Physics and Applications of Epsilon-Near-Zero Materials

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

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Canada Excellence Research
Chair (CERC) in Nonlinear Quantum Optics

Research interest:
Nonlinear optics, quantum optics,
integrated photonics, meta-materials, etc.

Physics and Applications of Epsilon-Near-Zero Materials

- Physics of ENZ Materials
- Huge NLO Response of ENZ Materials and Metastructures
- Non-perturbative Nature of the NLO Response
- Some Applications of ENZ Materials

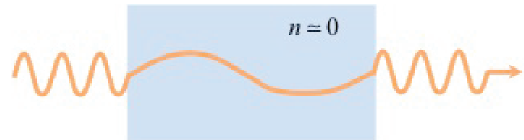
Physics of Epsilon-Near-Zero (ENZ) Materials

- 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)

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



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).

- 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 (for $\mu = 1$)

Stimulated emission rate = B times EM field energy density

$B = B_{\mathrm{vac}} \; / \; n^2$

Optical gain is very large!

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

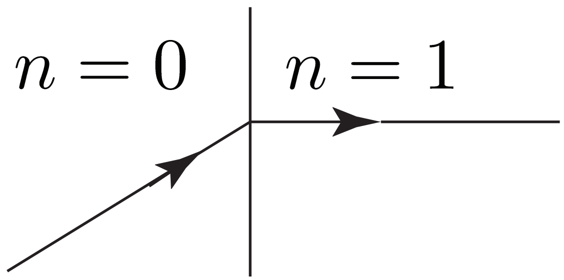
Terminology: ZIM= zero-index material; ENZ=epsilon near zero; EMNZ= epsilon and mu near zero

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

- Snell's law leads to intriguing predictions

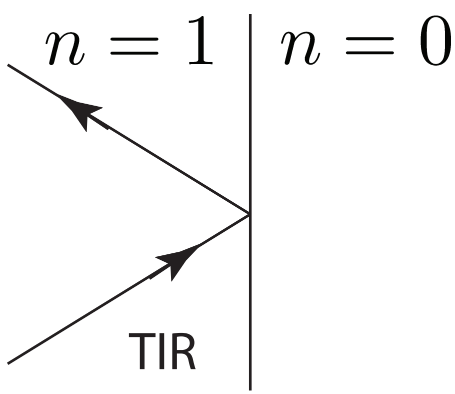
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

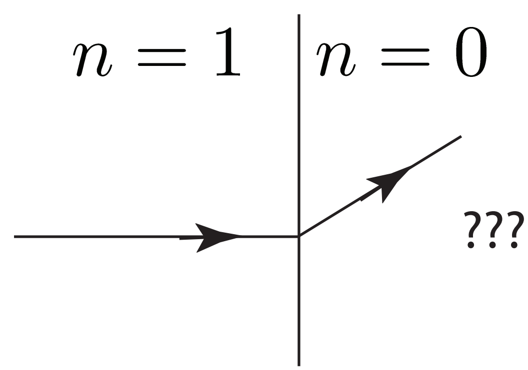
- Light always leaves perpendicular to surface of ENZ material!



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!

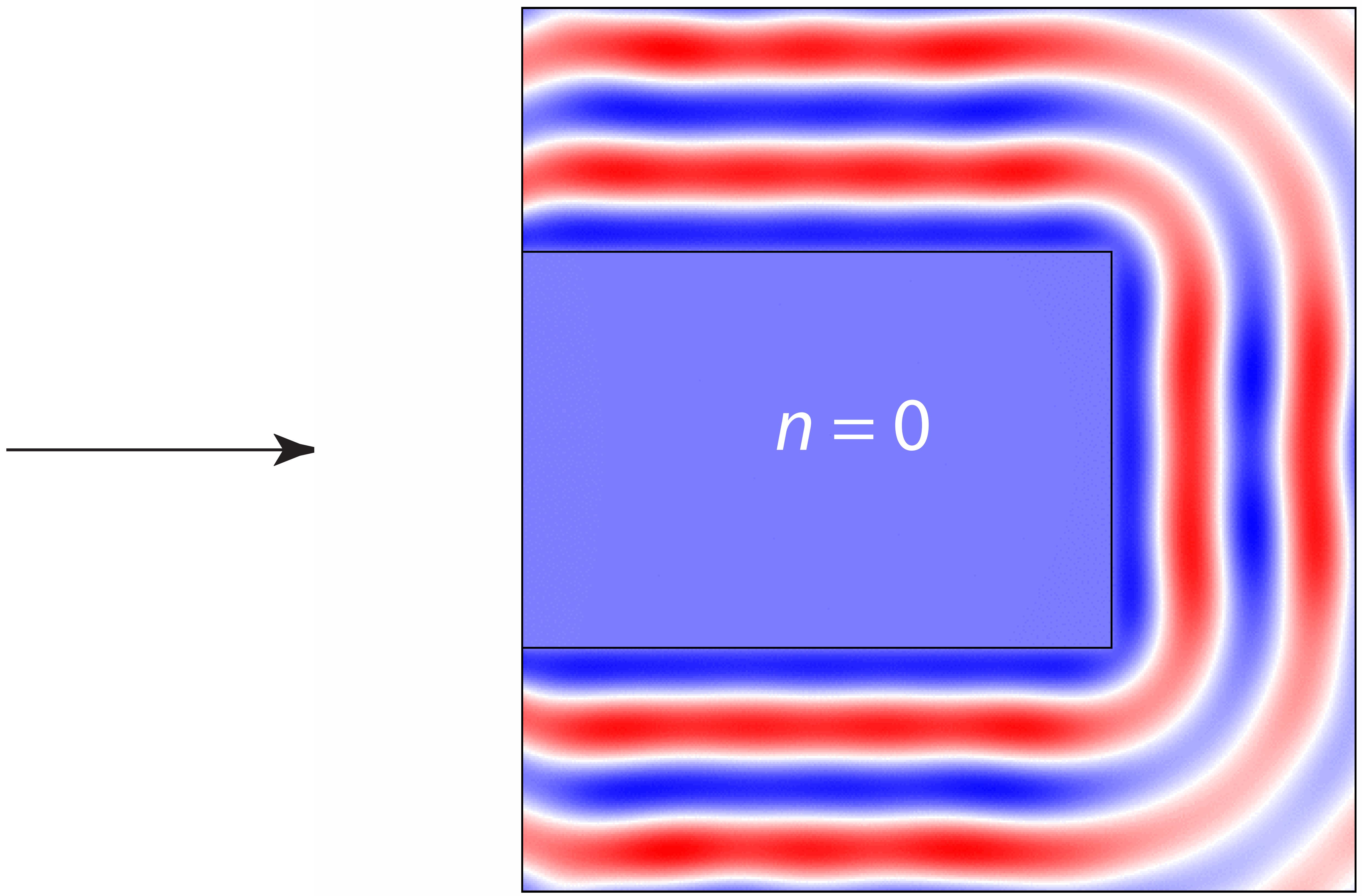




Y. Li, et al., Nat. Photonics 9, 738, 2015.

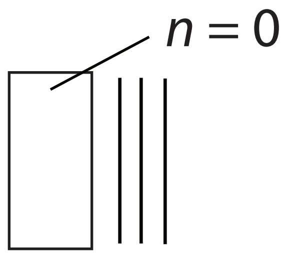
Maxwell Equations Prediction

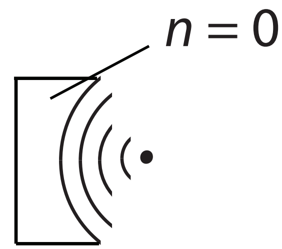
- light enters slab at normal incidence



Some Consequences of ENZ Behaviour - 1

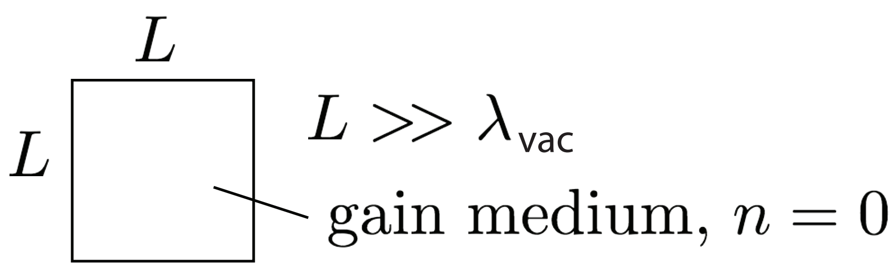
- Funny lenses





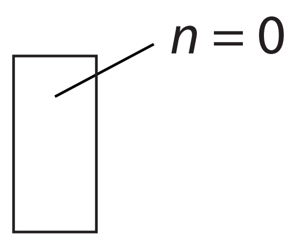
A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

