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)

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

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)

\[ \omega \rightarrow \chi^{(2)} \rightarrow 2\omega \]

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

\[ \omega \rightarrow \chi^{(3)} \rightarrow 3\omega \]

THG

FWM

NL index

\[ n = n_0 + n_2 I \]

where \( n_2 = \frac{3}{4n_0^2 \varepsilon_0 c} \chi^{(3)} \)
Some Fundamental Nonlinear Optical Processes: I

Second-Harmonic Generation

\[ \omega \rightarrow \chi^{(2)} \rightarrow 2\omega \]

Dolgaleva, Lepeshkin, and Boyd
Sum-Frequency Generation

\[ \omega_3 = \omega_1 + \omega_2 \]
Some Fundamental Nonlinear Optical Processes: III

Difference-Frequency Generation

\[ \chi^{(2)} \]

\[ \omega_1 \rightarrow \omega_2 \rightarrow \omega_3 = \omega_1 - \omega_2 \]

amplified!

 Optical Parametric Oscillation

\[ \omega_1 = \omega_2 + \omega_3 \] (pump)

\[ \omega_2 \text{ (signal)} \]

\[ \omega_3 \text{ (idler)} \]
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)
(b) Quantum technologies (e.g., secure communications)
Third-Harmonic Generation
Some Fundamental Nonlinear Optical Processes: IV

Intensity-Dependent Index of Refraction

• Single beam: self-phase modulation

\[ E(\omega) \rightarrow \chi^{(3)} \rightarrow E(\omega) e^{i\phi_{NL}} \]

• Two beam: cross-phase modulation

\[ E(\omega) \rightarrow E(\omega) e^{i\phi_{NL}} \]
\[ E(\omega') \rightarrow E(\omega') e^{i\phi_{NL}} \]

The refractive index is given by

\[ n = n_0 + n_2 I \quad \text{where} \quad n_2 = \frac{3}{4n_0^2\varepsilon_0 c} \chi^{(3)} \]
$\chi^{(2)}$ vanishes identically for a material possessing a center of inversion symmetry (a centrosymmetric medium).

**non-centrosymmetric medium**

- Actual potential
- Parabola

**centrosymmetric medium**

- Actual potential
- Parabola

---

**Applied Field**

**Linear Response**

**Nonlinear, Centrosymmetric Medium**

**Nonlinear, Noncentrosymmetric Medium**
2. Coupled Wave Equations and Harmonic Generation
Let
\[ \tilde{E}_1(z, t) = E_1(z)e^{-i\omega t} + \text{c.c.} = A_1e^{i(k_1z-\omega t)} + \text{c.c.} \]  
(1)
\[ \tilde{E}_2(z, t) = E_2(z)e^{-i2\omega t} + \text{c.c.} = A_2(z)e^{i(k_2z-2\omega t)} + \text{c.c.} \]  
(2)
where \( k_1 = n_1\omega/c \) and \( k_2 = n_22\omega/c \).

We have assumed that the pump wave \( E_1 \) at frequency \( \omega \) 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
\[ \tilde{P}_2(t) = P_2 e^{-i2\omega t} \]  
where \( P_2 = \epsilon_0\chi^{(2)}E_1^2 = \epsilon_0\chi^{(2)}A_1^2 e^{i2k_1z} \)  
(3)
The generation of the wave at \( 2\omega \) is governed by the wave equation
\[ \nabla^2\tilde{E}_2 - \frac{n^2}{c^2} \frac{\partial^2\tilde{E}_2}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2\tilde{P}_2}{\partial t^2}. \]  
(4)
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 \textit{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 \(\Delta k\) is known as the phase (or wavevector) mismatch factor, and it is crucially important in determining the efficiency of nonlinear optical interactions.
For the case $\Delta k = 0$, Eq. (8) becomes

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

with solution evaluated at $z = L$ of

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

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

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

(11)

Note that $\Delta k L$ must be kept much smaller than $\pi$ 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

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

where $\omega_1 + \omega_2 = \omega_3$

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** ($n_e > n_0$)
  - Type I: $n_3^0\omega_3 = n_1^e\omega_1 + n_2^e\omega_2$
  - Type II: $n_3^0\omega_3 = n_1^o\omega_1 + n_2^o\omega_2$

- **Negative uniaxial** ($n_e < n_0$)
  - Type I: $n_3^e\omega_3 = n_1^o\omega_1 + n_2^o\omega_2$
  - Type II: $n_3^e\omega_3 = n_1^e\omega_1 + n_2^o\omega_2$
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?

\begin{align*}
    \text{(too loose)} & & \text{(just right)} & & \text{(too tight)}
\end{align*}

