New Materials and Interactions for Nonlinear Optics

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Presented at Naval Air Systems Command, June 26, 2002

Prospectus

Introduction to Nonlinear Optics

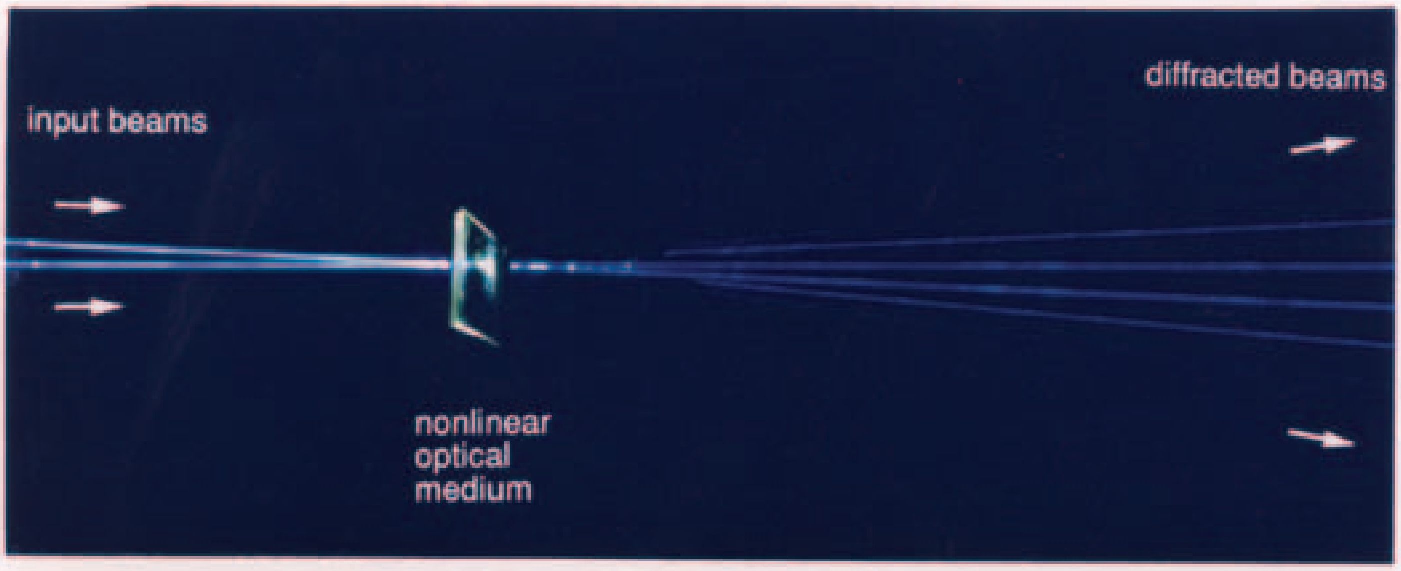
Development of New NLO Materials

EIT Techniques for Squeezed Light Generation

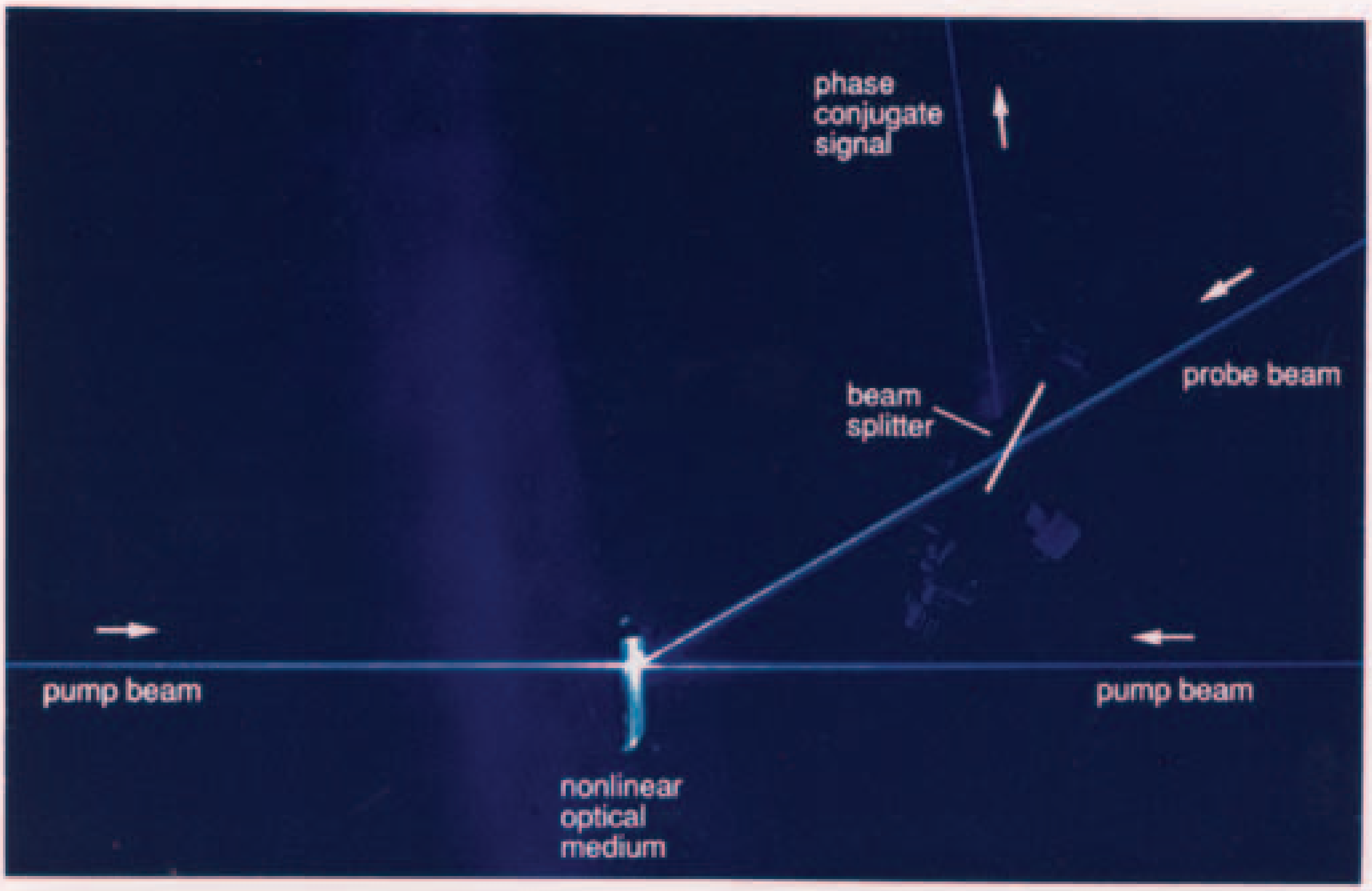
Some Underlying Issues in Nonlinear Optics

Nonlinear Optical Interactions

Light-by-Light Scattering



Phase Conjugation by Degenerate Four-Wave Mixing



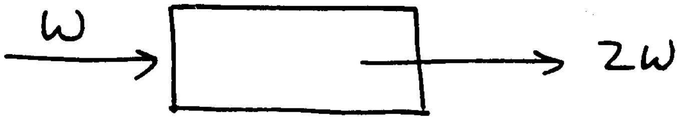
What is Nonlinear Optics?

$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots$$

dipole moment per unit volume

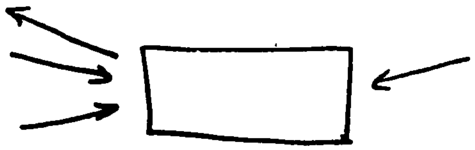
$\chi^{(1)}$: linear optics, eg 

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



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

four-wave mixing



Intensity-dependent refractive index

$$n = n_0 + n_2 I$$

$$n_2 = \frac{12\pi^2}{n_0^2 c} \chi^{(3)}$$

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.

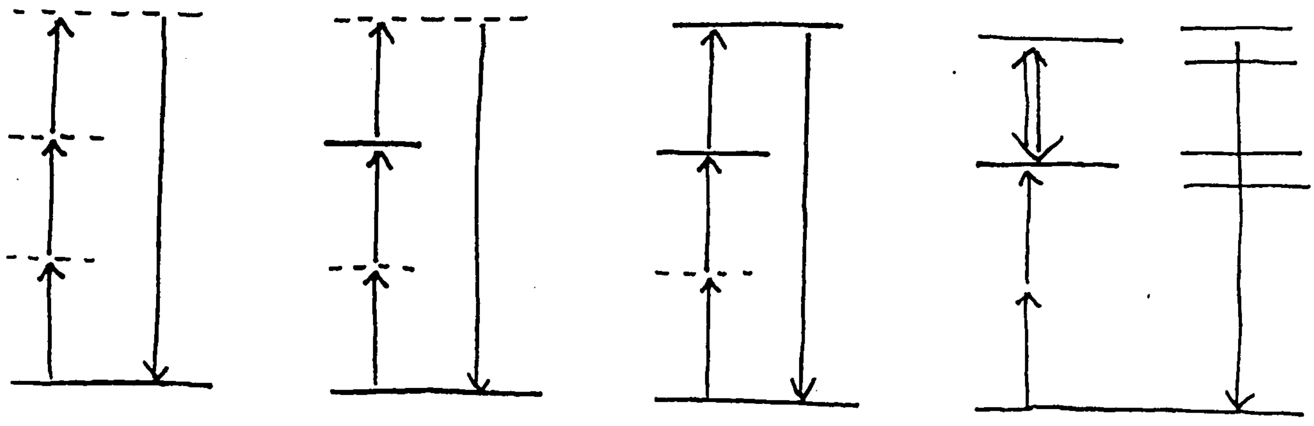
Approaches to the Development of Improved NLO Materials

- 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!

Electromagnetically Induced Transparency



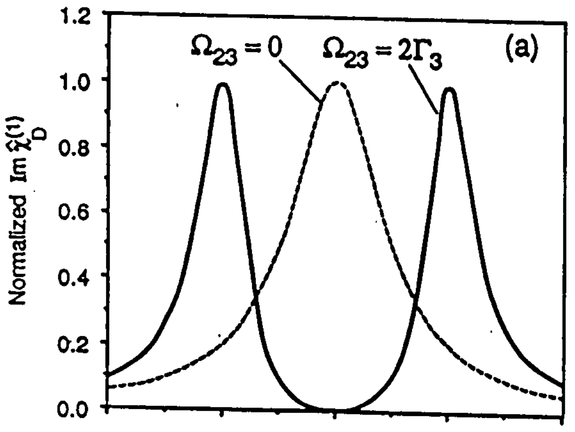
too weak

better (2-photon resonance)

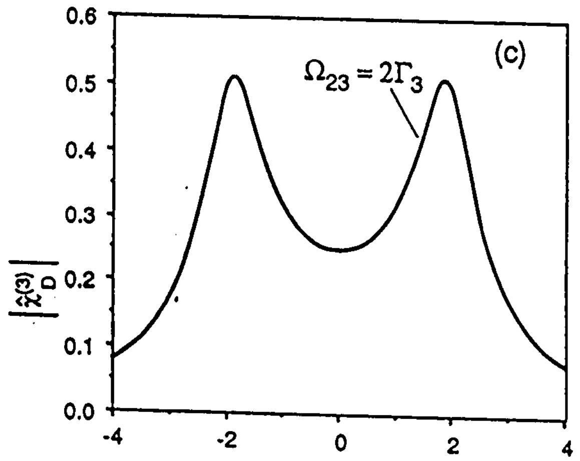
still better (but absorption!)

The EIT concept

- EIT Predictions



Absorption of the generated field vanishes.

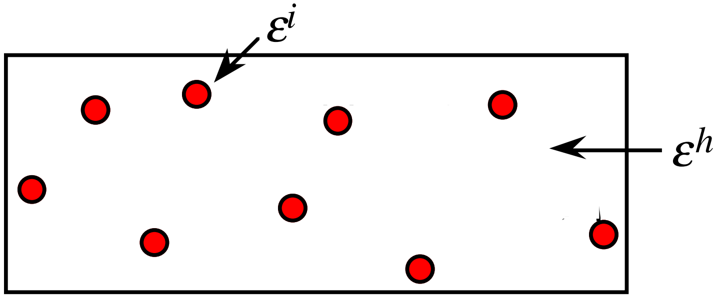


But the nonlinearity remains large!

