How Light Behaves When The Refractive Index Vanishes
Physics and Applications of Epsilon-Near-Zero materials
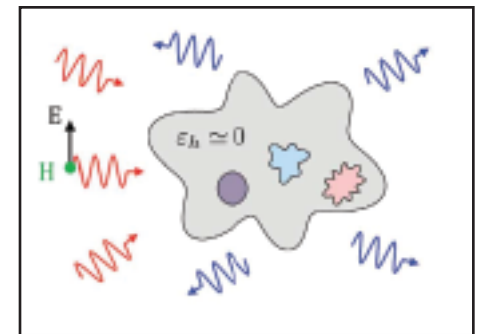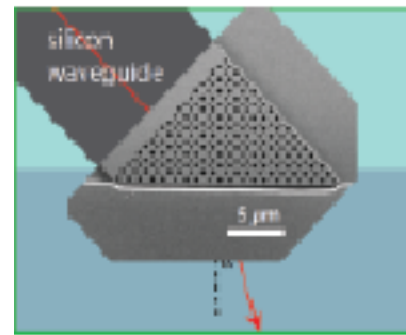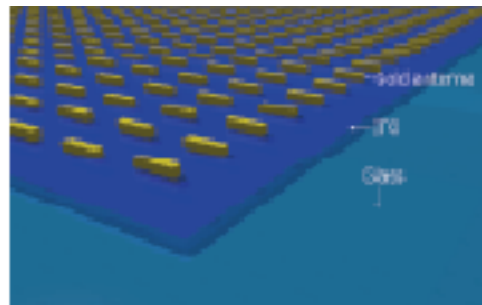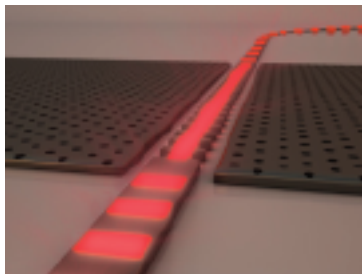
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The visuals of this talk are posted at boydnlo.ca/presentations

Presented at the Tampere University "International Day of Light" Conference, May 21, 2021.

How Light Behaves when the Refractive Index Vanishes

- Physics of Near-Zero Index (NZI) and Epsilon-Near Zero (ENZ) Materials

- Nonlinear Optical Properties of NZI and ENZ Materials

- Metamaterials for NZI and ENZ Studies

- Applications of NZI and ENZ Materials

Giant Nonlinear Response of ENZ Metastructures

- Nonlinear Optics is important for a variety of reasons:

  Photonic Devices
  - All-optical switching, buffers and routers based on slow light

  Used to create quantum states of light for
  - Quantum Computing/Communications/Imaging

  Fundamental understanding of light-matter interactions
  - Not "just" Lorentz oscillator formalism
 - Understand rogue waves
  - Control filamentation process

- However, the nonlinear response is usually much weaker than the linear response

- Means to enhance the nonlinear response

  Resonance interactions (atomic vapors)
  Plasmonic systems
  Electromagnetically induced transparency (EIT)
  Metamaterials (composite materials)

- Our approach: Use epsilon-near-zero (ENZ) materials and metamaterials

Physics of Near-Zero-Index (NZI) and Epsilon-Near-Zero (ENZ) Materials

- The wavelength of light is given by

$$\lambda = \lambda_{\mathrm{vac}}/n$$

and is significantly lenthened in a NZI material. The wavelength approaches infinity as *n* approaches zero.
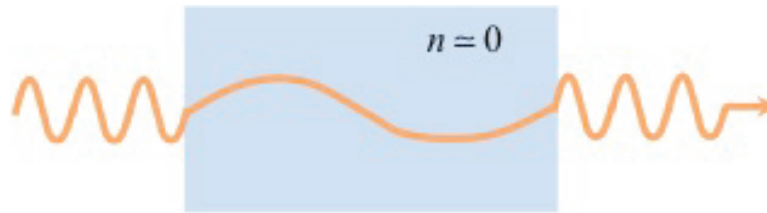
- The phase velocity of light is given by

$$v = c/n$$

and also approaches infinity as n approaches zero.

- For n approaching zero, the field oscillates in time but not in space; oscillations are in phase everywhere

Brown, Proc. IEE 100, 5 (1953).
Ziolkowski, Phys. Rev. E 70, 046608 (2004).
Silveirinha and Engheta, Phys. Rev. Lett. 97, 157403 (2006).

Some Details from Electromagnetic Theory

- The linear response of any material to electromagnetic radation can be described by

    - The dielectric permittivity (dielectric constant) ε define through the relation

    $$\mathbf{D} = \epsilon_0 \epsilon \mathbf{E}$$

    where **D**, known as the dielectric displacement, and **E**, known as the electric field, are the two fieldsthat describe the material response to an electric field.

    - The magnetic permeability μ define through the relation

    $$\mathbf{B} = \mu_0 \mu \mathbf{H}$$

    where **B,** known as the magnetic field, and **H**, known as the magnetic intensity, are the two fields that describe the magnet response of a material to an applied field.

- It is straightforward to shown from the equations of electromagnetism that

$$n = \sqrt{\epsilon \mu}$$

- Thus, *n*=0 when either ε =0 or μ=0 (or both ε and μ equal zero).

- Terminology:
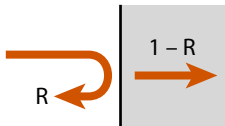   ENZ: epsilon near zero
    MNZ: mu near zero
    EMNZ: epsilon and mu near zero

Surface Reflection

- There is a problem getting light into a zero-index material.

- There is always reflection from the boundary between two materials

- The impedance and surface reflectivity are given by

$$Z = \sqrt{\mu/\epsilon} \qquad\qquad R = \left| \frac{Z-1}{Z+1} \right|^2$$

- Thus the reflectivity will be 100% if $\epsilon = 0$ unless $\mu = 0$ as well.

- This is one reason for the interest in developing EMNZ materials (epsilon and mu near zero materials.

Physics of Epsilon-Near-Zero (ENZ) Materials

- Radiative processes are modified in ENZ materials

  Einstein *A* coefficient (spontaneous emission lifetime = 1/*A*)
  $$A = n\, A_{\text{vac}}$$
  We can control (inhibit!) spontaneous emission!

  Einstein *B* coefficient
  Stimulated emission rate = *B* times EM field energy density
  $$B = B_{\text{vac}}\, / (n\, n_{\text{g}})$$
  Optical gain is very large!

  Einstein, Physikalische Zeitschrift 18, 121 (1917).
  Milonni, Journal of Modern Optics 42, 1991 (1995).

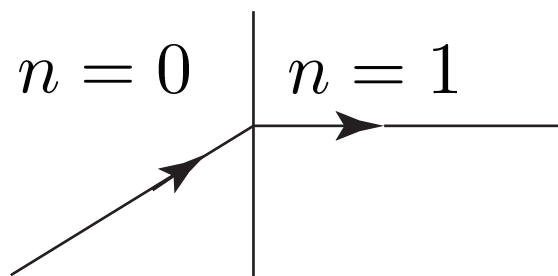 Equations are shown for nonmagnetic (μ = 1) materials

- Implications:

  - If we can inhibit spontaneous emission, we can build thresholdless lasers.
  - Expect superradiance effects to be pronounced in ENZ materials.

Optics of Zero-Index Materials
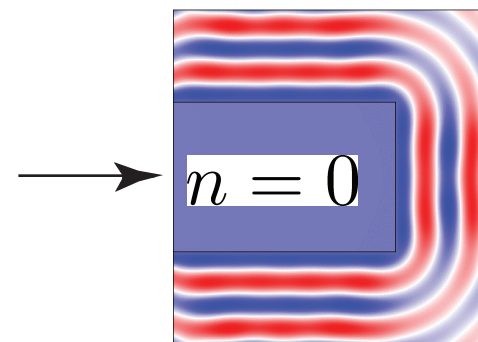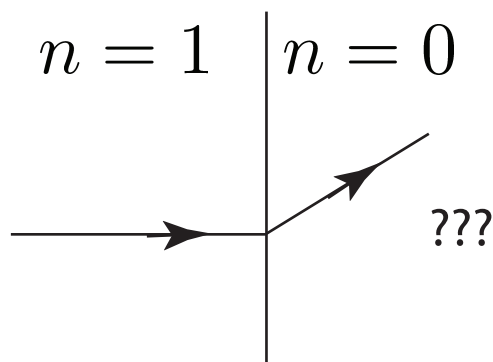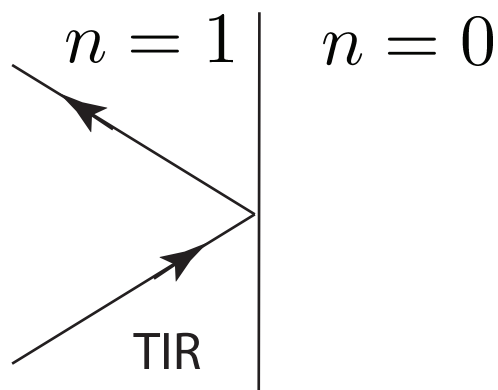
- Snell's law leads to intriguing predictions

$$n_1 \sin \theta_1 = n_2 \sin \theta_2$$

- Light always leaves perpendicular to surface of ENZ material!

$$n = 0 \quad | \quad n = 1$$

Y. Li, et al., Nat. Photonics 9, 738, 2015; D. I. Vulis, et al., Opt. Express 25, 12381, 2017.

- Thus light can enter an ENZ material only at normal incidence!

$$n = 1 \quad | \quad n = 0$$

TIR

$$n = 1 \quad | \quad n = 0$$

???

$$n = 0$$

Light enters at normal incidence but leaves in all directions.

