Introduction to Nonlinear Optics

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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.
1. What is Nonlinear Optics?
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):

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

\[ n = n_0 + n_2 I \quad \text{where} \quad n_2 = \frac{3}{4n_0^2\varepsilon_0c} \chi^{(3)} \]
Some Fundamental Nonlinear Optical Processes: I

Second-Harmonic Generation

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

Dolgaleva, Lepeshkin, and Boyd
Some Fundamental Nonlinear Optical Processes: II

Sum-Frequency Generation

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

\[ \omega_1 \rightarrow \omega_2 \rightarrow \chi^{(2)} \rightarrow \omega_3 = \omega_1 + \omega_2 \]

Diagram:

- \( \omega_1 \) to \( \omega_2 \) to \( \chi^{(2)} \) to \( \omega_3 = \omega_1 + \omega_2 \)
- \( \omega_2 \) and \( \omega_1 \) are inputs, \( \omega_3 \) is the output
- \( \omega_3 \) represents the sum frequency generated

Diagram:

- \( \omega_1 \) and \( \omega_2 \) are inputs, \( \omega_3 \) is the output
- \( \omega_3 \) represents the sum frequency generated
Some Fundamental Nonlinear Optical Processes: III

Difference-Frequency Generation

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

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

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)
(a) 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

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

- Two beam:
  cross-phase modulation

\[ E(\omega) \quad (\text{strong wave}) \]
\[ \xrightarrow{\chi^{(3)}} E(\omega') \quad (\text{probe wave}) \]

The refractive index is given by

\[ n = n_0 + n_2 I \]

where

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

**non-centrosymmetric medium**

**centrosymmetric medium**
2. Coupled Wave Equations and Harmonic Generation
Let
\[ \tilde{E}_1(z, t) = E_1(z)e^{-i\omega t} + \text{c.c.} = A_1 e^{i(k_1 z - \omega t)} + \text{c.c.} \tag{1} \]
\[ \tilde{E}_2(z, t) = E_2(z)e^{-i2\omega t} + \text{c.c.} = A_2(z)e^{i(k_2 z - 2\omega t)} + \text{c.c.} \tag{2} \]
where \( k_1 = n_1 \omega / c \) and \( k_2 = n_2 2\omega / c \).

We have assumed that the pump wave \( E_1 \) at frequency \( \omega \) 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 \( P_2 = \epsilon_0 \chi^{(2)} E_1^2 = \epsilon_0 \chi^{(2)} A_1^2 e^{i2k_1 z} \tag{3} \)

The generation of the wave at \( 2\omega \) is governed by the wave equation
\[ \nabla^2 \tilde{E}_2 - \frac{n^2}{c^2} \frac{\partial^2 \tilde{E}_2}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \tilde{P}_2}{\partial t^2}. \tag{4} \]
Treatment of Second-Harmonic Generation – II

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

\[ \nabla^2 \tilde{E}_2 = \left[ \frac{\partial^2 \tilde{A}_2}{\partial z^2} + 2ik_2 \frac{\partial \tilde{A}_2}{\partial z} - k_2^2 A_2 \right] e^{i(k_2z - 2\omega t)} \]  

(5)

\[ \approx \left[ 2ik_2 \frac{\partial \tilde{A}_2}{\partial z} - k_2^2 A_2 \right] e^{i(k_2z - 2\omega t)} \]  

(6)

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

Note also that

\[ \frac{\partial^2 \tilde{A}_2}{\partial t^2} = -(2\omega)^2 A_2 \ e^{i(k_2z - 2\omega t)} \quad \frac{\partial^2 \tilde{P}_2}{\partial t^2} = -(2\omega)^2 P_2 \ e^{i(2k_1z - 2\omega t)} \]  

(7)

By combining the above equations, we obtain

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

(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

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

with solution evaluated at $z = L$ of

$$A_2(L) = \frac{2i\omega}{n_2c} \chi^{(2)} A_1^2 L \quad \text{or} \quad |A_2(L)|^2 = \frac{4\omega^2}{n_2^2c^2} [\chi^{(2)}]^2 |A_1|^4 L^2. \quad (10)$$

