# Quantum Nonlinear Optics: Nonlinear Optics Meets the Quantum World

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Presented at the Nonlinear Optics Lecture Series of the Ottawa Chapter of the IEEE Photonics Society, September 14, 2016.

# Quantum Nonlinear Optics:
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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

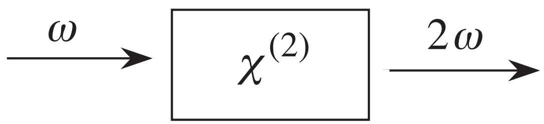
# Simple Formulation of the Theory of Nonlinear Optics

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

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

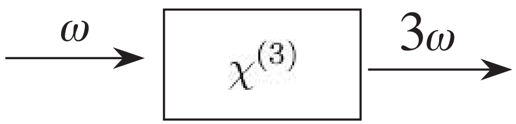
$\chi^{(1)}$ describes linear optics, e.g., how lenses work:

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

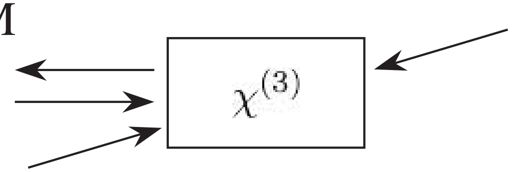



$\chi^{(3)}$ describes third-order effects such as third-harmonic generation, four-wave mixing, and the intensity dependence of the index of refraction.

THG




FWM




NL index

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

# Intense Field and Attosecond Physics

E 70, NUMBER 11 PHYSICAL REVIEW LETTERS 15 MARCH

**Above Threshold Ionization Beyond the High Harmonic Cutoff**

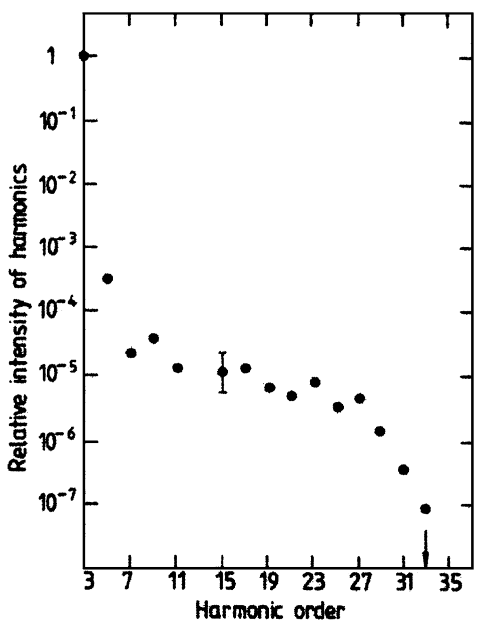
K. J. Schafer,[1] Baorui Yang,[2] L. F. DiMauro,[2] and K. C. Kulander[1]
[1] *Lawrence Livermore National Laboratory, Livermore, California 94550*
[2] *Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973*
(Received 2 December 1992)

VOLUME 71, NUMBER 13 PHYSICAL REVIEW LETTERS 27 SEPTEMBER 1993

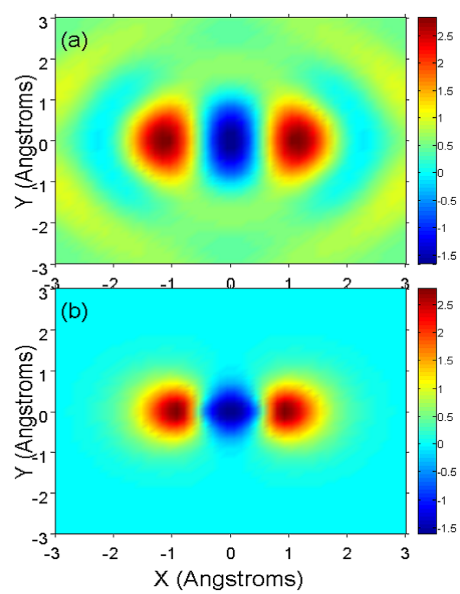
**Plasma Perspective on Strong-Field Multiphoton Ionization**

P. B. Corkum
*National Research Council of Canada, Ottawa, Ontario, Canada K1A 0R6*
(Received 9 February 1993)

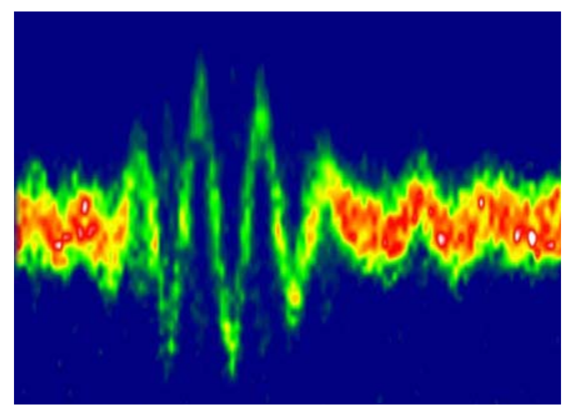

$I > 10^{15}\ W/cm^2$




High-harmonic generation




Measuring the molecular nitrogen wavefunction



Attosecond pulses to sample a visible E-field

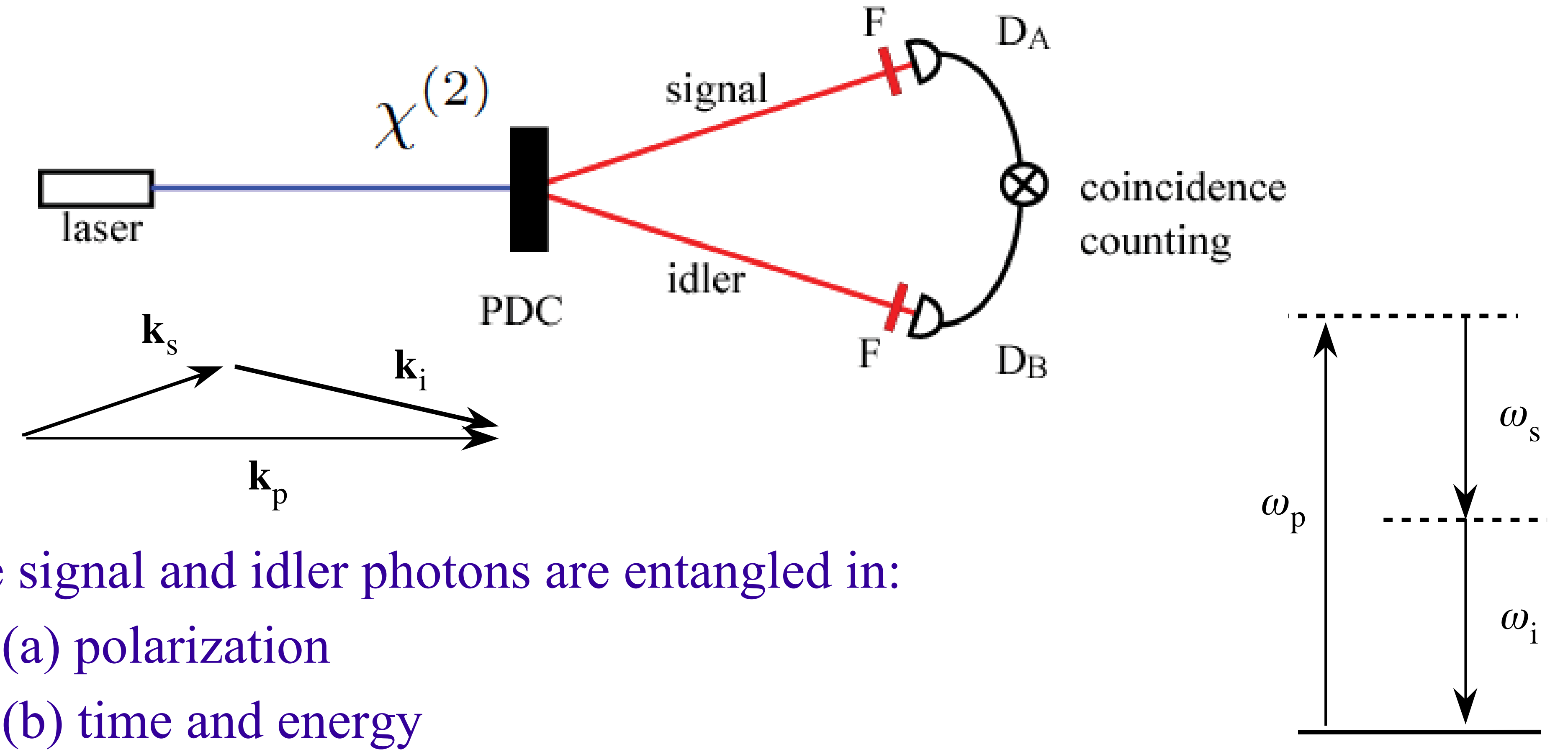
# Why Interest in Quantum Nonlinear Optics?

