Tutorial on Nonlinear Optics

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The visuals of this talk are available at boydnlo.ca/presentations/

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Why Study Nonlinear Optics?

It is good fundamental physics.

It leads to important applications.

It is a lot of fun.

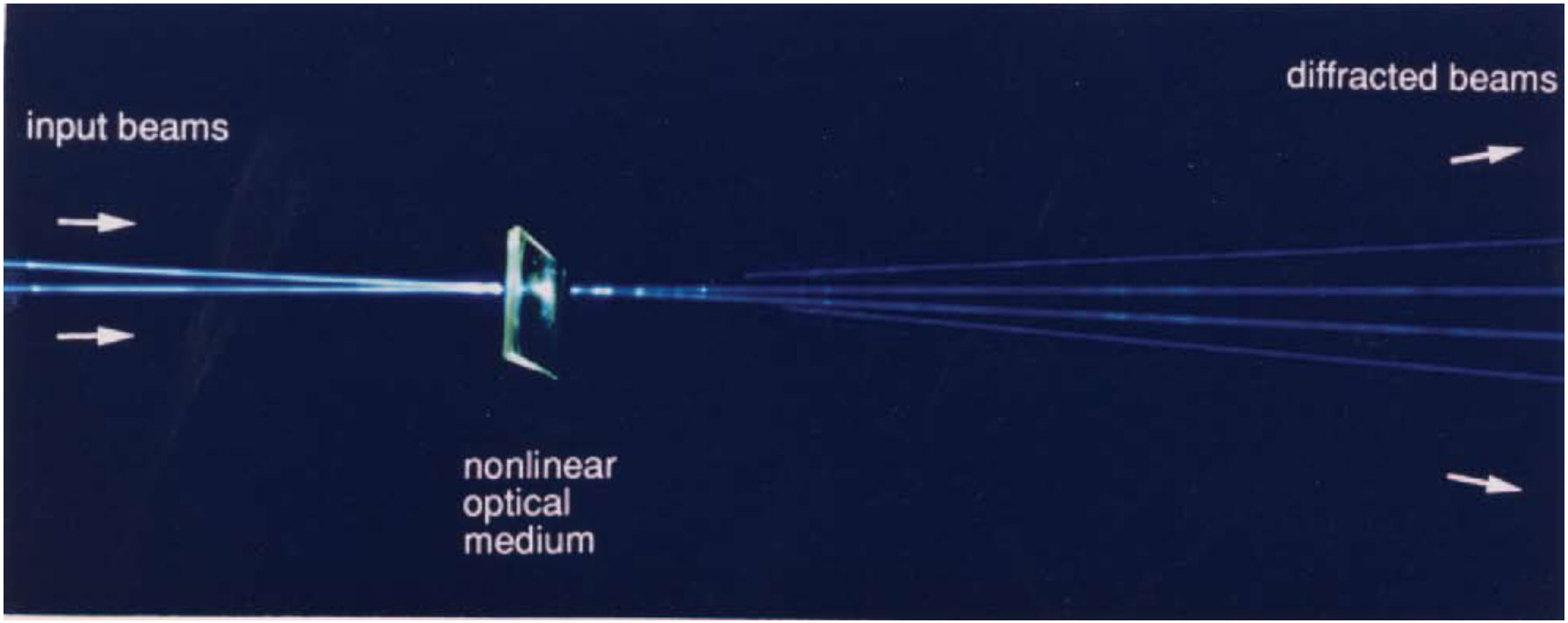
Demonstrate these features with examples in remainder of talk.

Tutorial on Nonlinear Optics

1. What is Nonlinear Optics?
2. Coupled Wave Equations and Harmonic Generation
3. Mechanisms of Optical Nonlinearity
4. Self-Action Effects in Nonlinear Optics
5. NLO of Epsilon-Near Zero Materials
6. Local-Field Effects in Nonlinear Optics
7. Composite Materials and Metamaterials for NLO
8. Slow and Fast Light
9. Spontaneous and Stimulated Light Scattering

1. What is Nonlinear Optics?

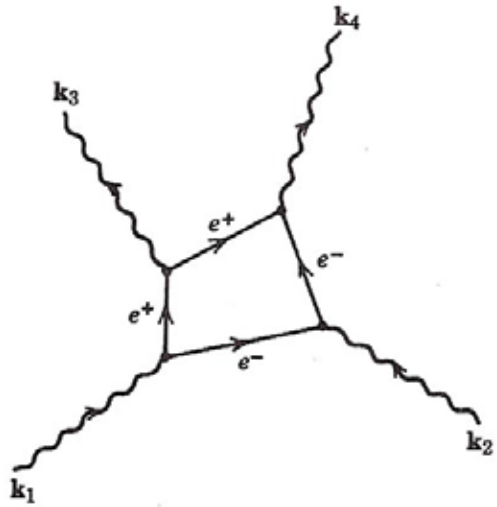
Nonlinear Optics and Light-by-Light Scattering



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)

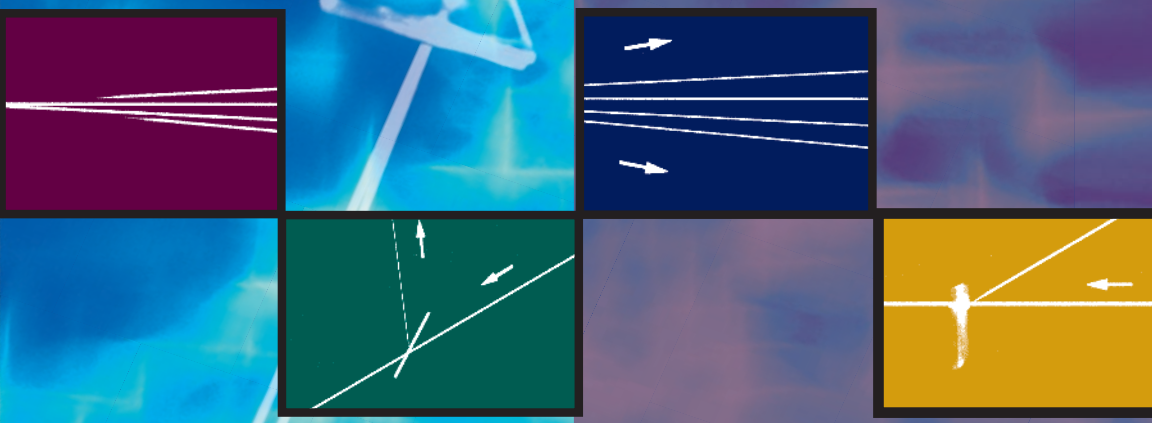
n_2 (FBAG) $\approx 10^{14}$ n_2(silica) [But very slow response!]



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

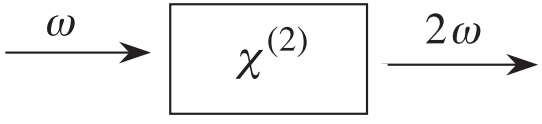
Simple Formulation of the Theory of Nonlinear Optics

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

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

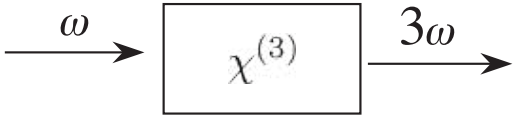
$\chi^{(1)}$ describes linear optics, e.g., how lenses work:

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

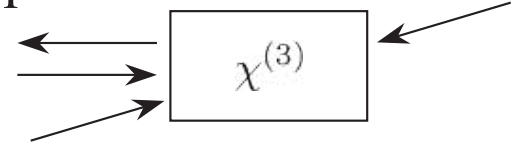


$\chi^{(3)}$ describes third-order effects such as third-harmonic generation, four-wave mixing, and the intensity dependence of the index of refraction.

THG



FWM

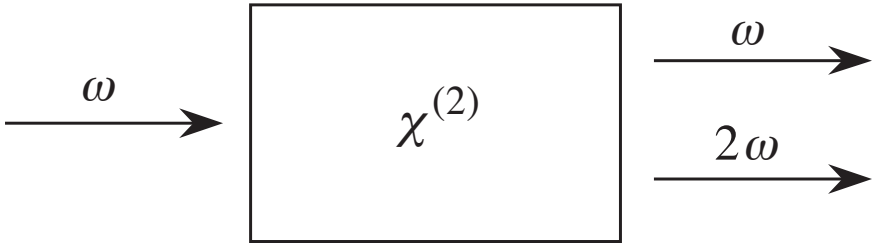


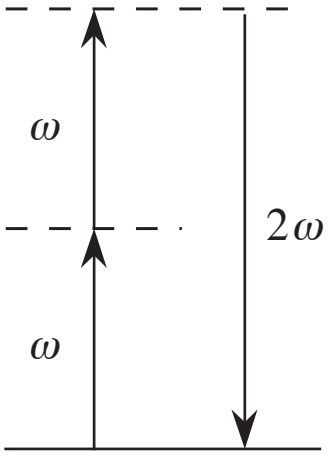
NL index

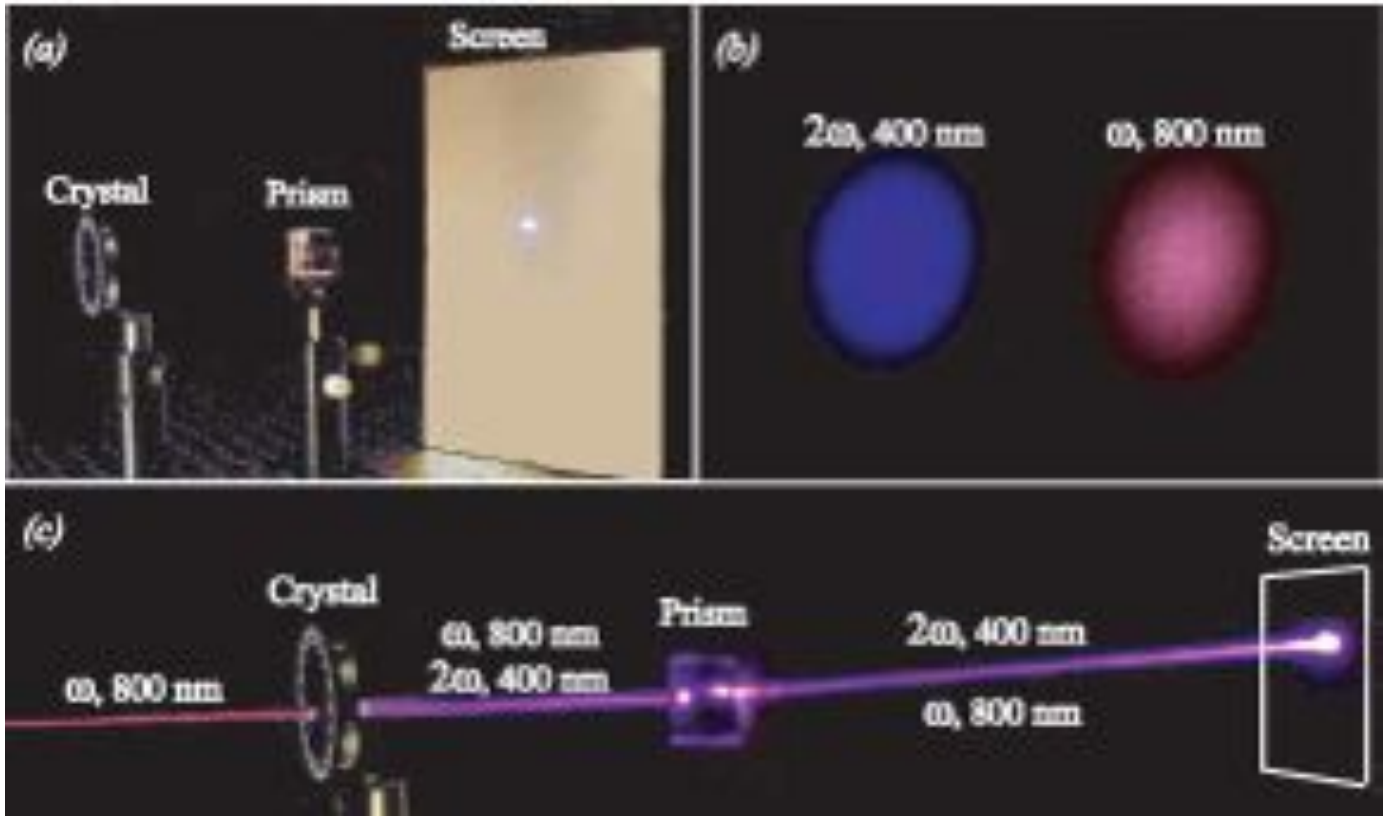
$$n = n_0 + n_2 I \quad \text{where} \quad n_2 = \frac{3}{4n_0^2\epsilon_0 c}\chi^{(3)}$$

Some Fundamental Nonlinear Optical Processes: I

Second-Harmonic Generation



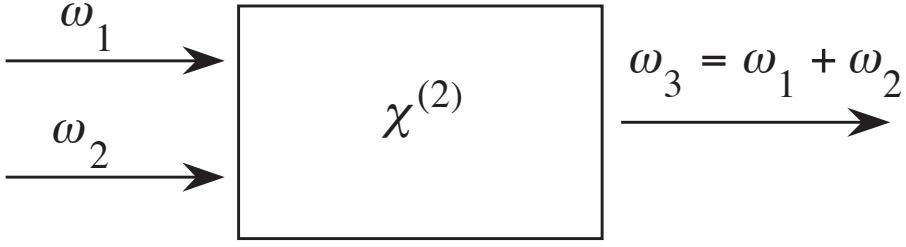


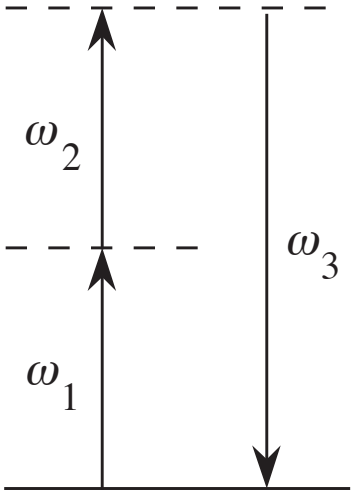


Dolgaleva, Lepeshkin, and Boyd

Some Fundamental Nonlinear Optical Processes: II

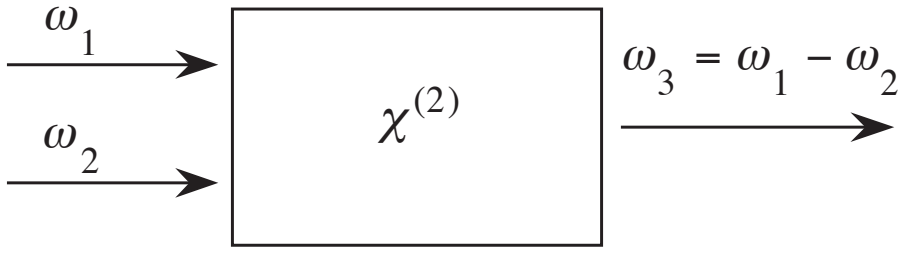
Sum-Frequency Generation

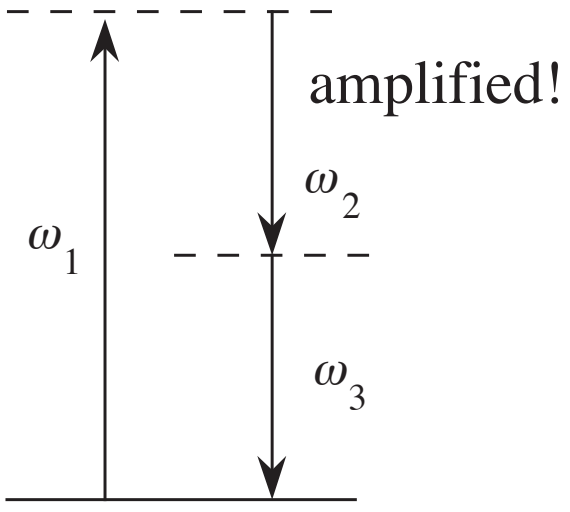




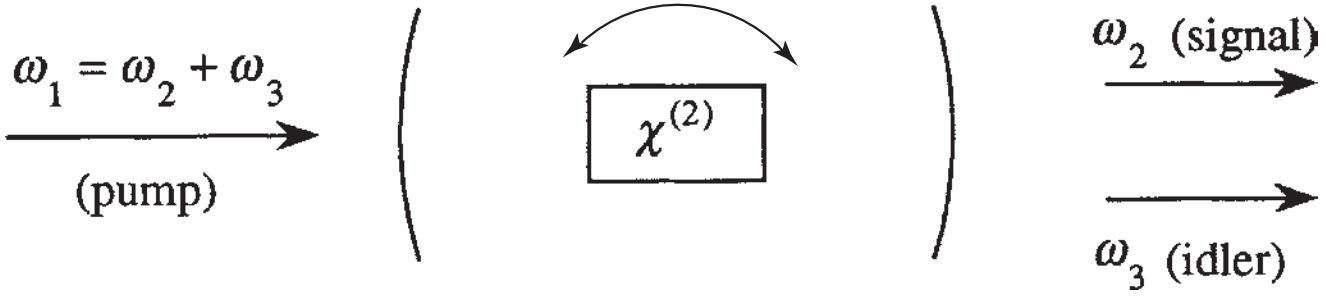
Some Fundamental Nonlinear Optical Processes: III

Difference-Frequency Generation

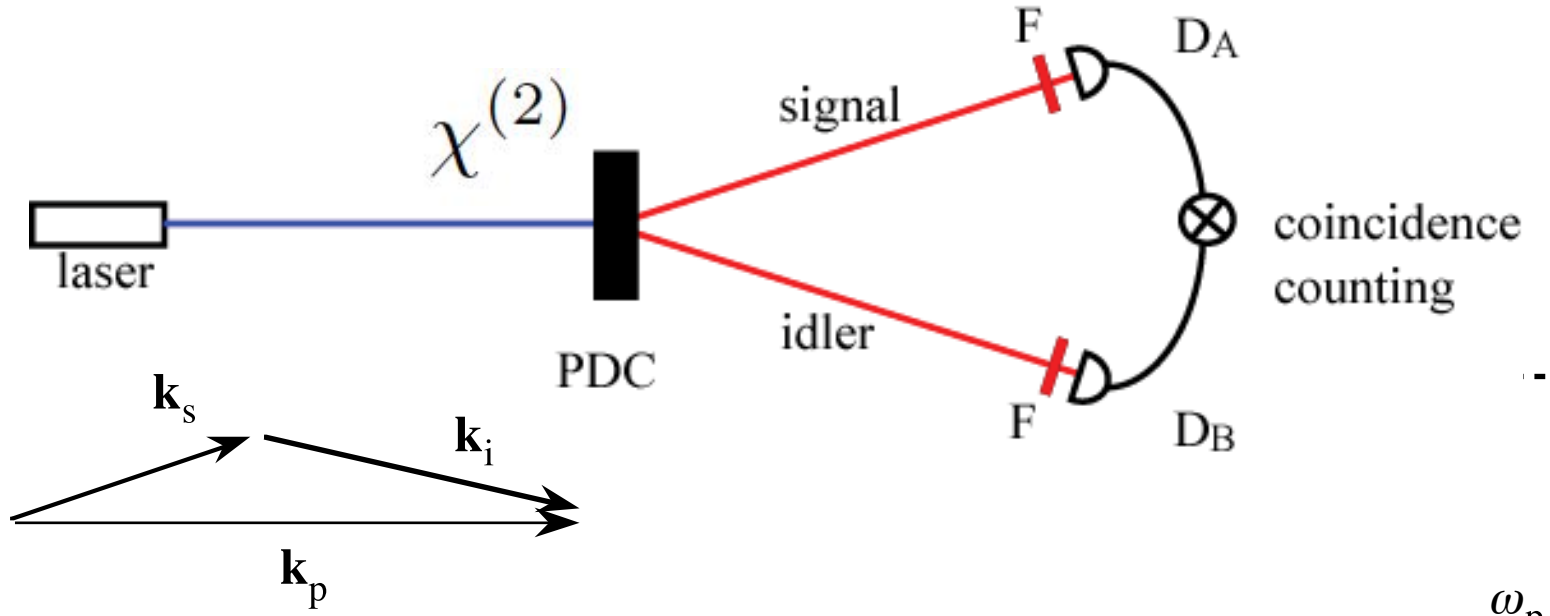


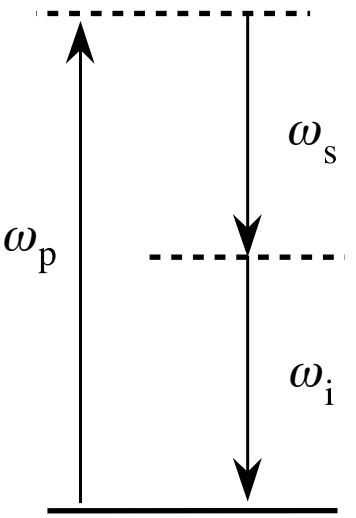


Optical Parametric Oscillation



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

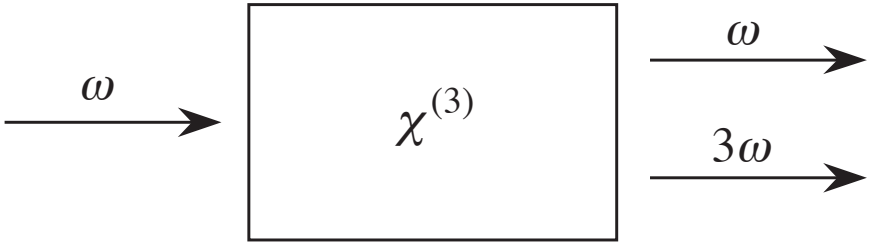
Entanglement is important for:

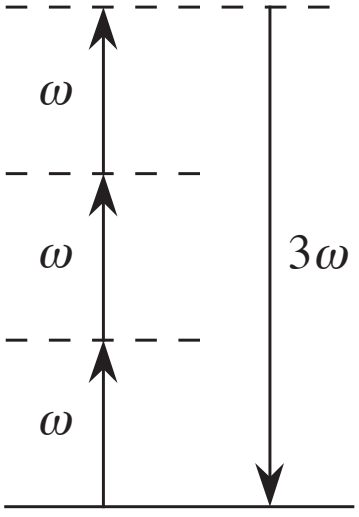
(a) Fundamental tests of QM (e.g., nonlocality)

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

Some Fundamental Nonlinear Optical Processes: III

Third-Harmonic Generation

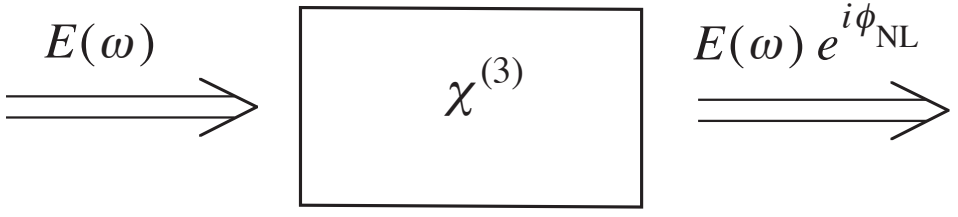




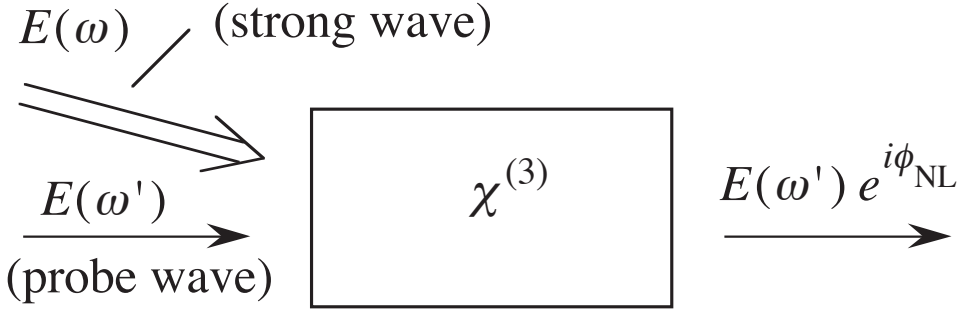
Some Fundamental Nonlinear Optical Processes: IV

Intensity-Dependent Index of Refraction

- Single beam:
  self-phase modulation



- Two beam:
  cross-phase modulation



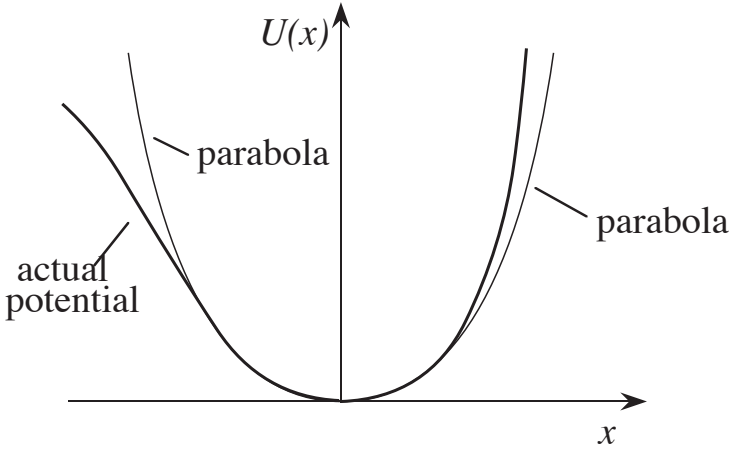
The refractive index is given by

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

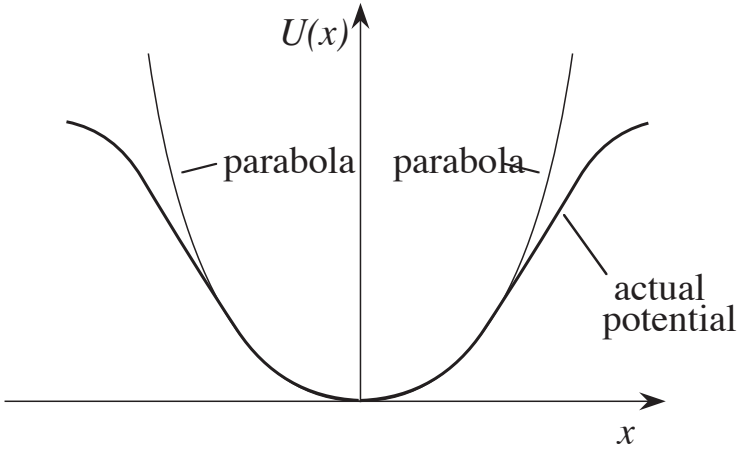
Role of Material Symmetry in Nonlinear Optics

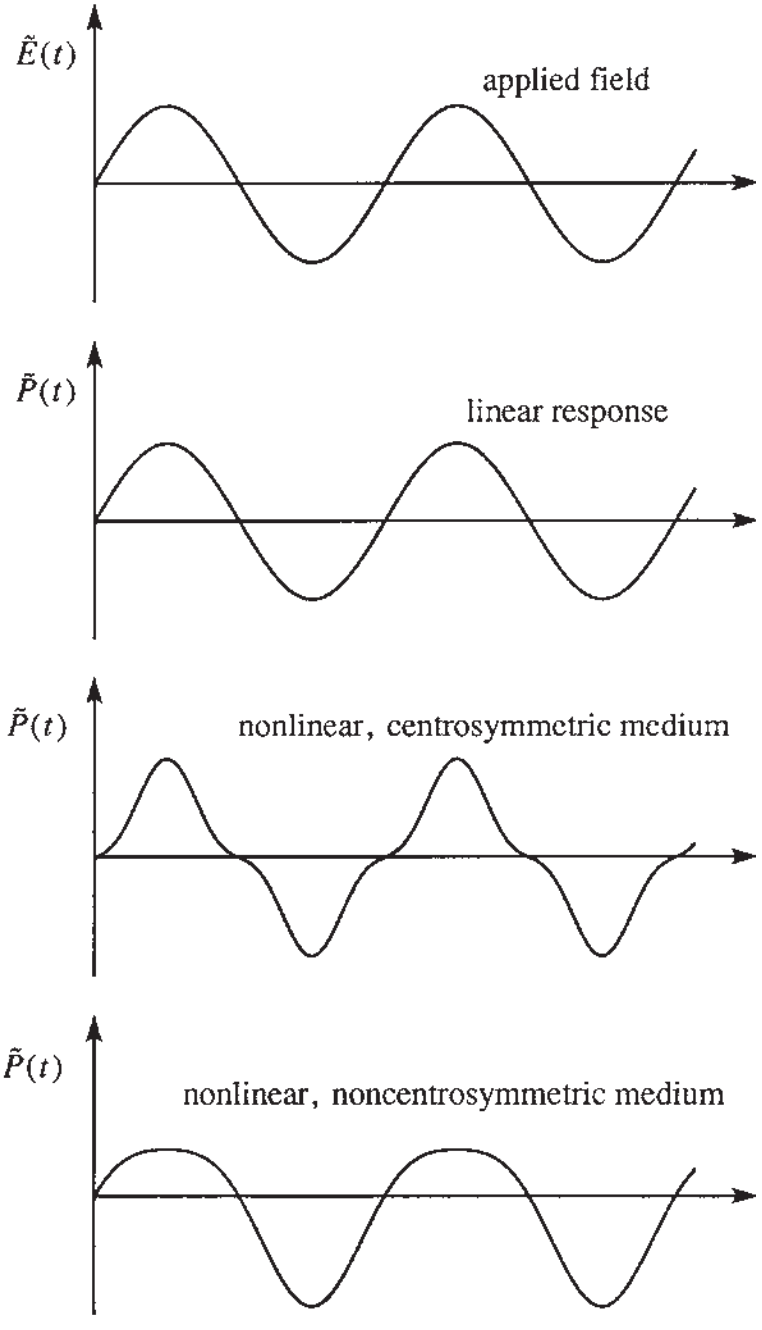
$\chi^{(2)}$ vanishes identically for a material possessing a center of inversion symmetry (a centrosymmetric medium).

non-centrosymmetric medium



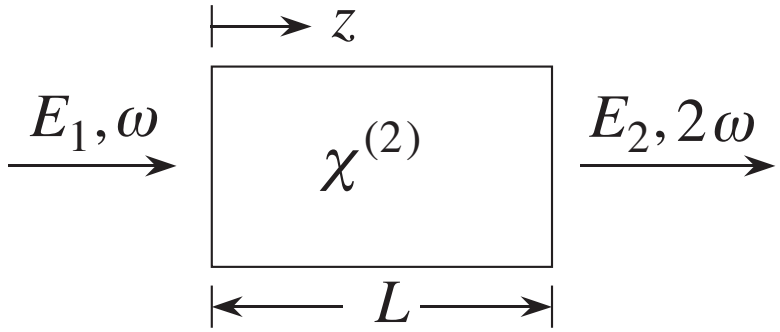
centrosymmetric medium





2. Coupled Wave Equations and Harmonic Generation

Treatment of Second-Harmonic Generation – I



Let

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

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

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

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

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

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

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

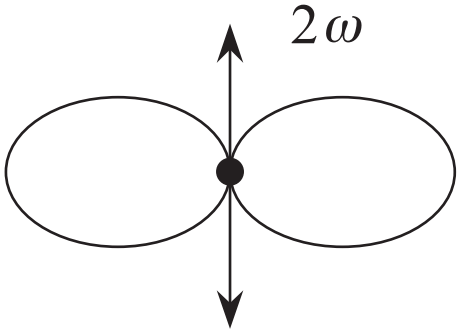
For the case $\Delta k = 0$, Eq. (8) becomes

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

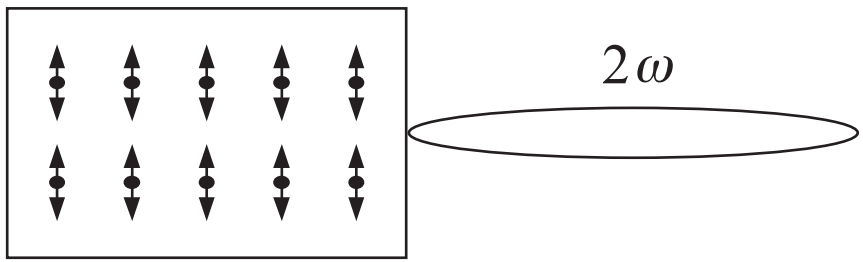
with solution evaluated at $z = L$ of

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

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



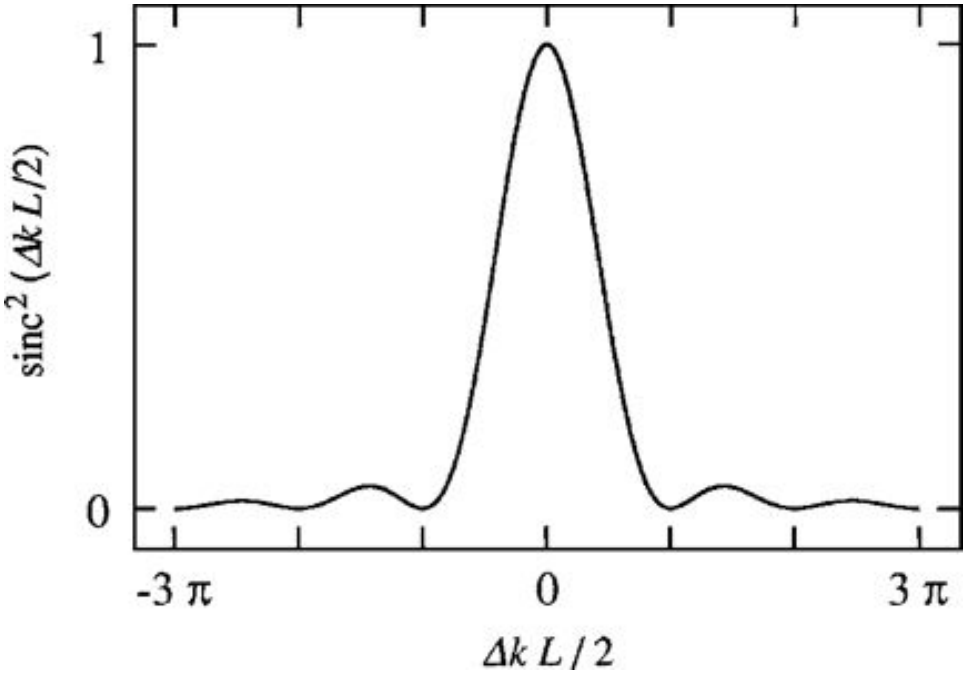
phased array of dipoles

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!

