







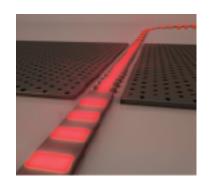
Physics and Applications of Epsilon-Near-Zero Materials

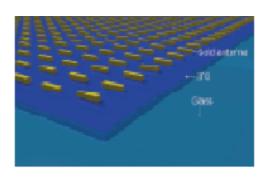
How Light Behaves When the Refractive Index Vanishes

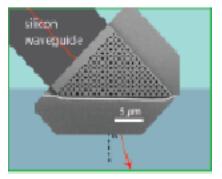
Robert W. Boyd

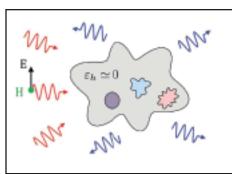
Department of Physics and
Max-Planck Centre for Extreme and Quantum Photonics
University of Ottawa

Institute of Optics and Department of Physics and Astronomy
University of Rochester









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

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

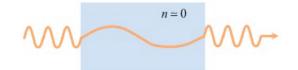
Recall that

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

where ϵ is the permittivity and μ is the magnetic permeability

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

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



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).

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).

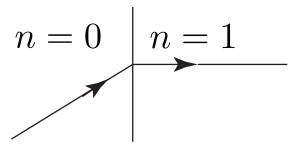
- Implications:
- If we can inhibit spontaneous emission, we can build thresholdless lasers.
- Expect superradiance effects to be pronounced in ENZ materials.

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.

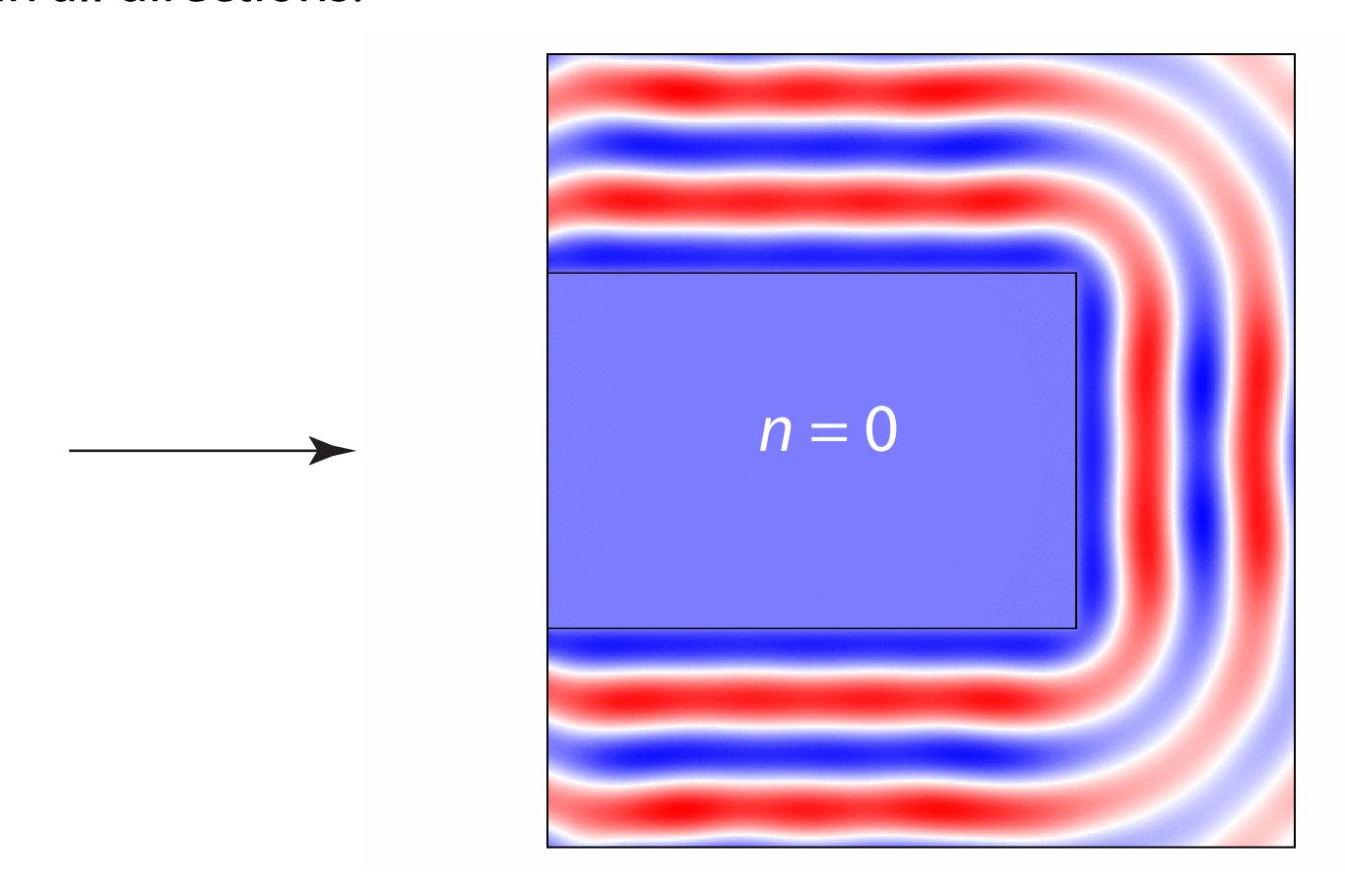
Thus light can enter an ENZ material only at normal incidence!



