Nanostructured Materials and Devices for Photonics

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Prospectus

Introduction to Nonlinear Optics

Development of New NLO Materials

Generation of Quantum States of Light

Some Underlying Issues in Nonlinear Optics

Light-by-Light Scattering



Phase Conjugation by Degenerate Four-Wave Mixing



What is Nonlinear Optics ?

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

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

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

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

$$W = 2w$$

$$\chi^{(3)}: \text{ third-order effects, eg}$$

$$\text{ four-wave mixing}$$

$$\text{ Intensity -dependent}$$

$$\text{ refractive index}$$

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

$$N_{2} = \frac{12\pi^{2}}{N_{0}^{2}C} \chi^{(3)}$$

.

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

• Bruggeman (interdispersed)





• Fractal Structure



• Layered



scale size of inhomogeneity << 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.

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.

Z-Scan Measurement of X(3)







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.



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



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



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.

"Slow" Light in Nanostructured Devices Robert W. Boyd with John Heebner, Nick Lepeshkin, Aaron Schweinsberg, and Q-Han Park

The Institute of Optics, University of Rochester, Rochester, NY 14627

Nanofabrication

- Materials (artificial materials)
- Devices

(distinction?)

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



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

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.



Microdisk Resonator Design



J. E. Heebner and R. W. Boyd

Photonic Device Fabrication Procedure



RWB - 10/4/01

Nonlinear Optical Loop-De-Loop



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

Disk Resonator and Optical Waveguide in PMMA Resist



AFM

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r



Photonic Devices in GaAs/AlGaAs









Generation of Quantum States of Light by Use of Electromagetically Induced Transparency

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

- Quantum states of light useful for applications including precision measurements and secure communications
- EIT enables the efficient creation of quantum states of light by eliminating spontaneous emission background noise.





Harris, Field and Imamoglu, PRL 64 1107

1 1107 1990

Application of EIT to Squeezed-Light Generation

• Squeezing by self-phase modulation



Blow, Loudon, and Phoenix, J. Mod. Opt., 40, 2515, 1993

• EIT allows phase shifts large enough to produce significant squeezing, and prevents signal-beam absorption which can degrade the squeezing.

Source of Polarized, Single-Photons on Demand

- Useful for secure communication by quantum cryptgraphy
- 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



Incident unpolarized light

Experimental procedure Implementation with S. Lukishova



Single-molecule fluorescence

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.

R. W. Boyd and M. O. Scully, Appl. Phys. Lett. 77, 3559, 2000.

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).

$$i\frac{\partial U}{\partial \xi} = \operatorname{sgn}(\beta_2)\frac{1}{2}\frac{\partial^2 U}{\partial \tau^2} - N^2|U|^2 U$$

- 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



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



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.

Experiment in Self Assembly



Joe Davis, MIT

Experimental Configuration



Conical Emission Patterns

Single input beam

Two input beams (equal intensity) (parallel polarization) Two input beams (unequal intensity) (parallel polarization)







Two cones formed, each centered on other beam.

Only stronger input beam can act as pump for cone generation.

Generated in carbon disulfide

Honey Comb Pattern Formation

Output from cell with single gaussian beam input



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.

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



Kauranen et al, Opt. Lett. 16, 943, 1991; Kauranen and Boyd, Phys. Rev. A, 47, 4297, 1993.

Efficient Far IR and THz Imaging by use of EIT R W. Boyd and M. O. Scully

- EIT concepts allow "upconversion" of IR images to the visible with high quantum efficiency (approaching unit efficiency).
- Upconversion is a "noise-free" process; only noise in output is (quantum) noise of IR signal.
- Technique holds promise of unprecedented sensitivity of FIR and THz detection (detection of single THz quanta)!
- Applications include FIR astronomy and THz imaging of biological tissue.
- Pitfall: very narrow spectral acceptance bandwidth.