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

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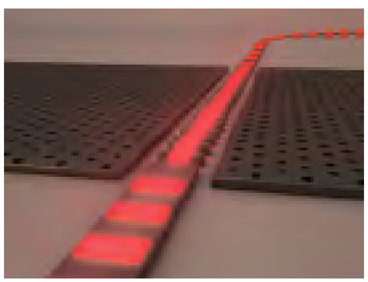
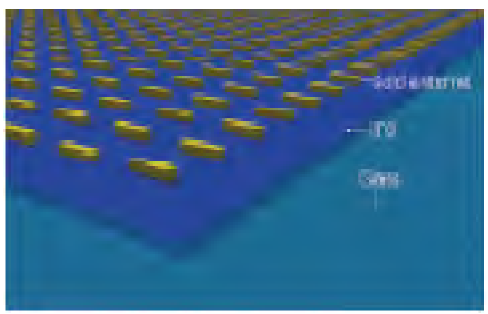
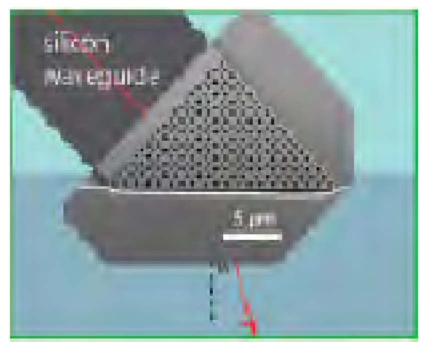
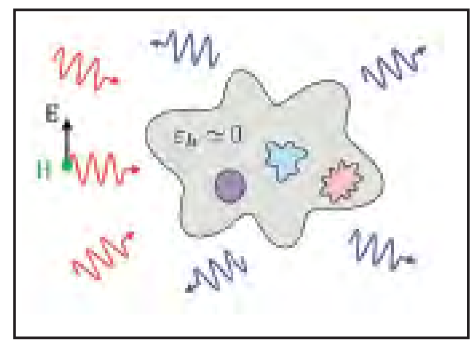








The visuals of this talk are posted at boydnlo.ca/presentations

Presented at the Optica Society's Conference on Advanced Photonics, Québec City, July 31, 2024.

Giant Nonlinear Response of ENZ Materials

- 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

Implications of ENZ Behavior for Nonlinear Optics

Here is the intuition for why the ENZ condition is of interest in NLO

ENZ means epsilon near zero. Here epsilon is the dielectric permittivity.
Recall that $n = \text{sqrt(epsilon)}$

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

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

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.

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

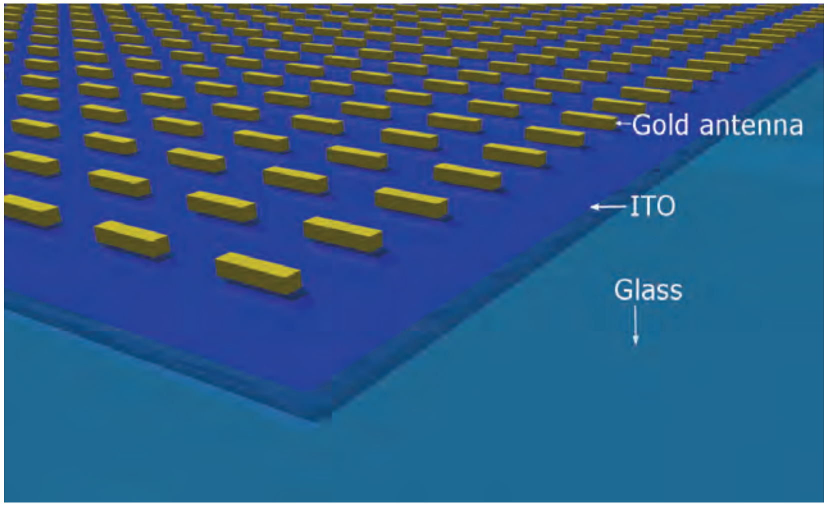
- Note that aluminum-doped zinc oxide (AZO), another transparent conducting oxide, also has strong nonlinear response at its ENZ wavelength [2].

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

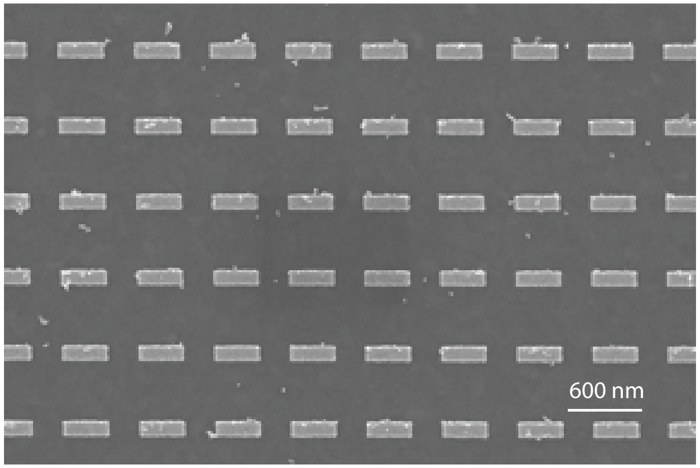
An ENZ Metasurface

- 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:

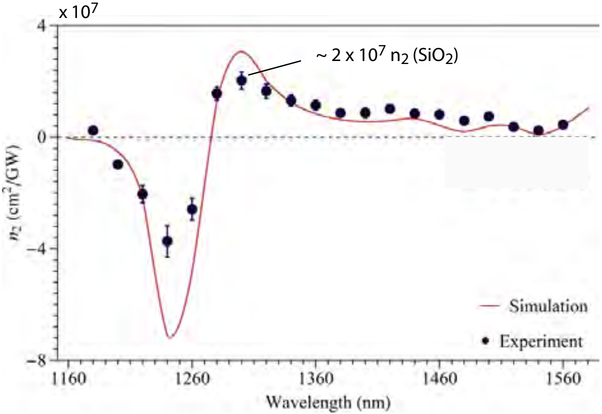


SEM:



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.

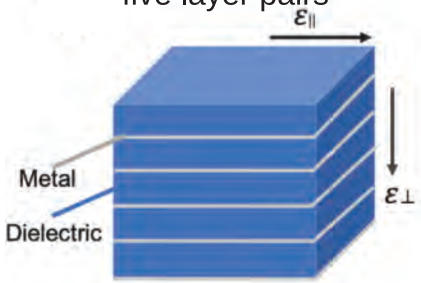
Alam, Schulz, Upham, De Leon and Boyd, Nature Photonics 12, 79-83 (2018).

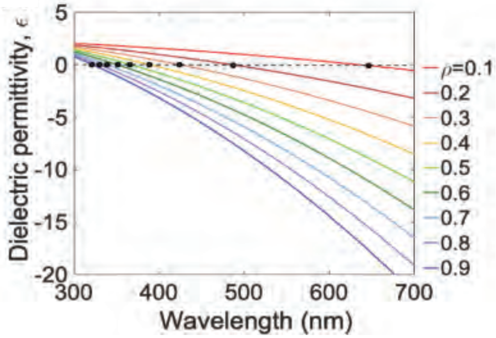
Nonlinear Optical Properties of a Layered Metamaterial in its ENZ Region

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

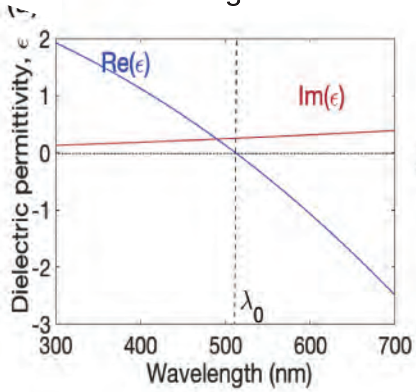
Can we use an effective-medium value of epsilon to determine the ENZ wavelength?

