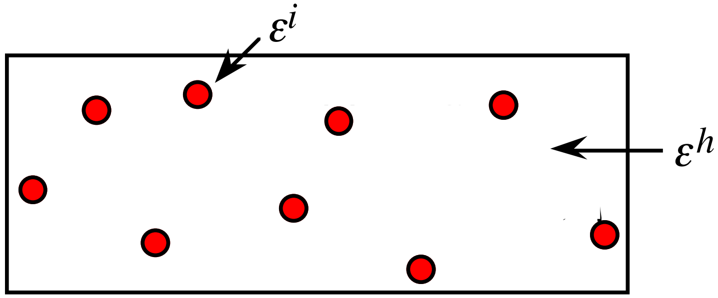
$$L(\omega) = \frac{\epsilon(\omega) + 2}{3}$$

For the typical value $n = 2$, $L = 2$, and $L^4 = 16$. Local field effects can be very large in nonlinear optics! But can we tailor them for our benefit?

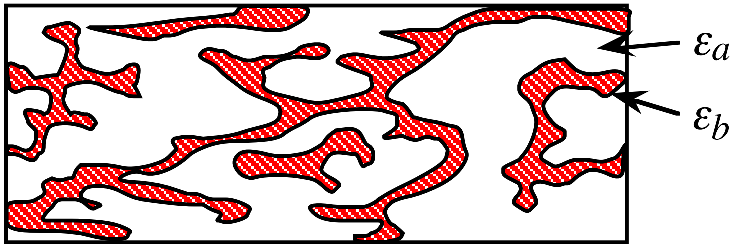
We have been developing new photonic materials with enhanced NLO response by using composite structures that exploit local field effects.

Nanocomposite Materials for Nonlinear Optics

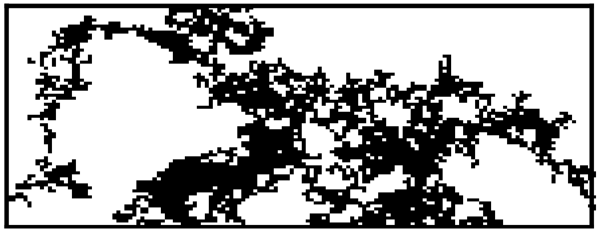
• Maxwell Garnett



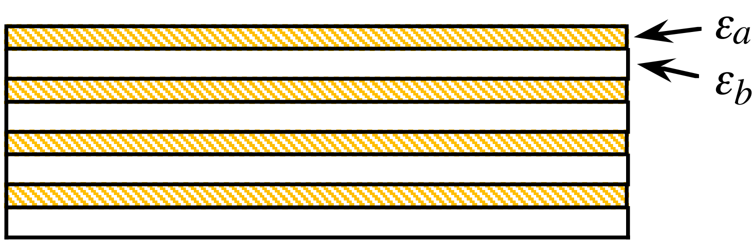
• Bruggeman (interdispersed)



• Fractal Structure



• Layered



- In each case, scale size of inhomogeneity << optical wavelength

- Thus all optical properties, such as n and $\chi^{(3)}$, can be described by effective (volume averaged) values

Recent review: Dolgaleva and Boyd, Advances in Optics and Photonics 4, 1–77 (2012).

Local Field Enhancement of the NLO Response

- Under very general conditions, we can express the NL response as

$$\chi_{\text{eff}}^{(3)} = f\,L^2|L|^2\chi^{(3)}$$

where f is the volume fraction of nonlinear material and L is the local-field factor, which is different for each material geometry.

- Under appropriate conditions, the product $f\,L^2|L|^2$ can exceed unity.

- For a homogeneous material $$L = \frac{\varepsilon + 2}{3}$$

- For a spherical particle of dielectric constant ε_m embedded in a host of dielectic constant ε_h

$$L = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h}$$

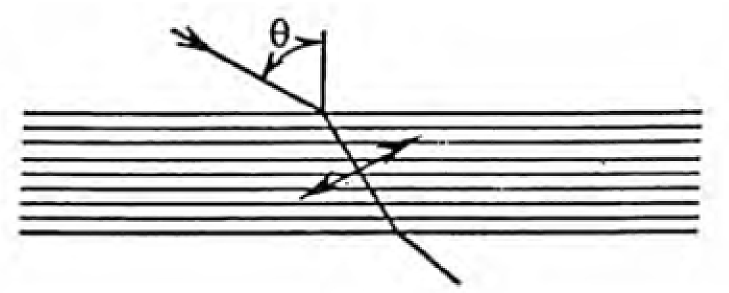
- For a layered geometry with the electric field perpendicular to the plane of the layers, the local field factor for component a is given by

$$L = \frac{\varepsilon_{\text{eff}}}{\varepsilon_{\text{a}}} \qquad \frac{1}{\varepsilon_{\text{eff}}} = \frac{f_{\text{a}}}{\varepsilon_{\text{a}}} + \frac{f_{\text{b}}}{\varepsilon_{\text{b}}}$$

Enhanced NLO Response from Layered Composite Materials

A composite material can display a larger NL response than its constituents!

Alternating layers of TiO_2 and the conjugated polymer PBZT.



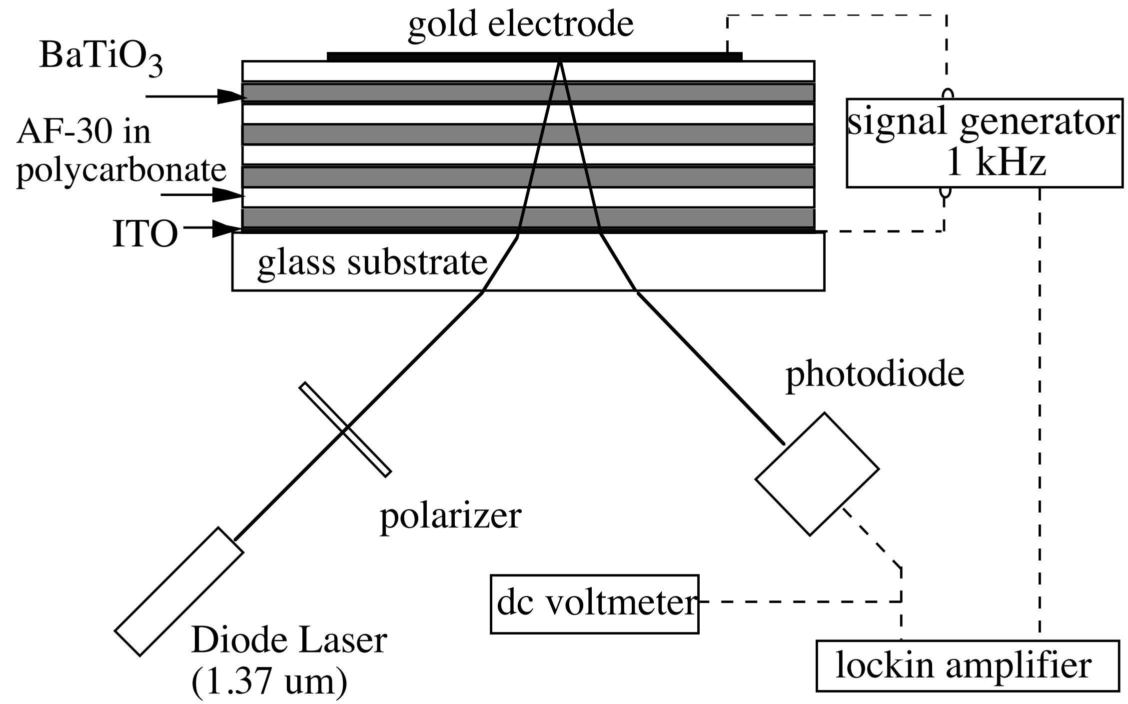
$\nabla \cdot \mathbf{D} = 0$ implies that $(\varepsilon \mathbf{E})_\perp$ is continuous.

Measure NL phase shift as a function of angle of incidence.

35% enhancement in $\chi^{(3)}$

Fischer, Boyd, Gehr, Jenekhe, Osaheni, Sipe, and Weller-Brophy, Phys. Rev. Lett. 74, 1871 (1995).

Quadratic EO effect

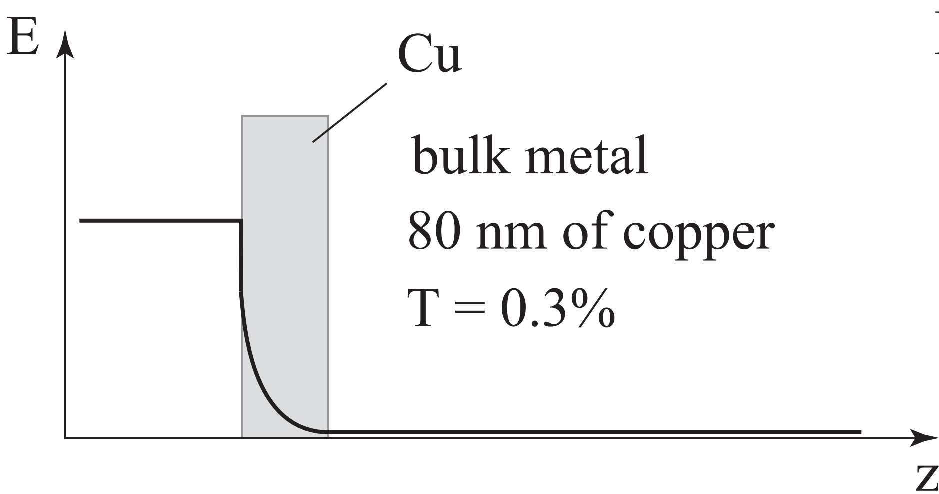


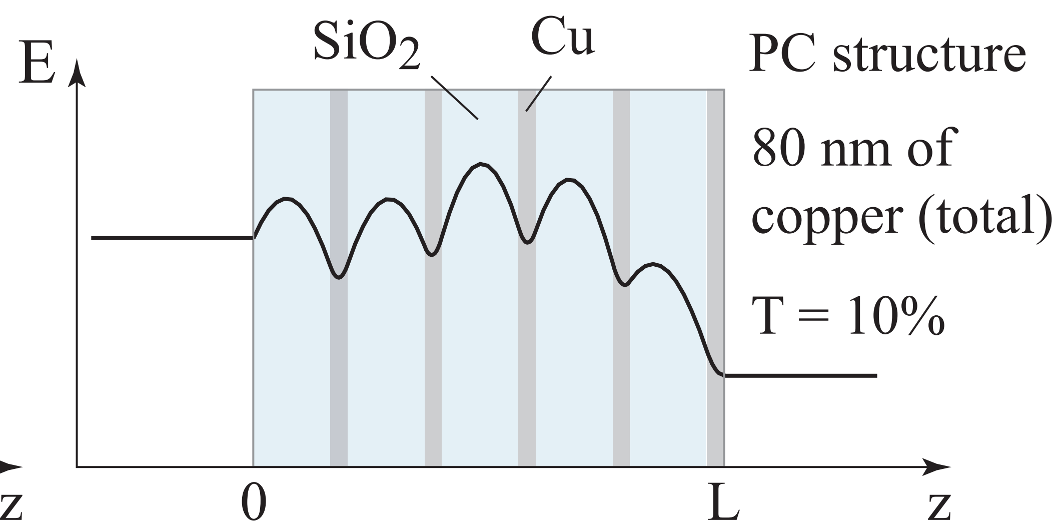
3.2 times enhancement!

