New Materials and Interactions for Nonlinear Optics

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Prospectus

Introduction to Nonlinear Optics

Development of New NLO Materials

EIT Techniques for Squeezed Light Generation

Some Underlying Issues in Nonlinear Optics
Nonlinear Optical Interactions

Light-by-Light Scattering

Phase Conjugation by Degenerate Four-Wave Mixing
What is Nonlinear Optics?

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

\( \chi^{(1)} \): Linear optics, e.g.

\( \chi^{(2)} \): Second-order effects, e.g., second-harmonic generation

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

\( \chi^{(3)} \): Third-order effects, e.g.

Four-wave mixing

Intensity-dependent refractive index

\[ n = n_0 + n_2 E \]

\[ n_2 = \frac{12 \pi^2}{n_0^2 c} \chi^{(3)} \]
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!
Electromagnetically Induced Transparency

too weak

better (2-photon resonance)

still better (but absorption!)

The EIT concept

EIT Predictions

Absorption of the generated field vanishes.

But the nonlinearity remains large!

Harris, Field and Imamoglu, PRL 64 1107 1990
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.

Z-Scan Measurement of $\chi^{(3)}$

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

Measures NL change in absorption. ($\Im \chi^{(3)}$)

Sheik Bahae, van Stryland, et al.
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

TWO GREAT IRONIES OF NONLINEAR OPTICS

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

Fiber NLO

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

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

Dilute colloid

$\chi^{(3)}_{\text{silver}} \approx 10^{-8} \text{ esu}$
Metal / Dielectric Composites

Very large local field effects

\[ \varepsilon_n \quad \varepsilon_m \quad \uparrow E_0 \quad E_{in} = \frac{3\varepsilon_n}{\varepsilon_m + 2\varepsilon_n} E_0 = 2E_0 \]  
(\(\varepsilon_m\) is negative !)

At resonance

\[ \lambda = \frac{3\varepsilon_n}{\varepsilon_m + 2\varepsilon_n} \rightarrow \frac{3\varepsilon_n}{i\varepsilon_m''} \approx (3 \text{ to } 30) i \]

\[ \chi^{(3)}_{\text{eff}} = f \lambda^2 |z|^2 \chi^{(3)}_m + (1-f) \chi^{(3)}_h \]
Countertuitive Consequence of Local Field Effects

gold nanoparticles in a liquid dye solution (HITC1)

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

Effective NL susceptibility of composite

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

\( z = \frac{3 \varepsilon_h}{E_m + 2 \varepsilon_h} = \text{pure imaginary at resonance!} \)

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

\[
\text{Normalized Transmittance}
\]

Accessing the Optical Nonlinearity of Metals with Metal-Dielectric PBG

- 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!

R.S. Bennink, Y.K. Yoon, R.W. Boyd, and J. E. Sipe
“Slow” Light in Nanostructured Devices

Robert W. Boyd

with

John Heebner, Nick Lepeshkin,
Aaron Schweinsberg, and Q-Han Park

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Nanofabrication

• Materials (artificial materials)
• Devices

(distinction?)
NLO of SCISSOR Devices
(Side-Coupled Integrated Spaced Sequence of Resonators)

Shows slow-light, tailored dispersion, and enhanced nonlinearity.

Optical solitons described by nonlinear Schrödinger equation.

- Weak pulses spread because of dispersion.

- But intense pulses form solitons through balance of dispersion and nonlinearity.
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: 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
Microdisk Resonator Design
(Not drawn to scale)
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

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

J.E. Heebner and R.W.B.
Photonic Devices Written into PMMA Resist
Pattern Etched Into Silica Mask
Photonic Devices in GaAs/AlGaAs
Generation of Squeezed Light by use of EIT

Robert W. Boyd and C. R. Stroud, Jr., University of Rochester

Three Approaches

Fundamental idea: EIT eliminates linear absorption so that there is no spontaneous emission background noise.
Application of EIT to Squeezed-Light Generation

- Squeezing by self-phase modulation


EIT allows phase shifts large enough to produce significant squeezing, and prevents signal-beam absorption which can degrade the squeezing.
Strong Absorption-Free Nonlinearity by Dark-State EIT

Saturation-Induced Extra Resonances

- Dark-State ($\Lambda$) Resonances
- Two-Level Atom Resonance
- V-System Resonances (saturation induced)

Pump-probe detuning [GHz]

- $10^{14}$ cm$^{-2}$
- Na

Transmitted probe power
Generation of Quantum States of Light by Two-Beam Excited Conical Emission

Efficient Far IR and THz Imaging by use of EIT

Basic concept of our approach. Because of strong saturation of the lower transitions, upconversion occurs with essentially unit efficiency.

