# How light behaves when the Refractive Index Vanishes

## Physics and Applications of Epsilon-Near-Zero Materials

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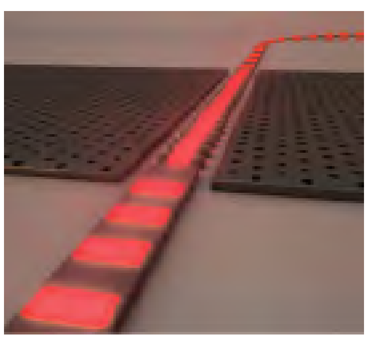
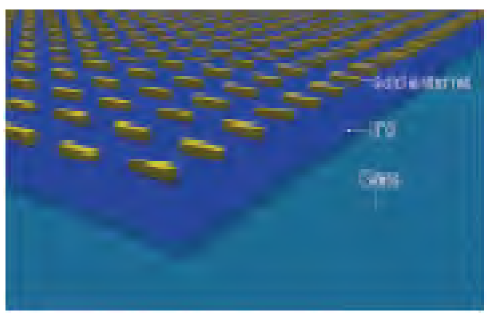
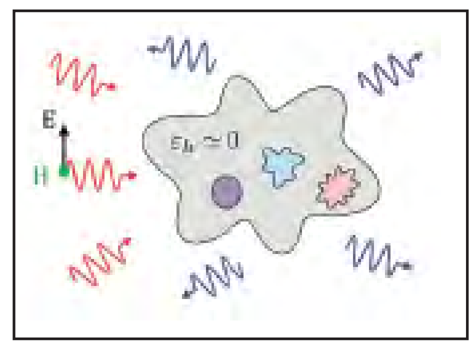





The visuals of this talk will be posted at boydnlo.ca/presentations

Presented at Frontiers in Optics, Washington DC, September 17, 2019

# 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

# Physics of Epsilon-Near-Zero (ENZ) Materials

- ENZ materials possess exotic electromagnetic properties

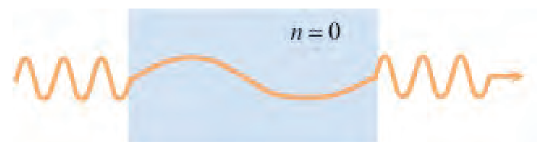
Recall that

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

where $\epsilon$ is the permittivity and $\mu$ is the magnetic permeability

Many opportunities in photonics are afforded by ENZ materials and ZIM (zero-index 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

Silveirinha and Engheta, Phys. Rev. Lett. 97, 157403 (2006).

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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

$B = B_{\text{vac}} \,/\, n^2$

Optical gain is very large!

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

- Implications:

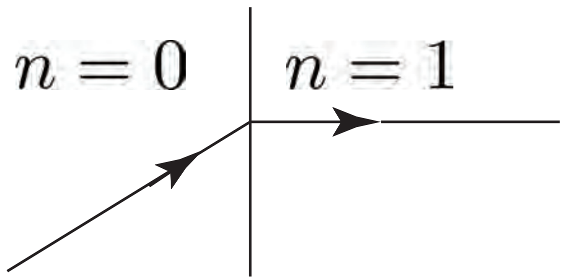
- If we can inhibit spontaneous emission, we can build thresholdless lasers.
- Expect superradiance effects to be pronounced in ENZ materials.

# Physics of Epsilon-Near-Zero (ENZ) Materials -- More

- Snell's law leads to intriguing predictions

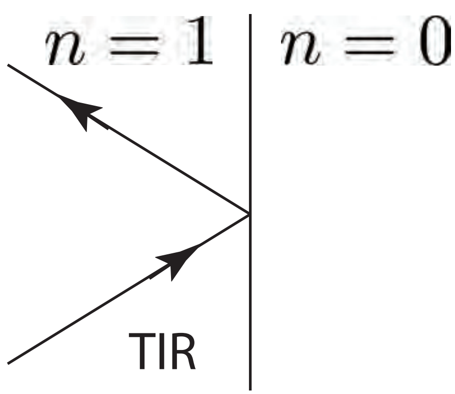
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

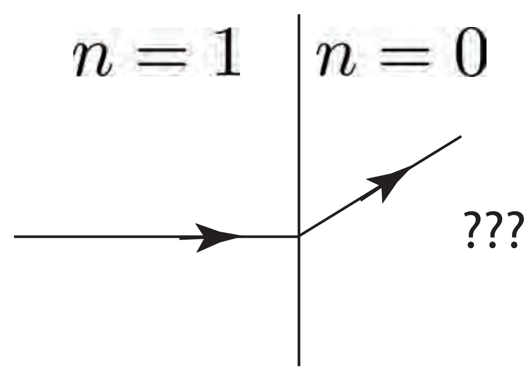
- Light always leaves perpendicular to surface of ENZ material!




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!

