



Quantum Nonlinear Optics: Nonlinear Optics Meets the Quantum World

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Quantum Nonlinear Optics:

Nonlinear Optics Meets the Quantum World

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

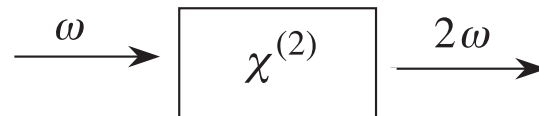
Simple Formulation of the Theory of Nonlinear Optics

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

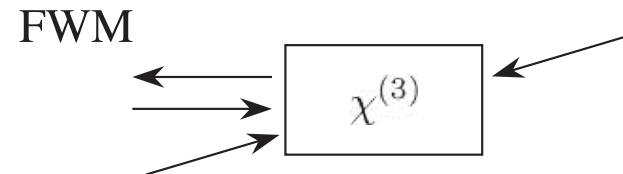
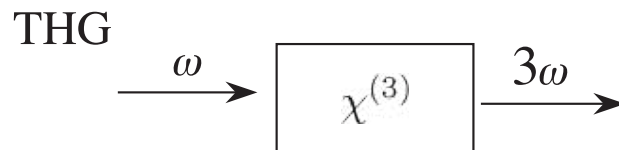
Here P is the induced dipole moment per unit volume and E is the field amplitude

$\chi^{(1)}$ 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

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

Intense Field and Attosecond Physics

PHYSICAL REVIEW LETTERS

PHYSICAL REVIEW LETTERS

13 MARCH

Above Threshold Ionization Beyond the High Harmonic Cutoff

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

⁽¹⁾Lawrence Livermore National Laboratory, Livermore, California 94550

⁽²⁾Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973

(Received 2 December 1992)

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PHYSICAL REVIEW LETTERS

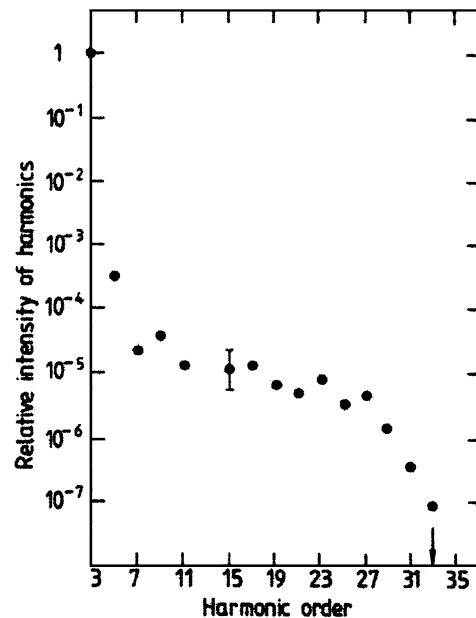
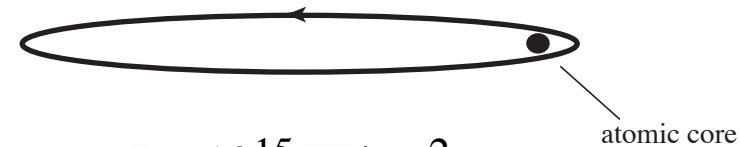
27 SEPTEMBER 1993

Plasma Perspective on Strong-Field Multiphoton Ionization

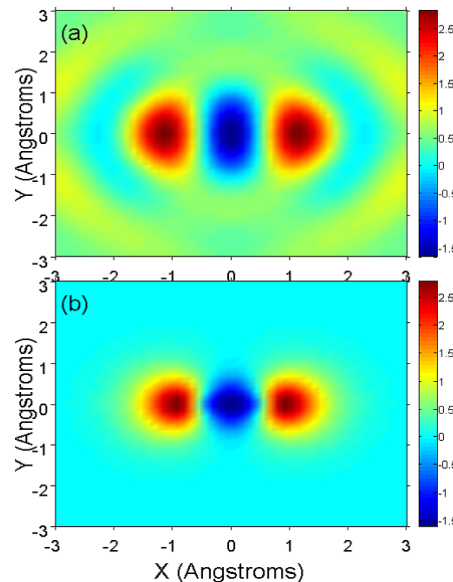
P. B. Corkum

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

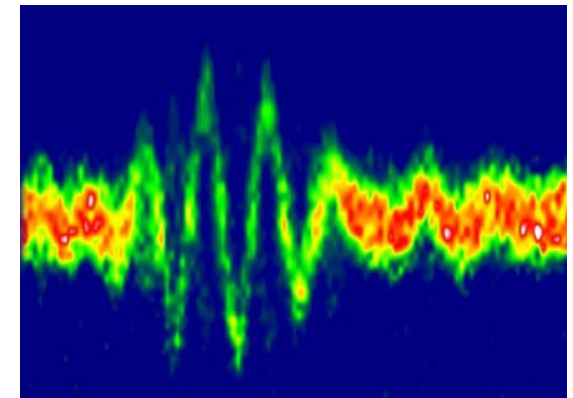
(Received 9 February 1993)



High-harmonic generation



Measuring the molecular nitrogen wavefunction



Attosecond pulses to sample a visible E-field



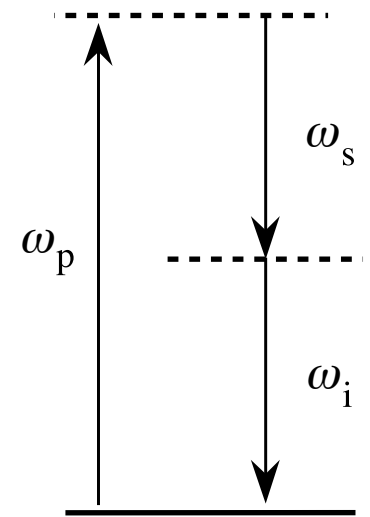
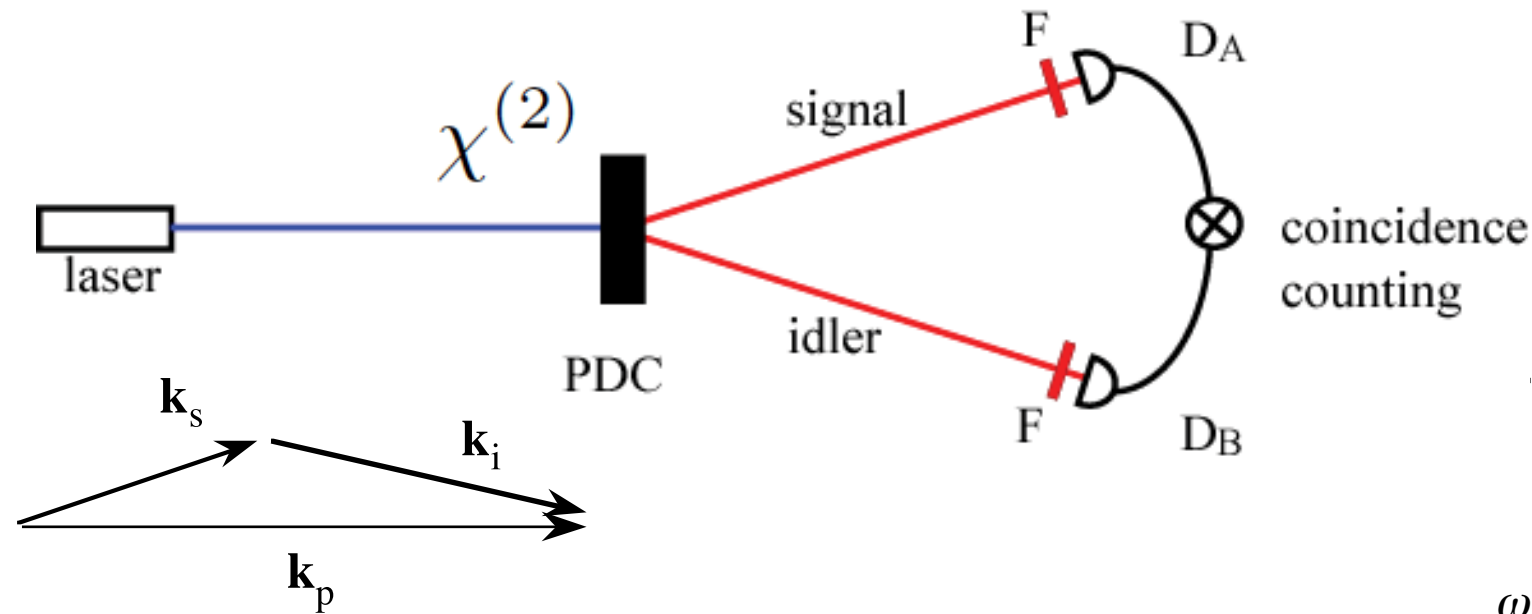
Why Interest in Quantum Nonlinear Optics?

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



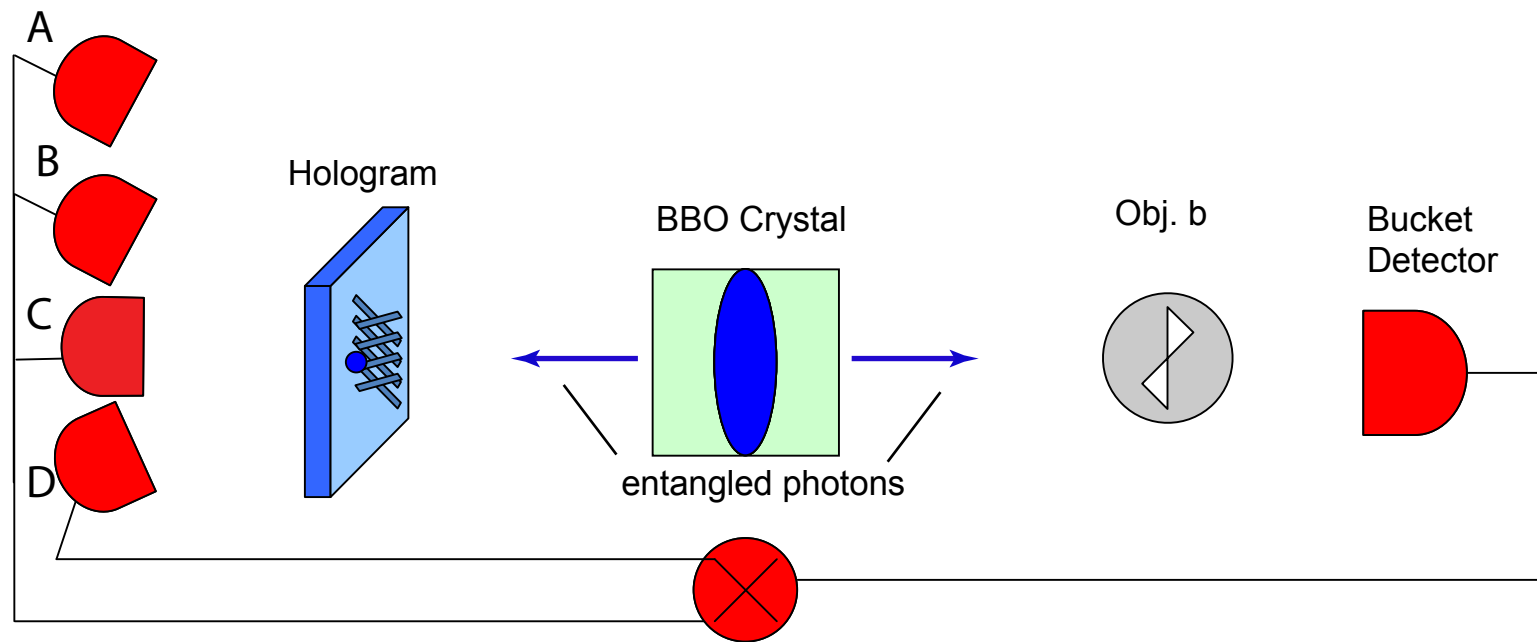
The signal and idler photons are entangled in:

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

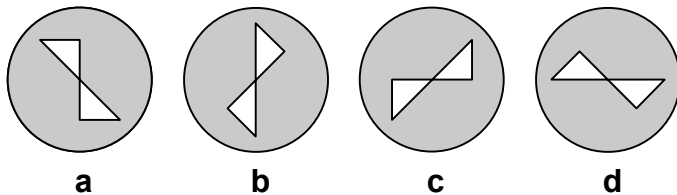
Entanglement is important for:

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

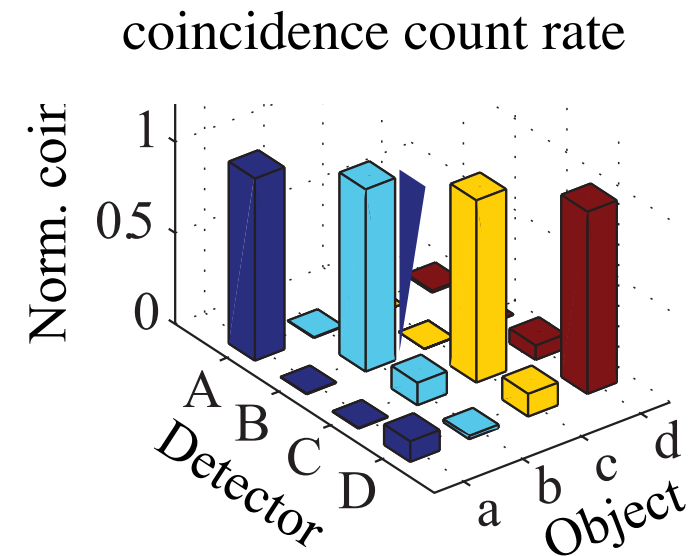
Single-Photon Coincidence Imaging



- 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.