Y. Li, et al., Nat. Photonics 9, 738, 2015.
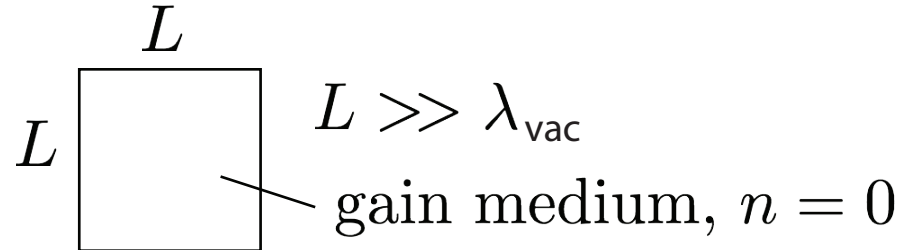
(wave-optics simulation - O. Reshef)

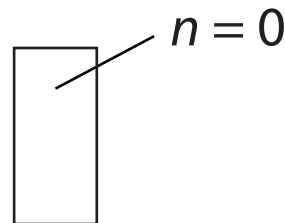# Some Consequences of ENZ Behavior - 1

- "Funny" lenses

$n = 0$

$n = 0$

A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

- Large-area single-transverse-mode surface-emitting lasers

L

L

$L \gg \lambda_{vac}$

gain medium, $n = 0$

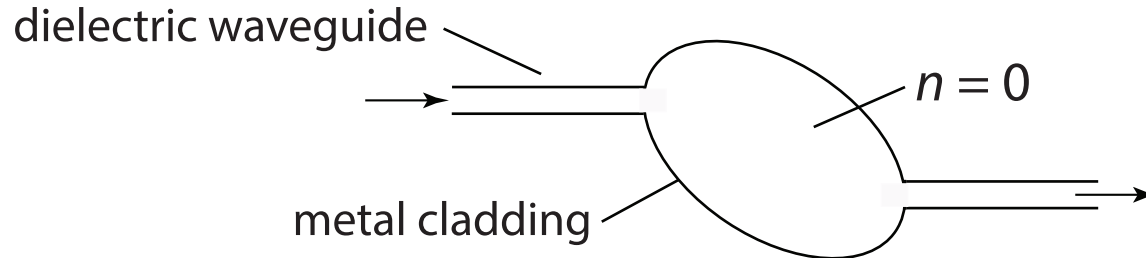J. Bravo-Abad et al., Proc. Natl. Acad. Sci. USA 109, 976, 2012.

- No Fabry-Perot interference

$n = 0$
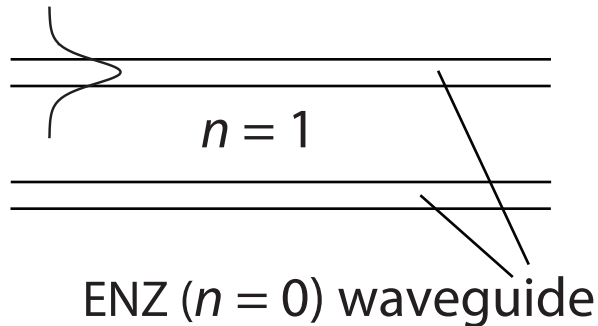
O. Reshef et al., ACS Photonics 4, 2385, 2017.

Some Consequences of ENZ Behavior - 2

- Super-coupling (of waveguides)

  dielectric waveguide

  $n = 0$

  metal cladding

  M. G. Silveirinha and N. Engheta, Phys. Rev. B 76, 245109, 2007; B. Edwards et al., Phys. Rev. Lett. 100, 033903, 2008.

- Coupling between two distant waveguides

  $n = 1$

  ENZ ($n = 0$) waveguide

  Mode of upper waveguide beams into the lower waveguide even for large separation

  Recall that k = n ω / c vanishes in an ENZ medium.

- Automatic phase matching of NLO processes

  - Recall that we need $\Delta k = 0$, but when $n=0$ $k = n\omega/c$ vanishes and so does Δk.

  - We have observed this effect in a Dirac-cone, zero-index metamaterial.

  - Usual four-wave mixing process

    in   out

  - With zero-index materials we can have

    in   out

H. Suchowski et al., Science 342, 1223, 2013.

How Light Behaves when the Refractive Index Vanishes

- Physics of Near-Zero Index (NZI) Materials

- Nonlinear Optical Properties of NZI Materials

- Meta-materials for NZI Studies

- Applications of NZI Materials

Nonlinear Optics and Optical Switching

- An important application in photonic technologies is optical switching.

control (for on/off switching)

in

out
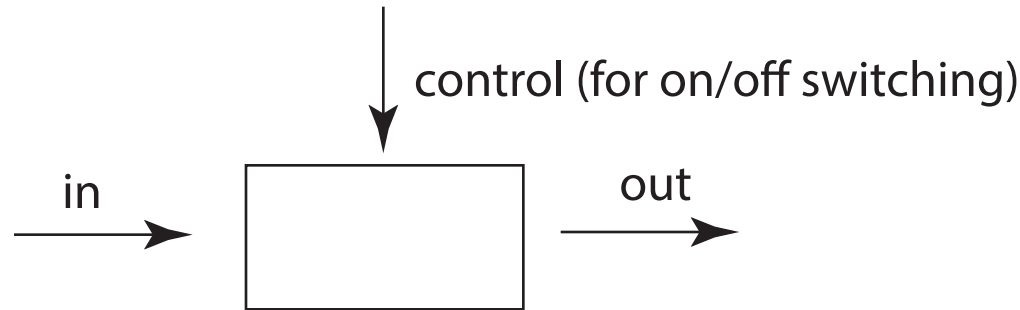
- One wants a switch with fast switching times and that operates with weak control fields.

- One needs a nonlinear interaction in order for one optical field to control another field.

- A strong nonlinear response is needed. How does one quantify the strength of a nonlinear response? Two standard methods:

$$n = n_0 + n_2 I$$

$$P^{\mathrm{NL}} = 3\chi^{(3)} |E|^2 E$$

- The nonlinear coefficients are n_2 and $\chi^{(3)}$

Implications of ENZ Behavior for Nonlinear Optics

Here is the intuition for why the ENZ condition is 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 under ENZ conditions the denominator becomes very small, leading to a very large value of n_2

Footnote:

Standard notation for perturbative NLO

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

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

Also, the refractive index changes according to

$$n = n_0 + n_2 I + n_4 I^2 + \ldots$$

How to Choose an Epsilon-Near-Zero Materials

- Electrical conductors

  All conductors display ENZ behavior at their (reduced) plasma frequency

  Recall the Drude formula

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

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

  ENZ wavelength restricted to a limited range in the visible.

- Electrical insulators (dielectrics)

  Dielectrics can show ENZ behavior at their (optical) phonon resonance.
  ENZ wavelength restricted to a limited range in the mid-IR.

- Metamaterials
  Can design the material so that the ENZ or EMNZ wavelengths are at
  any desired value.

- Challenge (for any material system). For low loss, we want Im ε as small
  as possible at the wavelength where Re ε =0.

Nonlinear Optics of Indium Tin Oxide (ITO)

- We recently reported that, at its ENZ wavelength, ITO possesses a nonlinear coefficient n_2 that is 100 times larger than those of previously reported materials [1].

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

- ITO has a large density of free electrons, and a bulk plasma frequency corresponding to a wavelength of approximately 1.24 μm.

- Dielectric properties of ITO are well described by the Drude formula.

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

- Note that aluminum-doped zinc oxide (AZO), another transparent conducting oxide, also has strong nonlinear response at its ENZ wavelength [2].

1. Alam, De Leon and Boyd, Science 352, 795–797 (2016)
2. Caspani, Shalaev, Boltasseva, Faccio et al., Phys. Rev. Lett. 116, 233901 (2016).

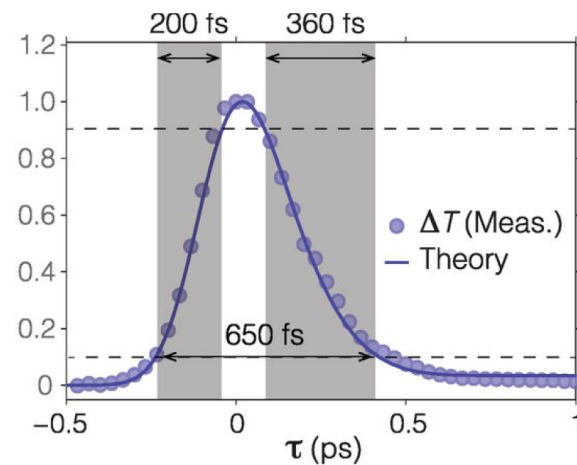# Huge, Fast NLO Response of Indium Tin Oxide at its ENZ Wavelength

- ellipsometry

$\lambda = 1240$ nm
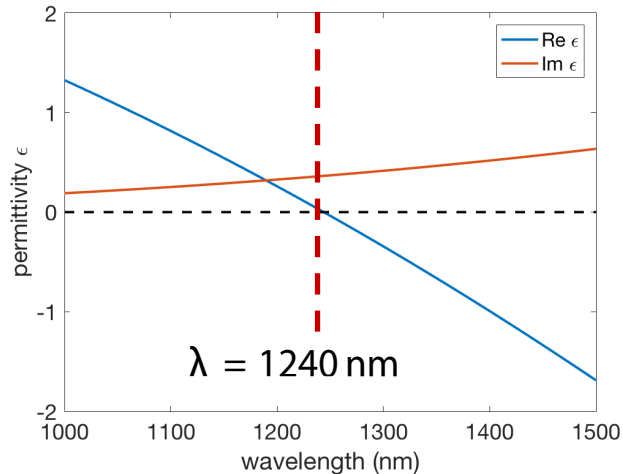
- n_2 can be 3.4 x10^5 times larger than that of silica glass

