Physics and Applications of Epsilon-Near-Zero Materials

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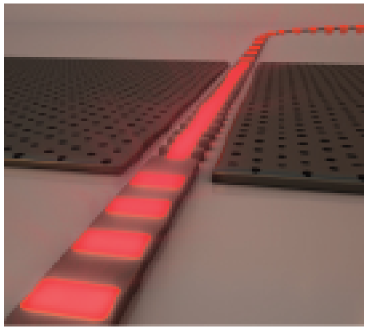
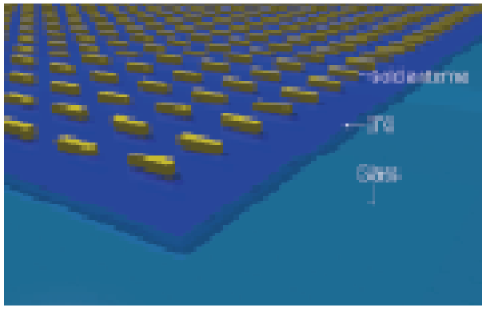
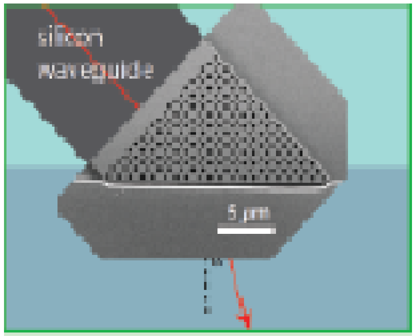
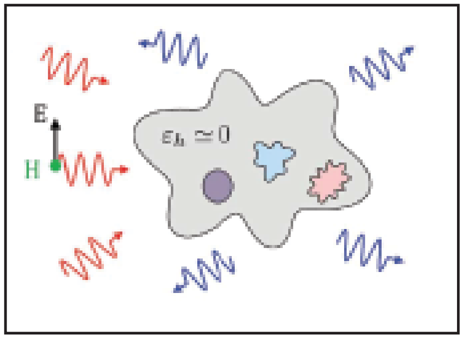








The visuals of this talk are posted at BoydNLO.ca/presentations

Presented at the Nonlinear Photonics virtual meeting, a part of the OSA Advanced Photonics Congress, 13-16 July, 2020.

Physics and Applications of Epsilon-Near-Zero Materials

- Physics of ENZ Materials
- Huge NLO Response of ITO and ITO Metastructures
- Applications of 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 Epsilon-Near-Zero (ENZ) Materials

- ENZ materials possess exotic electromagnetic properties

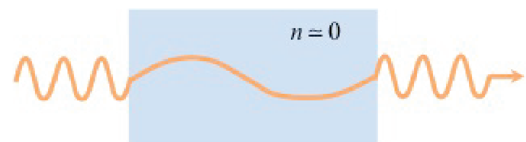
Recall that

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

where ϵ is the permittivity and μ is the magnetic permeability

- Many opportunities in photonics are afforded by ENZ and ZMI materials

$$\lambda = \lambda_{\mathrm{vac}}/n \qquad v = c/n$$



For $n = 0$ the wavelength is stretched and the phase velocity becomes infinite

- Light 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).

Terminology: ZIM= zero-index material; ENZ=epsilon near zero; EMNZ= epsilon and mu near zero

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 ($\mu = 1$) materials

- Implications:

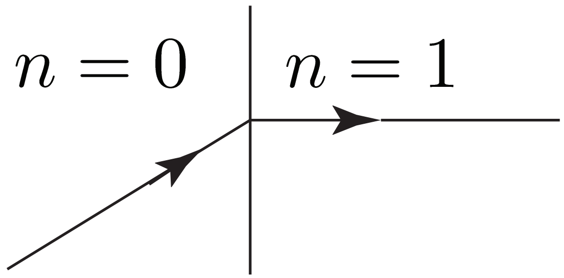
- If we can inhibit spontaneous emission, we can build thresholdless lasers.
- Expect superradiance effects to be pronounced in ENZ materials.