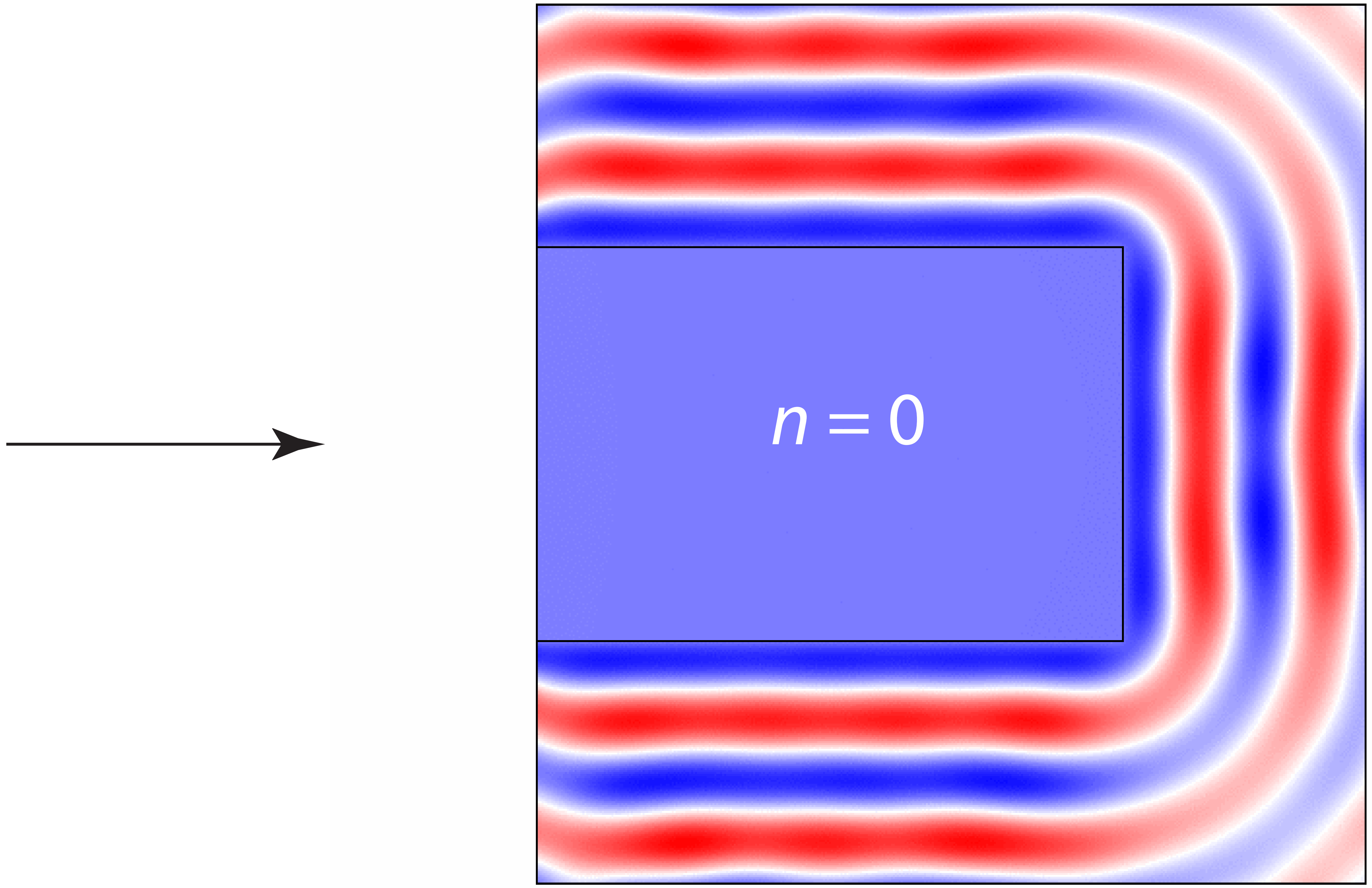



Y. Li, et al., Nat. Photonics 9, 738, 2015.

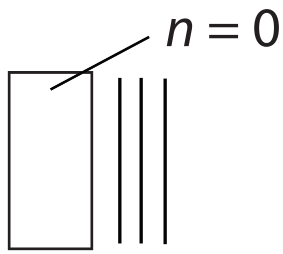
# Maxwell Equations Prediction

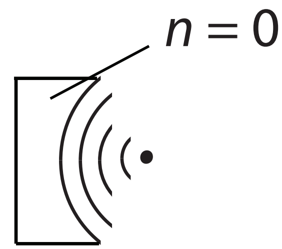
- light enters slab at normal incidence
- but leaves in all directions!



# Some Consequences of ENZ Behaviour - 1

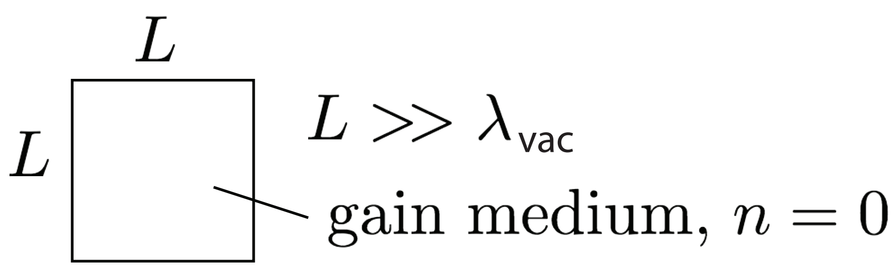
- Funny lenses



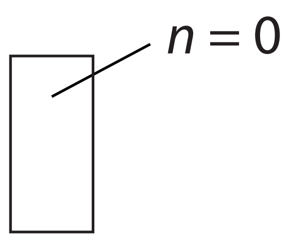

A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

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




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

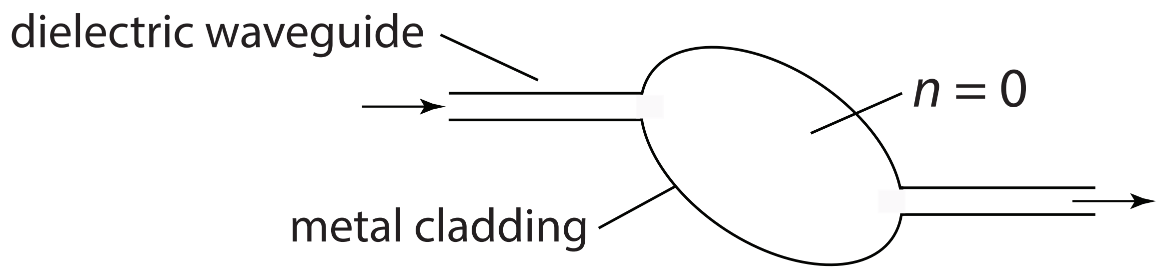
- No Fabry-Perot interference




O. Reshef et al., ACS Photonics 4, 2385, 2017.

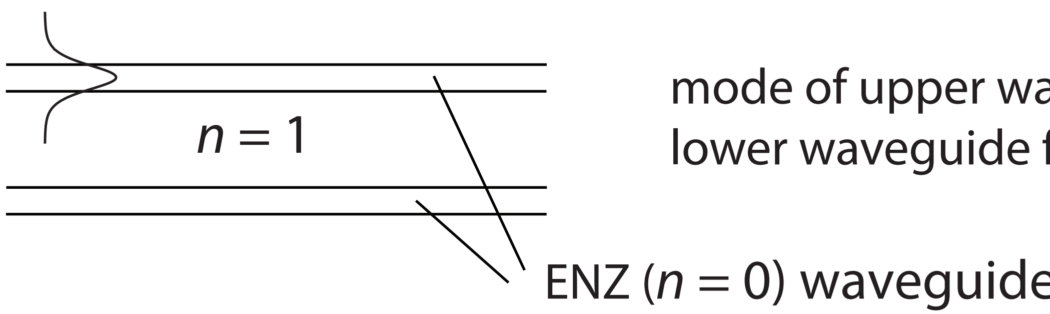
# Some Consequences of ENZ Behavior - 2

- 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.

