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

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Light-by-Light Scattering





SECOND EDITION



NONLINEAR Optics

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What is Nonlinear Optics?

$$P = \chi^{(1)} E + \chi^{(2)} E^{2} + \chi^{(3)} E^{3} + \cdots$$
dipole moment per unit volume

$$\chi^{(1)}: \text{ linear optics, eg } \int$$

$$\chi^{(2)}: \text{ Second-order effects, eg,}$$

$$\text{ second-harmonic generation}$$

$$\frac{\omega}{\sqrt{3}}: \text{ third-order effects, eg}$$

$$four - wave mixing$$

$$\text{Intensity - dependent}$$

$$\text{ refractive index}$$

$$N = N_{0} + N_{2} E$$

$$N_{2} = \frac{(2\pi^{2})}{n_{0}^{2}c} \chi^{(3)}$$

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-Phasematched Materials, etc
 - (b) Nanocomposite Materials

These approaches are not incompatible and in fact can be exploited synergistically!

Nanocomposite Materials for Nonlinear Optics

• Maxwell Garnett



• Fractal Structure



• Bruggeman (interdispersed)



• Layered



scale size of inhomogeneity << optical wavelength

Gold-Doped Glass: A Maxwell-Garnett Composite



Red Glass Caraffe, Nurenberg, 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 TiO₂ and the conjugated polymer PBZT.
 - - $\nabla \cdot \mathbf{D} = 0$ implies that $(\boldsymbol{\varepsilon} \mathbf{E})_{\perp}$ is continuous.

Thus field is concentrated in *lower* index material.

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



Fischer, Boyd, Gehr, Jenekhe, Osaheni, Sipe, and Weller-Brophy, Phys. Rev. Lett. 74, 1871, 1995. Gehr, Fischer, Boyd, and Sipe, Phys. Rev. A 53, 2792 1996.

Enhanced EO Response of Layered Composite Materials



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

- AF-30 (10%) in polycarbonate (spin coated) n=1.58 $\epsilon(dc) = 2.9$
- barium titante (rf sputtered) n=1.98 $\epsilon(dc) = 15$ $\chi^{(3)}_{zzzz} = (3.2 + 0.2i) \times 10^{-21} (m/V)^2 \pm 25\%$ $\approx 3.2 \chi^{(3)}_{zzzz}$ (AF-30/polycarbonate)

3.2 times enhancement in agreement with theory

R. L. Nelson, R. W. Boyd, Appl. Phys. Lett. 74, 2417, 1999.

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 enhancment of NLO response is predicted!

R.S. Bennink, Y.K. Yoon, R.W. Boyd, and J. E. Sipe Opt. Lett. 24, 1416, 1999.

Z-Scan Comparison of M/D PBG and Bulk Sample

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

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

 $\lambda = 640 \text{ nm}$

T_{norm} 1.00 Cu 0.75 0.50 PBG

 $\frac{\delta \phi_{\mathsf{PBG}}''}{\delta \phi_{\mathsf{Cu}}''} \cong 35$

z,mm

100 150 200 250

50

0

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 given by $V_{\overline{3}} = \frac{dW}{dR}$ For $k = \frac{n\omega}{c}$ $\frac{dk}{d\omega} = \frac{1}{c} \left(n + \omega \frac{dn}{d\omega} \right)$

Thus

 $V_{g} = \frac{c}{n + \omega \frac{dn}{d\omega}} = \frac{c}{n_{g}}$

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

- Want Vg very different from Vp Need very large dispersion Study resonances of atomic vapor

 $V_{\overline{g}} = \frac{c}{n + \omega \frac{dn}{d\omega}}$



How to Produce Slow Light ? Group index can be as large as $n_g \sim 1 + \frac{W Sn(max)}{\chi}$ Use nonlinear optics to (1) decrease line width Y (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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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 $dn/d\omega$. (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 $T_2 \ll T_1$.



inhomogeneously broadened medium homogeneously broadened medium (or inhomogeneously broadened)

PRL 90,113903(2003); see also news story in Nature.

Spectral Holes in Homogeneously Broadened Materials

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



Boyd, Raymer, Narum and Harter, Phys. Rev. A24, 411, 1981.

OBSERVATION OF A SPECTRAL HOLE DUE TO POPULATION OSCILLATIONS IN A HOMOGENEOUSLY BROADENED OPTICAL ABSORPTION LINE

Lloyd W. HILLMAN, Robert W. BOYD, Jerzy KRASINSKI and C.R. STROUD, Jr. The Institute of Optics, University of Rachester, Rochester, NY 14627, USA



Spectral Holes Due to Population Oscillations



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 oscillation 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 $\beta = (1 / T_1) (1 + \Omega^2 T_1 T_2)$

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



No pulse distortion!

Alexandrite Displays both Saturable and Inverse-Saturable Absorption



Inverse-Saturable Absorption Produces Superluminal Propagation in Alexandrite

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

Artifical 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.



J. E. Heebner and R. W. Boyd, Opt. Lett. 24, 847, 1999. (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



Nanofabrication

- Materials (artificial materials)
- Devices

(distinction?)

Microdisk Resonator Design

All dimensions in microns



Photonic Device Fabrication Procedure



Disk Resonator and Optical Waveguide in PMMA Resist



AFM





All-Pass Racetrack Microresonator



Five-Cell SCISSOR with Tap Channel



Resonator-Enhanced Mach-Zehnder Interferometers



~100 nanometer 500 nanometer 2.5 micron gaps guides height

Laboratory Characterization of Photonic Structures

- Characterization of fiber ring-resonator devices (Proof of principle studies)
- Characterization of nanofabricated devices

Fiber-Resonator Optical Delay Line

Fiber optical delay line:

 $\bigcirc]$

First study one element of optical delay line:





Transmission Characteristics of Fiber Ring Resonator



Phase Characteristics of Fiber Ring Resonator



Phase Characteristics of Fiber Ring Resonator



"Fast" (Superluminal) Light in SCISSOR Structures

Requires loss in resonator structure



Laboratory Characterization of Photonic Structures

- Characterization of fiber ring-resonator devices (Proof of principle studies)
- Characterization of nanofabricated devices

Microresonator-Based Add-Drop Filter



Phase Characteristics of Micro-Ring Resonator





transmission

induced phase shift



All-Optical Switching in a Microresonator-Enhanced Mach-Zehnder Interferometer



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

Real Summary

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.

Alliance for Nanomedical Technologies

Photonic Devices for Biosensing

Simulation of device operation:

Objective:

Obtain high sensitivity, high specificity detection of pathogens through optical resonance

Approach:

Utilize high-finesse whispering-gallerymode disk resonator.

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



Intensity distribution in absense of absorber.



Intensity distribution in presence of absorber.



Deposition of Surface Binding Layer



Demonstration of Selective Binding onto GaAs



n

Photonic Structures --What Next?

Performance of SCISSOR as Optical Delay Line



Frequency Dependence of GVD and SPM Coefficients



Soliton Propagation



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









Optical Power Limiting in a Nonlinear Mach-Zehnder Interferometer



Spectral Dependence of the Nonlinear Response



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