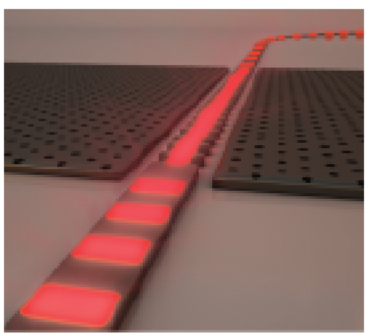
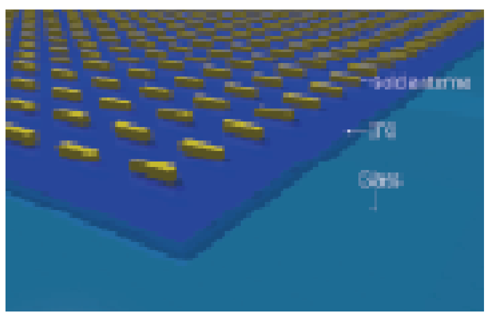
Nonlinear Photonics with Low-Index Materials

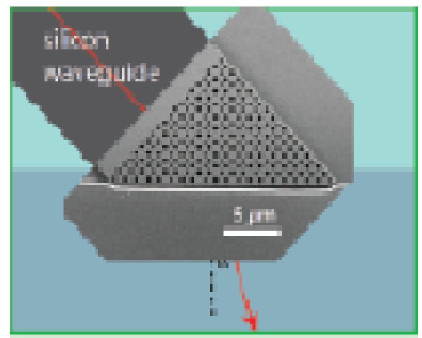
Robert W. Boyd

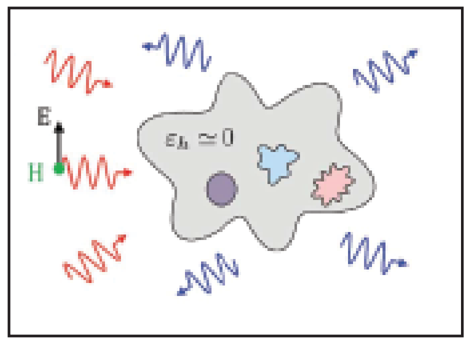
Department of Physics and
Max-Planck Centre for Extreme and Quantum Photonics
University of Ottawa

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









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

Presented at the OSA Laser Congress 2020, October 13, 2020.

Implications of Low-Index Behavior for Nonlinear Optics

Here is the intuition for why the low-index 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 low-index 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 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)

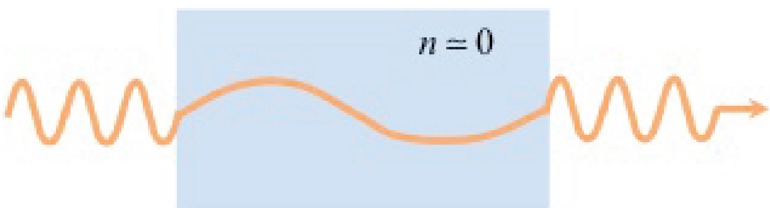
- Our approach: Use epsilon-near-zero (ENZ) materials and metamaterials

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 radiation 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:

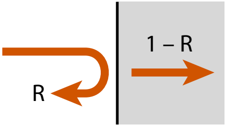
  ENZ: epsilon near zero
  MNZ: mu near zero
  EMNZ: epsilon and mu near zero

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 (with ϵ/μ finite).



- 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 = $1/A$)

$A = n\, A_{\mathrm{vac}}$

We can control (inhibit!) spontaneous emission!

Einstein *B* coefficient

Stimulated emission rate = *B* times EM field energy density

$B = B_{\mathrm{vac}} \,/\, (n\, n_{\mathrm{g}})$

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:

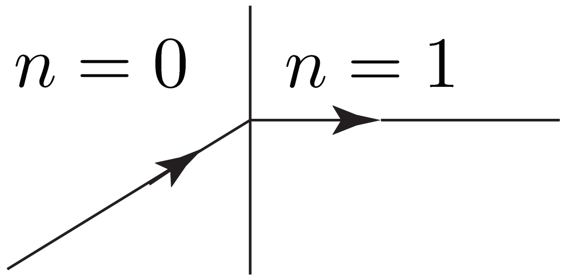
- If we can inhibit spontaneous emission, we can build thresholdless lasers.
- Expect superradiance effects to be pronounced in ENZ materials.

Optics of Zero-Index Materials

- Snell's law leads to intriguing predictions

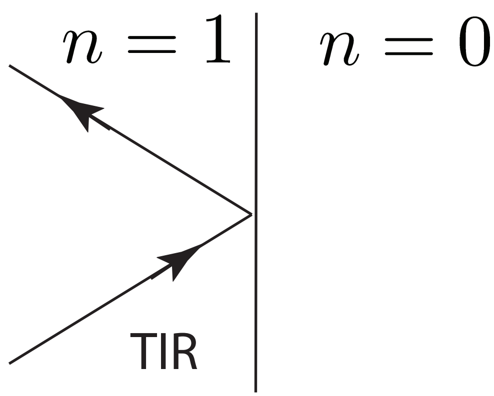
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

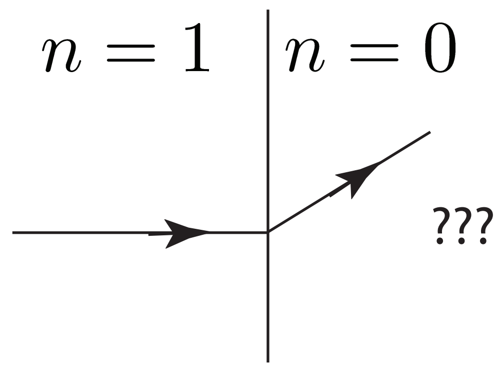
- Light always leaves perpendicular to surface of ENZ material!

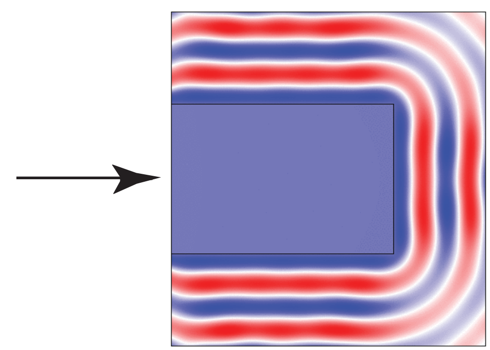


Y. Li, et al., Nat. Photonics 9, 738, 2015; D. I. Vulis, et al., Opt. Express 25, 12381, 2017.

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







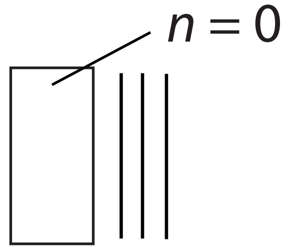
Light enters at normal incidence but leaves in all directions.

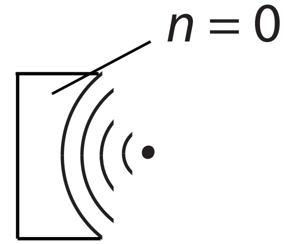
Y. Li, et al., Nat. Photonics 9, 738, 2015.

(wave-optics simulation - O. Reshef)

Some Consequences of ENZ Behavior - 1

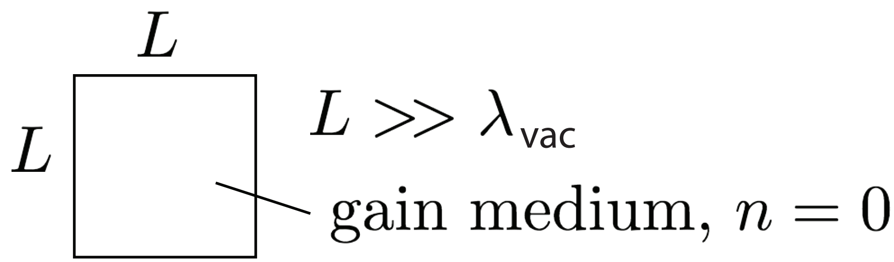
- “Funny” lenses





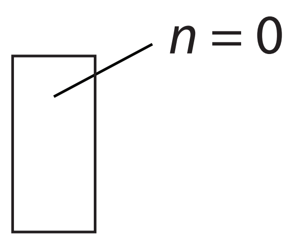
A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

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



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

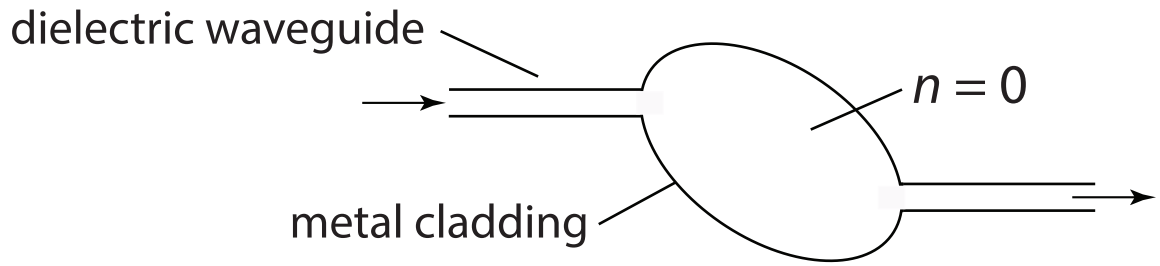
- No Fabry-Perot interference



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

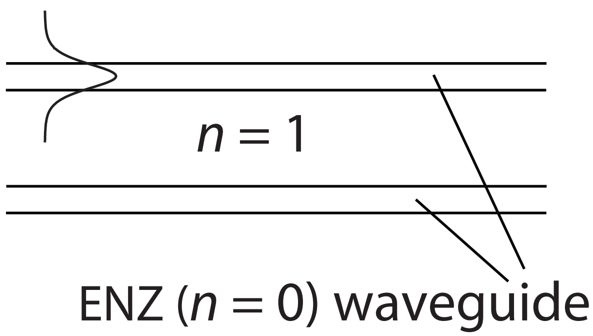
Some Consequences of ENZ Behavior - 2

- 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.

- 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.

