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

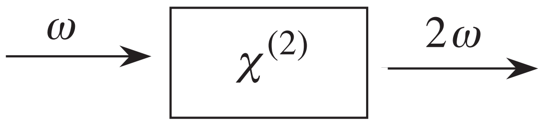
Simple Formulation of the Theory of Nonlinear Optics

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

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

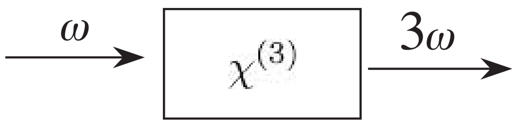
$\chi^{(1)}$ describes linear optics, e.g., how lenses work: 

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

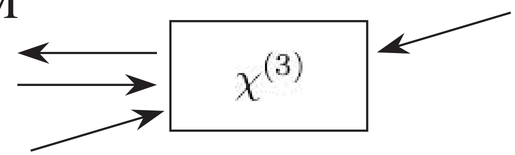


$\chi^{(3)}$ describes third-order effects such as third-harmonic generation, four-wave mixing, and the intensity dependence of the index of refraction.

THG



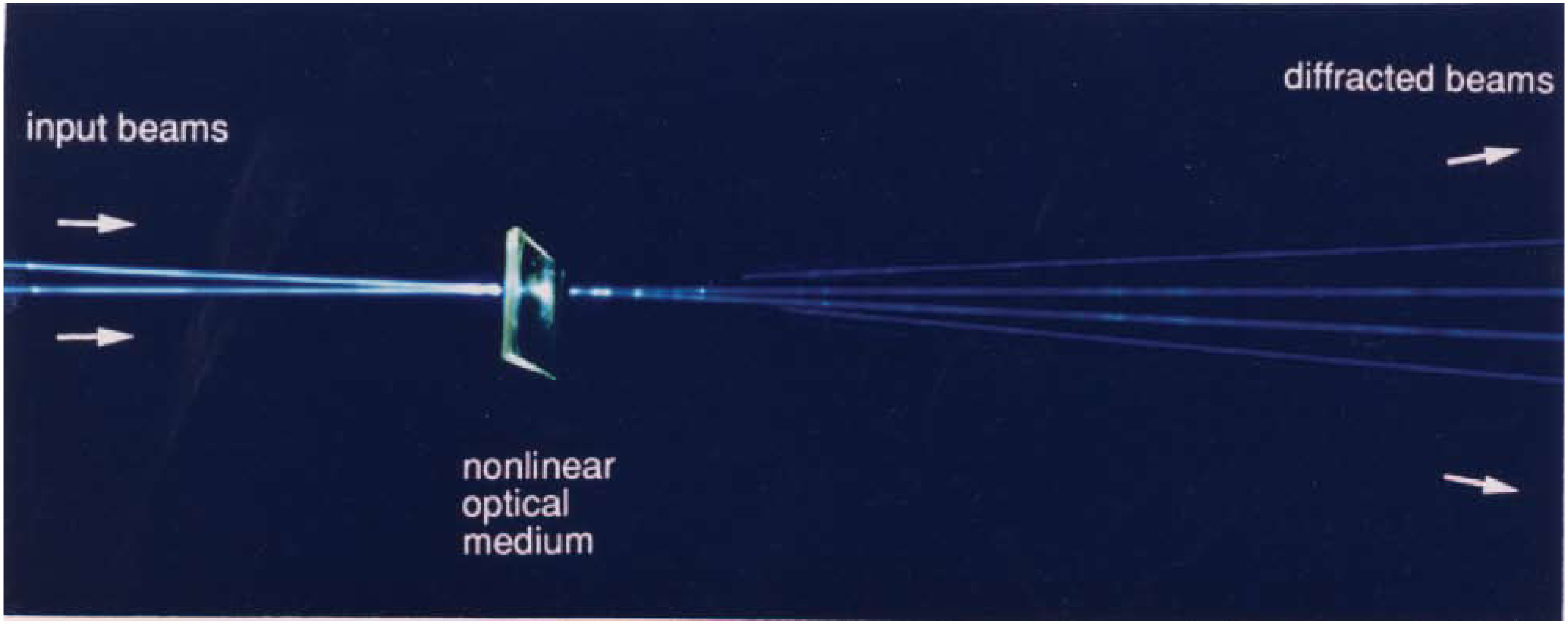
FWM



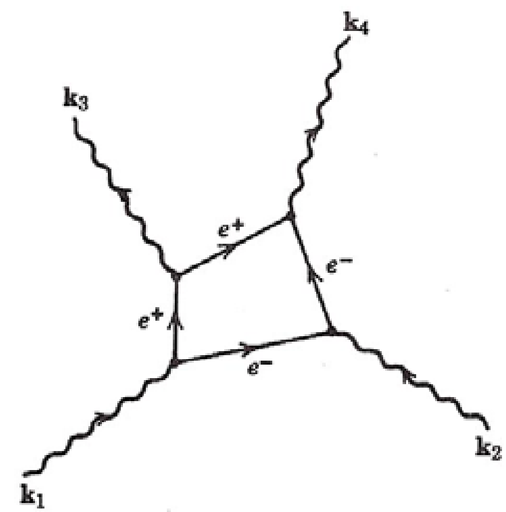
NL index

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

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(FBAG) $\approx 10^{14}$ n_2(silica)   [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.

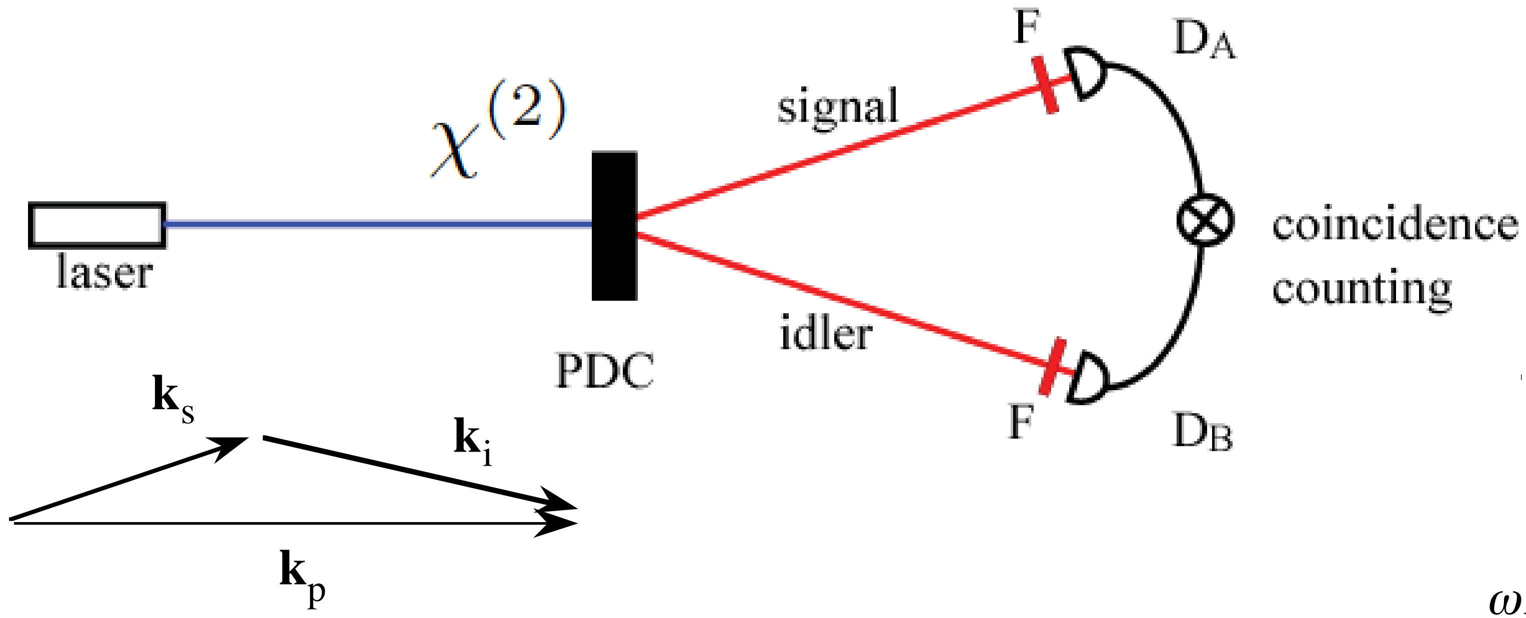
Why Interest in Quantum Nonlinear Optics?

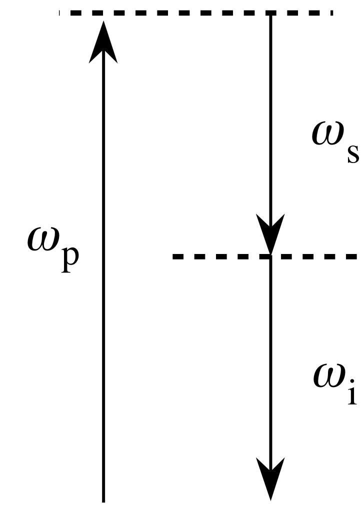
Explore the relation between traditional nonlinear optics (NLO) and phenomena in quantum information science (QIS).

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

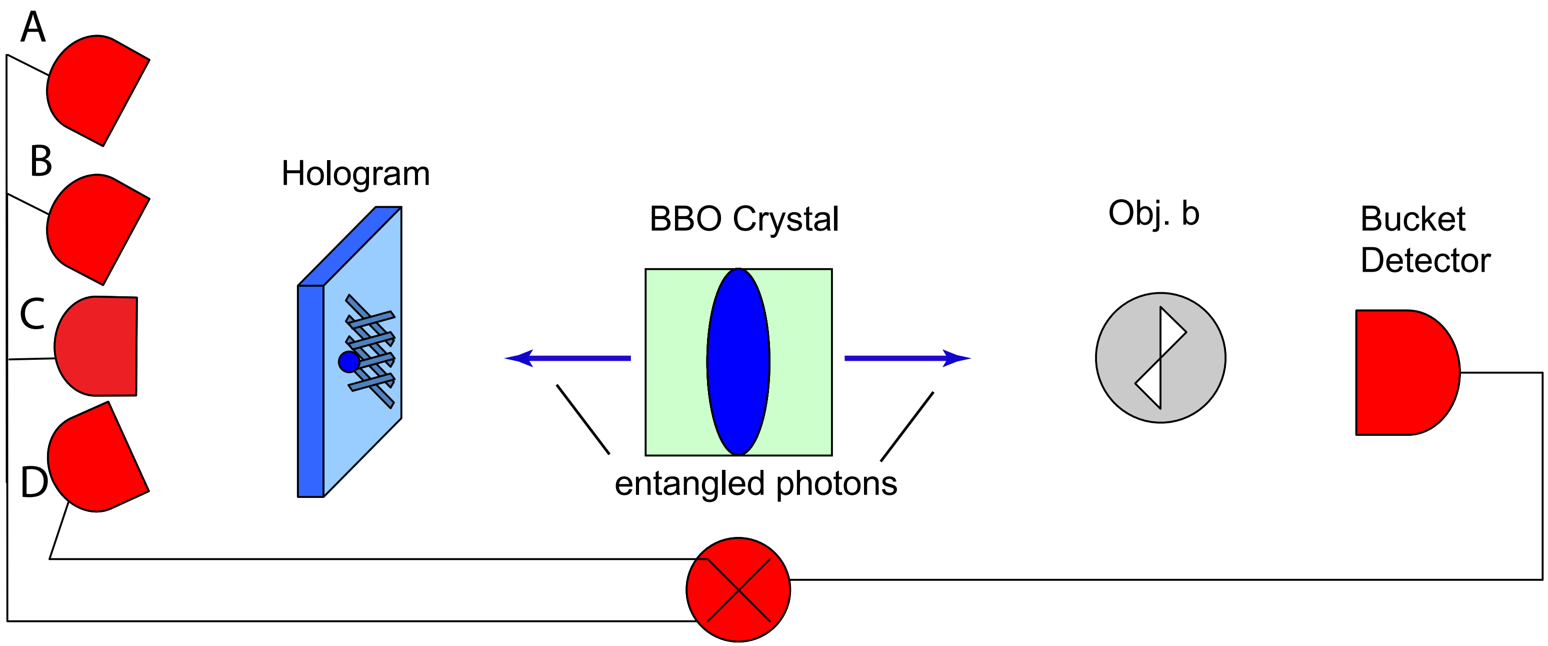
(d) angular position and orbital angular momentum

Entanglement is important for:

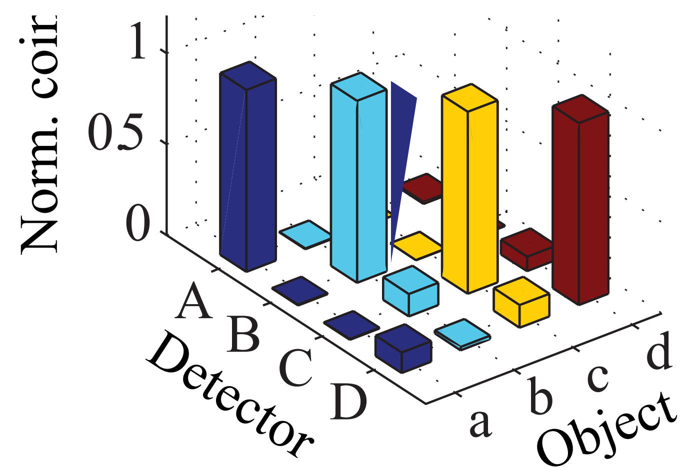
(a) Fundamental tests of QM (e.g., nonlocality, Bell tests)

(a) Quantum technologies (e.g., secure communications, Q teleportation)

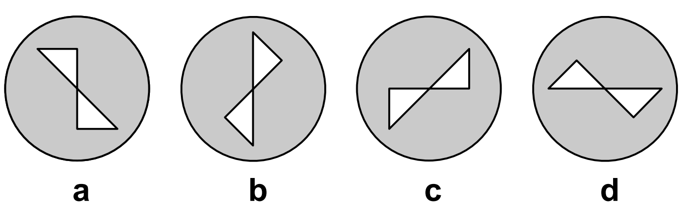
Single-Photon Coincidence Imaging



coincidence count rate



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



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

Malik, Shin, O'Sullivan. Zerom, and Boyd, Phys. Rev. Lett. 104, 163602 (2010).

