New Concepts and Materials for Nonlinear Optics

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NONLINEAR OPTICS

Robert W. Boyd

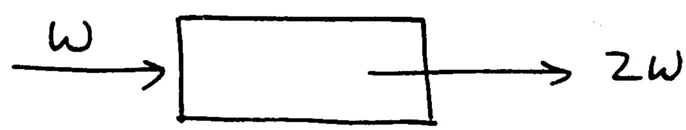
What is Nonlinear Optics?

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

(P:) dipole moment per unit volume

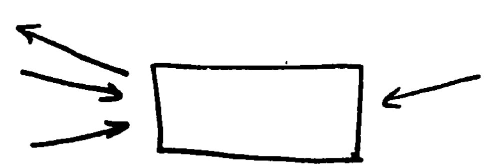
$\chi^{(1)}$: linear optics, eg 

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



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

four-wave mixing



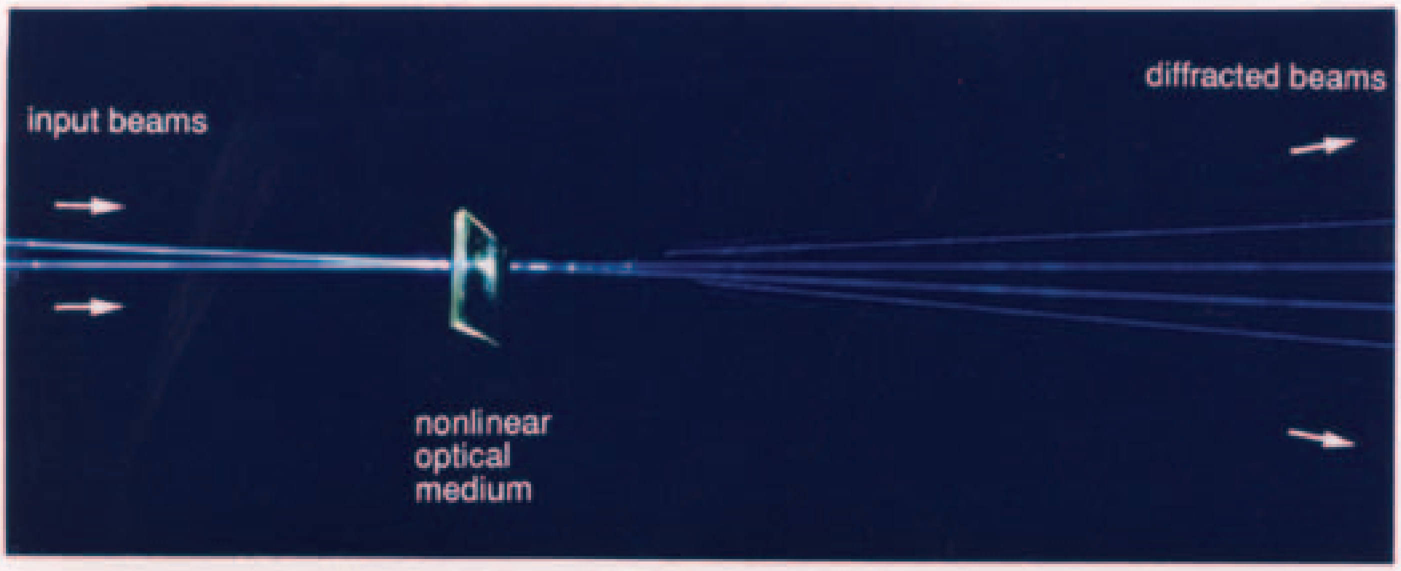
Intensity-dependent refractive index

$$n = n_0 + n_2 I$$

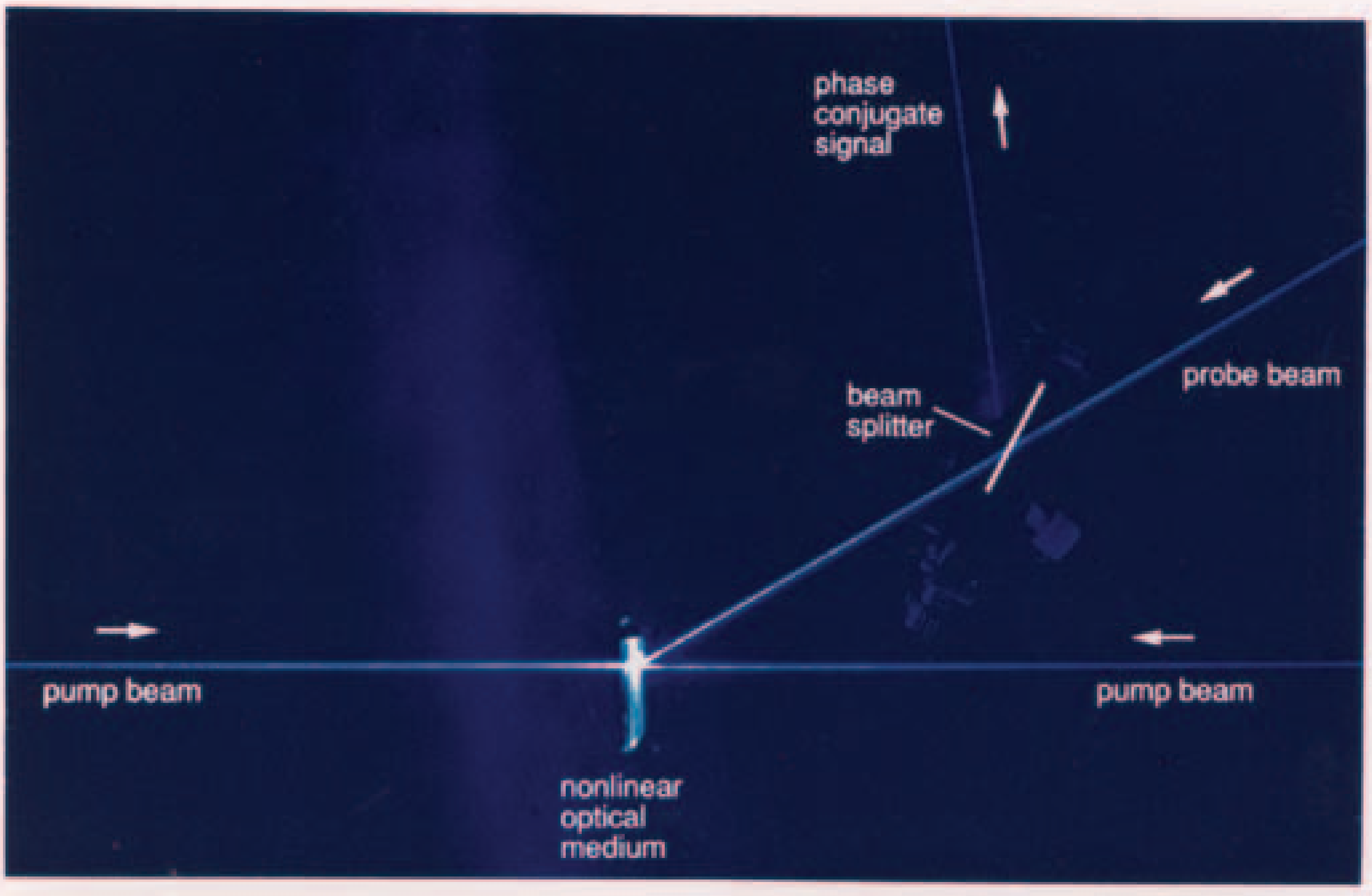
$$n_2 = \frac{12\pi^2}{n_0^2 c} \chi^{(3)}$$

Nonlinear Optical Interactions

Light-by-Light Scattering



Phase Conjugation by Degenerate Four-Wave Mixing



Nonresonant Electronic Nonlinearities

Estimate size:

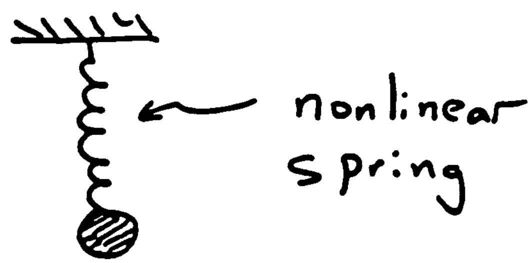
$$P = \chi^{(1)} E + \chi^{(3)} E^3$$

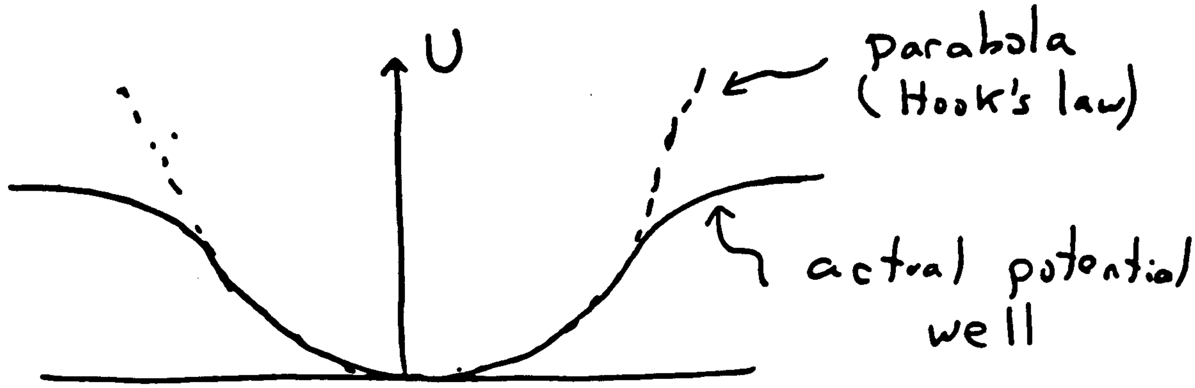
nonlinear term becomes comparable to linear term when

$$E \sim E_{at} \equiv \frac{e}{a_0^2}$$ ← Bohr radius

$$\Rightarrow \quad \chi^{(3)} \approx \frac{\chi^{(1)}}{E_{at}^2} \approx \frac{1}{E_{at}^2} = 10^{-16} \text{ esu}$$

Must generalize the Lorentz model of atom to allow a nonlinearity in restoring force.





Order-of-Magnitude Estimate of the Nonlinear Optical Susceptibility*

$$\chi^{(2)} = \hbar^4/8m^2e^5 = 7.29 \times 10^{-9} \text{ cm (sV)}^{-1}$$

$$\chi^{(3)} = \hbar^8/8m^4e^{10} = 4.25 \times 10^{-16} \text{ cm}^2 \text{ (sV)}^{-2}$$

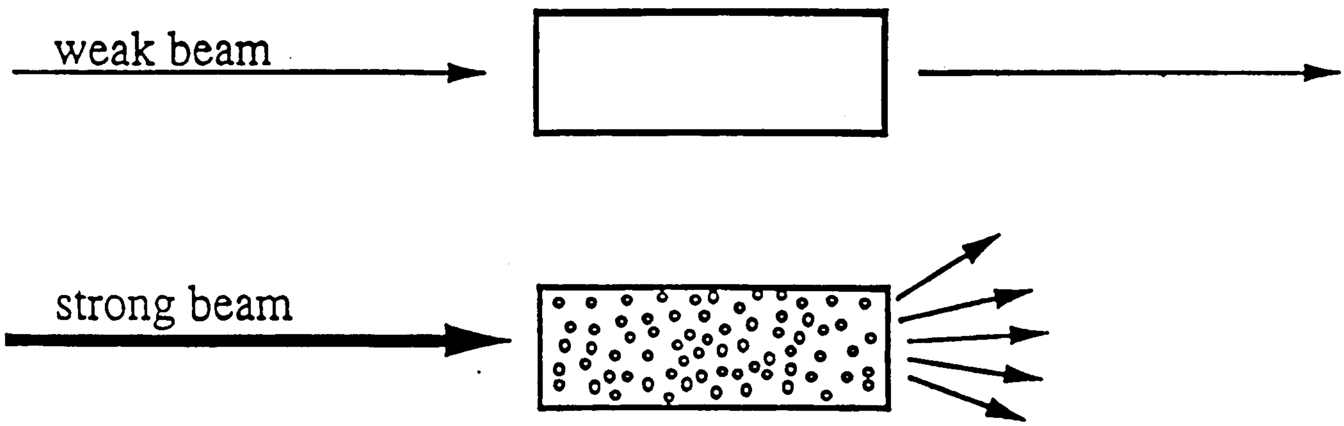
Remarkably, the electronic, Raman, and electrostrictive contributions to $\chi^{(3)}$ are all of the same order of magnitude!

* R. W. Boyd, Journal of Modern Optics. 46, 367, 1999.

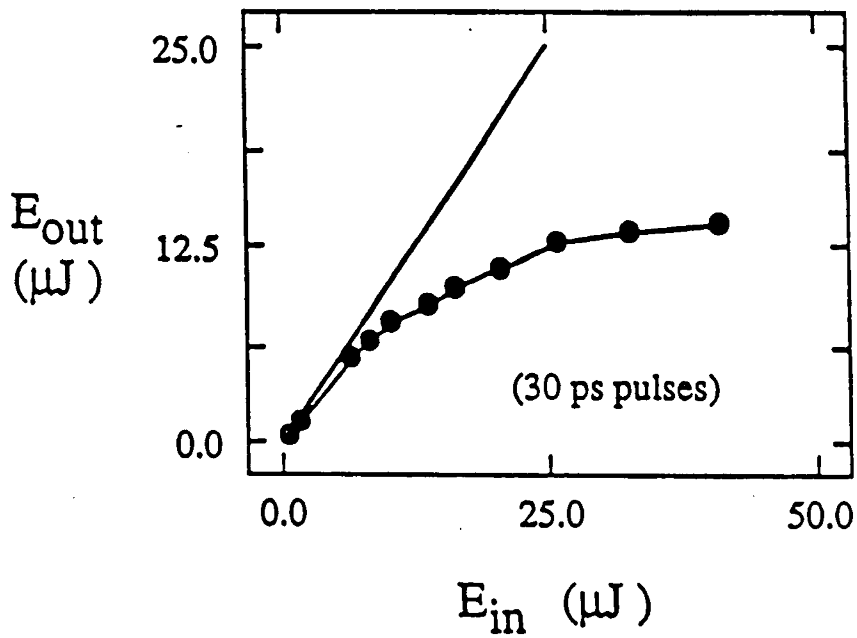
Optical Power Limiter

We want a filter that will transmit weak light, but block an intense laser beam.