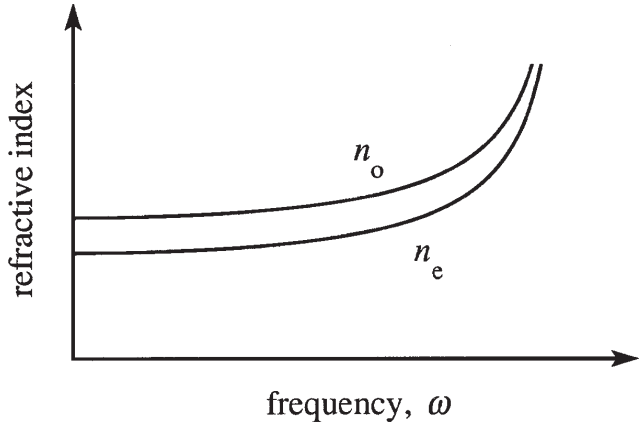
Curley et al., not Curly et al.

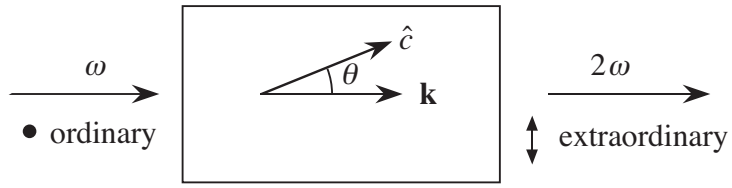
How to Achieve Phase Matching: Birefringence Phase Matching

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

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

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

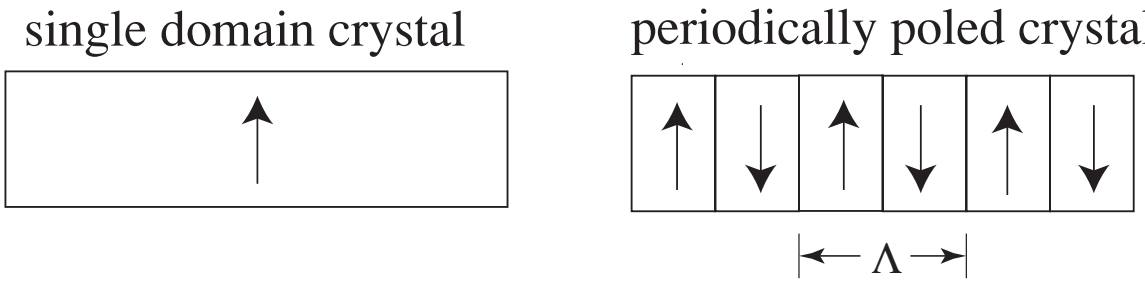




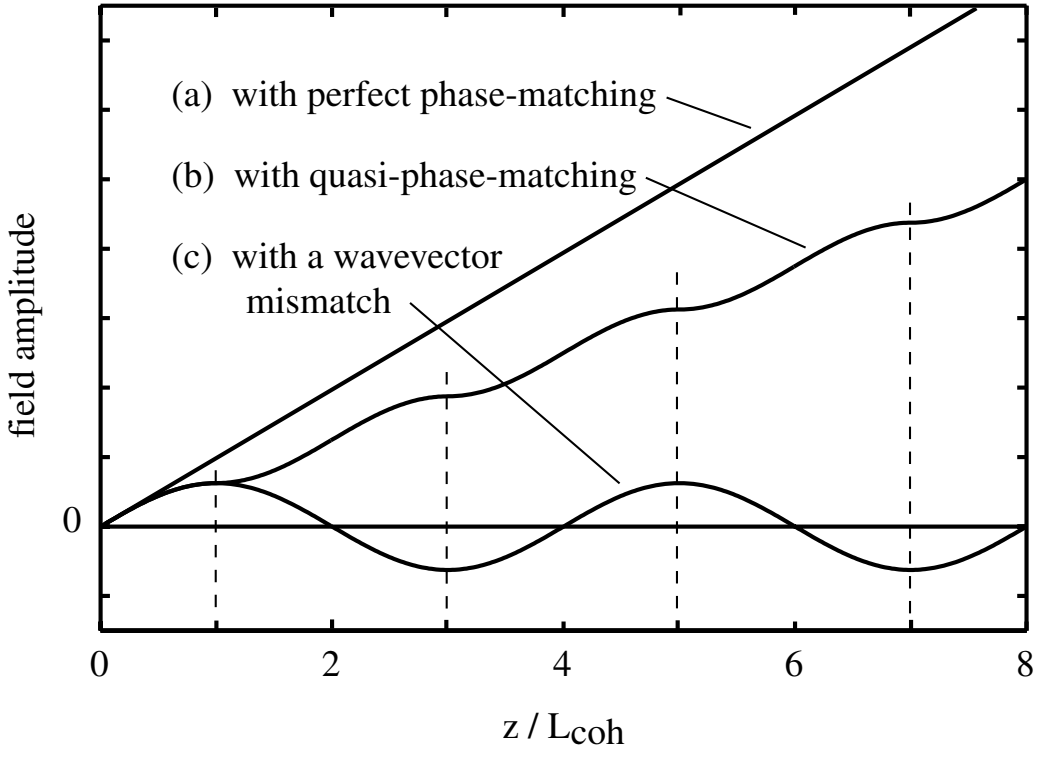
Midwinter and Warner showed that there are two ways to achieve phase matching:

	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



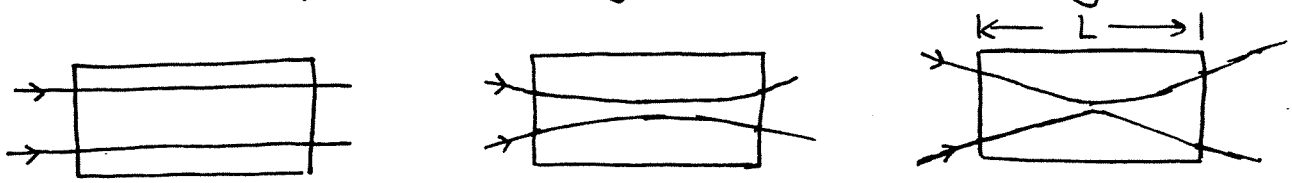
Sign of $\chi^{(2)}$ is periodically inverted to prevent reverse power flow.



Nonlinear Optics with Focused Laser Beams

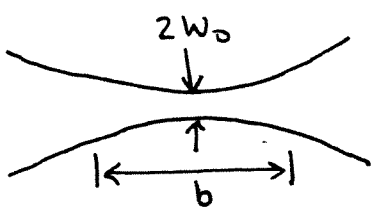
(Real experiments don't use infinite plane waves)

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)	$\chi^{(3)}_{1111}$ ($\mathrm{m}^2/\mathrm{V}^2$)	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

$$\chi^{(2)}_{ijk}(\omega_p+\omega_q,\omega_q,\omega_p)$$

$$= \frac{N}{2\,\epsilon_0\hbar^2}\sum_{lmn}\rho^{(0)}_{ll}\left\{\frac{\mu^i_{ln}\mu^j_{nm}\mu^k_{ml}}{[(\omega_{nl}-\omega_p-\omega_q)-i\gamma_{nl}][(\omega_{ml}-\omega_p)-i\gamma_{ml}]}\right. \qquad (a_1)$$

$$+\frac{\mu^i_{ln}\mu^k_{nm}\mu^j_{ml}}{[(\omega_{nl}-\omega_p-\omega_q)-i\gamma_{nl}][(\omega_{ml}-\omega_q)-i\gamma_{ml}]} \qquad (a_2)$$

$$+\frac{\mu^k_{ln}\mu^i_{nm}\mu^j_{ml}}{[(\omega_{mn}-\omega_p-\omega_q)-i\gamma_{mn}][(\omega_{nl}+\omega_p)+i\gamma_{nl}]} \qquad (a'_1)$$

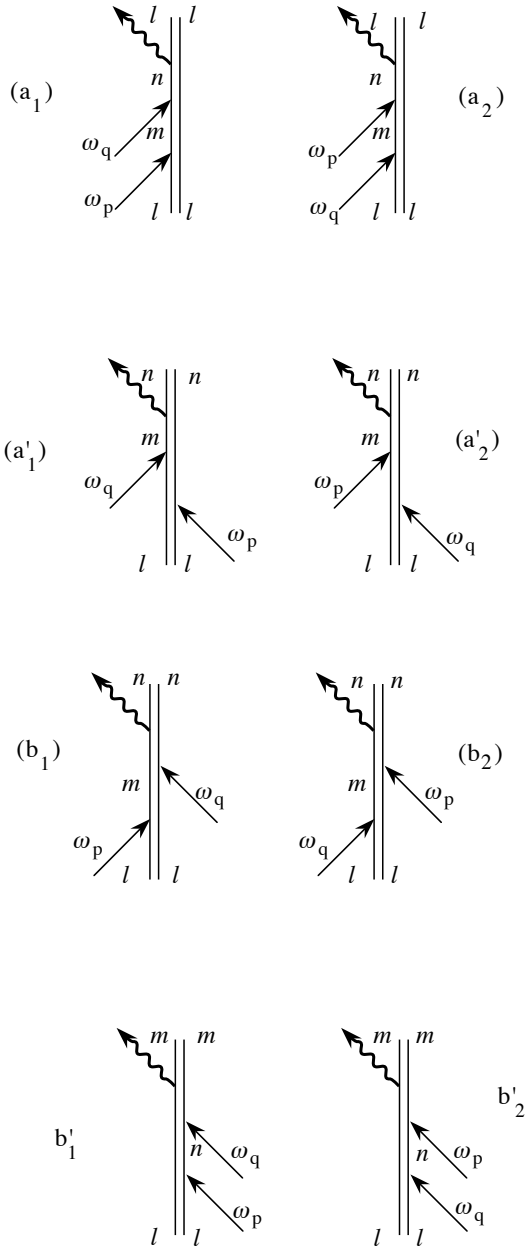
$$+\frac{\mu^j_{ln}\mu^i_{nm}\mu^k_{ml}}{[(\omega_{mn}-\omega_p-\omega_q)-i\gamma_{mn}][(\omega_{nl}+\omega_q)+i\gamma_{nl}]} \qquad (a'_2)$$

$$+\frac{\mu^j_{ln}\mu^i_{nm}\mu^k_{ml}}{[(\omega_{nm}+\omega_p+\omega_q)+i\gamma_{nm}][(\omega_{ml}-\omega_p)-i\gamma_{ml}]} \qquad (b_1)$$

$$+\frac{\mu^k_{ln}\mu^i_{nm}\mu^j_{ml}}{[(\omega_{nm}+\omega_p+\omega_q)+i\gamma_{nm}][(\omega_{ml}-\omega_q)-i\gamma_{ml}]} \qquad (b_2)$$

$$+\frac{\mu^k_{ln}\mu^j_{nm}\mu^i_{ml}}{[(\omega_{ml}+\omega_p+\omega_q)+i\gamma_{ml}][(\omega_{nl}+\omega_p)+i\gamma_{nl}]} \qquad (b'_1)$$

$$\left.+\frac{\mu^j_{ln}\mu^k_{nm}\mu^i_{ml}}{[(\omega_{ml}+\omega_p+\omega_q)+i\gamma_{ml}][(\omega_{nl}+\omega_q)+i\gamma_{nl}]}\right\} \qquad (b'_2)$$



Nonresonant Electronic Nonlinearities

Estimate size:

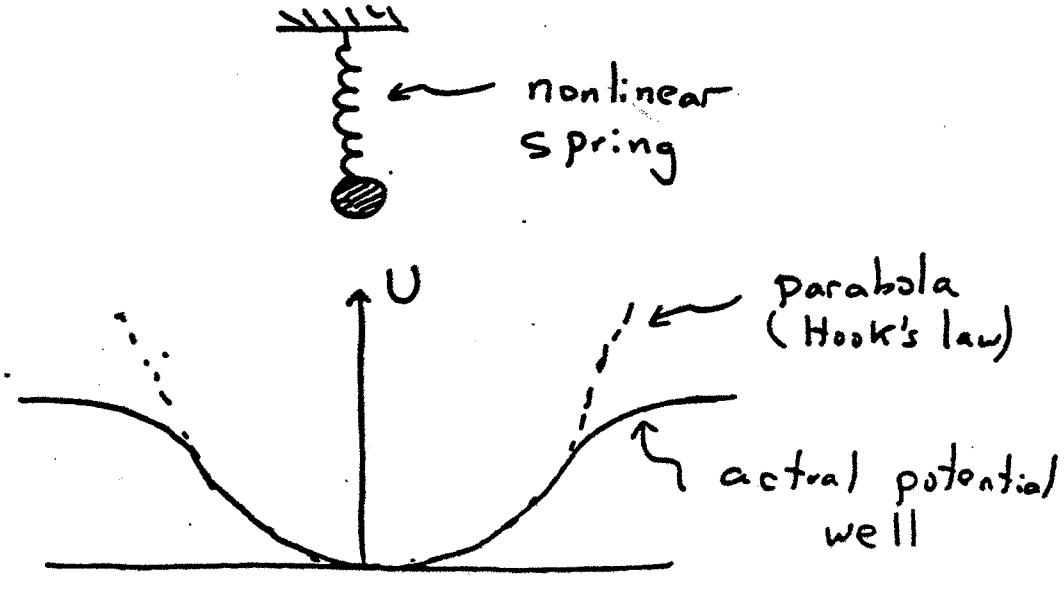
$$P = \chi^{(1)} E + \chi^{(3)} E^3$$

nonlinear term becomes comparable to linear term when

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

$$\Rightarrow \chi^{(3)} \approx \frac{\chi^{(1)}}{E_{at}^2} \approx \frac{1}{E_{at}^2} = 3.8 \times 10^{-24} \ m^2/V^2$$

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

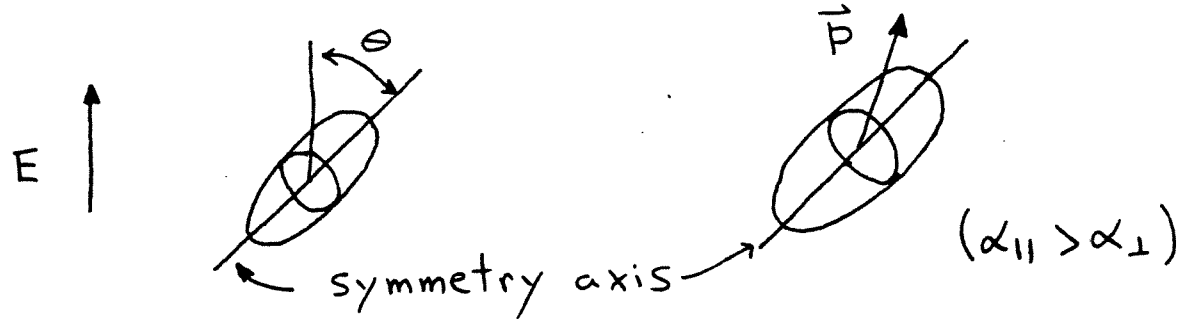


Molecular Orientation Effect

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

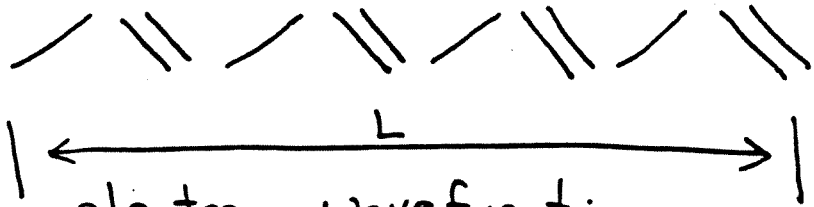
$$\vec{\tau} = \vec{p} \times \vec{E}$$

which tends to align the molecule with field.

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

How to Increase $\chi^{(3)}$

Use a conjugated polymer



electron wavefunction delocalized over length of polymer

linear polarizability $\alpha \sim L^3$

nonlinear hyperpolarizability $\gamma \sim L^5$

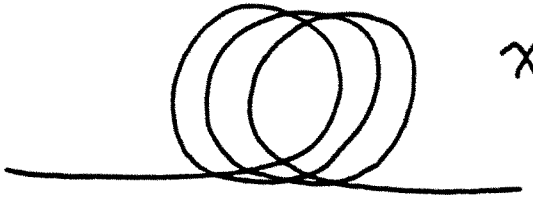
typically $\chi^{(3)} \simeq 10^{-11}$ esu

for conjugated polymer

TWO GREAT IRONIES OF NONLINEAR OPTICS

1. Silica has a small $\chi^{(3)}$, but the largest known $\chi^{(3)}/\alpha$.

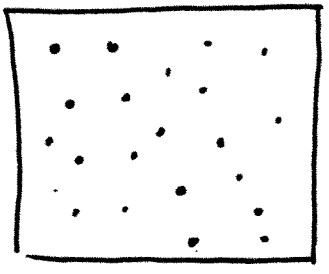
Fiber NLO



$\chi^{(3)} \simeq 1.8 \times 10^{-14}$ esu

2. Silver and gold have very large $\chi^{(3)}$, but are nearly opaque

dilute colloid →



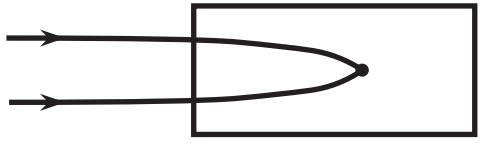
$\chi^{(3)}_{\text{silver}} \simeq 10^{-8}$ esu

4. Self-Action Effects in Nonlinear Optics

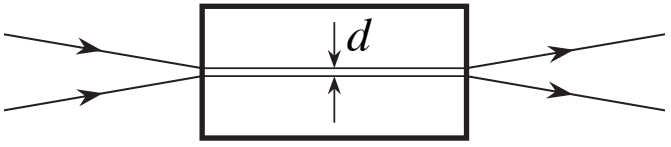
Self Action Effects in Nonlinear Optics

Self-action effects: light beam modifies its own propagation

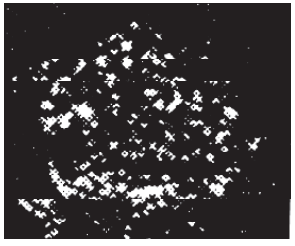
self focusing



self trapping



small-scale filamentation



Why Care About Self-Focusing and Filamentation?

- Optical switching
- Laser modelocking
- Directed energy
  - prevent filamentation
  -controlled self focusing

SOVIET PHYSICS JETP VOLUME 15, NUMBER 6 DECEMBER, 1962

EFFECTS OF THE GRADIENT OF A STRONG ELECTROMAGNETIC BEAM ON ELECTRONS AND ATOMS

G. A. ASKAR'YAN

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor December 22, 1961

J. Exptl. Theoret. Phys. (U.S.S.R.) **42**, 1567-1570 (June, 1962)

It is shown that the transverse inhomogeneity of a strong electromagnetic beam can exert a strong effect on the electrons and atoms of a medium. Thus, if the frequency exceeds the natural frequency of the electron oscillations (in a plasma or in atoms), then the electrons or atoms will be forced out of the beam field. At subresonance frequencies, the particles will be pulled in, the force being especially large at resonance. It is noted that this effect can create either a rarefaction or a compression in the beam and at the focus of the radiation, maintain a pressure gradient near an opening from an evacuated vessel to the atmosphere, and create a channel for the passage of charged particles in the medium.

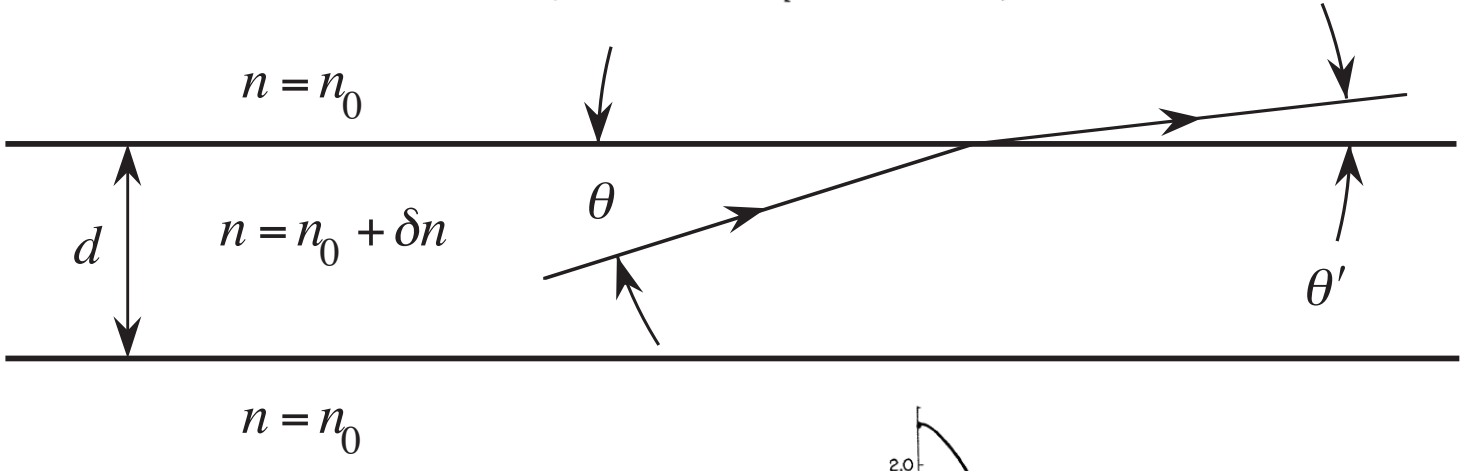
It is shown that the strong thermal ionizing and separating effects of the ray on the medium can be used to set up waveguide propagation conditions and to eliminate divergence of the beam (self-focusing). It is noted that hollow beams can give rise to directional flow and ejection of the plasma along the beam axis for plasma transport and creation of plasma current conductors. The possibilities of accelerating and heating plasma electrons by a modulated beam are indicated.

Prediction of Self Trapping

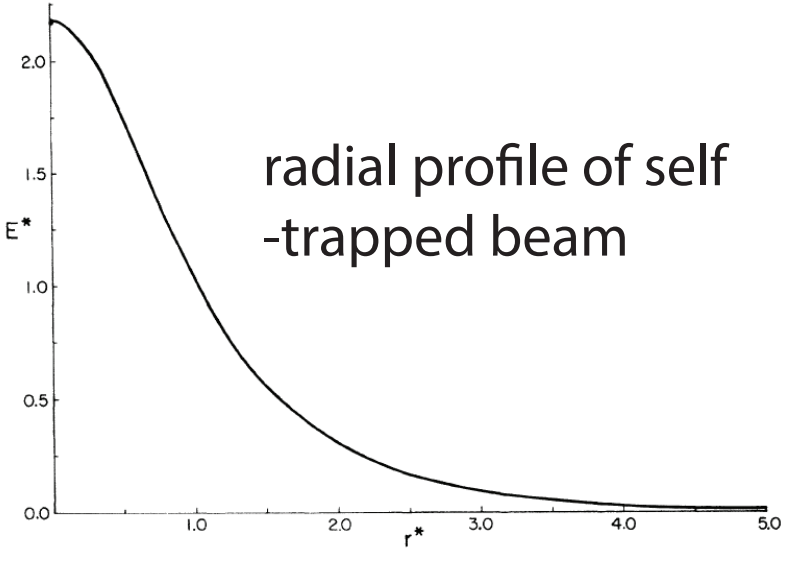
VOLUME 13, NUMBER 15 PHYSICAL REVIEW LETTERS 12 OCTOBER 1964

SELF-TRAPPING OF OPTICAL BEAMS*

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

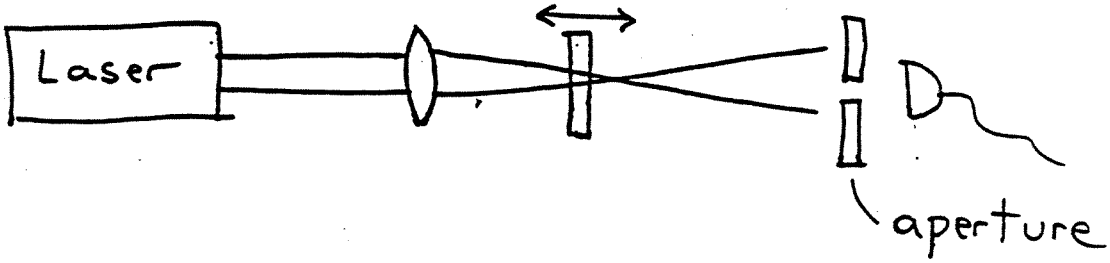


$$P_{\mathrm{cr}} = \frac{\pi(0.61)^2\lambda_0^2}{8n_0n_2}$$



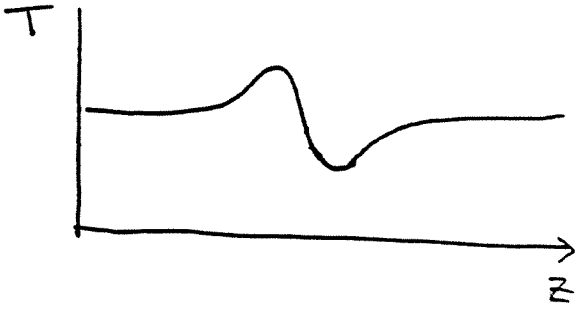
radial profile of self -trapped beam

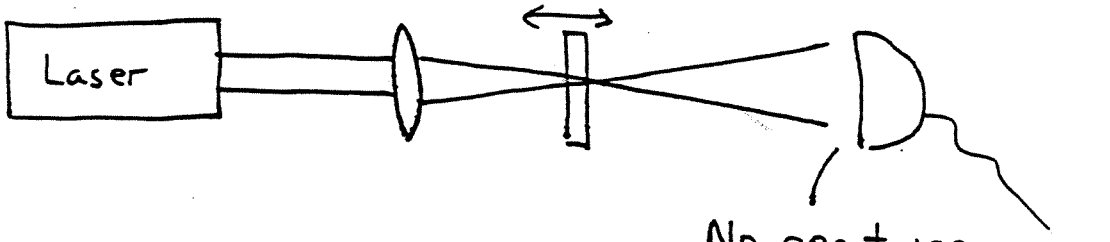
Z-Scan Measurement of $\chi^{(3)}$



Measures NL change in refraction

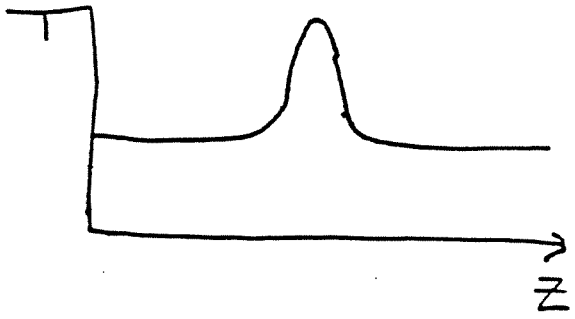
(Re $\chi^{(3)}$)



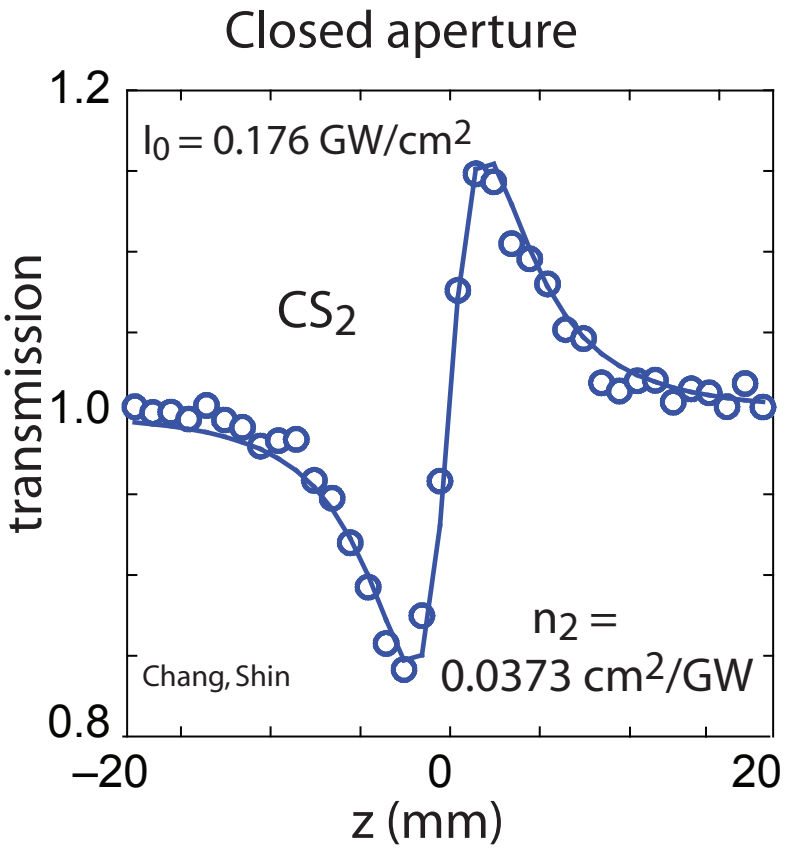


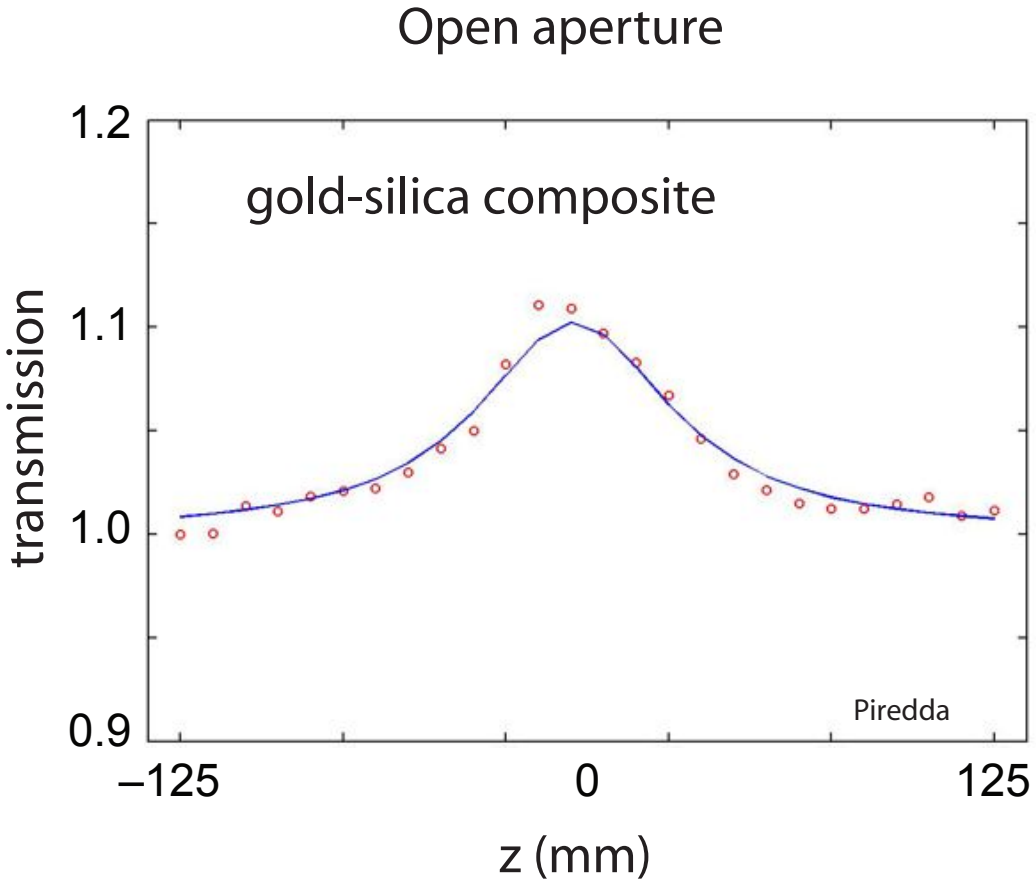
Measures NL change in absorption.

