An Embarrassment of Riches: What to Do With a Material One Million Times More Nonlinear Than Silica

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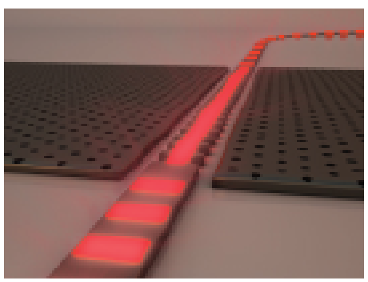
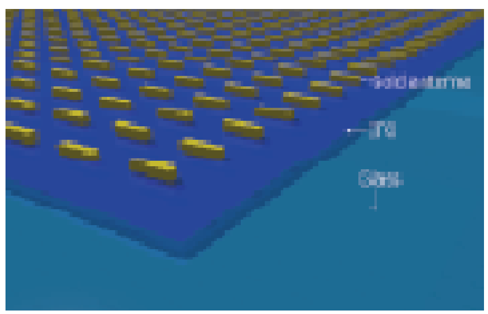
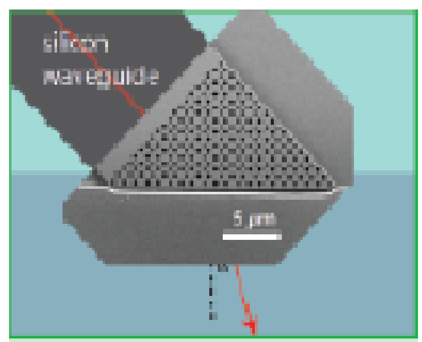
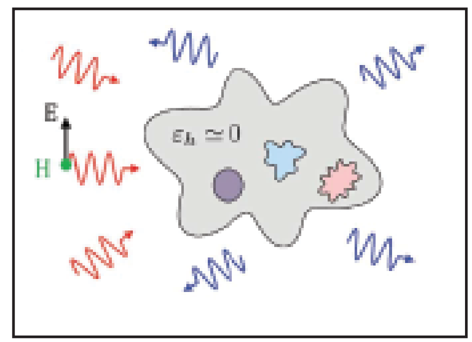






The visuals of this talk are posted at boydnlo.ca/presentations

Presented at Photonics West, San Francisco, January 28, 2025.

How Light Behaves when the Refractive Index Vanishes

- Physics of Near-Zero Index (NZI) and Epsilon-Near Zero (ENZ) Materials
- Nonlinear Optical Properties of ENZ and NZI Materials
- Metamaterials for ENZ and NZI Studies
- Applications of ENZ and NZI Materials

Spoiler Alert: 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

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

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

  Induce and control filamentation processes

- 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)

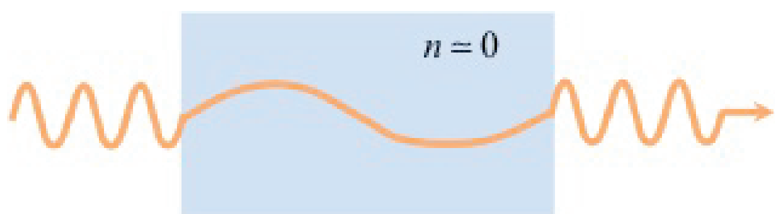
- Our approach: Use epsilon-near-zero (ENZ) materials and metamaterials

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

- The wavelength of light is given by

$$\lambda = \lambda_{\text{vac}}/n$$

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).

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.

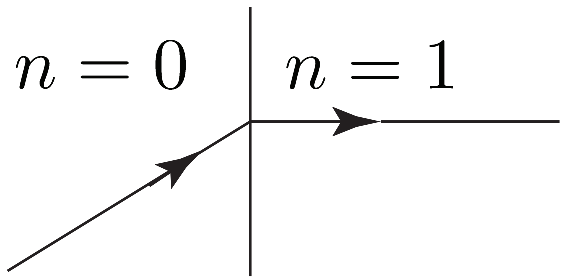
* Lobet, Liberal, Knall, Alam, Reshef, Boyd, Engheta, and Mazur, ACS Photonics 7, 1965-1970 (2020).

Optics of Zero-Index Materials

- Snell's law leads to intriguing predictions

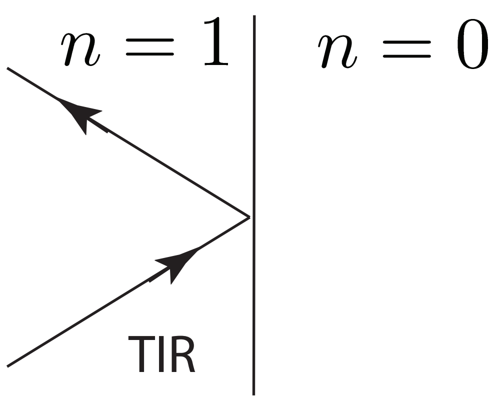
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

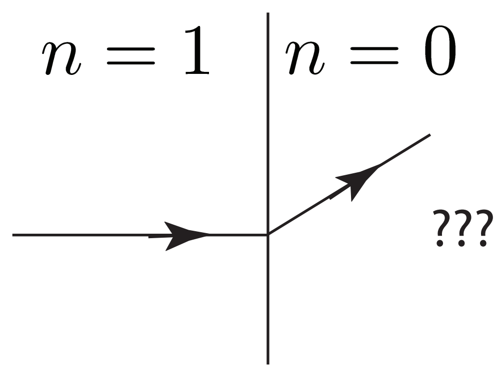
- Light always leaves perpendicular to surface of ENZ material!

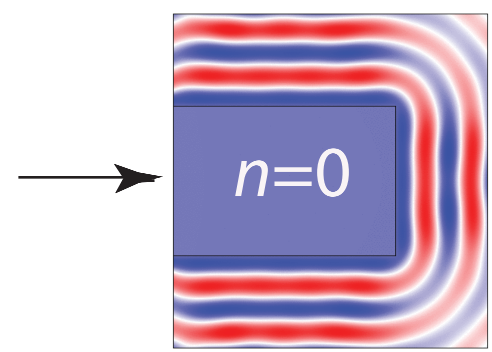


Y. Li, et al., Nat. Photonics 9, 738, 2015; D. I. Vulis, et al., Opt. Express 25, 12381, 2017.

- 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.

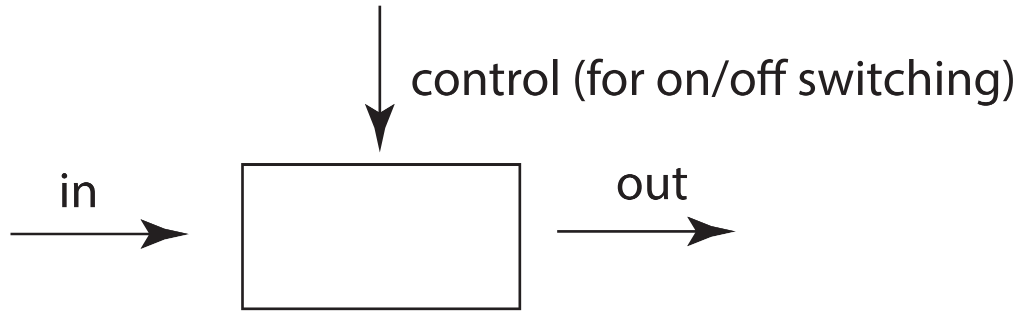
(wave-optics simulation - O. Reshef)

How Light Behaves when the Refractive Index Vanishes

- Physics of Near-Zero Index (NZI) Materials
- Nonlinear Optical Properties of NZI Materials
- Meta-materials for NZI Studies
- Applications of NZI Materials

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 $\chi^{(3)}$

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.

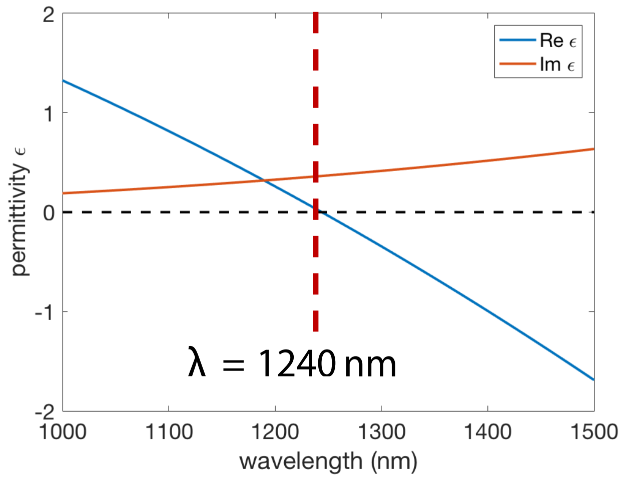
$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

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

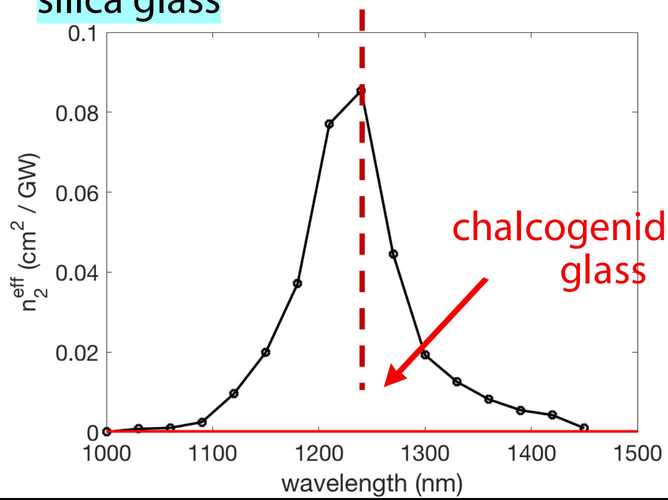
1. Alam, De Leon and Boyd, Science 352, 795–797 (2016)
2. Caspani, Shalaev, Boltasseva, Faccio et al., Phys. Rev. Lett. 116, 233901 (2016).