- Large evanescent tails for waveguide coupling

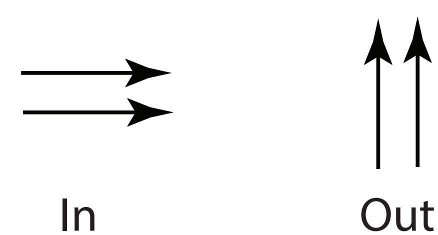



mode of upper waveguide extends to lower waveguide for any separation

- Automatic phase matching of NLO processes

Recall that $k = n\,\omega / c$ vanishes in an ENZ medium.

For example, the following 4WM proces is allowed




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

# Some Consequences of ENZ Behaviour - 3

- How is the theory of self-focusing modified?
- Does the theory of Z-scan need to be modified?
- How is the theory of blackbody radiation modified?
- Do we expect very strong superradiance effects?
- More generally, how is any NLO process modified when $n_0 = 0$?

# Physics and Applications of Epsilon-Near-Zero Materials

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- Huge NLO Response of ITO and ITO Metastructures
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# Epsilon-Near-Zero Materials

- Metamaterials
  Materials tailor-made to display ENZ behaviour

- Homogeneous materials
  All materials display ENZ behaviour 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 $\omega = \omega_p/\sqrt{\epsilon_\infty} \equiv \omega_0$.

- Challenge: Obtain low-loss ENZ materials
  Want Im $\varepsilon$ as small as possible at the frequency where Re $\varepsilon = 0$.

- We are examining a several materials
  ITO: indium tin oxide
  AZO: aluminum zinc oxide
  FTO: fluorine tin oxide

# 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

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

# Optical Properties of Indium Tin Oxide (ITO)

- ITO is a degenerate semiconductor (so highly doped as to be metal-like).
- It has a 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 $\omega = \omega_p/\sqrt{\epsilon_\infty} \equiv \omega_0$.

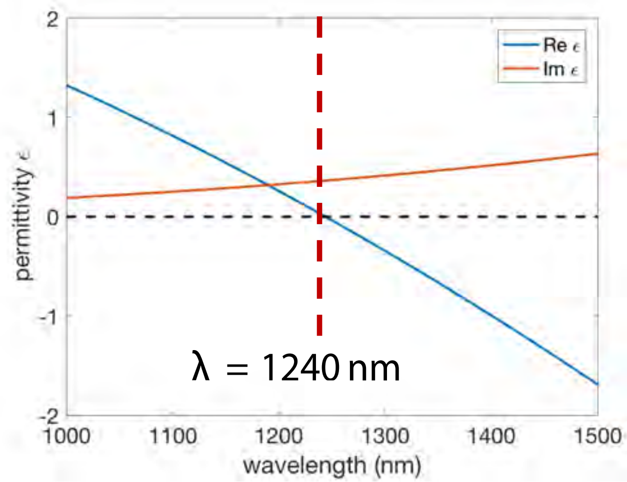
The region near $\omega_0$ is known as the epsilon-near-zero (ENZ) region.

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

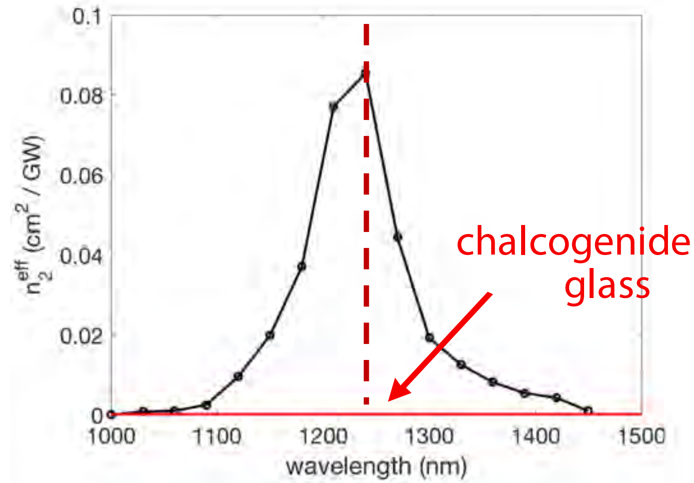
H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, Science 342, 1223 (2013).
C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, Phys. Rev. B 85, 045129 (2012).
S. Campione, D. de Ceglia, M. A. Vincenti, M. Scalora, and F. Capolino, Phys. Rev. B 87, 035120 (2013).
A. Ciattoni, C. Rizza, and E. Palange, Phys. Rev. A 81,043839 (2010).