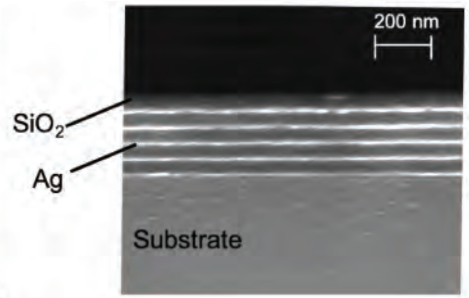
- By controlling the metallic fill fraction ρ, we can set the ENZ wavelength to be anywhere from 300 to 700 nm. We use ρ = 0.2, which corresponds to 500 nm. We deposit five layer pairs



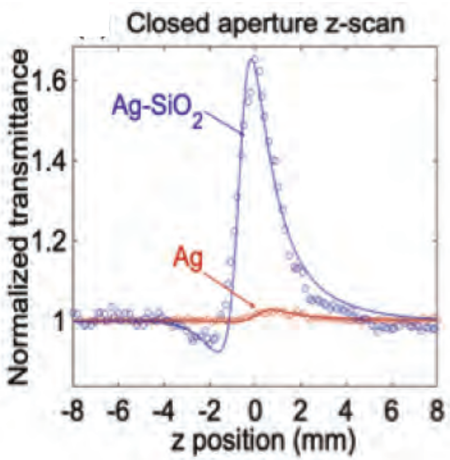


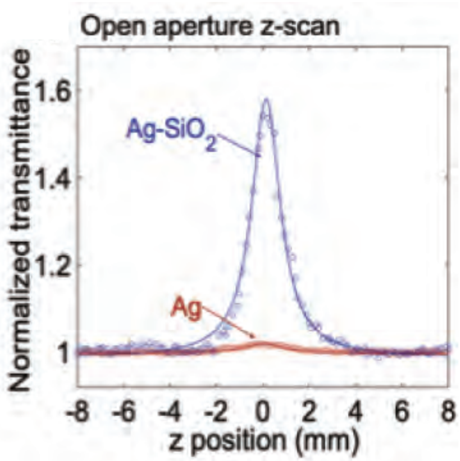
- Note that the real part of epsilon vanishes at 508 nm, close to the design wavelength. The SEM shows our structure. Ag thickness = 16 nm; silica thicness = 65 nm



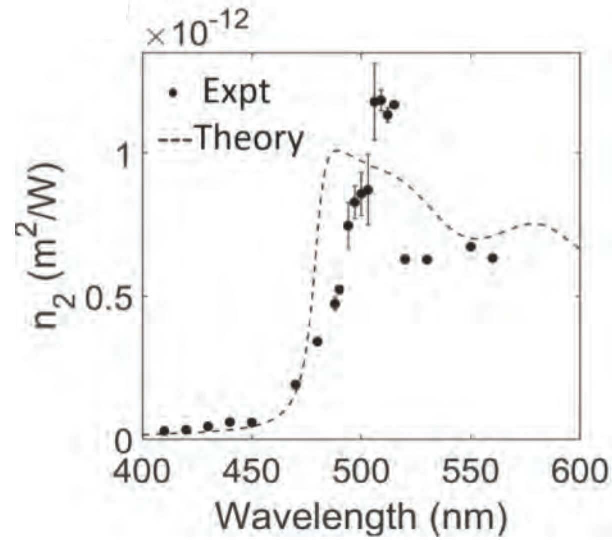


- We perform Z-scan measurements on the sample. Note the enhanced response of the composite as compared to a single layer of silver.



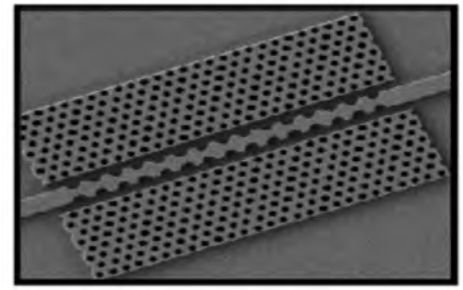


- Note the pronounced peak in the value of n_2 around the ENZ wavelength. We find a good but not perfect agreement with a simple effective medium theory.



Suresh, Reshef, Alam, Upham, Karimi and Boyd, ACS Photonics 8, 125–129 (2021)

Relaxed Phase-Matching Requirements in ENZ Media

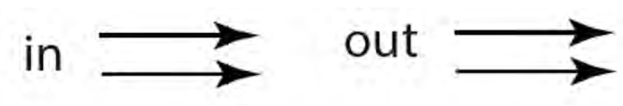


- We study four-wave mixing in a zero-index waveguide

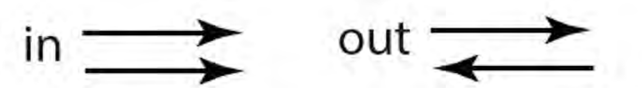
$$2\omega_p = \omega_s + \omega_i$$

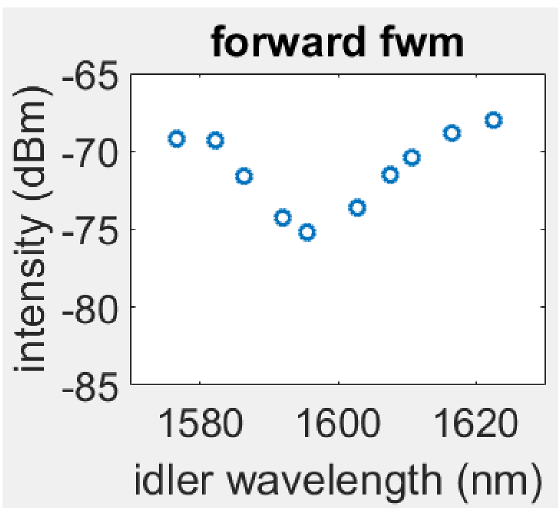
- We find that an idler field is generated in both the forward and backward directions!
- Recall that we need $\Delta k = 0$, but when $n = 0$, $k = n\,\omega/c$ vanishes for each of the interacting waves and thus so does Δk.