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

$$|A_2(L)|^2 = \frac{4\omega^2}{n_2^2 c^2} \left[\chi^{(2)}\right]^2 |A_1|^4 L^2 \text{sinc}^2(\Delta k L / 2)$$  \hspace{1cm} (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

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

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:

<table>
<thead>
<tr>
<th></th>
<th>Positive uniaxial</th>
<th>Negative uniaxial</th>
</tr>
</thead>
<tbody>
<tr>
<td>(positive uniaxial)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(negative uniaxial)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Type I</td>
<td>$n^0_3\omega_3 = n^e_1\omega_1 + n^e_2\omega_2$</td>
<td>$n^e_3\omega_3 = n^0_1\omega_1 + n^0_2\omega_2$</td>
</tr>
<tr>
<td>Type II</td>
<td>$n^0_3\omega_3 = n^o_1\omega_1 + n^e_2\omega_2$</td>
<td>$n^e_3\omega_3 = n^e_1\omega_1 + n^o_2\omega_2$</td>
</tr>
</tbody>
</table>
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?

![Diagram showing too loose, just right, and too tight focusing](image)

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

$$\frac{P(2w)}{P(w)} = 1.068 \left( \frac{128 \pi^2 W_0^2 \text{def}_{eff} L}{c^4 n_1 n_2} \right) P(w)$$
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>$n_2^a$</th>
<th>$\chi_{1111}^{(3)}$</th>
<th>Response time</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(cm²/W)</td>
<td>(m²/V²)</td>
<td>(sec)</td>
</tr>
<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 Susceptibility

\[
\chi_{ijk}^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p) = \frac{N}{2 \varepsilon_0 \hbar^2} \sum_{lmn} \rho_{ll}^{(0)} \left\{ \frac{\mu_{ln}^i \mu_{nm}^j \mu_{ml}^k}{[(\omega_{nl} - \omega_p - \omega_q) - i\gamma_{nl}][(\omega_{ml} - \omega_p) - i\gamma_{ml}]} \right. \\
+ \frac{\mu_{ln}^i \mu_{nm}^j \mu_{ml}^k}{[(\omega_{nl} - \omega_p - \omega_q) - i\gamma_{nl}][(\omega_{ml} - \omega_p) - i\gamma_{ml}]} \\
+ \frac{\mu_{ln}^i \mu_{nm}^j \mu_{ml}^k}{[(\omega_{ml} - \omega_p + \omega_q) + i\gamma_{mn}][(\omega_{nl} + \omega_q) + i\gamma_{nl}]} \\
+ \frac{\mu_{ln}^i \mu_{nm}^j \mu_{ml}^k}{[(\omega_{ml} - \omega_p + \omega_q) + i\gamma_{mn}][(\omega_{ml} - \omega_p - i\gamma_{ml}]} \\
+ \frac{\mu_{ln}^i \mu_{nm}^j \mu_{ml}^k}{[(\omega_{ml} + \omega_p + \omega_q) + i\gamma_{nl}][(\omega_{nl} + \omega_q) + i\gamma_{nl}]} \left\} 
\]
Nonresonant Electronic Nonlinearities

Estimate size:

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

nonlinear term becomes comparable to linear term when

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

\[ \Rightarrow \chi^{(3)} \approx \frac{\chi^{(1)}}{E_a^2} \approx \frac{1}{E_a^2} = 10^{-16} \text{ esu} \]

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

\[ \text{nonlinear spring} \]

[Diagram of potential energy with parabolic and non-parabolic regions labeled.]
Molecular Orientation Effect

Dominant nonlinear effect in most organic liquids.

Due to optical-field induced alignment of anisotropic molecules.

Picosecond response time

\[ E \]

\[ \theta \]

\[ \tilde{P} \]

\[ (\alpha_{11} > \alpha_{1}) \]

\[ \alpha_{11} = \text{polarizability parallel to symmetry axis} \]
\[ \alpha_{1} = \text{polarizability perpendicular to symmetry axis} \]

The induced dipole moment

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

is not parallel to the applied electric field.