chalcogenide glass
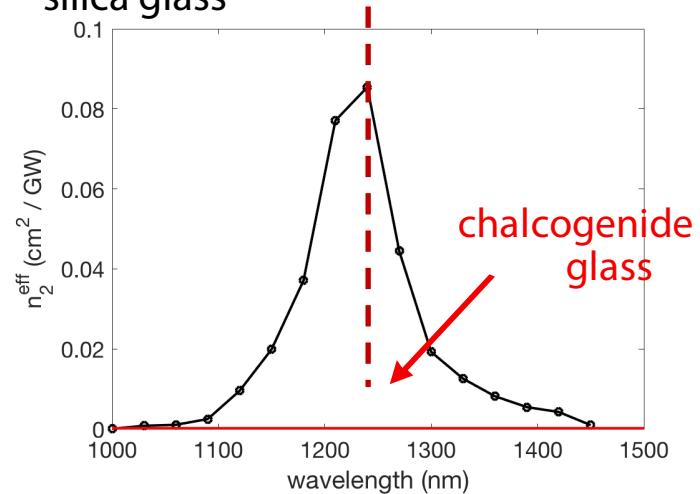
- overall change in refractive index of 0.8
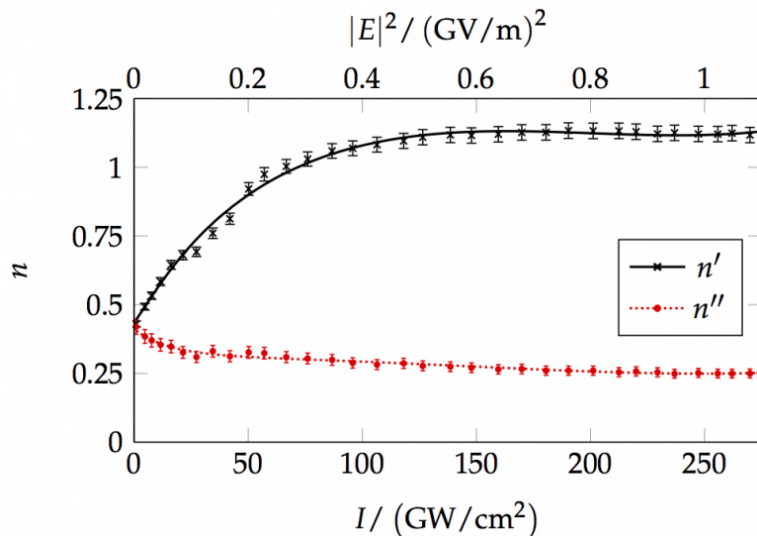
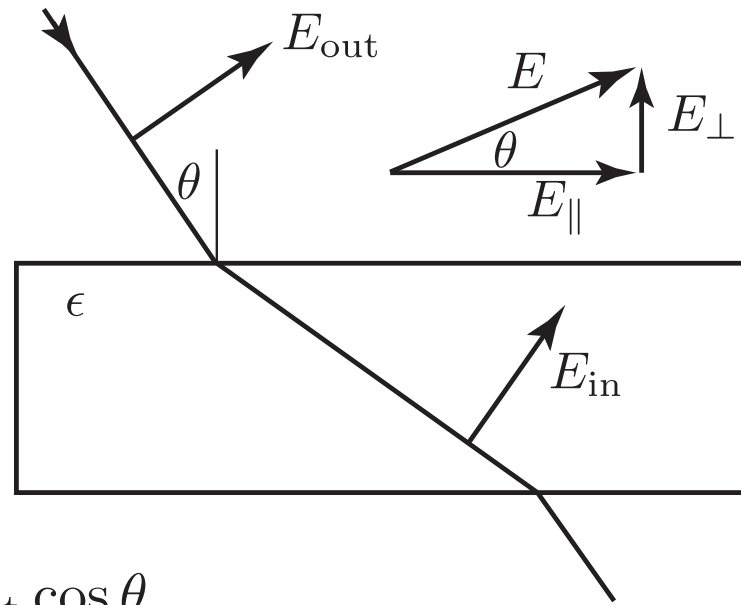

- sub picosecond reponse time

M. Z. Alam et al., Science 352, 795-797 (2016)

The NLO Response Is Larger For 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_{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

Wavelength dependence of n_2 — $n_{2\,(eff)}$ (cm²/GW) vs Wavelength (µm); θ increases; 5×10^{-5} cm²/GW
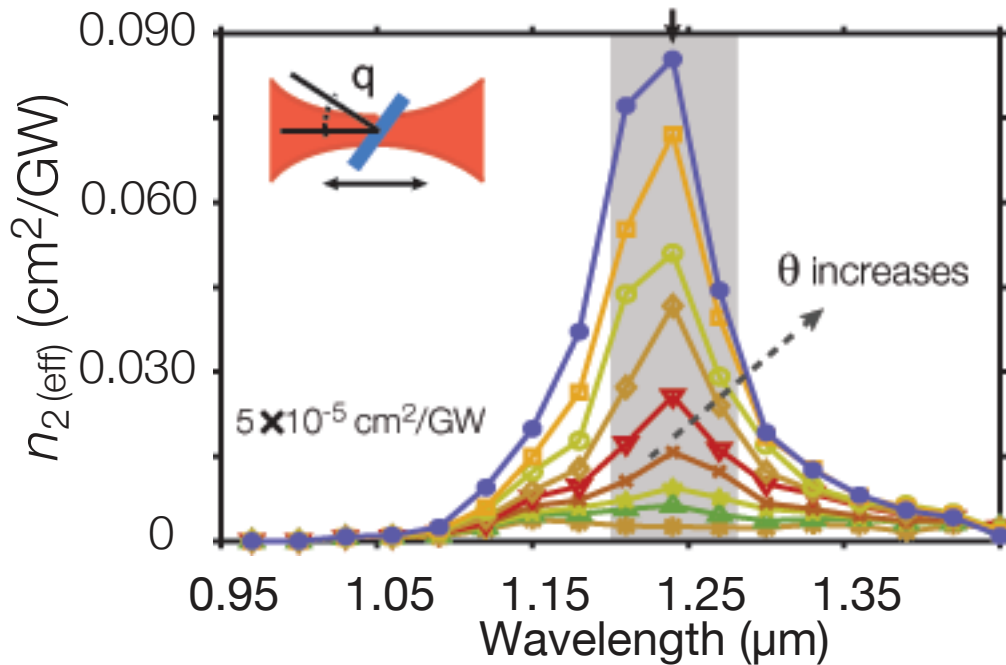
Variation with incidence angle — wavelength maximum values are shown; $\beta_{(eff)} \times 10^{-3}$ (cm/GW) vs Angle of incidence θ (deg); peak laser intensity was 50 GW cm^{-2}
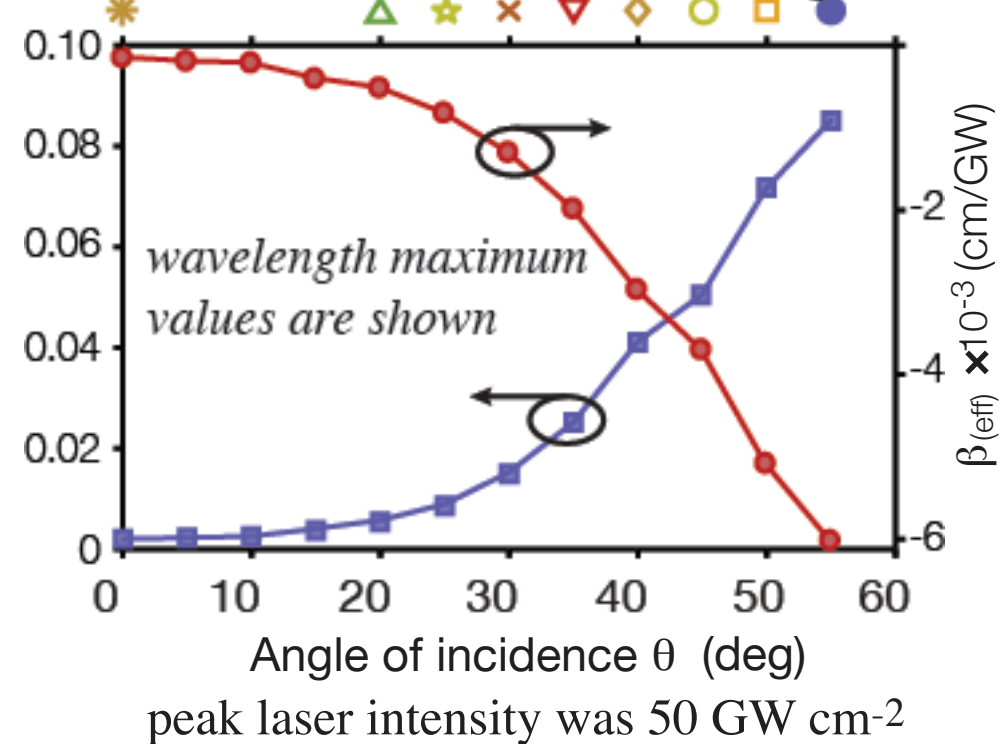
- Note that n_2 is positive (self focusing) and β is negative (saturable absorption).

- Both n_2 and nonlinear absorption increase with angle of incidence

- n_2 shows a maximum value of 0.11 cm²/GW = 1.1×10^{-10} cm²/W at 1.25 µm and 60 deg. This value is 2000 times larger than that away from ENZ region.

Why is n_2 so large for ITO?

The short-wavelength (away from the ENZ resonance) value of n_2 of ITO is 5×10^{-5} cm^2/GW, which is 150 times larger that of fused silica (3.2×10^{-7} cm^2/GW).

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

Thus $n_2 = 0.01$ cm^2/GW, which is 3.4×10^5 times that of fused silica.

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

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

Why Does ENZ Lead to Large NLO Response?

1. From form of n_2
$$n_2 = \frac{3\chi^{(3)}}{4\epsilon_0 c\, n_0\, \mathrm{Re}(n_0)}$$

2. From simple math: $n = n_b + \Delta n$ and $\epsilon = \epsilon_b + \Delta\epsilon$

$$n = \sqrt{\epsilon_b + \Delta\epsilon} \approx \sqrt{\epsilon_b}\left(1 + \frac{\Delta\epsilon}{2\epsilon_b}\right) = n_b + \frac{\Delta\epsilon}{2n_b} \quad \text{and thus} \quad \Delta n = \frac{\Delta\epsilon}{2n_b}$$

3. Note behavior of wave equation for $\epsilon = 0$

$$\nabla \times \nabla \times \mathbf{E} + \frac{\epsilon\mu}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E} = -\mu\frac{\partial^2 \mathbf{P}^{\mathrm{NL}}}{\partial t^2}$$

4. From Maxwell's equations, it is easy to show that the nonlinear response scales as

$$\frac{\left.\frac{dH_x}{dz'}\right|_{\mathrm{nl}}}{|H_x|} \propto \sqrt{\frac{\mu_r}{\epsilon_r}}$$

5. Detailed numerical integration confirms this behavior.

Dependence of Second-Harmonic Generation on the Linear Dielectric Permittivity

- We solve the standard equations for second-harmonic generation

$$\frac{dA_1}{dz} = i\frac{\eta_1\omega_1\chi^{(2)}}{c}A_2(z)A_1^*(z)e^{-i\Delta kz},$$

$$\frac{dA_2}{dz} = i\frac{\eta_2\omega_2\chi^{(2)}}{2c}A_1^2(z)e^{i\Delta kz},$$

- We take Δk = 0 and plot the solution for various values of the permittivity ε.

