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# Quantum Nonlinear Optics: Nonlinear Optics Meets the Quantum World

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The visuals of this talk will be posted at [boydnlo.ca/presentations](http://boydnlo.ca/presentations)

Presented at Hunan University, Changsha, P.R. China, May 28-29, 2018.



## Canada Excellence Research Chair (CERC) in Nonlinear Quantum Optics

Research interest:

Nonlinear optics, quantum optics,  
integrated photonics, meta-materials, etc.

# Quantum Nonlinear Optics:

## Nonlinear Optics Meets the Quantum World

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Outlook: NLO is a superb platform from which to explore new physical processes and to develop photonics applications.

### Prospectus

1. Introduction to Nonlinear Optics and Quantum NLO
2. New Applications of “Slow Light”
3. Möbius Strips of Polarization
4. Huge Optical Nonlinearity in Epsilon-Near-Zero Materials
5. Quantum Communication with Multiple Bits per Photon



# Why Study Nonlinear Optics?

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It is good fundamental physics.

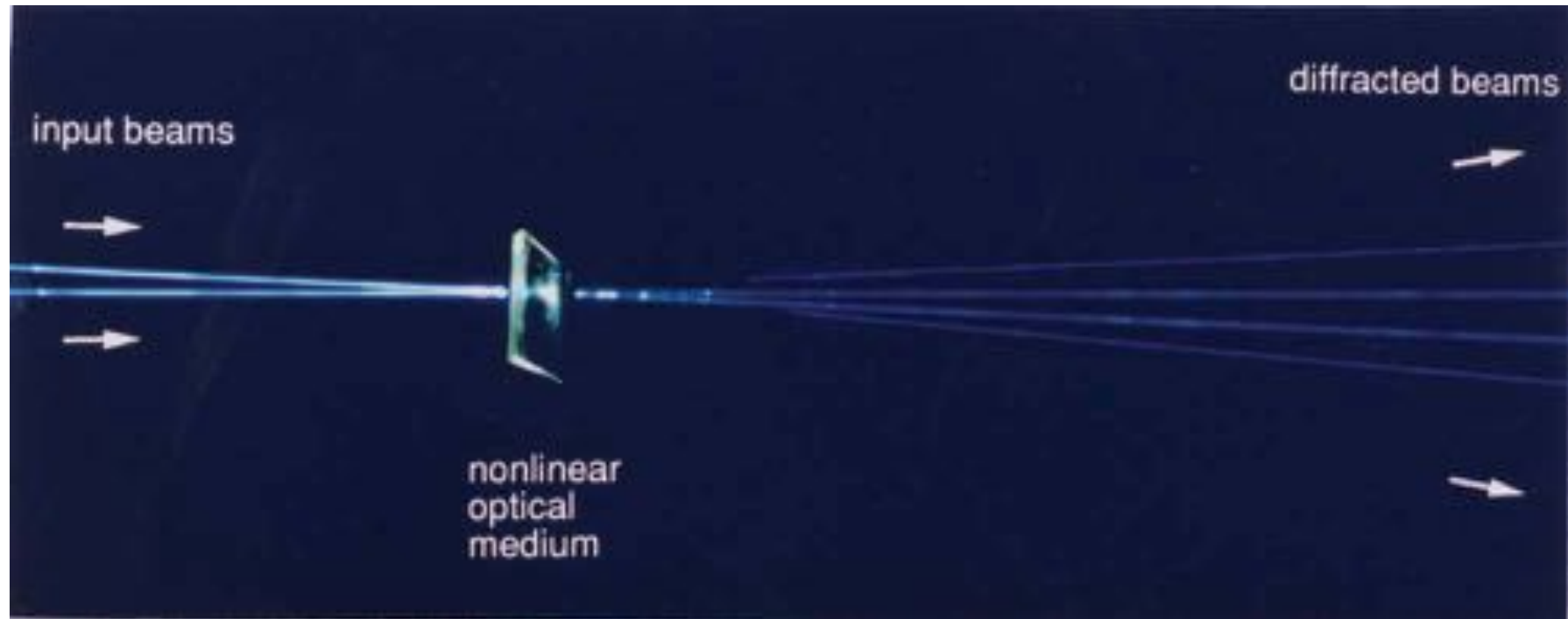
It leads to important applications.

It is a lot of fun.

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Demonstrate these features with examples in remainder of talk.

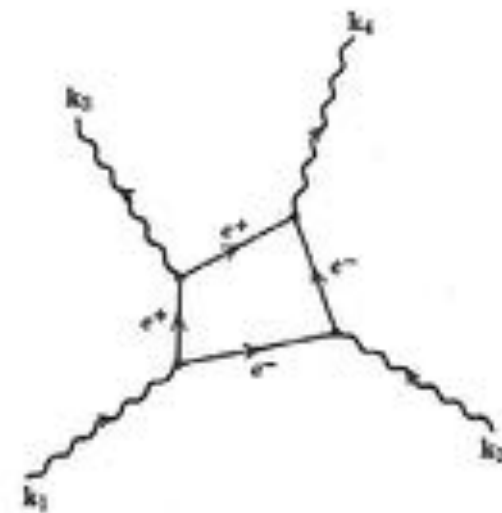
# Nonlinear Optics and Light-by-Light Scattering



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.

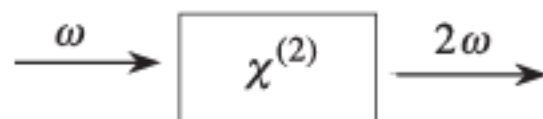
# Simple Formulation of the Theory of Nonlinear Optics

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

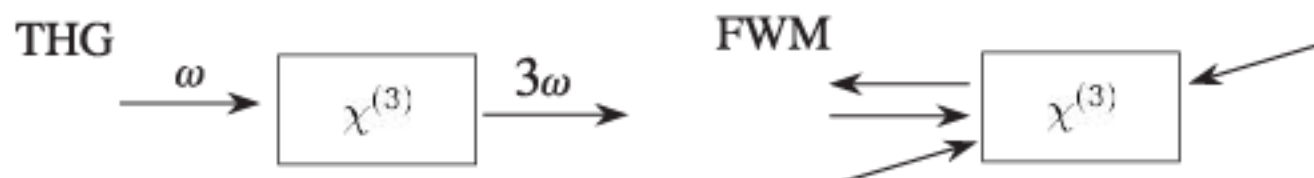
Here  $P$  is the induced dipole moment per unit volume and  $E$  is the field amplitude

$\chi^{(1)}$  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.



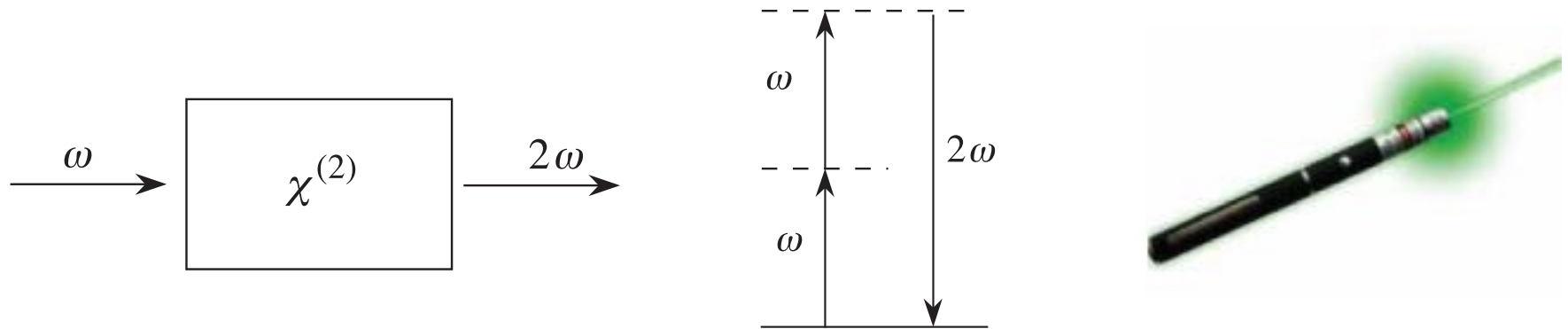
NL index

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





# Second-Harmonic Generation: The Prototypical Nonlinear Optical Process



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

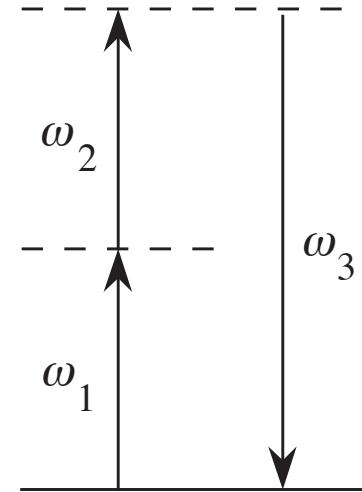
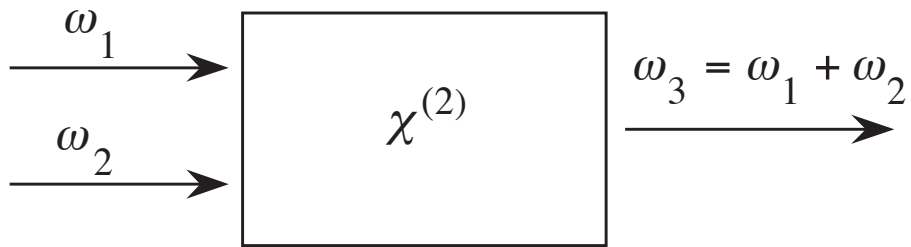
(Received July 21, 1961)



# Some Fundamental Nonlinear Optical Processes: II

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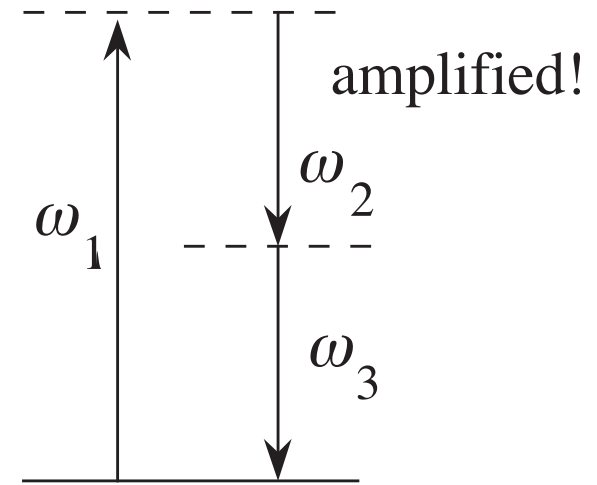
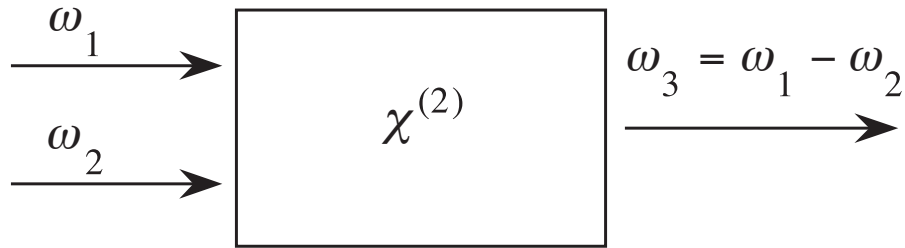
## Sum-Frequency Generation





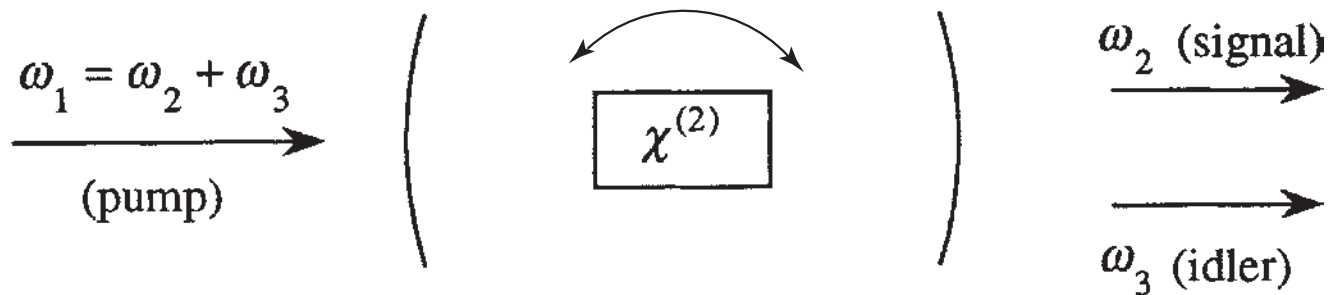
# Difference-Frequency Generation and Optical Parametric Amplification