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Controlling the Velocity of Light

“Slow,” “Fast” and “Backwards” Light

– Light can be made to go:

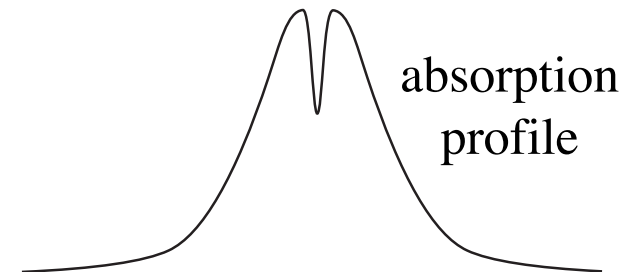
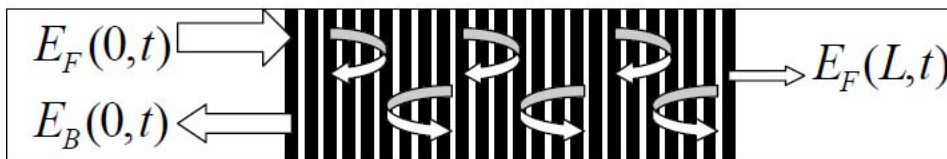
slow: $v_g \ll c$ (as much as 10^6 times slower!)

fast: $v_g > c$

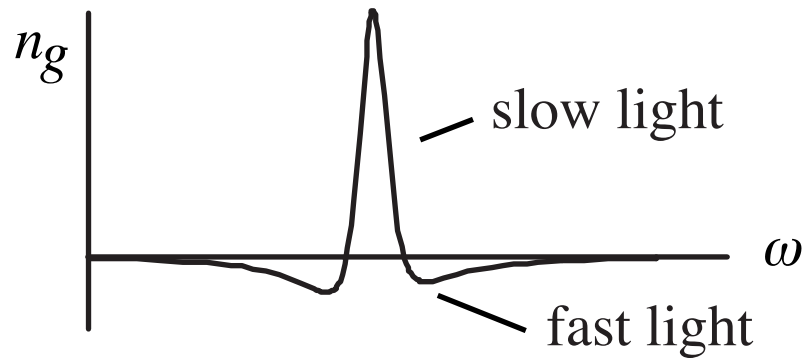
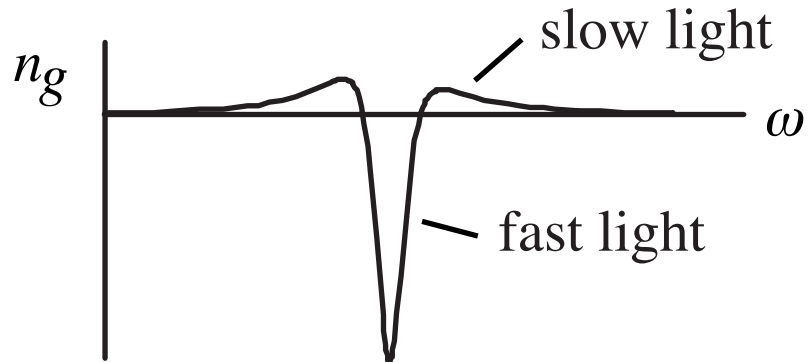
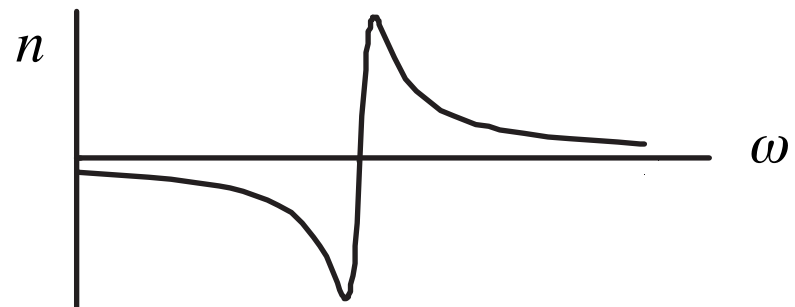
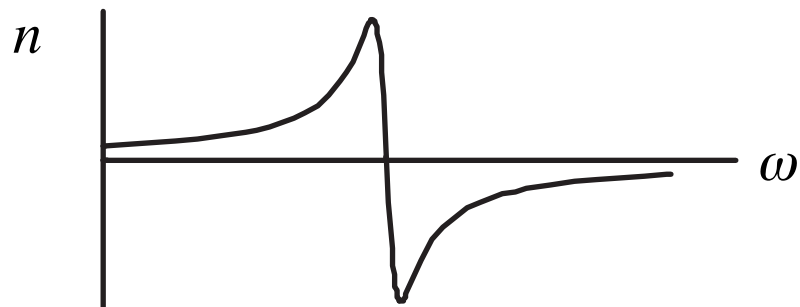
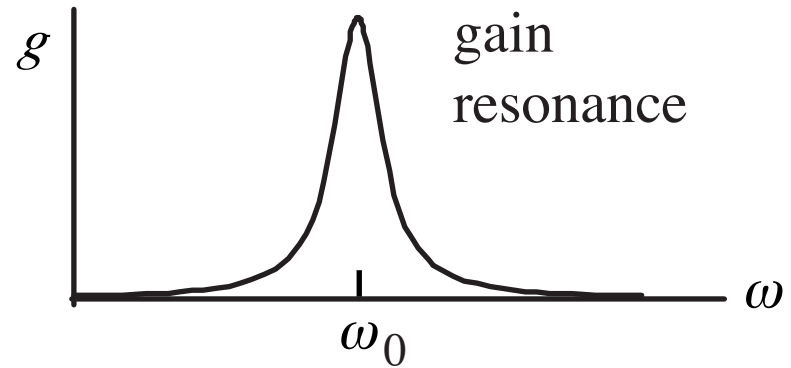
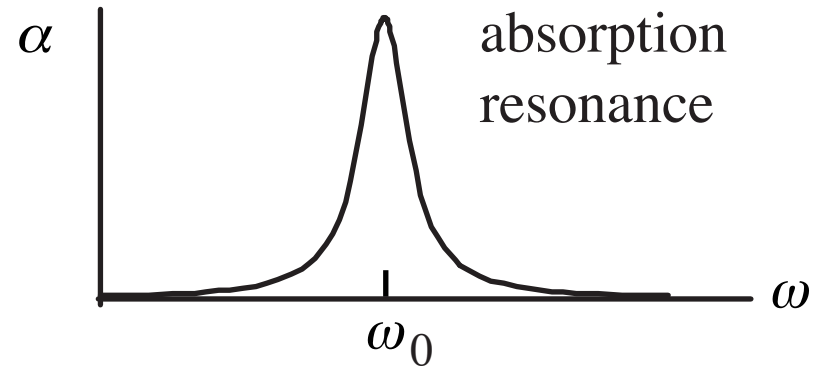
backwards: v_g negative

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



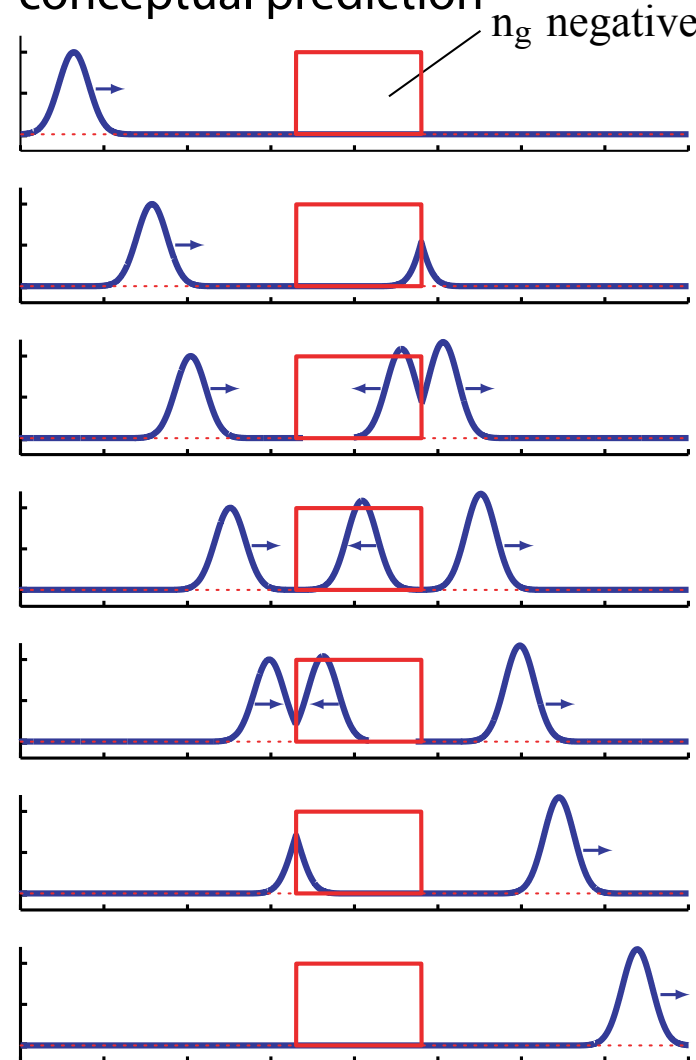
$$n_g = n + \omega (dn/d\omega)$$

Observation of Superluminal and “Backwards” Pulse Propagation



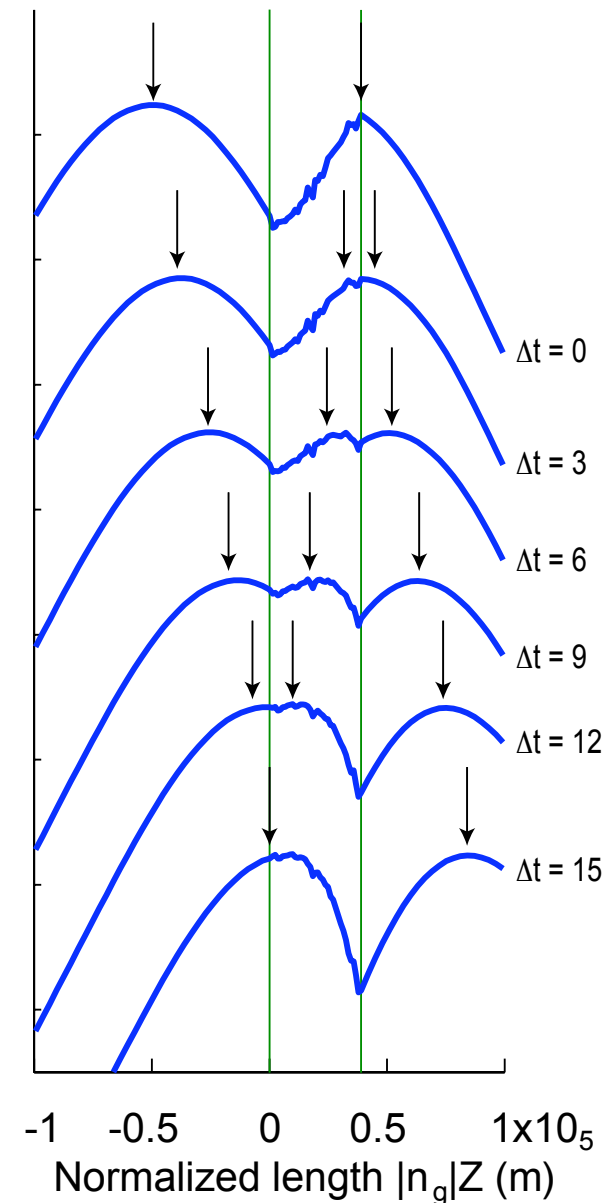
- A strongly counterintuitive phenomenon
- But entirely consistent with established physics
- Predicted by Garrett and McCumber (1970) and Chiao (1993).
- Observed by Gehring, Schweinsberg, Barsi, Kostinski, and Boyd Science 312, 985 2006.

- conceptual prediction



propagation distance

- laboratory results

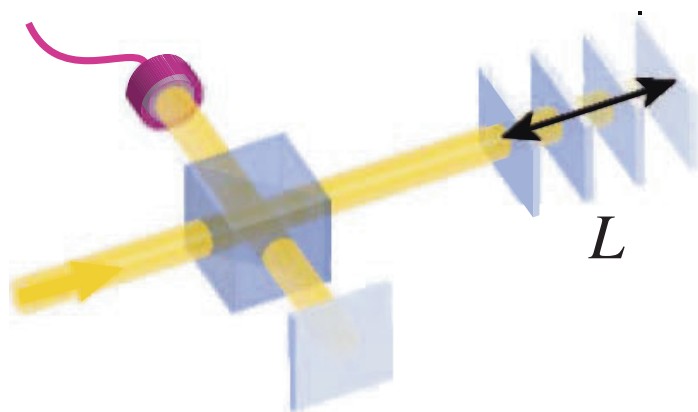


Development of Miniaturized, Chip-Scale Spectrometers

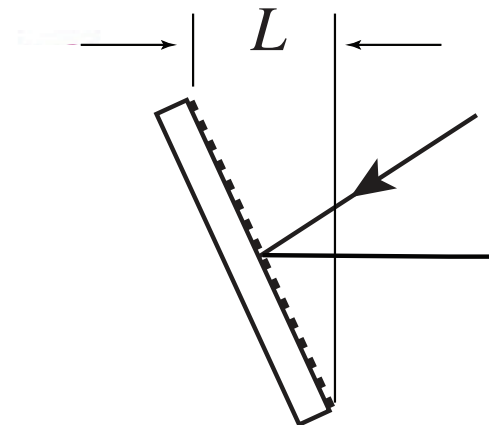
Can We Beat the $1/L$ Resolution Limit of Standard Spectrometers?

- 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



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

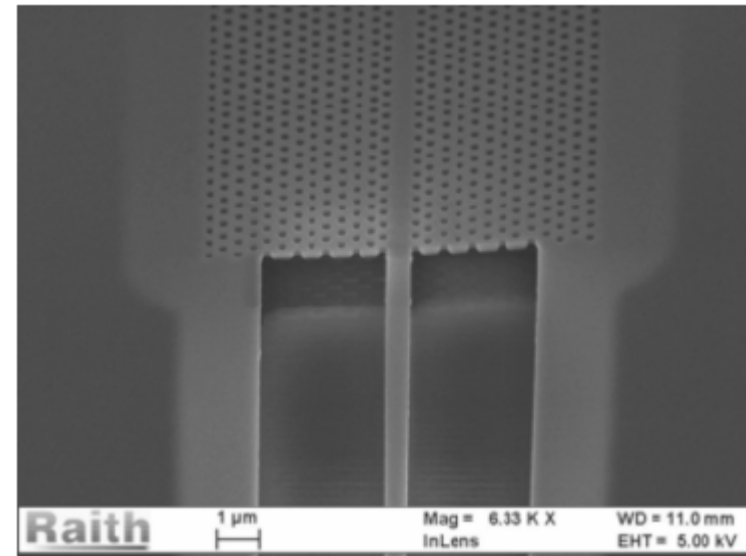
- We use slow-light methods to design spectrometers with resolution that exceeds this conventional limit by a factor as large as the group index.
- This ability allows us to miniaturize spectrometers with no loss of resolution, for “lab-on-a-chip” applications.