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



Some Actual Z-Scan Data





For closed aperture z-scan

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

where

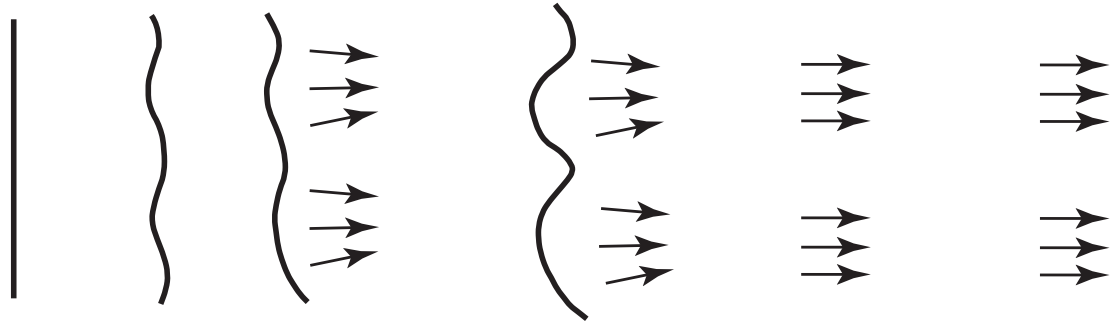
$$\Phi^{NL} = n_2\,(\omega/c)\,I_0\,L$$

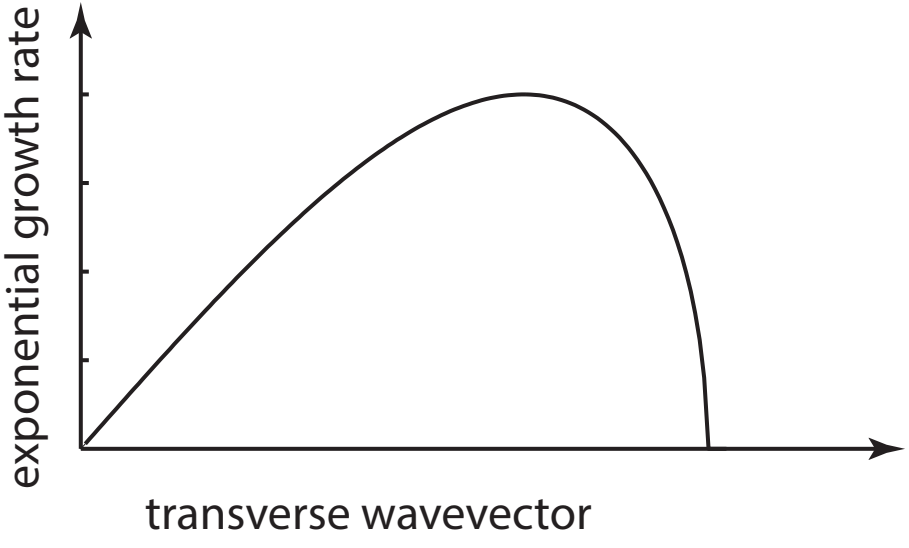
M. Sheik-Bahae et al., IEEE J. Quantum Electron. 26 760 (1990).

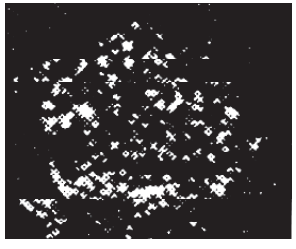
Beam Breakup by Small-Scale Filamentation

Predicted by Bespalov and Talanov (1966)

Exponential growth of wavefront imperfections by four-wave mixing processes





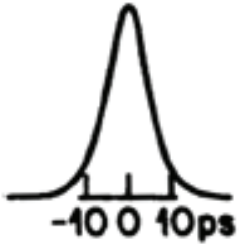


Optical Solitons

Field distributions that propagate without change of form

Temporal solitons (nonlinearity balances gvd)

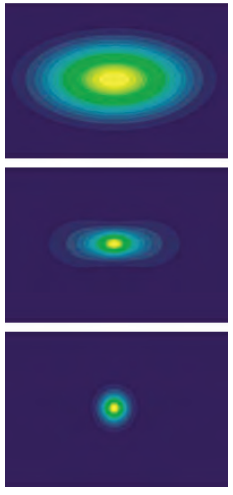
$$\frac{\partial \tilde{A}_s}{\partial z} + \tfrac{1}{2} i k_2 \frac{\partial^2 \tilde{A}_s}{\partial \tau^2} = i\gamma |\tilde{A}_s|^2 \tilde{A}_s.$$



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)

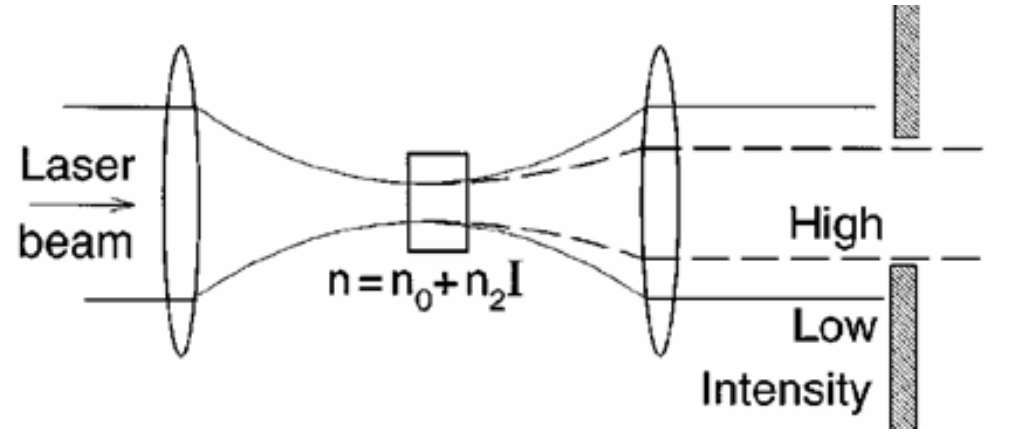
Solitons and self-focussing in Ti:Sapphire

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



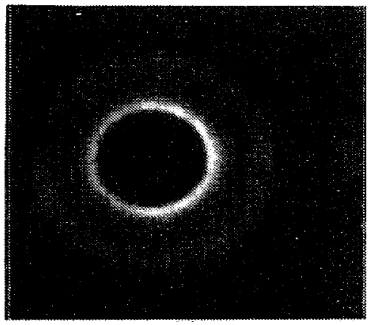
Diffraction-management controls the spatial self-focussing

Dispersion-management controls the temporal self-focussing

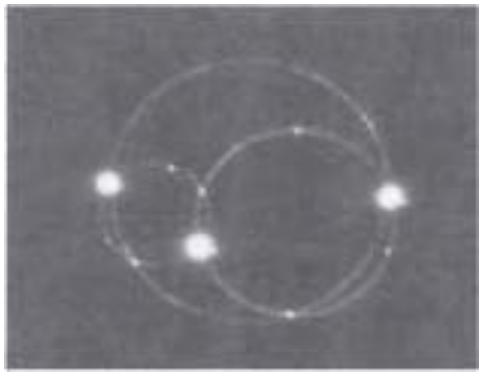
Self-Focusing Can Produce Unusual Beam Patterns

Pattern depends sensitively upon initial conditions

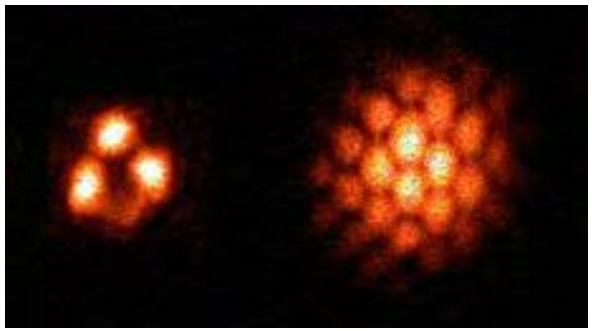
- Conical emission
  Harter et al., PRL 46, 1192 (1981)



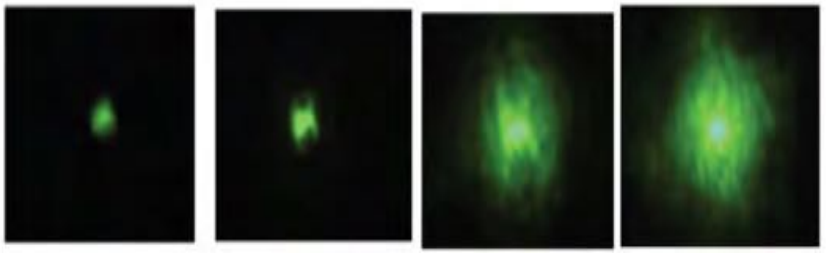
- Multiple ring patterns
  Kauranen et al, Opt. Lett. 16, 943, 1991;



- Honeycomb pattern formation
  Bennink et al., PRL 88, 113901 2002.



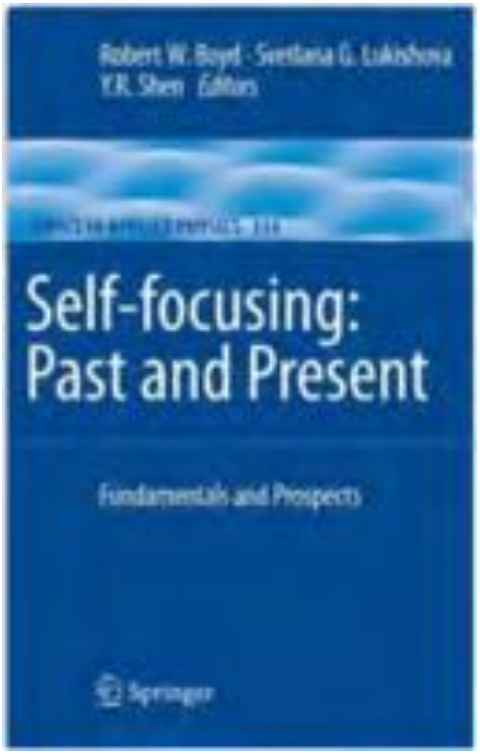
- Loss of spatial coherence
  Schweinsberg et al., Phys. Rev. A 84, 053837 (2011).



Summary

- Even more than 50 years after their inceptions, self-focusing and filamentation remain fascinating topics for investigation.

- If you want to learn more:



5. NLO of Epsilon-Near Zero Materials

Epsilon-Near-Zero (ENZ) Materials

- Physics of Epsilon-Near-Zero (ENZ) Materials
- Huge NLO Response of ENZ Materials and Metastructures
- Non-perturbative nature of the NLO Response (usual power series do not converge)

With Special Thanks To:

M. Zahirul Alam, Orad Reshef, Enno Giese, and Jeremy Upham, University of Ottawa
Israel De Leon, Tecnologico de Monterrey, Mexico
Sebastian Schulz, Cork Institute of Technology, Ireland

Physics of Epsilon-Near-Zero (ENZ) Materials

- ENZ materials possess exotic electromagnetic properties

  Silveirinha, Engheta, Phys. Rev. Lett. 97, 157403, 2006.

- If the dielectric permittivity ε is nearly zero, then refractive index $n = \text{sqrt}(\varepsilon)$ is nearly zero.

  Thus $v_{phase} = c / n$ is nearly infinite

  $\lambda = \lambda_{vac} / n$ is nearly infinite

  Light oscillates in time but not in space; everyhing is in phase

  Light "oscillates" but does not "propagate."

- Radiative processes are modified in ENZ materials

  Einstein *A* coefficient (spontaneous emission lifetime = 1/*A*)

  $A = n A_{vac}$

  We can control (inhibit!) spontaneous emission!

  Einstein *B* coefficient

  Stimulated emission rate = *B* times EM field energy density

  $B = B_{vac} / n^2$

  Optical gain is very large!

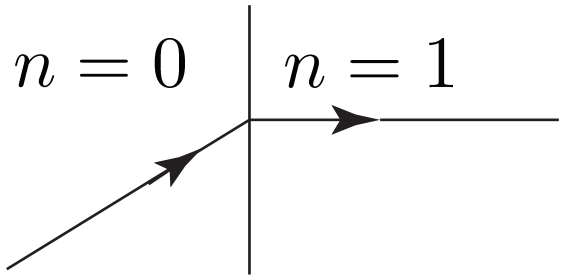
  Einstein, Physikalische Zeitschrift 18, 121 (1917).
  Milonni, Journal of Modern Optics 42, 1991 (1995).

Physics of Epsilon-Near-Zero (ENZ) Materials -- More

- Snell's law leads to intriguing predictions

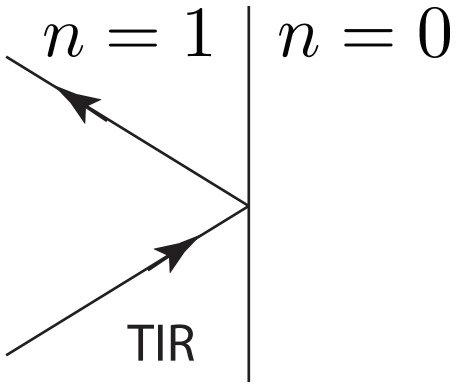
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

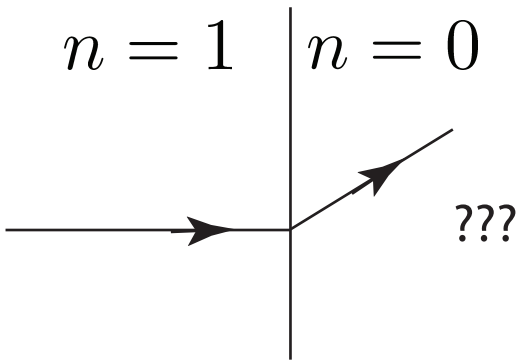
- Light always leaves perpendicular to surface of ENZ material!



Y. Li, et al., Nat. Photonics 9, 738, 2015; D. I. Vulis, et al., Opt. Express 25, 12381, 2017.

- Thus light can enter an ENZ material only at normal incidence!





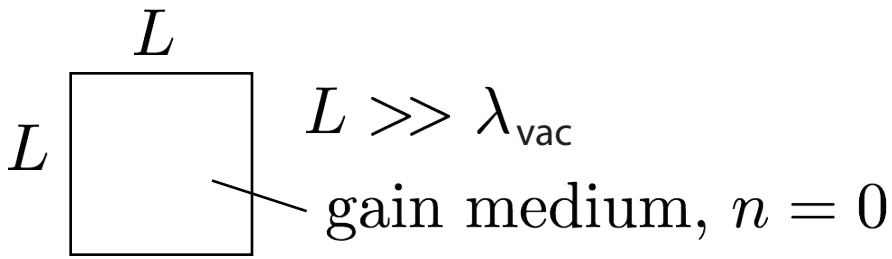
Y. Li, et al., Nat. Photonics 9, 738, 2015.

Some Consequences of ENZ Behaviour - 1

- Funny lenses

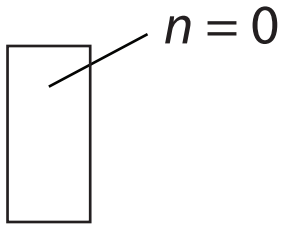
A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

- Large-area single-transverse-mode surface-emitting lasers



J. Bravo-Abad et al., Proc. Natl. Acad. Sci. USA 109, 976, 2012.

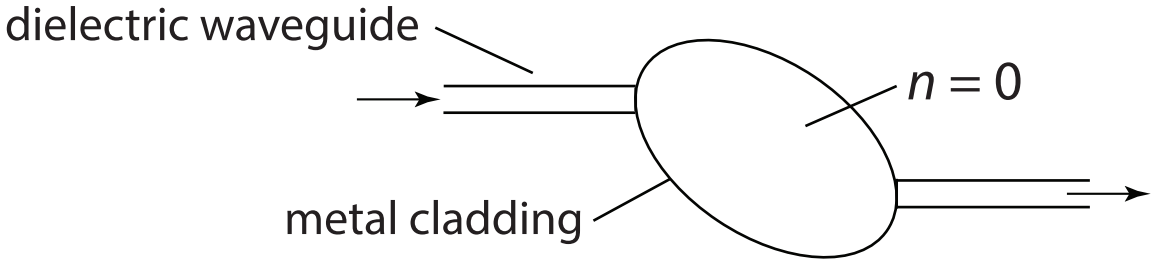
- No Fabry-Perot interference



O. Reshef et al., ACS Photonics 4, 2385, 2017.

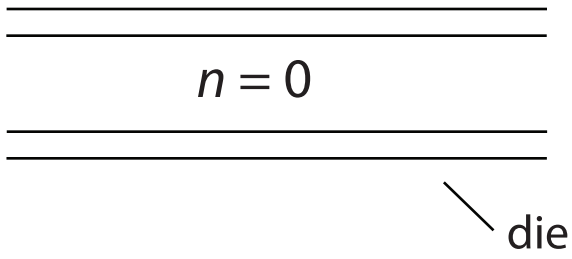
Some Consequences of ENZ Behaviour - 2

- Super-coupling (of waveguides)



M. G. Silveirinha and N. Engheta, Phys. Rev. B 76, 245109, 2007; B. Edwards et al., Phys. Rev. Lett. 100, 033903, 2008.

- Large evanescent tails for waveguide coupling

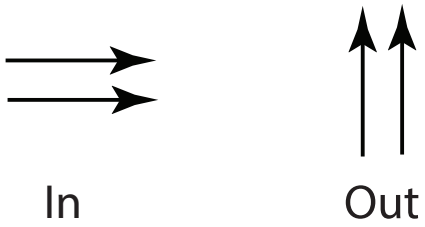


transverse profile of upper waveguide extends to lower waveguide for any distance

- Automatic phase matching of NLO processes

Recall that $k = n \omega / c$ vanishes in an ENZ medium.

For example, the following 4WM proces is allowed



H. Suchowski et al., Science 342, 1223, 2013.

Some Consequences of ENZ Behaviour - 3

- How is the theory of self-focusing modified?
- Does the theory of Z-scan need to be modified?
- How is the theory of blackbody radiation modified?
- Do we expect very strong superradiance effects?
- More generally, how is any NLO process modified when $n_0 = 0$?

Epsilon-Near-Zero Materials

- Metamaterials
  Materials tailor-made to display ENZ behaviour

- Homogeneous materials
  All materials display ENZ behaviour at their (reduced) plasma frequency

  Recall the Drude formula

$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

$$\text{Note that } \operatorname{Re}\epsilon = 0 \text{ for } \omega = \omega_p/\sqrt{\epsilon}_\infty \equiv \omega_0.$$

- Challenge: Obtain low-loss ENZ materials
  Want Im ε as small as possible at the frequency where Re $\varepsilon = 0$.

- We are examining a several materials
  ITO: indium tin oxide
  AZO: aluminum zinc oxide
  FTO: fluorine tin oxide

Epsilon-Near-Zero Materials for Nonlinear Optics

- We need materials with a much larger NLO response
- We recently reported a material (indium tin oxide, ITO) with an n_2 value 100 time larger than those previously reported.
- This material utilizes the strong enhancement of the NLO response that occurs in the epsilon-near zero (ENZ) spectral region.

Large optical nonlinearity of indium tin oxide in its epsilon-near-zero region,
M. Zahirul Alam, I. De Leon, R. W. Boyd, Science 352, 795 (2016).

Implications of ENZ Behavior for Nonlinear Optics

Here is the intuition for why the ENZ conditions are of interest in NLO

Recall the standard relation between n_2 and $\chi^{(3)}$

$$n_2 = \frac{3\chi^{(3)}}{4\epsilon_0 c\, n_0\, \mathrm{Re}(n_0)}$$

Note that for ENZ conditions the denominator becomes very small, leading to a very large value of n_2

Optical Properties of Indium Tin Oxide (ITO)

ITO is a degenerate semiconductor (so highly doped as to be metal-like).

It has a very large density of free electrons, and a bulk plasma frequency corresponding to a wavelength of approximately 1.24 μm.

Recall the Drude formula

$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

Note that $\mathrm{Re}\,\epsilon = 0$ for $\omega = \omega_p/\sqrt{\epsilon_\infty} \equiv \omega_0$.

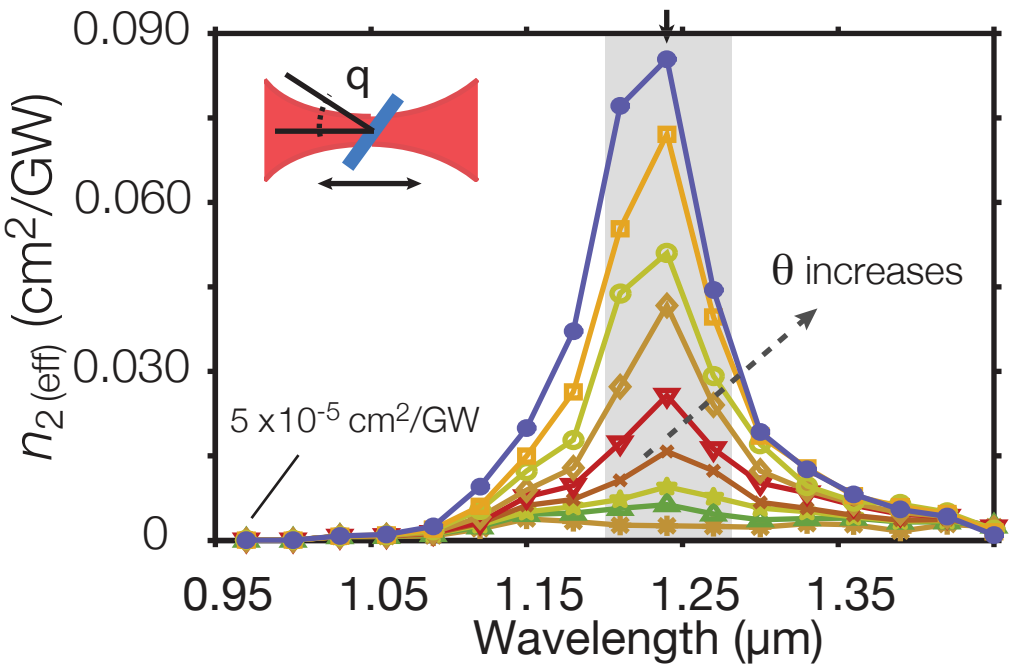
The region near ω_0 is known as the epsilon-near-zero (ENZ) region.

There has been great recent interest in studies of ENZ phenomena:

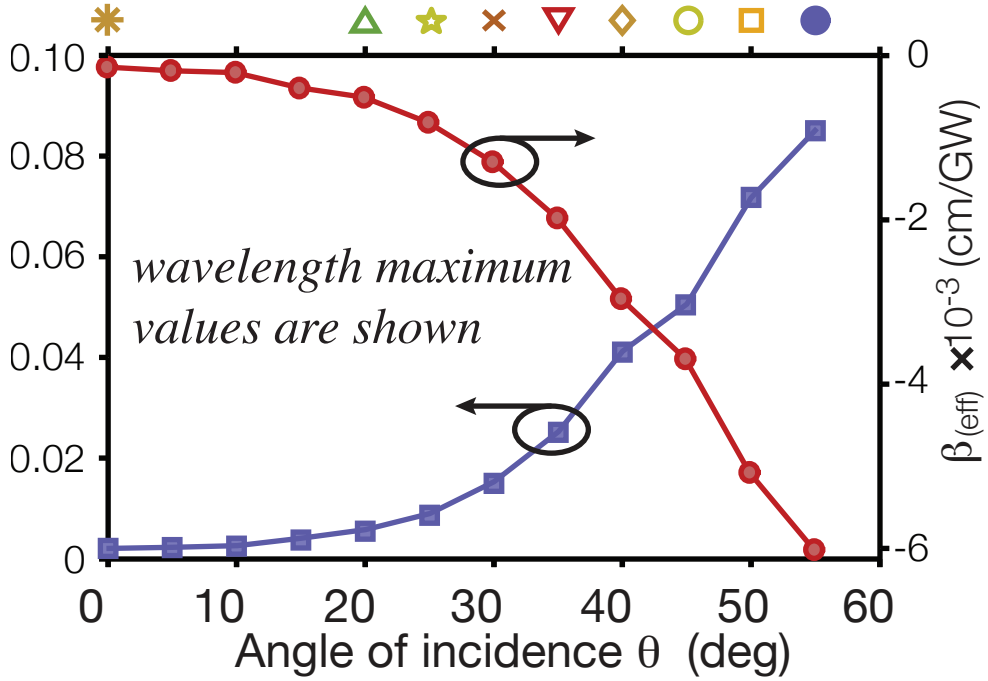
H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, Science 342, 1223 (2013).
C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, Phys. Rev. B 85, 045129 (2012).
S. Campione, D. de Ceglia, M. A. Vincenti, M. Scalora, and F. Capolino, Phys. Rev. B 87, 035120 (2013).
A. Ciattoni, C. Rizza, and E. Palange, Phys. Rev. A 81,043839 (2010).

Huge Nonlinear Optical Response Measured by Z-scan

Wavelength dependence of n_2

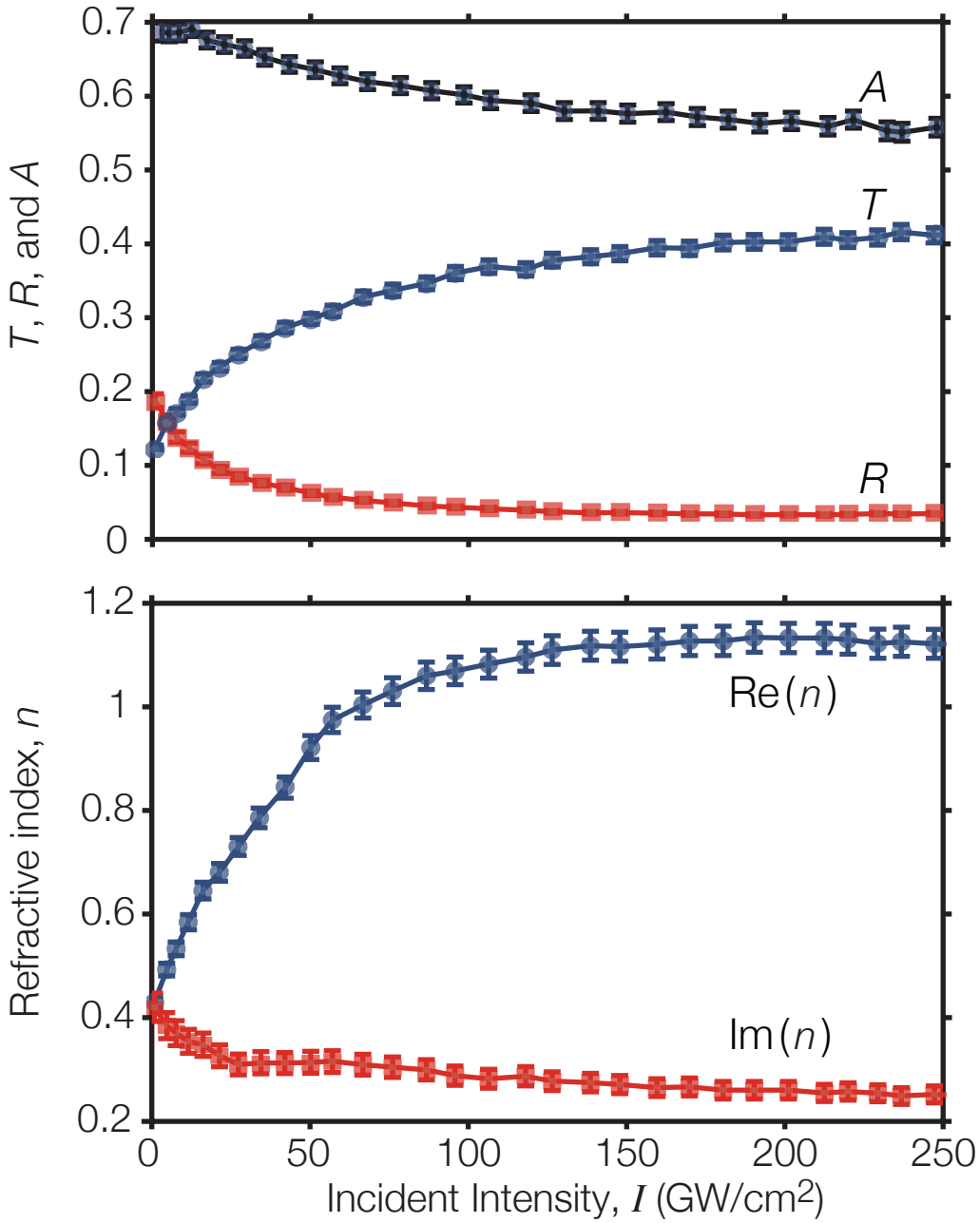


Variation with incidence angle



- Note that n_2 is positive (self focusing) and β is negative (saturable absorption)
- Both n_2 and nonlinear absorption increase with angle of incidence
- n_2 shows a maximum value of 0.11 cm^2/GW = 1.1 x 10^{-10} cm^2/W at 1.25 μm and 60 deg. This value is 2000 times larger than that away from ENZ region.
- n_2 is 3.4 x 10^5 times larger than that of fused silica
  n_2 is 200 times larger than that of chalcogenide glass

Beyond the $\chi^{(3)}$ limit



The nonlinear change in refractive index is so large as to change the transmission, absorption, and reflection!