The molecule hence experiences a torque

\[ \tilde{T} = \tilde{p} \times \tilde{E} \]

which tends to align the molecule with field.

\[ \chi^{(1)} = N\left(\frac{1}{3}\alpha_{11} + \frac{2}{3}\alpha_{1}\right) \]
\[ \chi^{(3)} = \frac{2N}{45} \frac{(\alpha_{11} - \alpha_{1})^2}{kT} \]
TWO GREAT IRONIES OF NONLINEAR OPTICS

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

2. Silver and gold have very large $X^{(3)}$, but are nearly opaque.
6. Self-Action Effects in Nonlinear Optics
Self-Action Effects in Nonlinear Optics

Self-action effects: light beam modifies its own propagation

- self focusing

- self trapping

- small-scale filamentation
**Self-Focusing**

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

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

\[
OPL = \left[ \sqrt{z_f^2 + \rho_0^2} \right] \left[ n_0 + \frac{1}{2} n_2 I_0 \right]
\]

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

Equate two optical path lengths:

\[
z_f = \rho_0 \sqrt{\frac{n_0}{n_2 I_0}}
\]

This result can be expressed equivalently as

\[
z_f = \rho_0^2 \sqrt{\frac{\pi n_0}{n_2 P}} = \frac{2\sqrt{2} n_0 \rho_0^2}{0.61 \lambda} \frac{1}{\sqrt{P/P_{cr}}}
\]
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

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

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)

\[ n = n_0 + \delta n \]

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

radial profile of self-trapped beam
Properties and Applications of Spatial Solitons

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

\[ n = n_0 + n_2 I \]
\[ n_2 > 0 \]

- Diffraction
- Self-focusing
- Spatial soliton
Optical Solitons

Field distributions that propagate without change of form

Temporal solitons (nonlinearity balances gvd)

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

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

Spatial solitons (nonlinearity balances diffraction)

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

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
Beam Breakup by Small-Scale Filamentation

Predicted by Bespalov and Talanov (1966)

Exponential growth of wavefront imperfections by four-wave mixing processes

Sidemode amplitude grows as

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

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

$$\gamma = n_2 (\omega / c) I_0, \quad q = |k_\perp|$$

$$q_{\text{max}} = \sqrt{2k\gamma}, \quad \theta_{\text{max}} = \frac{q_{\text{max}}}{k}$$
Honeycomb Pattern Formation

Output from cell with a single gaussian input beam

At medium input power

At high input power

Quantum statistics?

Input power 100 to 150 mW
Input beam diameter 0.22 mm

Sodium vapor cell  T = 220° C
Wavelength = 588 nm
Bennink et al., PRL 88, 113901 2002.
Optical Radiance Limiter Based on Spatial Coherence Control

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

Alternative to standard approaches to optical power limiting

4. 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

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

\[ \alpha = \frac{c f \Gamma_0 \lambda_0 e / 4\pi}{\omega_0 - \omega - i \gamma} \]

\[ \Rightarrow \]

\[ \varepsilon = 1 + \frac{N f \Gamma_0 \lambda_0 c}{\omega_0 + \Delta \omega_L - \omega - i \gamma} \]

\[ \Delta \omega_L = -\frac{1}{3} N f \Gamma_0 \lambda_0 c \]

Lorentz red shift

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

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

$I_{Dop} \sim 1 \text{ GHz}$

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

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

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

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

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

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

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

- Maxwell Garnett
  - Fractal Structure
  - Layered

- Bruggeman (interdispersed)

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

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

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

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

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

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

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

- For a homogeneous material

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

- For a spherical particle of dielectric constant \( \varepsilon_m \) 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

\[ L = \frac{\varepsilon_{\text{eff}}}{\varepsilon_a} \]

\[ \frac{1}{\varepsilon_{\text{eff}}} = \frac{f_a}{\varepsilon_a} + \frac{f_b}{\varepsilon_b} \]
Enhanced NLO Response from Layered Composite Materials

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

Alternating layers of TiO$_2$ and the conjugated polymer PBZT.