Our Goal

Replace this:



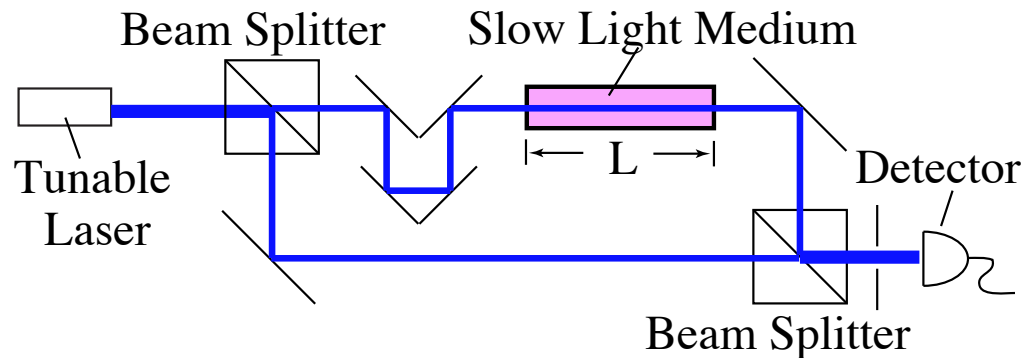
with this:



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:

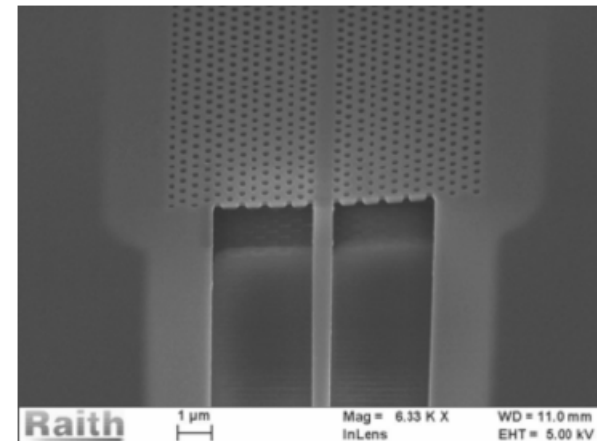


Simple analysis

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

- 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.

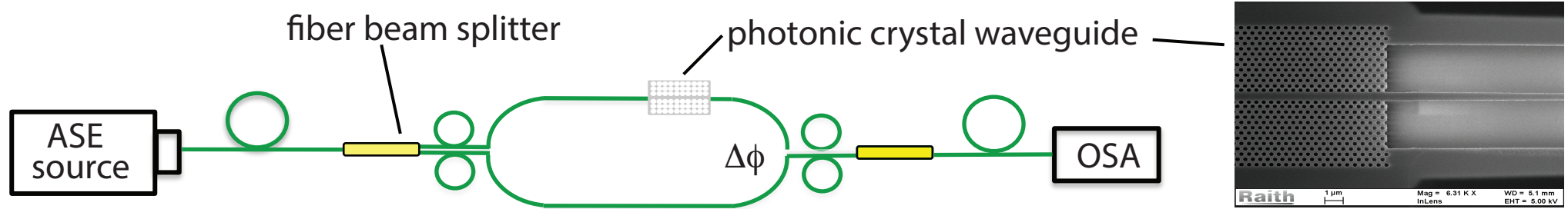


Shi, Boyd, Gauthier, and Dudley, Opt. Lett. 32, 915 (2007)

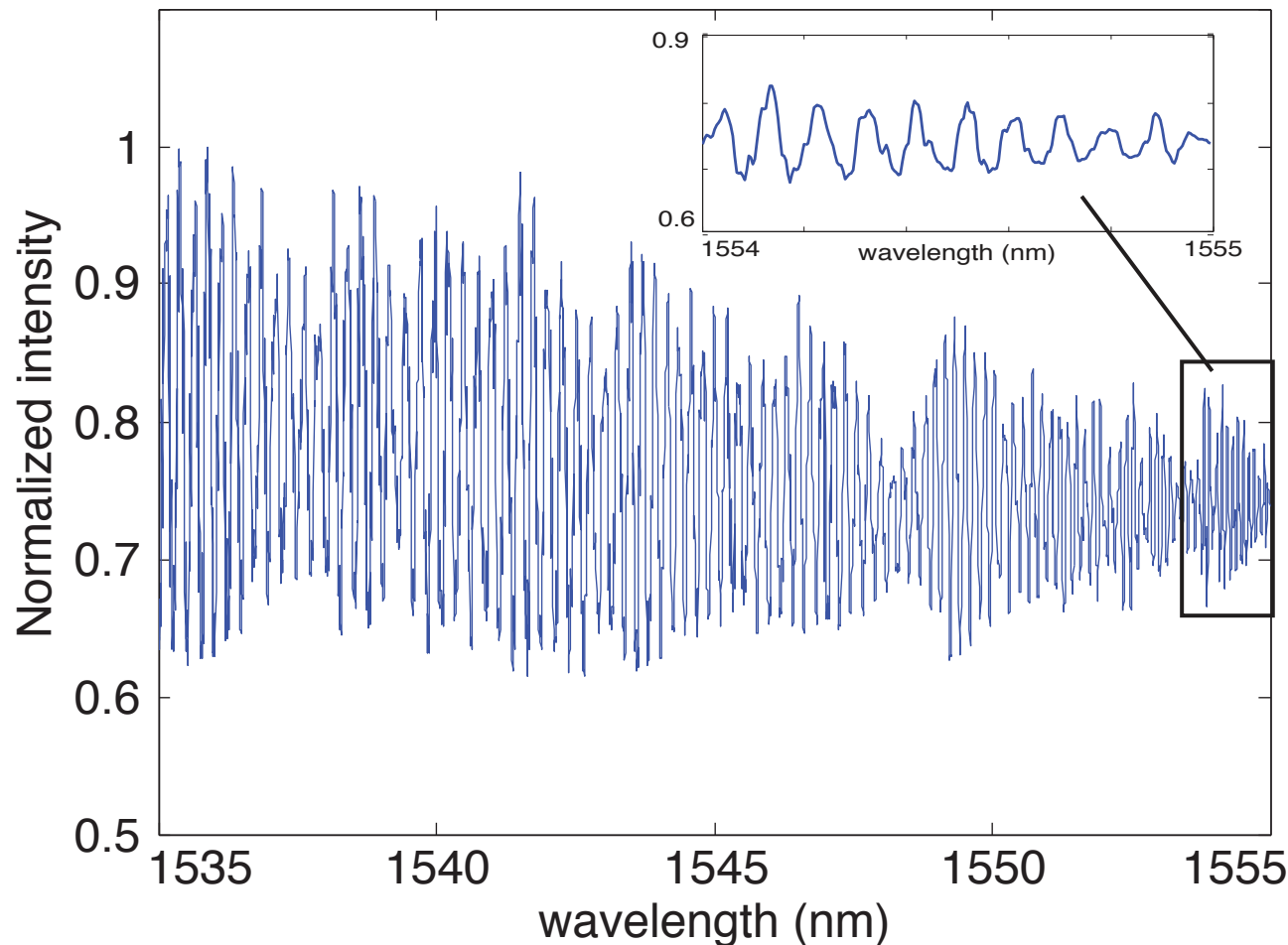
Shi, Boyd, Camacho, Vudiyasetu, and Howell, PRL. 99, 240801 (2007)

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

Laboratory Characterization of the Slow-Light Mach-Zehnder Interferometer



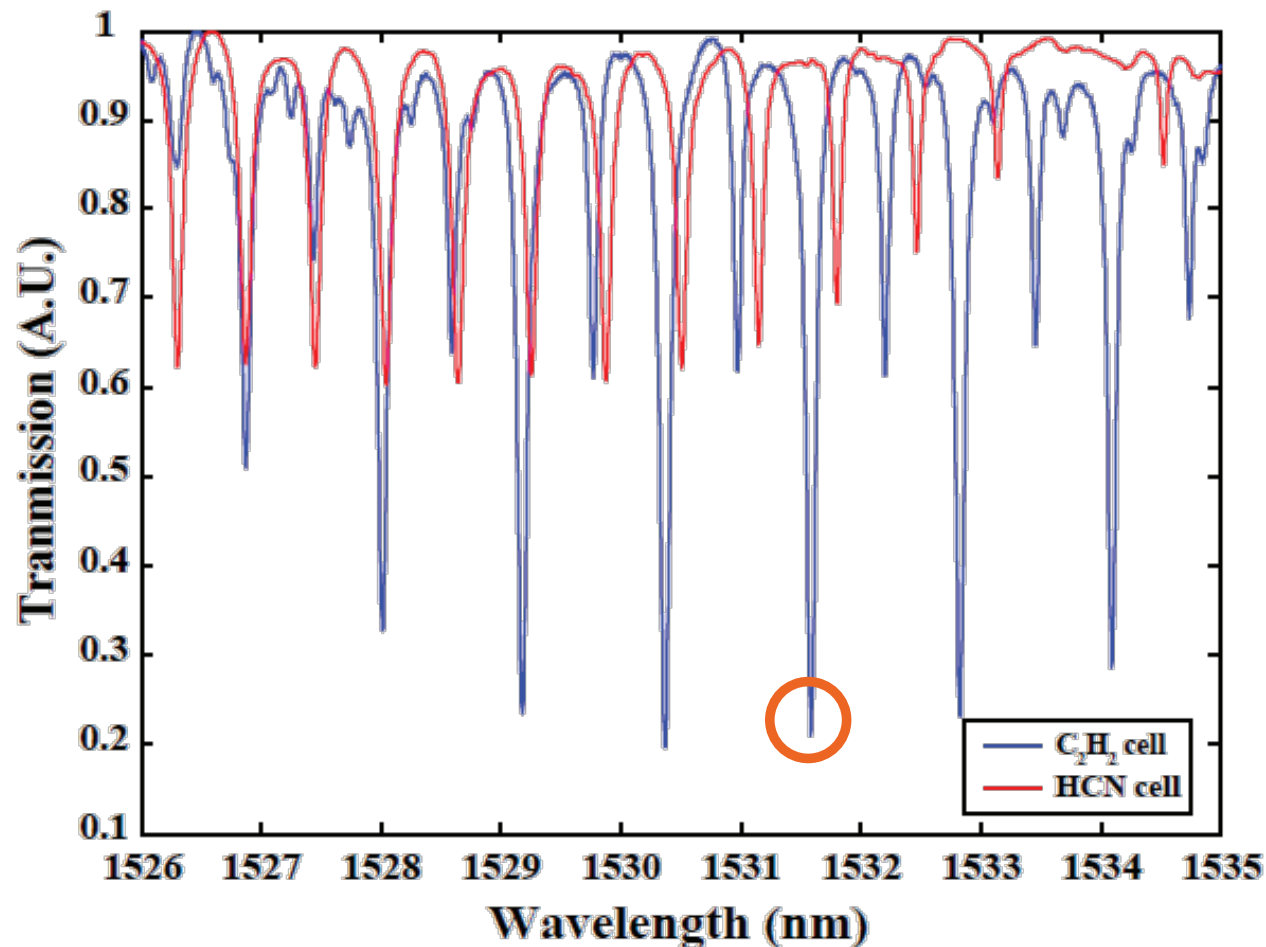
- Interference fringes



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

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

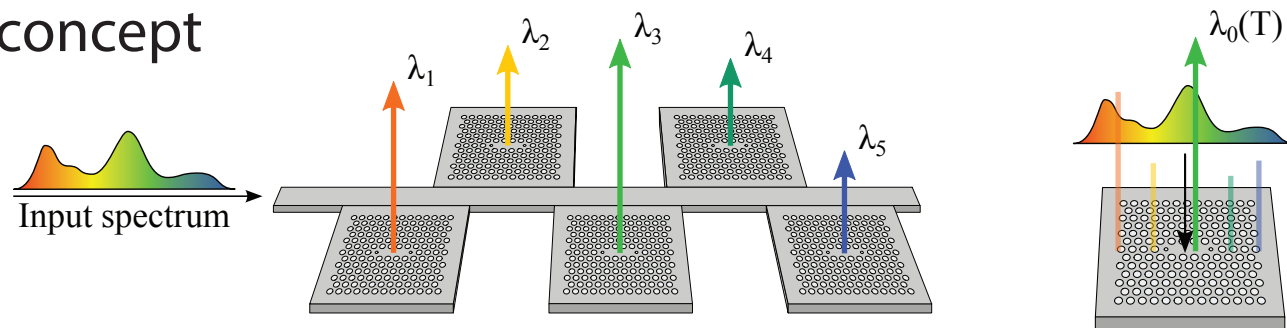
Challenge: Fabricate a chip-scale spectrometer that can discriminate acetylene (H_2C_2) from hydrogen cyanide (HCN)?