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

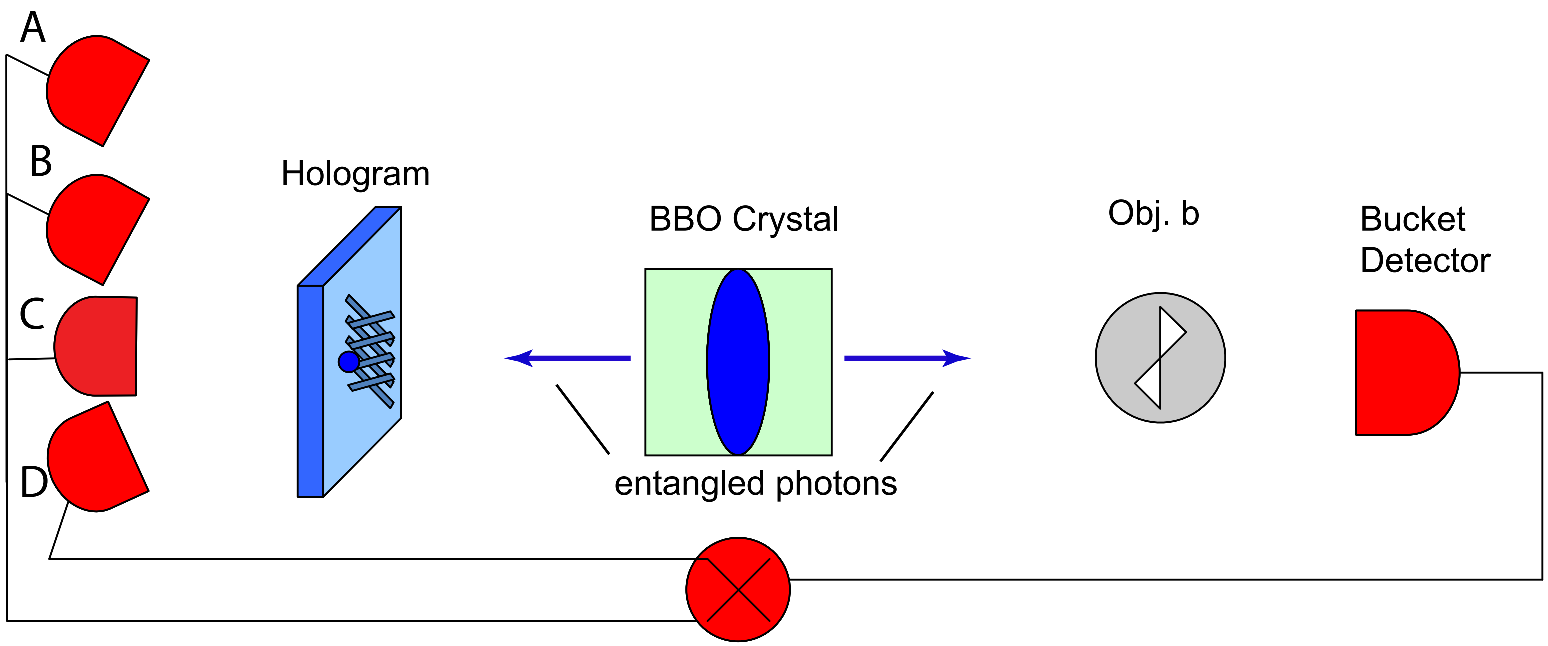
(d) angular position and orbital angular momentum

Entanglement is important for:

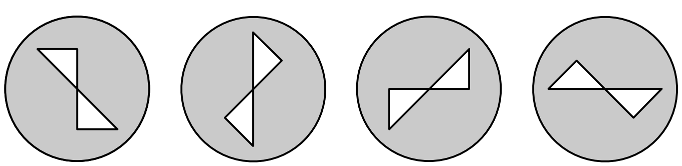
(a) Fundamental tests of QM (e.g., nonlocality, Bell tests)

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

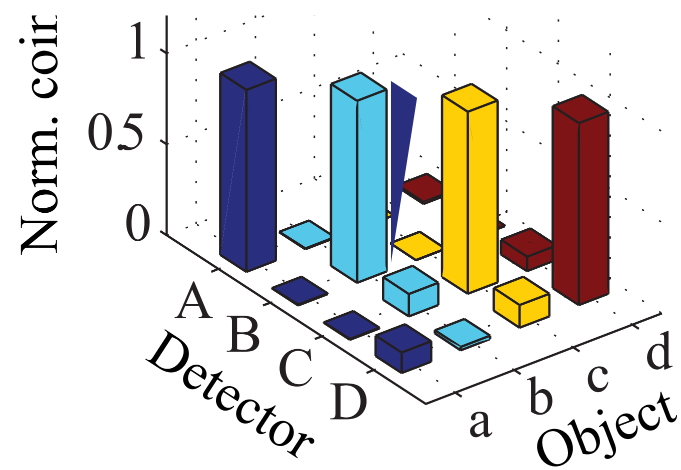
# Single-Photon Coincidence Imaging




- We discriminate among four orthogonal images using single-photon 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

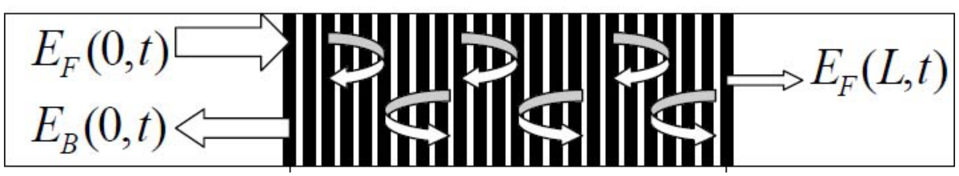
## "Slow," "Fast" and "Backwards" Light

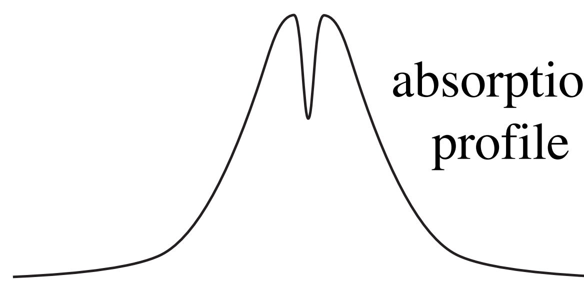
– Light can be made to go:

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: $v_g = c/n_g \quad n_g = n + \omega\,(dn/d\omega)$

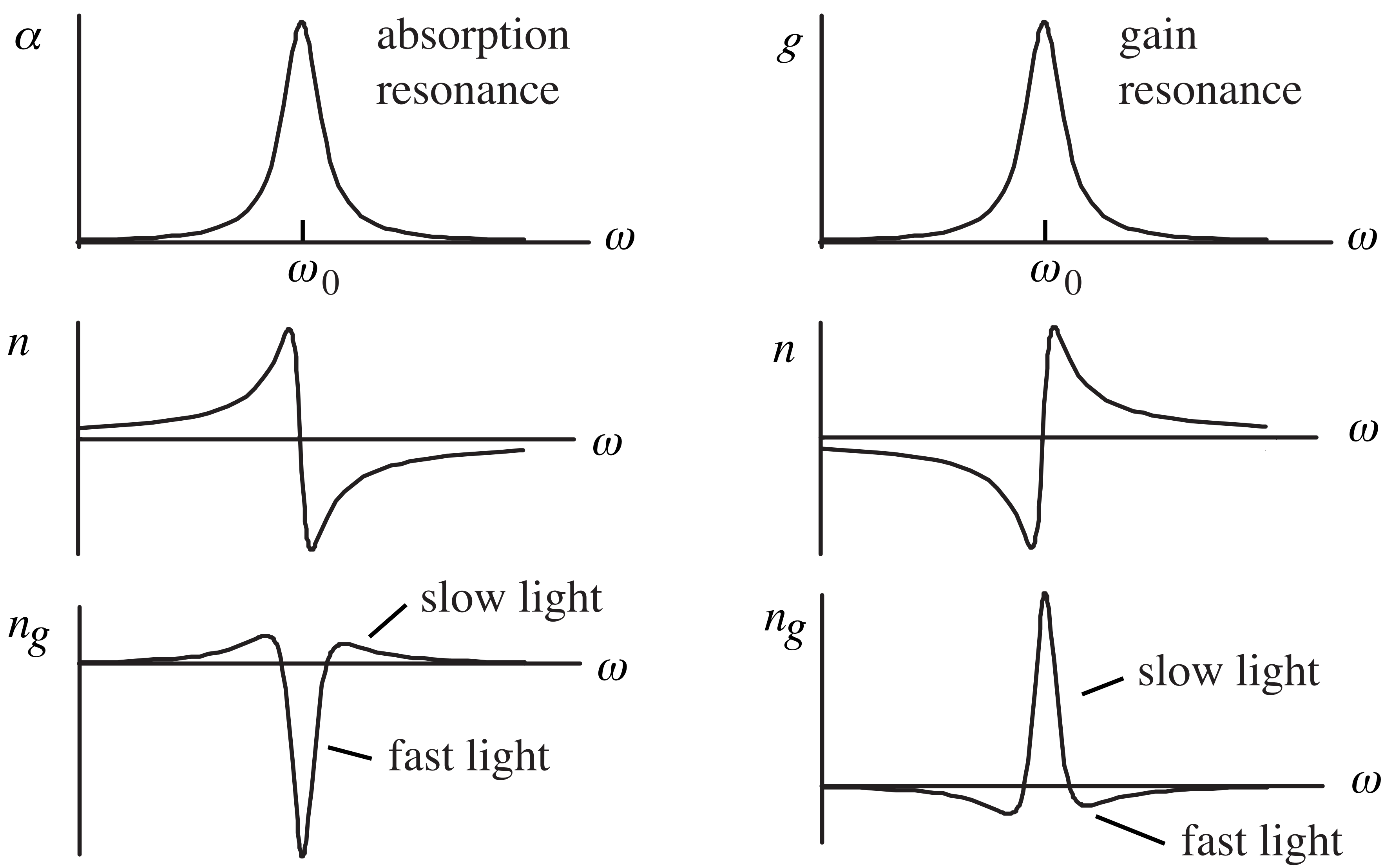
– Velocity controlled by structural or material resonances