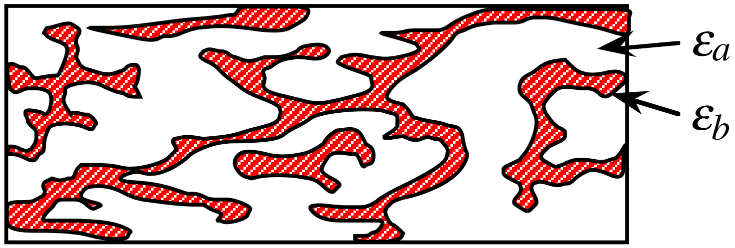
Harris, Field and Imamoglu, PRL 64 1107 1990

Nanocomposite Materials for Nonlinear Optics

- Maxwell Garnett

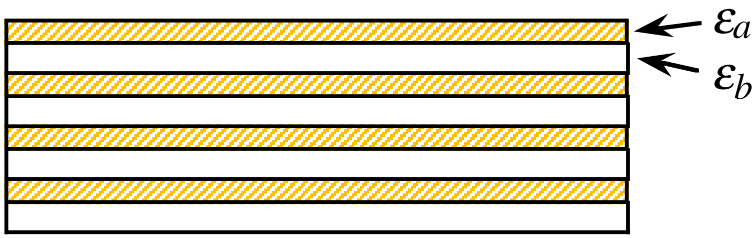


- Bruggeman (interdispersed)



- Fractal Structure

- Layered



scale size of inhomogeneity << 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 the constituents.*

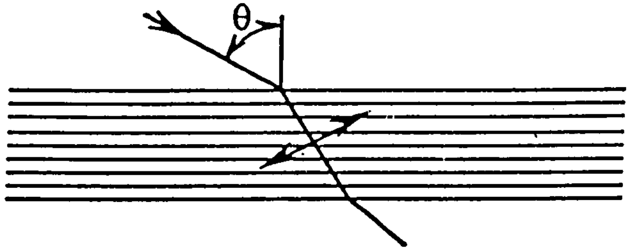
- Enhancement of $\chi^{(3)}$ can be understood in terms of local field effects

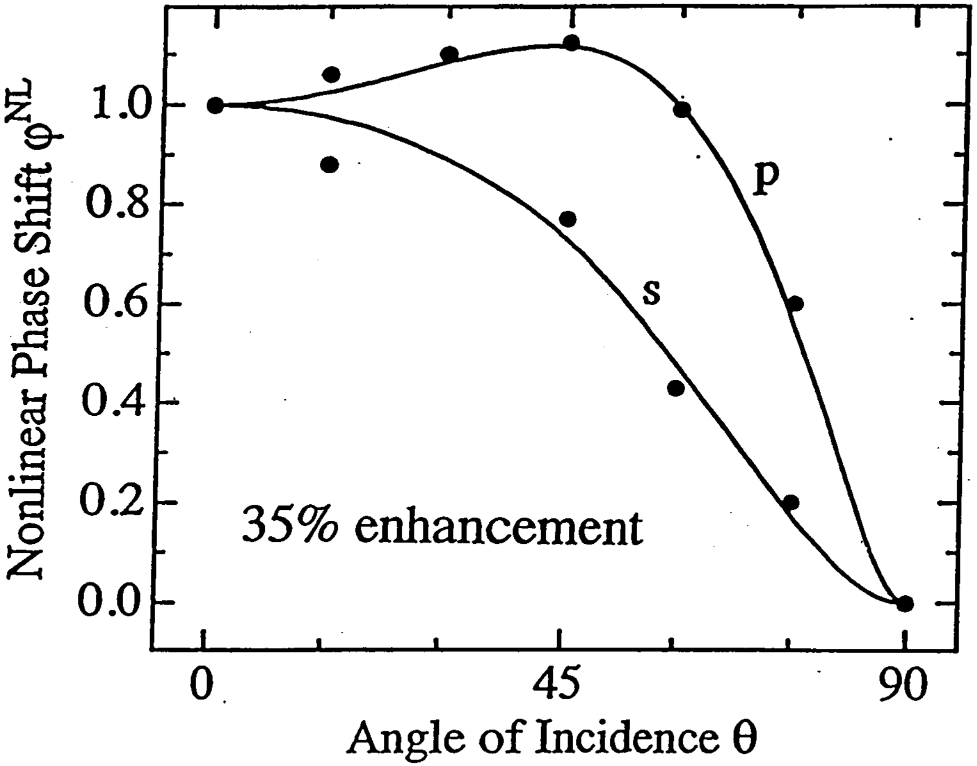
* RWB and J. E. Sipe, US patent #5,253,103; PRL 74, 1871, 1995.

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.





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 $\chi^{(3)}$

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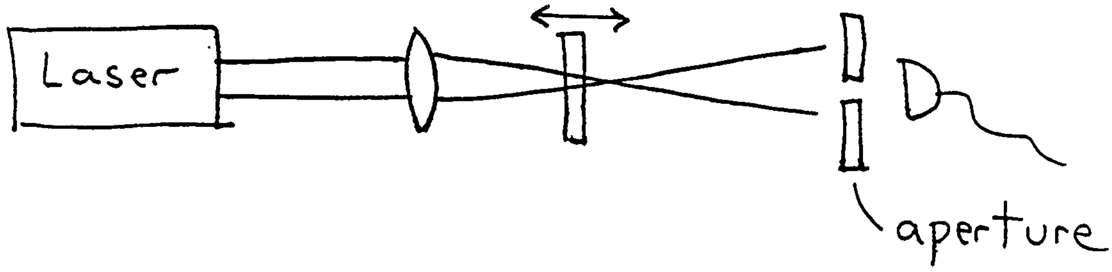
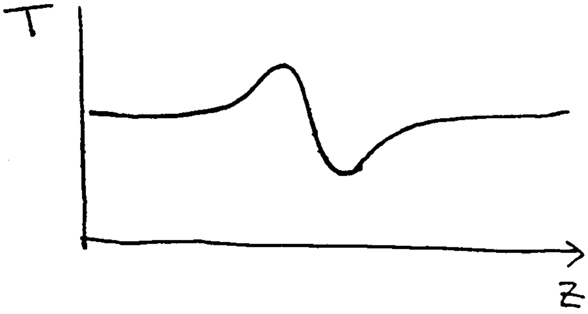


Measures NL change in refraction

($\mathrm{Re}\,\chi^{(3)}$)



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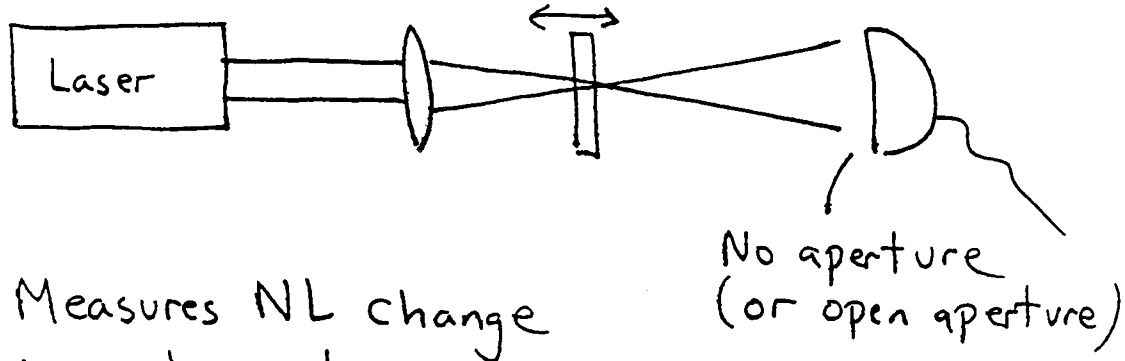
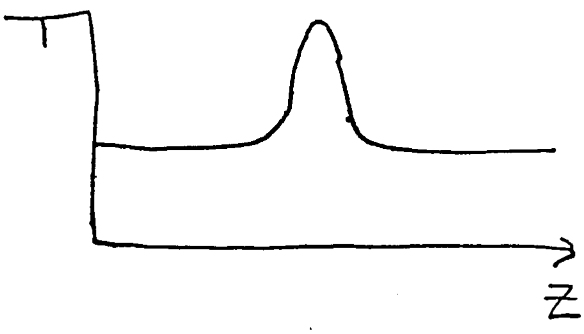


Measures NL change in absorption.

($\mathrm{Im}\,\chi^{(3)}$)



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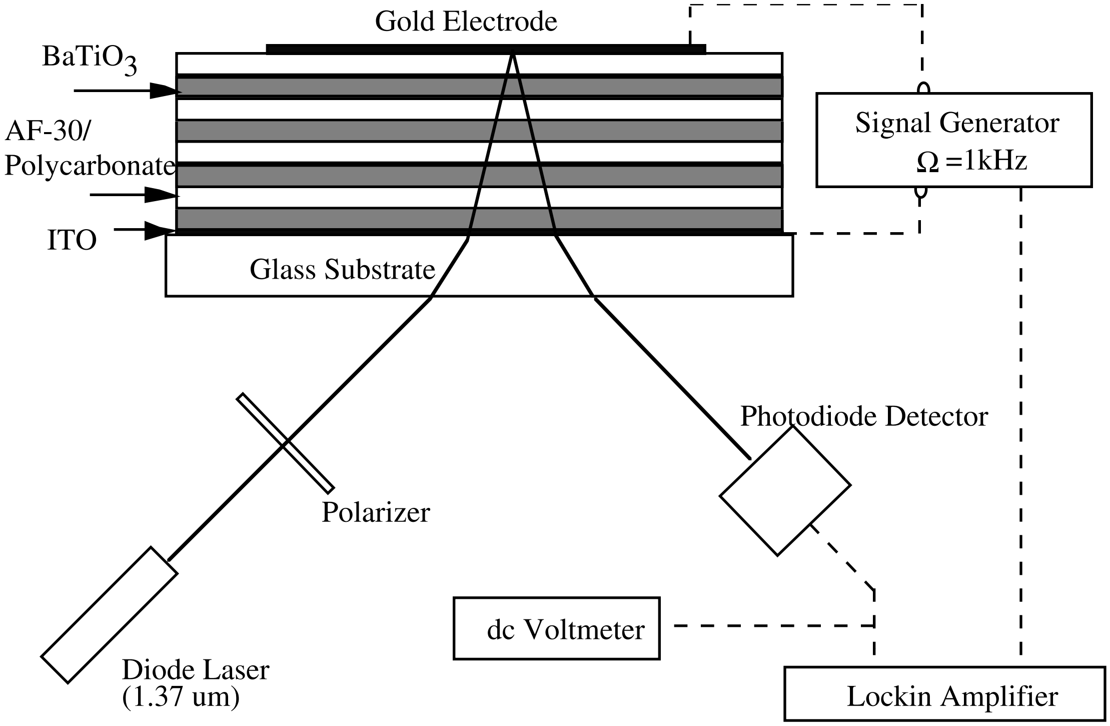
Sheik Bahae, van Stryland, et al.

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 ε(dc) = 2.9

- barium titante (rf sputtered)

  n=1.98 ε(dc) = 15

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

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

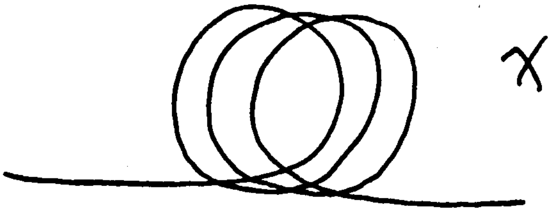
3.2 times enhancement in agreement with theory

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

TWO GREAT IRONIES OF NONLINEAR OPTICS

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

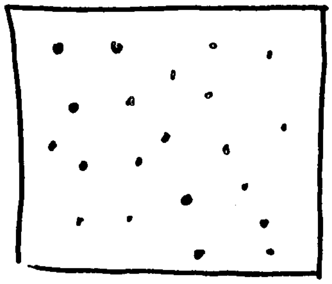
Fiber NLO



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

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

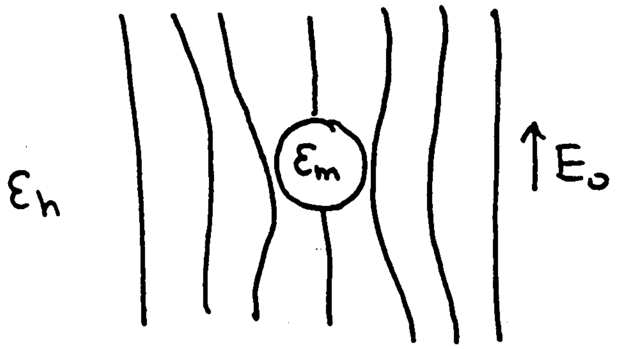
dilute colloid →



$\chi^{(3)}_{\text{silver}} \simeq 10^{-8}$ esu

Metal / Dielectric Composites

Very large local field effects



$$E_{in} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} E_0$$

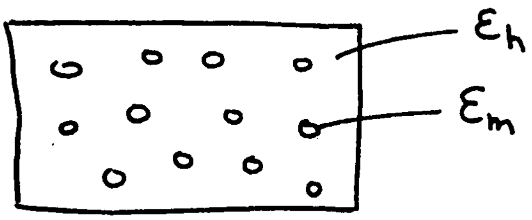
$$\equiv \mathcal{L} E_0$$

(ε_m is negative !)

