







# 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

Quantum Communication with Multiple Bits per Photon
 Huge Optical Nonlinearity in Epsilon-Near-Zero Materials
 New Applications of "Slow Light"

# 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 As a diagnostic, we need to be able to measure the statevector of OAM states

#### Single Photon States

Laguerre-Gaussian Basis 
$$\ell = -$$





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



### Mode Sorting

#### A mode sorter



# Sorting OAM using Phase Unwrapping

Optically implement the transformation  $\phi \rightarrow x$ 



 $e\phi$   $y\phi + x \log r - x$   $-\exp(-x) \cos(y)$ 

Position of spot determines OAM

Experimental Results (CCD images in output plane)



-Can also sort angular position states.

-Limited by the overlap of neighboring states.



\*Berkhout *et al. PRL* **105,** 153601 (2010). O. Bryngdahl, *J. Opt. Soc. Am.* **64**, 1092 (1974).



# **Our Laboratory Setup**



# 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,

# Next Step: gigabit-per-second OAM-based QKD system

• Use direct modulation of laser diode to encode at gigabits per sec.



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1. Quantum Communication with Multiple Bits per Photon

2. Huge Optical Nonlinearity in Epsilon-Near-Zero Materials

3. New Applications of "Slow Light"

- We want all-optical switches that work at the single-photon level
- We need photonic materials with a much larger NLO response
- I report a new NLO material with an n<sub>2</sub> value 100 times larger than any previously reported results (but with background absorption).

M. Z. Alam et al., Science 352, 795 (2016).

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,  $n_2 = 1.1 \times 10^{-10} \text{ cm}^2/\text{W}$  and  $\Delta n^{(max)} = 0.8$ 

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

## 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  $1.24 \mu m$ .

Recall the Drude formula

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

Note that  $\operatorname{Re} \epsilon = 0$  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



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

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  $n_2$ 

# The NLO Response Is Even Larger at Oblique Incidence



Thus the total field inside of the medium is given by

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

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

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

# Huge Nonlinear Optical Response of ITO

Z-scan measurements for various angles of incidence



- Note that n<sub>2</sub> is positive (self focusing) and  $\beta$  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<sup>2</sup>/GW = 1.1 × 10<sup>-10</sup> cm<sup>2</sup>/W at 1.25 µm and 60 deg.

The short-wavelength value of  $n_2$  of ITO is 6 x 10<sup>-5</sup> cm<sup>2</sup>/GW, which is 190 times larger that of fused silica (3.2 x 10<sup>-7</sup> cm<sup>2</sup>/GW).

There is a 43x enhancement from working at the ENZ wavelength and an additional 43x enhancement from using non-normal incidence.

Thus  $n_2 = 0.11 \text{ cm}^2/\text{GW}$ , which is 3.4 x 10<sup>5</sup> times that of fused silica.

Incidentally, for arsenic trisulfide glass,  $n_2 = 2.4 \times 10^{-4} \text{ cm}^2/\text{GW}$ . which is 750 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  $1.2 \ \mu m$ .
- Data shows a rise time of no longer than 200 fs and a recover time of of 360 fs.
- 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<sup>5</sup> 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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# **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:  $v_g = c/n_g$   $n_g = n + \omega (dn/d\omega)$
- 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



## **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.





SIL(0

# 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 wavenumbers) by the inverse of a characteristic dimension *L* of the spectrometer

Fourier-transform spectrometer



Grating spectrometer



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

- 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 spectrometers



• 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<sup>-1</sup>
- (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



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

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

• 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<sub>2</sub> 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)



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

# Boyd Name Origin



#### (Road outside Glasgow)



# Why We Shouldn't Always Trust Google



#### Images for robert boyd

Report images



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#### Boyd Group : Institute of Optics : University of Rochester www.optics.rochester.edu/workgroups/boyd/ -

Boyd Quantum Photonics Research Group ... JOSA B July 2014; Robert Boyd awarded honorary doctorate by the University of Glasgow July 2014; Robert Boyd ...

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#### Robert W. Boyd

Robert William Boyd is an American physicist noted for his work in optical physics and especially in nonlinear optics. Wikipedia

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