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

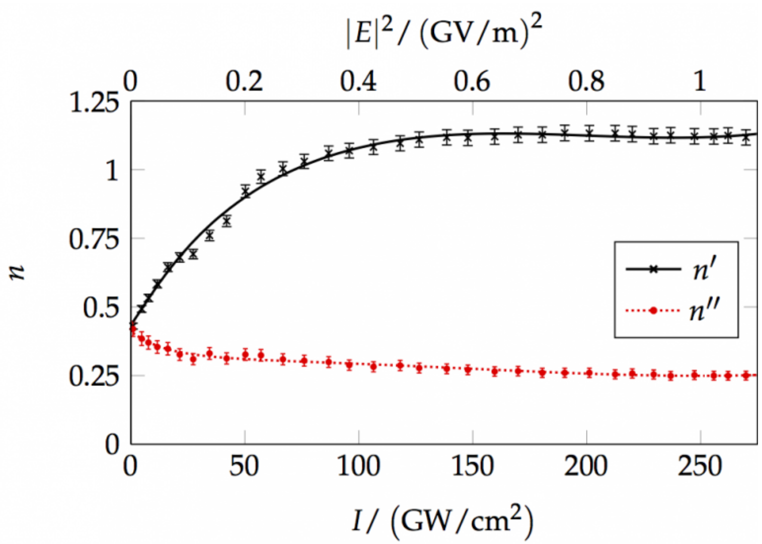
- ellipsometry



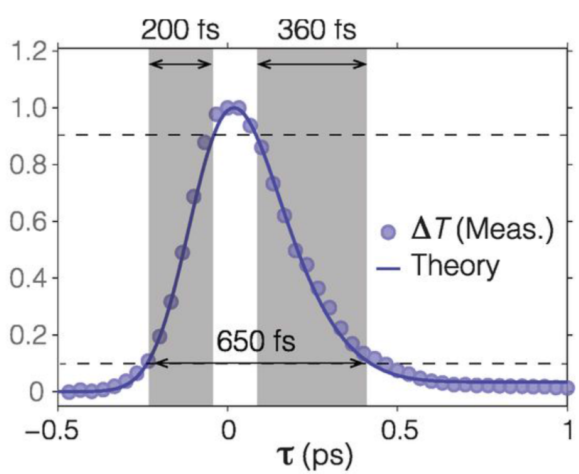
- n_2 can be 3.4 x10^5 times larger than that of silica glass



- overall change in refractive index of 0.8



- sub picosecond reponse time

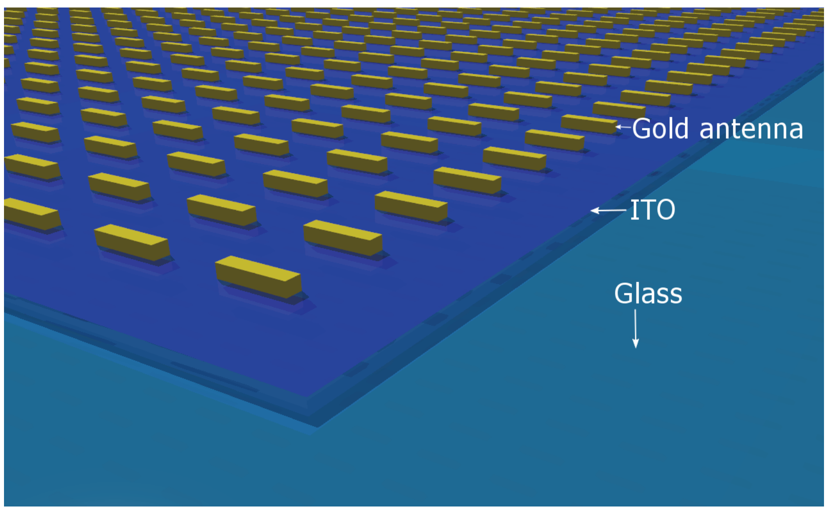


M. Z. Alam et al., Science 352, 795-797 (2016)

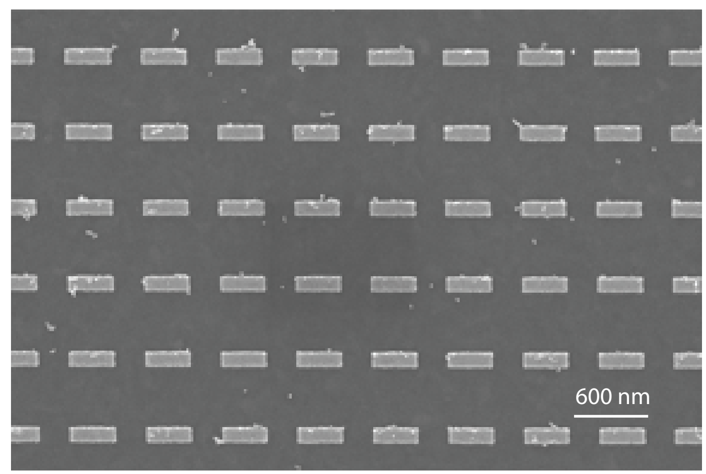
An ENZ Metasurface

- We functionalize ITO by creating a photonic metasurface
- We obtain an even larger NLO response by placing a gold antenna array on top of ITO.
  - Lightning rod effect: antennas concentrate the field within the ITO
  - Coupled resonators: ENZ resonance and nano-antennas

Concept:

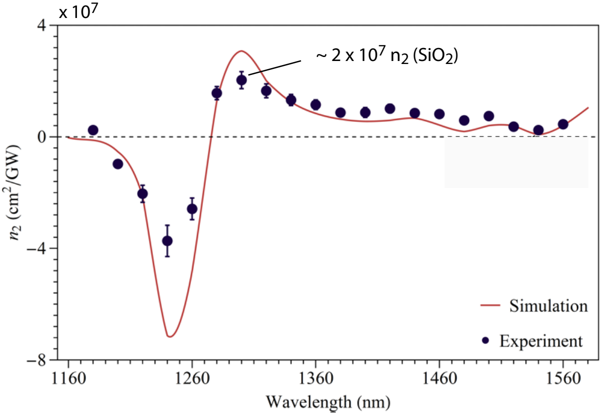


SEM:



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

NLO response of the coupled antenna-ENZ system



The 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.

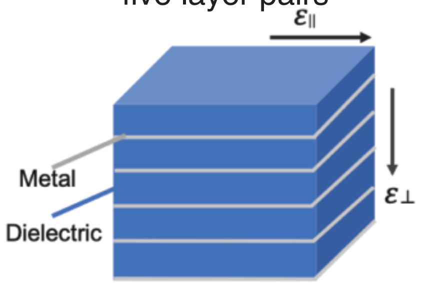
Alam, Schulz, Upham, De Leon and Boyd, Nature Photonics 12, 79-83 (2018).

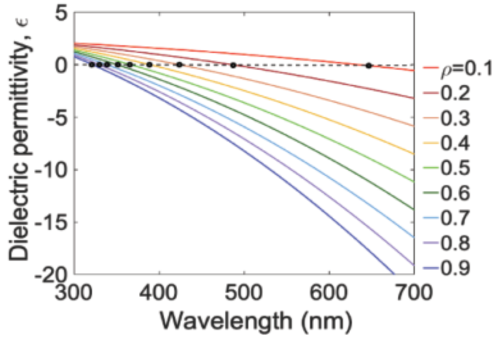
Nonlinear Optical Properties of a Layered Metamaterial in its ENZ Region

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

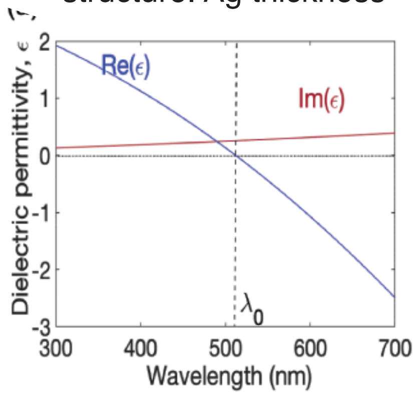
Can we use an effective-medium value of epsilon to determine the ENZ wavelength?