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

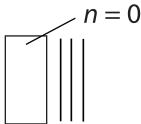
Maxwell Equations Prediction

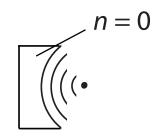
- light enters slab at normal incidence
- but leaves in all directions!



Some Consequences of ENZ Behaviour - 1

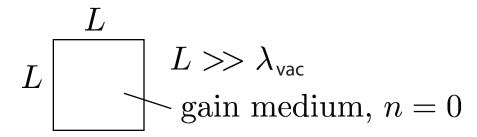
Funny lenses





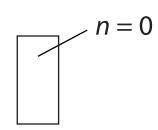
A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

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



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

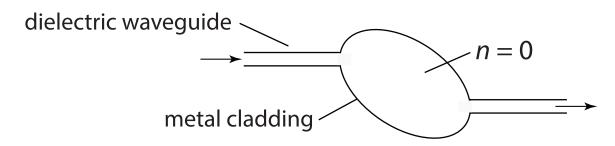
No Fabry-Perot interference



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

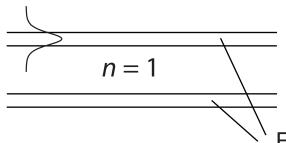
Some Consequences of ENZ Behavior - 2

• 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



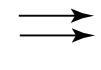
mode of upper waveguide extends to lower waveguide for any separation

ENZ (n = 0) waveguide

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

- Physics of ENZ Materials
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New Nonlinear Optical Material for Quantum Photonics

- 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 those previously reported (but with some background absorption).
- Material makes use of strong enhancement that occurs in the epsilon-near zero (ENZ) spectral region.
- 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).

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
$$\operatorname{Re} \epsilon = 0$$
 for $\omega = \omega_p / \sqrt{\epsilon_\infty} \equiv \omega_0$.

- Challenge: Obtain low-loss ENZ materials Want Im ϵ as small as possible at the frequency where Re $\epsilon=0$.
- 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 $\chi^{(3)}$

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

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

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

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

Also, the refractive index changes according to

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

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

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

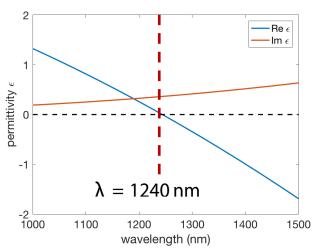
Note that $\operatorname{Re} \epsilon = 0$ for $\omega = \omega_p / \sqrt{\epsilon_\infty} \equiv \omega_0$.

The region near ω_0 is known as the epsilon-near-zero (ENZ) region.

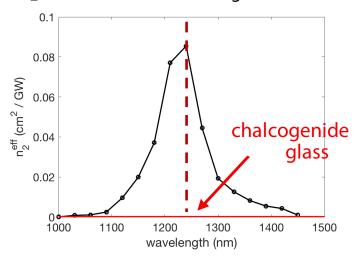
- There has been great recent interest in studies of ENZ phenomena:
- H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, Science 342, 1223 (2013).
- C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, Phys. Rev. B 85, 045129 (2012).
- S. Campione, D. de Ceglia, 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

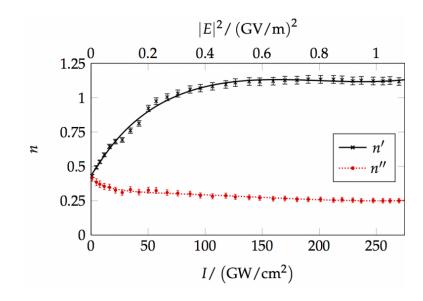




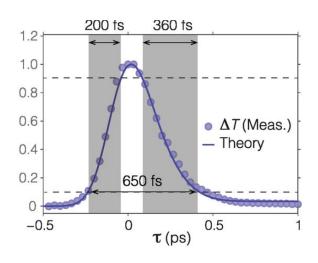
• n₂ can be 3.4 x10⁵ times larger than that of silica glass