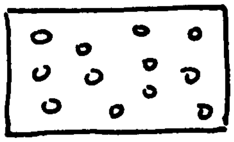
At resonance

$$\mathcal{L} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} \longrightarrow \frac{3\varepsilon_h}{i\varepsilon_m''} \approx (3 \text{ to } 30)\, i$$

$$\chi^{(3)}_{eff} = f\, \mathcal{L}^2 |\mathcal{L}|^2 \chi^{(3)}_m + (1-f)\, \chi^{(3)}_h$$



Counterintuitive Consequence of Local Field Effects



gold nanoparticles in a liquid dye solution (HITCl)

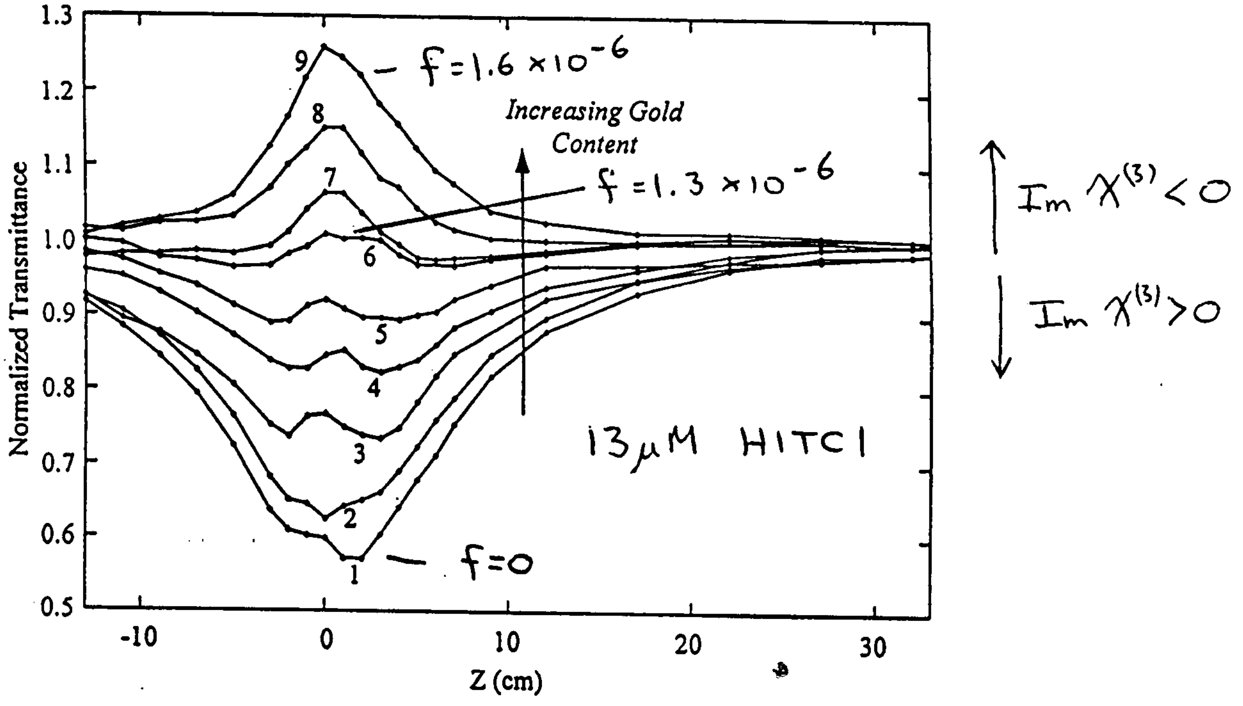
Both constituents are reverse saturable absorbers $\Rightarrow$ $\text{Im}\,\chi^{(3)} > 0$

Effective NL susceptibility of composite

$$\chi^{(3)}_{eff} = f\,\mathcal{L}^2|\mathcal{L}|^2\chi^{(3)}_{Au} + (1-f)\,\chi^{(3)}_{\text{dye sol'n}}$$

$$\mathcal{L} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} = \text{pure imaginary at resonance!}$$

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

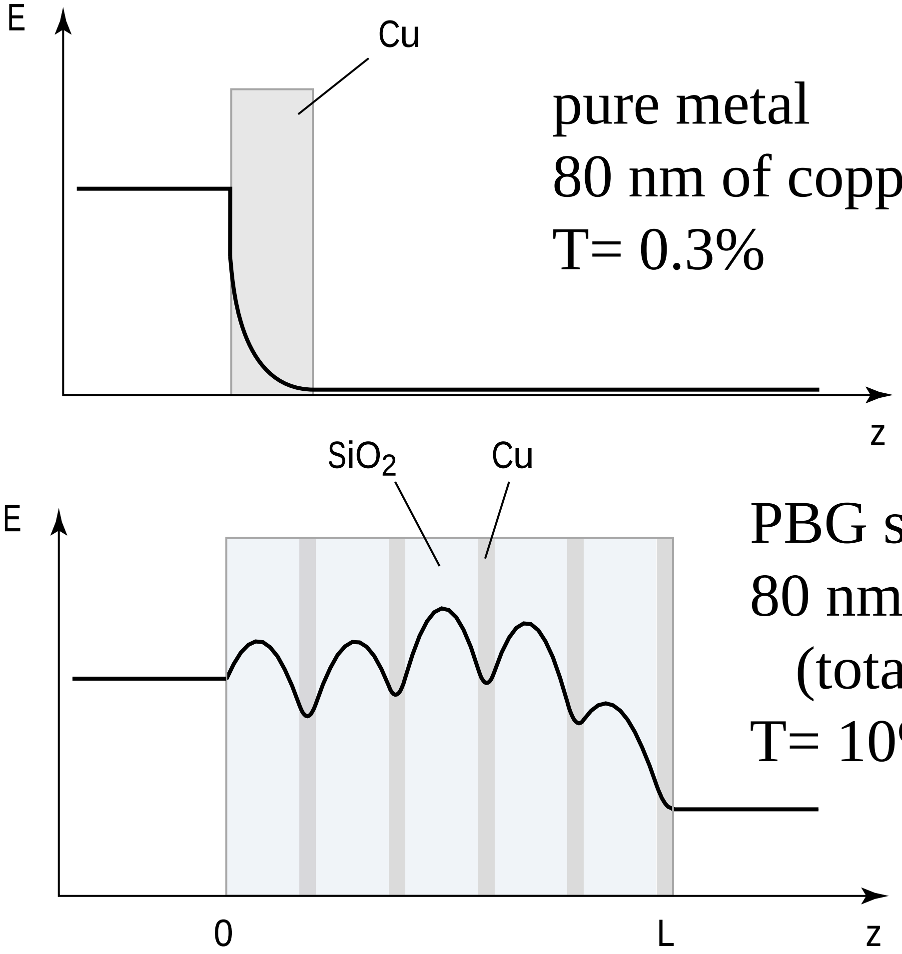


Smith et al, JOSA B 14 1625 1997.

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)



pure metal
80 nm of copper
T= 0.3%

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

- 40 times enhancement 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

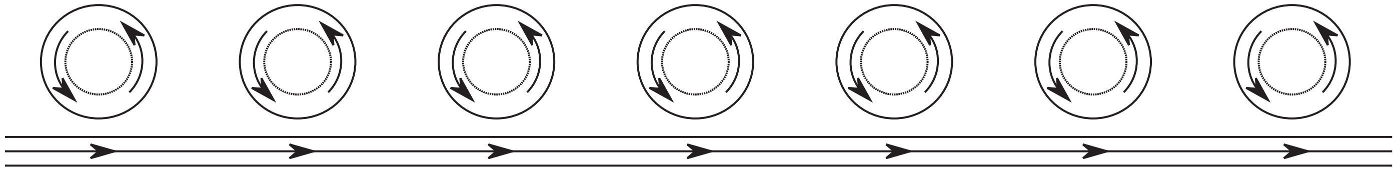
Nanofabrication

- Materials (artificial materials)
- Devices

(distinction?)

NLO of SCISSOR Devices

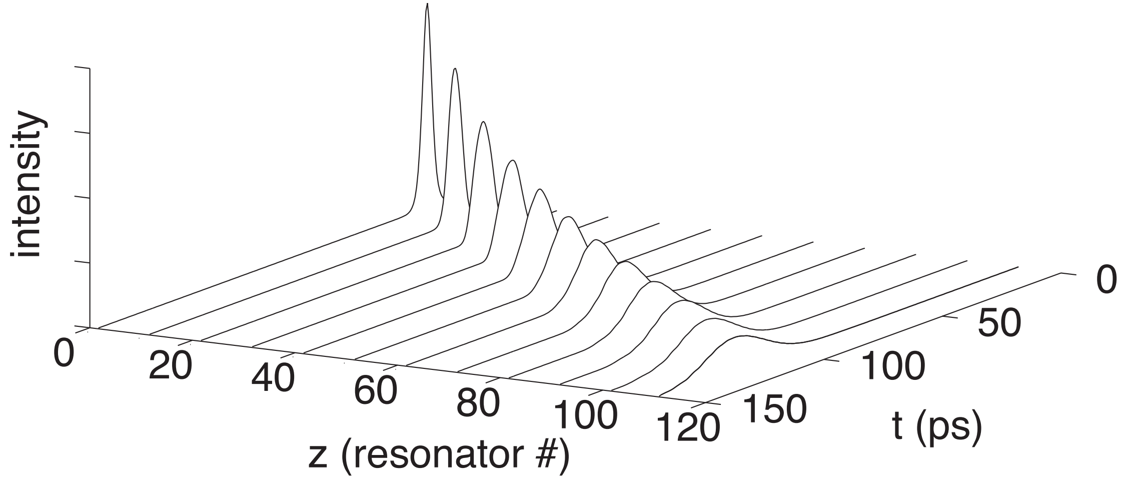
(Side-Coupled Integrated Spaced Sequence of Resonators)



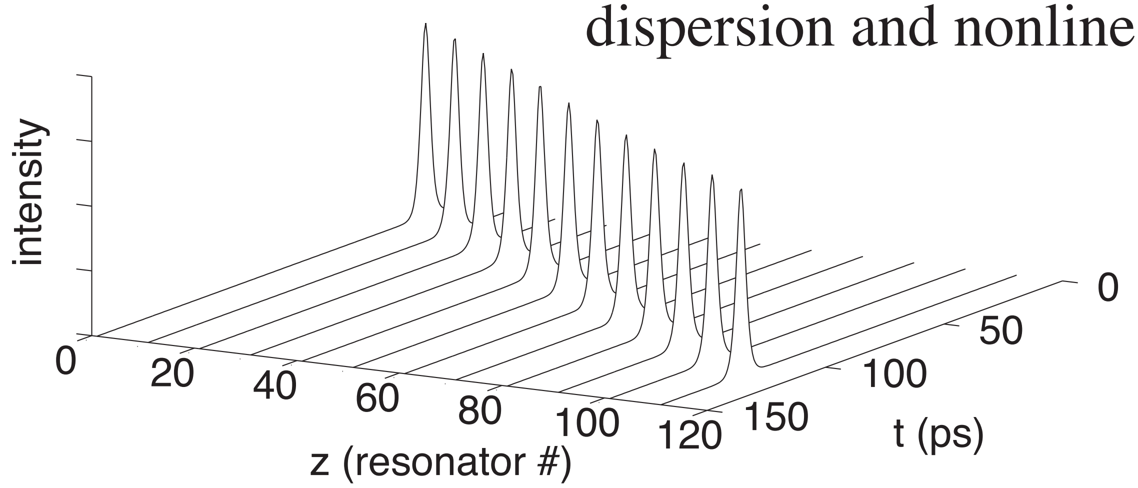
Shows slow-light, tailored dispersion, and enhanced nonlinearity

Optical solitons described by nonlinear Schrodinger equation

- Weak pulses spread because of dispersion

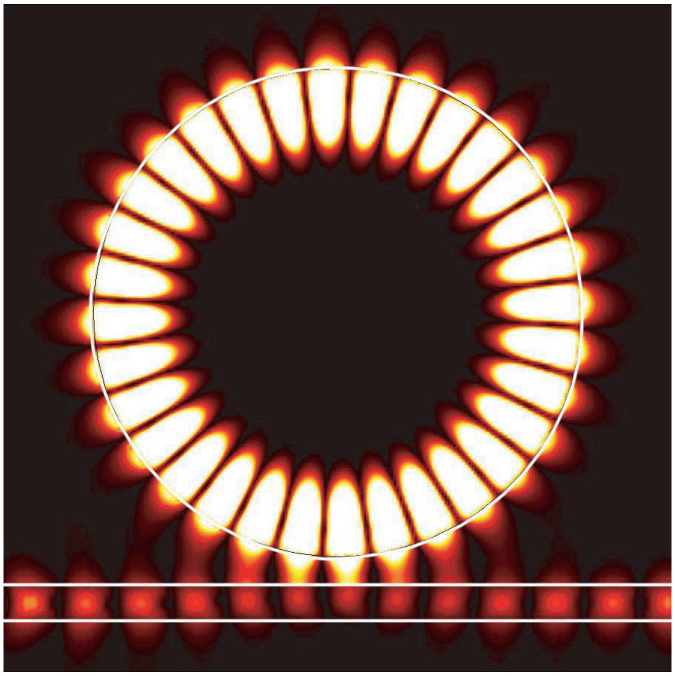


- But intense pulses form solitons through balance of dispersion and nonlinearity.

