Ultra-Slow and Superluminal Light and Enhanced Optical Nonlinearities based on Quantum Coherence and on Artificial Optical Materials

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What is **Nonlinear Optics**?

\[ P = X^{(1)} E + X^{(2)} E^2 + X^{(3)} E^3 + \ldots \]

- \( X^{(1)} \): linear optics, eg

- \( X^{(2)} \): second-order effects, eg, second-harmonic generation

\[ \omega \rightarrow \text{rectangular} \rightarrow 2\omega \]

- \( X^{(3)} \): third-order effects, eg

  four-wave mixing

  intensity-dependent refractive index

\[ n = n_0 + n_2 E \]

\[ n_2 = \frac{12 \pi^2}{n_0^2 c} X^{(3)} \]
The Promise of Nonlinear Optics

Nonlinear optical techniques hold great promise for applications including:

- Photonic Devices
- Quantum Imaging
- Quantum Computing/Communications
- Optical Switching
- Optical Power Limiters
- All-Optical Image Processing

But the lack of high-quality photonic materials is often the chief limitation in implementing these ideas.
Approaches to the Development of Improved NLO Materials

- New chemical compounds
- Quantum coherence (EIT, etc.)
- Composite Materials:
  (a) Microstructured Materials, e.g. Photonic Bandgap Materials, Quasi-Phase matched Materials, etc
  (b) Nanocomposite Materials

These approaches are not incompatible and in fact can be exploited synergistically!
Nanocomposite Materials for Nonlinear Optics

- Maxwell Garnett
- Bruggeman (interdispersed)
- Fractal Structure
- Layered

scale size of inhomogeneity $\ll$ optical wavelength
Gold-Doped Glass: A Maxwell-Garnett Composite

Red Glass Caraffe, Nuremberg, ca. 1700
Bielefeld museum.

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 strongly at the surface plasmon frequency, which is in the blue.
Demonstration of Enhanced NLO Response

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

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

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

Thus field is concentrated in lower index material.

Enhanced EO Response of Layered Composite Materials

\[ \chi_{ijkl}^{(\text{eff})}(\omega';\omega,\Omega_1,\Omega_2) = f_a \left( \frac{\varepsilon_{\text{eff}}(\omega')}{\varepsilon_a(\omega')} \right) \left( \frac{\varepsilon_{\text{eff}}(\omega)}{\varepsilon_a(\omega)} \right) \left( \frac{\varepsilon_{\text{eff}}(\Omega_1)}{\varepsilon_a(\Omega_1)} \right) \left( \frac{\varepsilon_{\text{eff}}(\Omega_2)}{\varepsilon_a(\Omega_2)} \right) \chi_{ijkl}^{(a)}(\omega';\omega,\Omega_1,\Omega_2) \]

- AF-30 (10%) in polycarbonate (spin coated)
  \[ n = 1.58 \quad \varepsilon(\text{dc}) = 2.9 \]
- Barium titante (rf sputtered)
  \[ n = 1.98 \quad \varepsilon(\text{dc}) = 15 \]
  \[ \chi_{zzzz}^{(3)} = (3.2 + 0.2i) \times 10^{-21} \ (m / V)^2 \pm 25\% \]
  \[ \approx 3.2 \chi_{zzzz}^{(3)} \text{(AF-30 / polycarbonate)} \]

3.2 times enhancement in agreement with theory

Accessing the Optical Nonlinearity of Metals with Metal-Dielectric PBG Structures

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

40 times enhancement of NLO response is predicted!

Open-aperture Z-scan
(measures $\text{Im} \chi^{(3)}$)

$I = 500 \text{ MW/cm}^2$
$\lambda = 640 \text{ nm}$

$$\frac{\delta \phi_{\text{PBG}}}{\delta \phi_{\text{Cu}}} \approx 35$$
Interest in Slow Light

Fundamentals of optical physics

Intrigue: Can (group) refractive index really be $10^6$?

