

# Nonlinear Optics: Past Successes and Future Challenges

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From Photonics Spectra Magazine

## **Making Ottawa the world's photonics center**

OTTAWA, Ontario, Canada –

A \$55 million photonics center that was expected to break ground on the University of Ottawa campus in March will help fulfill the school's goal to make Ottawa the global hub of photonics.

Our research interests include

Nanophotonics

Plasmonics

Photonic crystals

Photonic device

Applications of slow and fast light

Quantum nonlinear optics

Optical methods for quantum information





# Why Study Nonlinear Optics?

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It is good fundamental physics.

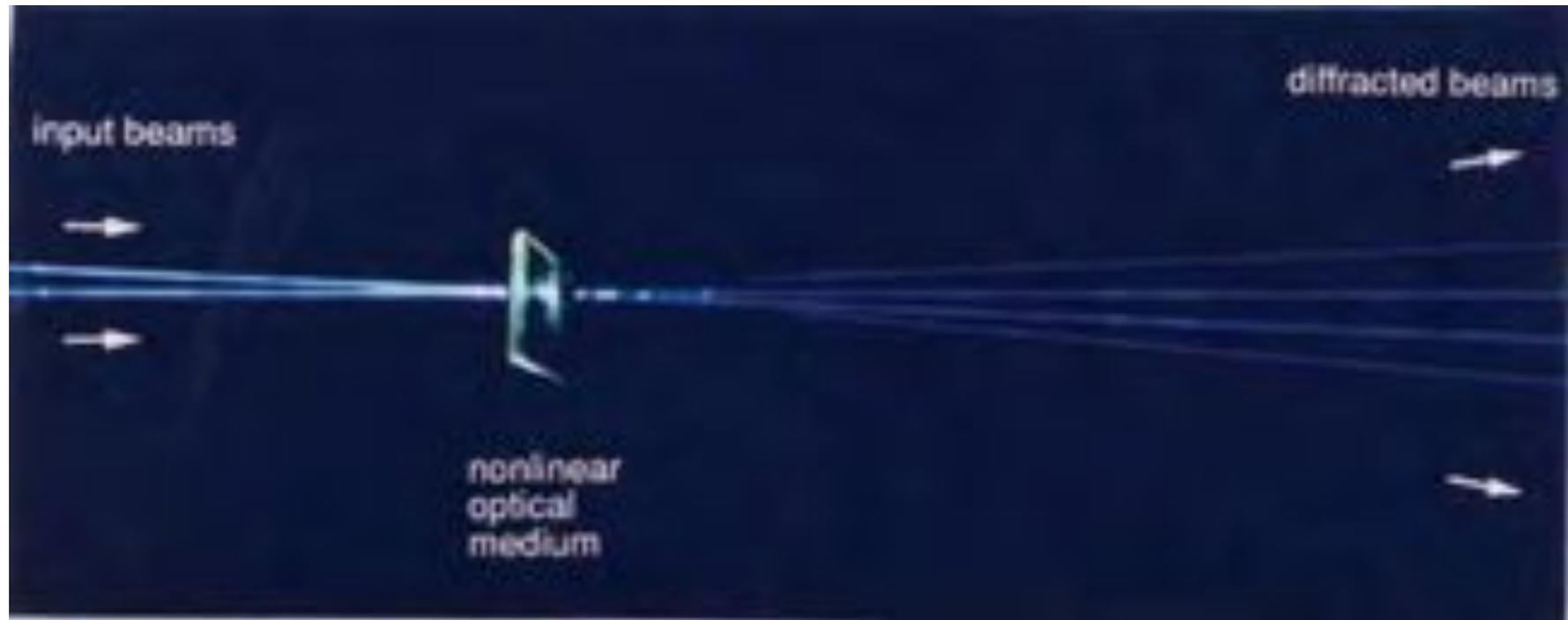
It leads to important applications.

It is a lot of fun.

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Demonstrate these features with examples in remainder of talk.

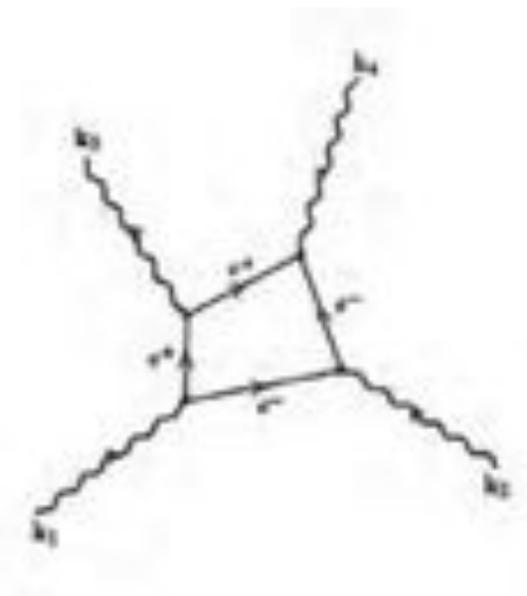
# Nonlinear Optics and Light-by-Light Scattering



The elementary process of light-by-light scattering has never been observed in vacuum, but is readily observed using the nonlinear response of material systems.

Nonlinear material is fluorescein-doped boric acid glass (FBAG)

$$n_2(\text{FBAG}) \approx 10^{14} n_2(\text{silica}) \quad [\text{But very slow response!}]$$



M. A. Kramer, W. R. Tompkin, and R. W. Boyd, Phys. Rev. A, 34, 2026, 1986.

W. R. Tompkin, M. S. Malcuit, and R. W. Boyd, Applied Optics 29, 3921, 1990.

# Brief Introduction to Nonlinear Optics

# Nonlinear Optics

THIRD EDITION



Robert W. Boyd

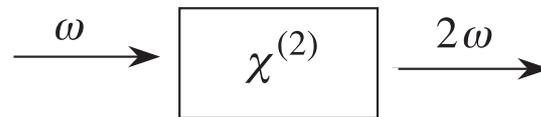
# Simple Formulation of the Theory of Nonlinear Optics

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

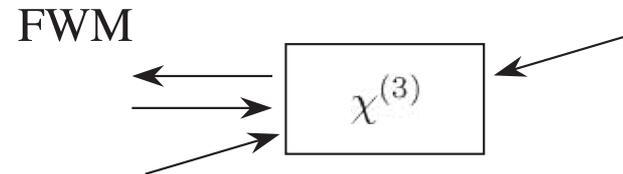
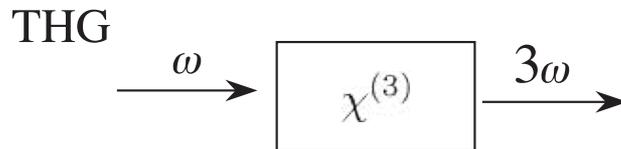
Here  $P$  is the induced dipole moment per unit volume and  $E$  is the field amplitude

$\chi^{(1)}$  describes linear optics, e.g., how lenses work: 

$\chi^{(2)}$  describes second-order effects, e.g., second-harmonic generation (SHG)



$\chi^{(3)}$  describes third-order effects such as third-harmonic generation, four-wave mixing, and the intensity dependence of the index of refraction.



NL index

$$n = n_0 + n_2 I \quad \text{where} \quad n_2 = \frac{3}{4n_0^2 \epsilon_0 c} \chi^{(3)}$$

# Timeline – The Early Days of Nonlinear Optics

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1958 - Schawlow and Townes; optical maser (laser) proposed

1960 - Maiman, ruby laser demonstrated

1961 - Franken, Hill, Peters, and Weinreich, second-harmonic generation (SHG) observed

1962 - Armstrong, Bloembergen, Ducuing, and Pershan; systematic formulation of NLO

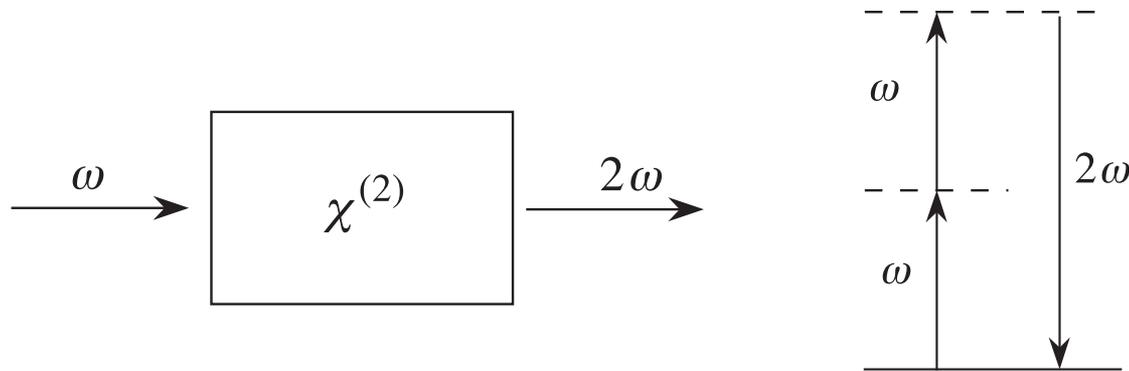
1962 - McClung, Hellwarth, Woodbury, and Ng, stimulated Raman scattering (SRS)

1964 - Chiao, Townes, and Stoicheff, stimulated Brillouin scattering (SBS)

1964 - Chiao, Garmire, and Townes, self-trapping of light

# Nonlinear Optics: Past Successes

# Second-Harmonic Generation: The Prototypical Nonlinear Optical Process



\$1 online

VOLUME 7, NUMBER 4

PHYSICAL REVIEW LETTERS

AUGUST 15, 1961

## GENERATION OF OPTICAL HARMONICS\*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich

The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan

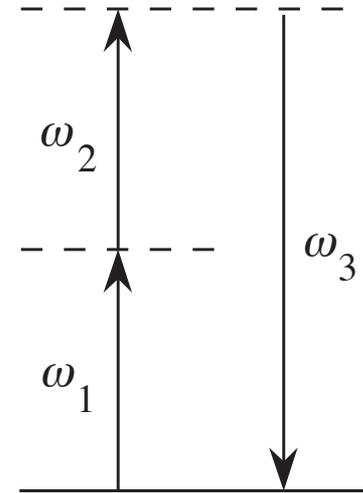
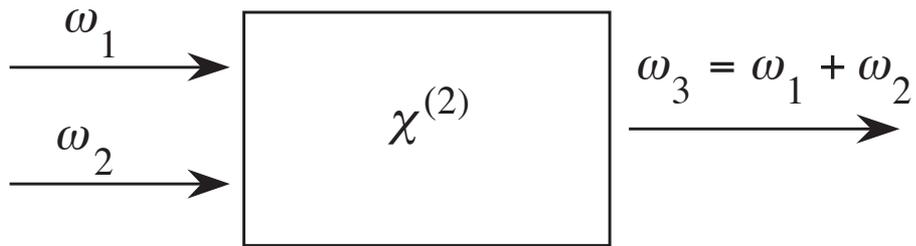
(Received July 21, 1961)



# Some Fundamental Nonlinear Optical Processes: II

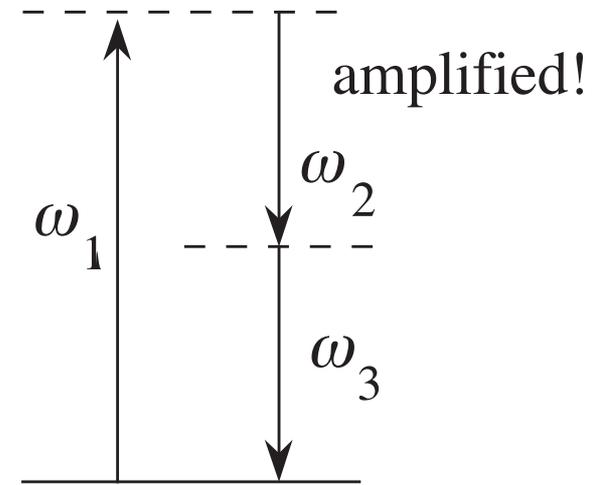
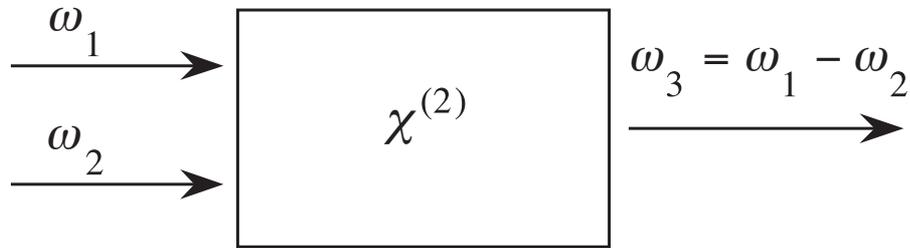
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## Sum-Frequency Generation