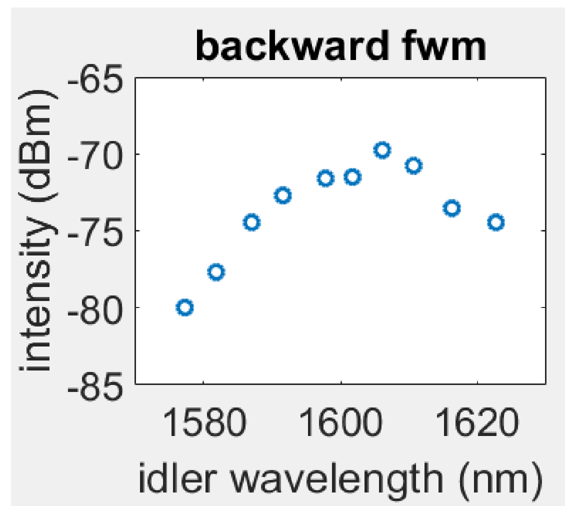
“forward” process



“backward” process





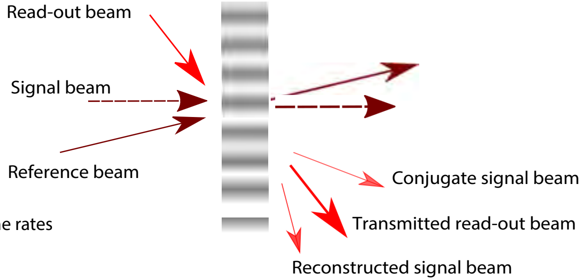


- Significance: Nonlinear optical processes that were previously believed to be too weak to be useful can be excited through use of ENZ materials.

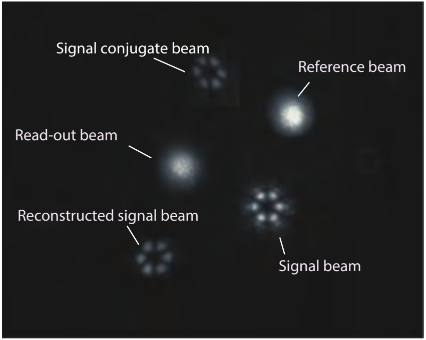
"Relaxed Phase-Matching Constraints in Zero-Index Waveguides," Gagnon et al., Phys. Rev. Lett. 128, 203902 (2022).

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)



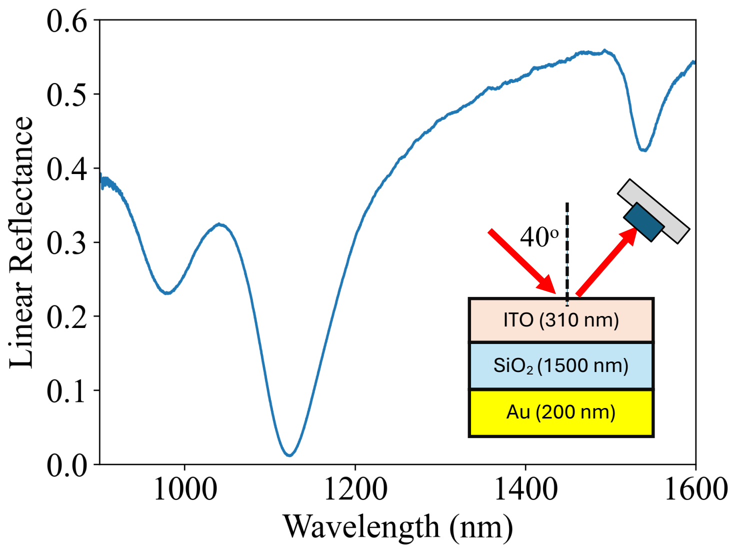
Alam, Fickler, Reshef, Giese, Upham, and Boyd

Cavity-Enhanced ENZ-Based DFWM

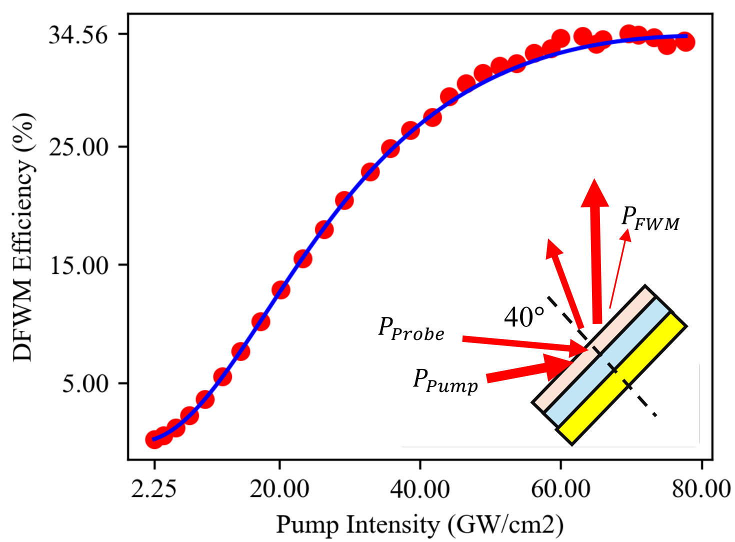
Theng-Loo Lim
NpTh3C.5 • 15:00

- 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

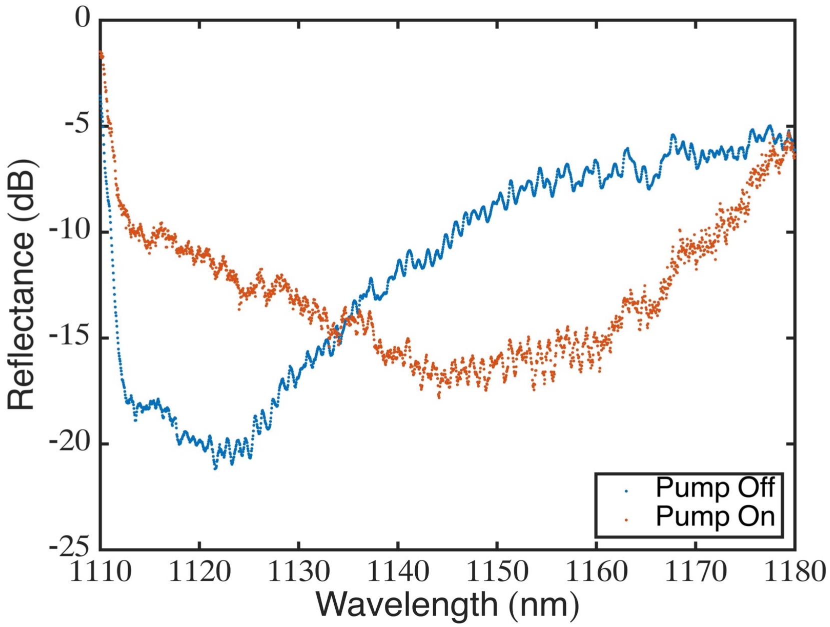


Cavity-Enhanced ENZ-Based Ultrafast Optical Switching

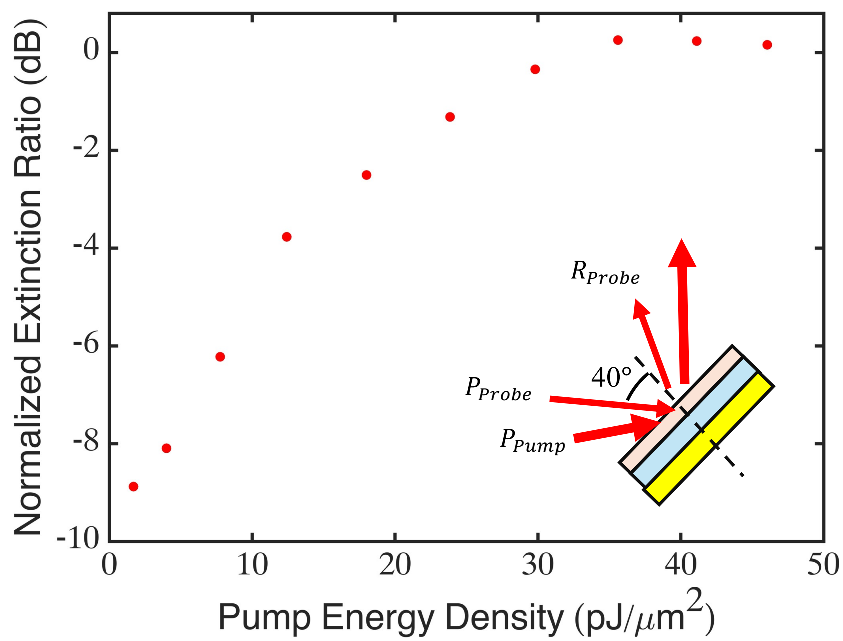
Yaswant Vaddi
NpTh3C.1 • 14:00

- 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.