Optical delay lines, optical storage, optical memories

Implications for quantum information
Slow Light

group velocity ≠ phase velocity
Group Velocity

Pulse (wave packet) \[ \rightarrow \mathbf{v}_g \]

Group velocity given by \[ \mathbf{v}_g = \frac{d\mathbf{w}}{d\mathbf{k}} \]

For \[ \mathbf{k} = \frac{n\mathbf{w}}{c} \] \[ \frac{d\mathbf{k}}{d\mathbf{w}} = \frac{1}{c} (n + \mathbf{w} \frac{dn}{d\mathbf{w}}) \]

Thus \[ \mathbf{v}_g = \frac{c}{n + \mathbf{w} \frac{dn}{d\mathbf{w}}} = \frac{c}{n_g} \]

Thus \( n_g \neq n \) in a dispersive medium!
Want $U_g$ very different from $U_p$

Need very large dispersion

Study resonances of atomic vapor

\[ U_g = \frac{c}{n + w \frac{dn}{dw}} \]
Light Propagation in Atomic Vapors

\[ n = \sqrt{\varepsilon} = \sqrt{1 + 4\pi \chi} \quad \chi = \frac{Ne^2/2m\omega_0}{(\omega_0 - \omega) - i\gamma} \]

For \( N \) not too large, \( n = n' + in'' \approx 1 + 2\pi \chi \)

\[ n' = 1 + \frac{\pi Ne^2}{m\omega_0} \frac{\omega_0 - \omega}{(\omega_0 - \omega)^2 + \gamma^2} \]

\[ n'' = \frac{\pi Ne^2}{2m\omega_0\gamma} \frac{\gamma^2}{(\omega_0 - \omega)^2 + \gamma^2} \]

\[ n_g = n' + \omega \frac{dn'}{d\omega} \]

\[ \frac{\omega \frac{dn'(\max)}{d\omega}}{\gamma} \approx \frac{2\pi (5 \times 10^{14})(0.1)}{2\pi (1 \times 10^9)} = 5 \times 10^4 \sim (1) \]

\( n_g \) can range from \( +5 \times 10^4 \) to \( -5 \times 10^4 \).

(But with lots of absorption)
How to Produce Slow Light?

Group index can be as large as

$$n_g = 1 + \frac{W \, S_{\text{max}}}{\gamma}$$

Use Nonlinear optics to

1. decrease line width $\gamma$
   (produce sub-Doppler linewidth)

2. decrease absorption
   (so transmitted pulse is detectable)
Slow Light in Atomic Media

Slow light propagation in atomic media (vapors and BEC), facilitated by quantum coherence effects, has been successfully observed by many groups.
Light speed reduction to 17 metres per second in an ultracold atomic gas

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\[
v_g = \frac{c}{n(\omega_p) + \frac{dn}{d\omega_p}} = \frac{\hbar c e_0}{2 \omega_p} \frac{|\Omega_c|^2}{|\mu_1|^2 N}
\]

\[
T = 450 \text{ nK}
\]
\[
\tau_{\text{Delay}} = 7.05 \pm 0.05 \mu s
\]
\[
L = 229 \pm 3 \mu m
\]
\[
v_g = 32.5 \pm 0.5 \text{ m s}^{-1}
\]
Challenge/Goal

Slow light in room-temperature solid-state material.

• Slow light in room temperature ruby 
  (facilitated by a novel quantum coherence effect)

• Slow light in a structured waveguide
Slow Light in Ruby

Need a large $dn/dω$. (How?)

Kramers-Kronig relations:
   Want a very narrow absorption line.

Well-known (to the few people how know it well) how to do so:

Make use of “spectral holes” due to population oscillations.

Hole-burning in a homogeneously broadened line; requires $T_2 << T_1$.