Optics of Zero-Index Materials

- Snell's law leads to intriguing predictions

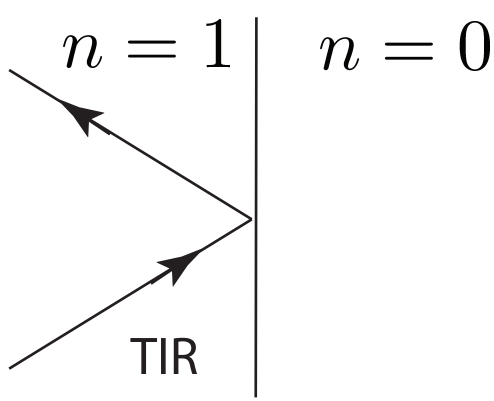
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

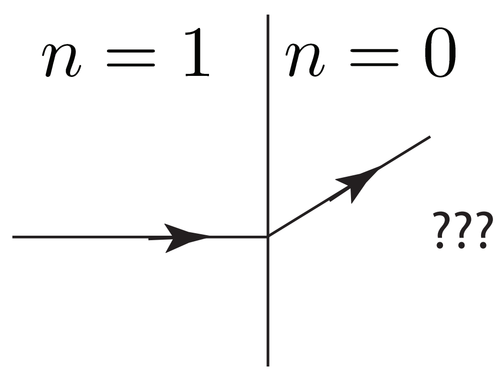
- Light always leaves perpendicular to surface of ENZ material!

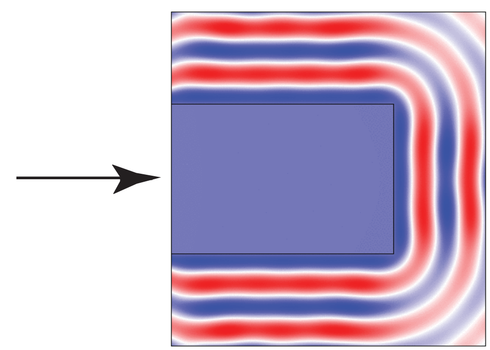


Y. Li, et al., Nat. Photonics 9, 738, 2015; D. I. Vulis, et al., Opt. Express 25, 12381, 2017.

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







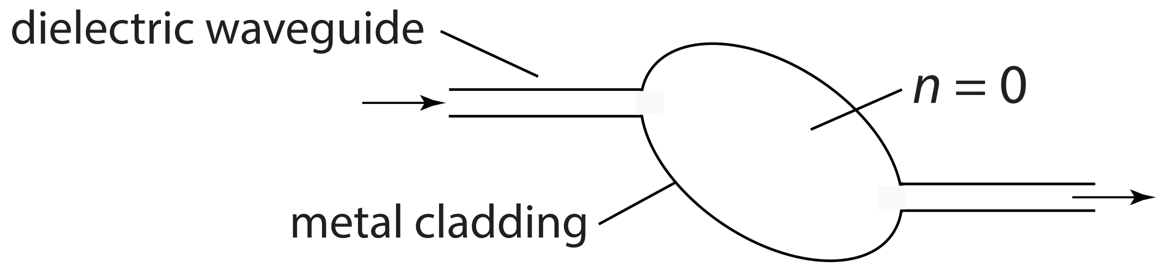
Light enters at normal incidence but leaves in all directions.

Y. Li, et al., Nat. Photonics 9, 738, 2015.

(wave-optics simulation - O. Reshef)

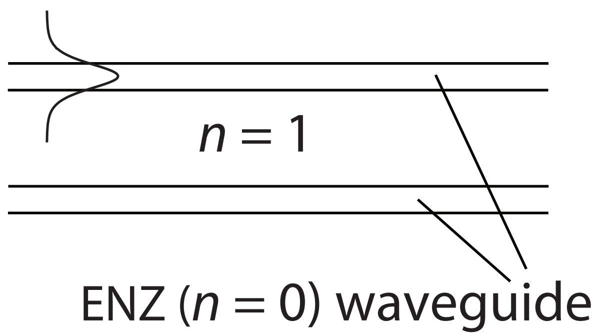
Some Consequences of ENZ Behavior - 2

- Super-coupling (of waveguides)



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



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

Recall that $k = n\,\omega / c$ 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

  - With zero-index materials we can have

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

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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 Material

- 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 usually 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.