Our approach:
Nonlinear Christiansen filter:



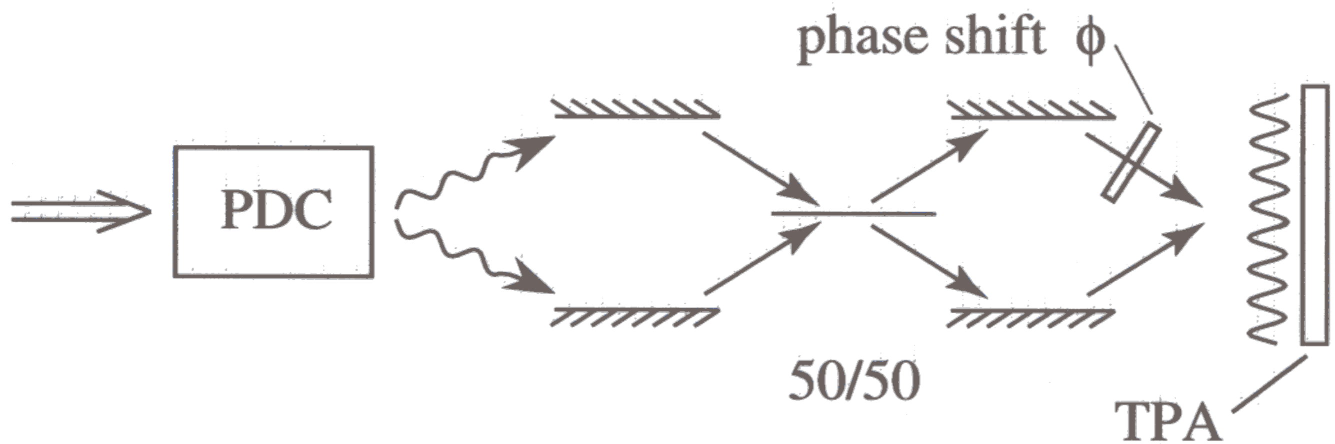
We have constructed such a filter using ground glass in a mixture of acetone and CS_2



G. L. Fischer, R. W. Boyd, T. R. Moore, and J. E. Sipe, Optics Letters, 21, 1643-1645, 1996.

Quantum Lithography and Microscopy

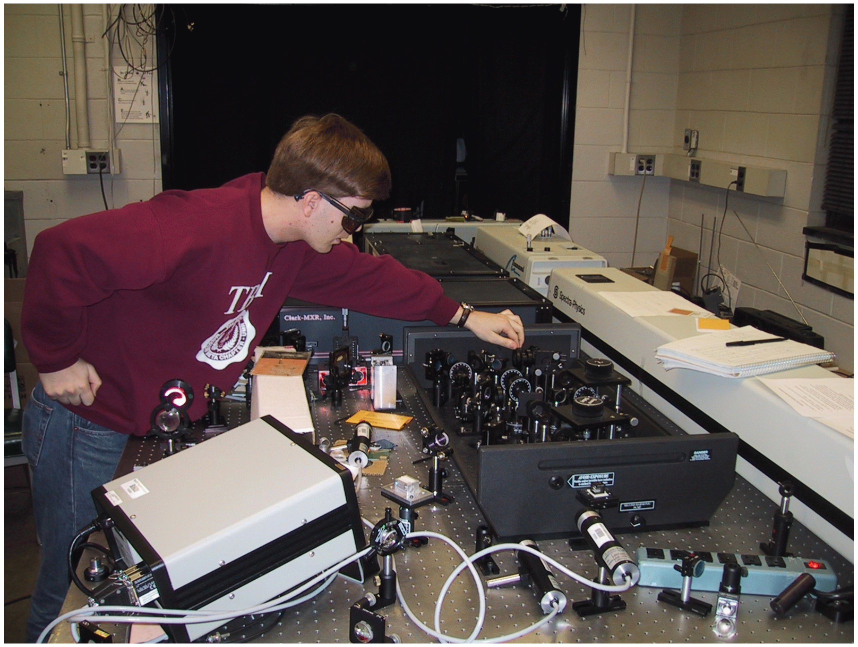
- Entangled photons can be used to form interference patterns with detail finer than the Rayleigh limit
- Process "in reverse" performs sub-Rayleigh microscopy

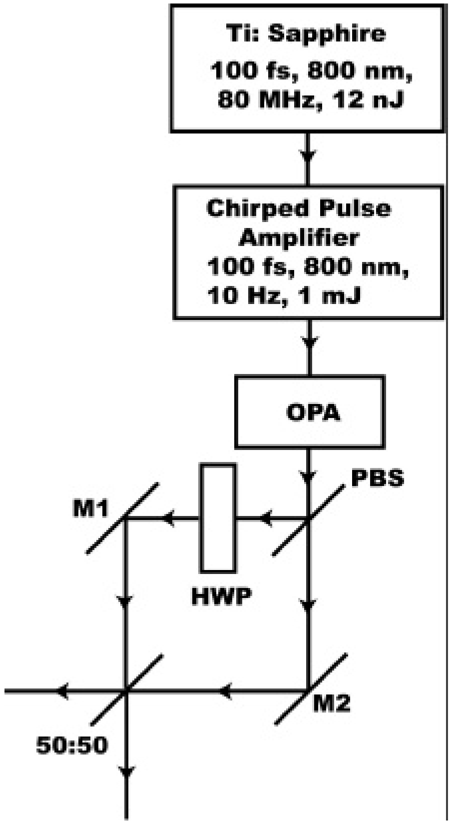


Boto et al, Phys. Rev. Lett. 85, 2733, 2000.

QUANTUM LITHOGRAPHY PROPOSAL

Experimental Layout





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.

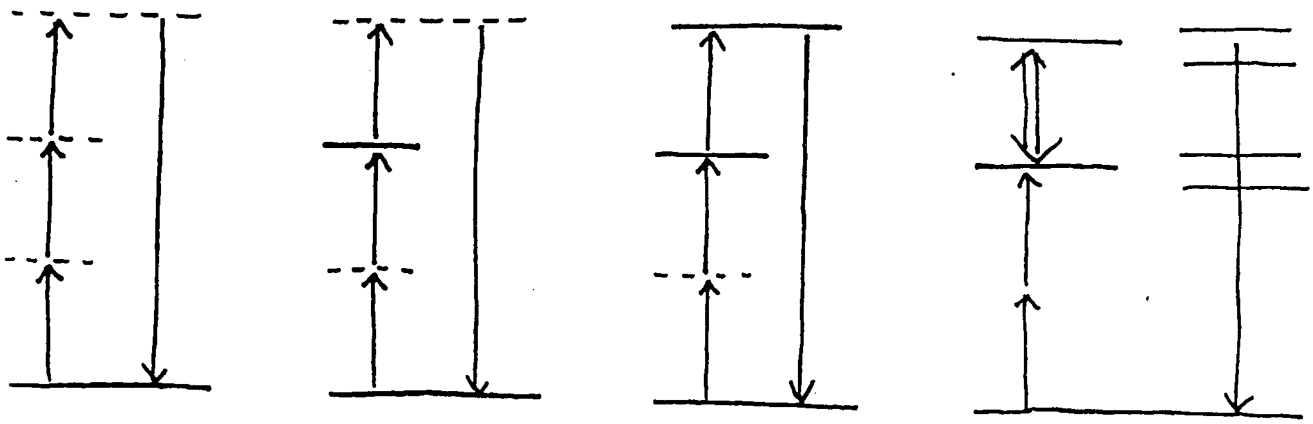
Approaches to the Development of Improved NLO Materials

- 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



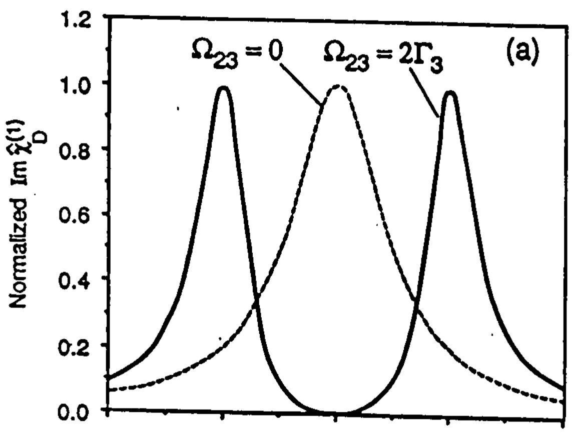
too weak

better (2-photon resonance)

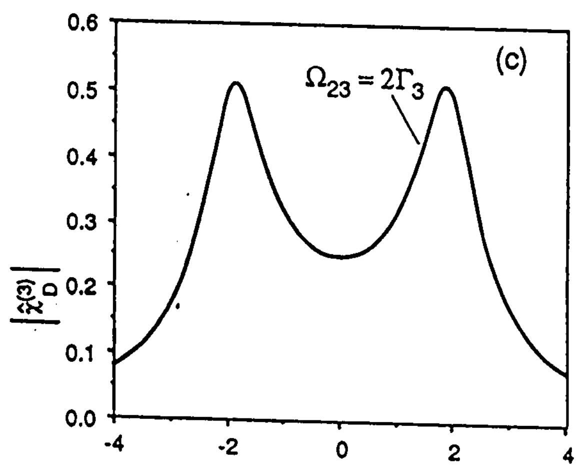
still better (but absorption!)

The EIT concept

- EIT Predictions



Absorption of the generated field vanishes.

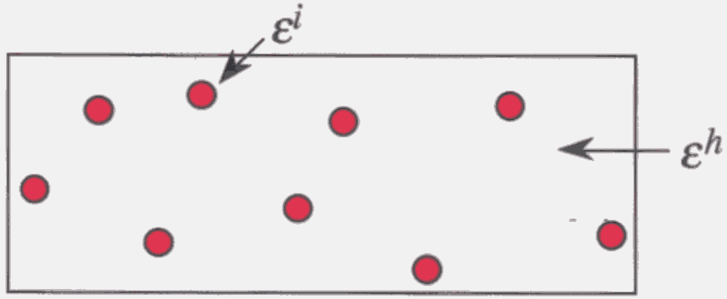


But the nonlinearity remains large!

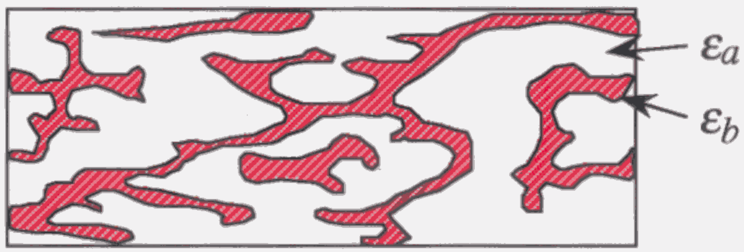
Harris, Field and Imamoglu, PRL 64 1107 1990

Nanocomposite Materials for Nonlinear Optics

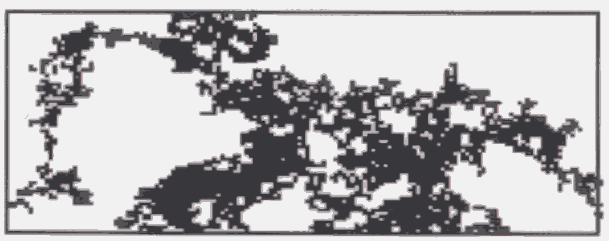
- Maxwell Garnett



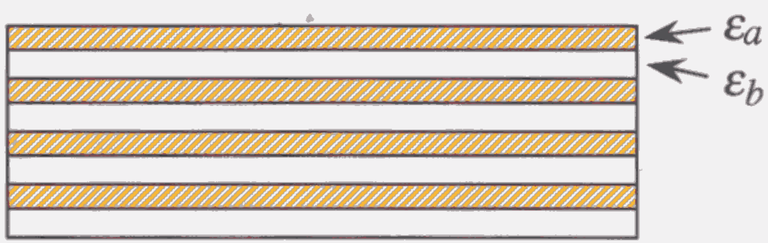
- Bruggeman (interdispersed)



- Fractal Structure



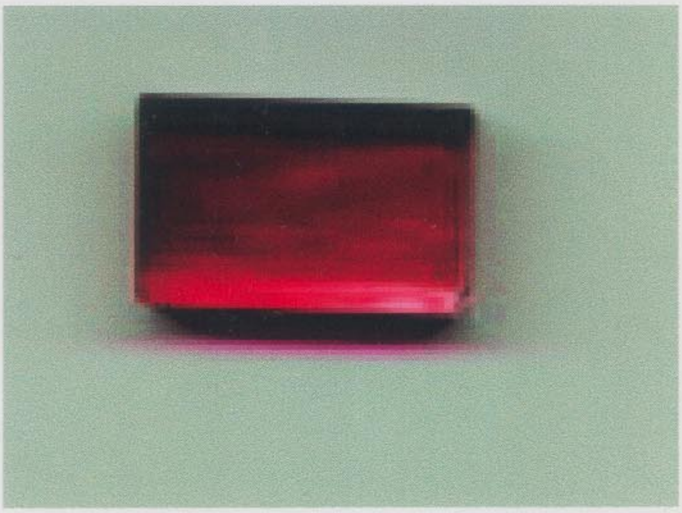
- Layered



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.*

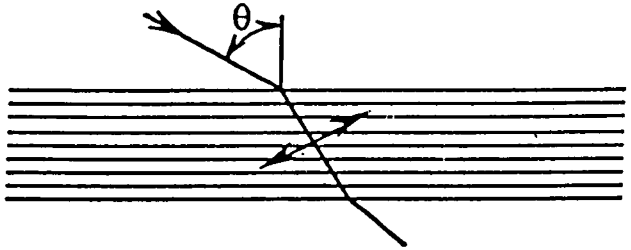
- Enhancement of $\chi^{(3)}$ can be understood in terms of local field effects

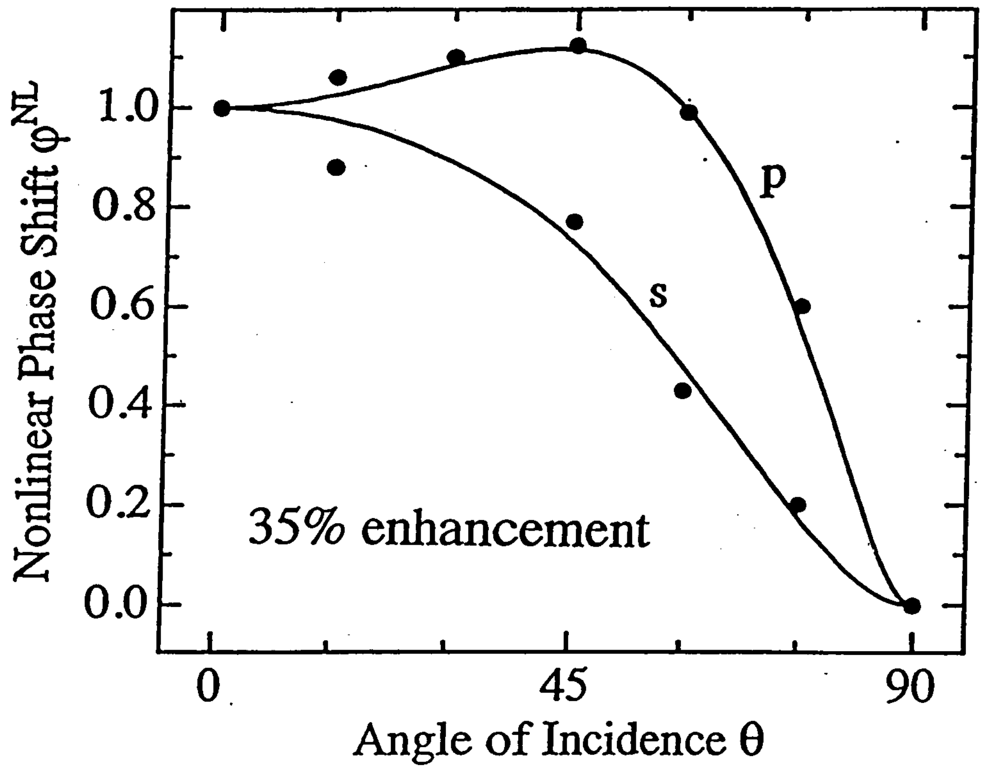
* RWB and J. E. Sipe, US patent #5,253,103; PRL 74, 1871, 1995.

First Demonstration of Enhanced NLO Response

Alternating layers of TiO_2 and the conjugated polymer PBZT.

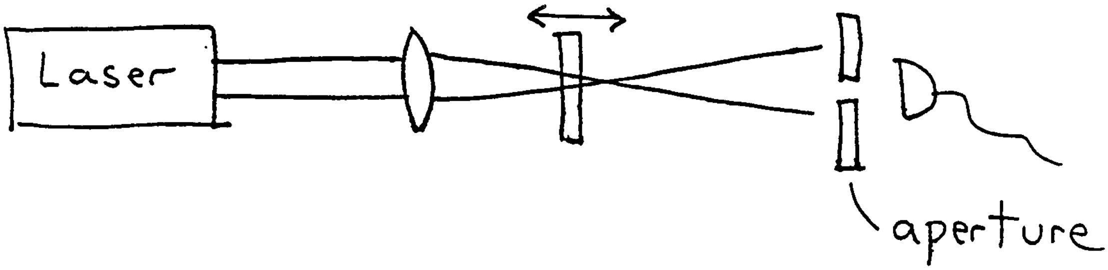
Measure NL phase shift as a function of the angle of incidence.





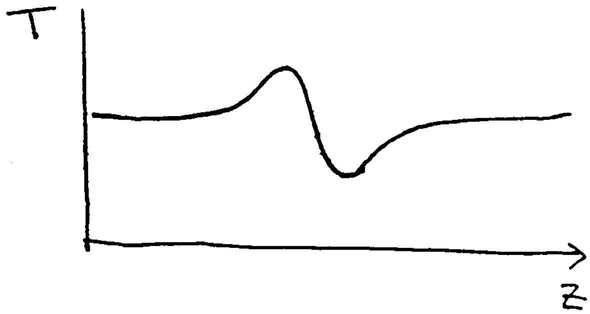
Fischer, Boyd, Gehr, Jenekhe, Osaheni, Sipe and Weller-Brophy, Phys. Rev. Lett 74, 1871 (1995).
Gehr, Fischer, Boyd and Sipe, Phys. Rev. A 53, 2792 (1996).