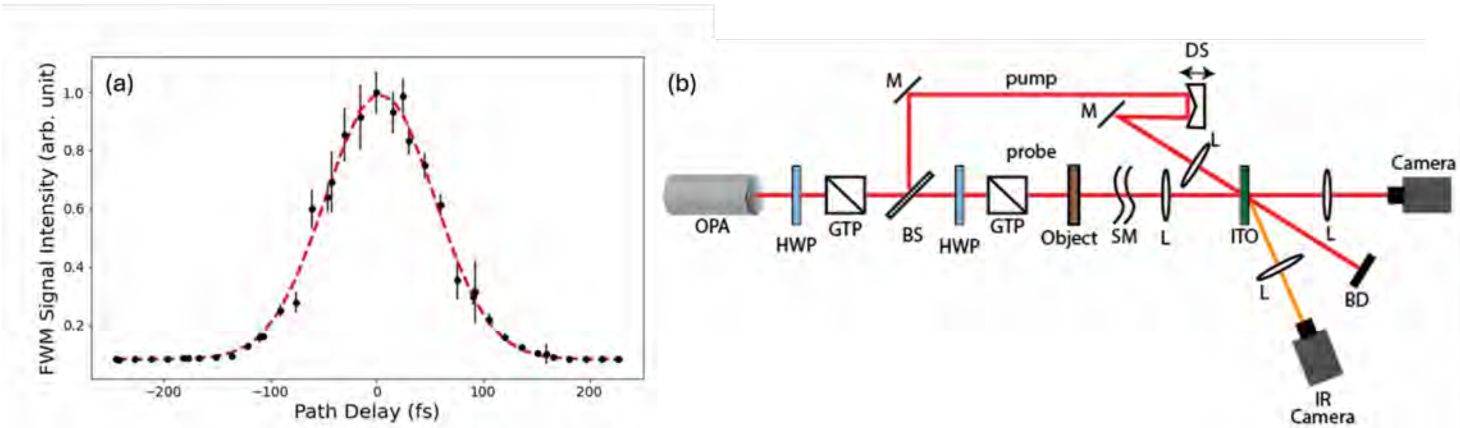
Probe reflectance with and without pump presence



Switching contrast wrt pump energy density



Imaging Through Scattering Media with Ultrafast Spatiotemporal Gating Using Epsilon-Near-Zero Materials

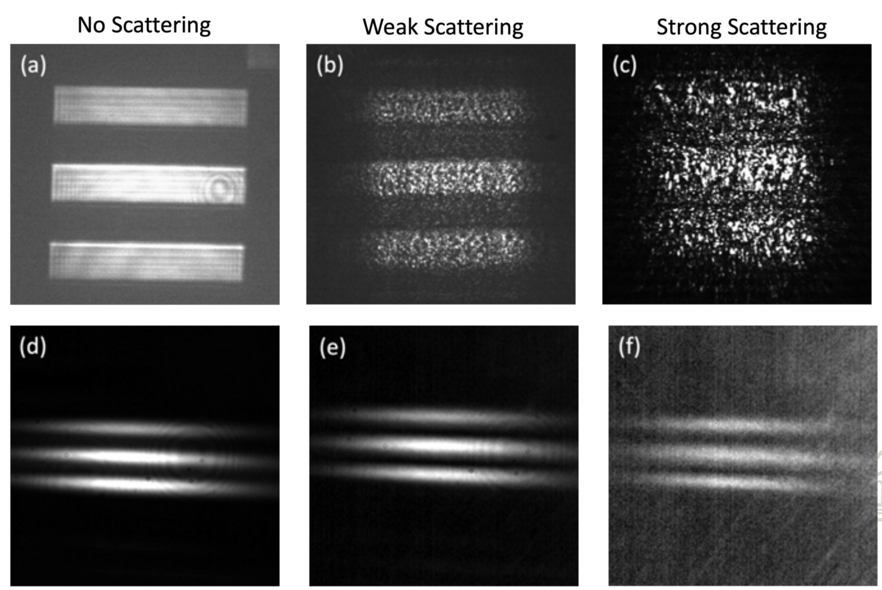


Only first-arriving photons are transmitted through the FWM gate

Requires a large chi-3 with a fast response: ITO

Direct image

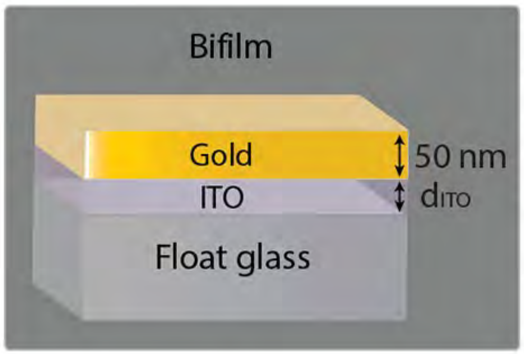
FWM image

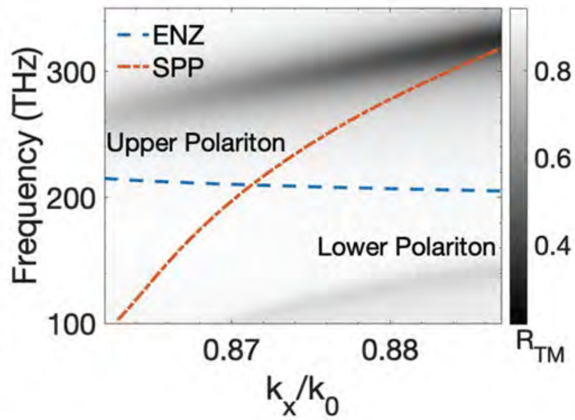


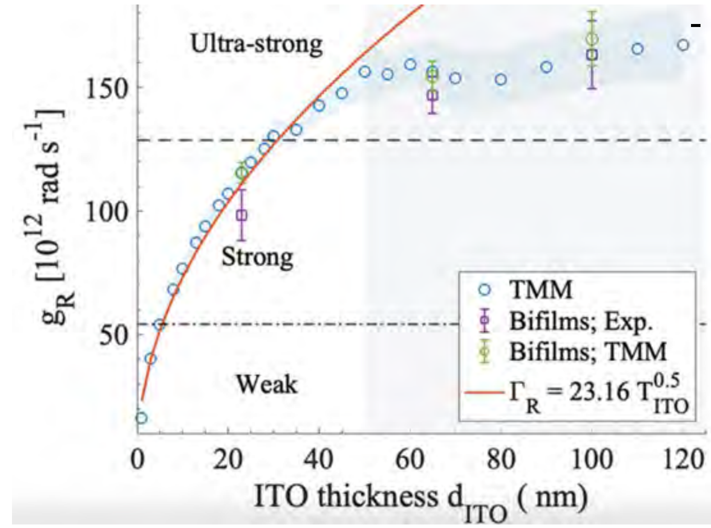
with Yang Xu, Saumya Choudhary, and Zahirul Alam

Strong and ultra-strong coupling of ENZ and surface plasmon modes

- Dispersion of SPP modes shows strong modification in the presence of an ENZ layer.
- Coupled SPP-ENZ modes can reach ultra-strong coupling.