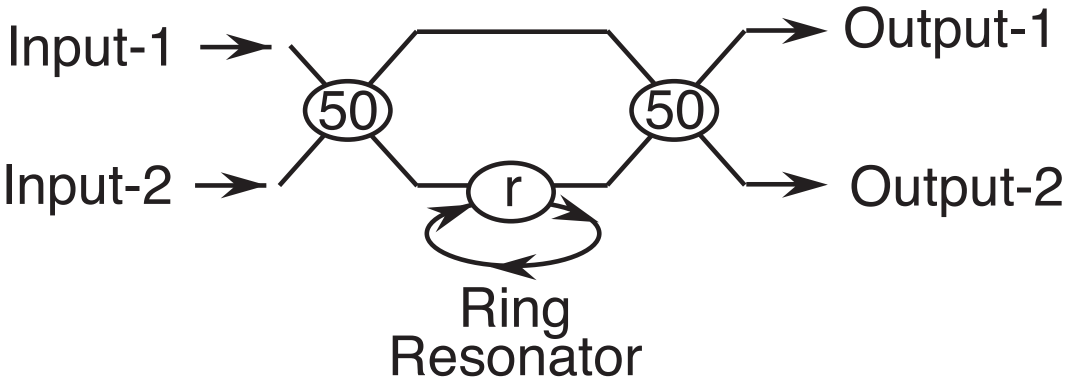


Ultrafast All-Optical Switch Based On Arsenic Triselenide Chalcogenide Glass

- 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 ≈ 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

Photonic Devices for Biosensing

Objective:

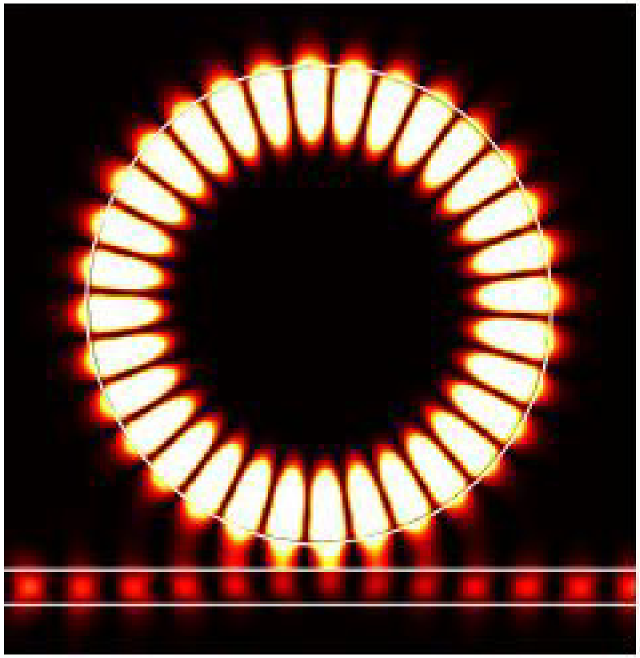
Obtain high sensitivity, high specificity detection of pathogens through optical resonance

Approach:

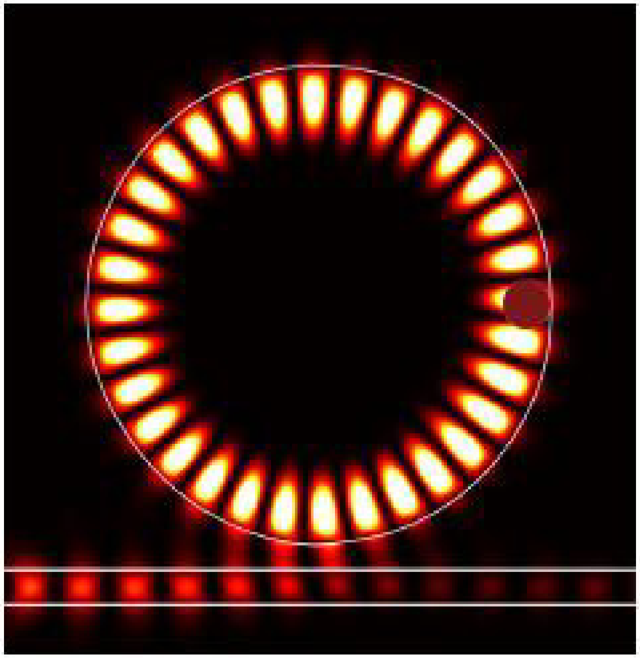
Utilize high-finesse whispering-gallery-mode disk resonator.

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

Simulation of device operation:



Intensity distribution in absense of absorber.

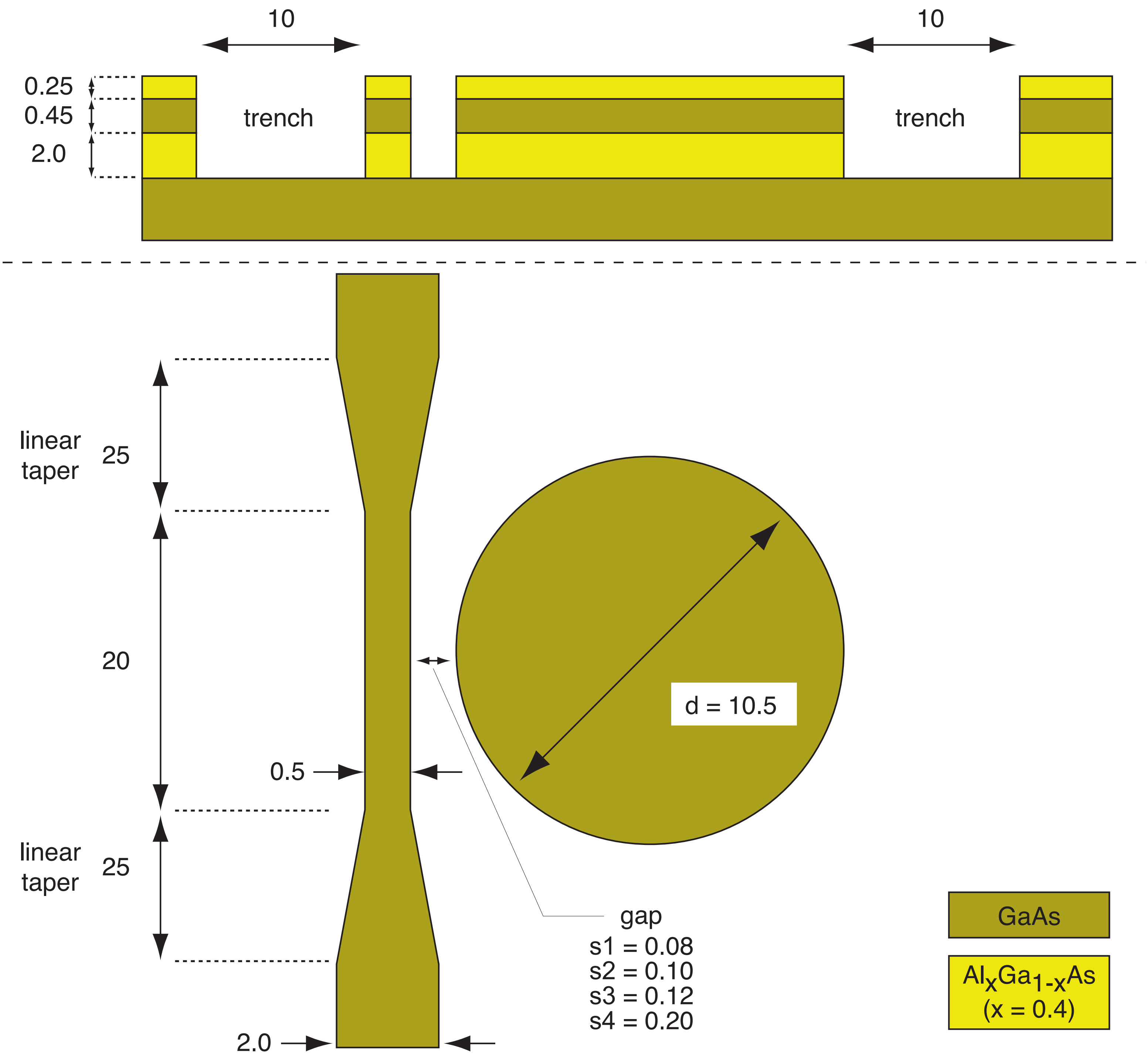


Intensity distribution in presence of absorber.

FDTD

Microdisk Resonator Design

(Not drawn to scale)
All dimensions in microns



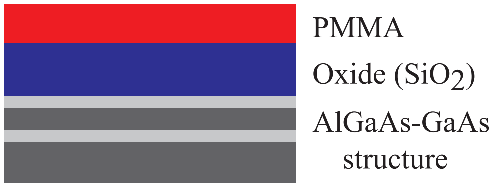
J. E. Heebner and R. W. Boyd

Photonic Device Fabrication Procedure

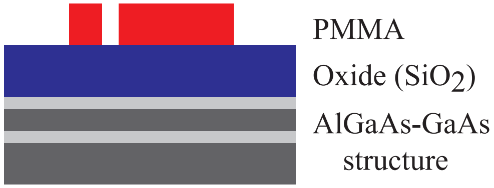
(1) MBE growth

(2) Deposit oxide

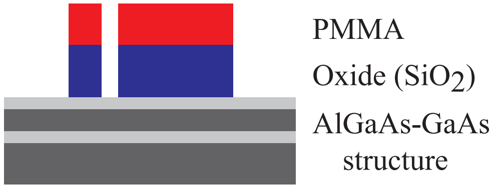
(3) Spin-coat e-beam resist



(4) Pattern inverse with e-beam & develop



(5) RIE etch oxide



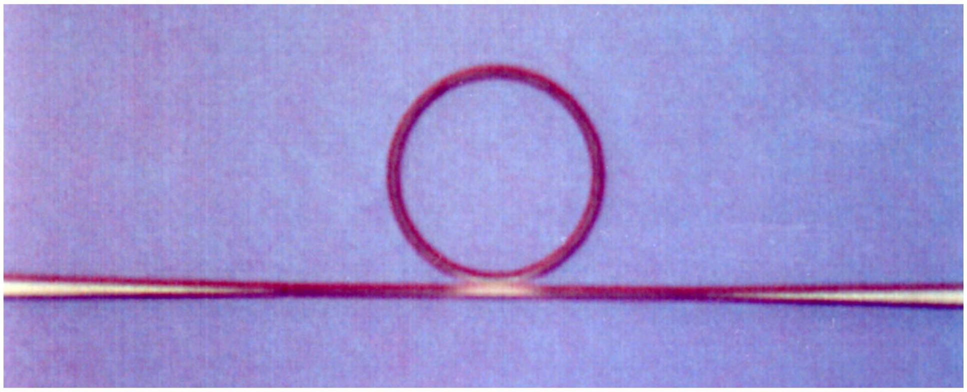
(6) Remove PMMA

(7) CAIBE etch AlGaAs-GaAs

(8) Strip oxide

RWB - 10/4/01

Nonlinear Optical Loop-De-Loop



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

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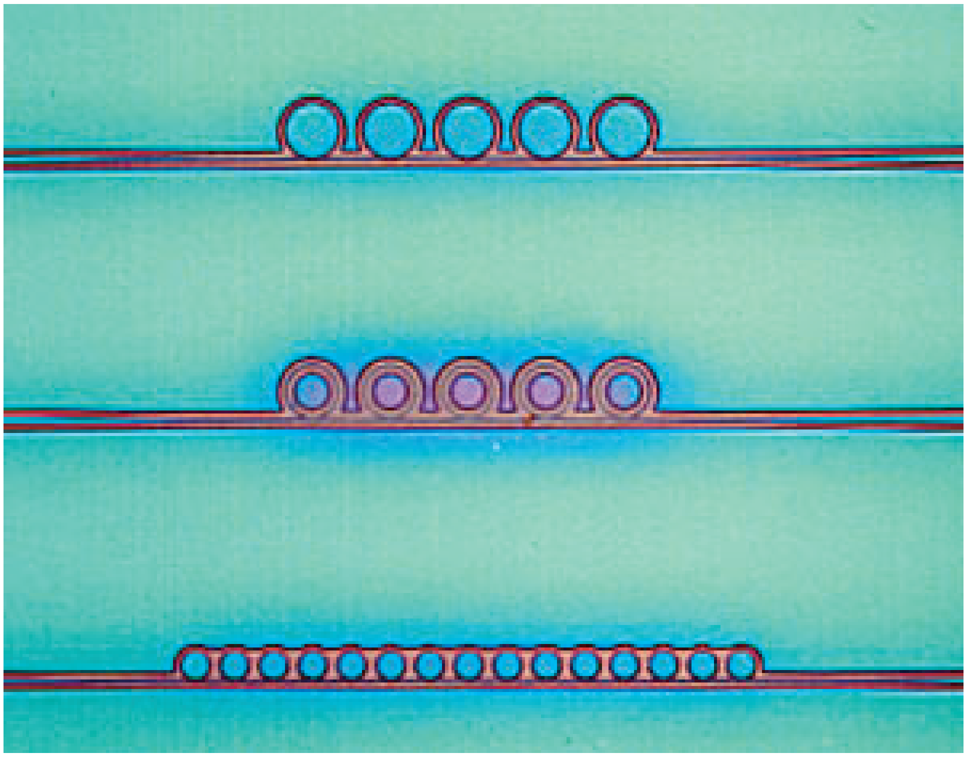
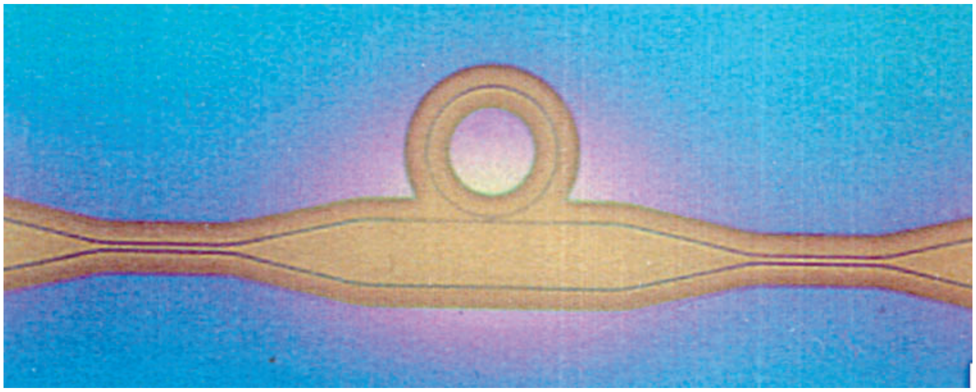
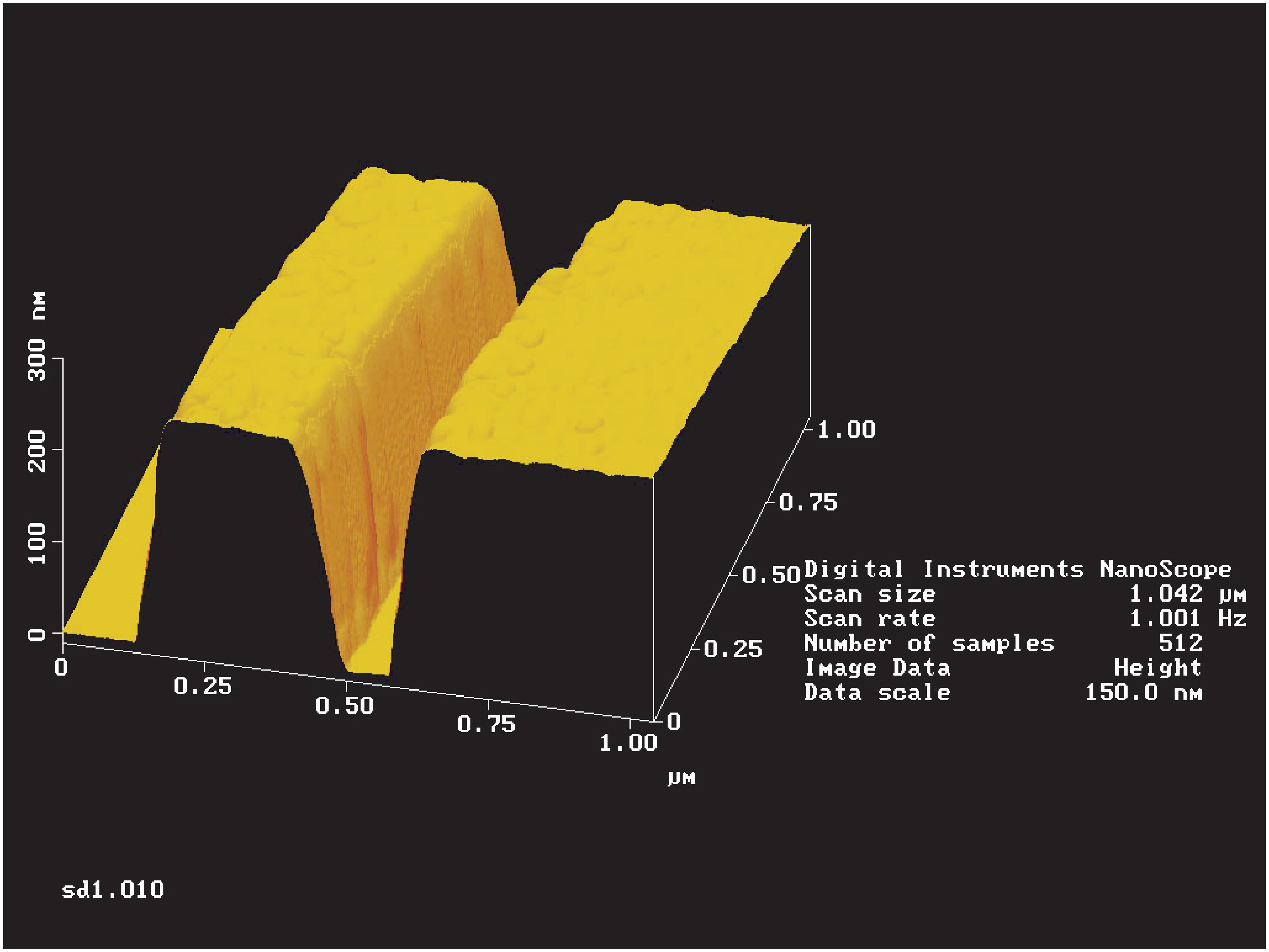