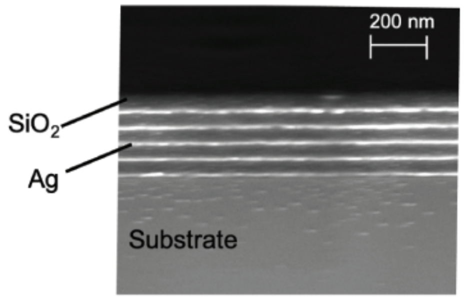
- By controlling the metallic fill fraction ρ, we can set the ENZ wavelength to be anywhere from 300 to 700 nm. We use ρ = 0.2, which corresponds to 500 nm. We deposit five layer pairs



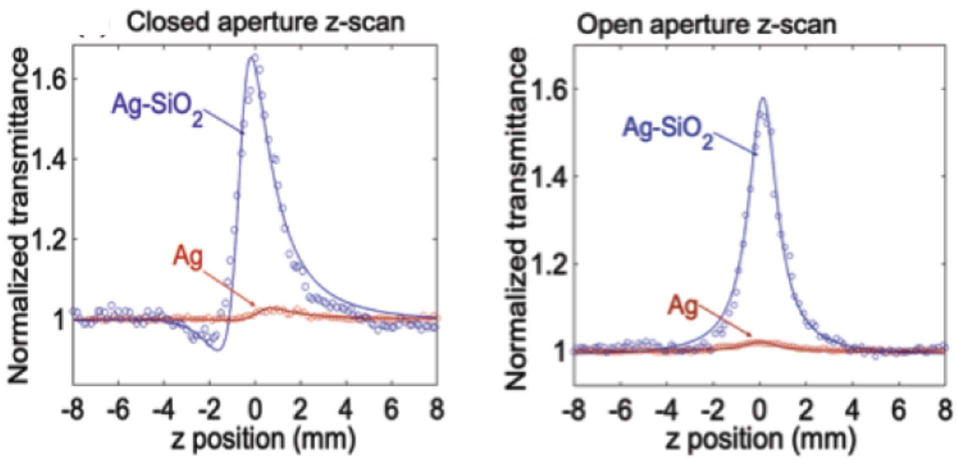


- Note that the real part of epsilon vanishes at 508 nm, close to the design wavelength. The SEM shows our structure. Ag thickness = 16 nm; silica thicness = 65 nm

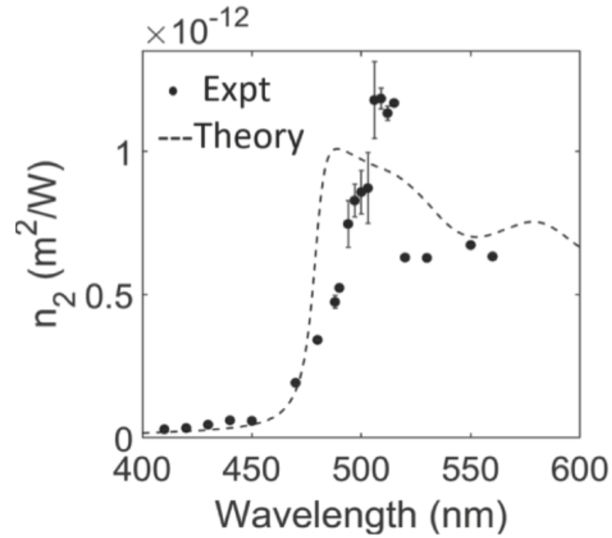




- We perform Z-scan measurements on the sample. Note the enhanced response of the composite as compared to a single layer of silver.



- 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.

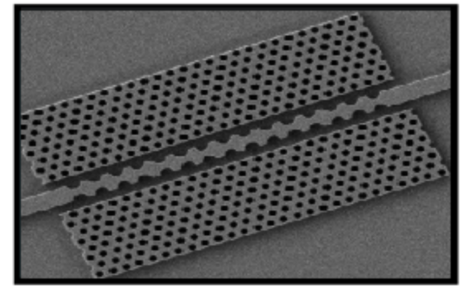


Suresh, Reshef, Alam, Upham, Karimi and Boyd

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

Relaxed Phase-Matching Requirements in ENZ Media



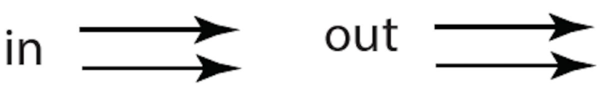
- We study four-wave mixing in a zero-index waveguide

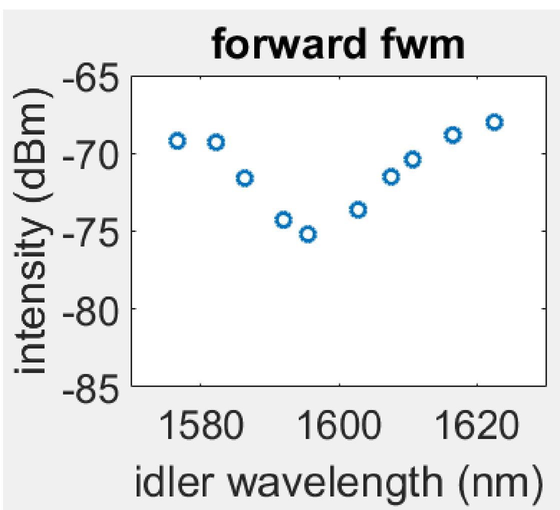
$$2\omega_p = \omega_s + \omega_i$$

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

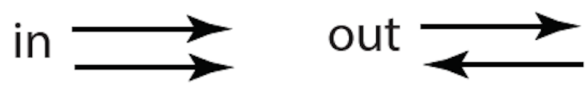
- Recall that we need Δk = 0, but when n = 0, k = n ω /c vanishes for each of the interacting waves and thus so does Δk.

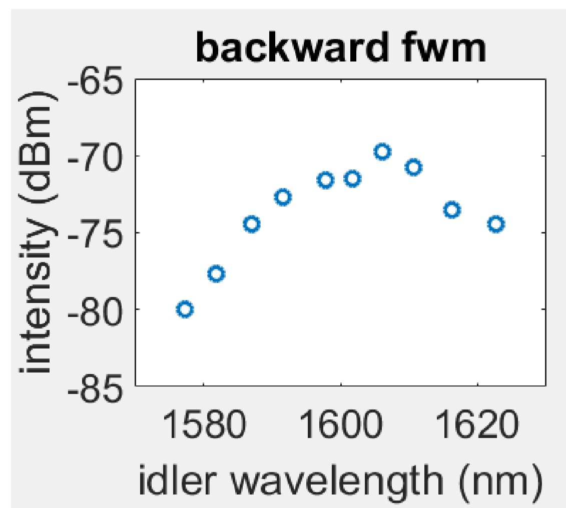
“forward” process





“backward” process

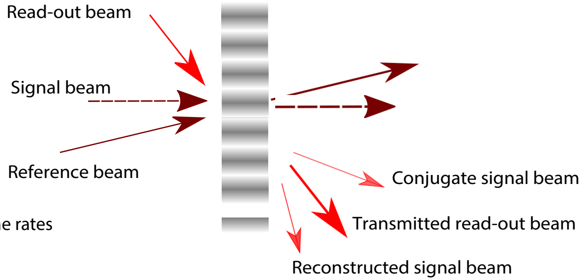




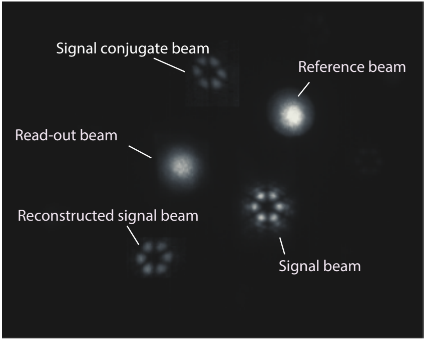
- Significance: Nonlinear optical processes that were previously believed to be too weak to be useful can be excited through use of ENZ materials.