## Difference-Frequency Generation



## Optical Parametric Amplification

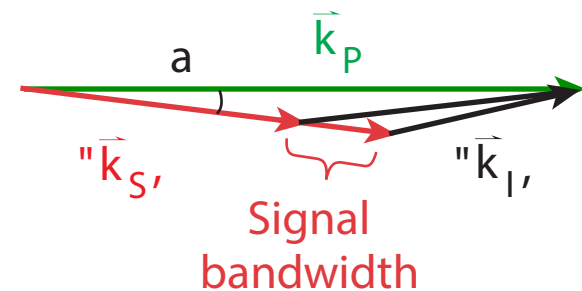
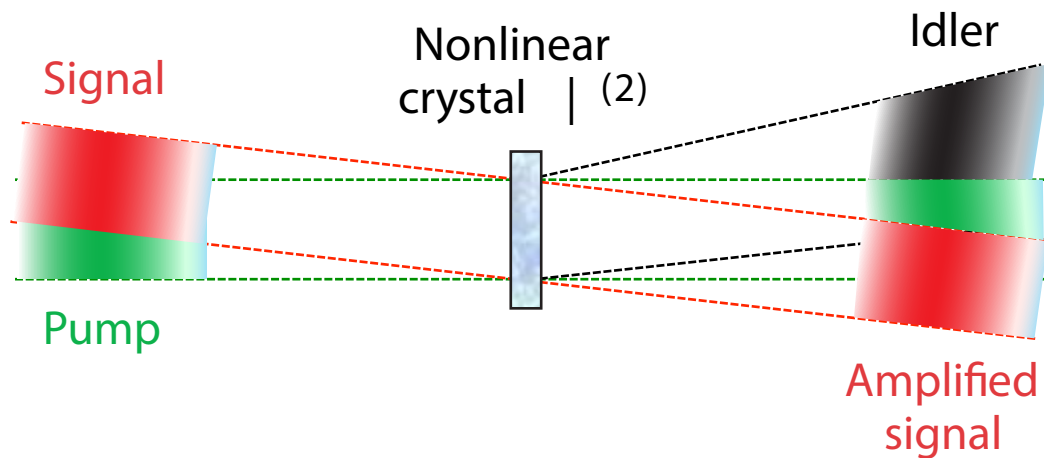
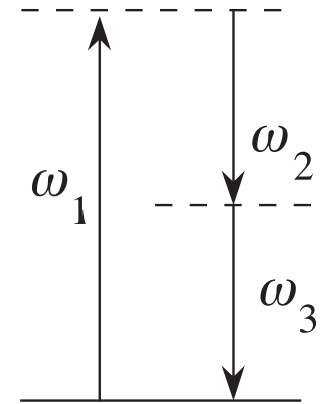
## Optical Parametric Oscillator (very broadly tunable)



# 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).



# Why Interest in Quantum Nonlinear Optics?

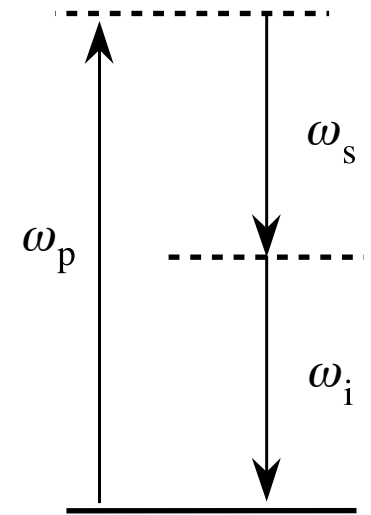
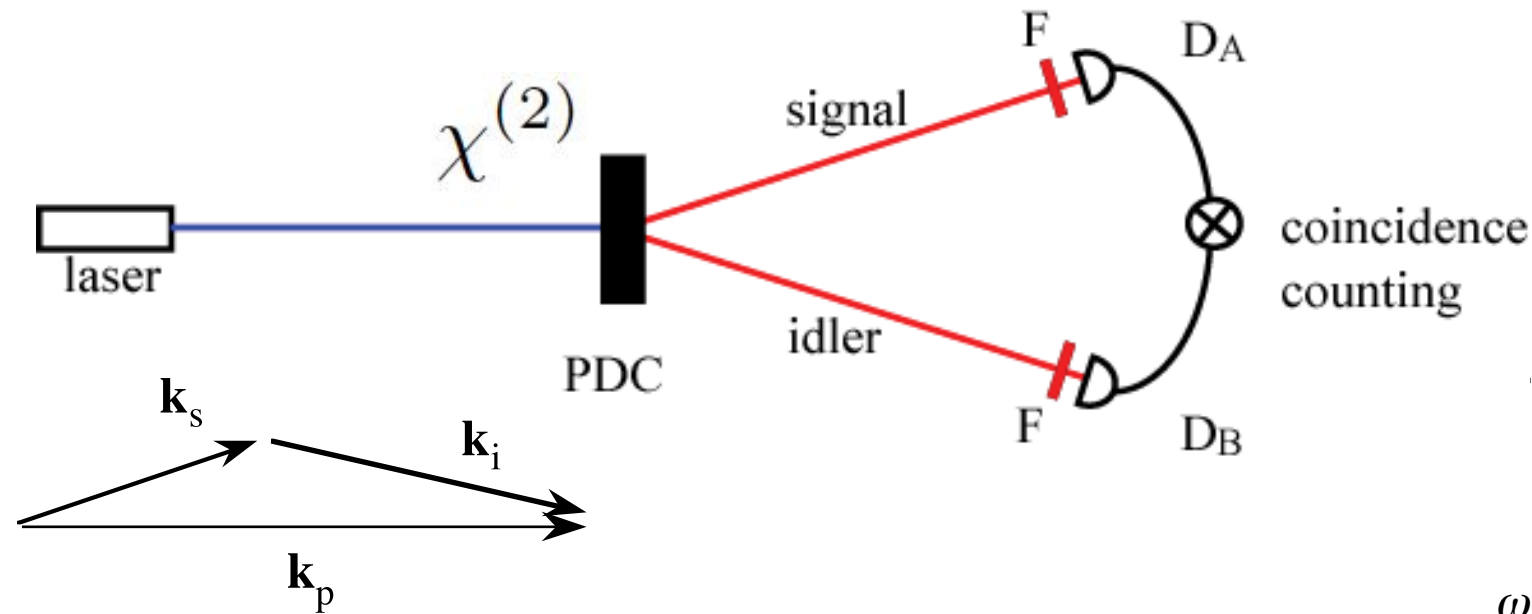
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Explore the relation between traditional nonlinear optics (NLO) and phenomena in quantum information science (QIS).

QIS holds great promise for secure communication, quantum logic, quantum computing, etc.

Many processes in QIS rely on nonlinear optical interactions.

# Parametric Downconversion: A Source of Entangled Photons



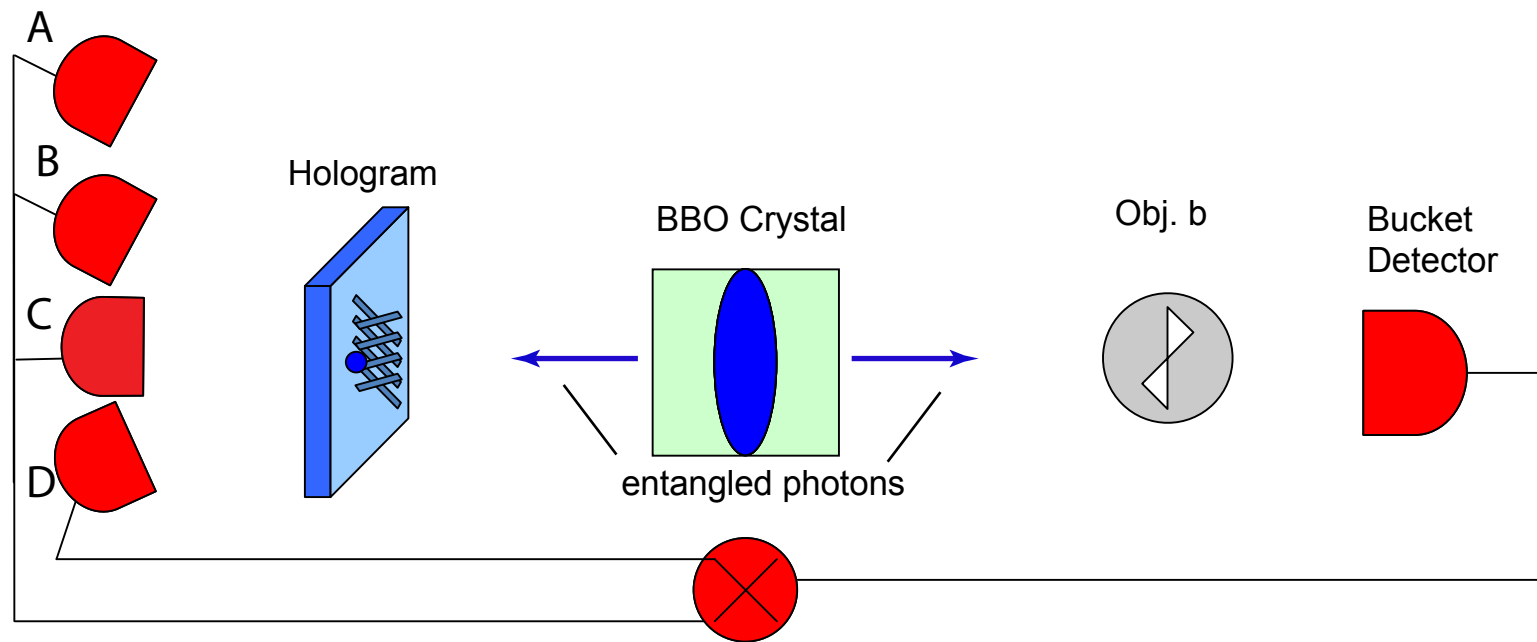
The signal and idler photons are entangled in:

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

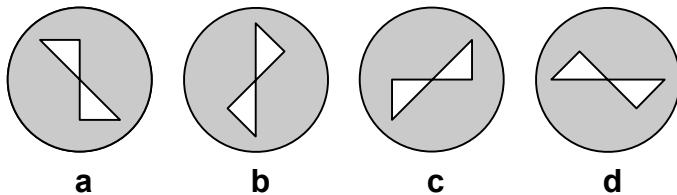
Entanglement is important for:

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

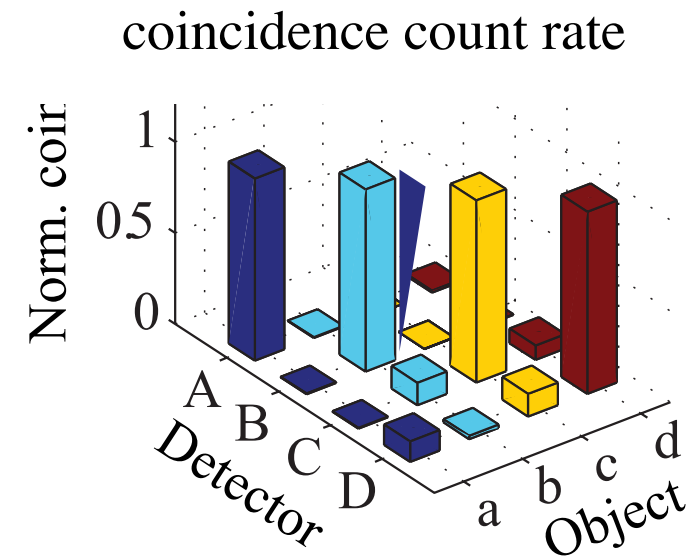
# Single-Photon Coincidence Imaging



- We discriminate among four orthogonal images using single-photon interrogation in a coincidence imaging configuration.

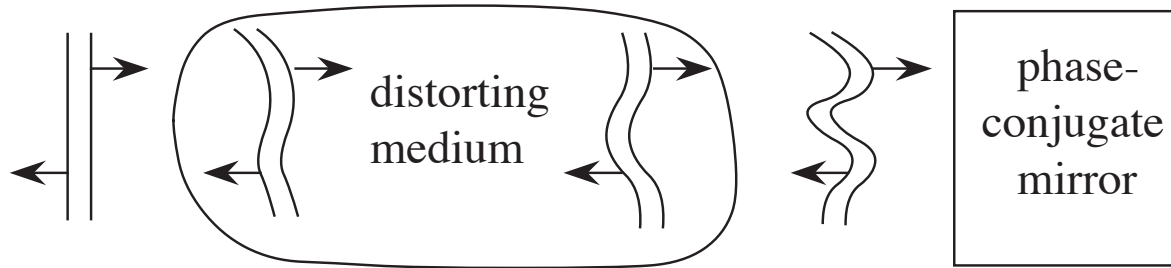


- Note that a single photon can carry more than one bit of information.



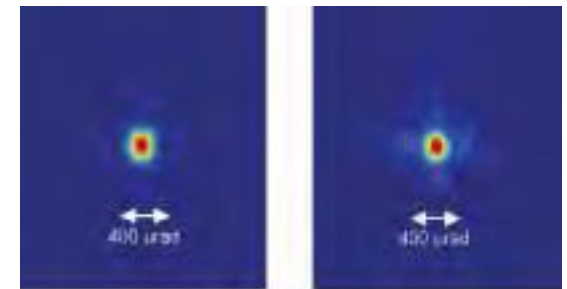
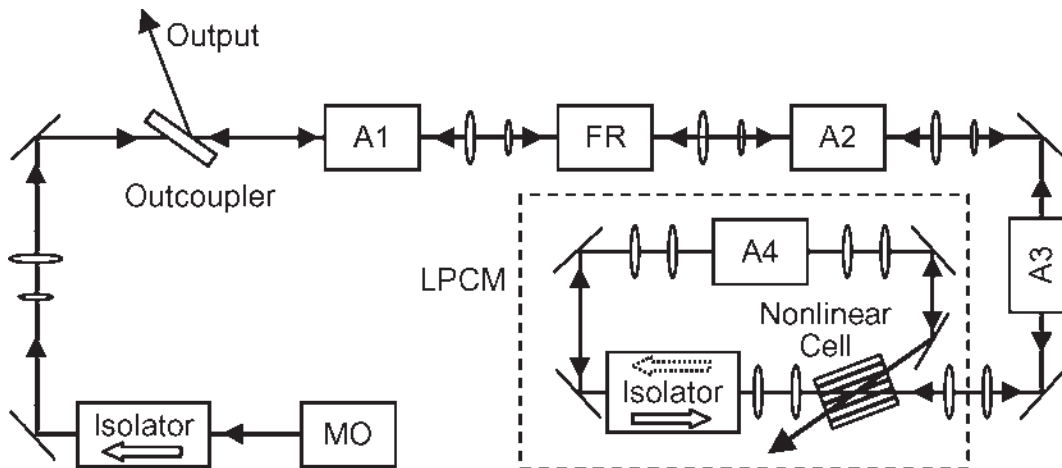
# Optical Phase Conjugation: A Nonlinear Optics Success Story

- 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

# Theory of nonlinear optics

PHYSICAL REVIEW

VOLUME 127, NUMBER 6

SEPTEMBER 15, 1962

## Interactions between Light Waves in a Nonlinear Dielectric\*

J. A. ARMSTRONG, N. BLOEMBERGEN, J. DUCUING,† AND P. S. PERSHAN

*Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts*

(Received April 16, 1962)

THE interaction between electromagnetic waves and atomic matter was carried out to higher orders of perturbation theory in the early years of modern quantum mechanics.<sup>1-3</sup> The interest in the absorption of two or more light quanta and scattering processes, in which three or more light quanta are involved, has recently been revived,<sup>4-7</sup> because intense light fluxes available from laser sources have made possible the experimental observation of such higher order processes in the laboratory.