Nelson and Boyd, APL 74 2417 (1999)

Accessing the Optical Nonlinearity of Metals with Metal-Dielectric Photonic Crystal Structures

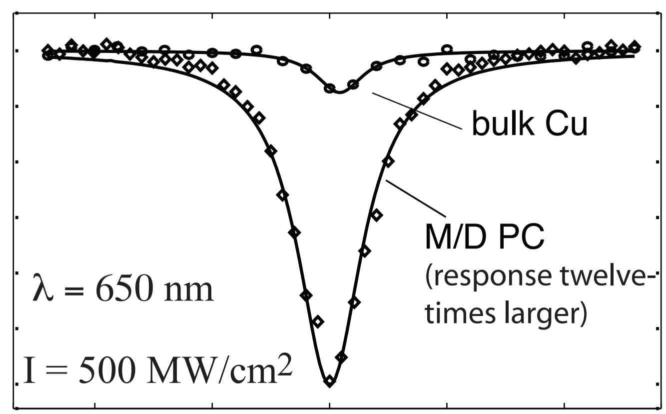
- Metals have very large optical nonlinearities but low transmission
- Low transmission because metals are highly reflecting (not because they are absorbing!)
- Solution: construct metal-dielectric photonic crystal structure
  (linear properties studied earlier by Bloemer and Scalora)





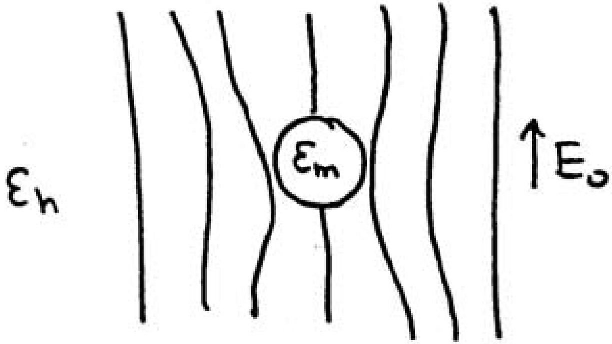
Bennink, Yoon, Boyd, and Sipe, Opt. Lett. 24, 1416 (1999).

Lepeshkin, Schweinsverg, Piredda, Bennink, and Boyd, Phys. Rev. Lett. 93, 123902 (2004).



Metal / Dielectric Composites

Very large local field effects



$$E_{in} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} E_o$$

$$\equiv \alpha E_o$$

(ε_m is negative !)

At resonance

$$\alpha = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} \longrightarrow \frac{3\varepsilon_h}{i\varepsilon_m''} \approx (3 \text{ to } 30)\, i$$

Gold-Doped Glass: A Maxwell-Garnett Composite

Developmental Glass, Corning Inc.

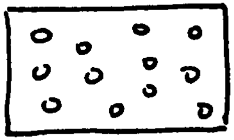
Red Glass Caraffe
Nurenberg, ca. 1700

Huelsmann Museum, Bielefeld

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

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

Counterintuitive Consequence of Local Field Effects



gold nanoparticles in a liquid dye solution (HITCI)

Both constituents are reverse saturable absorbers $\Rightarrow$ $\mathrm{Im}\,\chi^{(3)} > 0$

Effective NL susceptibility of composite

$$\chi^{(3)}_{eff} = f\, \mathcal{L}^2 |\mathcal{L}|^2 \chi^{(3)}_{Au} + (1-f)\, \chi^{(3)}_{dye\ sol'n}$$

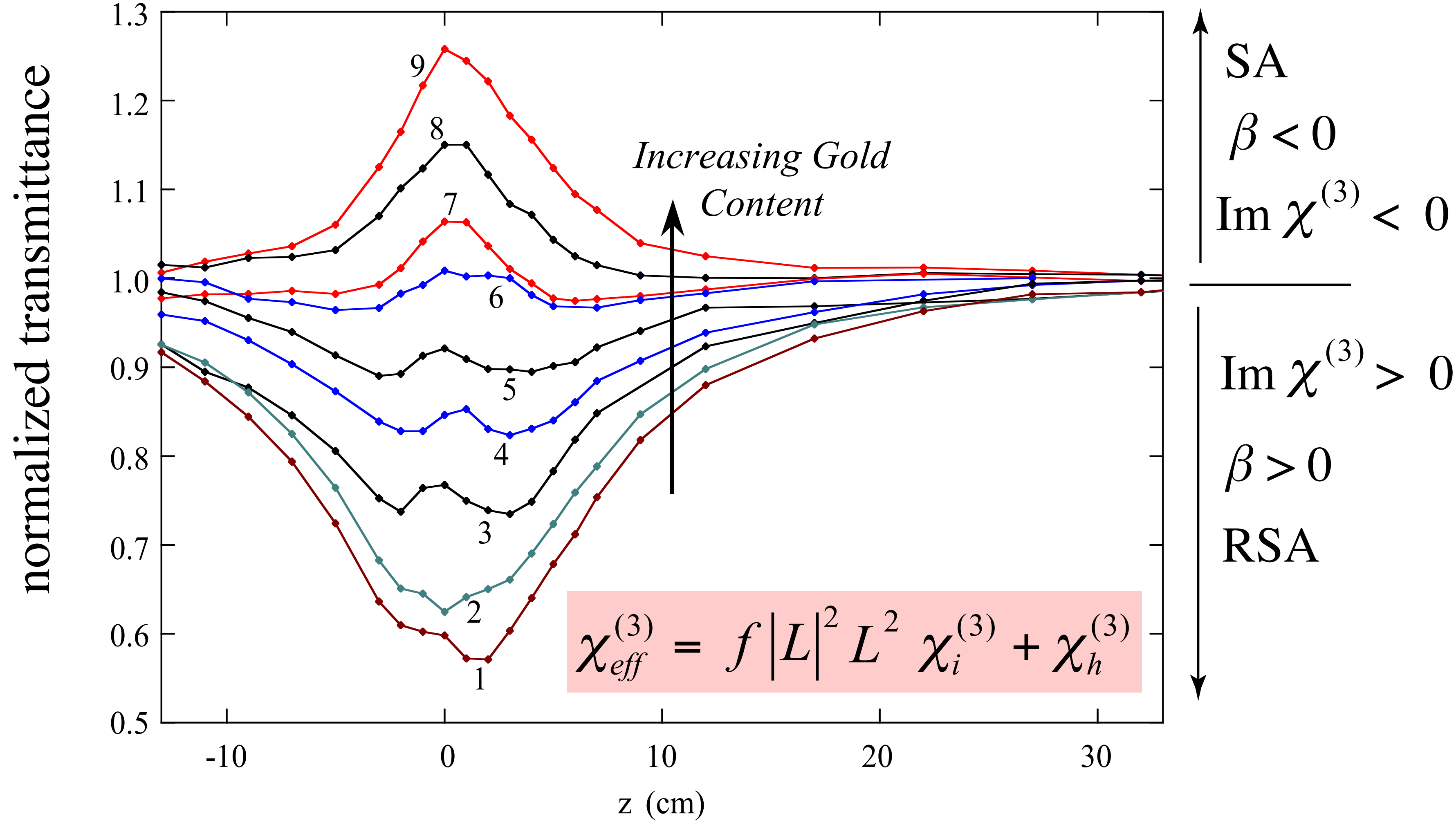
$$\mathcal{L} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} = \text{pure imaginary at resonance!}$$

A cancellation of the two contributions to $\chi^{(3)}$ can occur, even though they have same sign.

Counterintuitive Consequence of Local Field Effects

Cancellation of two contributions that have the same sign

Gold nanoparticles in a reverse saturable absorber dye solution (13 μM HITCI)

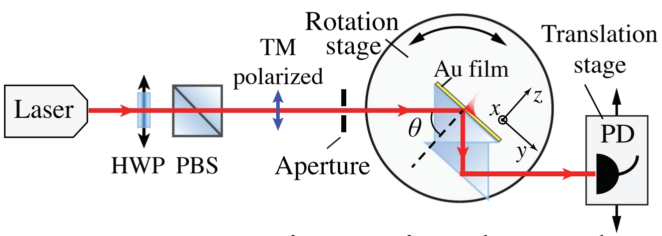


D.D. Smith, G. Fischer, R.W. Boyd, D.A.Gregory, *JOSA B* **14**, 1625, 1997.

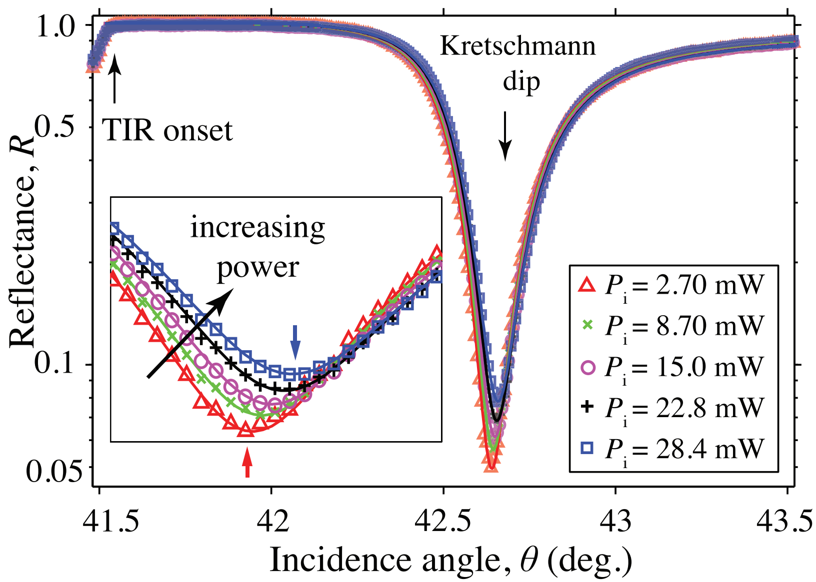
NLO in Plasmonics (Photonics Using Metals)

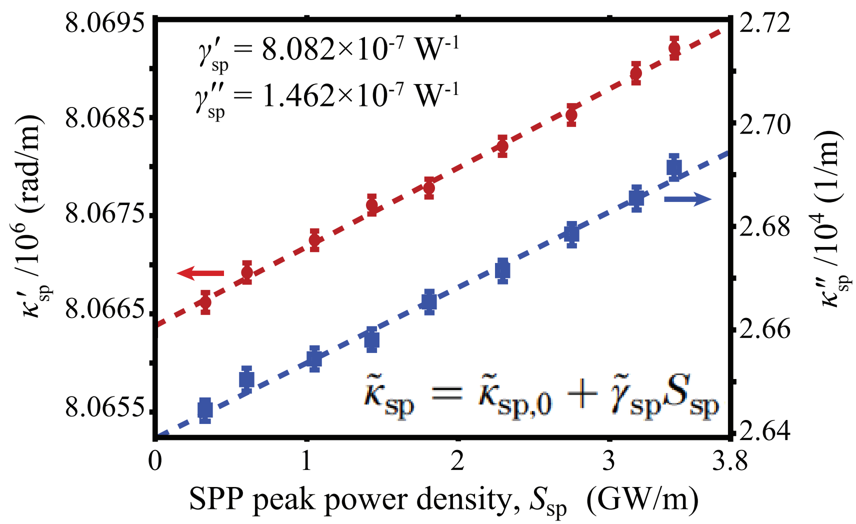
Is there an intrinsic nonlinear reponse to surface plasmon polaritons (SPPs)?

- A nonlinar response would be useful for photonics applications
- Metals are highly nonlinear (n_2 is10^6 times that of silica)
- High (sub-wavelength) field confinement in an SPP
- But SPPs tend to show high loss



Measure intensity dependence of the Kretschmann angle





$$\tilde{\chi}^{(3)}_{Au} = (4.67 + i3.03) \times 10^{-19} \ \mathrm{m^2/V^2}$$

power varies from 2 to 18 mW;
intensity varies from 2 to 22 GW/cm
laser wavelength is 796.5 nm
laser pulse duration is 100 fs

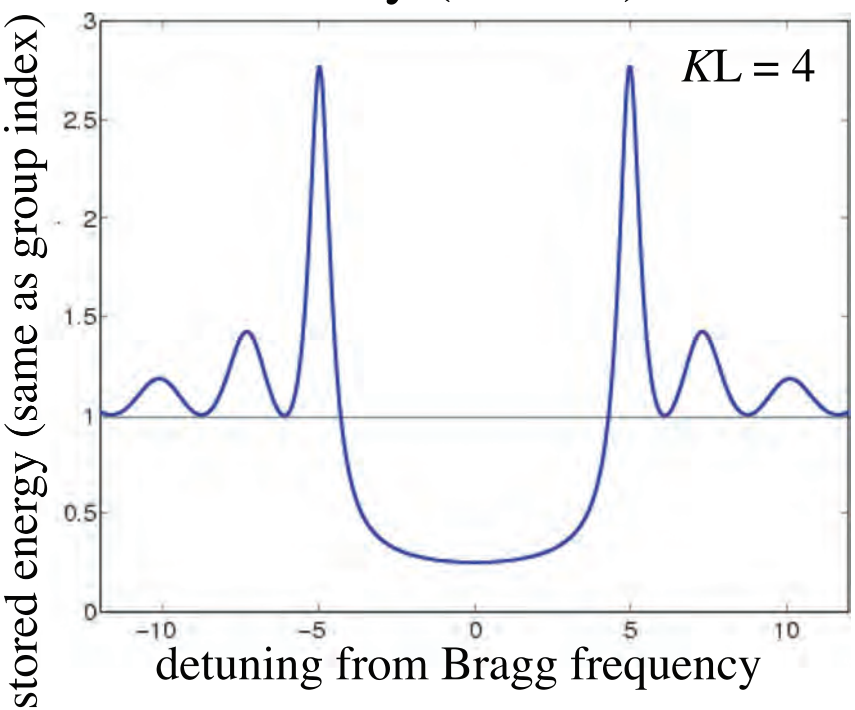
I. De Leon, Z. Shi, A. Liapis and R.W.Boyd, Optics Letters 39, 2274 (2014)