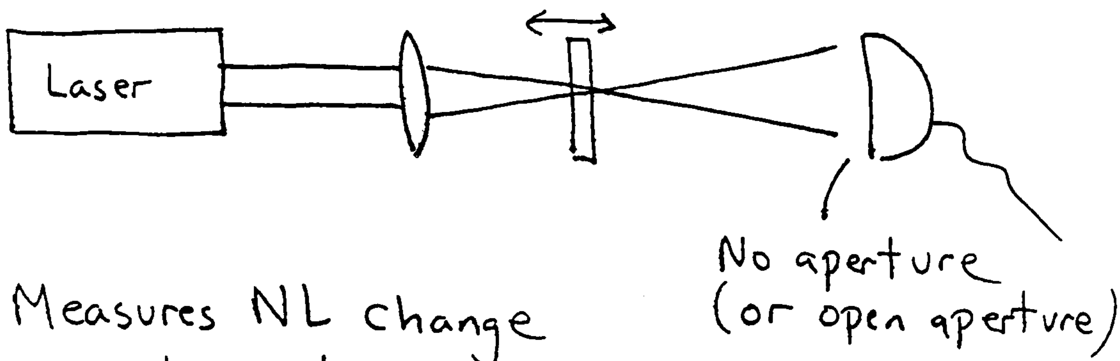
Z-Scan Measurement of $\chi^{(3)}$



Measures NL change in refraction

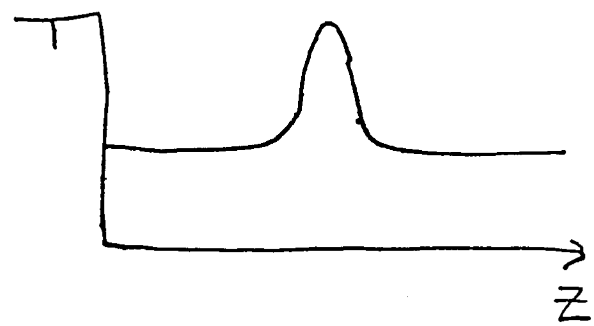
($\mathrm{Re}\,\chi^{(3)}$)





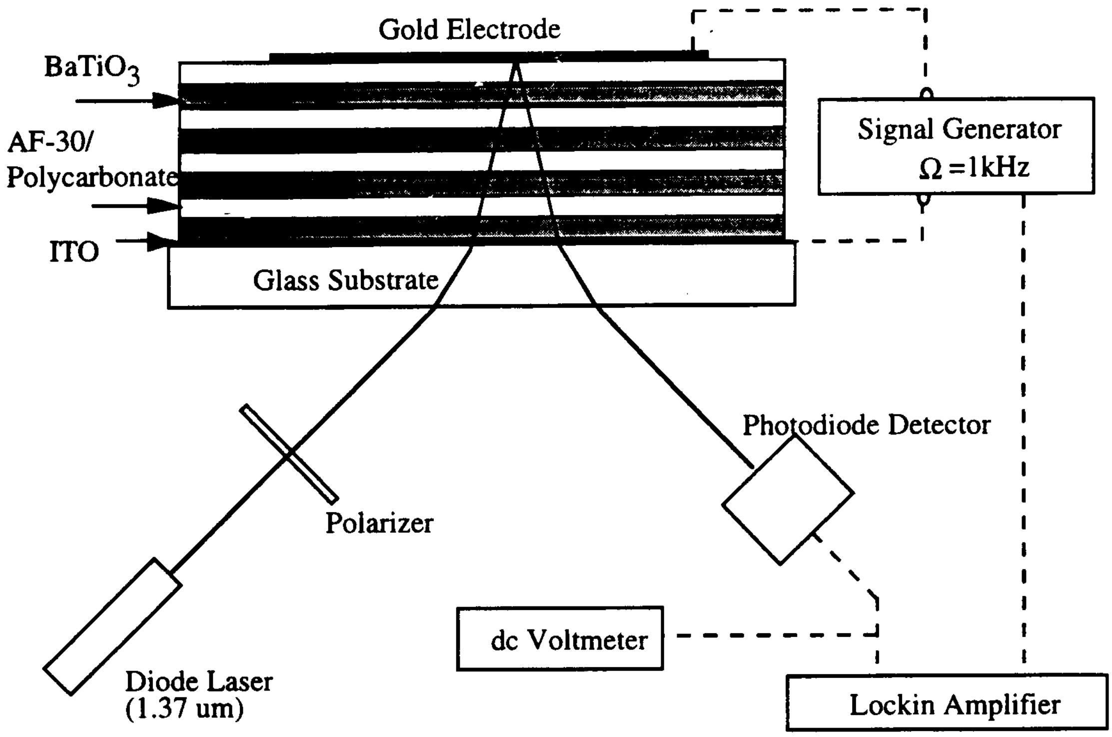
Measures NL change in absorption.

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



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

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

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

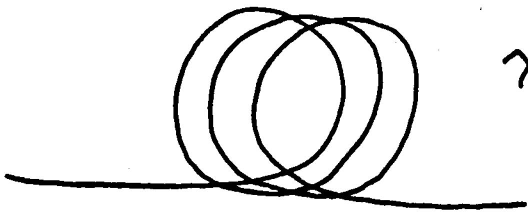
3.2 times enhancement in agreement with theory

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 $\chi^{(3)}/\alpha$.

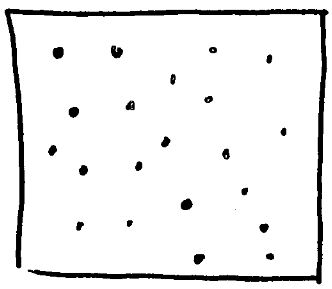
Fiber NLO



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

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

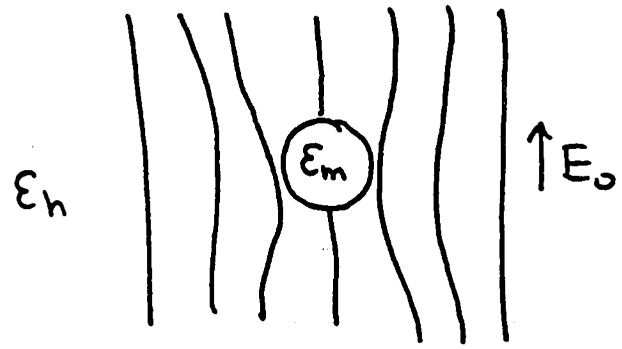
dilute colloid →



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

Metal / Dielectric Composites

Very large local field effects



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

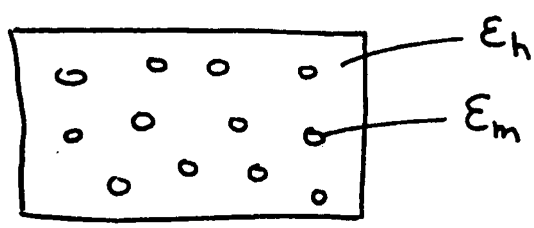
$$\equiv \mathcal{L} E_0$$

(ε_m is negative!)

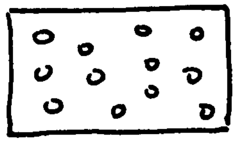
At resonance

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

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



Counterintuitive Consequence of Local Field Effects



gold nanoparticles in a liquid dye solution (HITCl)

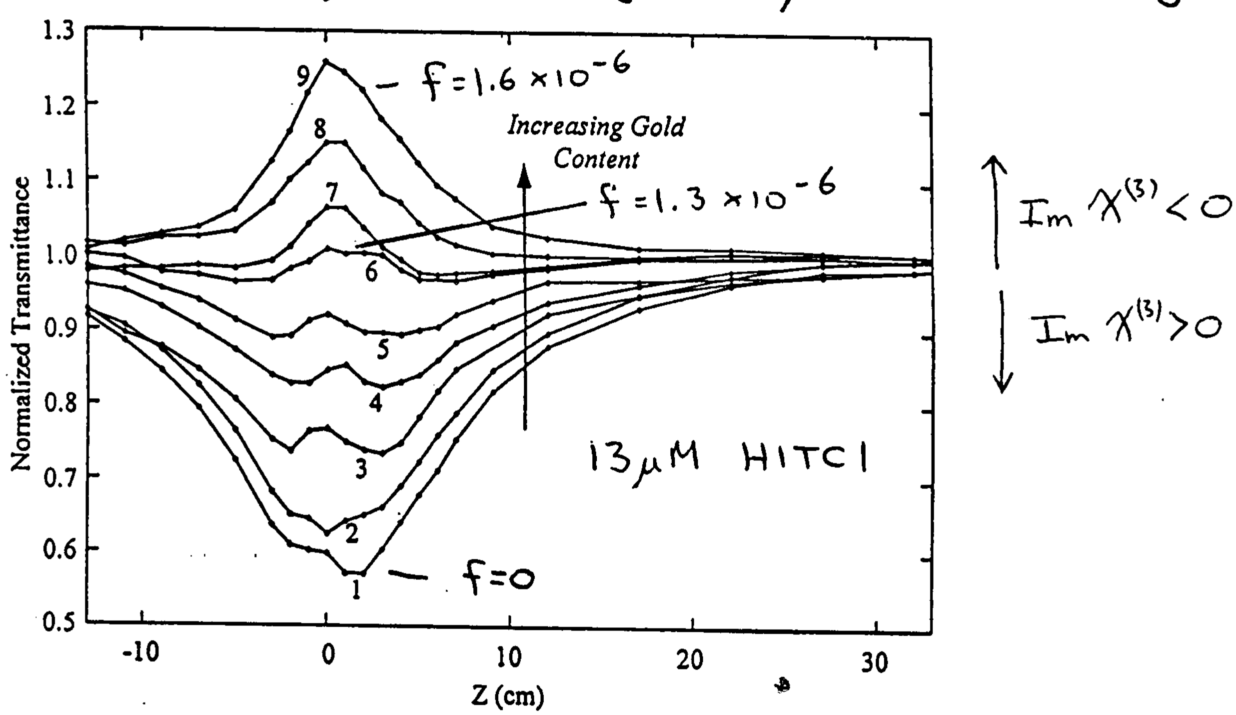
Both constituents are reverse saturable absorbers $\Rightarrow$ $\text{Im}\,\chi^{(3)} > 0$

Effective NL susceptibility of composite

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

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

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

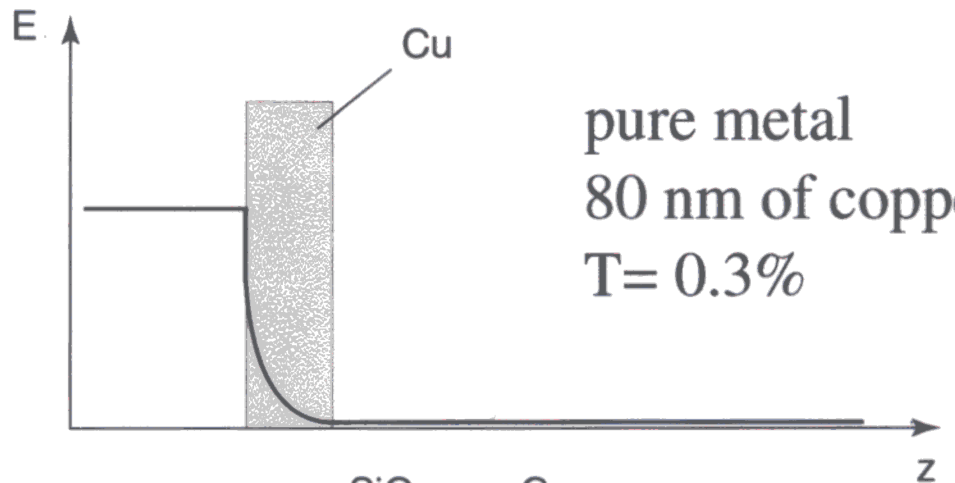


Smith et al, JOSA B 14 1625 1997.

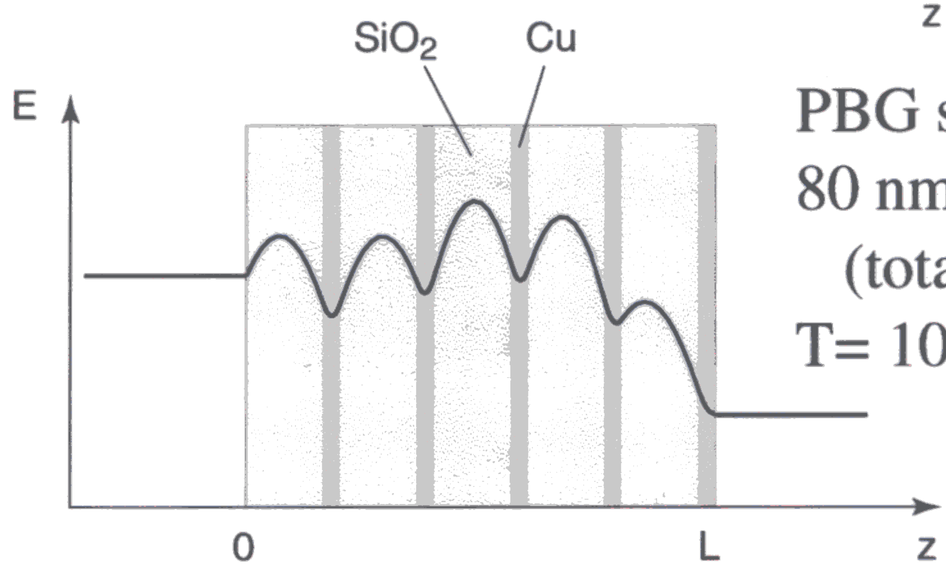
Accessing the Optical Nonlinearity of Metals with Metal-Dielectric PBG Structures

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

- 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!

“Slow” Light in Nanostructured Devices

Robert W. Boyd

with

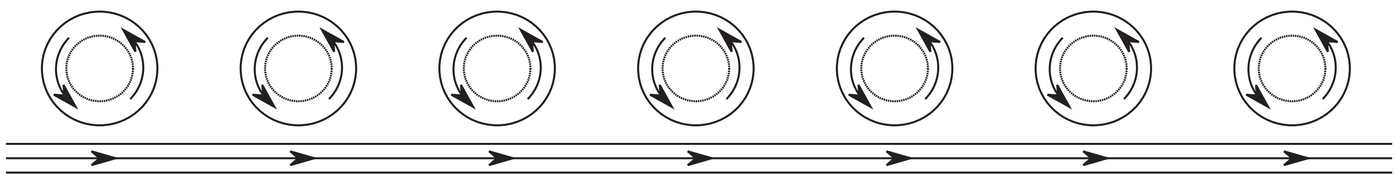
**John Heebner, Nick Lepeshkin,
Aaron Schweinsberg, and Q-Han Park**

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

Presented at PQE-2002

NLO of SCISSOR Devices

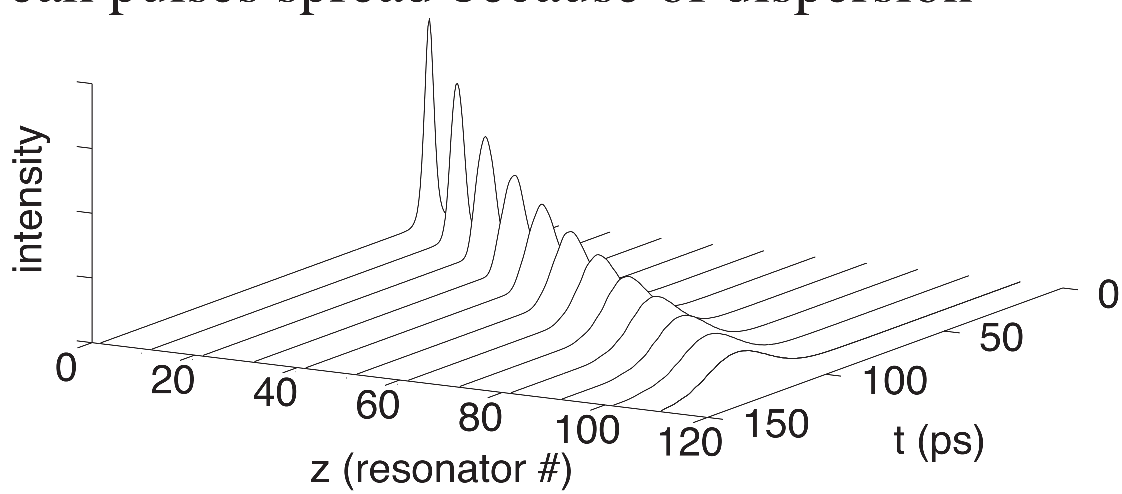
(Side-Coupled Integrated Spaced Sequence of Resonators)