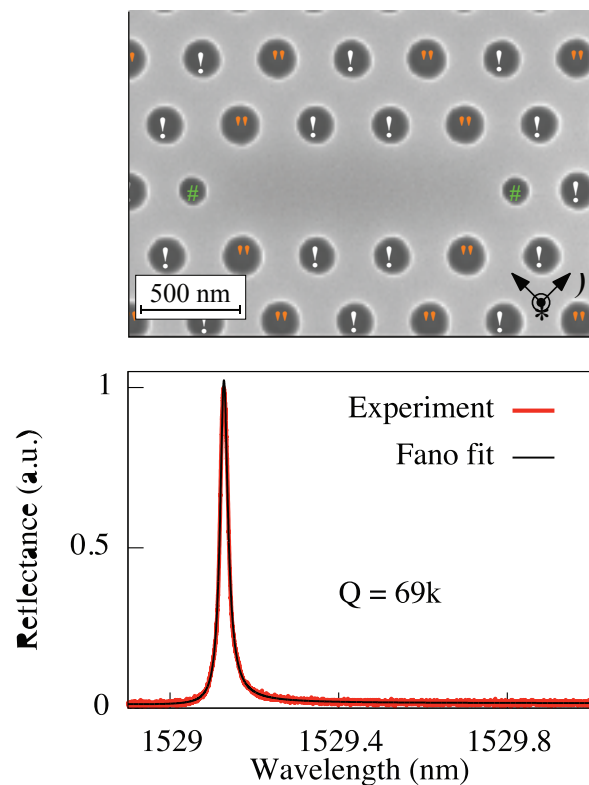
(data from our own lab)

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

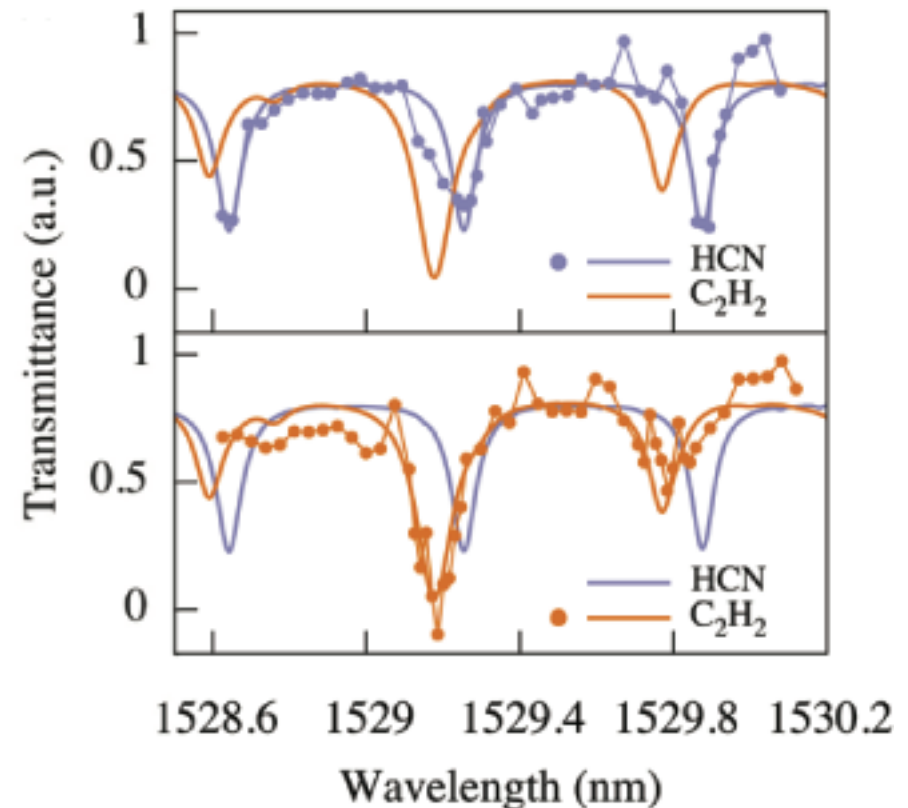
- The concept



- Cavity design



- Spectroscopy results

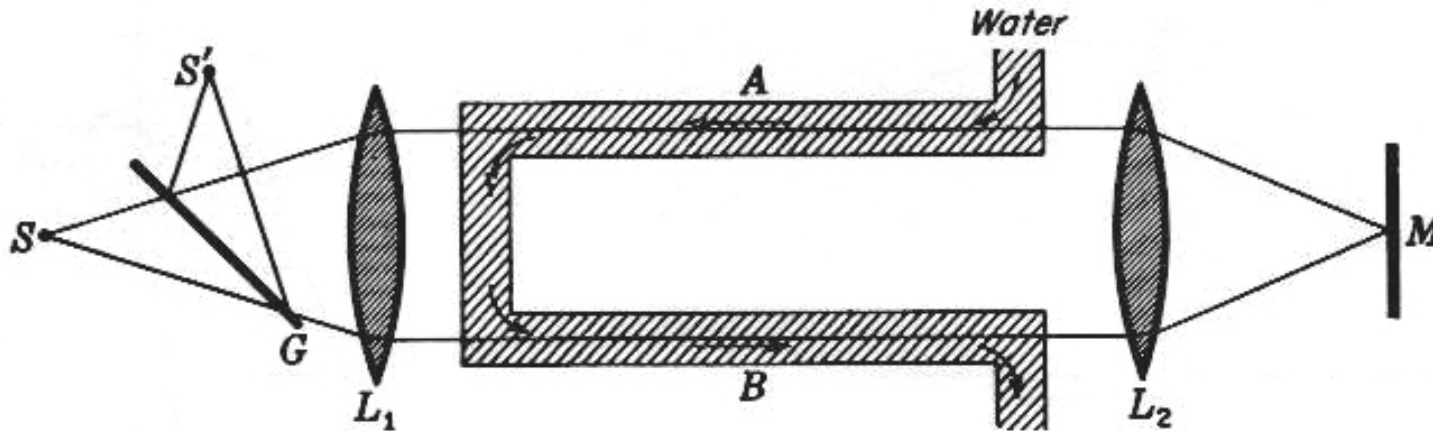


The Velocity of Light in Moving Matter: Fresnel Drag (or Ether Drag) Effects

- 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?

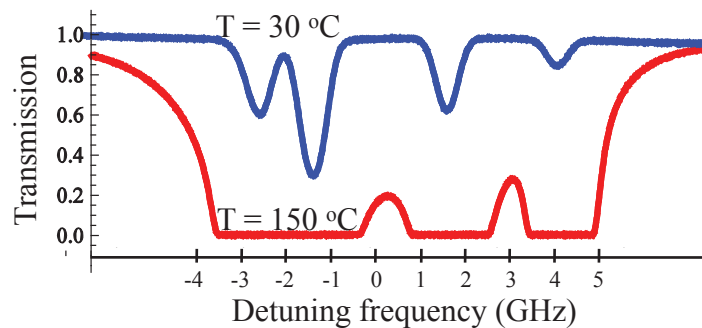
Fresnel Drag in a Highly Dispersive Medium

Light Drag in a Slow Light Medium (Lorentz)

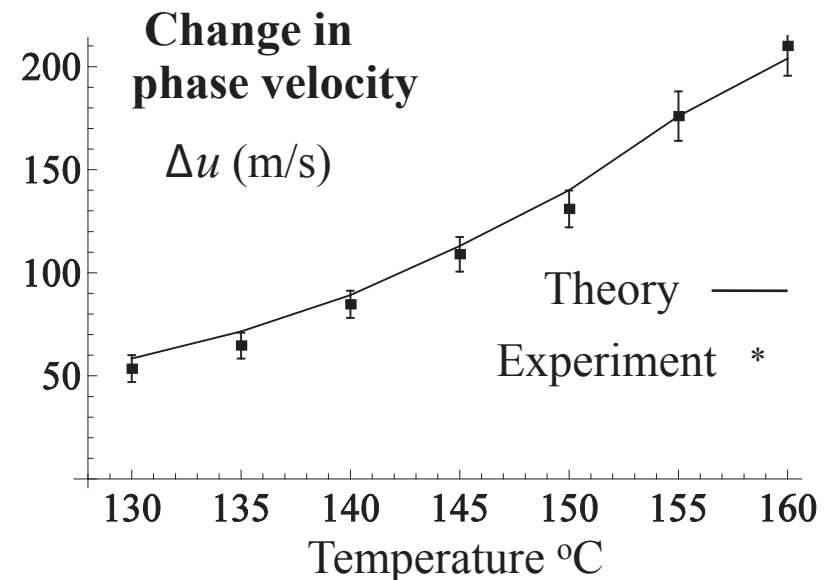
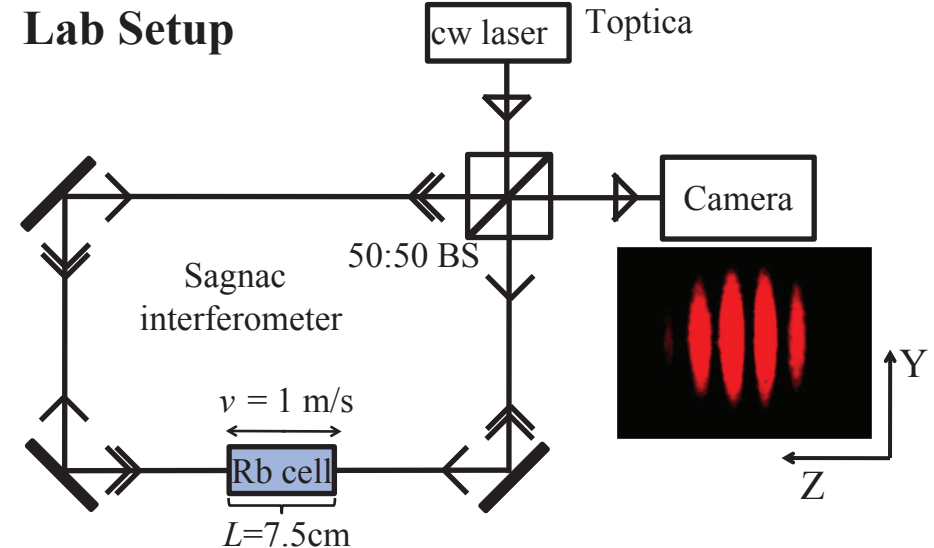
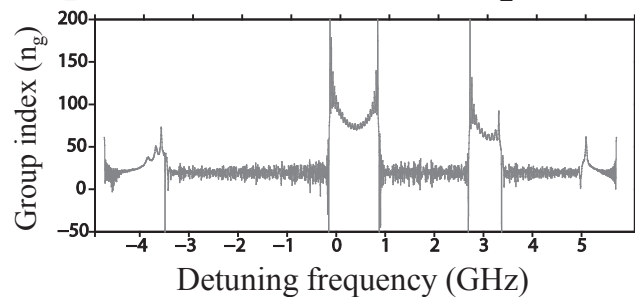
$$u \approx \frac{c}{n} \pm v \left(1 - \frac{1}{n^2} + \frac{n_g - n}{n^2} \right)$$

We Use Rubidium as Our Slow Light Medium

- Transmission spectrum of Rb around D₂ transition:



- Group index of Rb around D₂ line at T=130



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

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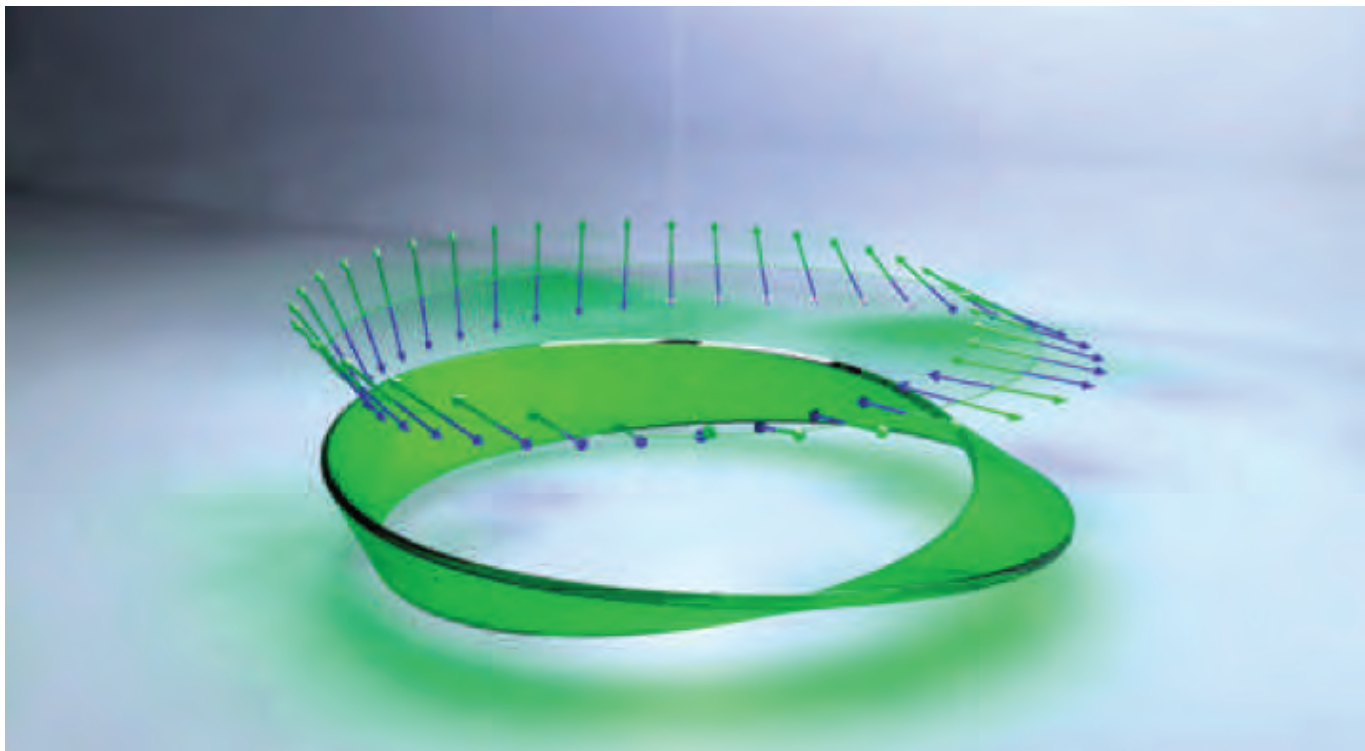
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Observation of Optical Polarization Möbius Strips

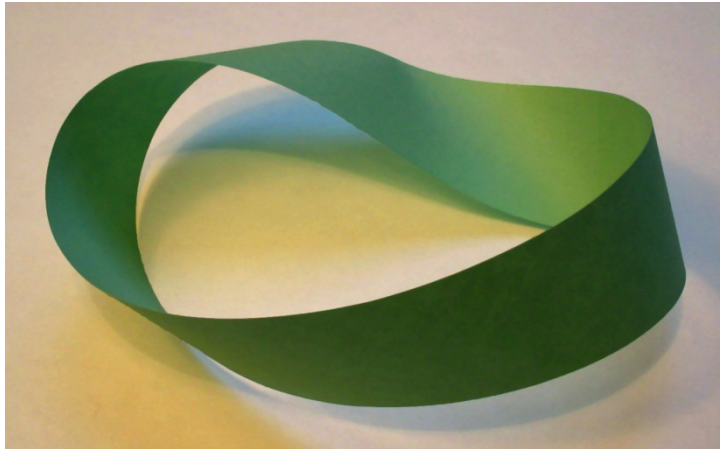
- Möbius strips are familiar geometrical structures, but their occurrence in nature is extremely rare.
- We generate such structures in the nanoscale in tightly focused vector light beams and confirm experimentally their Möbius topology.