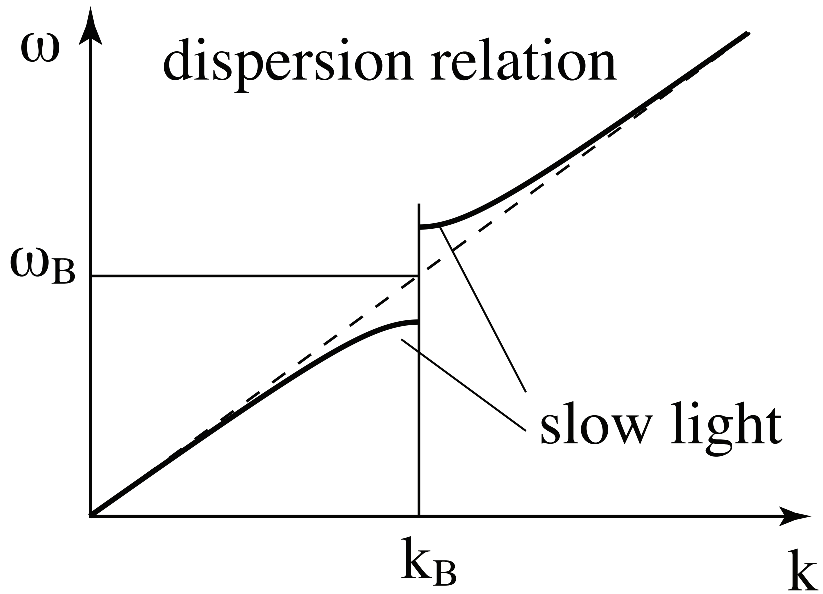
Slow Light in a Fiber Bragg Grating (FBG) Structure

(Can describe properties of FBGs by means of analytic expressions)

forward and backward waves are strongly coupled

theory (Winful)





- Enhanced NLO response

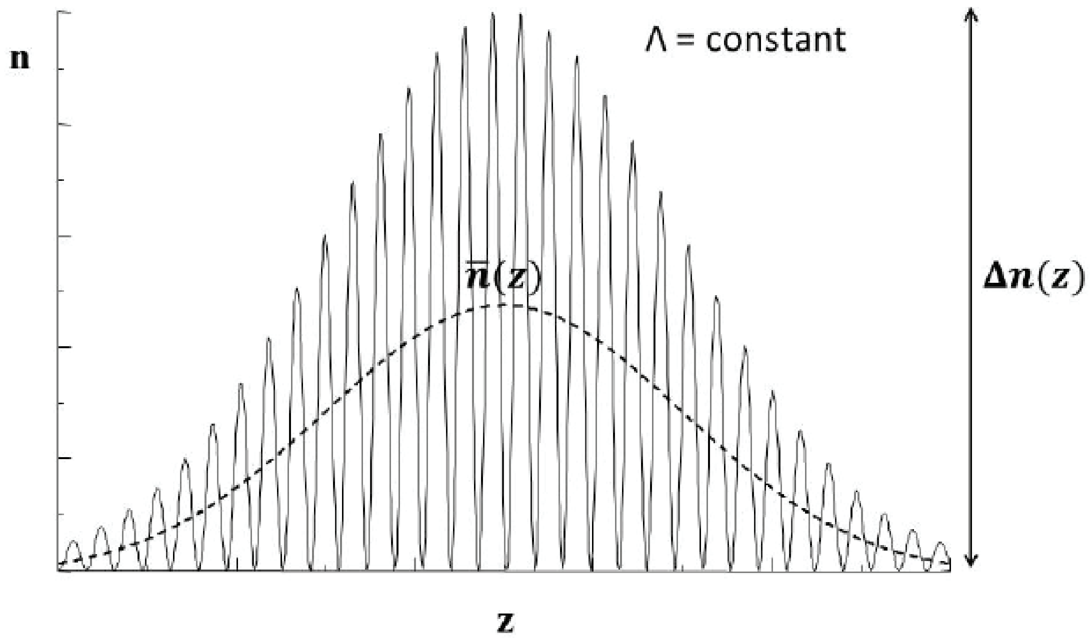
Bhat and Sipe showed that the nonlinear coeficient is given by

$$\Gamma = \left(\frac{3 - S^{-2}}{2} \right) S^2 \gamma_0$$

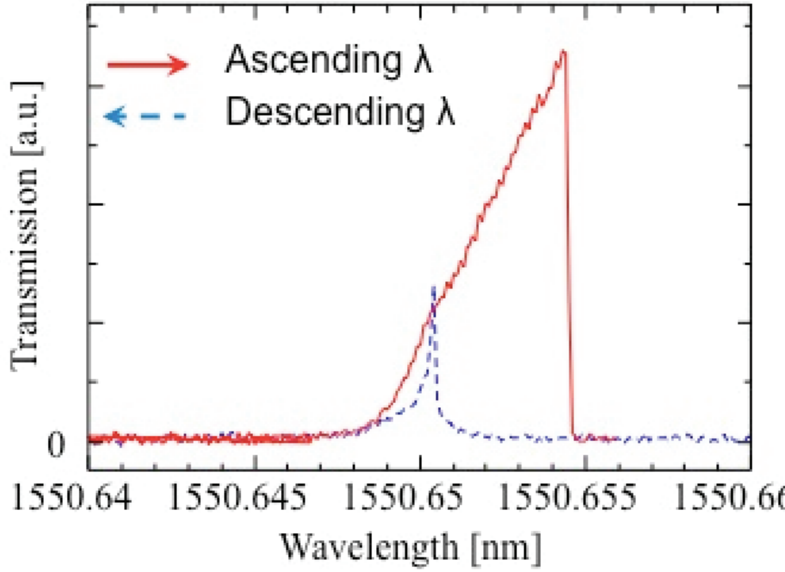
where the slow-down factor $S = n_g/n$

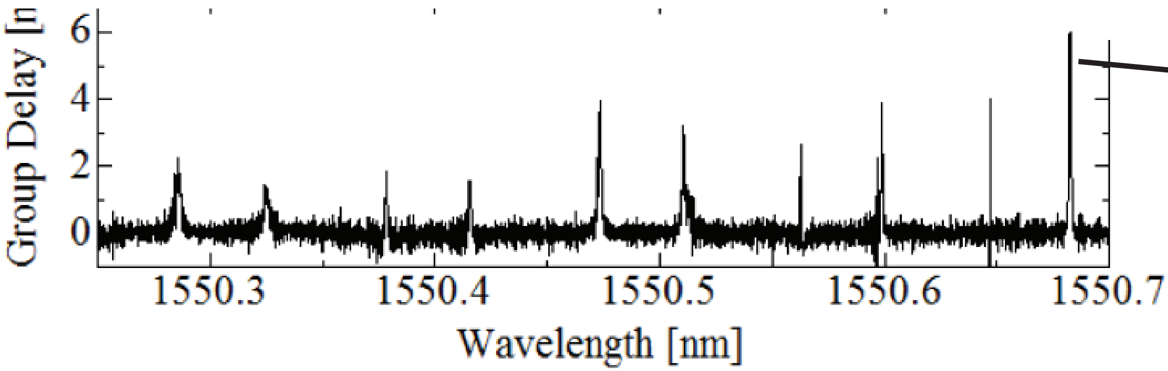
Improved Slow-Light Fiber Bragg Grating (FBG) Structure

Much larger slow-down factors possible with a Gaussian-profile grating



Observation of (thermal) optical bistability at mW power levels





group index approximately 140

H. Wen, M. Terrel, S. Fan and M. Digonnet, IEEE Sensors J. 12, 156-163 (2012).

J. Upham, I. De Leon, D. Grobnic, E. Ma, M.-C. N. Dicaire, S.A. Schulz, S. Murugkar, and R.W. Boyd, Optics Letters 39, 849-852 (2014).

The other Lake Como

(Buffalo, New York)

5. Slow and Fast Light

Controlling the Velocity of Light

"Slow," "Fast" and "Backwards" Light

– Light can be made to go:

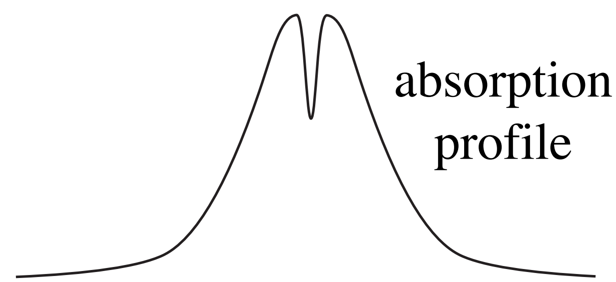
slow: $v_g << c$ (as much as 10^6 times slower!)

fast: $v_g > c$

backwards: v_g negative

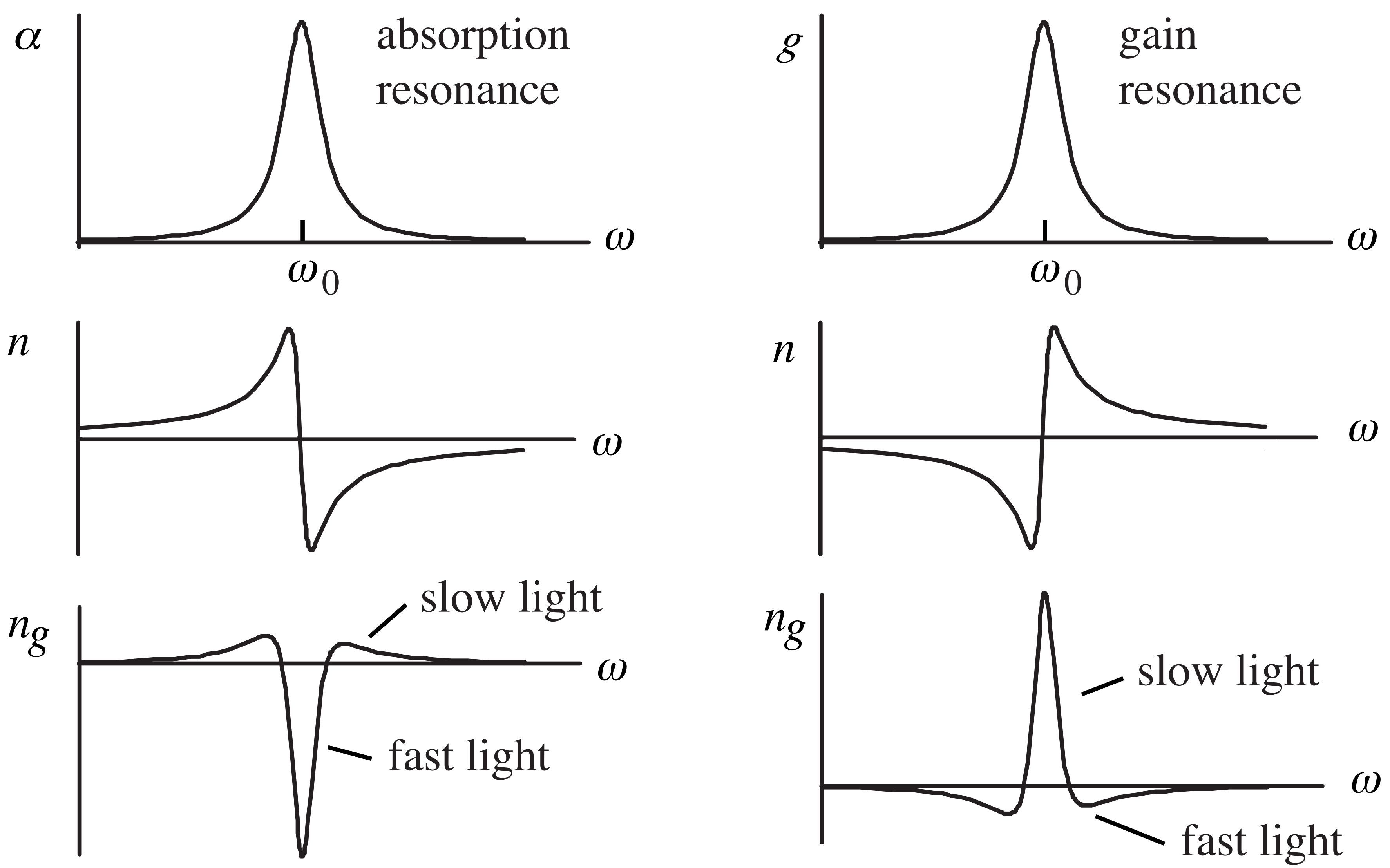
Here v_g is the group velocity: $v_g = c/n_g$ $n_g = n + \omega\,(dn/d\omega)$

– Velocity controlled by structural or material resonances



Review article: Boyd and Gauthier, Science 326, 1074 (2009).