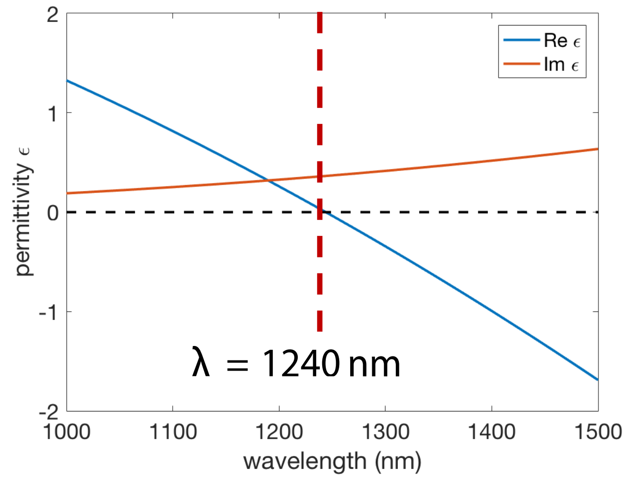
$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

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

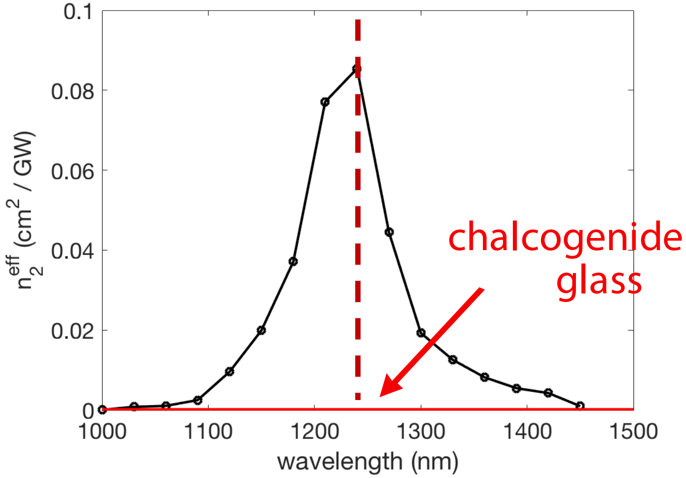
1. Alam, De Leon and Boyd, Science 352, 795–797 (2016)
2. Caspani, Shalaev, Boltasseva, Faccio et al., Phys. Rev. Lett. 116, 233901 (2016).

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

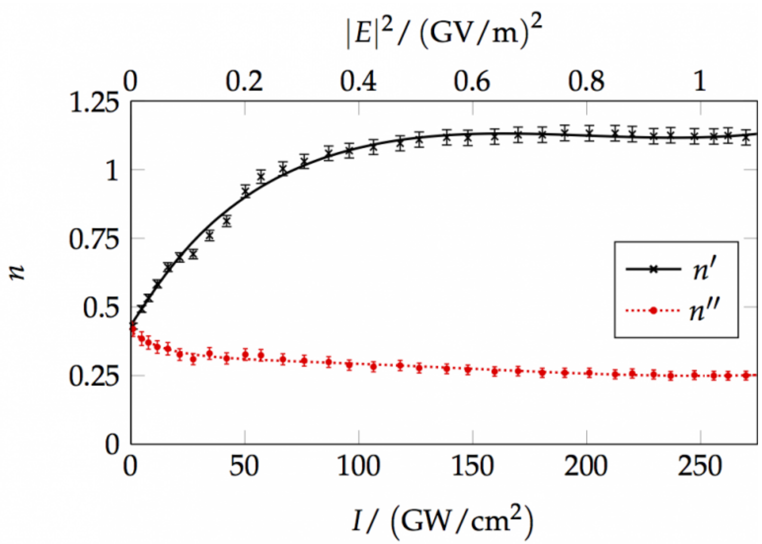
- ellipsometry



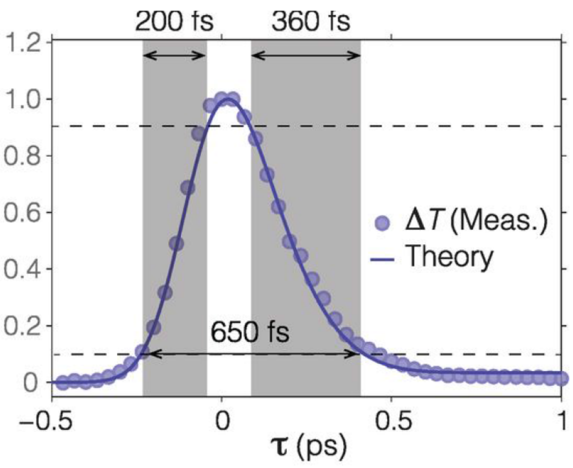
- n_2 can be 3.4 x10^5 times larger than that of silica glass