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

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)
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)


Metal / Dielectric Composites

Very large local field effects

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

\[ = \approx 3E_0 \]  \( (\varepsilon_m \text{ is negative!}) \)

At resonance

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

Red Glass Caraffe
Nuremberg, 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
Counter-intuitive Consequence of Local Field Effects

Gold nanoparticles in a liquid dye solution (HITCl)

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

Effective NL susceptibility of composite

\[
\chi_{\text{eff}}^{(3)} = f \, 2^2 |f|^2 \chi^{(3)}_{Au} + (1-f) \chi^{(3)}_{\text{dye soln}}
\]

\[
f = \frac{3E_h}{E_m + 2E_h} = \text{pure imaginary at resonance!}
\]

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

---

Smith et al., JOSA B 14 1625 1997.
Counterintuitive Consequence of Local Field Effects

Cancellation of two contributions that have the same sign
Gold nanoparticles in a reverse saturable absorber dye solution (13 μM HITCI)

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

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

- A nonlinear response would be useful for photonics applications
- Metals are highly nonlinear (n₂ is 10⁶ times that of silica)
- High (sub-wavelength) field confinement in an SPP
- But SPPs tend to show high loss

Measure intensity dependence of the Kretschmann angle

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

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

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

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

\[ KL = 4 \]

- 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 \( S = \frac{n_g}{n} \)
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

5. Slow and Fast Light
Controlling the Velocity of Light

“Slow,” “Fast” and “Backwards” Light

– Light can be made to go:
  
  slow: \( v_g << c \) (as much as \( 10^6 \) times slower!)
  fast: \( v_g > c \)
  backwards: \( v_g \) negative

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

– Velocity controlled by structural or material resonances

Slow and Fast Light Using Isolated Gain or Absorption Resonances

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

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Note also related work by Chu, Wong, Welch, Scully, Budker, Ketterle, and many others
Goal: Slow Light in a Room-Temperature Solid-State Material

Crucial for many real-world applications

We have identified two preferred methods for producing slow light

1. Slow light \textit{via} coherent population oscillations (CPO)
2. Slow light \textit{via} stimulated Brillouin scattering (SBS)
Slow Light by Stimulated Brillouin Scattering (SBS)

\[ \frac{dI_S}{dz} = -gI_L I_S \]

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

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 \[ G = g I_L L \] and \[ \Gamma_B \] is the Brillouin linewidth.

Ground state population oscillates at beat frequency $\delta$ (for $\delta < 1/T_1$).

$$K_{ba} = \frac{1}{T_1}$$

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!
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

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

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.
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
8. Spontaneous and Stimulated Light Scattering
Spontaneous vs. Stimulated Light Scattering

Light scattering can occur only due to fluctuations in the optical properties of materials. Consider a completely homogeneous medium:

\[ \frac{\lambda}{2 \sin \theta} \]

incident plane wave

complete destructive interference!

Spontaneous light scattering

Fluctuations (e.g., in $\mathbf{E}$) 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 → scattering medium
Intensity $I_0$
Frequency $\nu_0$

Scattered light

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

Stokes ← | → Anti-Stokes

<table>
<thead>
<tr>
<th>Process</th>
<th>Shift</th>
<th>Linewidth</th>
<th>Relaxation Time</th>
<th>Gain (cm/NW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raman</td>
<td>1000 cm$^{-1}$</td>
<td>5 cm$^{-1}$</td>
<td>$10^{-12}$ s</td>
<td>$5 \times 10^{-3}$</td>
</tr>
<tr>
<td>Brillouin</td>
<td>0.1 - 1</td>
<td>$5 \times 10^{-3}$</td>
<td>$10^{-9}$ s</td>
<td>$10^{-2}$</td>
</tr>
<tr>
<td>Rayleigh</td>
<td>0</td>
<td>$5 \times 10^{-4}$</td>
<td>$10^{-8}$ s</td>
<td>$10^{-4}$</td>
</tr>
<tr>
<td>Rayleigh-Wing</td>
<td>0</td>
<td>10</td>
<td>$10^{-12}$ s</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:

\[ G = \frac{4 \pi^3 N c^2}{\hbar \omega_L \omega_s^2 n_s^2} \left( \frac{\omega}{\omega_s} \frac{\omega}{\omega_s} \frac{\omega}{\omega_s} \right) I_L \]

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

- Spontaneous Brillouin scattering is a very weak process.