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Controlling the Velocity of Light

“Slow,” “Fast” and “Backwards” Light

– Light can be made to go:

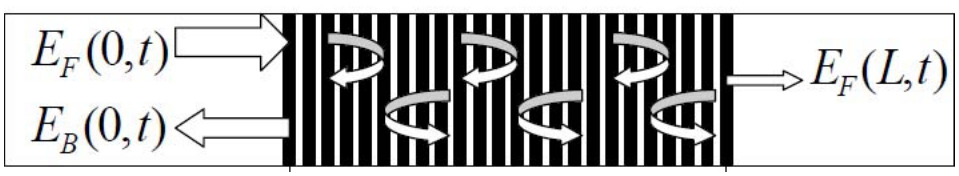
slow: $v_g << c$ (as much as 10^6 times slower!)

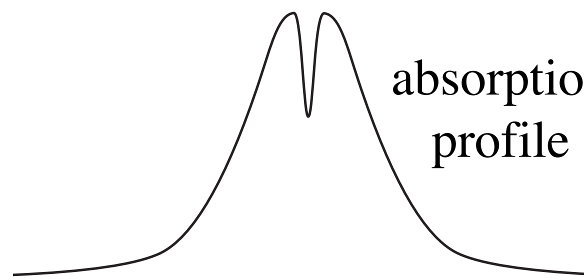
fast: $v_g > c$

backwards: v_g negative

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

– Velocity controlled by structural or material resonances

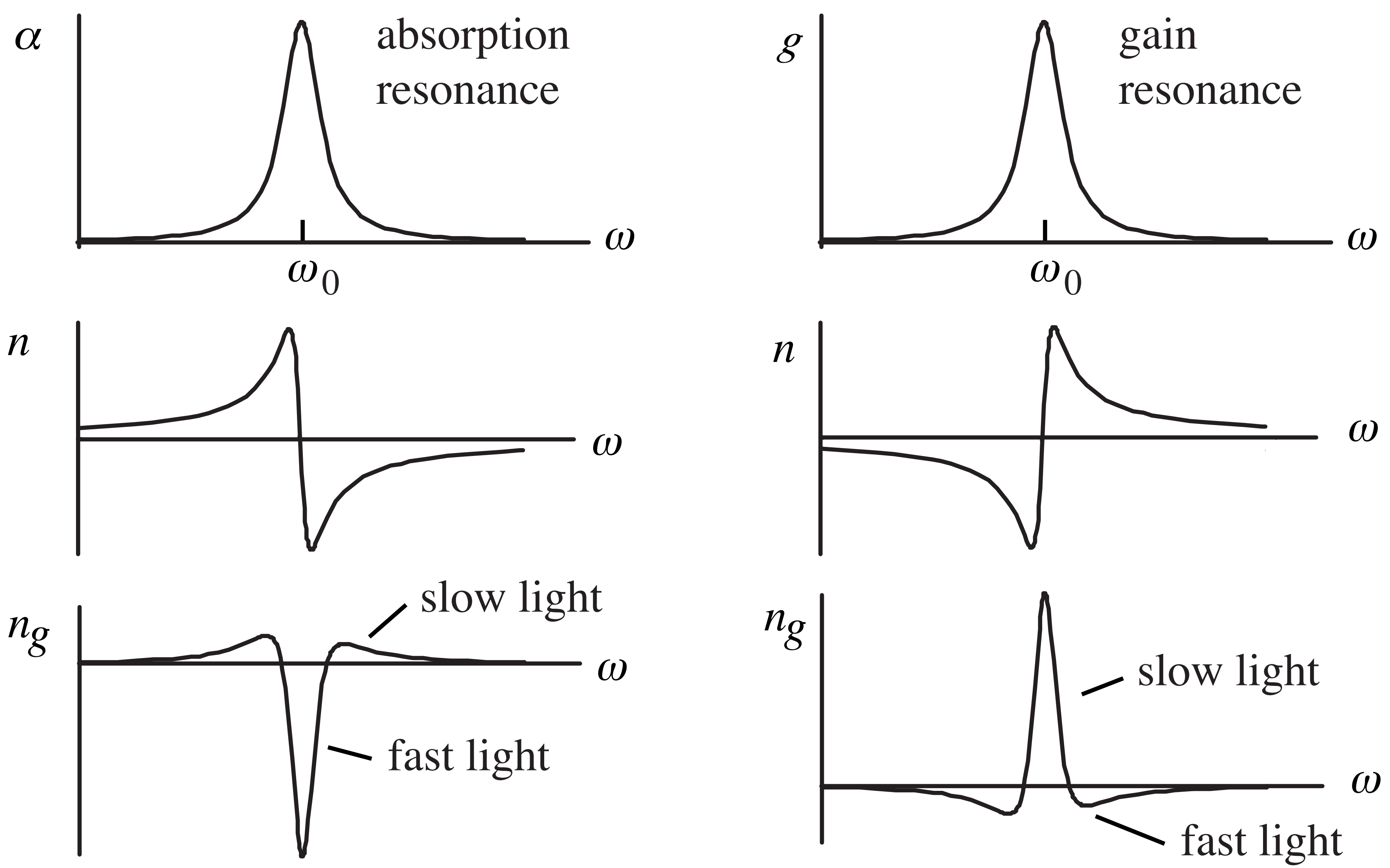




absorption
profile

Review article: Boyd and Gauthier, Science 326, 1074 (2009).

Slow and Fast Light Using Isolated Gain or Absorption Resonances

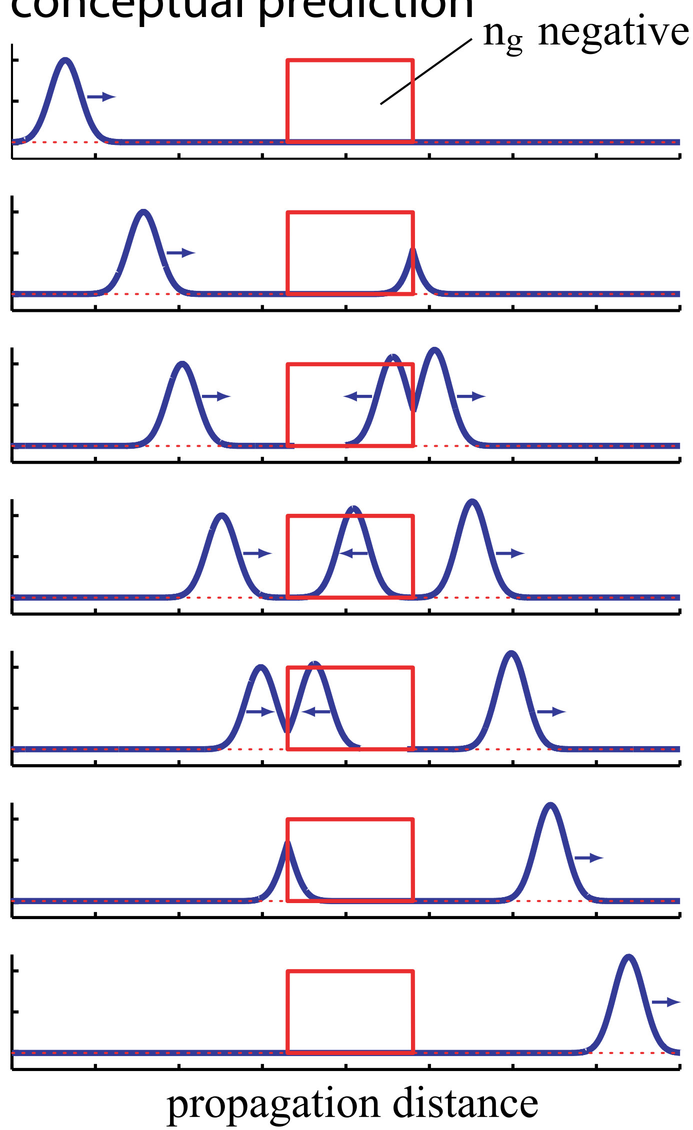


$$n_g = n + \omega\, (dn/d\omega)$$

Observation of Superluminal and "Backwards" Pulse Propagation

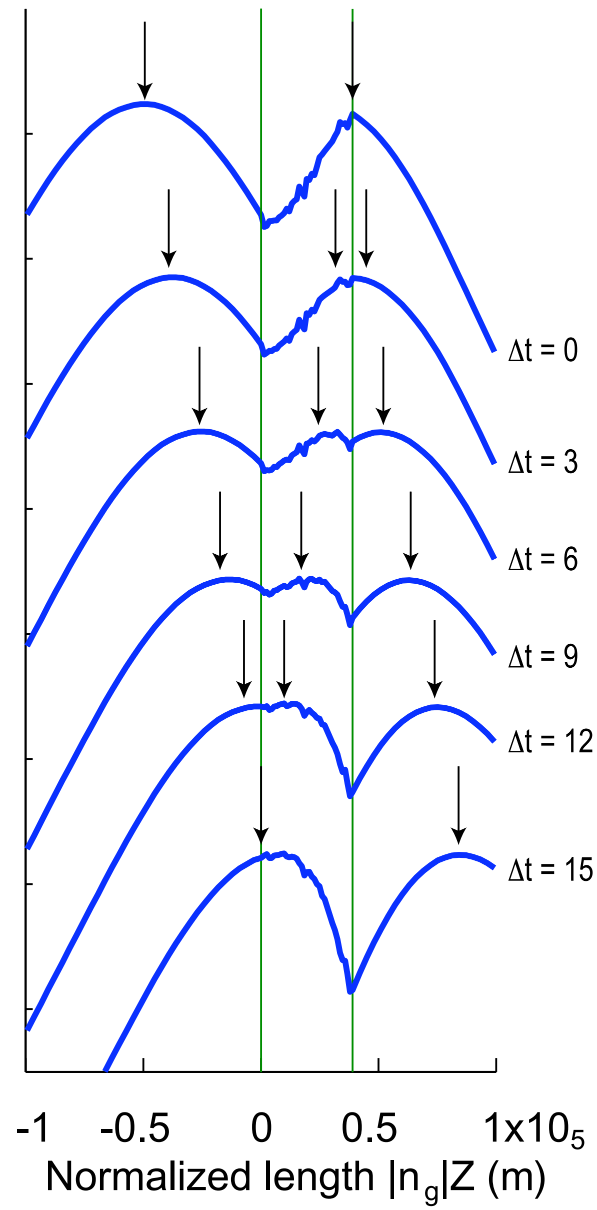
- A strongly counterintuitive phenomenon
- But entirely consistent with established physics
- Predicted by Garrett and McCumber (1970) and Chiao (1993).
- Observed by Gehring, Schweinsberg, Barsi, Kostinski, and Boyd Science 312, 985 2006.

\- conceptual prediction



$$\frac{\partial A}{\partial z} - \frac{1}{v_g}\frac{\partial A}{\partial t} = 0$$

\- laboratory results

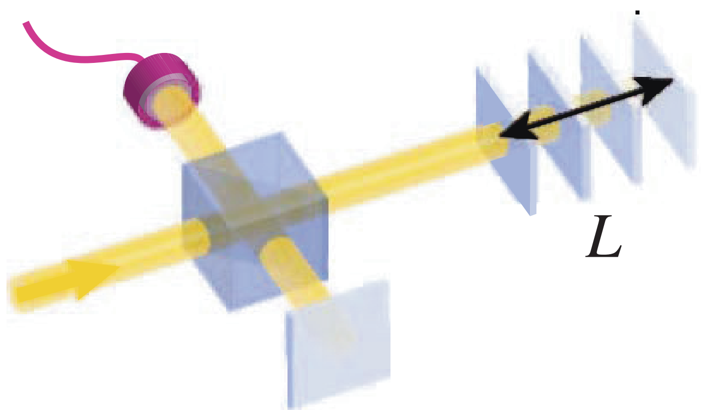


Development of Miniaturized, Chip-Scale Spectrometers

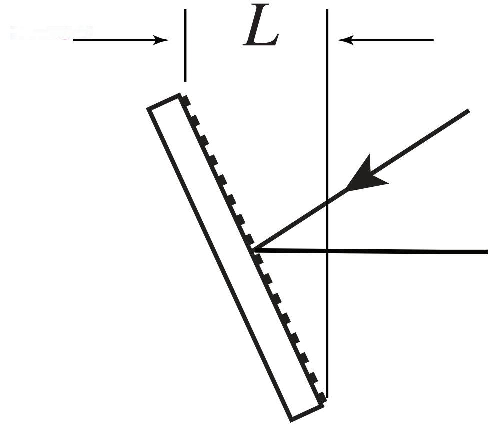
Can We Beat the $1/L$ Resolution Limit of Standard Spectrometers?