How Light Behaves when the Refractive Index Vanishes

- Physics of Near-Zero Index (NZI) Materials
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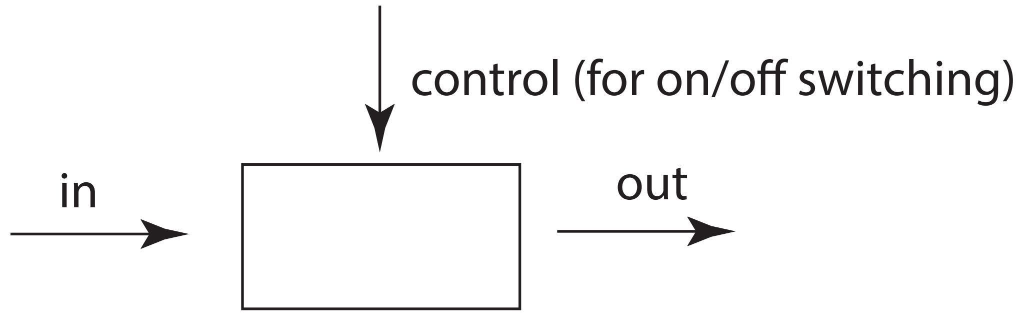
Nonlinear Optics and Optical Switching

- An important application in photonic technologies is optical switching.



- 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 $\chi^{(3)}$

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Also, the refractive index changes according to

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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.

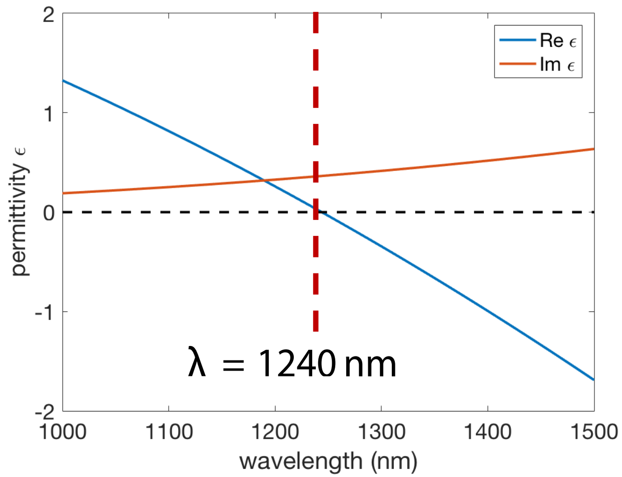
$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

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

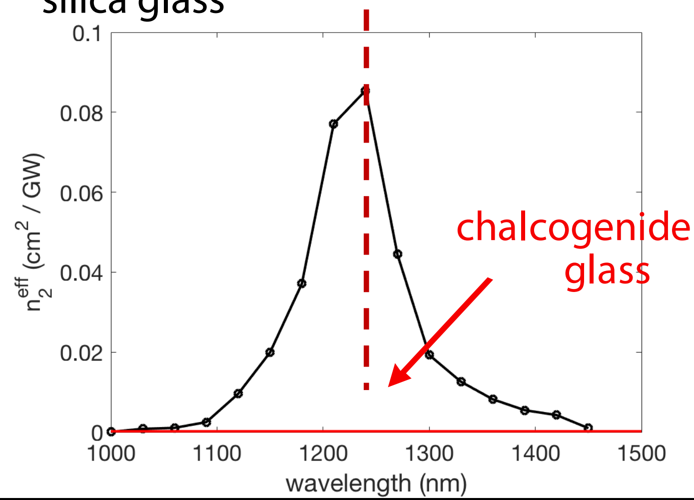
1. Alam, De Leon and Boyd, Science 352, 795–797 (2016)
2. Caspani, Shalaev, Boltasseva, Faccio et al., Phys. Rev. Lett. 116, 233901 (2016).

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

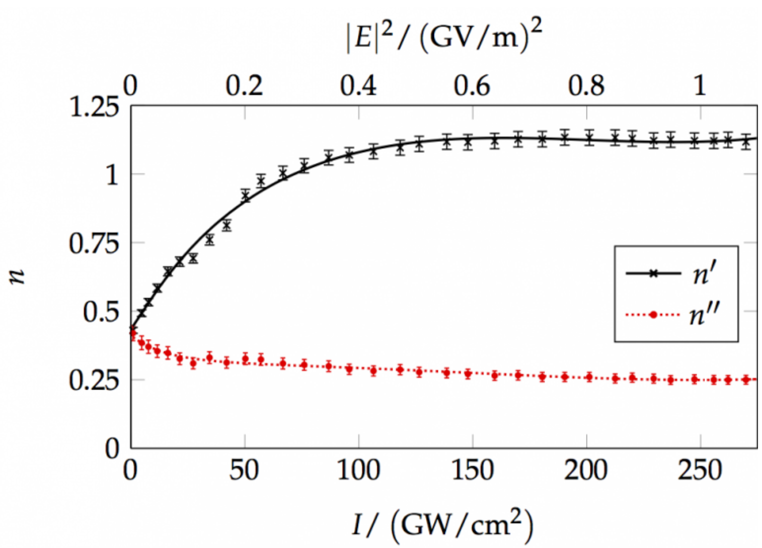
- ellipsometry



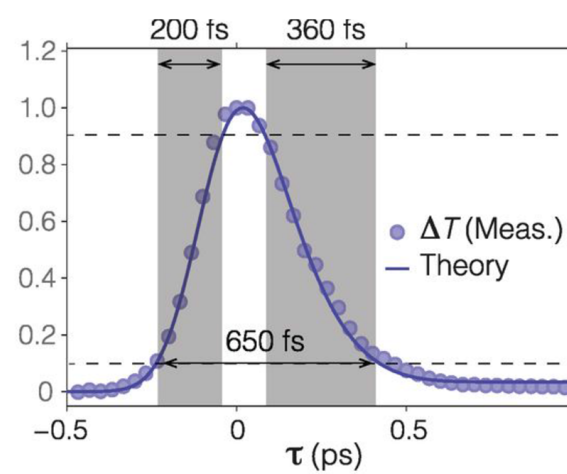
- n2 can be 3.4 x10^5 times larger than that of silica glass



- overall change in refractive index of 0.8

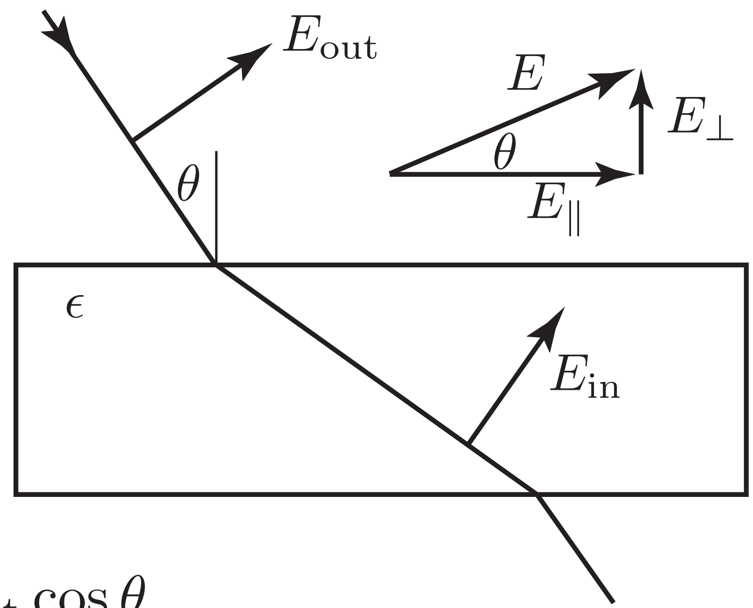


- sub picosecond reponse time



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

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

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

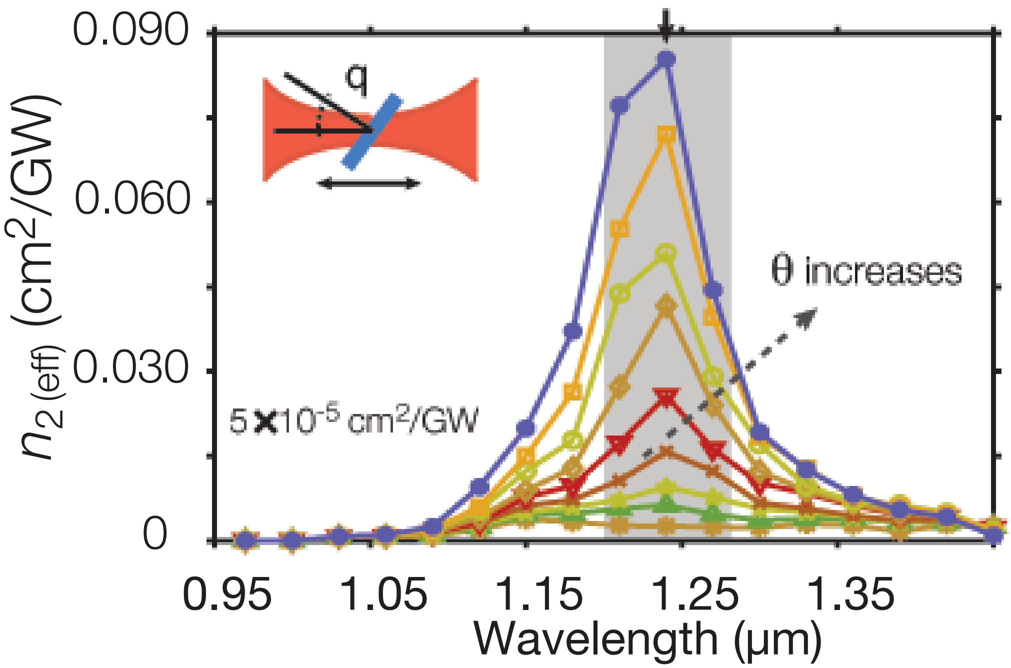
Note that, for $\epsilon < 1$, E_{in} exceeds E_{out} for $\theta \neq 0$.