• overall change in refractive index of 0.8



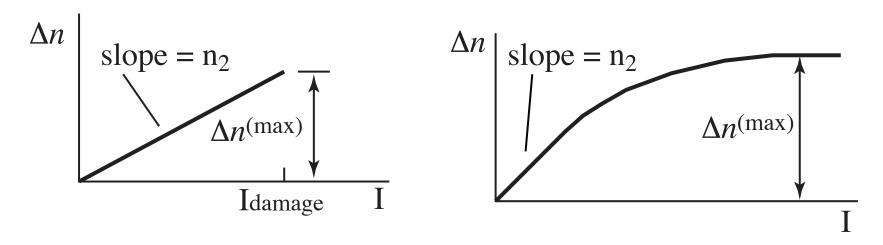
• sub picosecond reponse time



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

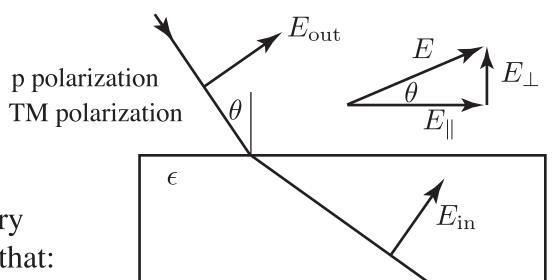
What Makes a Good (Kerr-Effect) Nonlinear Optical Material?

• We 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 $\Delta n^{(\max)}$



- For ITO at ENZ wavelength, both n_2 and $\Delta n^{(\text{max})}$ are extremely large $(n_2 = 1.1 \times 10^{-10} \text{ cm}^2/\text{W} \text{ and } \Delta n^{(\text{max})} = 0.8)$
- n_2 is 3.4 x 10⁵ times larger than that of silica glass $\Delta n^{(\text{max})}$ is 2700 times larger that that of silica glass (For silica glass $n_2 = 3.2$ x 10⁻¹⁶ cm²/W, $I_{\text{damage}} = 1$ TW/cm², and thus $\Delta n_{(\text{max})} = 3$ x 10⁻⁴)

NLO Response Increases 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

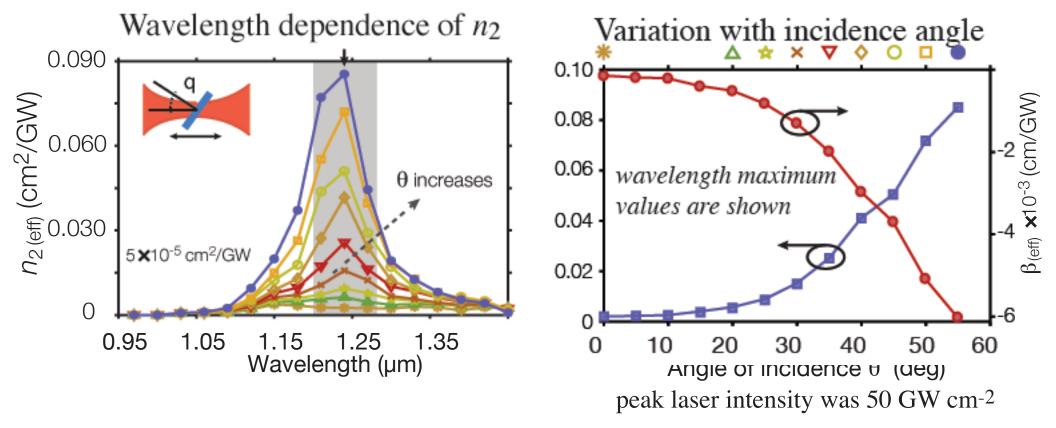
$$E_{\rm in} = E_{\rm out} \sqrt{\cos^2 \theta + \frac{\sin^2 \theta}{\epsilon}}$$

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

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

Huge Nonlinear Optical Response of ITO

• Z-scan measurements for various angles of incidence

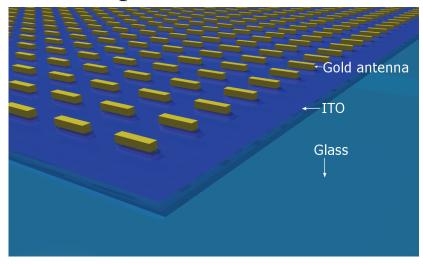


- Note that n_2 is positive (self focusing) and β is negative (saturable absorption).
- Both n_2 and nonlinear absorption increase with angle of incidence
- n_2 shows a maximum value of 0.11 cm²/GW = 1.1 × 10⁻¹⁰ cm²/W at 1.25 µm and 60 deg. This value is 2000 times larger than that away from ENZ region.