The Nobel Prize in Physics 1981

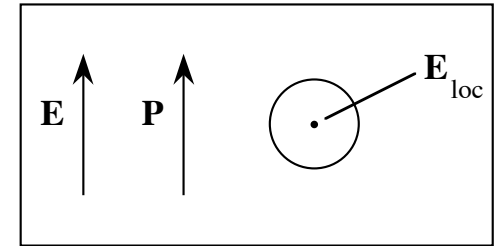
Nicolaas Bloembergen, Arthur L. Schawlow, Kai M. Siegbahn



# 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

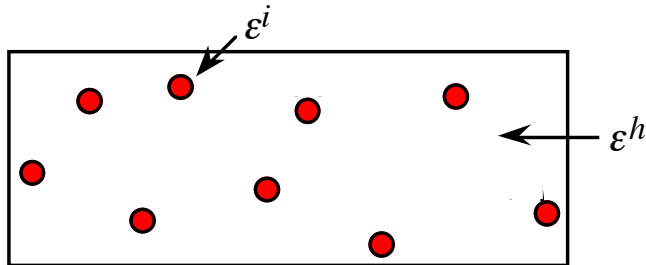
$$L(\omega) = \frac{\epsilon(\omega) + 2}{3}$$

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?

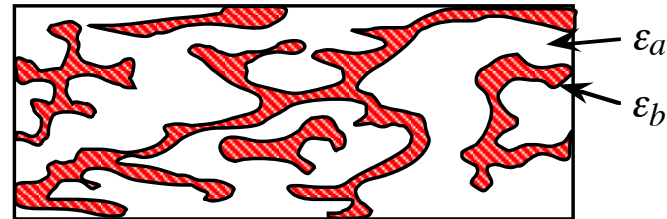
We have been developing new photonic materials with enhanced NLO response by using composite structures that exploit local field effects.

# Metamaterials and Nanocomposite Materials for Nonlinear Optics

- 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

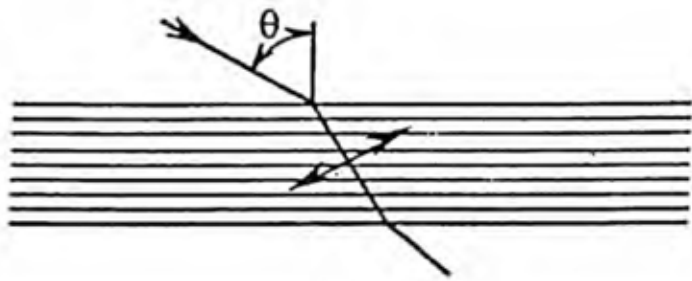
V. M. Shalaev and M. I. Stockman, Z. Phys. D 10, 71 (1988);

J. E. Sipe and R. W. Boyd, Phys. Rev. A 46, 1614 (1992).

# 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.

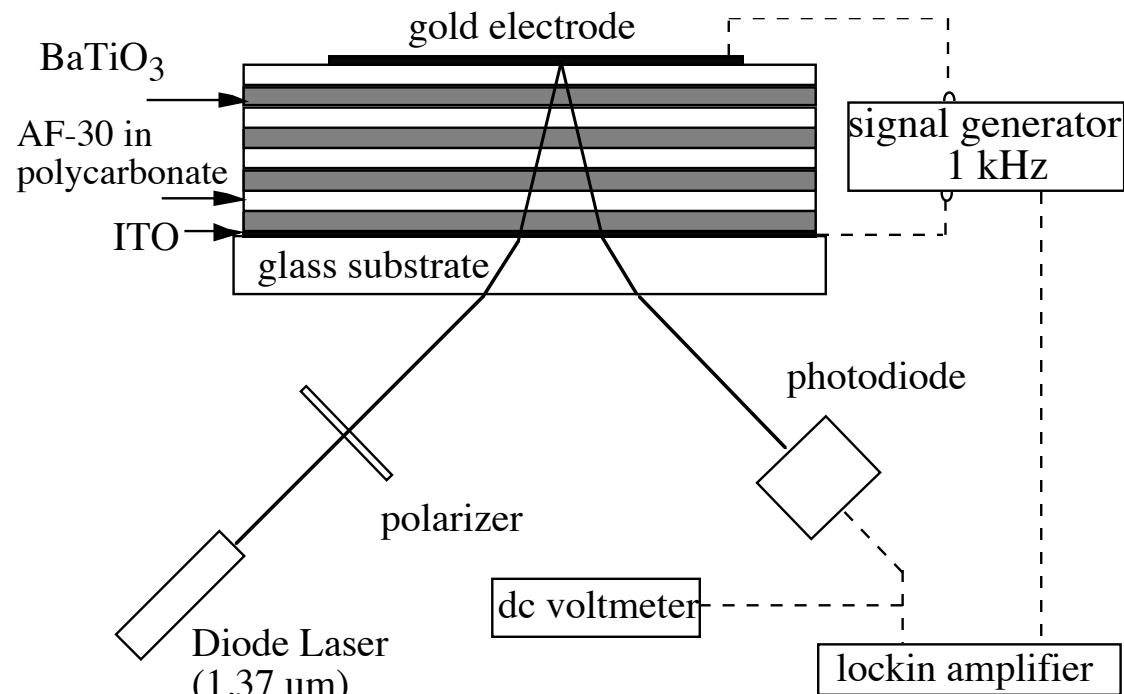


$\nabla \cdot \mathbf{D} = 0$  implies that  $(\epsilon \mathbf{E})_{\perp}$  is continuous.

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

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

## Quadratic EO effect



3.2 times enhancement!

# 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

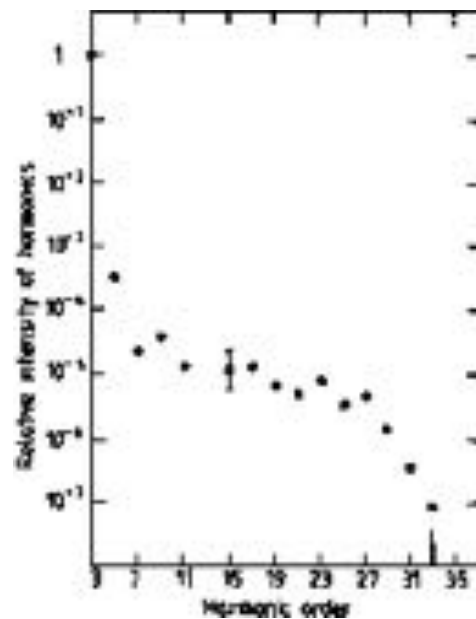
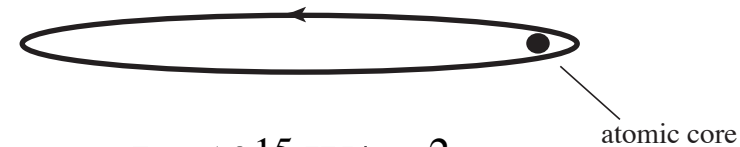
27 SEPTEMBER 1993

## Plasma Perspective on Strong-Field Multiphoton Ionization

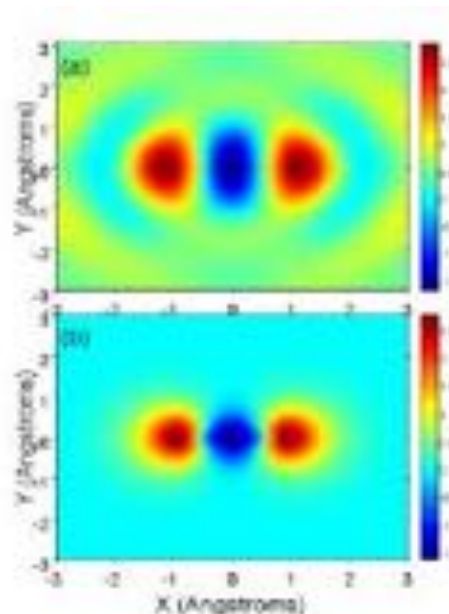
P. B. Corkum

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

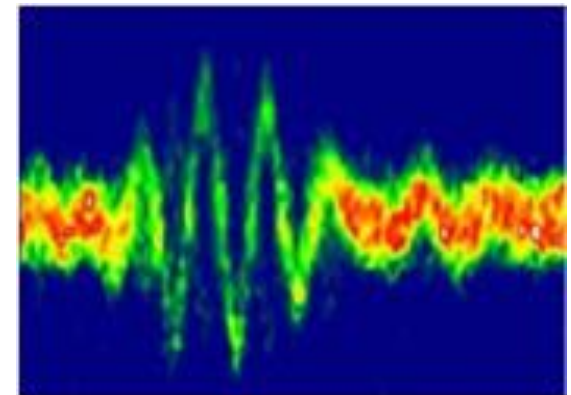
(Received 9 February 1993)



High-harmonic generation



Measuring the molecular nitrogen wavefunction



Attosecond pulses to sample a visible E-field; F. Krausz

# Quantum Nonlinear Optics:

## Nonlinear Optics Meets the Quantum World

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Outlook: NLO is a superb platform from which to explore new physical processes and to develop photonics applications.

### Prospectus

1. Introduction to Nonlinear Optics and Quantum NLO
2. New Applications of “Slow Light”
3. Möbius Strips of Polarization
4. Huge Optical Nonlinearity in Epsilon-Near-Zero Materials
5. Quantum Communication with Multiple Bits per Photon

# Controlling the Velocity of Light

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

– Light can be made to go:

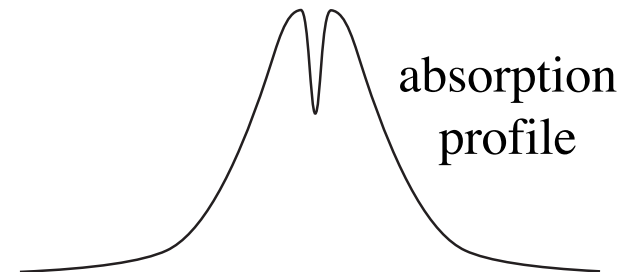
slow:  $v_g \ll c$  (as much as  $10^6$  times slower!)

fast:  $v_g > c$

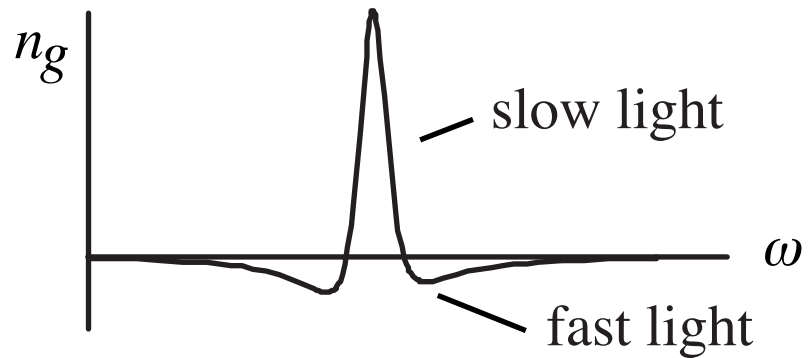
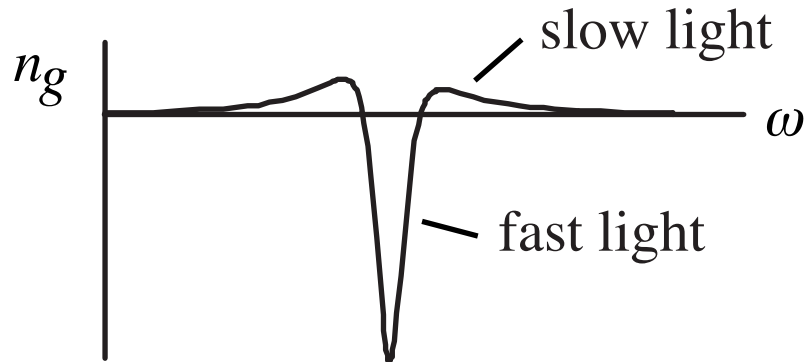
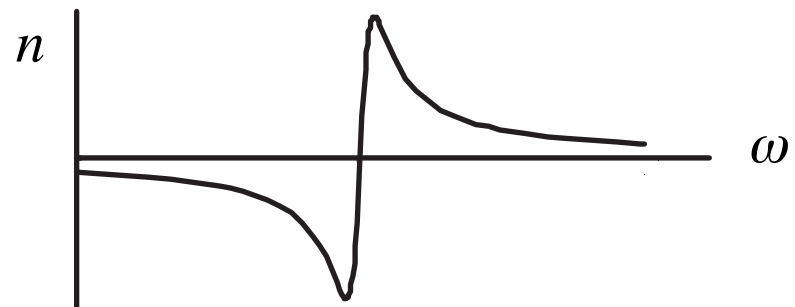
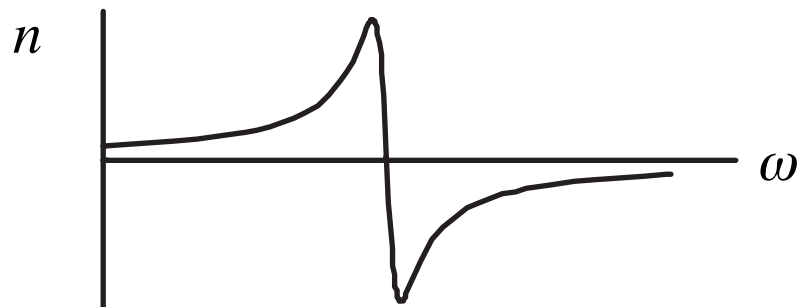
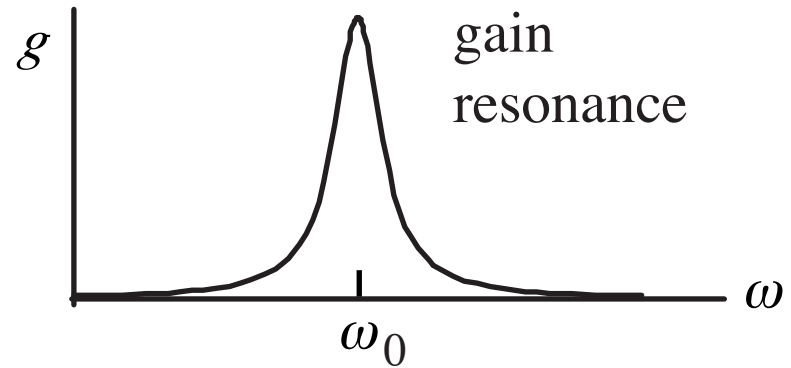
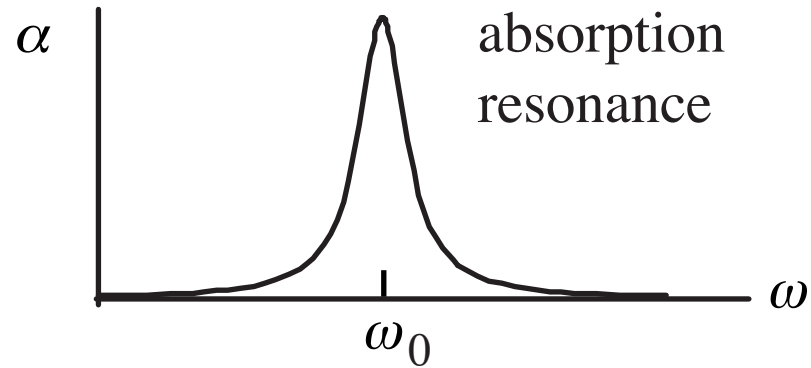
backwards:  $v_g$  negative