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

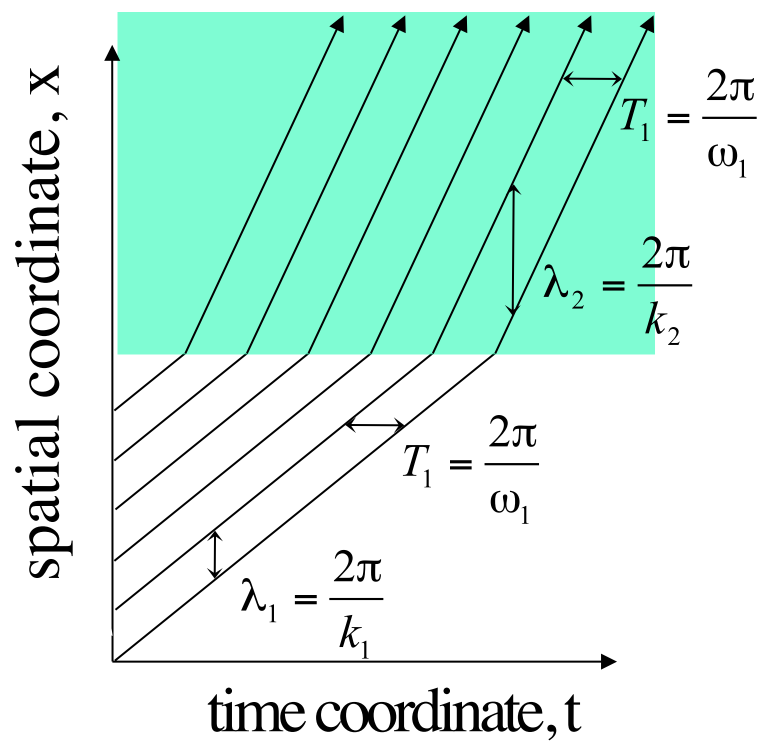
Spatial and temporal refraction

Spatial (ordinary) refraction.

Refractive index changes at some spatial coordinate.

Frequency remains unchanged, but the wavelength changes according to

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

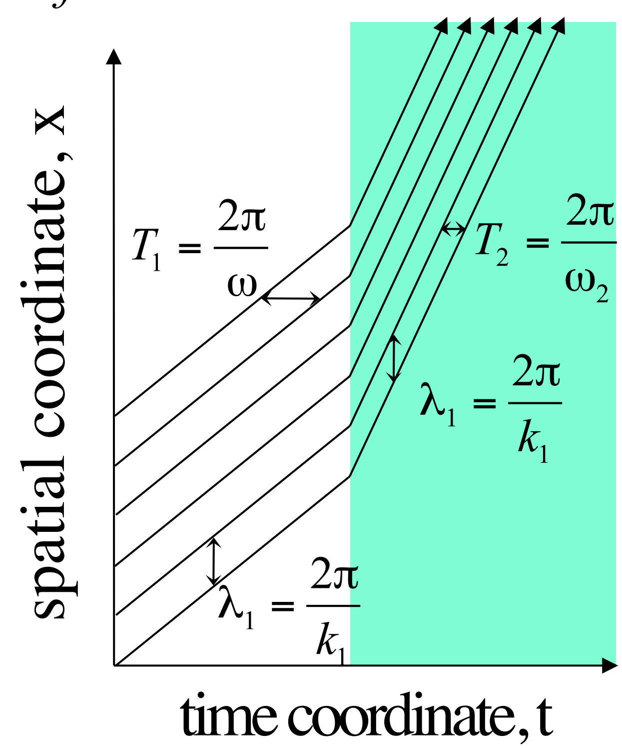


Time refraction.

Refractive index changes at some moment in time

Wavelength remains unchanged, but the frequency changes according to

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

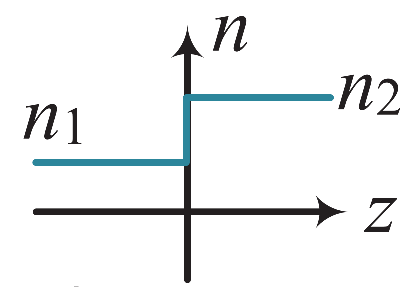


Salem, Foster, and Gaeta, Advances in Optics and Photonics 5, pp. 274-317 (2013).

Adiabatic Wavelength Conversion through Time Refraction

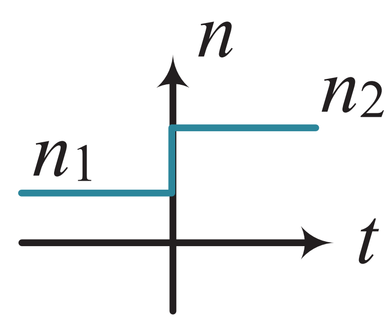
- Space refraction (e.g., Snell's law)

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



- Time refraction (analog of Snell's law)

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

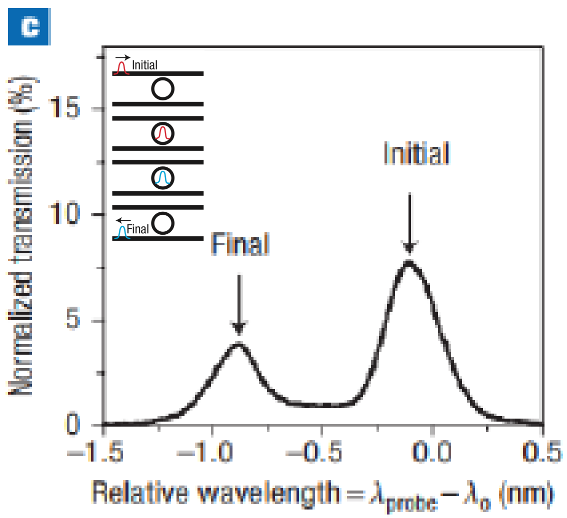


Photon frequency (energy) is changed because of the temporal change in index, but the wavelength (inverse of momentum) is conserved in the absence of any spatial asymmetry

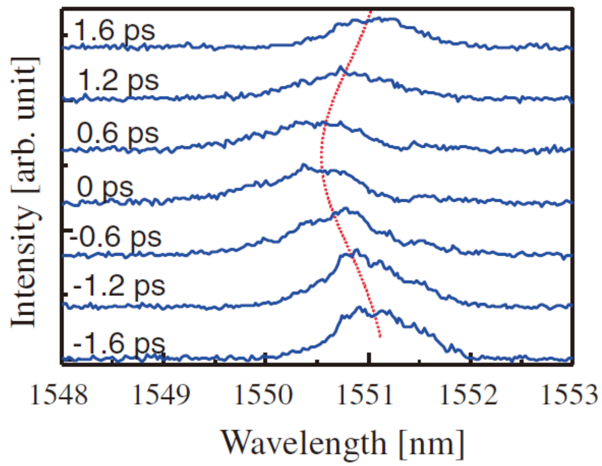
- Time refraction is an alternative way of understanding frequency broadening and shifting by self-phase modulation:

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

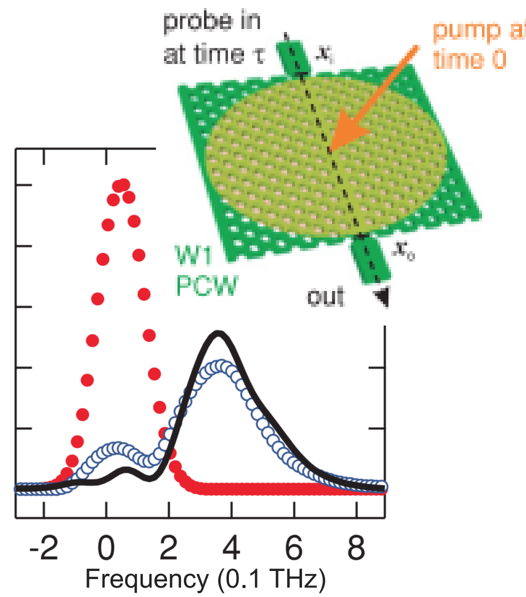
Previous work on adiabatic wavelength conversion



Nat. Photonics, 2007, 1(5): 293



Appl. Phys. Ex., 2010, 3(6): 062001.