- 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



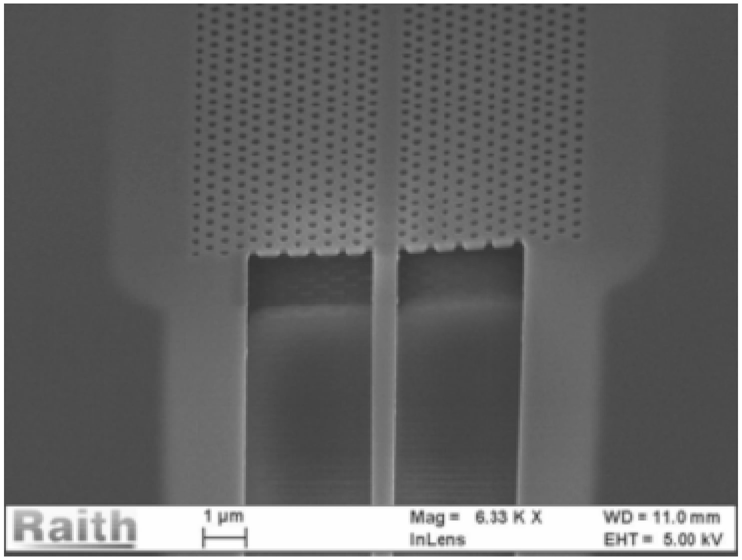
$$\Delta\nu(\text{res}) \approx 1/L$$

- 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:



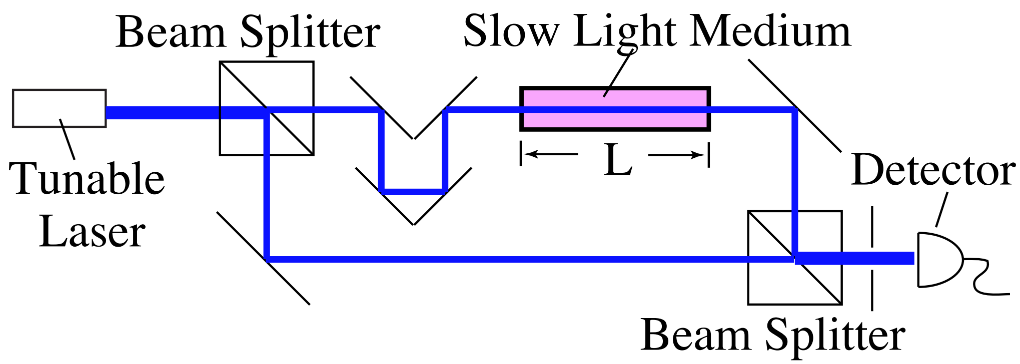
Our Approach: Chip-Scale Slow-Light Spectrometer

- 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 resoluation as large laboratory spectromters

- Here is why it works:

Slow-light interferometer:

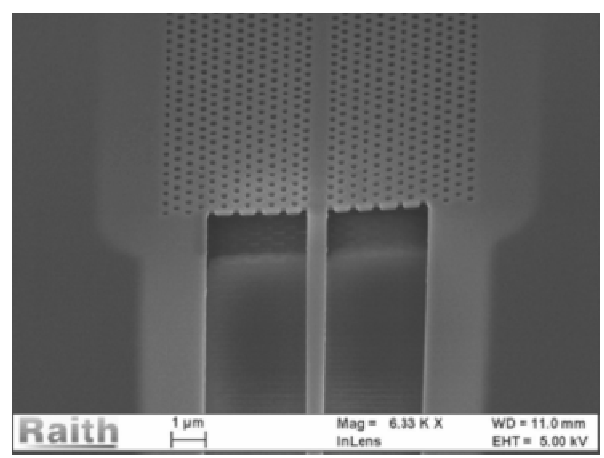


Simple analysis

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

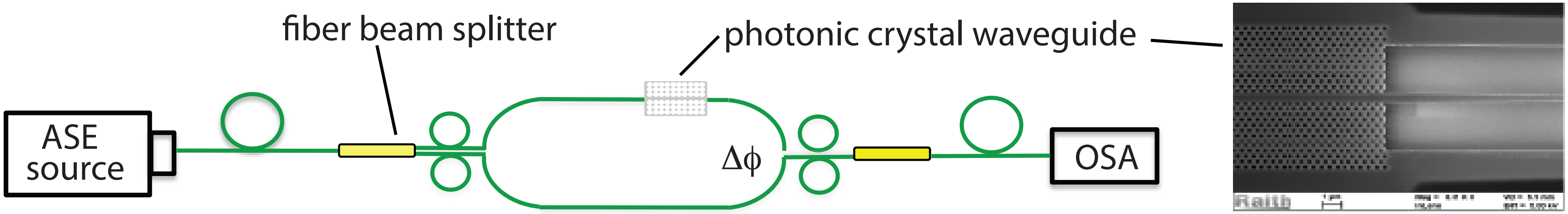
- We use line-defect waveguides in photonic crystals as our slow light mechanism

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

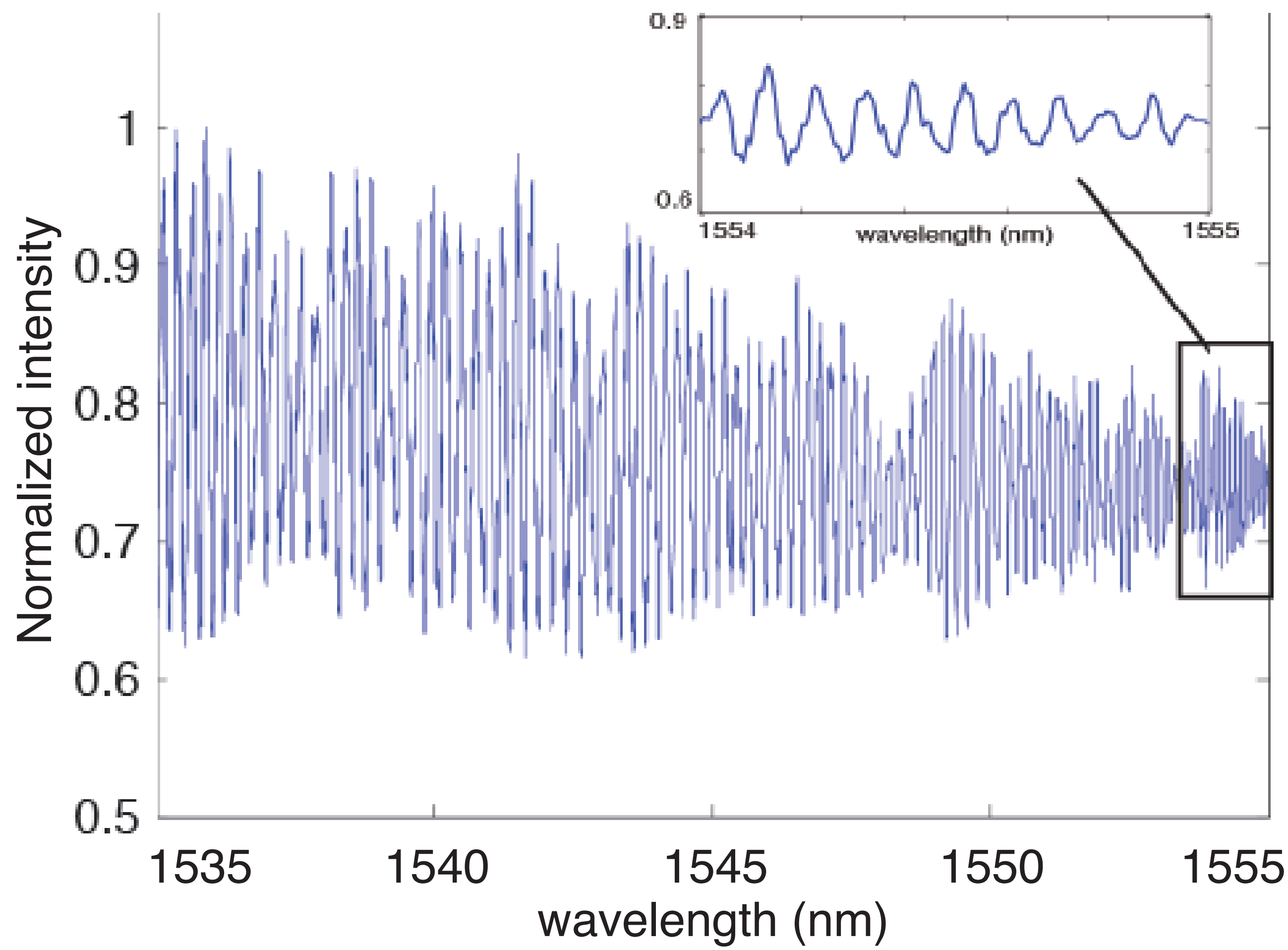


Shi, Boyd, Gauthier, and Dudley, Opt. Lett. 32, 915 (2007)
Shi, Boyd, Camacho, Vudyasetu, and Howell, PRL. 99, 240801 (2007)
Shi and Boyd, J. Opt. Soc. Am. B 25, C136 (2008).

Laboratory Characterization of the Slow-Light Mach-Zehnder Interferometer



- Interference fringes

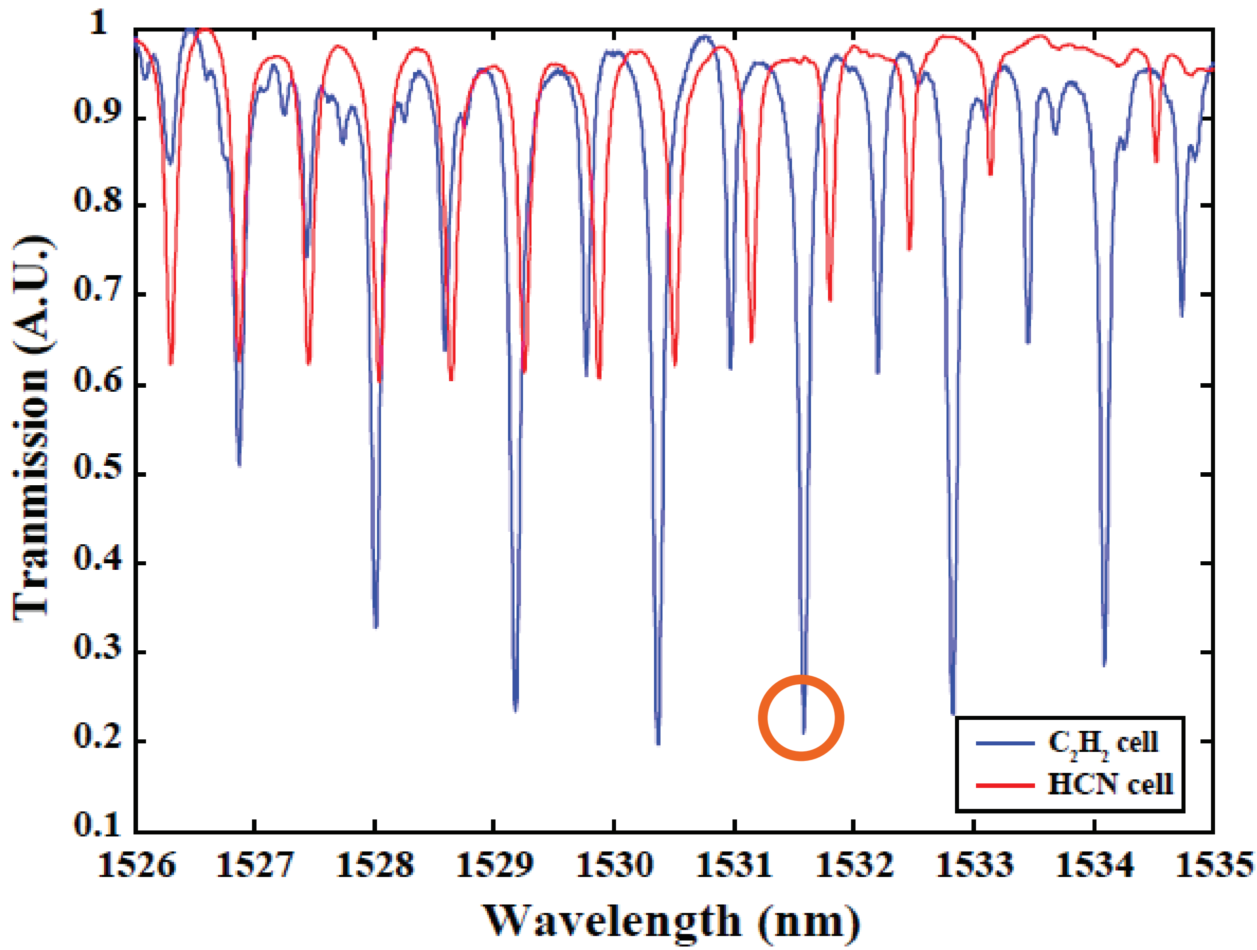


- Resolution (quarter wave) is 17 pm or 2.1 GHz or 0.071 cm^{-1}

- (Slow-light waveguide is only 1 mm long!)