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



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

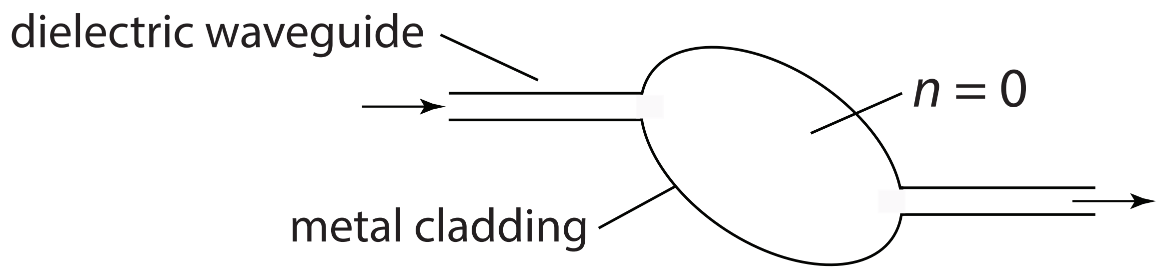
- No Fabry-Perot interference



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

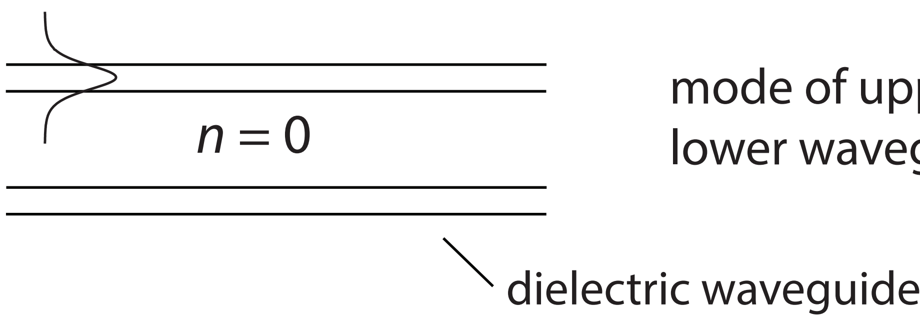
Some Consequences of ENZ Behavior - 2

- 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

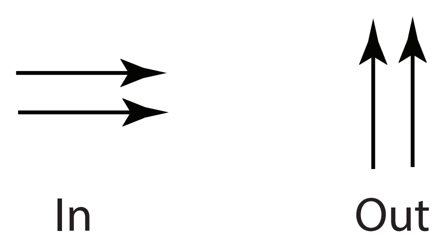


mode of upper waveguide extends to lower waveguide for any separation

- 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 Implications of Near-Zero Index

- Wavelength is stretched ($\lambda = \lambda_0 / n$).

- Phase velocity becomes very large ($v = c / n$).
  Frequency is unchanged (for static fields)

- The field becomes static in space, yet remains dynamic in time.
  Space and time decouples.

- k-vector is undefined (has zero length) in the medium.

- Light exits a zero-index medium with k-vector perpendicular to the interface.

- Phase matching conditions of nonlinear optics is relaxed.

- Propagation nearly always nonparaxial ($\lambda >> $ beam diameter).

- E field is enhanced due to the boundary condition.

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 $n_0 = 0$?

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 ε 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

Epsilon-Near-Zero Materials for Nonlinear Optics

- We need materials with a much larger NLO response
- We recently reported a material (indium tin oxide, ITO) with an n_2 value 100 time larger than those previously reported.
- This material utilizes the strong enhancement of the NLO response that occurs in the epsilon-near zero (ENZ) spectral region.

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).

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

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

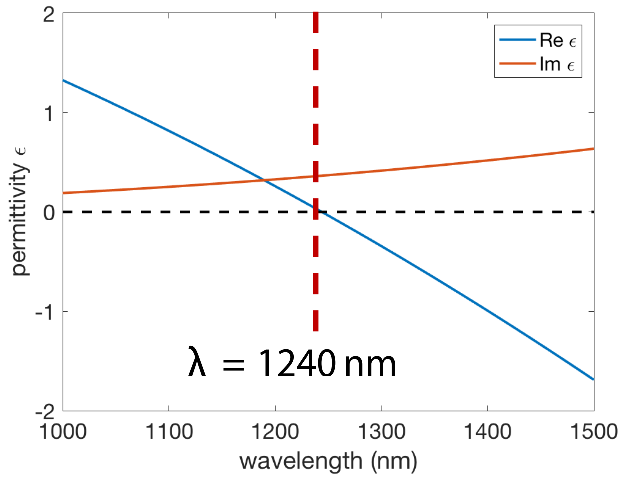
P is the induced dipole moment per unit volume and E is the field amplitude.

Also, the refractive index changes according to

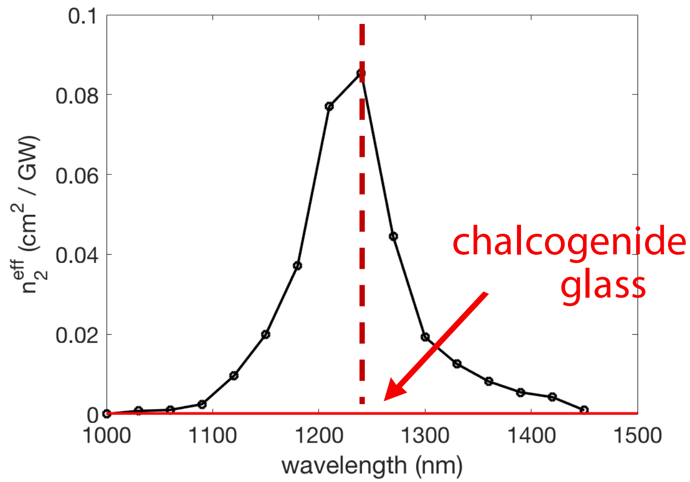
$$n = n_0 + n_2 I + n_4 I^2 + \ldots$$

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

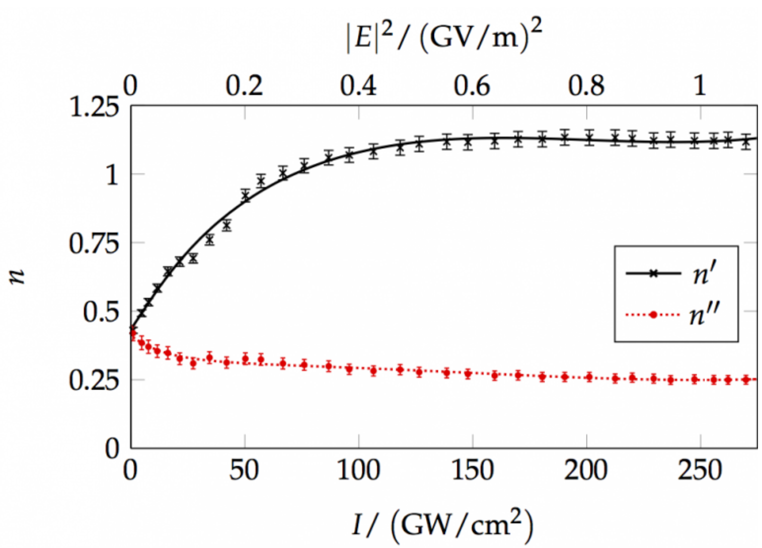
- ellipsometry



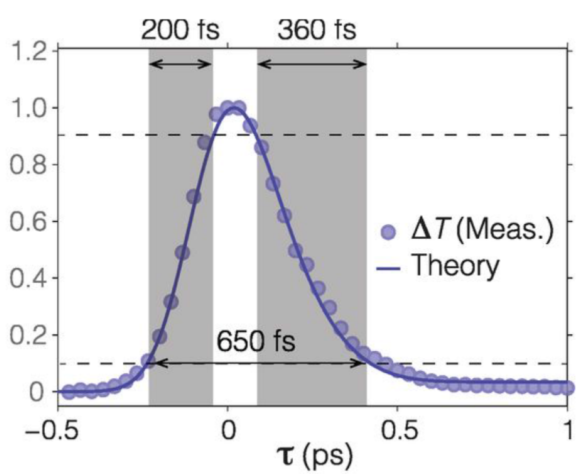
- n_2 is 3.4 x10^5 times larger than that of silica glass