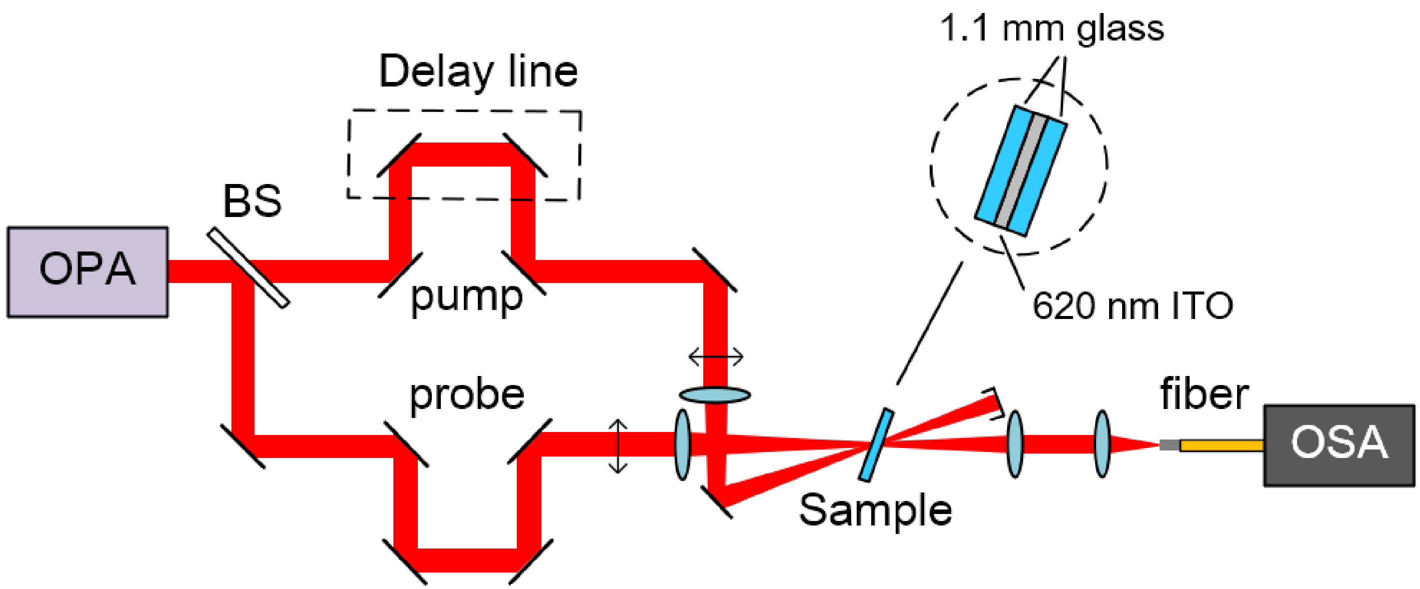
Phys. Rev. A., 2010, 81: 043837.

For small change in index $\Delta n \sim 10^{-4}$, one gets a small change in frequency (e.g., 0.06%)

But with ENZ materials we can obtain a much larger Δn

Laboratory Study of Wavelength Conversion by Time Refraction

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



OPA = optical parametric amplifier

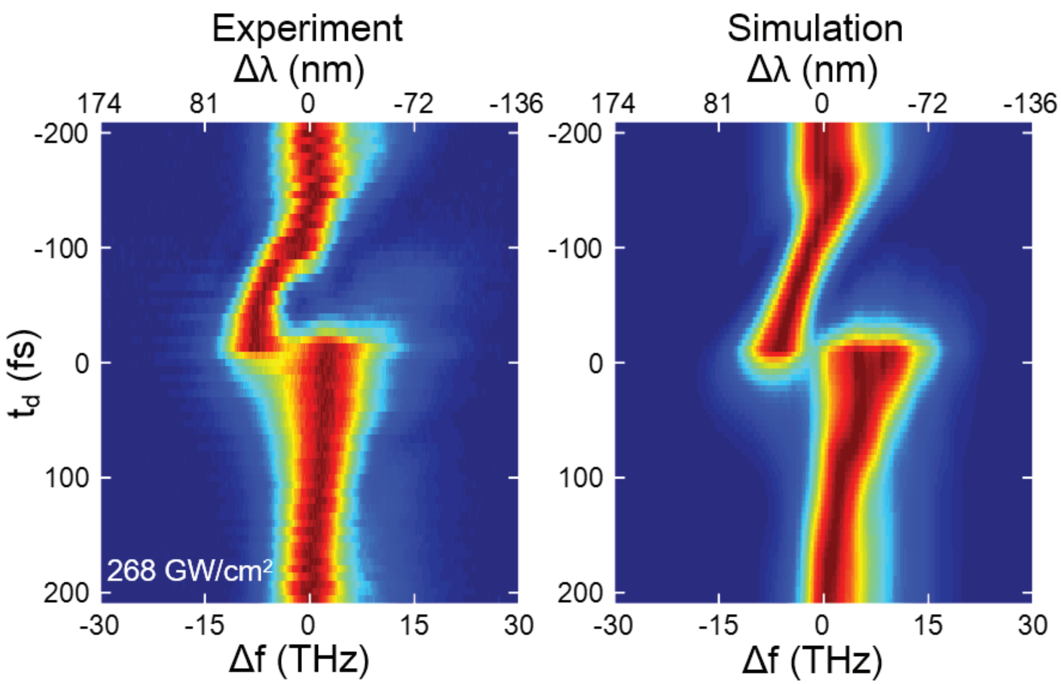
wavelength = 1240 nm

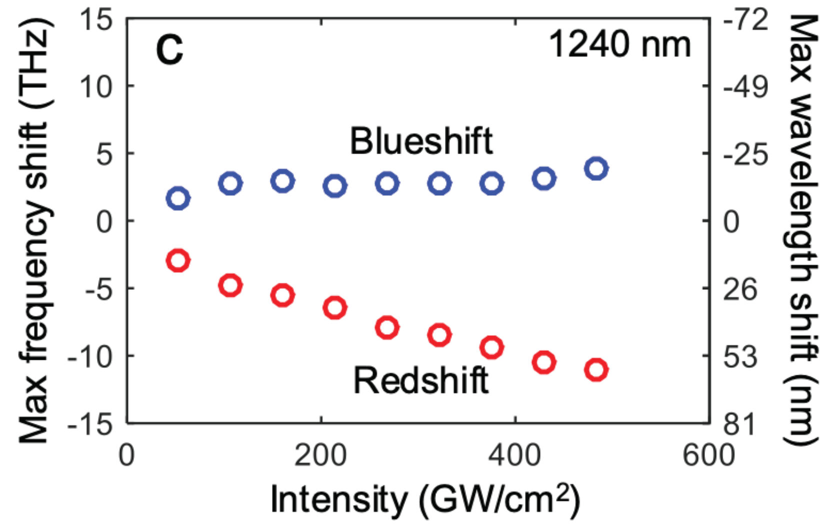
pulse duration = 120 fs

OSA = optical spectrum analyzer

Results: Adiabatic Wavelength Conversion by Time Refraction

Experimental results at 1240 nm





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

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

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

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

Imaging through a Strongly Scattering Medium

We want an imaging method that

- Preserves the spatial resolution of the object
- Is background free
- Ideally converts image to a desirable wavelength

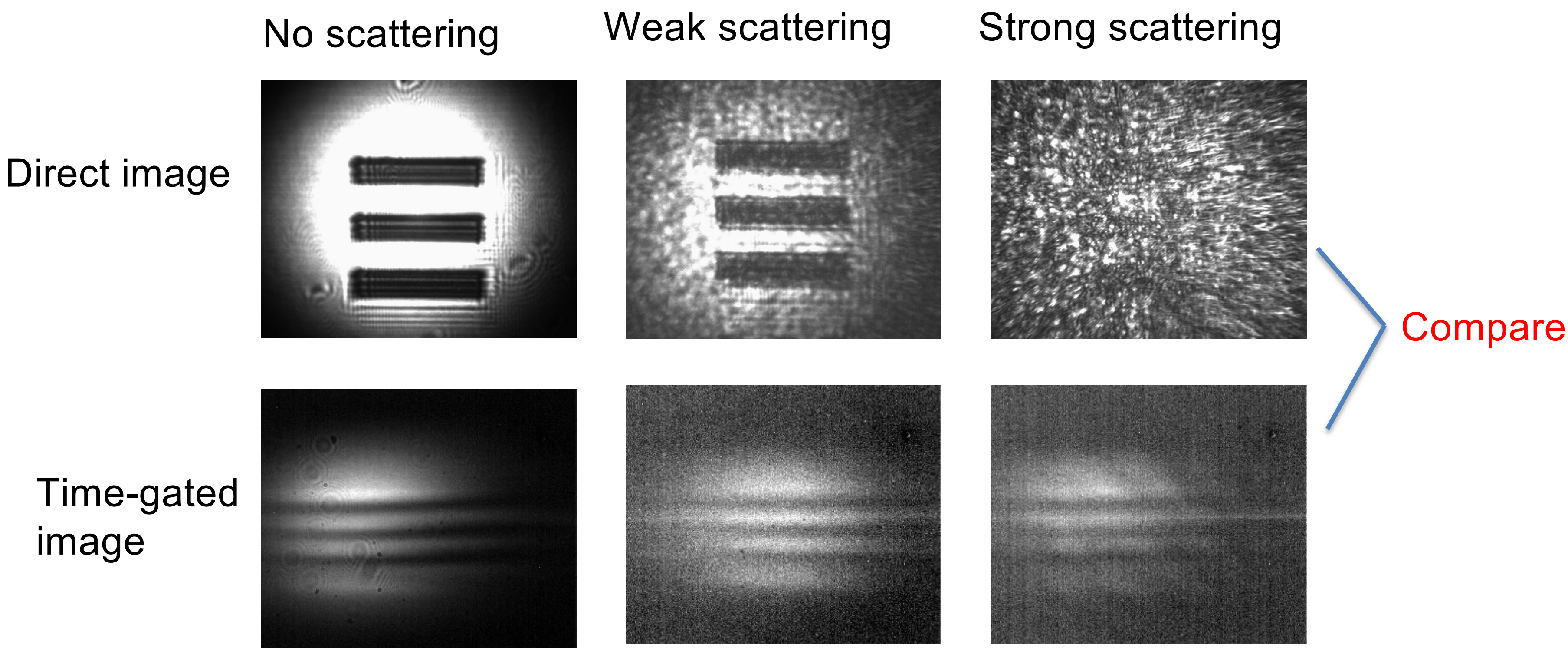
Our approach involves time-gating using a highly nonlinear ENZ material