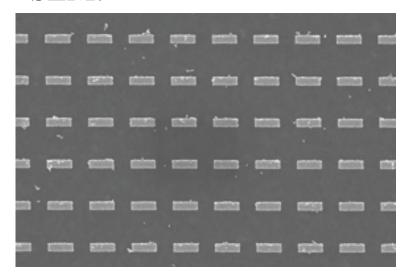
An ENZ Metasurface

- 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

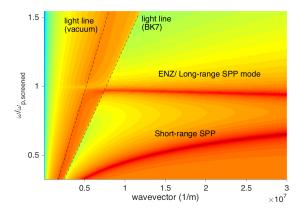
Concept:



SEM:



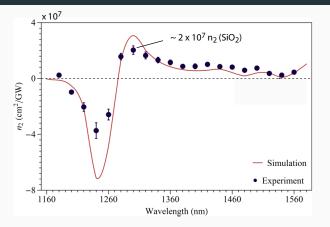
A thin ENZ medium supports a bulk plasma mode.



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,

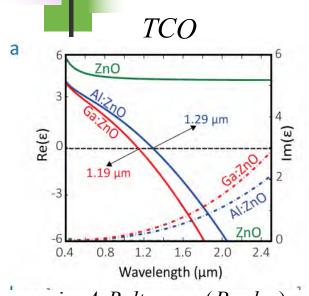
Alam, Schulz, Upham, De Leon and Boyd, Nature Photonics 12, 79-83 (2018).

Physics and Applications of Epsilon-Near-Zero Materials

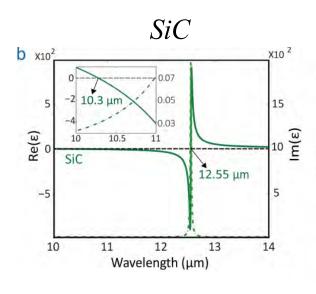
- Physics of ENZ Materials
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Epsilon-Near-Zero (ENZ) and Near-Zero-Index (NZI) Examples

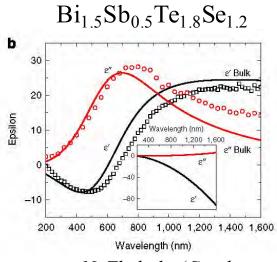




A. Boltasseva (Purdue) Kim et al., Optica (2016)

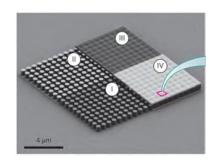


J. Caldwell (Vanderbilt) Kim et al., Optica (2016)

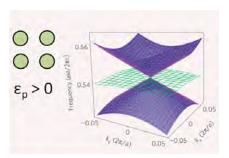


N. Zheludev(Southmapton)
Ou et al., Nat. Commun. (2014)

Metamaterials



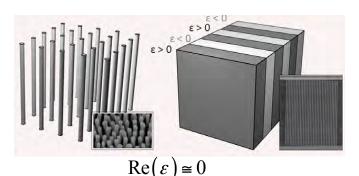
E. Mazur Li et al., Nat. Photon. (2015)