Slow and Fast Light Using Isolated Gain or Absorption Resonances



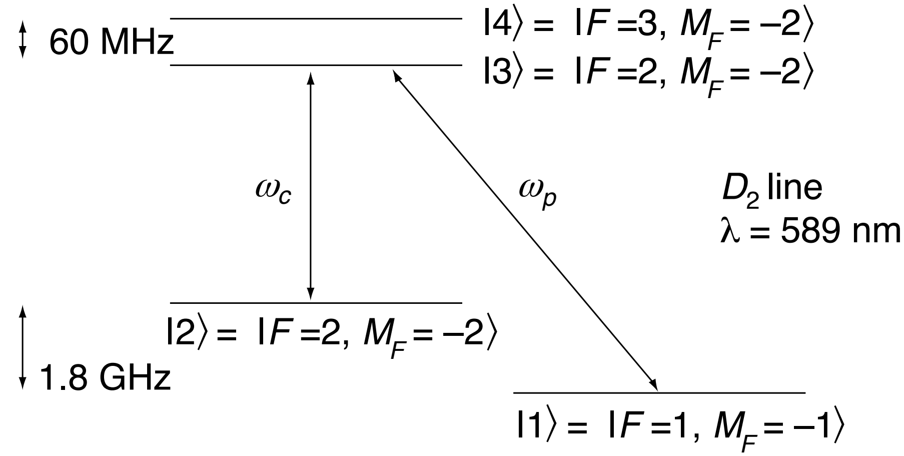
$$n_g = n + \omega\,(dn/d\omega)$$

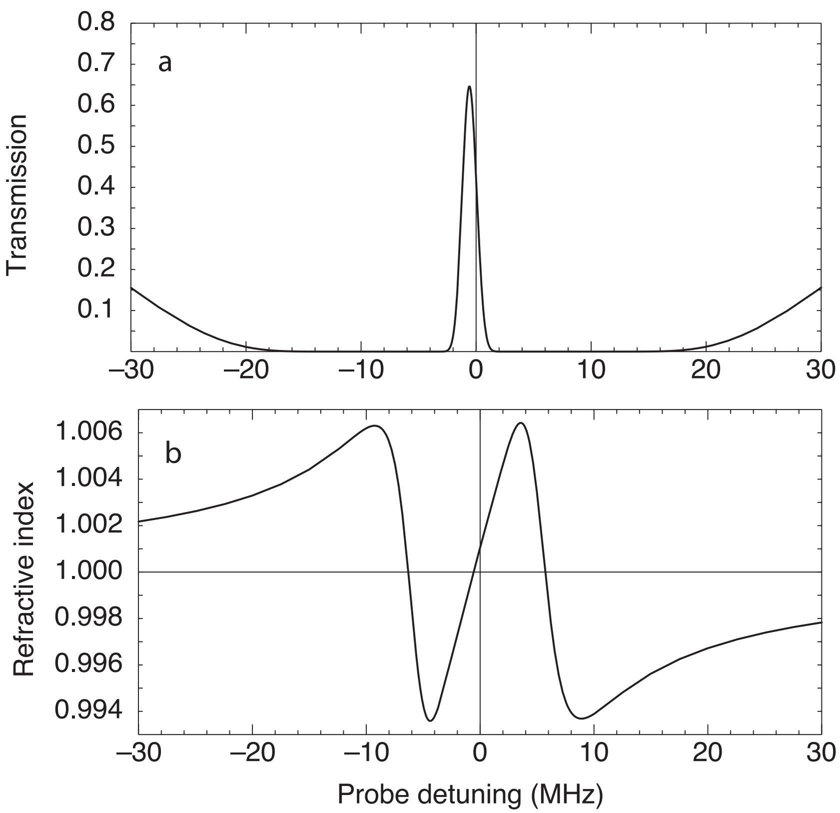
Light speed reduction to 17 metres per second in an ultracold atomic gas

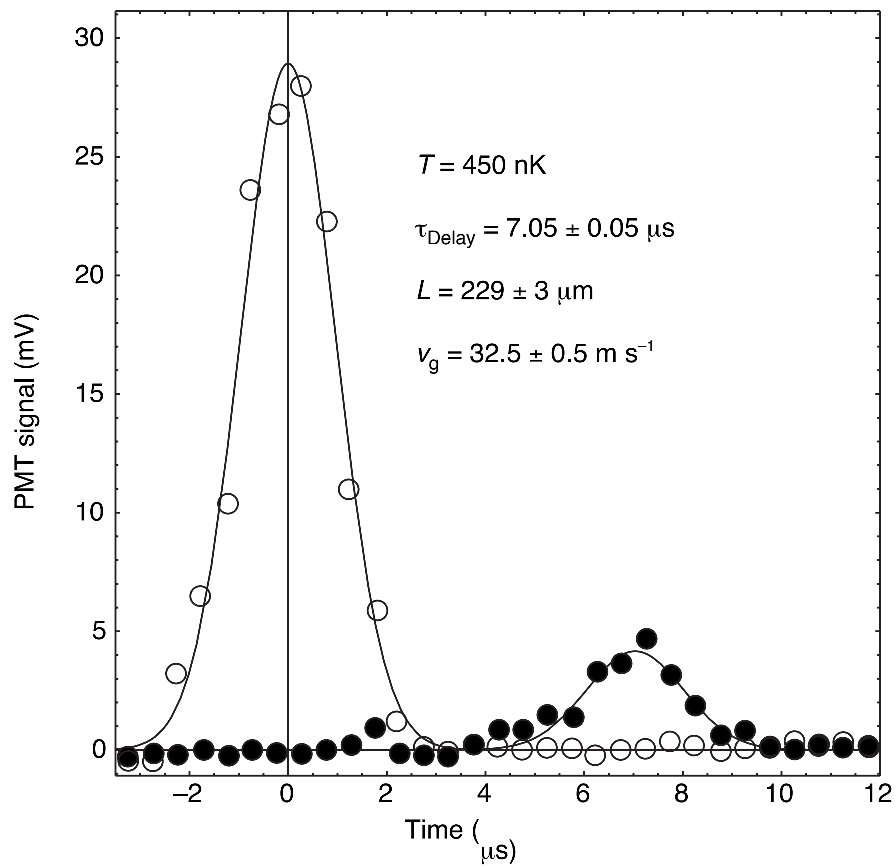
Lene Vestergaard Hau*[2], S. E. Harris[3], Zachary Dutton*[2] & Cyrus H. Behroozi*§

* Rowland Institute for Science, 100 Edwin H. Land Boulevard, Cambridge, Massachusetts 02142, USA
[2] Department of Physics, § Division of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA
[3] Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA

NATURE | VOL 397 | 18 FEBRUARY 1999 |www.nature.com







Note also related work by Chu, Wong, Welch, Scully, Budker, Ketterle, and many others

Goal: Slow Light in a Room-Temperature Solid-State Material

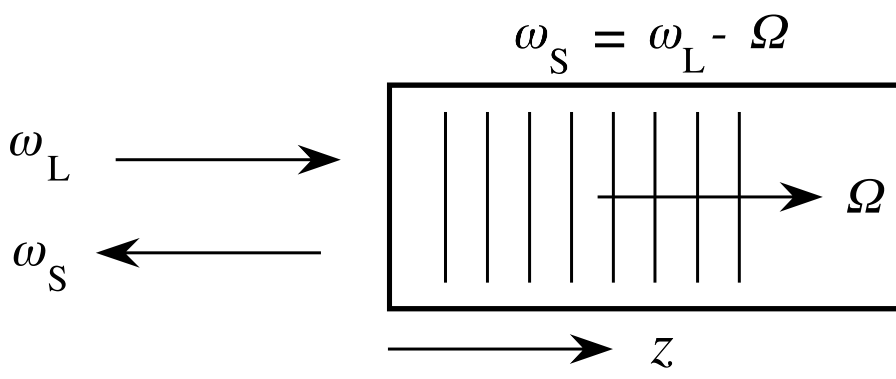
Crucial for many real-world applications

We have identified two preferred methods for producing slow light

(1) Slow light *via* coherent population oscillations (CPO)

(2) Slow light *via* stimulated Brillouin scattering (SBS)

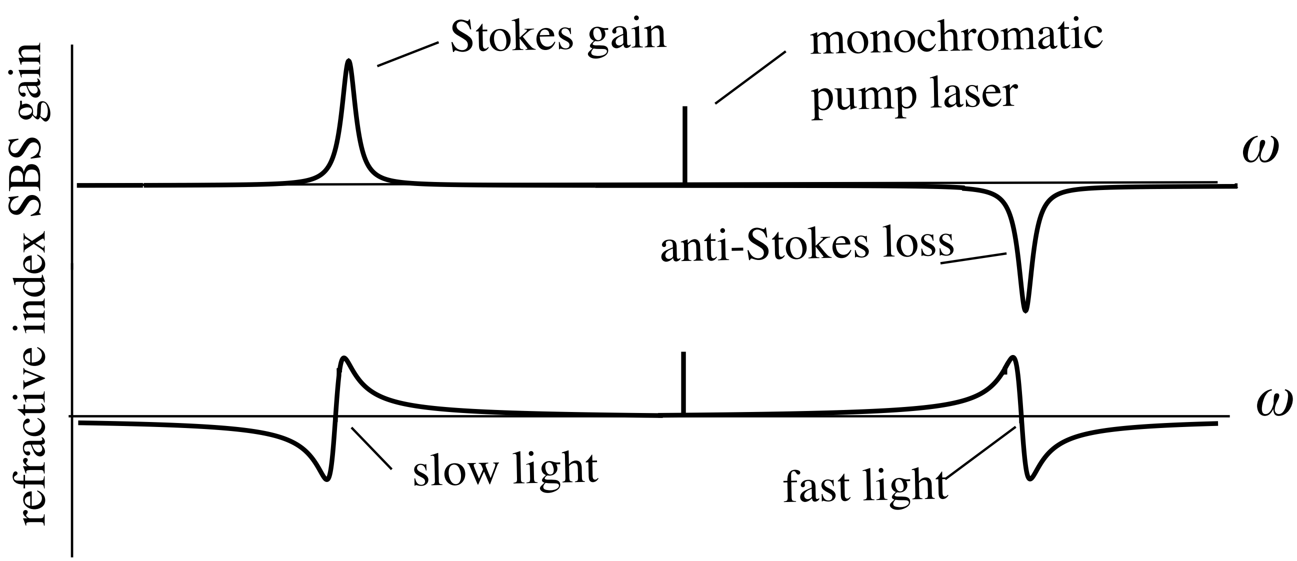
Slow Light by Stimulated Brillouin Scattering (SBS)

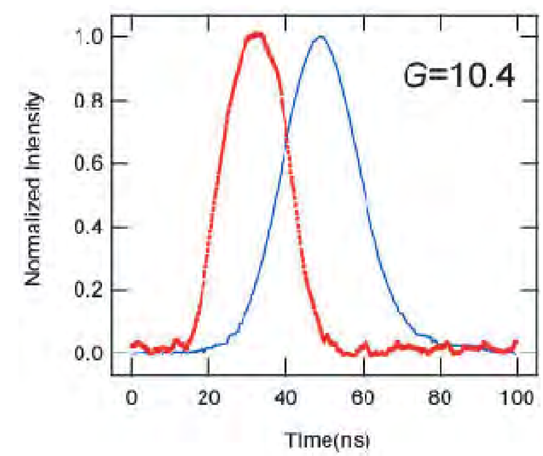


$$\frac{dI_S}{dz} = -gI_LI_S$$

$$g = \frac{\gamma_e^2 \omega^2}{nv\, c^3 \rho_0 \Gamma_B}.$$

We often think of SBS as a pure gain process, but it also leads to a change in refractive index