- Time-gate transmits only the unscattered photons, which contain the image information

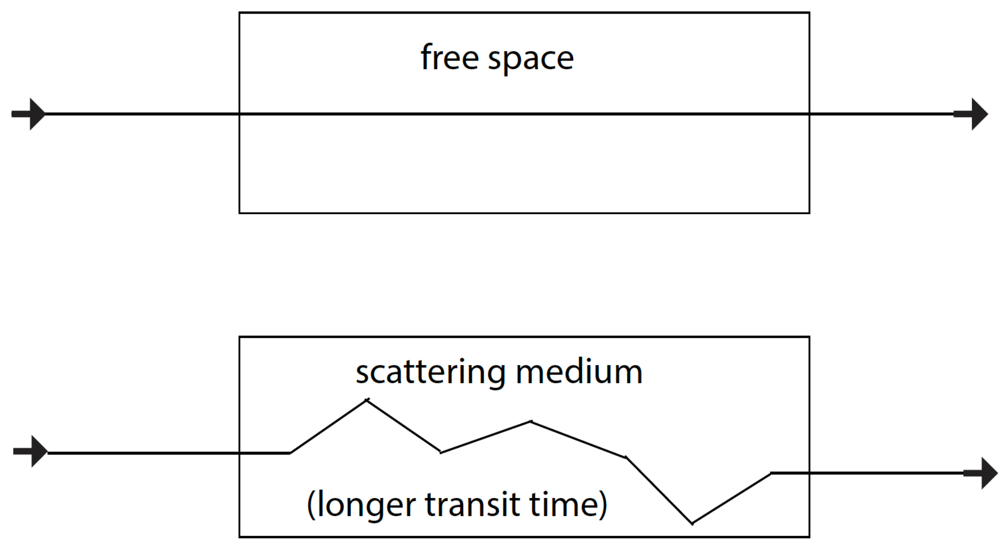
Useful for

- Non-invasive biomedical imaging and tomography
- Optical including OAM-based communication through atmospheric turbulence

We use time-gating to measure only the first-arriving photons



Need material with a strong, fast nonlinear optical response to construct gate. Use ITO.

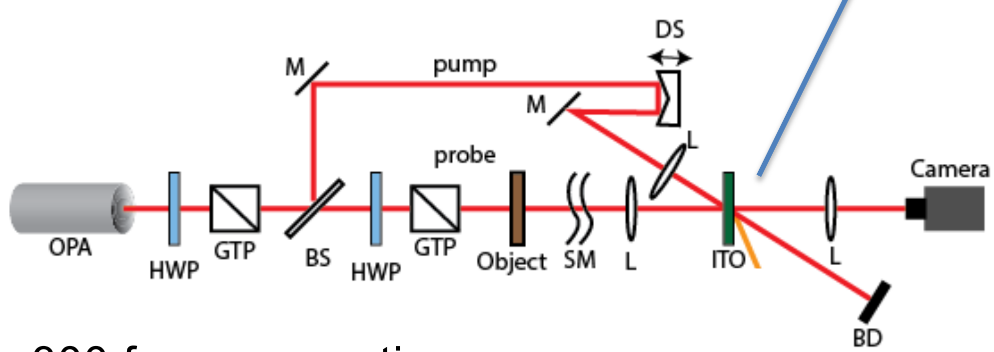


See also Wang et al (Alfano group) Science 253, 769 (1991),

Experiment Setups

Nonlinear Transmission Setup
(using a Kerr Gate)

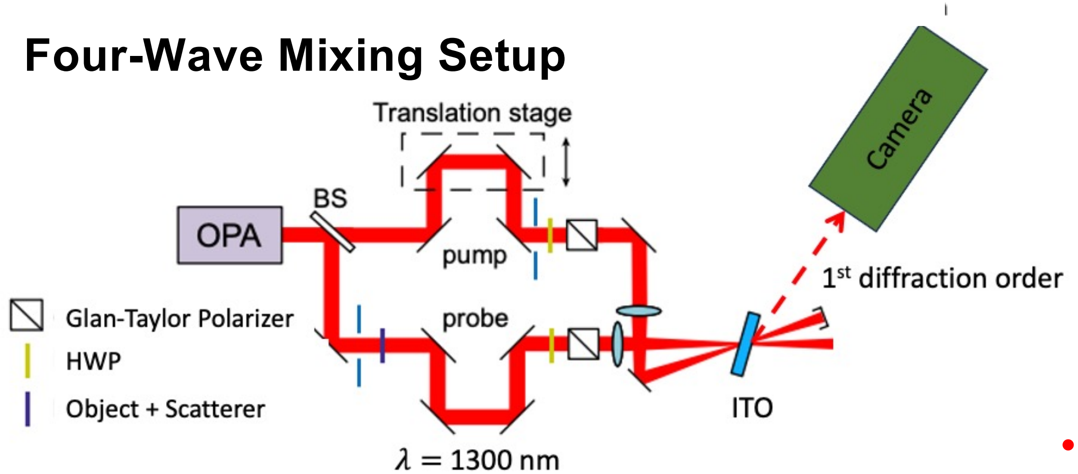
Probe transmission shuttered by pump pulse in a "Kerr gate."



300 fs response time

GTP: Glan-Taylor polarizer
SM: scattering media
BS: beam splitter
BD: beam dump
DS: delay stage
L: lens
M: mirror

Four-Wave Mixing Setup



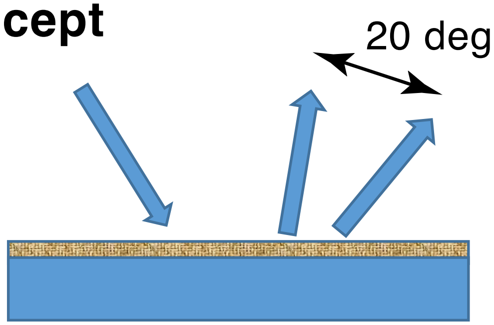
120 fs response time

Pump and probe are both centered at the ENZ wavelength (1240 nm) of the 310-nm-thick ITO plate

- We use the four-wave mixing setup because it gives a shorter gating time and suppresses background.

All-Optical, Nanoscale, Sub-Picosecond Beam Steering

- **Concept**



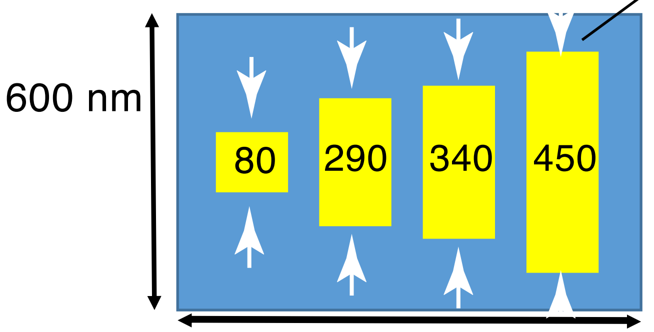
Tune output direction by
+/- 20 degrees under
optical or electrical control