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

\[
    b = \frac{L}{2.84} \quad \Delta k = \frac{3.2}{L}
\]

because of Gouy phase shift!

and is given by

\[
    \frac{P(2w)}{P(w)} = 1.068 \left[ \frac{128\pi^2 W_1^2 d_{eff}^2 L}{c^4 n_1 n_2} \right] P(w)
\]

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Additional Studies of Wave Propagation Effects
3. Mechanisms of Optical Nonlinearity
## Typical Values of the Nonlinear Refractive Index

<table>
<thead>
<tr>
<th>Mechanism</th>
<th>$n_2 ,^a$ (cm$^2$/W)</th>
<th>$\chi_{1111}^{(3)}$ (m$^2$/V$^2$)</th>
<th>Response time (sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electronic polarization</td>
<td>$10^{-16}$</td>
<td>$10^{-22}$</td>
<td>$10^{-15}$</td>
</tr>
<tr>
<td>Molecular orientation</td>
<td>$10^{-14}$</td>
<td>$10^{-20}$</td>
<td>$10^{-12}$</td>
</tr>
<tr>
<td>Electrostriction</td>
<td>$10^{-14}$</td>
<td>$10^{-20}$</td>
<td>$10^{-9}$</td>
</tr>
<tr>
<td>Saturated atomic absorption</td>
<td>$10^{-10}$</td>
<td>$10^{-16}$</td>
<td>$10^{-8}$</td>
</tr>
<tr>
<td>Thermal effects</td>
<td>$10^{-6}$</td>
<td>$10^{-12}$</td>
<td>$10^{-3}$</td>
</tr>
<tr>
<td>Photorefractive effect$^b$</td>
<td>(large)</td>
<td>(large)</td>
<td>(intensity-dependent)</td>
</tr>
</tbody>
</table>

$^a$ 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 \varepsilon_0 \hbar^2} \sum_{lmn} \rho_{ll}^{(0)} \left\{ \begin{array}{c}
\mu^i_{ln} \mu^j_{nm} \mu^k_{ml} \frac{[\omega_{nl} - \omega_p - \omega_q - i\gamma_{nl}][\omega_{ml} - \omega_p - i\gamma_{ml}]}{[\omega_{nl} - \omega_p - \omega_q - i\gamma_{nl}][\omega_{ml} - \omega_p - i\gamma_{ml}]}

(a_1)
\end{array} \right. \\
+ \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}]}

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

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

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

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

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

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

(b'_2)
\right. 
\]
Nonresonant Electronic Nonlinearities

Estimate size:

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

The nonlinear term becomes comparable to linear term when

\[ E \sim E_{at} = \frac{e}{a^2} \sim \text{Bohr radius} \]

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

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

\[ \alpha_{\parallel} = \text{polarizability parallel to symmetry axis} \]
\[ \alpha_{\perp} = \text{polarizability perpendicular to symmetry axis} \]

The induced dipole moment

\[ \bar{p} = \alpha \cdot \bar{E} \]

is not parallel to the applied electric field.

The molecule hence experiences a torque

\[ \bar{\tau} = \bar{p} \times \bar{E} \]

which tends to align the molecule with field.

\[ \chi^{(1)} = N \left( \frac{1}{3} \alpha_{\parallel} + \frac{2}{3} \alpha_{\perp} \right) \]
\[ \chi^{(3)} = \frac{2N}{45} \frac{(\alpha_{\parallel} - \alpha_{\perp})^2}{kT} \]
How to Increase $\chi^{(3)}$

Use a conjugated polymer

\[ \begin{array}{c}
\downarrow \downarrow \downarrow \downarrow \downarrow \downarrow \downarrow \downarrow \\
\text{electron wavefunction}
\end{array} \begin{array}{c}
\xleftarrow{L} \\
delocalized over length
\text{of polymer}
\end{array} \]

linear polarizability $\alpha \sim L^3$

nonlinear hyperpolarizability $\gamma \sim L^5$

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

for conjugated polymer
TWO GREAT IRONIES OF NONLINEAR OPTICS

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

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

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

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


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

SELF-TRAPPING OF OPTICAL BEAMS

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

\[ n = n_0 + \delta n \]

\[ P_{cr} = \frac{\pi (0.61)^2 \lambda_0^2}{8 n_0 n_2} \]

radial profile of self-trapped beam
Z-Scan Measurement of $\chi^{(3)}$

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

Measures NL change in absorption.
($\text{Im } \chi^{(3)}$)
Some Actual Z-Scan Data

Closed aperture

$\Delta T_{pv} = 0.406 \Phi_{NL}$

where

$\Phi_{NL} = n_2 (\omega/c) I_0 L$

Open aperture

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)