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

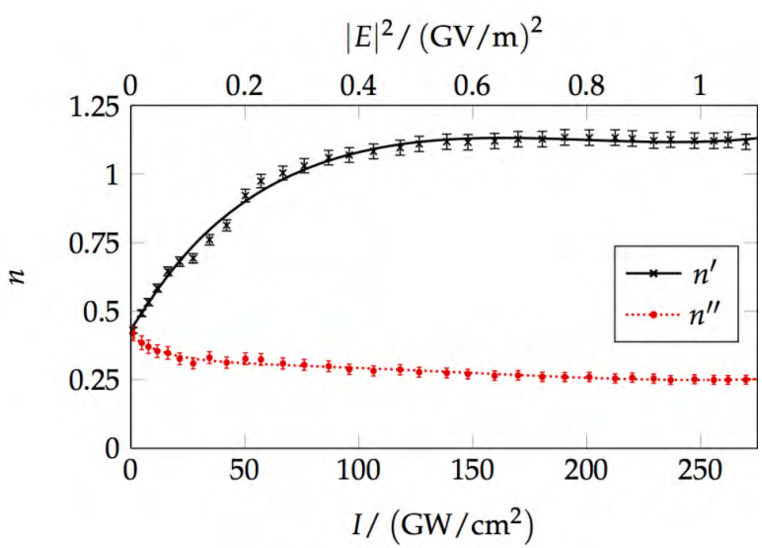
- ellipsometry



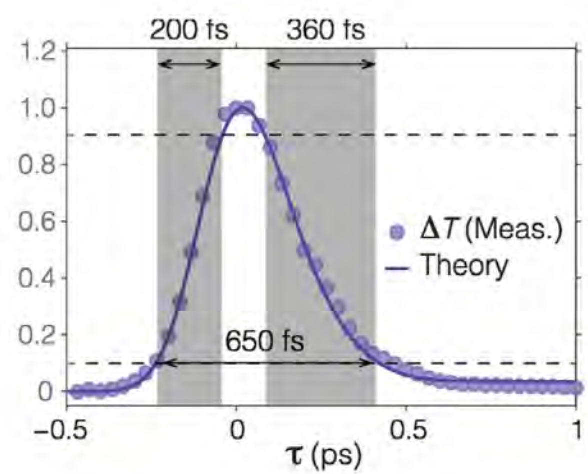

- $n_2$ can be 3.4 x$10^5$ times larger than that of silica glass




- overall change in refractive index of 0.8




- sub picosecond reponse time

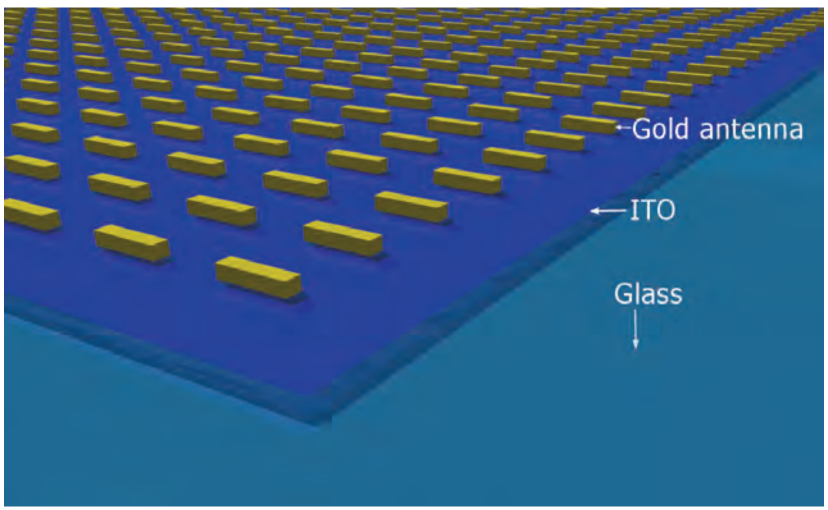



*M. Z. Alam et al., Science 352, 795-797 (2016)*

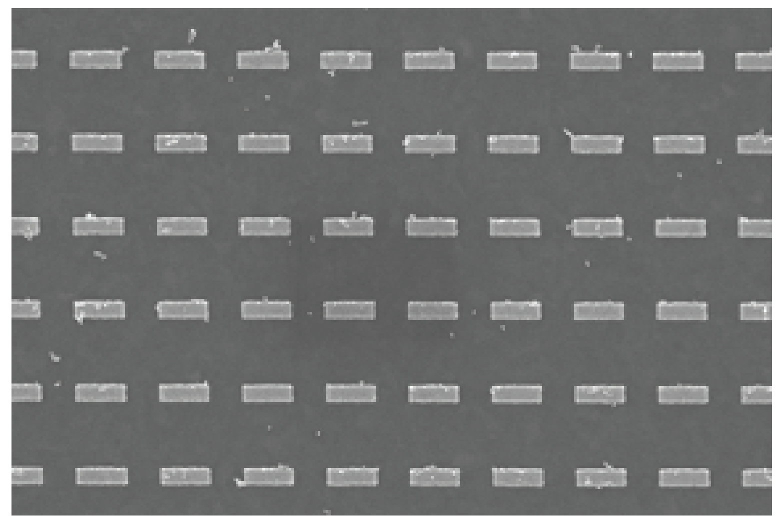
# An ENZ Metasurface

- 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:

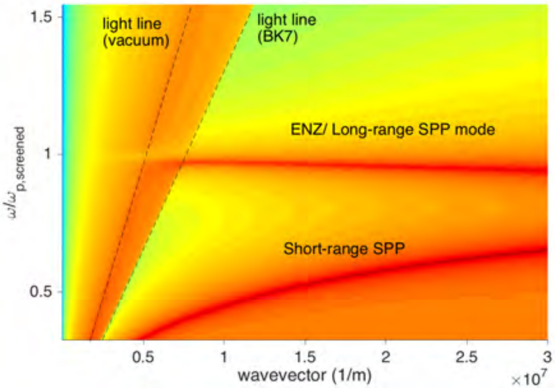



SEM:



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

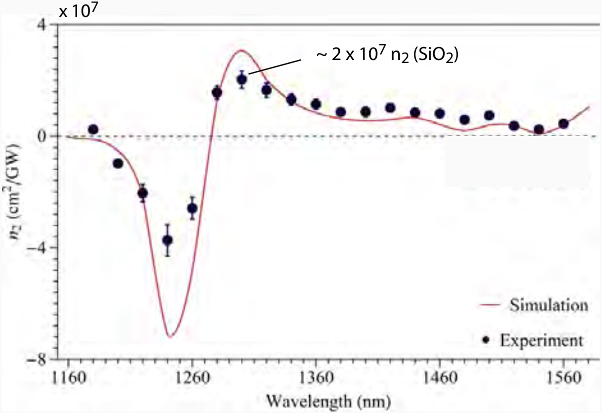
# A thin ENZ medium supports a bulk plasma mode.




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 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).

# Nonperturbative Nature of the NLO Response

1. The conventional equation $n = n_0 + n_2 I$ is not applicable to ENZ and other low-index materials. The nonlinear response is nonperturbative.
2. The problem is that $n_2$ is not a reliable metric under ENZ conditions. But $n_2$ is the standard way for quantifying intensity-dependent index changes. What can we do?
3. The nonlinear response can be accurately modeled in the $\chi^{(3)}$ limit by