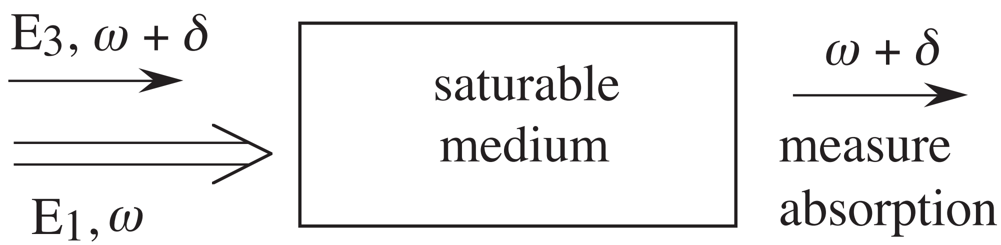


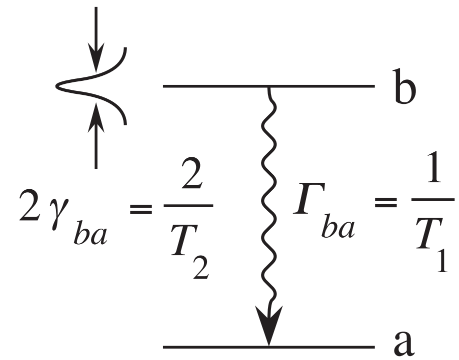


The induced time delay is $\Delta T_d \approx \dfrac{G}{\Gamma_B}$ where $G = g\, I_P\, L$ and Γ_B is the Brillouin linewidth

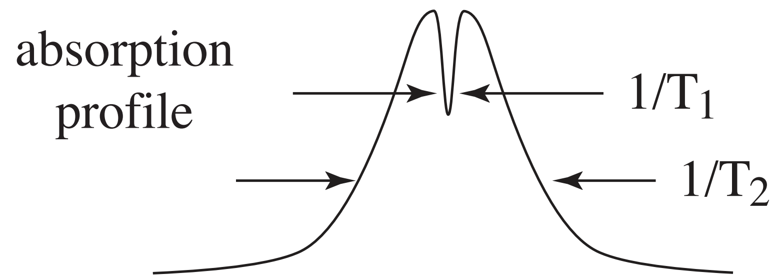
Okawachi, Bigelow, Sharping, Zhu, Schweinsberg, Gauthier, Boyd, and Gaeta Phys. Rev. Lett. 94, 153902 (2005).
Related results reported by Song, González Herráez and Thévenaz, Optics Express 13, 83 (2005).

Slow Light via Coherent Population Oscillations





$n_g = n + \omega(dn/d\omega)$ $\quad T_2 << T_1$

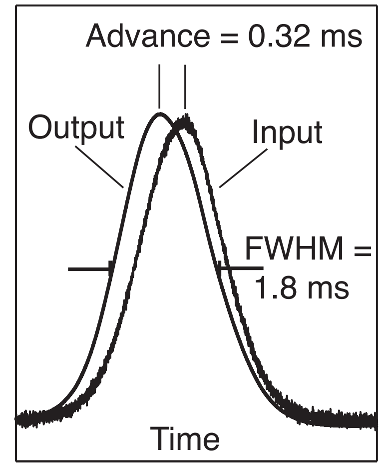


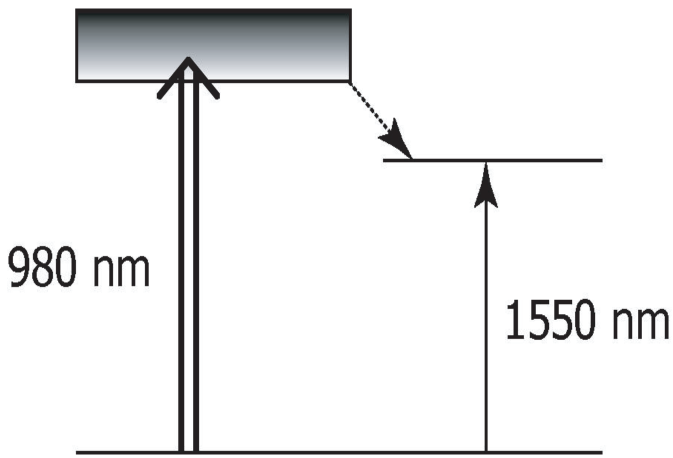
- Want a narrow feature in absorption profile to give a large $dn/d\omega$
- Ground state population oscillates at beat frequency δ (for $\delta < 1/T_1$).
- Population oscillations lead to decreased probe absorption (by explicit calculation), even though broadening is homogeneous.
- Ultra-slow light ($n_g > 10^6$) observed in ruby and ultra-fast light ($n_g = -4 \times 10^5$) observed in alexandrite.
- Slow and fast light effects occur at room temperature!

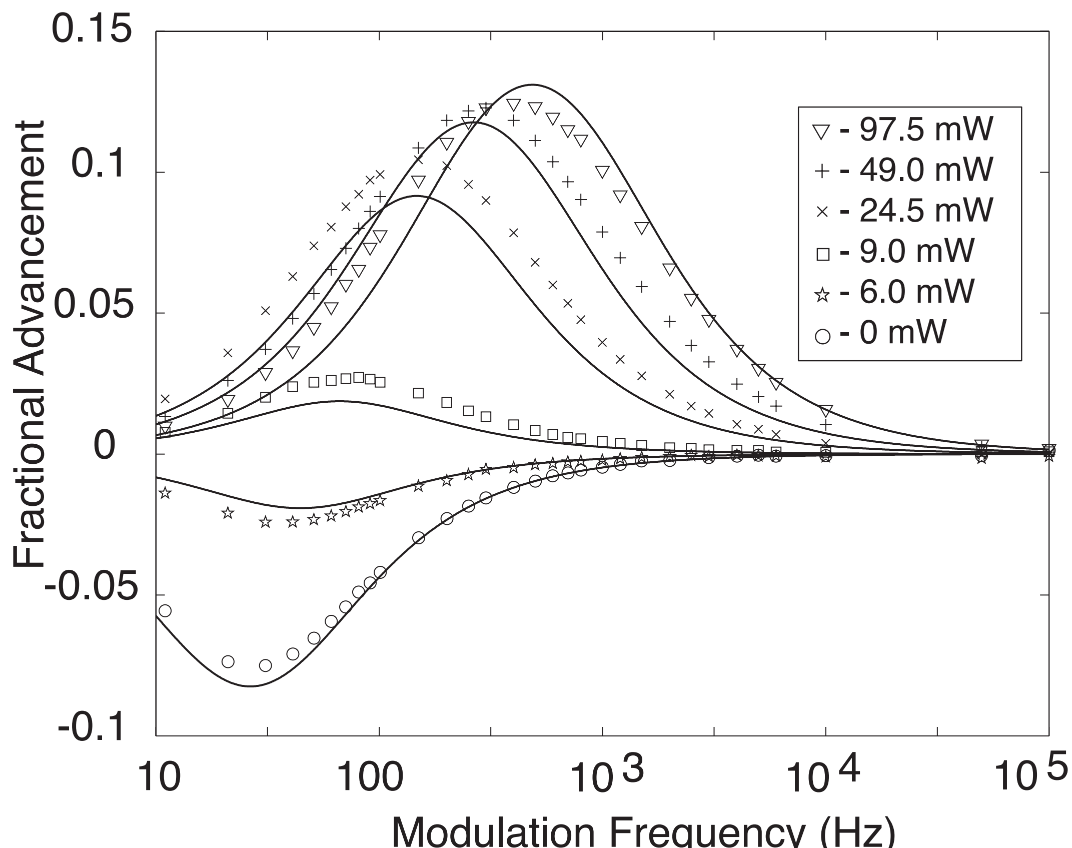
PRL 90,113903(2003); Science, 301, 200 (2003)

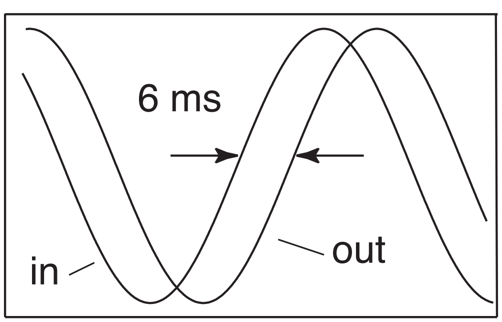
Slow and Fast Light in an Erbium Doped Fiber Amplifier

- Fiber geometry allows long propagation length
- Saturable gain or loss possible depending on pump intensity



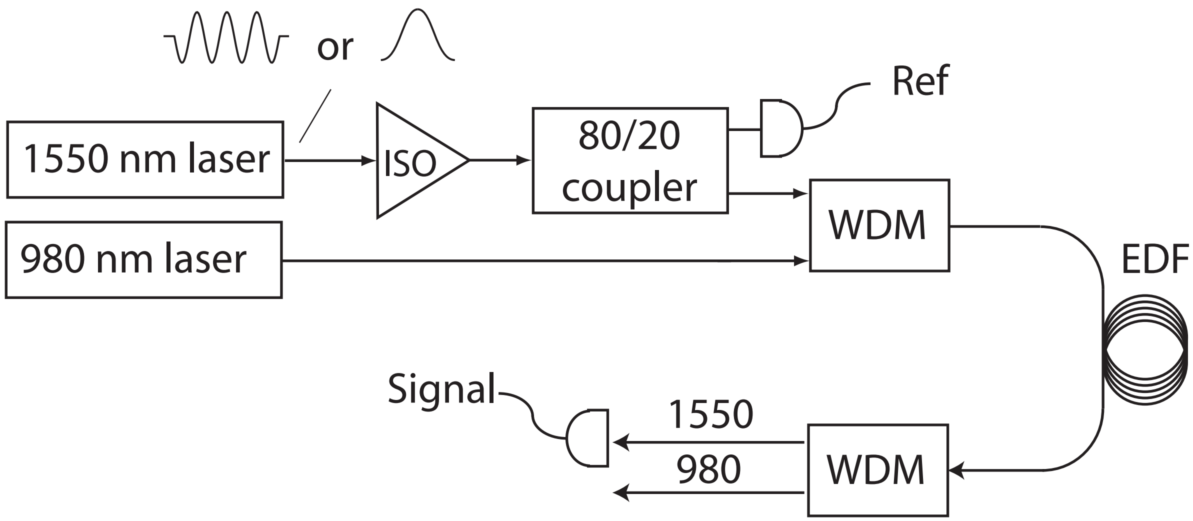






Schweinsberg, Lepeshkin, Bigelow, Boyd, and Jarabo, Europhysics Letters, 73, 218 (2006).

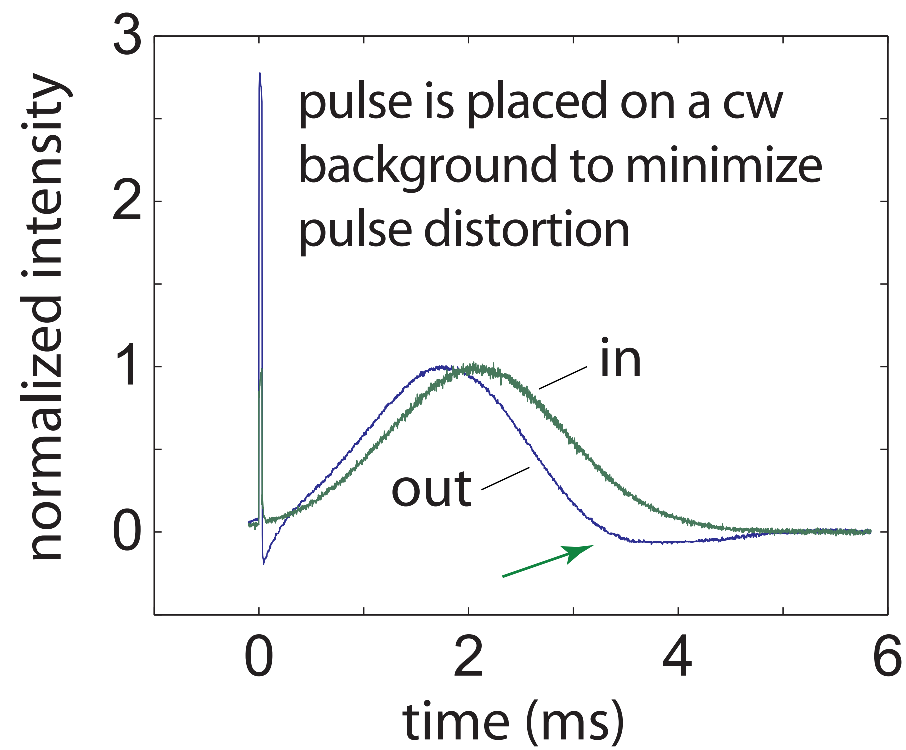
Observation of Backward Pulse Propagation in an Erbium-Doped-Fiber Optical Amplifier



We time-resolve the propagation of the pulse as a function of position along the erbium-doped fiber.