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

# Slow and Fast Light Using Isolated Gain or Absorption Resonances



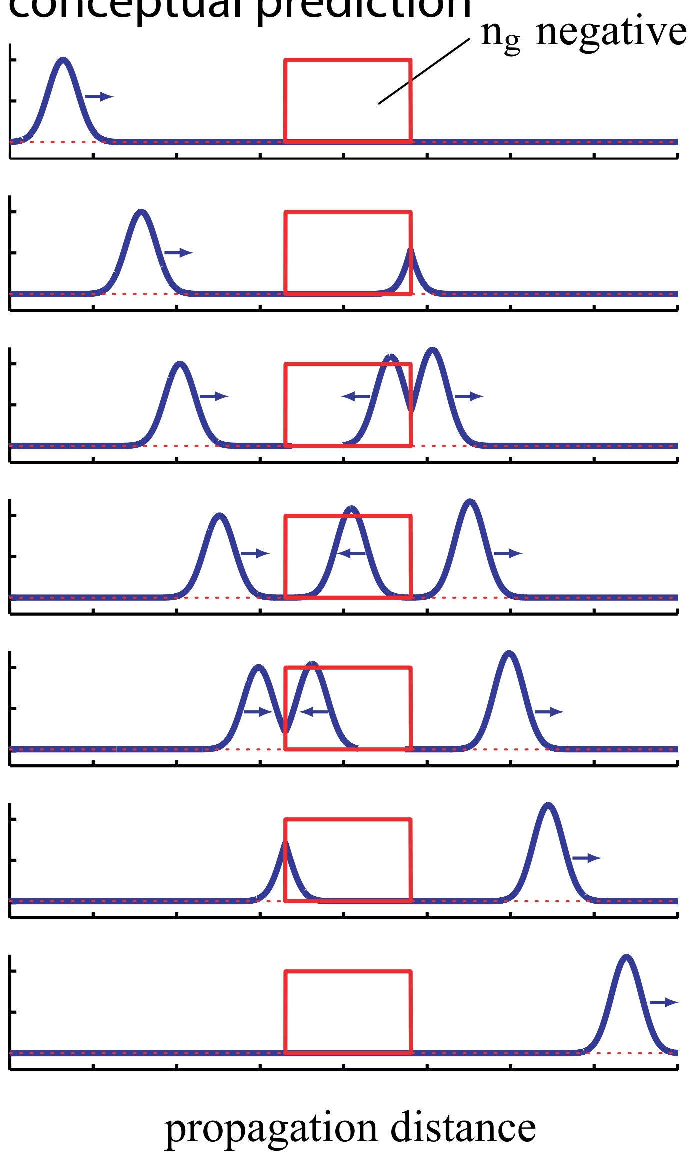

$$n_g = n + \omega\,(dn/d\omega)$$

# Observation of Superluminal and "Backwards" Pulse Propagation

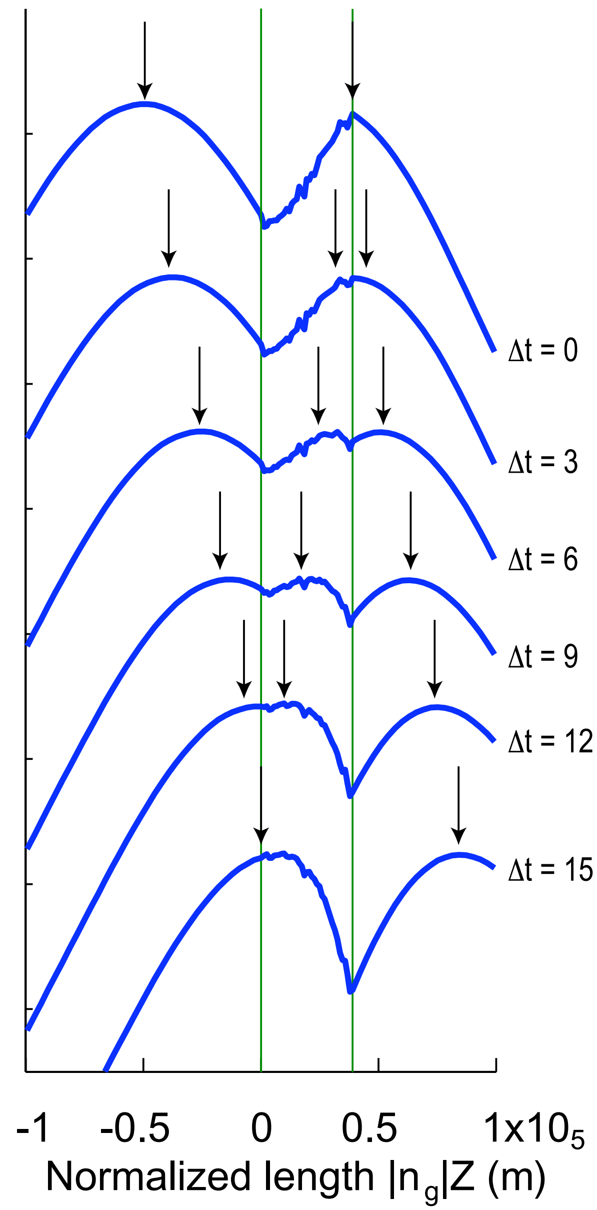
SLOW LIGHT

- 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

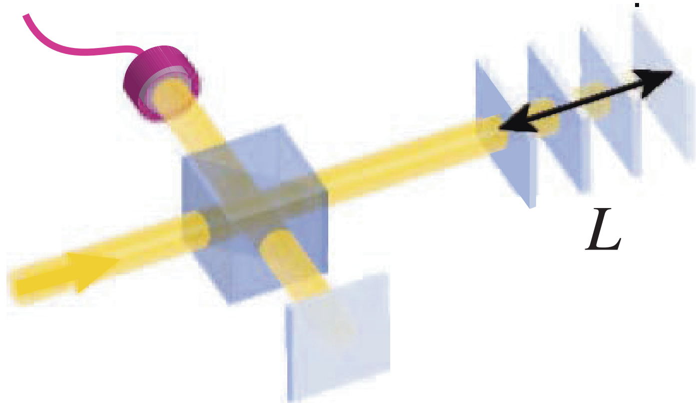


# Development of Miniaturized, Chip-Scale Spectrometers

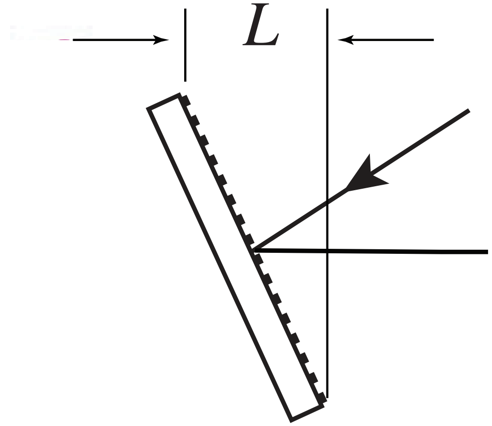
## Can We Beat the $1/L$ Resolution Limit of Standard Spectrometers?

- 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*




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

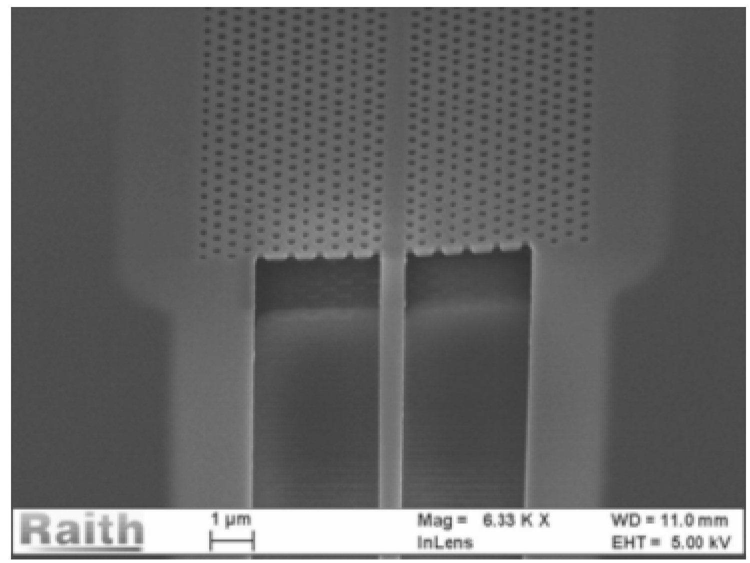
- We use slow-light methods to design spectrometers with resolution that exceeds this conventional limit by a factor as large as the group index.

- This ability allows us to miniaturize spectrometers with no loss of resolution, for "lab-on-a-chip" applications.

# Our Goal

Replace this:

with this:



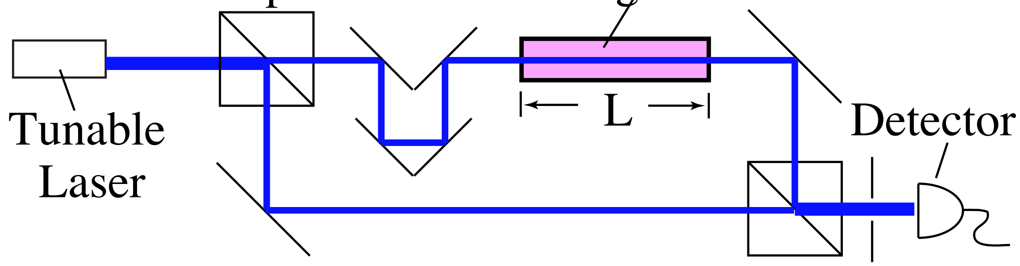
# Our Approach: Chip-Scale Slow-Light Spectrometer

- 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:

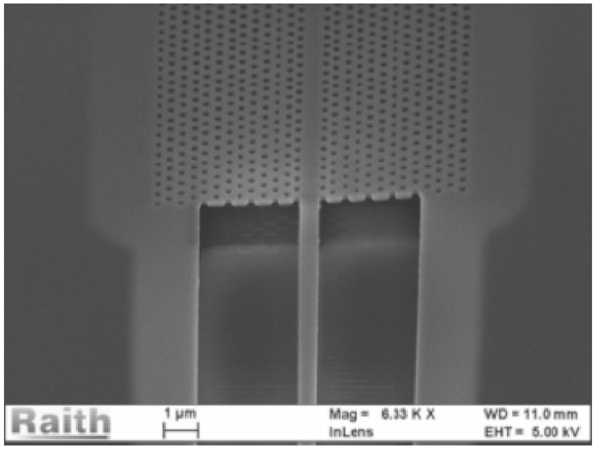



Simple analysis

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

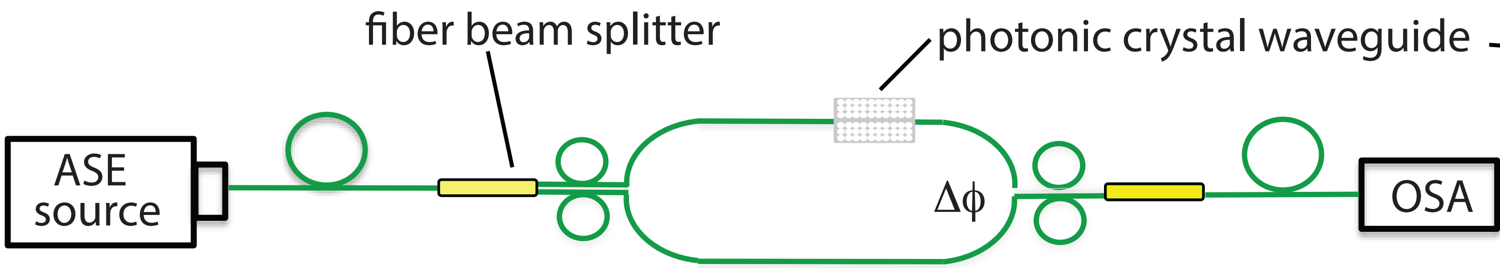
- We use line-defect waveguides in photonic crystals as our slow light mechanism

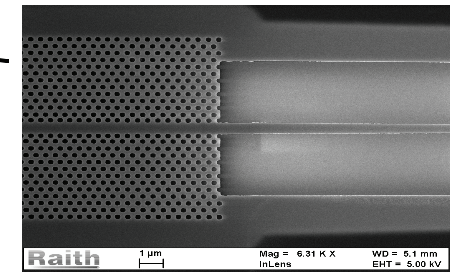
Slow-down factors of greater than 100 have been observed in such structures.



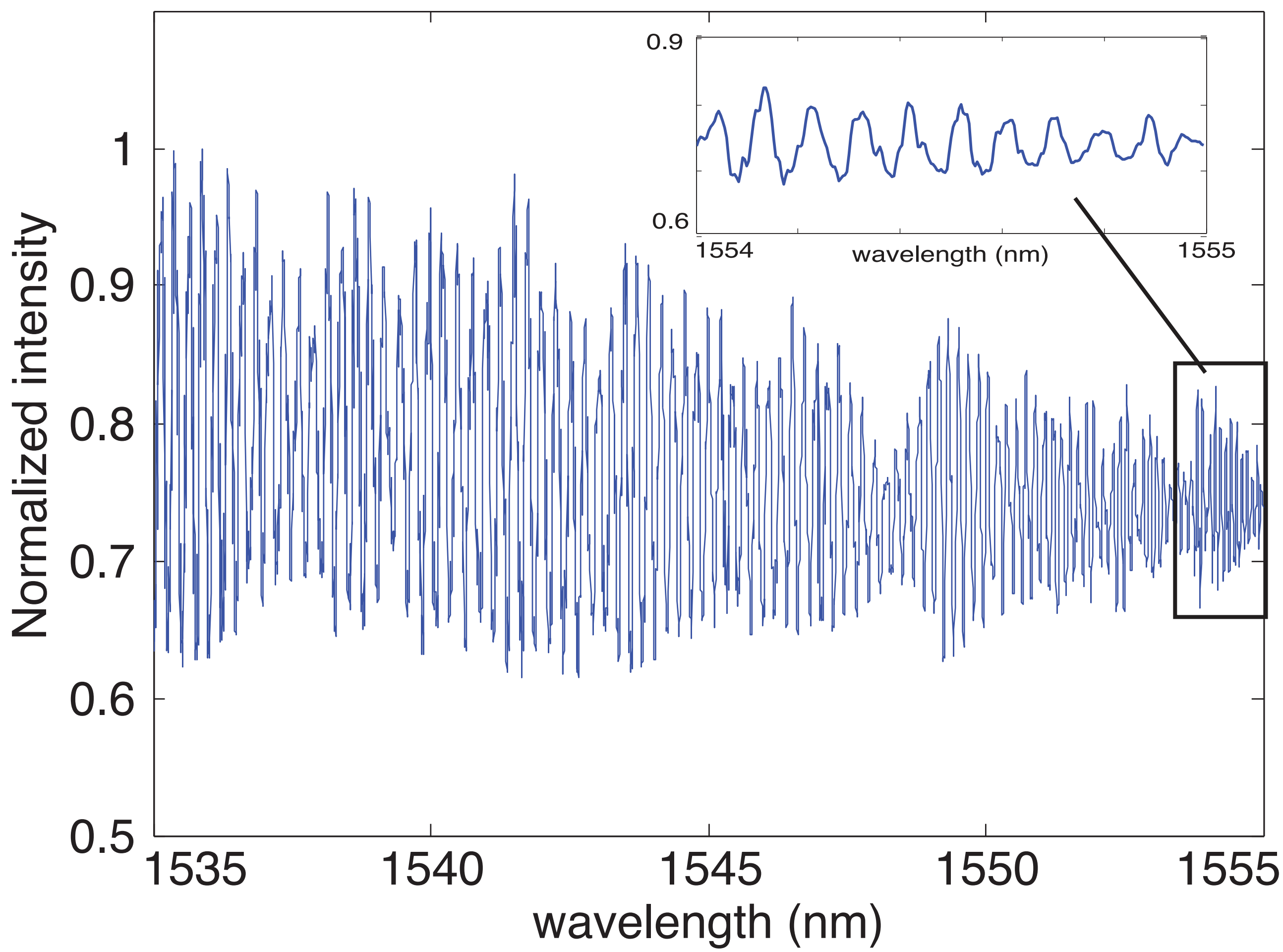

Shi, Boyd, Gauthier, and Dudley, Opt. Lett. 32, 915 (2007)
Shi, Boyd, Camacho, Vudyasetu, and Howell, PRL. 99, 240801 (2007)
Shi and Boyd, J. Opt. Soc. Am. B 25, C136 (2008).

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



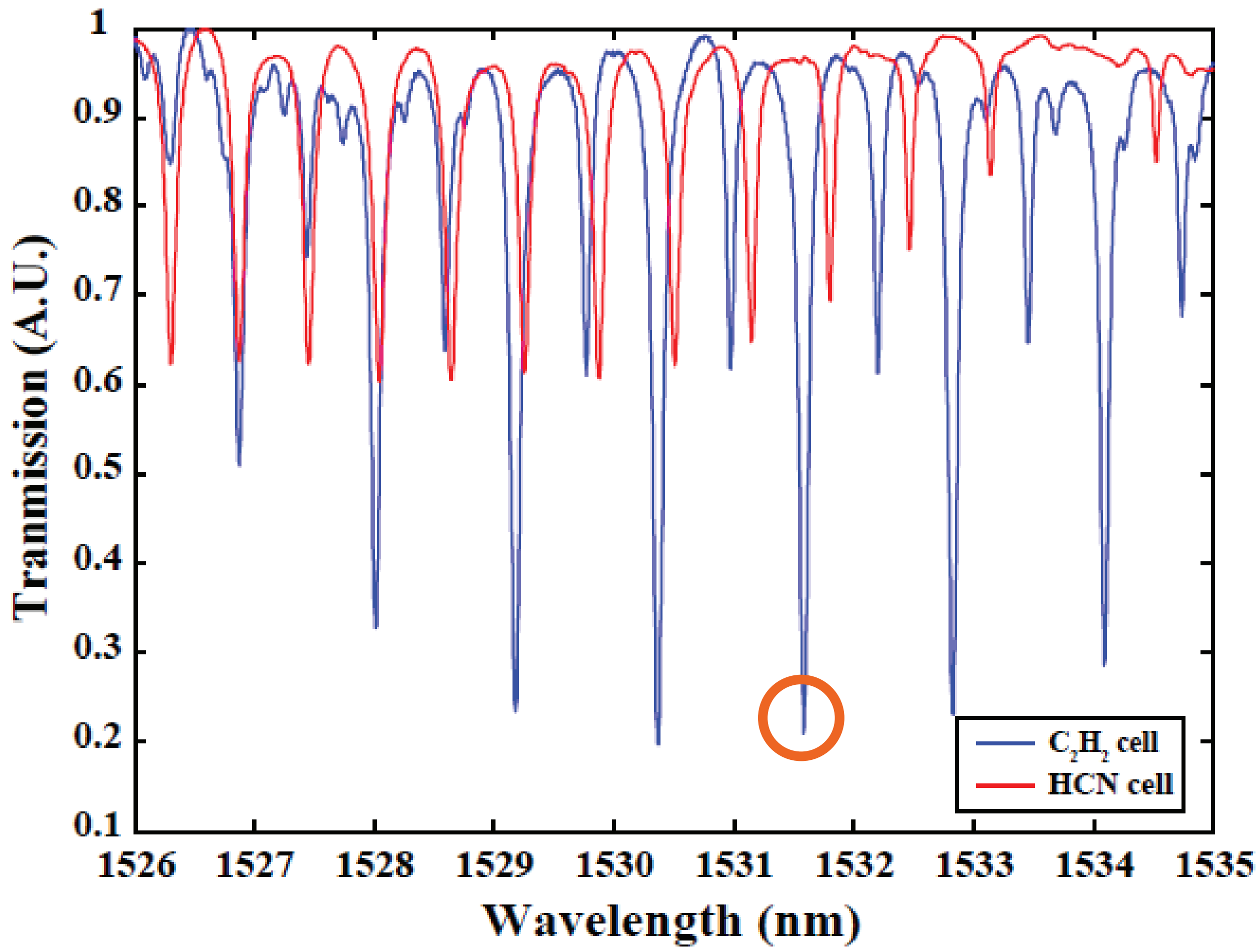

- Interference fringes




- 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, in review.