e i s



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Digital Instruments NanoScope
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Scan rate                1.001 Hz
Number of samples          512
Image Data              Height
Data scale             150.0 nM
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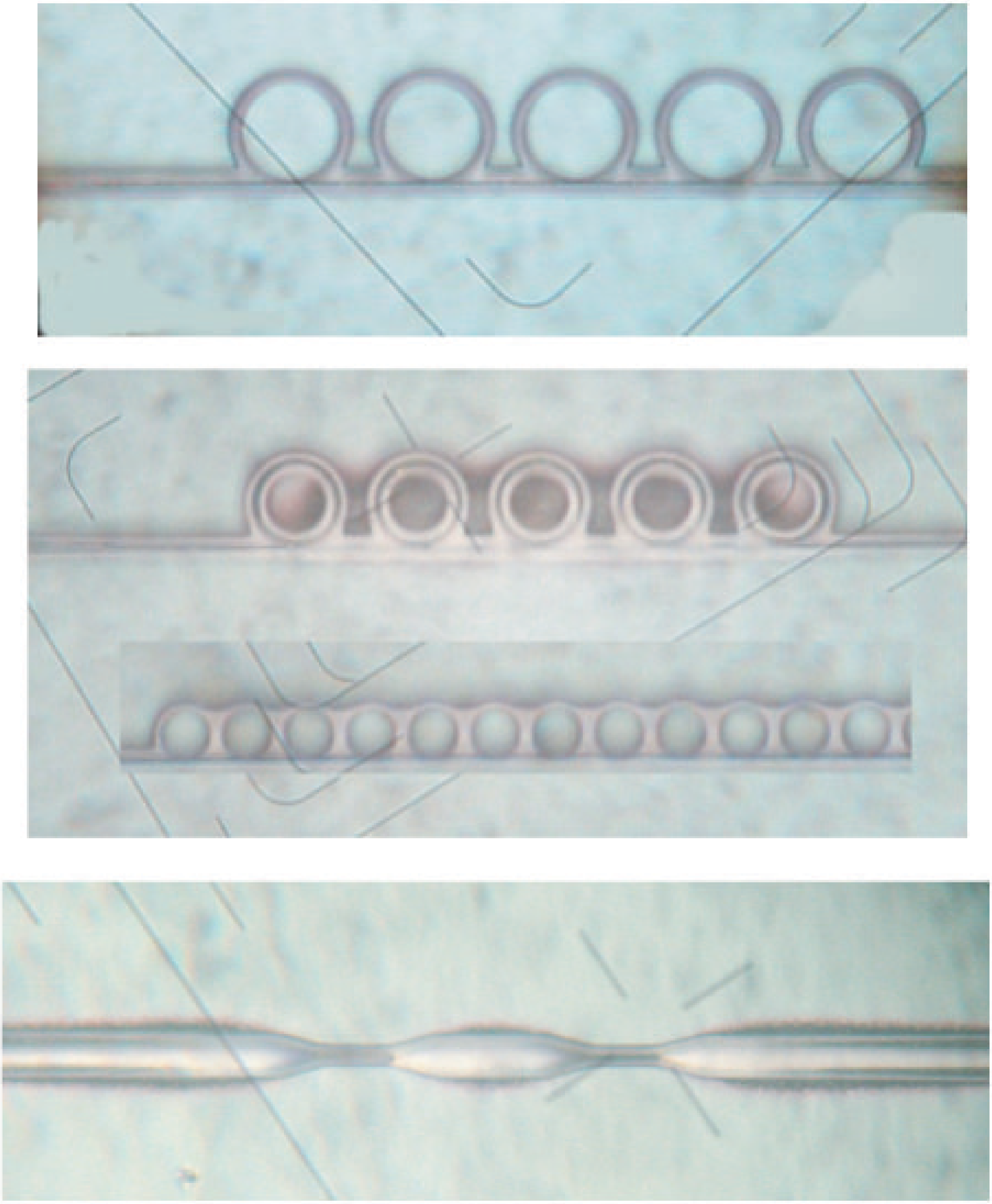
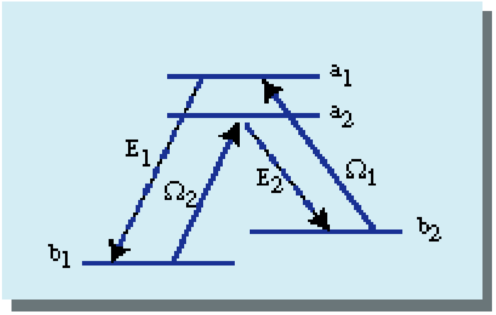
Photonic Devices in GaAs/AlGaAs

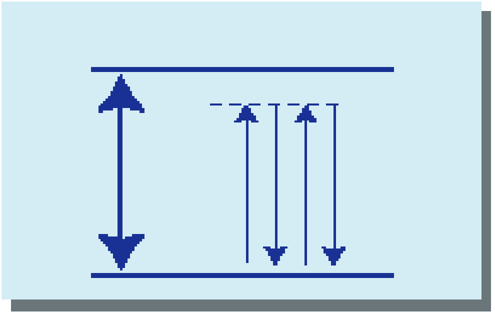


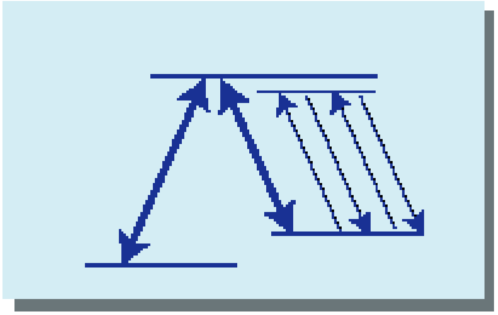
Generation of Squeezed Light by use of EIT

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

Three Approaches





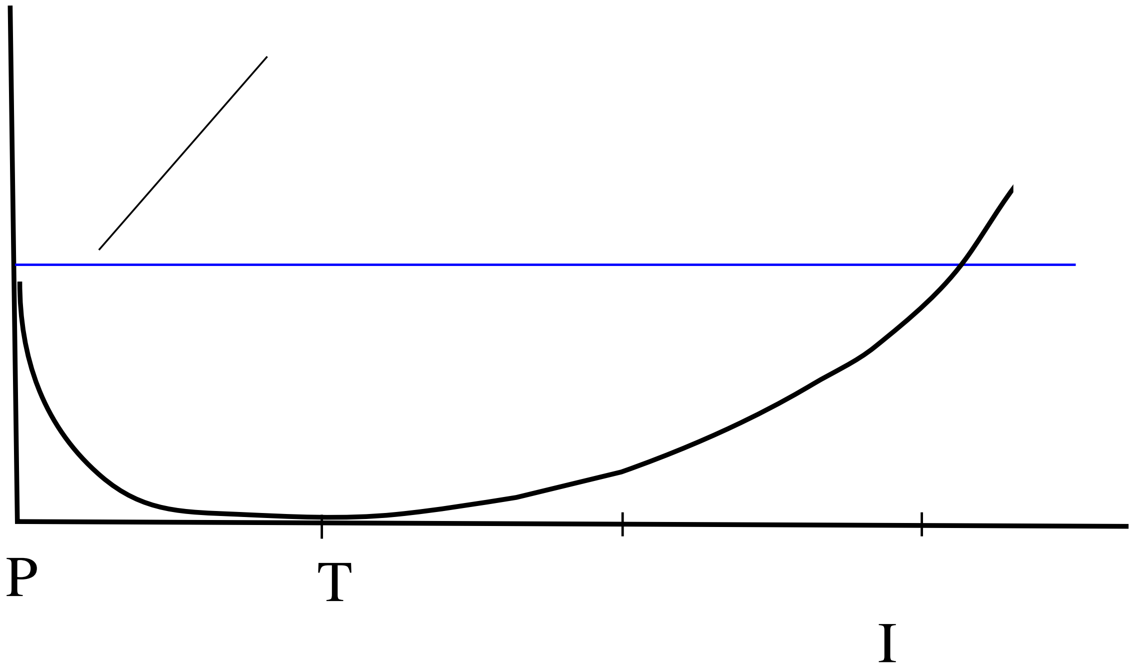


Fundamental idea: EIT eliminates linear absorption so that there is no spontaneous emission background noise.