- overall change in refractive index of 0.8



- sub picosecond reponse time

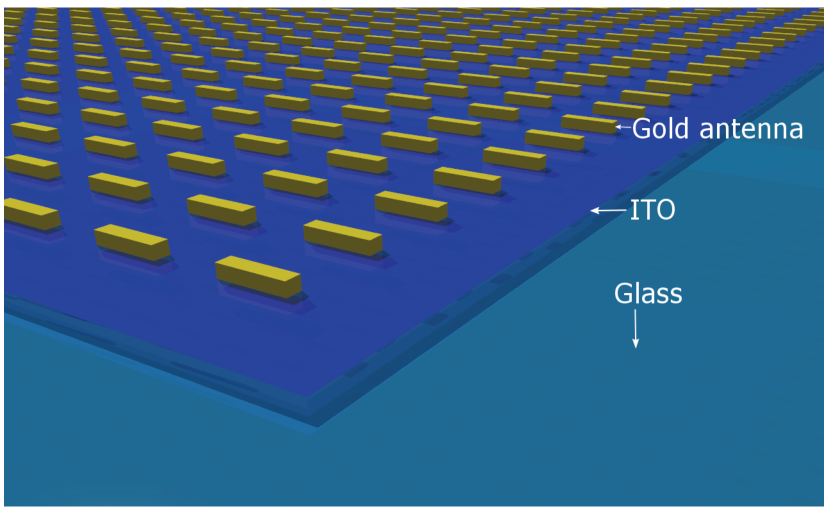


M. Z. Alam et al., Science 352, 795-797 (2016)

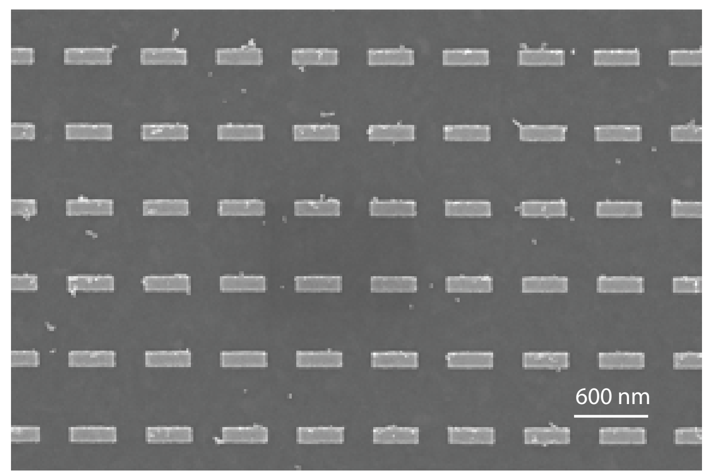
An ENZ Metasurface

- 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 concetrate the field within the ITO
  - Coupled resonators: ENZ resonance and nano-antennas

Concept:

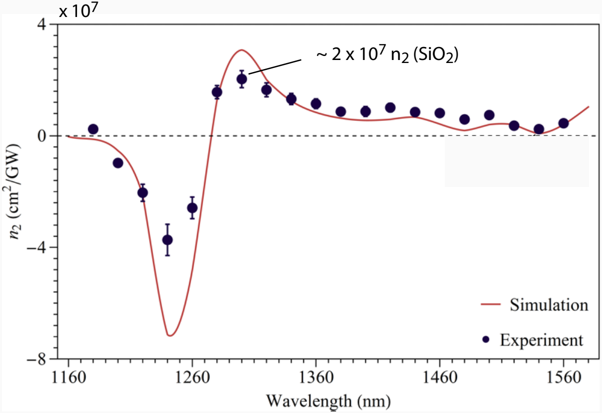


SEM:



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

NLO response of the coupled antenna-ENZ system



The material exhibits extremely large n_2 over a broad spectral range. The magnitude of the on-resonance value is 7 orders of magnitude larger than that of SiO_2.

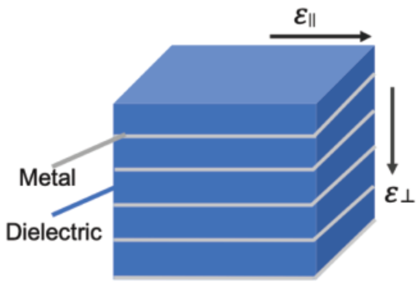
Alam, Schulz, Upham, De Leon and Boyd, Nature Photonics 12, 79-83 (2018).