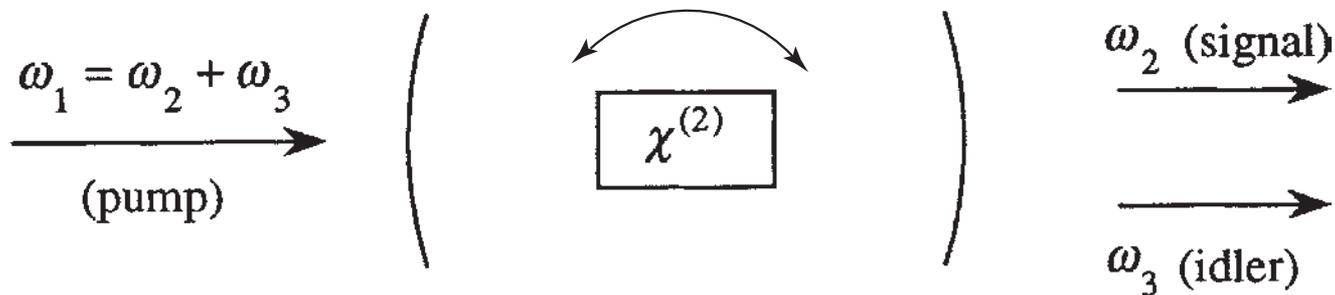
# Difference-Frequency Generation and Optical Parametric Amplification

## Difference-Frequency Generation



## Optical Parametric Amplification

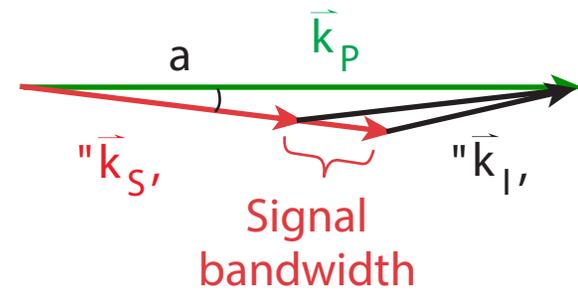
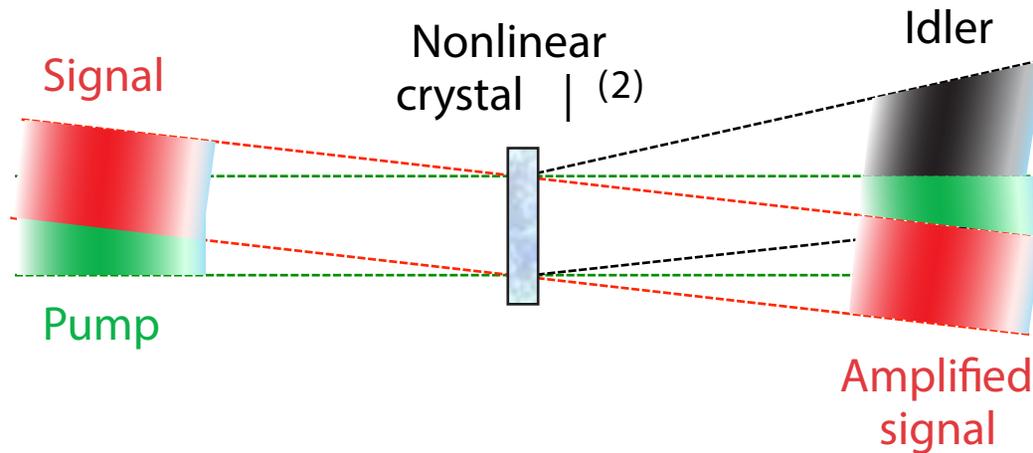
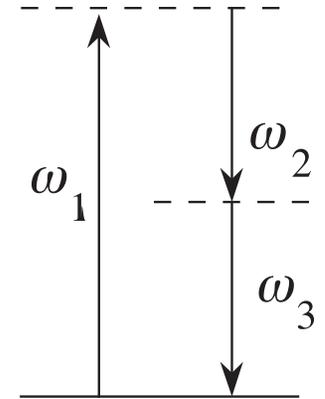
## Optical Parametric Oscillator (very broadly tunable)



# Optical Parametric Amplification Can Amplify Extremely Broadband Pulses

Can amplify extremely short laser pulses or broadband chirped pulses.

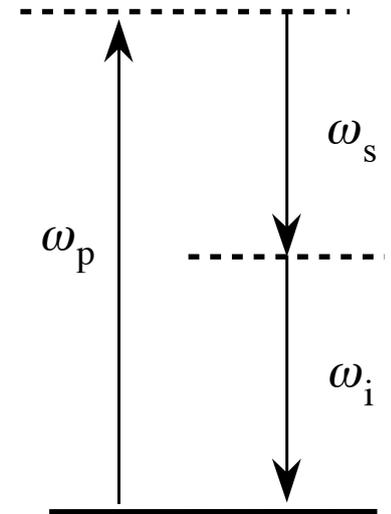
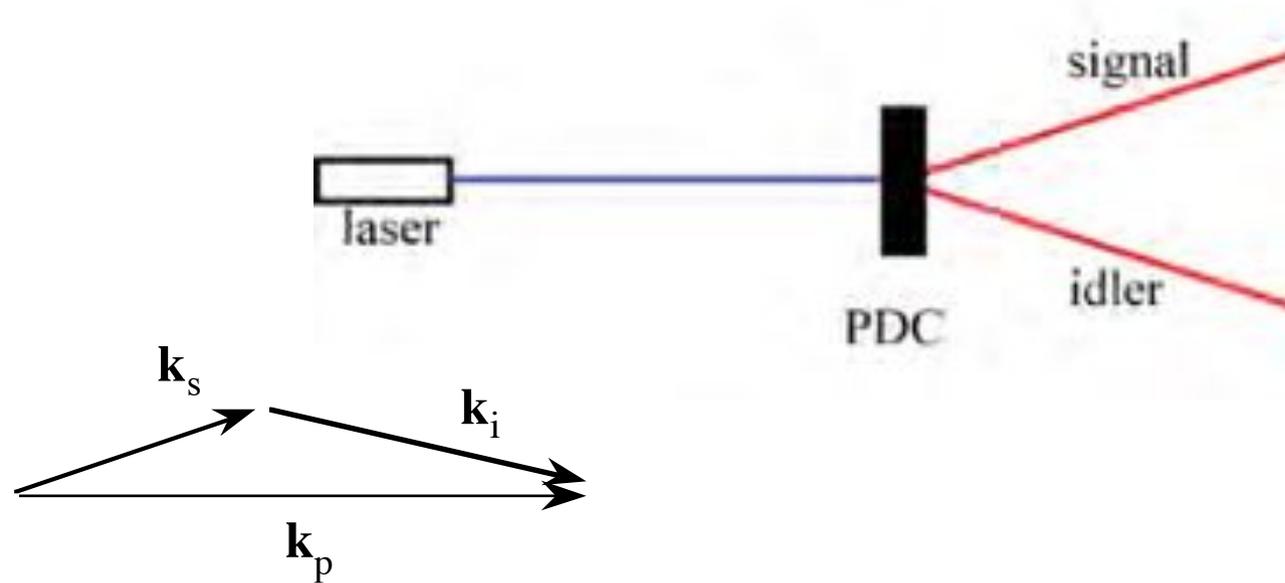
Goal: Design laser source capable of reaching focused intensities as large as  $10^{24}$  W/cm<sup>2</sup>.



Work of Jake Bromage and others at U. Rochester LLE.

See also Lozhkarev et al. Laser Phys. Lett. 4, 421 (2007) and Y. Tang et al. Opt. Lett. 33, 2386 (2008).

# Parametric Downconversion: A Source of Entangled Photons



The signal and idler photons are entangled in:

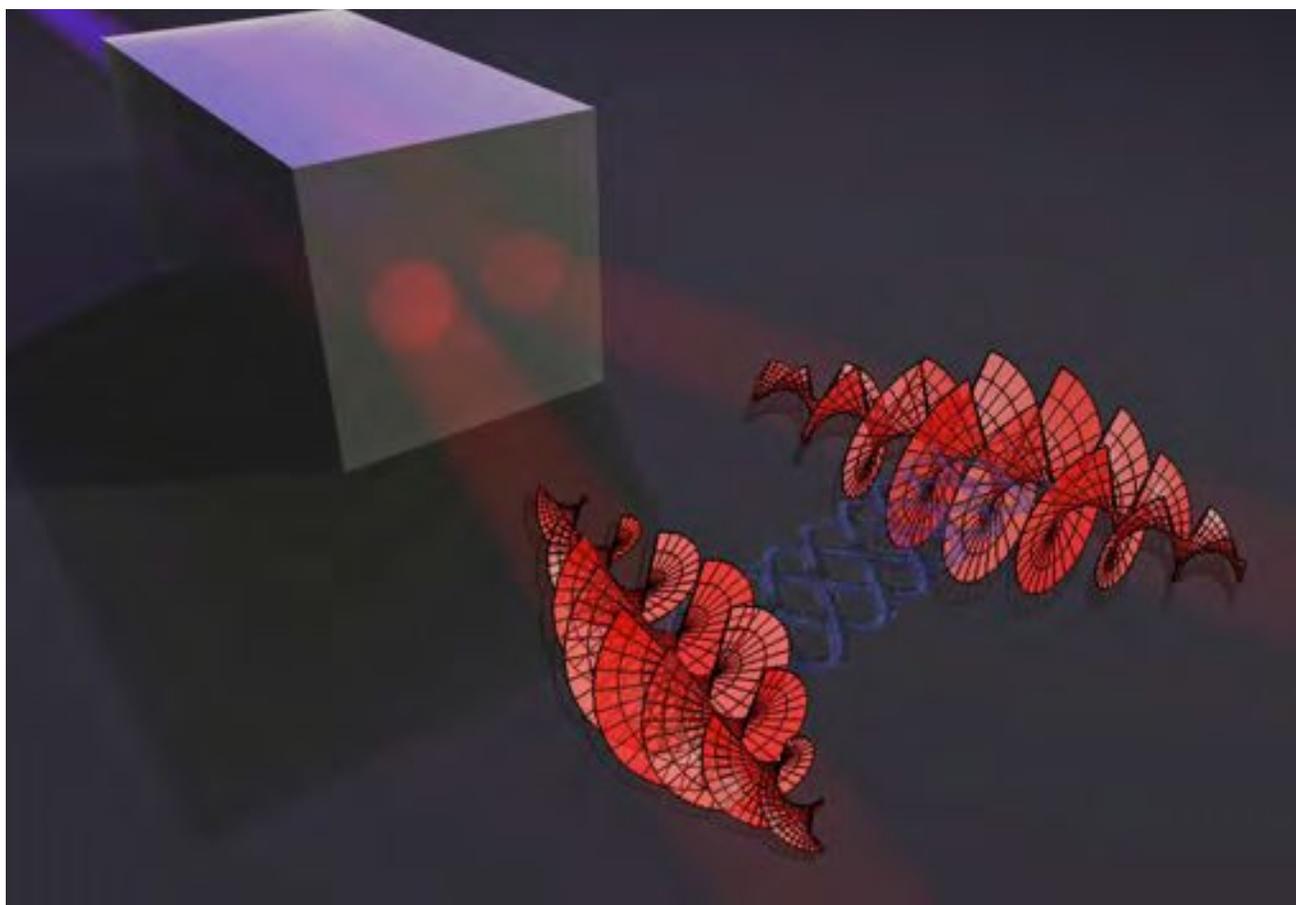
- (a) polarization
- (b) time and energy
- (c) position and transverse momentum
- (d) angular position and orbital angular momentum

Entanglement is important for:

- (a) Fundamental tests of QM (e.g., nonlocality)
- (a) Quantum technologies (e.g., secure communications)

# Nonlinear Optics Leads to Creation of Entangled Photons

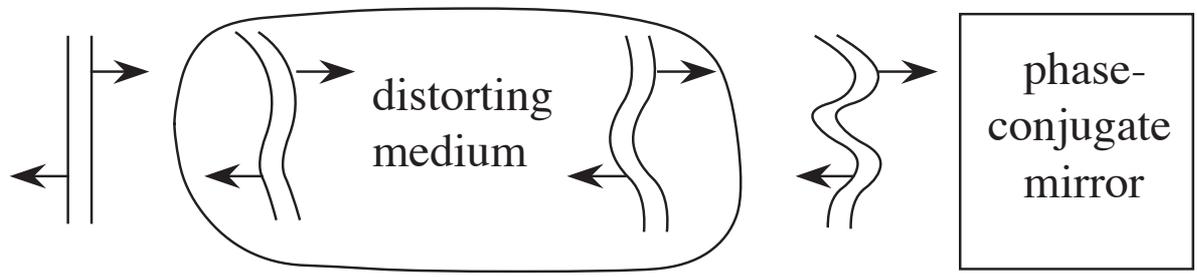
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Quantum Correlations in Optical Angle-Orbital Angular Momentum Variables,  
Leach et al., Science 329, 662 (2010).