$$n = \sqrt{n_0^2 + 2n_0 n_2 I}$$

where

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

and

$$I = 2\mathrm{Re}(n_0)\epsilon_0 c|E|^2$$

4. More generally, the intensity dependent refractive index can be described by

$$n = \sqrt{\epsilon^{(1)} + 3\chi^{(3)}|E|^2 + 10\chi^{(5)}|E|^4 + \cdots}$$

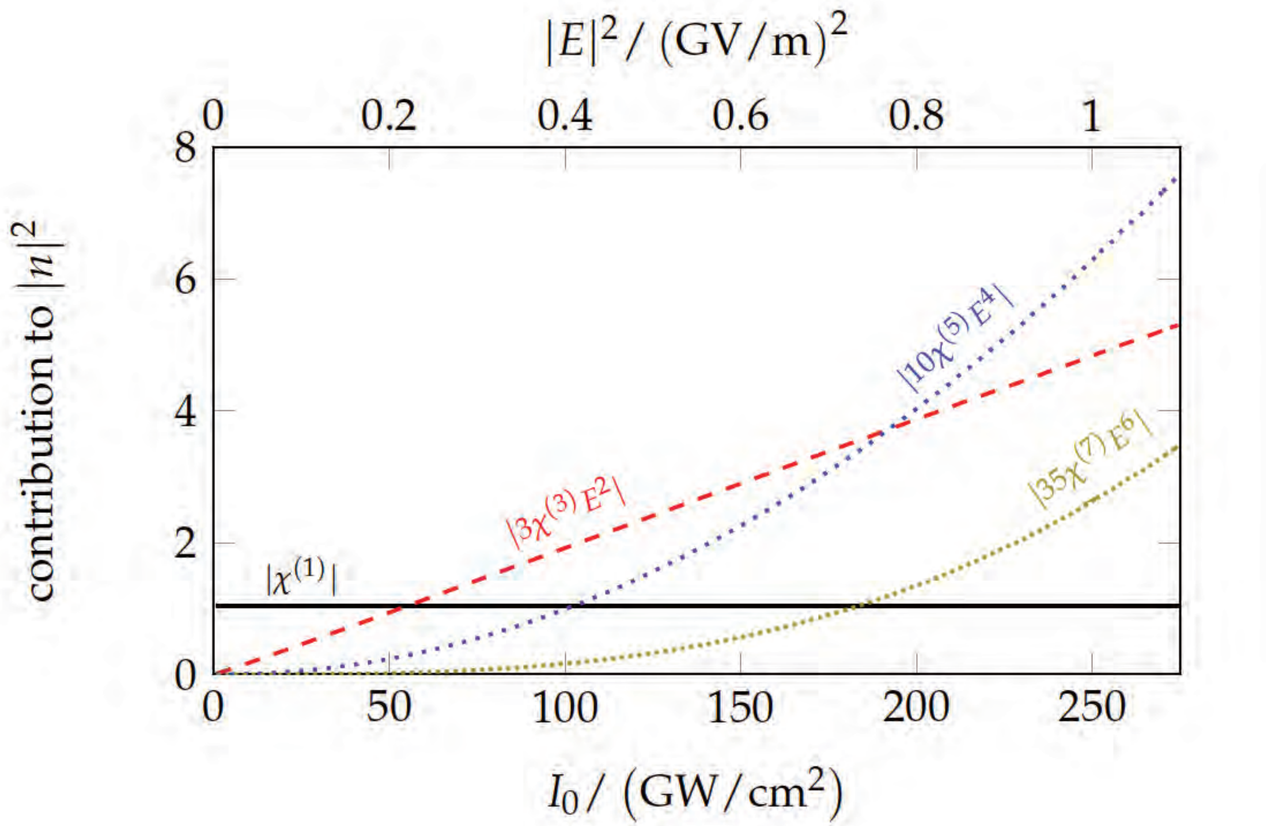
Reshef, Giese, Alam, De Leon, Upham, Boyd, Opt. Lett. 42, 3225 (2017).

# Is Nonlinear Response of ITO Nonperturbative?

- Dependence of the polarization P on the electric field obeys the usual power series expansion.

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

- However, the dependence of the refractive index *n* on the intensity $|E|^2$ is nonperturbative.



# Physics and Applications of Epsilon-Near-Zero Materials

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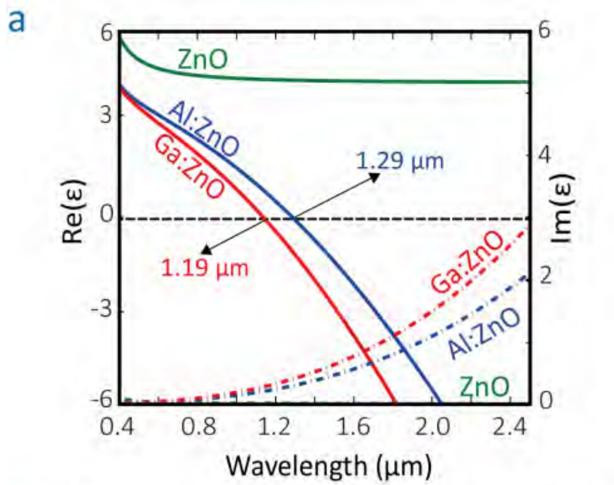
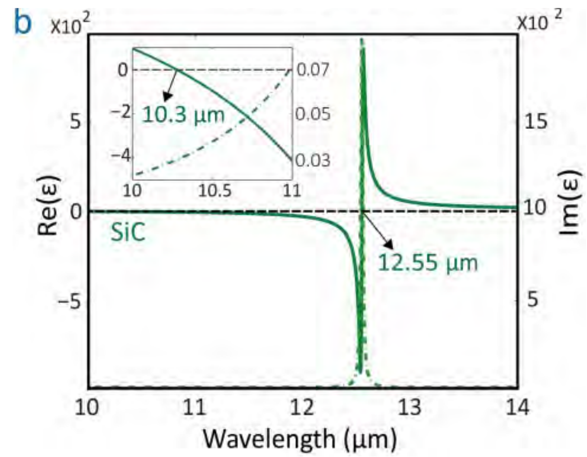
# Epsilon-Near-Zero (ENZ) and Near-Zero-Index (NZI) Examples

TCO



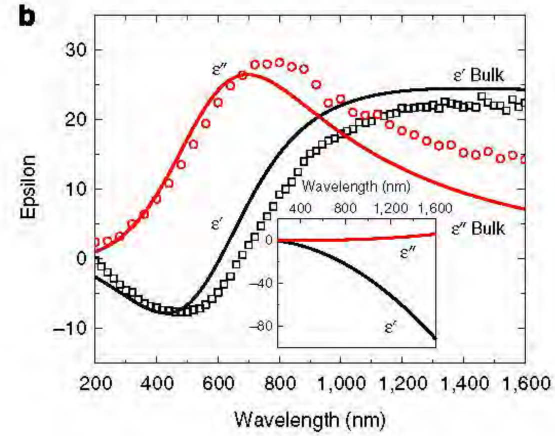

*A. Boltasseva (Purdue)*
*Kim et al., Optica* (2016)

SiC



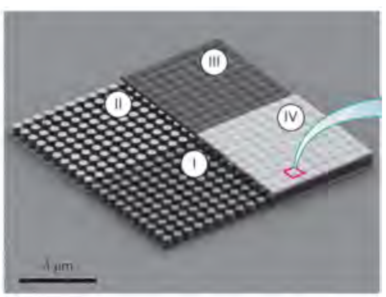

*J. Caldwell (Vanderbilt)*
*Kim et al., Optica* (2016)