Beam steerer mcan be made
of one or many cells

Sub-picosecond response time

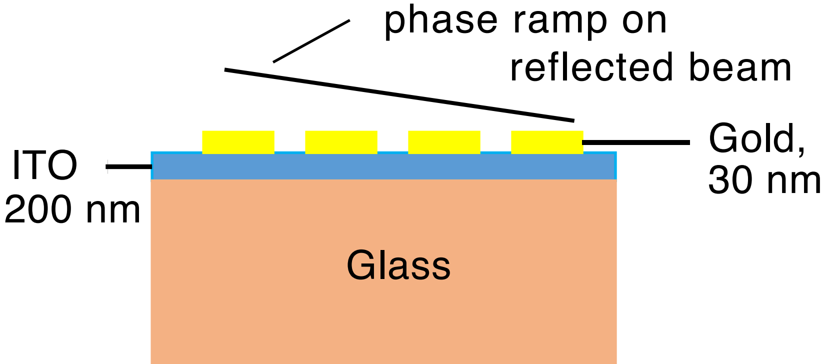
- **Design**

Top View of Cell



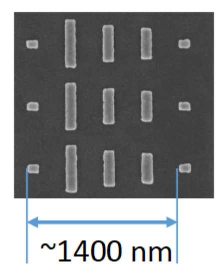
nanoantennas

Nonlinear response
depends on
antenna length

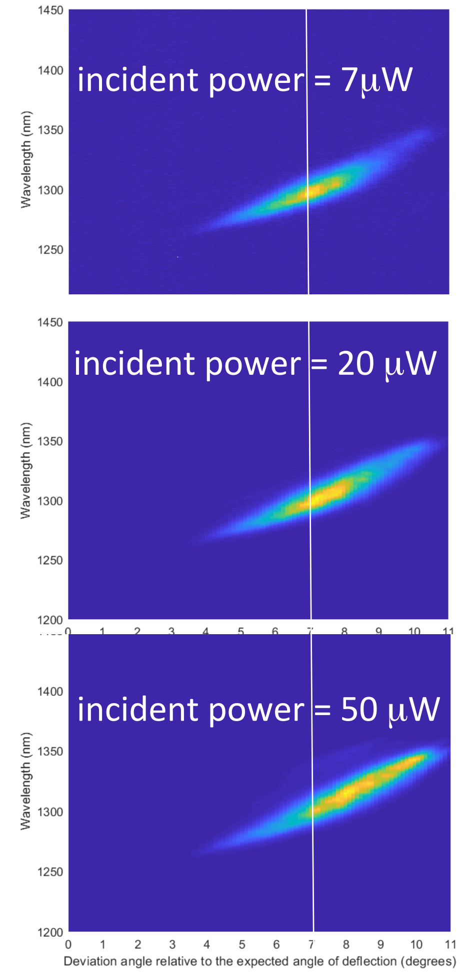


Side View of Cell

- SEM



- **Results**



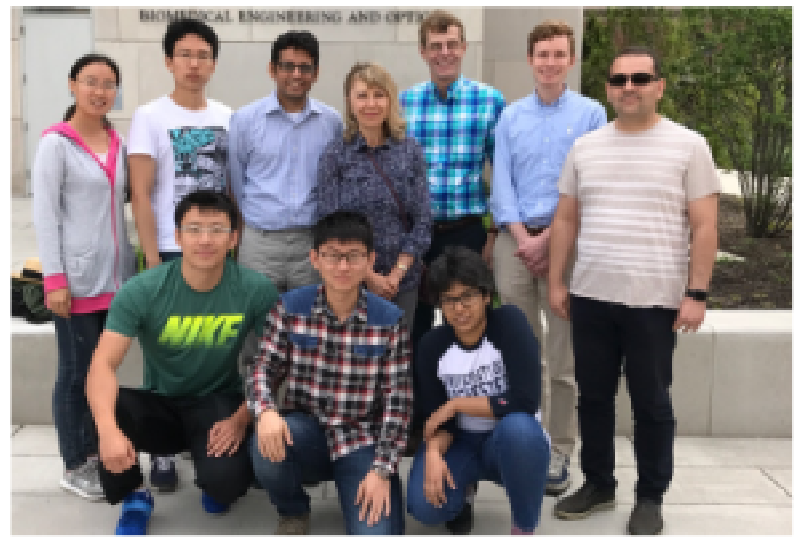
One application: Mode-division multiplexing for
telecommunications

M. Karimi, Alam, Upham, Reshef and Boyd, Nanophotonics (2023).

Special thanks to:

- Nader Engheta, Eric Mazur and Alan Willner for close collaboration throughout this project.
- DARPA, ARO, and NSERC for financial support.
- And especially to my students and postdocs

Rochester Group



Ottawa Group

Why Does ENZ Lead to Large NLO Response?

1. From form of n_2 $\quad n_2 = \dfrac{3\chi^{(3)}}{4\epsilon_0 c\, n_0\, \mathrm{Re}(n_0)}$

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

$$n = \sqrt{\epsilon_b + \Delta\epsilon} \approx \sqrt{\epsilon_b}\left(1 + \frac{\Delta\epsilon}{2\epsilon_b}\right) = n_b + \frac{\Delta\epsilon}{2n_b} \text{ and thus } \Delta n = \frac{\Delta\epsilon}{2n_b}$$

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. From Maxwell's equations, it is easy to show that the nonlinear response scales as

$$\frac{\left.\dfrac{dH_x}{dz'}\right|_{\mathrm{nl}}}{|H_x|} \propto \sqrt{\frac{\mu_r}{\epsilon_r}}$$

5. Detailed numerical integration confirms this behavior.

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

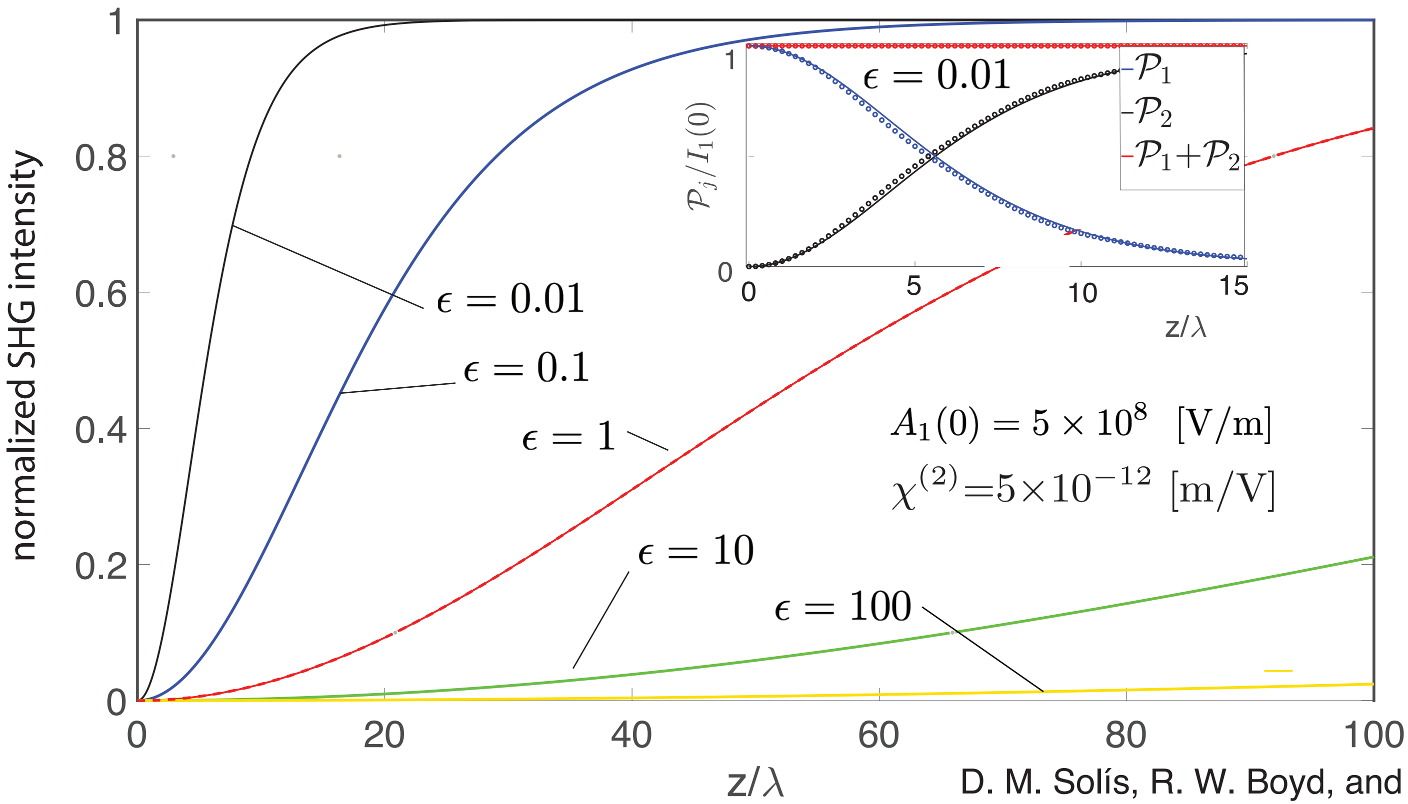
- We solve the standard equations for second-harmonic generation

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

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

- 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.



D. M. Solís, R. W. Boyd, and N. Engheta, Laser & Photonics Rev., 2100032 (2021).

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

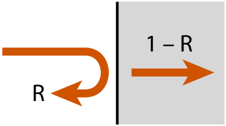
- There is a problem getting light into a zero-index material.

- There is always reflection from the boundary between two materials

- The impedance and surface reflectivity are given by

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

- Thus the reflectivity will be 100% if $\epsilon = 0$ unless $\mu = 0$ as well.



- This is one reason for the interest in developing EMNZ materials (epsilon and mu near zero materials.