PRL 90,113903(2003); see also news story in Nature.
Spectral Holes in Homogeneously Broadened Materials

Occurs only in collisionally broadened media ($T_2 \ll T_1$)

Fig. 3. Attenuation of the modulated component (probe beam) is plotted as a function of modulation frequency. The probe beam experiences decreased absorption at low modulation frequencies. The width of this hole is 37 Hz for low laser powers. The spectral hole is power broadened at high laser powers.
**Spectral Holes Due to Population Oscillations**

\[ 2 \gamma_{ba} = \frac{2}{T_2} \]

\[ \Gamma_{ba} = \frac{1}{T_1} \]

\[ E_3, \omega + \delta \]

\[ \rightarrow \]

\[ E_1, \omega \]

\[ \text{atomic medium} \]

\[ \omega + \delta \]

\[ \rightarrow \]

\[ \text{measure absorption} \]

Population inversion:

\[ (\rho_{bb} - \rho_{aa}) = w \]

\[ w(t) \approx w^{(0)} + w^{(-\delta)} e^{i\delta t} + w^{(\delta)} e^{-i\delta t} \]

population inversion terms important only for \( \delta \leq 1 / T_1 \)

Probe-beam response:

\[ \rho_{ba}(\omega + \delta) = \frac{\mu_{ba}}{\hbar} \frac{1}{\omega - \omega_{ba} + i / T_2} \left[ E_3 w^{(0)} + E_1 w^{(\delta)} \right] \]

Probe-beam absorption:

\[ \alpha(\omega + \delta) \propto \left[ w^{(0)} - \frac{\Omega^2 T_2}{T_1} \frac{1}{\delta^2 + \beta^2} \right] \]

linewidth \( \beta = \frac{1}{T_1} \left( 1 + \Omega^2 T_1 T_2 \right) \)
Experimental Setup Used to Observe Slow Light in Ruby

7.25 cm ruby laser rod (pink ruby)
Measurement of Delay Time for Harmonic Modulation

For 1.2 ms delay, $v = 60$ m/s and $n_g = 5 \times 10^6$
Gaussian Pulse Propagation Through Ruby

$v = 140 \text{ m/s}$

$ng = 2 \times 10^6$

No pulse distortion!
Alexandrite Displays both Saturable and Inverse-Saturable Absorption

Mirror Sites:

Inversion Sites:

$T_{1,m} = 260 \mu s$

$T_{1,i} \sim 50 \text{ ms}$
At 476 nm, alexandrite is an inverse saturable absorber.

Negative time delay of 50 μs corresponds to a velocity of -800 m/s.
Slow and Fast Light -- What Next?

Longer fractional delay
(saturate deeper; propagate farther)

Find material with faster response
(technique works with shorter pulses)
Artificial Materials for Nonlinear Optics

Artificial materials can produce
Large nonlinear optical response
Large dispersive effects

Examples
Fiber/waveguide Bragg gratings
PBG materials
CROW devices (Yariv et al.)
SCISSOR devices
Motivation

To exploit the ability of microresonators to enhance nonlinearities and induce strong dispersive effects for creating structured waveguides with exotic properties.

A cascade of resonators side-coupled to an ordinary waveguide can exhibit:

- slow light propagation
- induced dispersion
- enhanced nonlinearities
Ultrafast All-Optical Switch Based On Arsenic Triselenide Chalcogenide Glass

- We excite a whispering gallery mode of a chalcogenide glass disk.

- The nonlinear phase shift scales as the square of the finesse $F$ of the resonator. ($F \approx 10^2$ in our design)

- Goal is 1 pJ switching energy at 1 Tb/sec.