Legend

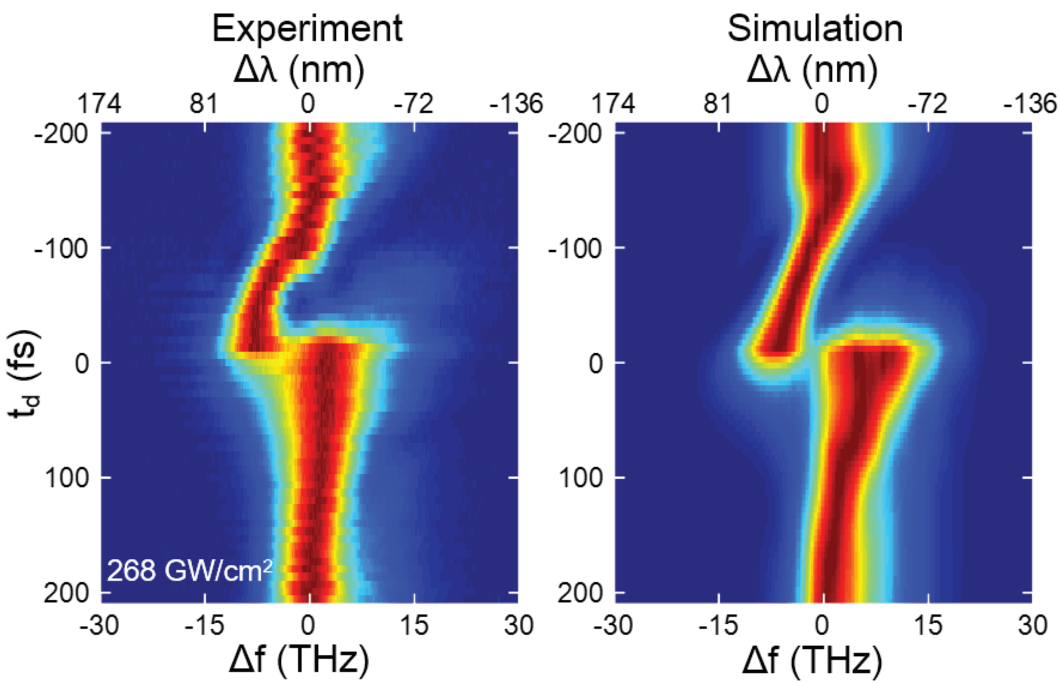
"ENZ" is the (bare) epsilon-near-zero mode
"SPP" is the (bare) surface-plasmon-polariton mode
"Upper Polariton" is the Upper branch of the coupled ENZ-SPP mode
"Lower Polariton" is the Lower branch of the coupled ENZ-SPP mode

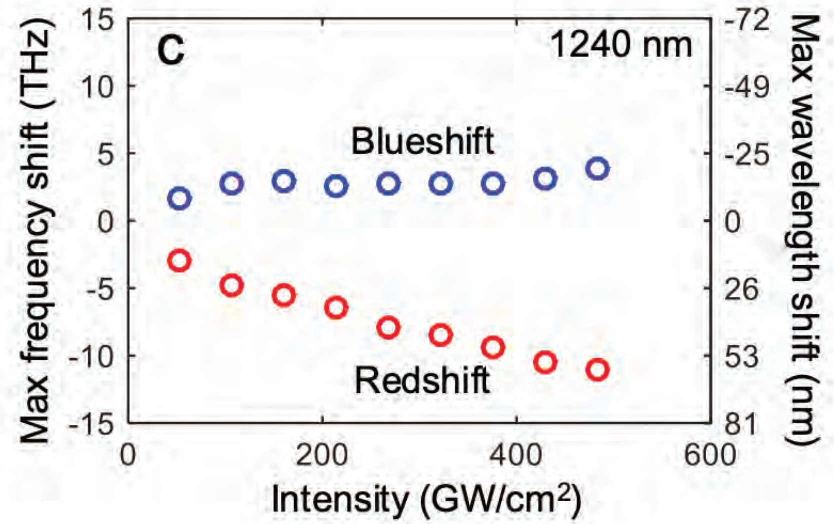
- A Gold-ITO bifilm is a platform for studying the ultralow-power nonlinear physics of surface waves.
- Observing strong nonlinear modifications of propagating surface waves is nearly impossible without the thin ENZ layer, despite the large third-order susceptibility of gold (the maximum Δn of gold is not large enough).

Choudhary et at., ACS Photonics 2023 10 (1), 162-169

Adiabatic Wavelength Conversion by Time Refraction

Experimental results at 1240 nm

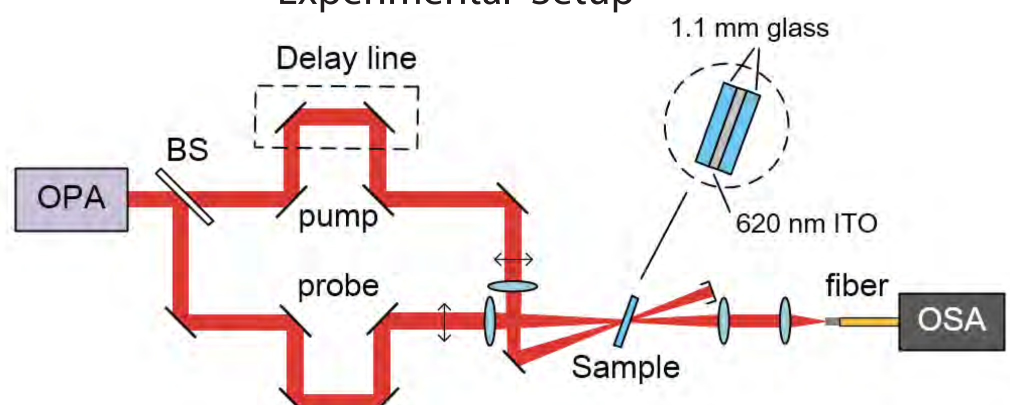




The wavelength shift can be controlled by the pump intensity and the sign of the time delay.