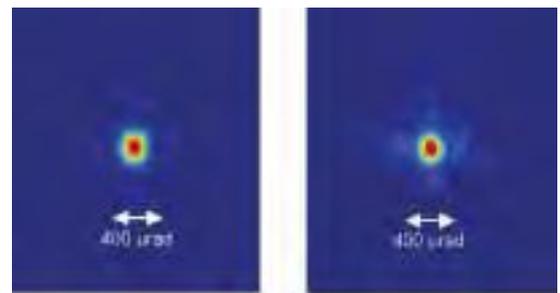
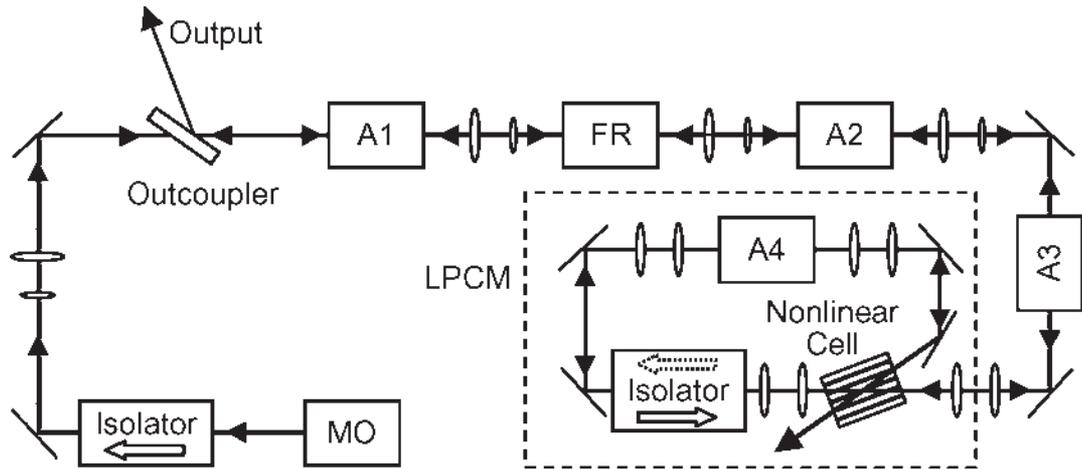
# Optical Phase Conjugation: A Nonlinear Optics Success Story

- A phase conjugate mirror (a nonlinear optical device) can remove the influence of aberrations in double pass.



(Zeldovich, Pilipetsky, Shkunov, Yariv, Hellwarth, Fisher, 1980s).

- Phase conjugation is extremely useful in high power laser systems  
 2-kW average power phase-conjugate master oscillator power amplifier



near-diffraction-limited output

Zakharenkov, Clatterbuck, Shkunov, Betin, Filgas, Ostby, Strohkendl, Rockwell, and Baltimore, IEEE JSTQE (2007).

# Intense Field and Attosecond Physics

PHYSICAL REVIEW LETTERS

PHYSICAL REVIEW LETTERS

13 MARCH

## Above Threshold Ionization Beyond the High Harmonic Cutoff

K. J. Schafer,<sup>(1)</sup> Baorui Yang,<sup>(2)</sup> L. F. DiMauro,<sup>(2)</sup> and K. C. Kulander<sup>(1)</sup>

<sup>(1)</sup>Lawrence Livermore National Laboratory, Livermore, California 94550

<sup>(2)</sup>Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973

(Received 2 December 1992)

VOLUME 71, NUMBER 13

PHYSICAL REVIEW LETTERS

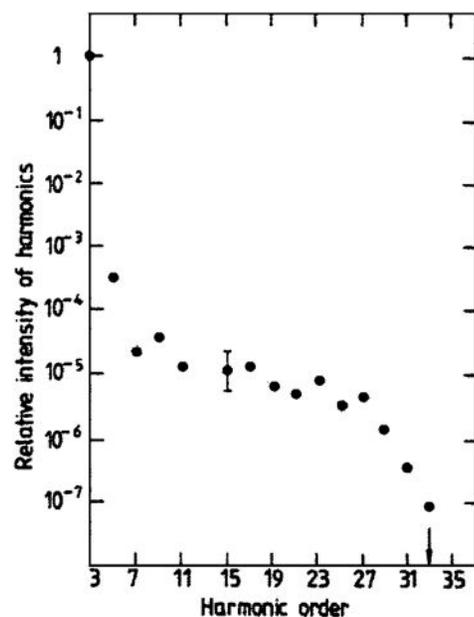
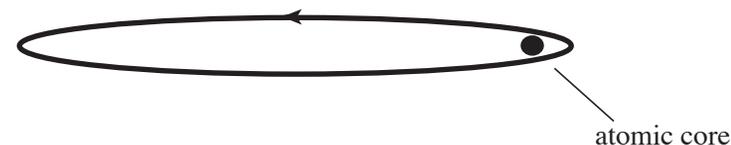
27 SEPTEMBER 1993

## Plasma Perspective on Strong-Field Multiphoton Ionization

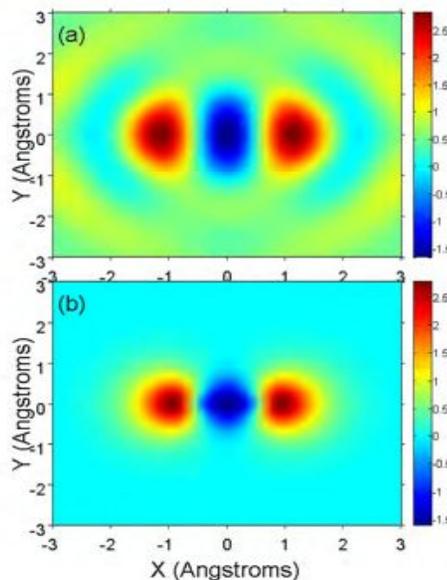
P. B. Corkum

National Research Council of Canada, Ottawa, Ontario, Canada K1A 0R6

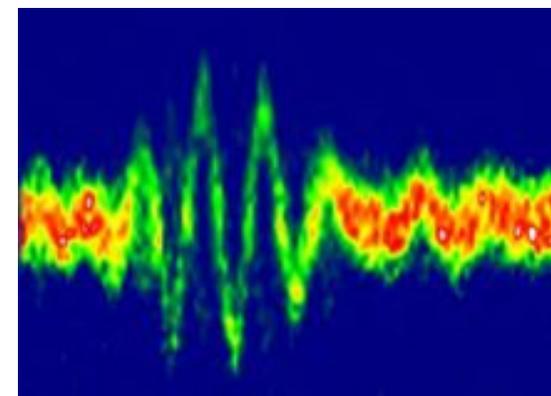
(Received 9 February 1993)



High-harmonic generation



Measuring the molecular nitrogen wavefunction



Attosecond pulses to sample a visible E-field

# Theory of nonlinear optics



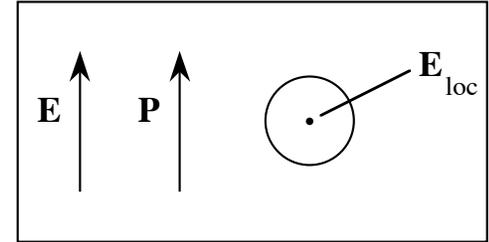
The Nobel Prize in Physics 1981

Nicolaas Bloembergen, Arthur L. Schawlow, Kai M. Siegbahn

# Local Field Effects in Nonlinear Optics

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Bloembergen (1962, 1965) showed that



$$\chi^{(3)}(\omega = \omega + \omega - \omega) = N\gamma^{(3)}|L(\omega)|^2[L(\omega)]^2.$$

where  $\gamma^{(3)}$  is the second hyperpolarizability and where

$$L(\omega) = \frac{\epsilon(\omega) + 2}{3}$$

For the typical value  $n = 2$ ,  $L = 2$ , and  $L^4 = 16$ . Local field effects can be very large in nonlinear optics! But can we tailor them for our benefit?

We have been developing new photonic materials with enhanced NLO response by using composite structures that exploit local field effects.

# Enhanced NLO Response from Layered Composite Materials

A composite material can display a larger NL response than its constituents!

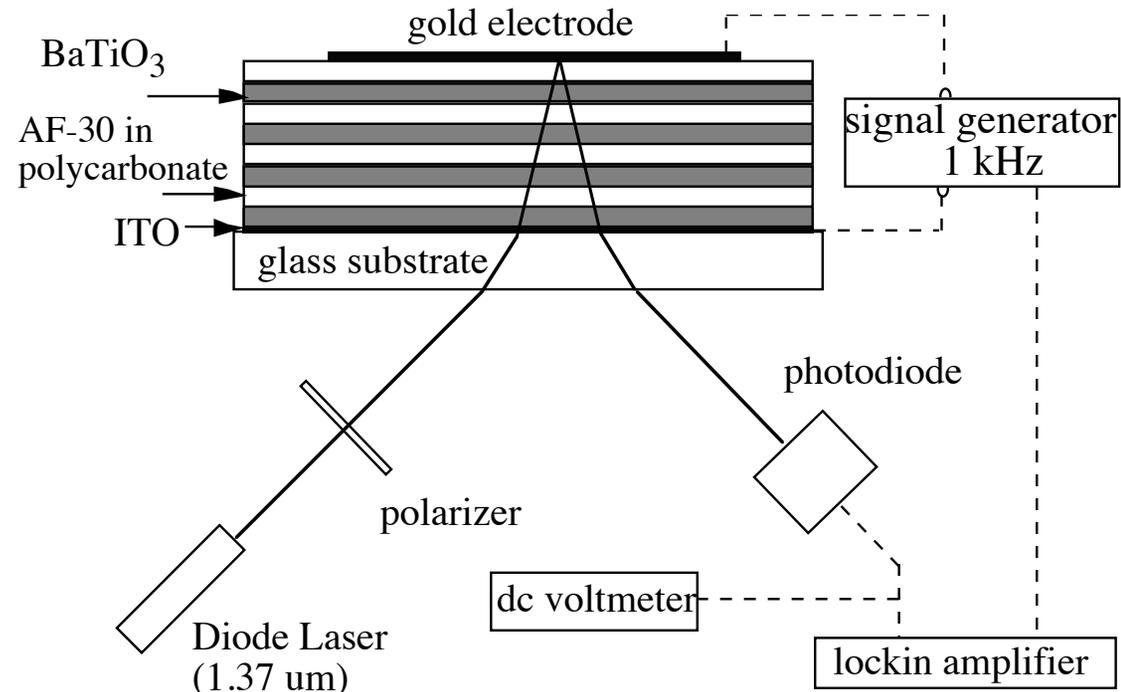
Alternating layers of  $\text{TiO}_2$  and the conjugated polymer PBZT.

$\nabla \cdot \mathbf{D} = 0$  implies that  $(\epsilon \mathbf{E})_{\perp}$  is continuous.

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

35% enhancement in  $\chi^{(3)}$

## Quadratic EO effect

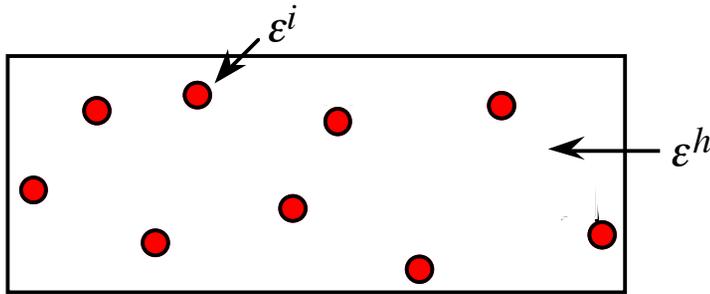


3.2 times enhancement!