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



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

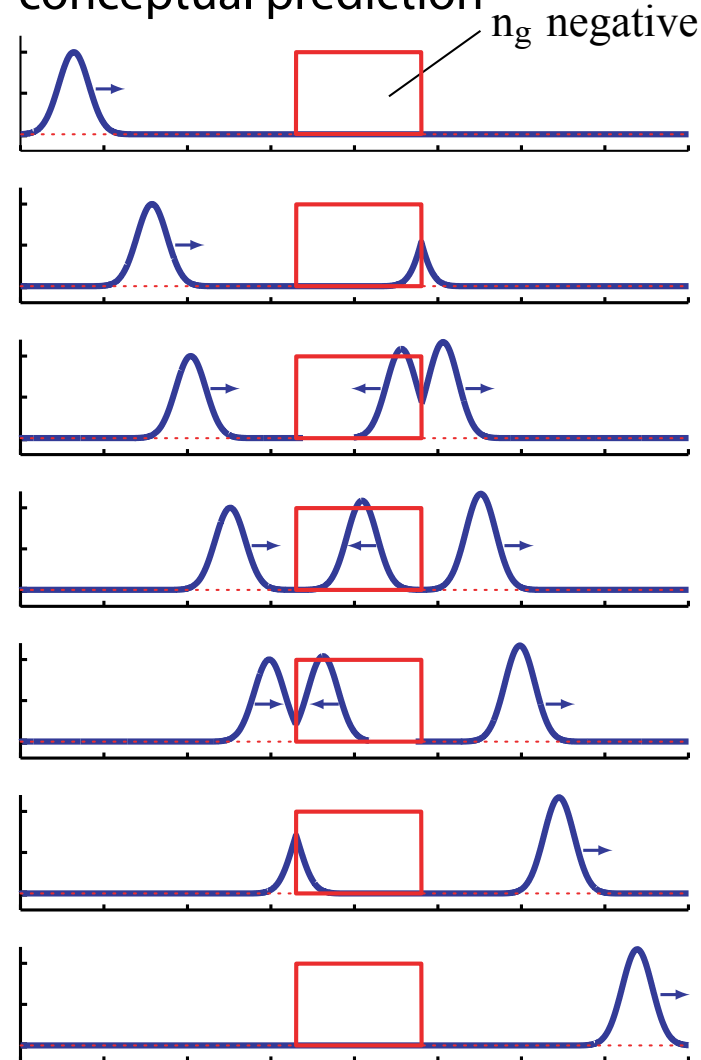


# Observation of Superluminal and “Backwards” Pulse Propagation

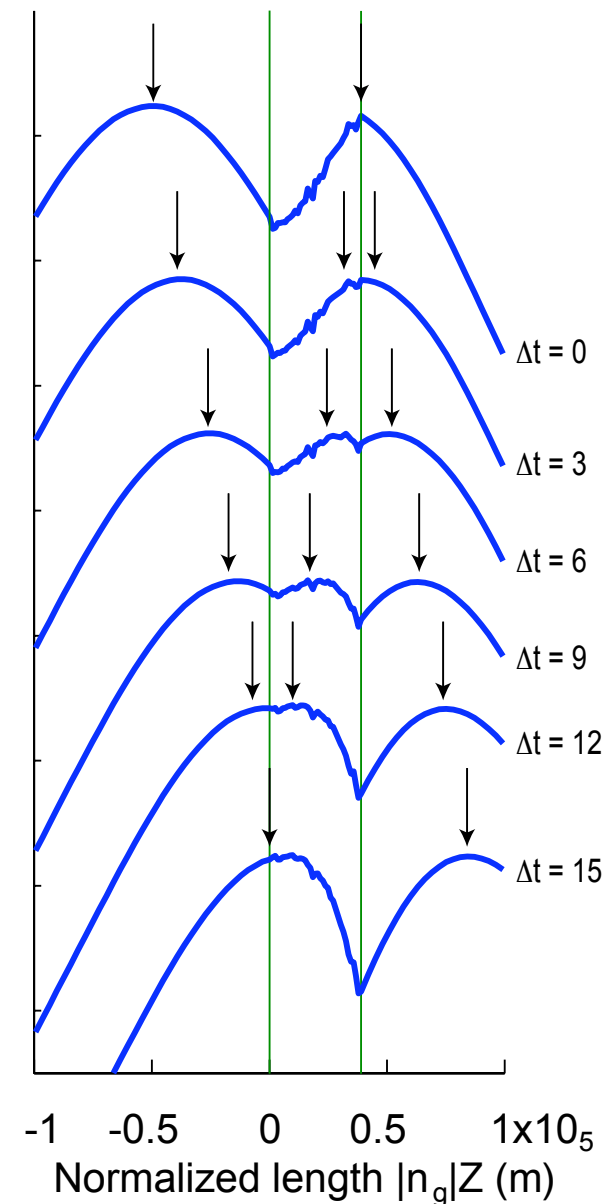


- A strongly counterintuitive phenomenon
- But entirely consistent with established physics
- Predicted by Garrett and McCumber (1970) and Chiao (1993).
- Observed by Gehring, Schweinsberg, Barsi, Kostinski, and Boyd Science 312, 985 2006.

- conceptual prediction



- laboratory results

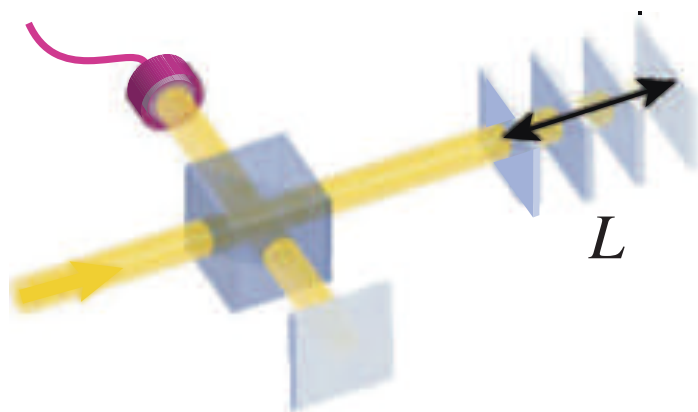


# Development of Miniaturized, Chip-Scale Spectrometers

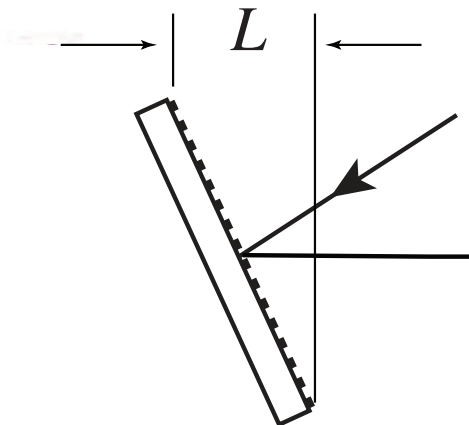
## Can We Beat the $1/L$ Resolution Limit of Standard Spectrometers?

- The limiting resolution of a broad class of spectrometers is given (in wave-numbers) by the inverse of a characteristic dimension  $L$  of the spectrometer

*Fourier-transform spectrometer*



*Grating spectrometer*



$$\Delta\nu(\text{res}) \approx 1/L$$

- We use slow-light methods to design spectrometers with resolution that exceeds this conventional limit by a factor as large as the group index.
- This ability allows us to miniaturize spectrometers with no loss of resolution, for “lab-on-a-chip” applications.

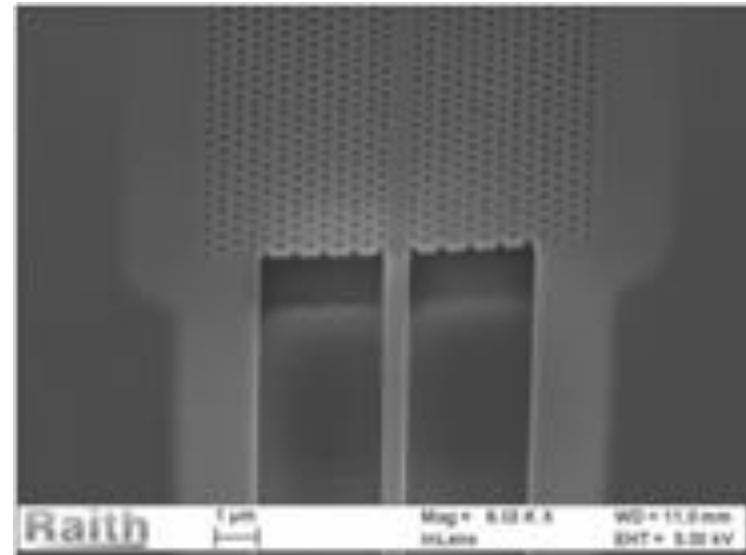
# Our Goal

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Replace this:



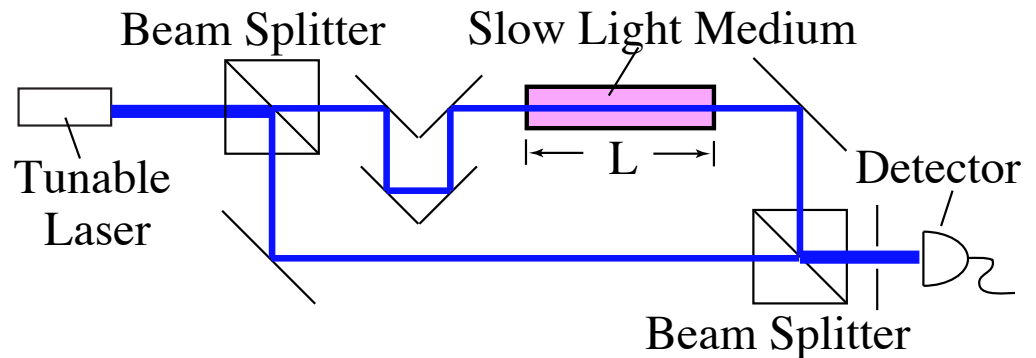
with this:



# Our Approach: Chip-Scale Slow-Light Spectrometer

- 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:

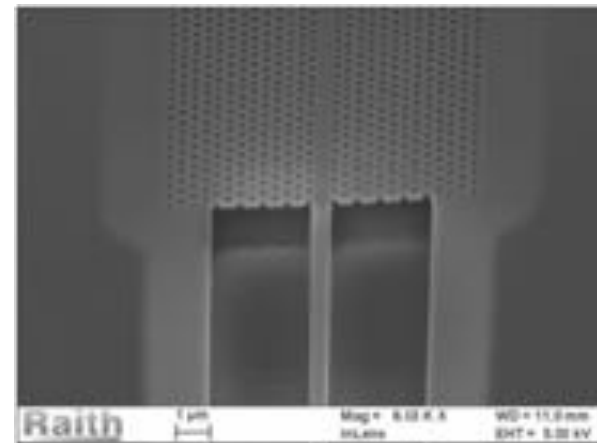


Simple analysis

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

- 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.