- We find that the growth rate increases dramatically as the permittivity is decreased.

$A_1(0) = 5 \times 10^8$ [V/m]

$\chi^{(2)} = 5 \times 10^{-12}$ [m/V]

Solís, Boyd and Engheta, to be published
See also Solís, CLEO Poster JW2D.15
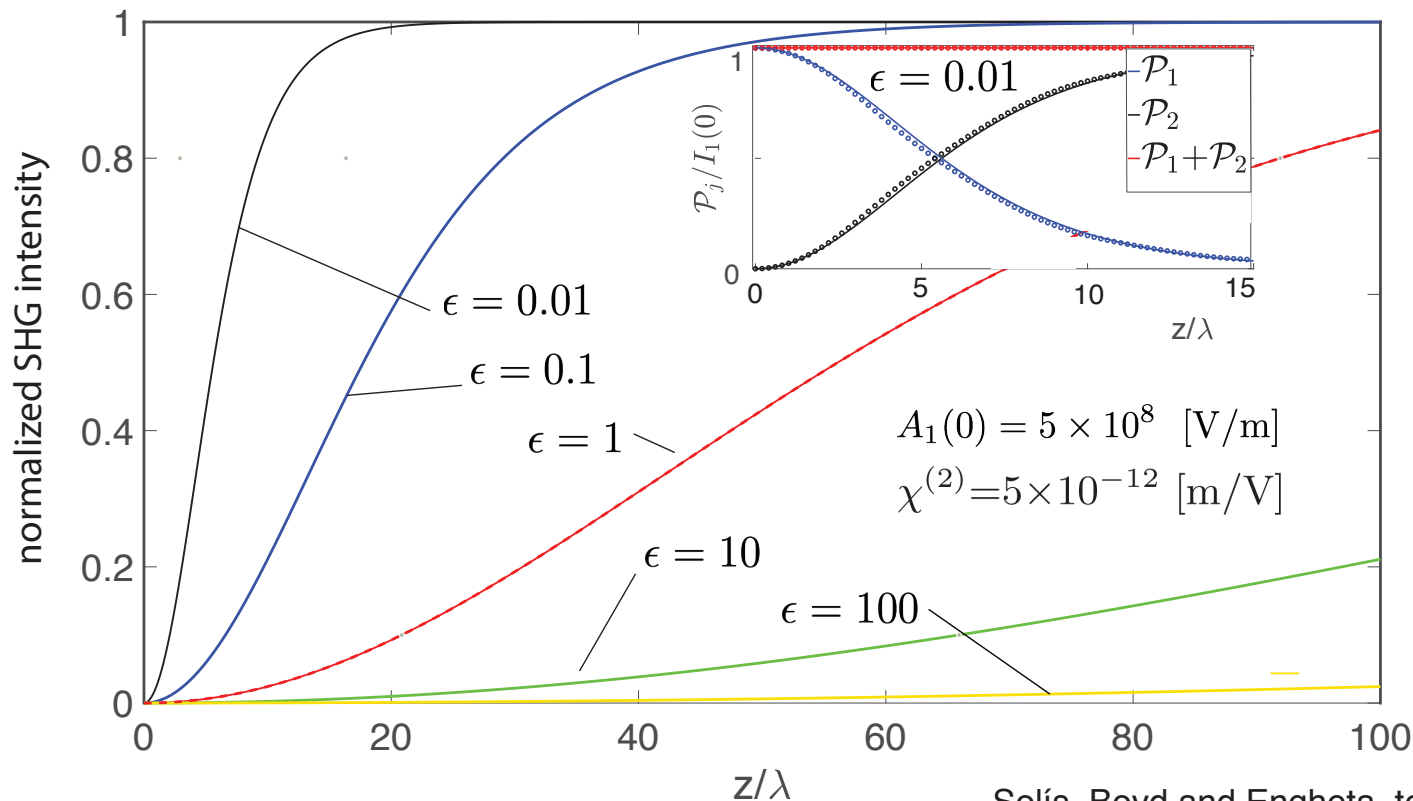
An ENZ Metasurface

- We functionalize ITO by creating a photonic metasurface

- We obtain an even larger NLO response by placing a gold antenna array on top of ITO.

  - Lightning rod effect: antennas concentrate the field within the ITO

  - Coupled resonators: ENZ resonance and nano-antennas

Concept:

SEM:

600 nm
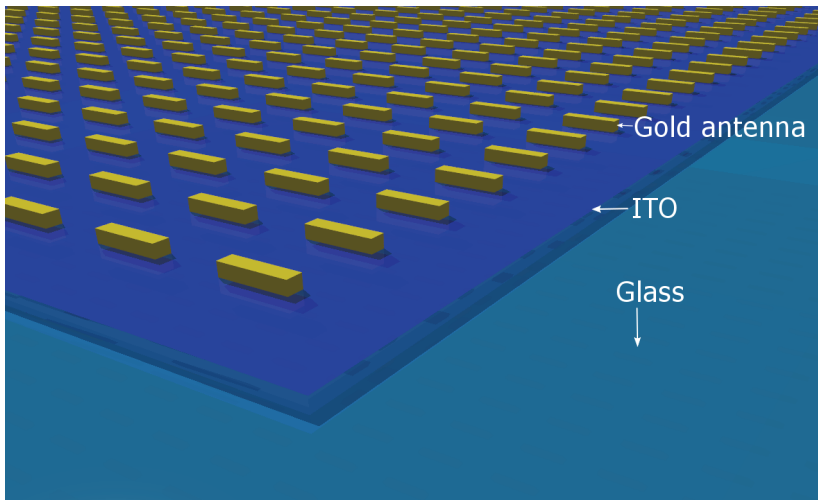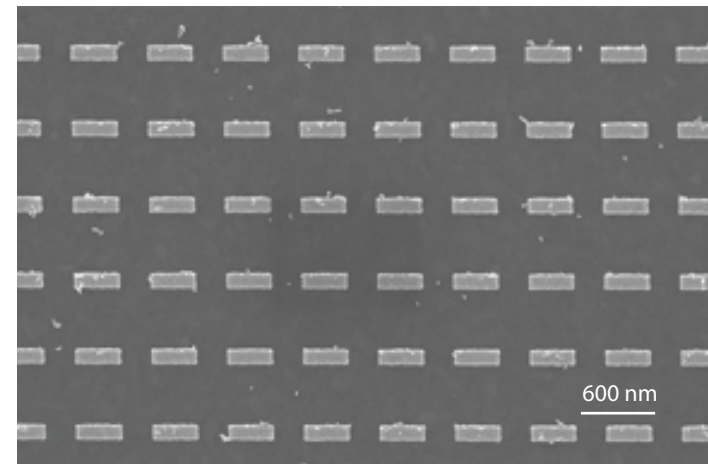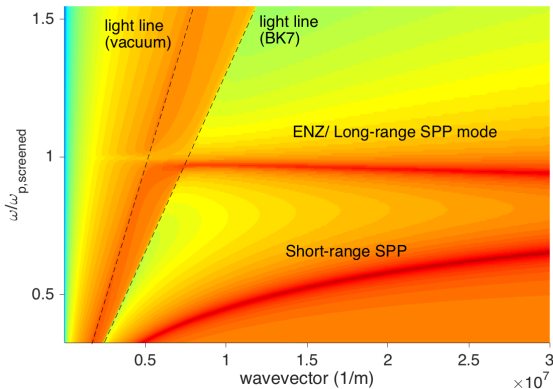
A thin layer of ITO supports two modes
  • the bulk plasma mode, also called the ENZ or long-range SPP mode
  • the short range surface plasmon polariton (SPP) mode

NLO response of the coupled antenna-ENZ system
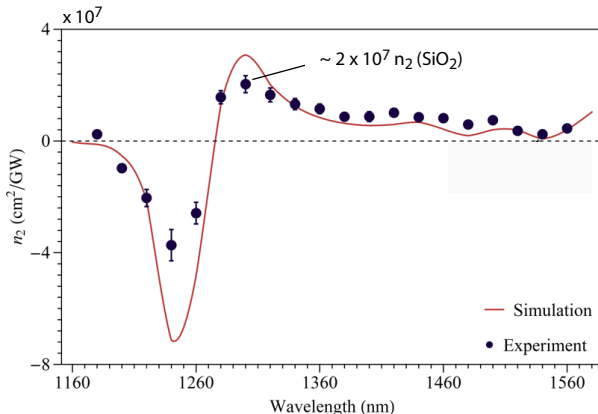
The structure exhibits and extremely large n2 value over a broad spectral range. The on-resonance n2 value is seven orders of magnitude larger than that of silica glass.

Alam, Schulz, Upham, De Leon and Boyd, Nature Photonics 12, 79-83 (2018).

Physics and Applications of Epsilon-Near-Zero Materials

- Physics of ENZ Materials

- Huge NLO Response of ITO and ITO Metastructures

- Materials for ENZ

- Applications of ENZ Materials

Epsilon-Near-Zero (ENZ) and Near Zero-Index (NZI) Materials

Homogeneous materials

TCO
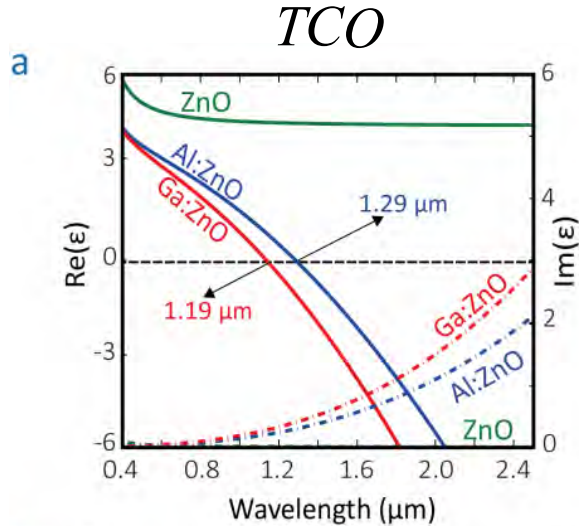
A. Boltasseva (Purdue)
Kim et al., Optica (2016)