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

1973: Hasegawa & Tappert
1980: Mollenauer, Stolen, Gordon

Spatial solitons (nonlinearity balances diffraction)

\[ 2i k_0 \frac{\partial A}{\partial z} + \frac{\partial^2 A}{\partial x^2} = -3 \chi^{(3)} \frac{\omega^2}{c^2} |A|^2 A \]

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

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

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

- Loss of spatial coherence
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)

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  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

- If the dielectric permittivity $\varepsilon$ is nearly zero, then refractive index $n = \sqrt{\varepsilon}$ is nearly zero.
  Thus $\nu_{\text{phase}} = c / n$ is nearly infinite
  $\lambda = \lambda_{\text{vac}} / n$ is nearly infinite
  Light oscillates in time but not in space; everything is in phase
  Light “oscillates” but does not “propagate.”

- Radiative processes are modified in ENZ materials
  Einstein $A$ coefficient (spontaneous emission lifetime $= 1/A$)
  $A = n A_{\text{vac}}$
  We can control (inhibit!) spontaneous emission!
  Einstein $B$ coefficient
  Stimulated emission rate $= B$ times EM field energy density
  $B = B_{\text{vac}} / n^2$
  Optical gain is very large!
  Einstein, Physikalische Zeitschrift 18, 121 (1917).

- 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!
  \[ n = 0 \quad | \quad n = 1 \]


- Thus light can enter an ENZ material only at normal incidence!
  \[ n = 1 \quad | \quad n = 0 \]
  TIR

Some Consequences of ENZ Behaviour - 1

- Funny lenses

\[ n = 0 \]


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

\[ L \quad L \gg \lambda_{\text{vac}} \]


- No Fabry-Perot interference

\[ n = 0 \]

O. Reshef et al., ACS Photonics 4, 2385, 2017.
Some Consequences of ENZ Behaviour - 2

- Super-coupling (of waveguides)

- Large evanescent tails for waveguide coupling

- Automatic phase matching of NLO processes

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

For example, the following 4WM process is allowed


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
  \[ \epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \]
  Note that \( \text{Re}\ \epsilon = 0 \) for \( \omega = \omega_p/\sqrt{\epsilon_\infty} \equiv \omega_0 \).

- Challenge: Obtain low-loss ENZ materials
  Want \( \text{Im}\ \epsilon \) as small as possible at the frequency where \( \text{Re}\ \epsilon = 0 \).

- 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.

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 $\chi^{(3)}$

$$n_2 = \frac{3\chi^{(3)}}{4\varepsilon_0 c n_0 \text{Re}(n_0)}$$

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 $\mu$m.

Recall the Drude formula

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

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

The region near $\omega_0$ is known as the epsilon-near-zero (ENZ) region.

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

- Note that $n_2$ is positive (self focusing) and $\beta$ is negative (saturable absorption).
- Both $n_2$ and nonlinear absorption increase with angle of incidence.
- $n_2$ shows a maximum value of $0.11 \text{ cm}^2/\text{GW} = 1.1 \times 10^{-10} \text{ cm}^2/\text{W}$ at 1.25 $\mu\text{m}$ and 60 deg. This value is 2000 times larger than that away from ENZ region.
- $n_2$ is $3.4 \times 10^5$ 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

$$n_2 = \frac{3\chi^{(3)}}{4n_0 \text{Re}(n_0) \varepsilon_0 c}.$$  

and

$$I = 2\text{Re}(n_0) \varepsilon_0 c |E|^2.$$  

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

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

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

\[
  n_2 = \frac{3\chi^{(3)}}{4n_0 \text{Re}(n_0) \epsilon_0 c}.
\]

and

\[
  I = 2\text{Re}(n_0) \epsilon_0 c |E|^2.
\]

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

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

Nonlinear Response of ITO is Nonperturbative
6. Local-Field Effects in Nonlinear Optics
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

\[ E_{\text{loc}} = E + \frac{4}{3}\pi P \quad \text{where} \quad P = \chi^{(1)}E \]

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

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

\[ \lambda = \frac{c f \Gamma e \lambda_0 e / 4\pi}{\omega_0 - \omega - i \gamma} \]

\[ \Rightarrow \]

\[ \varepsilon = 1 + \frac{N f \Gamma e \lambda_0 c}{\omega_0 + \Delta \omega_L - \omega - i \gamma} \]

\[ \Delta \omega_L = -\frac{1}{3} N f \Gamma e \lambda_0 c \]

\[ \text{Lorentz red shift} \]