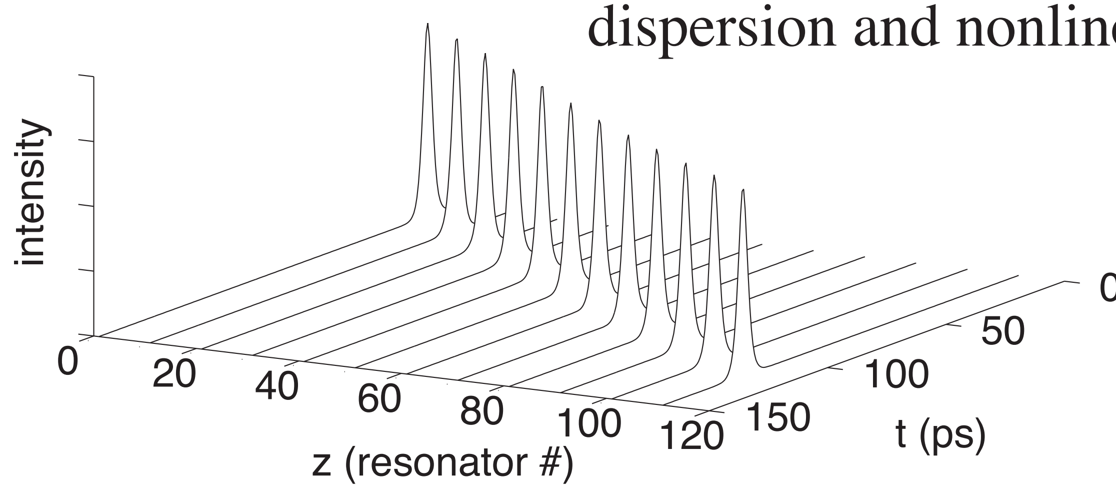
Shows slow-light, tailored dispersion, and enhanced nonlinearity

Optical solitons described by nonlinear Schrodinger equation

- Weak pulses spread because of dispersion

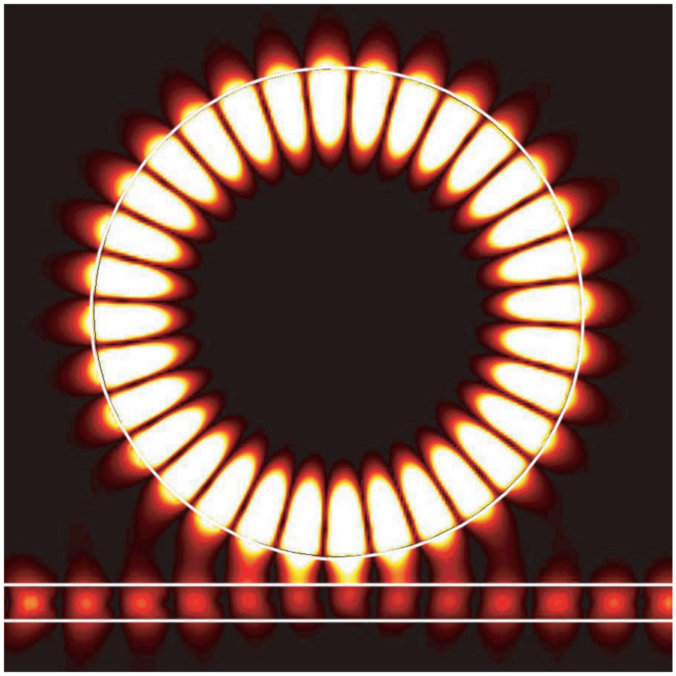


- But intense pulses form solitons through balance of dispersion and nonlinearity.

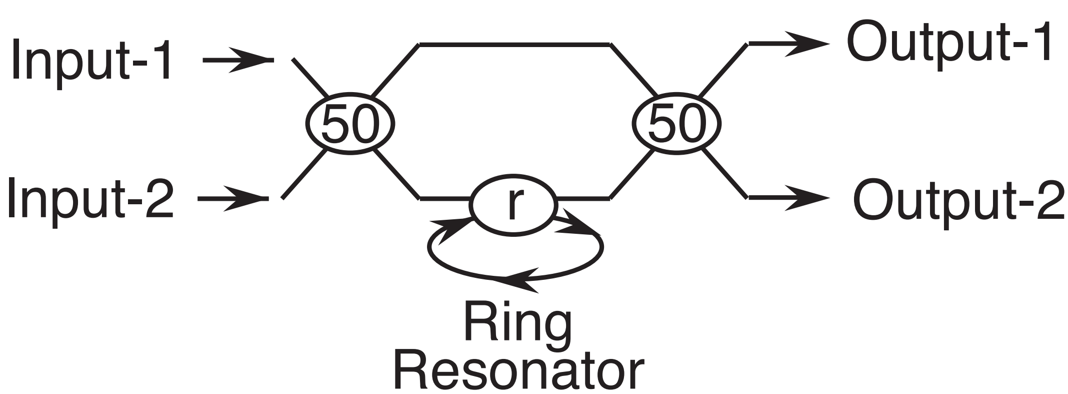


Ultrafast All-Optical Switch Based On Arsenic Triselenide Chalcogenide Glass

- 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. ($F \approx 10^2$ 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

Photonic Devices for Biosensing

Objective:

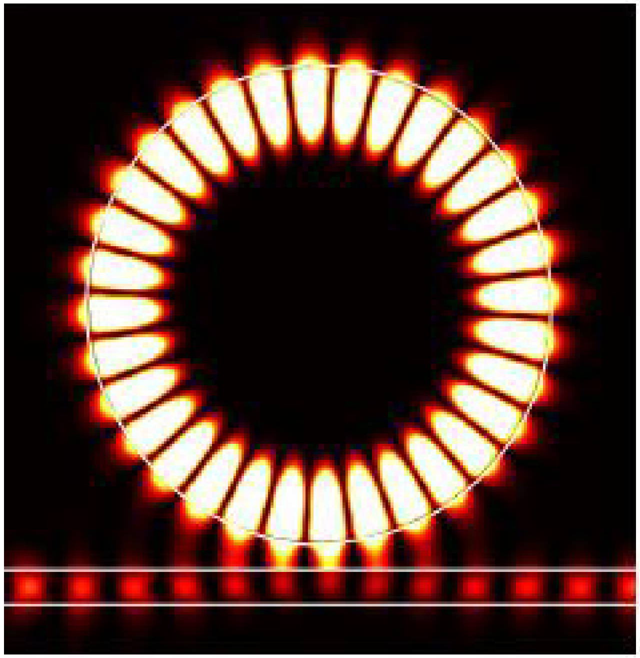
Obtain high sensitivity, high specificity detection of pathogens through optical resonance

Approach:

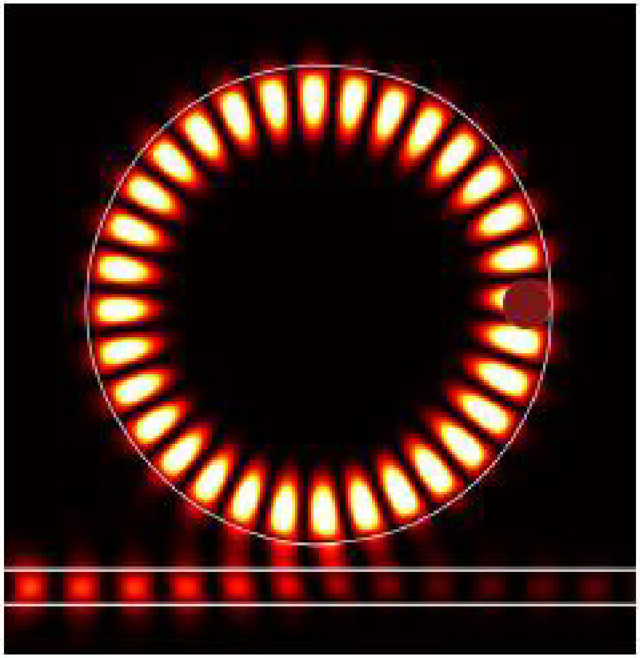
Utilize high-finesse whispering-gallery-mode disk resonator.

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

Simulation of device operation:



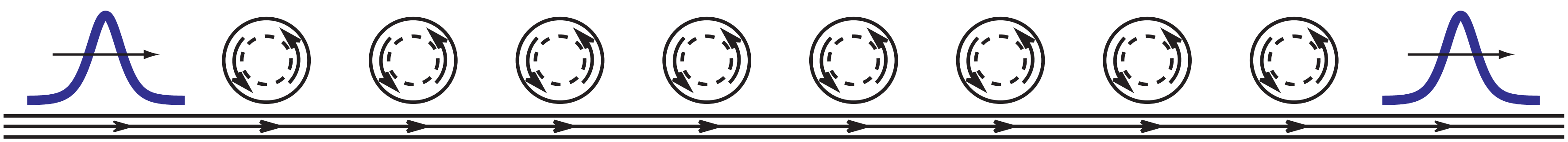
Intensity distribution in absense of absorber.



Intensity distribution in presence of absorber.

FDTD

Slow Light, Induced Dispersion, Enhanced Nonlinearity, Self-Steepening, and Optical Solitons in a Side-Coupled Integrated Spaced Sequence Of Resonators



SCISSOR

John E. Heebner
Q-Han Park
Robert W. Boyd

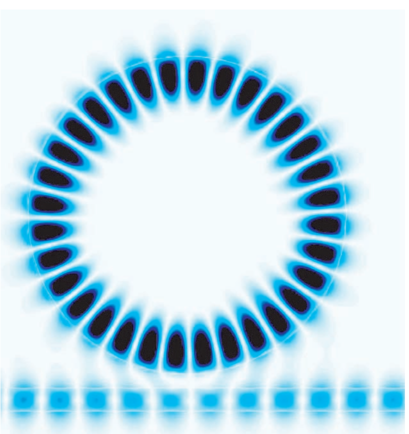
The Institute of Optics
Nonlinear Optics
University of Rochester

Motivation

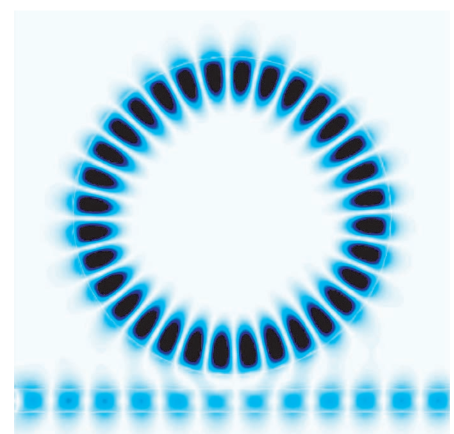
To exploit the ability of microresonators to enhance nonlinearities and induce strong dispersive effects for creating structured waveguides with exotic properties.

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:

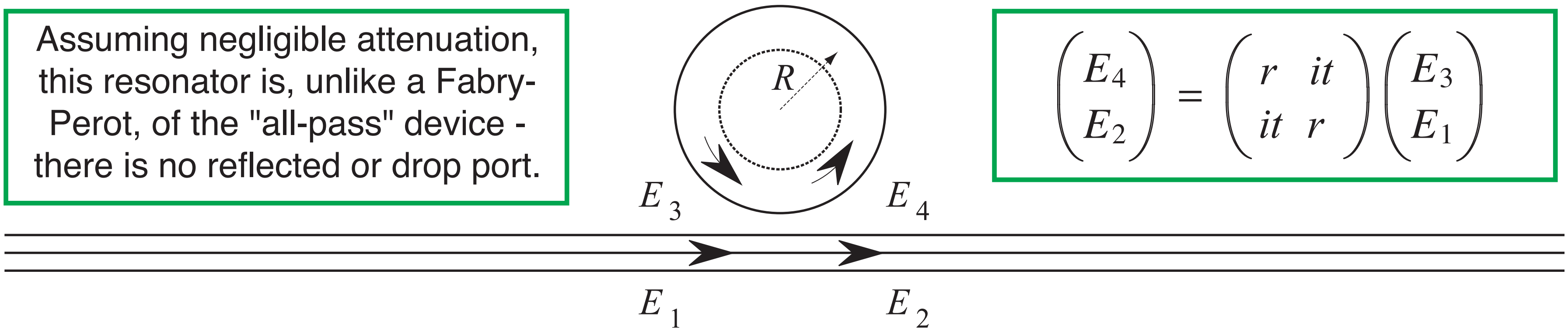


- slow light propagation
- induced dispersion
- enhanced nonlinearities



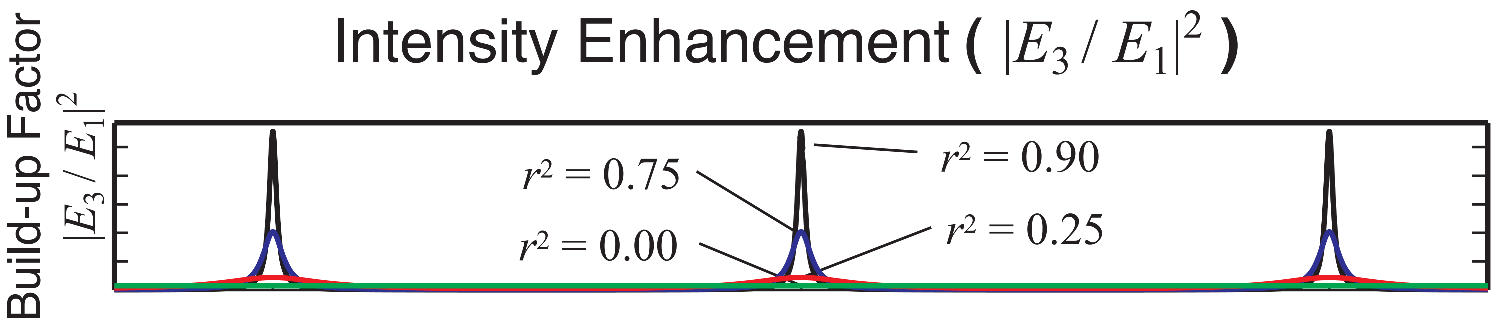
Properties of a Single Microresonator

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

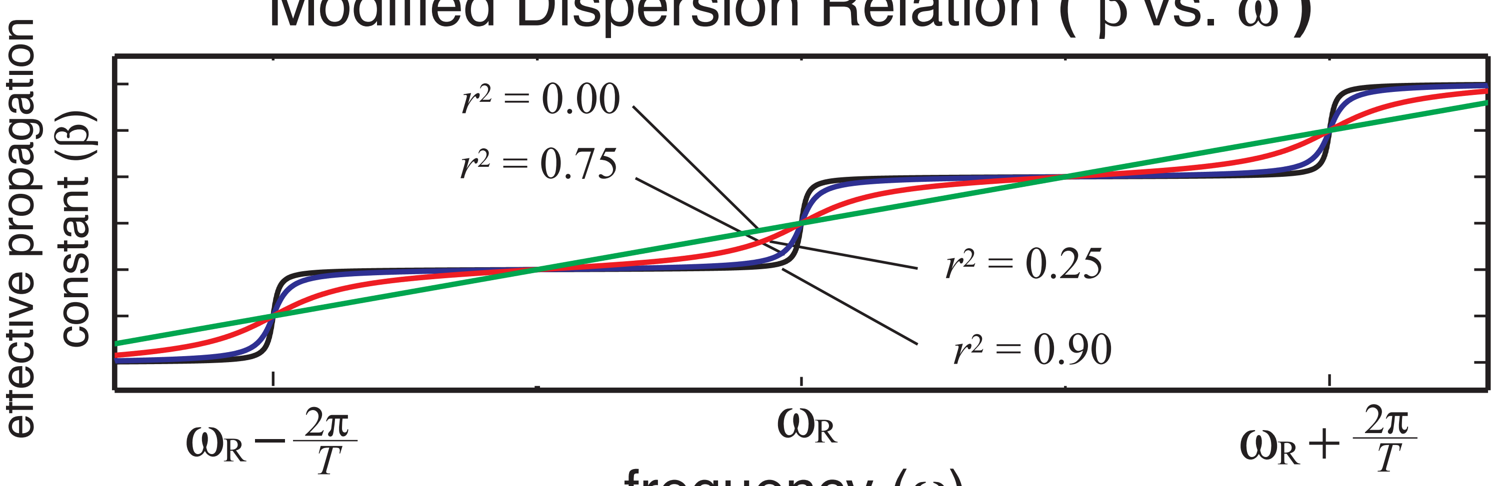


$$\begin{pmatrix} E_4 \\ E_2 \end{pmatrix} = \begin{pmatrix} r & it \\ it & r \end{pmatrix} \begin{pmatrix} E_3 \\ E_1 \end{pmatrix}$$

Intensity Enhancement ($|E_3 / E_1|^2$)



Modified Dispersion Relation (β vs. ω)



Definitions

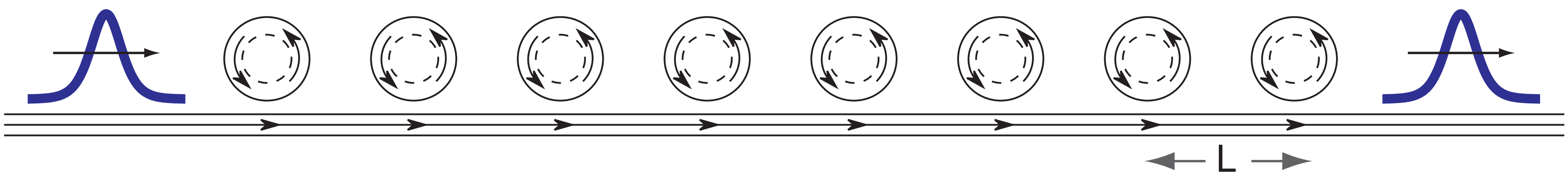
Finesse

$$F = \frac{\pi}{1-r}$$

Transit Time

$$T = \frac{n 2\pi R}{c}$$

Propagation Equation for a SCISSOR



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.