- overall change in refractive index of 0.8



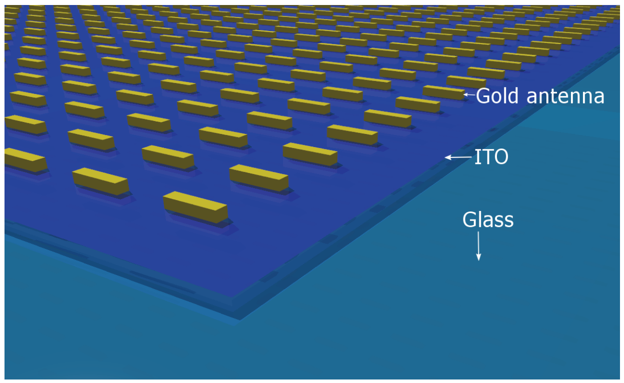
- sub picosecond reponse time

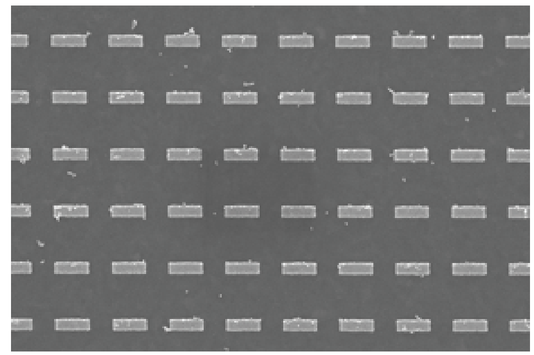


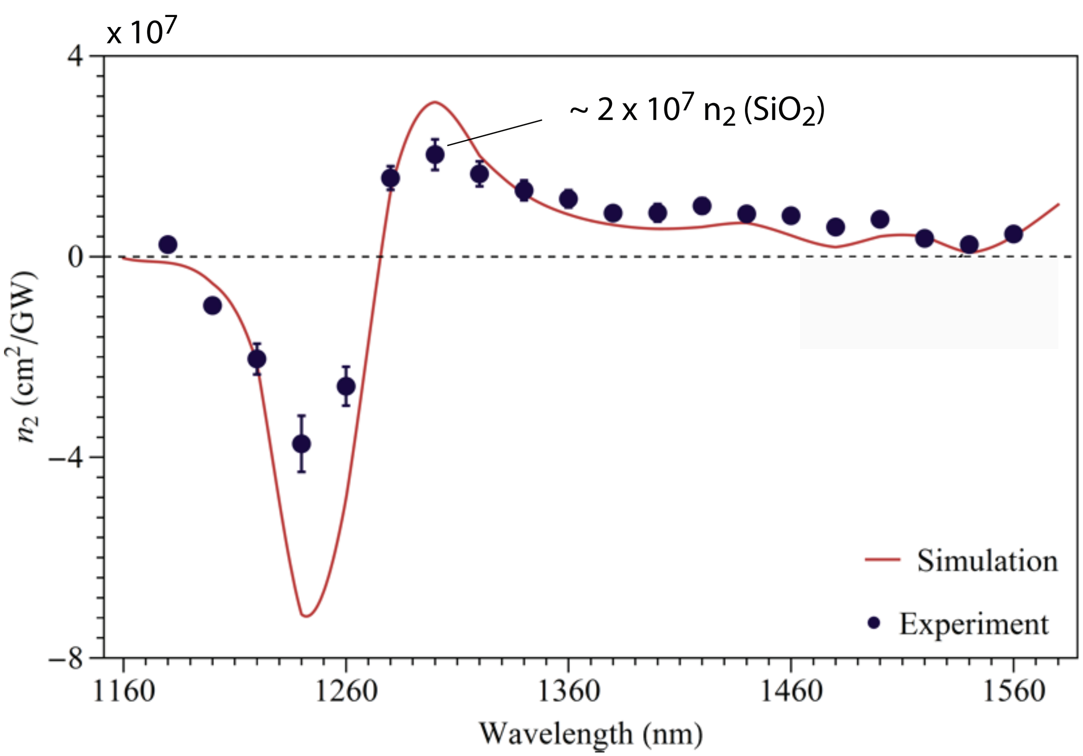
M. Z. Alam et al., Science 352, 795-797 (2016)

ENZ Metasurface: Gold Nanoantennas on ITO

- 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







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

Giant Nonlinear Response of ENZ Metastructures: Our Team

Robert Boyd

- Professor of Optics and Physics at UR; Canada Excellence Research Chair in U Ottawa.
- Research interests include nonlinear optics, light-matter interaction, fabrication of nanophotonic devices and metastructures.
- Awards include the Townes Award of OSA and the Schawlow Prize of APS; Honorary Doctorate from the University of Glasgow.

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

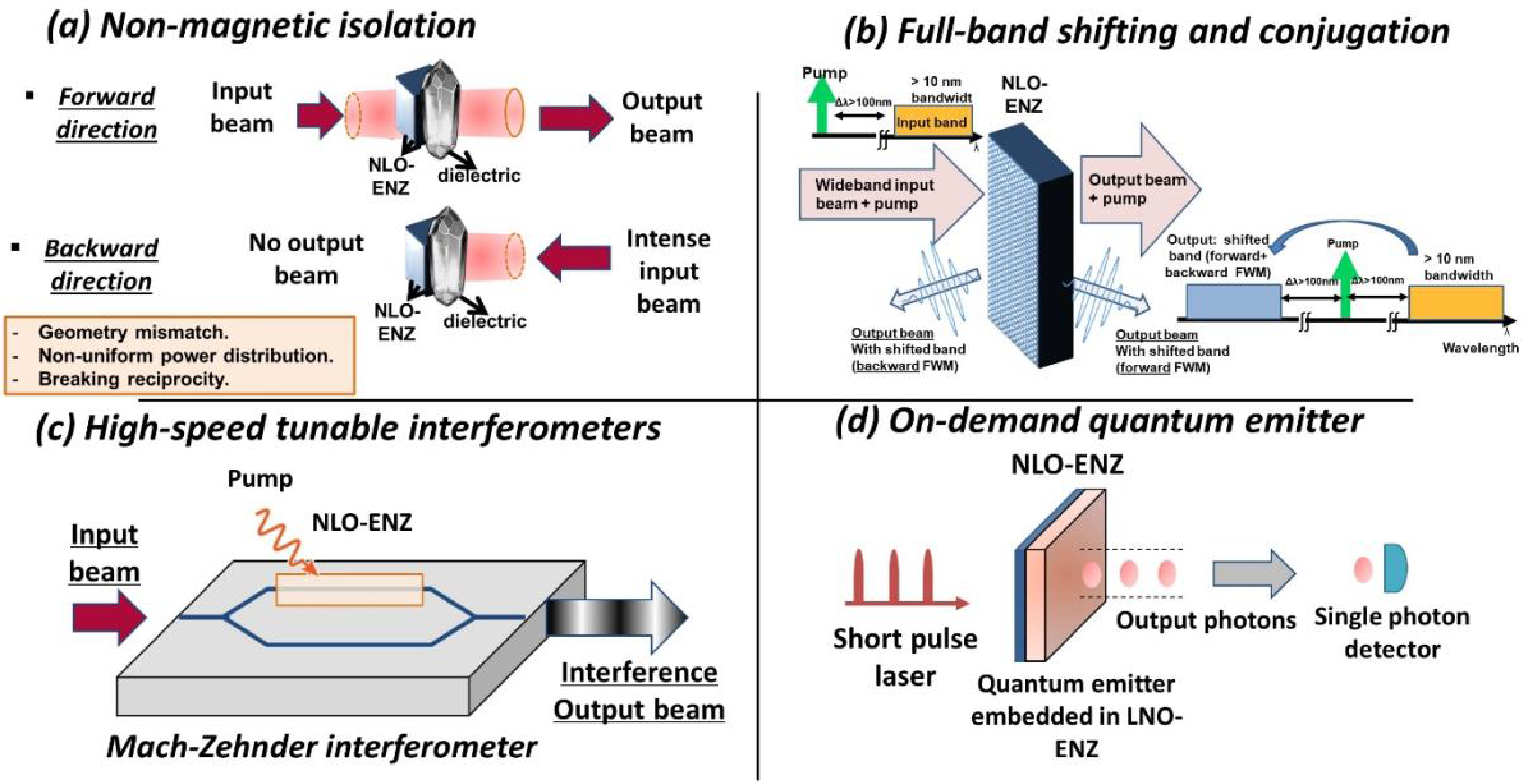
Eric Mazur

- 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

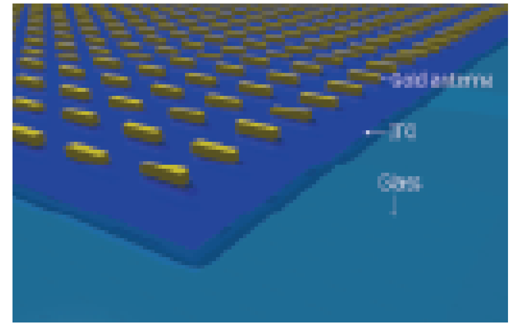
- Steven & Kathryn Sample Chair in Engineering at the University of Southern California.
- Ph.D. Columbia University
- Honors include Member of US National Academy of Engineering; Int'l Fellow of UK Royal Academy of Engineering; President of OSA and of IEEE Photonics Society.
- Activities include using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

Some Potential Applications of ENZ Behavior

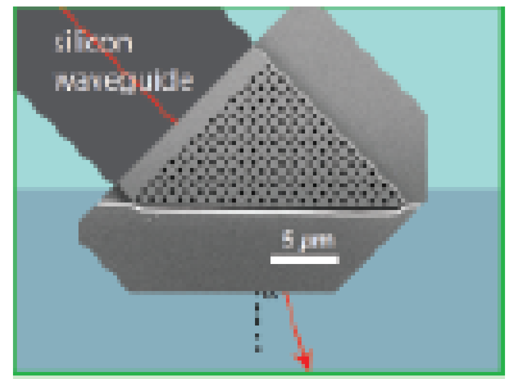


Three Material Platforms Under Investigation

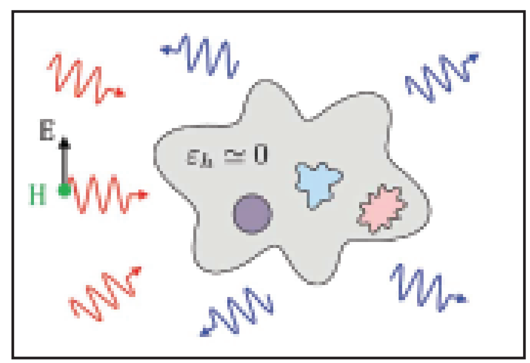
- Nanoantennas coupled to ENZ substrate
  (out of plane; free-space coupling)
  (Rochester)