$Bi_{1.5}Sb_{0.5}Te_{1.8}Se_{1.2}$

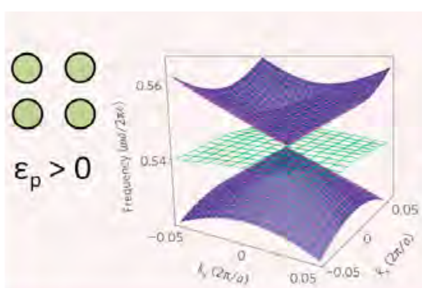



*N. Zheludev (Southmapton)*
*Ou et al., Nat. Commun.* (2014)

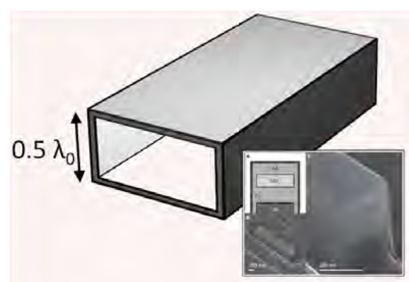
Metamaterials



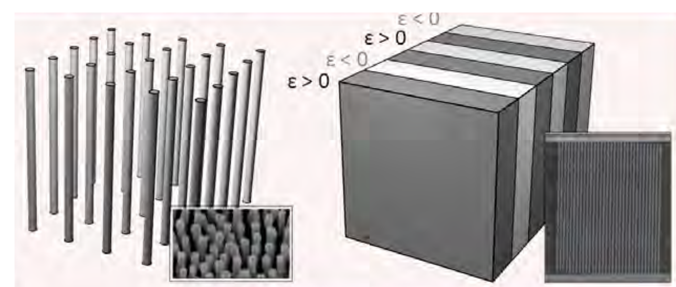
*E. Mazur Li et al., Nat. Photon.* (2015)




*Chan, Huang et al., Nat. Mater.* (2011)




*SEM from: Polman's & Engheta's*
*Vesseur et al., PRL* (2013)




$\mathrm{Re}(\varepsilon) \cong 0$

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

# Giant Nonlinear Response of ENZ Metastructures:  Our Team

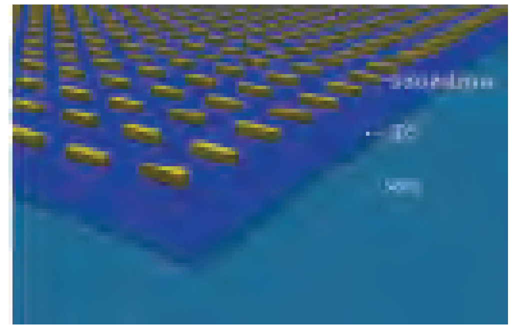
**Nader Engheta**
- H. Nedwill Ramsey Professor at the University of Pennsylvania
- B.S. degree from the University of Tehran and his M.S and Ph.D. from Caltech.
- Activities include ENZ, photonics, metamaterials, nano-optics, graphene optics, electrodynamics, microwave and optical antennas, studies 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.

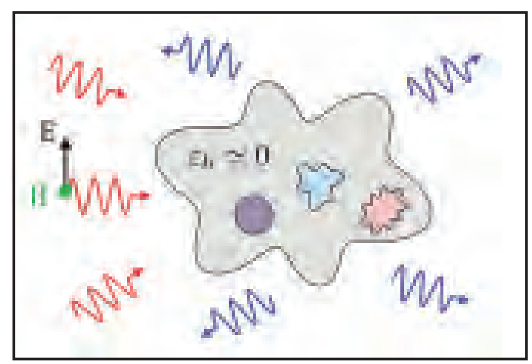
# Three Material Platforms Under Investigation

- 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)



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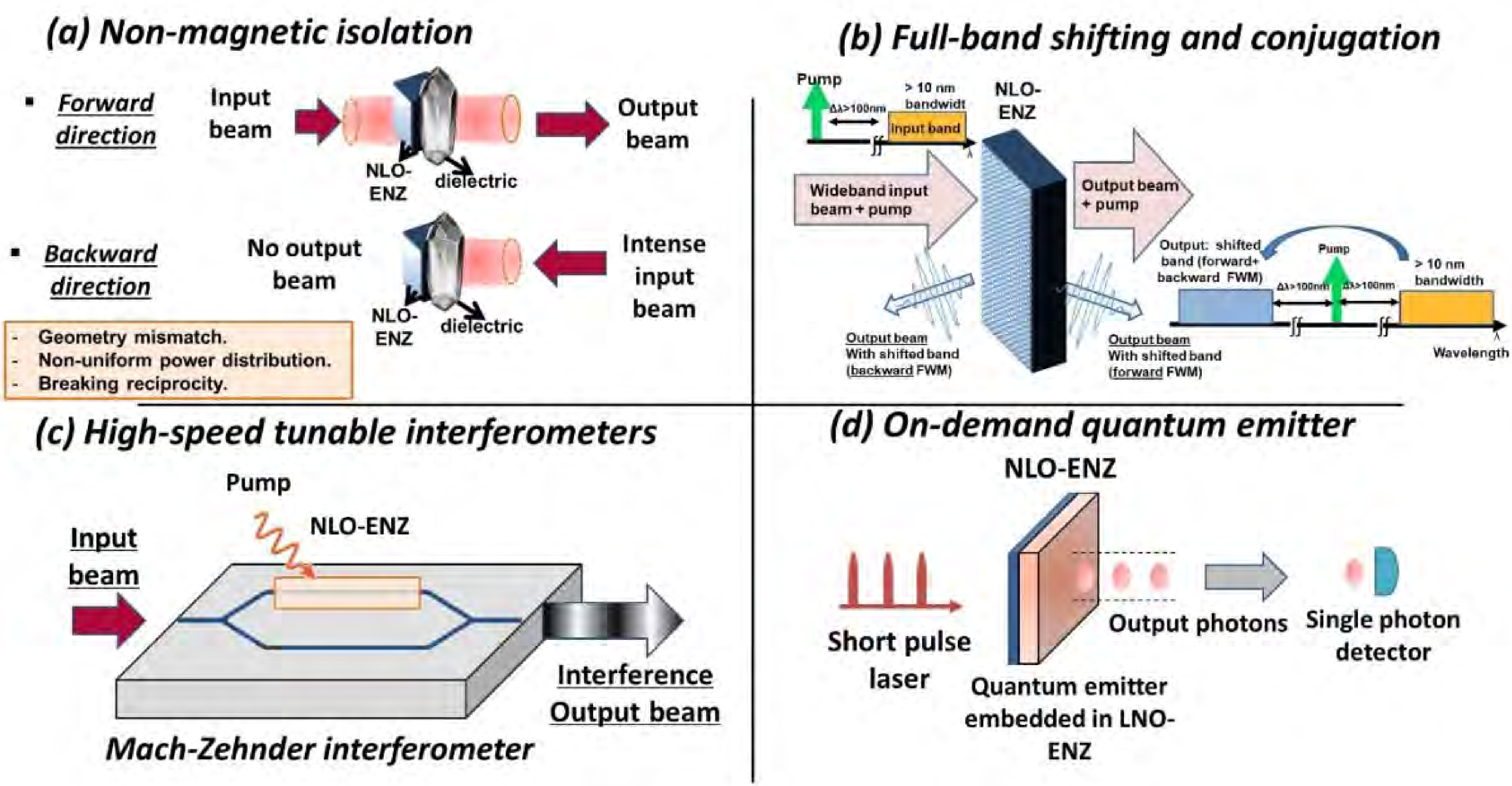
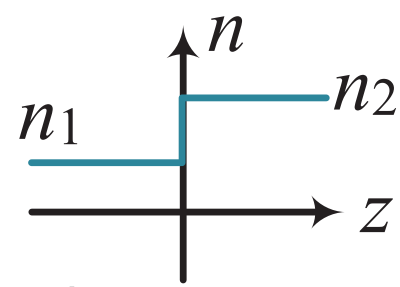
# Some Potential Applications of ENZ Behavior