Note that transmission is increased at high intensity.

Here is the refractive index extracted from the above data.

Note that the total nonlinear change in refractive index is $\Delta n = 0.8$.

The absorption decreases at high intensity, allowing a predicted NL phase shift of 0.5 radians.

Nonperturbative Nature of the NLO Response

1. The conventional equation $n = n_0 + n_2 I$ is not applicable to ENZ and other low-index materials. The nonlinear response is nonperturbative.

2. The nonlinear response can be accurately modeled in the $\chi^{(3)}$ limit by

$$n = \sqrt{n_0^2 + 2n_0 n_2 I}$$

where

$$n_2 = \frac{3\chi^{(3)}}{4n_0 \mathrm{Re}(n_0)\epsilon_0 c}.$$

and

$$I = 2\mathrm{Re}(n_0)\epsilon_0 c|E|^2$$

3. More generally, the intensity dependent refractive index can be described by

$$n = \sqrt{\epsilon^{(1)} + 3\chi^{(3)}|E|^2 + 10\chi^{(5)}|E|^4 + \cdots}$$

Reshef, Giese, Alam, De Leon, Upham, Boyd, Opt. Lett. 42, 3225 (2017).

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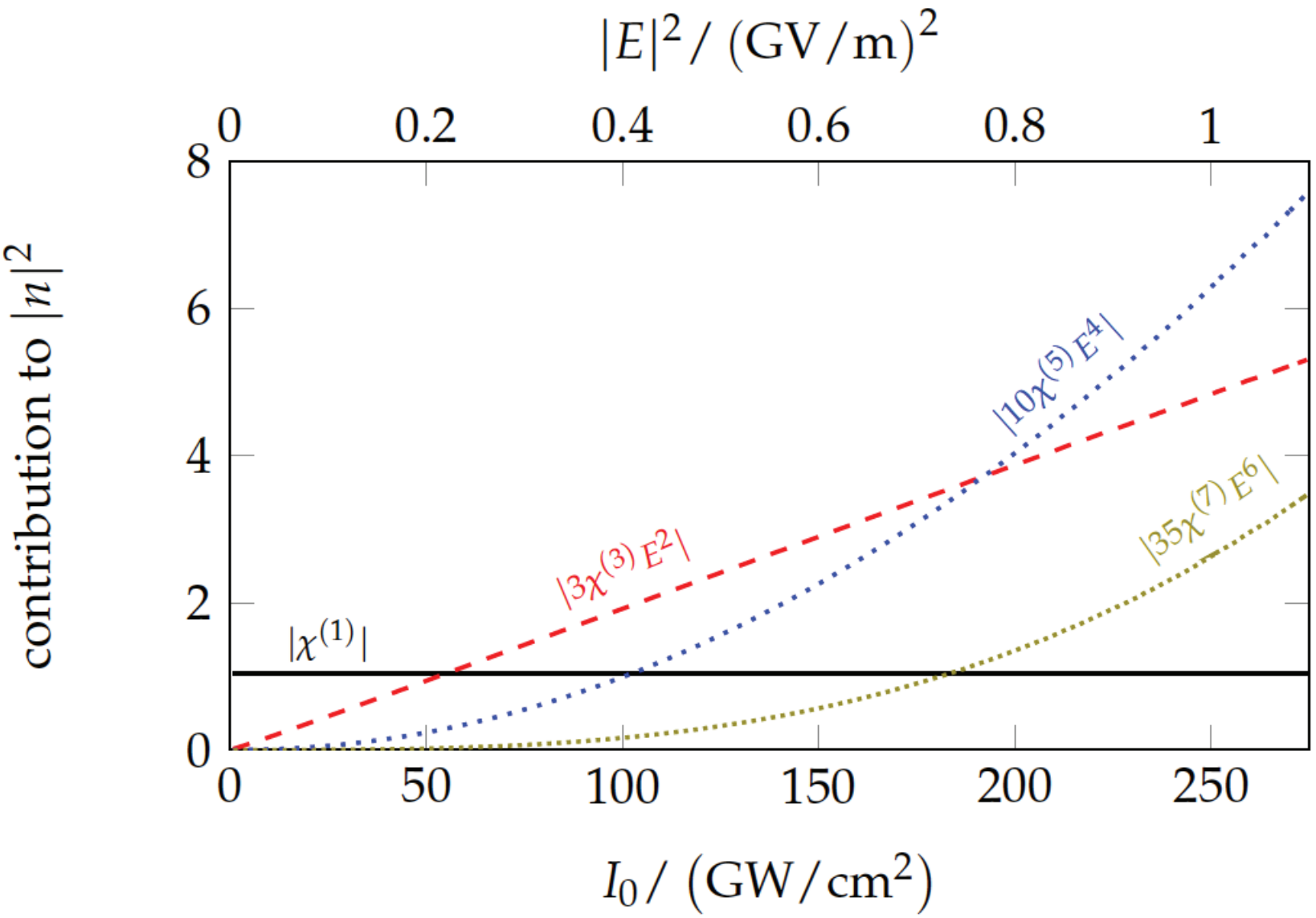
Reshef, Giese, Alam, De Leon, Upham, Boyd, Opt. Lett. 42, 3225 (2017).

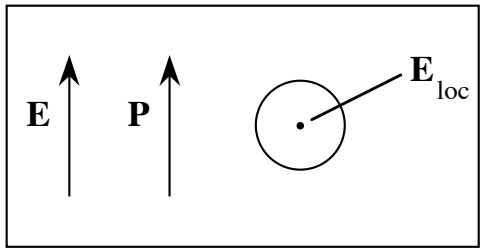
Nonlinear Response of ITO is Nonperturbative



6. 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

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

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

$$\Rightarrow$$

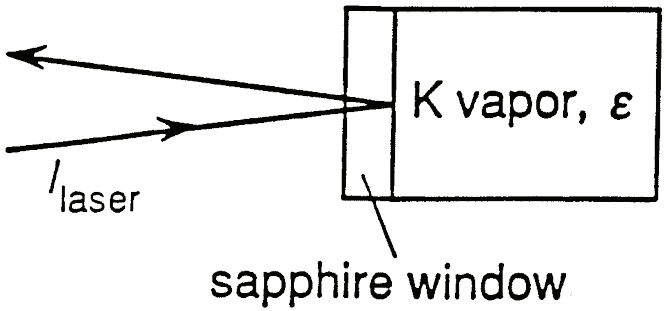
$$\varepsilon = 1 + \frac{N f r_e \lambda_0 c}{\omega_0 + \Delta\omega_L - \omega - i\gamma}$$

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

↑ Lorentz red shift

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

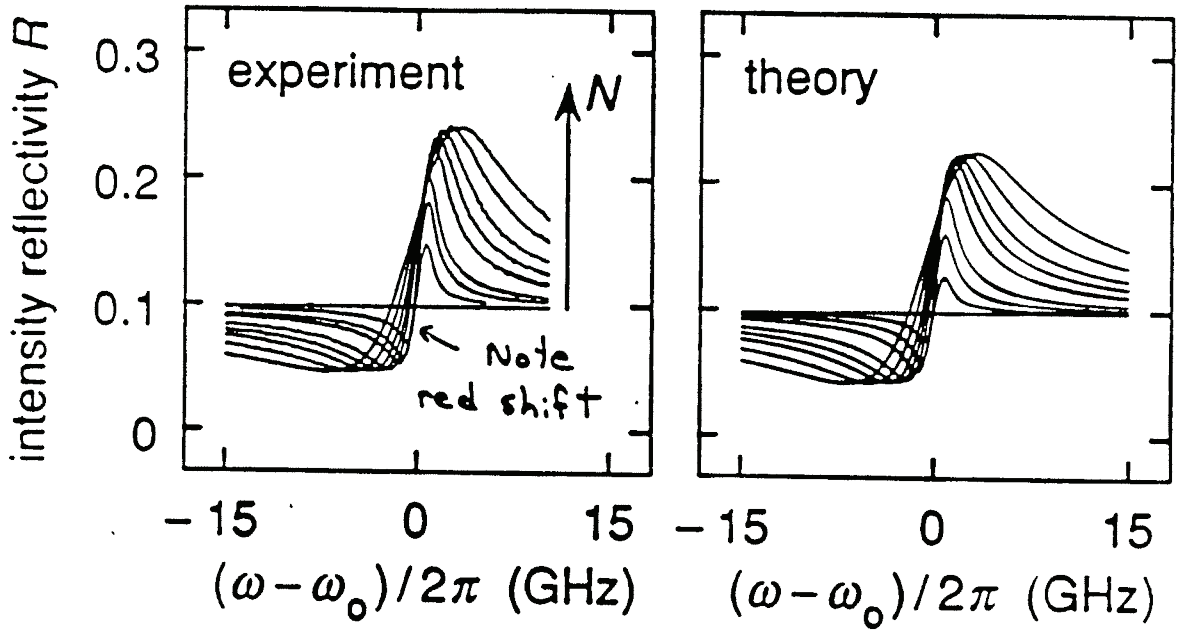
Observation of the Lorentz Red Shift



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

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

$|\varepsilon - 1| \sim 1$ 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

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

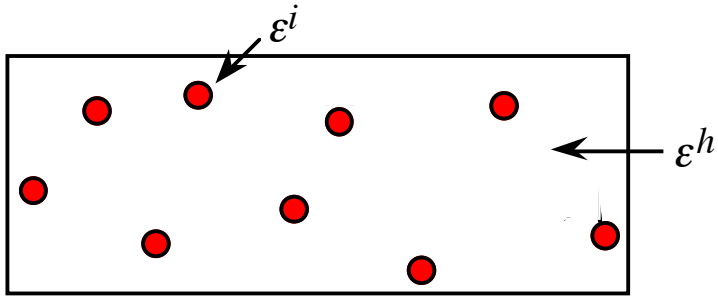
For the typical value $n = 2$, $L = 2$, and $L^4 = 16$. Local field effects can be very large in nonlinear optics! But can we tailor them for our benefit?

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

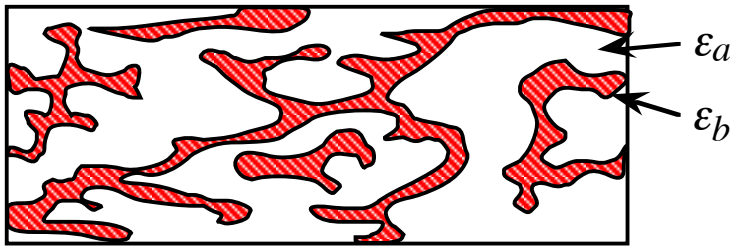
7. Composite Materials and Metamaterials for NLO

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

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

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 $L = \dfrac{\varepsilon + 2}{3}$

- 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

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

Material Systems for Composite NLO Materials

All-dielectric composite materials

Minimum loss, but limited NL response

Metal-dielectric composite materials

Larger loss, but larger NL response

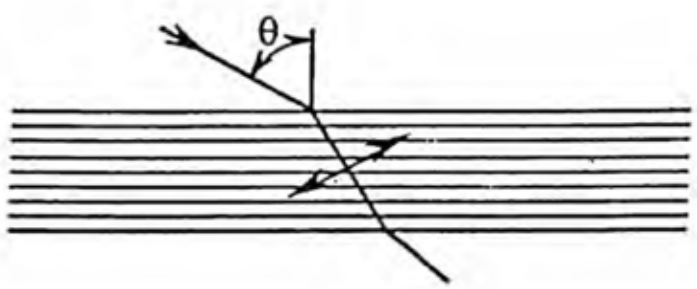
Note that $\chi^{(3)}$ of gold $\approx 10^6$ $\chi^{(3)}$ of silica glass!

Also, metal-dielectric composites possess surface plasmon resonances, which can further enhance the NL response.

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.



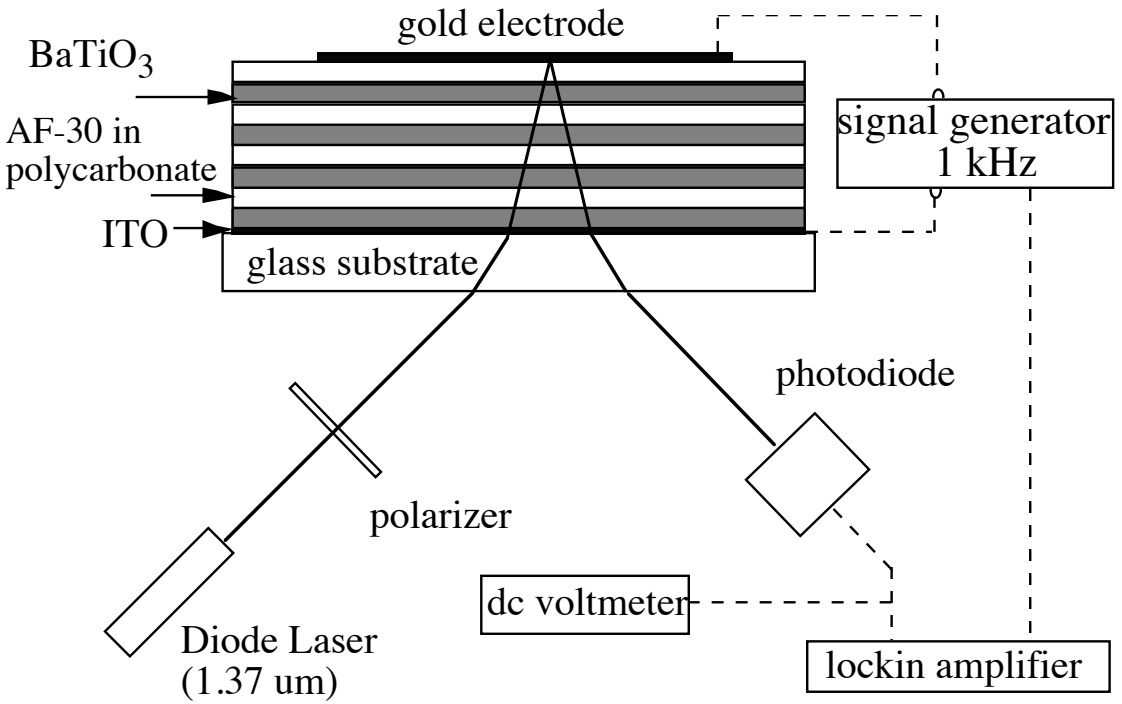
$\nabla \cdot \mathbf{D} = 0$ implies that $(\varepsilon \mathbf{E})_\perp$ is continuous.

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

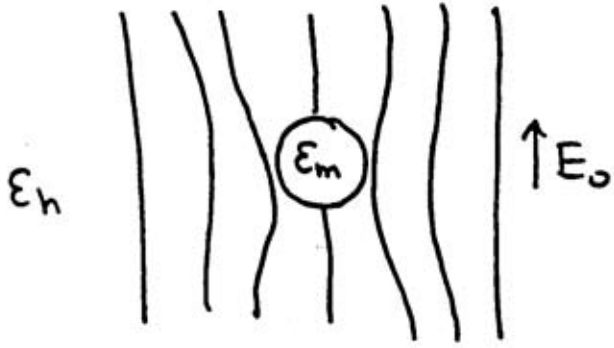


3.2 times enhancement!

Nelson and Boyd, APL 74 2417 (1999)

Metal / Dielectric Composites

Very large local field effects



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

$$\equiv \mathcal{L} E_o$$

(ε_m is negative !)

At resonance

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

Gold-Doped Glass: A Maxwell-Garnett Composite

Developmental Glass, Corning Inc.

gold volume fraction approximately 10^{-6}

gold particles approximately 10 nm diameter

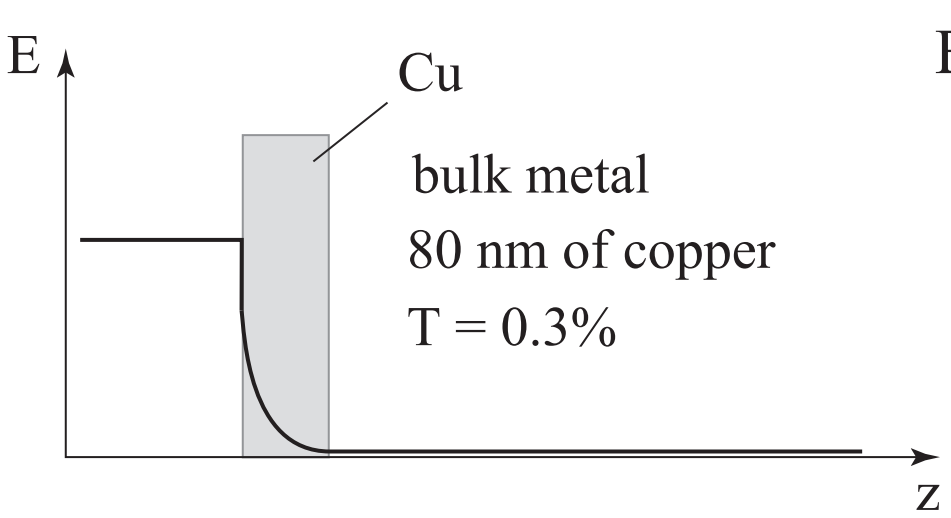
Red Glass Caraffe
Nurenberg, ca. 1700

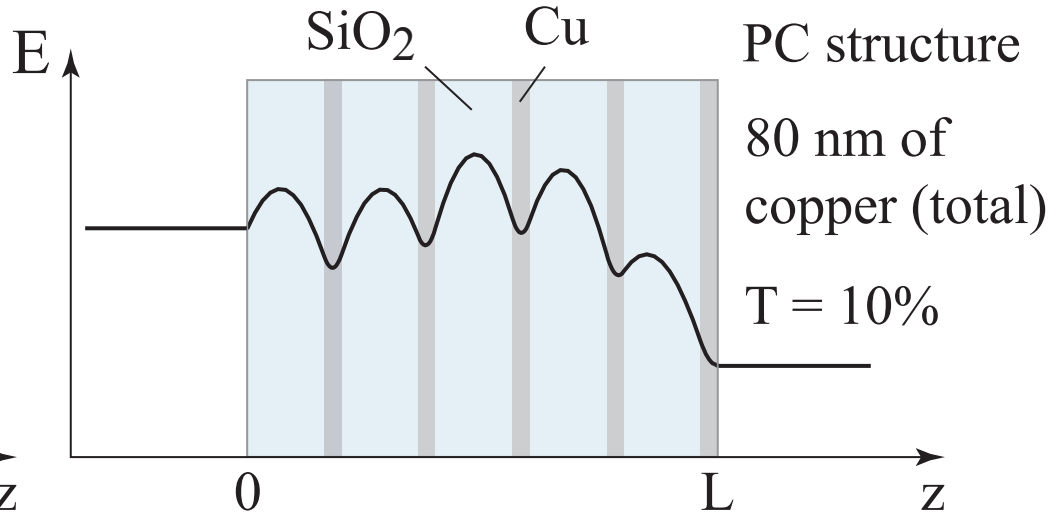
Huelsmann Museum, Bielefeld

- 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

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

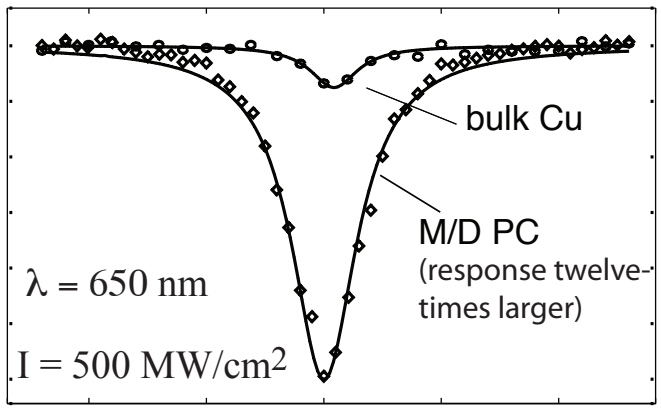
- Metals have very large optical nonlinearities but low transmission
- Low transmission because metals are highly reflecting (not because they are absorbing!)
- Solution: construct metal-dielectric photonic crystal structure
  (linear properties studied earlier by Bloemer and Scalora)





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

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



8. Slow and Fast Light

Controlling the Velocity of Light

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

– Light can be made to go:

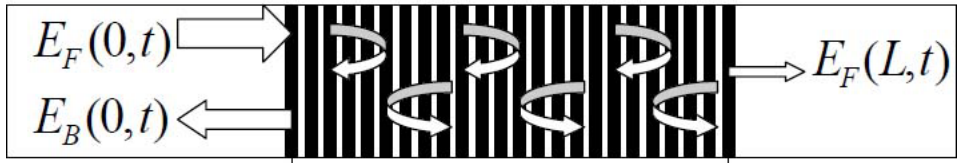
slow: $v_g << c$ (as much as 10^6 times slower!)

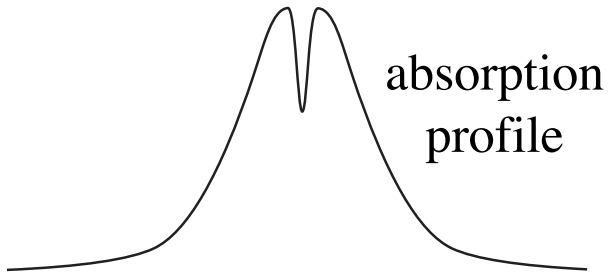
fast: $v_g > c$

backwards: v_g negative

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

– Velocity controlled by structural or material resonances

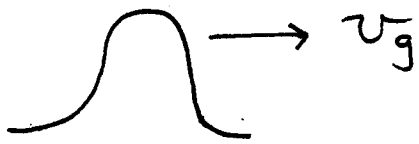




Review article: Boyd and Gauthier, Science 326, 1074 (2009).

Group Velocity

Pulse
(wave packet)



Group velocity given by $v_g = \frac{d\omega}{dk}$

For $k = \frac{n\omega}{c}$ $\quad \frac{dk}{d\omega} = \frac{1}{c}\left(n + \omega \frac{dn}{d\omega}\right)$

Thus

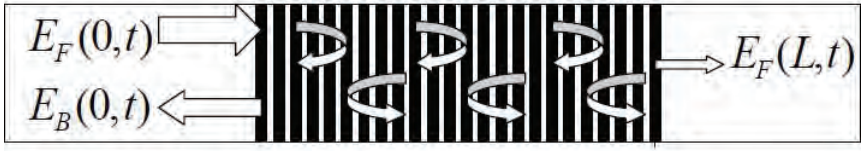
$$v_g = \frac{c}{n + \omega \frac{dn}{d\omega}} \equiv \frac{c}{n_g}$$

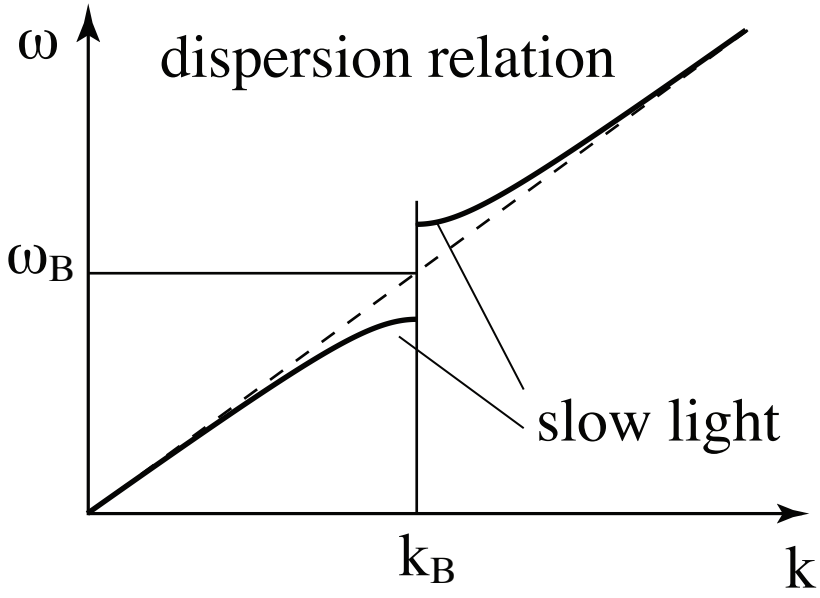
Thus $n_g \neq n$ in a dispersive medium!

Slow Light in a Fiber Bragg Grating (FBG) Structure

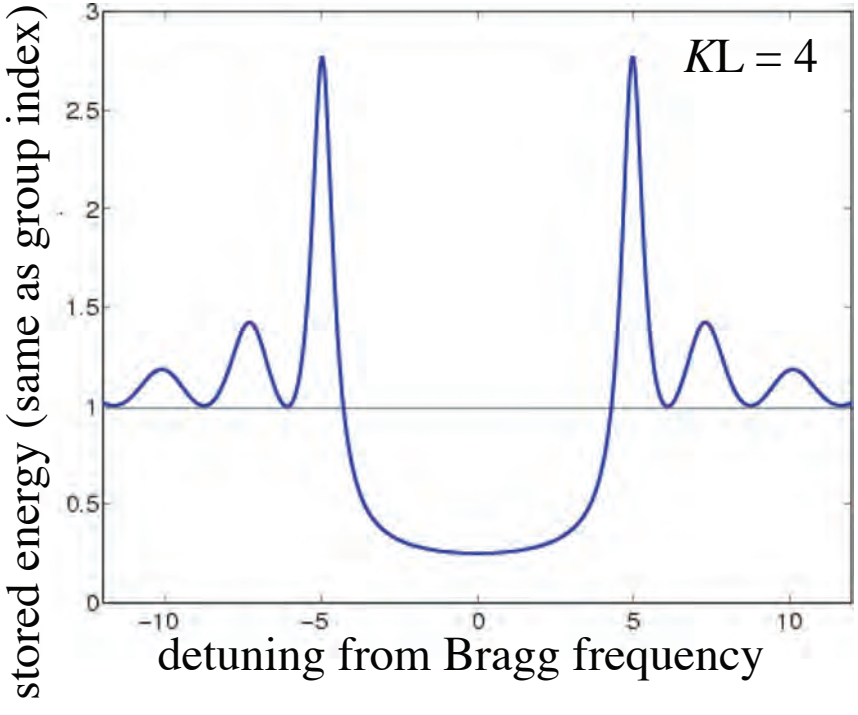
(Can describe properties of FBGs by means of analytic expressions)

forward and backward waves are strongly coupled





theory (Winful)



- Enhanced NLO response

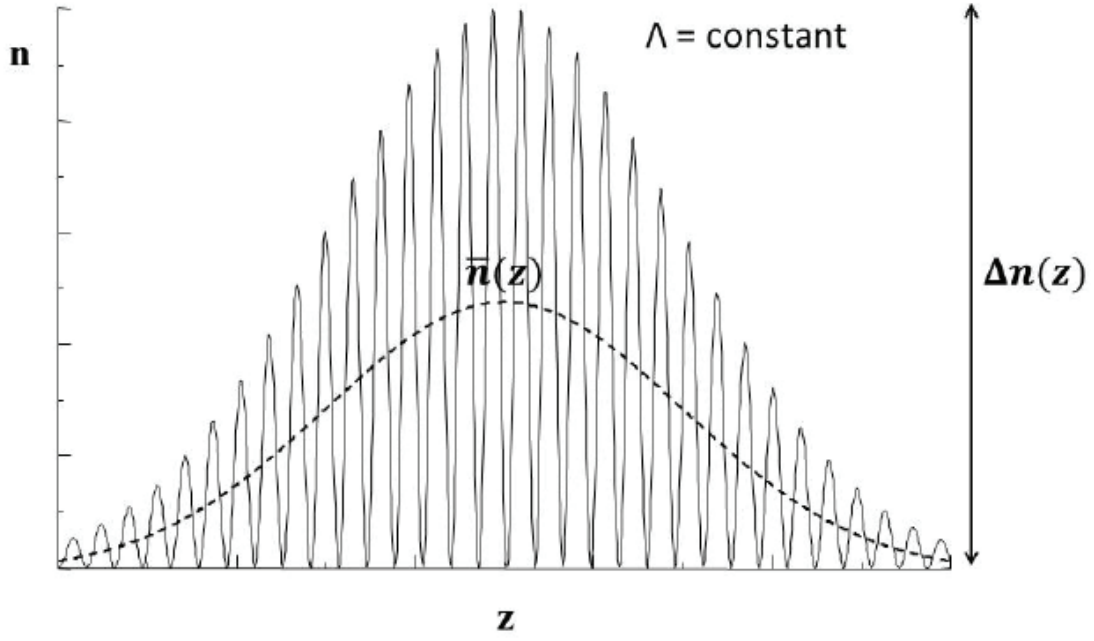
Bhat and Sipe showed that the nonlinear coeficient is given by