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

Observation of the Lorentz Red Shift

\[ N \sim 10^{17} \text{cm}^{-3} \]
\[ \Gamma_{\text{Dop}} \sim 1 \text{GHz} \]
\[ |\varepsilon - 1| \sim 1 \quad \text{and} \quad |\mathcal{L}| \sim 4/3 \]

\[ {}^2S_{1/2} \leftrightarrow {}^2P_{1/2} \]
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 $\ll$ optical wavelength
- Thus all optical properties, such as $n$ and $\chi^{(3)}$, can be described by effective (volume averaged) values

Recent review: Dolgaleva and Boyd, Advances in Optics and Photonics 4, 1–77 (2012).
Local Field Enhancement of the NLO Response

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

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

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

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

- For a homogeneous material

\[ L = \frac{\varepsilon + 2}{3} \]

- For a spherical particle of dielectric constant \( \varepsilon_m \) embedded in a host of dielectric constant \( \varepsilon_h \)

\[ L = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} \]

- 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

\[ L = \frac{\varepsilon_{\text{eff}}}{\varepsilon_a} \quad \frac{1}{\varepsilon_{\text{eff}}} = \frac{f_a}{\varepsilon_a} + \frac{f_b}{\varepsilon_b} \]
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.

\[ \nabla \cdot \mathbf{D} = 0 \text{ implies that} \hspace{0.5cm} (\varepsilon \mathbf{E})_\perp \text{ is continuous.} \]

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

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

3.2 times enhancement!


Nelson and Boyd, APL 74 2417 (1999)
**Metal / Dielectric Composites**

Very large local field effects

\[
\varepsilon_h \quad \varepsilon_m \quad \uparrow E_0
\]

\[
E_{in} = \frac{3 \varepsilon_h}{\varepsilon_m + 2 \varepsilon_h} \quad E_0
\]

\[
= \varepsilon \cdot E_0
\]

(\(\varepsilon_m\) is negative!)

At resonance

\[
\varepsilon = \frac{3 \varepsilon_h}{\varepsilon_m + 2 \varepsilon_h} \quad \rightarrow \quad \frac{3 \varepsilon_h}{i \varepsilon_m''} \quad \approx (3 \text{ to } 30) i
\]
Gold-Doped Glass: A Maxwell-Garnett Composite

Red Glass Caraffe
Nurenberg, ca. 1700
Huelsmann Museum, Bielefeld

Developmental Glass, Corning Inc.
gold volume fraction approximately $10^{-6}$
gold particles approximately 10 nm diameter

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


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:

\[
v_g = \frac{c}{n_g} \quad n_g = n + \omega \left(\frac{dn}{d\omega}\right)
\]

- Velocity controlled by structural or material resonances

Group Velocity

Pulse (wave packet) \[ \rightarrow u_g \]

Group velocity given by \[ u_g = \frac{d\omega}{dk} \]

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

Thus \[ u_g = \frac{c}{n + \omega \frac{dn}{d\omega}} \equiv \frac{c}{n_g} \]

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

\[ E_F(0,t) \rightarrow E_F(L,t) \]
\[ E_B(0,t) \leftarrow E_B(L,t) \]

Theory (Winful)

\[ KL = 4 \]

Dispersion relation

\[ \omega \]
\[ k \]

\[ \omega_B \]
\[ k_B \]

Slow light

- Enhanced NLO response

Bhat and Sipe showed that the nonlinear coefficient is given by

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

Where the slow-down factor \( S = n_g/n_B \)
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


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

\[ n_g = n + \omega \left( \frac{dn}{d\omega} \right) \]
Light speed reduction to 17 metres per second in an ultracold atomic gas

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² Department of Physics, § Division of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA
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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
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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)

\[ \omega_S = \omega_L - \Omega \]

\[ \frac{dI_S}{dz} = -gI_LI_S \]

\[ g = \frac{\gamma_e^2 \omega^2}{nv c^3 Q_0 \Gamma_B} . \]

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 \( \Gamma_B \) is the Brillouin linewidth

Slow Light via Coherent Population Oscillations

\[ E_3, \omega + \delta \]
\[ \rightarrow \]
\[ \text{saturable medium} \]
\[ E_1, \omega \]

\[ \frac{\omega + \delta}{\text{measure absorption}} \]

\[ n_g = n + \omega \frac{dn}{d\omega} \quad T_2 << T_1 \]

- Want a narrow feature in absorption profile to give a large \( \frac{dn}{d\omega} \)
- Ground state population oscillates at beat frequency \( \delta \) (for \( \delta < \frac{1}{T_1} \)).
- 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 $T_{\text{pulse}} \ll T_1 = \text{time scale for saturation changes}$
  - Then absorption decreases with time during pulse due to saturation
Slow Light in Ruby

![Graph showing delay vs. modulation frequency](image_url)