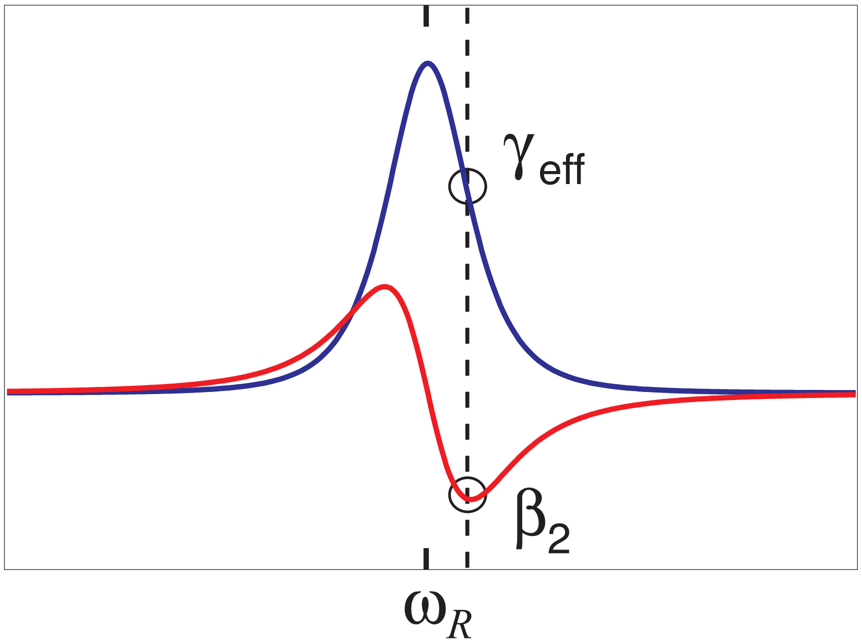
Nonlinear Schrödinger Equation (NLSE)

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

Fundamental Soliton Solution

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

Balancing Dispersion & Nonlinearity



soliton amplitude

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

adjustable by controlling ratio of transit time to pulse width

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

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

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

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

Soliton Propagation

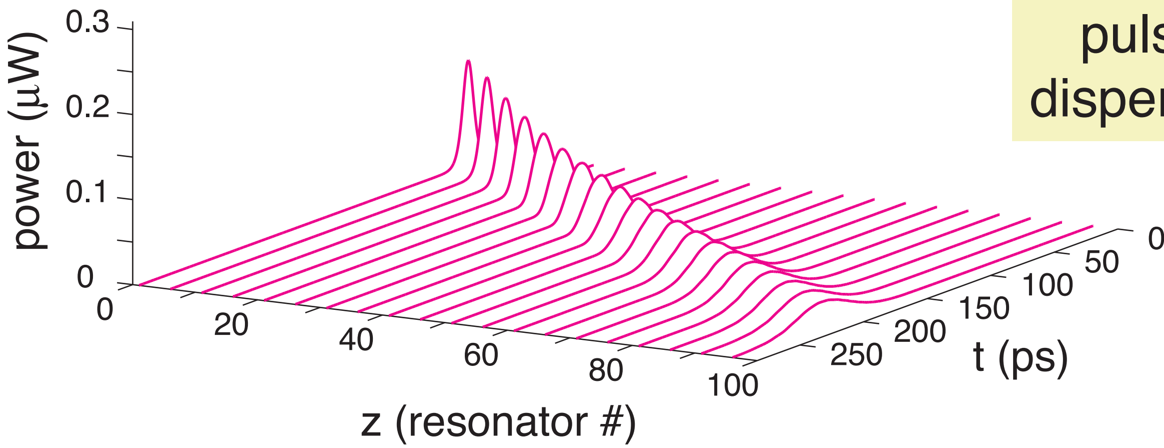
5 μm diameter resonators with a finesse of 30

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

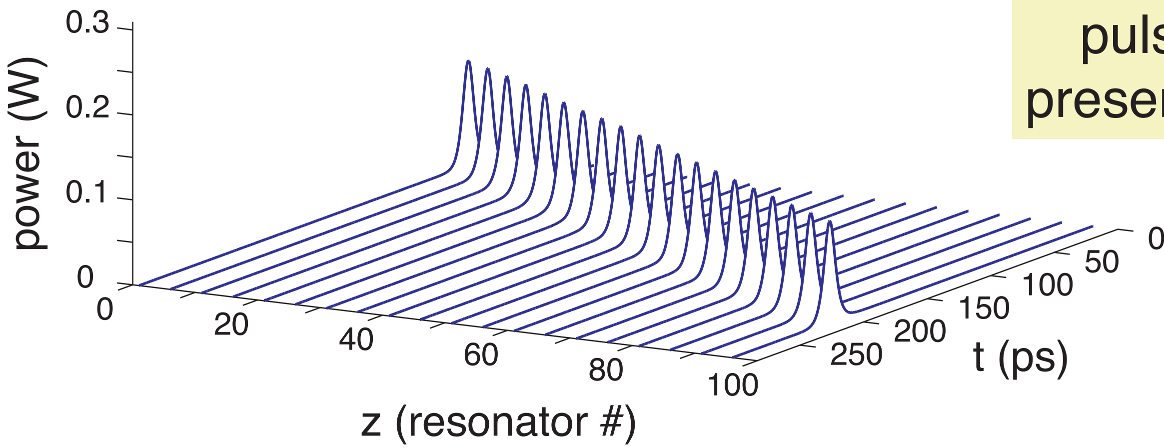
simulation assumes a chalcogenide/GaAs-like nonlinearity

Weak Pulse



pulse disperses

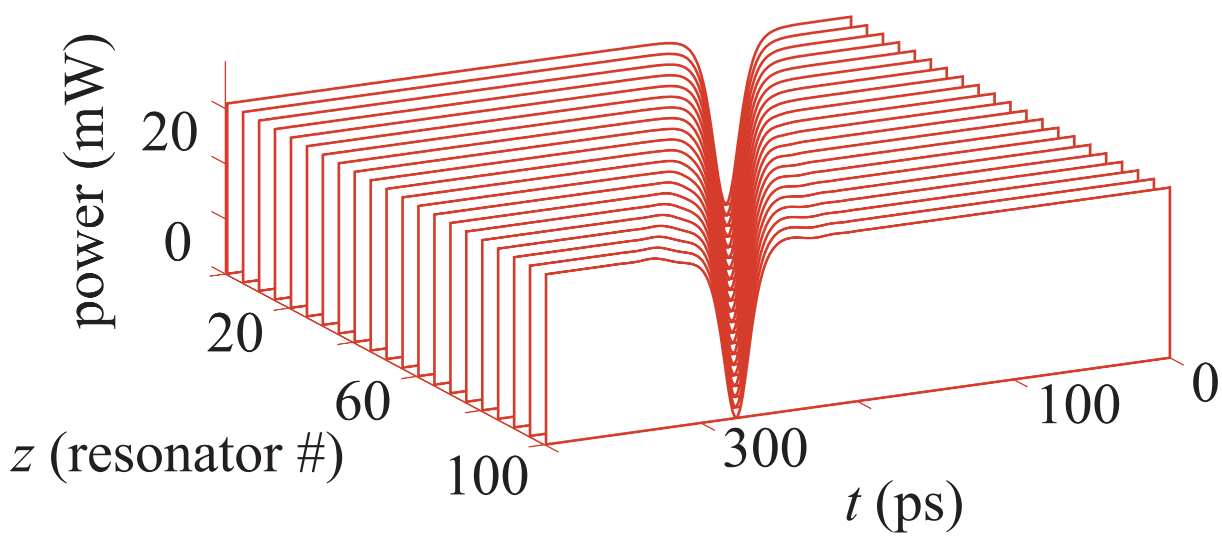
Fundamental Soliton



pulse preserved

Dark Solitons

SCISSOR system also supports the propagation of dark solitons.



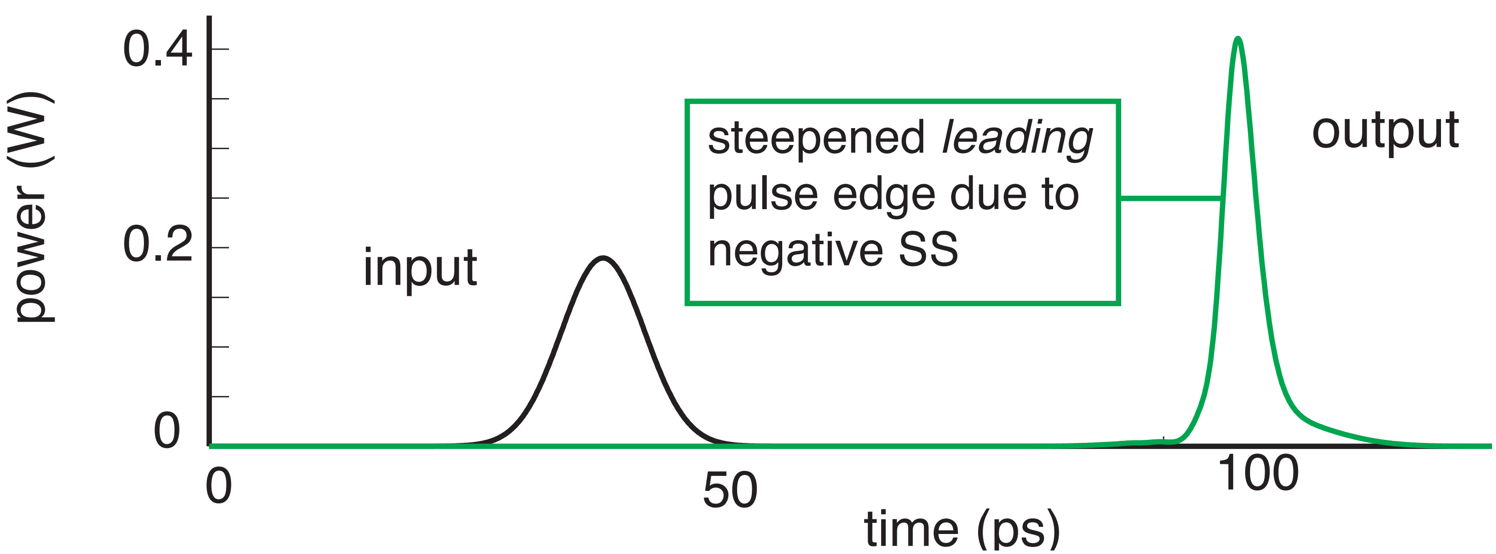
Higher-Order Effects - Self-Steepening

Higher order dispersive terms such as β_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:

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

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



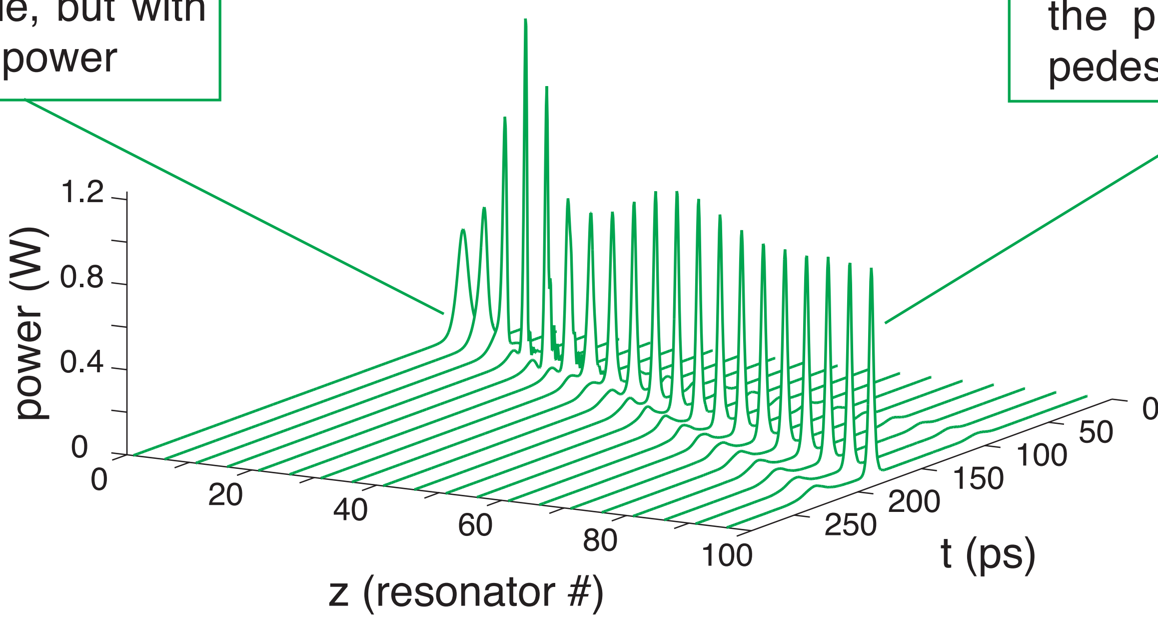
Soliton Splitting and Compression

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^{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.