# Adiabatic Wavelength Conversion through Time Refraction

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

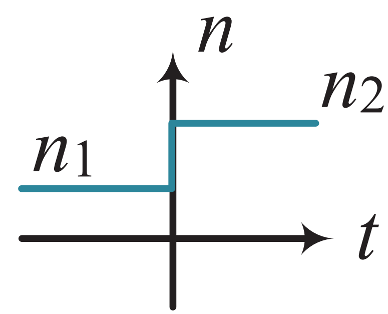
$$\frac{c}{f} = n \cdot \lambda \longrightarrow n_1\lambda_1 = n_2\lambda_2$$




index vs. position

- Time refraction (analog of Snell's law)

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



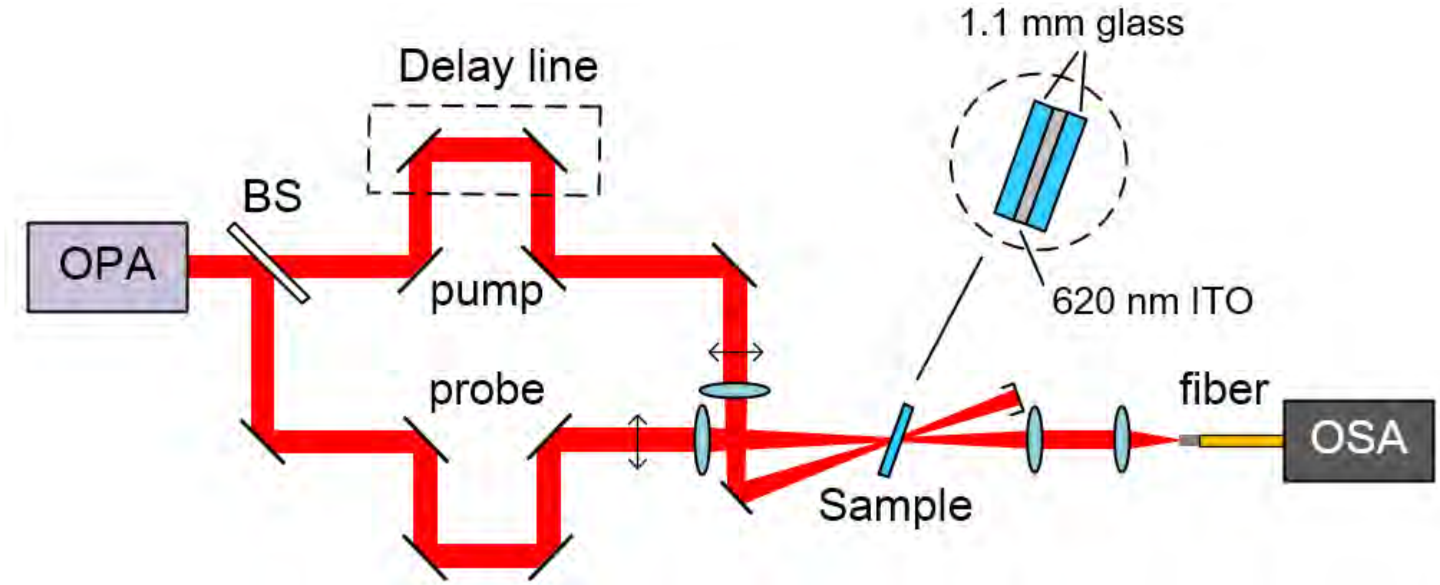

index vs. time

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:

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

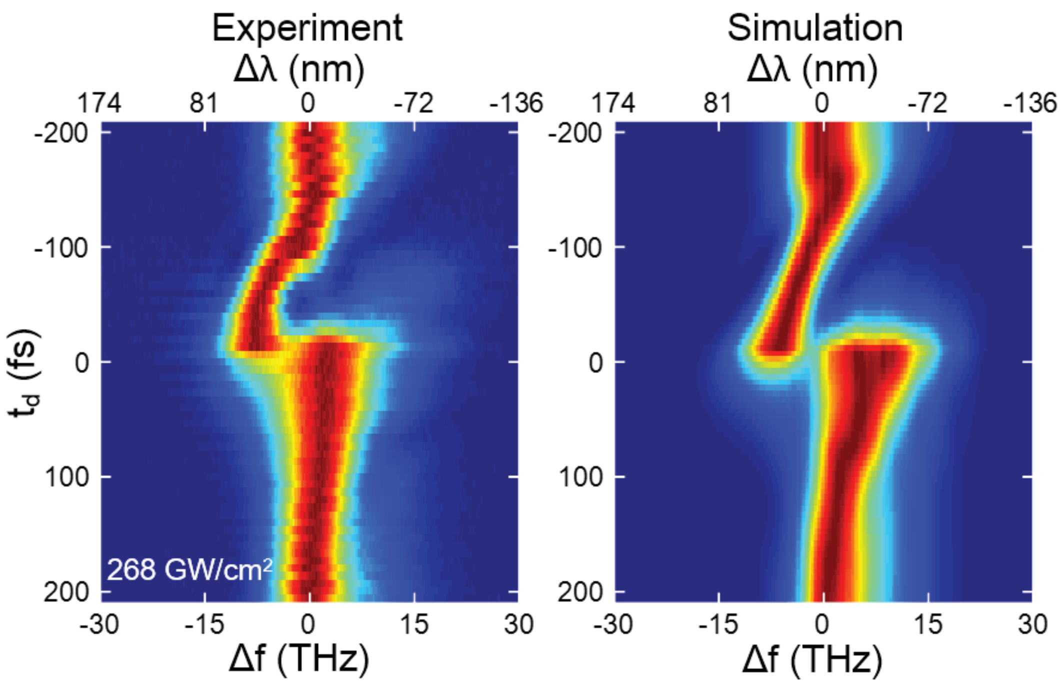
# Schematic of pump-probe experiment

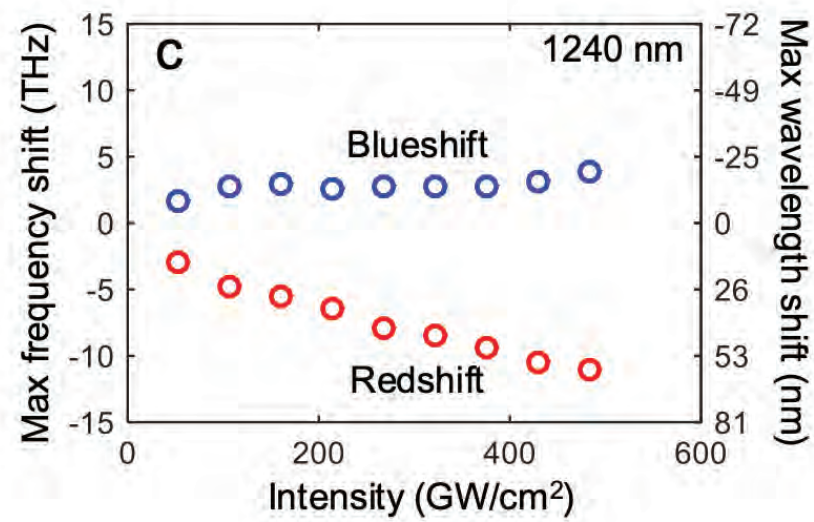



- Pump and probe beams are degenerate and *p*-polarized. Pulse width ~120 fs.
- Angle of incidence: pump 15 degrees, probe 10 degrees.
- Focused Airy disk diameter: pump ~90 um, probe ~30 um.

# Results: Adiabatic Wavelength Conversion by Time Refraction

Experimental results at 1240 nm




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

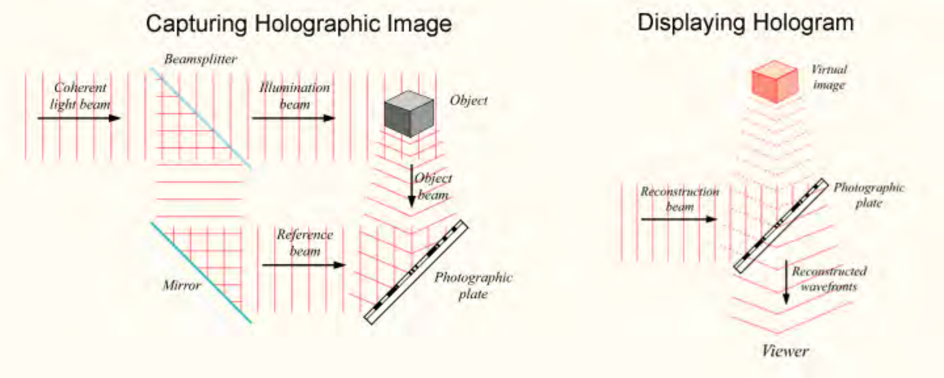
The wavelength shift can be controlled by the pump intensity and the sign of the time delay.

- The observed effect is 100 times larger than previous reports of AWC.
- Application: wavelength-division multiplexing for telecom

# Ultrafast Real-Time Holography with an Epsilon-Near-Zero Material

Can we perform holography at video (or faster) frame rates?

In optical holography an interference pattern is stored in a material to be read later by another light beam.



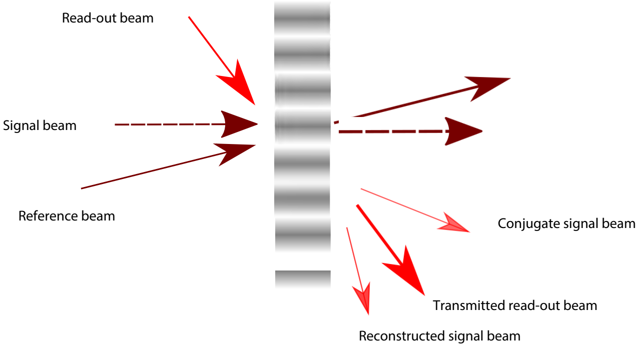

Gabor (1948); Leith and Upatnieks(1964).

Photorefractive and photochromatic materials are typical holographic materials.

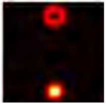
See also works by Lohman, Goodman, Yariv, and Peyghambarian.

**Standard holographic methods have very slow write rates.**

# Holography using ITO

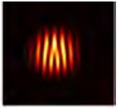



OAM beam



Gaussian beam

Transient hologram written on ITO

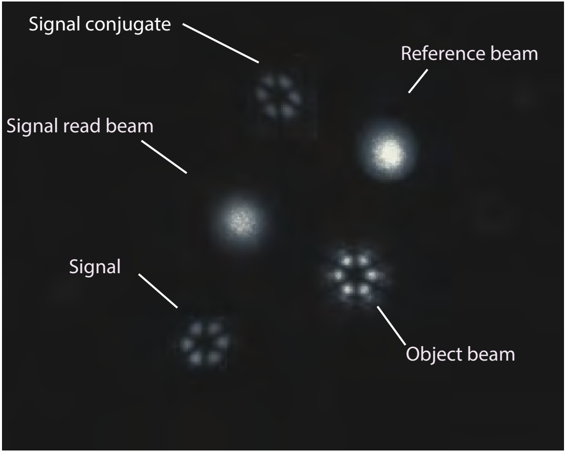


– The interference pattern gets written on the refractive index variation in ITO due to its large intensity-dependent changes in refractive index.

– A second gaussian beam of same or different wavelength can be used to read out the transient hologram.

# Holography Using ITO: Experimental Results

- Measured diffraction efficiency is 25%.
- 9-12 orders of magnitude larger refresh rate than conventional methods



# Special Thanks To My Students and Postdocs!

## Ottawa Group

## Rochester Group