- Delay (ms) vs. Modulation Frequency (Hz)
- $n_g = 5 \times 10^6$

**Equipment:**
- Argon Ion Laser
- Function Generator
- EO modulator
- Diffuser
- Reference Detector
- Digital Oscilloscope

**References:**
- Bigelow et al, PRL 2003
Alexandrite Displays both Saturable and Reverse-Saturable Absorption

- Both slow and fast propagation observed in alexandrite

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

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Observation of Superluminal and “Backwards” Pulse Propagation

- A strongly counterintuitive phenomenon
- But entirely consistent with established physics
- Observed by Gehring, Schweinsberg, Barsi, Kostinski, and Boyd Science 312, 985 2006.

 normalized length $|n_g|Z (m)$

\[
\frac{\partial A}{\partial z} - \frac{1}{n_g} \frac{\partial A}{\partial t} = 0
\]
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

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 nonanalyticity 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

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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?

"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 resolution as large laboratory spectrometers.

- Here is why it works:

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

- 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.

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!)

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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

\[ S = \frac{1}{2} n \epsilon_0 c E^2 \]  \hspace{1cm} (intensity)

\[ u = \frac{1}{2} n n_g \epsilon_0 E^2 \]  \hspace{1cm} (energy density)

where

\[ v_g = c/n_g \]  \hspace{1cm} (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)
\[
P = E \times H / c^2 \quad \text{(EM momentum density)}
\]
\[
p = (\hbar \omega / c)(1/n_g) \quad \text{(photon momentum)}
\]

Minkowski form (for light in matter)
\[
P = D \times B \quad \text{(EM momentum density)}
\]
\[
p = (\hbar \omega / c)(n^2/n_g) \quad \text{or} \quad p = (\hbar \omega / c) n \quad \text{photon momentum}
\]

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

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
  \[ \Delta z = (n_g - 1)L \frac{\hbar \omega}{M c^2} \]
- Simple kinematic argument shows that momentum of photon in block is
  \[ p = \frac{\hbar \omega}{n_g c} \quad \text{Abraham form!} \]
Fermi’s Argument Supports the Minkowski Form

- Photon in medium of refractive index $n(\omega)$
- Atom with mass $m$ and resonance frequency $\omega_0$

- Fermi describes Doppler effect in terms of atomic recoil (RMP, 1932)
- Atom can absorb only if $\omega \approx \omega_0 (1 - n\nu/c)$
- Conservation of energy and momentum
  
  Initial energy $= \hbar\omega + \frac{1}{2}mv^2$
  Final energy $= \hbar\omega_0 + \frac{1}{2}mv'^2$
  Initial momentum $= p + mv$
  Final momentum $= mv'$

- Solve: find photon momentum $p$ in medium given by
  
  $p = n \hbar\omega/c$ 
  Minkowski form!
Which is Correct, Abraham or Minkowski?

Resolution of the Abraham-Minkowski Dilemma

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(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

\[ p^\text{med}_{\text{kin}} + p^\text{Abr} = p^\text{med}_{\text{can}} + p^\text{Min}. \]

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)
\[
P = E \times H / c^2 \quad (\text{EM momentum density})
\]
\[
p = (\hbar \omega / c)(1/n_g) \quad (\text{photon momentum})
\]
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)
\[
P = D \times B \quad (\text{EM momentum density})
\]
\[
p = (\hbar \omega / c)(n^2/n_g) \quad \text{or} \quad p = (\hbar \omega / c) n \quad (\text{photon momentum})
\]
It is the canonical momentum (as in \( h/\lambda_{\text{deBroglie}} \))
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

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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.

\[ V = 700 \, \text{cm/sec}; \ L = 150 \, \text{cm}; \ \text{displacement of 0.5 fringe}. \]

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

Fresnel “drag” coefficient

• Modern theory: relativistic addition of velocities

- But what about slow-light media?
Fresnel Drag in a Highly Dispersive Medium

Light Drag in a Slow Light Medium (Lorentz)

\[ u \approx \frac{c}{n} \pm v \left( 1 - \frac{1}{n^2} + \frac{n_g - n}{n^2} \right) \]

We Use Rubidium as Our Slow Light Medium
- Transmission spectrum of Rb around D\(_2\) transition:

\[ T = 30 \, ^\circ\text{C} \]
\[ T = 150 \, ^\circ\text{C} \]

- Group index of Rb around D\(_2\) line at T=130

\[ \Delta u \ (\text{m/s}) \]

\[ \text{Temperature } ^\circ\text{C} \]

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

Safari, De Leon, Mirhosseini, Magana-Loaiza, and Boyd
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 (ng as high as 10^7).
Transverse Photon Drag

\[ \Delta x = \left( \frac{vL}{c} \right) \left( n_g - \frac{1}{n_\phi} \right) \]

For \( L = 25 \text{ mm}, v = 2000 \text{ cm/s}, \) displacement = 6 nm.