Magaña-Loaiza, Gao, Schulz, Awan, Upham, Dolgaleva, and Boyd, Opt. Lett. 41, 1431 (2016).

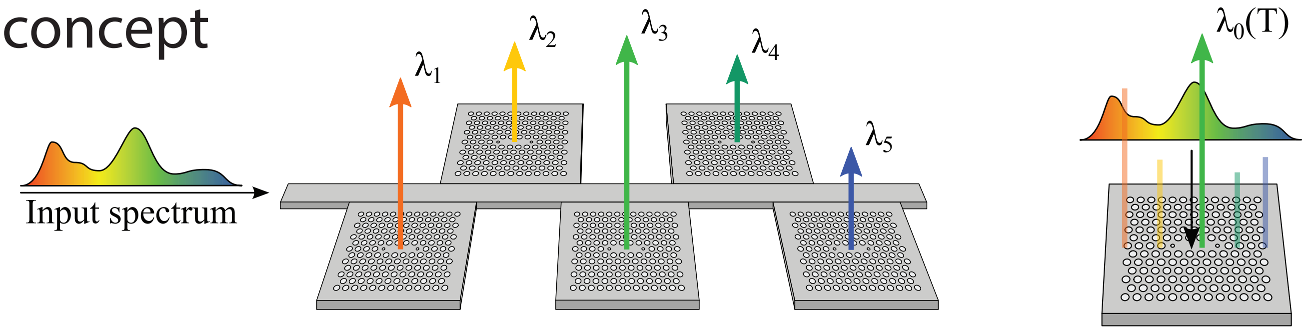
Challenge: Fabricate a chip-scale spectrometer that can discriminate acetylene (H_2C_2) from hydrogen cyanide (HCN)?



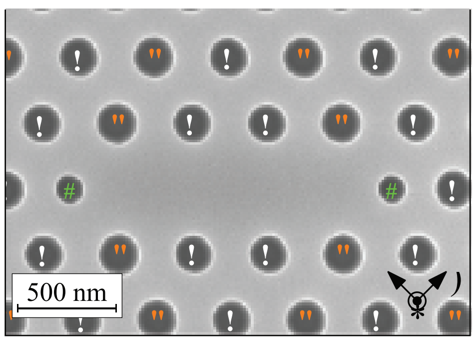
(data from our own lab)

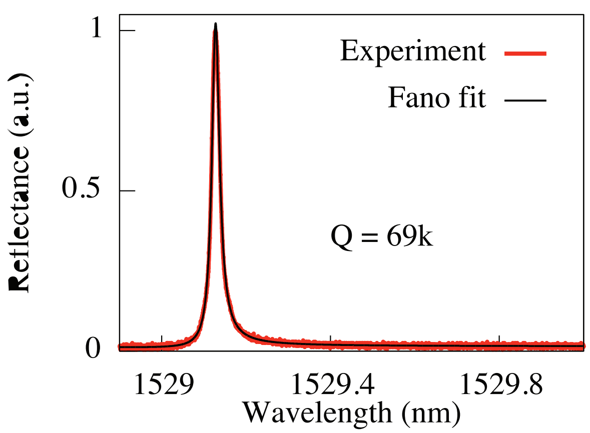
On-chip spectrometer based on high-Q photonic crystal cavities

- The concept

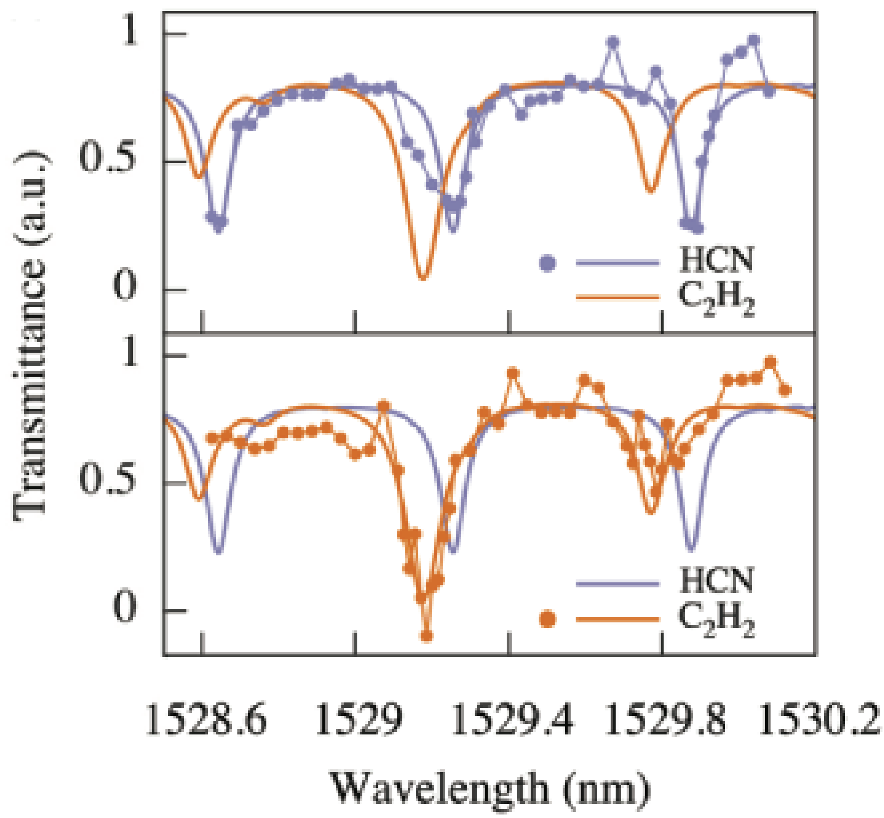


- Cavity design





- Spectroscopy results



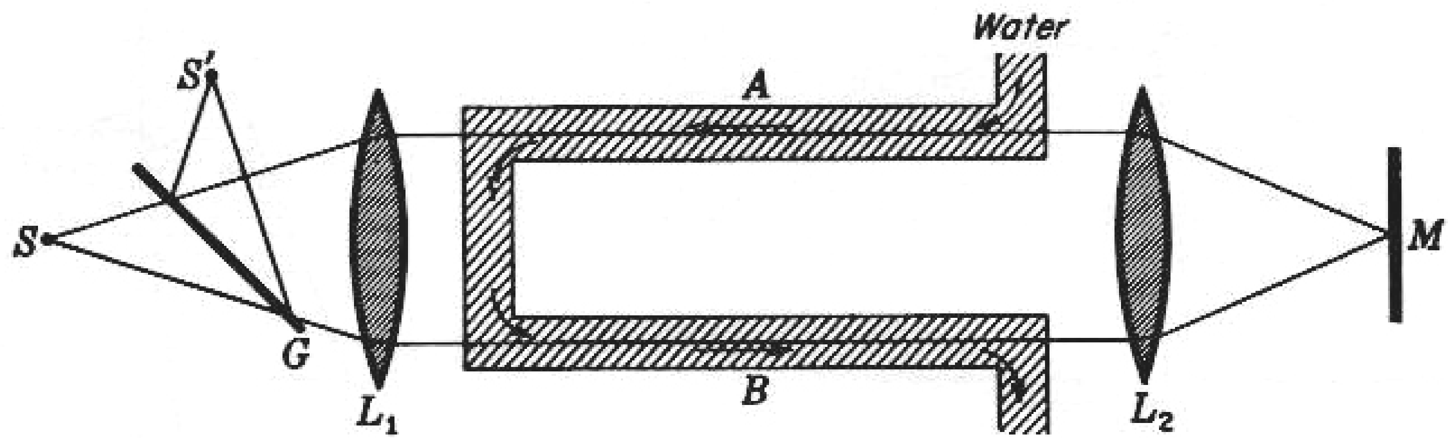
Liapis, Gao, Siddiqui, Shi, Boyd, Appl. Phys. Lett. 108, 021105 (2016).

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 cm/sec; L = 150 cm; displacement of 0.5 fringe.



- Modern theory: relativistic addition of velocities

$$\mathrm{v} = \frac{\mathrm{c}/n + V}{1 + (V/\mathrm{c})(1/n)} \approx \frac{c}{n} + V\left(1 - \frac{1}{n^2}\right)$$

Fresnel "drag" coefficient

- But what about slow-light media?

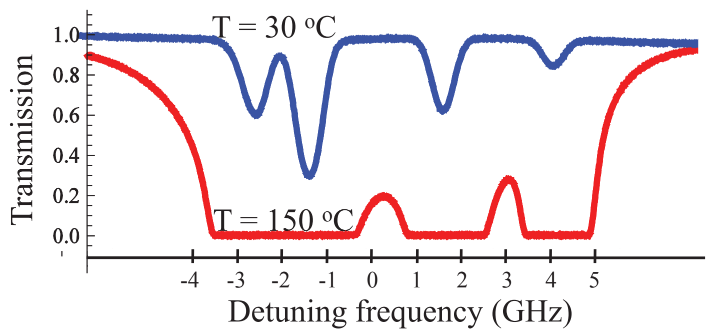
Fresnel Drag in a Highly Dispersive Medium

Light Drag in a Slow Light Medium (Lorentz)

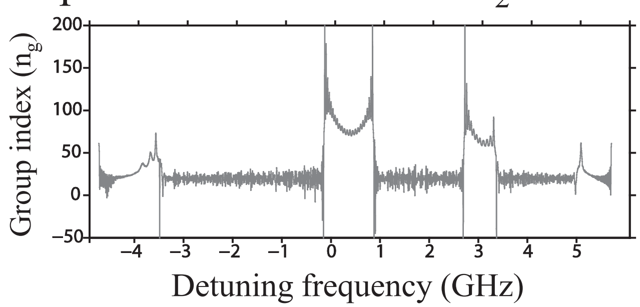
$$u \simeq \frac{c}{n} \pm v\left(1 - \frac{1}{n^2} + \frac{n_g - n}{n^2}\right)$$

We Use Rubidium as Our Slow Light Medium

- Transmission spectrum of Rb around D_2 transition:

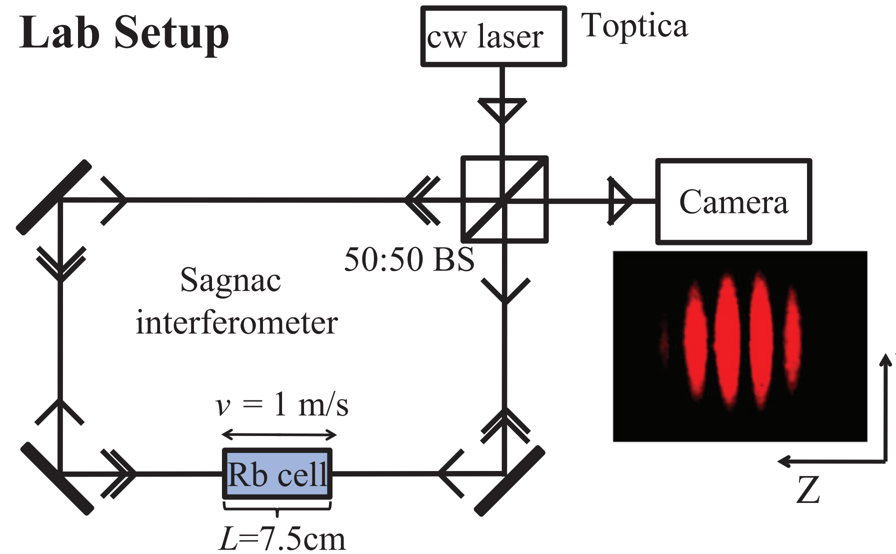


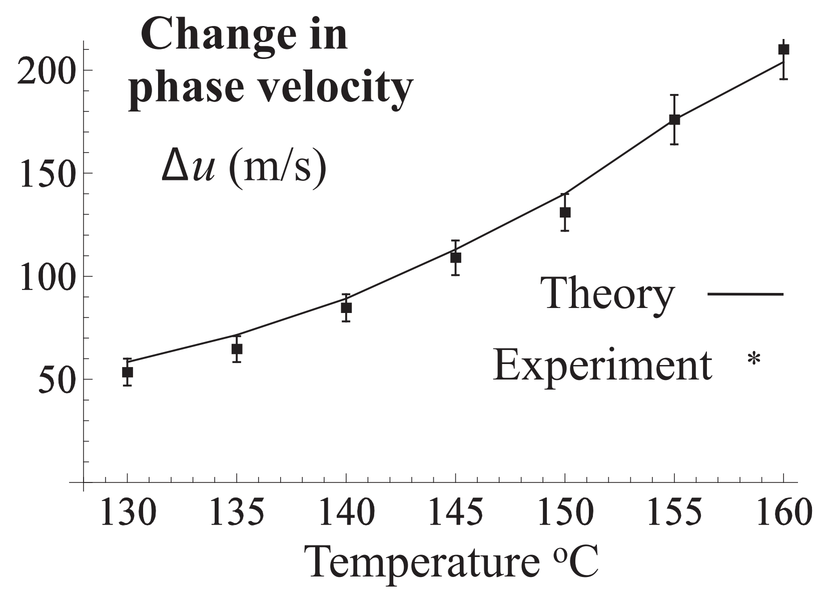
- Group index of Rb around D_2 line at T=130