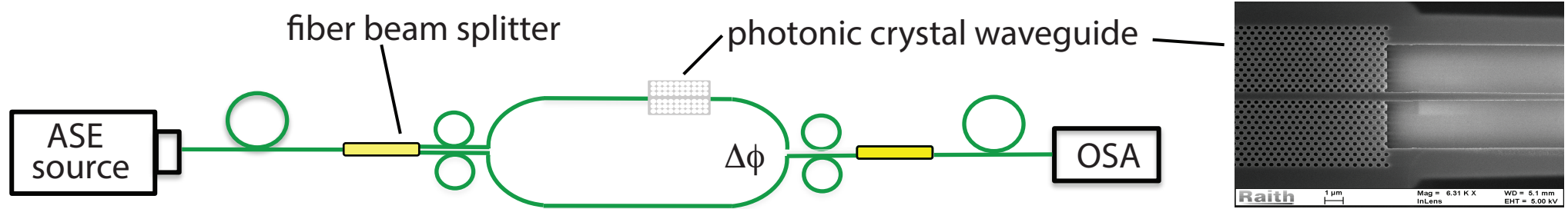


Shi, Boyd, Gauthier, and Dudley, Opt. Lett. 32, 915 (2007)

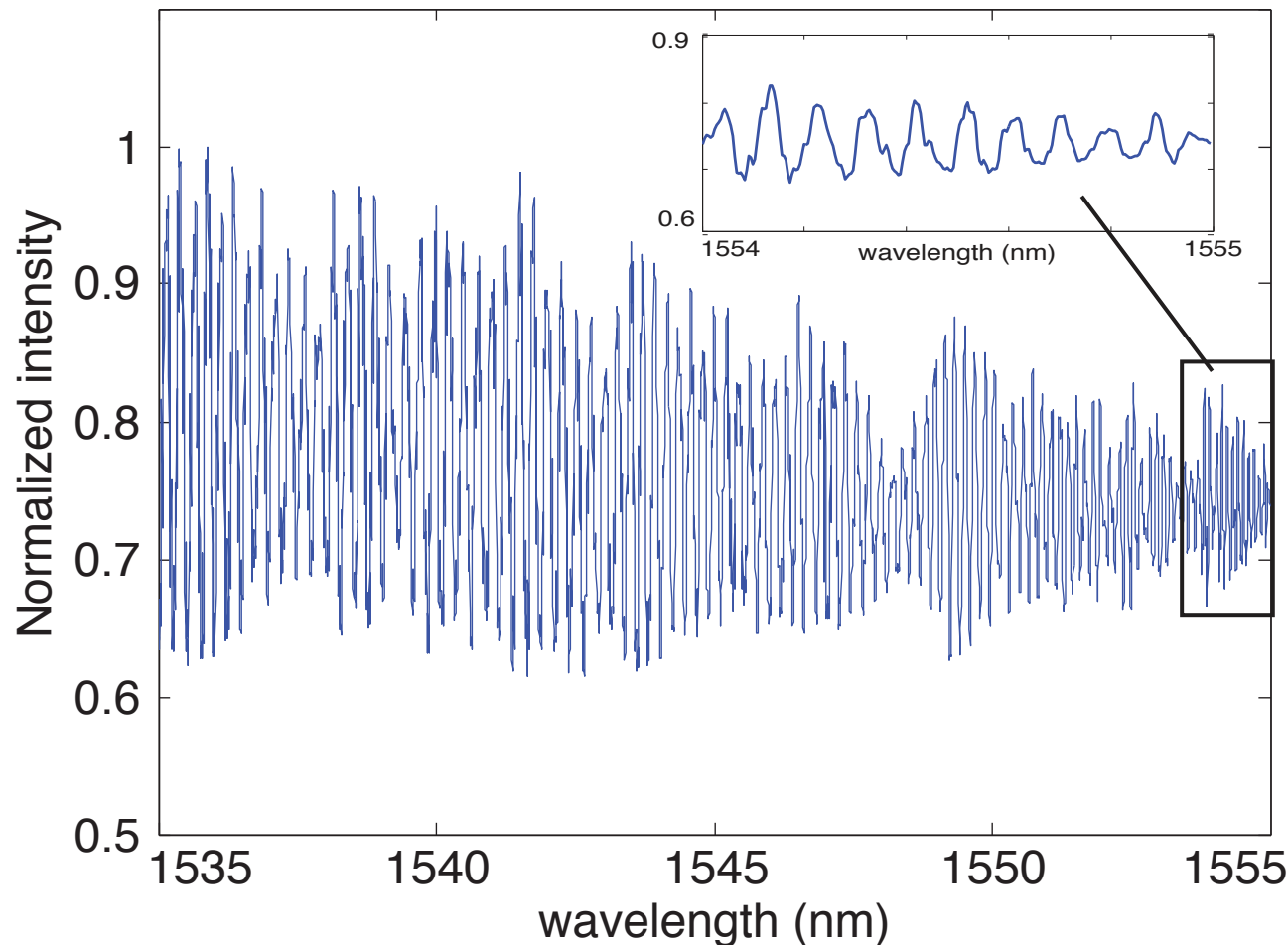
Shi, Boyd, Camacho, Vudiyasetu, and Howell, PRL. 99, 240801 (2007)

Shi and Boyd, J. Opt. Soc. Am. B 25, C136 (2008).

# Laboratory Characterization of the Slow-Light Mach-Zehnder Interferometer



- Interference fringes

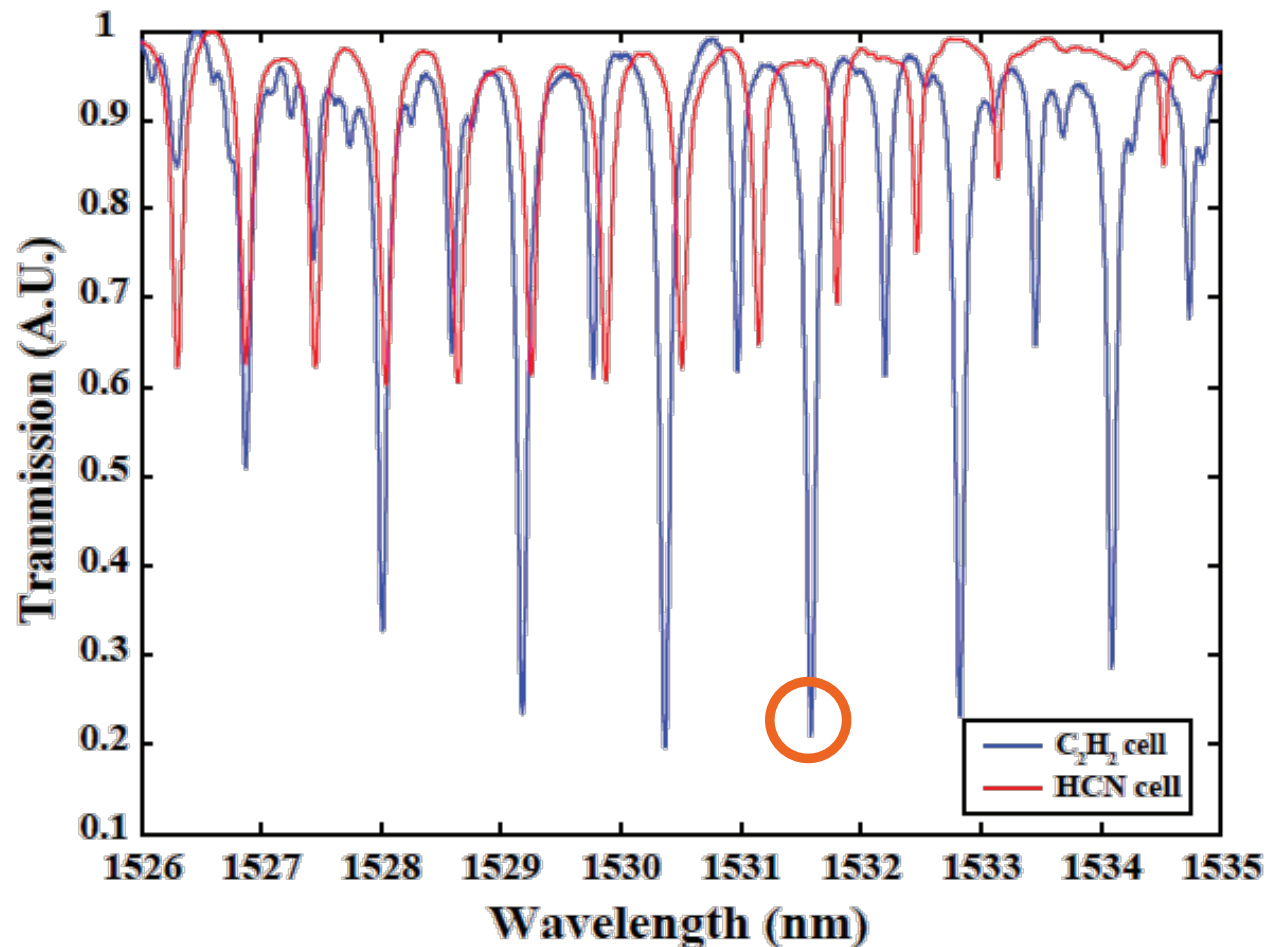


- Resolution (quarter wave) is 17 pm or 2.1 GHz or  $0.071 \text{ cm}^{-1}$
- (Slow-light waveguide is only 1 mm long!)

Magaña-Loaiza, Gao, Schulz, Awan, Upham, Dolgaleva, and Boyd, in review.

Challenge: Fabricate a chip-scale spectrometer that can discriminate acetylene ( $\text{H}_2\text{C}_2$ ) from hydrogen cyanide ( $\text{HCN}$ )?

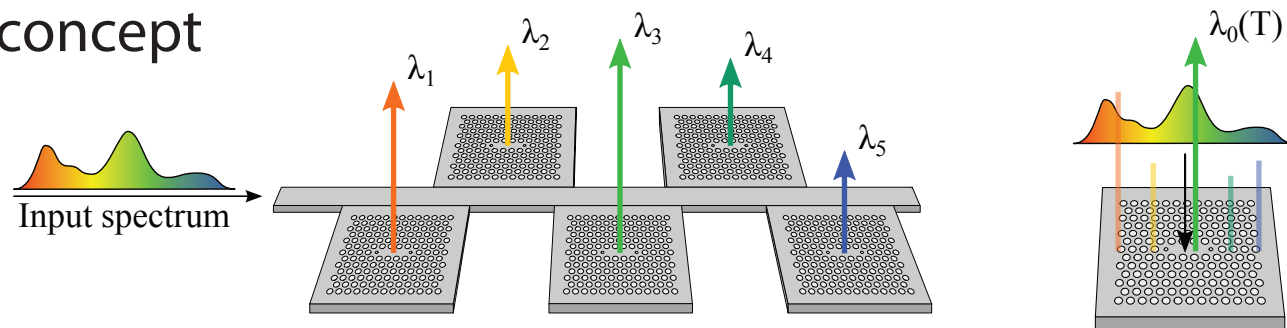
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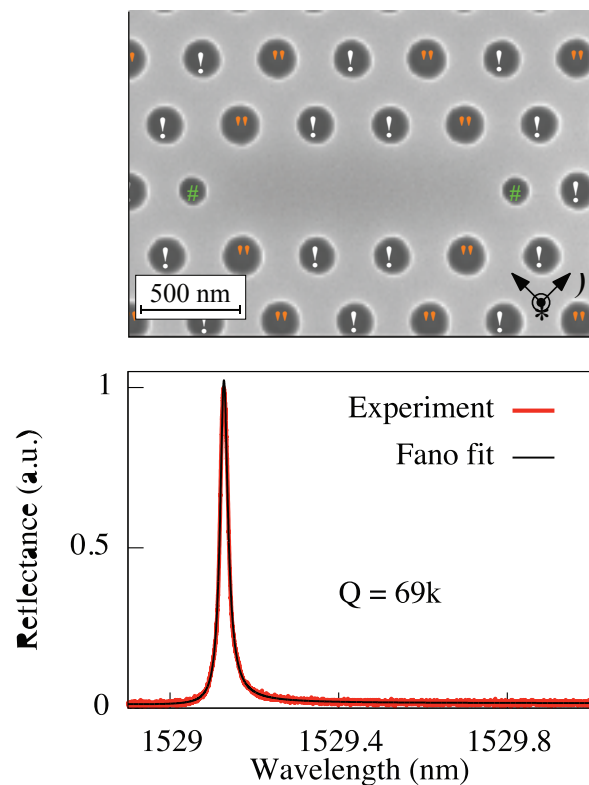
(data from our own lab)