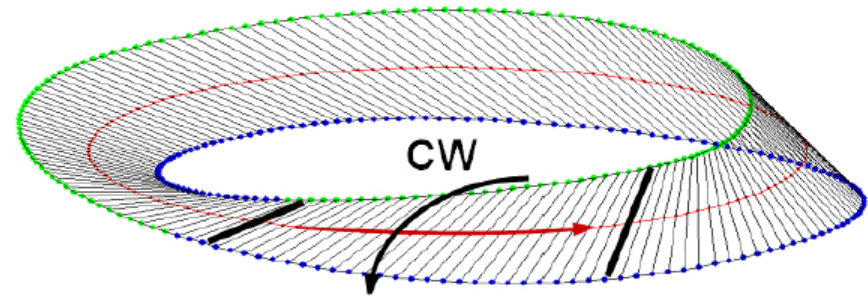
Bauer, Banzer, Karimi, Orlov, Rubano, Marrucci, Santamato, Boyd and Leuchs,
Science, 347, 964 (2015)

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)

¹ Wikipedia

² 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)

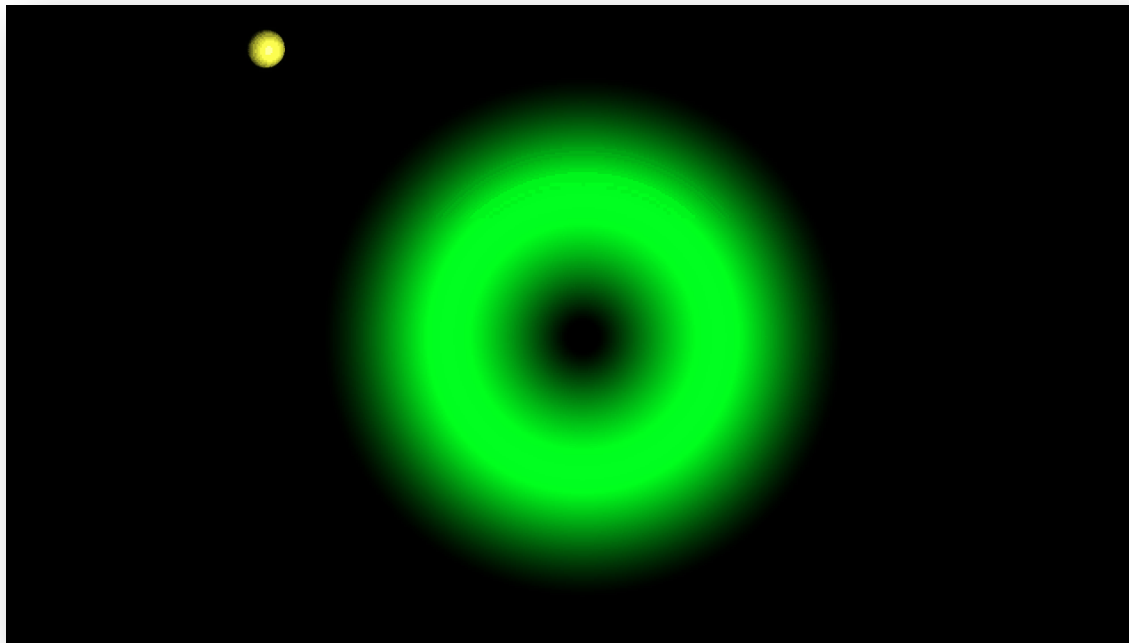
Isaac Freund, Opt. Lett. 35, 148-150 (2010)

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

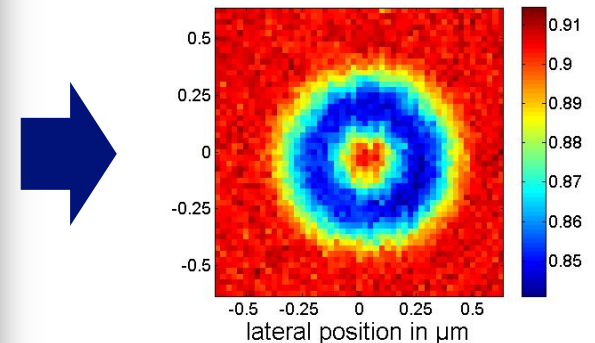
Full vectorial beam measurement on the nanoscale

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



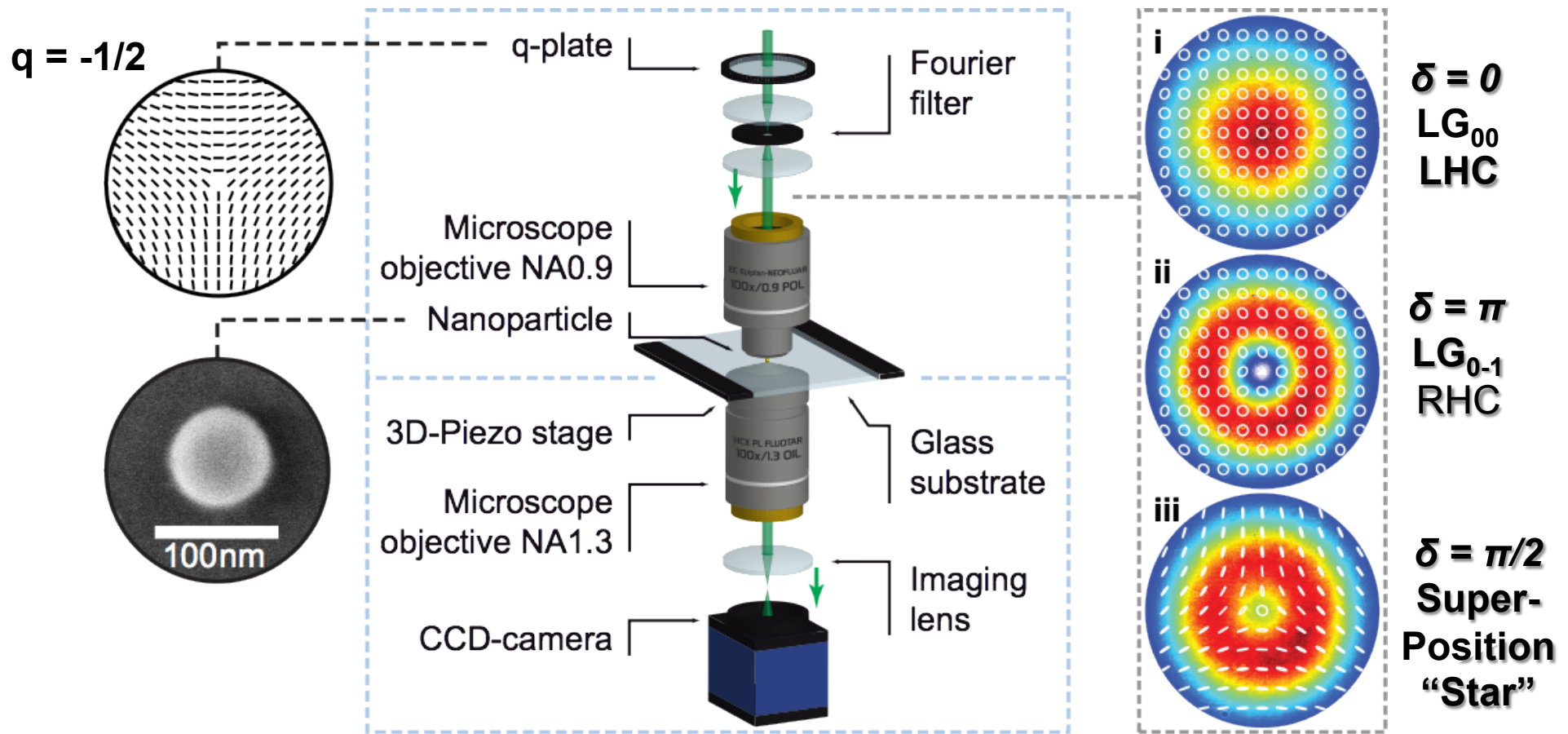
measured intensity
(can also measure
polarization and phase)



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).

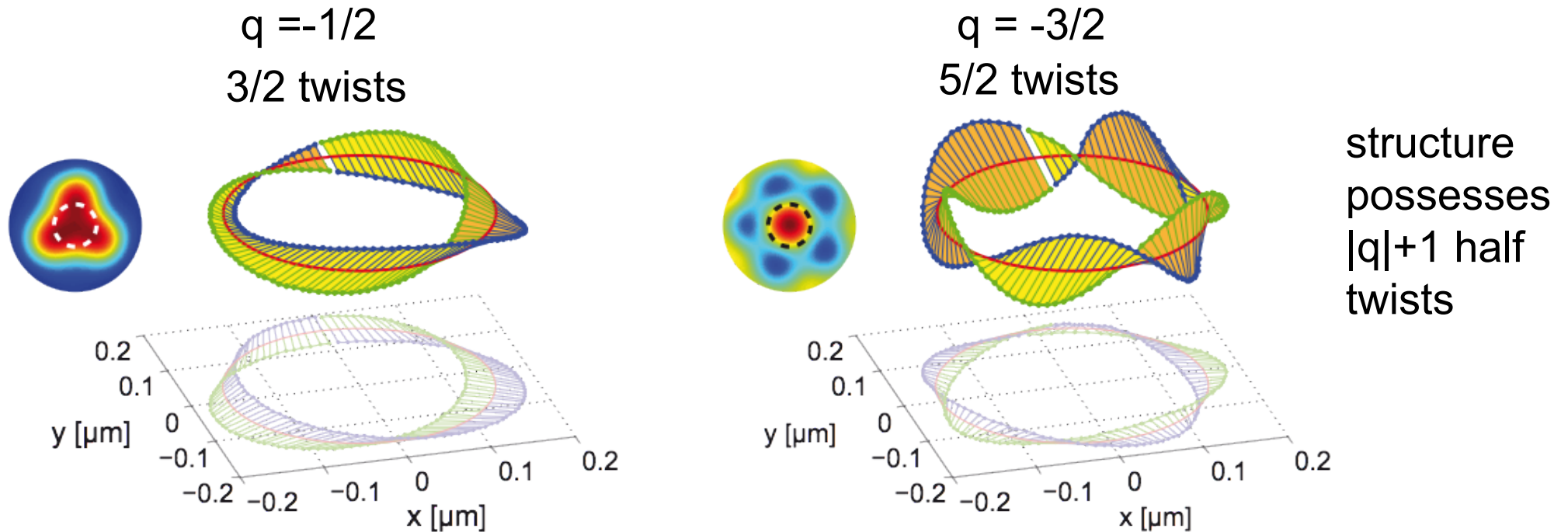
Lab Setup to Observe a Polarization Möbius Strip



- 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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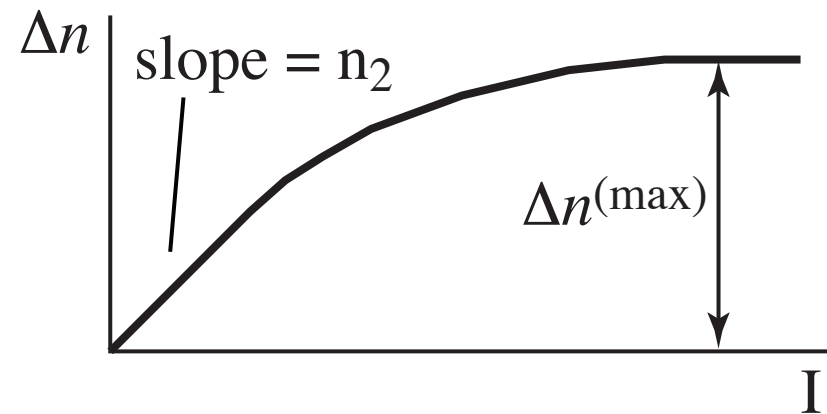
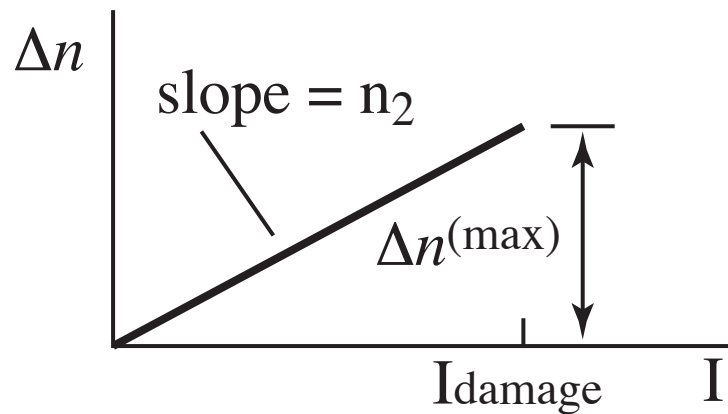
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What Makes a Good (Kerr-Effect) Nonlinear Optical Material?

Want n_2 large; and also want $\Delta n^{(\max)}$ large.

These are distinct concepts! Damage and saturation can limit $\Delta n^{(\max)}$



We report a material for which both n_2 and $\Delta n^{(\max)}$ are extremely large!

For ITO at ENZ wavelength, $n_2 = 8 \times 10^{-11} \text{ cm}^2/\text{W}$ and $\Delta n^{(\max)} = 0.8$

(For silica glass $n_2 = 3.2 \times 10^{-16} \text{ cm}^2/\text{W}$, $I_{\text{damage}} = 1 \text{ TW}/\text{cm}^2$, and thus $\Delta n^{(\max)} = 3 \times 10^{-4}$)

Nonlinear Optical Properties of Indium Tin Oxide (ITO)

ITO is a degenerate semiconductor (so highly doped as to be metal-like).

It has a very large density of free electrons, and a bulk plasma frequency corresponding to a wavelength of approximately $1.24 \mu\text{m}$.

Recall the Drude formula

$$\epsilon(\omega) = \epsilon_{\infty} - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

Note that $\text{Re } \epsilon = 0$ for $\omega = \omega_p / \sqrt{\epsilon_{\infty}} \equiv \omega_0$.

The region near ω_0 is known as the epsilon-near-zero (ENZ) region.