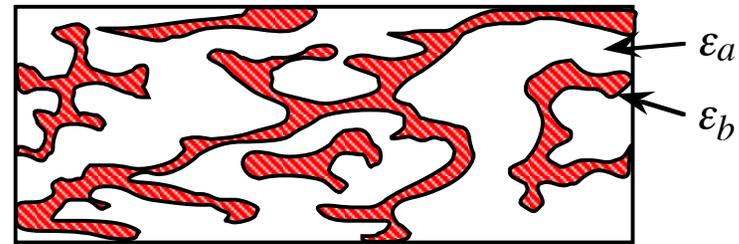
# Metamaterials and Nanocomposite Materials for Nonlinear Optics

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- Maxwell Garnett



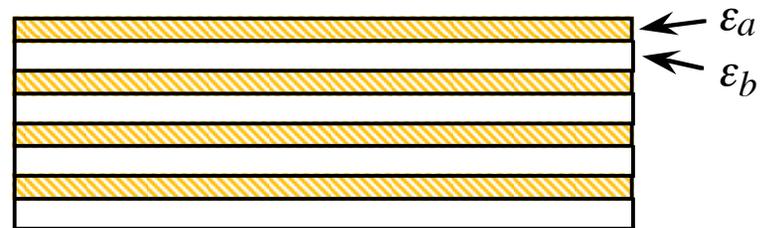
- Bruggeman (interdispersed)



- Fractal Structure



- Layered



- In each case, scale size of inhomogeneity  $\ll$  optical wavelength
- Thus all optical properties, such as  $n$  and  $\chi^{(3)}$ , can be described by effective (volume averaged) values

# Slow Light, Fast Light, and their Applications

# Controlling the Velocity of Light

## “Slow,” “Fast” and “Backwards” Light

– Light can be made to go:

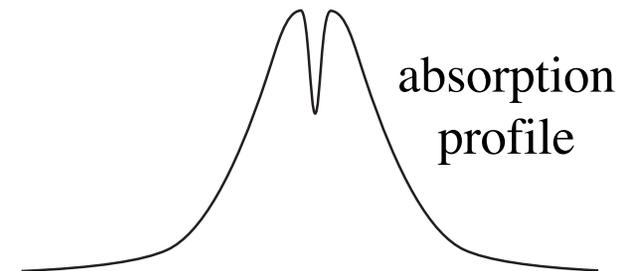
slow:  $v_g \ll c$  (as much as  $10^6$  times slower!)

fast:  $v_g > c$

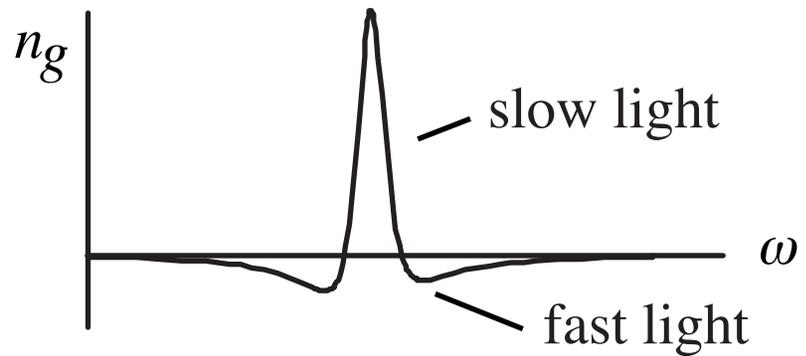
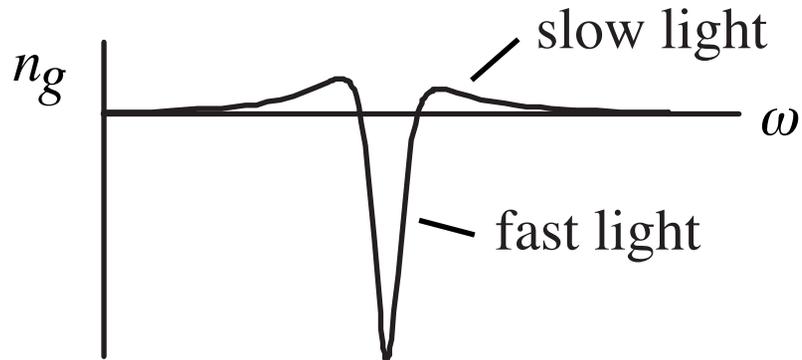
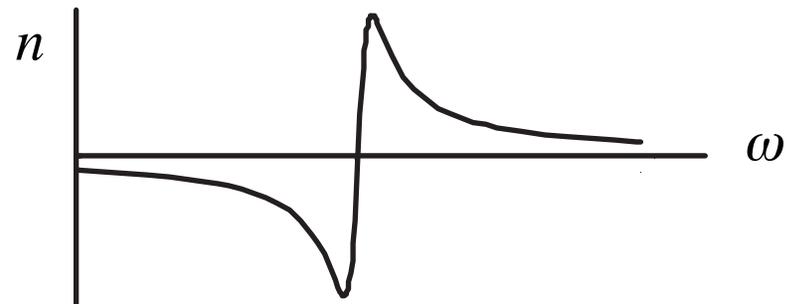
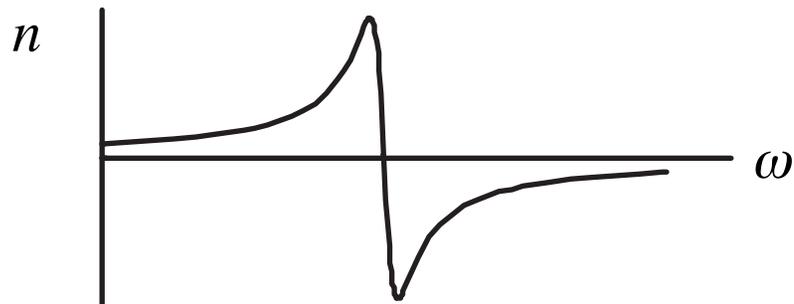
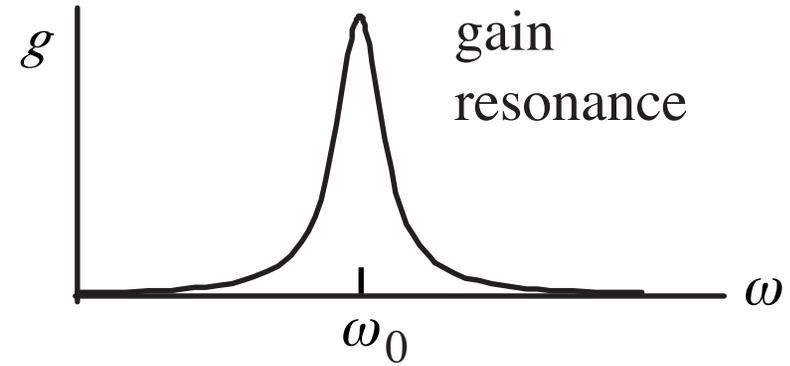
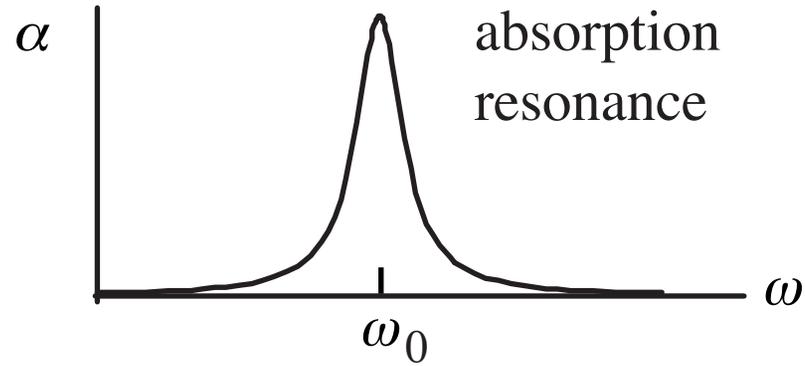
backwards:  $v_g$  negative

Here  $v_g$  is the group velocity:  $v_g = c/n_g$   $n_g = n + \omega (dn/d\omega)$

– Velocity controlled by structural or material resonances



# Slow and Fast Light Using Isolated Gain or Absorption Resonances



$$n_g = n + \omega (dn/d\omega)$$

# Light speed reduction to 17 metres per second in an ultracold atomic gas

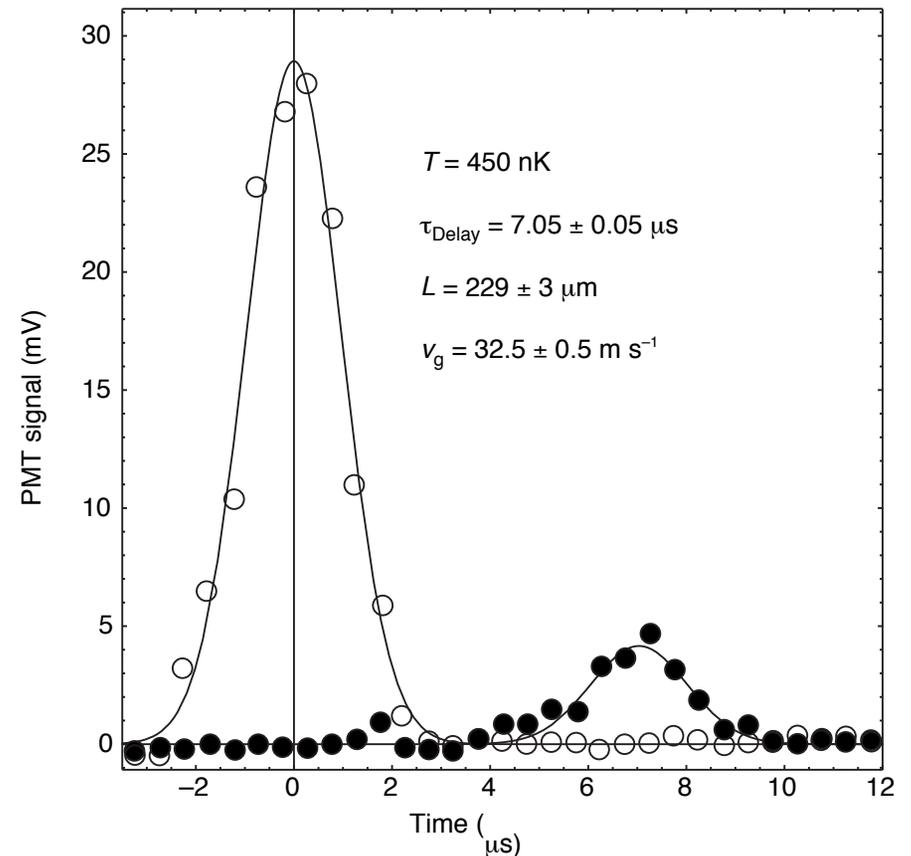
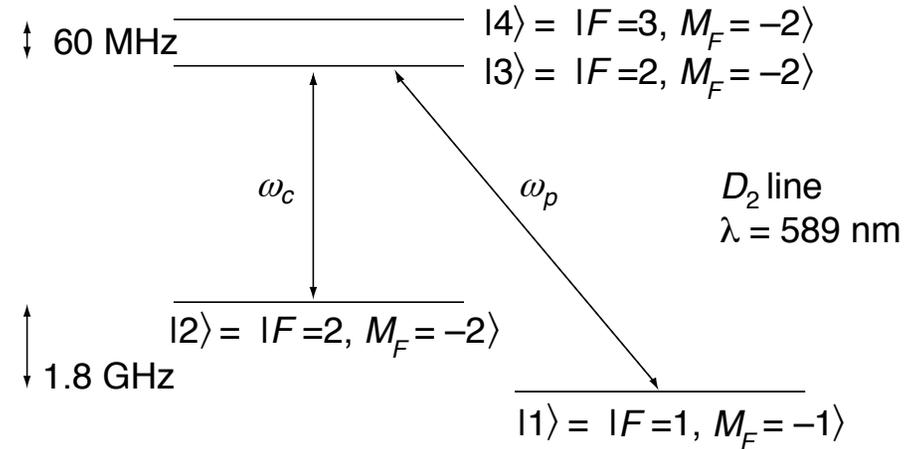
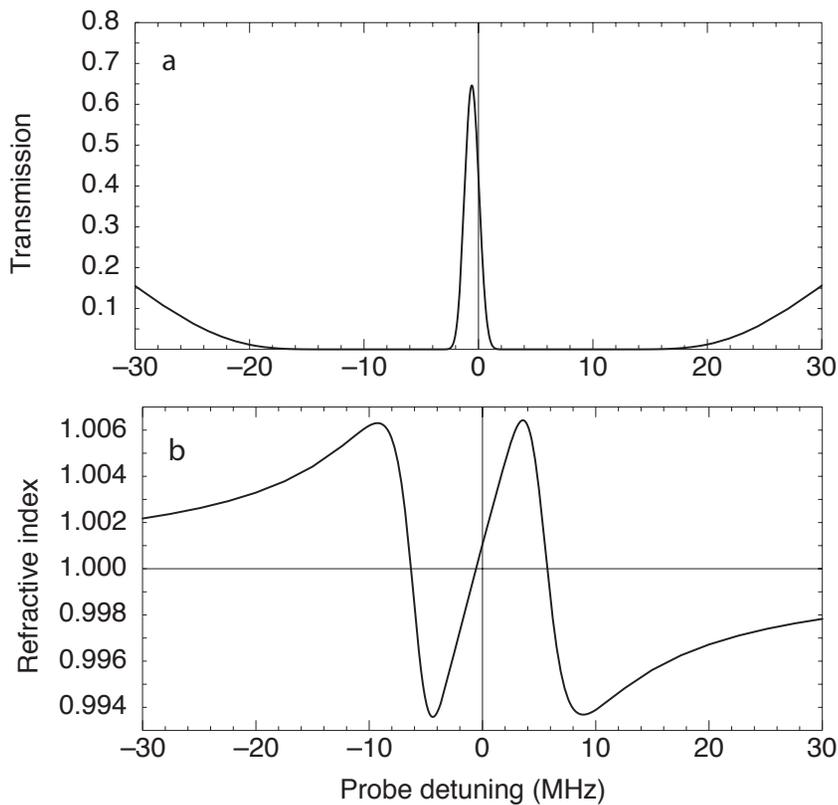
Lene Vestergaard Hau<sup>\*2</sup>, S. E. Harris<sup>3</sup>, Zachary Dutton<sup>\*2</sup>  
& Cyrus H. Behroozi<sup>\*§</sup>

<sup>\*</sup> Rowland Institute for Science, 100 Edwin H. Land Boulevard, Cambridge, Massachusetts 02142, USA