SiC
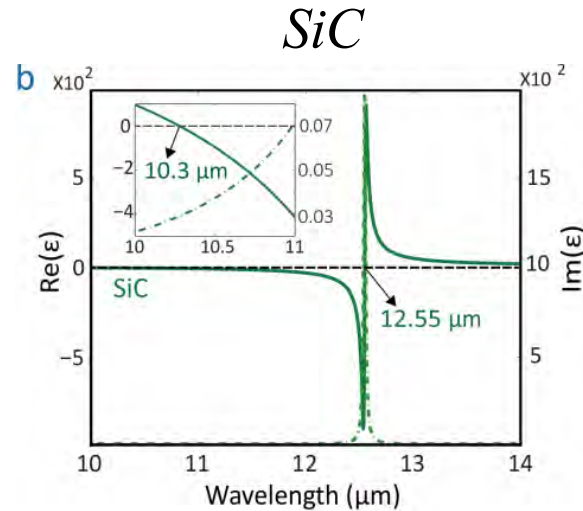
J. Caldwell (Vanderbilt)
Kim et al., Optica (2016)

$Bi_{1.5}Sb_{0.5}Te_{1.8}Se_{1.2}$
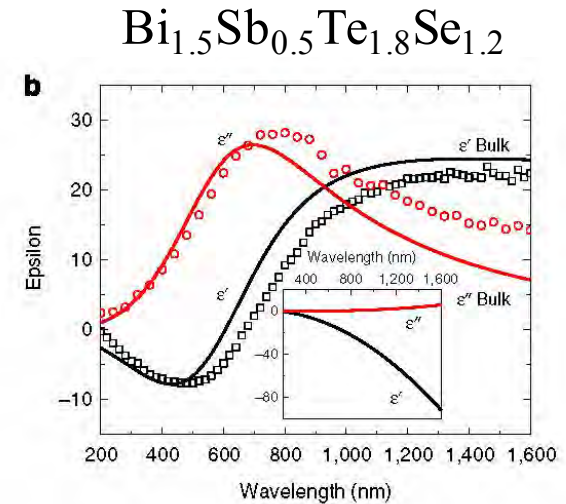
N. Zheludev (Southmapton)
Ou et al., Nat. Commun. (2014)

Metamaterials
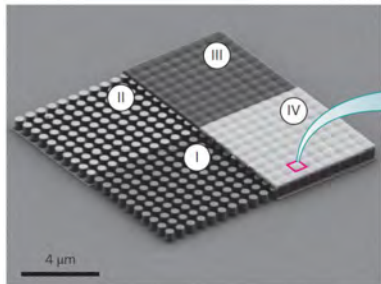
E. Mazur Li et al.,
Nat. Photon. (2015)
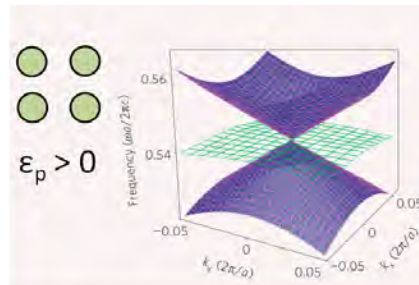
Chan, Huang et al.,
Nat. Mater. (2011)
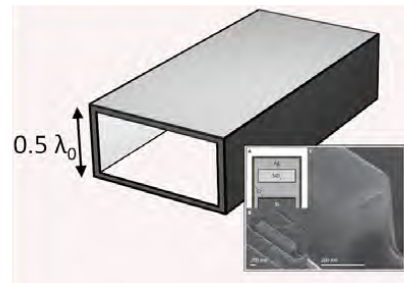
SEM from : Polman's & Engheta's
Vesseur et al., PRL (2013)
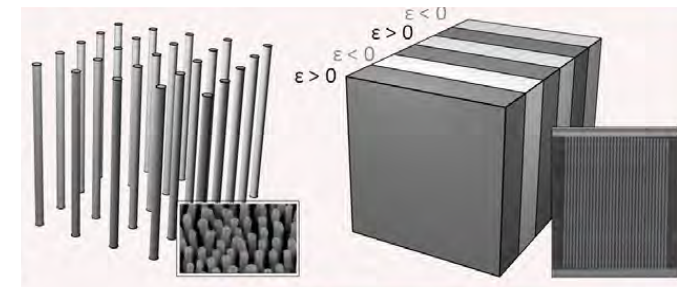
$Re(\varepsilon) \cong 0$

Wire SEM from : Zayat & Podolskiy
Pollard et al., PRL (2009)
Stack SEM from : Polman & Engheta
Mass et al., Nat. Photon. (2013)

Giant Nonlinear Response of ENZ Metastructures: Our Team

Nader Engheta
• H. Nedwill Ramsey Professor at the University of Pennsylvania
• B.S. degree from the University of Tehran and his M.S and Ph.D. from Caltech.
• Activities include ENZ, photonics, metamaterials, nano-optics, graphene optics,
   electrodynamics, microwave and optical antennas, studies of fields and waves.
• Many awards including the Streifer Award of IEEE and the Gold Medal from SPIE

Eric Mazur
• Balkanski Professor of Physics and Applied Physics at Harvard University
• Ph.D. University of Leiden.
• Activities include light-matter interactions with ultrashort laser pulses, nonlinear optics
   at the nanoscale, and zero-index dielectric metamaterials.
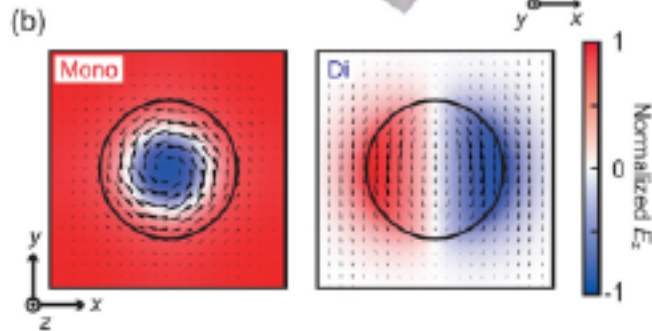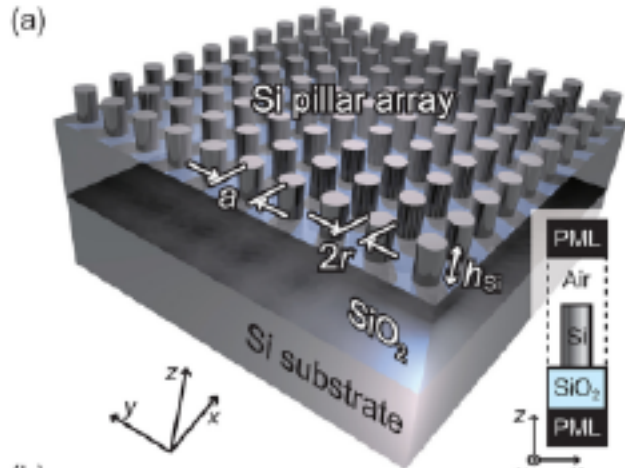• Awards include the Beller Award of OSA and the Millikan Medal of the AAPT

Alan Willner
• Steven & Kathryn Sample Chair in Engineering at the University of Southern California.
• Ph.D. Columbia University
• Honors include Member of US National Academy of Engineering; Int'l Fellow of UK Royal
   Academy of Engineering; President of OSA and of IEEE Photonics Society.
• Activities include using nonlinearity for signal processing and wave manipulation.

Three Metamaterial Platforms Under Investigation

• Nanoantennas coupled to ENZ substrate
  (out of plane; free-space coupling)
  (Rochester)

• Dirac cone metamaterials
  (in plane; compatible with integrated optics)
  (Harvard)

• Photonically doped metamaterials
  (out of plane; free-space coupling)
  (Penn)

Dirac Cone Metamaterials

(a) Si pillar array
a, 2r, h_{Si}
Si substrate
SiO₂
PML / Air / Si / SiO₂ / PML

(b) Mono, Di, Normalized E_z

(c) Wavelength (nm), Mono, Di, Wavenumber k_y (π/a), Wavenumber k_x (π/a)
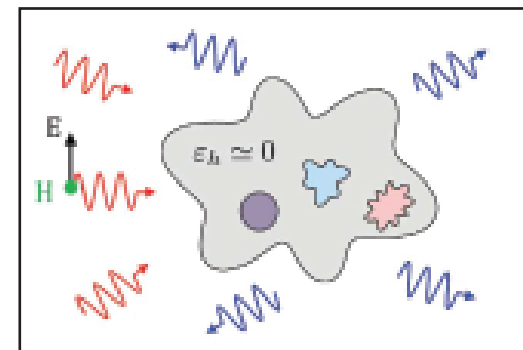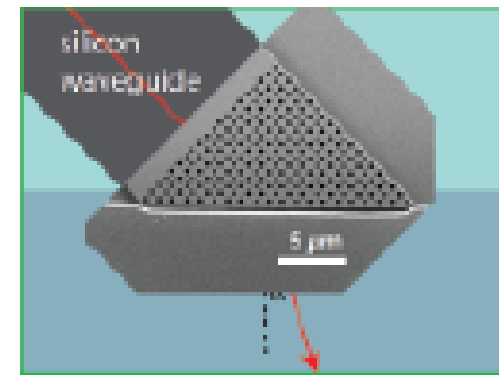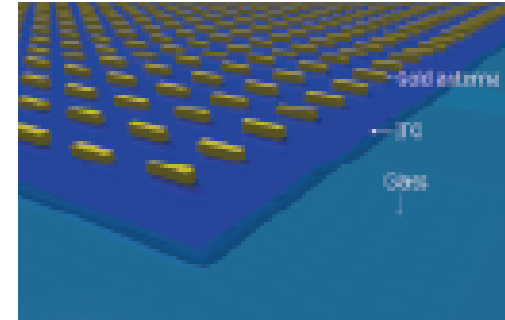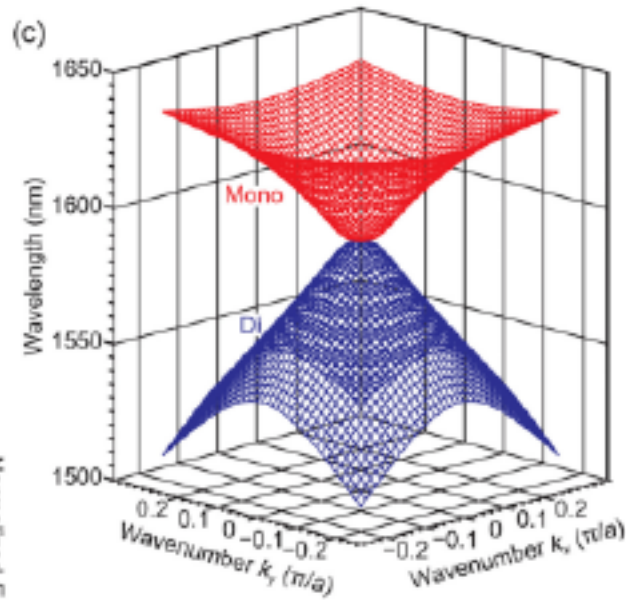
An EMNZ (epsilon and mu near zero) metamaterial