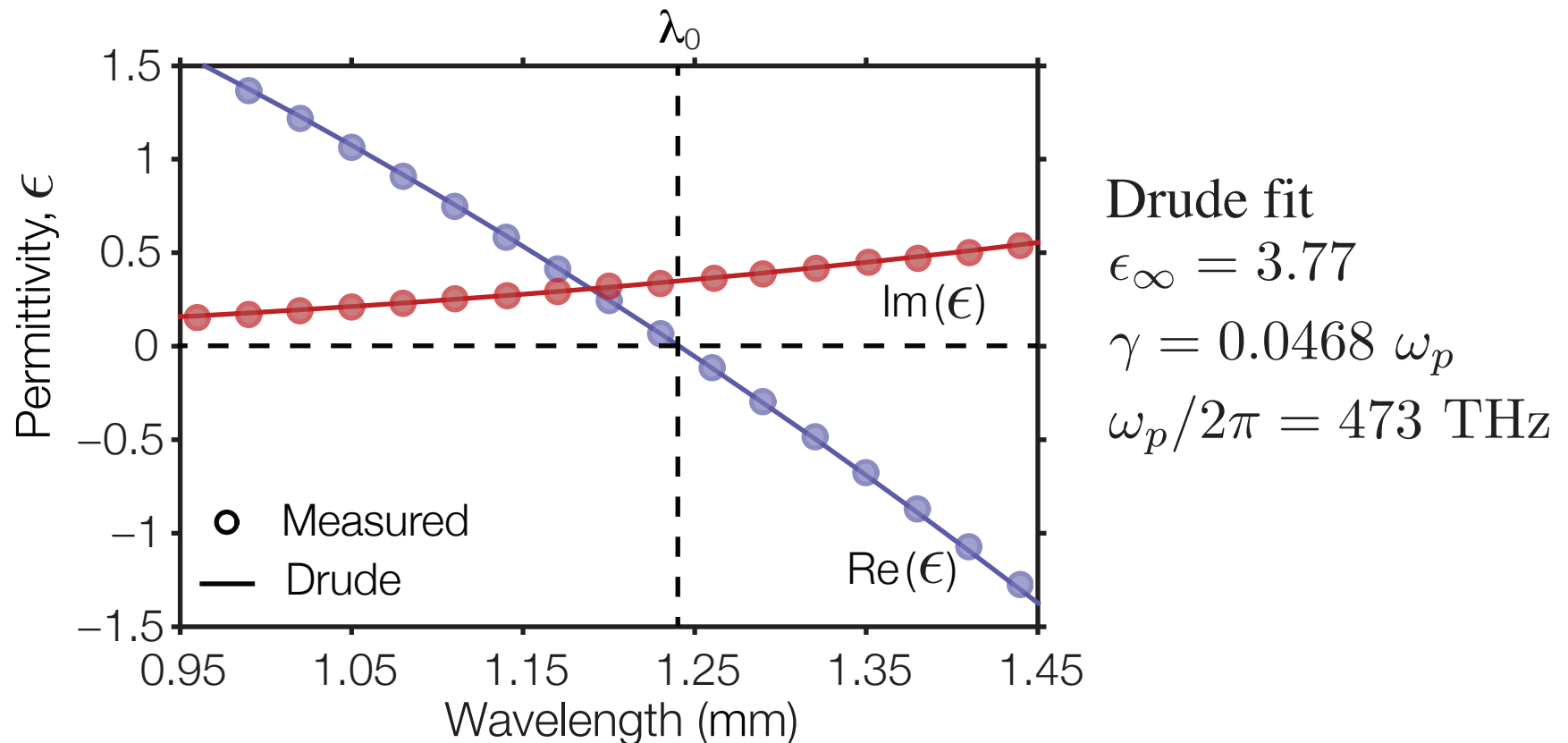
There has been great recent interest in studies of ENZ phenomena:

- H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, *Science* 342, 1223 (2013).
- C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, *Phys. Rev. B* 85, 045129 (2012).
- S. Campione, D. de Ceglia, M. A. Vincenti, M. Scalora, and F. Capolino, *Phys. Rev. B* 87, 035120 (2013).
- A. Ciattoni, C. Rizza, and E. Palange, *Phys. Rev. A* 81, 043839 (2010).

The Epsilon-Near-Zero (ENZ) region of Indium Tin Oxide (ITO)

Measured real and imaginary parts of the dielectric permittivity.

Commercial ITO sample, 310 nm thick on a glass substrate



Note that $\text{Re}(\epsilon)$ vanishes at 1.24 mm, but that the loss-part $\text{Im}(\epsilon)$ is non-zero.

Implications of ENZ Behavior for Nonlinear Optics

Here is the intuition for why the ENZ conditions are of interest in NLO

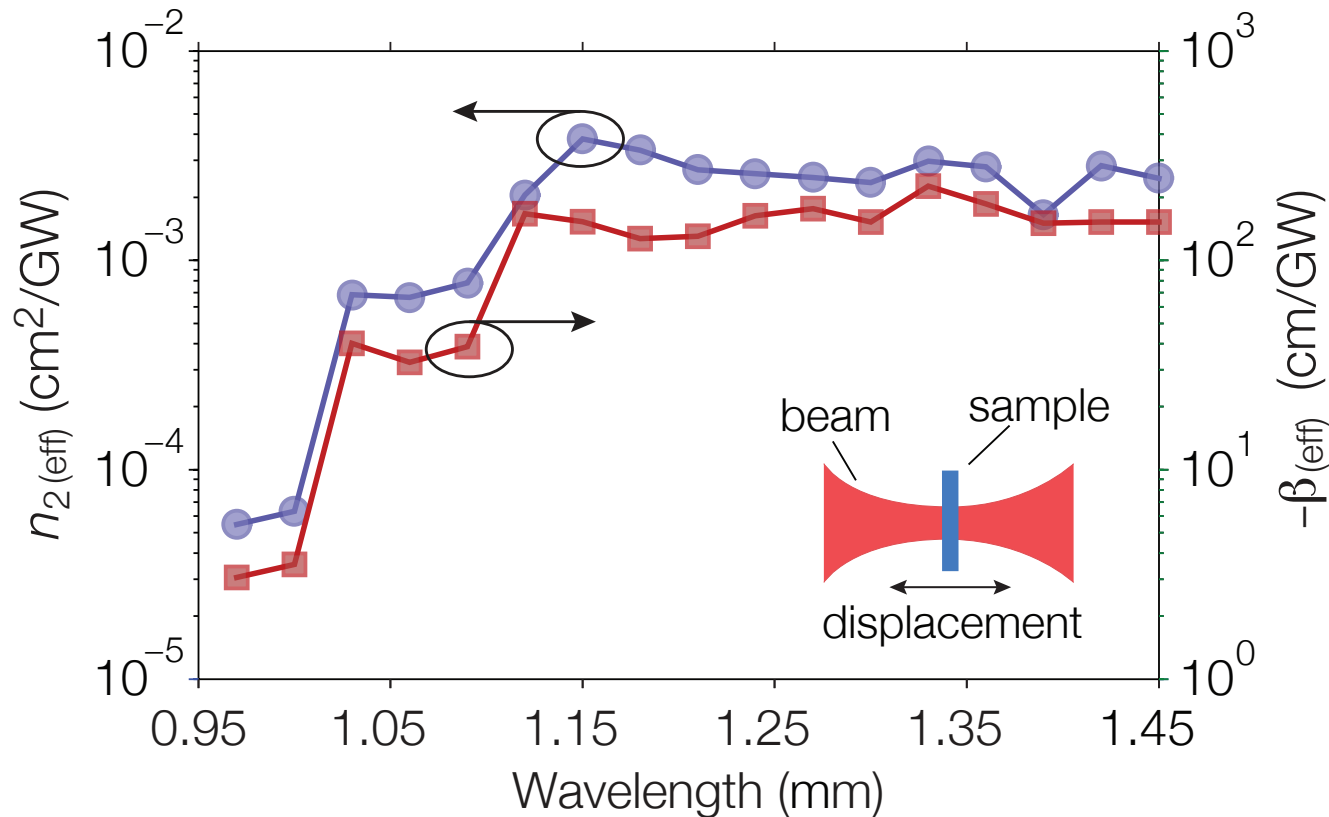
Recall the standard relation between n_2 and $\chi^{(3)}$

$$n_2 = \frac{3\chi^{(3)}}{4\epsilon_0 c n_0 \operatorname{Re}(n_0)}$$

Note that for ENZ conditions the denominator becomes very small, leading to a very large value of n_2

Z-Scan Measurements of the NLO Response

NLO response at *normal incidence*

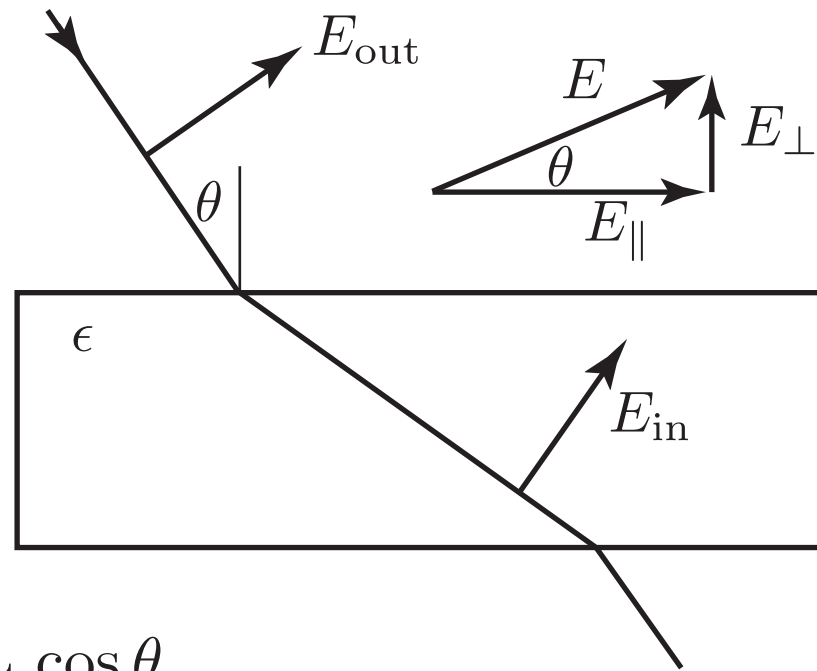


Maximum measured value of n_2 is $3 \times 10^{-3} \text{ cm}^2/\text{GW} = 3 \times 10^{-12} \text{ cm}^2/\text{W}$

Note that n_2 is positive (self focusing) and β is negative (saturable absorption).

The NLO Response Is Even Larger at Oblique Incidence

Standard boundary conditions show that:



$$E_{\text{in},\parallel} = E_{\text{out},\parallel} = E_{\text{out}} \cos \theta$$

$$D_{\text{in},\perp} = D_{\text{out},\perp} \Rightarrow E_{\text{in},\perp} = E_{\text{out},\perp} / \epsilon = E_{\text{out}} \cos \theta / \epsilon$$

Thus the total field inside of the medium is given by

$$E_{\text{in}} = E_{\text{out}} \sqrt{\cos^2 \theta + \frac{\sin^2 \theta}{\epsilon}}$$

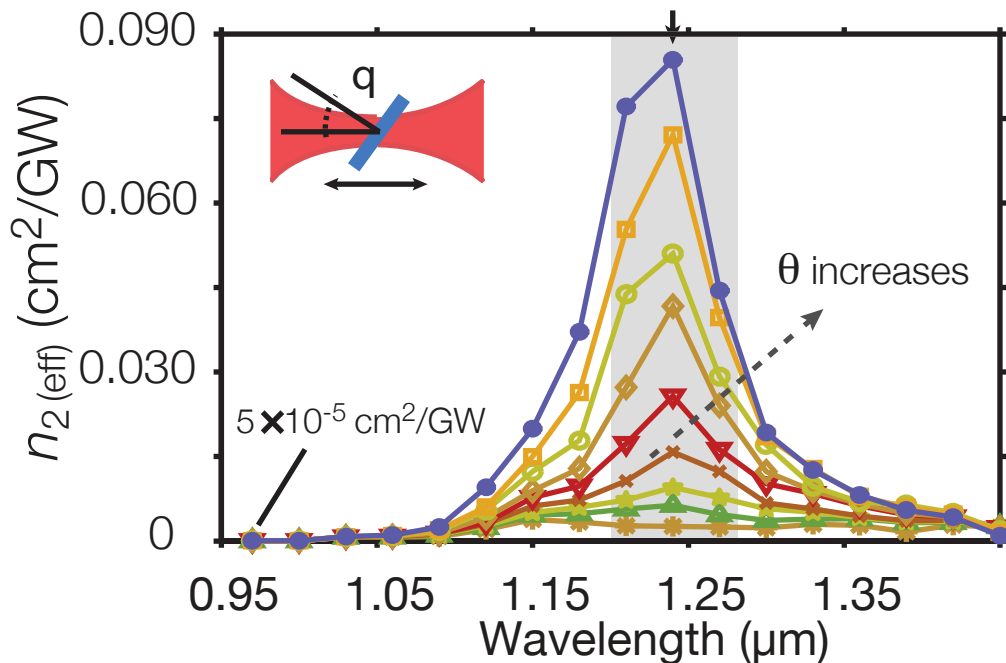
Note that, for $\epsilon < 1$, E_{in} exceeds E_{out} for $\theta \neq 0$.

Note also that, for $\epsilon < 1$, E_{in} increases as θ increases.