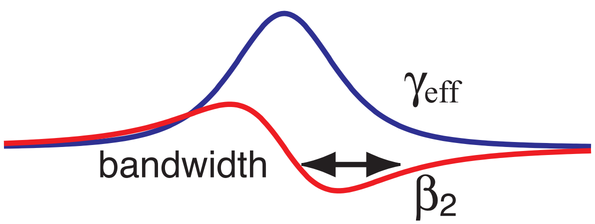
Limitations

Bandwidth

A 5 μm diamater disk with a finesse of 200 possesses a bandwidth of 100 GHz

Cascading N resonators further decreases the available bandwidth;

fortunately the scaling is governed by β_4 effects which results in a reduction of $N^{1/4}$



Manufacturability

Tolerances on resonator diameter and gap spacing to be met within 200 nm

variance in gap spacing analogous to hyperfine distribution

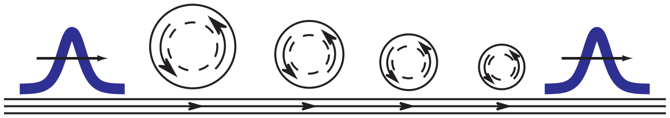
variance in radius/index analogous to Doppler broadening

Attenuation - dominated by scattering losses

may be counterbalanced by:

1) gain

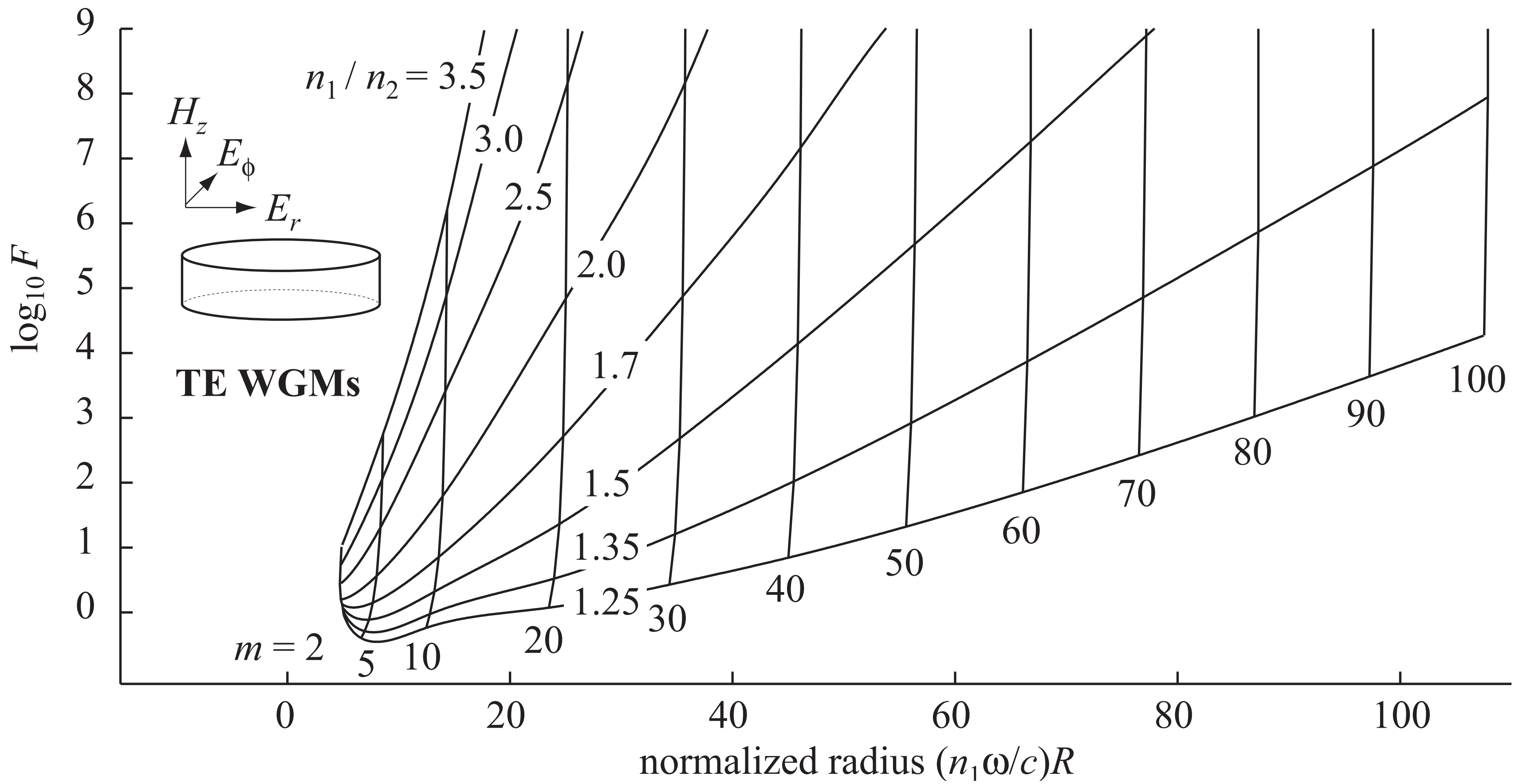
2) dispersion decreasing SCISSOR :



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.



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

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

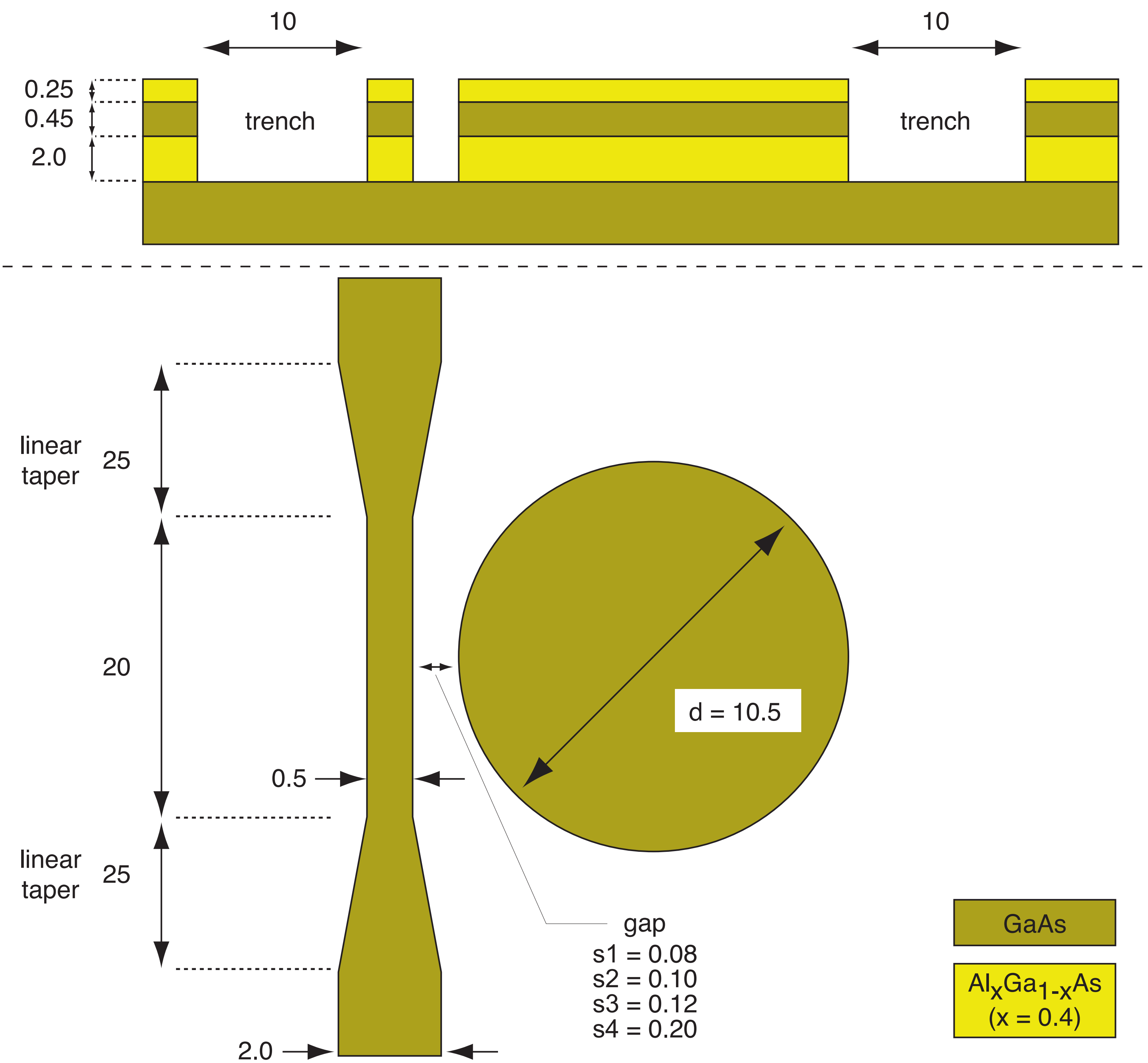
Nanofabrication

- Materials (artificial materials)
- Devices

(distinction?)

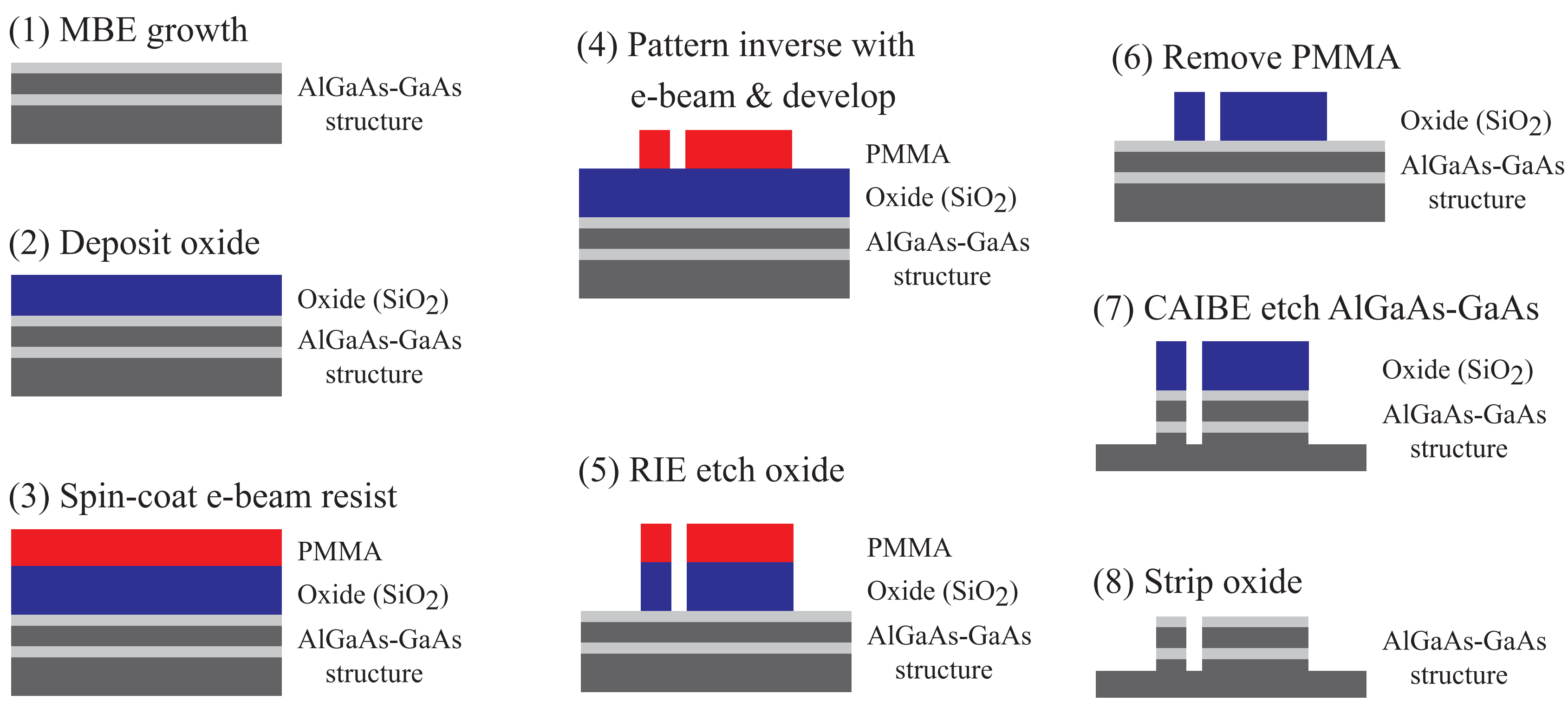
Microdisk Resonator Design

(Not drawn to scale)
All dimensions in microns



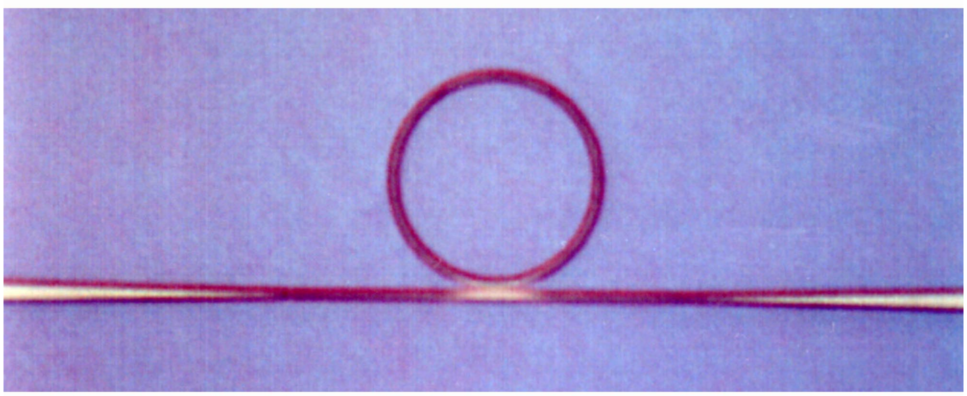
J. E. Heebner and R. W. Boyd

Photonic Device Fabrication Procedure



RWB - 10/4/01

Nonlinear Optical Loop-De-Loop



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

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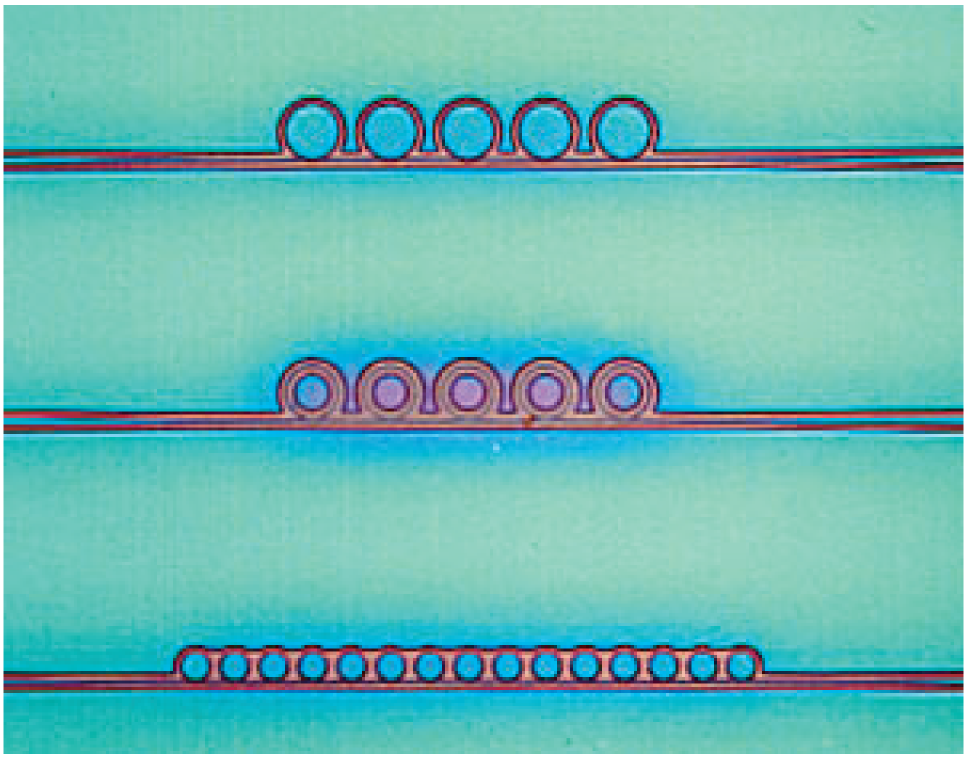