<sup>2</sup> Department of Physics, <sup>§</sup> Division of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA

<sup>3</sup> Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA

NATURE | VOL 397 | 18 FEBRUARY 1999 | www.nature.com



Note also related work by Chu, Wong, Welch, Scully, Budker, Ketterle, and many others

# Goal: Slow Light in a Room-Temperature Solid-State Material

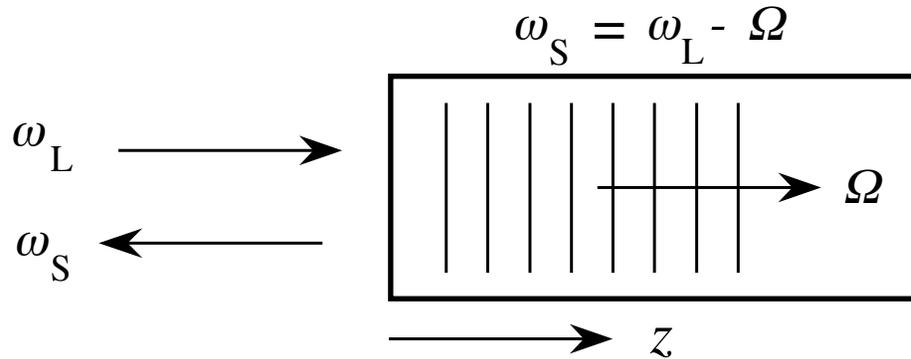
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Crucial for many real-world applications

We have identified two preferred methods for producing slow light

- (1) Slow light *via* coherent population oscillations (CPO)
- (2) Slow light *via* stimulated Brillouin scattering (SBS)

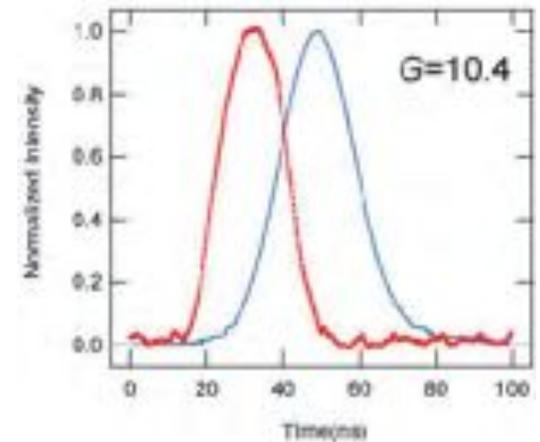
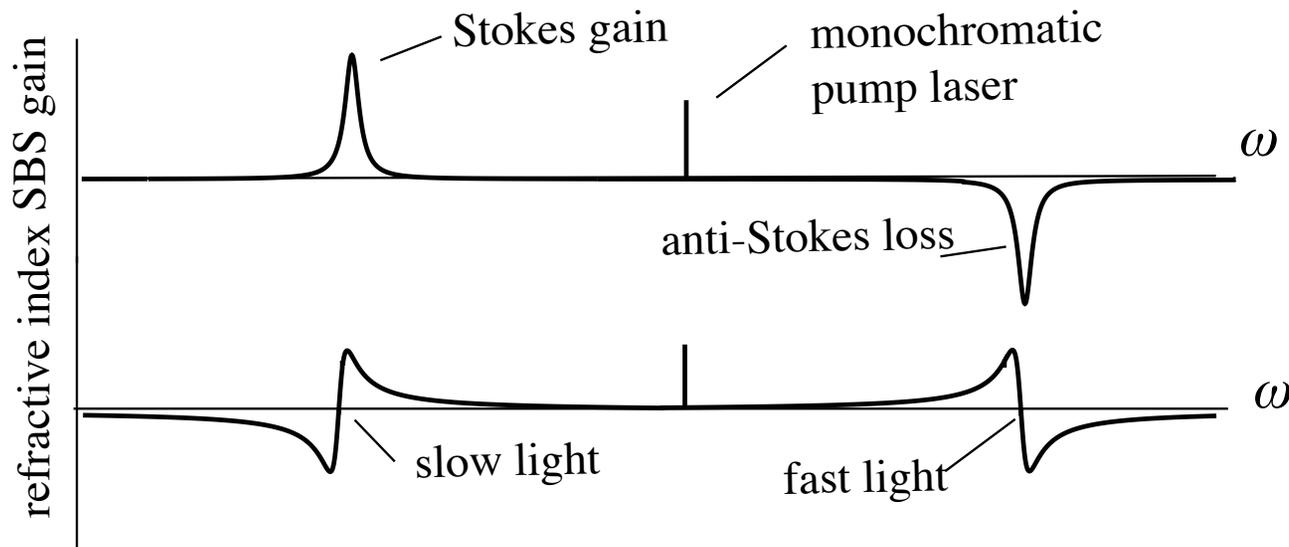
# Slow Light by Stimulated Brillouin Scattering (SBS)



$$\frac{dI_S}{dz} = -gI_L I_S$$

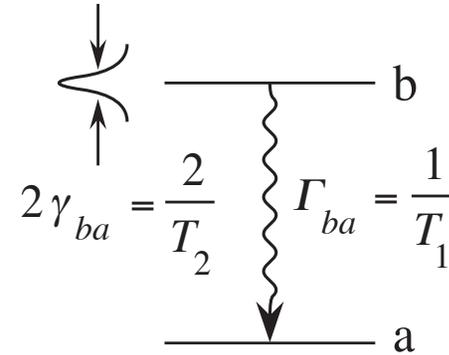
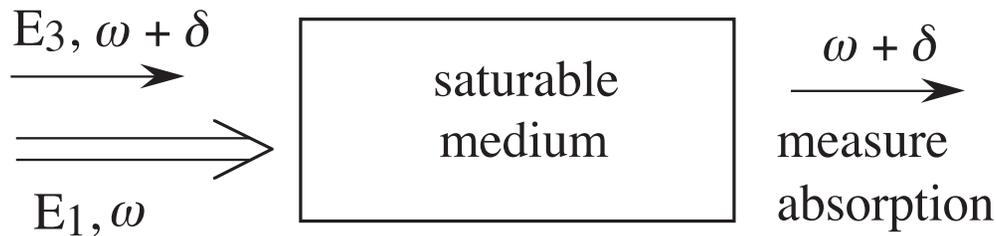
$$g = \frac{\gamma_e^2 \omega^2}{nv c^3 Q_0 \Gamma_B}$$

We often think of SBS as a pure gain process, but it also leads to a change in refractive index

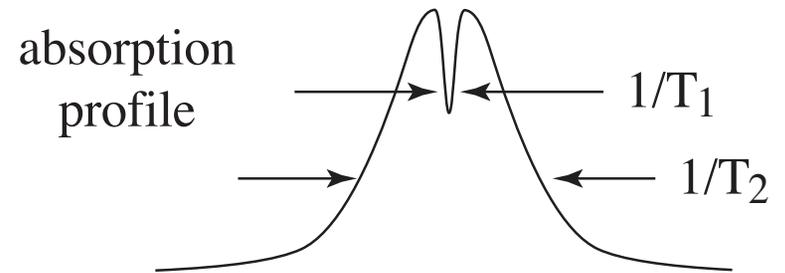


The induced time delay is  $\Delta T_d \approx \frac{G}{\Gamma_B}$  where  $G = g I_p L$  and  $\Gamma_B$  is the Brillouin linewidth

# Slow Light via Coherent Population Oscillations



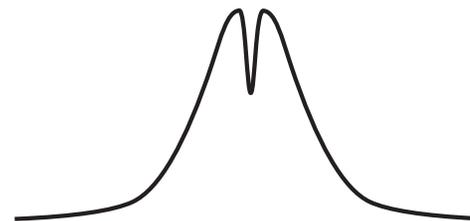
$$n_g = n + \omega(dn/d\omega) \quad T_2 \ll T_1$$



- Want a narrow feature in absorption profile to give a large  $dn/d\omega$
- Ground state population oscillates at beat frequency  $\delta$  (for  $\delta < 1/T_1$ ).
- Population oscillations lead to decreased probe absorption (by explicit calculation), even though broadening is homogeneous.
- Ultra-slow light ( $n_g > 10^6$ ) observed in ruby and ultra-fast light ( $n_g = -4 \times 10^5$ ) observed in alexandrite.
- Slow and fast light effects occur at room temperature!

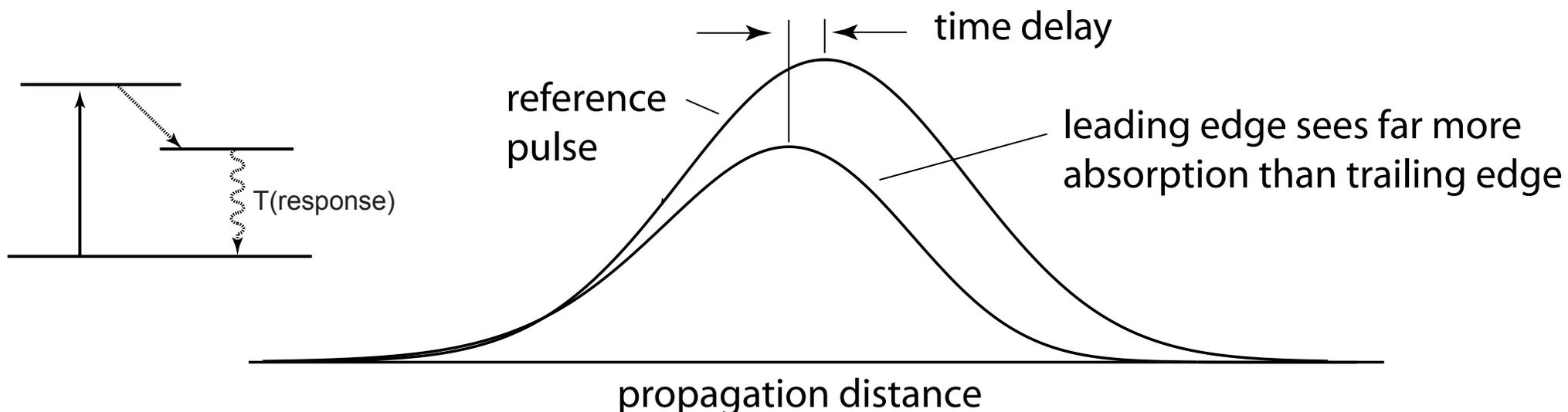
# Relation of CPO to the Basov Mechanism

- CPO slow light: a strong pump beam creates a narrow transparency window, and strong spectral variation of the refractive index leads to a large group index.