a e it

s M g

s M

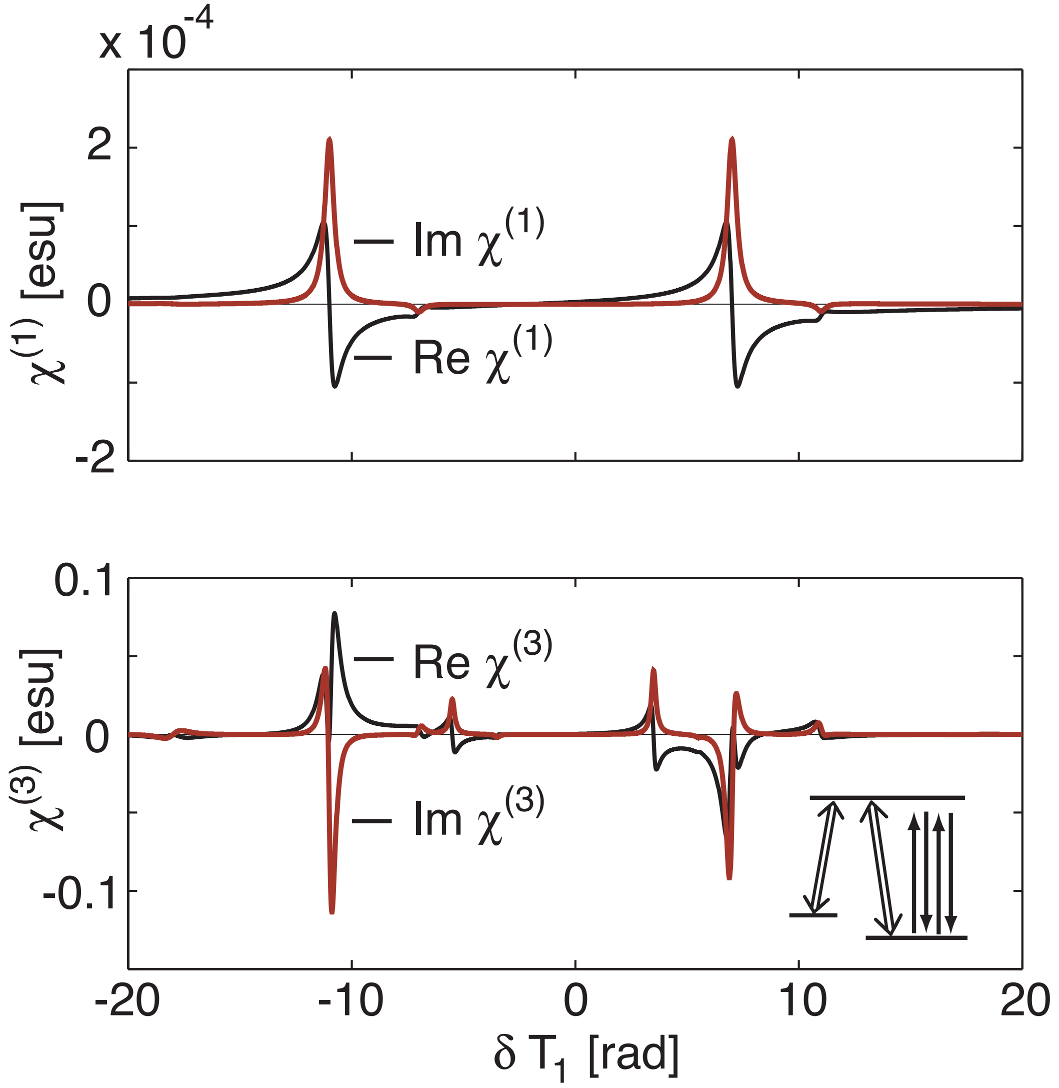


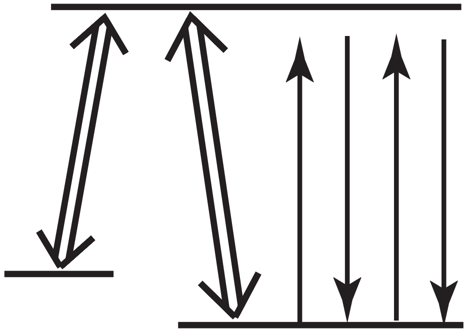
b Ll L p L m o LTPL L S

e it

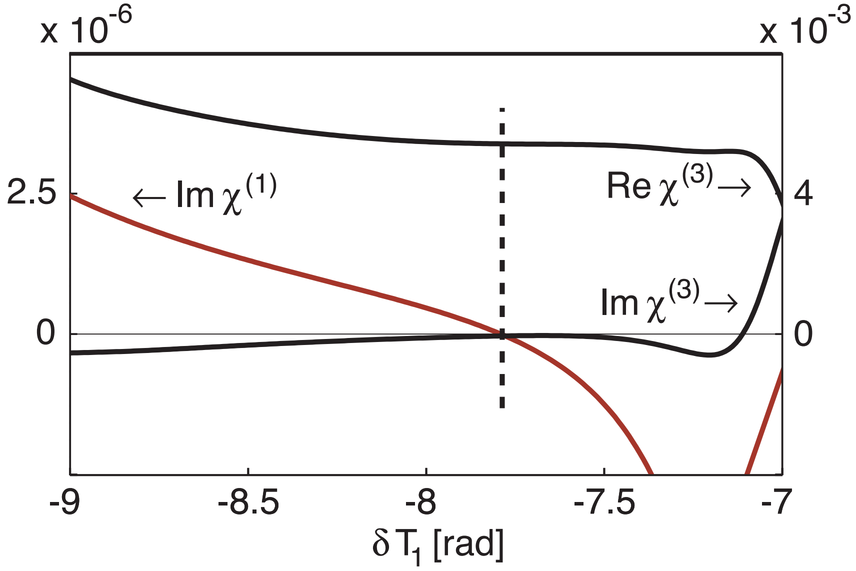
L M

Strong Absorption-Free Nonlinearity by Dark-State EIT



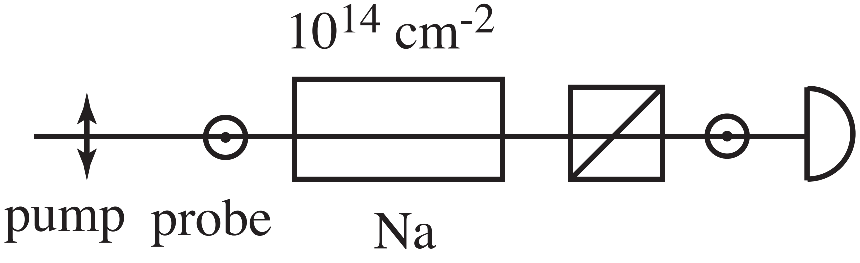


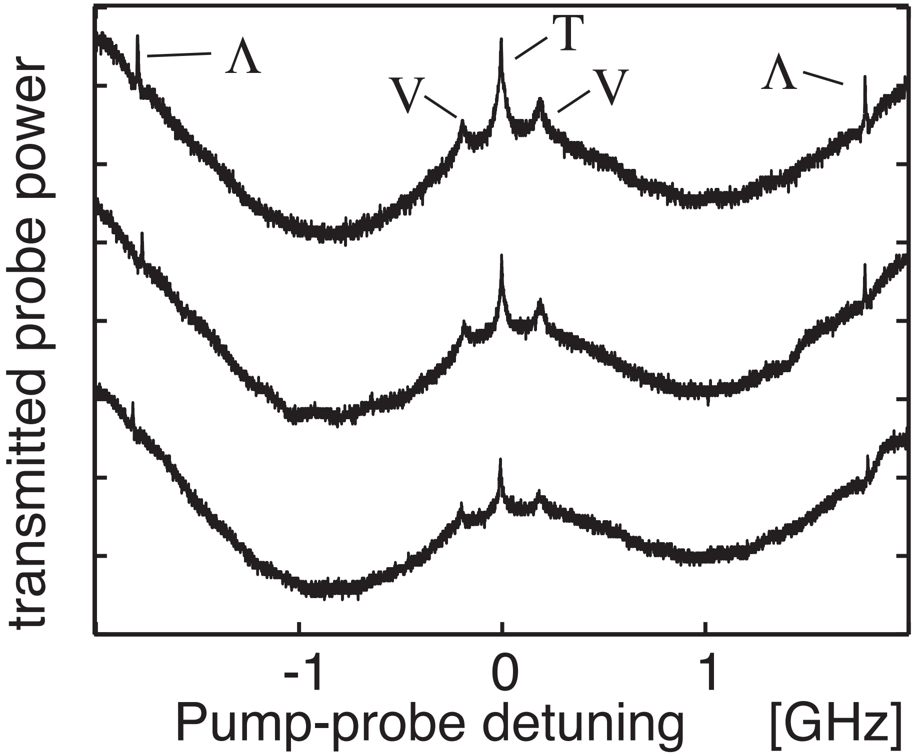
DETAIL:



Wong, Boyd, Stroud et al, Phys. Rev. A, 65, 013810-1 (2001)

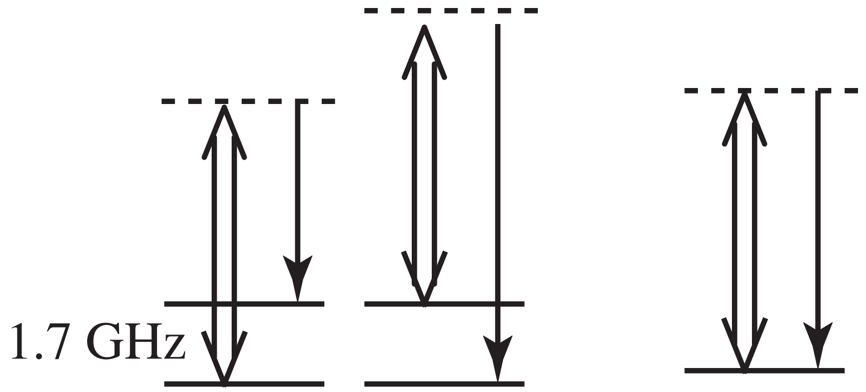
Saturation-Induced Extra Resonances



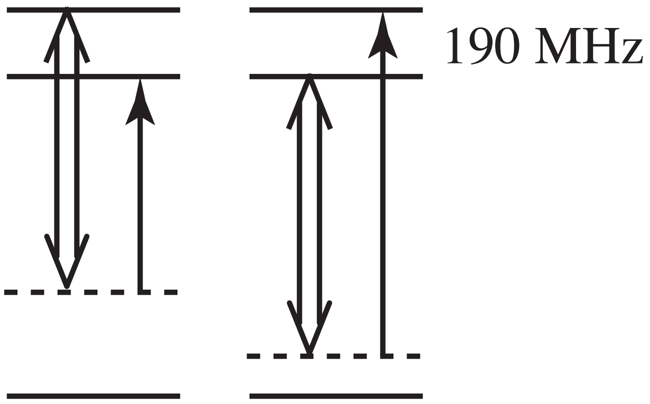


Dark-State (Λ)
Resonances

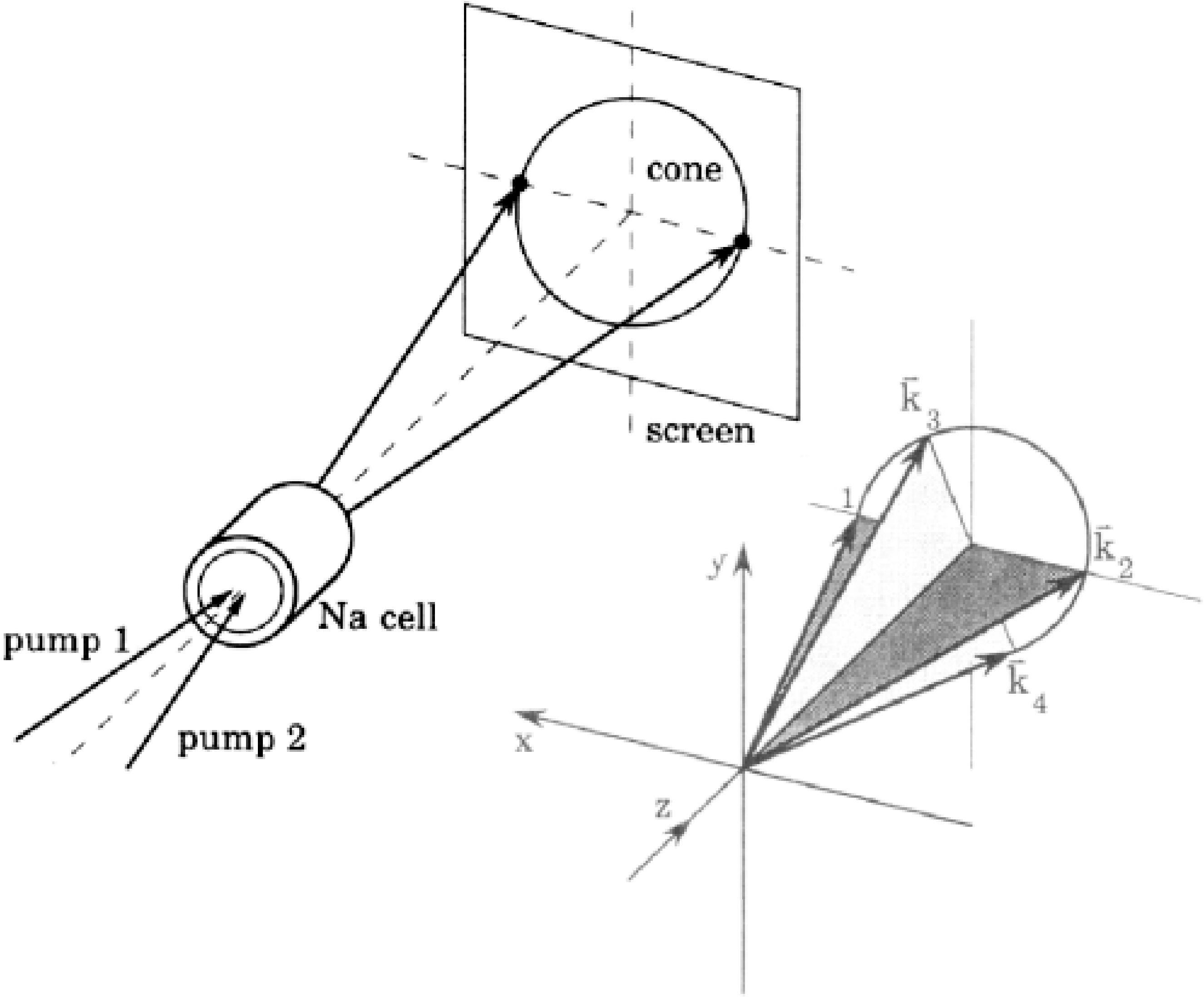
Two-Level Atom
Resonance

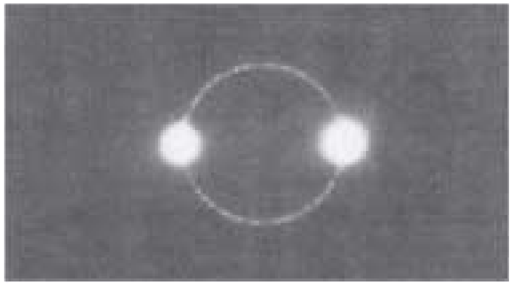


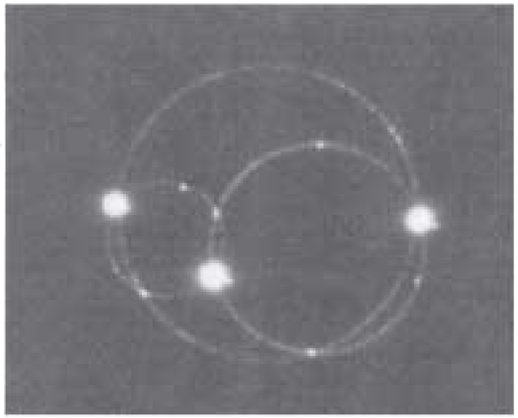
V-System Resonances
(saturation induced)