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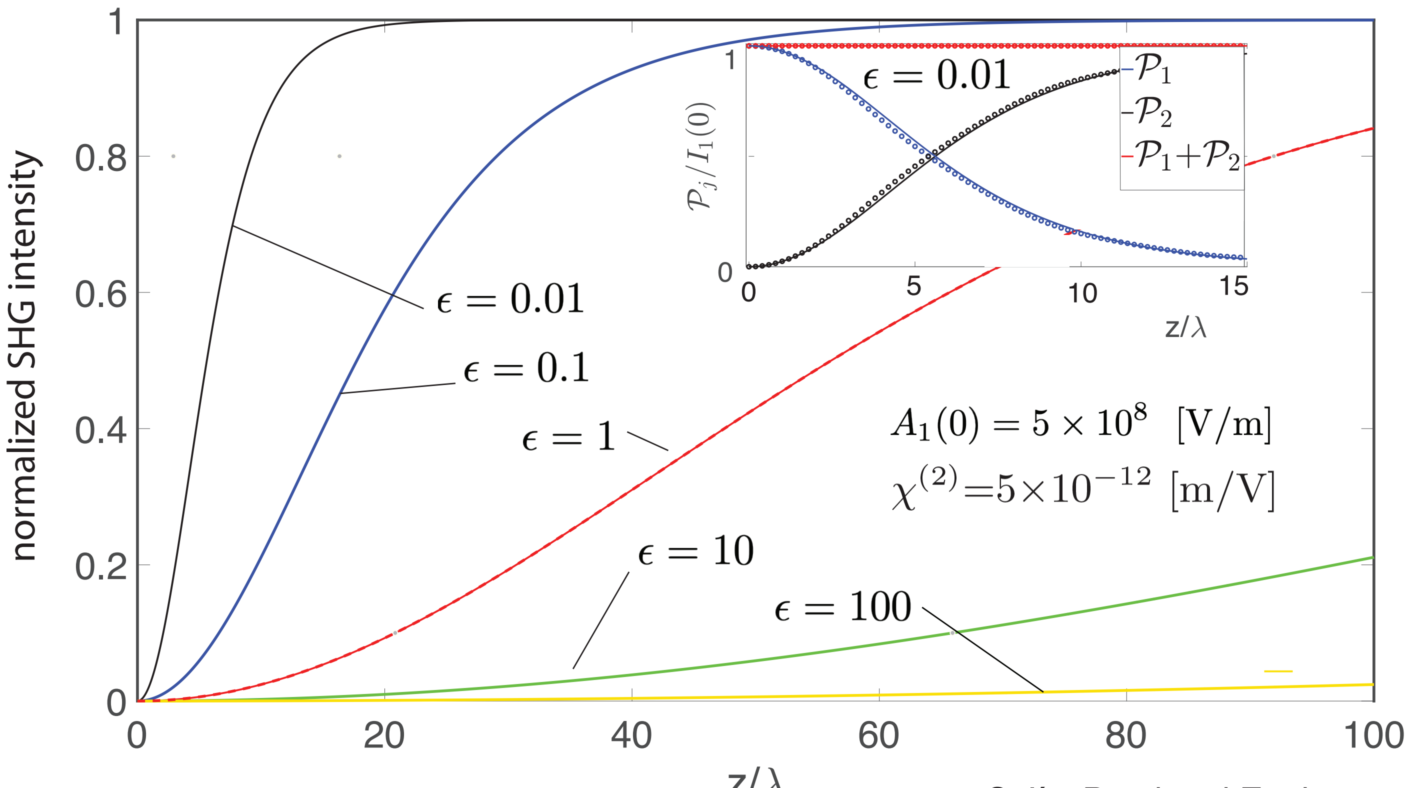


Solís, Boyd and Engheta, to be published

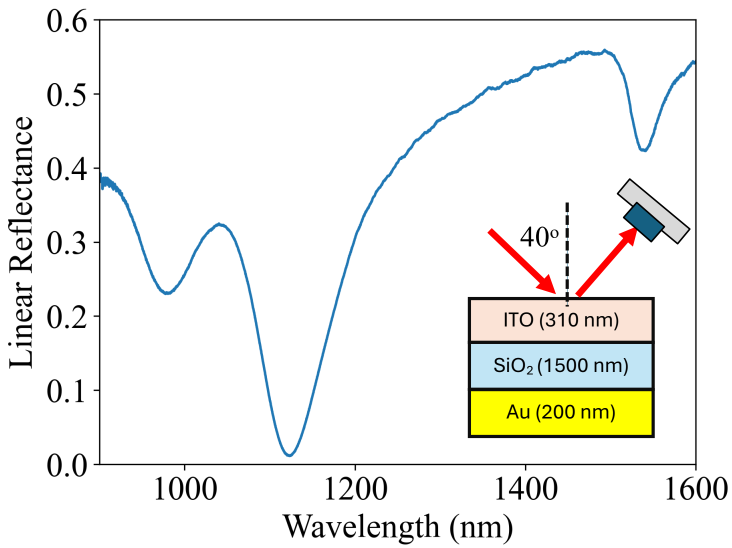
See also Solís, CLEO Poster JW2D.15

Cavity-Enhanced ENZ-Based DFWM

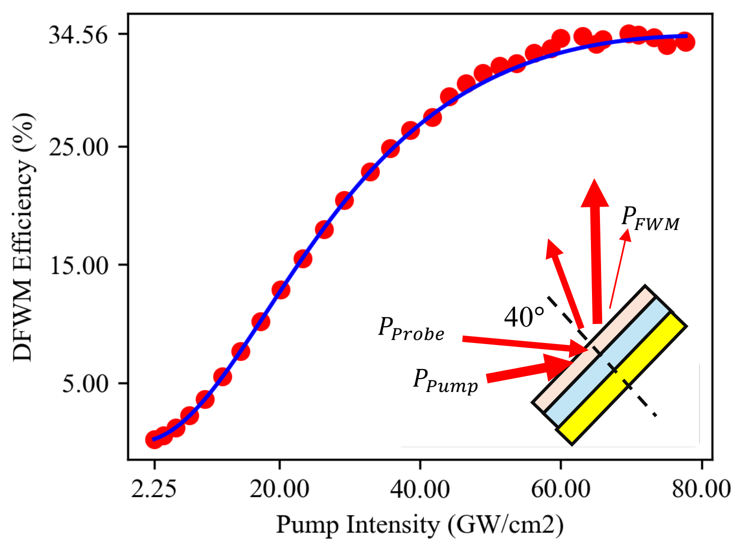
Theng-Loo Lim
NpTh3C.5 • 15:00

- We fabricated an ENZ-based nanocavity and studied the enhanced degenerate four-wave mixing (DFWM) from the device.
- The linear measurement suggests that the resonance mode of the cavity is located at 1130 nm with an angle of incidence of 40 degrees.
- The observed maximum DFWM efficiency is ~34% in nonlinear measurements.

Linear measurement



Nonlinear measurement

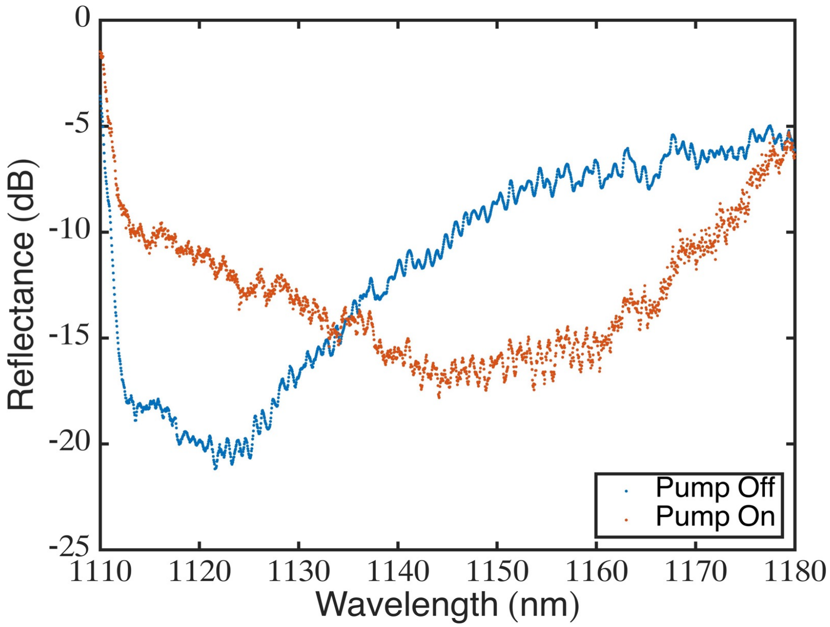


Cavity-Enhanced ENZ-Based Ultrafast Optical Switching

Yaswant Vaddi
NpTh3C.1 • 14:00

- We experimentally demonstrate an ultrafast all-optical switch using a 1D, nonlinear nanocavity with an epsilon-near-zero mirror. The switch exhibits a 10 dB modulation depth over a large spectral range.

Probe reflectance with and without pump presence



Switching contrast wrt pump energy density