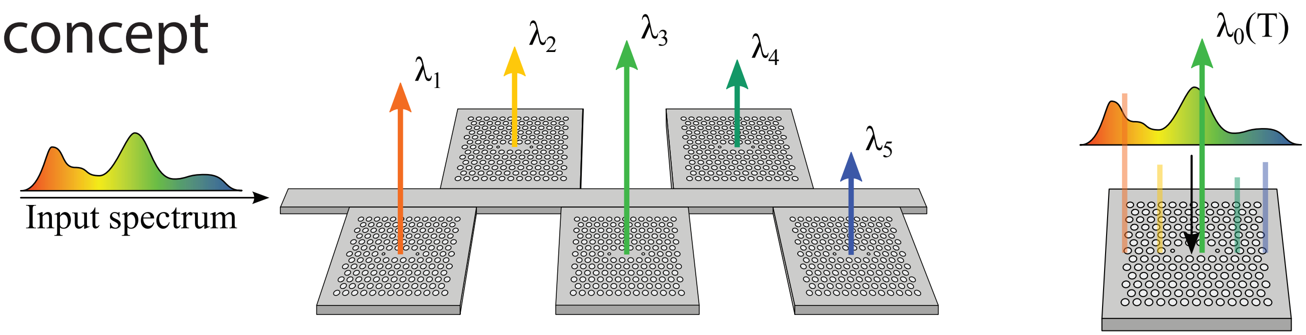
# Challenge: Fabricate a chip-scale spectrometer that can discriminate acetylene ($H_2C_2$) from hydrogen cyanide (HCN)?



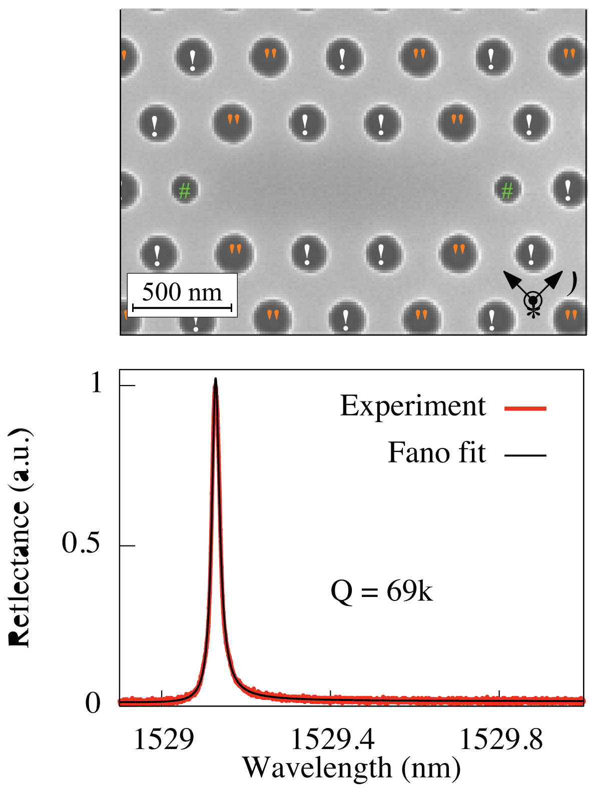

(data from our own lab)

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

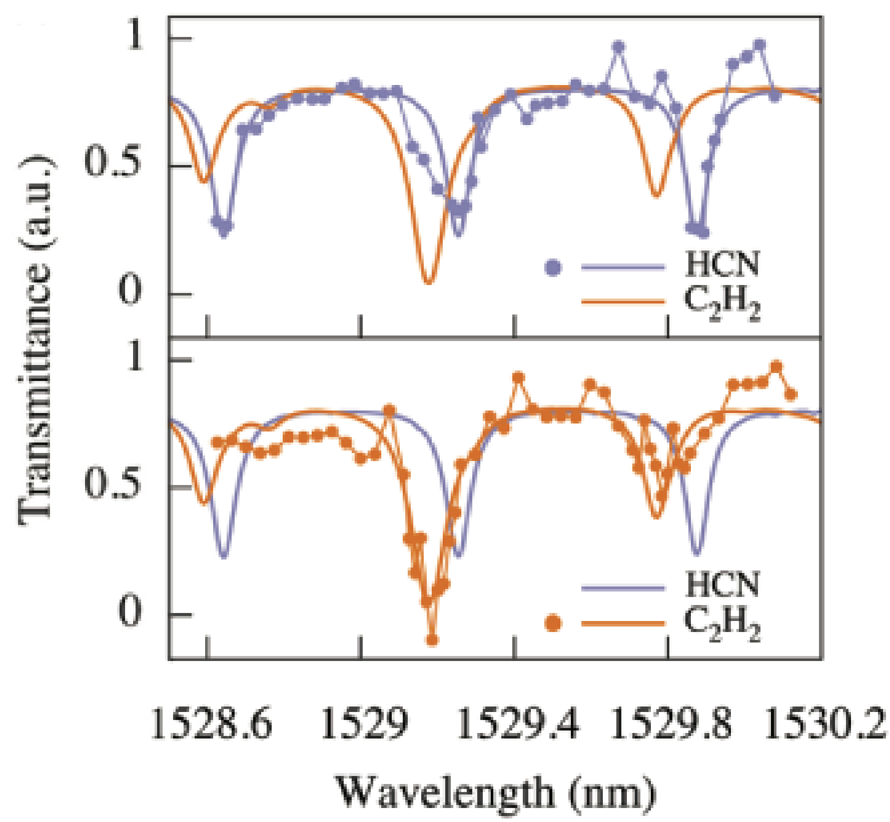
- The concept




- Cavity design




- Spectroscopy results



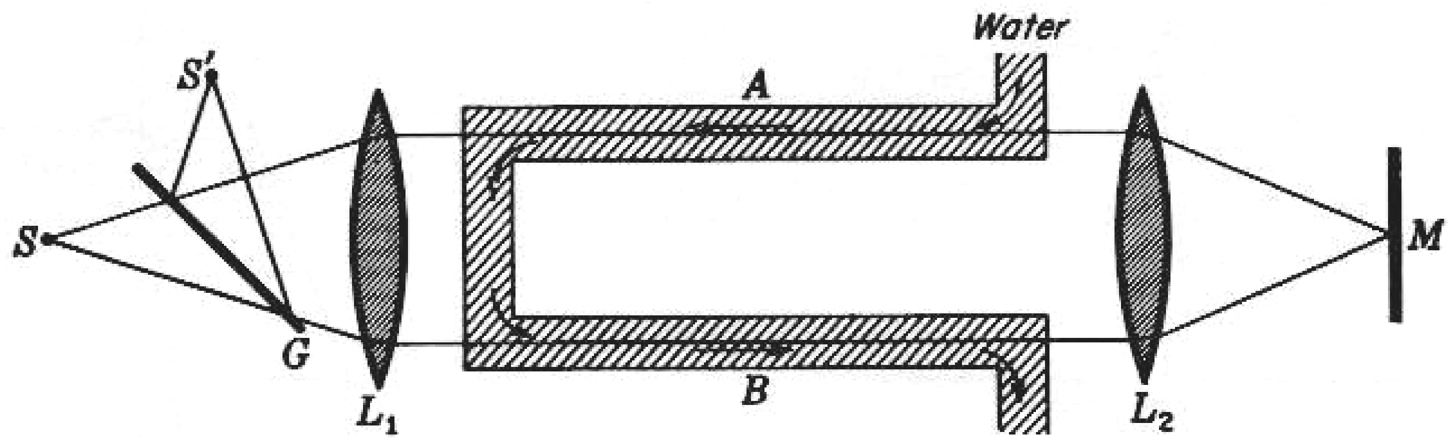

Liapis, Gao, Siddiqui, Shi, Boyd, Appl. Phys. Lett. 108, 021105 (2016).

# 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?

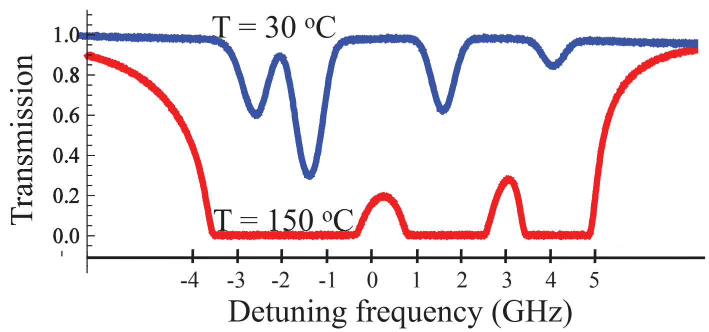
# Fresnel Drag in a Highly Dispersive Medium

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

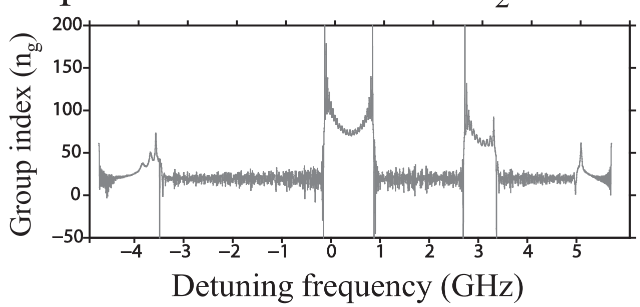
$$u \simeq \frac{c}{n} \pm v\left(1 - \frac{1}{n^2} + \frac{n_g - n}{n^2}\right)$$

**We Use Rubidium as Our Slow Light Medium**

- Transmission spectrum of Rb around $D_2$ transition:

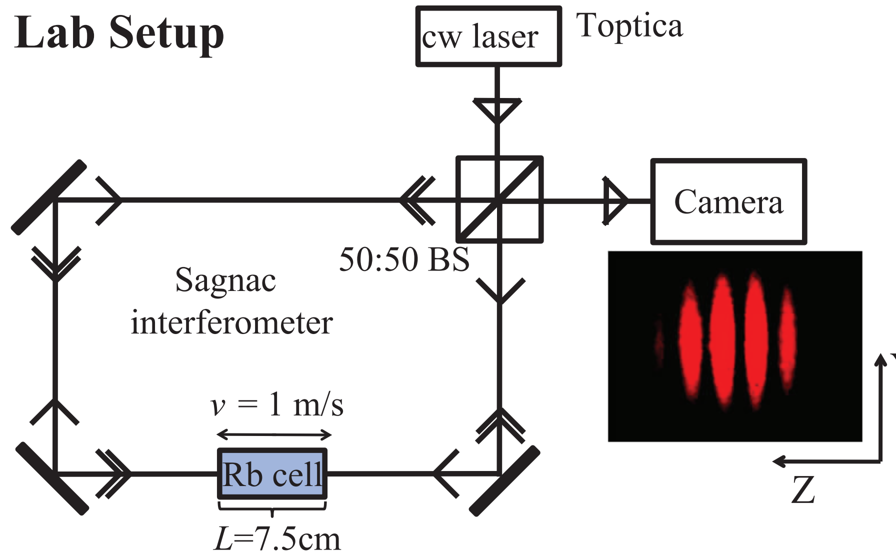


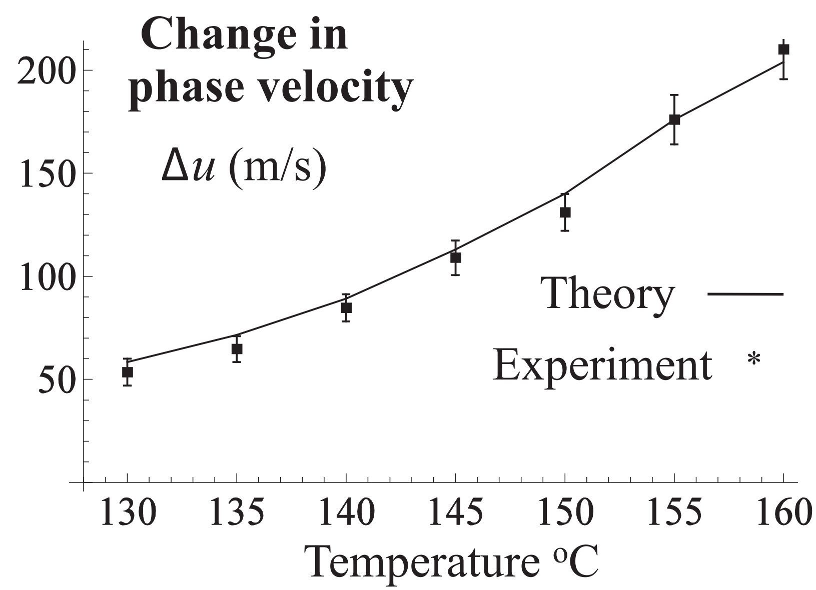

- Group index of Rb around $D_2$ line at T=130




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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# New Nonlinear Optical Material for Quantum Information Processing

- 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).

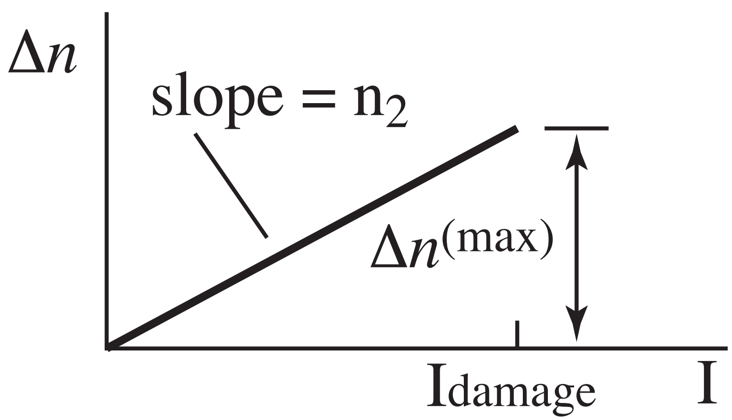
- A potential game changer for the field of photonics

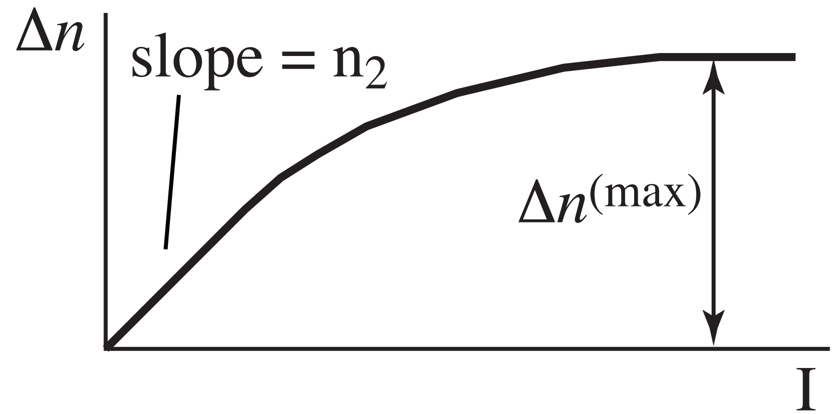
Large optical nonlinearity of indium tin oxide in its epsilon-near-zero region, M. Zahirul Alam, I. De Leon, R. W. Boyd, Science 352, 795 (2016).

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

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!
(M. Z. Alam et al., Science 10.1126/science.aae0330 2016.)

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 $\Delta n^{(max)} = 3 \times 10^{-4}$ )

# 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 $\omega_0$ is known as the epsilon-near-zero (ENZ) region.

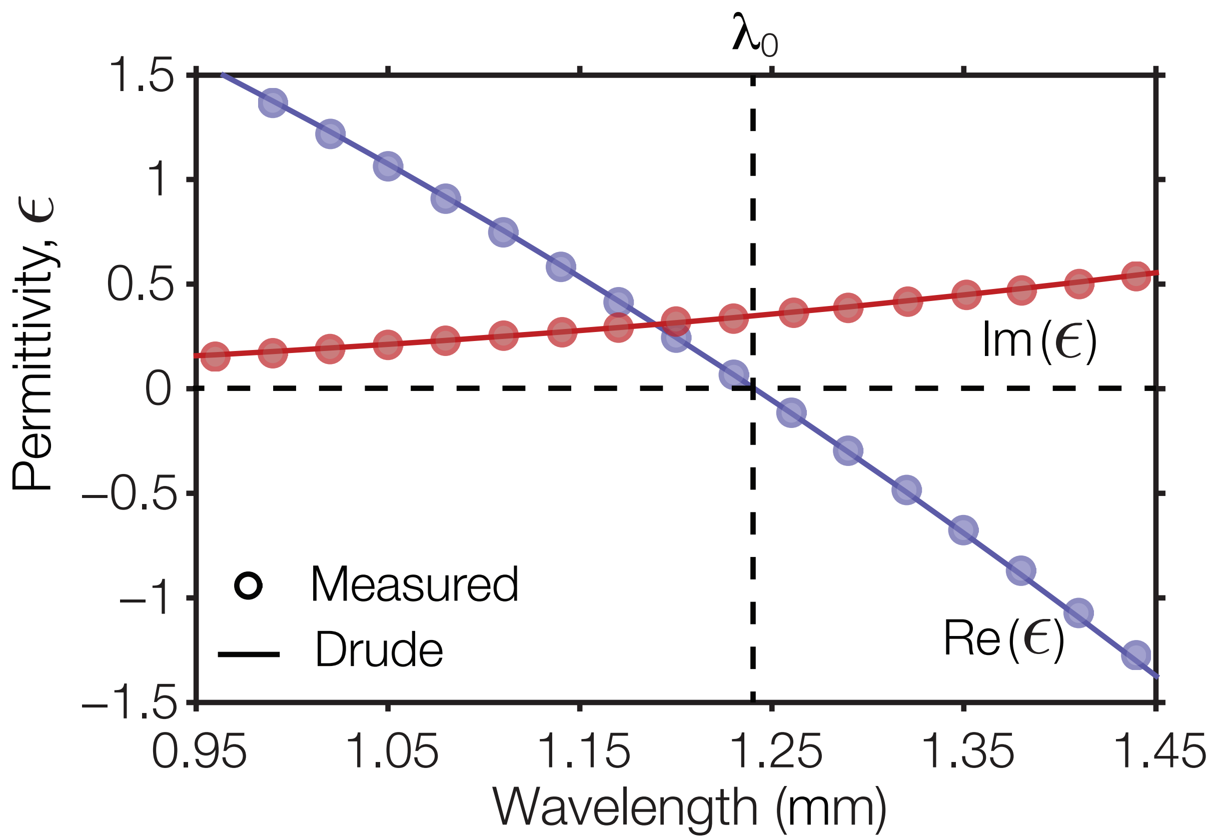
There has been great recent interest in studies of ENZ phenomena:

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

# Implications of ENZ Behavior for Nonlinear Optics

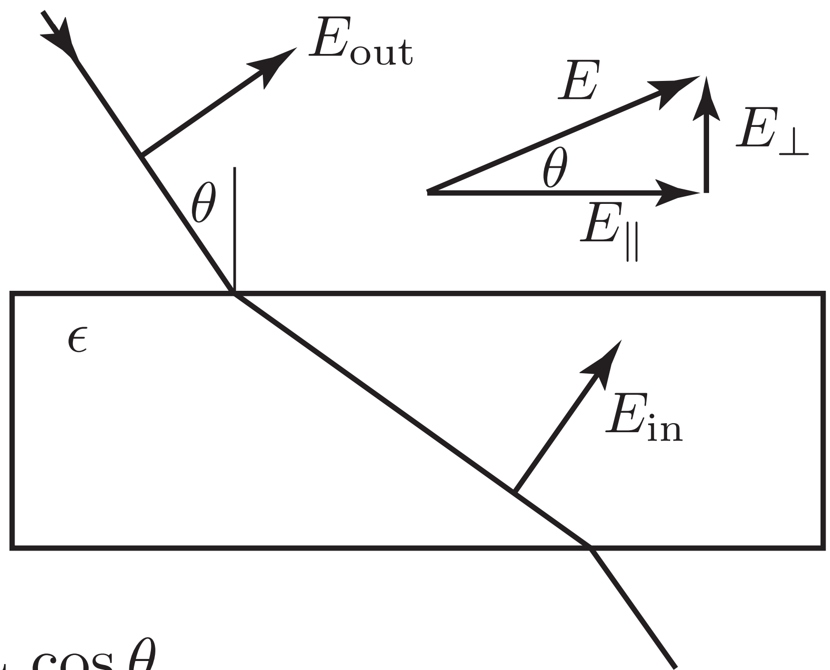
Here is the intuition for why the ENZ conditions are of interest in NLO