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

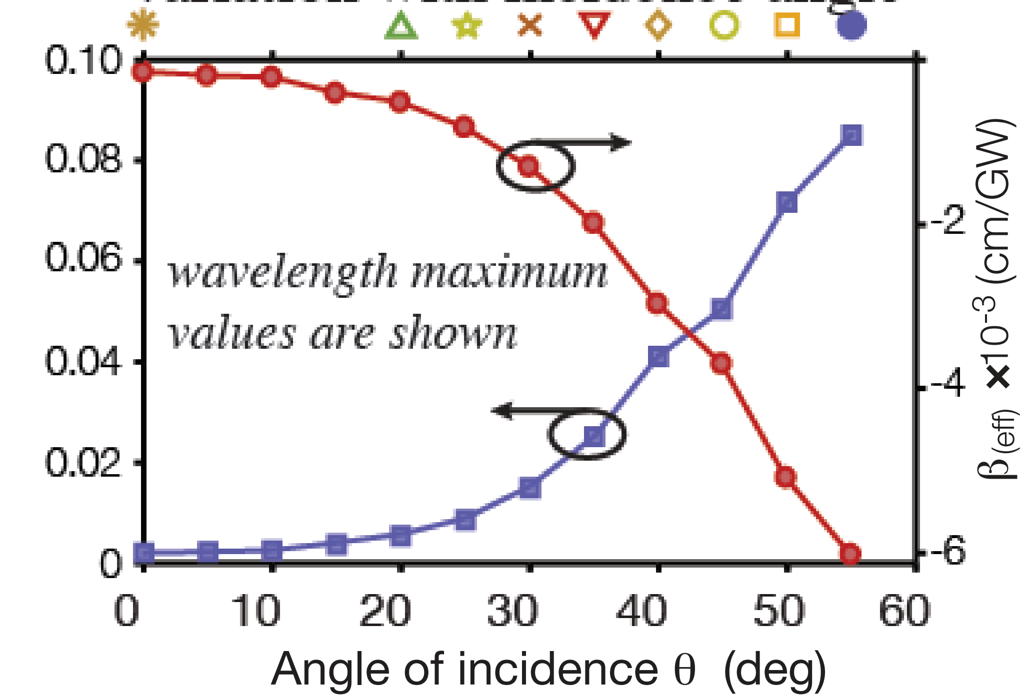
Huge Nonlinear Optical Response of ITO

- Z-scan measurements for various angles of incidence

Wavelength dependence of n_2



Variation with incidence angle



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 cm^2/GW = 1.1×10^{-10} cm^2/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 the form of n_2 $$n_2 = \frac{3\chi^{(3)}}{4\epsilon_0 c\, n_0 \,\mathrm{Re}(n_0)}$$

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

then: $$\Delta n = \frac{\Delta\epsilon}{2n_b}$$

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. When a collimated laser beam enters a material its intensity remains constant. Recall that

$$I = \tfrac{1}{2} n \epsilon_0 c\, |E|^2$$

Thus for small *n* the electric field strength is increased, giving a large nonlinear response

5. Detailed numerical integration confirms this behavior.

Dependence of Second-Harmonic Generation on the Linear Dielectric Permittivity

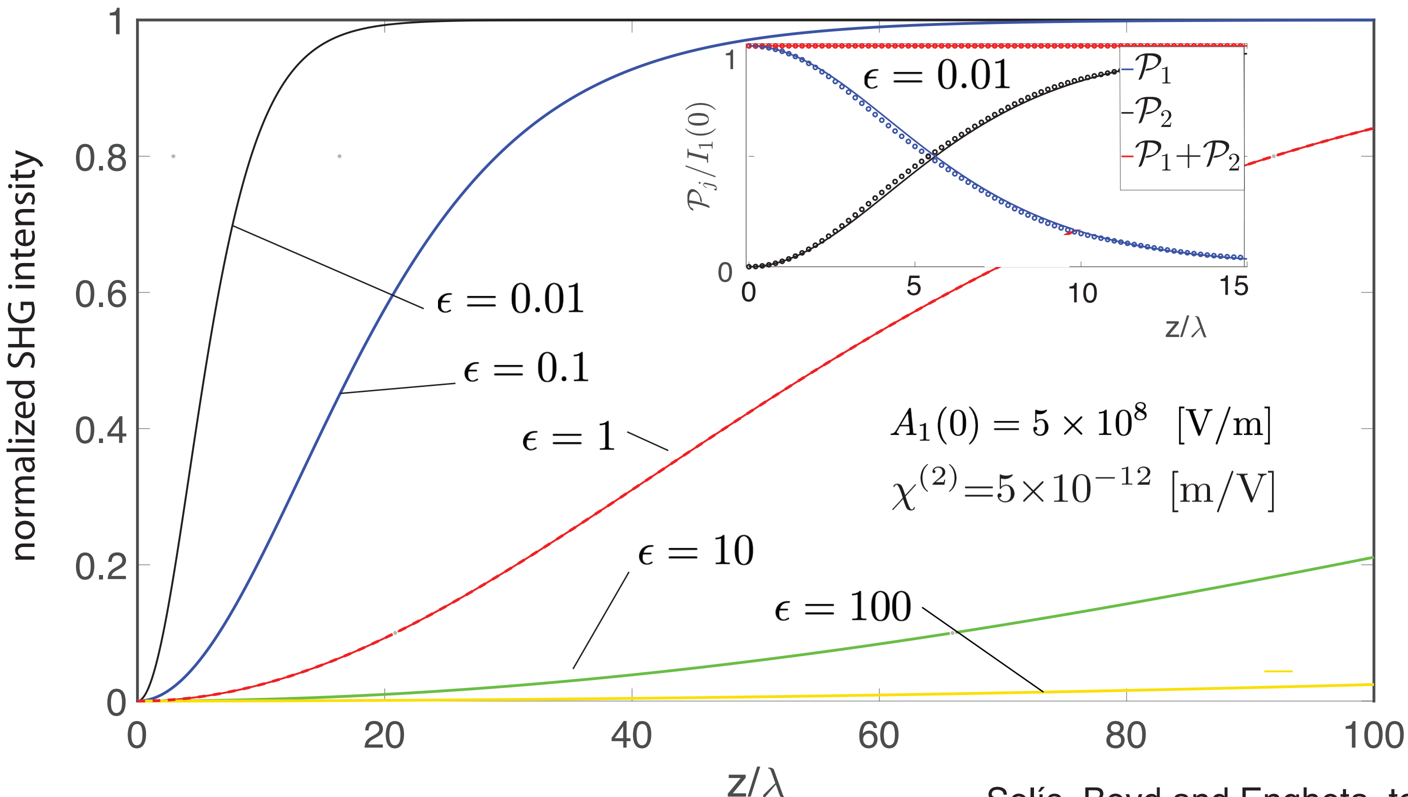
- We solve the standard equations for second-harmonic generation

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

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

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

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



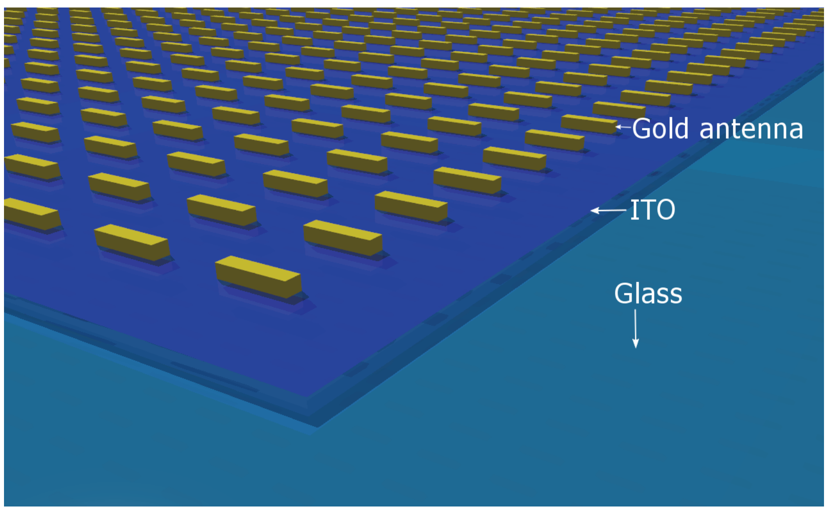
Solís, Boyd and Engheta, to be published

See also Solís, CLEO Poster JW2D.15

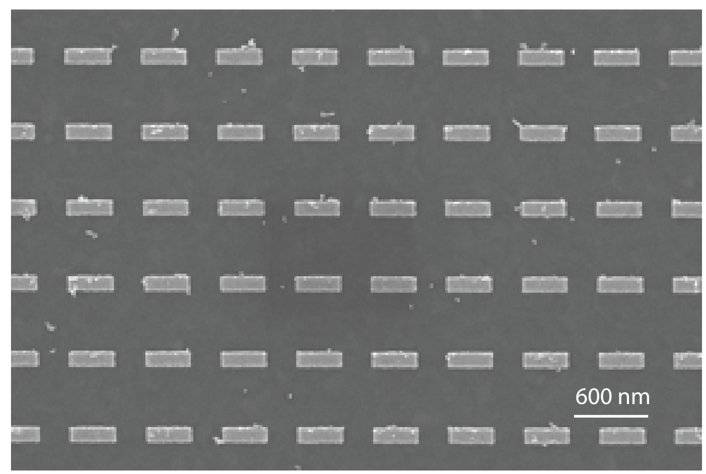
An ENZ Metasurface

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

Concept:

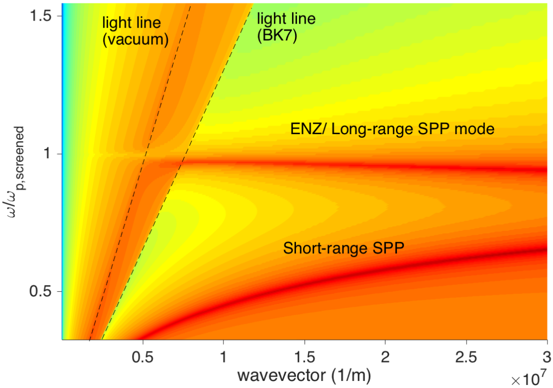


SEM:



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

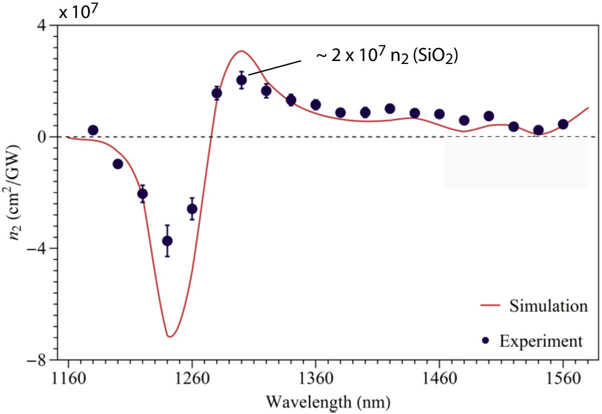
A thin ENZ medium supports a bulk plasma mode.



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 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).