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

Experimental Setup

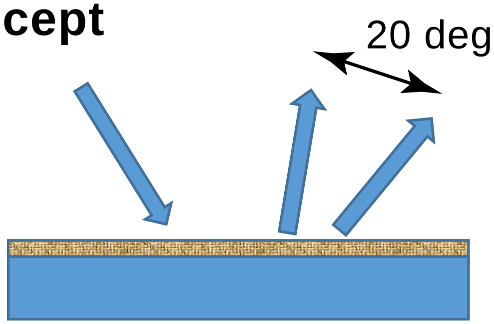


- The observed effect is 100 times larger with almost 100 times smaller propagation distance than previous reports of AWC.
- Application: wavelength-division multiplexing for telecom

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

All-Optical, Nanoscale, Sub-Picosecond Beam Steering

- **Concept**

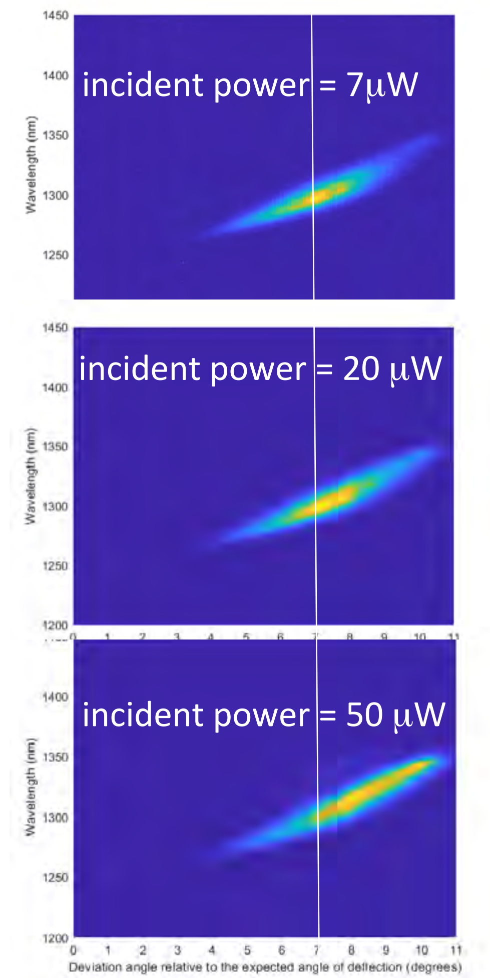


Tune output direction by
+/- 20 degrees under
optical or electrical control

Beam steerer mcan be made
of one or many cells

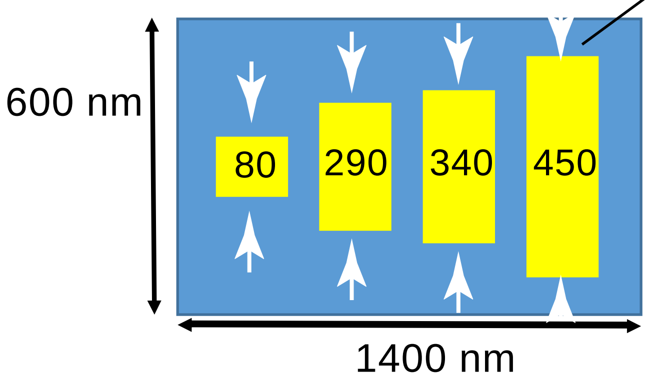
Sub-picosecond response time

- **First results**



- **Design**

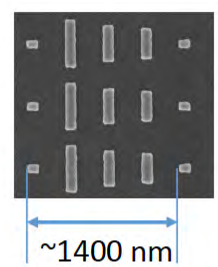
Top View of Cell

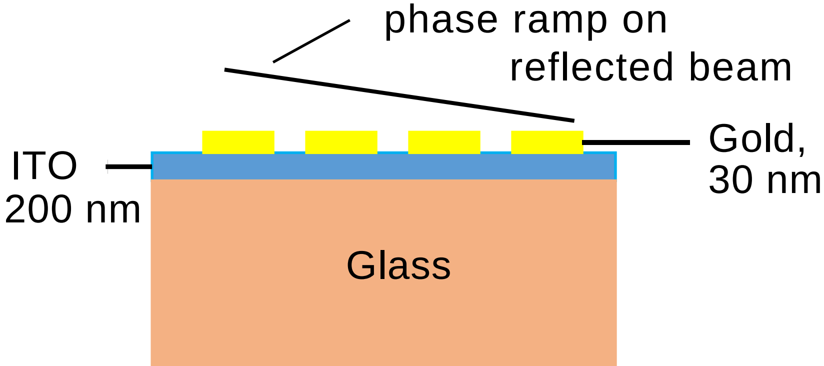


nanoantennas

Nonlinear response
depends on
antenna length

- SEM





Side View of Cell

One application: Mode-division multiplexing for telecommunications

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

Influence of pulse duration and nonlinear Fresnel reflection on the NL Response of ITO

- For materials with large Δn_{NL}, interfacial nonlinearities become comparable to bulk effects
- A modified Z-scan theory accounts for the new contribution due to intensity-dependent Fresnel reflection to accurately measure n_2 and β
- Also observe larger nonlinearity for longer pulses due to the integrating nature of ITO's nonlinearity

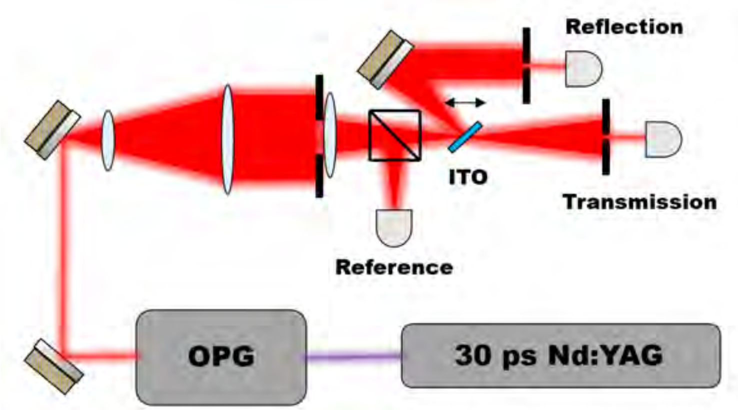


Fig. 2: Resolving competing nonlinear effects in ITO by acquiring both transmission and reflection Z-scan traces

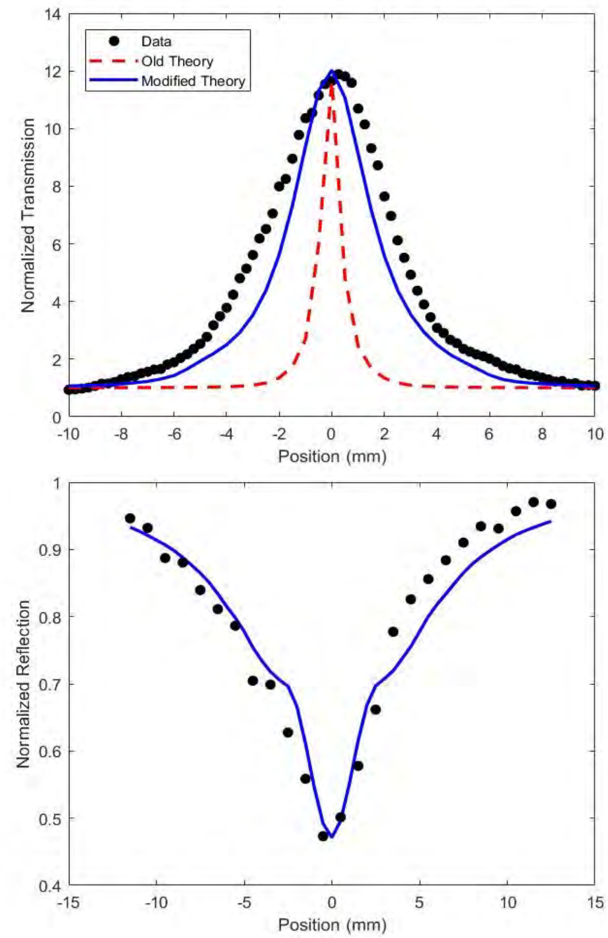


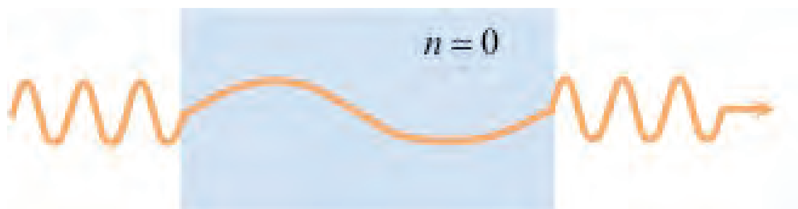
Fig. 1: Open aperture traces for ITO at 1240 nm. Observe ~10x increase in n_2 and β over fs values

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).