Safari, De Leon, Mirhosseini, Magana-Loaiza, and Boyd
Phys. Rev. Lett. 116, 013601 (2016)

Lab Setup





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

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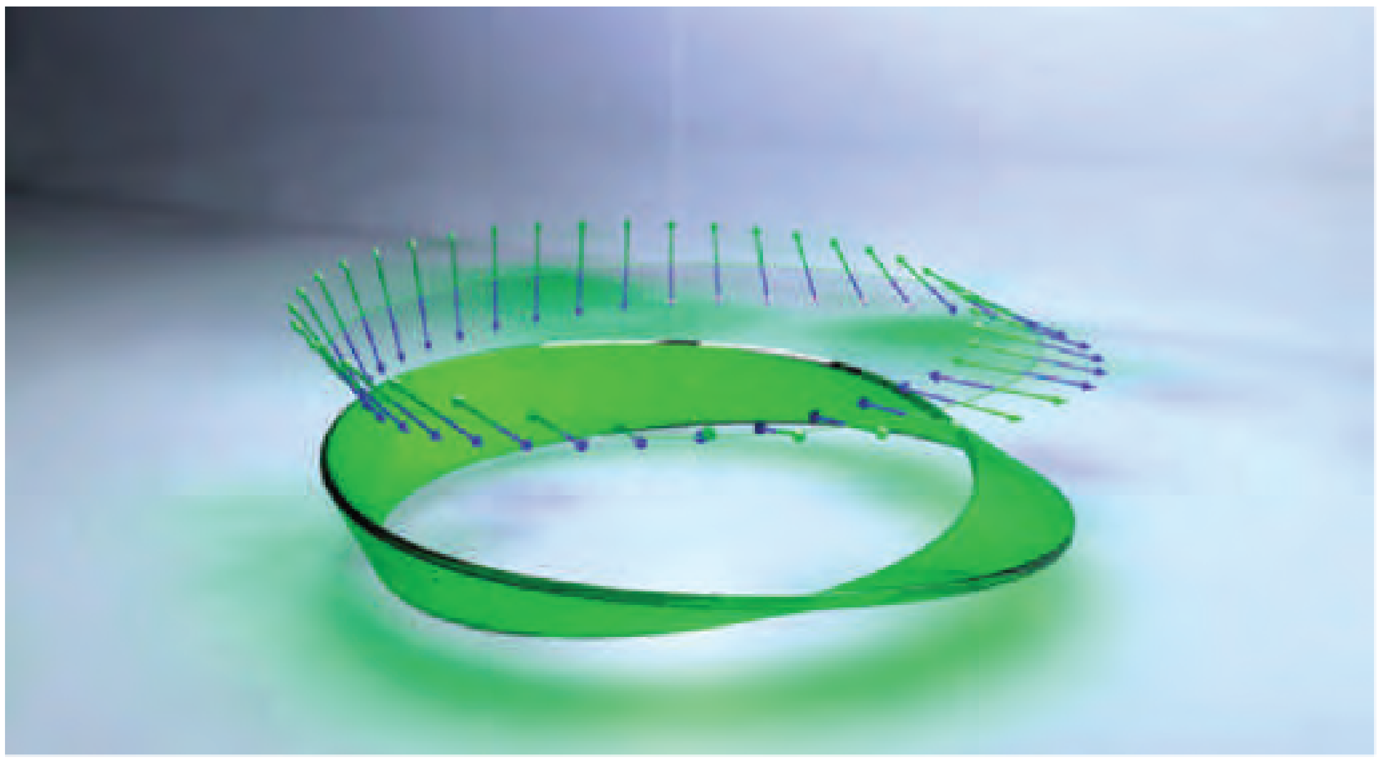
Observation of Optical Polarization Möbius Strips

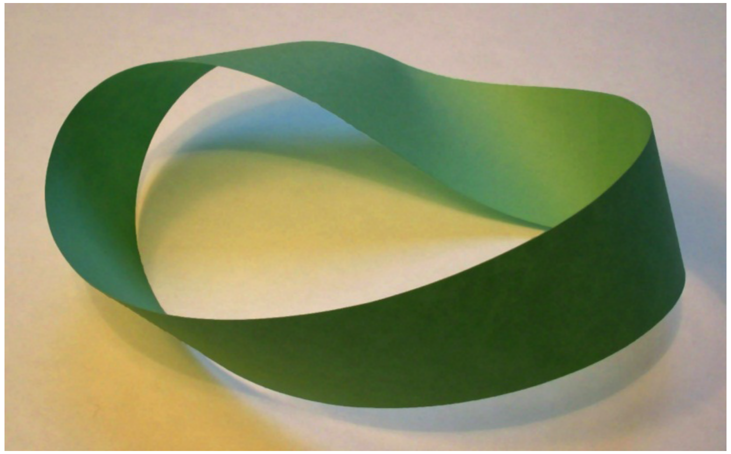
- Möbius strips are familiar geometrical structures, but their occurrence in nature is extremely rare.
- We generate such structures in the nanoscale in tightly focused vector light beams and confirm experimentally their Möbius topology.



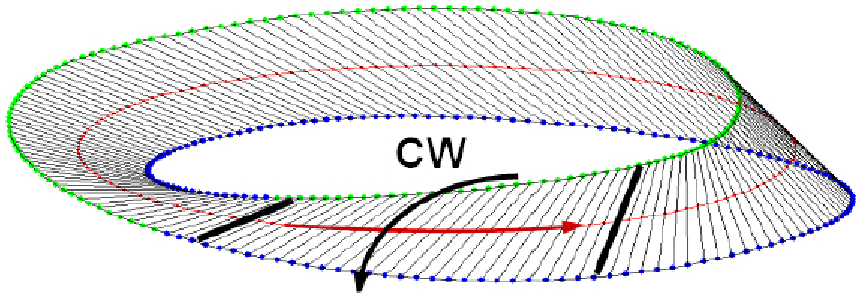
Bauer, Banzer, Karimi, Orlov, Rubano, Marrucci, Santamato, Boyd and Leuchs, Science, 347, 964 (2015)

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

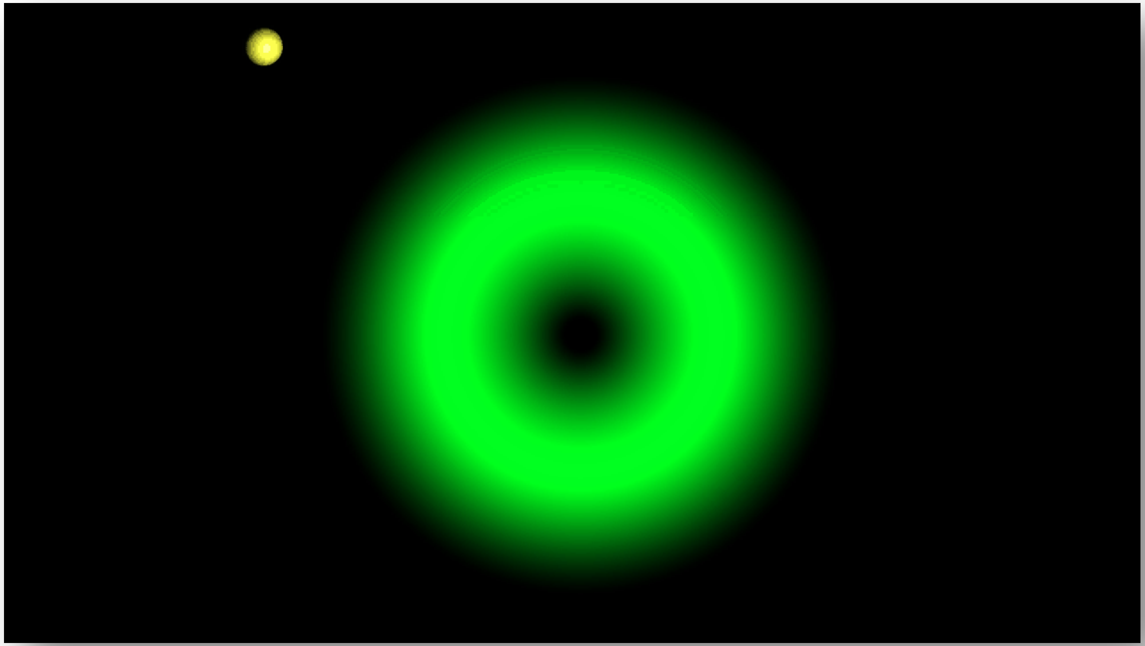
[2] Isaac Freund, Bar-Ilan Univ., Talk: *Optical Moebius Strips and Twisted Ribbons*, Conf. on Singular Optics, ICTP Trieste, Part II, 30 May 2011

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)

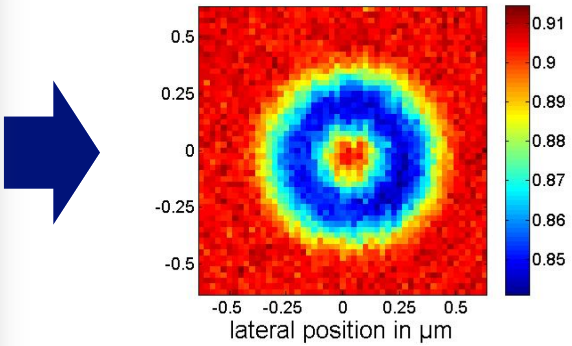
Full vectorial beam measurement on the nanoscale

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

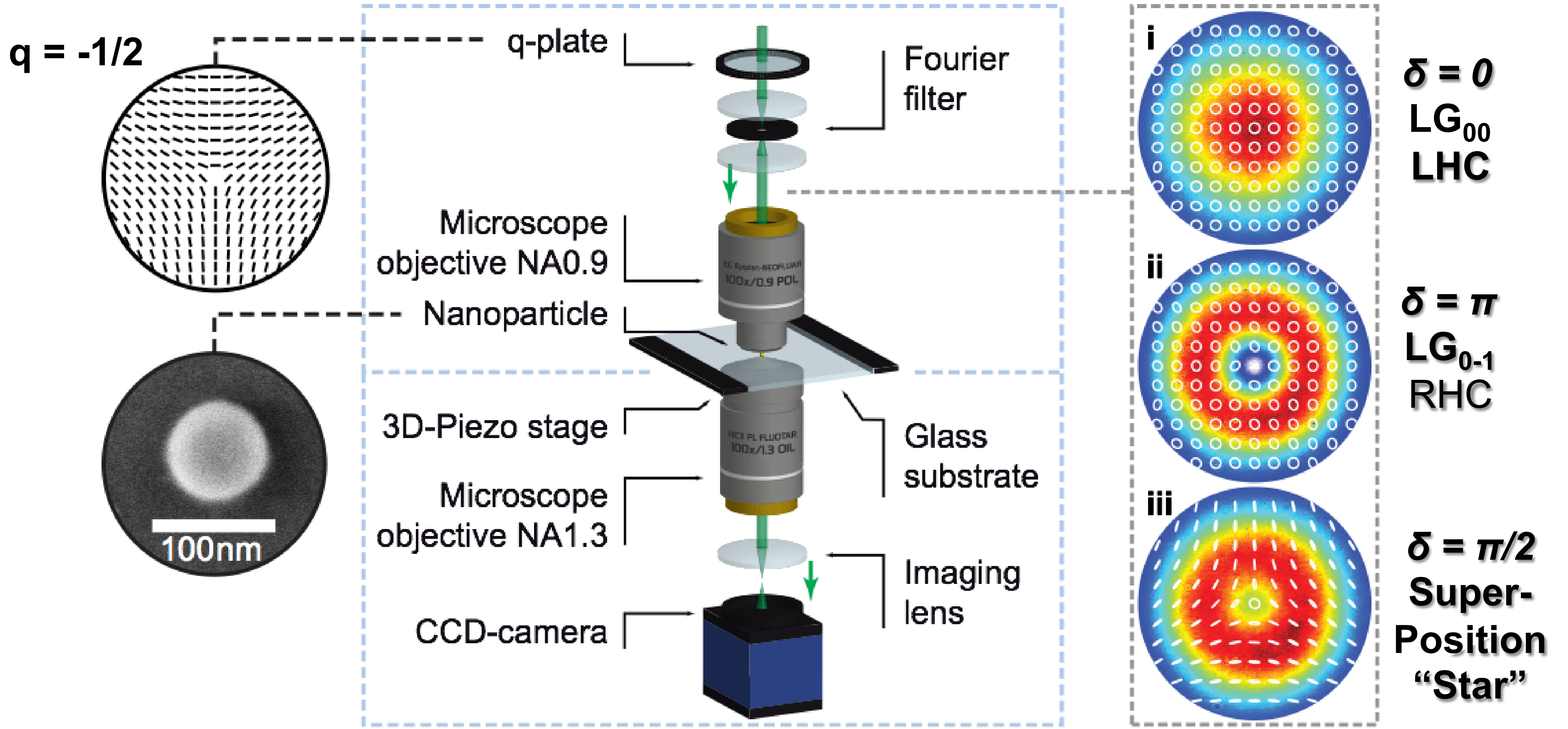


measured intensity
(can also measure
polarization and phase)



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

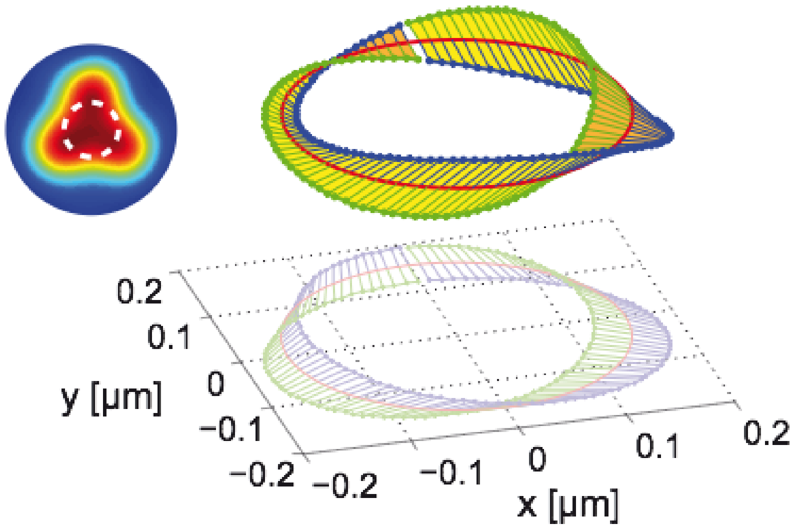


- q-plate: waveplate with a spatially varying orientation
  (q is the topological charge)
- output beam has a spatially varying state of polarization
  (vector beam, Poincaré beam, etc.)