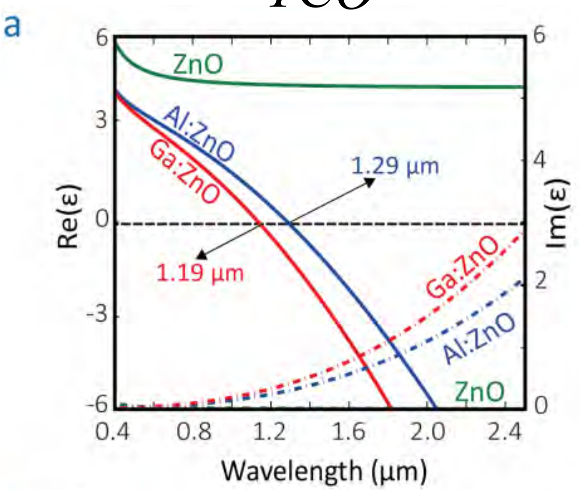
Physics and Applications of Epsilon-Near-Zero Materials

- 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

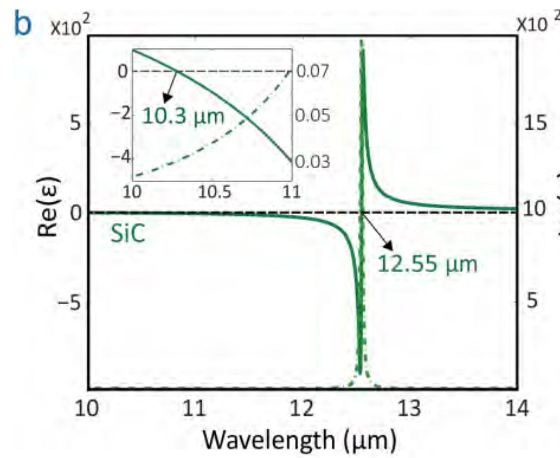
Homogeneous materials

TCO



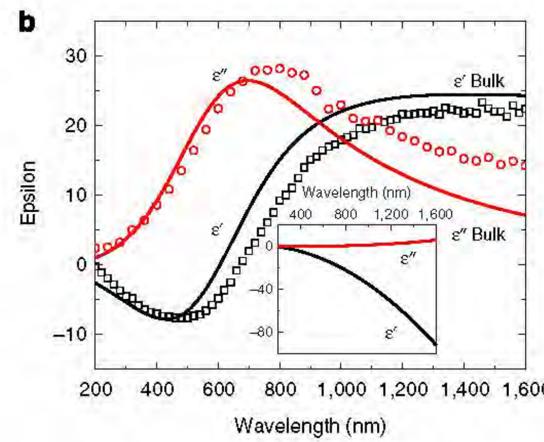
A. Boltasseva (Purdue)
Kim et al., Optica (2016)

SiC



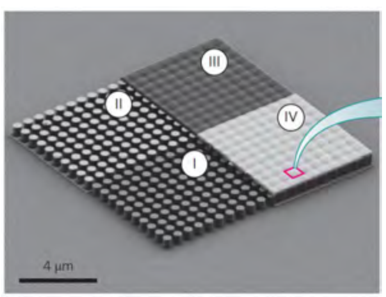
J. Caldwell (Vanderbilt)
Kim et al., Optica (2016)

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

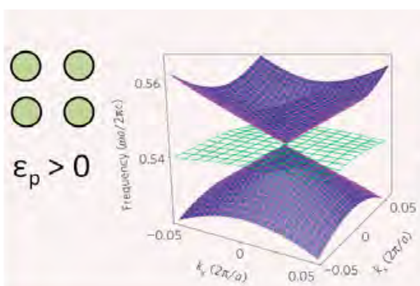


N. Zheludev (Southmapton)
Ou et al., Nat. Commun. (2014)

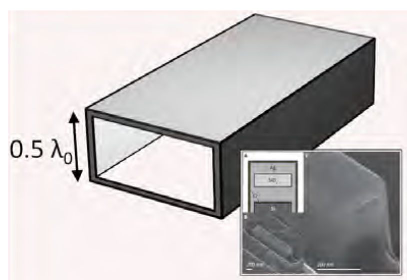
Metamaterials



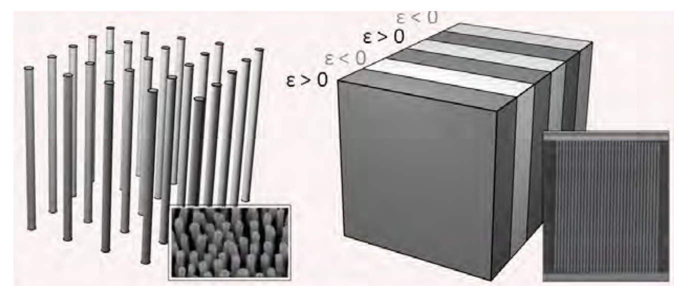
E. Mazur Li et al.,
Nat. Photon. (2015)



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



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



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

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

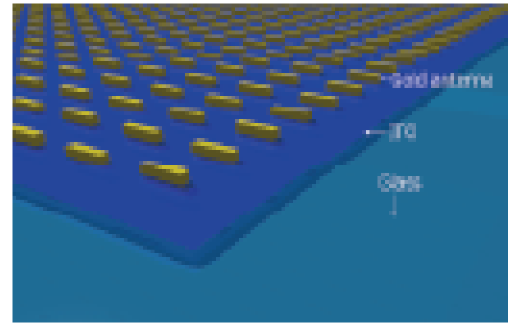
- Balkanski Professor of Physics and Applied Physics at Harvard University
- Ph.D. University of Leiden.
- Activities include light-matter interactions with ultrashort laser pulses, nonlinear optics at the nanoscale, and zero-index dielectric metamaterials.
- Awards include the Beller Award of OSA and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

Alan Willner

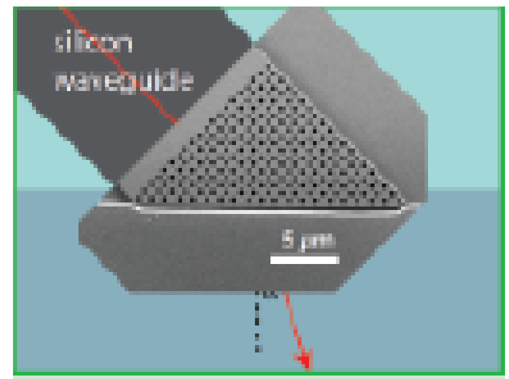
- Steven & Kathryn Sample Chair in Engineering at the University of Southern California.
- Ph.D. Columbia University
- Honors include Member of US National Academy of Engineering; Int'l Fellow of UK Royal Academy of Engineering; President of OSA and of IEEE Photonics Society.
- Activities include using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

Three Metamaterial Platforms Under Investigation

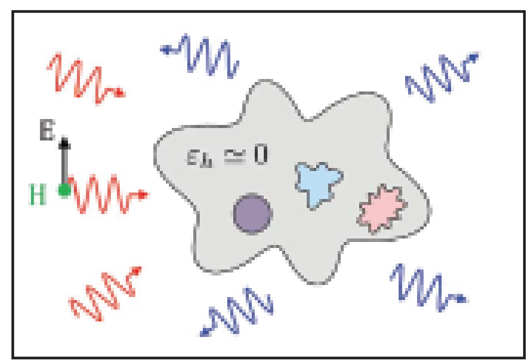
• Nanoantennas coupled to ENZ substrate
(out of plane; free-space coupling)
(Rochester)



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



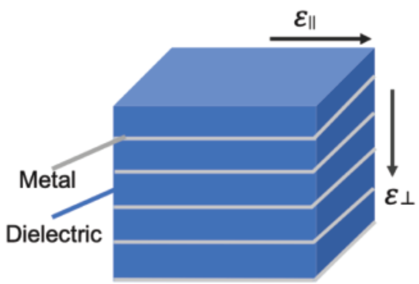
• Photonically doped metamaterials
(out of plane; free-space coupling)
(Penn)

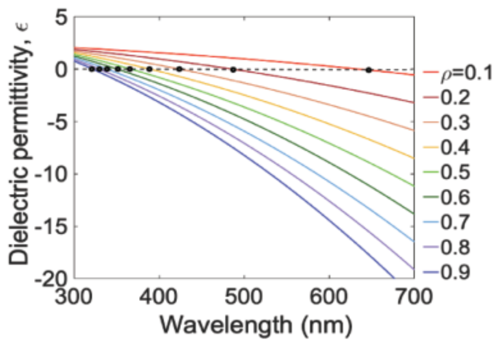


Nonlinear Optical Properties of a Layered Metamaterial in its ENZ Region

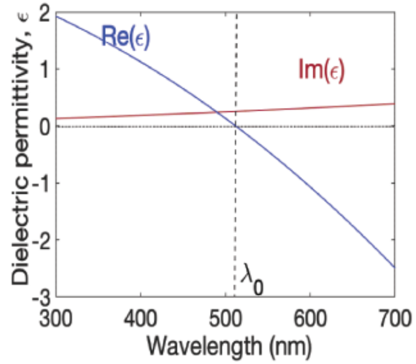
Do layered metamaterials also show enhanced NLO response at ENZ wavelength?

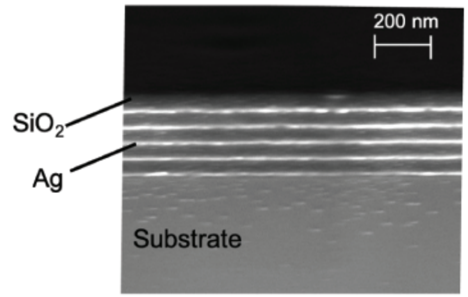
- By controlling the metallic fill fraction ρ, we can set the ENZ wavelength to be anywhere from 300 to 700 nm. We use ρ = 0.2, which corresponds to 500 nm. We deposit five layer pairs



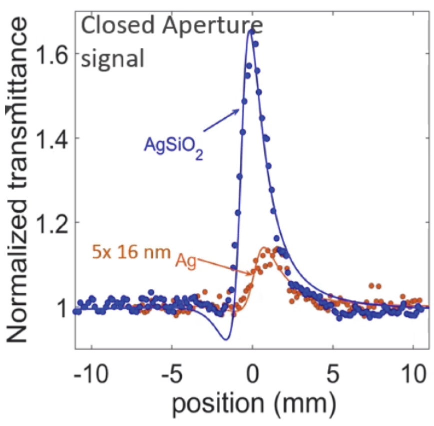


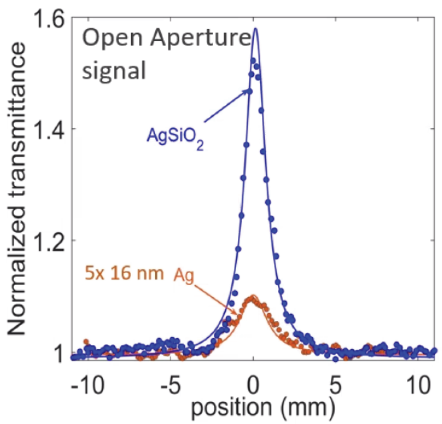
- Note that the real part of epsilon vanishes at 508 nm, close to the design wavelength. The SEM shows our structure. Ag thickness = 16 nm; silica thicness = 65 nm



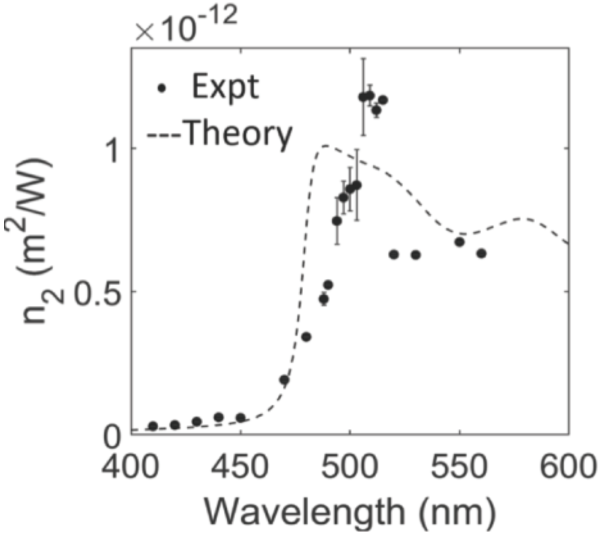


- We perform Z-scan measurements on the sample. Note the enhanced response of the composite as compared to a single layer of silver.