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

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

\( P \geq 20 \text{ kW} \)

\( T \geq 5 \text{ nsec} \)

organic liquid (CS\(_2\))
Theory of SBS (backward SBS)

\[ \begin{array}{c}
E_1, \omega_1, \vec{k}_1 \\
\downarrow \\
E_2, \omega_2, \vec{k}_2 \\
\downarrow \\
\rho, \Omega, \vec{q} \\
\downarrow \\
\text{Sound wave}
\end{array} \]

Positive feedback:
- laser + Stokes \rightarrow sound
- laser + sound \rightarrow Stokes

Frequencies:
\[
\omega_2 = \omega_1 - \Omega_B, \quad \Omega_B = \left| \frac{\vec{q}}{\vec{k}_1} \right| \nu \\
\vec{q} = \vec{k}_1 - \vec{k}_2 \approx 2 \vec{k}_1
\]

\[ \Rightarrow \Omega_B = 2\omega \frac{\nu}{c/\eta} \]  
\( (\Omega_B / 2\pi \approx 5 \text{ GHz}) \)

Coupling among waves: electrostriction

Material tends to be drawn into regions of high field.

\[
P_{st} = -\gamma e \frac{E^2}{8\pi}
\]

\[
\gamma e = \rho \frac{\partial E}{\partial \rho} \approx 1 \text{ for condensed matter}
\]

\[
\frac{\Delta \rho}{\rho} = -C P_{st}
\]

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

\[ \tilde{E}_1(z,t) = A_1(z,t) e^{i(k_1z - \omega_1 t)} + \text{cc} \]

\[ \tilde{E}_2(z,t) = A_2(z,t) e^{i(k_1z - \omega_2 t)} + \text{cc} \]

\[ \bar{\rho}(z,t) = \rho_0 + \left[ \rho(z,t) e^{i(\theta z - \Omega t)} + \text{cc} \right] \]

\[ \begin{align*}
\mathcal{B} & = 2k_1 \\
\Omega & = \omega_1 - \omega_2 \\
\end{align*} \]

density \( \bar{\rho} \) obeys acoustic wave equation

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

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

\[ \bar{f} = \nabla \bar{P}_{st} \quad \bar{P}_{st} = -\gamma_e \frac{\langle \tilde{E}^2 \rangle}{8\pi} \]

Thus

\[ \nabla \cdot \bar{f} = \frac{\gamma_e \varepsilon^2}{4\pi} \left[ A_1 A_2^* e^{i(\theta z - \Omega t)} + \text{cc} \right] \]

In the SVAA, acoustic wave equation becomes

\[ -2i \Omega \frac{\partial \rho}{\partial t} + (\Omega_e^2 - \Omega^2 - i\Omega \Gamma_e) \rho - 2i\gamma \nu^2 \frac{\partial \rho}{\partial z} = \frac{\gamma_e \varepsilon^2}{4\pi} A_1 A_2^* \]

where

\[ \Gamma_e = \varepsilon^2 \Gamma' \quad \Omega_e = 2\omega \frac{\nu}{c/n} \]
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

$$\rho(z,t) = \frac{Y e \varepsilon}{4\pi} \frac{A_1 A_2^*}{\Omega_b^2 - \Omega^2 - i\Omega \Gamma_b}$$

- 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

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

$$= \frac{1}{4\pi \rho_0} Y e (\tilde{P} - \rho_0) \tilde{E}$$

Thus

$$\frac{\partial A_1}{\partial z} + \frac{1}{c/n} \frac{\partial A_1}{\partial t} = \frac{i\omega Y e}{2\pi c \rho_0} \rho A_2$$

$$- \frac{\partial A_2}{\partial z} + \frac{1}{c/n} \frac{\partial A_2}{\partial t} = \frac{i\omega Y e}{2\pi c \rho_0} \rho^* A_1$$