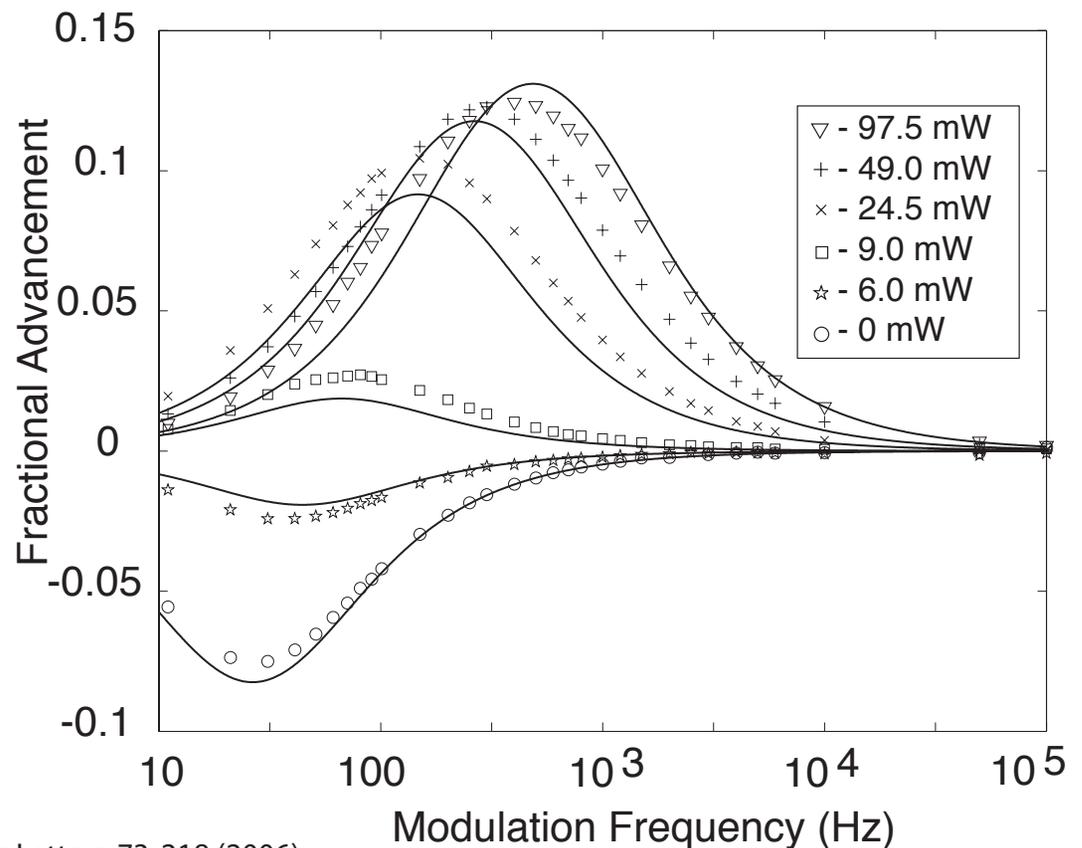
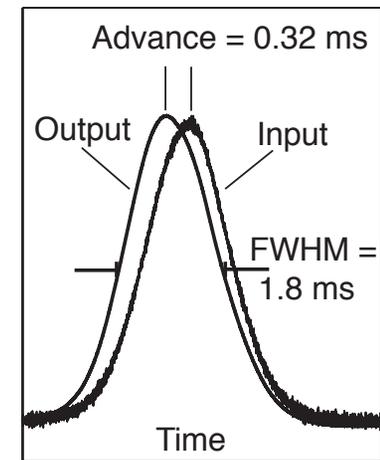
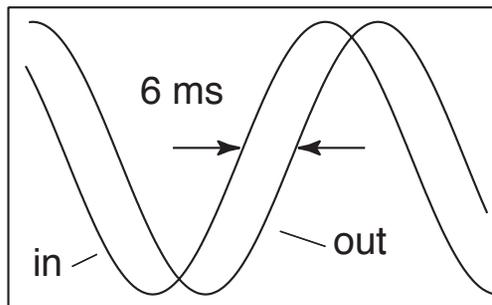
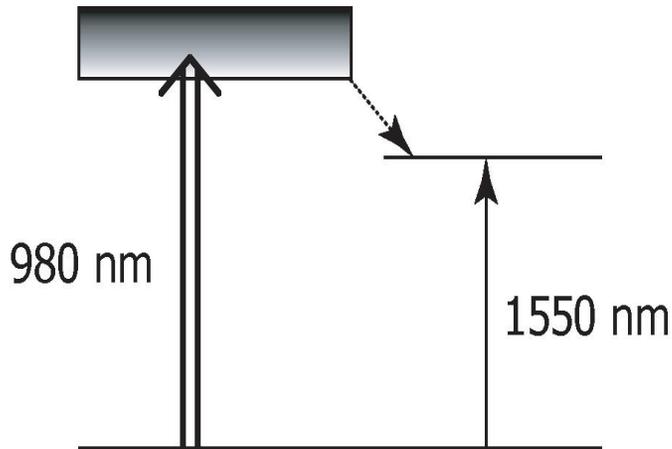
- Basov mechanism: an isolated intense pulse passing through a saturable material experiences a time delay.

- Assume that  $T_{\text{pulse}} \ll T_1$  = time scale for saturation changes
- Then absorption decreases with time during pulse due to saturation



# Slow and Fast Light in an Erbium Doped Fiber Amplifier

- Fiber geometry allows long propagation length
- Saturable gain or loss possible depending on pump intensity



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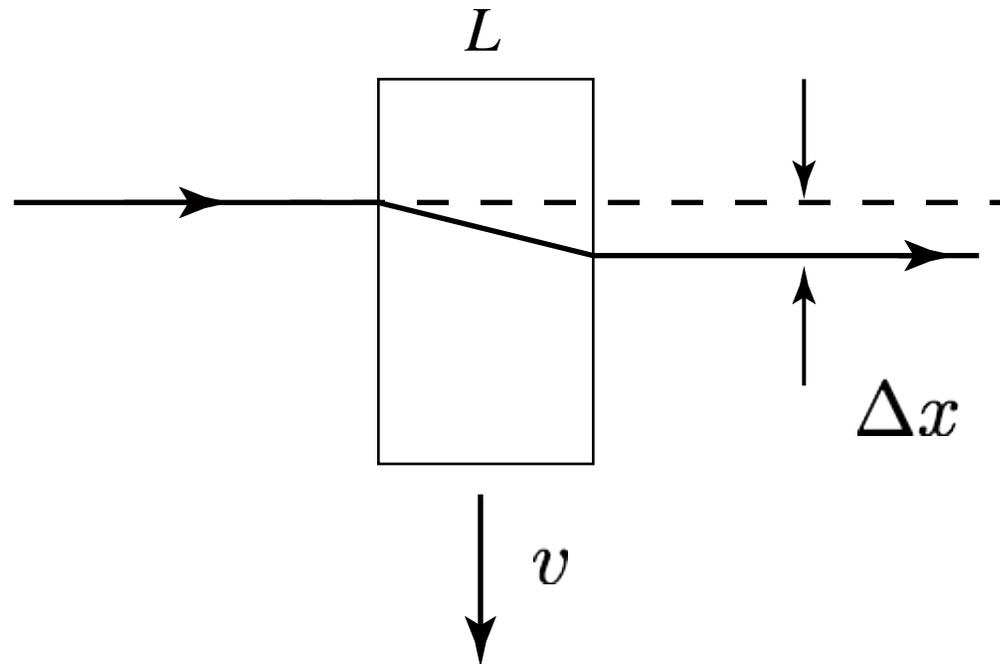
# Fun Physics with Slow and Fast Light

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Topic 1: Photon Drag Effects with Slow Light

# Transverse Photon Drag

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$$\Delta x = (vL/c)(n_g - 1/n_\phi)$$

For  $L = 25$  mm,  $v = 2000$  cm/s, displacement = 6 nm.

Measured by R.V. Jones, 1972.

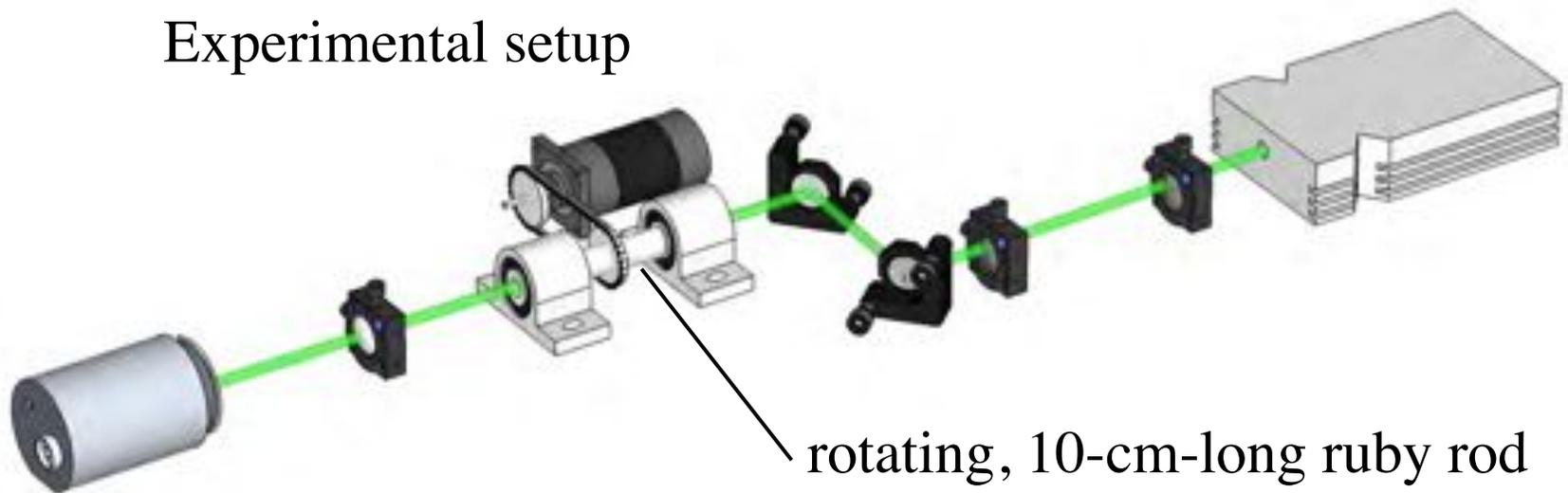
# Observation of Rotary Photon Drag

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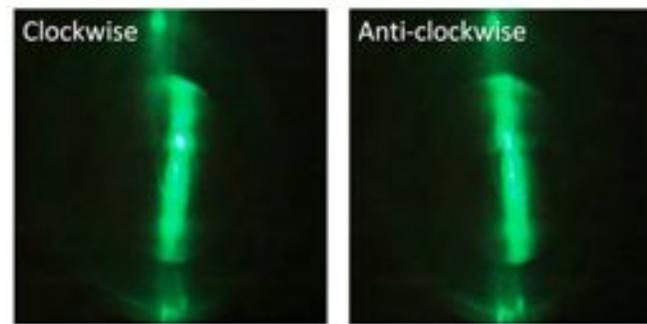
The world as seen through a spinning window.

(Laser-excited ruby has a group index of  $10^6$ .)

Experimental setup



Effect clearly visible by eye!



Franke-Arnold, Gibson, Boyd and Padget, Science, 2011

(See also the earlier work of Leach et al., 2008.)

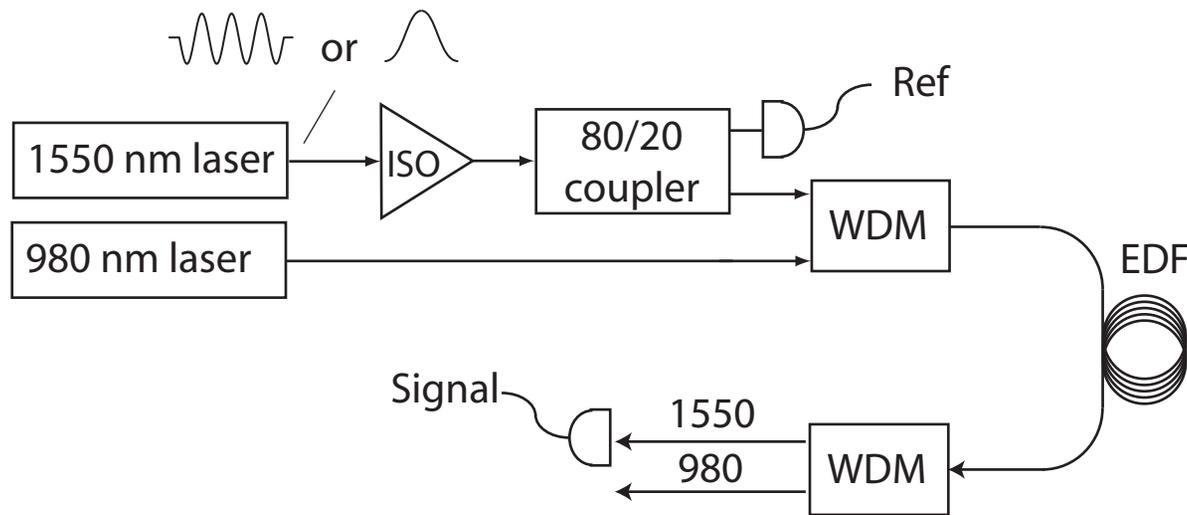
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# Fun Physics with Slow and Fast Light

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Topic 2: “Backwards” Propagation and  
Negative Group Velocities

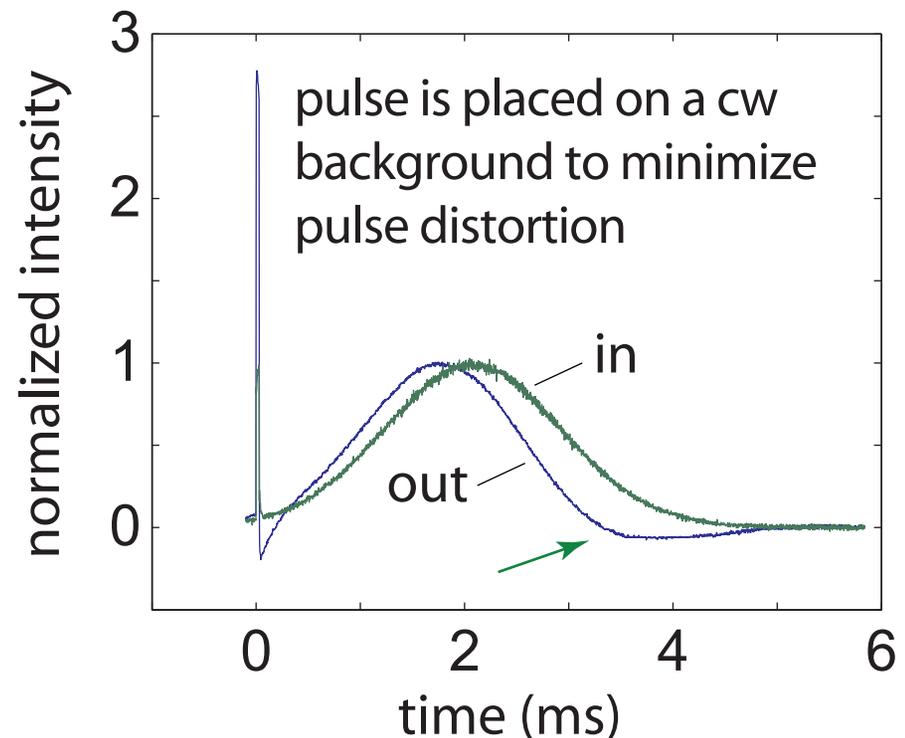
# Observation of Backward Pulse Propagation in an Erbium-Doped-Fiber Optical Amplifier



We time-resolve the propagation of the pulse as a function of position along the erbium-doped fiber.