- 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.



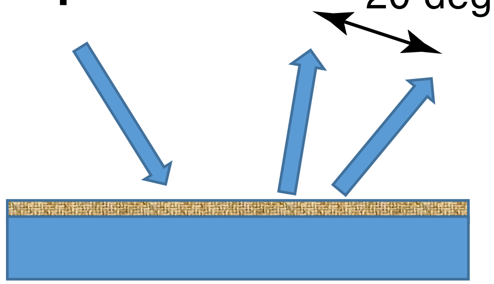
Suresh, Reshef, Alam, Upham, Karimi and Boyd

Physics and Applications of Epsilon-Near-Zero Materials

- Physics of ENZ Materials
- Huge NLO Response of ITO and ITO Metastructures
- Materials for ENZ
- Applications of ENZ Materials

All-Optical, Nanoscale, Sub-Picosecond Beam Steering

- **Concept**



Tune output direction by
+/- 20 degrees under
optical or electrical control

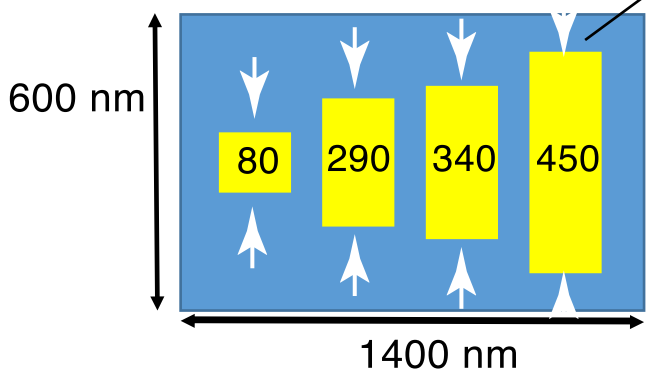
Beam steerer mcan be made
of one or many cells

Sub-picosecond response time

- **Design**

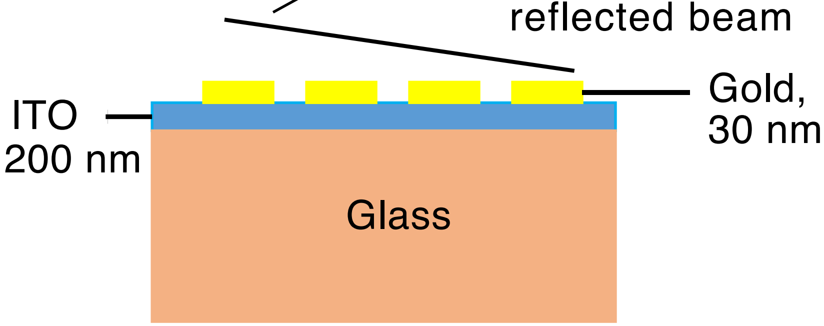
Top View of Cell

nanoantennas



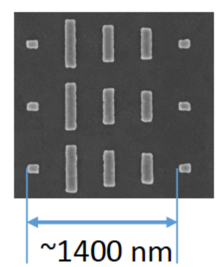
Nonlinear response
depends on
antenna length

phase ramp on
reflected beam

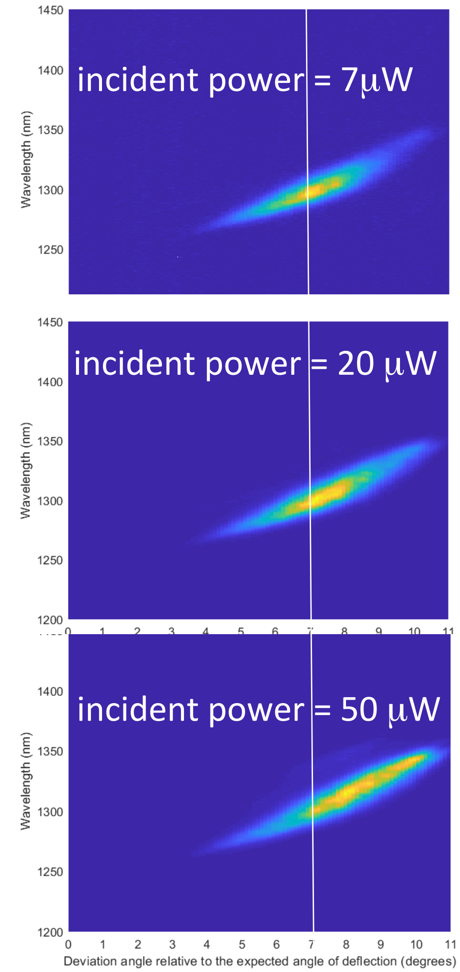


Side View of Cell

- SEM

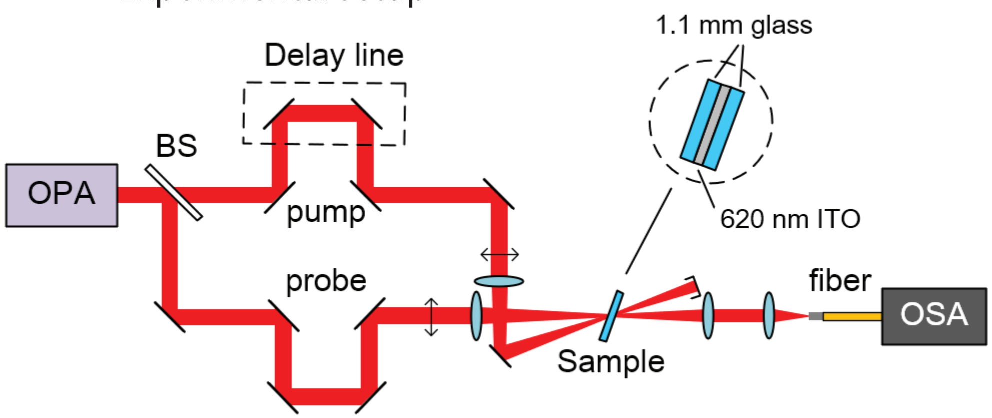


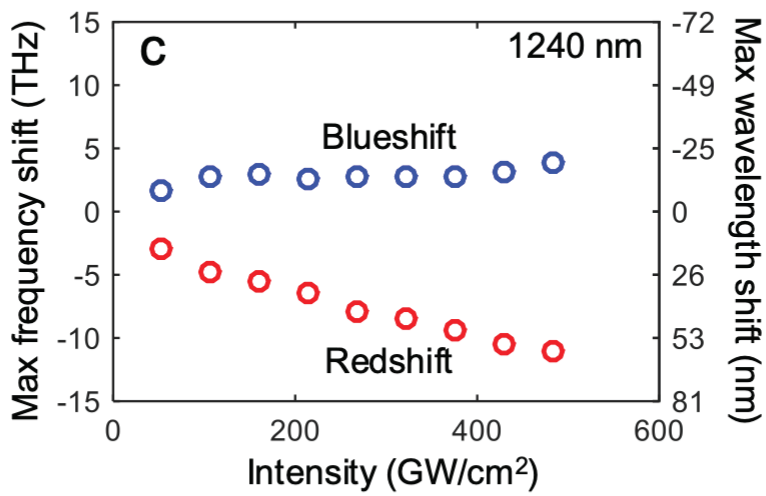
- **First results**

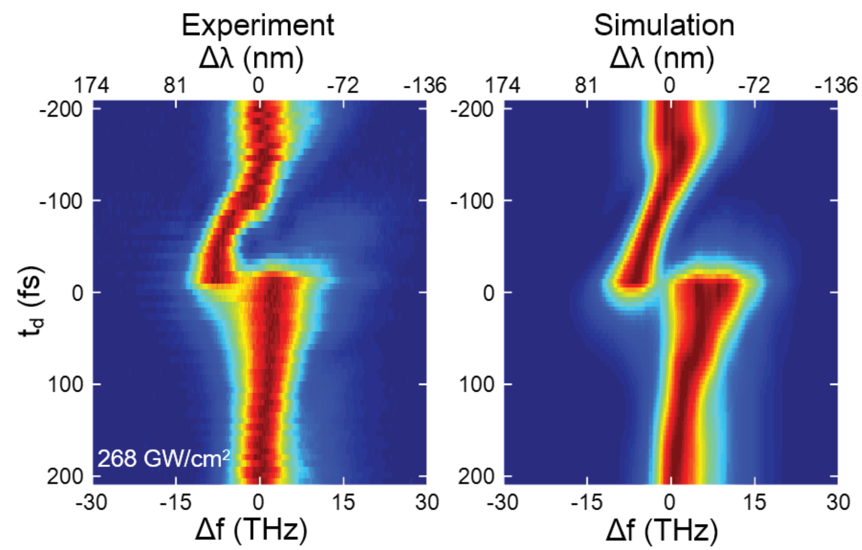


One application: Mode-division multiplexing
for telecommunications

Adiabatic Wavelength Conversion by Time Refraction





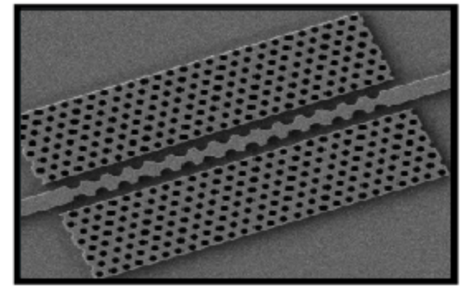


Probe phase and amplitude are measured by frequency-resolved optical gating (FROG)

- 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 Communications 11, 2180 (2020).