# On-chip spectrometer based on high-Q photonic crystal cavities

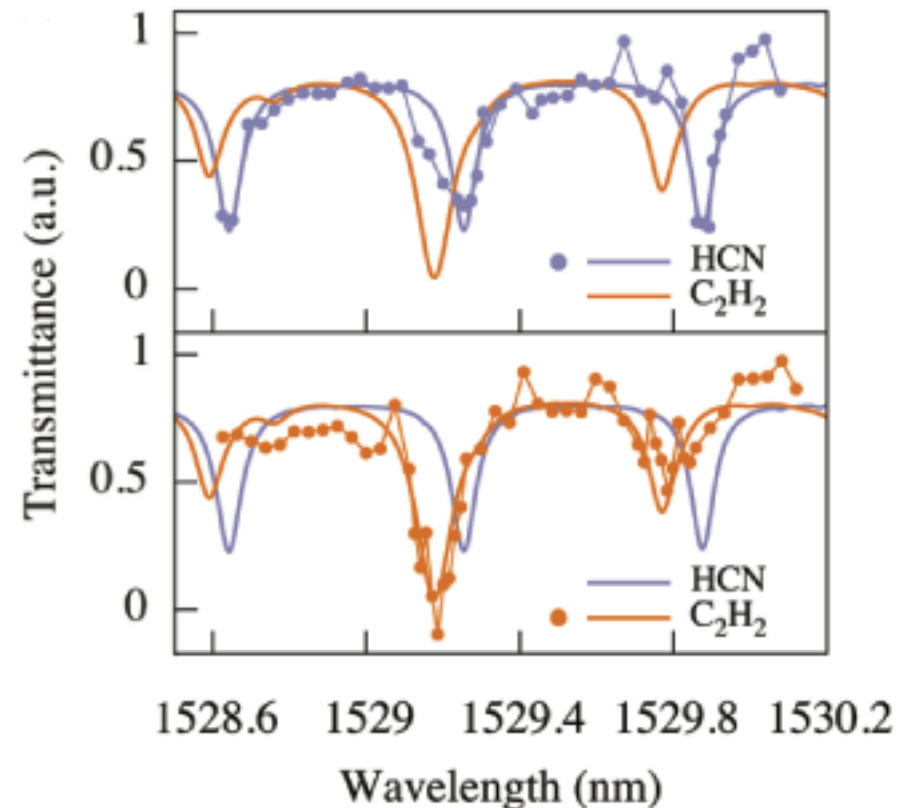
- The concept



- Cavity design



- Spectroscopy results





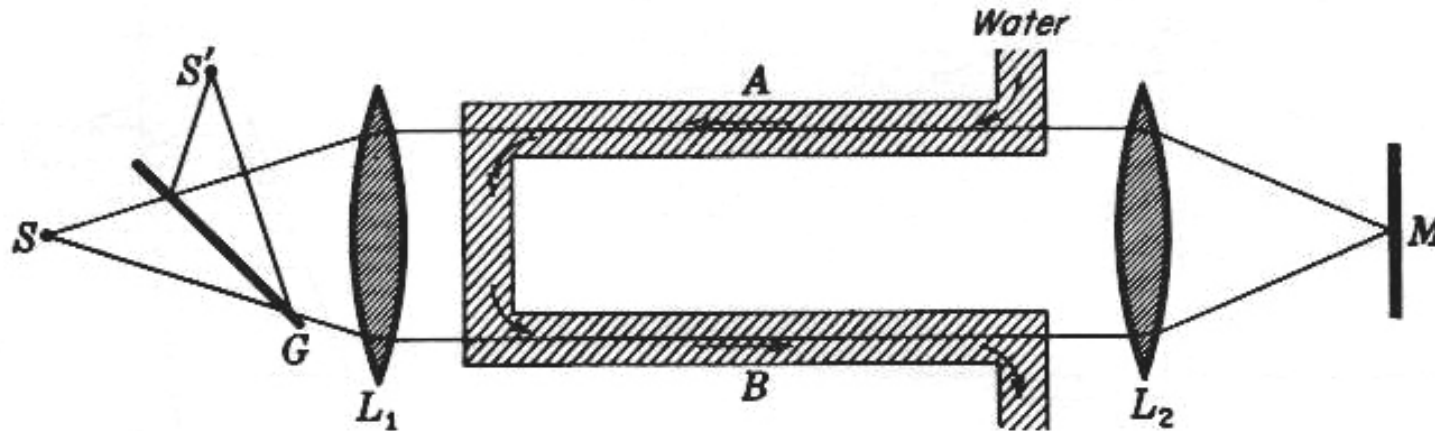
# The Velocity of Light in Moving Matter: Fresnel Drag (or Ether Drag ) Effects

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- Fizeau (1859): Longitudinal photon drag:

Velocity of light in flowing water.

$V = 700 \text{ cm/sec}$ ;  $L = 150 \text{ cm}$ ; displacement of 0.5 fringe.



- Modern theory: relativistic addition of velocities

$$v = \frac{c/n + V}{1 + (V/c)(1/n)} \approx \frac{c}{n} + V \left( 1 - \frac{1}{n^2} \right) \quad \text{— Fresnel “drag” coefficient}$$

- But what about slow-light media?

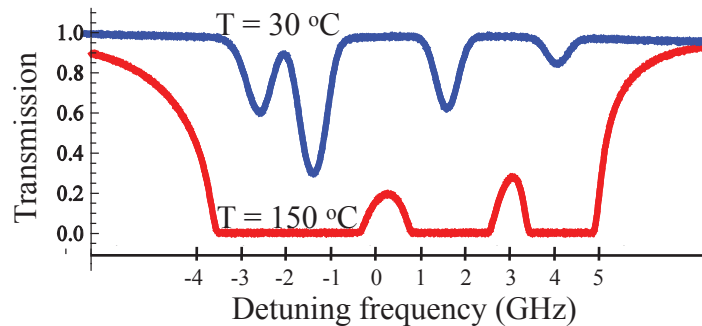
# Fresnel Drag in a Highly Dispersive Medium

## Light Drag in a Slow Light Medium (Lorentz)

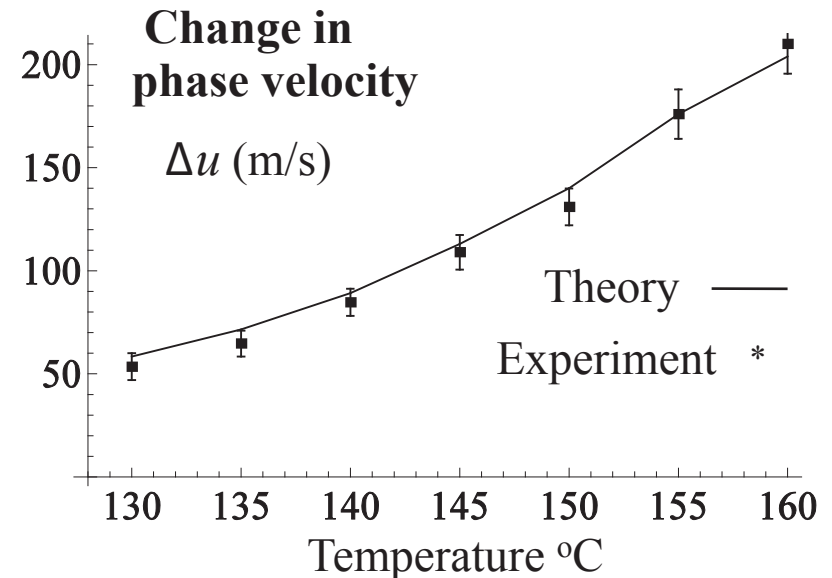
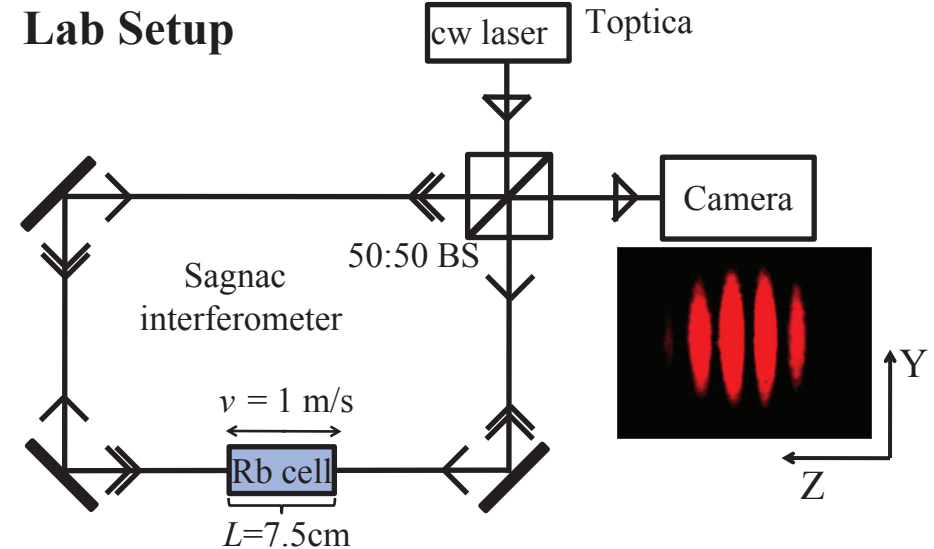
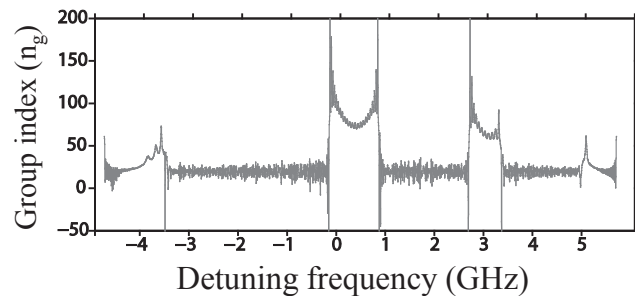
$$u \approx \frac{c}{n} \pm v \left( 1 - \frac{1}{n^2} + \frac{n_g - n}{n^2} \right)$$

## We Use Rubidium as Our Slow Light Medium

- Transmission spectrum of Rb around D<sub>2</sub> transition:



- Group index of Rb around D<sub>2</sub> line at T=130



- Change in phase velocity is much larger than velocity of rubidium cell. Implications for new velocimeters?

# Quantum Nonlinear Optics:

## Nonlinear Optics Meets the Quantum World

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Outlook: NLO is a superb platform from which to explore new physical processes and to develop photonics applications.

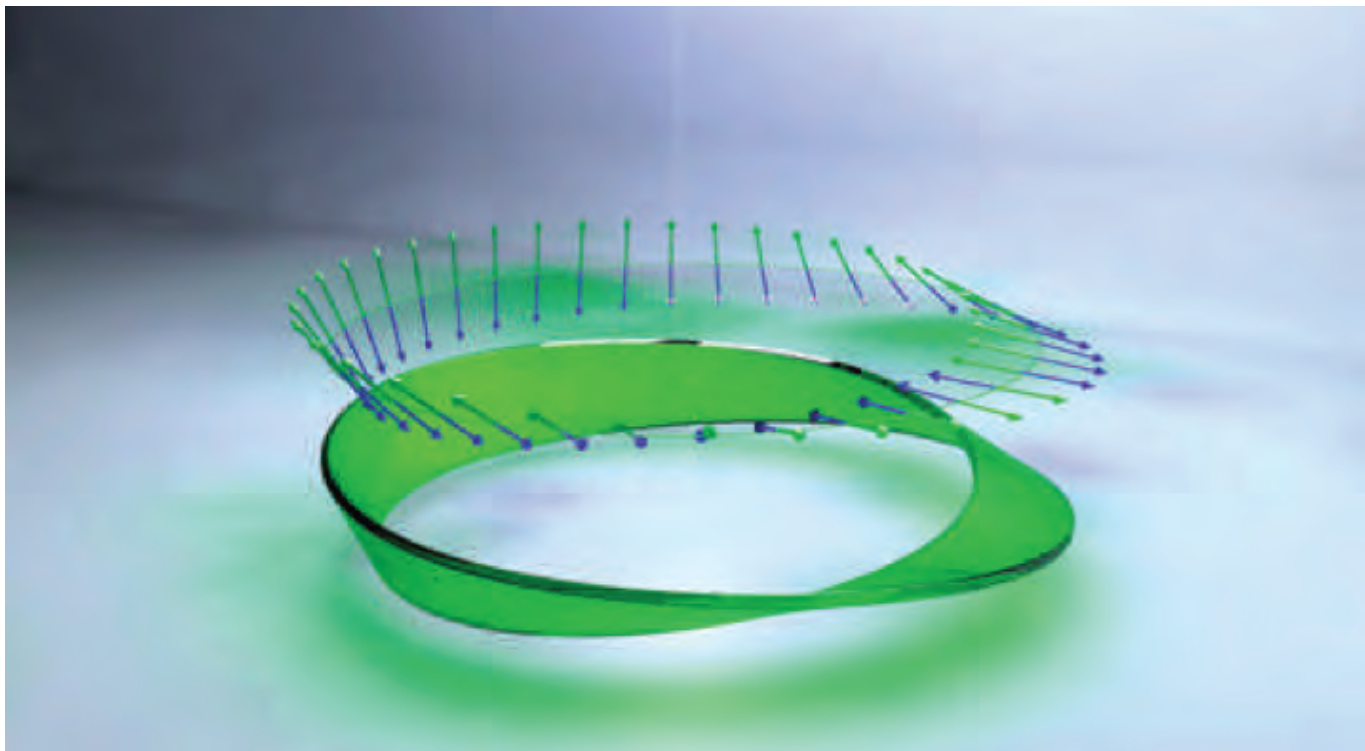
### Prospectus

1. Introduction to Nonlinear Optics and Quantum NLO
2. New Applications of “Slow Light”
3. Möbius Strips of Polarization
4. Huge Optical Nonlinearity in Epsilon-Near-Zero Materials
5. Quantum Communication with Multiple Bits per Photon

# Observation of Optical Polarization Möbius Strips

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- Möbius strips are familiar geometrical structures, but their occurrence in nature is extremely rare.
- We generate such structures in the nanoscale in tightly focused vector light beams and confirm experimentally their Möbius topology.