Procedure

- cutback method
- couplers embedded in fiber

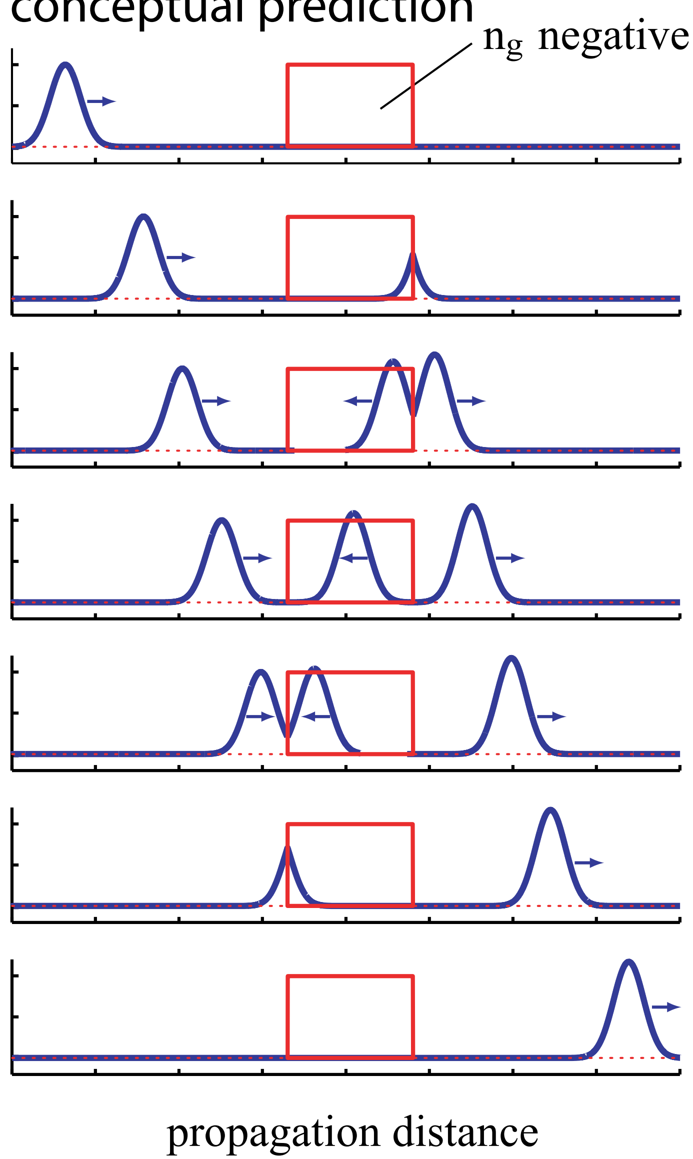


G. M. Gehring, A. Schweinsberg, C. Barsi, N. Kostinski, R. W. Boyd, Science 312, 985 2006.

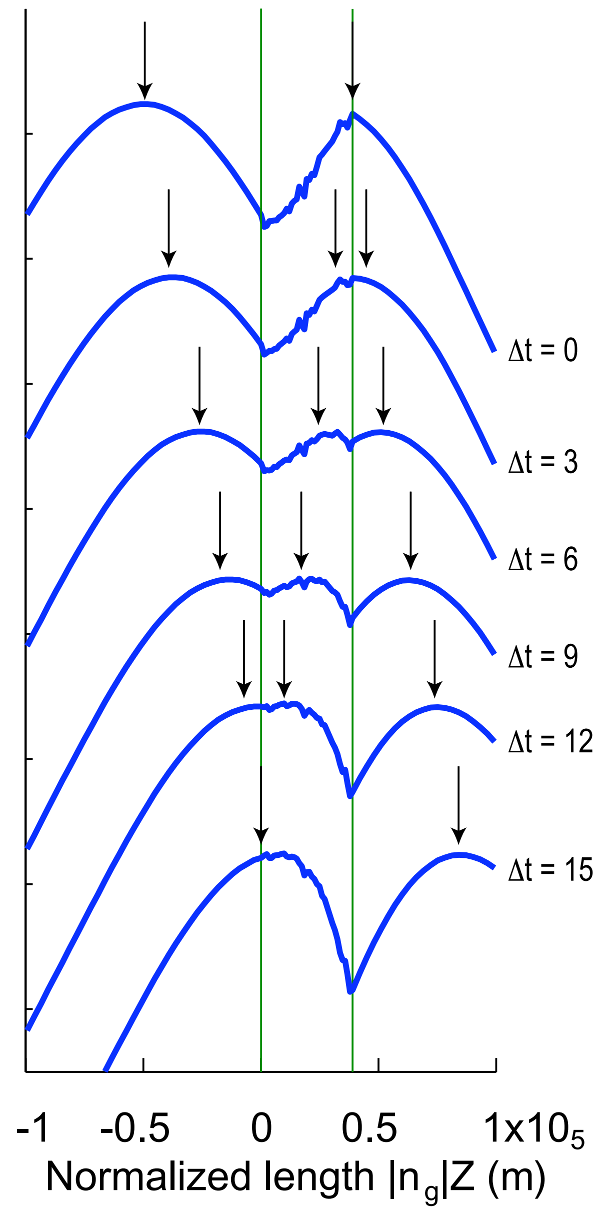
Observation of Superluminal and "Backwards" Pulse Propagation

- A strongly counterintuitive phenomenon
- But entirely consistent with established physics
- Predicted by Garrett and McCumber (1970) and Chiao (1993).
- Observed by Gehring, Schweinsberg, Barsi, Kostinski, and Boyd Science 312, 985 2006.

\- conceptual prediction



\- laboratory results

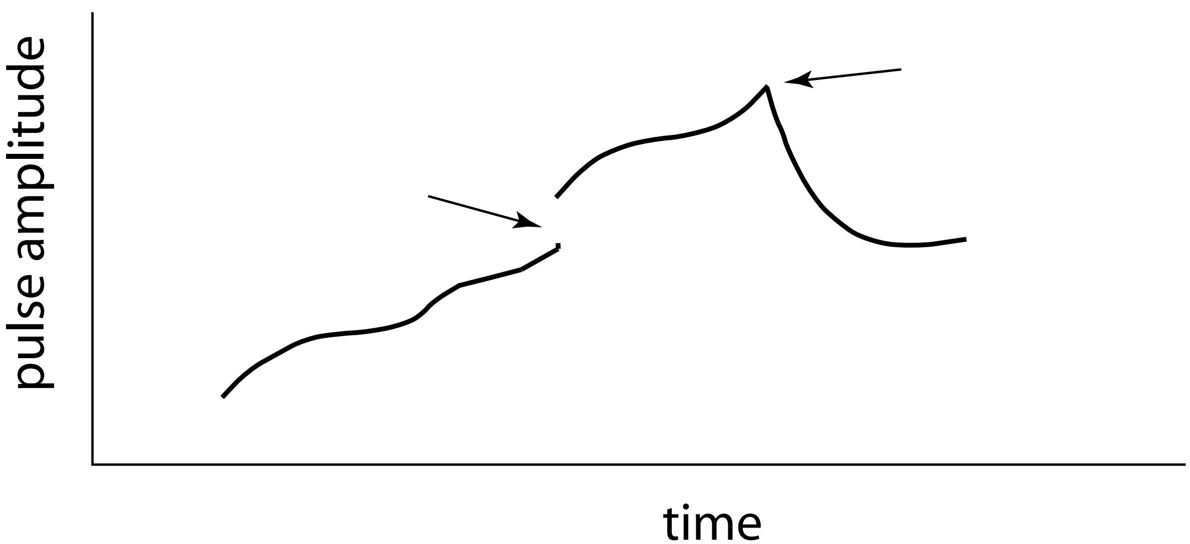


Causality?

- Superluminal ($v_g > c$) and backwards (v_g negative) propagation may seem counterintuitive but are fully compatible with causality.

- The group velocity is the velocity at which peak of pulse moves; it is not the "information velocity."

- It is believed that information is carried by points of nonalyticity of a waveform



- broad spectral content at points of discontinuity
- disturbance moves at vacuum speed of light

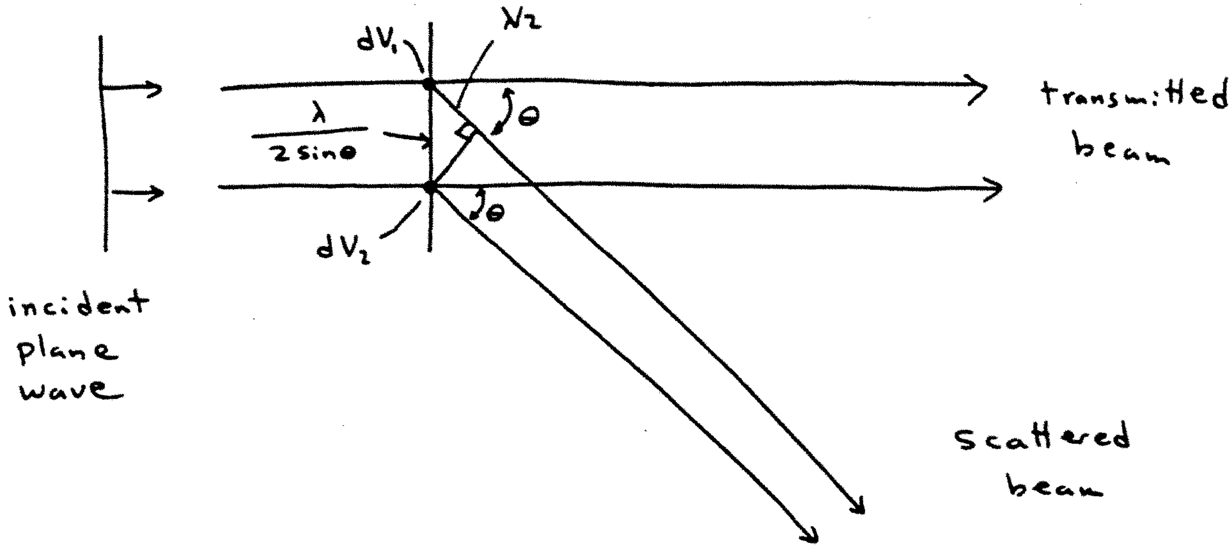
see, for instance, R. Y. Chiao

8. Spontaneous and Stimulated Light Scattering

Spontaneous vs. Stimulated Light Scattering

Light scattering can occur only due to fluctuations in the optical properties of materials.

Consider a completely homogeneous medium:



complete destructive interference!

Spontaneous light scattering

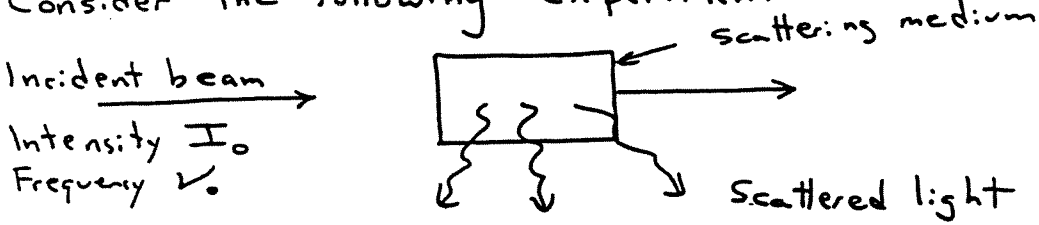
Fluctuations (e.g., in ε) occur due to thermal (or quantum mechanical zero-point) excitation.