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



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



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

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

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

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

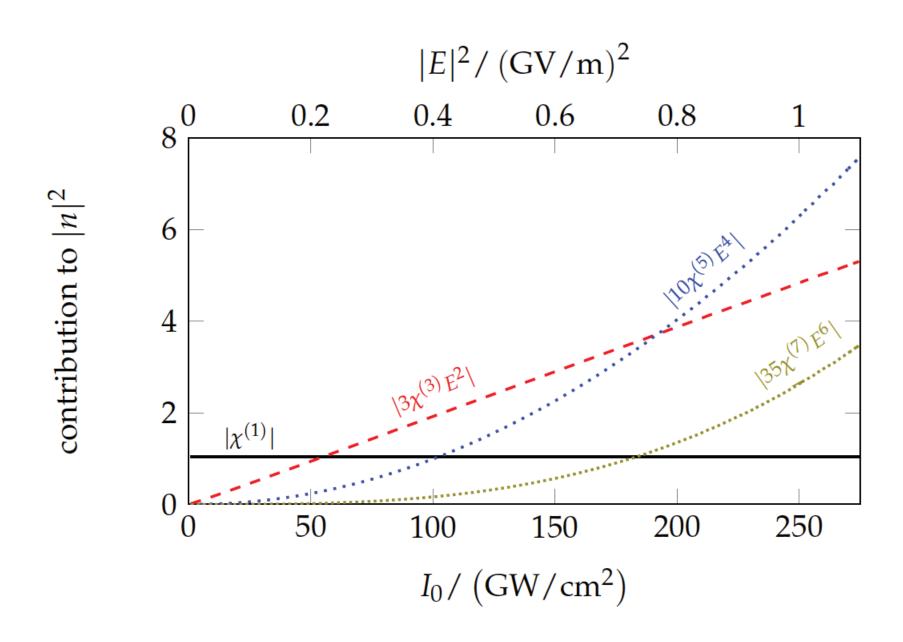
where

and

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

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

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



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

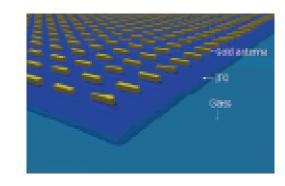


Alan Willner

- 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.

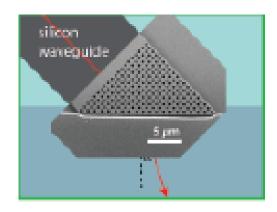
Three Material Platforms Under Investigation

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

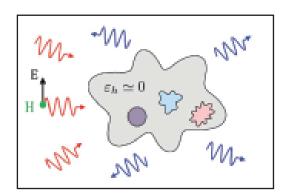


Dirac cone metamaterials

 (in plane; compatible with integrated optics)
 (Harvard)



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



Surface Reflection

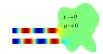
There is a problem getting light into a zero-index material.

Recall that the imdedance and surface reflectivity are given by

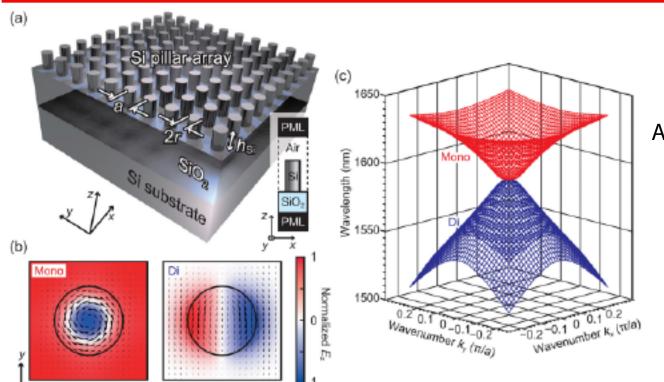
$$Z = \sqrt{\mu/\epsilon}$$
 $R = \left| \frac{Z-1}{Z+1} \right|^2$

Thus the reflectivity will be 100% if ϵ = 0 unless μ = 0 as well.





Dirac Cone Metamaterials



An EMNZ (epsilon and mu near zero) metamaterial

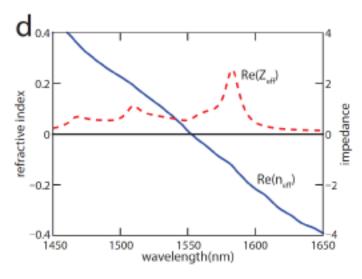
Opt Express 25, 8326 (2017)

It is also a ZIM (zero index material)

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

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

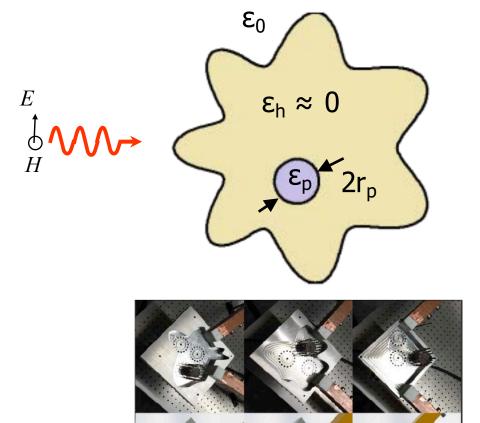
$$R = \frac{1 - Z}{1 + Z}$$





Photonic Doping of ENZ





$$\varepsilon_{\text{eff}} \simeq 0$$

$$\mu_{\text{eff}} = \frac{1}{A} \left[A_h + \frac{2\pi r_p}{k_p} \frac{J_1(k_p r_p)}{J_0(k_p r_p)} \right]$$

I. Liberal, A. Mahmoud, Y. Li, B. Edwards and N. Engheta, Science, 355, March 10, 2017

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Frequency conversion through time refraction using an epsilon-near-zero material

Yiyu Zhou¹, Mohammad Karimi², Jeremy Upham², Orad Reshef², Cong Liu³, Alan E. Willner³, M. Zahirul Alam², and Robert W. Boyd^{1,2}