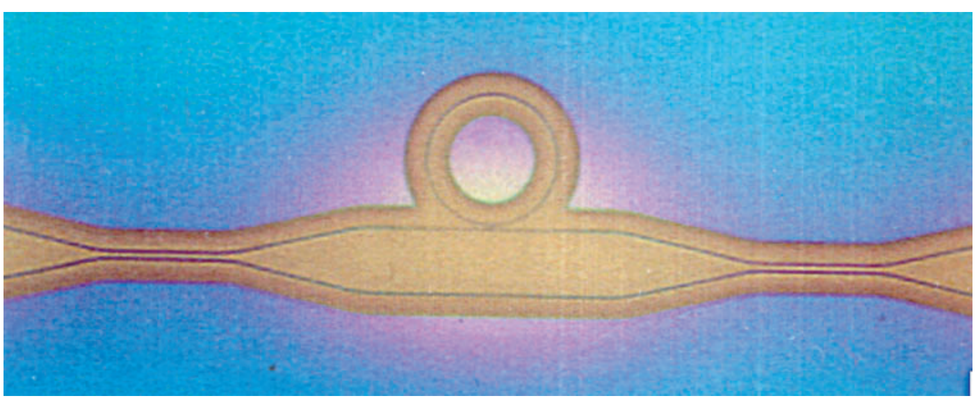

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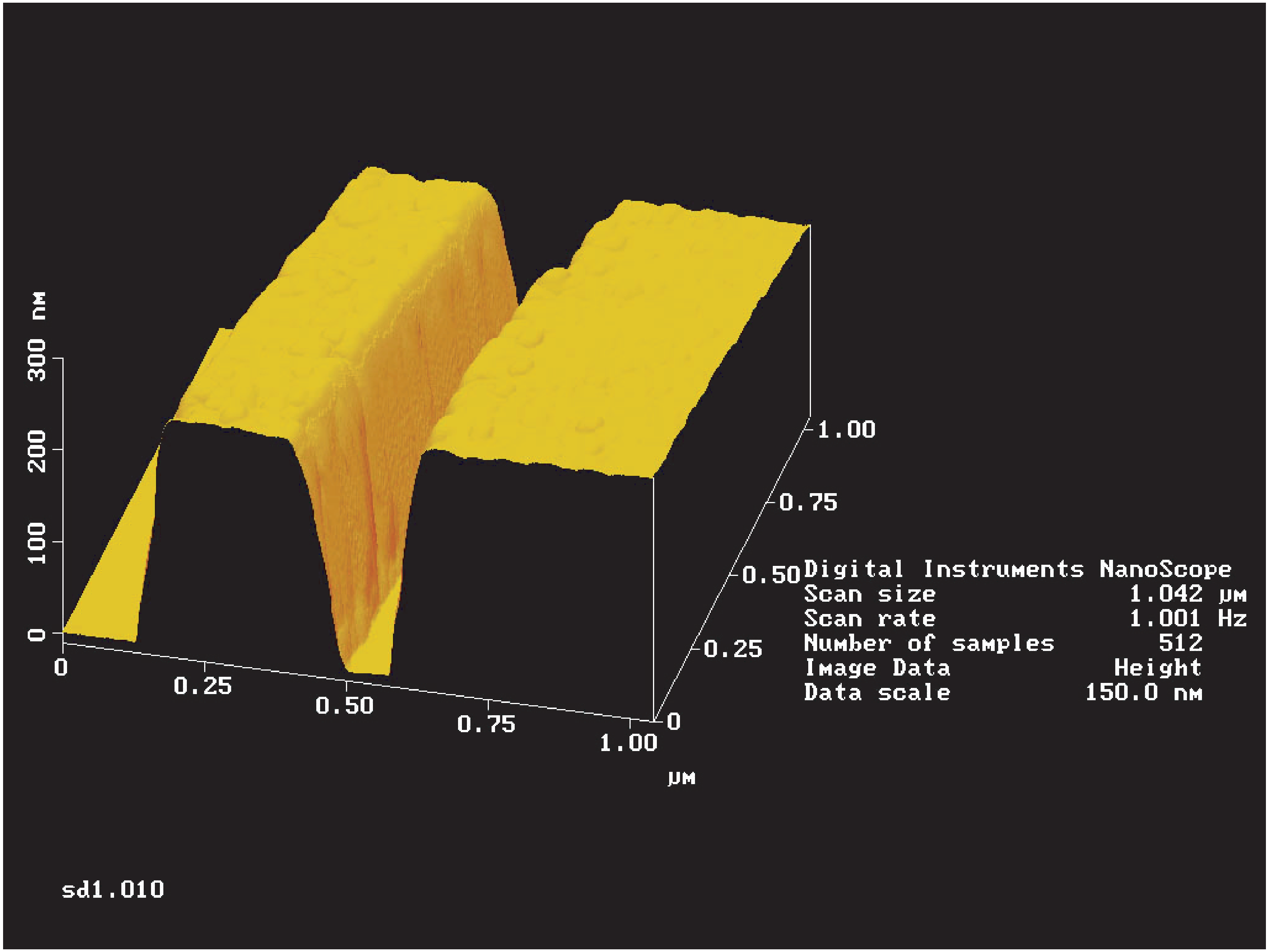
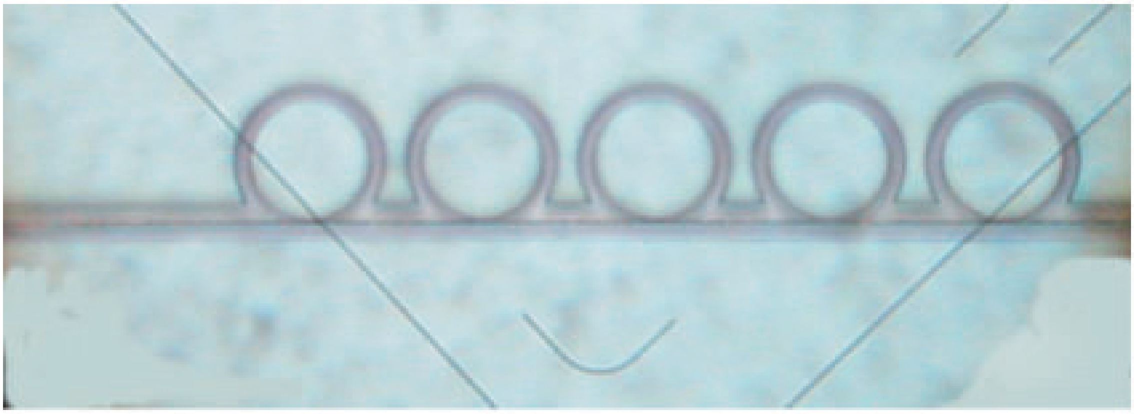
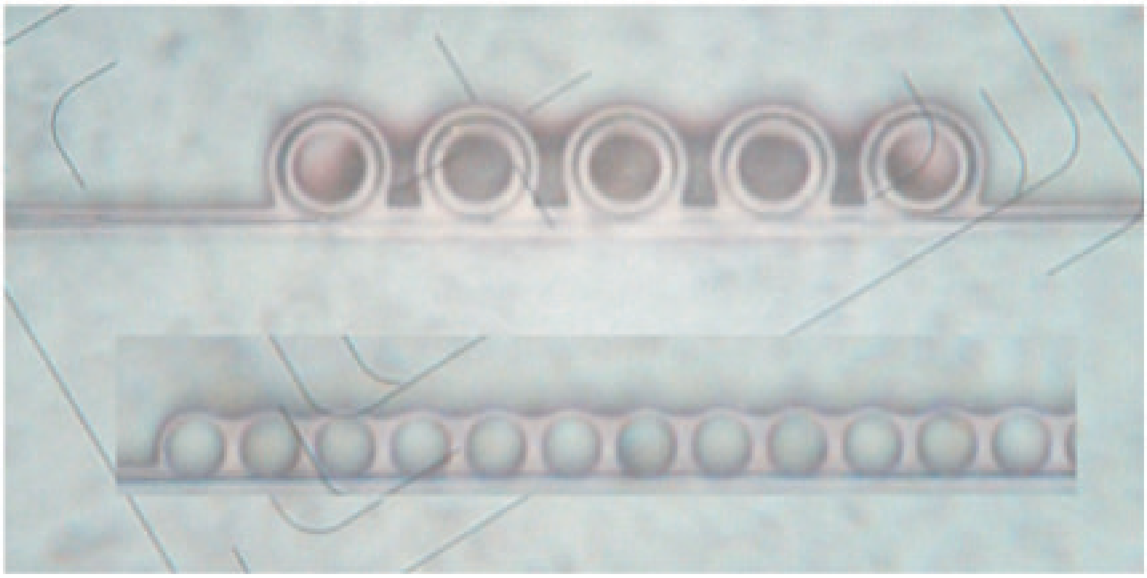


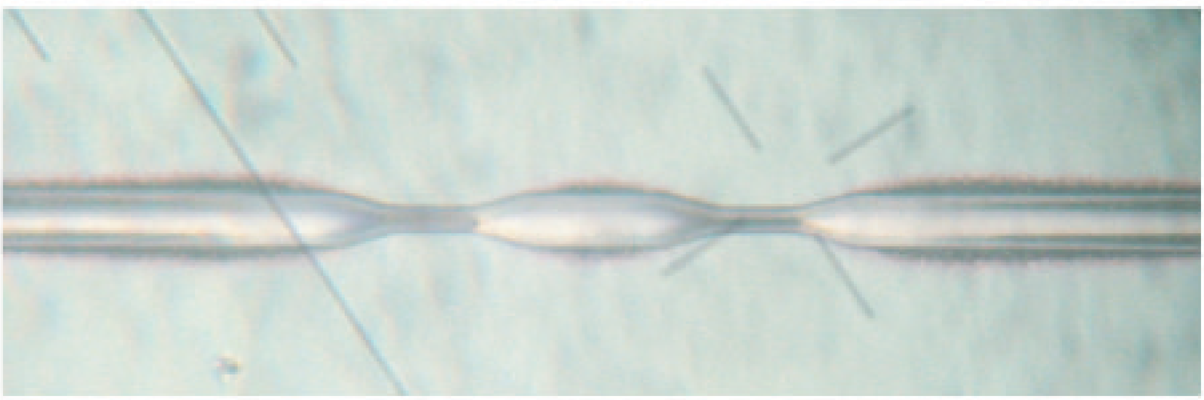
300
200
100
0
nm
0
0.25
0.50
0.75
1.00
μm
0
0.25
0.50
0.75
1.00
Digital Instruments NanoScope
Scan size 1.042 μm
Scan rate 1.001 Hz
Number of samples 512
Image Data Height
Data scale 150.0 nm
sd1.010

a

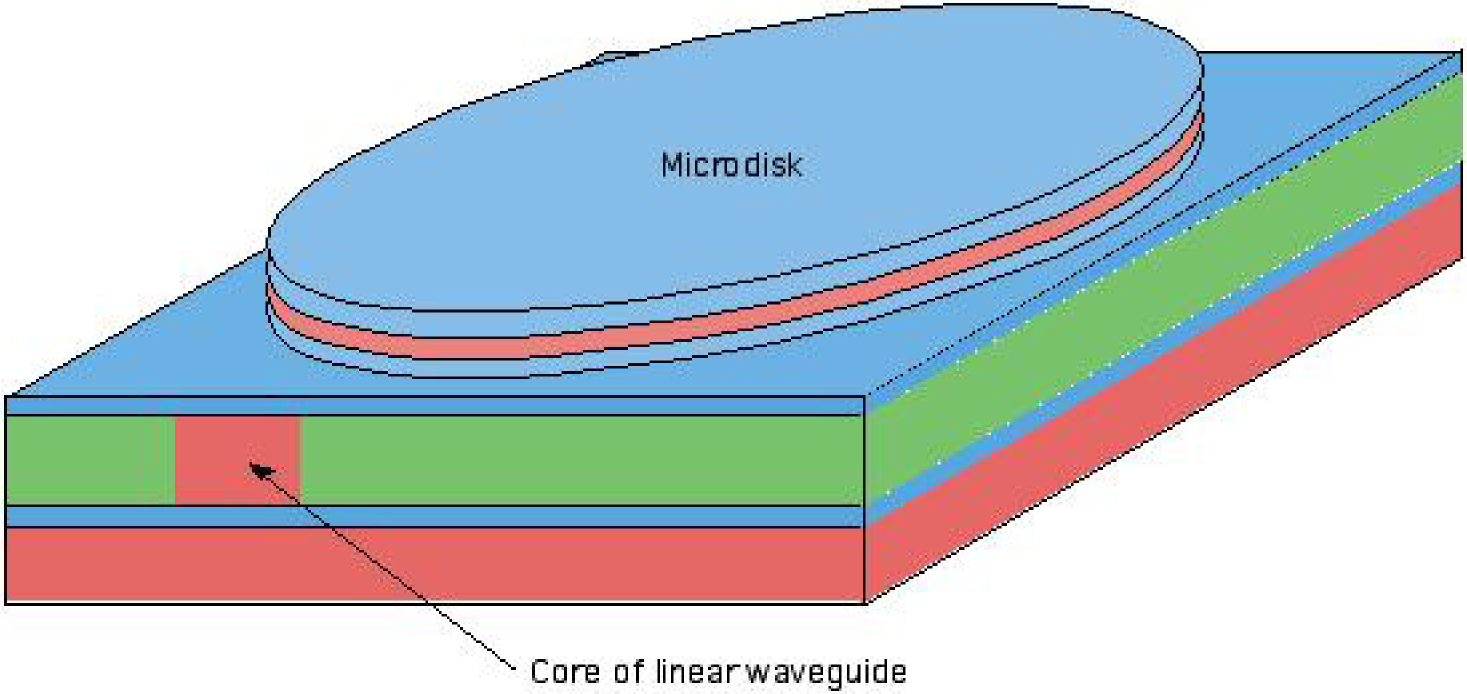
Photonic Devices in GaAs/AlGaAs







Microdisk Resonator Design / Vertically Coupled Structure



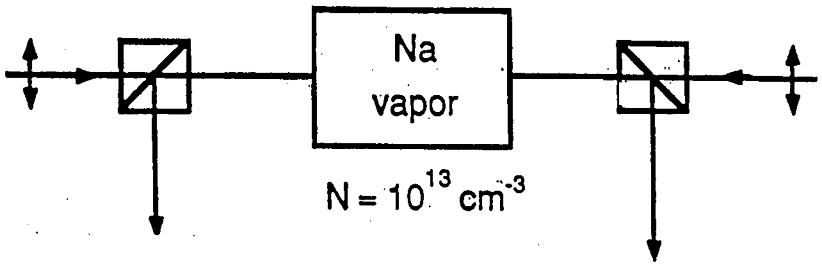
High refractive index (GaAs or AlGaAs superlattice)
Medium refractive index (intermixed superlattice)
Low refractive index (AlGaAs)

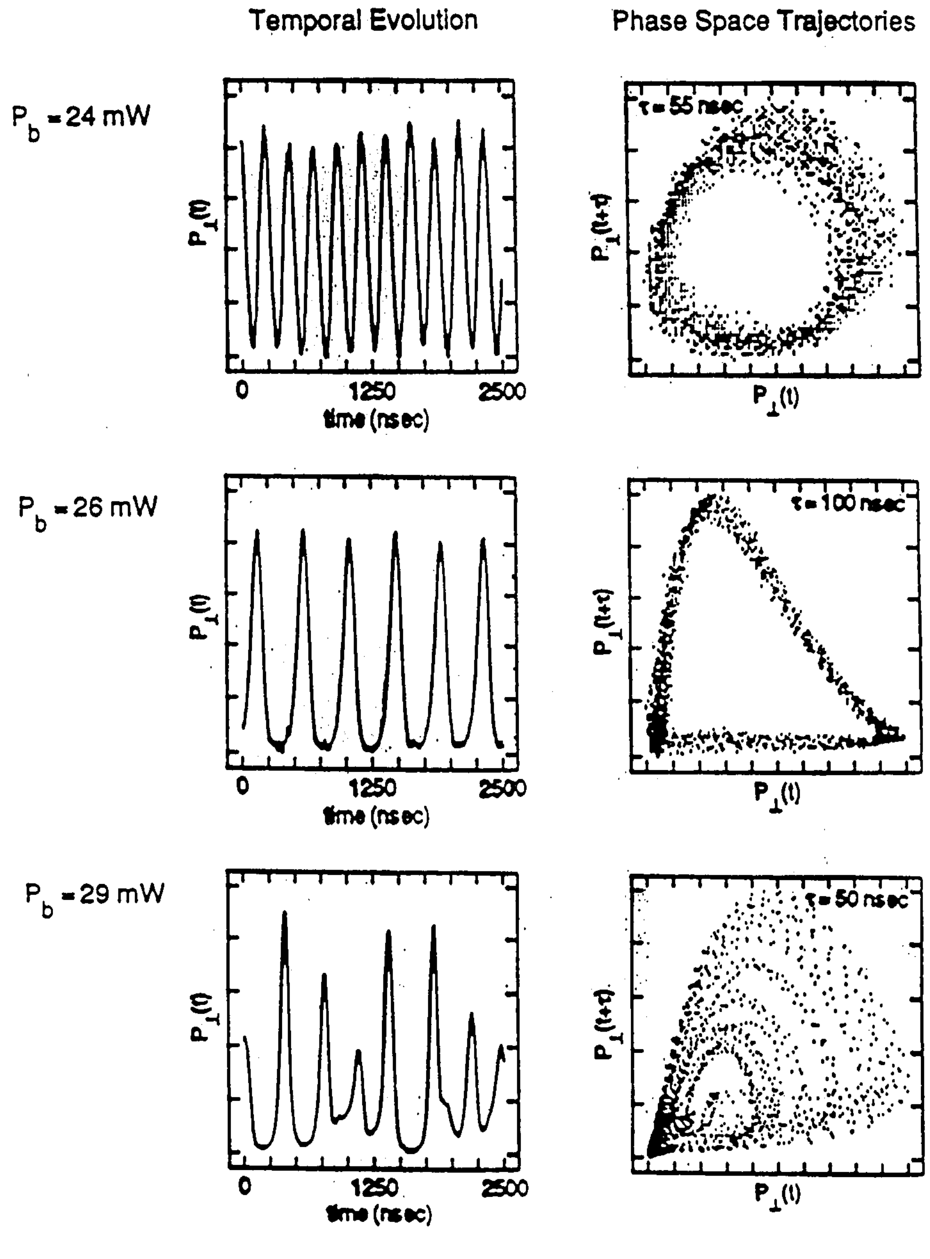
GWW - 10/8/01

Some Underlying Issues in Nonlinear Optics

- Self-Assembly/Self-Organization in Nonlinear Systems
- Stability vs. Instability (and Chaos) in Nonlinear Systems

Chaos in Sodium Vapor

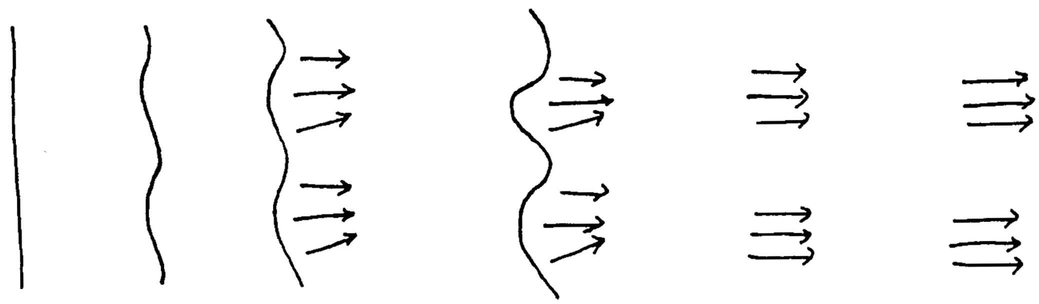




PRL 58, 2432 (1987); 61, 1827 (1988); 64 1721 (1990).