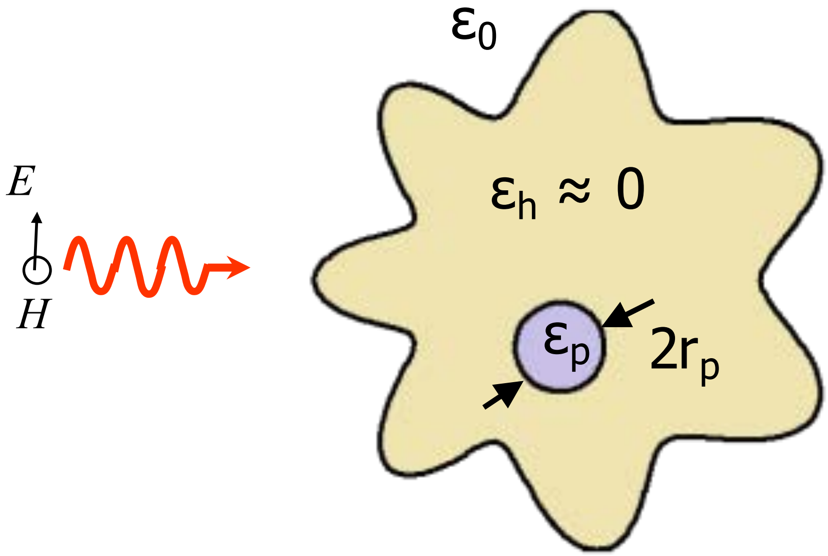
- Dirac cone metamaterials
  (in plane; compatible with integrated optics)
  (Harvard)



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

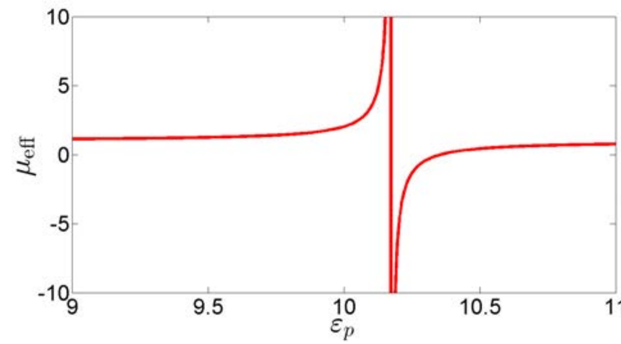


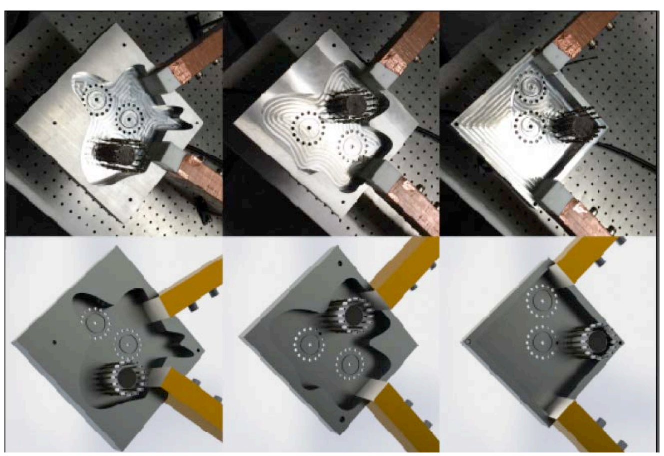
Photonic Doping of ENZ

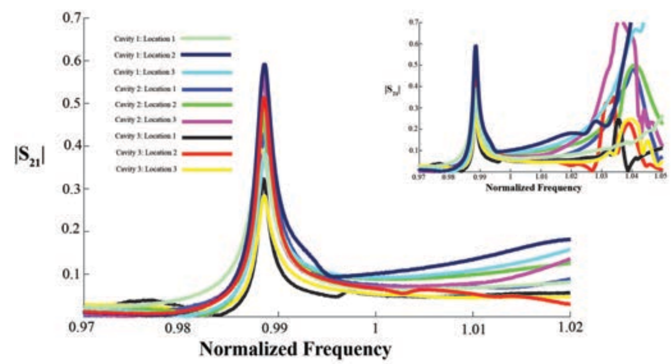


$$\varepsilon_{\text{eff}} \simeq 0$$

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

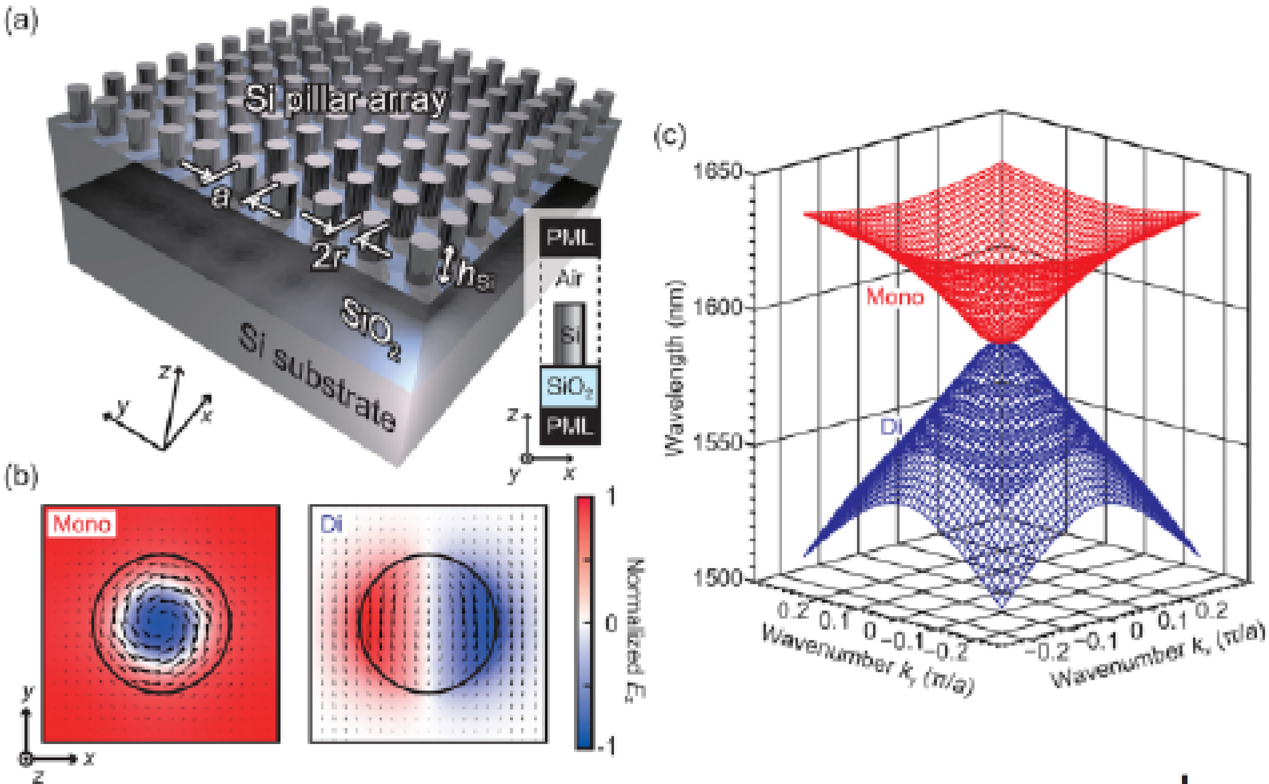






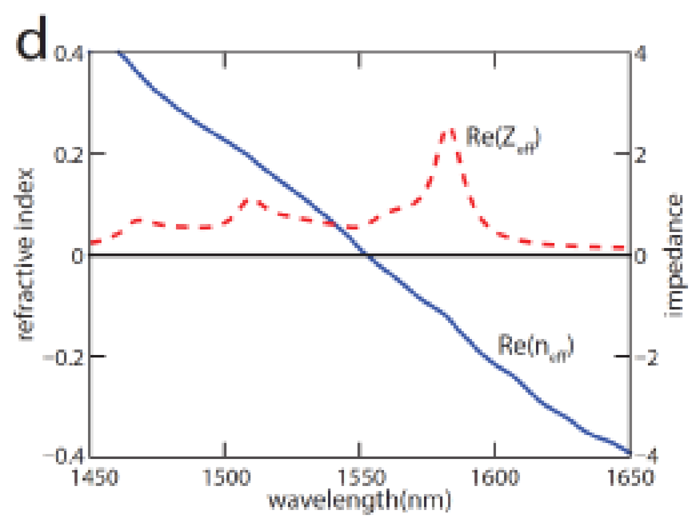
I. Liberal, A. Mahmoud, Y. Li, B. Edwards and N. Engheta, Science, 355, March 10, 2017

Dirac Cone Metamaterials



An EMNZ (epsilon and mu near zero) metamaterial

Opt Express 25, 8326 (2017)



It is also a ZIM
(zero index material)