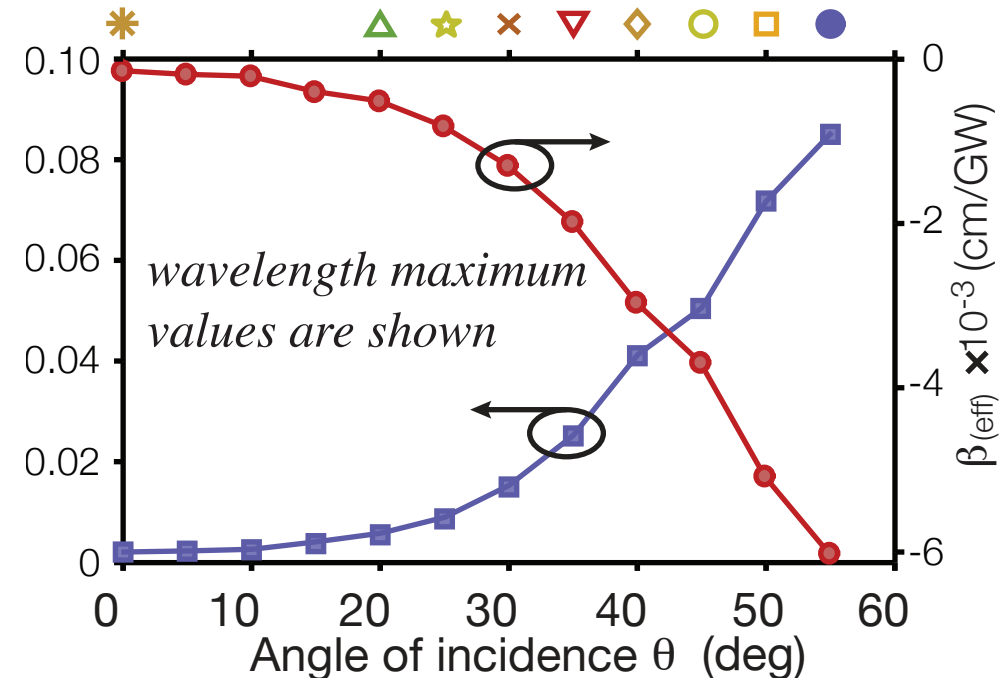
Nonlinear Optical Response at Oblique Incidence

Z-scan measurements for non-normal incidence

Wavelength dependence of n_2



Variation with incidence angle



- Both n_2 and nonlinear absorption increase with angle of incidence
- n_2 achieves a maximum value of $0.08 \text{ cm}^2/\text{GW} = 8 \times 10^{-11} \text{ cm}^2/\text{W}$ at $1.25 \mu\text{m}$ and 60 deg .

Why is n_2 so large?

The short-wavelength value of n_2 of ITO is $5 \times 10^{-5} \text{ cm}^2/\text{GW}$, which is 150 times larger than that of fused silica ($3.2 \times 10^{-7} \text{ cm}^2/\text{GW}$).

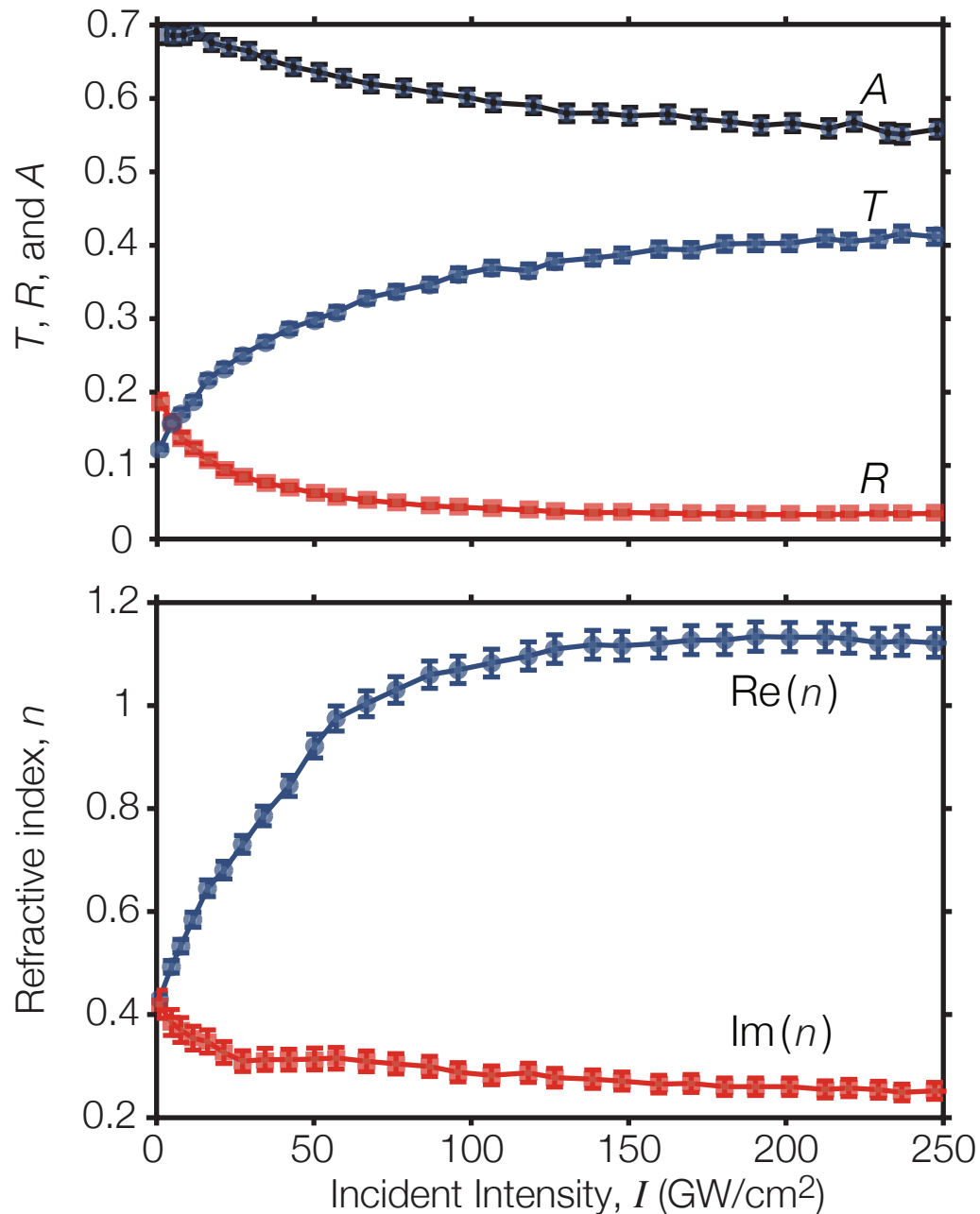
There is a 50x enhancement from working at the ENZ wavelength and an additional 30x enhancement from using non-normal incidence.

Thus $n_2 = 8 \times 10^{-2} \text{ cm}^2/\text{GW}$, which is 2.5×10^5 times that of fused silica.

Incidentally, for arsenic trisulfide glass, $n_2 = 2.4 \times 10^{-4} \text{ cm}^2/\text{GW}$, which is 800 times larger than that of fused silica.

R.E. Slusher et al., J. Opt. Soc. Am. B 21, 1146 (2004).

Beyond the $\chi^{(3)}$ limit



The nonlinear change in refractive index is so large as to change the transmission, absorption, and reflection!

Note that transmission is increased at high intensity.

Here is the refractive index extracted from the above data.

Note that the total nonlinear change in refractive index is $\Delta n = 0.8$.

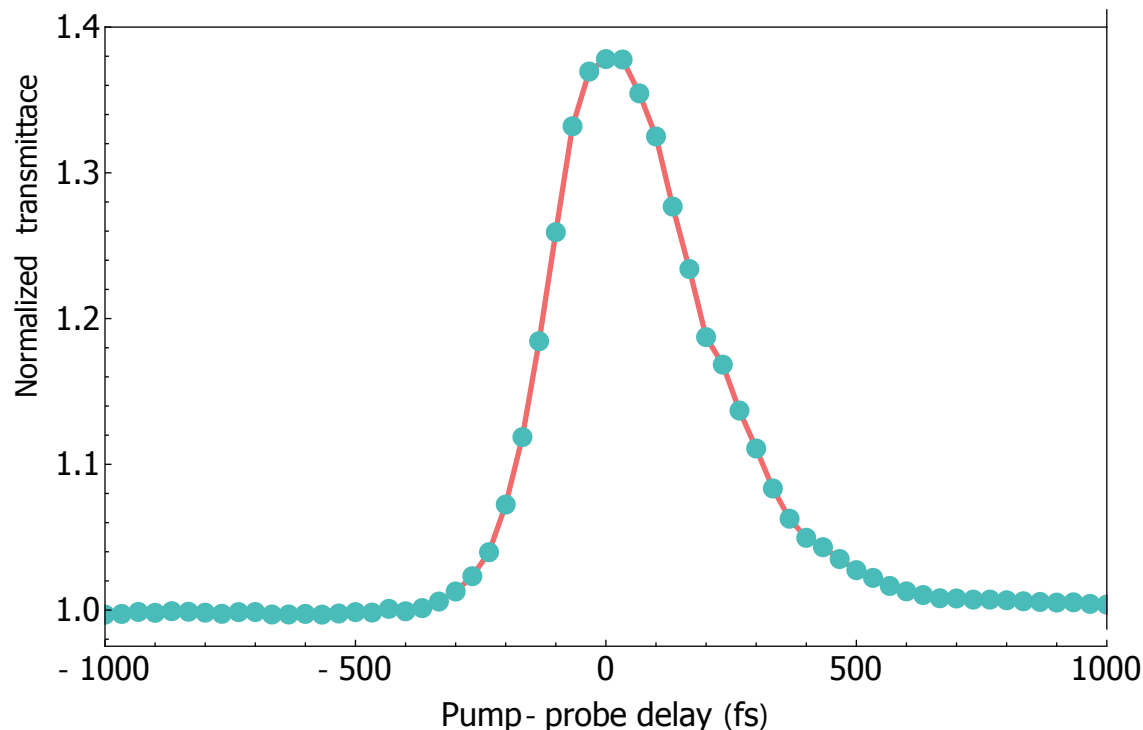
The absorption decreases at high intensity, allowing a predicted NL phase shift of 0.5 radians.

Measurement of Response Time of ITO

We have performed a pump-probe measurement of the response time.
Both pump and probe are 100 fs pulses at 1.2 μm .

Data suggests a response time of 270 fs.

ITO will support THz switching speeds



Implications of the Large NLO Response of ITO

Indium Tin Oxide at its ENZ wavelength displays enormously strong NLO properties:

n_2 is 2.5×10^5 times that of fused silica

nonlinear change in refractive index as large as 0.8

Note that the usual “power-series” description of NLO is not adequate for describing this material. (We can have fun reformulating the laws of NLO!)



Some possible new effects




Waveguiding outside the “weakly-guiding” regime

Efficient all-optical switching

No need for phase-matching

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Quantum Nonlinear Optics:

Nonlinear Optics Meets the Quantum World

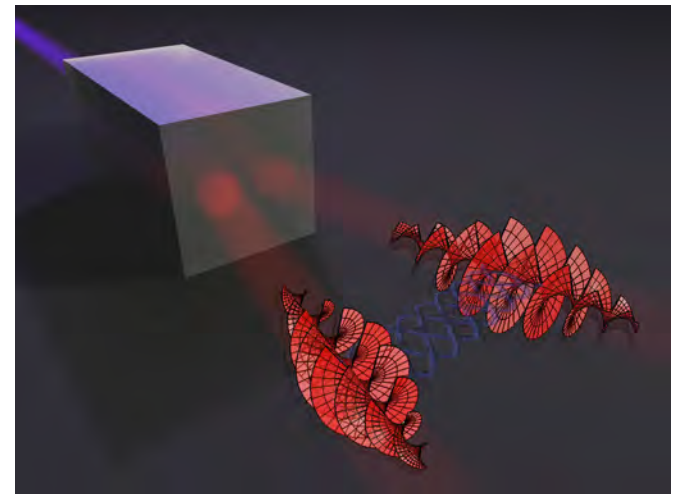
Outlook: NLO is a superb platform from which to explore new physical processes and to develop photonics applications.

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

Use of Quantum States for Secure Optical Communication

- 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

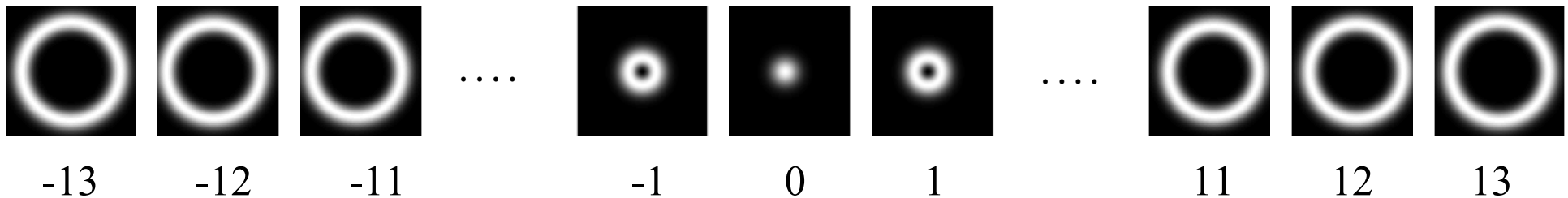
We are constructing a QKD system in which each photon carries many bits of information

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

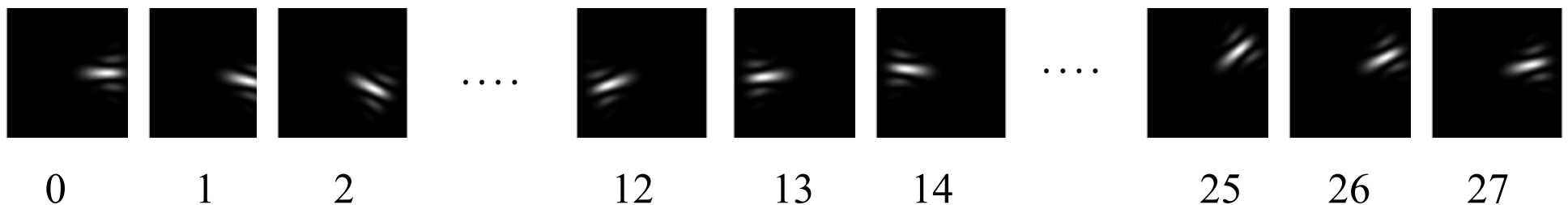
We also need a second basis composed of linear combinations of these states