Relaxed Phase-Matching Requirements in ENZ Media



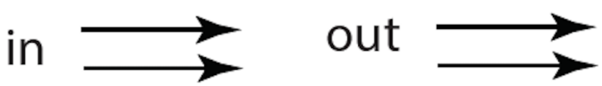
- We study four-wave mixing in a zero-index waveguide

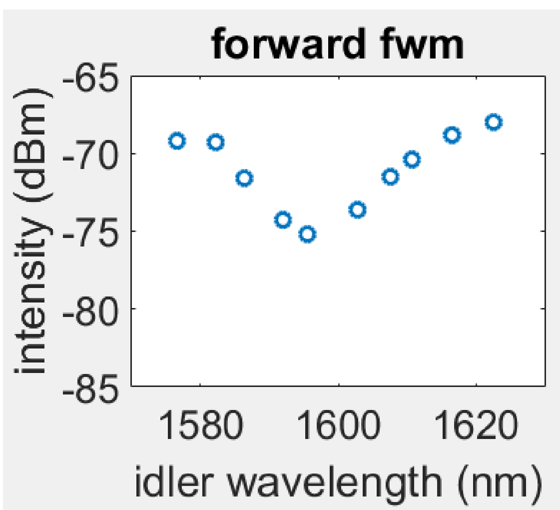
$$2\omega_p = \omega_s + \omega_i$$

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

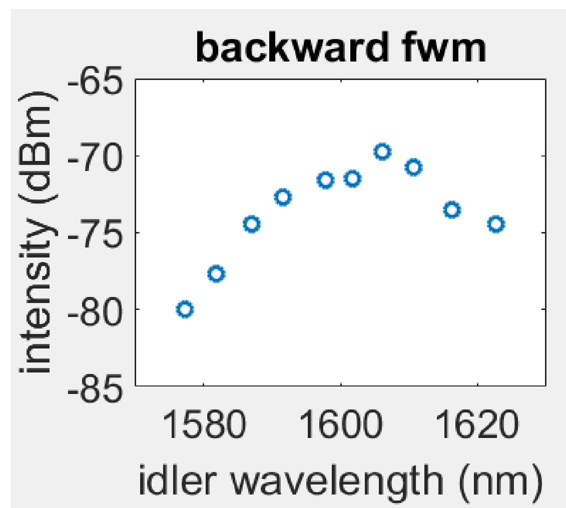
- Recall that we need $\Delta k = 0$, but when $n = 0$, $k = n\,\omega/c$ vanishes for each of the interacting waves and thus so does Δk.

"forward" process



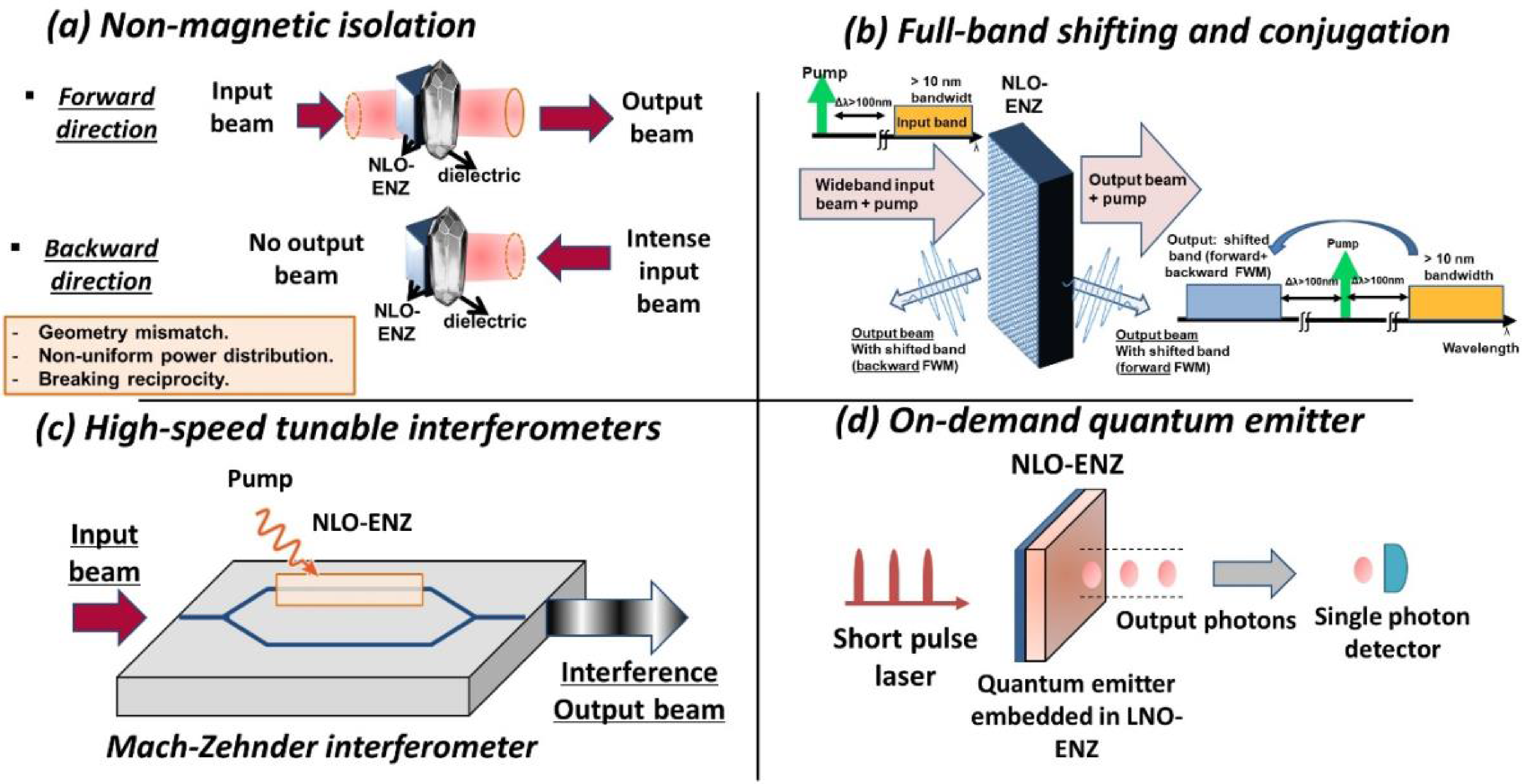


"backward" process



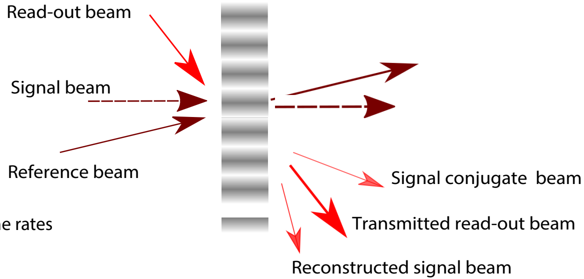
- Significance: Nonlinear optical processes that were previously believed to be too weak to be useful can be excited through use of ENZ materials.

Some Potential Applications of ENZ Behavior

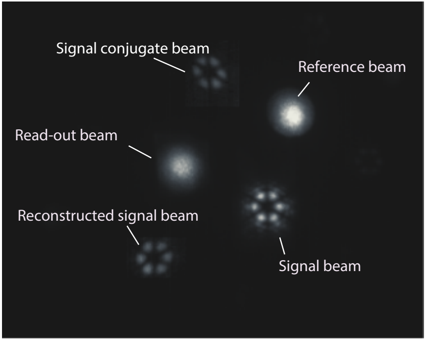


Real-Time Holography with THz Refresh Rates

- 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)

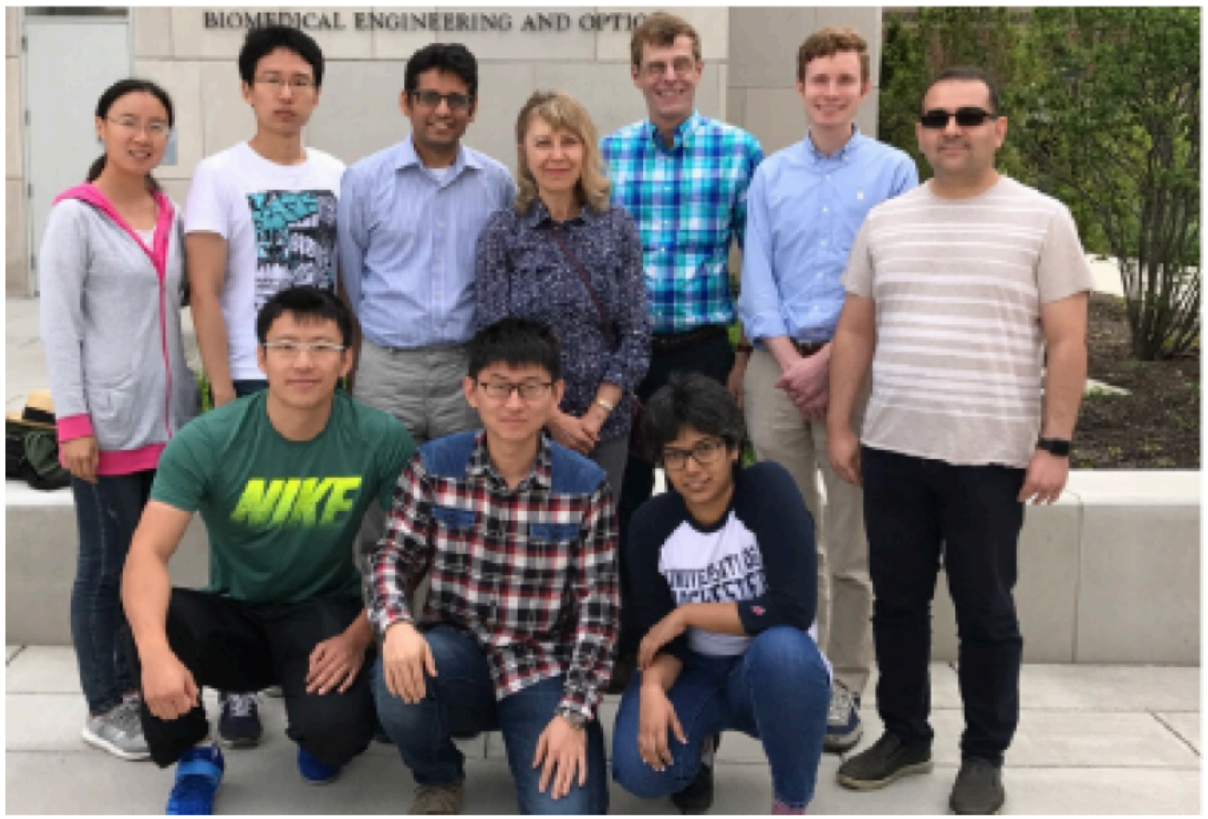


Alam, Fickler, Reshef, Giese, Upham, and Boyd

Special Thanks To My Students and Postdocs!