¹The Institute of Optics, University of Rochester, Rochester, NY, 14627, USA
²Department of Physics, University of Ottawa, Ottawa ON K1N 6N5, Canada
³Department of Electrical Engineering, University of Southern California, Los Angeles, CA, 90089, USA

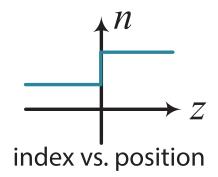




Space Refraction and Time Refraction

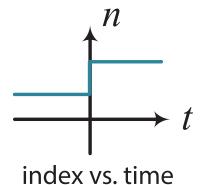
Space refraction (e.g., Snell's law)

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



Time refraction (analog of Snell's law)

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

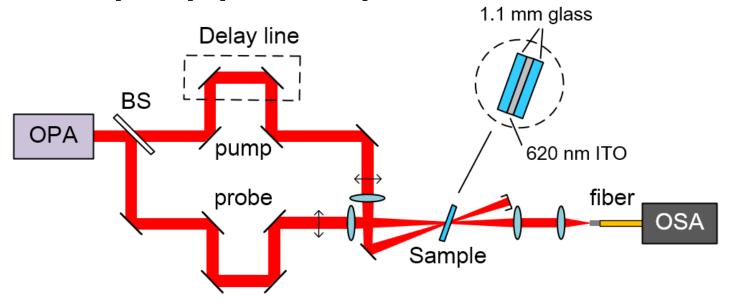


Photon frequency (energy) is changed because of the temporal change in index, but the wavelength (inverse of momentum) is conserved in the absence of any spatial asymmetry

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

$$\delta\omega(t) = \frac{d}{dt}\phi_{\rm NL} = \frac{d}{dt}[n_2I(t)\omega/c]$$

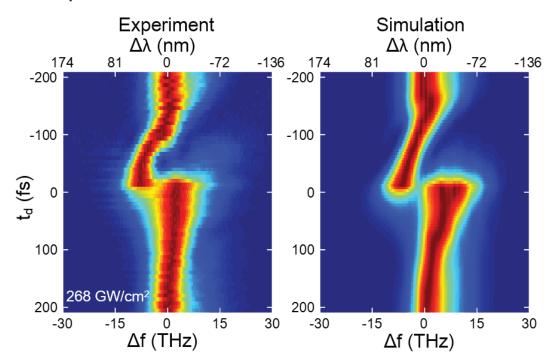
Schematic of pump-probe experiment



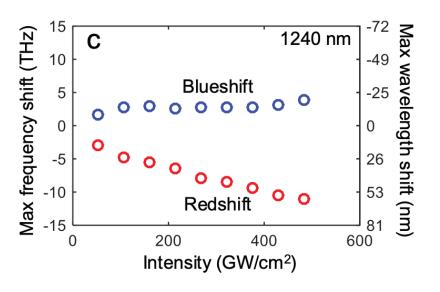
- Pump and probe beams are degenerate and p-polarized. Pulse width ~120 fs.
- Angle of incidence: pump 15 degrees, probe 10 degrees.
- Focused Airy disk diameter: pump ~90 um, probe ~30 um.

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 with almost 100 times smaller propagation distance than previous reports of AWC.
- Application: wavelength-division multiplexing for telecom

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

<u>M. Zahirul Alam</u>, Robert Fickler, Orad Reshef, Enno Giese, Jeremy Upham, Robert W. Boyd

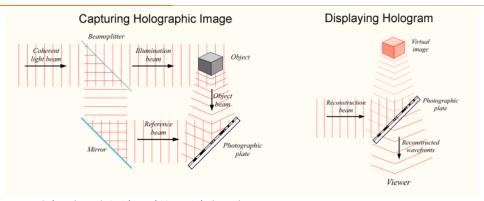
May 19, 2019

Department of Physics, University of Ottawa, Ottawa, Canada

Motivation:



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

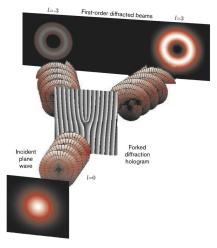


Gabor (1948); Leith and Upatnieks(1964).

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.

Orbital angular momentum (OAM) modes: a quick introduction.

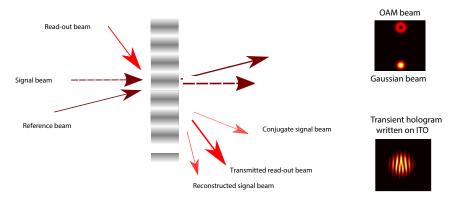


A forked diffraction hologram can convert a gaussian beam to an OAM-carrying beam. An OAM beam has an azimuthal phase factor of $\exp(il\phi)$

Orbital angular momentum (OAM) modes: a quick introduction.