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

rotating, 10-cm-long ruby rod

Effect clearly visible by eye!

Franke-Arnold, Gibson, Boyd and Padget, Science, 2011
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Applications of Slow and Fast Light

- Buffers and regenerators for telecom
- Slow/fast light for interferometry
- 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:

![Diagram showing light scattering](image)

complete destructive interference!

Spontaneous light scattering
Fluctuations (e.g., in $E$) 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:

Incident beam
Intensity $I_0$
Frequency $\nu_0$

scattering medium

Scattered light

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

$\nu_0$

<table>
<thead>
<tr>
<th>Process</th>
<th>Shift</th>
<th>Linewidth</th>
<th>Relaxation time</th>
<th>Gain (cm/mW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raman</td>
<td>$100\text{cm}^{-1}$</td>
<td>$5\text{cm}^{-1}$</td>
<td>$10^{-12.5}$</td>
<td>$5 \times 10^{-3}$</td>
</tr>
<tr>
<td>Brillouin</td>
<td>$0.1-1$</td>
<td>$5 \times 10^{-3}$</td>
<td>$10^{-9.5}$</td>
<td>$10^{-2}$</td>
</tr>
<tr>
<td>Rayleigh</td>
<td>$0$</td>
<td>$5 \times 10^{-4}$</td>
<td>$10^{-8.5}$</td>
<td>$10^{-4}$</td>
</tr>
<tr>
<td>Rayleigh-Wing</td>
<td>$0$</td>
<td>$10$</td>
<td>$10^{-12.5}$</td>
<td>$10^{-3}$</td>
</tr>
</tbody>
</table>
Relation Between Spontaneous and Stimulated Light Scattering

\[ \frac{dm_s}{dz} = D m_L (m_s + 1) \]

# of laser photons per mode  # 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:

\[ G = \frac{4\pi^3 N e^2}{h \omega_L \omega_S^2 n_S^2} \left( \frac{\varepsilon^2 \sigma}{\omega_S \omega_L \Omega} \right) I_L \]

Stimulated Brillouin Scattering

- Spontaneous Brillouin scattering is a very weak process.

\[ \text{\[\sim 1 \text{ part in } 10^5 \text{ of incident scattered per cm of liquid.}\]} \]

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

\[ P \geq 20 \text{ kW} \]
\[ T \geq 5 \text{ nsec} \]

organic liquid \((\text{CS}_2)\)
Theory of SBS (backward SBS)

\[
\begin{align*}
\text{laser-Stokes} & \quad \frac{E_1, \omega_1, \vec{k}_1}{E_2, \omega_2, \vec{k}_2} \quad \rightarrow \quad \text{Sound wave} \\
\text{sound} & \quad \rightarrow \quad \text{laser-Stokes} \\
\rightarrow & \quad \text{Positive feedback:} \\
& \quad \text{laser + Stokes} \quad \rightarrow \quad \text{sound} \\
& \quad \text{laser + sound} \quad \rightarrow \quad \text{Stokes}
\end{align*}
\]

Frequencies:

\[
\begin{align*}
\omega_2 &= \omega_1 - \Omega_B \quad \text{where} \quad \Omega_B = |\mathbf{\Omega}_B| \quad \Omega \quad \text{velocity of sound} \\
\mathbf{\Omega}_B &= |\mathbf{\Omega}_B| \quad \text{velocity of sound} \\
\overrightarrow{\Omega}_B &= \overrightarrow{k}_1 - \overrightarrow{k}_2 \approx 2 \overrightarrow{k}_1 \\
\Rightarrow \quad \Omega_B &= 2 \omega \frac{v}{c/n} \quad (\Omega_B/2\pi \approx 5 \text{ GHz})
\end{align*}
\]

Coupling among waves: electrostriction

Material tends to be drawn into regions of high field.

\[
P_{st} = -Y_e \frac{E^2}{8\pi} \\
Y_e = \rho \frac{\partial E}{\partial \rho} \approx 1 \quad \text{for condensed matter}
\]

\[
\frac{\Delta \rho}{\rho} = -C \rho_p \\
\Delta \varepsilon = \left( \rho \frac{\partial E}{\partial \rho} \right) \frac{1}{\rho} \Delta \rho
\]
Theory of SBS

\[ \tilde{E}_1(z,t) = A_1(z,t) e^{i(k_1 z - \omega_1 t)} + cc \]
\[ \tilde{E}_2(z,t) = A_2(z,t) e^{i(k_2 z - \omega_2 t)} + cc \]
\[ \tilde{\rho}(z,t) = \rho_0 + [\rho(z,t) e^{i(\delta z - \Omega t)} + cc] \]

\[ \delta = 2k_1 \]
\[ \Omega = \omega_1 - \omega_2 \]