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

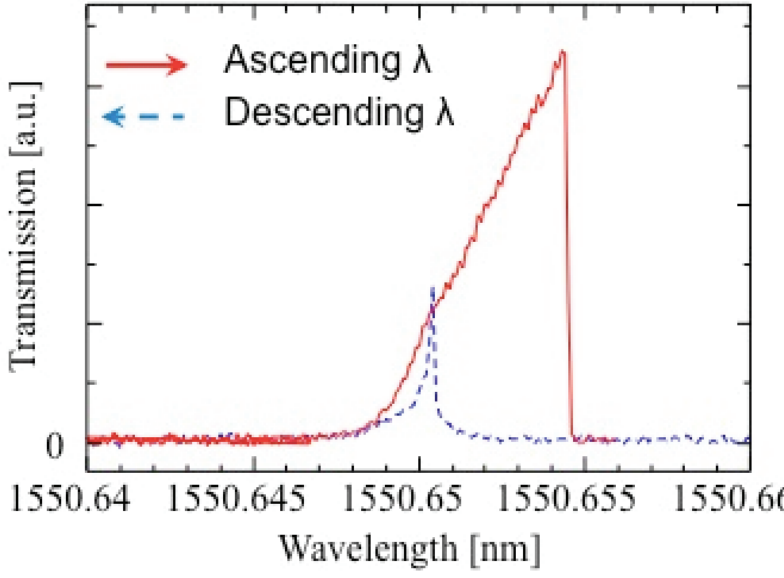
where the slow-down factor $S = n_g/n$

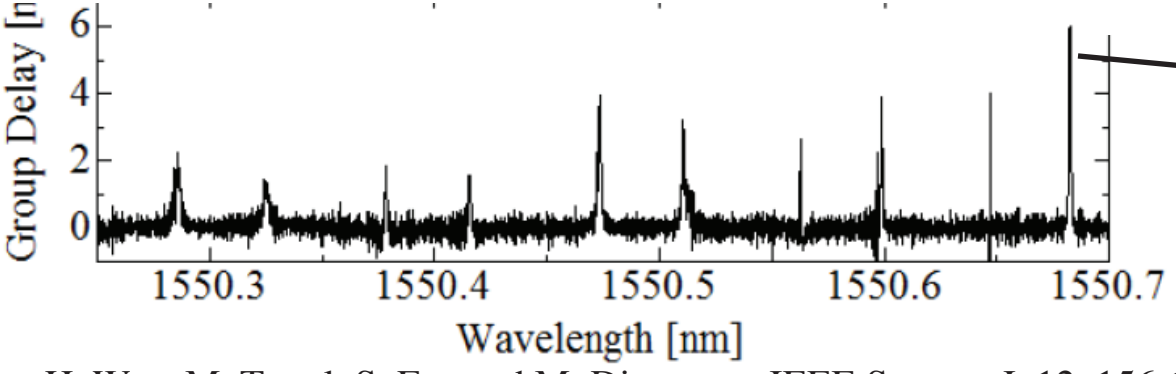
Improved Slow-Light Fiber Bragg Grating (FBG) Structure

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



Observation of (thermal) optical bistability at mW power levels



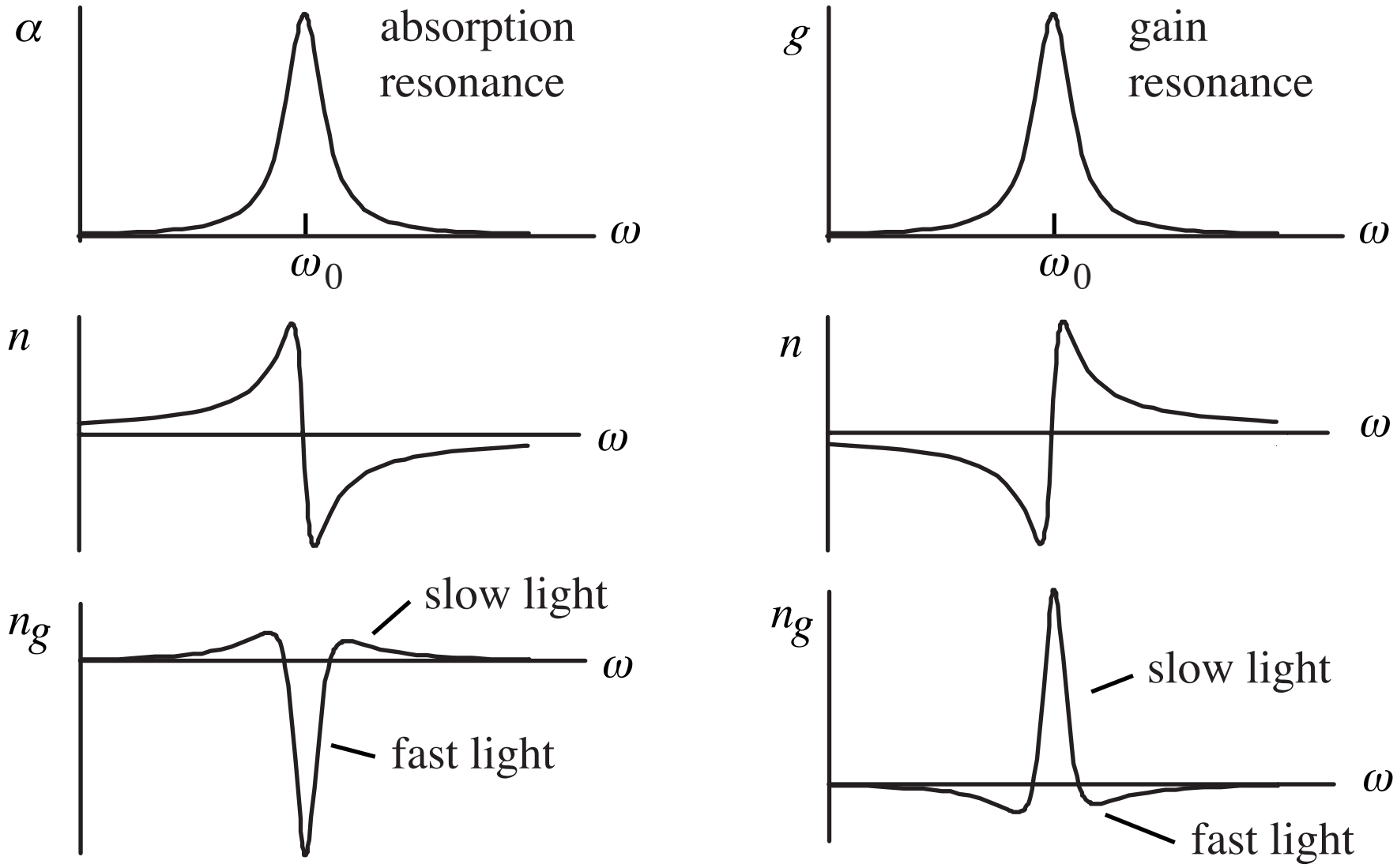


group index approximately 140

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).

Slow Light Fundamentals: How to Create Slow and Fast Light I Use Isolated Gain or Absorption Resonance



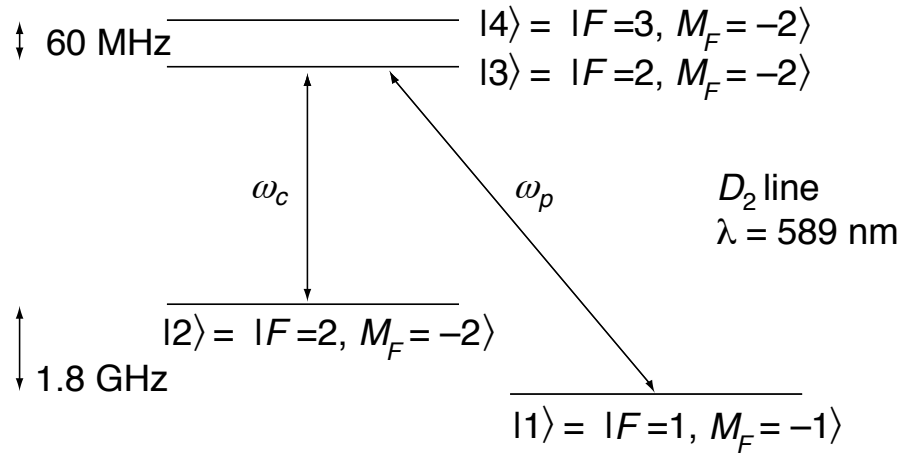
$$n_g = n + \omega\,(dn/d\omega)$$

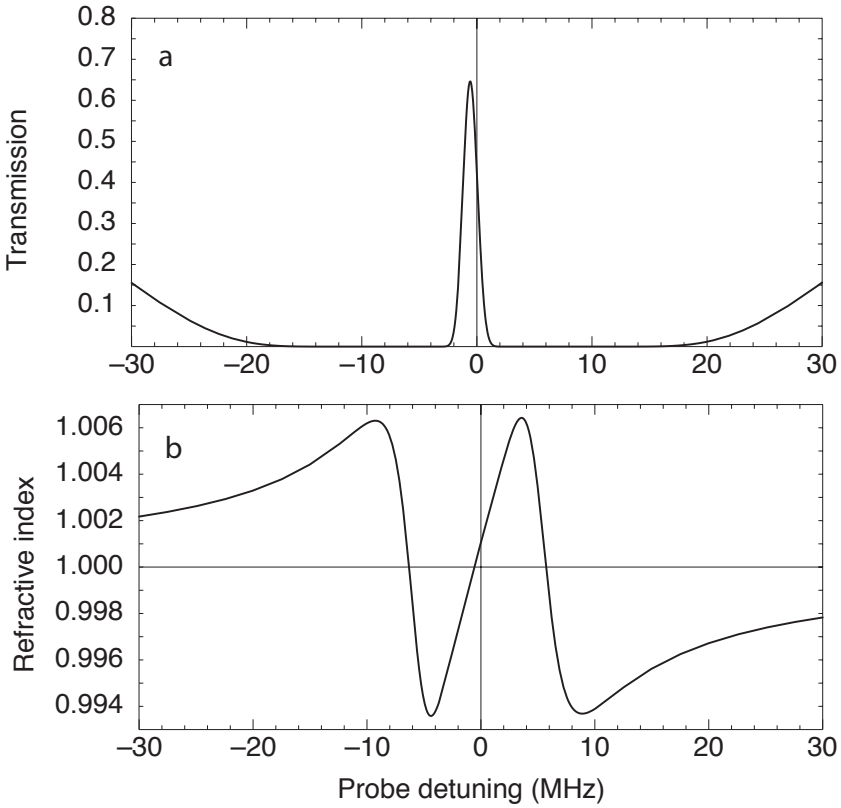
Light speed reduction to 17 metres per second in an ultracold atomic gas

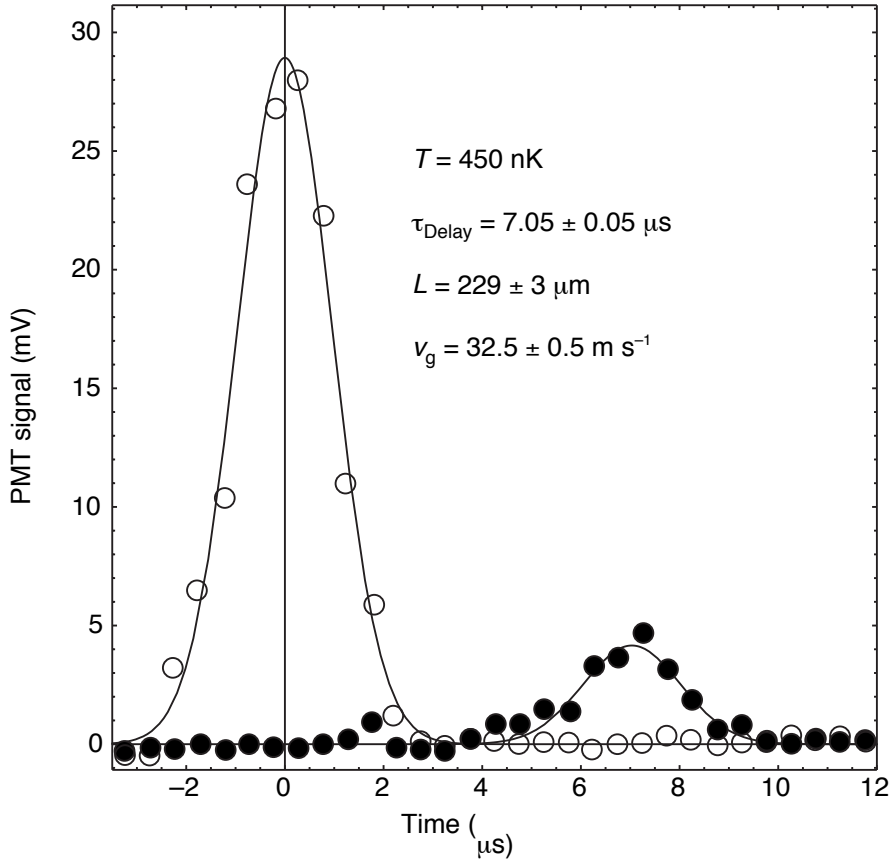
Lene Vestergaard Hau*², S. E. Harris³, Zachary Dutton*² & Cyrus H. Behroozi*§

* 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

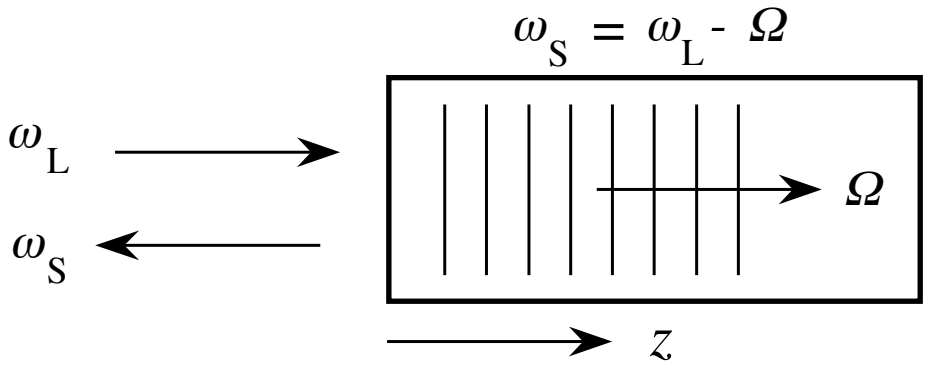
Slow Light and Fast Light: A Tutorial Review

1. Introduction to slow light
2. Mechanisms of slow light
3. Backwards light and causality
4. Slow light spectrometer
5. Optical forces
6. Fresnel drag
7. Other applications

Slow Light in Room-Temperature Solid-State Materials

- Crucial for many real-world applications
- Two examples
  - Slow light *via* stimulated Brillouin scattering (SBS)
  - Slow light *via* coherent population oscillations (CPO)

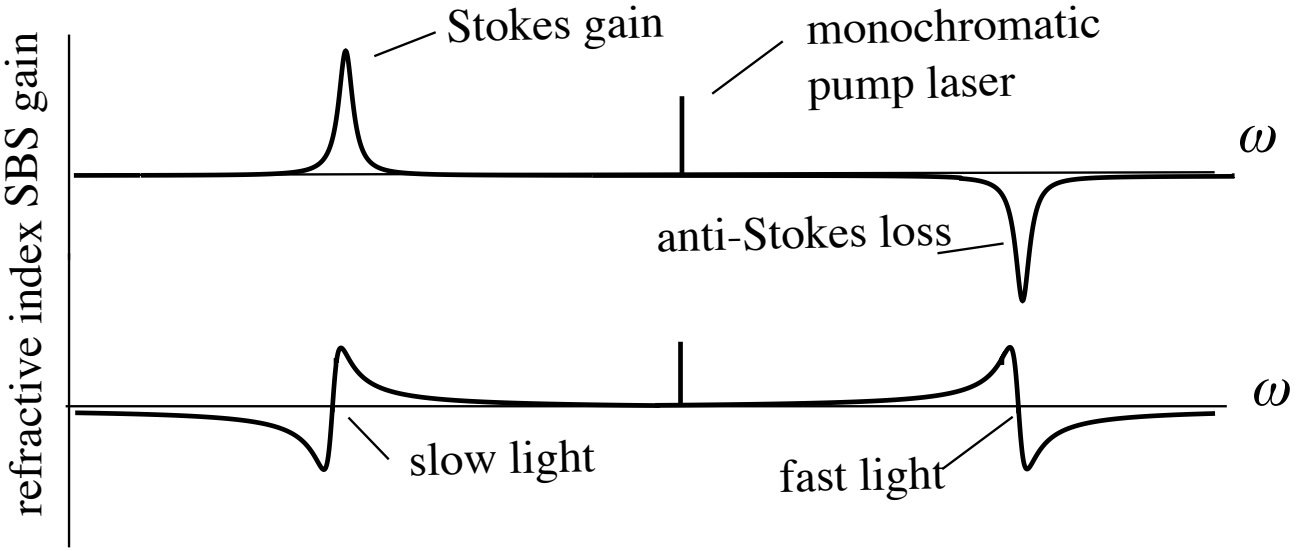
Slow Light by Stimulated Brillouin Scattering (SBS)

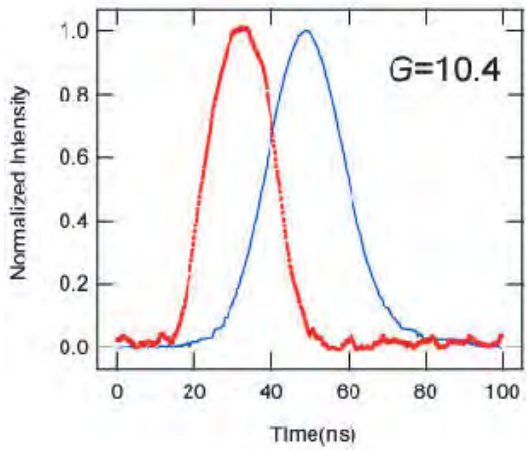


$$\frac{dI_S}{dz} = -gI_L I_S$$

$$g = \frac{\gamma_e^2 \omega^2}{nv\, c^3 \varrho_0 \Gamma_B}.$$

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 $\Delta T_d \approx \frac{G}{\Gamma_B}$ where $G = g\, I_p\, L$ and Γ_B is the Brillouin linewidth

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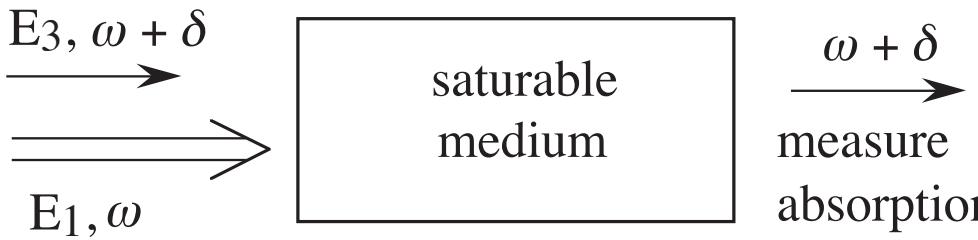
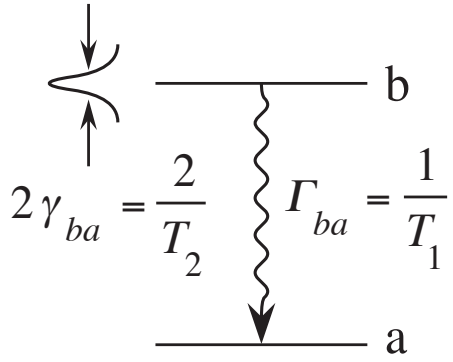
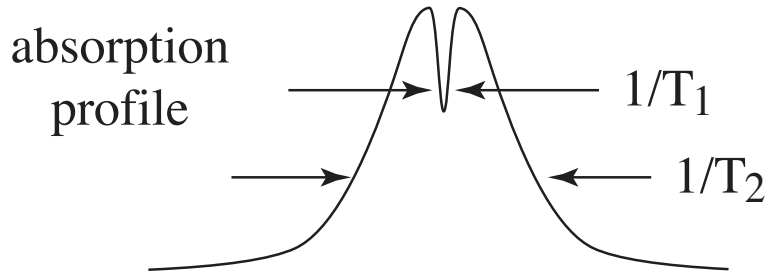
Okawachi, Bigelow, Sharping, Zhu, Schweinsberg, Gauthier, Boyd, and Gaeta Phys. Rev. Lett. 94, 153902 (2005).
Related results reported by Song, González Herráez and Thévenaz, Optics Express 13, 83 (2005).

Slow Light via Coherent Population Oscillations





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

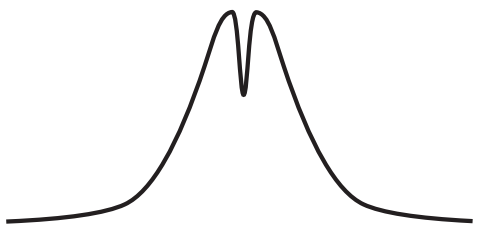


- Want a narrow feature in absorption profile to give a large $dn/d\omega$
- Ground state population oscillates at beat frequency δ (for $\delta < 1/T_1$).
- Population oscillations lead to decreased probe absorption (by explicit calculation), even though broadening is homogeneous.
- Ultra-slow light ($n_g > 10^6$) 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!

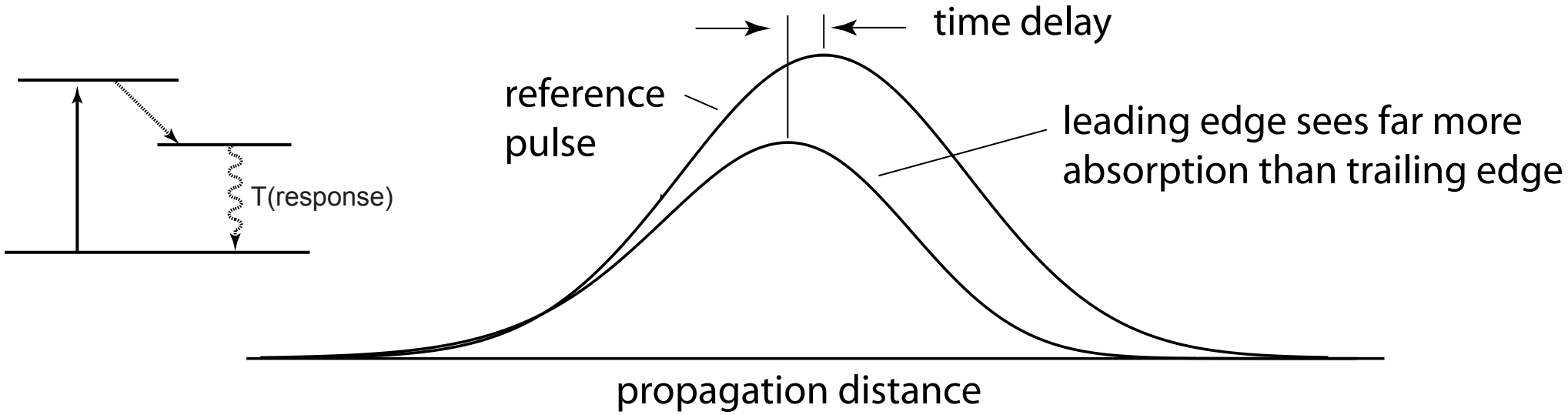
PRL 90,113903(2003); Science, 301, 200 (2003)

Relation of CPO to the Basov Mechanism

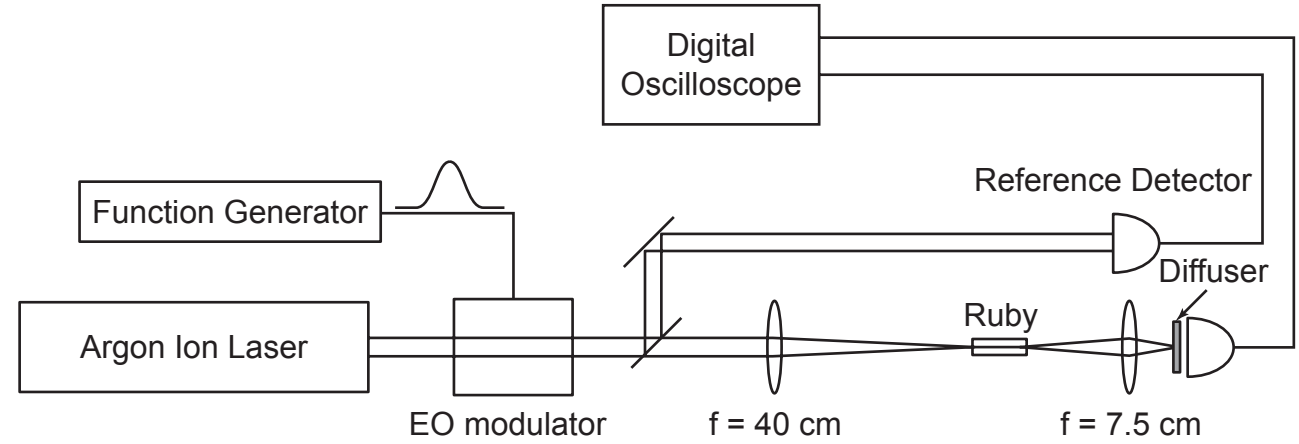
- CPO slow light: a strong pump beam creates a narrow transparency window, and strong spectral variation of the refractive index leads to a large group index.

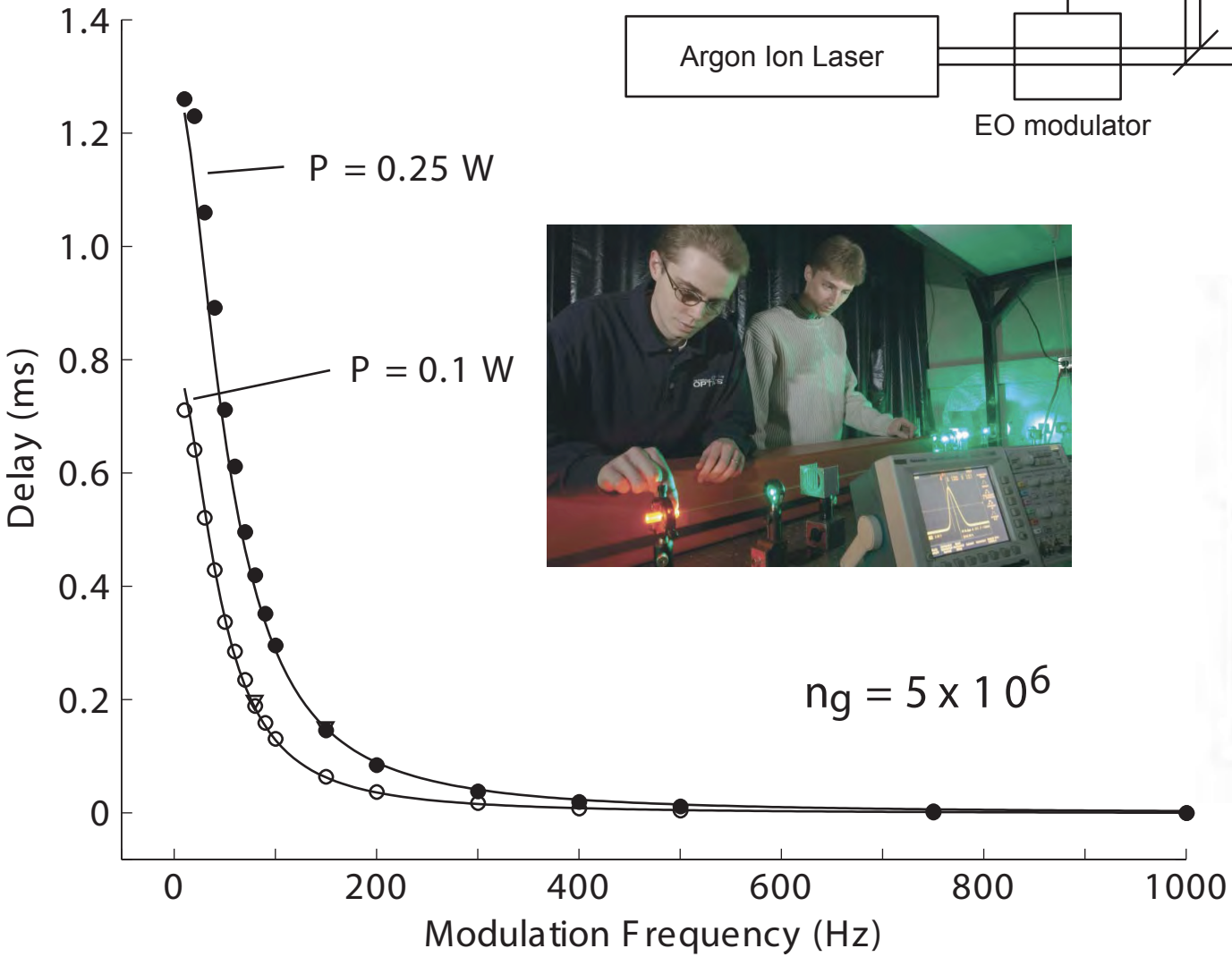


- Basov mechanism: an isolated intense pulse passing through a saturable material experiences a time delay.
  - Assume that $T_{pulse} << T_1$ = time scale for saturation changes
  - Then absorption decreases with time during pulse due to saturation