(implementation with Dick Slusher, Lucent)
A Real Whispering Gallery

St. Paul's Cathedral, London
NLO of SCISSOR Devices

(Side-Coupled Integrated Spaced Sequence of Resonators)

Shows slow-light, tailored dispersion, and enhanced nonlinearity

Optical solitons described by nonlinear Schrodinger equation

- Weak pulses spread because of dispersion

- But intense pulses form solitons through balance of dispersion and nonlinearity.
Slow Light and SCISSOR Structures

\[ \text{group index} = \frac{2\pi F_n}{nR} \]

\[ k_{\text{eff}} - k_0 = \frac{2\pi}{L} \]

\[ \text{slope} = \frac{F_n}{c} \]

\[ \frac{c}{F_n R} \]

\[ \text{build-up} \]

\[ \frac{2\pi F}{c} \]

\[ 0 \]

\[ \text{frequency, } \omega \]
Nanofabrication

- Materials (artificial materials)
- Devices

(d distinction?)
Microdisk Resonator Design

All dimensions in microns

GaAs
Al_{x}Ga_{1-x}As
(x = 0.4)

J. E. Heebner and R. W. Boyd
Photonic Device Fabrication Procedure

1. MBE growth
2. Deposit oxide
3. Spin-coat e-beam resist
4. Pattern inverse with e-beam & develop
5. RIE etch oxide
6. Remove PMMA
7. CAIBE etch AlGaAs-GaAs
8. Strip oxide
Disk Resonator and Optical Waveguide in PMMA Resist
Photonic Devices Written Into PMMA Resist
All-Pass Racetrack Microresonator

- 10 microns
- 100 nanometer gap
- 500 nanometer guide widths
- 2.5 micron height
Five-Cell SCISSOR with Tap Channel

5 microns  
100 nanometer gaps
500 nanometer guides
2.5 micron height
Resonator-Enhanced Mach-Zehnder Interferometers

~100 nanometer gaps
500 nanometer guides
2.5 micron height
Laboratory Characterization of Photonic Structures

- Characterization of fiber ring-resonator devices (Proof of principle studies)
- Characterization of nanofabricated devices
Fiber optical delay line:

First study one element of optical delay line:

variable wavelength pulse

- 27 ns (delay)
- 51 ns (FWHM)
Transmission Characteristics of Fiber Ring Resonator

Measure transmission vs. $\lambda$ for various values of the finesse

circumference = 31 cm

variable coupling

undercoupled

critically coupled

overcoupled
Phase Characteristics of Fiber Ring Resonator

Place ring resonator inside Mach-Zhender interferometer and measure transmission versus wavelength.

\[
\begin{align*}
\mathcal{F} &= 51 \\
\mathcal{F} &= 40 \\
\mathcal{F} &= 30 \\
\mathcal{F} &= 17 \\
\mathcal{F} &= 12 \\
\mathcal{F} &= 7.3 \\
\mathcal{F} &= 6.8 \\
\mathcal{F} &= 4.4
\end{align*}
\]

\[
\begin{align*}
\text{transmission} \\
\text{transmission} \\
\text{transmission} \\
\text{transmission} \\
\text{transmission} \\
\text{transmission} \\
\text{transmission} \\
\text{transmission}
\end{align*}
\]

\[
\begin{align*}
0 & \quad 10 & \quad 20 & \quad 30 & \quad 40 \\
\Delta \text{wavelength (pm)}
\end{align*}
\]

variable coupler

undercoupled

critically coupled

overcoupled
Phase Characteristics of Fiber Ring Resonator

Extracted phase structure

variable coupler

undercoupled

critically coupled

overcoupled

$F = 51$

$F = 40$

$F = 30$

$F = 17$

$F = 4.4$

$F = 12$

$F = 7.3$

$F = 6.8$
"Fast" (Superluminal) Light in SCISSOR Structures

Requires loss in resonator structure

\[
\frac{k_{\text{eff}} - k_0}{L} = 2\pi
\]

\[f - f_0\]

Frequency detuning, \(\omega - \omega_R\)

Power (arb. units)

Propagation through SCISSOR

Propagation through vacuum

Delay (ps)
Laboratory Characterization of Photonic Structures

- Characterization of fiber ring-resonator devices (Proof of principle studies)
- Characterization of nanofabricated devices
Microresonator-Based Add-Drop Filter