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

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

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

# The NLO Response Is Even Larger at Oblique Incidence




Standard boundary conditions show that:

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

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

Thus the total field inside of the medium is given by

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

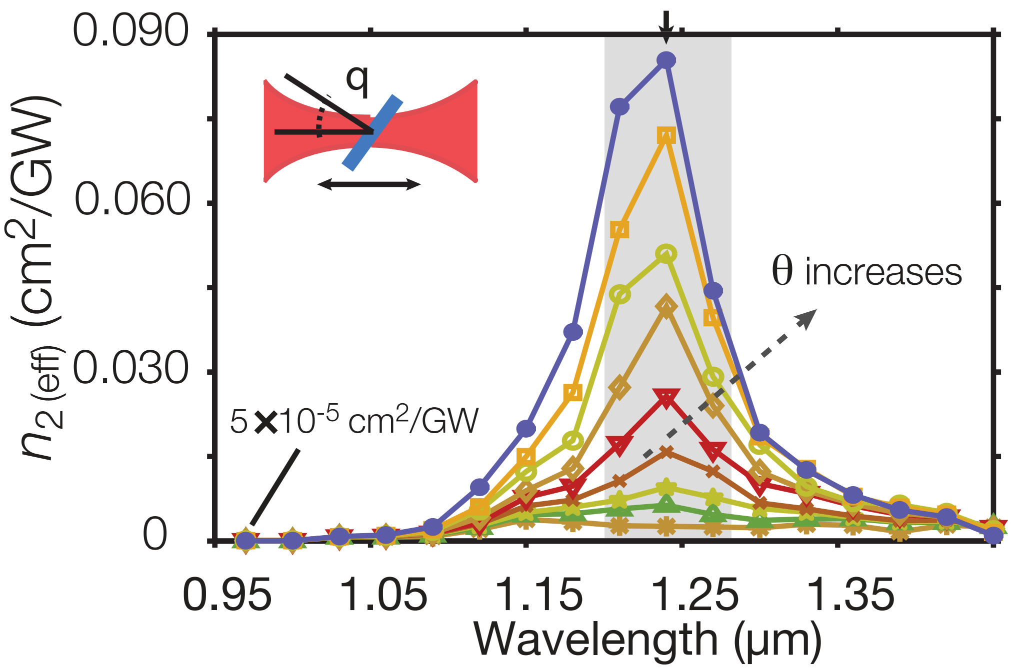
Note that, for $\epsilon < 1$, $E_{\text{in}}$ exceeds $E_{\text{out}}$ for $\theta \neq 0$.

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

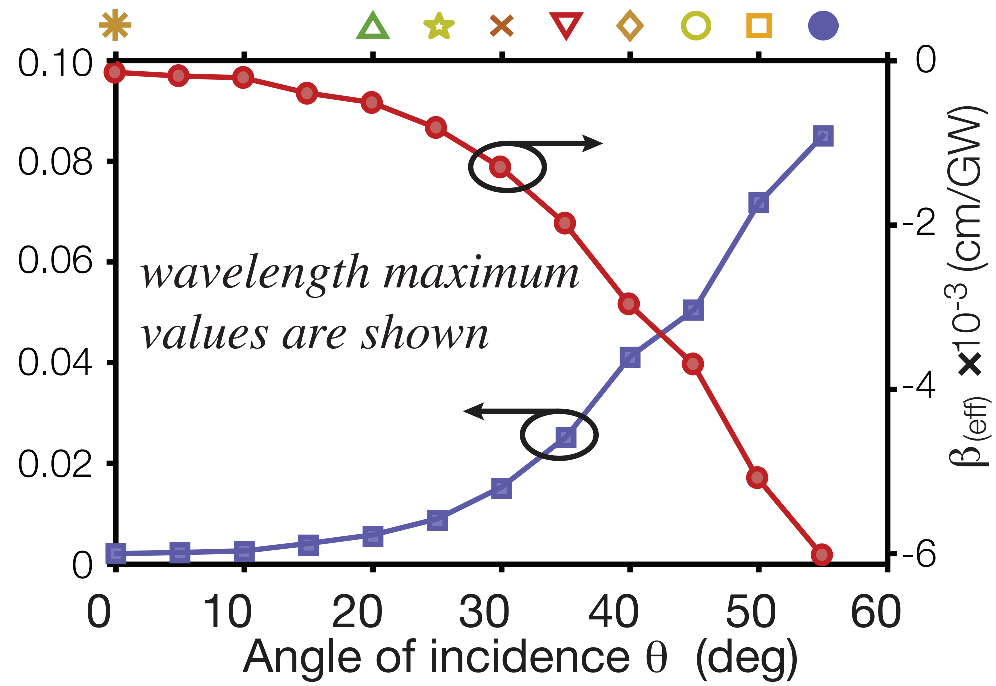
# Huge Nonlinear Optical Response of ITO

Z-scan measurements for various angles of incidence

Wavelength dependence of $n_2$

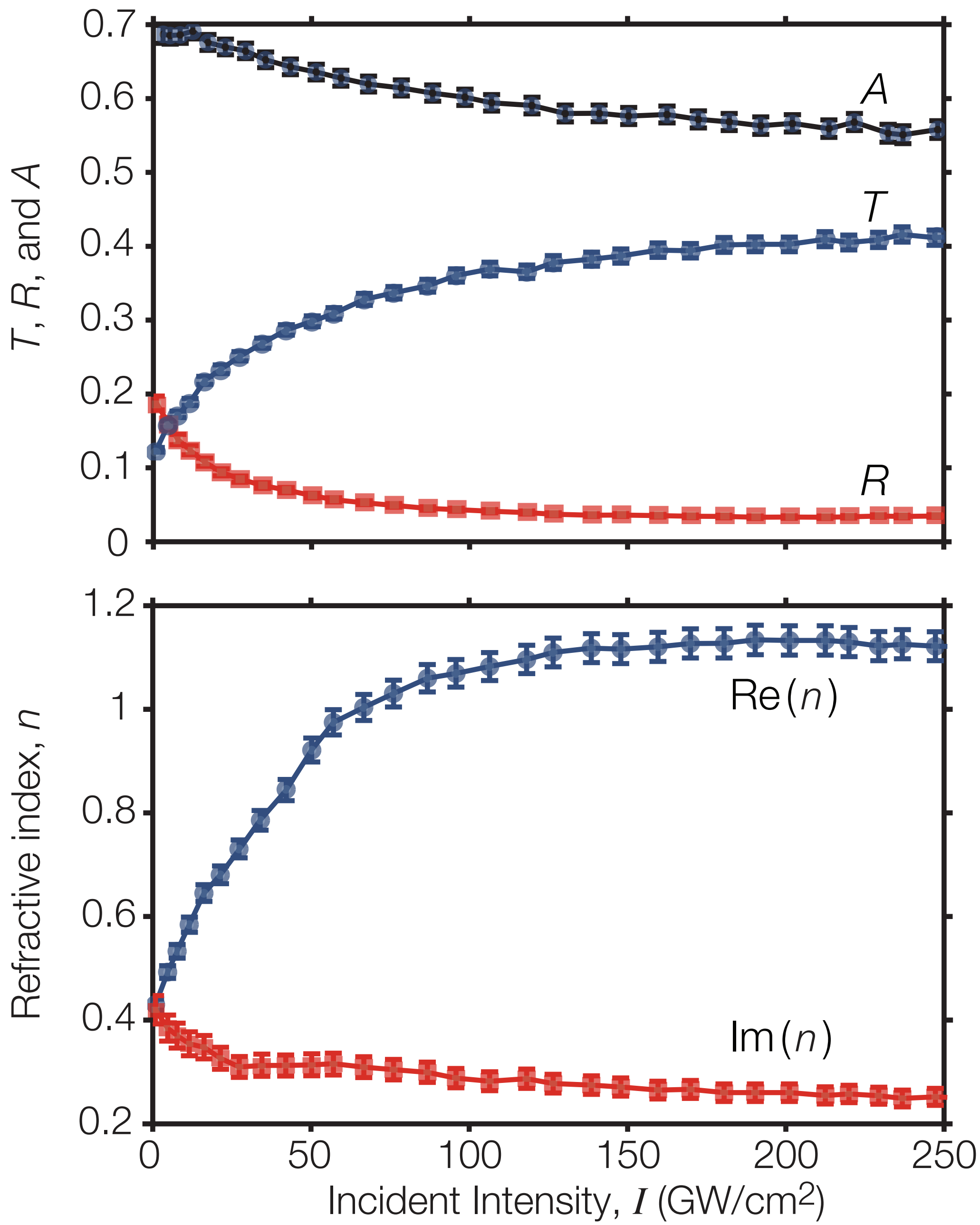



Variation with incidence angle




- 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.