Density \( \tilde{\rho} \) obeys acoustic wave equation

\[ \frac{\partial^2 \tilde{\rho}}{\partial t^2} - \Gamma \nabla^2 \frac{\partial \tilde{\rho}}{\partial t} - U^2 \nabla^2 \tilde{\rho} = \nabla \cdot \tilde{f} \]

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

\[ \tilde{f} = \nabla \tilde{P}_{st}, \quad \tilde{P}_{st} = -\gamma e \frac{\langle \tilde{E}_1^2 \rangle}{8\pi} \]

Thus

\[ \nabla \cdot \tilde{f} = \frac{\gamma e \delta^2}{4\pi} \left[ A_1 A_2^* e^{i(\delta z - \Omega t)} + cc \right] \]

In the SVAA, acoustic wave equation becomes

\[-2i \Omega \frac{\partial \rho}{\partial t} + (\Omega_B^2 - \Omega^2 - i \Omega \Gamma_B) \rho - 2i q U^2 \frac{\partial \rho}{\partial z} = \frac{\gamma e \delta^2}{4\pi} A_1 A_2^* \]

where

\[ \Gamma_B = \delta^2 \Gamma' \quad \Omega_B = 2\omega \frac{\nu}{c/n} \]
Theory of SBS (continued)

Simplifications: (1) \( \partial P / \partial t = 0 \) in steady state
(2) \( \partial P / \partial z = 0 \) (phonons strongly damped)
\((\Delta s \leq 10 \mu m)\)

Then

\[
\rho(z, t) = \frac{\gamma \epsilon z}{4\pi} \frac{A_1 A_2^*}{\Omega_{yb}^2 - \Omega^2 - i \Omega \Delta_B}
\]

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

\[
P^{nl} = \Delta X \tilde{E} = \frac{\Delta E}{4\pi} \tilde{E} = \frac{1}{4\pi \rho_0} \left( \rho \frac{\partial E}{\partial \rho} \right) (\tilde{P} - \rho_0) \tilde{E}
\]

\[
= \frac{1}{4\pi \rho_0} \gamma \epsilon (\tilde{P} - \rho_0) \tilde{E}
\]

Thus

\[
\frac{\partial A_1}{\partial z} + \frac{1}{c/n} \frac{\partial A_1}{\partial t} = \frac{i\omega \gamma \epsilon}{2\pi n_c \rho_0} \rho A_2
\]

\[-\frac{\partial A_2}{\partial z} + \frac{1}{c/n} \frac{\partial A_2}{\partial t} = \frac{i\omega \gamma \epsilon}{2\pi n_c \rho_0} \rho^* A_1
\]

At the Brillouin resonance \((\Omega = \Omega_{yb})\) and in steady state

\[
\frac{dA_1}{dz} = -\frac{\omega \gamma \epsilon \gamma^2}{8\pi n_c \rho_0 \Delta B} (A_2 A_2^*) A_1
\]

\[
\frac{dA_2}{dz} = -\frac{\omega \gamma \epsilon \gamma^2}{8\pi n_c \rho_0 \Delta B} (A_1 A_1^*) A_2
\]
Theory of SBS (continued)

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

\[
\begin{align*}
\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\nu c^3 \rho_0 \Gamma} \quad \text{SBS gain factor} \\
&\sim 0.01 \text{ cm/MW} \\
&\text{(recall that } \Gamma \sim \omega^2) \\

\text{Solution in constant-pump limit} \quad (I_1 \text{ constant}) \\
I_2(z) = I_2(L) e^{gI_1(L-z)} \\

\text{Total amplification} = \frac{I_2(0)}{I_2(L)} = e^{gI_1L}
\]
Phase Conjugation by SBS

SBS can produce high quality phase conjugation.

\[ \frac{dI_S}{dz} = -g I_L I_S \]

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**

\[ W_L \rightarrow \begin{array}{c}
\text{Raman medium} \\
\end{array} \rightarrow W_s = W_L - W_U \]

We can predict the Stokes output intensity:

\[ I_L \rightarrow \begin{array}{c}
\text{Raman medium} \\
\end{array} \rightarrow I_s(L) \]

\[ I_s(L) = I_s(0) e^{gL} \]

\[ g = \text{Raman gain factor} \approx 10^{-9} \text{ cm/W for most materials} \]
Example of Raman Frequency Shifter

