Nanostructured Materials and Devices for Nonlinear Optics

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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
- Bruggeman (interdispersed)
- Fractal Structure
- Layered

Scale size of inhomogeneity $\ll$ optical wavelength
Gold-Doped Glass

A Maxwell-Garnett Composite

gold volume fraction approximately $10^{-6}$
gold particles approximately 10 nm diameter

- Composite materials can possess properties very different from their constituents.
- Red color is because the material absorbs very strongly at the surface plasmon frequency (in the blue) -- a consequence of local field effects.
Composite Optical Materials

• Why composite materials?
  At least -- Obtain best features of each component
  At best -- Properties of composite superior to those of its components.

• Specific Goal: Find structures for which the effective $\chi^{(3)}$ exceeds those of the constituents.*

• Enhancement of $\chi^{(3)}$ can be understood in terms of local field effects

First Demonstration of Enhanced NLO Response

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

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

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)
  \[ n = 1.98 \quad \varepsilon(\text{dc}) = 15 \]

\[ \chi_{zzzz}^{(3)} = (3.2 + 0.2i) \times 10^{-21} \text{ (m/V)}^2 \pm 25\% \]

\[ \approx 3.2 \chi_{zzzz}^{(3)}(\text{AF-30 / polycarbonate}) \]

3.2 times enhancement in agreement with theory

TWO GREAT IRONIES OF NONLINEAR OPTICS

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

   Fiber NLO $X^{(3)} \approx 1.8 \times 10^{-14}$ esu

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

   DILUTE COLLOID $X^{(3)}_{\text{silver}} \approx 10^{-8}$ esu
Accessing the Optical Nonlinearity of Metals with Metal-Dielectric PBG Structures

R.S. Bennink, Y.K. Yoon, R.W. Boyd, and J. E. Sipe

- 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)

![Pure Metal](image.png)

- Pure metal
- 80 nm of copper
- \( T = 0.3\% \)

![PBG Structure](image2.png)

- PBG structure
- 80 nm of copper
- (total)
- \( T = 10\% \)

- 40 times enhancement of NLO response is predicted!
Nanofabrication

• Materials (artificial materials)

• Devices

(distinction?)
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.

A Real Whispering Gallery

St. Paul's Cathedral, London
### Objective:
Obtain high sensitivity, high specificity detection of pathogens through optical resonance.

### Approach/Features:
Construct high-finesse whispering-gallery-mode disk resonator.
Presence of pathogen on surface leads to dramatic decrease in finesse.

### Simulation of device operation: (FDTD)
- Intensity distribution in absence of absorber.
- Intensity distribution in presence of absorber.

### Progress:
Device design is complete.
Beginning fabrication.
NLO of SCISSOR Devices
(Side-Coupled Integrated Spaced Sequence of Resonators)

Displays slow-light, tailored dispersion, and optical solitons.
Description by NL Schrodinger eqn. in continuum limit.

- Pulses spread when only dispersion is present

- But form solitons through balance of dispersion and nonlinearity

(J.E. Heebner, Q-Han Park and RWB)
Slow Light, Induced Dispersion, Enhanced Nonlinearity, Self-Steepening, and Optical Solitons in a Side-Coupled Integrated Spaced Sequence Of Resonators

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Motivation

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

Currently, most of the work done in microresonators involves applications such as disk lasers, dispersion compensators and add-drop filters. There's not much nonlinear action!

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

- slow light propagation
- induced dispersion
- enhanced nonlinearities
Properties of a Single Microresonator

Assuming negligible attenuation, this resonator is, unlike a Fabry-Perot, of the "all-pass" device - there is no reflected or drop port.

\[
\begin{pmatrix}
E_4 \\
E_2 \\
E_3 \\
E_1
\end{pmatrix} = \begin{pmatrix}
 r & it \\
it & r
\end{pmatrix}
\begin{pmatrix}
E_3 \\
E_1
\end{pmatrix}
\]

Intensity Enhancement \( \left( \frac{|E_3|}{E_1} \right)^2 \)

\[
\text{Build-up Factor} = \left| \frac{E_3}{E_1} \right|^2
\]

\[
r^2 = 0.75 \quad r^2 = 0.90 \\
r^2 = 0.00 \quad r^2 = 0.25
\]

Modified Dispersion Relation \( (\beta \text{ vs. } \omega) \)

\[
\omega_R - \frac{2\pi}{T} \quad \omega_R \quad \omega_R + \frac{2\pi}{T}
\]

\[
r^2 = 0.00 \quad r^2 = 0.75 \\
r^2 = 0.25 \quad r^2 = 0.90
\]

Definitions

Finesse
\[
F = \frac{\pi}{1-r}
\]

Transit Time
\[
T = \frac{n2\pi R}{c}
\]
Nonlinear Schrödinger Equation (NLSE)

\[
\frac{\partial A}{\partial z} = -i \frac{1}{2} \beta_2 \frac{\partial^2 A}{\partial t^2} + i \gamma |A|^2 A
\]

Fundamental Soliton Solution

\[
A(z,t) = A_0 \text{sech} \left( \frac{t}{T_p} \right) e^{i \frac{1}{2} \gamma |A_0|^2 z}
\]

By arranging a spaced sequence of resonators, side-coupled to an ordinary waveguide, one can create an effective, structured waveguide that supports pulse propagation in the NLSE regime.