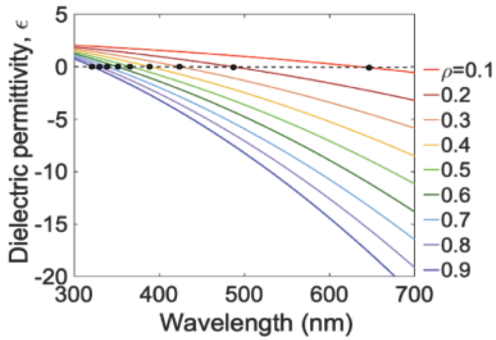
Nonlinear Optical Properties of a Layered Metamaterial in its ENZ Region

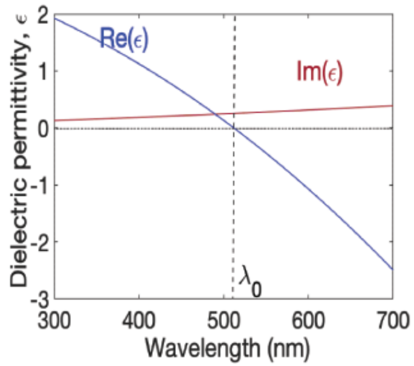
Do layered metamaterials also show enhanced NLO response at ENZ wavelength?

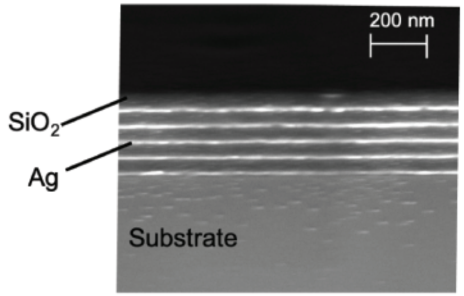
- By controlling the metallic fill fraction ρ, we can set the ENZ wavelength to be anywhere from 300 to 700 nm. We use ρ = 0.2, which corresponds to 500 nm. We deposit five layer pairs

- 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



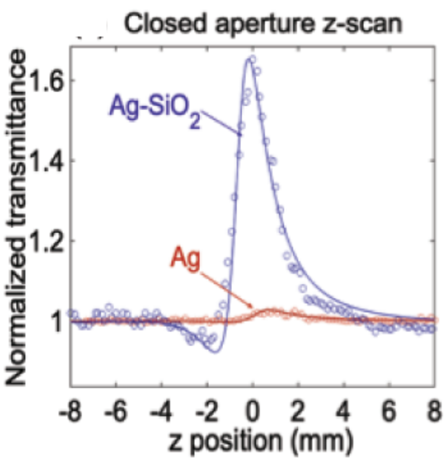


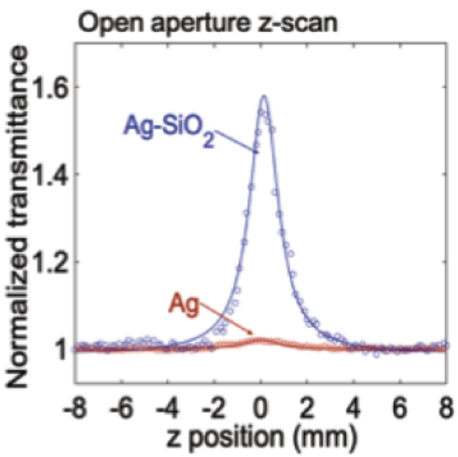


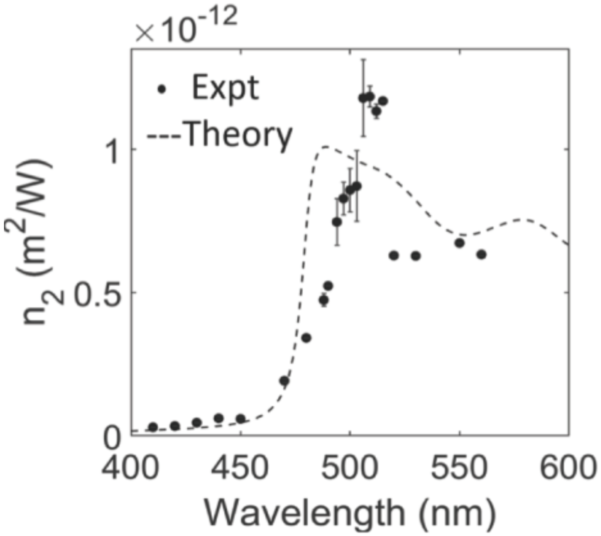


- We perform Z-scan measurements on the sample. Note the enhanced response of the composite as compared to a single layer of silver.