# 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.

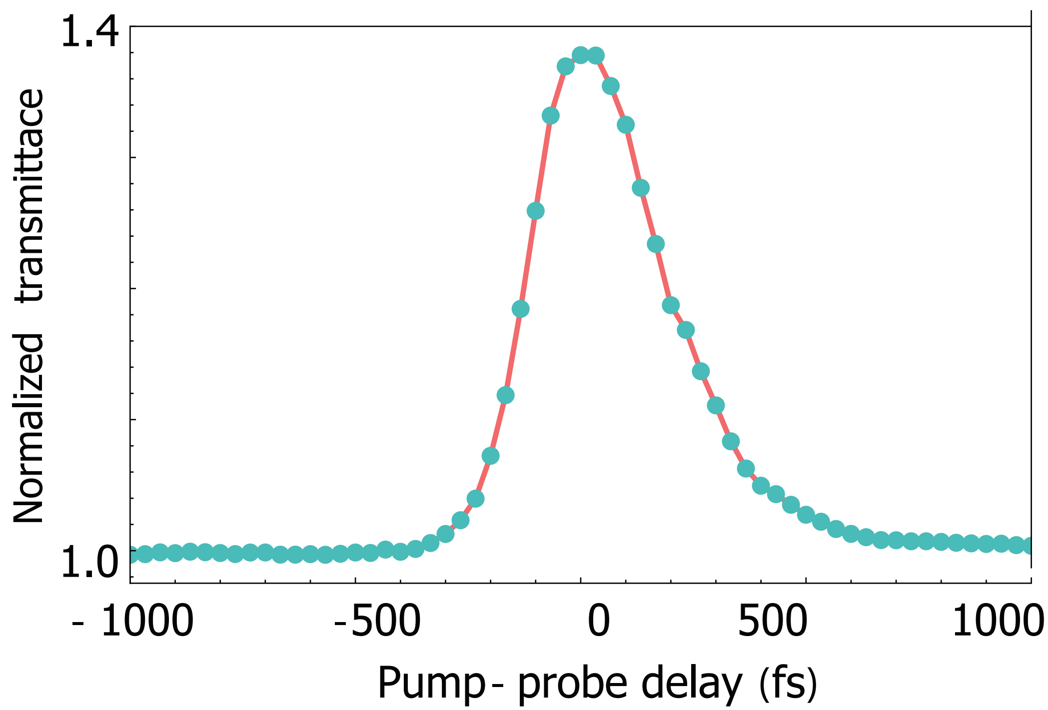
Here is the refractive index extracted from the above data.

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 $3.4 \times 10^5$ 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

# Quantum Nonlinear Optics:
# Nonlinear Optics Meets the Quantum World

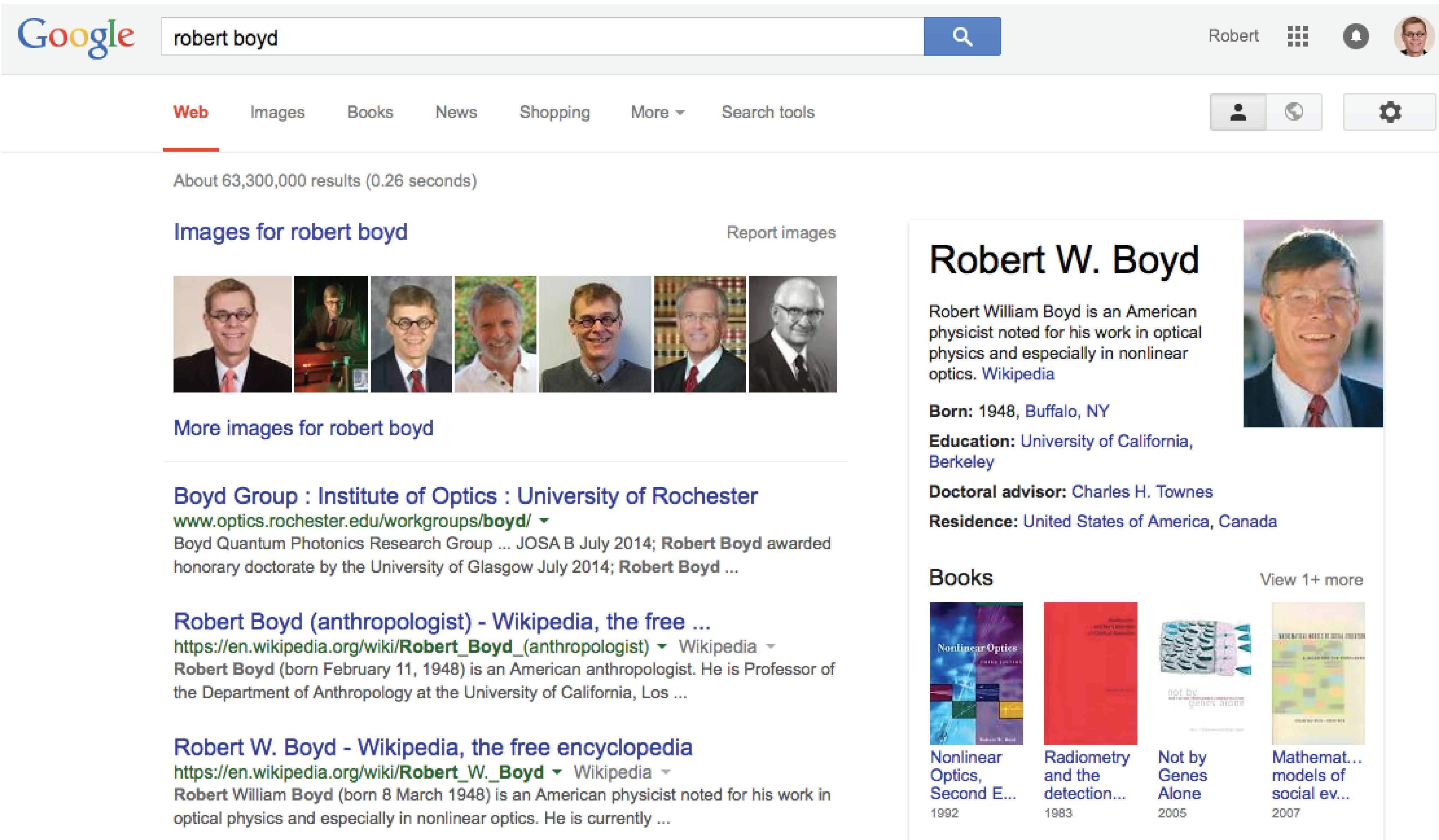
Outlook: NLO is a superb platform from which to explore new physical processes and to develop photonics applications.

**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

# Thank you for your attention!

# Why We Shouldn't Always Trust Google

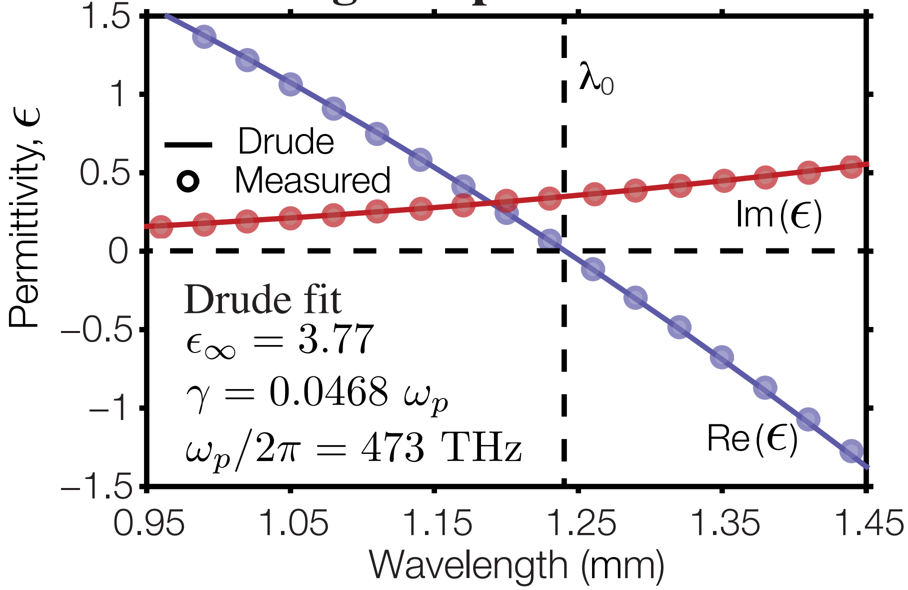


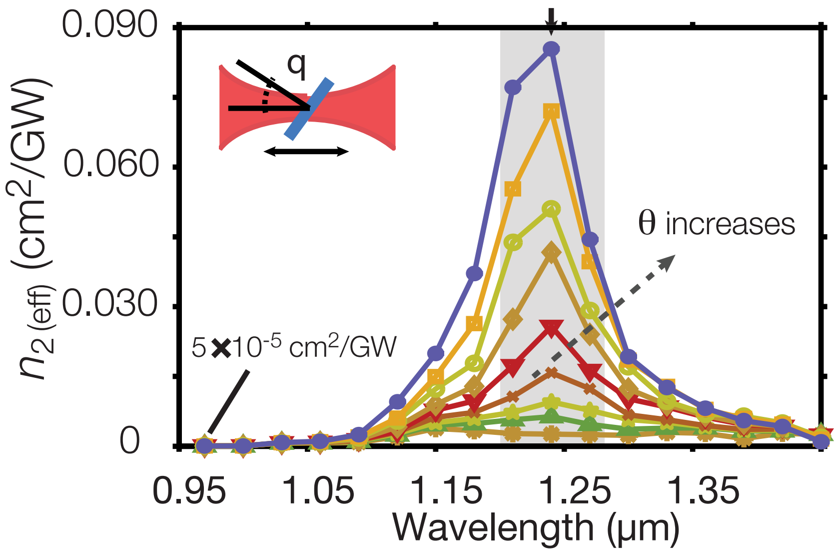
# Huge Nonlinear Optical Response of ITO near its Epsilon-Near-Zero Wavelength

Indium Tin Oxide (ITO) displays enormously strong NLO properties:

- $n_2$ is 2.5 x $10^5$ times that of fused silica
- nonlinear change in refractive index as large as 0.8
- response time of 270 fs

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




Some possible new effects

- Waveguiding outside the "weakly-guiding" regime
- Efficient all-optical switching
- No need for phase-matching

Alam, De Leon, Boyd