Propagation is unidirectional, and there is NO photonic bandgap to produce the enhancement. Feedback is intra-resonator and not inter-resonator.
Balancing Dispersion & Nonlinearity

Resonator-induced dispersion can be 5-7 orders of magnitude greater than the material dispersion of silica!

Resonator enhancement of nonlinearity can be 3-4 orders of magnitude!

An enhanced nonlinearity may be balanced by an induced anomalous dispersion at some detuning from resonance to form solitons

A characteristic length, the soliton period may as small as the distance between resonator units!

\[
A_0 = \sqrt{\frac{|\beta_2|}{\gamma T_p^2}} = \sqrt{\frac{T^2}{\sqrt{3} \gamma 2\pi R T_p^2}}
\]

soliton amplitude

adjustable by controlling ratio of transit time to pulse width
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
Dark Solitons

SCISSOR system also supports the propagation of dark solitons.
Higher order dispersive terms such as $\beta_3$ are present in the system and become more dominant as the pulsewidth becomes nearly as short as the cavity lifetime. Because the nonlinear enhancement is in fact frequency dependent, or (equivalently here) because the group velocity is intensity dependent, self-steepening of pulses is possible even for relatively long pulse widths.

A generalized NLSE:

$$\frac{\partial}{\partial z} A = -i \frac{1}{2} \beta_2 \frac{\partial^2}{\partial t^2} A - \frac{1}{6} \beta_3 \frac{\partial^3}{\partial t^3} A + i \gamma |A|^2 A - s \frac{\partial}{\partial t} |A|^2 A$$

Self-steepening of a 20 ps Gaussian pulse after 100 resonators

steepened *leading* pulse edge due to negative SS
Soliton Splitting and Compression

The dispersive nature of the nonlinear enhancement (self-steepening) leads to an intensity-dependent group velocity which splits an N-order soliton into N fundamental solitons of differing peak intensities and widths.

Here, a 2\textsuperscript{nd} - order "breathing" soliton splits into 2 fundamental solitons:

Same parameters as used for the previous soliton example, but with 4X the launch power

Note that one pulse is compressed and is uncorrupted by the presence of a pedestal.
Radiation Losses

Is it really possible to confine light within a wavelength-size structure?

- Radiation losses provide fundamental limit to the finesse of a WGM resonator.
- Radiation losses are analogous to fiber bending losses.
- But these losses can be rendered negligible through use of a large dielectric contrast.
Waveguide-coupled AlGaAs/GaAs microcavity ring and disk resonators with high finesse and 21.6-nm free spectral range

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We report the realization and demonstration of novel semiconductor waveguide-coupled microcavity ring and disk resonators. For a 10.5-μm-diameter disk resonator, we measure a finesse of 120, a resonant linewidth of 0.18 nm, and a free-spectral range of 21.6 nm in the 1.55-μm-wavelength region. We present the nanofabrication methods and the experimental results for 10.5- and 20.5-μm-diameter ring and disk resonators to show the feasibility of such devices. © 1997 Optical Society of America

Fig. 1. (a) Illustration of the fabricated ring or disk resonator geometry and (b)–(d) infrared camera images showing switching of light from port Z to port Y at the 1555.6-nm resonance of the 10.5-μm-diameter disk.

Fig. 2. SEM images of a 10.5-μm-diameter (a) disk and (b) ring.
Microdisk Resonator Design

(Not drawn to scale)
All dimensions in microns

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\[ \text{GaAs} \]
\[ \text{Al}_{x}\text{Ga}_{1-x}\text{As} \]
\( (x = 0.4) \)

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

(1) MBE growth
AlGaAs-GaAs structure

(2) Deposit oxide
Oxide (SiO₂)
AlGaAs-GaAs structure

(3) Spin-coat e-beam resist
PMMA
Oxide (SiO₂)
AlGaAs-GaAs structure

(4) Pattern inverse with e-beam & develop
PMMA
Oxide (SiO₂)
AlGaAs-GaAs structure

(6) Remove PMMA
Oxide (SiO₂)
AlGaAs-GaAs structure

(7) CAIBE etch AlGaAs-GaAs
Oxide (SiO₂)
AlGaAs-GaAs structure

(5) RIE etch oxide
PMMA
Oxide (SiO₂)
AlGaAs-GaAs structure

(8) Strip oxide
AlGaAs-GaAs structure

RWB - 10/4/01
Nonlinear Optical Loop-De-Loop

J.E. Heebner and R.W.B.
Conclusions

The SCISSOR exhibits strong dispersive & nonlinear properties and supports solitons. It incorporates the essence of photonic bandgap (PBG) enhancement found in structures such as CROWs & Bragg gratings without the gap itself.

Manufacturability will require some ingenuity, but already resonators are already in use as all-pass filters and tunable dispersion compensators.

SCISSORs have the potential for producing soliton propagation effects at the integrated photonics scale. The possibilities for structurally engineered waveguides of this type include:

- Pulse compression / imaging in an integrated device
- Optical Time Division Multiplexing (OTDM)
- Soliton-based optical switching (perhaps w/ a few photons)
- Slow-light propagation (group-matched acousto-optics)
- EO,TO - Tunable NLSE propagation (A theorist's dream)
- Just about any other generalized NLSE effect