Output for on-resonance input

Variable wavelength input

Output for off-resonance input

Transmission vs. Wavelength (micrometers)
Phase Characteristics of Micro-Ring Resonator

Transmission

Induced phase shift
All-Optical Switching in a Microresonator-Enhanced Mach-Zehnder Interferometer

Increasing pulse energy (~1 nJ)

Output intensity (arb. units)

Output position (x)

Output port #1

Output port #2
Summary

Artificial materials hold great promise for applications in photonics because of

• large controllable nonlinear response
• large dispersion controllable in magnitude and sign

Demonstration of slow light propagation in ruby
Nonlinear optics is an extremely exciting research area because it includes topics that range from fundamental physics to numerous applications.
Thank you for your attention.

Feliz Cinco de Mayo.
Objective:
Obtain high sensitivity, high specificity detection of pathogens through optical resonance

Approach:
Utilize high-finesse whispering-gallery-mode disk resonator.

Presence of pathogen on surface leads to dramatic decrease in finesse.

Simulation of device operation:

Intensity distribution in absence of absorber.

Intensity distribution in presence of absorber.

FDTD
Deposition of Surface Binding Layer

1) Bare device surface

GaAs or AlGaAs

2) SiO₂ layer deposited by PECVD

~300 nm

GaAs or AlGaAs

SiO₂

3) Silane coupling agent deposited on surface

~1.2 nm

SiO₂

GaAs or AlGaAs

3-Mercaptopropyl Trimethoxysilane (MPT)
(C₆H₁₆O₃SSi)

4) Antibodies washed over surface / adhere to MPT

~12 nm

SiO₂

GaAs or AlGaAs

5) Pathogen captured by antibody layer

~1-5 microns

target pathogen

SiO₂

GaAs or AlGaAs
Demonstration of Selective Binding onto GaAs

- biotin on GaAs
- streptavidin over biotin on GaAs
- biotin on silica-coated GaAs
- streptavidin over biotin on silica-coated GaAs
- biotin on microscope slide
- streptavidin over biotin on microscope slide

Notes: 1. false-color images of fluorescent intensity are shown
2. streptavidin is tagged with the dye Cy3

University of Rochester/Corning Collaboration
Photonic Structures -- What Next?
Performance of SCISSOR as Optical Delay Line

Input
(duty factor = 1/3)

Output --delayed one time slot by six resonators in linear limit

Output --delayed four time slots by 26 resonators in linear limit

Output --delayed four time slots by 26 resonators in nonlinear limit
Frequency Dependence of GVD and SPM Coefficients

\[
\begin{align*}
\text{(GVD)} & : \quad \frac{k''_{\text{eff}}}{(2FT/\pi)^2/L} \\
\text{(SPM)} & : \quad \frac{\gamma_{\text{eff}}}{\gamma (2F/\pi)^2 (2\pi R/L)}
\end{align*}
\]
Soliton Propagation

5 µm diameter resonators with a finesse of 30

SCISSOR may be constructed from 100 resonators spaced by 10 µm for a total length of 1 mm

soliton may be excited via a 10 ps, 125mW pulse

simulation assumes a chalcogenide/GaAs-like nonlinearity

Weak Pulse

Fundamental Soliton
Dark Solitons

SCISSOR system also supports the propagation of dark solitons.
SCISSOR Dispersion Relations

Single-Guide SCISSOR
No bandgap
Large intensity buildup

Double-Guide SCISSOR
Bandgaps occur
Reduced intensity buildup

separation = 1.5 \pi R

Bragg + resonator gap
resonator gap
Bragg gap
Optical Power Limiting in a Nonlinear Mach-Zehnder Interferometer
Spectral Dependence of the Nonlinear Response

![Graph showing spectral dependence of nonlinear response with peak wavelengths and bulk material comparison.]

**OPG:**
- $t = 25$ ps
- $Q = 2$ to $5$ mJ
- $I \approx 100$ MW/cm$^2$
Is This a Good Idea?

Protein detection by optical shift of a resonant microcavity