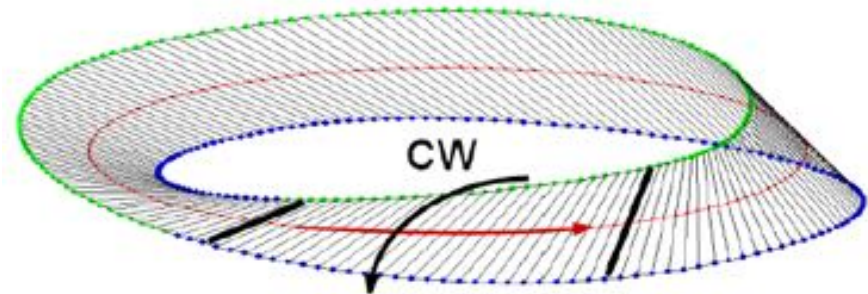
Bauer, Banzer, Karimi, Orlov, Rubano, Marrucci, Santamato, Boyd and Leuchs, Science, 347, 964 (2015)

# Prediction of Optical Möbius Strips

An “ordinary” Möbius strip



A polarization Möbius strip  
(introduced by Isaac Freund)



- Isaac Freund discovered, described, and investigated these unusual structures
- To observe these structures, one needs to create a very special field distribution (e.g., a Poincaré beam)
- One also needs to observe the field distribution in a very special way (measure polarization as a function of position around a very tightly focused light beam)

<sup>1</sup> Wikipedia

<sup>2</sup> Isaac Freund, Bar-Ilan Univ., Talk: *Optical Moebius Strips and Twisted Ribbons*, Conf. on Singular Optics, ICTP Trieste, Part II, 30 May 2011

Isaac Freund, Opt. Commun. 242, 65-78 (2004)

Isaac Freund, Opt. Commun. 249, 7-22 (2005)

Isaac Freund, Opt. Commun. 256, 220-241 (2005)

Isaac Freund, Opt. Commun. 283, 1-15 (2010)

Isaac Freund, Opt. Commun. 283, 16-28 (2010)

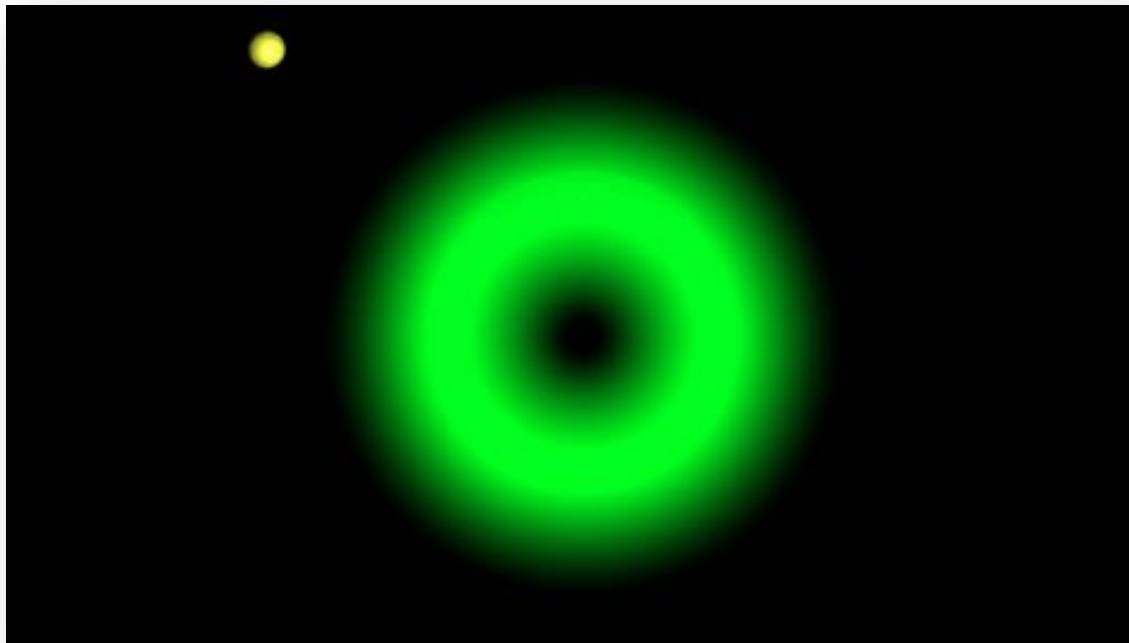
Isaac Freund, Opt. Lett. 35, 148-150 (2010)

Isaac Freund, Opt. Commun. 284, 3816-3845 (2011)

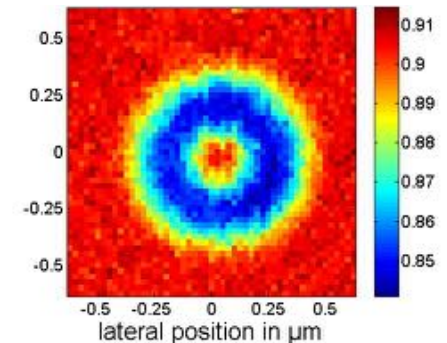
# Full vectorial beam measurement on the nanoscale

## Nanoparticle-based probing technique for vector beam reconstruction

1. A dipole-like spherical nanoparticle (90 nm diameter) is scanned through the beam
2. The forward- and backward-scattered light for each position of the nanoparticle relative to the beam in the focal plane is measured



measured intensity  
(can also measure  
polarization and phase)

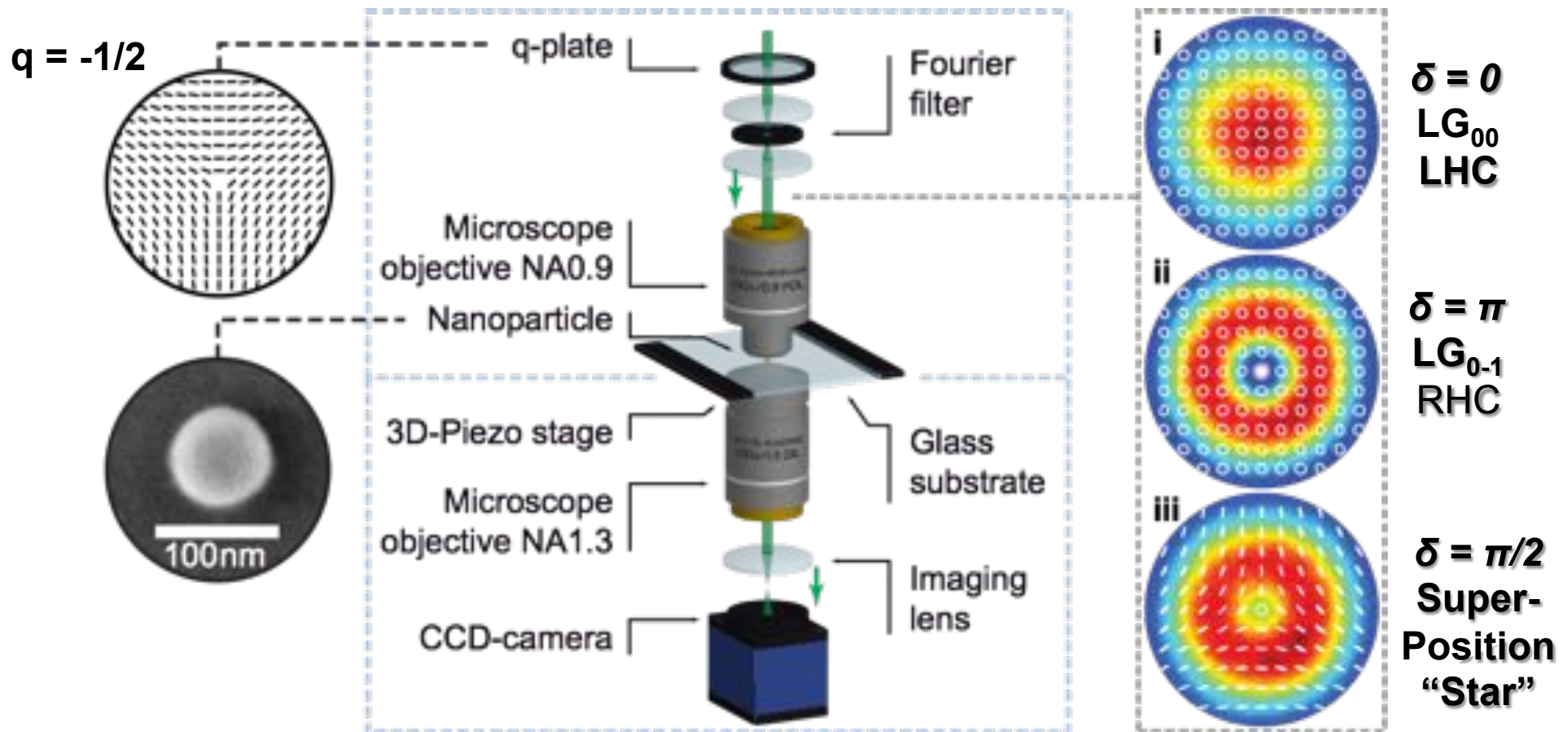


### Full amplitude and phase reconstruction scheme:

T. Bauer, S. Orlov, U. Peschel, P. B. and G. Leuchs, "Nanointerferometric Amplitude and Phase Reconstruction of Tightly Focused Vector Beams", Nat. Photon 8, 23 - 27 (2014).



# Lab Setup to Observe a Polarization Möbius Strip

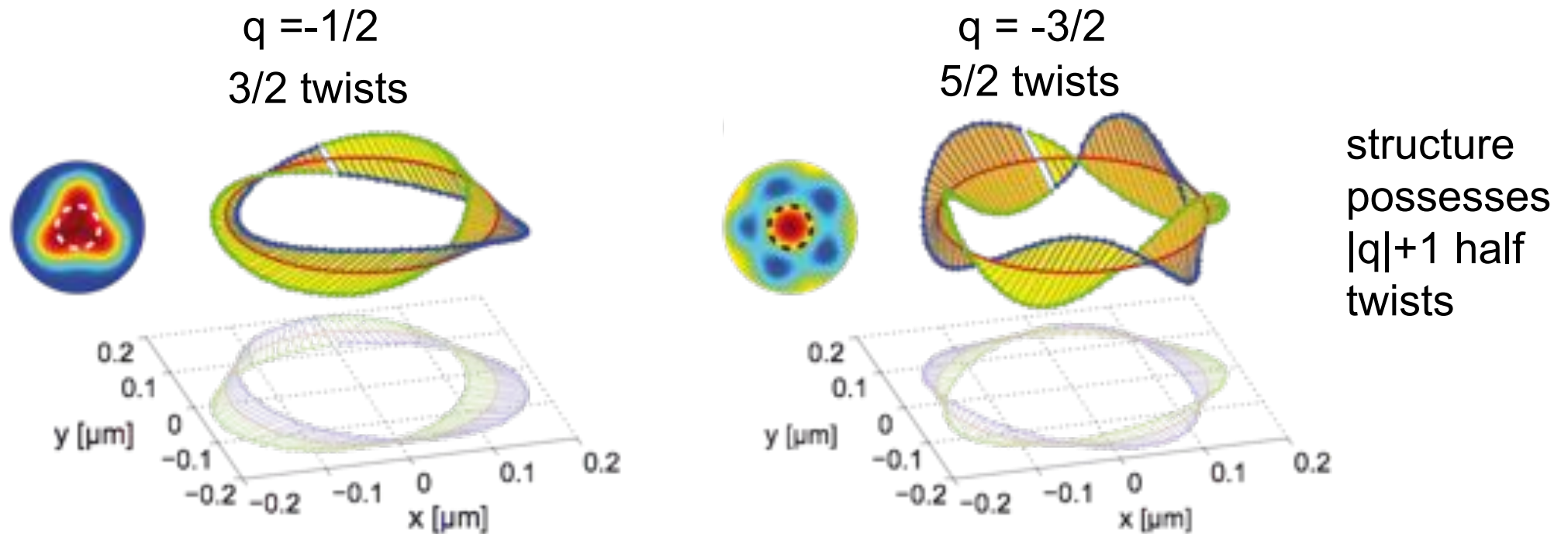


- q-plate: waveplate with a spatially varying orientation ( $q$  is the topological charge)
- output beam has a spatially varying state of polarization (vector beam, Poincaré beam, etc.)

Tight focusing enhances the Möbius effect, which depends on the  $z$  component of the field



# Observation of Polarization Möbius Strips



## Remarks

- First observation of a polarization Möbius strip
- Light fields can possess rich spatial structure on subwavelength scales
- Current technology is capable of controllably creating beams with such structures and measuring it at subwavelength distances.

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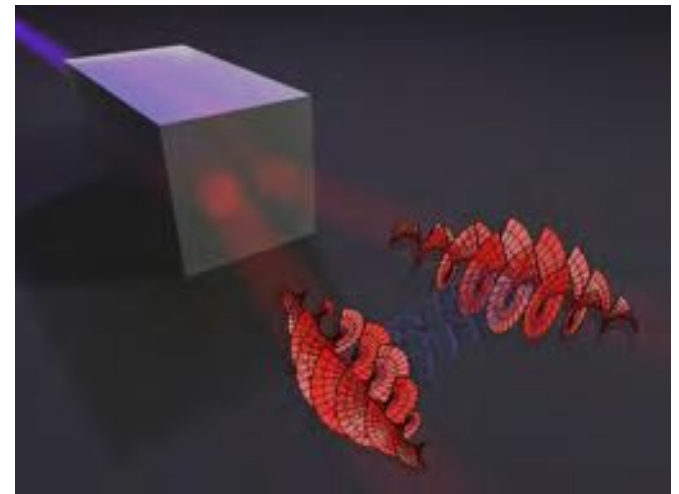
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# Use of Quantum States for Secure Optical Communication

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- The celebrated BB84 protocol for quantum key distribution (QKD) transmits one bit of information per received photon
- We have built a QKD system that can carry more than one bit per photon.
  - Note that in traditional telecom, one uses many photons per bit!
- Our procedure is to encode using beams that carry orbital angular momentum (OAM), such as the Laguerre-Gauss states, which reside in an infinite dimensional Hilbert space.