Sodium energy levels for the conversion of 100 micron radiation to the visible.

Source of Polarized, Single-Photons on Demand

- Useful for secure communication by quantum cryptography
- Embed isolated dye molecules in chiral nematic liquid crystal
- Host acts as self-assembled photonic bandgap material
- Host composition helps prevent dye from bleaching
- Fluorescence shows strong antibunching

Experimental procedure
Implementation with S. Lukishova

Single-molecule fluorescence
Some Underlying Issues in Nonlinear Optics

• Self-Assembly/Self-Organization in Nonlinear Systems

• Stability vs. Instability (and Chaos) in Nonlinear Systems
Experimental Study of Soliton Propagation through 40 km of Dispersion-Decreasing Fiber

Andrew J. Stentz, Robert W. Boyd, University of Rochester
Alan F. Evans, Corning Inc.

- Solitons propagate without spreading because of exact balance between group velocity dispersion (GVD) and self-phase modulation (SPM).
  \[
  i \frac{\partial U}{\partial \xi} = \text{sgn}(\beta_2) \frac{1}{2} \frac{\partial^2 U}{\partial \tau^2} - N^2 |U|^2 U
  \]

- Even the small attenuation (0.2 dB/km) of communications fibers can upset this local balance and lead to pulse spreading.

- Solution is to use a tapered fiber (15% in 40 km) so that the GVD decreases at the same rate as the pulse energy.
Chaos in Sodium Vapor

Temporal Evolution

\[ P_b = 24 \text{ mW} \]

Phase Space Trajectories

\[ t = 55 \text{ nsec} \]

\[ P_{\perp}(t) \]

\[ P_{\parallel}(t) \]

\[ P_{\parallel}(t) \]

\[ P_{\perp}(t) \]

\[ \tau = 100 \text{ nsec} \]

\[ P_{\perp}(t) \]

\[ P_{\parallel}(t) \]

\[ \tau = 50 \text{ nsec} \]

\[ P_{\perp}(t) \]

\[ P_{\parallel}(t) \]

\[ P_{\perp}(t) \]

\[ P_{\parallel}(t) \]

PRL 58, 2432 (1987); 61, 1827 (1988); 64 1721 (1990).
Laser Beam Filamentation

Spatial growth of wavefront perturbations

Fig. 17.2 Image of small-scale filaments at the exit windows of a CS$_2$ cell created by self-focusing of a multimode laser beam. [After S. C. Abbi and H. Mahr, Phys. Rev. Lett. 26, 604 (1971).]
Honey Comb Pattern Formation

Output from cell with single gaussian beam input

Exiting beam  Far field pattern

Quantum image?

Input power 150 mW
Input beam diameter 0.22 mm
$\lambda = 588.995$ nm

Sodium vapor cell
$T = 220^\circ$ C
Bennink et al., PRL 88, 113901 2002.
A sodium vapor may be thought of as a medium composed of two-level atoms. Light whose frequency is near the atomic transition frequency experiences a refractive index $n$ which depends strongly on the intensity $I$:

$$n = 1 + n_f \frac{I}{I_{\text{sat}}}$$

Since light refracts in the direction of increasing index, in a medium with negative saturable nonlinearity it refracts toward regions of higher intensity. This causes smooth beams to narrow or self-focus. But it also tends to destabilize a beam as small amplitude fluctuations grow due to local self-focusing. Thus beams with even small amplitude noise can spontaneously split into two or more separate beams.

For sodium at 200ºC, $c_0 \approx -0.05$ and $I_{\text{sat}} \approx 6 \text{ mW/cm}^2$.

A simulation of spontaneous break-up into 3 stable beams:

Experimental observation of spontaneous break-up resulting in a striking far-field pattern:

Pictures taken by R. Bennink, S. Lukisbova, and V. Wong.
Experiment in Self Assembly

Joe Davis, MIT