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

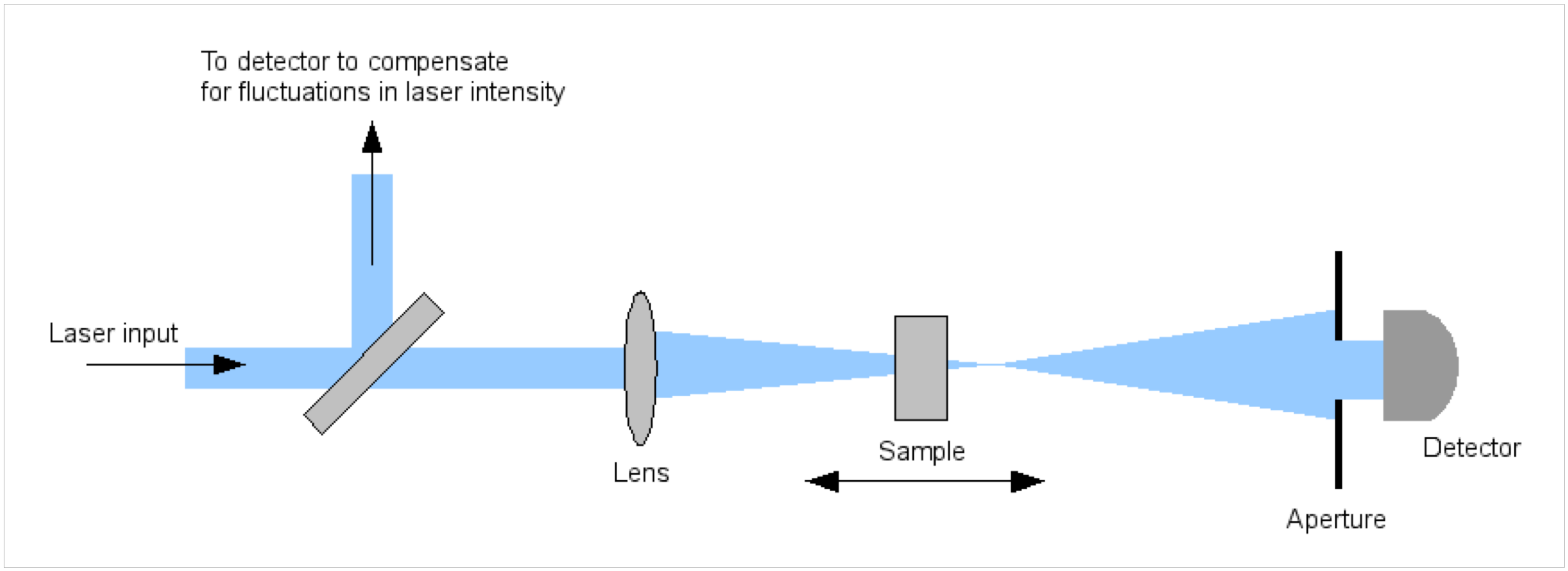
$$Z = \sqrt{\mu/\epsilon}$$

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

Nonlinearity of Dirac-cone zero-index metamaterials

We believe that the nonlinearity of these metamaterials will enhanced just as is the case for ITO and similar ENZ materials

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

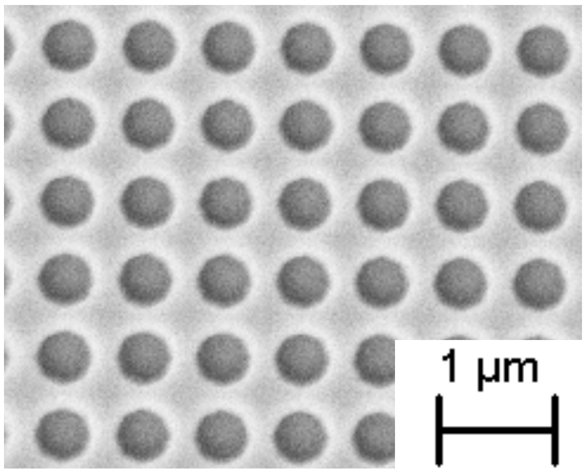


Z-scan measurement setup
(from Wikipedia)

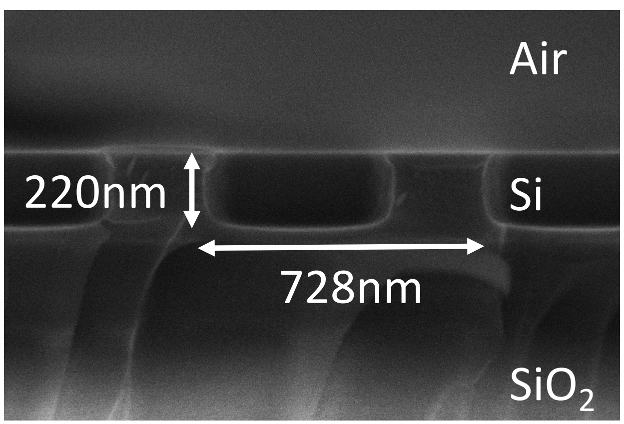
This nonlinear characterization will be done by Rochester team. First, we need to determine the zero-index wavelength for each sample.

Determining the zero-index wavelength

Zero-index Dirac cone metamaterial

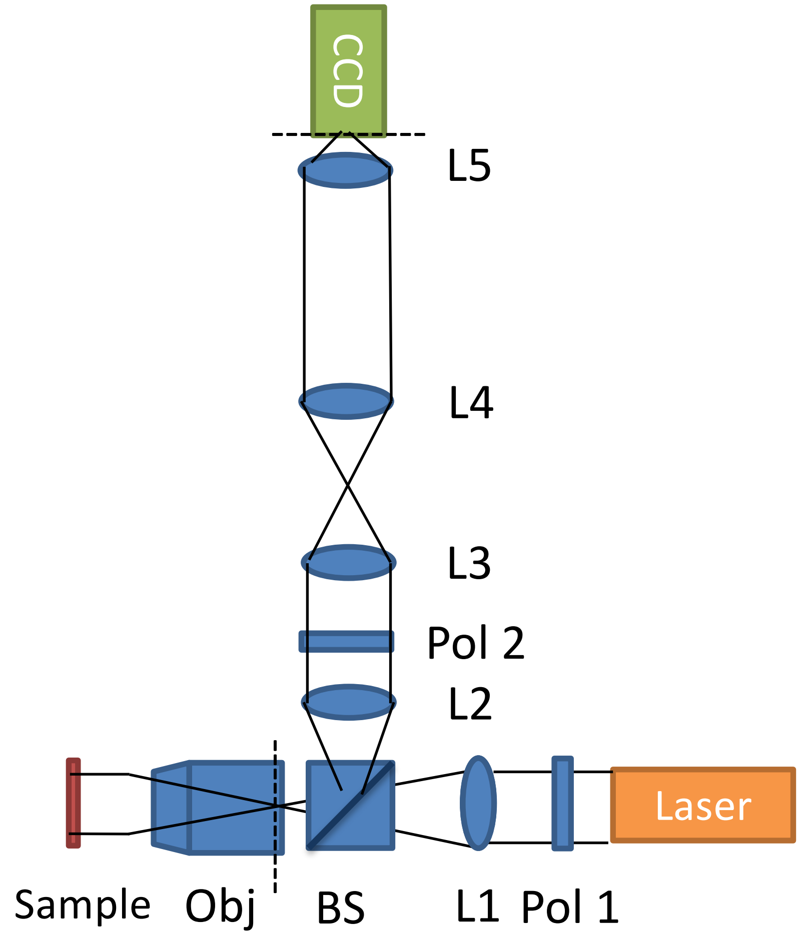


(top view)