Single Photon States

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

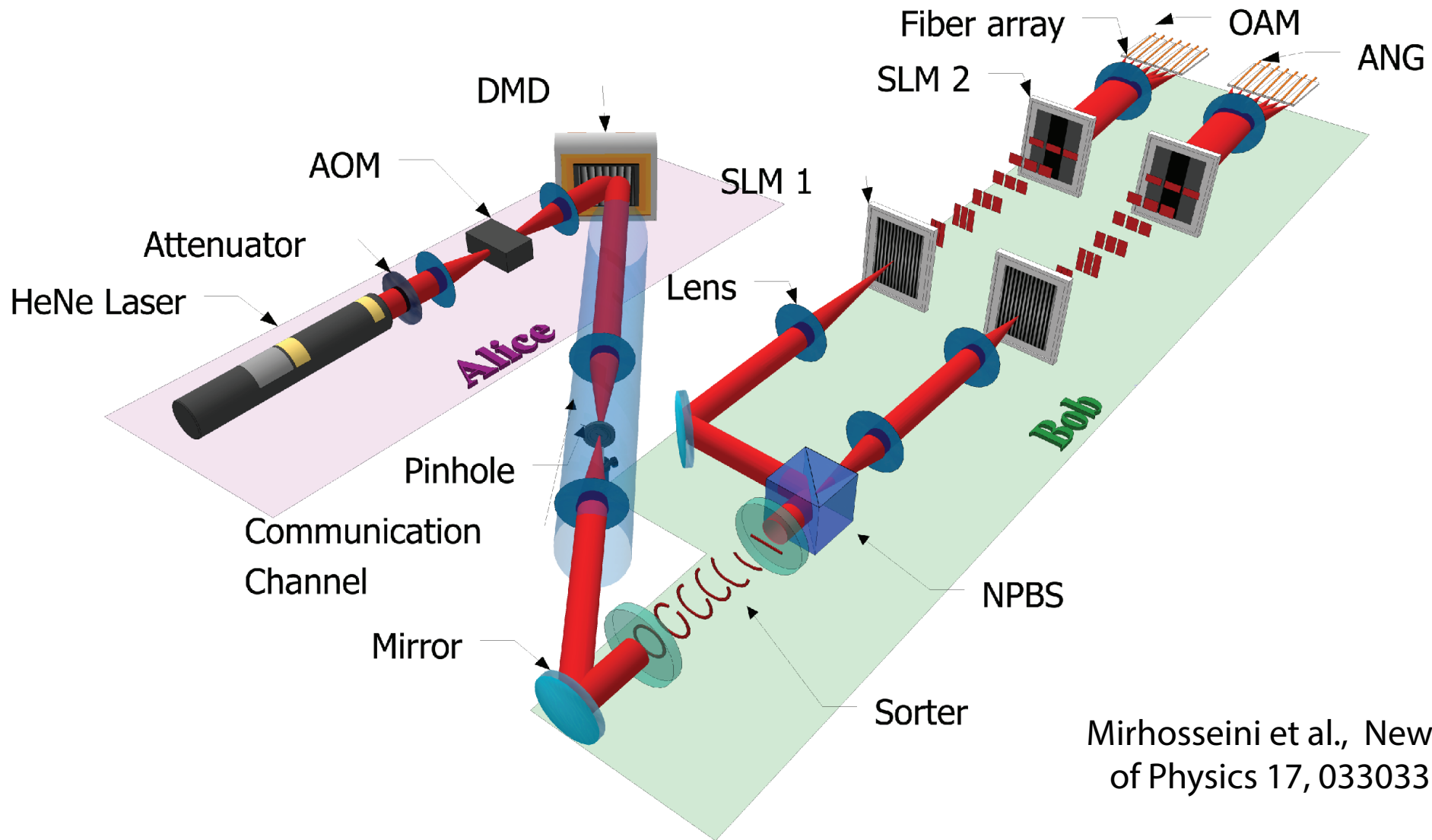


“Angular” Basis (mutually unbiased with respect to LG)



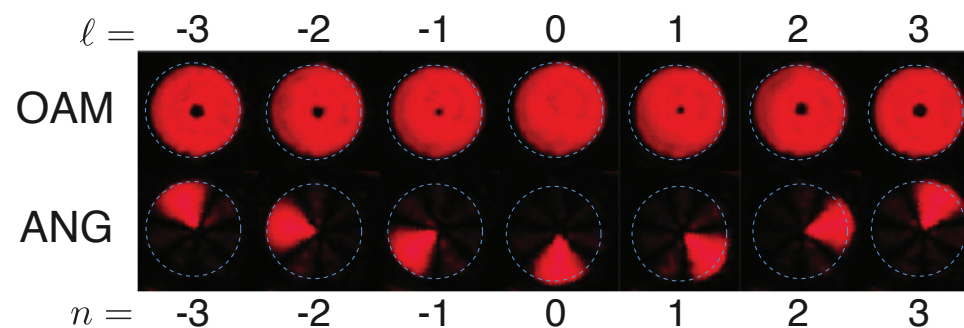
$$\Psi_{AB}^N = \frac{1}{\sqrt{27}} \sum_{l=-13}^{13} \text{LG}_{l,0} \exp(i2\pi Nl/27)$$

Our Laboratory Setup

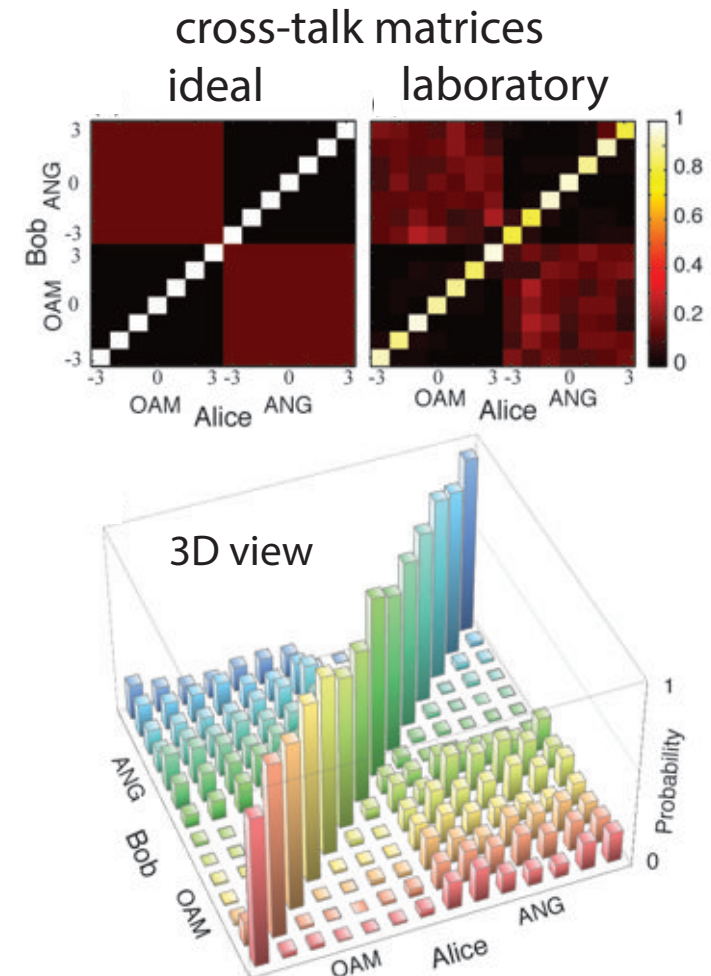
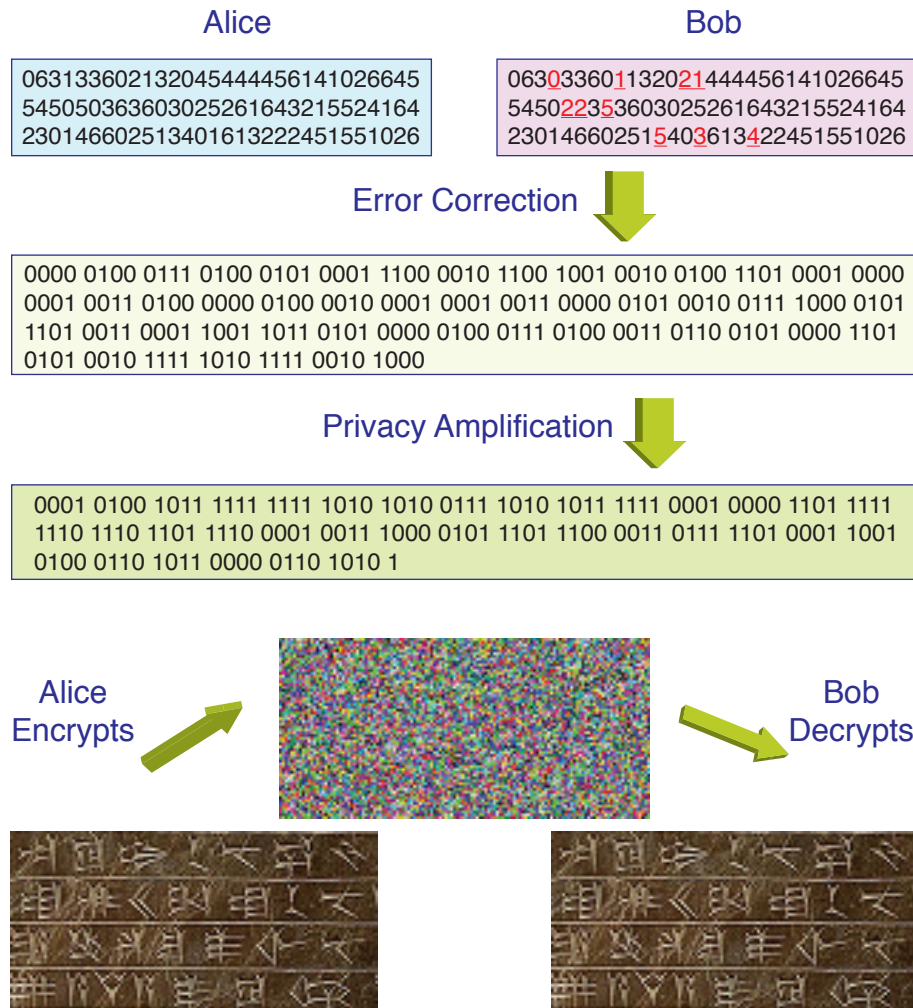


Mirhosseini et al., New Journal of Physics 17, 033033 (2015).

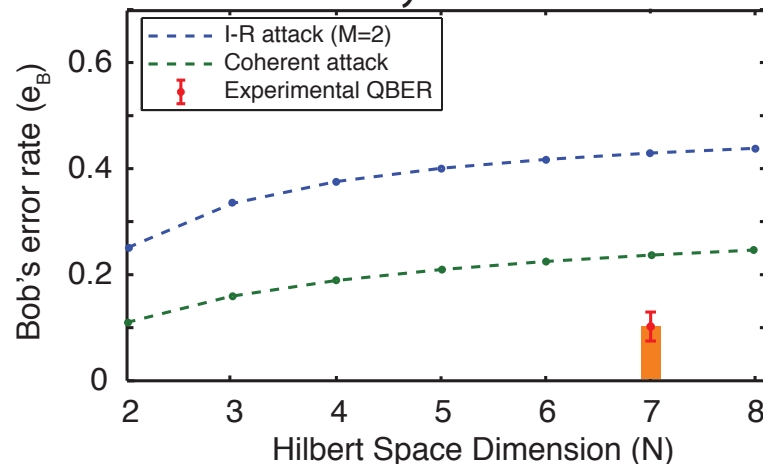
We use a seven-dimensional state space.



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,

Quantum Nonlinear Optics: Nonlinear Optics Meets the Quantum World

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



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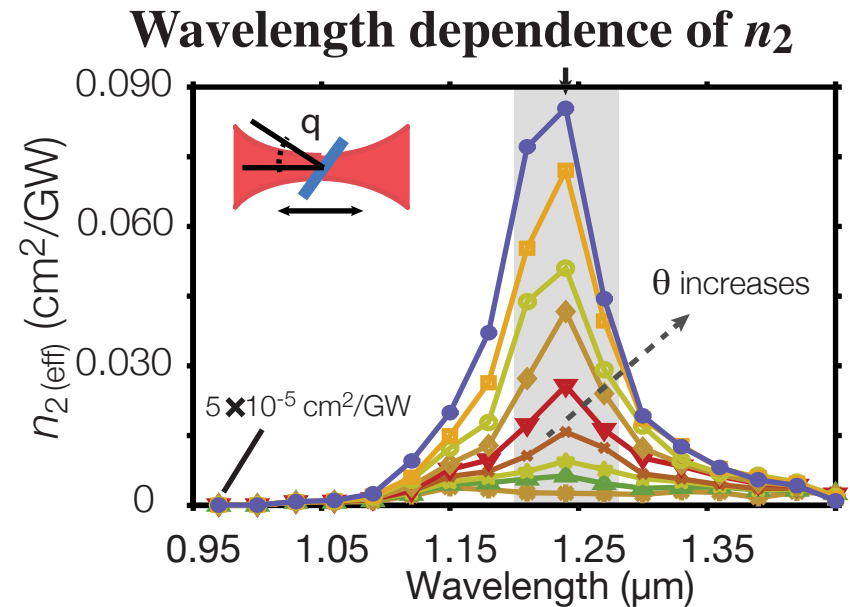
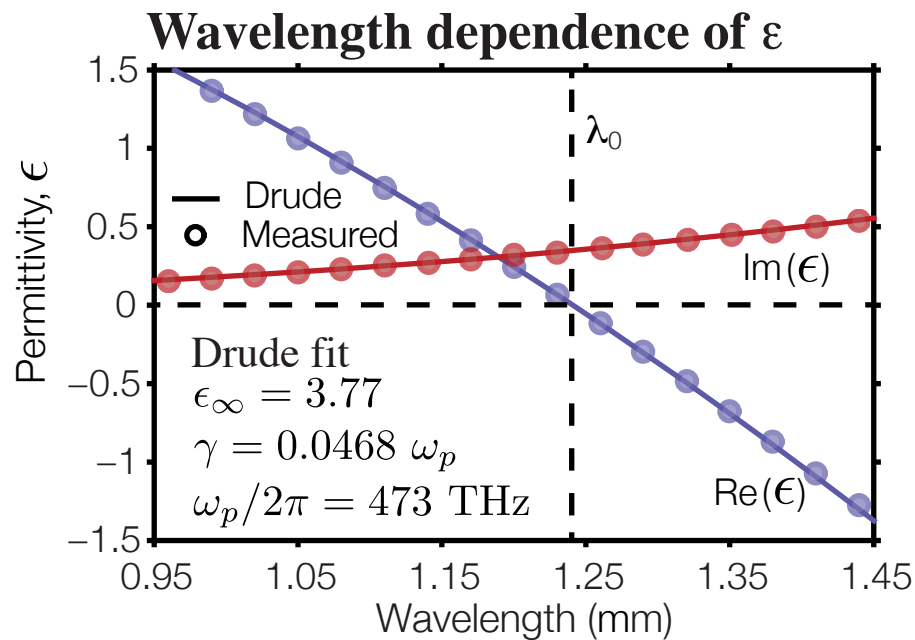
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- Efficient all-optical switching
- No need for phase-matching