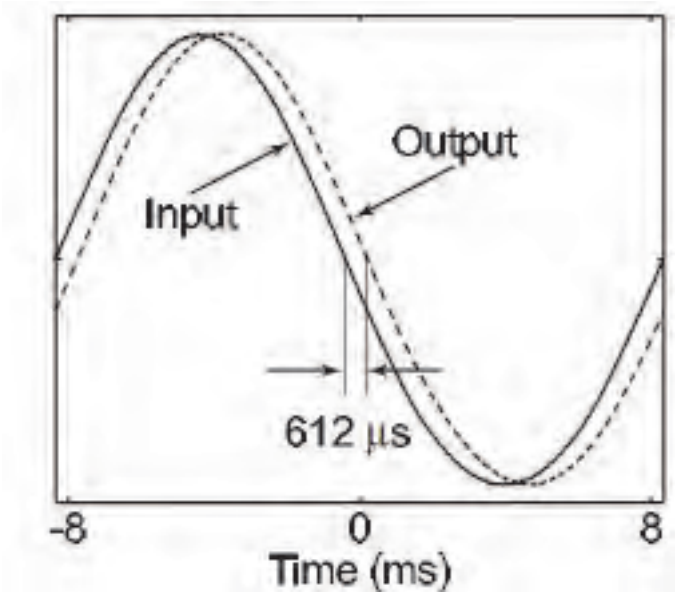


Slow Light in Ruby





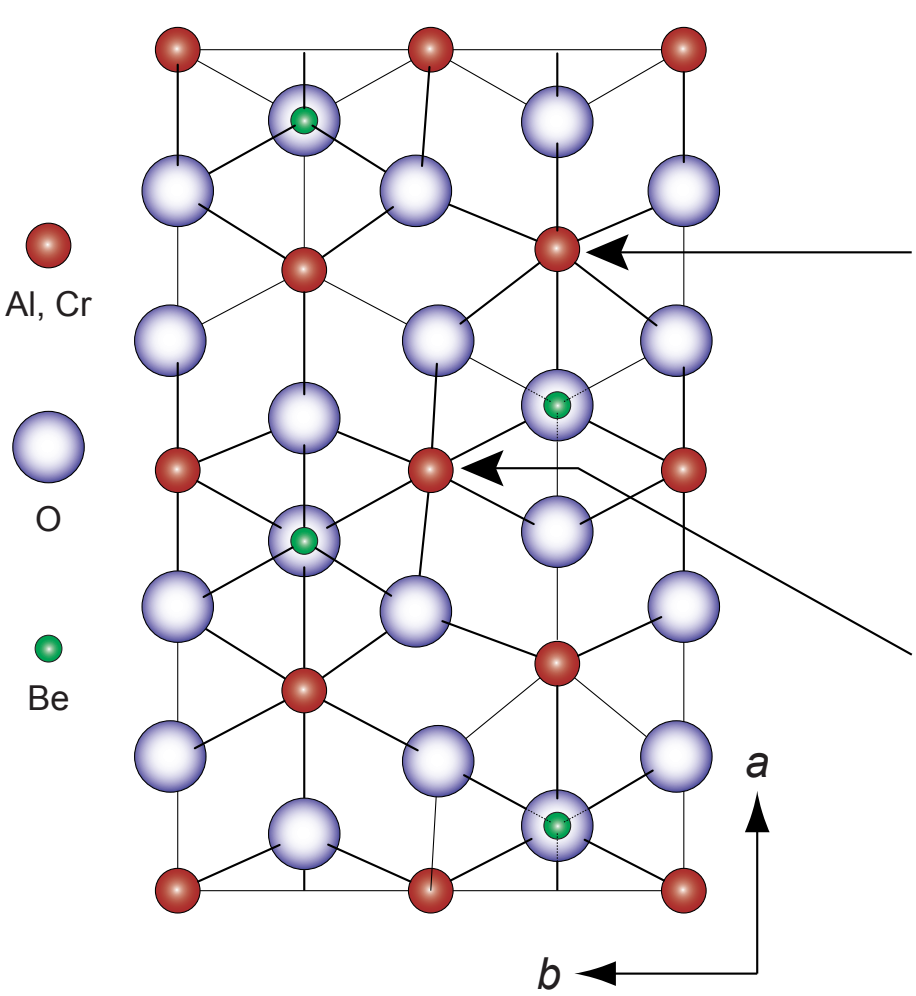
$n_g = 5 \times 10^6$



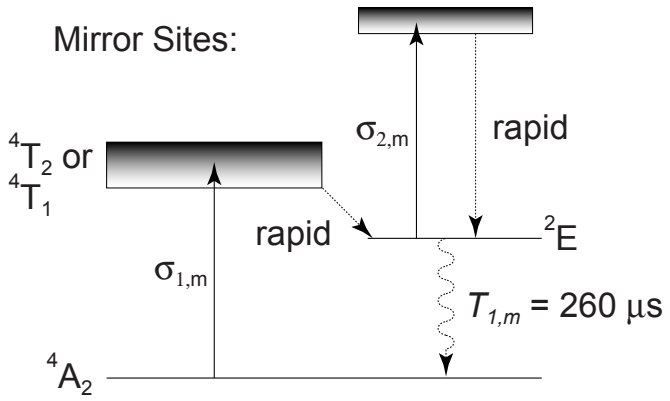
Bigelow et al, PRL 2003

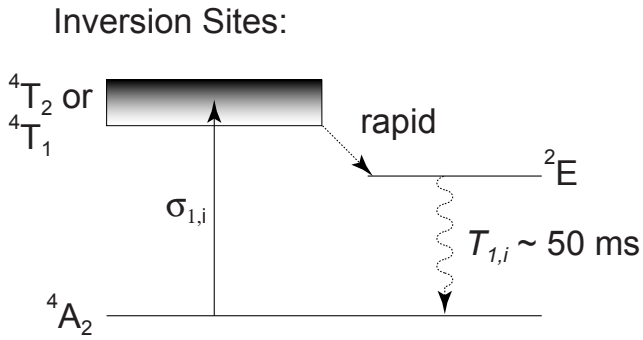
Alexandrite Displays both Saturable and Reverse-Saturable Absorption

- Both slow and fast propagation observed in alexandrite



vg = -800 m/s ng = -3.8 x 10^5



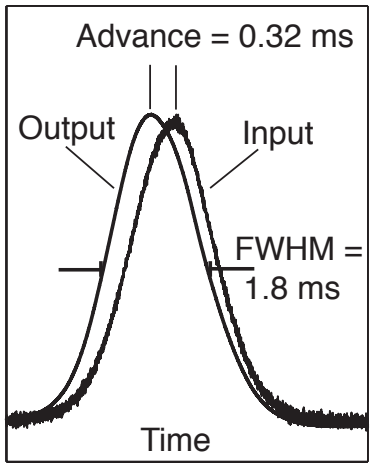


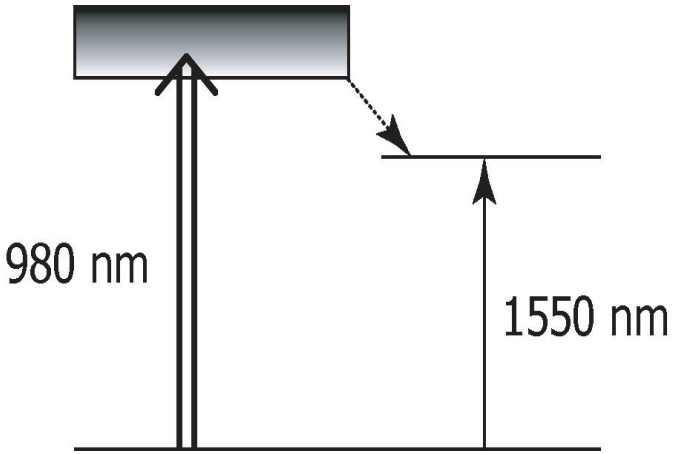
vg = 91 m/s ng = 3.3 x 10^6

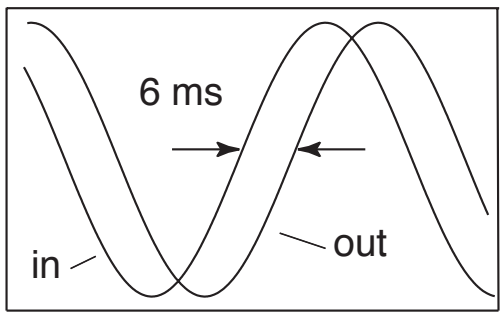
Bigelow, Lepeshkin, and Boyd, Science 301, 200 (2003).

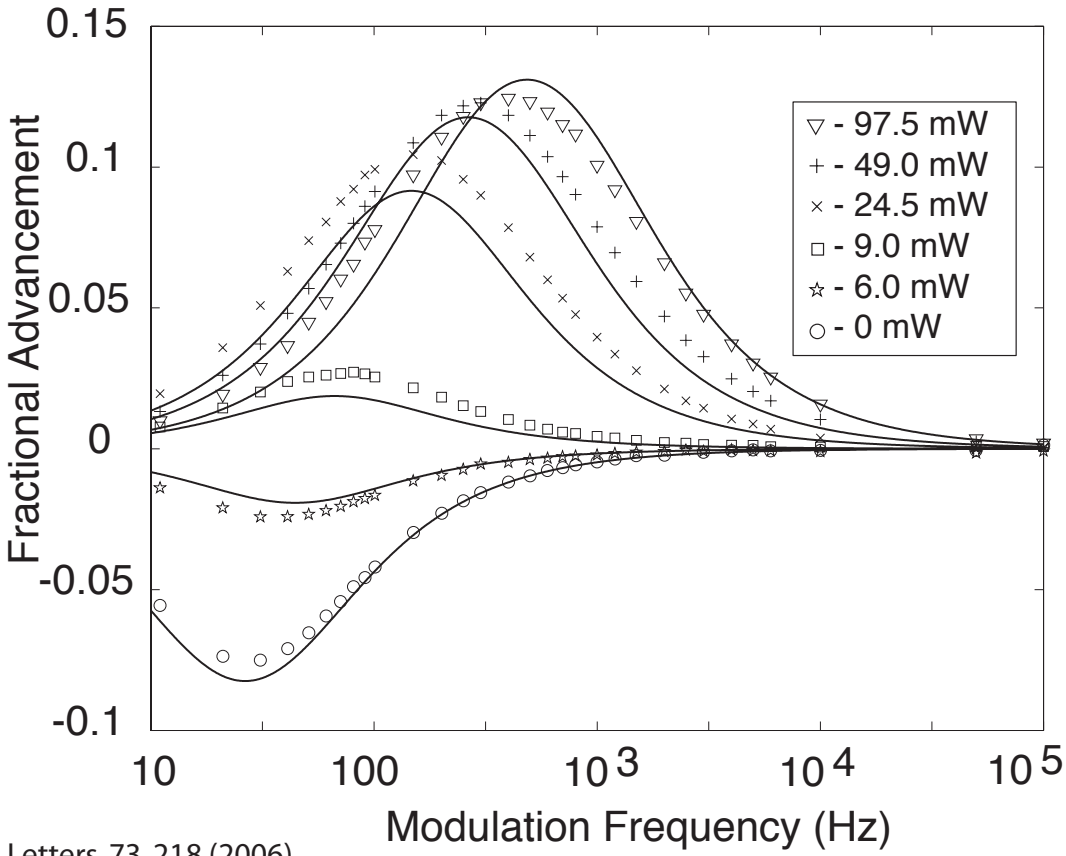
Slow and Fast Light in an Erbium Doped Fiber Amplifier

- 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).

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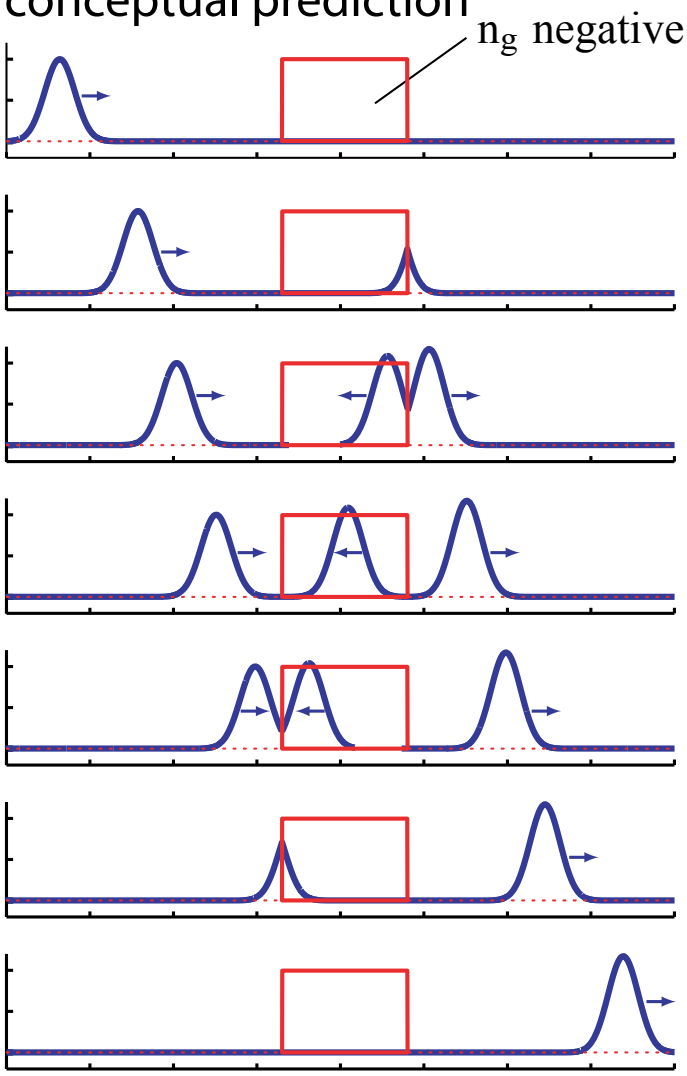
Observation of Superluminal and "Backwards" Pulse Propagation

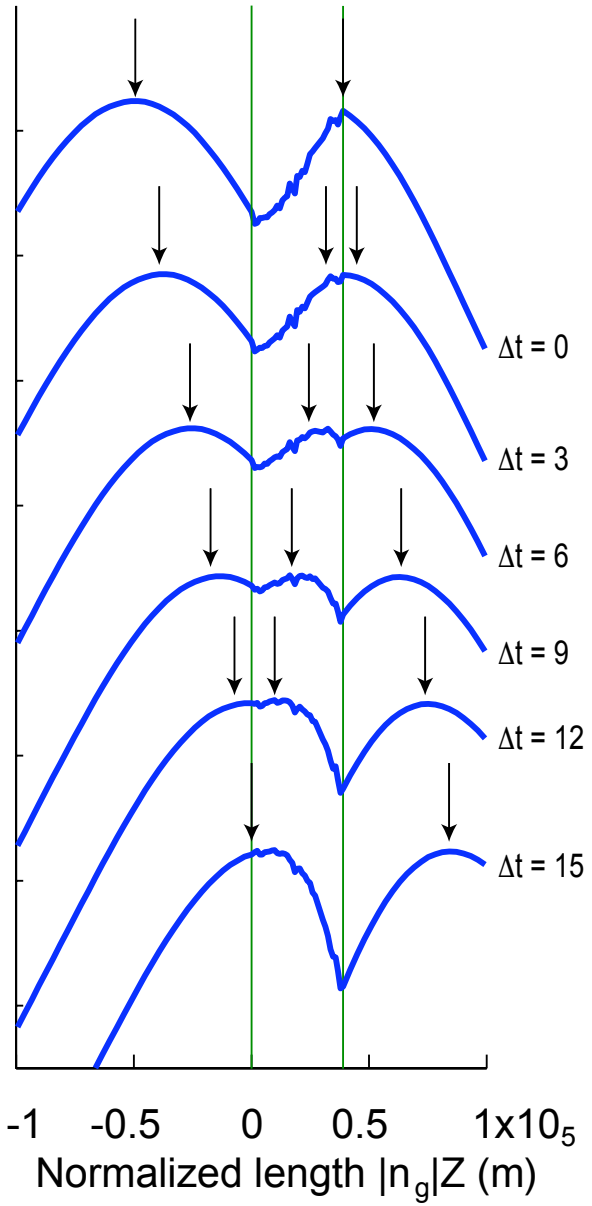
- A strongly counterintuitive phenomenon
- But entirely consistent with established physics
- Predicted by Garrett and McCumber (1970) and Chiao (1993).
- Observed by Gehring, Schweinsberg, Barsi, Kostinski, and Boyd Science 312, 985 2006.

- conceptual prediction

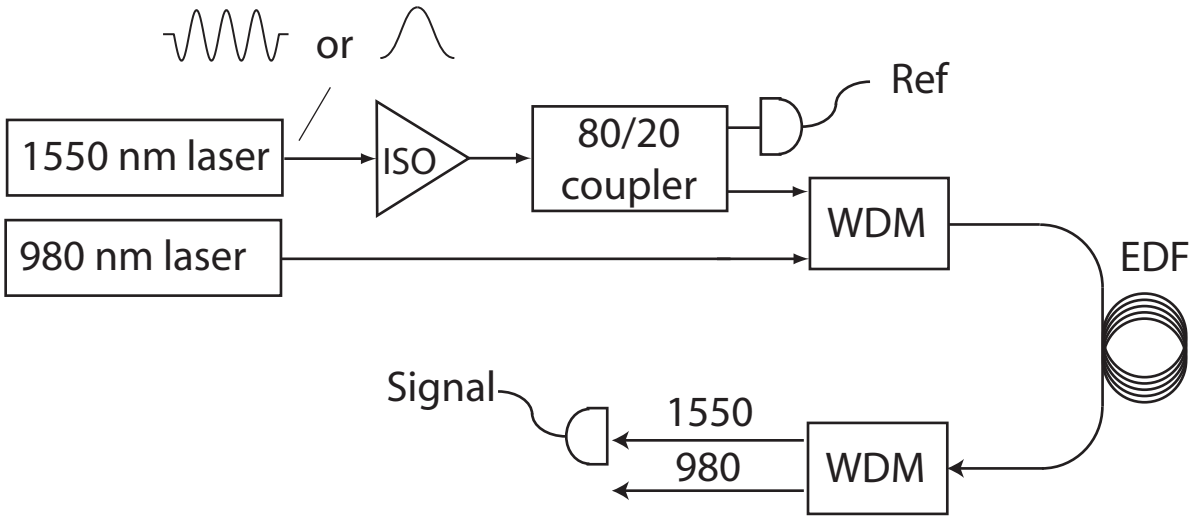


$$\frac{\partial A}{\partial z} - \frac{1}{v_g}\frac{\partial A}{\partial t} = 0$$

- laboratory results



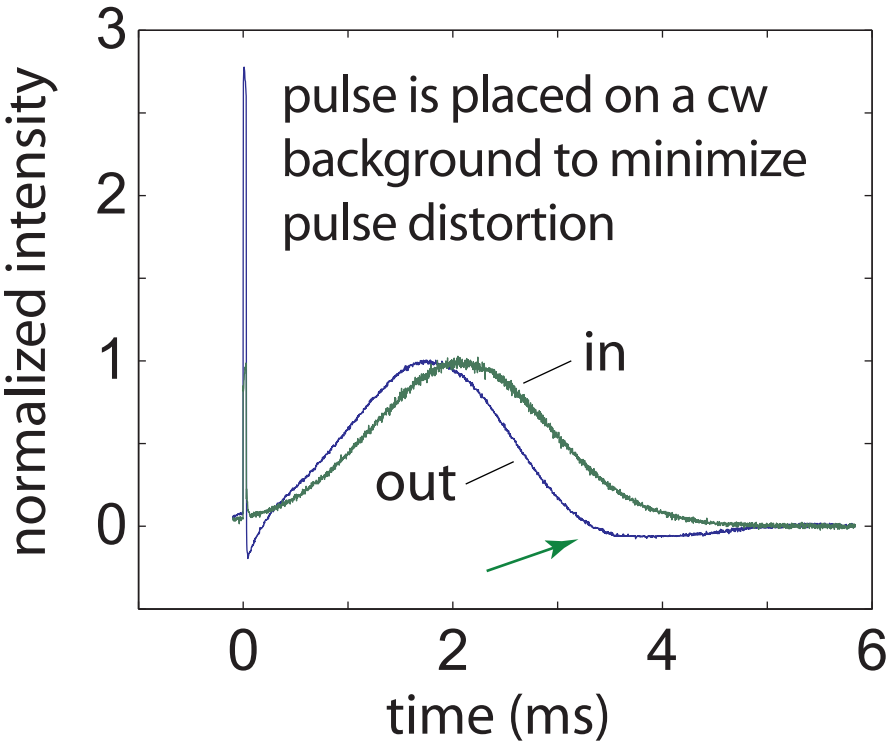
Observation of Backward Pulse Propagation in an Erbium-Doped-Fiber Optical Amplifier



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



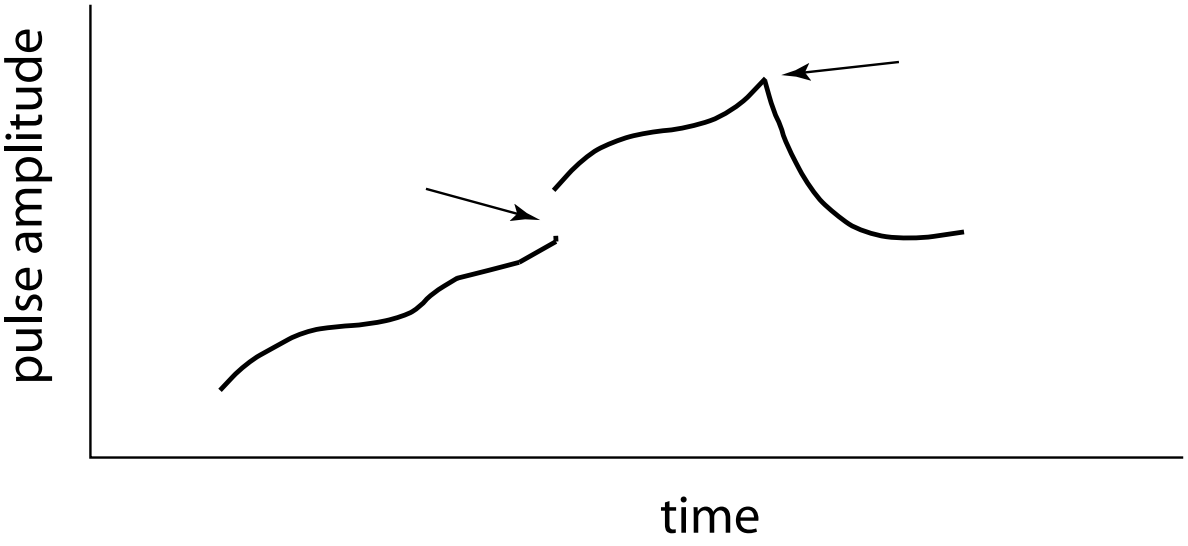
G. M. Gehring, A. Schweinsberg, C. Barsi, N. Kostinski, R. W. Boyd, Science 312, 985 2006.

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."

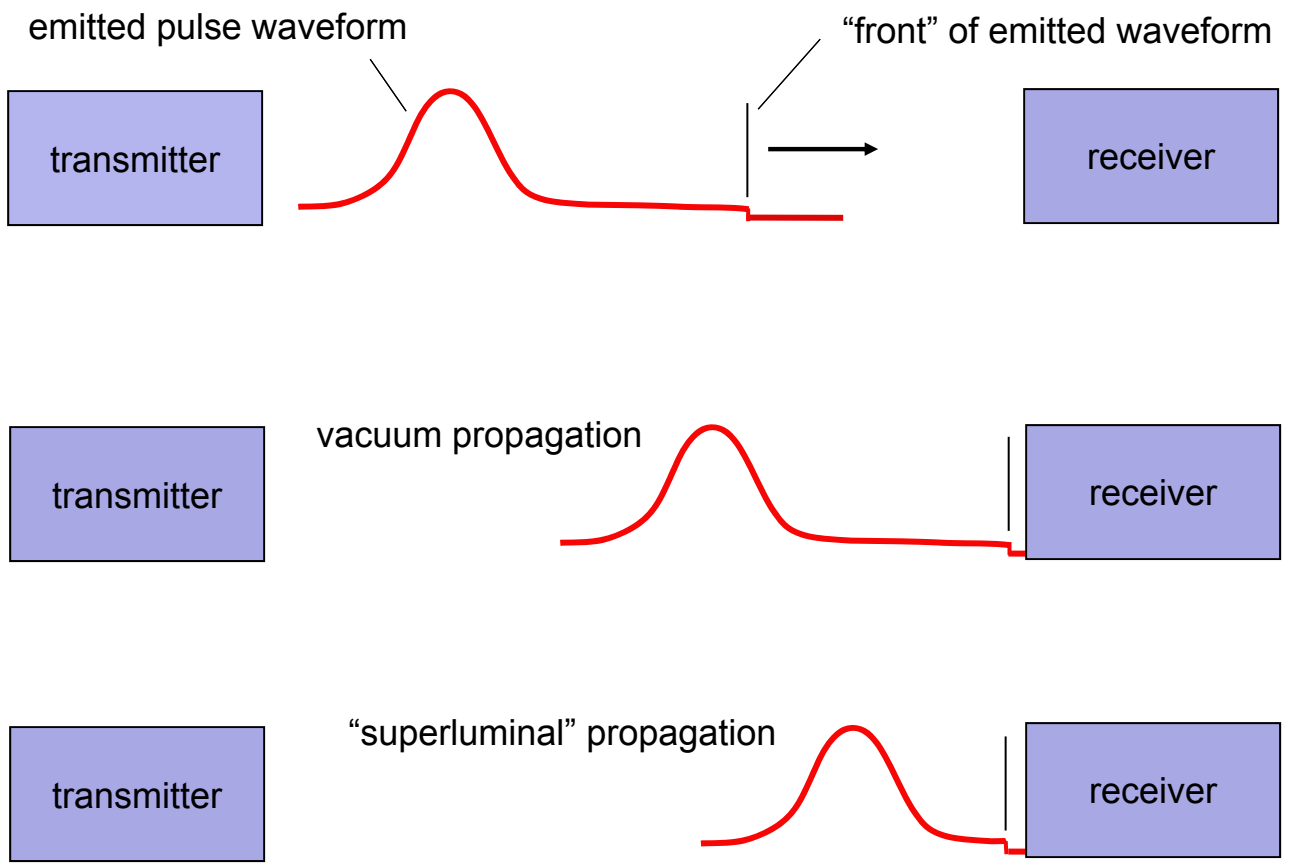
- It is believed that information is carried by points of nonalyticity of a waveform



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

see, for instance, R. Y. Chiao

How to Reconcile Superluminality with Causality



Stenner, Gauthier, and Neifeld, Nature, 425, 695, 2003.

Bigelow, Lepeshkin, Shin, and Boyd, J. Phys: Condensed Matter, 3117, 2006.

Gauthier and Boyd, Photonics Spectra, p. 82 January 2007.

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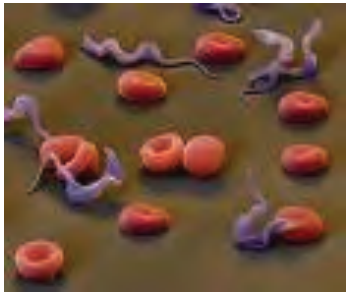
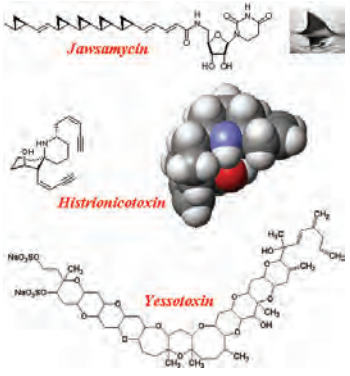
Chip-Scale Spectrometers for Chem-Bio Identification

- Spectroscopy is the standard laboratory procedure for identifying chemical species.
- Can we fabricate miniaturized, chip-scale spectrometers without sustaining a loss in resolution?

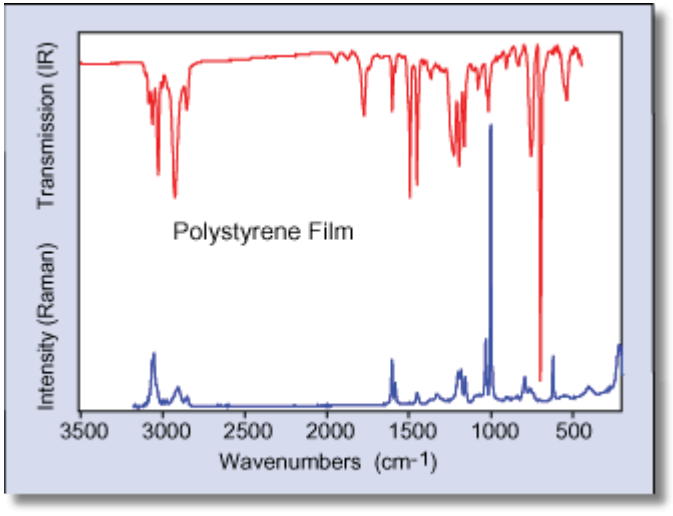
pathogens



toxins



"fingerprint" Raman signature spectrum

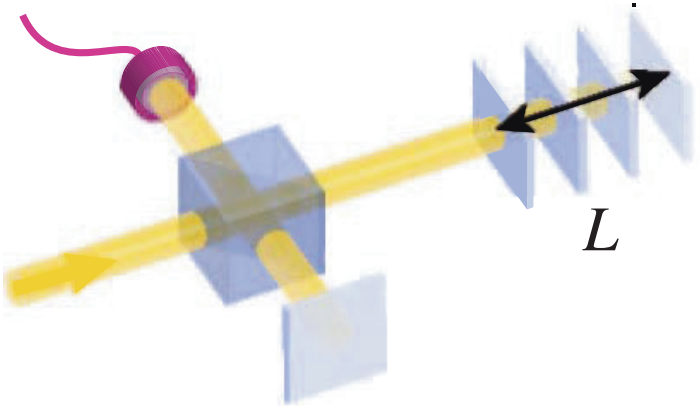


Development of Miniaturized, Chip-Scale Spectrometers

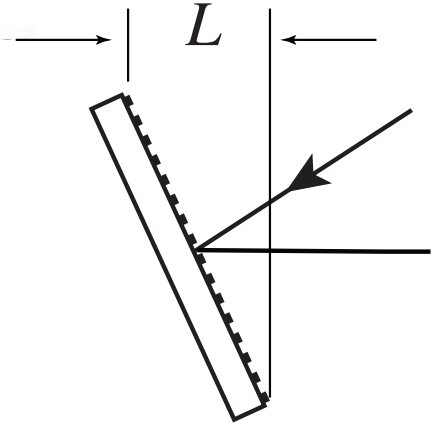
Can We Beat the $1/L$ Resolution Limit of Standard Spectrometers?

- The limiting resolution of a broad class of spectrometers is given (in wave-numbers) by the inverse of a characteristic dimension L of the spectrometer

Fourier-transform spectrometer



Grating spectrometer



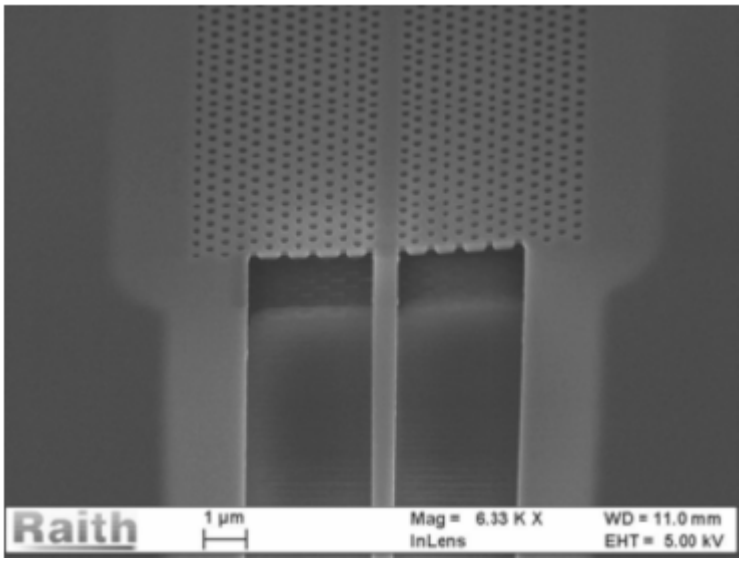
$$\Delta\nu(\text{res}) \approx 1/L$$

- We use slow-light methods to design spectrometers with resolution that exceeds this conventional limit by a factor as large as the group index.
- This ability allows us to miniaturize spectrometers with no loss of resolution, for "lab-on-a-chip" applications.