Need 1.9 μm light

\[ 1.06 \mu m \xrightarrow{4155 \text{ cm}^{-1}} \text{H}_2 \xrightarrow{1.9 \mu m} \]

Experimental setup

\[ \lambda = 1.06 \mu m \]
\[ 30 \text{ps pulse length} \]
\[ 3 \text{ mJ pulse energy} \]

\[ \lambda = 1.9 \mu m \]
\[ 150 \mu J \text{ energy} \]
\[ 50\% \text{ conversion} \]
<table>
<thead>
<tr>
<th>Typical Raman media</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>material</td>
<td>$\Delta \nu$ (cm$^{-1}$)</td>
<td>gain g x $10^3$ (cm/MW)</td>
</tr>
<tr>
<td>LIQUIDS</td>
<td></td>
<td></td>
</tr>
<tr>
<td>benzene</td>
<td>992</td>
<td>3</td>
</tr>
<tr>
<td>water</td>
<td>3290</td>
<td>0.14</td>
</tr>
<tr>
<td>$N_2$</td>
<td>2326</td>
<td>17</td>
</tr>
<tr>
<td>$O_2$</td>
<td>1555</td>
<td>16</td>
</tr>
<tr>
<td>GASES</td>
<td></td>
<td></td>
</tr>
<tr>
<td>methane</td>
<td>2916</td>
<td>0.66 (10 atm, 500 nm)</td>
</tr>
<tr>
<td>hydrogen</td>
<td>4155 (vibrational)</td>
<td>1.5 (above 10 atm)</td>
</tr>
<tr>
<td></td>
<td>450 (rotational)</td>
<td>0.5 (above 0.5 atm)</td>
</tr>
<tr>
<td>deuterium</td>
<td>2991 (vibrational)</td>
<td>1.1 (above 10 atm)</td>
</tr>
<tr>
<td>$N_2$</td>
<td>2326</td>
<td>0.071 (10 atm, 500 nm)</td>
</tr>
<tr>
<td>$O_2$</td>
<td>1555</td>
<td>0.016 (10 atm, 500 nm)</td>
</tr>
</tbody>
</table>

Configurations for Stimulated Light Scattering

**Generator**
Apply only laser frequency

\[ W_L \rightarrow W_S \]

Stokes signal grows from noise.

or

\[ W_L \rightarrow W_S \]

(Initiation due to thermal or quantum fluctuations)

(SBS occurs only in backward direction)
(SRS occurs in both forward and backward directions)
Stokes radiation is always emitted at the frequency of maximum gain.

**Amplifier**
Apply both laser and Stokes frequency

\[ W_L \leftrightarrow W_S \]

Stokes "seed"

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)

\[
\begin{align*}
\omega_L, \vec{k}_L & \quad \rightarrow \quad \Omega, \hat{\vec{q}} \\
\omega_s, \vec{k}_s & \quad \leftarrow \\
\omega_s = \omega_L - \Omega & \\
\Omega = \frac{\hat{q} \cdot v}{\hat{q}} & = \frac{\vec{k}_L - \vec{k}_s}{\hat{q}}
\end{align*}
\]

**Stimulated Rayleigh Scattering**
interaction with nonpropagating density (temperature) waves (coupling can be electrostrictive or thermal)

\[
\begin{align*}
\omega_L & \quad \rightarrow \\
\omega_s = \omega_L - \Omega & \quad \rightarrow
\end{align*}
\]

**Stimulated Raman Scattering**
interaction with vibrational degree of freedom

\[
\begin{align*}
\omega_L & \quad \rightarrow \\
\omega_s = \omega_L - \omega_v & \quad \rightarrow \\
\omega_s = \omega_L - \omega_v
\end{align*}
\]

**Stimulated Rayleigh-Wing Scattering**
interaction with orientational degree of freedom

\[
\begin{align*}
\omega_L & \quad \rightarrow \\
\omega_s = \omega_L - \Omega & \quad \rightarrow \\
\Omega & \sim 1/\tau_{\text{orientation}}
\end{align*}
\]
Uses of Stimulated Light Scattering

Optical Phase Conjugation (by SBS)

\[ W_s \leftrightarrow W_l \]

Laser Frequency Shifting (by SRS)

\[ W_l \rightarrow W_l - W_r \]

Can shift UV excimer laser radiation to visible

Laser Beam Combining (by SRS)

\[ W_l \rightarrow W_s \]

White-Light Generation (?)

\[ \text{ps laser pulse} \rightarrow \text{White light} \]

(broad frequency spectrum)

Thermal Blooming (Stimulated Thermal Ryleigh Scattering)

(new components experience growth by stimulated scattering)
Experiment in Self Assembly

Joe Davis, MIT
Thank you for your attention!