Laser Beam Filamentation

Spatial growth of wavefront perturbations



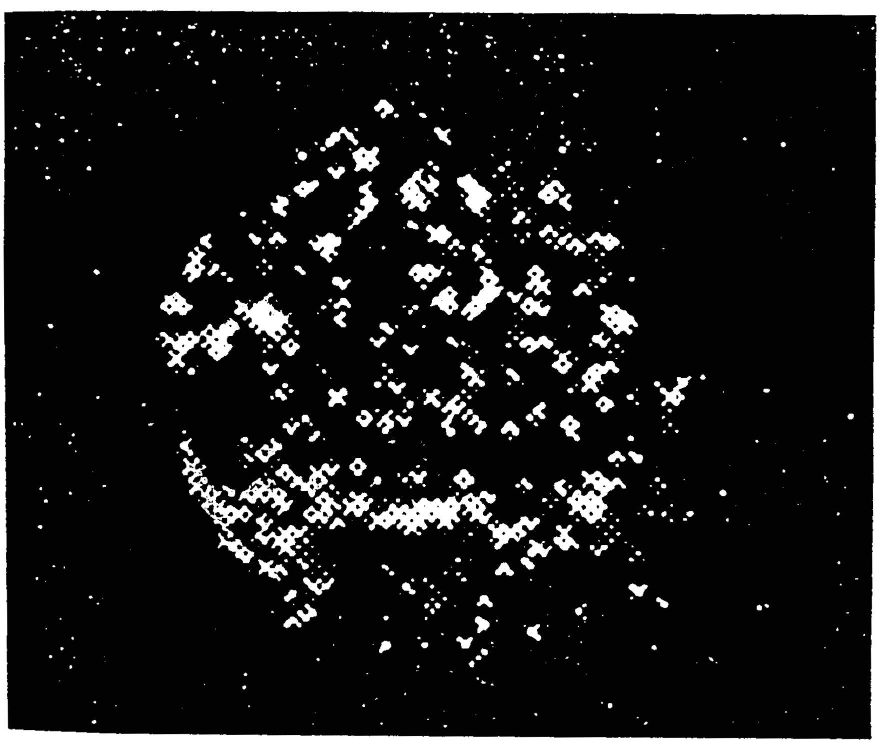
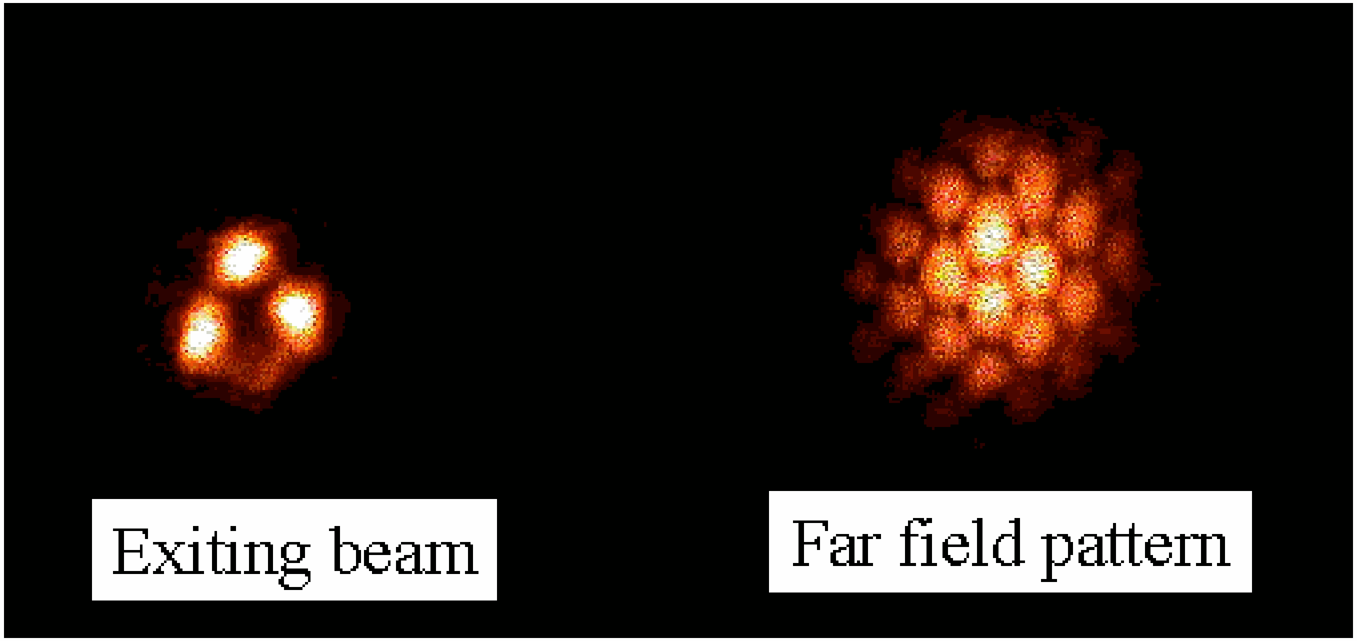


Fig. 17.2 Image of small-scale filaments at the exit windows of a CS_2 cell created by self-focusing of a multimode laser beam. [After S. C. Abbi and H. Mahr, *Phys. Rev. Lett.* 26, 604 (1971).]

Honey Comb Pattern Formation

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

Output from cell with single gaussian beam input



Quantum image?

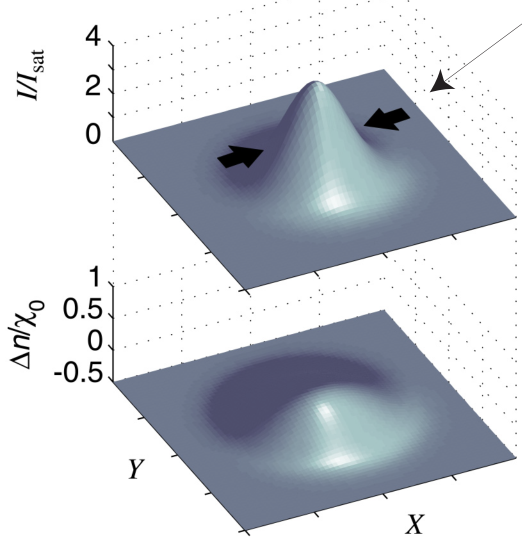
Input power 150 mW
Input beam diameter 0.22 mm
$\lambda = 588.995$ nm

Sodium vapor cell
$T = 220°$ C

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



$$n = 1 + \ n$$

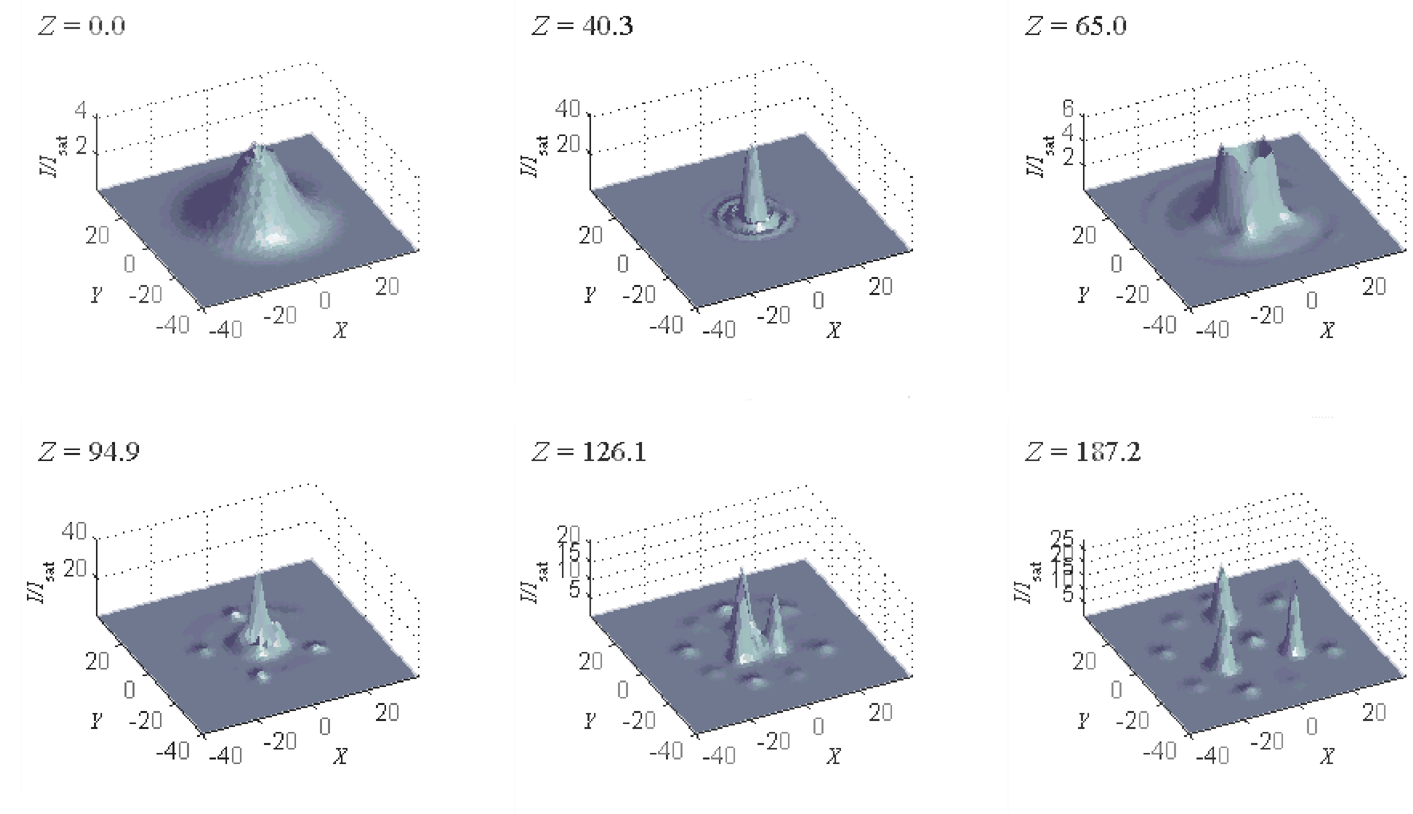
$$1 + \frac{1}{2}\,\frac{0}{1 + I/I_{sat}}$$

induced refractive index is like a positive lens

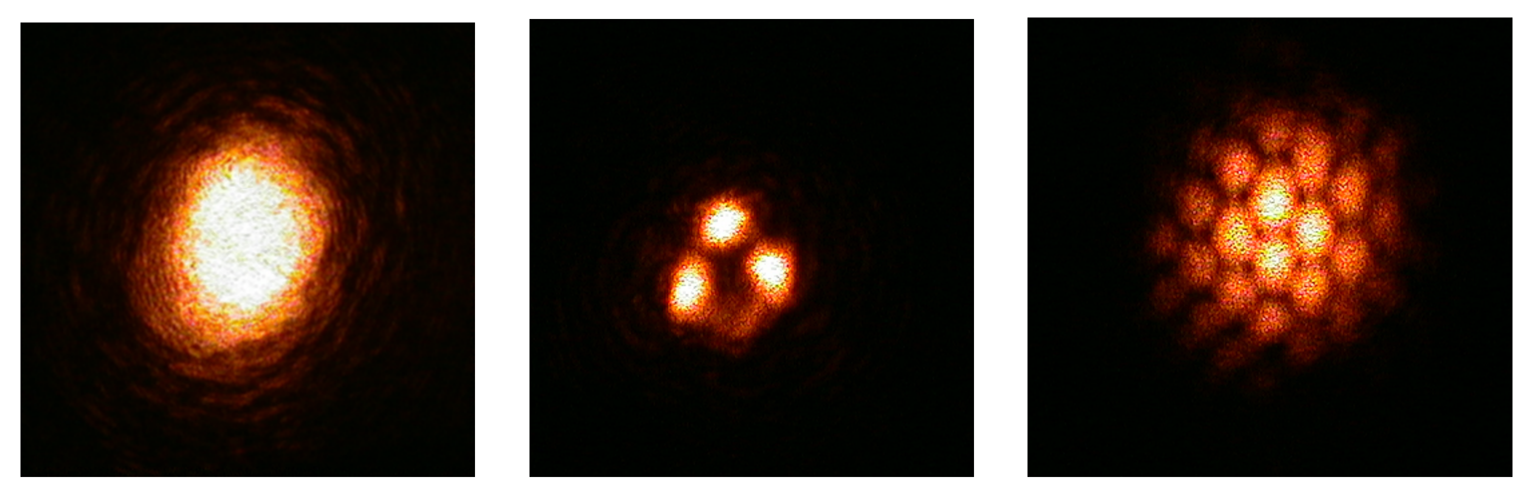
Since light refracts in the direction of increasing index, in a medium with negative saturable nonlinearity it refracts toward regions of higher intensity. This causes smooth beams to narrow or self-focus. But it also tends to destabilize a beam as small amplitude fluctuations grow due to local self-focusing.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



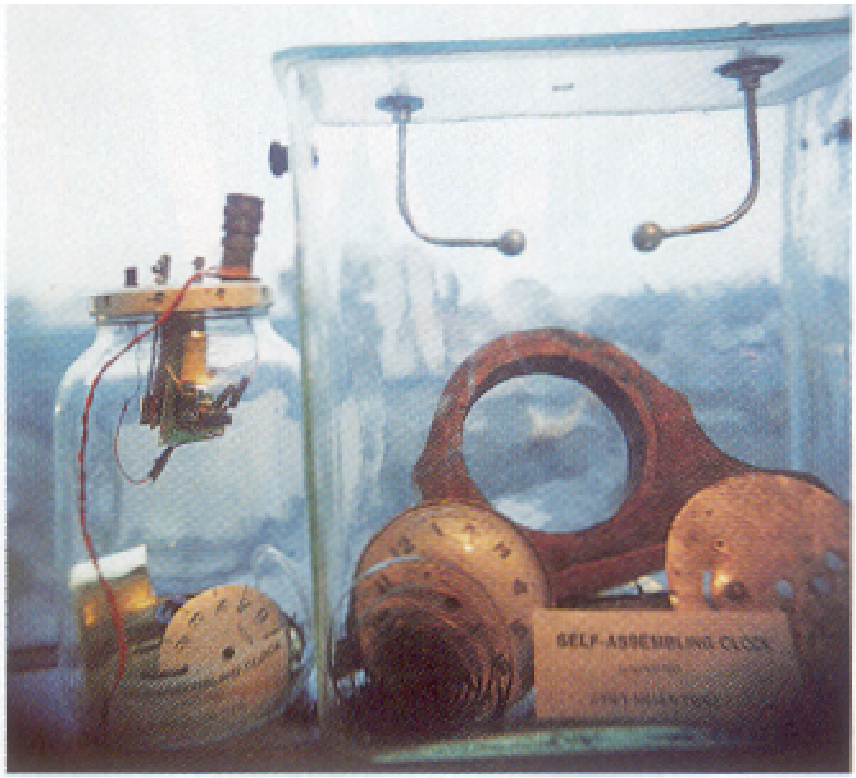
Experimental observation of spontaneous break-up resulting in a striking far-field pattern:



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