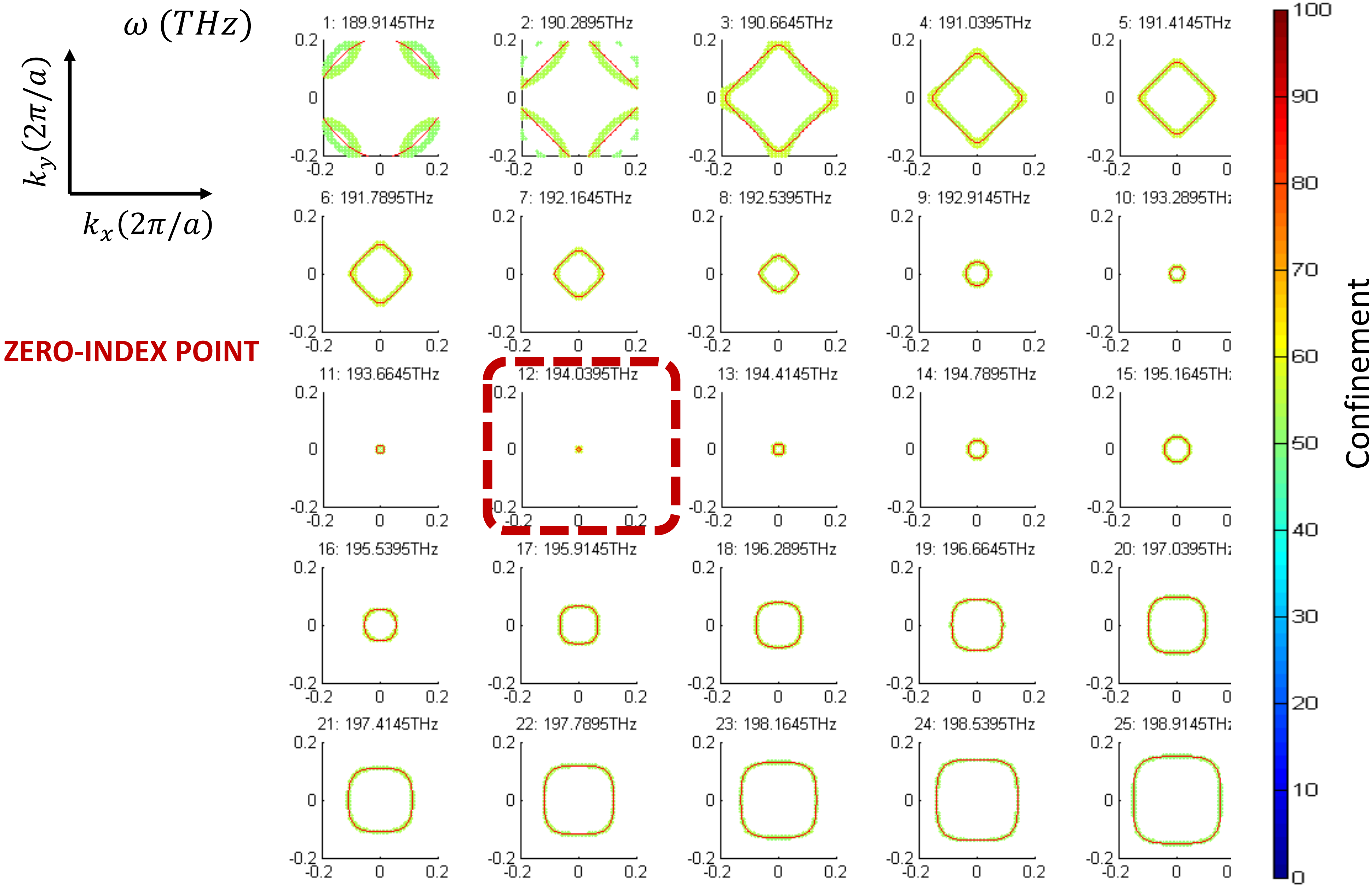
(cross-sectional view)

Isofrequency contour measurement setup



Regan, E. C. *et al.* Direct Imaging of Iso-frequency Contours in Photonic Crystal Slabs. *Sci. Adv.* **1,** 1–2 (2016)

Isofrequency Contour Simulation

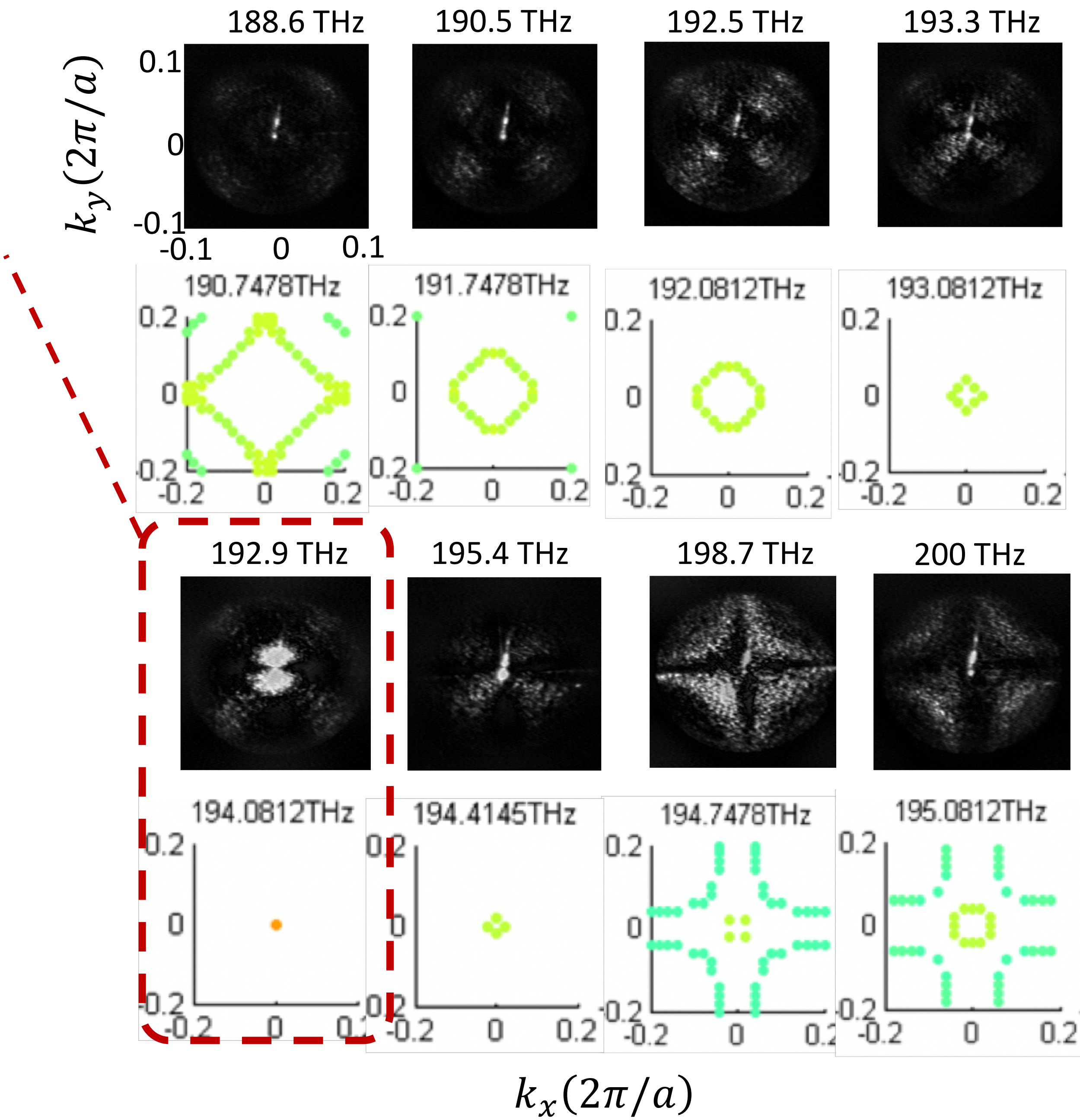


Measurement and Simulation

Zero-index point measured at 192.9 THz (experiment) and 194.04 THz (numerical)

Experimental results are shifted relative to simulations due to fabrication errors.

High intensity at zero-index point correlates with a large quality factor of the resonance.



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

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

where

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

and

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

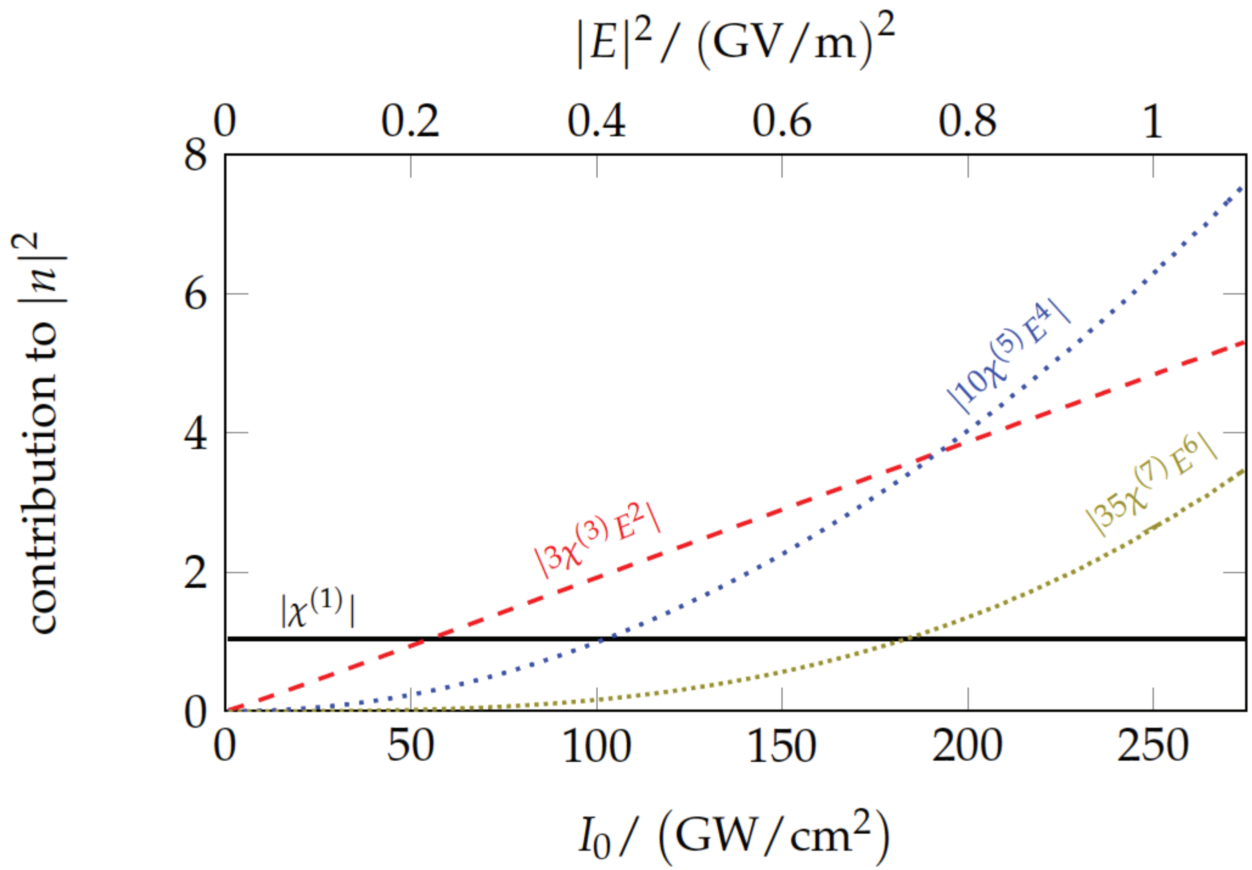
4. More generally, the intensity dependent refractive index can be described by