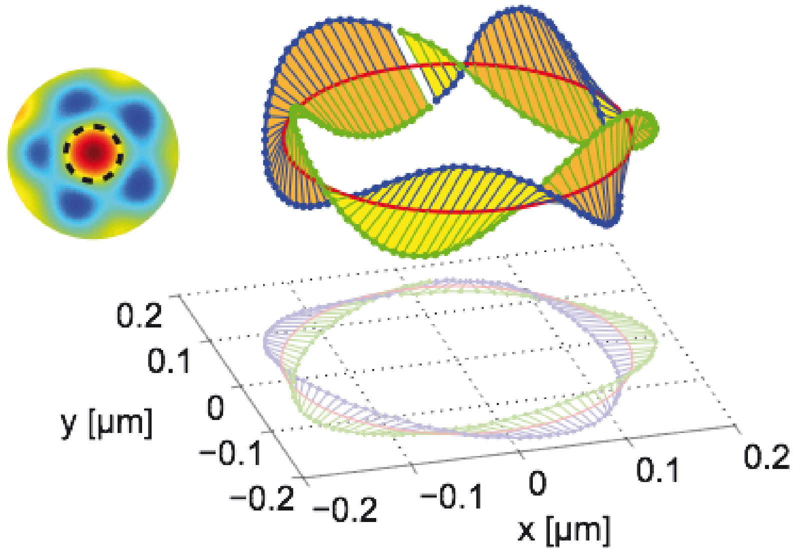
Tight focusing enhances the Möbius effect,which depends on the z component of the field

Observation of Polarization Möbius Strips

q =-1/2
3/2 twists



q = -3/2
5/2 twists



structure possesses |q|+1 half twists

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.

Bauer T, Banzer P, Karimi E, Orlovas S, Rubano A, Marrucci L, Santamato E, Boyd RW, and Leuchs G. Science, 2015.

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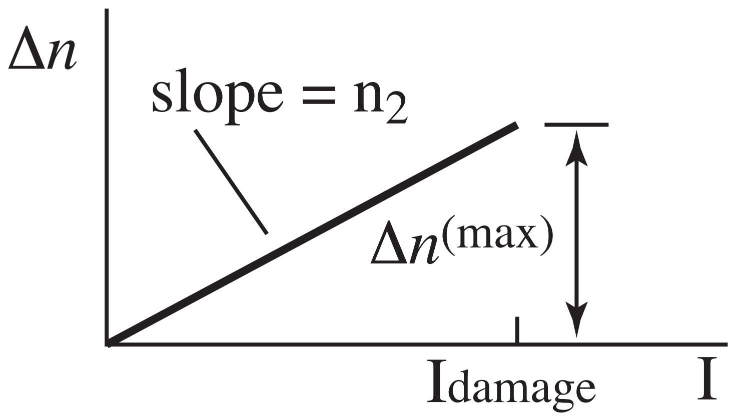
New Nonlinear Optical Material for Quantum Technologies

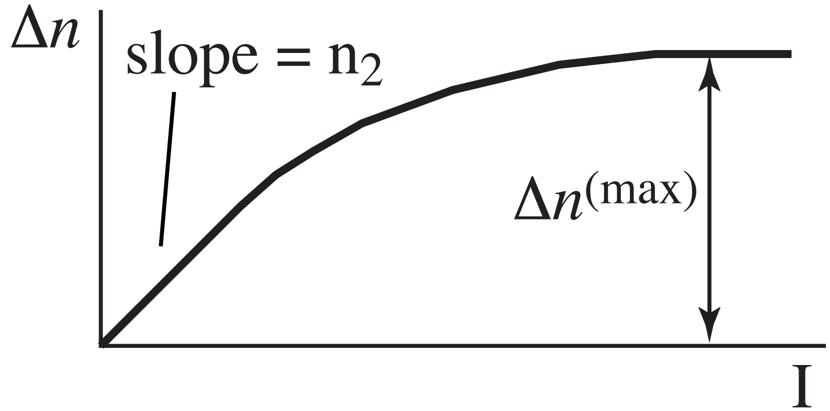
- We want all-optical switches that work at the single-photon level
- We need photonic materials with a much larger NLO response
- We recently reported a new NLO material with an n_2 value 100 times larger than any previously reported results (but with some background absorption).
- A potential game changer for the field of photonics

Large optical nonlinearity of indium tin oxide in its epsilon-near-zero region, M. Zahirul Alam, I. De Leon, R. W. Boyd, Science 352, 795 (2016).

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

- We want n_2 large ($\Delta n = n_2 I$). We 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, $n_2 = 1.1 \times 10^{-10}$ cm^2/W and $\Delta n^{(\max)} = 0.8$
  (For silica glass $n_2 = 3.2 \times 10^{-16}$ cm^2/W, $I_{damage} = 1$ TW/cm^2, and thus $\Delta n^{(\max)} = 3 \times 10^{-4}$)
- Thus n_2 is 3.4×10^5 times larger than that of silica glass
  $\Delta n^{(\max)}$ is 2700 times larger that that of silica glass

M. Z. Alam, I. De Leon, R. W. Boyd, Science 352, 795 (2016).

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 μm.

Recall the Drude formula

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

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

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

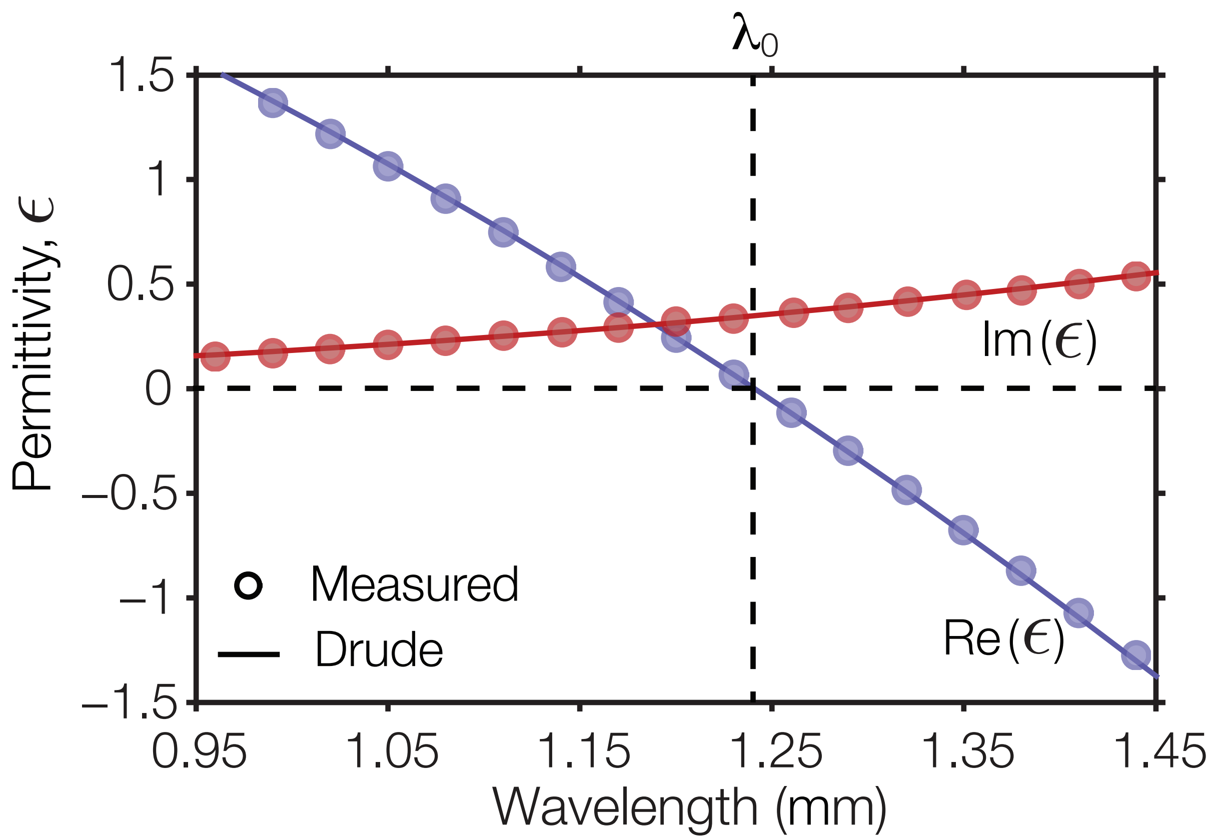
There has been great recent interest in studies of ENZ phenomena:

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



Drude fit

$\epsilon_\infty = 3.77$

$\gamma = 0.0468\ \omega_p$

$\omega_p/2\pi = 473$ THz

Note that Re(ϵ) vanishes at 1.24 mm, but that the loss-part Im(ϵ) 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 $\chi^{(3)}$

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

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

Why Does ENZ Lead to Large NLO Response?

+

Simple Math:

$$\epsilon = \epsilon_b + \Delta\epsilon \quad (\text{b} = \text{“bulk”})$$

$$n = \sqrt{\epsilon} = \sqrt{\epsilon_b + \Delta\epsilon}$$

Assume $\Delta\epsilon \ll \epsilon_b$ (this assumption can be violated).

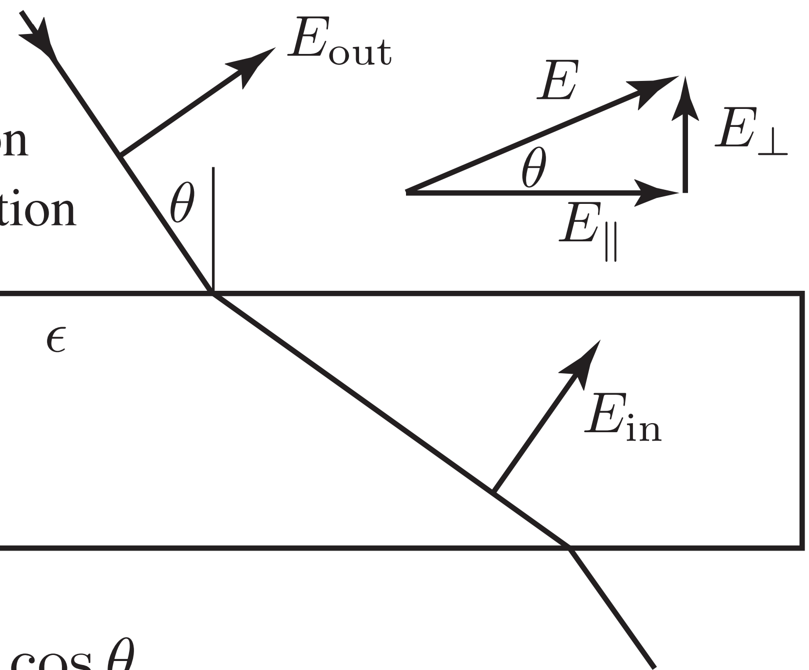
$$n = \sqrt{\epsilon_b}\left(1 + \frac{\Delta\epsilon}{2\epsilon_b} + \cdots\right) = \sqrt{\epsilon_b} + \frac{\Delta\epsilon}{2\sqrt{\epsilon_b}}$$

or

$$n = n_b + \Delta n \quad \text{where} \quad \Delta n = \frac{\Delta\epsilon}{2n_b}$$

The NLO Response Is Even Larger at Oblique Incidence

p polarization
TM polarization



Standard boundary conditions show that:

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

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

Thus the total field inside of the medium is given by

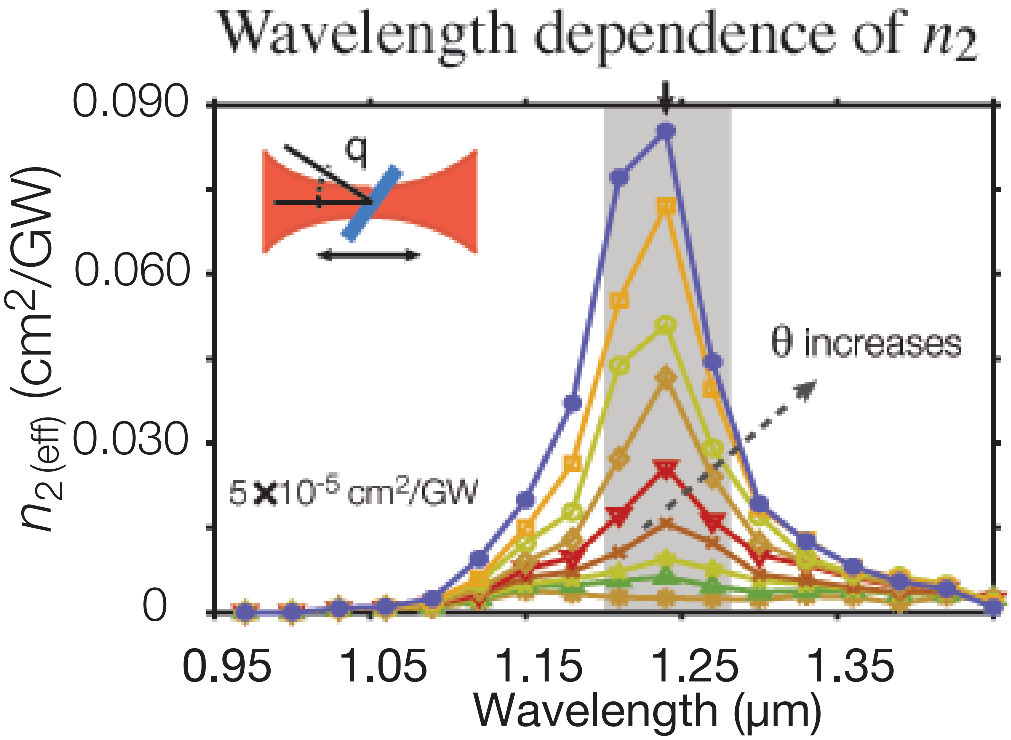
$$E_{\mathrm{in}} = E_{\mathrm{out}} \sqrt{\cos^2\theta + \frac{\sin^2\theta}{\epsilon}}$$

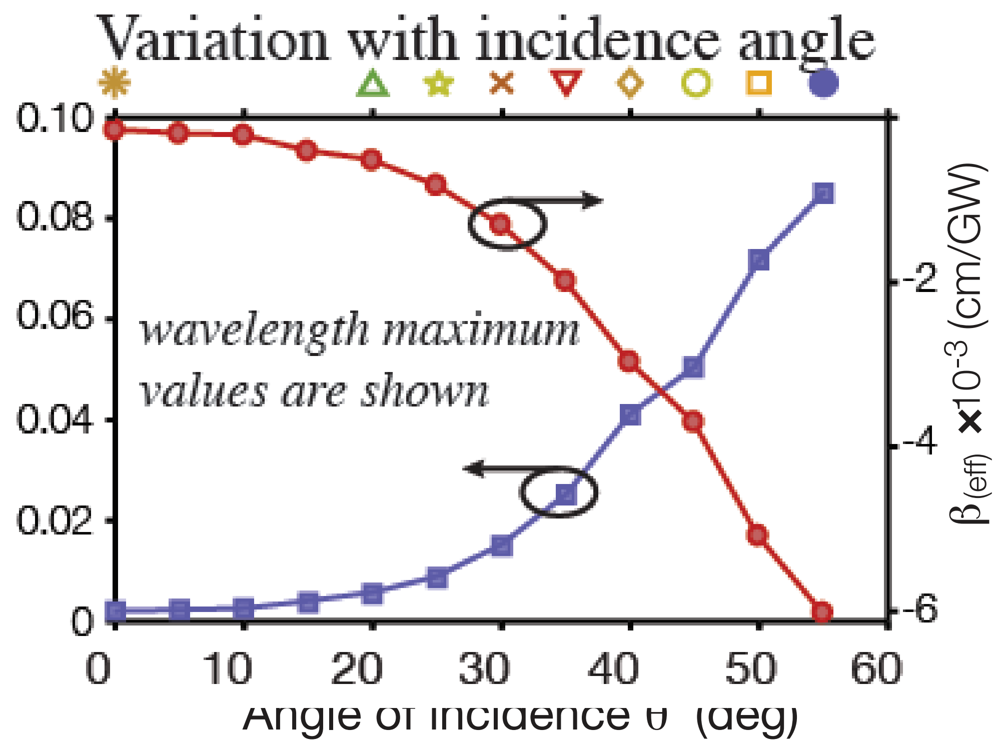
Note that, for $\epsilon < 1$, E_{in} exceeds E_{out} for $\theta \neq 0$.

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

Huge Nonlinear Optical Response of ITO

- Z-scan measurements for various angles of incidence





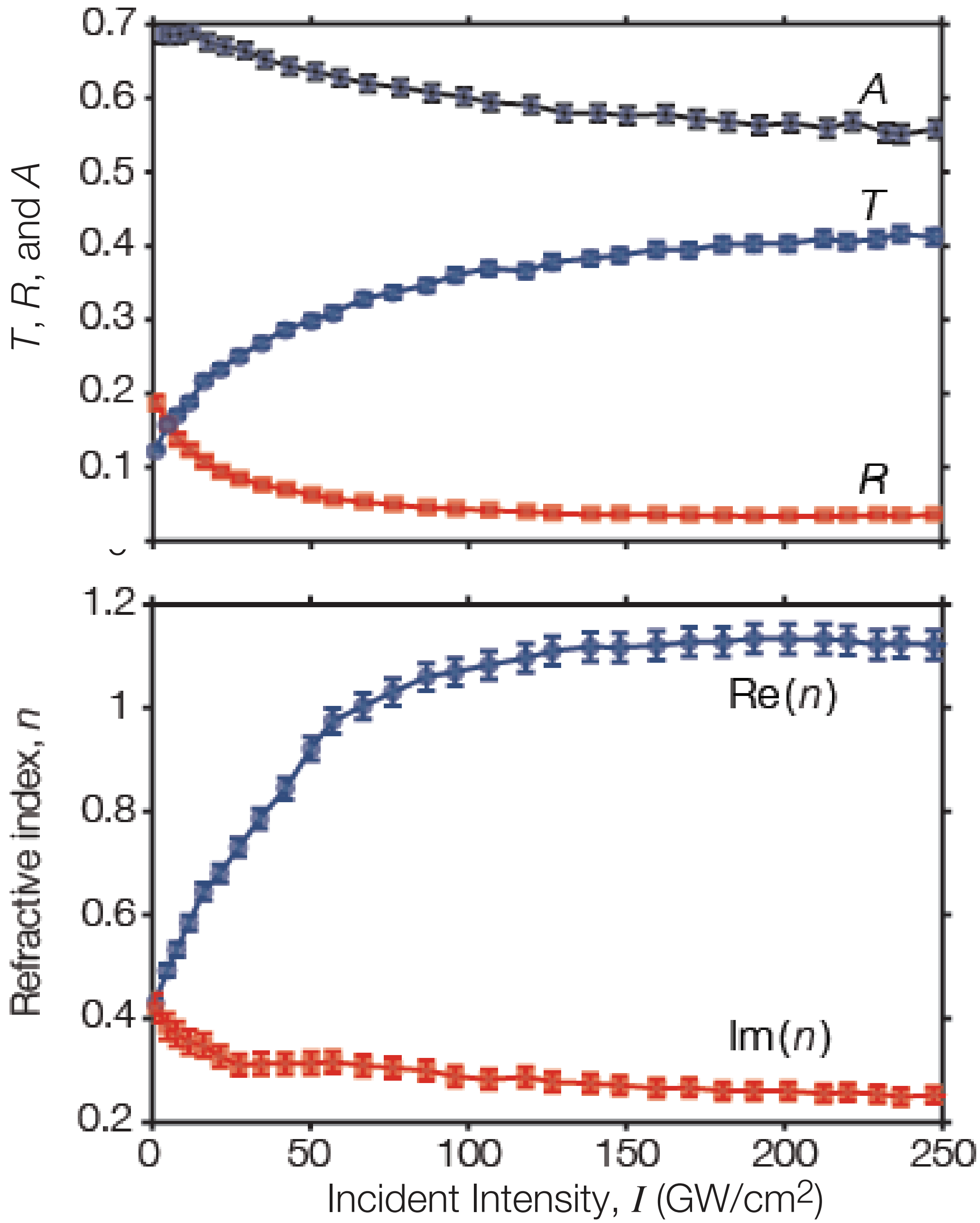
peak laser intensity was 50 GW cm^{-2}

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

- Both n_2 and nonlinear absorption increase with angle of incidence

- n_2 shows a maximum value of 0.11 cm^2/GW = 1.1 x 10^{-10} cm^2/W at 1.25 μm and 60 deg. This value is 2000 times larger than that away from ENZ region.

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.

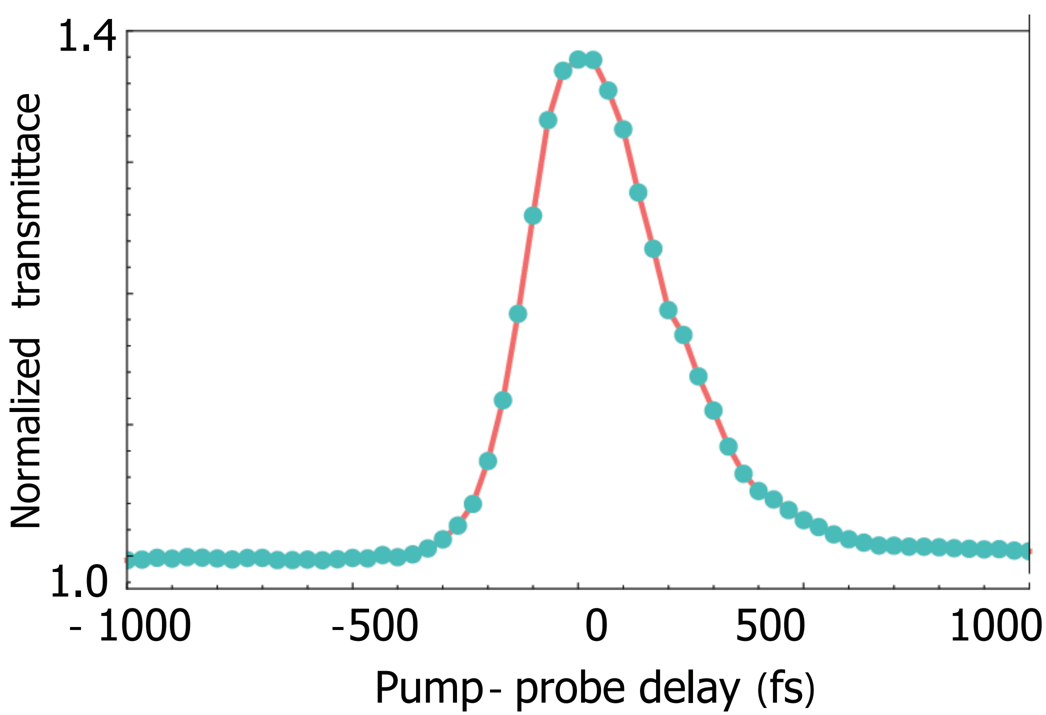
Here is the refractive index extracted from the above data.

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 shows a rise time of no longer than 200 fs and a recover time of of 360 fs.
- Results suggest a hot-electron origin of the nonlinear response
- ITO will support switching speeds as large as 1.5 THz



Implications of the Large NLO Response of ITO

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

n_2 is 3.4 x 10^5 times larger than that of fused silica
n_2 is 200 times larger than that of chalcogenide glass
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
Control of radiative processes

Other Reports of Highly Nonlinear Response in ENZ Material and Metamaterialc

Enhanced Nonlinear Refractive Index in epsilon-Near-Zero Materials,
L. Caspani, R. P. M. Kaipurath, M. Clerici, M. Ferrera, T. Roger, J. Kim, N. Kinsey,
M. Pietrzyk, A. D. Falco, V. M. Shalaev, A. Boltasseva and D. Faccio,
Phys. Rev. Lett. 116, 233901, 2016.

Giant nonlinearity in a superconducting sub-terahertz metamaterial,
V. Savinov, K. Delfanazari, V. A. Fedotov, and N. I. Zheludev
Applied Physics Letters 108, 101107 (2016); doi: 10.1063/1.4943649

Nano-optomechanical nonlinear dielectric metamaterials
Artemios Karvounis, Jun-Yu Ou, Weiping Wu, Kevin F. MacDonald, and Nikolay I. Zheludev
Applied Physics Letters 107, 191110 (2015); doi: 10.1063/1.4935795.