Our Goal

Replace this:

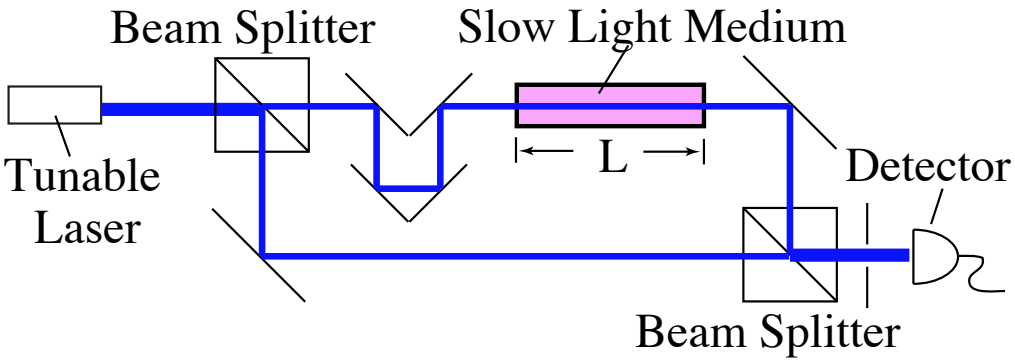
with this:



Our Approach: Chip-Scale Slow-Light Spectrometer

- The spectral sensitivity of an interferometer is increased by a factor as large as the group index of a material placed within the interferometer.
- We want to exploit this effect to build chip-scale spectrometers with the same resoluation as large laboratory spectromters
- Here is why it works:

Slow-light interferometer:

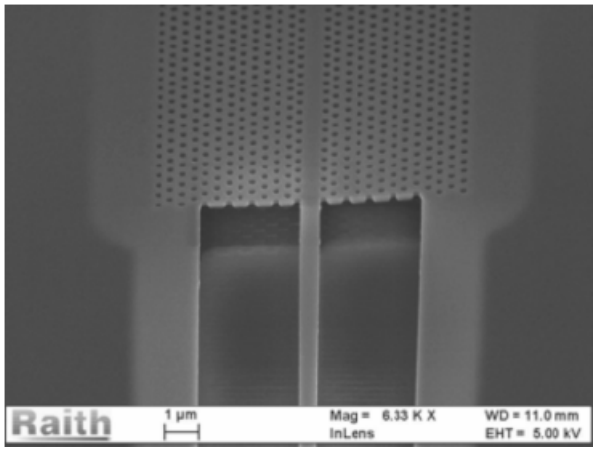


Simple analysis

$$\frac{d}{d\omega}\frac{\Delta\phi}{} = \frac{d}{d\omega}\frac{\omega n L}{c} = \frac{L}{c}\left(n + \omega\frac{dn}{d\omega}\right) = \frac{L n_g}{c}$$

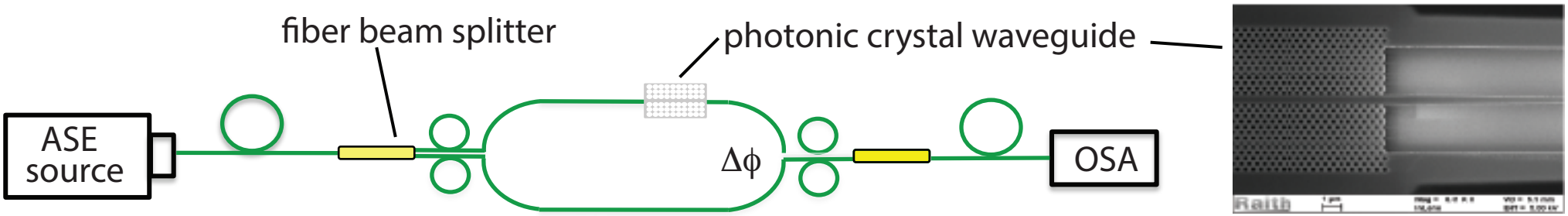
- We use line-defect waveguides in photonic crystals as our slow light mechanism

Slow-down factors of greater than 100 have been observed in such structures.

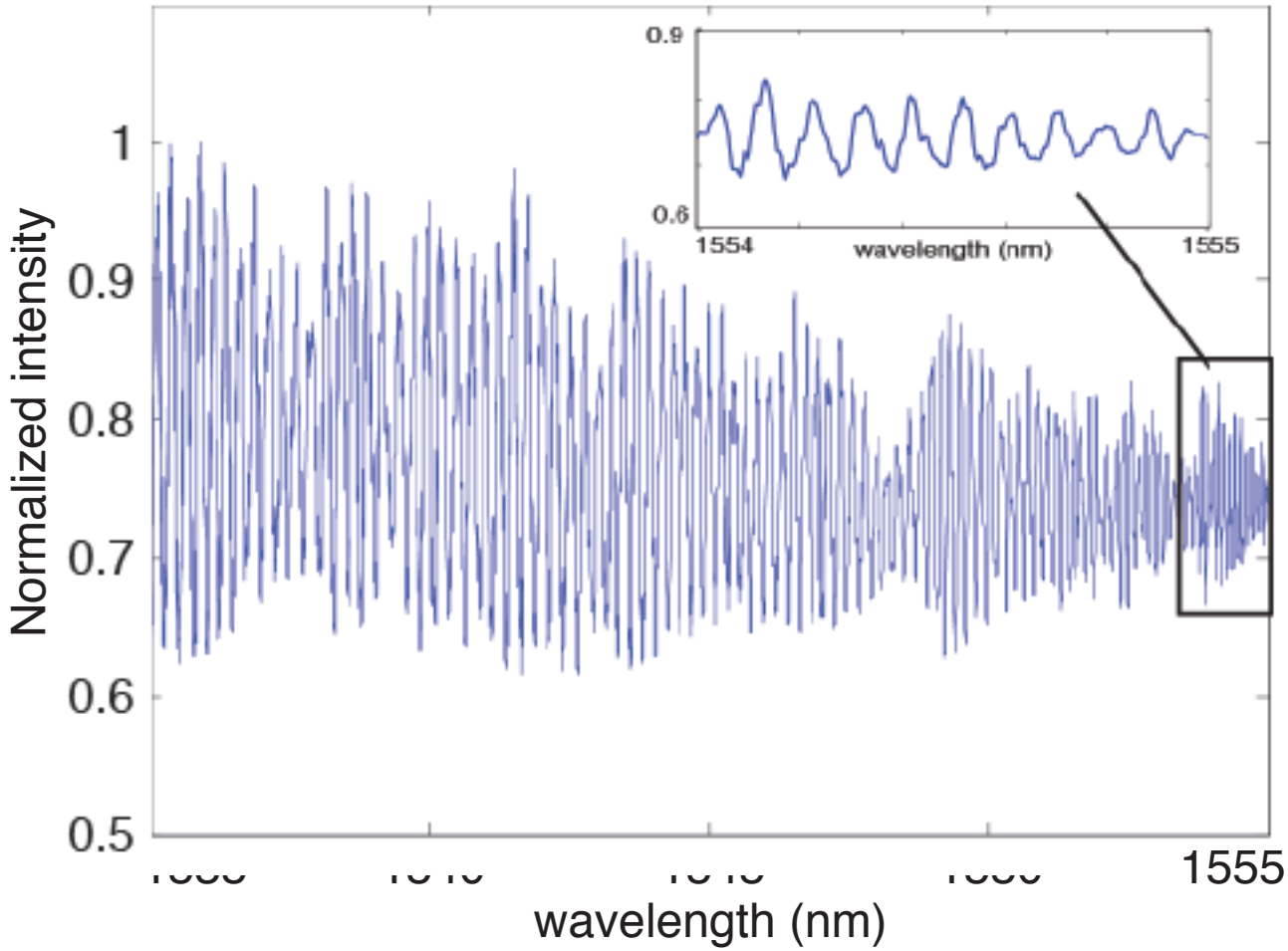


Shi, Boyd, Gauthier, and Dudley, Opt. Lett. 32, 915 (2007)
Shi, Boyd, Camacho, Vudyasetu, and Howell, PRL. 99, 240801 (2007)
Shi and Boyd, J. Opt. Soc. Am. B 25, C136 (2008).

Laboratory Characterization of the Slow-Light Mach-Zehnder Interferometer



- Interference fringes



- Resolution (quarter wave) is 17 pm or 2.1 GHz or 0.071 cm^{-1}
- (Slow-light waveguide is only 1 mm long!)

Magaña-Loaiza, Gao, Schulz, Awan, Upham, Dolgaleva, and Boyd, Opt. Lett. 41, 1431 (2016).

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Why Care about Optical Forces?

- Optical levitation
- Optical tweezers
- Optomechanical systems
- But can we ***control*** optical forces.

  - Yes! Photon momentum and optical forces depend on both refractive index and group index of optical materials.

  - The "slow light" community knows how to manipulate the group velocity of light.

Kinematic Properties of Slow and Fast Light

Poynting's Theorem when derived for a dispersive medium leads to the conclusion that

$$S = \tfrac{1}{2}\, n\, \epsilon_0\, c\, E^2 \quad \text{(intensity)}$$

$$u = \tfrac{1}{2}\, n\, n_g\, \epsilon_0\, E^2 \quad \text{(energy density)}$$

where

$$v_g = c/n_g \quad \text{(group velocity)}.$$

It thus follows that

$$S = u\, v_g.$$

Note:

Large enhancement of stored energy

But no enhancement of E!

See, e.g., Haus, Landau and Lifshitz, Milonni, or Harris and Hau

What is the Momentum of a Photon?

In vacuum: $p = (\hbar\omega/c)$

Abraham form (for light in matter)

$P = E \times H/c^2$ (EM momentum density)

$p = (\hbar\omega/c)(1/n_g)$ (photon momentum)

Minkowski form (for light in matter)

$P = D \times B$ (EM momentum density)

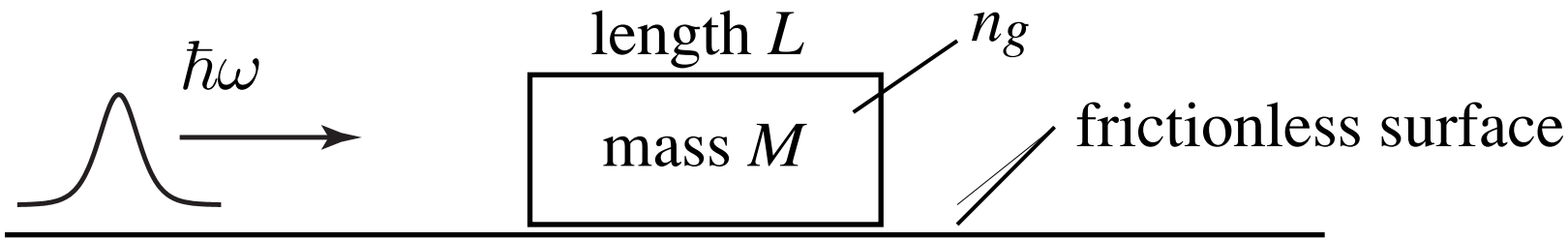
$p = (\hbar\omega/c)(n^2/n_g)$ or $p = (\hbar\omega/c)\, n$ photon momentum

One way or other, photon momentum very small in slow-light medium

See, e.g., Barnett, PRL (2010), Milonni and Boyd, AOP (2010).

rev 1/28/2014

Einstein-Balazs Argument Supports the Abraham Form



- Argue that center of mass-energy must move with a constant velocity
- When photon wavepack enters block, it slows down. Block thus receives a kick into the forward direction.
- When photon leaves block, block receives backward kick and returns to rest.
- Block undergoes longitudinal displacement of

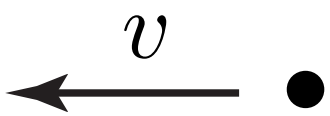
$$\Delta z = (n_g - 1) L \, \hbar\omega / (Mc^2)$$

- Simple kinematic argument shows that momentum of photon in block is

$$p = \hbar\omega / (n_g c) \qquad \text{Abraham form!}$$

Fermi's Argument Supports the Minkowski Form

photon in medium of
refractive index $n(\omega)$



atom with mass m and
resonance frequency ω_0

- Fermi describes Doppler effect in terms of atomic recoil (RMP, 1932)
- Atom can absorb only if $\omega \approx \omega_0(1 - nv/c)$
- Conservation of energy and momentum

$$\text{Initial energy} = \hbar\omega + \tfrac{1}{2}mv^2 \qquad \text{Final energy} = \hbar\omega_0 + \tfrac{1}{2}mv'^2$$

$$\text{Initial momentum} = p + mv \qquad \text{Final momentum} = mv'$$

- Solve: find photon momentum p in medium given by

$$p = n\,\hbar\omega/c \qquad \text{Minkowski form!}$$

Which is Correct, Abraham or Minkowski?

PRL 104, 070401 (2010) PHYSICAL REVIEW LETTERS week ending 19 FEBRUARY 2010

Resolution of the Abraham-Minkowski Dilemma

Stephen M. Barnett

Department of Physics, SUPA, University of Strathclyde, Glasgow G4 0NG, United Kingdom

(Received 7 October 2009; published 17 February 2010)

The dilemma of identifying the correct form for the momentum of light in a medium has run for a century and has been informed by many distinguished contributions, both theoretical and experimental. We show that *both* the Abraham and Minkowski forms of the momentum density are correct, with the former being the kinetic momentum and the latter the canonical momentum. This identification allows us to explain why the experiments supporting each of the rival momenta gave the results that they did. The inclusion of dispersion and absorption provides an interesting subtlety, but does not change our conclusion.

DOI: 10.1103/PhysRevLett.104.070401 PACS numbers: 03.50.De, 42.50.Nn, 42.50.Wk

$$\mathbf{p}_{\mathrm{kin}}^{\mathrm{med}} + \mathbf{p}_{\mathrm{Abr}} = \mathbf{p}_{\mathrm{can}}^{\mathrm{med}} + \mathbf{p}_{\mathrm{Min}}.$$

Total momentum (field plus material) the same in either treatment!

What is the Momentum of a Photon?

In vacuum: $p = (\hbar\omega/c)$

Abraham form (for matter)

$P = E \times H/c^2$ (EM momentum density)

$p = (\hbar\omega/c)(1/n_g)$ (photon momentum)

It is the kinetic (as in mv) momentum

It is the momentum of the field (alone)

It is what comes out of Balazs's moving block analysis

Minkowski form (for matter)

$P = D \times B$ (EM momentum density)

$p = (\hbar\omega/c)(n^2/n_g)$ or $p = (\hbar\omega/c)\, n$ photon momentum

It is the canonical momentum (as in $h/\lambda_{deBroglie}$)

It is the momentum of field and (at least part of that of the) matter

It is what comes out of a Doppler shift analysis

One way or other, photon momentum very small in slow-light medium

See, e.g., Barnett, PRL (2010), Milonni and Boyd, AOP (2010).

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Photon Drag Effects with Slow Light

We would like to use the dependence of the photon momentum on the group index as a means to control optical forces.

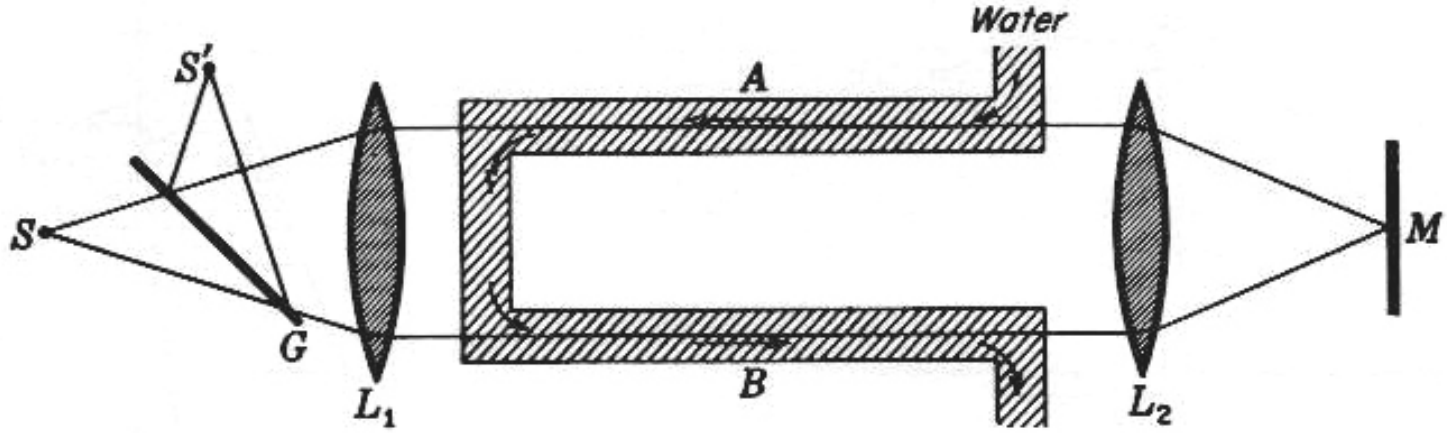
As a first step down this pathway, we are studying how to control photon drag effects using slow light.

The Velocity of Light in Moving Matter: Fresnel Drag (or Ether Drag) Effects

- Fizeau (1859): Longitudinal photon drag:

  Velocity of light in flowing water.

  V = 700 cm/sec; L = 150 cm; displacement of 0.5 fringe.



- Modern theory: relativistic addition of velocities

$$v = \frac{c/n + V}{1 + (V/c)(1/n)} \approx \frac{c}{n} + V\left(1 - \frac{1}{n^2}\right)$$

Fresnel "drag" coefficient

- But what about slow-light media?

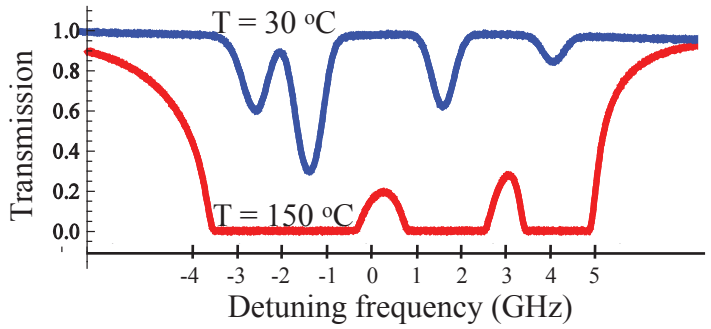
Fresnel Drag in a Highly Dispersive Medium

Light Drag in a Slow Light Medium (Lorentz)

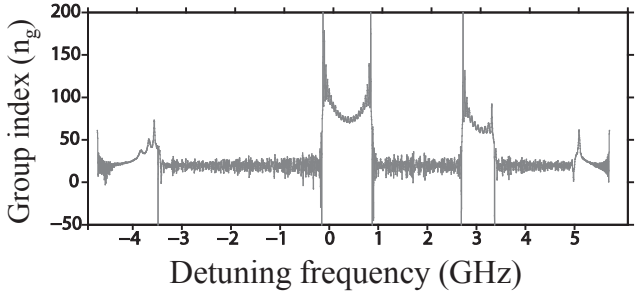
$$u \simeq \frac{c}{n} \pm v\left(1 - \frac{1}{n^2} + \frac{n_g - n}{n^2}\right)$$

We Use Rubidium as Our Slow Light Medium

- Transmission spectrum of Rb around D_2 transition:

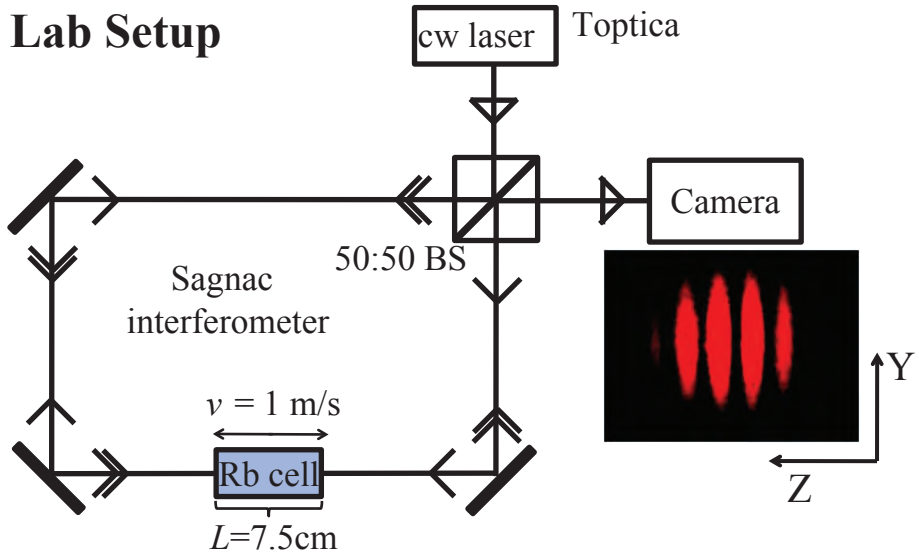


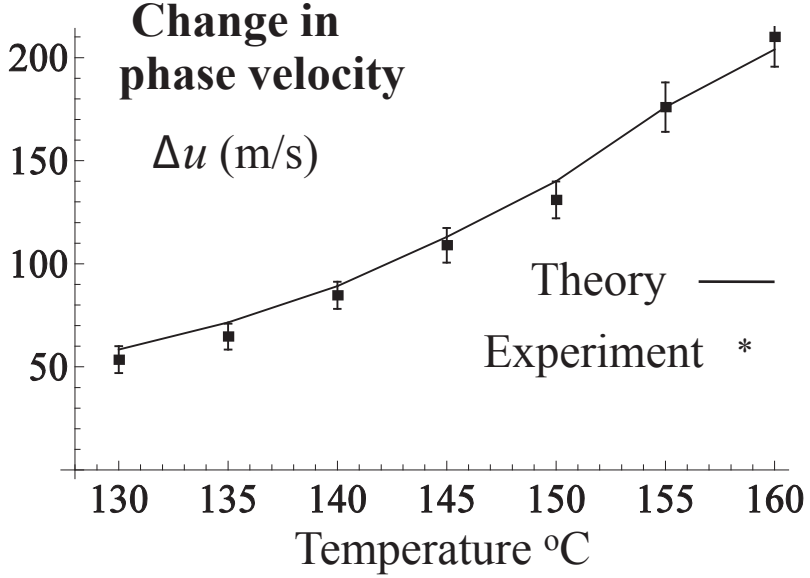
- Group index of Rb around D_2 line at T=130



Safari, De Leon, Mirhosseini, Magana-Loaiza, and Boyd
Phys. Rev. Lett. 116, 013601 (2016)

Lab Setup





- **Change in phase velocity is much larger than velocity of rubidium cell. Implications for new velocimeters?**

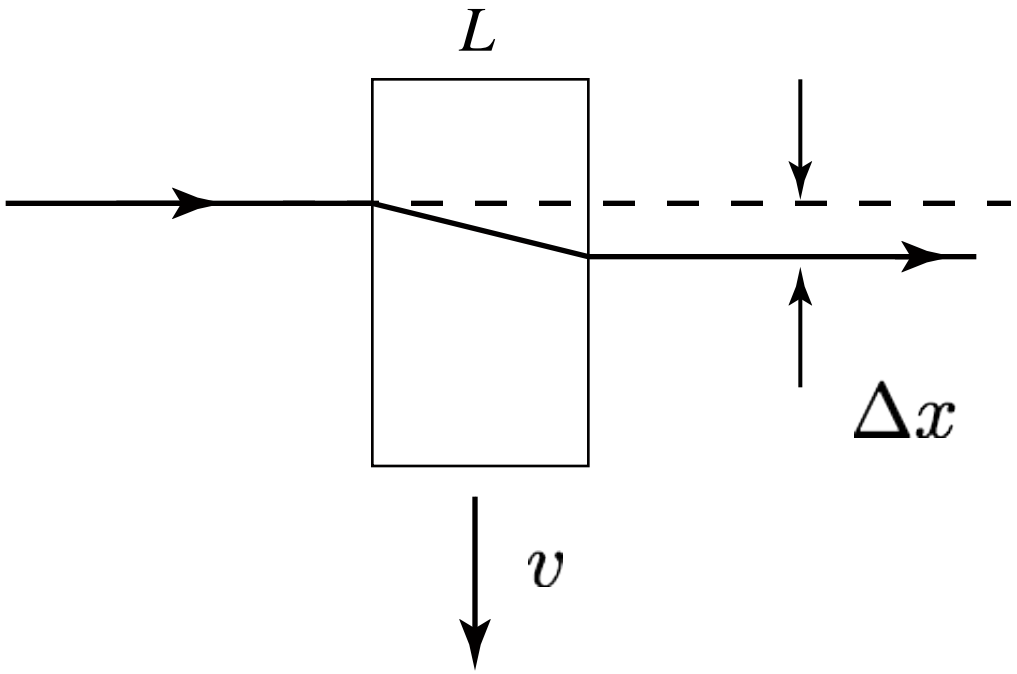
Conclusions

- A maximum drag speed of 205 m/s was measured in a highly dispersive medium (hot Rb vapor).

- This effect is at least two orders of magnitude larger than that observed to date.

- Much larger dispersion can be achieved in Rb atoms using electromagnetically induced transparency (n_g as high as 10^7).

Transverse Photon Drag

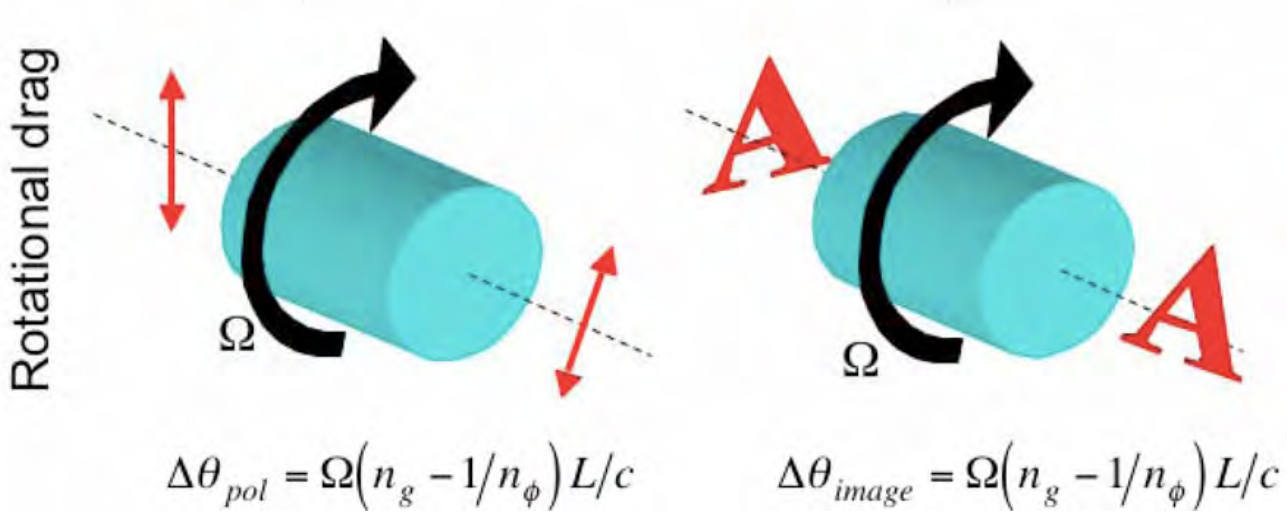


$$\Delta x = (vL/c)(n_g - 1/n_\phi)$$

For L = 25 mm, v = 2000 cm/s, displacement = 6 nm.

Measured by R.V. Jones, 1972.

Rotary Photon Drag: An image viewed through a spinning window



Theory says that transmitted image is rotated! (rotary photon drag)

(Polarization rotation measured earlier by Jones.)

Image rotation never previously observed
(although implied by work of Leach et al., PRL 2008.)

Effect scales as group index!

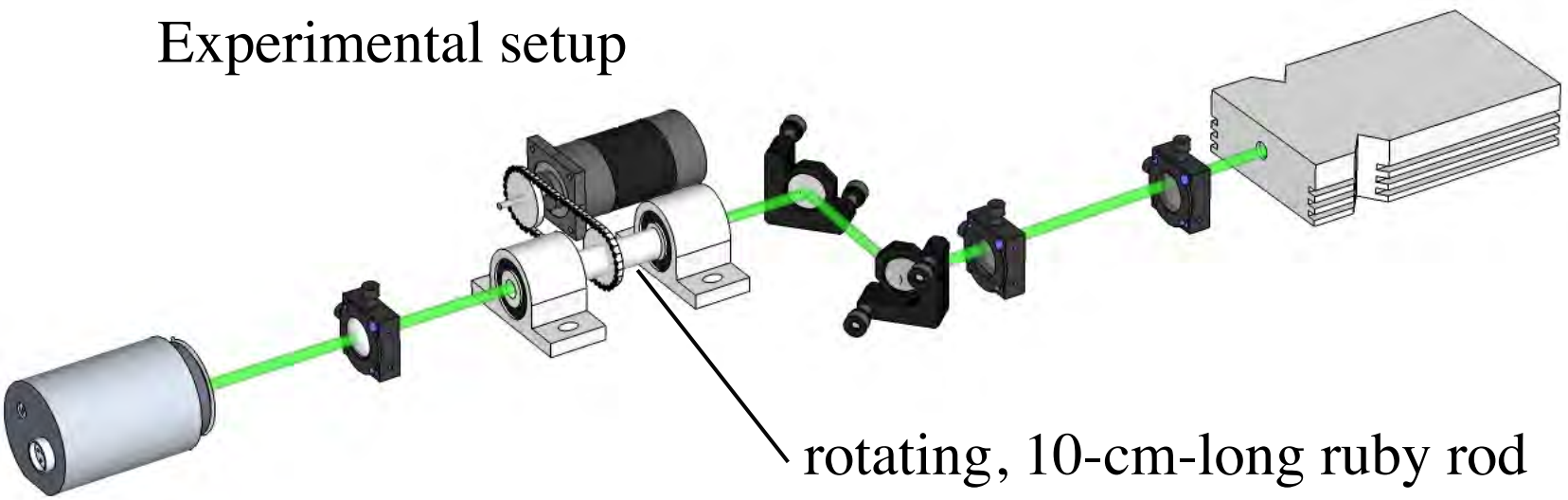
Franke-Arnold, Gibson, Boyd and Padgett, Science, 2011

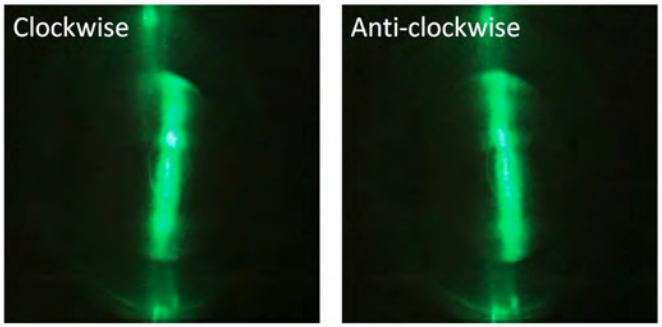
Observation of Rotary Photon Drag

The world as seen through a spinning window.

(Laser-excited ruby has a group index of 10^6.)

Experimental setup





Effect clearly visible by eye!