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

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

Nonperturbative Behavior of the NLO Response - 2

- Example of nonperturbative behavior (data from ITO Science paper)



- Summary
  - Dependence of refractive index *n* on intensity *I* is nonperturbative
  - But the dependence of P on E can be described by the usual power-series expansion

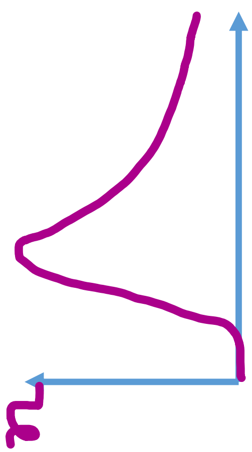
$$P = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \ldots$$

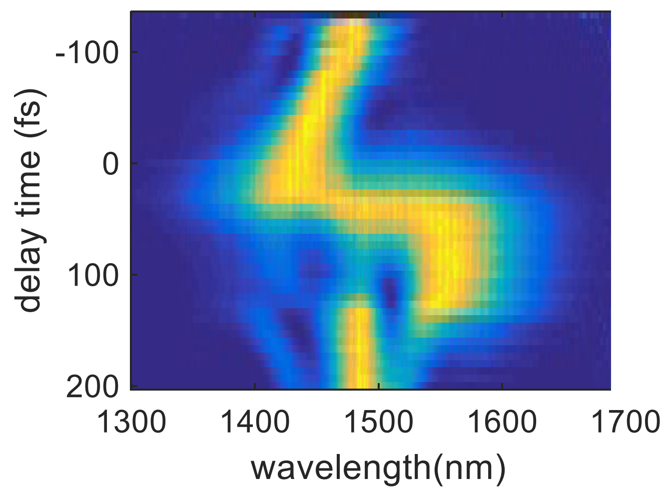
Adiabatic wavelength conversion

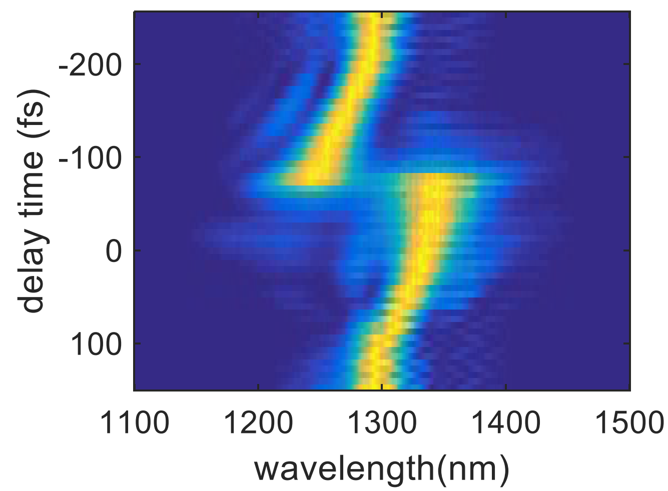
- A time-varying refractive index changes the wavelength of light passing through the medium. (See works by Lipson, Fan, Notomi, and Engheta.)

$$\Delta\omega = \mp \frac{\Delta n \omega_1}{\Delta n + n_1}$$

- We find that the rising edge of ITO's response redshifts the probe, whereas the falling edge leads to a blueshift.







(two different central wavelengths)

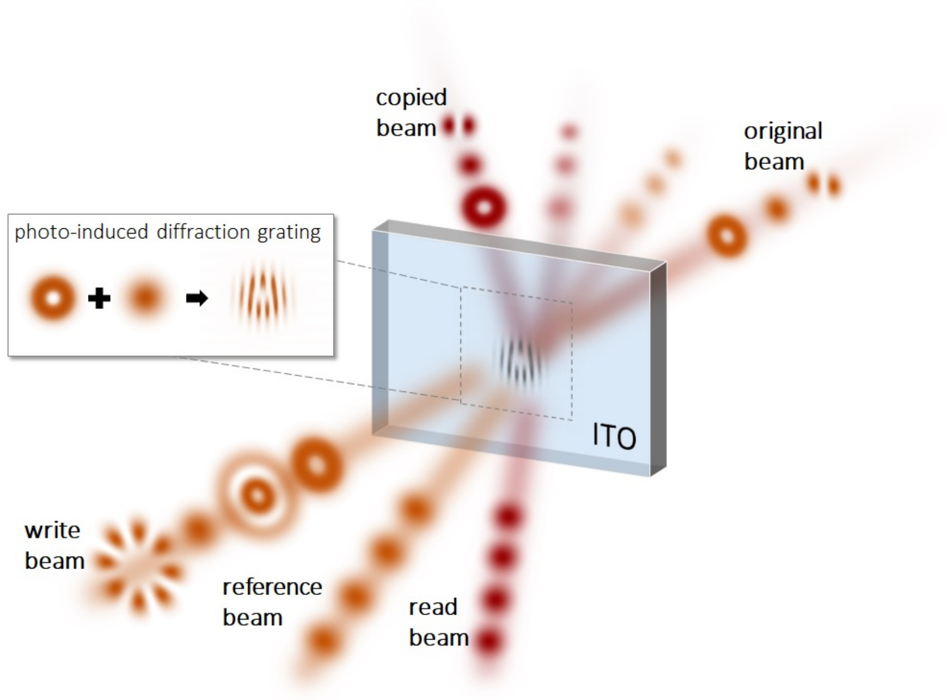
- The observed effect is 100 times larger with almost 100 times smaller propagation distance than previous reports of AWC.