Nanostructured Plasmonic Medium for Terahertz Bandwidth All-Optical Switching
Mengxin Ren , Baohua Jia , Jun-Yu Ou , Eric Plum, Jianfa Zhang , Kevin F. MacDonald ,
Andrey E. Nikolaenko , Jingjun Xu, Min Gu, and Nikolay I. Zheludev *
Adv. Mater. 2011, 23, 5540–5544 (2011).

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

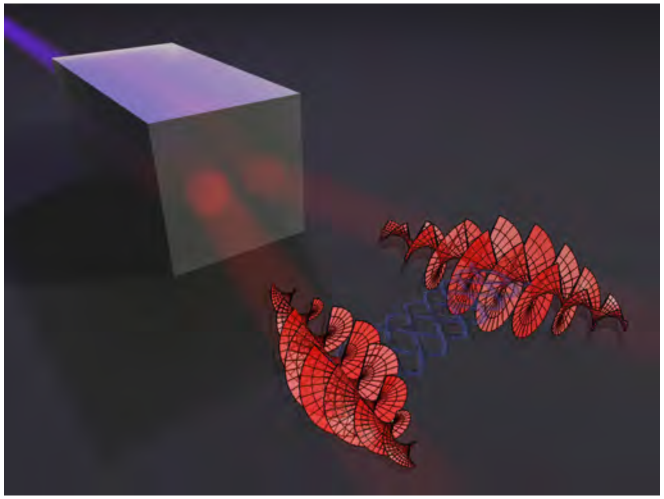
1. Introduction to Nonlinear Optics and Quantum NLO
2. New Applications of "Slow Light"
3. Möbius Strips of Polarization
4. Huge Optical Nonlinearity in Epsilon-Near-Zero Materials
5. Quantum Communication with Multiple Bits per Photon

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

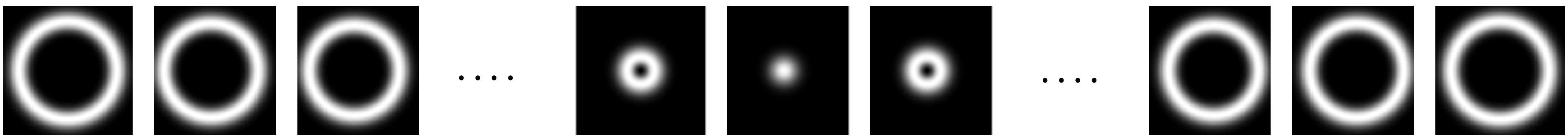
We are constructing a QKD system in which each photon carries many bits of information

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

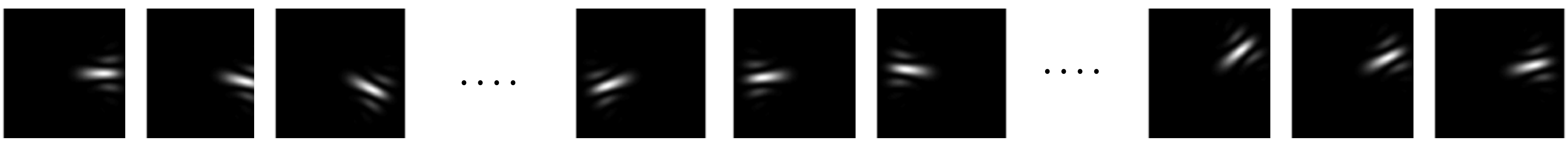
<u>Single Photon States</u>

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



-13 -12 -11 .... -1 0 1 .... 11 12 13

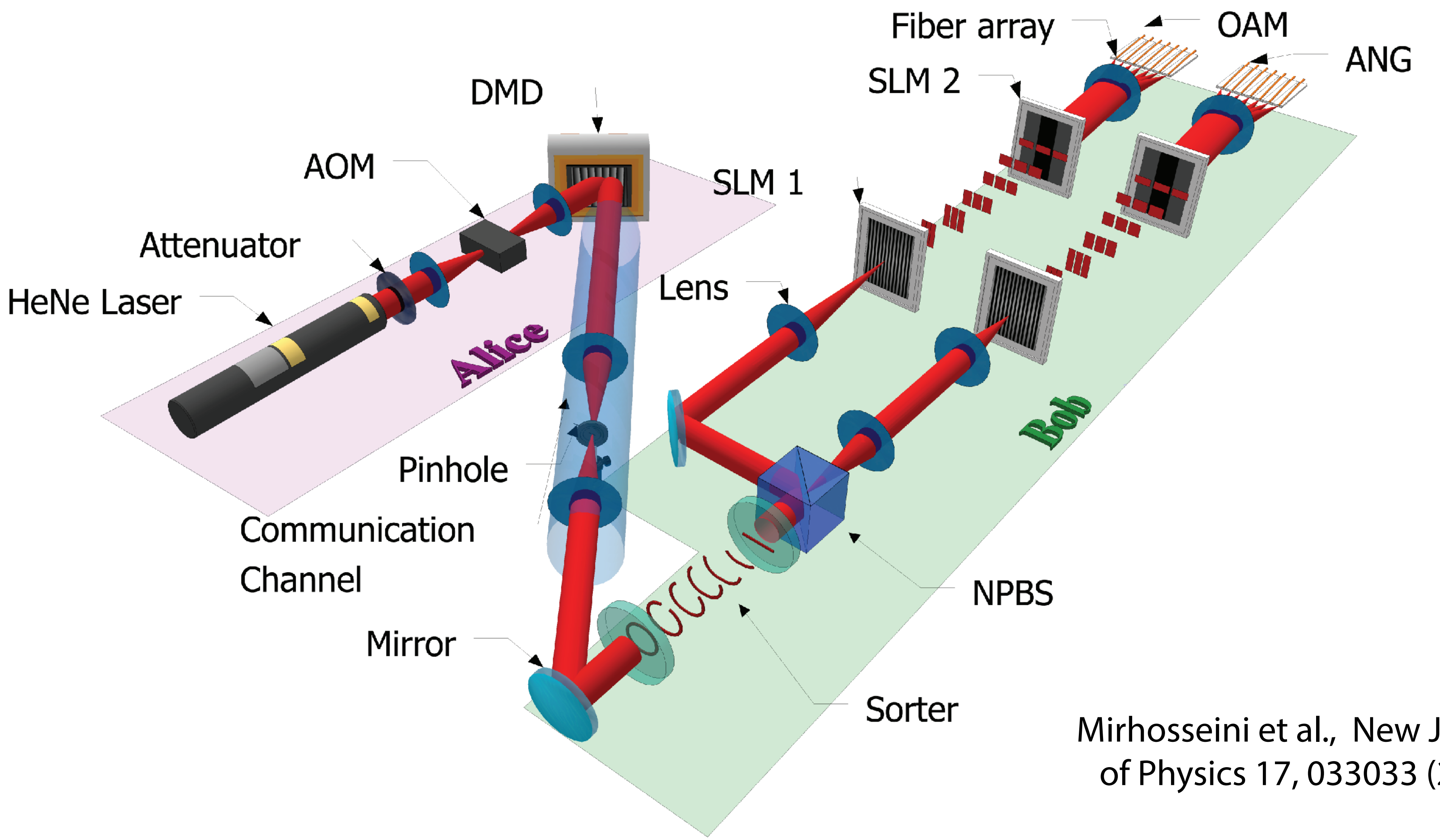
"Angular" Basis (mutually unbiased with respect to LG)



0 1 2 .... 12 13 14 .... 25 26 27

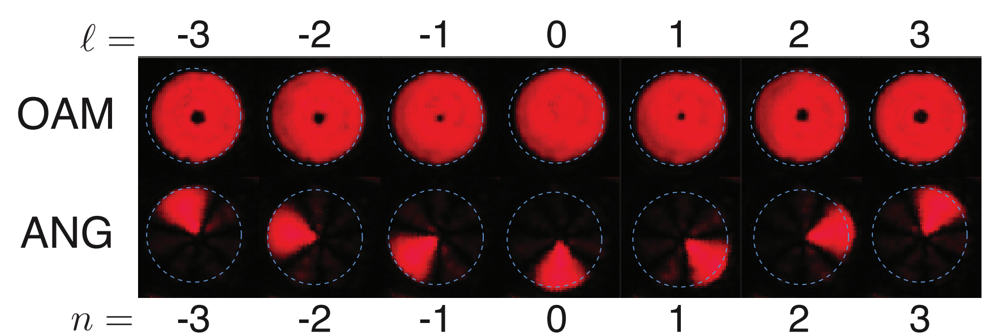
$$\Psi_{\mathrm{AB}}^{N} = \frac{1}{\sqrt{27}} \sum_{l=-13}^{13} \mathrm{LG}_{l,0} \exp\left(i 2\pi N l / 27\right)$$

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

Alice

063133602132045444456141026645
545050363603025261643215524164
230146602513401613222451551026

Bob

063033601132021444456141026645
545022353603025261643215524164
230146602515403613422451551026

Error Correction

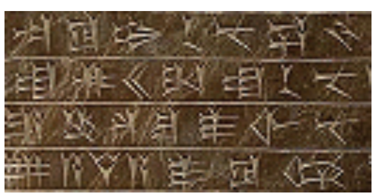
0000 0100 0111 0100 0101 0001 1100 0010 1100 1001 0010 0100 1101 0001 0000
0001 0011 0100 0000 0100 0010 0001 0001 0011 0000 0101 0010 0111 1000 0101
1101 0011 0001 1001 1011 0101 0000 0100 0111 0100 0011 0110 0101 0000 1101
0101 0010 1111 1010 1111 0010 1000

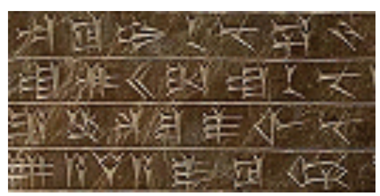
Privacy Amplification

0001 0100 1011 1111 1111 1010 1010 0111 1010 1011 1111 0001 0000 1101 1111
1110 1110 1101 1110 0001 0011 1000 0101 1101 1100 0011 0111 1101 0001 1001
0100 0110 1011 0000 0110 1010 1

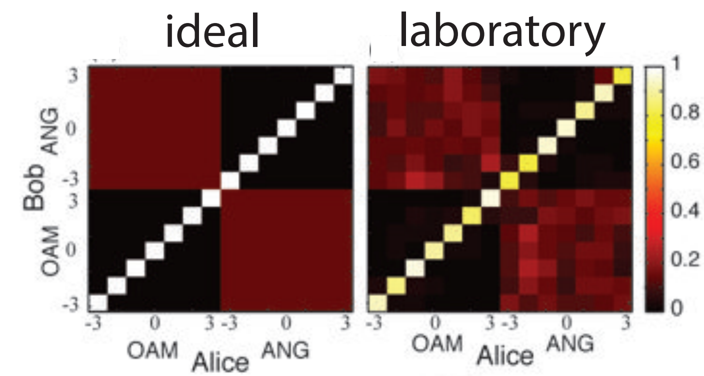
Alice Encrypts

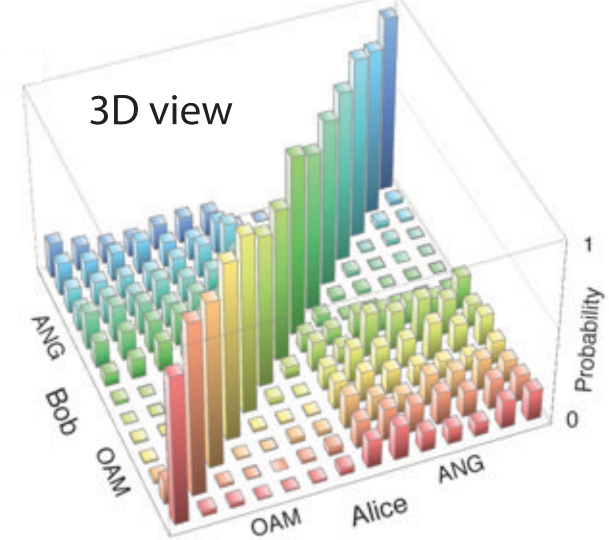
Bob Decrypts



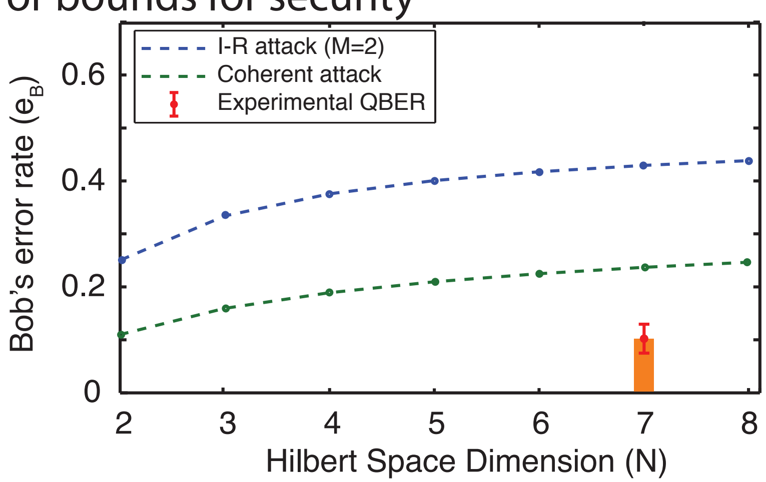


cross-talk matrices





- 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!