Opt Express 25, 8326 (2017)

$$n = \sqrt{\epsilon \mu}$$

$$Z = \sqrt{\mu / \epsilon}$$

$$R = \frac{1 - Z}{1 + Z}$$

It is also a ZIM
(zero index material)

(d) refractive index, impedance, Re(Z_{eff}), Re(n_{eff}), wavelength(nm)
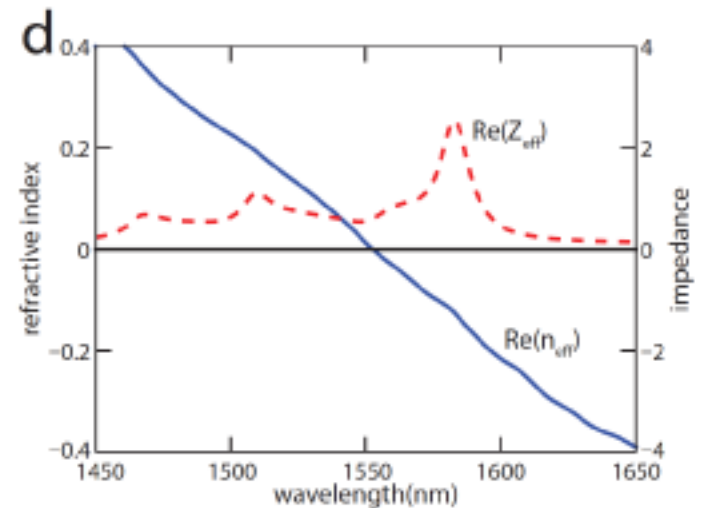
Nonlinear Optical Properties of a Layered Metamaterial in its ENZ Region

Do layered metamaterials also show enhanced NLO response at ENZ wavelength?

- By controlling the metallic fill fraction ρ, we can set the ENZ wavelength to be anywhere from 300 to 700 nm. We use $\rho = 0.2$, which corresponds to 500 nm. We deposit five layer pairs
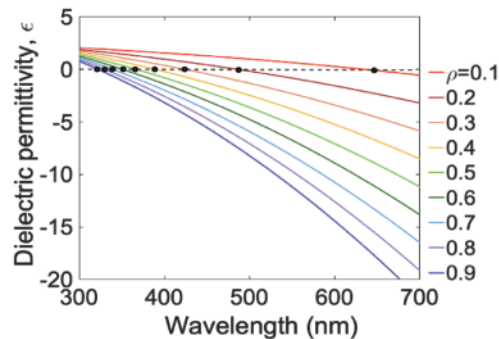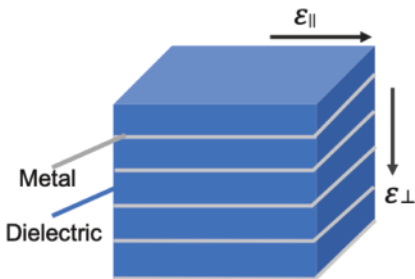
- Note that the real part of epsilon vanishes at 508 nm, close to the design wavelength. The SEM shows our structure. Ag thickness = 16 nm; silica thicness = 65 nm
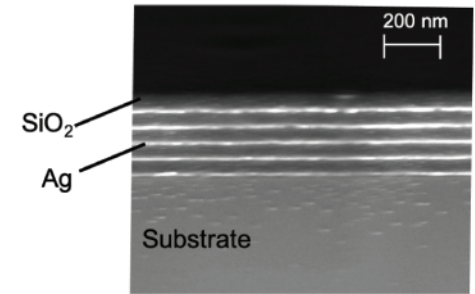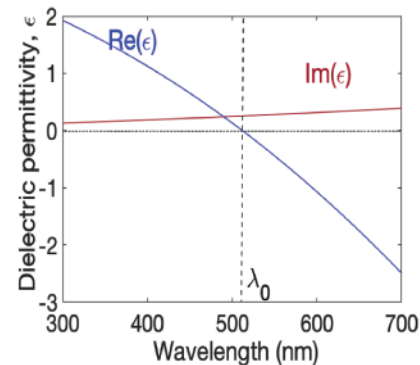
- We perform Z-scan measurements on the sample. Note the enhanced response of the composite as compared to a single layer of silver.

- Note the pronounced peak in the value of n_2 around the ENZ wavelength. We find a good but not perfect agreement with a simple effective medium theory.
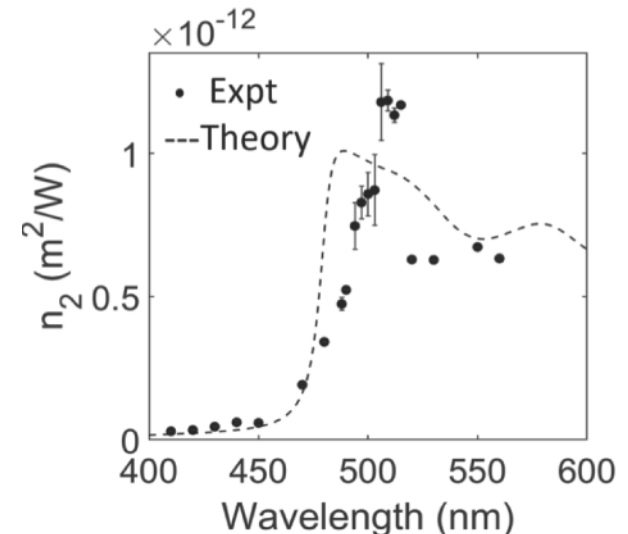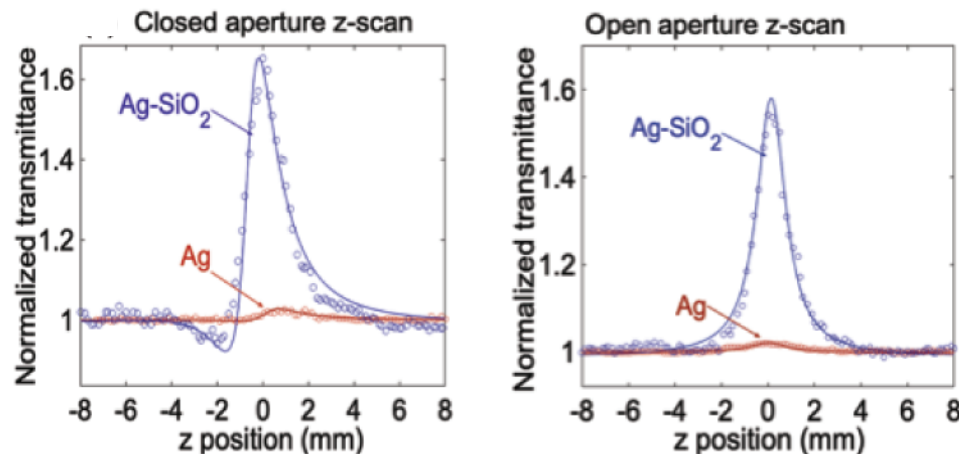
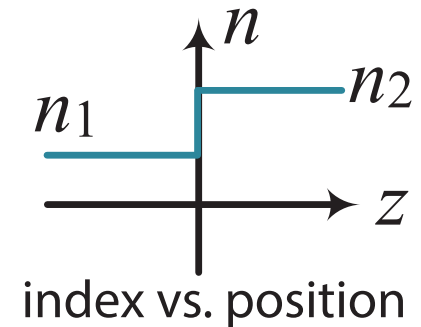Suresh, Reshef, Alam, Upham, Karimi and Boyd

Physics and Applications of Epsilon-Near-Zero Materials

- Physics of ENZ Materials

- Huge NLO Response of ITO and ITO Metastructures

- Materials for ENZ

- Applications of ENZ Materials

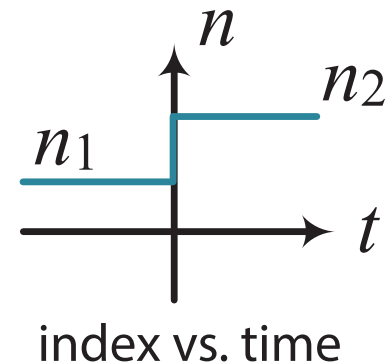# Adiabatic Wavelength Conversion through Time Refraction

- Space refraction (e.g., Snell's law)

$$\frac{c}{f} = n \cdot \lambda \quad \longrightarrow \quad n_1 \lambda_1 = n_2 \lambda_2$$

index vs. position

- Time refraction (analog of Snell's law)

$$\frac{c}{f} = n \cdot \lambda \quad \longrightarrow \quad \boxed{n_1 f_1 = n_2 f_2}$$

index vs. time

Photon frequency (energy) is changed because of the temporal change in index, but the wavelength (inverse of momentum) is conserved in the absence of any spatial asymmetry

- Time refraction is an alternative way of understanding frequency broadening and shifting by self-phase modulation:

$$\delta \omega(t) = \frac{d}{dt} \phi_{NL} = \frac{d}{dt}[n_2 I(t) \omega / c]$$