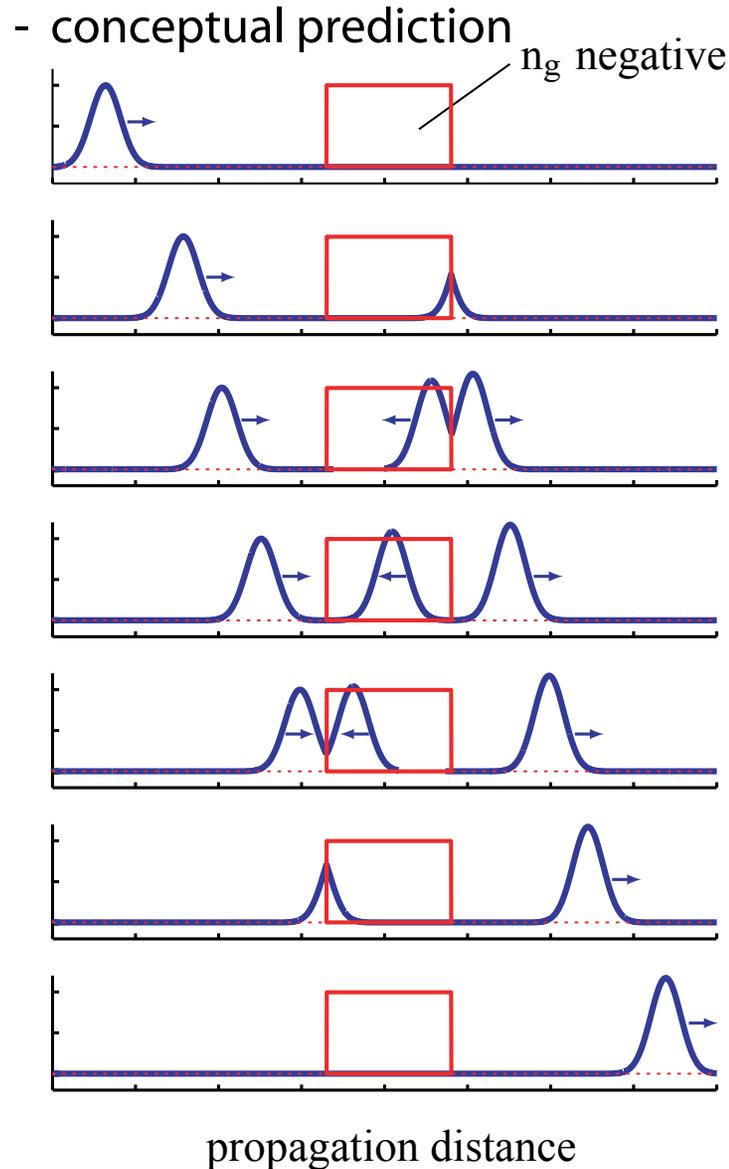
## Procedure

- cutback method
- couplers embedded in fiber

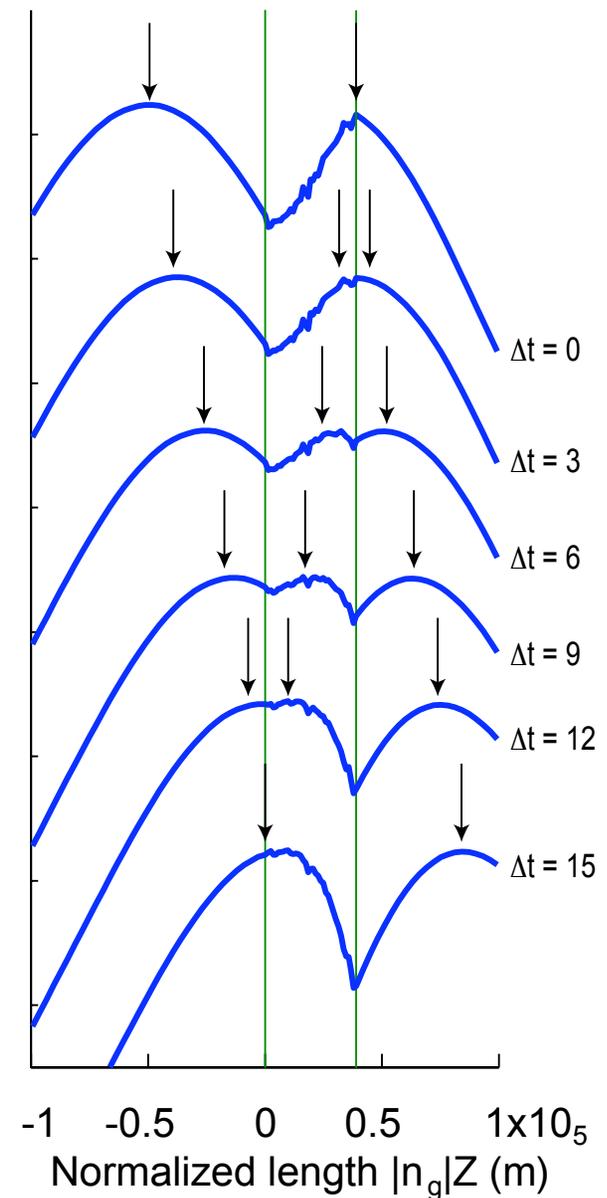


# Observation of Superluminal and "Backwards" Pulse Propagation

- A strongly counterintuitive phenomenon
- But entirely consistent with established physics
- Predicted by Garrett and McCumber (1970) and Chiao (1993).
- Observed by Gehring, Schweinsberg, Barsi, Kostinski, and Boyd Science 312, 985 2006.



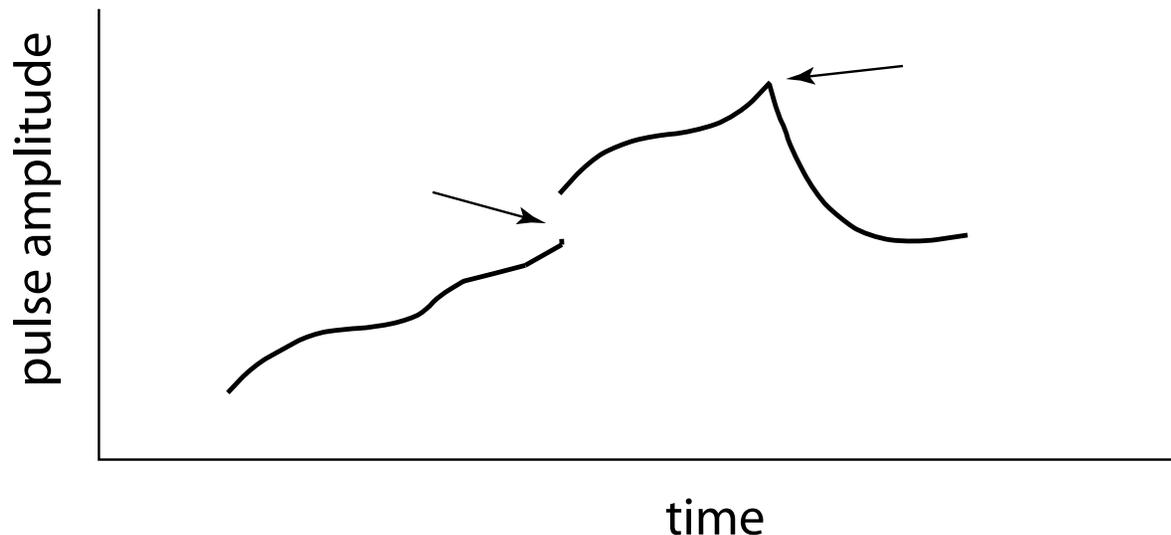
- laboratory results



# Causality?

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- Superluminal ( $v_g > c$ ) and backwards ( $v_g$  negative) propagation may seem counterintuitive but are fully compatible with causality.
- The group velocity is the velocity at which peak of pulse moves; it is not the “information velocity.”
- It is believed that information is carried by points of nonanalyticity of a waveform



- broad spectral content at points of discontinuity
- disturbance moves at vacuum speed of light

see, for instance, R. Y. Chiao

# Applications of Slow and Fast Light (where the action is now!)

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Buffers and regenerators for telecom

Slow/fast light for interferometry

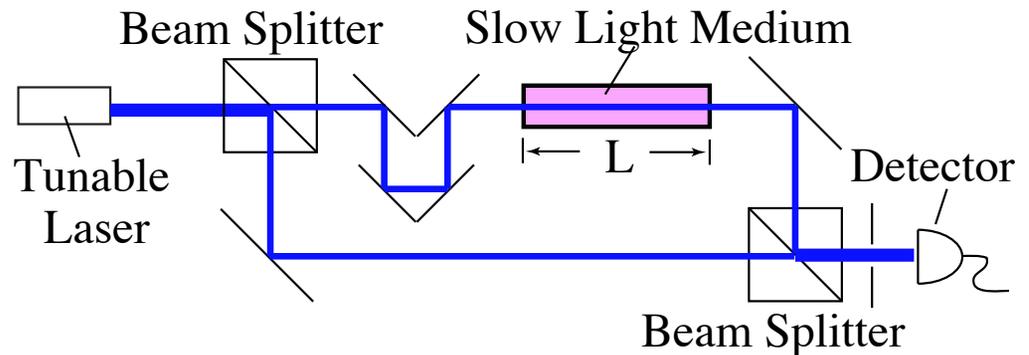
Phased- and synchronized-array laser radar

Construction of quantum memories

# Interferometry and Slow Light

- The spectral sensitivity of an interferometer is increased by a factor as large as the group index of a material placed within the interferometer.
- We want to exploit this effect to build chip-scale spectrometers with the same resolution as large laboratory spectrometers
- Here is why it works:

Slow-light interferometer:

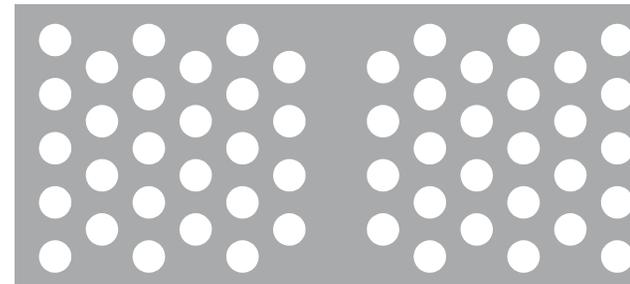


Simple analysis

$$\frac{d \Delta\phi}{d\omega} = \frac{d}{d\omega} \frac{\omega n L}{c} = \frac{L}{c} \left( n + \omega \frac{dn}{d\omega} \right) = \frac{L n_g}{c}$$

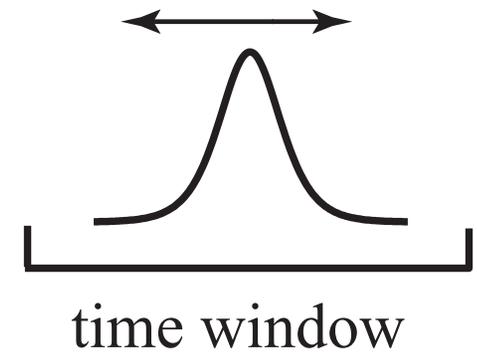
- We use line-defect waveguides in photonic crystals as our slow light mechanism

Slow-down factors of greater than 100 have been observed in such structures.