Physics of Epsilon-Near-Zero (ENZ) Materials

- Radiative processes are modified in ENZ materials *

Einstein *A* coefficient (spontaneous emission lifetime = 1/*A*)

$A = n A_{vac}$

We can control (inhibit!) spontaneous emission!

Einstein *B* coefficient

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

$B = B_{vac} / (n n_g)$

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.

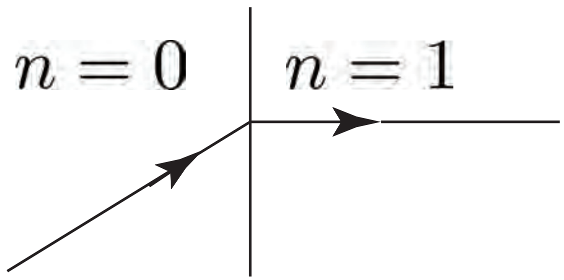
* Lobet, Liberal, Knall, Alam, Reshef, Boyd, Engheta, and Mazur, ACS Photonics 7, 1965-1970 (2020).

Optics of Zero-Index Materials

- Snell's law leads to intriguing predictions

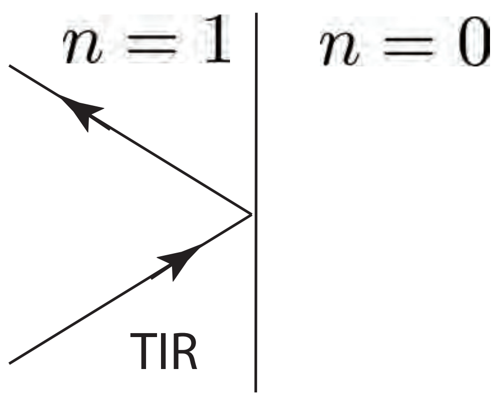
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

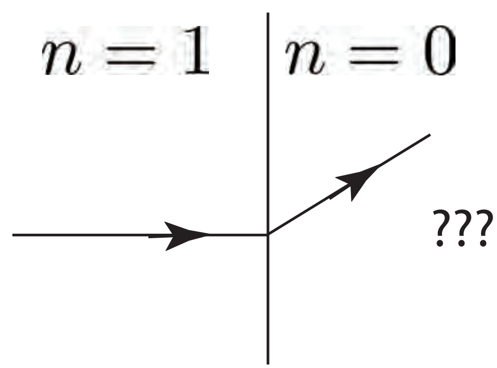
- Light always leaves perpendicular to surface of ENZ material!



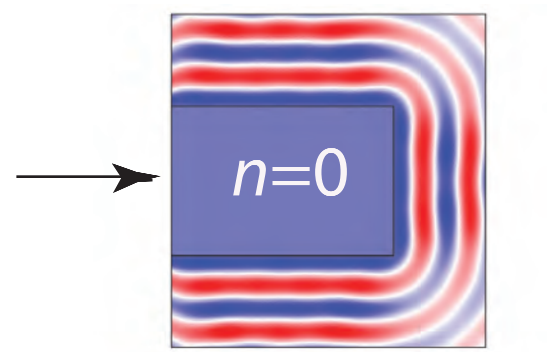
Y. Li, et al., Nat. Photonics 9, 738, 2015; D. I. Vulis, et al., Opt. Express 25, 12381, 2017.

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





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



Light enters at normal incidence but leaves in all directions.

(wave-optics simulation - O. Reshef)

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.

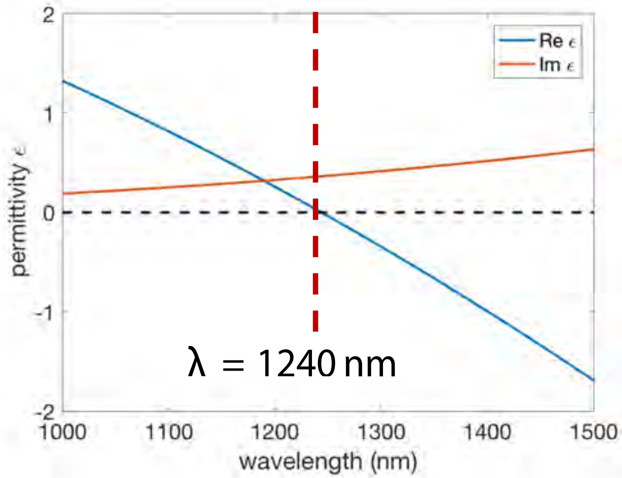
$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

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

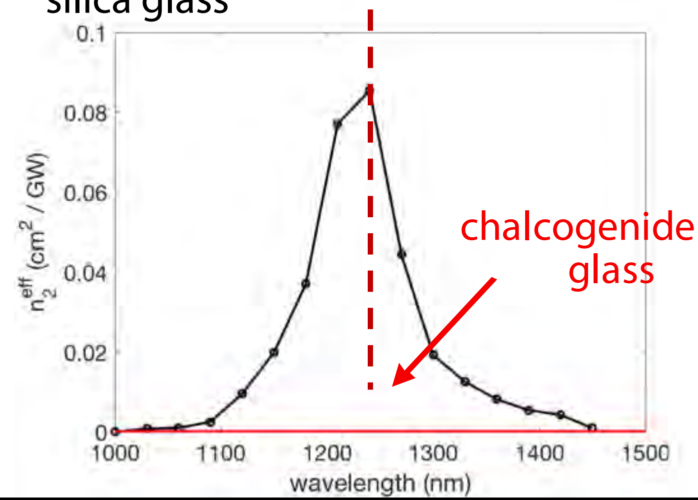
1. Alam, De Leon and Boyd, Science 352, 795–797 (2016)
2. Caspani, Shalaev, Boltasseva, Faccio et al., Phys. Rev. Lett. 116, 233901 (2016).