Ottawa Group

Rochester Group



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

Dependence of Second-Harmonic Generation on the Linear Dielectric Permittivity

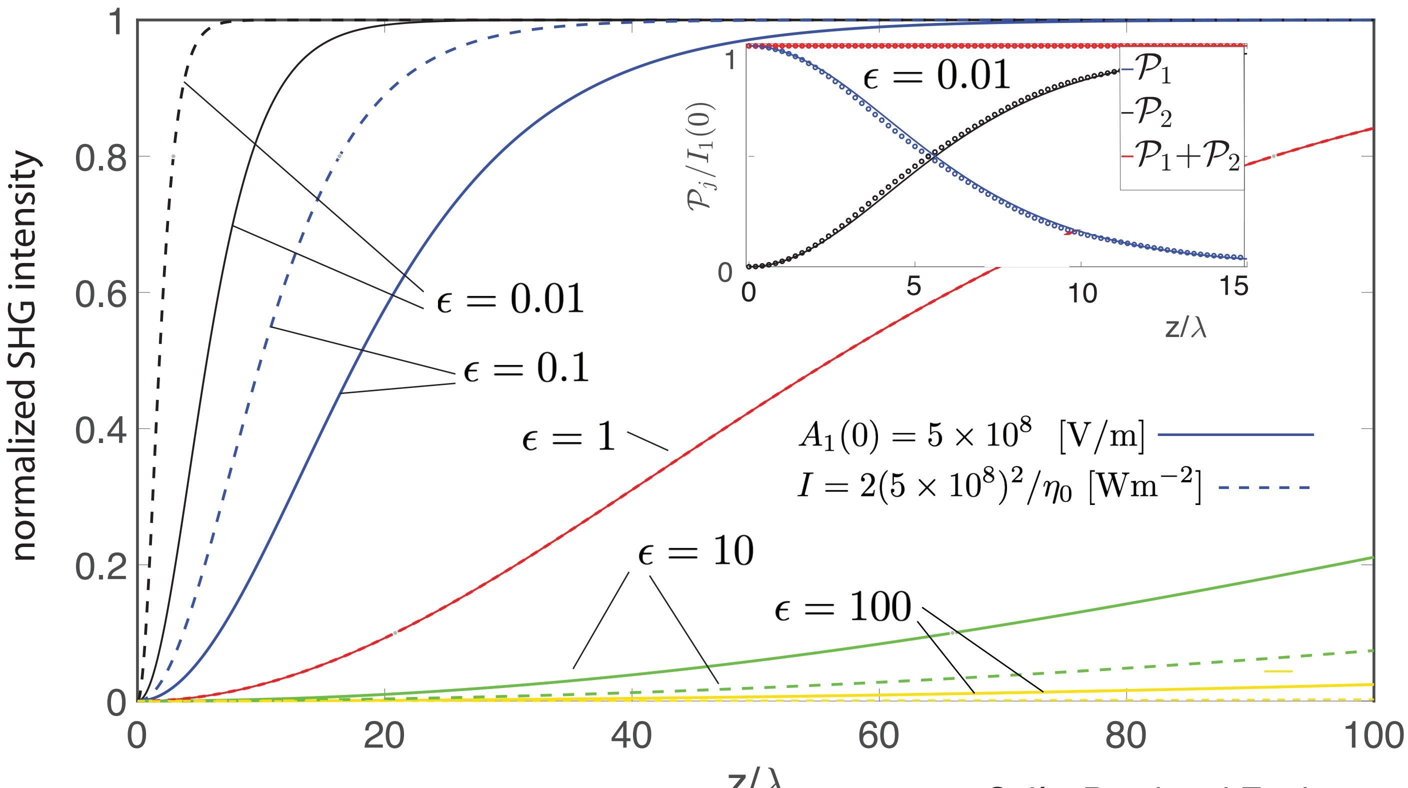
- We solve the standard equations for second-harmonic generation

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

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

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

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

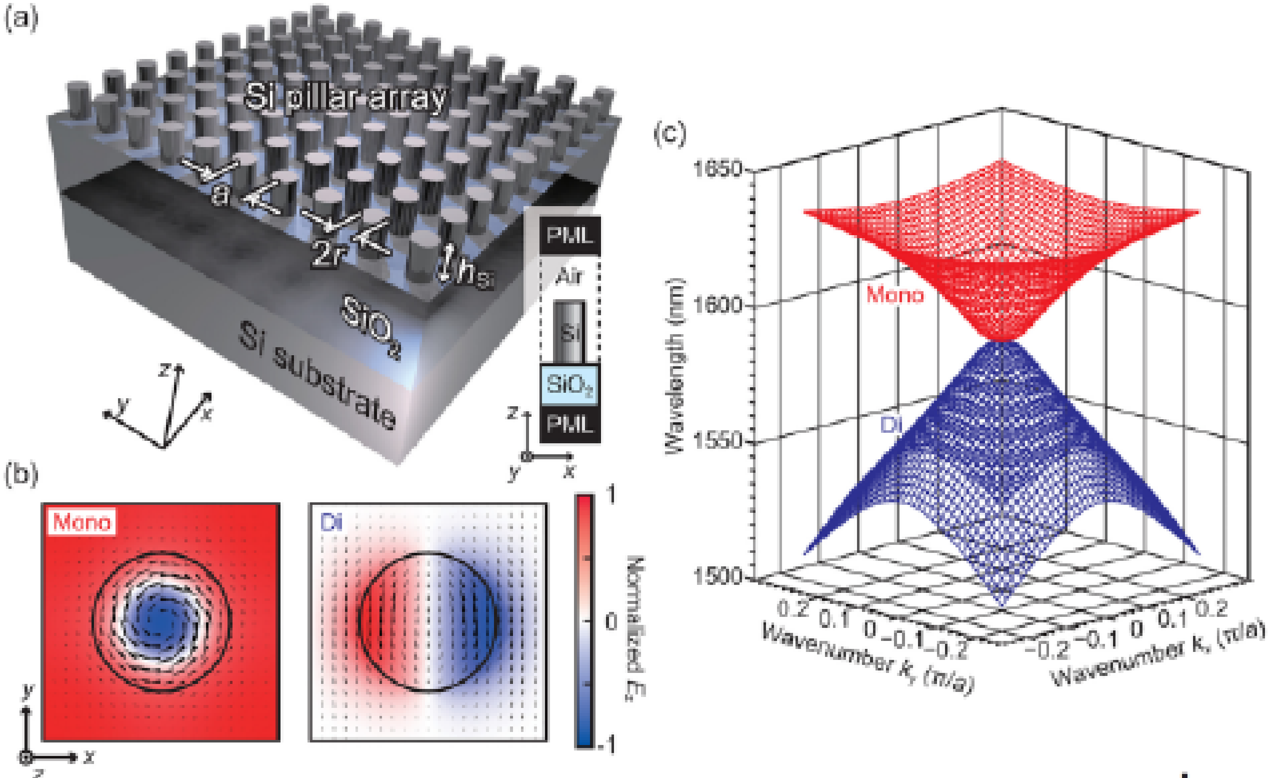


$\chi^{(2)}=5\times10^{-12}$ [m/V]

Solís, Boyd and Engheta, to be published

See also Solís, CLEO Poster JW2D.15

Dirac Cone Metamaterials



An EMNZ (epsilon and mu near zero) metamaterial

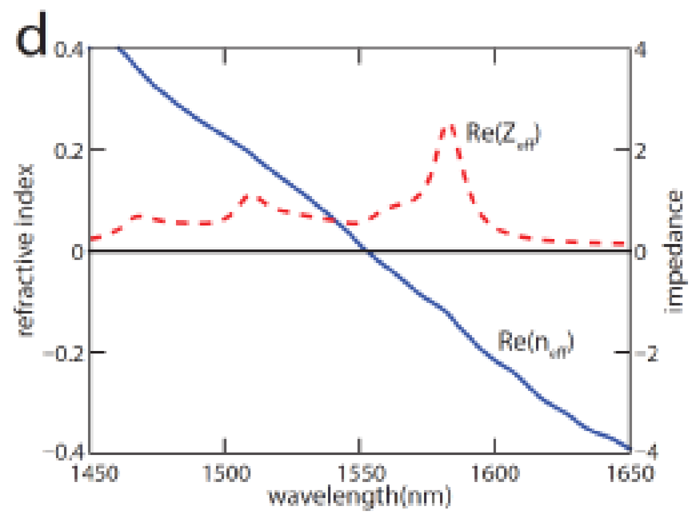
Opt Express 25, 8326 (2017)

It is also a ZIM
(zero index material)