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

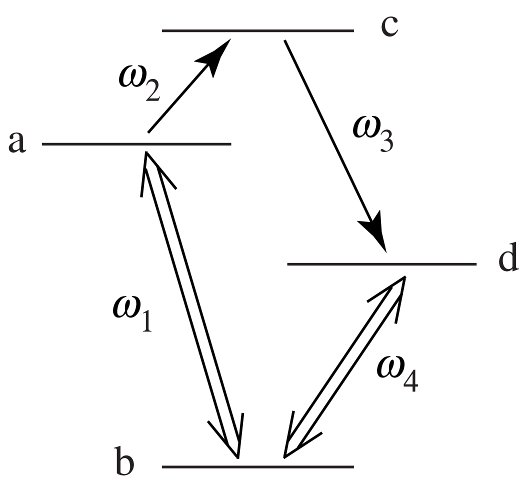




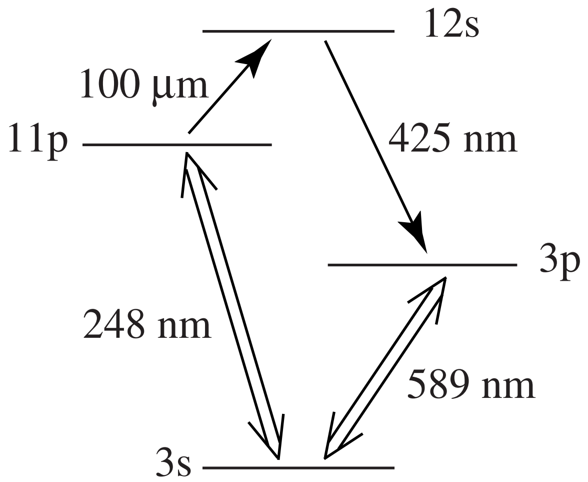


Kauranen et al, Opt. Lett. 16, 943, 1991; Kauranen and Boyd, Phys. Rev. A, 47, 4297, 1993.

Efficient Far IR and THz Imaging by use of EIT



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

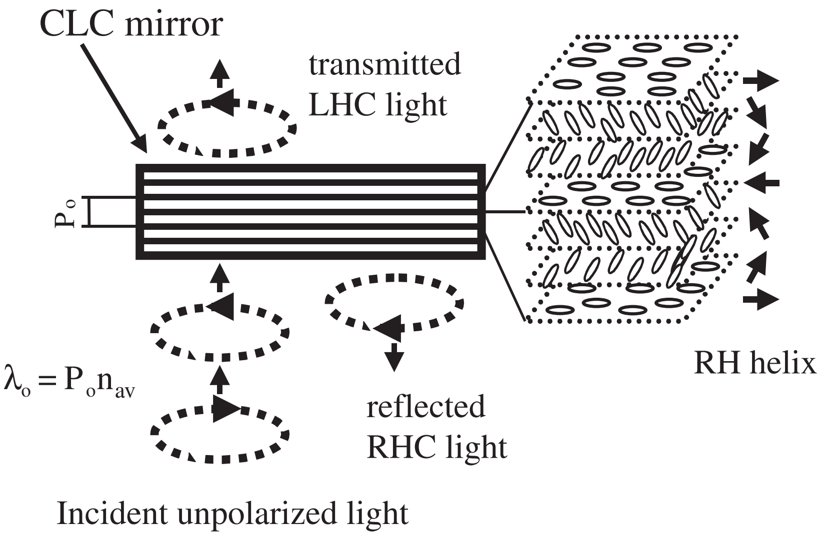


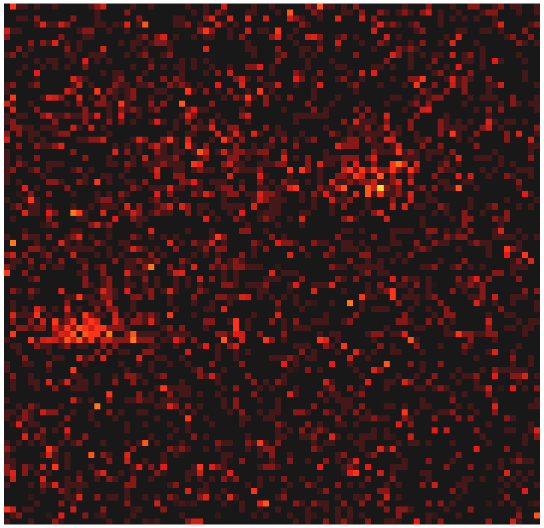
Sodium energy levels for the conversion of 100 micron radiation to the visible.

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

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





Experimental procedure

Single-molecule fluorescence

Implementation with S. Lukishova

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

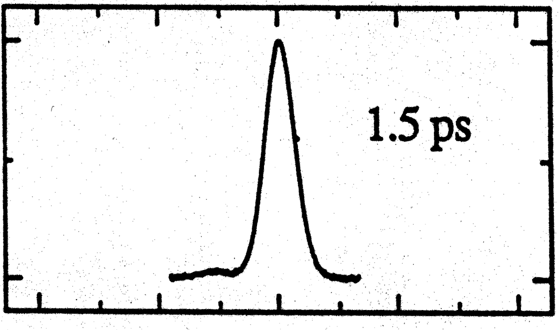
Andrew J. Stentz, Robert W. Boyd, University of Rochester
Alan F. Evans, Corning Inc.

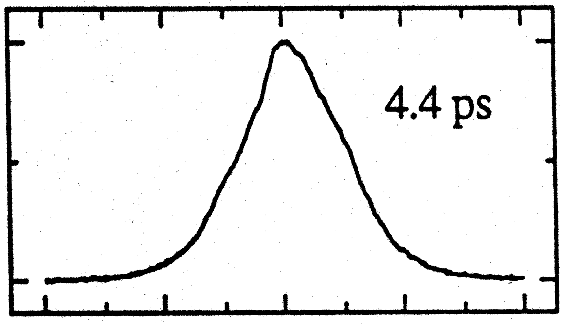
- Solitons propagate without spreading because of exact balance between group velocity dispersion (GVD) and self-phase modulation (SPM).

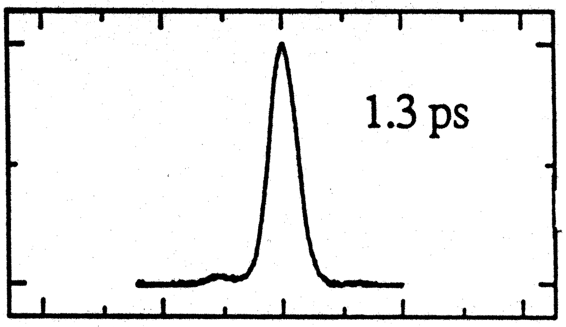
$$i\frac{\partial U}{\partial \xi} = \mathrm{sgn}(\beta_2)\frac{1}{2}\frac{\partial^2 U}{\partial \tau^2} - N^2|U|^2 U$$

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

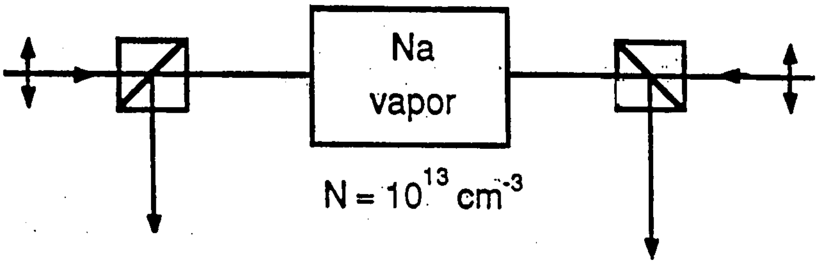
- Solution is to use a tapered fiber (15% in 40 km) so that the GVD decreases at the same rate as the pulse energy.