Franke-Arnold, Gibson, Boyd and Padget, Science, 2011

Slow Light and Fast Light: A Tutorial Review

1. Introduction to slow light
2. Mechanisms of slow light
3. Backwards light and causality
4. Slow light spectrometer
5. Optical forces
6. Fresnel drag
7. Other applications

Applications of Slow and Fast Light

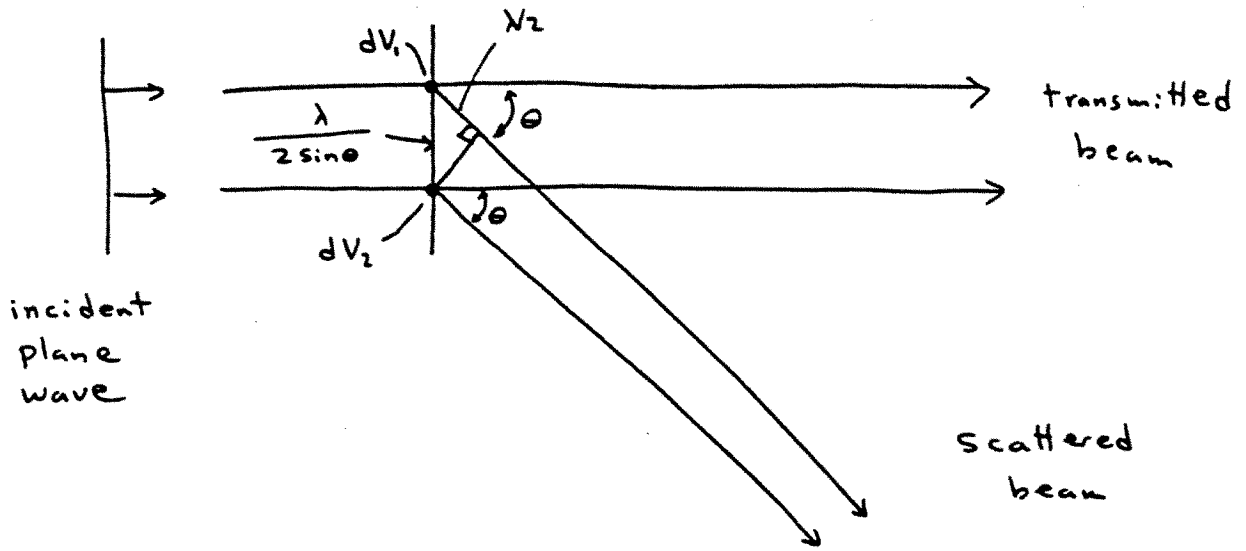
- Buffers and regenerators for telecom
- Slow/fast light for interferometery
- Phased- and synchronized-array laser radar
- Construction of quantum memories

9. 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

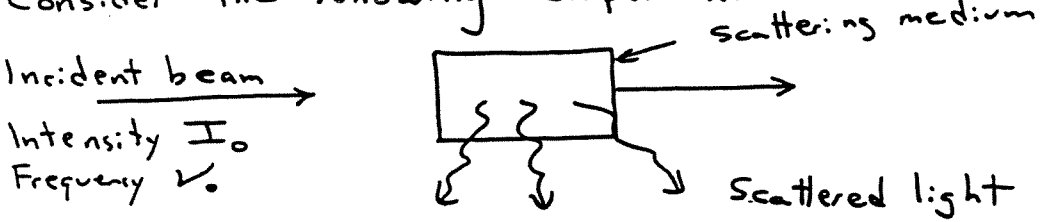
Fluctuations (e.g., in ε) occur due to thermal (or quantum mechanical zero-point) excitation.

Stimulated light scattering

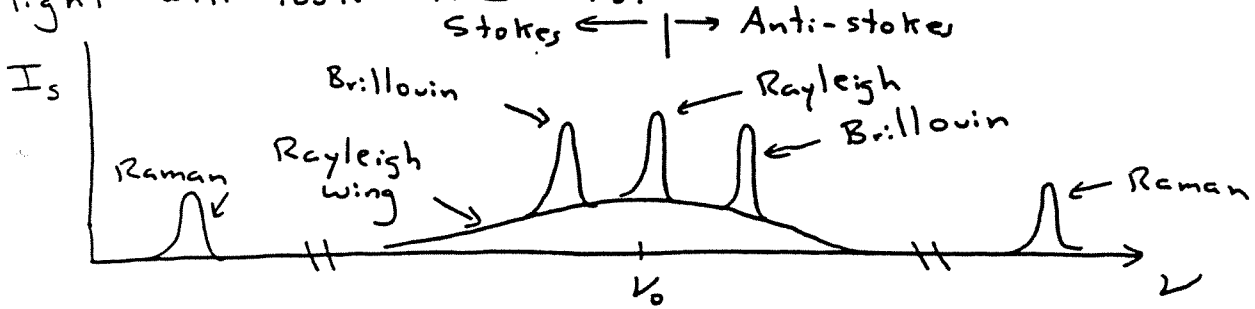
Fluctuations are induced by the incident laser field.

Spontaneous Light Scattering

Consider the following experiment:

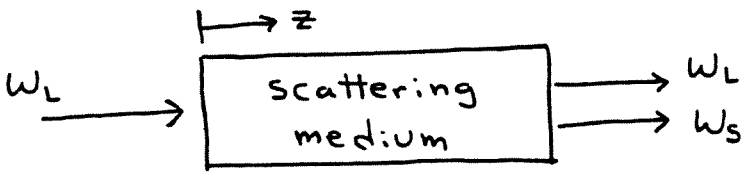


In the most general case, the spectrum of the scattered light will look like this:



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}

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+1): # 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

$$G = D\, m_L$$

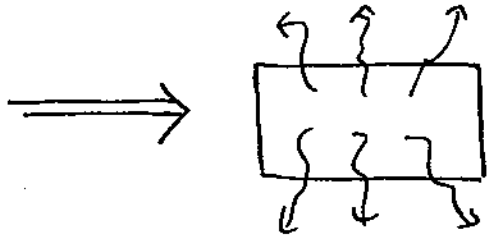
We can relate G to the spontaneous scattering cross section:

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

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

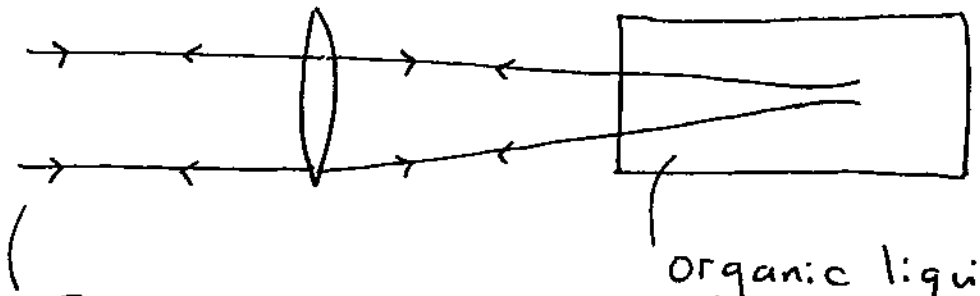
Stimulated Brillouin Scattering

- Spontaneous Brillouin scattering is a very weak process.

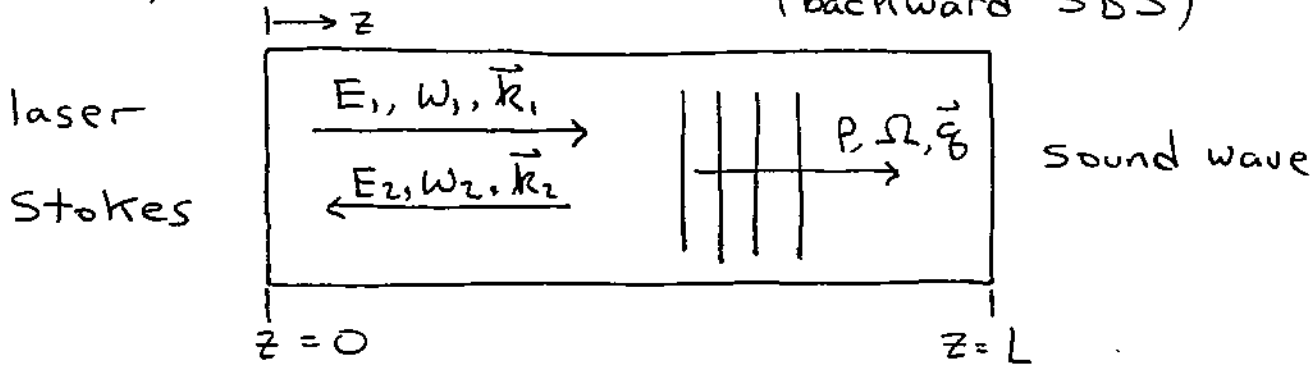


[~ 1 part in 10^5 of incident scattered per cm of liquid.]

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



Theory of SBS (backward SBS)



Positive feed back:

laser + Stokes ⟶ sound

laser + sound ⟶ Stokes

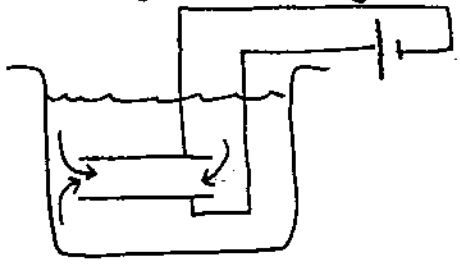
Frequencies:

$\omega_2 = \omega_1 - \Omega_B$ where $\Omega_B = |\vec{q}|\, v$ ← velocity of sound

$\vec{q} = \vec{k}_1 - \vec{k}_2 \simeq 2\vec{k}_1$ (since $v \ll c$, $\Omega \ll \omega_1$)

$\Rightarrow \Omega_B = 2\omega \frac{v}{c/n}$ $\left(\Omega_B/2\pi \approx 5\ \text{GHz}\right)$

Coupling among waves: electrostriction



Material tends to be drawn into regions of high field.

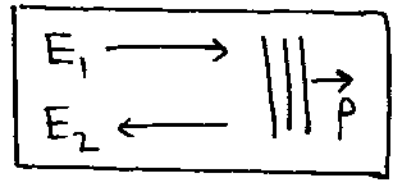
$$p_{st} = -\gamma_e \frac{E^2}{8\pi}$$

$\gamma_e \equiv \rho \frac{\partial \varepsilon}{\partial \rho} \approx 1$ for condensed matter.

$\frac{\Delta\rho}{\rho} = -C\, p_{st}$ where $C \equiv \frac{1}{\rho}\left(\frac{\partial \rho}{\partial p}\right)$ = compressibility

$\Delta\varepsilon = \left(\rho \frac{\partial \varepsilon}{\partial \rho}\right) \frac{1}{\rho}\ \Delta\rho$

Theory of SBS



$$\tilde{E}_1(z,t) = A_1(z,t)\, e^{i(k_1 z - \omega_1 t)} + cc$$

$$\tilde{E}_2(z,t) = A_2(z,t)\, e^{i(-k_2 z - \omega_2 t)} + cc$$

$$\tilde{\rho}(z,t) = \rho_0 + \left[\rho(z,t)\, e^{i(qz - \Omega t)} + cc\right] \qquad \begin{cases} q \equiv 2k_1 \\ \Omega \equiv \omega_1 - \omega_2 \end{cases}$$

density $\tilde{\rho}$ obeys acoustic wave equation

$$\frac{\partial^2 \tilde{\rho}}{\partial t^2} - \Gamma' \nabla^2 \frac{\partial \tilde{\rho}}{\partial t} - v^2 \nabla^2 \tilde{\rho} = +\nabla \cdot \vec{f}$$

$$\Gamma' = \frac{1}{\rho_0}\left[\frac{4}{3}\eta_s + \eta_b + \frac{K}{C_p}(\gamma - 1)\right]$$

$$\vec{f} = \nabla p_{st} \qquad p_{st} = -\gamma_e \frac{\langle \tilde{E}^2 \rangle}{8\pi}$$

Thus $$\nabla \cdot \vec{f} = \frac{\gamma_e q^2}{4\pi}\left[A_1 A_2^* e^{i(qz - \Omega t)} + cc\right]$$

In the SVAA, acoustic wave equation becomes

$$-2i\Omega \frac{\partial \rho}{\partial t} + \left(\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B\right)\rho - 2iqv^2 \frac{\partial \rho}{\partial z} = \frac{\gamma_e q^2}{4\pi} A_1 A_2^*$$

where $$\Gamma_B \equiv q^2 \Gamma' \qquad \Omega_B \equiv 2\omega \frac{v}{c/n}$$

Theory of SBS (continued)

Simplifications: (1) $\partial\rho/\partial t = 0$ in steady state

(2) $\partial\rho/\partial z = 0$ (phonons strongly damped) ($\alpha_s \lesssim 10\,\mu m$)

Then

$$\rho(z,t) = \frac{\gamma_e \varepsilon^2}{4\pi} \frac{A_1 A_2^*}{\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B}$$

- Optical field obeys

$$\frac{\partial^2 \tilde{E}_i}{\partial z^2} - \frac{1}{(c/n)^2}\frac{\partial^2 \tilde{E}_i}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 \tilde{P}_i}{\partial t^2}$$

where

$$P^{NL} = \Delta\chi\,\tilde{E} = \frac{\Delta\varepsilon}{4\pi}\tilde{E} = \frac{1}{4\pi\rho_0}\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)(\tilde{\rho}-\rho_0)\tilde{E}$$

$$= \frac{1}{4\pi\rho_0}\gamma_e(\tilde{\rho}-\rho_0)\tilde{E}$$

Thus

$$\frac{\partial A_1}{\partial z} + \frac{1}{c/n}\frac{\partial A_1}{\partial t} = \frac{i\omega\gamma_e}{2ncp_0}\rho A_2$$

$$-\frac{\partial A_2}{\partial z} + \frac{1}{c/n}\frac{\partial A_2}{\partial t} = \frac{i\omega\gamma_e}{2ncp_0}\rho^* A_1$$

At the Brillouin resonance ($\Omega = \Omega_B$) and in steady state

$$\frac{dA_1}{dz} = \frac{-\omega\gamma_e^2\varepsilon^2}{8\pi nc\rho_0\Omega\Gamma}(A_2 A_2^*)A_1$$

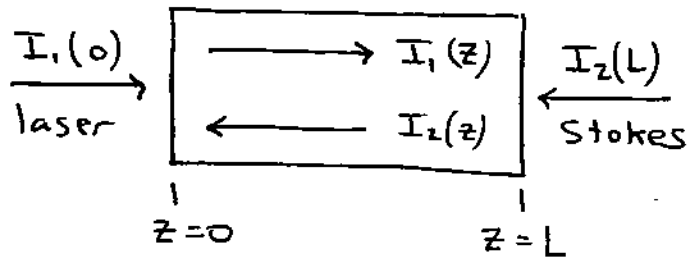
$$\frac{dA_2}{dz} = -\frac{\omega\gamma_e^2\varepsilon^2}{8\pi nc\rho_0\Omega\Gamma}(A_1 A_1^*)A_2$$

Theory of SBS (continued)

Can express in terms of intensities $I_i = (nc/2\pi)|A_i|^2$

$$\frac{dI_1}{dz} = -g_0 I_1 I_2$$

$$\frac{dI_2}{dz} = -g_0 I_1 I_2$$



$$g_0 = \frac{\gamma_e^2 \omega^2}{n v c^3 \rho_0 \Gamma}$$ ← SBS gain factor

~ 0.01 cm/MW

(recall that $\Gamma \sim \omega^2$)

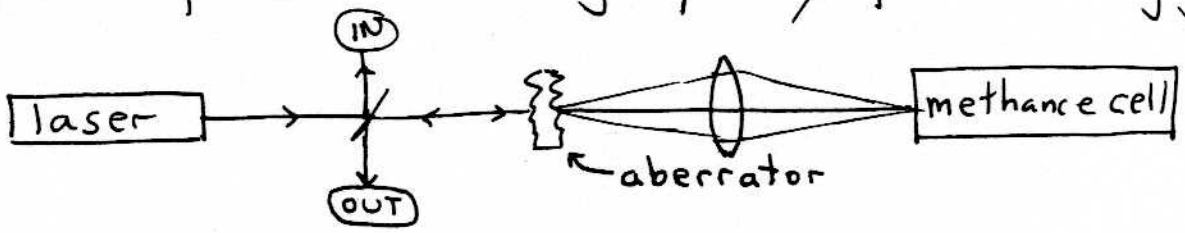
Solution in constant-pump limit (I_1 constant)

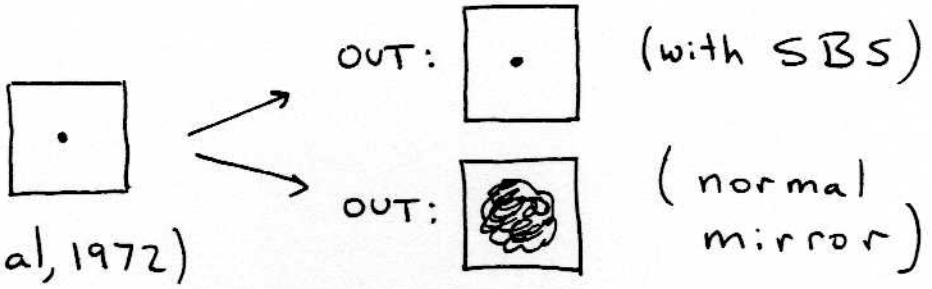
$$I_2(z) = I_2(L)\, e^{g I_1 (L-z)}$$

Total amplification $= I_2(0)/I_2(L) = e^{g I_1 L}$

Phase Conjugation by SBS

SBS can produce high quality phase conjugation

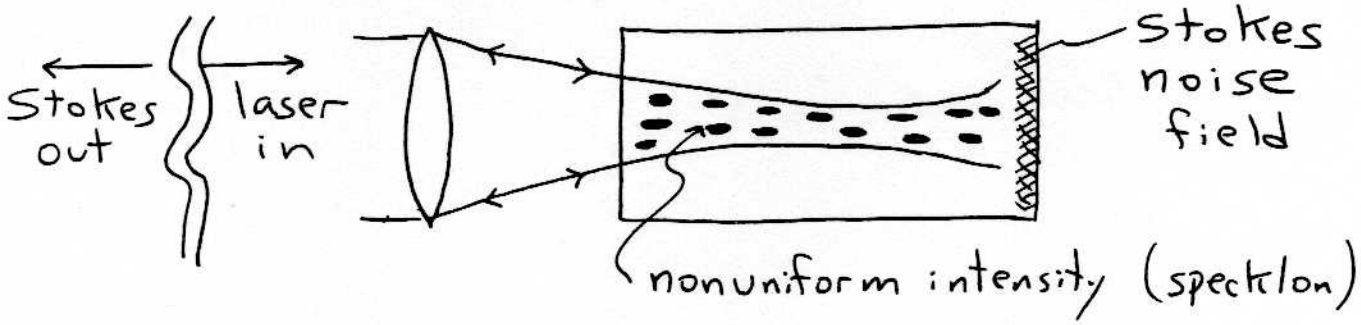




(Zel'dovich et al, 1972)

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.

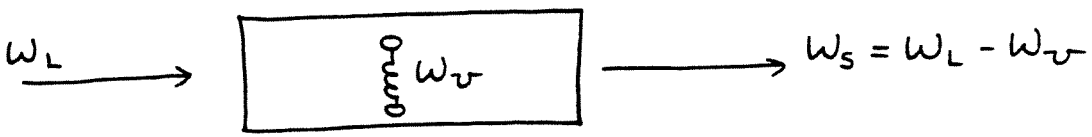
Stokes field is most efficiently amplified if it spatially overlaps the gain distribution.

Stokes field grows from noise

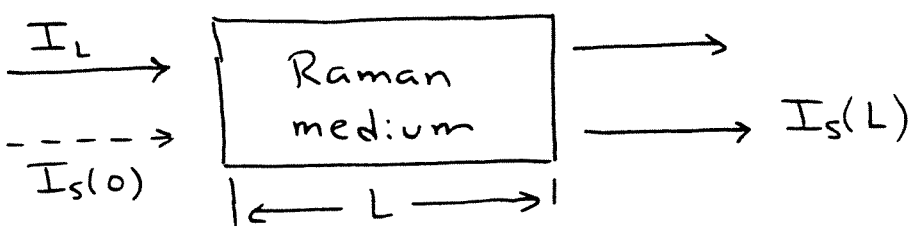
⇒ Stokes output wavefronts coincide with input laser wavefronts

⇒ $E_s \propto E_L^*$

Stimulated Raman Scattering



We can predict the Stokes output intensity:



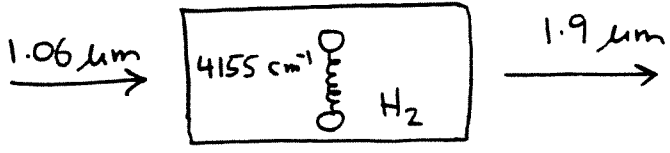
$$I_S(L) = I_S(0)\, e^{g I_L L}$$

g = Raman gain factor

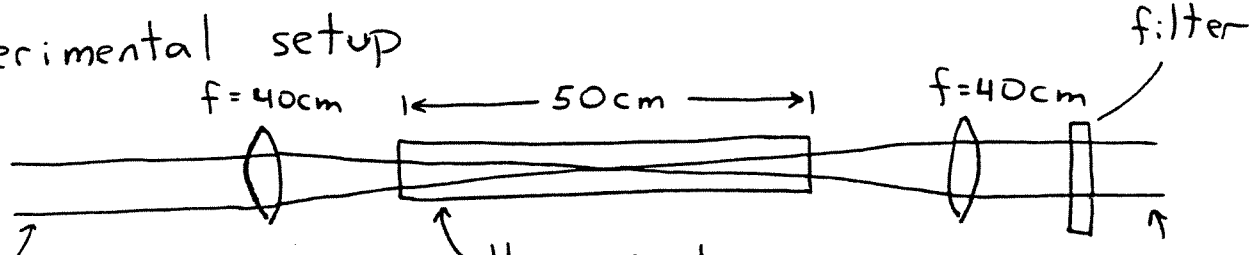
$\simeq 10^{-9}$ cm/W for most materials

Example of Raman Frequency Shifter

Need 1.9 μm light



Experimental setup



$\lambda = 1.06\,\mu m$

30 ps pulse length

3 mJ pulse energy

H_2 gas at 30 atm

$\lambda = 1.9\,\mu m$

150 μJ energy

5% conversion

Typical Raman media

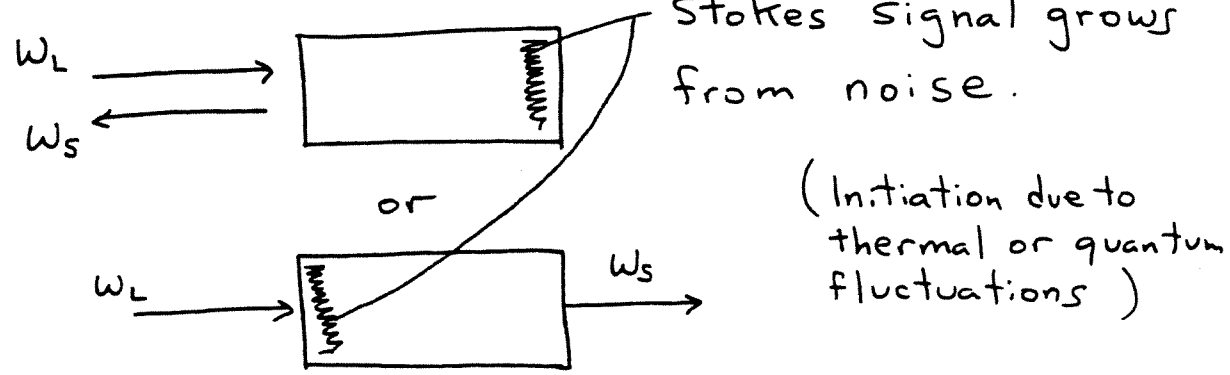
material	$\Delta\nu$ (cm^{-1})	gain g x 10^3 (cm/MW)
LIQUIDS		
benzene	992	3
water	3290	0.14
N_2	2326	17
O_2	1555	16
GASES		
methane	2916	0.66 (10 atm, 500 nm)
hydrogen	4155 (vibrational)	1.5 (above 10 atm)
	450 (rotational)	0.5 (above 0.5 atm)
deuterium	2991 (vibrational)	1.1 (above 10 atm)
N_2	2326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

Configurations for Stimulated Light Scattering

Generator

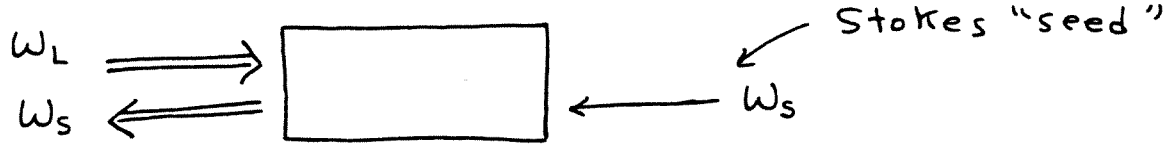
Apply only laser frequency



(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.

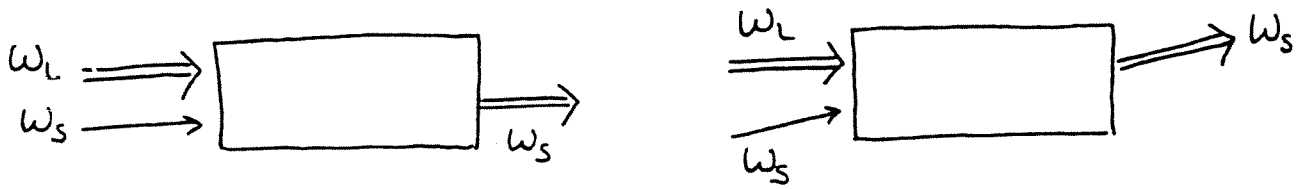
Amplifier

Apply both laser and Stokes frequency



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

Alternative geometries:



Stimulated Scattering Processes

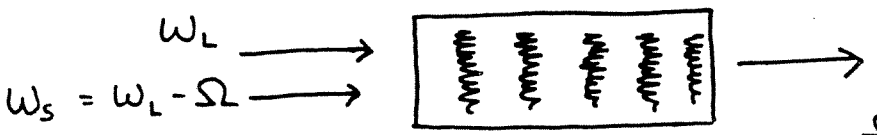
Stimulated Brillouin Scattering

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

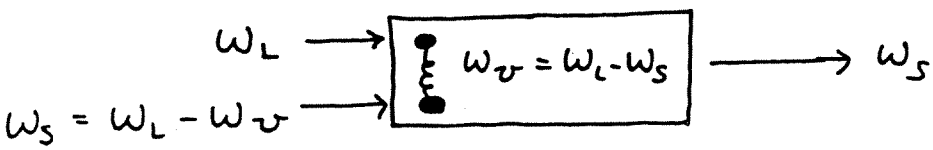
interaction with nonpropagating density (temperature) waves
(coupling can be electrostrictive of thermal)



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

Stimulated Raman Scattering

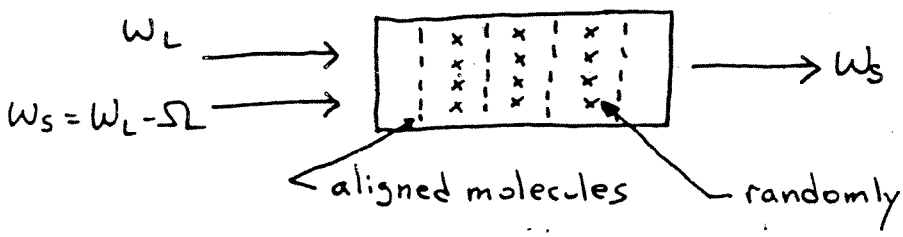
interaction with vibrational degree of freedom



Stimulated Rayleigh-Wing Scattering

interaction with orientational degree of freedom

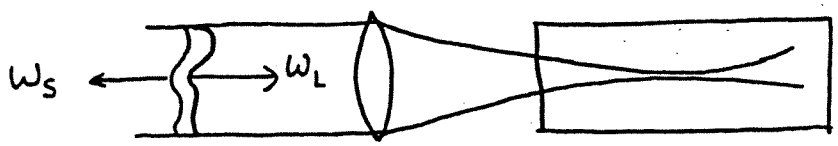
~2 ps



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

Uses of Stimulated Light Scattering

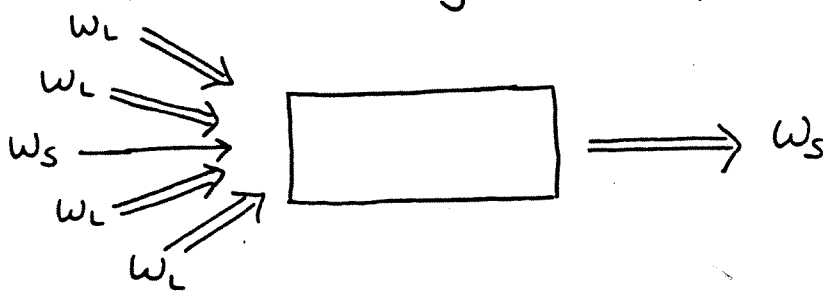
Optical Phase Conjugation (by SBS)



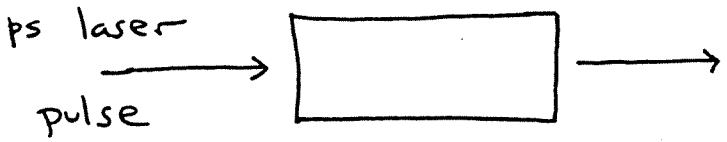
Laser Frequency Shifting (by SRS)

Can shift UV excimer laser radiation to visible

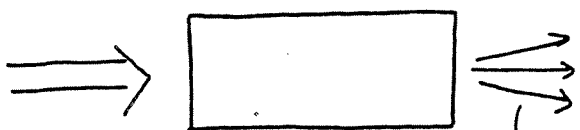
Laser Beam Combining (by SRS)



White-Light Generation (?)

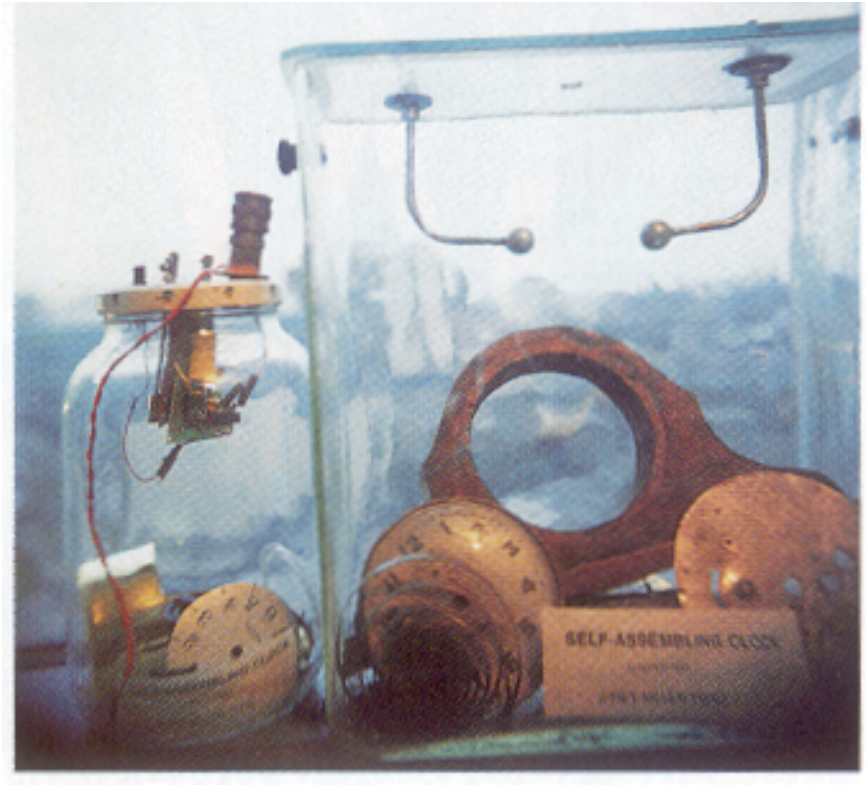


Thermal Blooming (Stimulated Thermal Rayleigh Scattering)



new components experience growth by stimulated scattering

Experiment in Self Assembly



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Thank you for your attention!