- Application: wavelength-division multiplexing for telecom

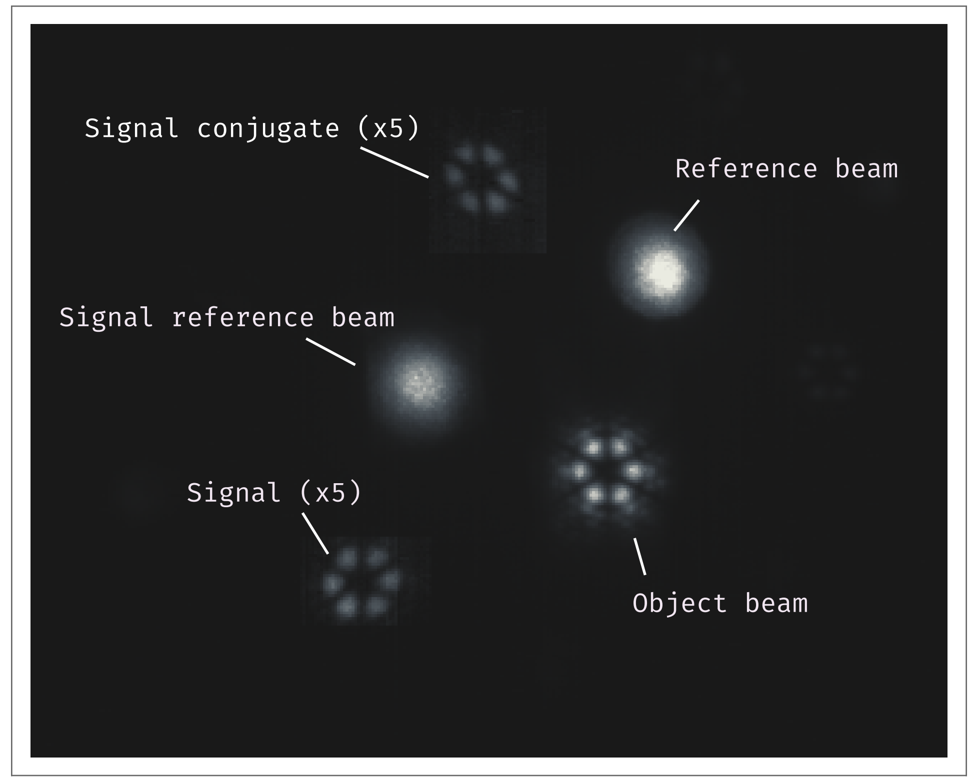
Ultrafast Holography and Beam Copying

- Real-time holography with sub-picosecond response time

- Schematic of beam-copying procedure

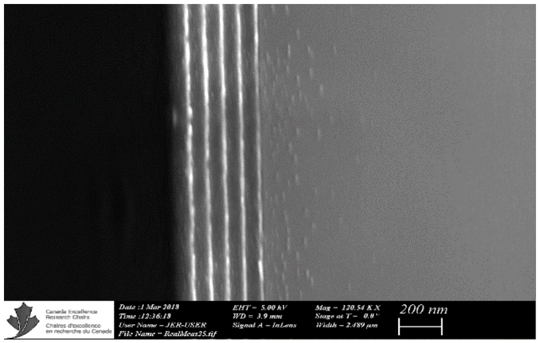


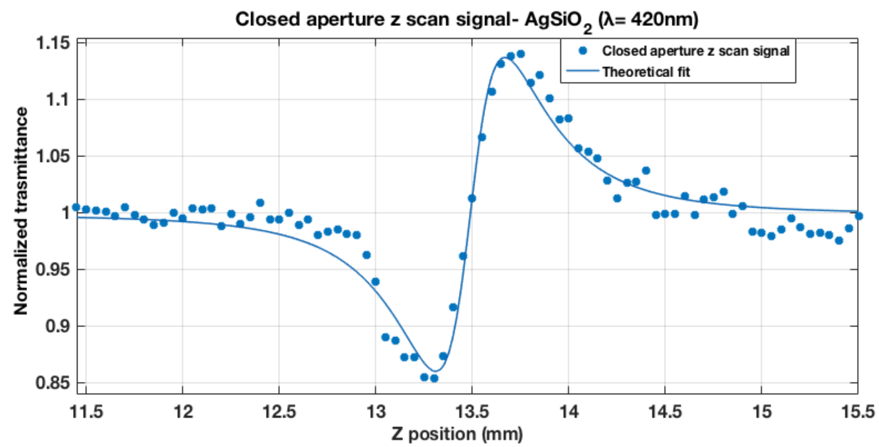
- Laboratory results



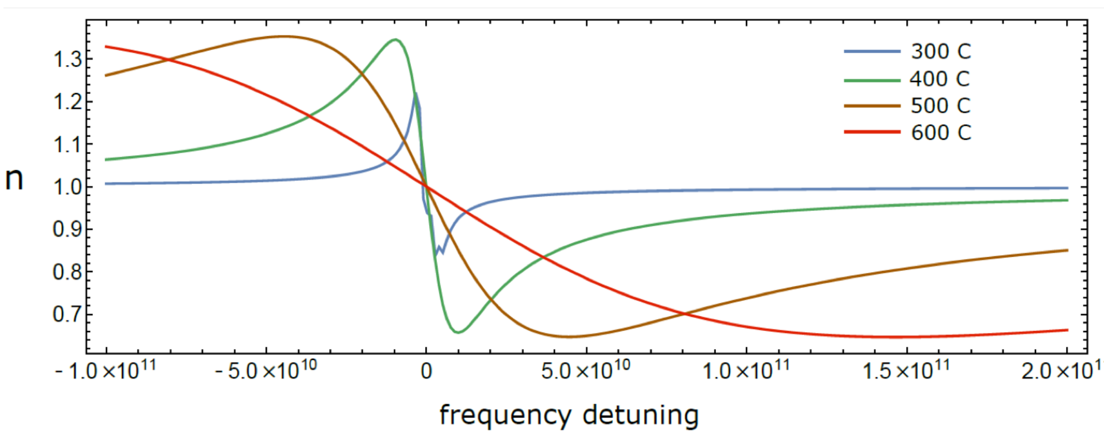
Z-Scan Measurements of ENZ Materials

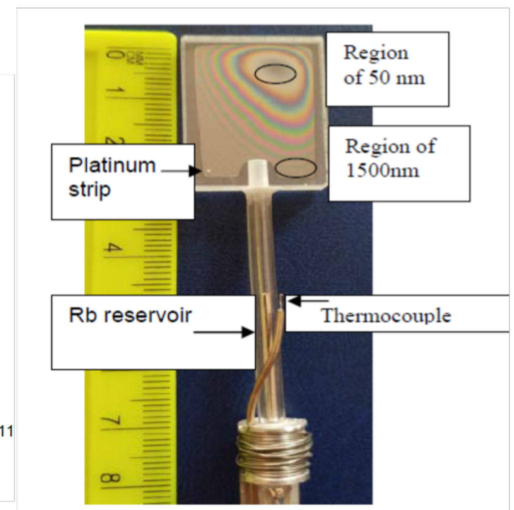
- Ag-SiO_2 multilayer stack





- Rb vapor nanocell

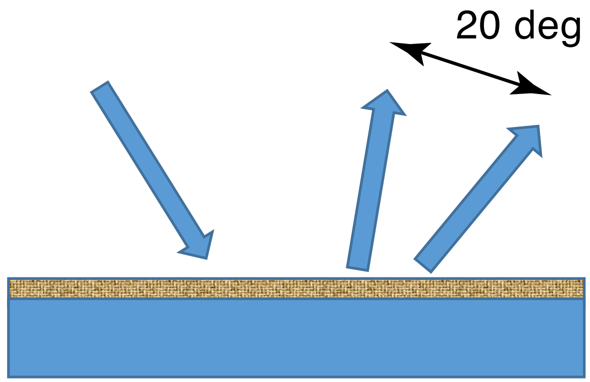




- ITO with 30 ps pulses
  - How does n_2 scale with pulse duration?
  - Modify Z-scan theory to include nonparaxiality and the large nonlinear Fresnel reflection

All-Optical, Nanoscale, Sub-Picosecond Beam Steering

- **Concept**



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

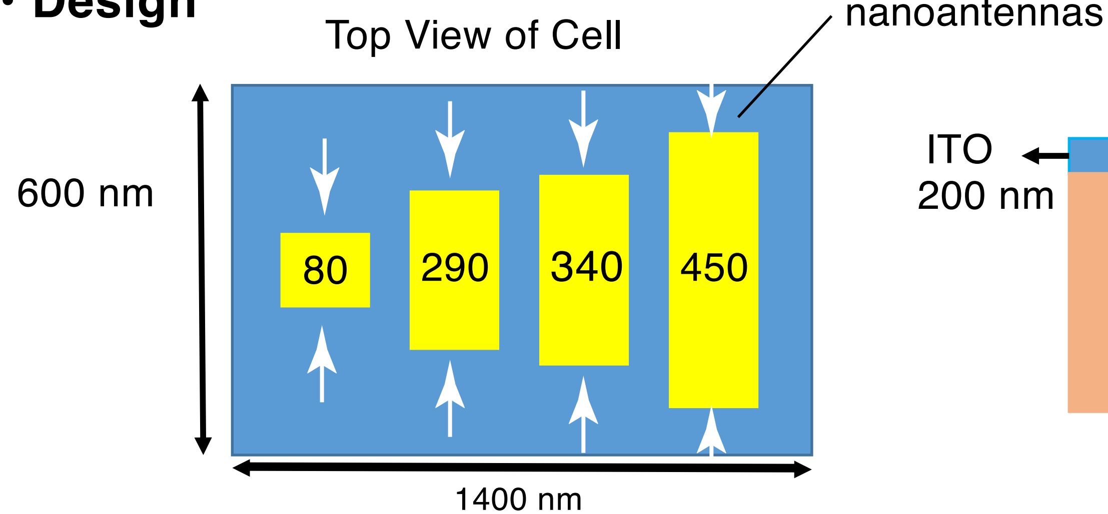
Sub-picosecond response time

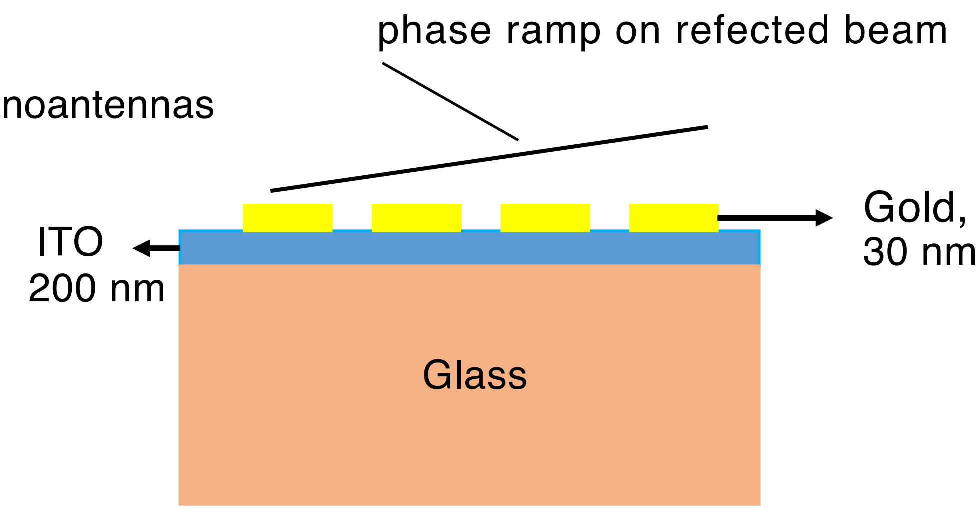
Beam steerer made of one or many cells

Application: Mode-division multiplexing for telecommunications

- **Design**

Top View of Cell





Side View of Cell

Nonlinear response depends on antenna length

- **Characterization**

We have fabricated this design and are currently testing it

Summary: Physics and Applications of ENZ Materials

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

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

Special Thanks To My Students and Postdocs!

Ottawa Group

Rochester Group