input


output of conventional fiber


output of dispersion-decreasing fiber

Chaos in Sodium Vapor



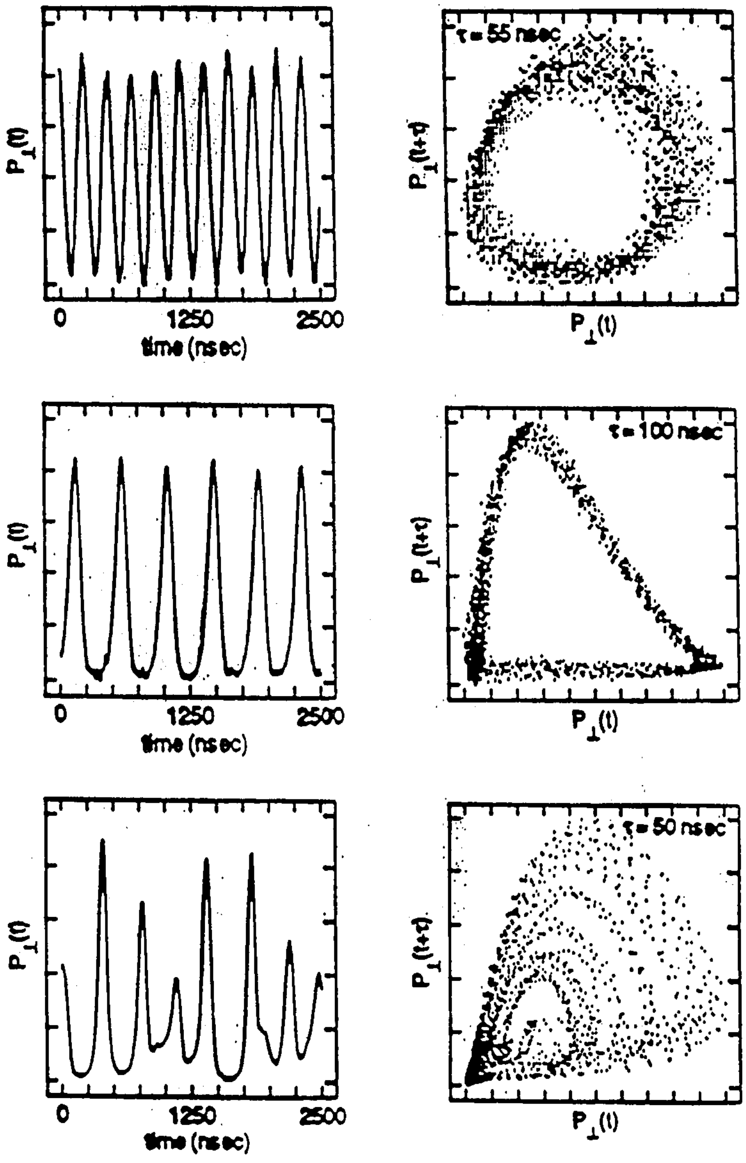
Temporal Evolution

Phase Space Trajectories

$P_b = 24$ mW

$P_b = 26$ mW

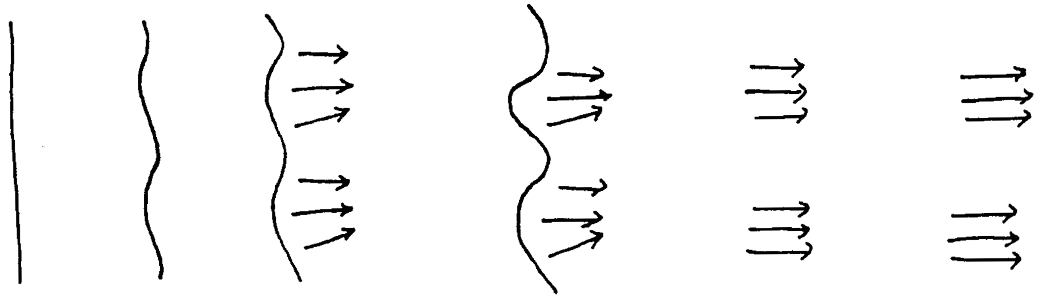
$P_b = 29$ mW



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

Laser Beam Filamentation

Spatial growth of wavefront perturbations



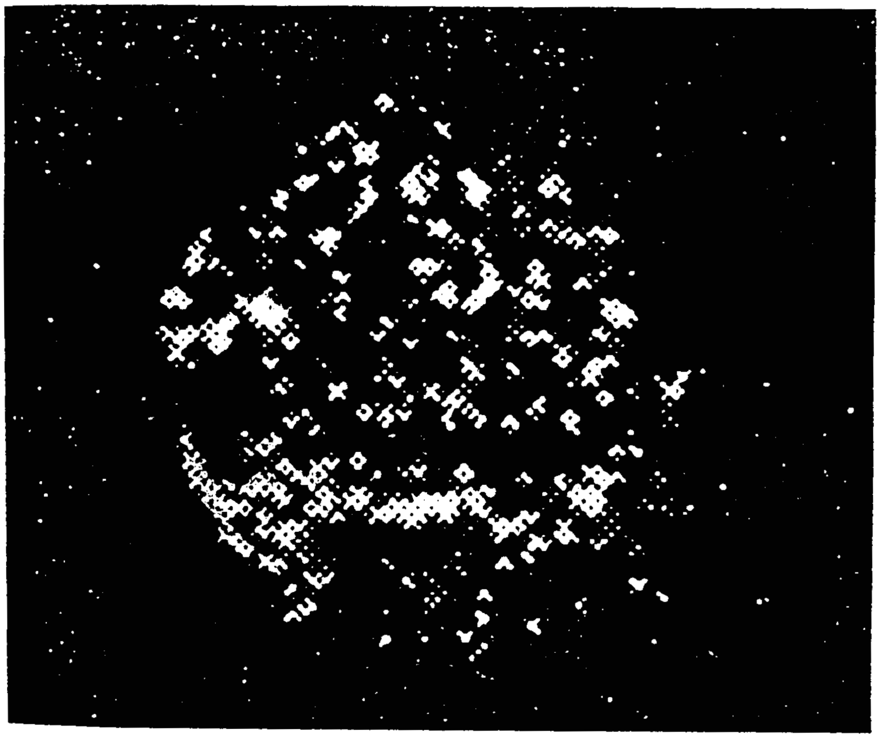
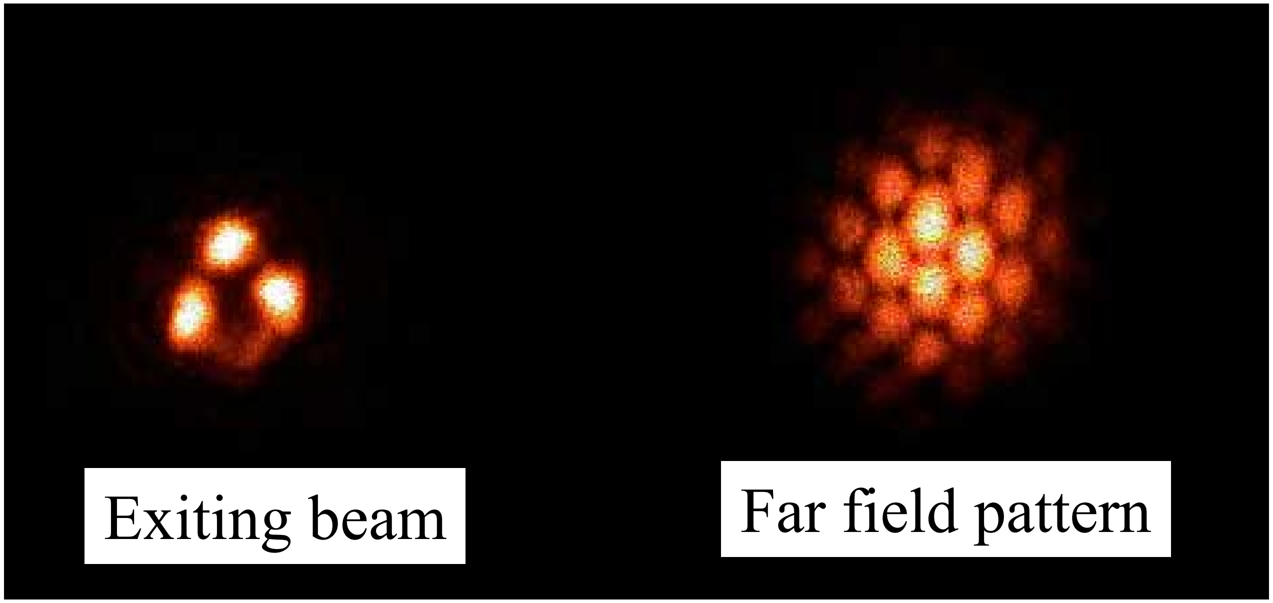


Fig. 17.2 Image of small-scale filaments at the exit windows of a CS_2 cell created by self-focusing of a multimode laser beam. [After S. C. Abbi and H. Mahr, *Phys. Rev. Lett.* 26, 604 (1971).]

Honey Comb Pattern Formation

Output from cell with single gaussian beam input



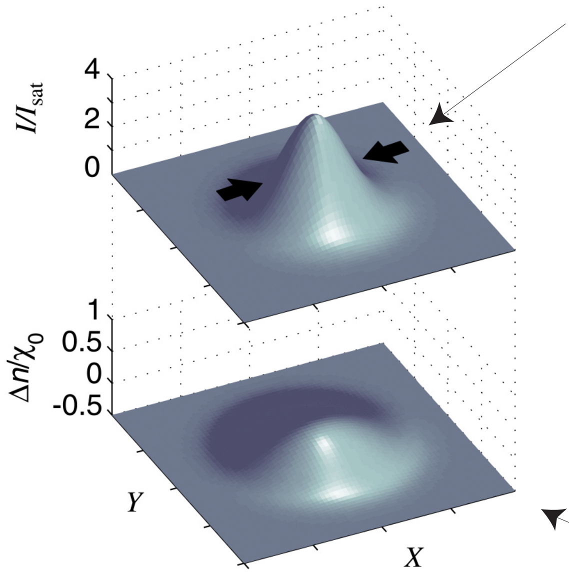
Quantum image?

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

Sodium vapor cell
T = 220° C
Bennink et al., PRL 88, 113901 2002.

Spontaneous Pattern Formation in Sodium Vapor

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:



beam will narrow due to lens-like refractive index profile

$$n = 1 + \quad n$$

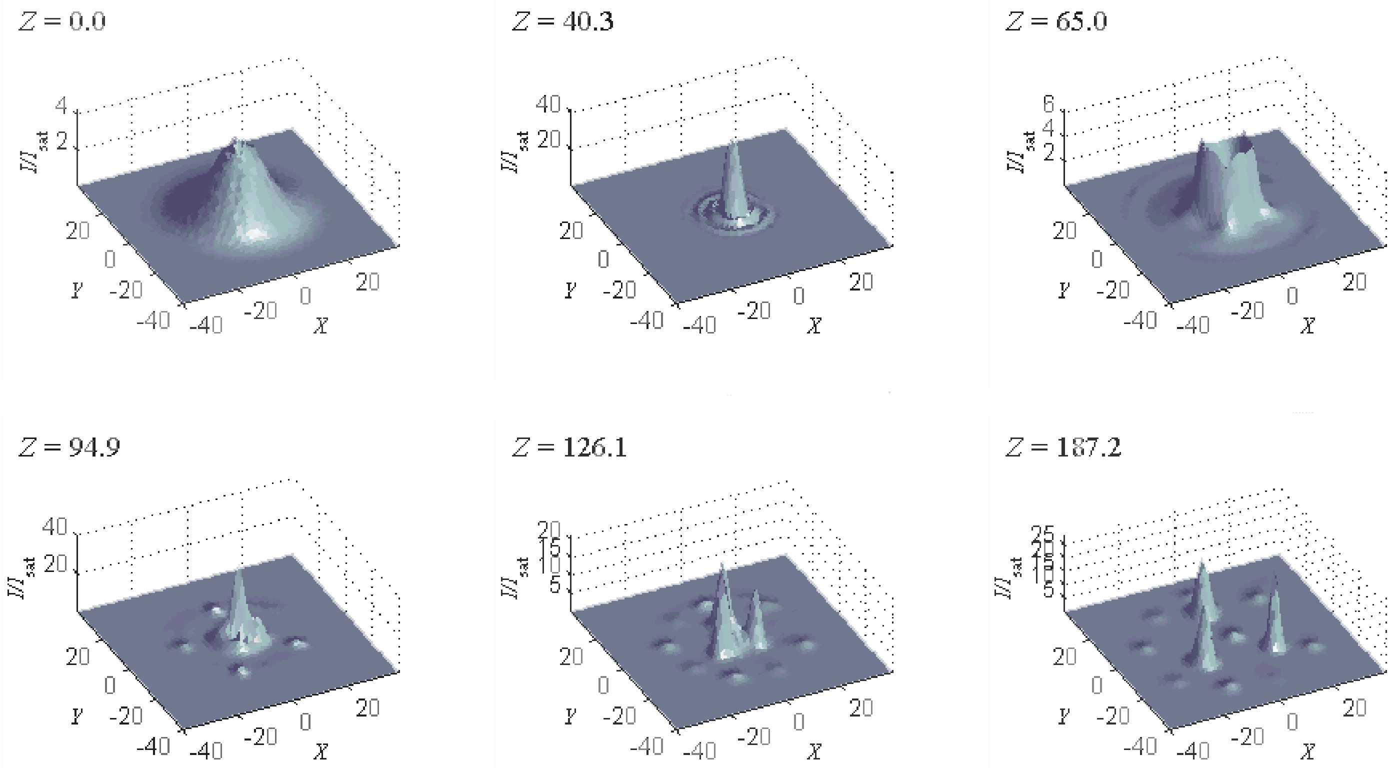
$$1 + \frac{1}{2}\,\frac{0}{1 + I/I_{sat}}$$

induced refractive index is like a positive lens

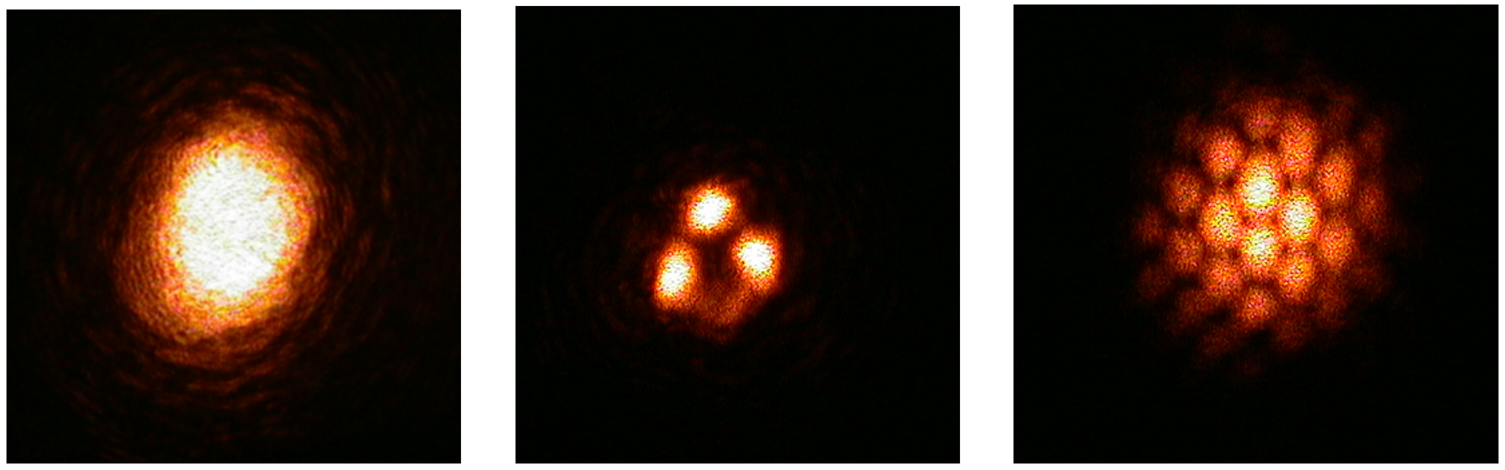
Since light refracts in the direction of increasing index, in a medium with negative saturable nonlinearity it refracts toward regions of higher intensity. This causes smooth beams to narrow or self-focus. But it also tends to destabilize a beam as small amplitude fluctuations grow due to local self-focusing.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

**For sodium at 200°C, $_0$ -0.05 and I_{sat} 6 mW/cm²*

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



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



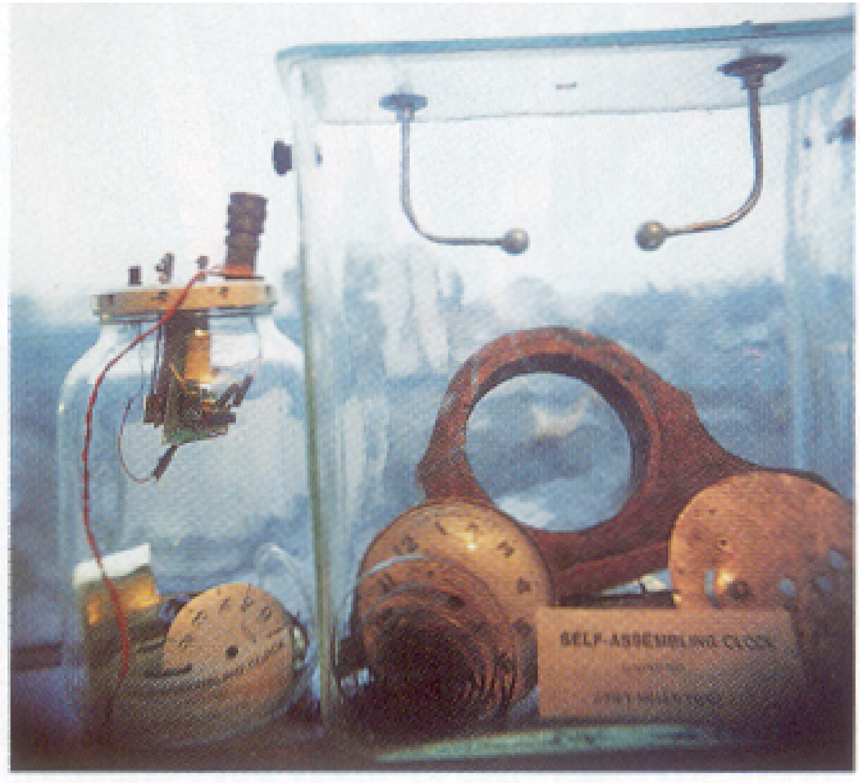
beam entering sodium

beam leaving sodium

far-field pattern

Pictures taken by R. Bennink, S. Lukishova, and V. Wong.

Experiment in Self Assembly



Joe Davis, MIT