- 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

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- Materials for ENZ
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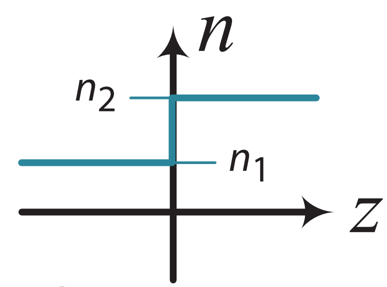
Wavelength Conversion by Time Refraction

- Recall that in space refraction (normal refraction) frequency is conserved but the wavelength is changed

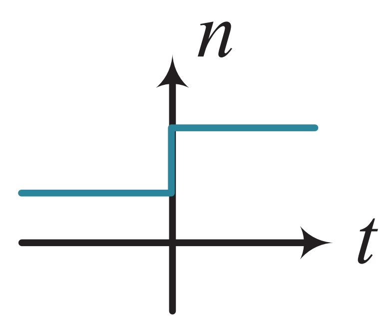
$$\frac{c}{f} = n \cdot \lambda \longrightarrow n_1 \lambda_1 = n_2 \lambda_2$$



index vs. position

- Time refraction (analog of space refraction)

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



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.

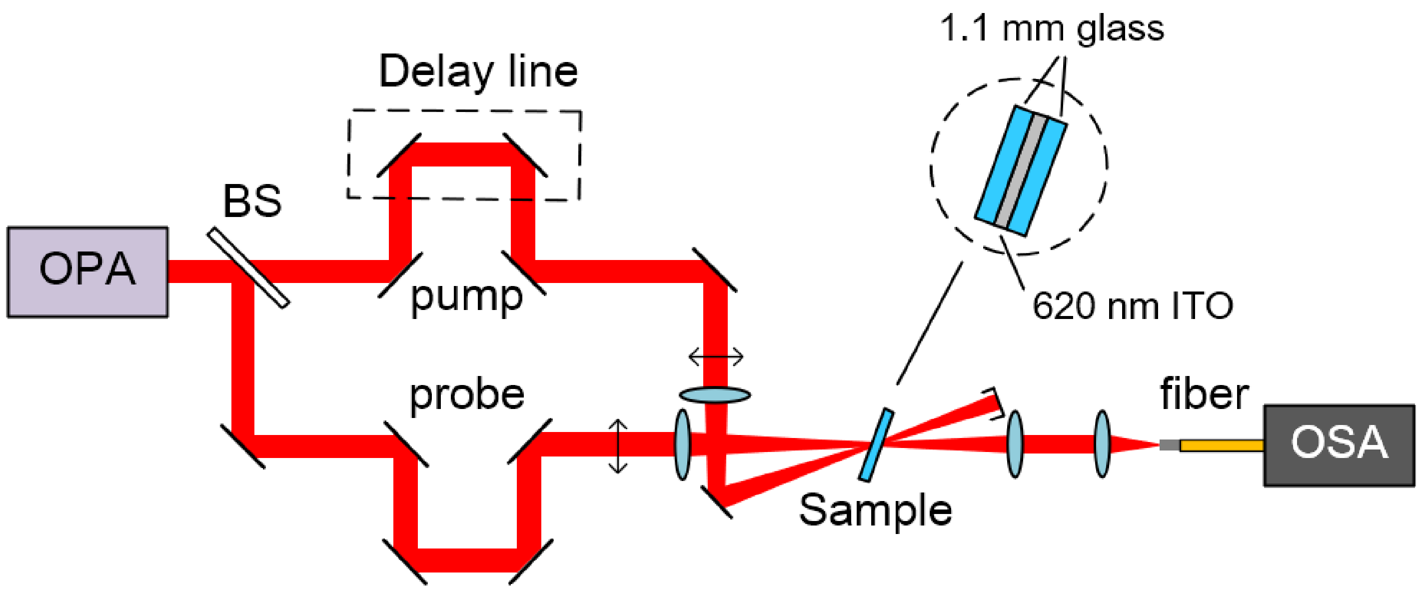
- Time refraction is an alternative way of understanding frequency broadening and shifting by self-phase modulation:

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

Review of time refraction: R. Salem, M. A. Foster, and A. L. Gaeta, Advances in Optics and Photonics 5, 274–317 (2013)