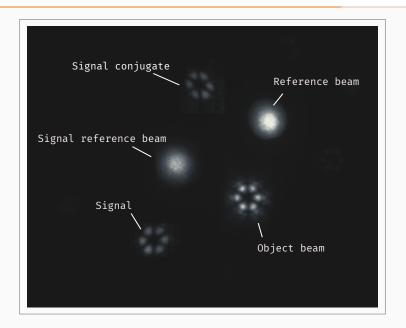


Holography using ITO



- 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.



Take home messages.

- The large nonlinear response of ENZ material can be exploited for real-time holography with an efficiency of 25%.
- The material is four orders of magnitude thinner (310-nm-thick) than a conventional holographic material.
- 9-12 orders of magnitude larger refresh rate. Limited by the sub-ps recovery time.
- Broadband response. 1000 1500 nm wavelength range with larger than 1 % diffraction efficiency.
- Might find applications in multimode communications and real-time signal processing such as edge detection, convolution, correlation, etc.
- Essentially we use structured light beams to temporally structure a surface to perform certain mathematical operations.

Special Thanks To My Students and Postdocs!

Ottawa Group



Rochester Group



Group ENZ Presentations at Photonics North

Zahirul Alam University of Ottawa,

Modification of the spontaneous emission rate of quantum emitters in near-zero refractive index media Date: Tuesday, May 21 Presentation time: 11:45 AM

Sisira Suresh University of Ottawa

Nonlinear optical properties of metal-dielectric stacks in their Epsilon-Near-Zero regime Date: Thursday, May 23 Presentation time: 09:30 AM

Ryan Hogan University of Ottawa; Institute of Optics, University of Rochester Tuning the dielectric constant zero crossing of vanadium dioxide (VO2) Date: Tuesday, May 21 Presentation time: 05:15 PM

Orad Reshef University of Ottawa

Waveguide-to-waveguide directional coupling beyond a free space wavelength Date: Wednesday, May 22 Presentation time: 02:30 PM

Mohammad Karimi University of Ottawa

Dynamic control of the reflection phase of a metasurface incorporating an epsilon-near-zero medium Date: Tuesday, May 21 Presentation time: 05:30 PM

Yiyu Zhou University of Ottawa

Adiabatic wavelength conversion in an epsilon-near-zero material by time refraction

Date: Tuesday, May 21 Presentation time: 05:00 PM

Additional Group Presentations

Alicia Sit University of Ottawa

The Ottawa River as an underwater communication channel

Date: Thursday, May 23 Presentation time: 04:25 PM Room: 207

Yiyu Zhou University of Rochester

Complete Characterization of Vector Beams Using Direct Measurement

Date: Wednesday, May 22 Presentation time: 08:45 AM Room: 2000 A

Samuel Lemieux University of Ottawa

Theory of Parametric Down-Conversion as an Absolute Standard for Radiometry

Date: Tuesday, May 21 Presentation time: 08:45 AM Room: 2101

Rioux University of Ottawa

Observing Spatial Quantum Correlations with an Electron-Multiplying Charge Coupled Device

Date: Thursday, May 23 Presentation time: 08:30 AM Room: 2000 B

Saad Bin Alam University of Ottawa

Multi-resonant high-Q Plasmonic Metasurface

Date: Wednesday, May 22 Presentation time: 04:45 PM Room: 2101

Payman Rasekh University of Ottawa

Modeling of Linear and Nonlinear Propagation of Broadband THz Pulses

Date: Tuesday, May 21 Presentation time: 03:35 PM Room: 2104 AB

Boris Braverman University of Ottawa

Nonlocal Quantum Aberration Cancellation

Date: Wednesday, May 22 Presentation time: 08:30 AM Room: 2000 A!