Stimulated light scattering

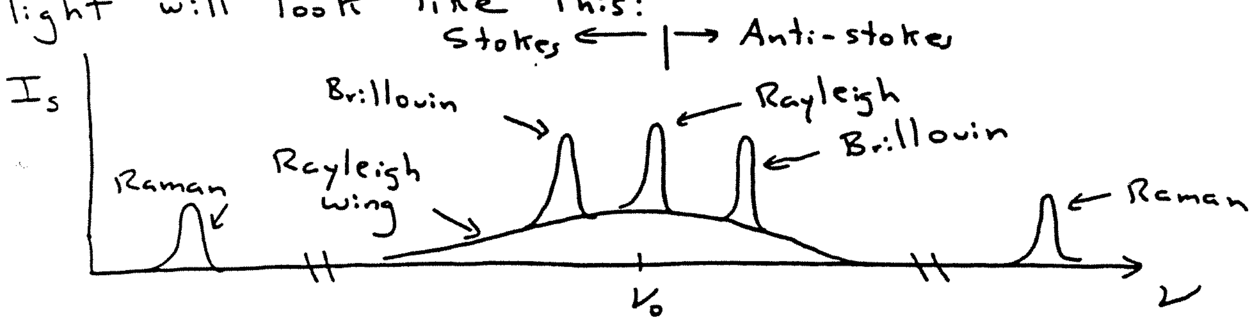
Fluctuations are induced by the incident laser field.

Spontaneous Light Scattering

Consider the following experiment:



In the most general case, the spectrum of the scattered light will look like this:

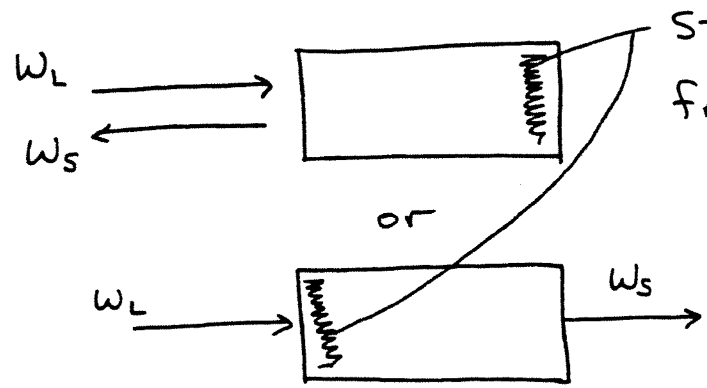


Process	Shift	Linewidth	Relaxation time	gain (cm/MW)
Raman	$1000\,cm^{-1}$	$5\,cm^{-1}$	$10^{-12}\,s$	5×10^{-3}
Brillouin	.1 - 1	5×10^{-3}	$10^{-9}\,s$	10^{-2}
Rayleigh	0	5×10^{-4}	$10^{-8}\,s$	10^{-4}
Rayleigh-wing	0	10	$10^{-12}\,s$	10^{-3}

Configurations for Stimulated Light Scattering

Generator

Apply only laser frequency



Stokes signal grows from noise.

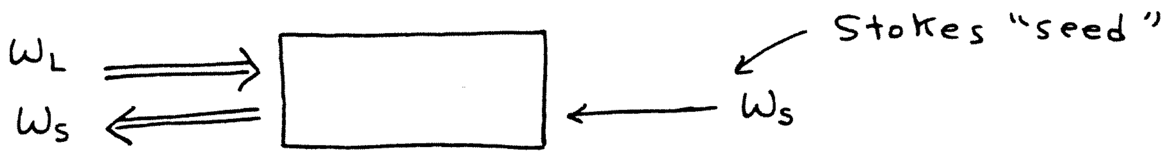
(Initiation due to thermal or quantum fluctuations)

(SBS occurs only in backward direction)
(SRS occurs in both forward and backward directions)

Stokes radiation is always emitted at the frequency of maximum gain.

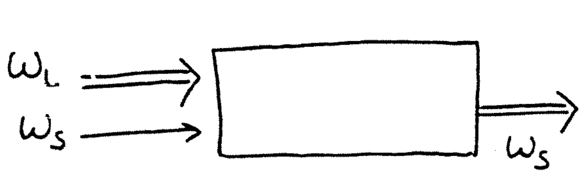
Amplifier

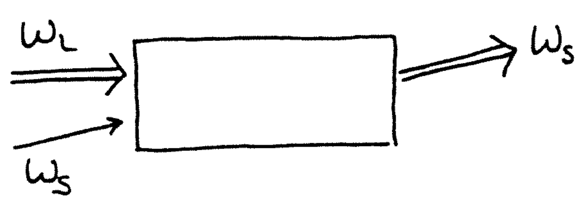
Apply both laser and Stokes frequency



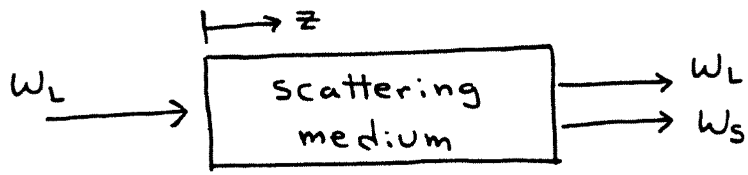
Stokes radiation is amplified at the expense of the laser (pump) radiation.

Alternative geometries:





Relation Between Spontaneous and Stimulated Light Scattering



$$\frac{dm_S}{dz} = D\, m_L\, (m_S + 1)$$

m_L: # of laser photons per mode; m_S: # of Stokes photons per mode

For $m_S \ll 1$

$$m_S(z) = m_S(0) + D\, m_L\, z$$

linear increase with length of scattering medium
$\Rightarrow$ spontaneous scattering

For $m_S \gg 1$

$$m_S(z) = m_S(0)\, e^{Gz}$$

exponential growth
$\Rightarrow$ stimulated scattering

$$G = D\, m_L$$

We can relate G to the spontaneous scattering cross section:

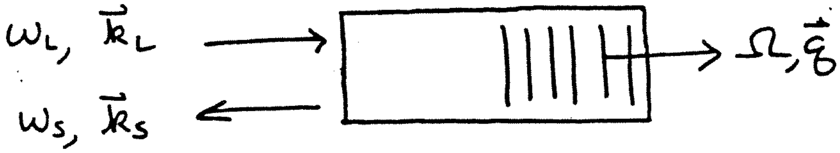
$$G = \frac{4\pi^3 N c^2}{\hbar\, \omega_L\, \omega_S^2\, n_S^2} \left(\frac{\partial^2 \sigma}{\partial \omega_S\, \partial \Omega} \right) I_L$$

(R.W. Hellwarth, Phys. Rev 130 1850, 1963)

Stimulated Scattering Processes

Stimulated Brillouin Scattering

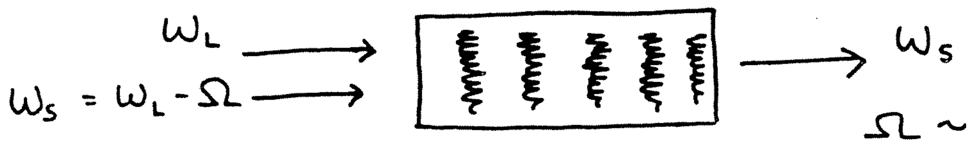
interaction of light with acoustic waves
(coupling can be electrostrictive or thermal)



$$\omega_S = \omega_L - \Omega$$
$$\Omega = q v$$
$$\vec{q} = \vec{k}_L - \vec{k}_S$$

Stimulated Rayleigh Scattering

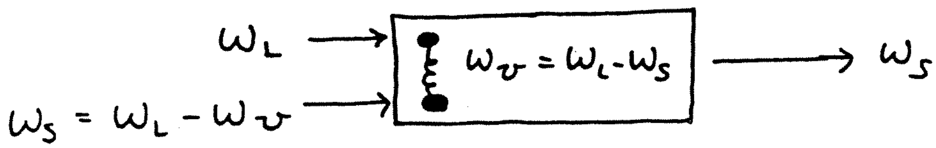
interaction with nonpropagating density (temperature) waves
(coupling can be electrostrictive of thermal)



$$\Omega \sim 1/\tau_{thermal}$$

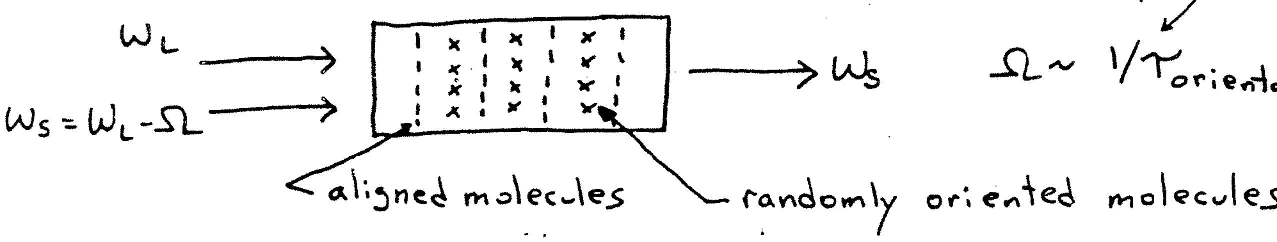
Stimulated Raman Scattering

interaction with vibrational degree of freedom



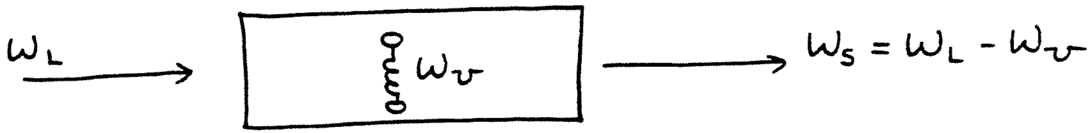
Stimulated Rayleigh-Wing Scattering

interaction with orientational degree of freedom

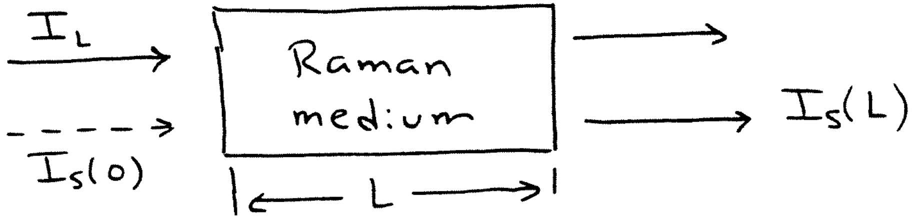


$$\Omega \sim 1/\tau_{orientation}$$ ($\tau_{orientation} \sim 2$ ps)

Stimulated Raman Scattering



We can predict the Stokes output intensity :



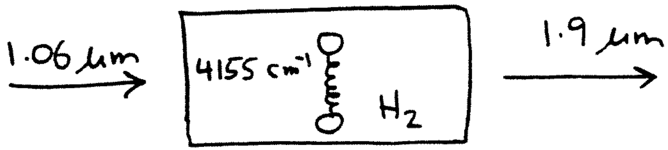
$$I_s(L) = I_s(0)\, e^{g I_L L}$$

g = Raman gain factor

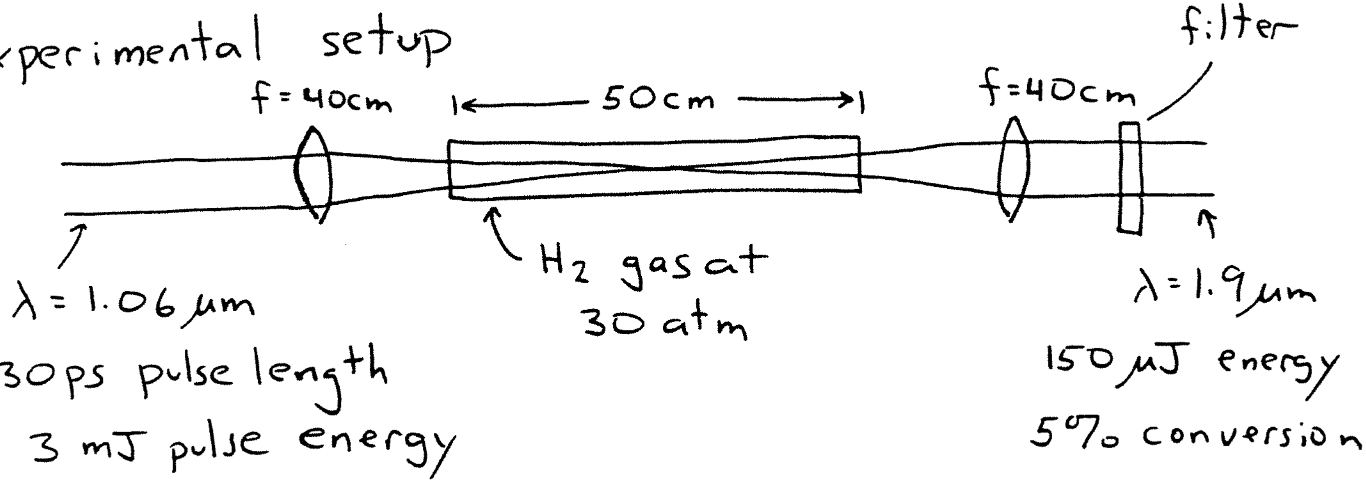
$\simeq 10^{-9}$ cm/W for most materials