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

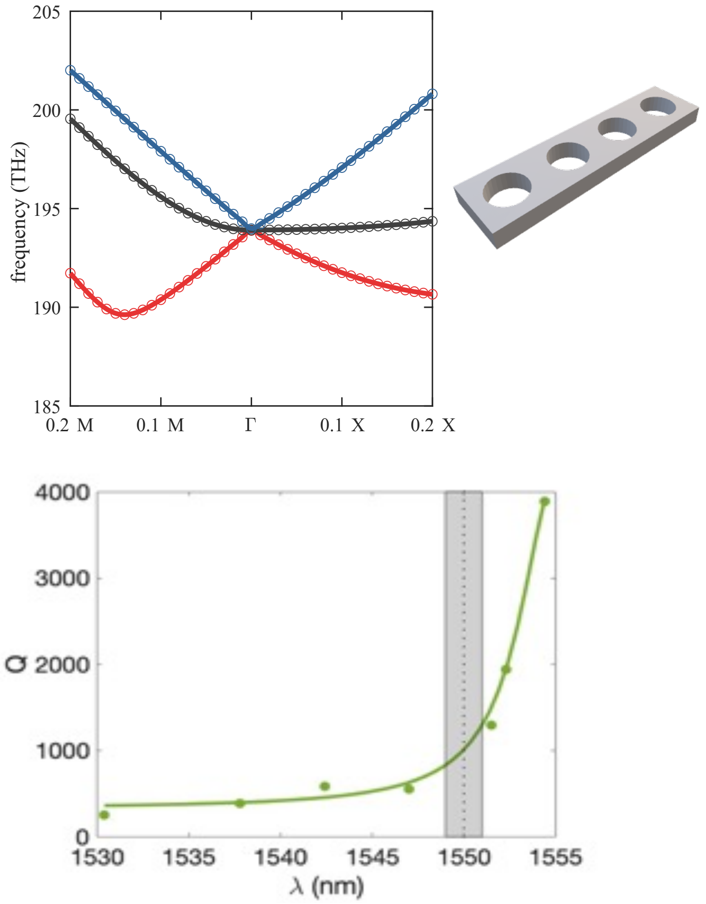
$$Z = \sqrt{\mu/\epsilon}$$

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



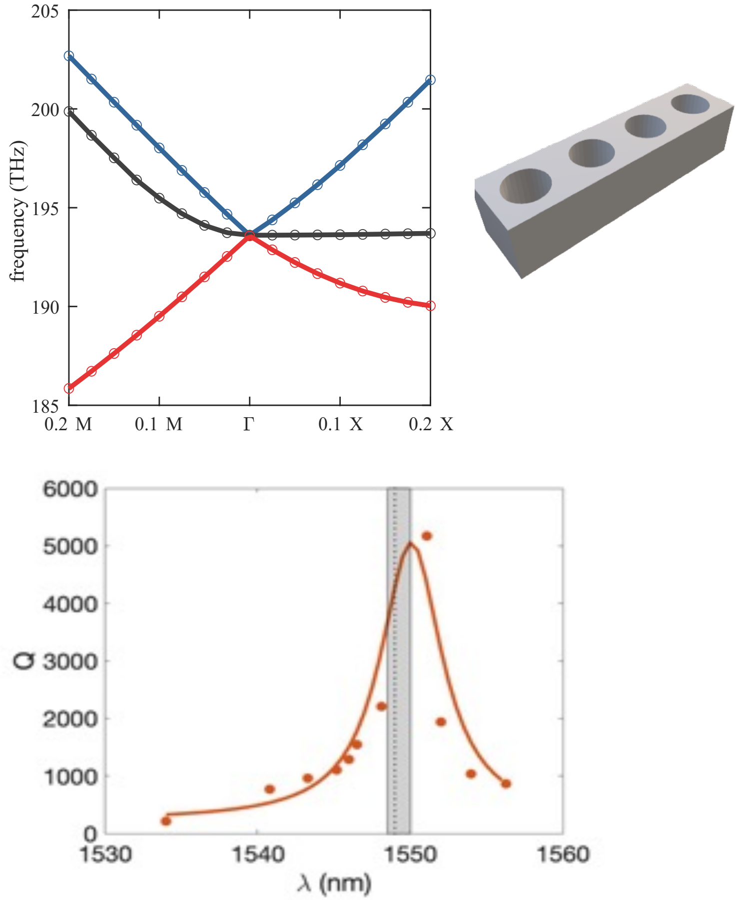
Low-Loss Zero-Index Metamaterials via a Bound State in the Continuum -- Mazur Group

TRADITIONAL ZIM DESIGN



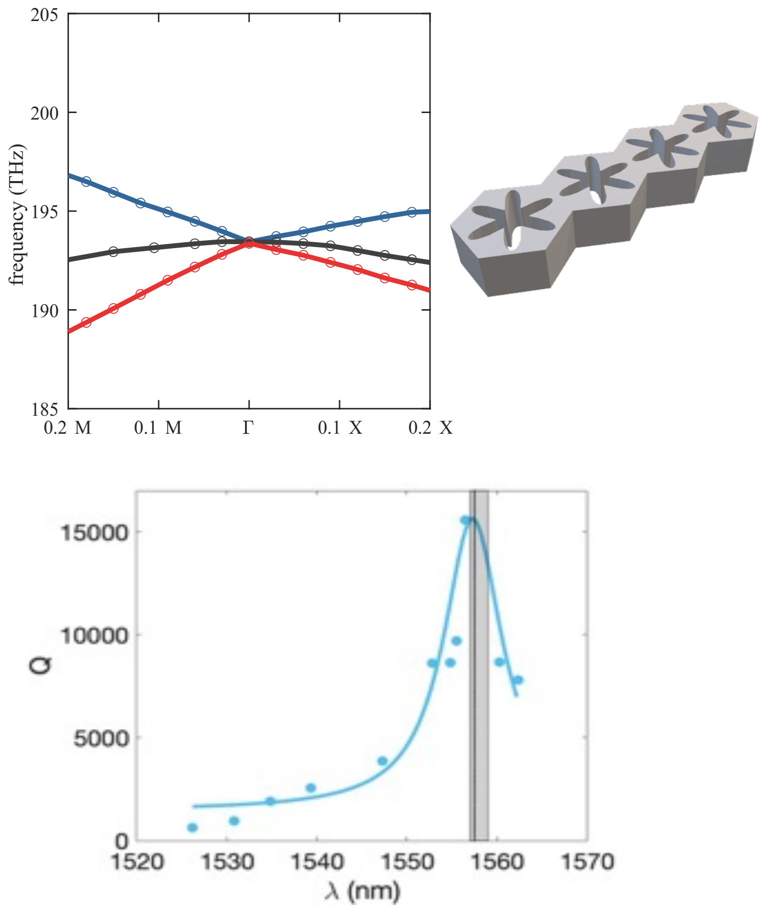
- ✓ ZERO REFRACTIVE INDEX
- ✕ LOW QUALITY FACTOR: ~1000
  - ✕ POOR MODE CONFINEMENT
  - ✕ LOW PROPAGATION LENGTH

FABRY-PÉROT ZIM



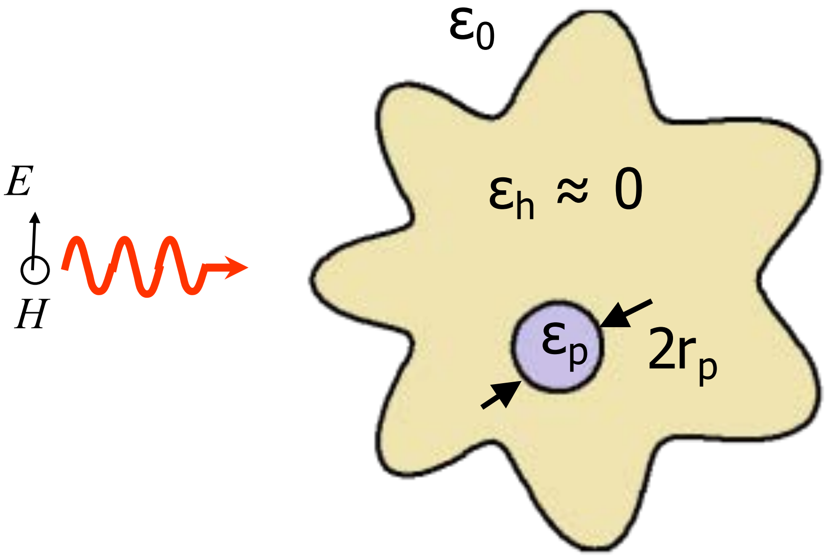
- ✓ ZERO REFRACTIVE INDEX
- ✓ QUALITY FACTOR: 5.1×10^3
- ✓ EASY TO FABRICATE

SYMMETRY-PROTECTED ZIM



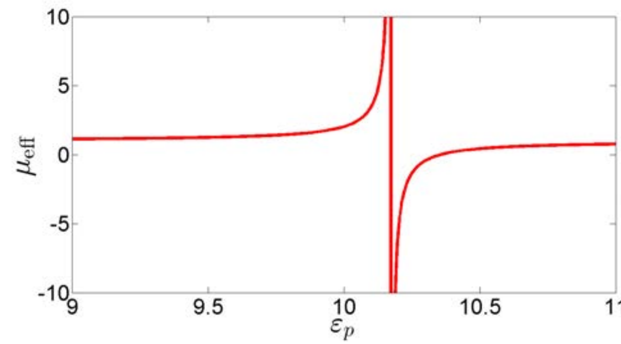
- ✓ ZERO REFRACTIVE INDEX
- ✓ QUALITY FACTOR: 1.6×10^4
- ✓ 10X PROPAGATION LENGTH

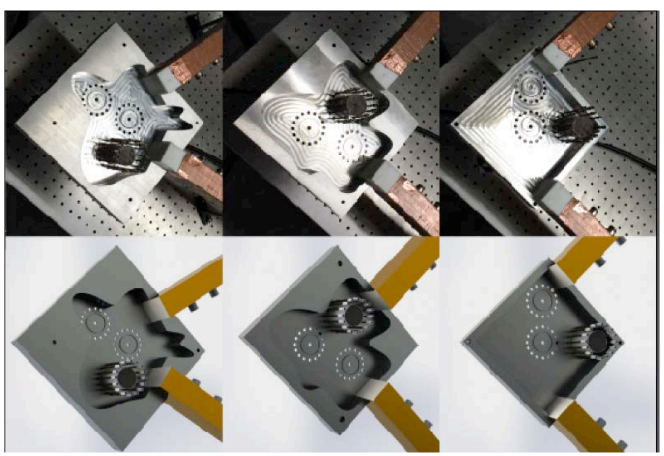
Photonic Doping of ENZ

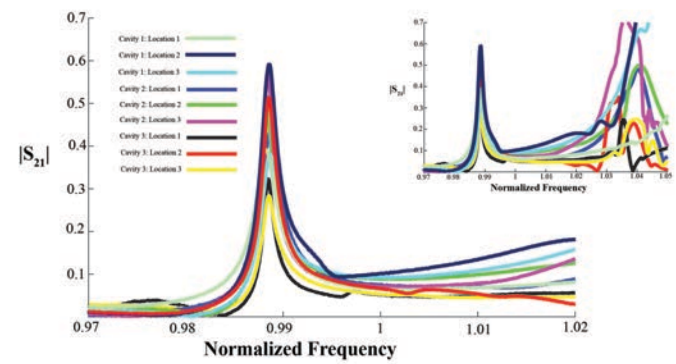


$$\varepsilon_{\text{eff}} \simeq 0$$

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







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