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

$$\frac{dA_1}{dz} = -\omega \frac{Y e \varepsilon}{8\pi n c \rho_0 \Delta \Gamma} (A_2 A_2^*) A_1$$

$$\frac{dA_2}{dz} = -\omega \frac{Y e \varepsilon}{8\pi n c \rho_0 \Delta \Gamma} (A_1 A_1^*) A_2$$
Theory of SBS (continued)

Can express in terms of intensities \( I_i = (ne^2 / 2\pi) |\mathbf{A}_i|^2 \)

\[
\frac{dI_1}{dz} = -g_0 I_1 I_2 \\
\frac{dI_2}{dz} = -g_0 I_1 I_2
\]

\( g_0 = \frac{\gamma e^2 \omega^2}{n v c^3 \rho_0 \Gamma} \) \( \sim \) SBS gain factor

\( \sim 0.01 \text{ cm/MW} \)

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

Solution in constant-pump limit (\( I_1 \) constant)

\[
I_2(z) = I_2(L) e^{g_0 I_1 (L-z)}
\]

Total amplification = \( I_2(0) / I_2(L) = e^{g_0 I_1 L} \)
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

\[ W_L \rightarrow \text{Raman medium} \rightarrow W_s = W_L - W_o \]

We can predict the Stokes output intensity:

\[ I_L \rightarrow \text{Raman medium} \rightarrow I_s(L) \]

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

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

Need 1.9 μm light

1.06 μm 4155 cm⁻¹ H₂ 1.9 μm

Experimental setup

f = 40 cm 50 cm

λ = 1.06 μm

30 ps pulse length

3 mJ pulse energy

H₂ gas at 30 atm

λ = 1.9 μm

150 μJ energy

50% conversion
## Typical Raman media

<table>
<thead>
<tr>
<th>Material</th>
<th>$\Delta v$ (cm$^{-1}$)</th>
<th>$g \times 10^3$ (cm/MW)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>LIQUIDS</strong></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><strong>GASES</strong></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

\[ W_L \rightarrow W_S \rightarrow \text{Stokes signal grows from noise.} \]

(Initiation due to thermal or quantum fluctuations)

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

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

**Amplifier**

Apply both laser and Stokes frequency

\[ W_L \leftrightarrow W_S \rightarrow \text{Stokes "seed"} \]

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

Alternative geometries:

1. \[ W_L \rightarrow W_S \rightarrow W_S \]
2. \[ W_L \rightarrow W_S \rightarrow W_S \]
Stimulated Scattering Processes

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

\[ \omega_L, \vec{k}_L \rightarrow \rightarrow \Omega, \vec{\gamma} \]
\[ \omega_S, \vec{k}_S \leftarrow \rightarrow \Omega = \frac{\omega}{\gamma} - \vec{k}_L - \vec{k}_S \]

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

\[ \omega_L \rightarrow \rightarrow \omega_S \]
\[ \omega_S = \omega_L - \Omega \]
\[ \Omega \sim \frac{1}{T_{\text{thermal}}} \]

Stimulated Raman Scattering
interaction with vibrational degree of freedom

\[ \omega_L \rightarrow \rightarrow \omega_S \]
\[ \omega_S = \omega_L - \omega_V \]

Stimulated Rayleigh-Wing Scattering
interaction with orientational degree of freedom \( \sim 2 \text{ps} \)

\[ \omega_L \rightarrow \rightarrow \omega_S \]
\[ \omega_S = \omega_L - \Omega \]
\[ \Omega \sim \frac{1}{T_{\text{orientation}}} \]
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 (?)

ps laser pulse → White light (broad frequency spectrum)

Thermal Blooming (Stimulated Thermal Rayleigh Scattering)

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

Joe Davis, MIT
Thank you for your attention!