Example of Raman Frequency Shifter

Need 1.9 μm light



Experimental setup



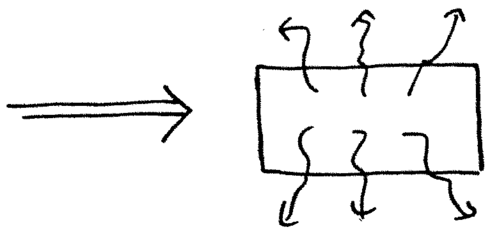
Typical Raman media

material	$\Delta\nu$ (cm^{-1})	gain g x 10^3 (cm/MW)
LIQUIDS		
benzene	992	3
water	3290	0.14
N_2	2326	17
O_2	1555	16
GASES		
methane	2916	0.66 (10 atm, 500 nm)
hydrogen	4155 (vibrational)	1.5 (above 10 atm)
	450 (rotational)	0.5 (above 0.5 atm)
deuterium	2991 (vibrational)	1.1 (above 10 atm)
N_2	2326	0.071 (10 atm, 500 nm)
O_2	1555	0.016 (10 atm, 500 nm)

U. Simon and F. K. Tittel, Nonlinear Optical Frequency Conversion Techniques, in *Methods of Experimental Physics, Vol. III (Lasers and Optical Devices)*, R. G. Hulet, and F. B. Dunning, eds., Academic Press, 1994.

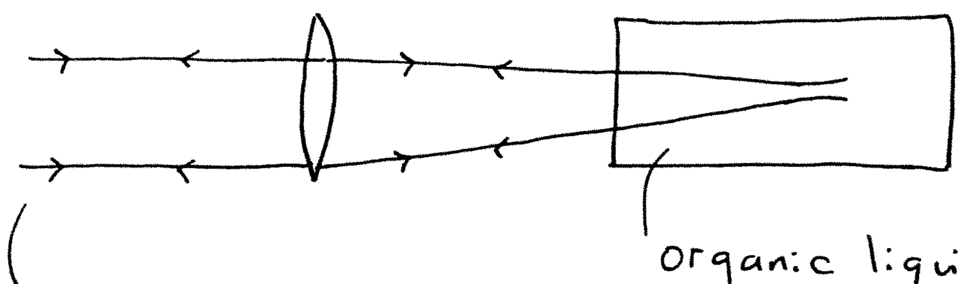
Stimulated Brillouin Scattering

- Spontaneous Brillouin scattering is a very weak process.



[~1 part in 10^5 of incident scattered per cm of liquid.]

- But Stimulated Brillouin scattering can be very efficient (>50%)



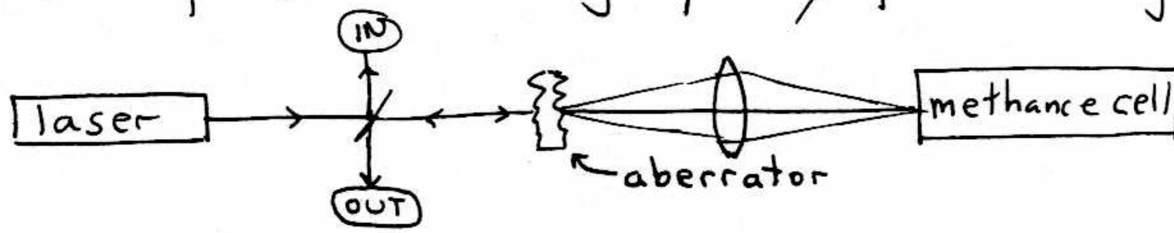
organic liquid (CS_2)

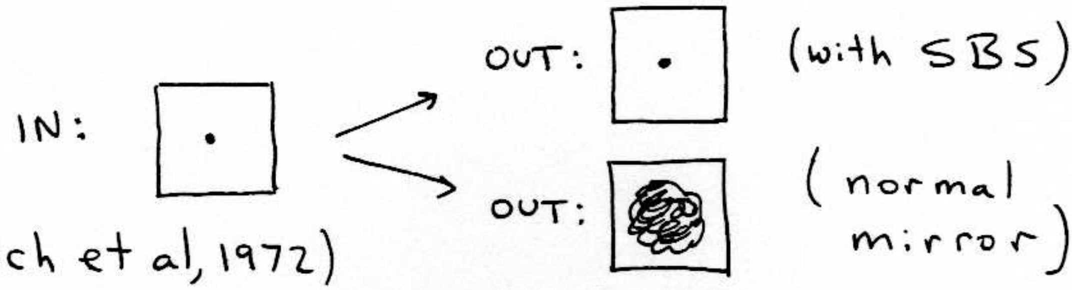
$P \gtrsim 20$ kW

$T \gtrsim 5$ nsec

Phase Conjugation by SBS

SBS can produce high quality phase conjugation



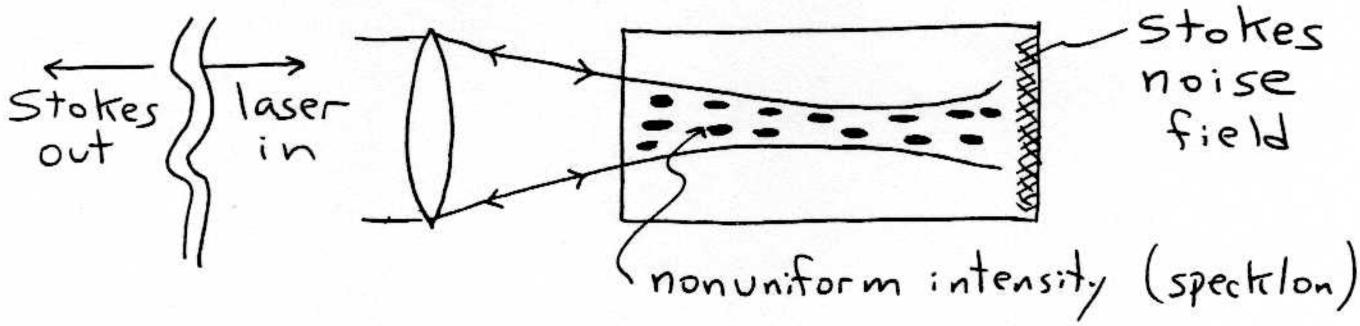


(Zel'dovich et al, 1972)

But Why?

$$\frac{dI_s}{dz} = -g\, I_L\, I_s$$

↖ No explicit dependence on the phase of laser!



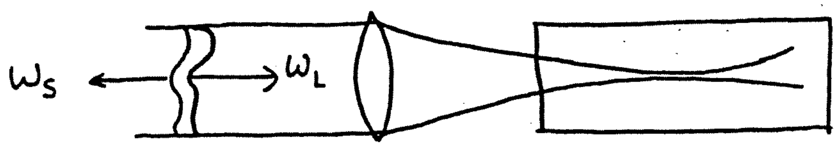
Non-uniform distribution of intensity
⇒ non-uniform distribution of gain.

Stokes field is most efficiently amplified if it spatially overlaps the gain distribution.
Stokes field grows from noise

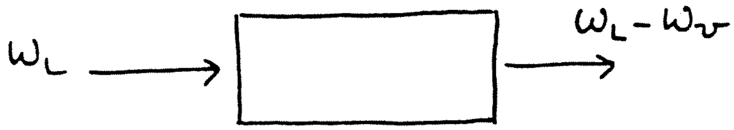
⇒ Stokes output wavefronts coincide with input laser wavefronts
⇒ $E_s \propto E_L^*$

Uses of Stimulated Light Scattering

Optical Phase Conjugation (by SBS)

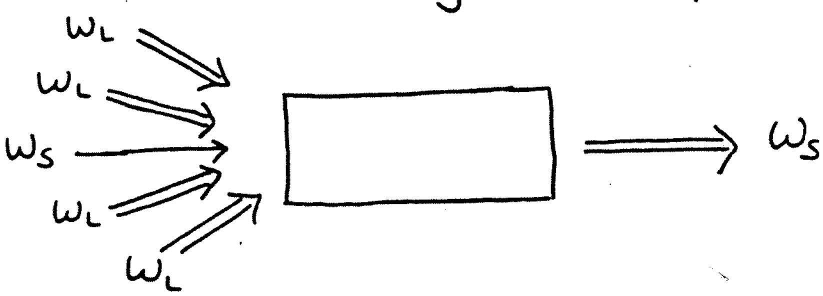


Laser Frequency Shifting (by SRS)

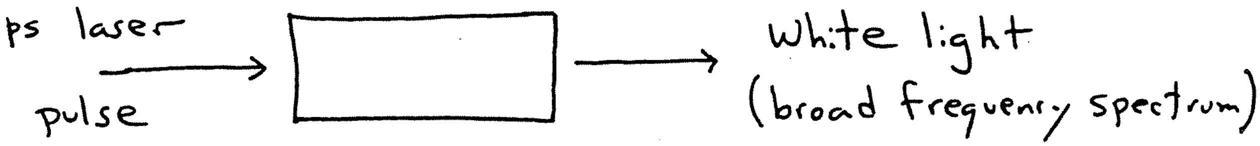


Can shift UV excimer laser radiation to visible

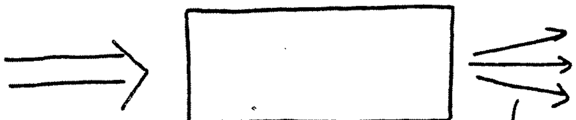
Laser Beam Combining (by SRS)



White-Light Generation (?)

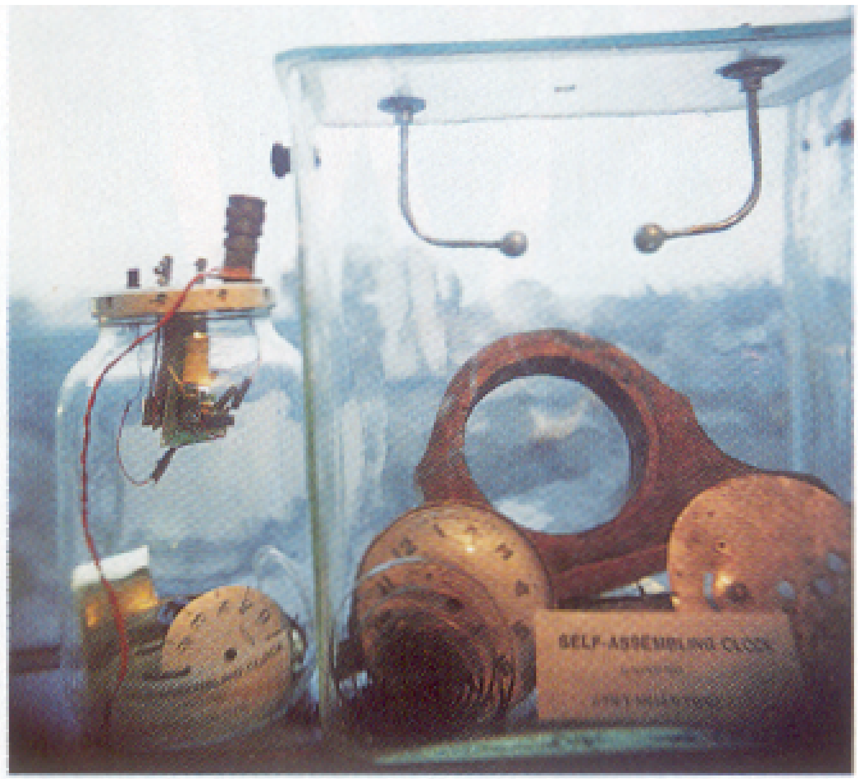


Thermal Blooming (Stimulated Thermal Rayleigh Scattering)



(new components experience growth by stimulated scattering

Experiment in Self Assembly



Joe Davis, MIT

Thank you for your attention!