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

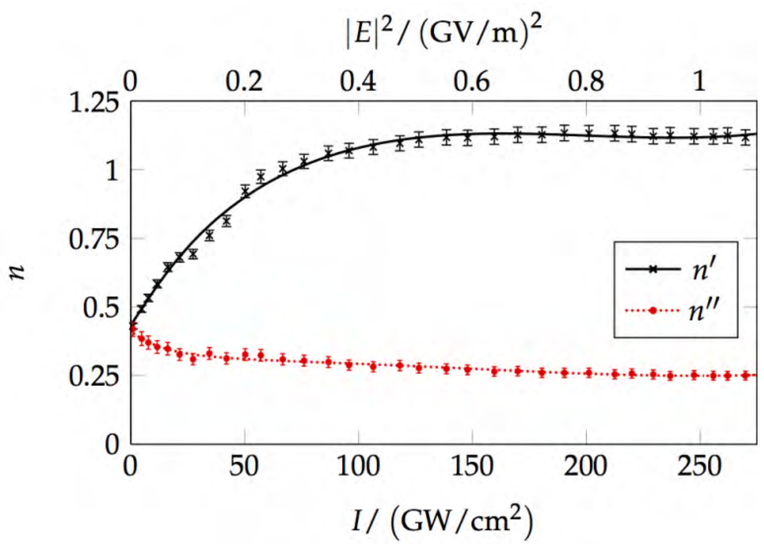
- ellipsometry



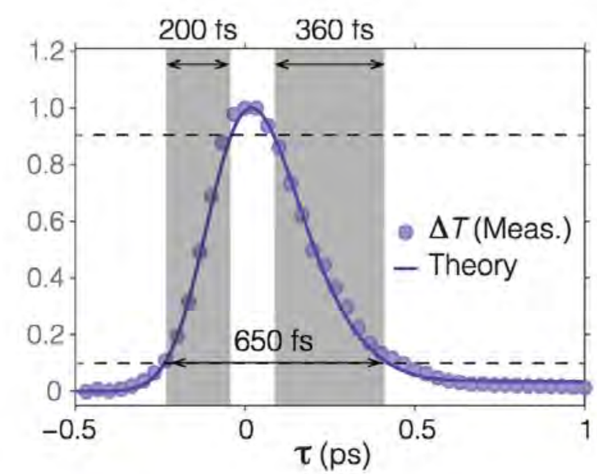
- n_2 can be 3.4 x10^5 times larger than that of silica glass



- overall change in refractive index of 0.8



- sub picosecond reponse time



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

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 $\epsilon = \epsilon_b + \Delta\epsilon$

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

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.

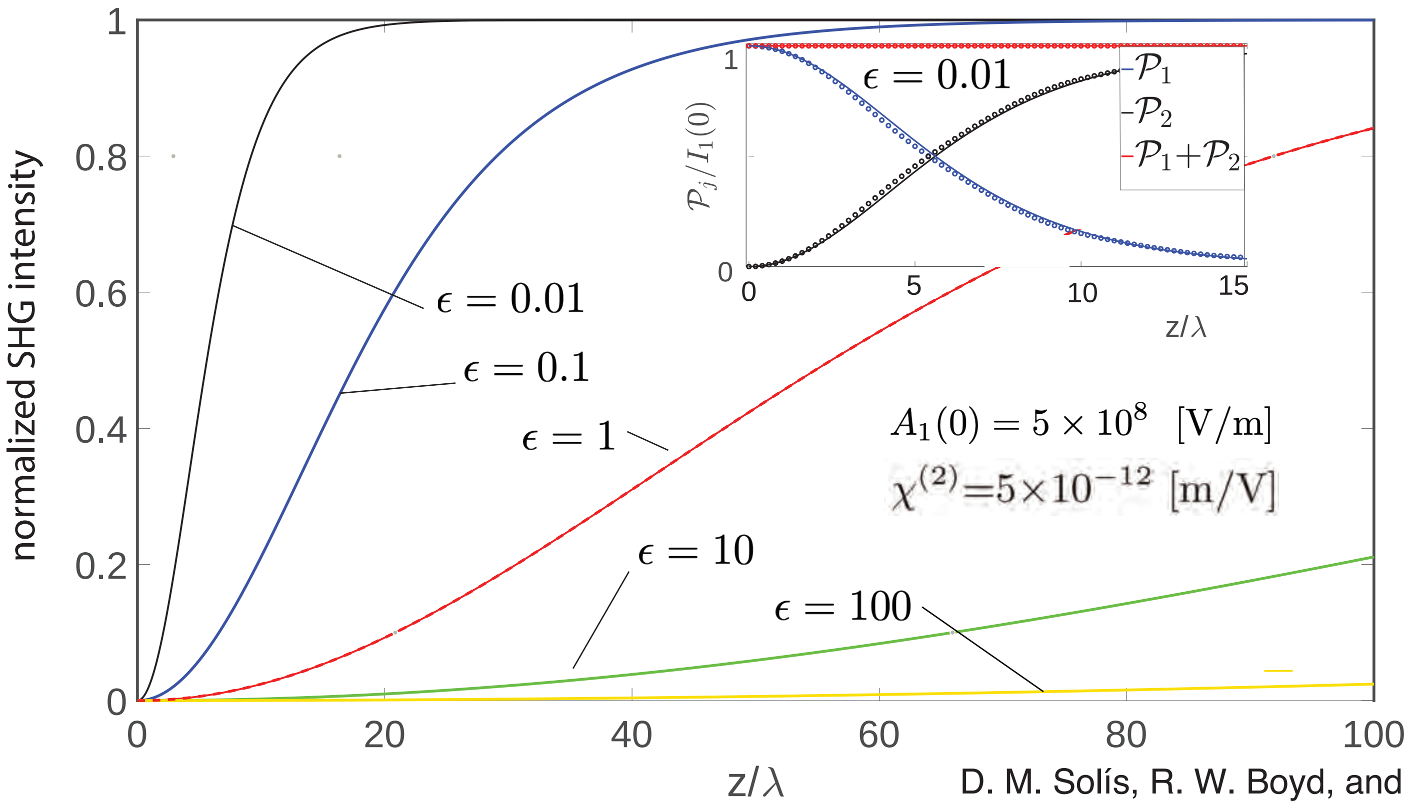
Dependence of Second-Harmonic Generation on the Linear Dielectric Permittivity

- We solve the standard equations for second-harmonic generation

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

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

- 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).

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

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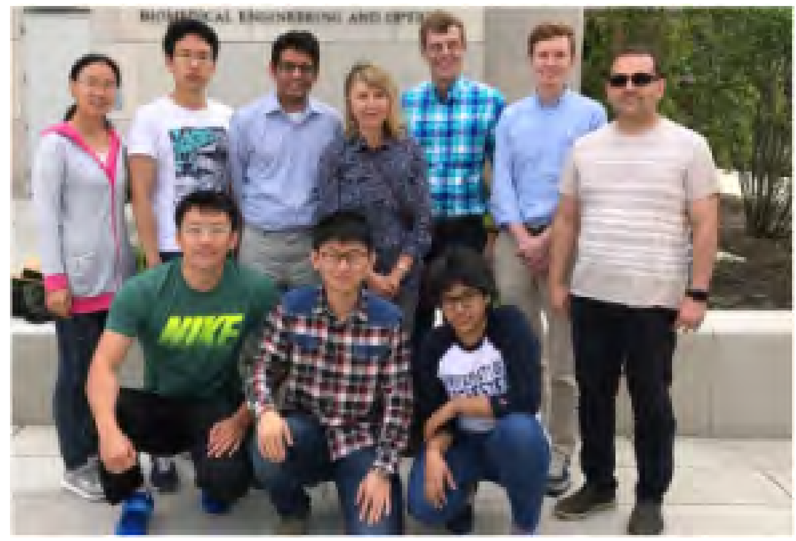

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