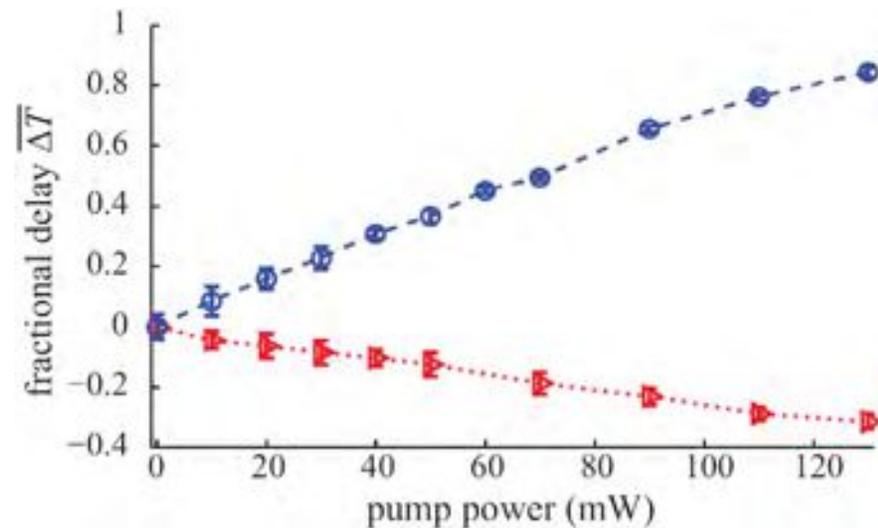
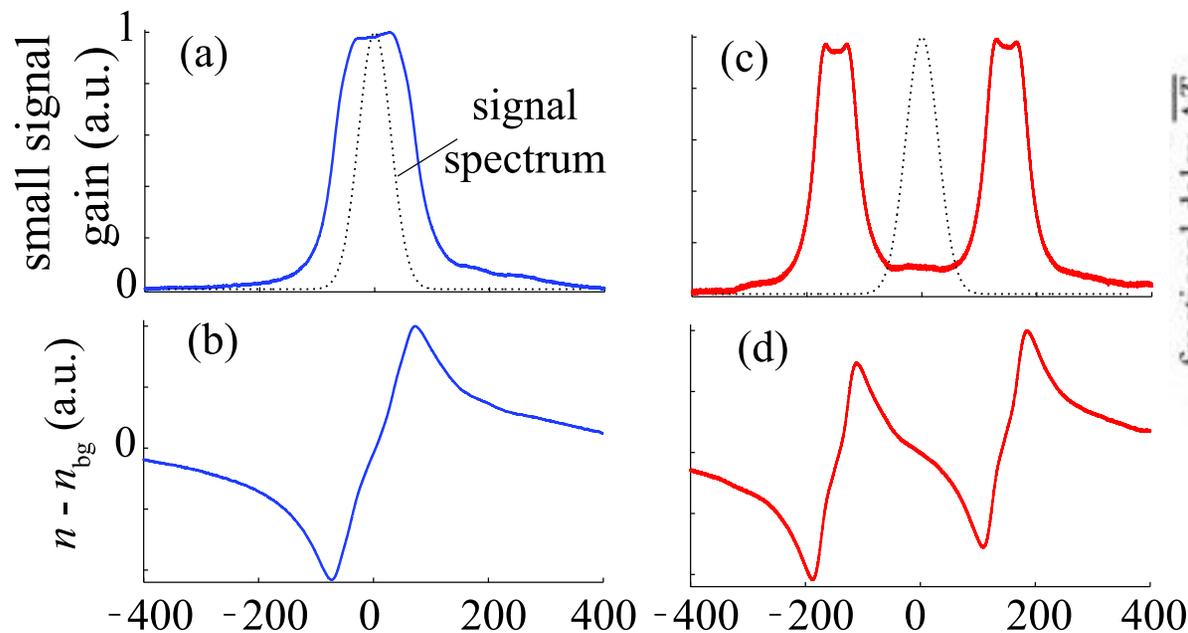


# Regeneration of Pulse Timing

Need to recenter each pulse in its time window  
Removes timing jitter caused by NL and environment effects in fiber  
Need only approximately  $\pm 1$  pulse width of delay!  
Most conveniently done by access to both slow and fast light



Recent implementation using SBS with EO-controlled gain spectrum





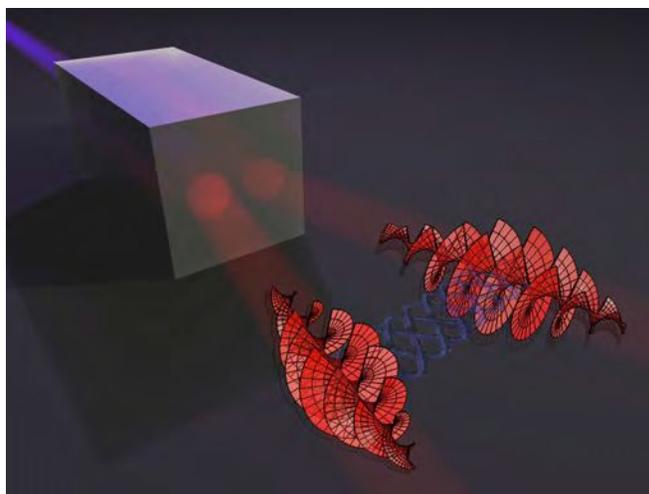
# Nonlinear Optics and Quantum Information Science

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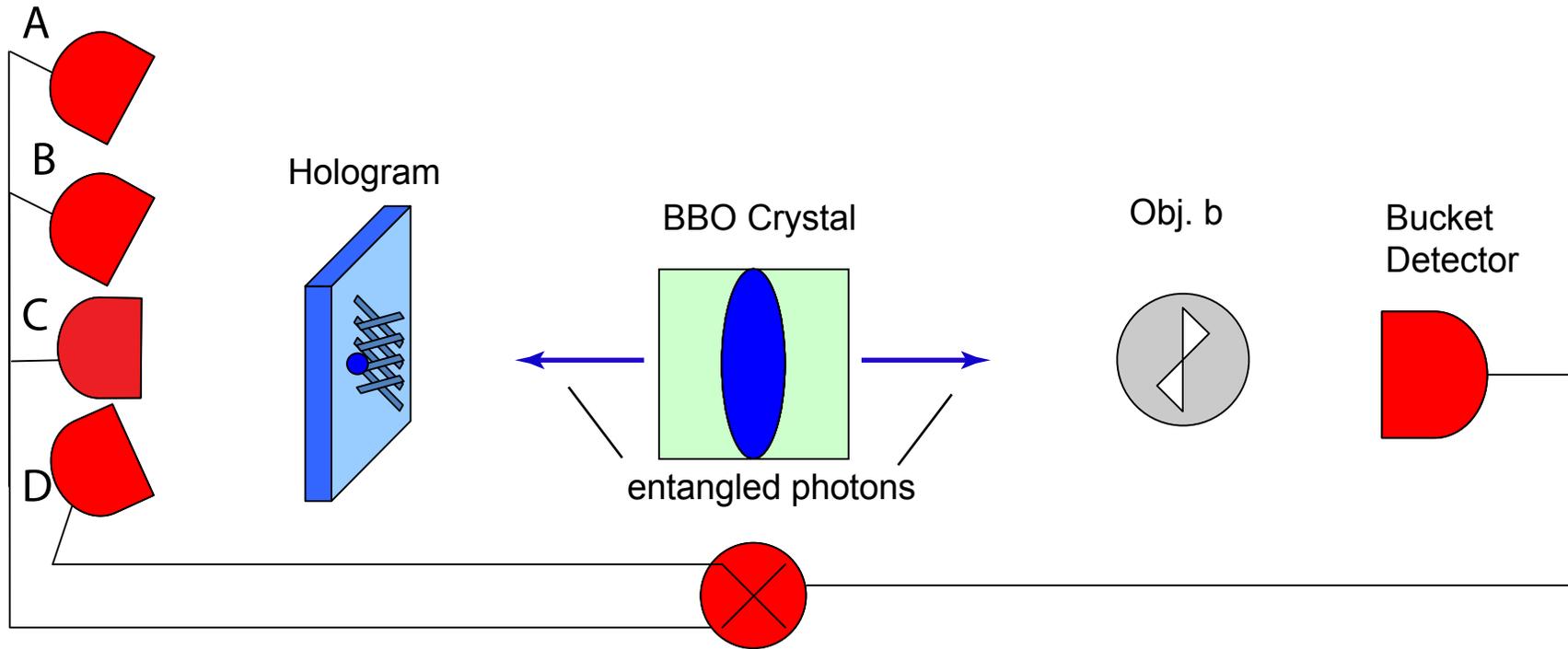
- We *use* nonlinear optics to create quantum states of light
- Some questions under investigation

How many bits of information can one photon carry?

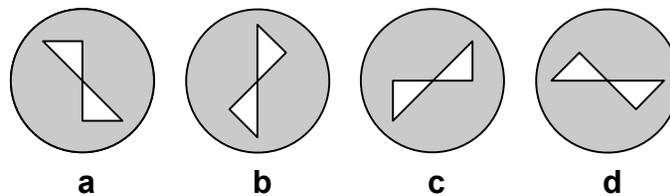
Can we perform quantum communications with more than one bit of classical information per photon?



# Single-Photon Coincidence Imaging



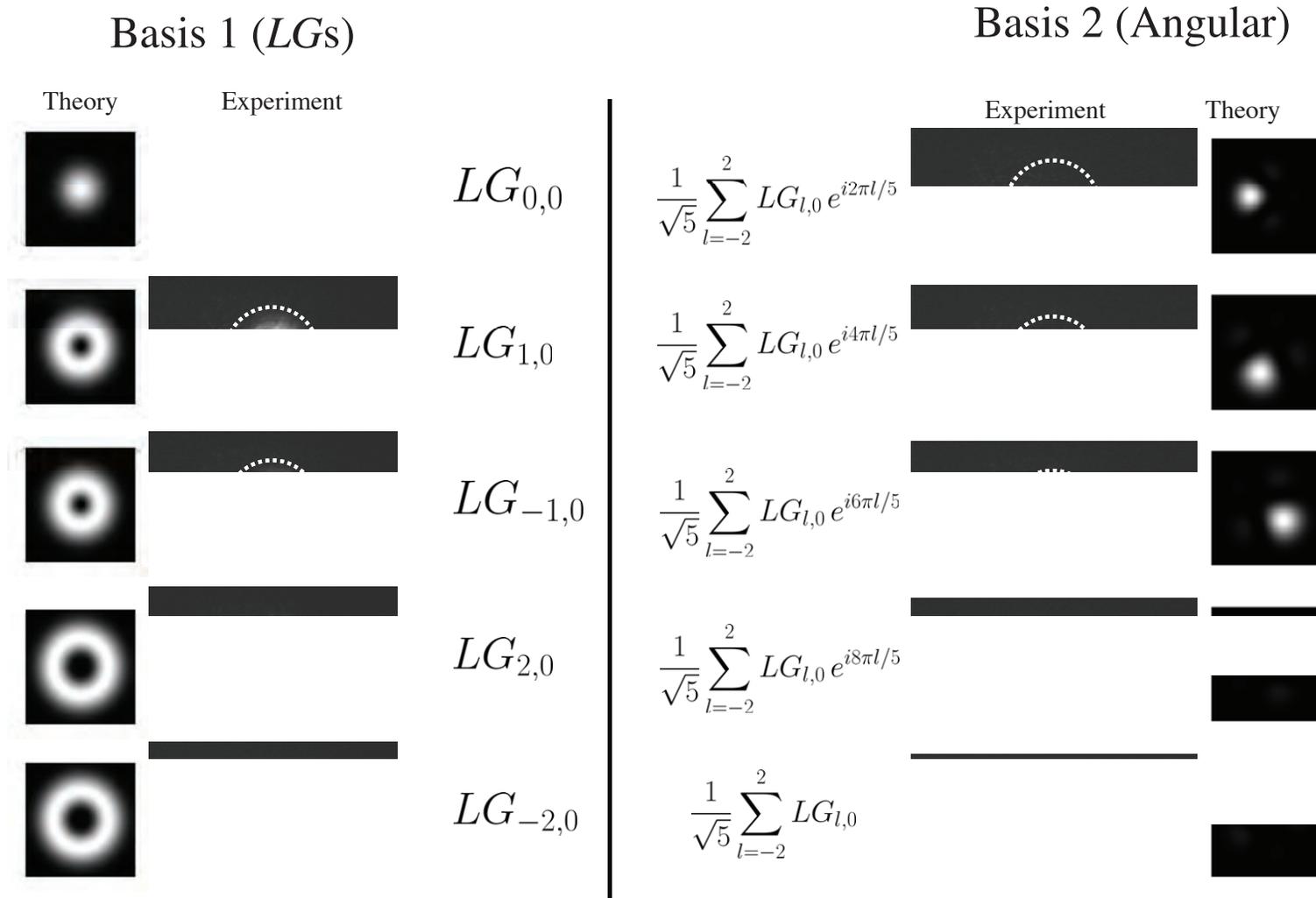
We discriminate between four orthogonal images at the single-photon level in a coincidence imaging configuration.



# Quantum Key Distribution with Many Bits Per Photon

Offers absolutely secure communications

Encode in a large alphabet (the Laguerre-Gauss modes)



$$\frac{1}{\sqrt{5}} \sum_{l=-2}^2 LG_{l,0} e^{i2\pi l/5}$$

$$\frac{1}{\sqrt{5}} \sum_{l=-2}^2 LG_{l,0} e^{i4\pi l/5}$$

$$\frac{1}{\sqrt{5}} \sum_{l=-2}^2 LG_{l,0} e^{i6\pi l/5}$$

$$\frac{1}{\sqrt{5}} \sum_{l=-2}^2 LG_{l,0} e^{i8\pi l/5}$$

$$\frac{1}{\sqrt{5}} \sum_{l=-2}^2 LG_{l,0}$$

# Closing Remarks

NLO is as exciting today as it was 51 years ago

NLO has spun off many new research fields, such as  
ultrafast phenomena, photonics, optical solitons, . . .

One exciting future direction is that of quantum nonlinear optics

Thank you for your attention!