Laboratory Study of Wavelength Conversion by Time Refraction

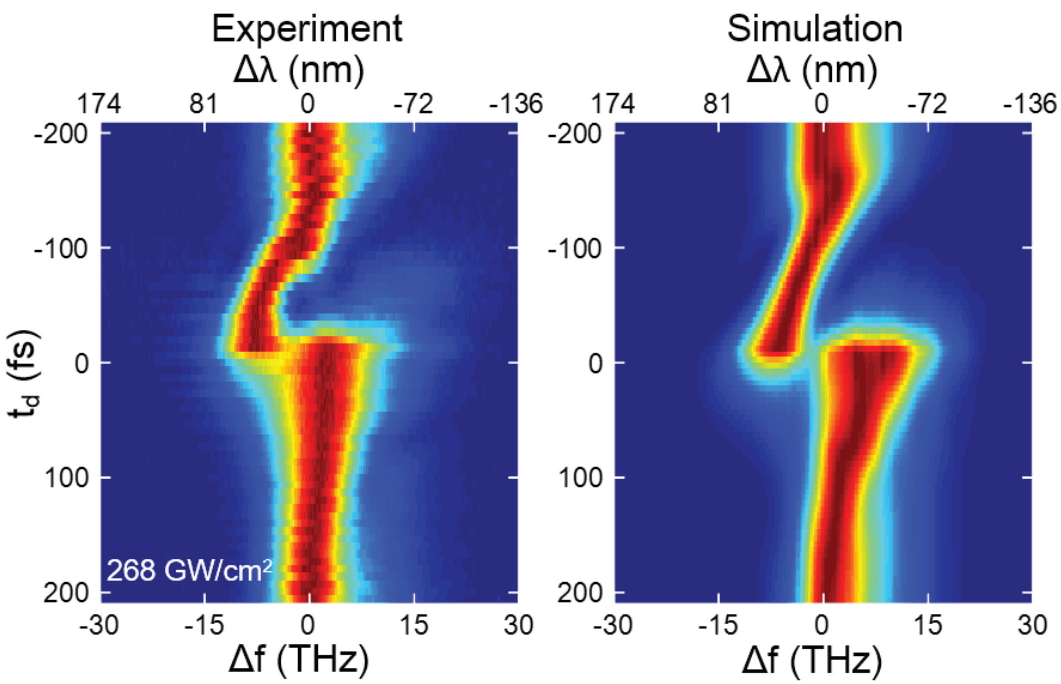
- Pump beam creates a time-varying refractive index in ITO sample
- Frequency of probe beam is thereby modified.

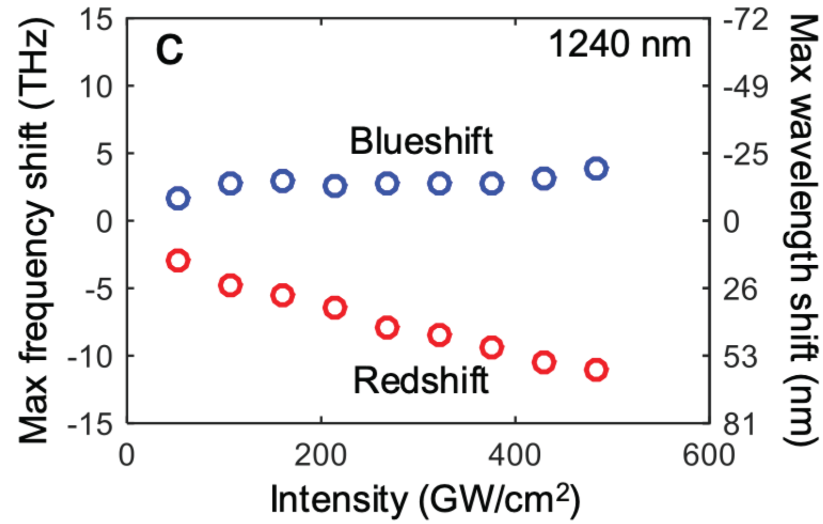


OPA = optical parametric amplifier
    wavelength = 1240 nm
    pulse duration = 120 fs
OSA = optical spectrum analyzer

Results: Adiabatic Wavelength Conversion by Time Refraction

Experimental results at 1240 nm





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

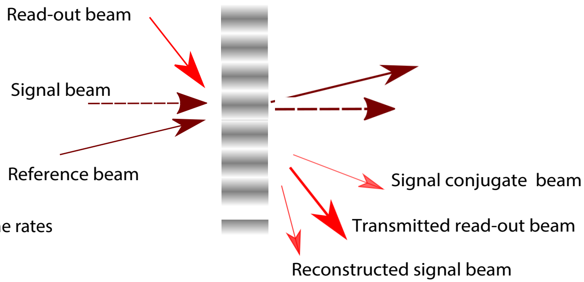
The wavelength shift can be controlled by the pump intensity and the sign of the time delay.