Laboratory Study of Wavelength Conversion by Time Refraction
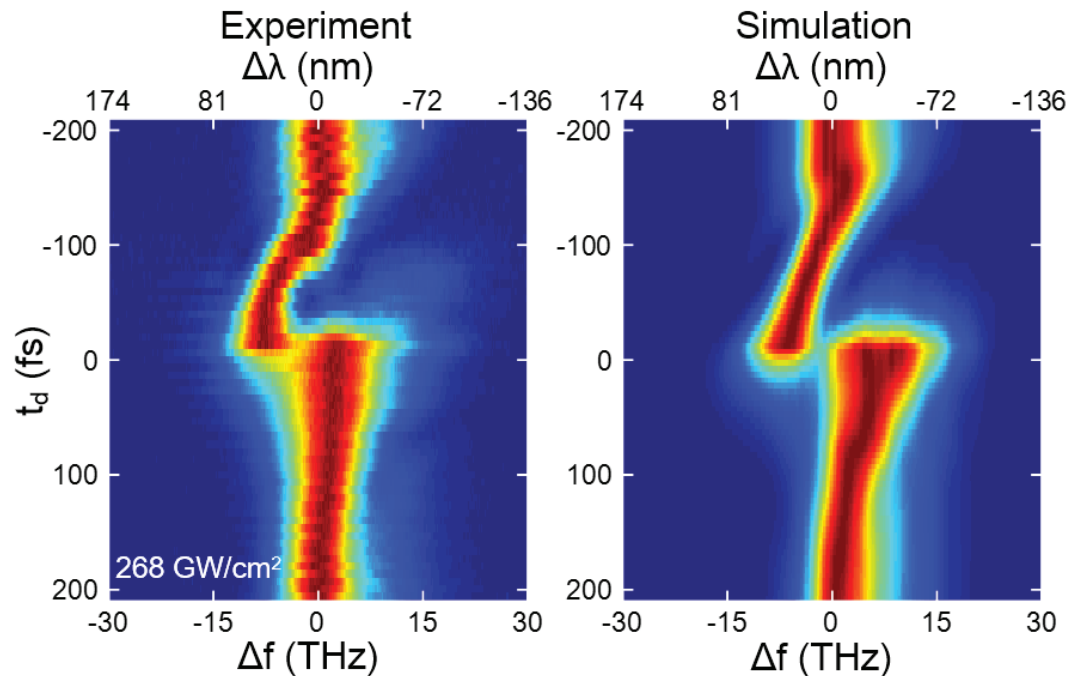
- Pump beam creates a time-varying refractive index in ITO sample
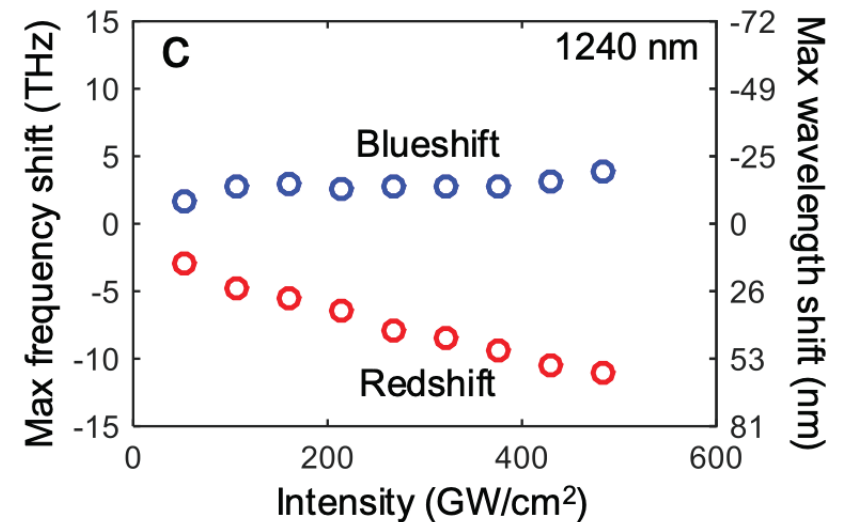
- Frequency of probe beam is thereby modified.
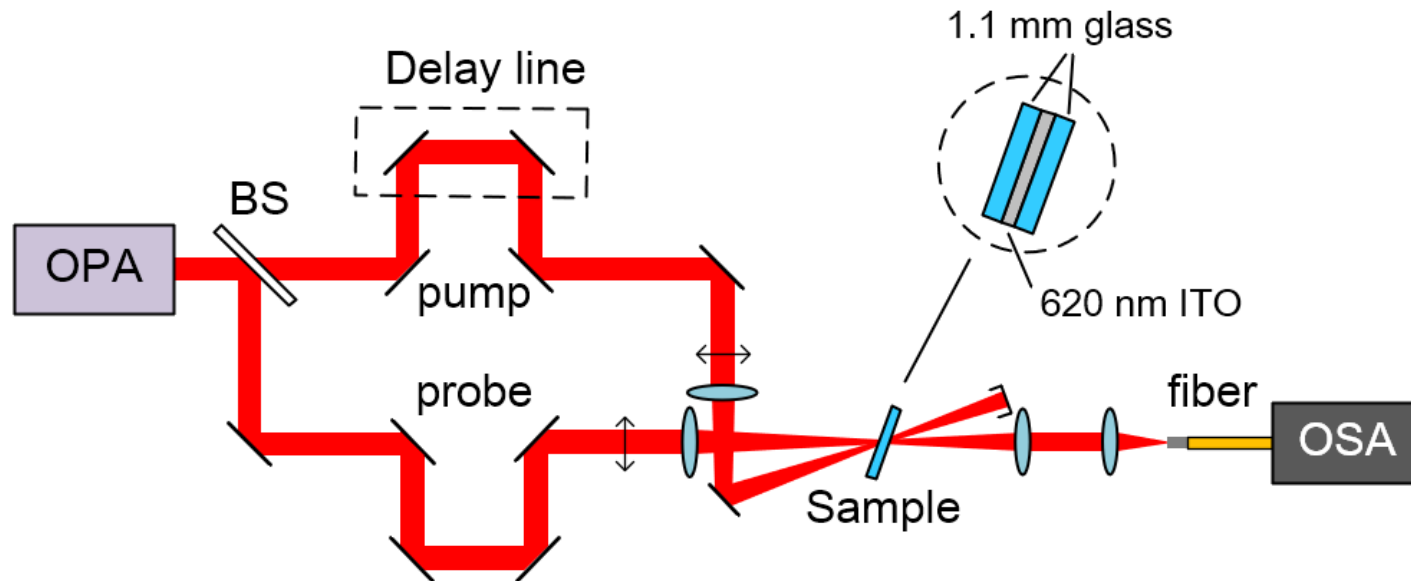
OPA = optical parametric amplifier
   wavelength = 1240 nm
   pulse duration = 120 fs
OSA = optical spectrum analyzer

Results: Adiabatic Wavelength Conversion by Time Refraction

Experimental results at 1240 nm

Probe phase and amplitude are measured
by frequency-resolved optical gating (FROG)

The wavelength shift can be controlled
by the pump intensity and the sign of
the time delay.

- The observed effect is 100 times larger with almost 100 times smaller propagation
  distance than previous reports of AWC.

- Application: wavelength-division multiplexing for telecom

Zhou, Alam, Karimi, Upham, Reshef, Liu, Willner and Boyd, Nature Commun. 11:2180 (2020)

Relaxed Phase-Matching Requirements in ENZ Media

- We study four-wave mixing in a zero-index waveguide

$$2\omega_p = \omega_s + \omega_i$$

- We find that an idler field is generated in both the forward and backward directions!

- Recall that we need $\Delta k = 0$, but when $n = 0$, $k = n\,\omega/c$ vanishes for each of the interacting waves and thus so does Δk.