My Group's Publications on ENZ

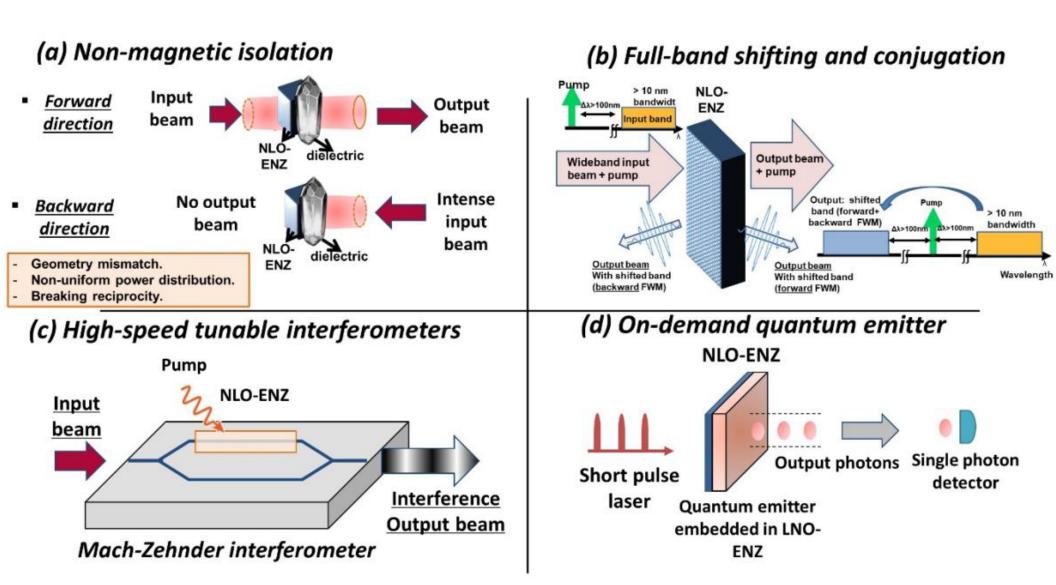
- Large Optical Nonlinearity of Indium Tin Oxide in its Epsilon-Near-Zero Region, M. Z. Alam, I. D. Leon, and R. W. Boyd, Science 352, 795 (2016).
- Optical Response of Dipole Antennas on an Epsilon-Near-Zero Substrate, S. A. Schulz, A. A. Tahir, M. Z. Alam, J. Upham, I. De Leon, and R. W. Boyd, Phys. Rev. A 93, 063846 (2016).
- Beyond the Perturbative Description of the Nonlinear Optical Response of Low-Index Materials, O. Reshef, E. Giese, M. Z. Alam, I. D. Leon, J. Upham, and R. W. Boyd, Opt. Lett. 42, 3225 (2017).
- Large Optical Nonlinearity of Nanoantennas Coupled to an Epsilon-Near-Zero material, M. Zahirul Alam, S. A. Schulz, J. Upham, I. De Leon and R. W. Boyd, Nature Photonics 12, 79–83 (2018).
- Nonlinear Optical Effects in Epsilon-Near-Zero Media, O. Reshef, I. De León, M. Z. Alam, and R.W. Boyd, Nat. Rev. Mat. in press (2019).

Summary: Physics and Applications of ENZ Materials

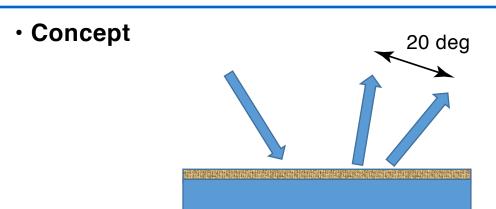
- Extremely interesting physical processes occur in ENZ materials
- ENZ materials, metamaterials, and metastructures display extremely large NLO response
- The huge, ultrafast NLO response of ENZ materials lend themselves to many important applications

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

Some Potential Applications of ENZ Behavior



All-Optical, Nanoscale, Sub-Picosecond Beam Steering

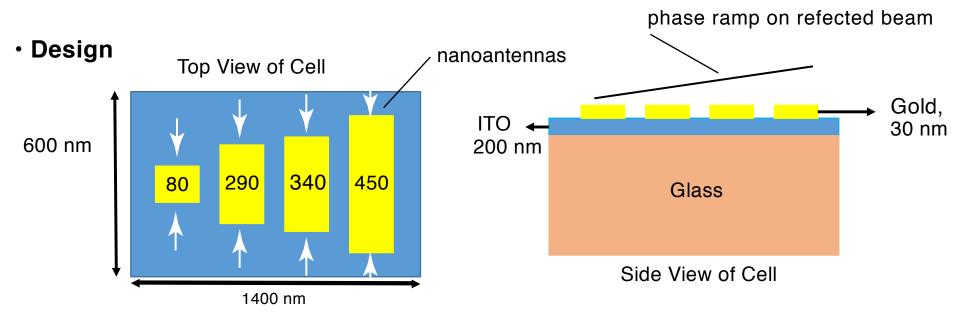


Vary output direction by +/- 20 degrees under all-optical control

Sub-picosecond response time

Beam steerer made of one or many cells

Application: Mode-division multiplexing for telecommunications



Nonlinear response depends on antenna length

Characterization

We have fabricated this design and are currently testing it