- 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

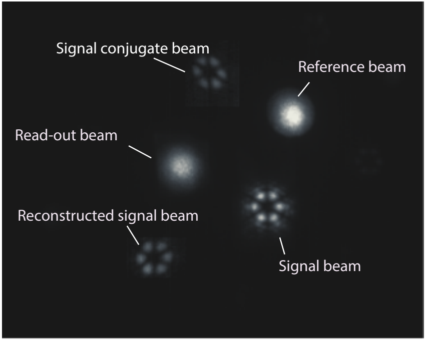
Zhou, Alam, Karimi, Upham, Reshef, Liu, Willner and Boyd, Nature Commun. 11:2180 (2020)

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

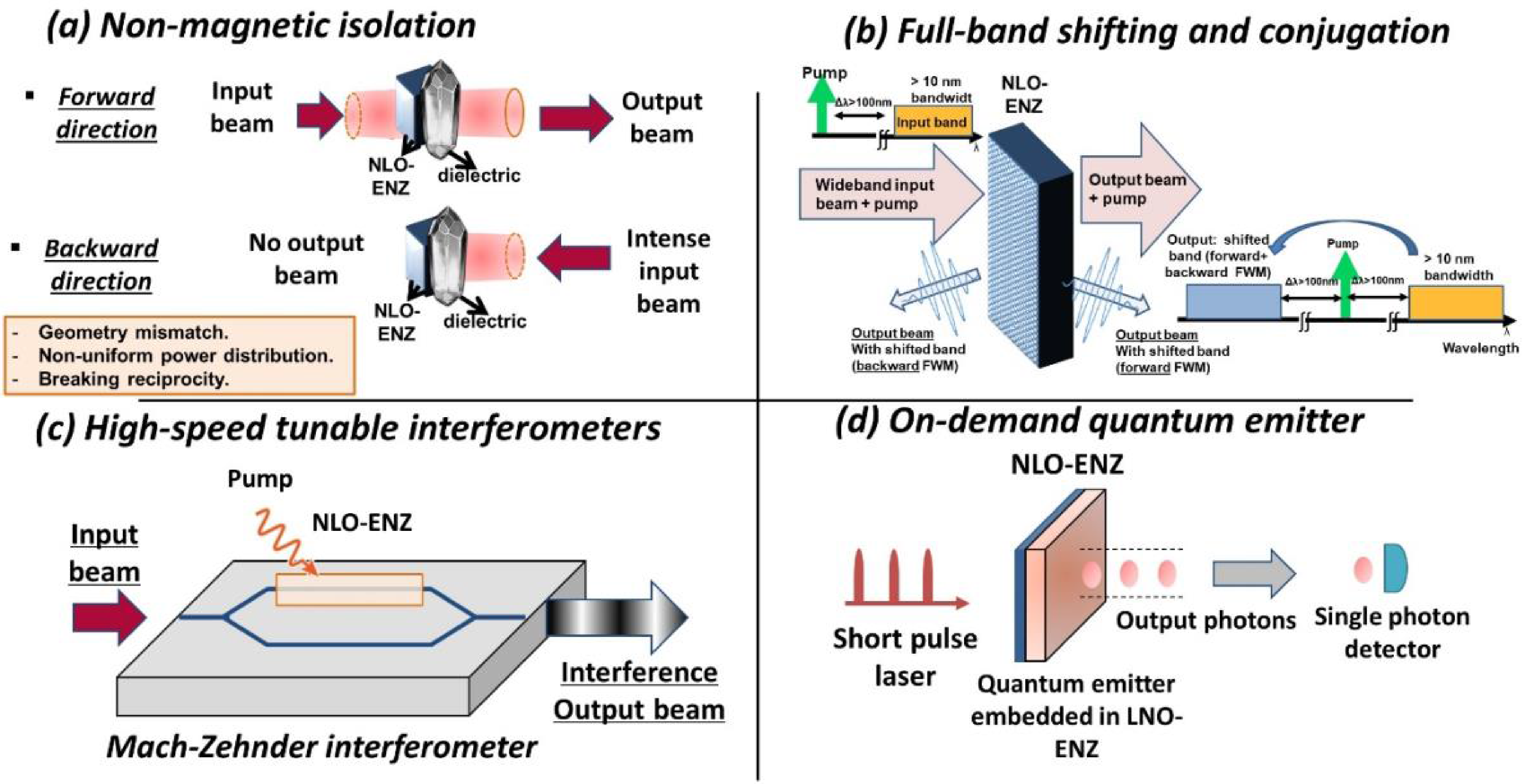


- Demonstration of image processing (edge enhancement)



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

Some Potential Applications of ENZ Behavior



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Rochester Group