# QKD System Carrying Many Bits Per Photon

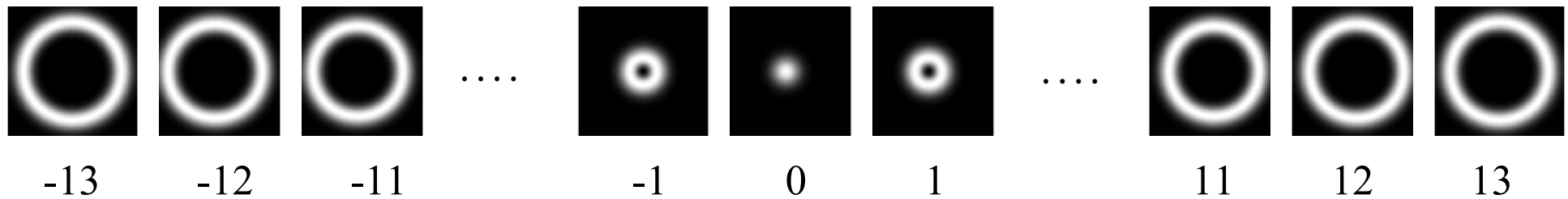
We are constructing a QKD system in which each photon carries many bits of information

We encode in states that carry OAM such as the Laguerre-Gauss states

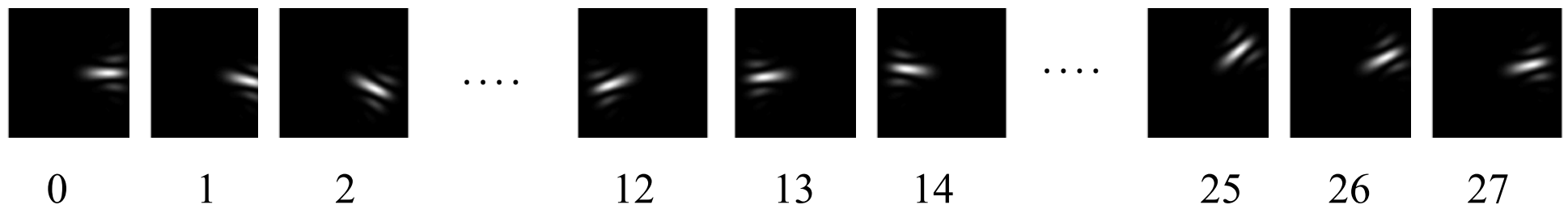
We also need a second basis composed of linear combinations of these states

## Single Photon States

*Laguerre-Gaussian Basis*  $\ell = -13, \dots, 13$

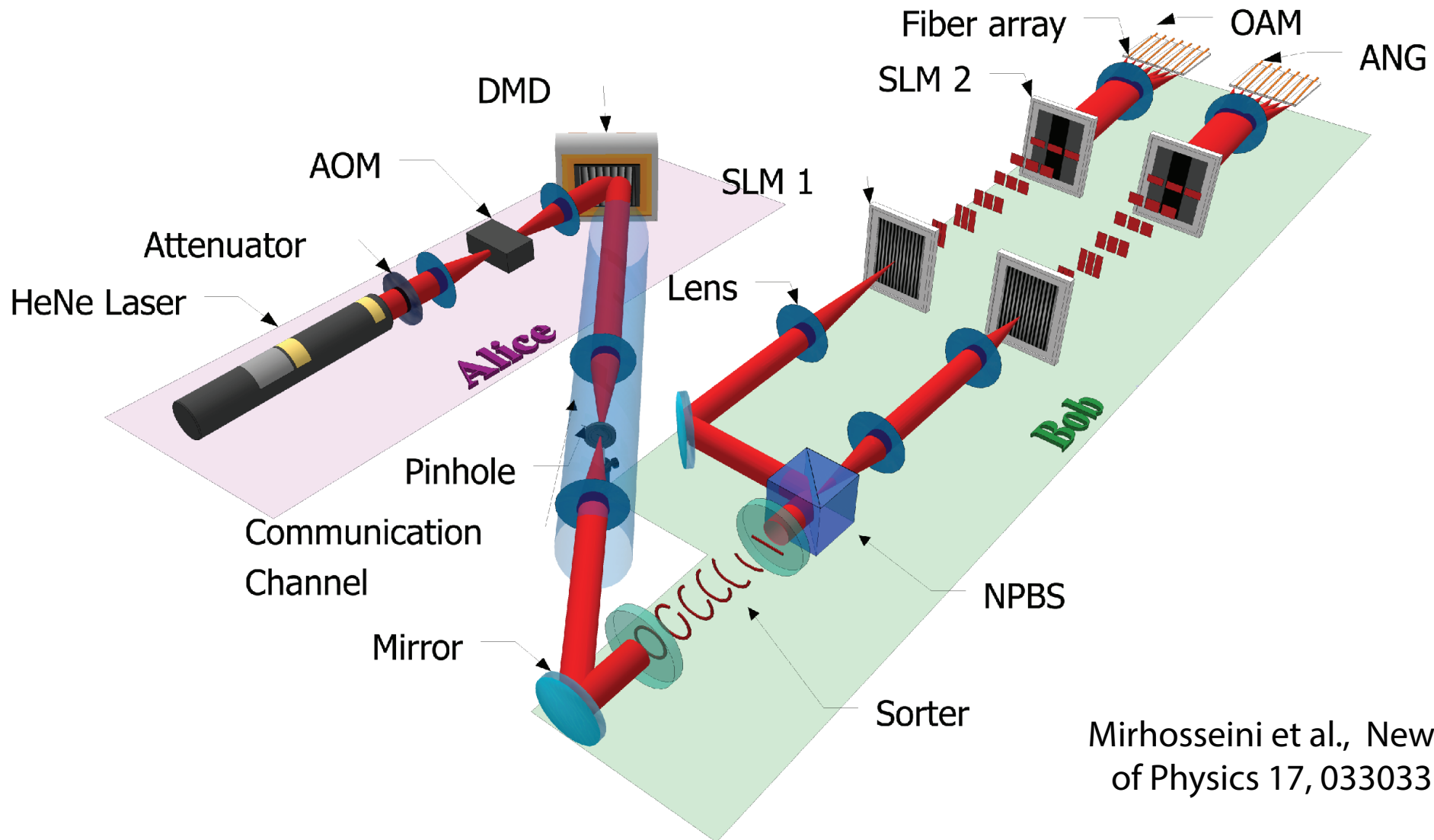


*“Angular” Basis (mutually unbiased with respect to LG)*



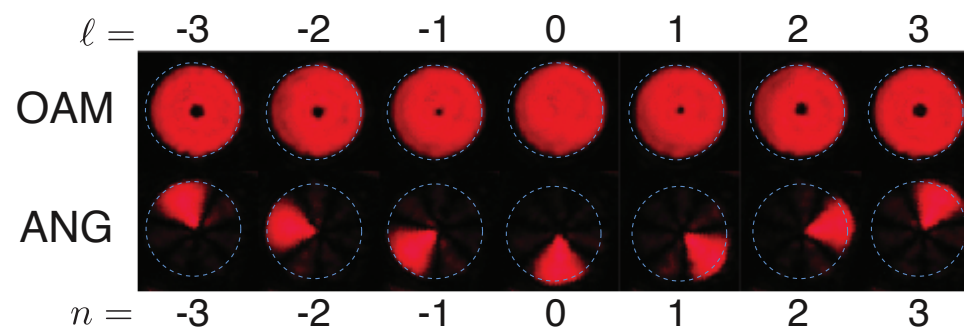
$$\Psi_{AB}^N = \frac{1}{\sqrt{27}} \sum_{l=-13}^{13} \text{LG}_{l,0} \exp(i2\pi Nl/27)$$

# Our Laboratory Setup

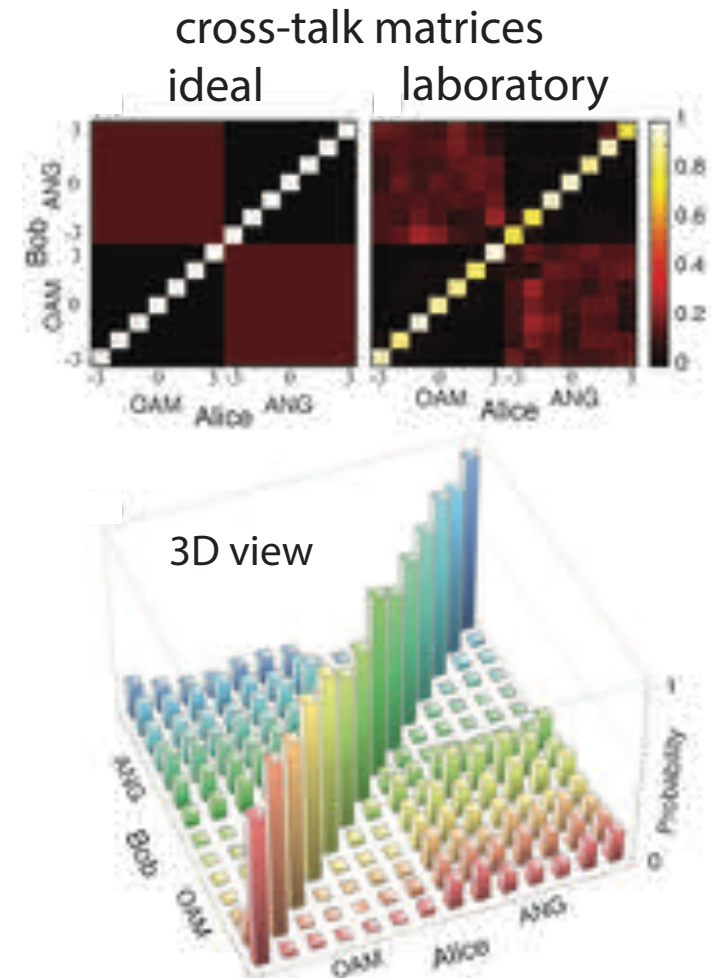
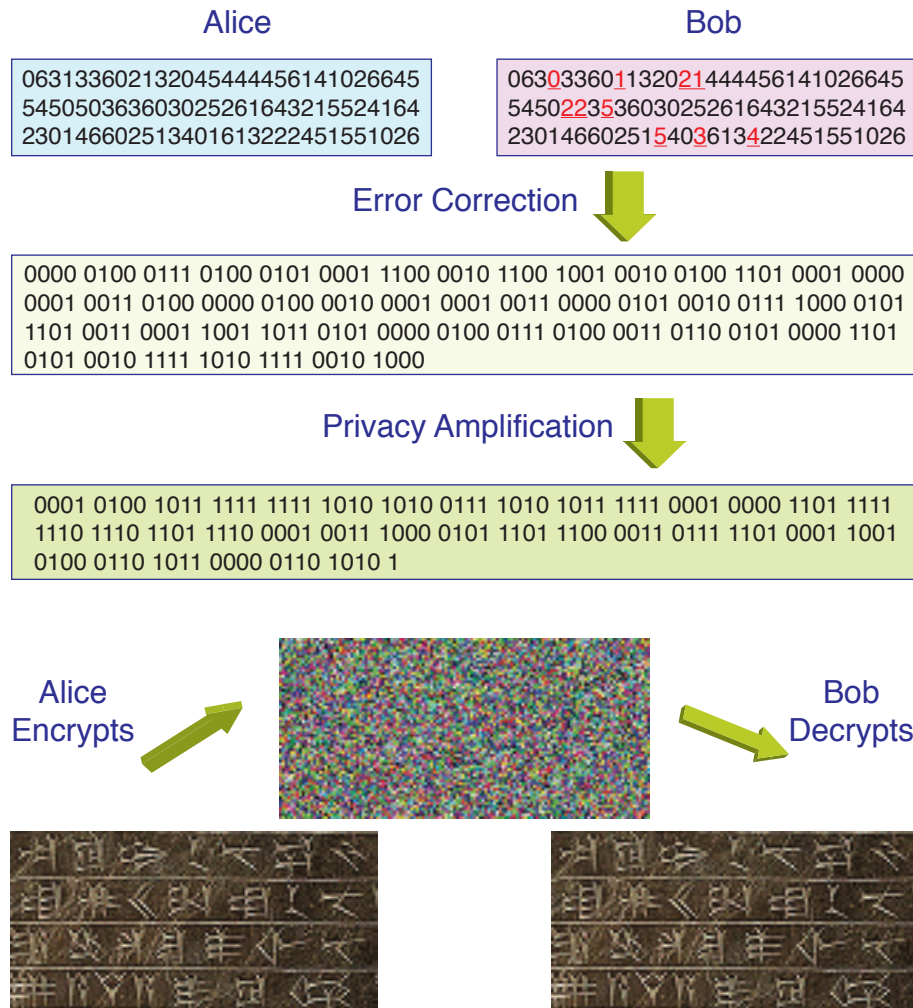


Mirhosseini et al., New Journal of Physics 17, 033033 (2015).

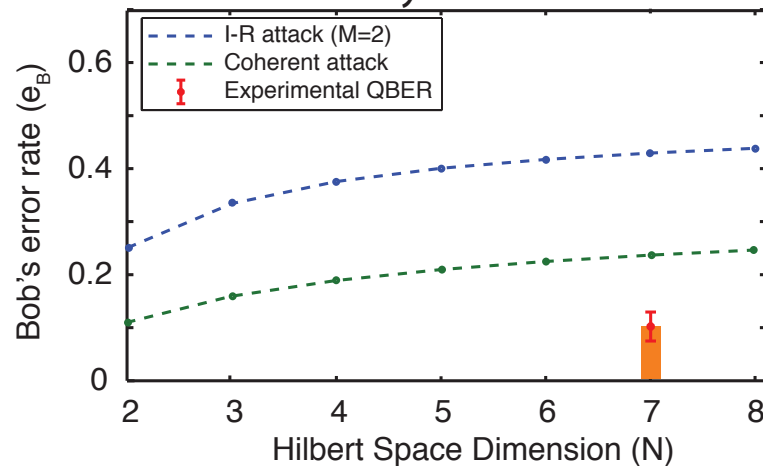
We use a seven-dimensional state space.



# Laboratory Results - OAM-Based QKD



- error bounds for security



We use a 7-letter alphabet, and achieve a channel capacity of 2.1 bits per sifted photon.

We do not reach the full 2.8 bits per photon for a variety of reasons, including dark counts in our detectors and cross-talk among channels resulting from imperfections in our sorter.

Nonetheless, our error rate is adequately low to provide full security,

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**Thank you for your attention!**