"forward" process

in $\Rightarrow$ out $\Rightarrow$

"backward" process

in $\Rightarrow$ out $\rightleftarrows$
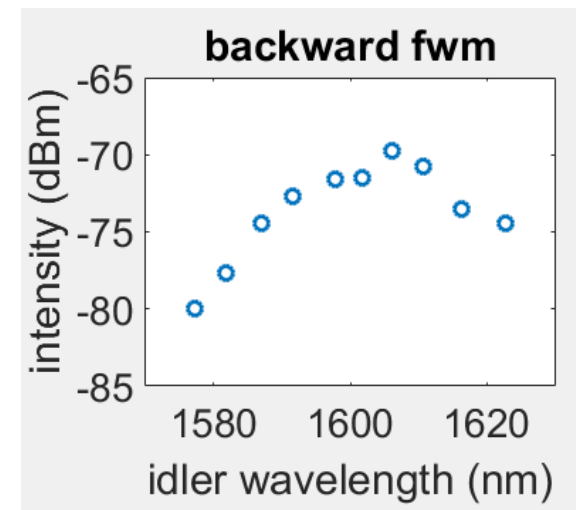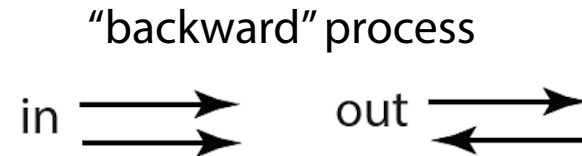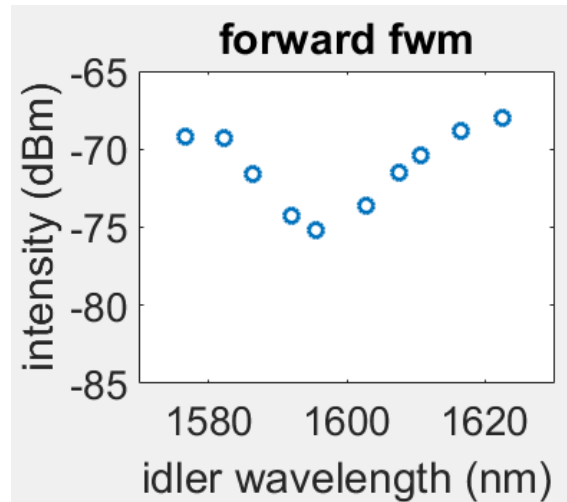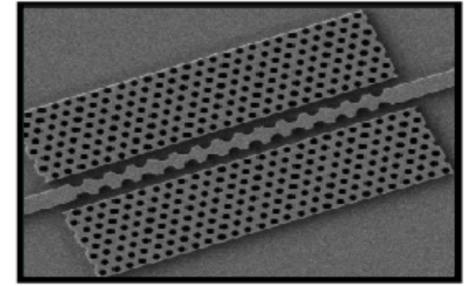
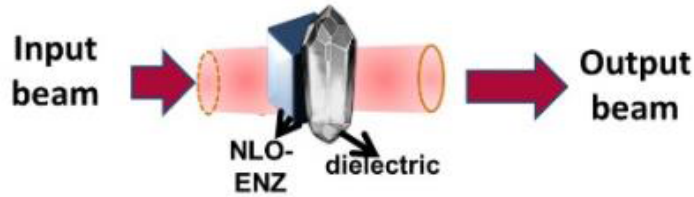
- Significance: Nonlinear optical processes that were previously believed to be too weak to be useful can be excited through use of ENZ materials.
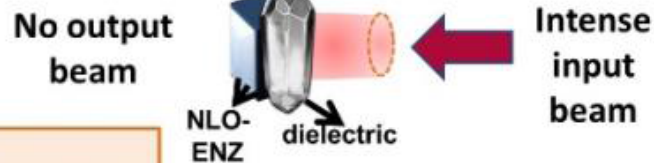
Some Potential Applications of ENZ Behavior
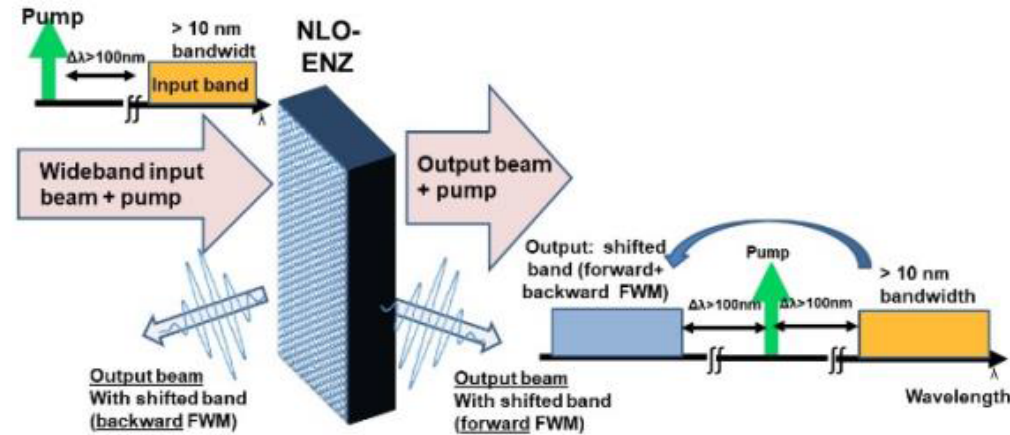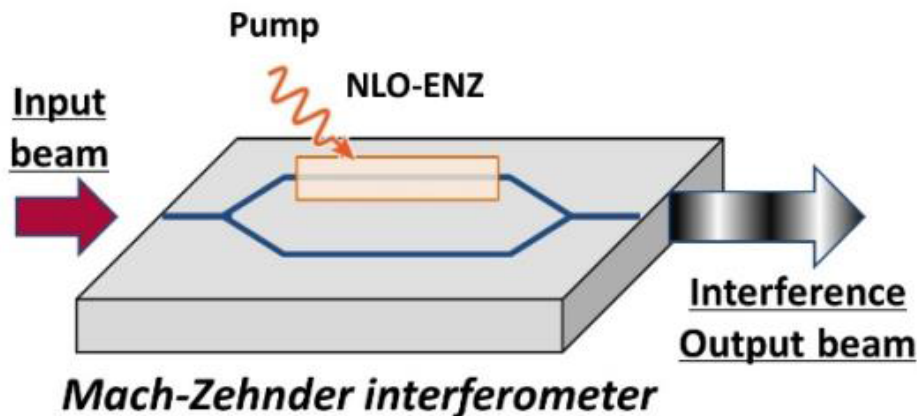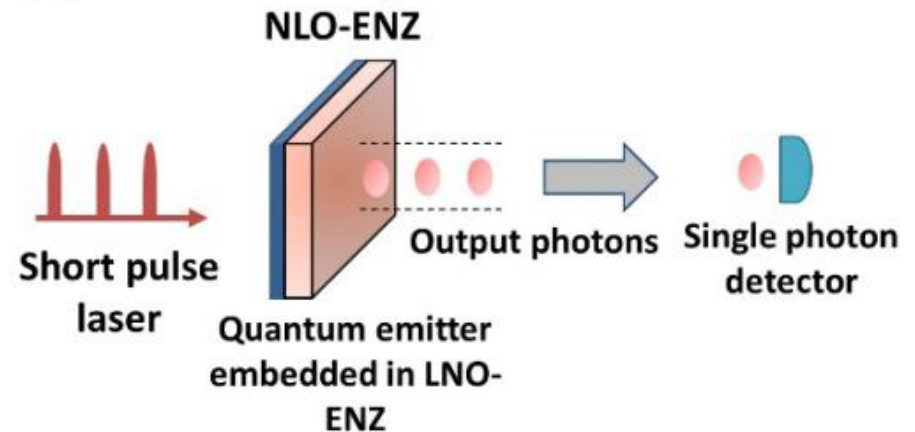
Real-Time Holography with THz Refresh Rates

- Goal: Real-time holography with video or much faster refresh rates.

- The ultrafast response of ITO permits THz refresh rates

- Important applications involve image processing and signal processing
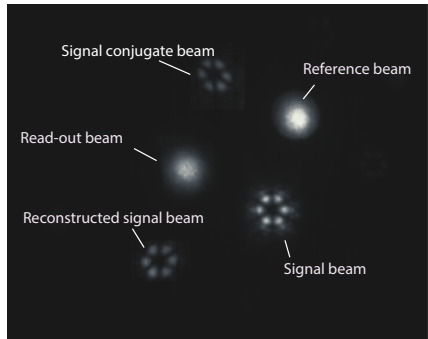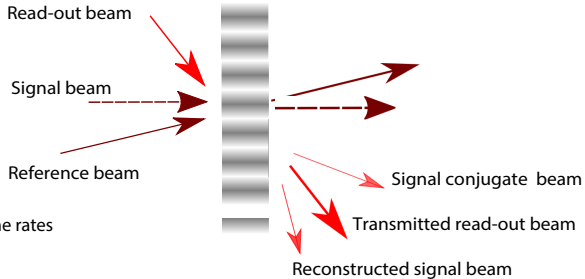
- Current real-time holographic materials cannot even support video frame rates

- Demonstration of image processing (edge enhancement)

Read-out beam

Signal beam

Reference beam

Signal conjugate beam

Transmitted read-out beam

Reconstructed signal beam

Signal conjugate beam

Reference beam

Read-out beam

Reconstructed signal beam

Signal beam

Alam, Fickler, Reshef, Giese, Upham, and Boyd

Summary: Physics and Applications of ENZ Materials

- Extremely interesting physical processes occur in ENZ materials

- ENZ materials, metamaterials, and metastructures display extremely large NLO response

- The huge, ultrafast NLO response of ENZ materials lend themselves to many important applications
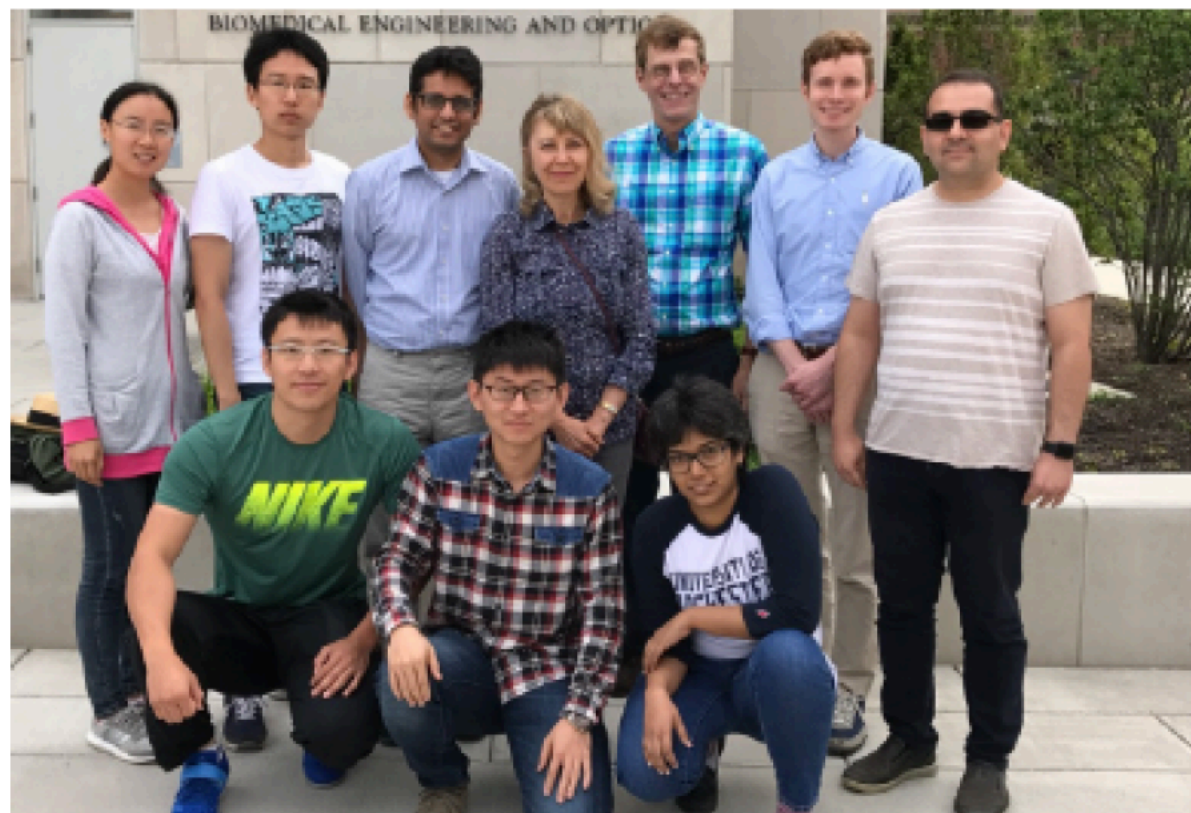
The visuals of this talk are posted at boydnlo.ca/presentations

Special Thanks To My Students and Postdocs!

Ottawa Group

Rochester Group

Dependence of Second-Harmonic Generation on the Linear Dielectric Permittivity

- We solve the standard equations for second-harmonic generation

$$\frac{dA_1}{dz} = i\frac{\eta_1 \omega_1 \chi^{(2)}}{c} A_2(z) A_1^*(z) e^{-i\Delta k z},$$

$$\frac{dA_2}{dz} = i\frac{\eta_2 \omega_2 \chi^{(2)}}{2c} A_1^2(z) e^{i\Delta k z},$$

- We take $\Delta k = 0$ and plot the solution for various values of the permittivity ε.

- We find that the growth rate increases dramatically as the permittivity is decreased.

$\chi^{(2)} = 5 \times 10^{-12}$ [m/V]

Solís, Boyd and Engheta, to be published
See also Solís, CLEO Poster JW2D.15