Nonlinear Optics of Time-Varying Media (especially epsilon-near-zero media)

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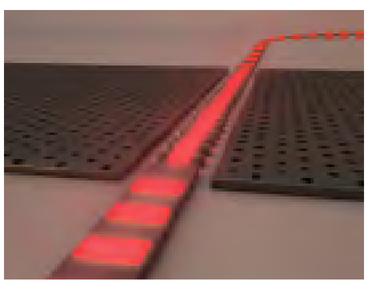
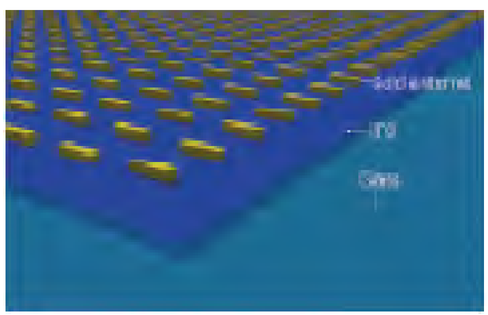
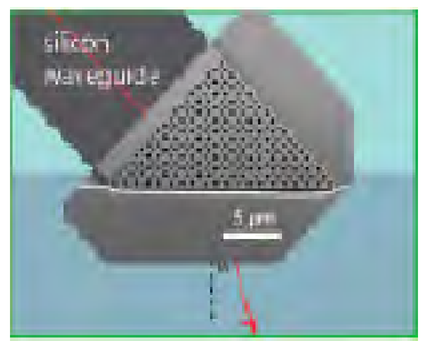
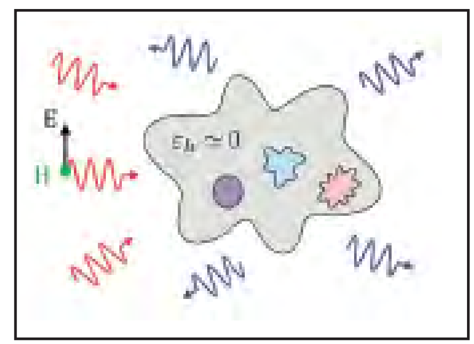








The visuals of this talk are posted at boydnlo.ca/presentations

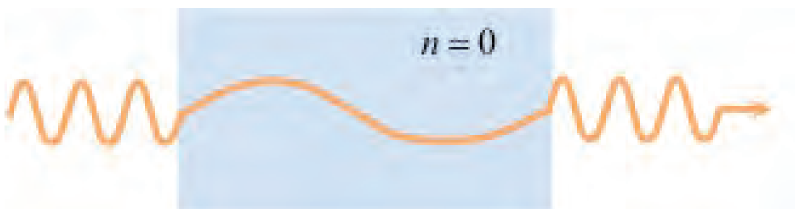
Presented at the virtual Workshop on Time-Varying Media, Imperial College, London, May 26, 2021.

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*)

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

We can control (inhibit!) spontaneous emission!

Einstein *B* coefficient

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

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

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).

Nonlinear Optical Properties of Epsilon-Near-Zero 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

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

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

How to Choose an Epsilon-Near-Zero Material

- 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.

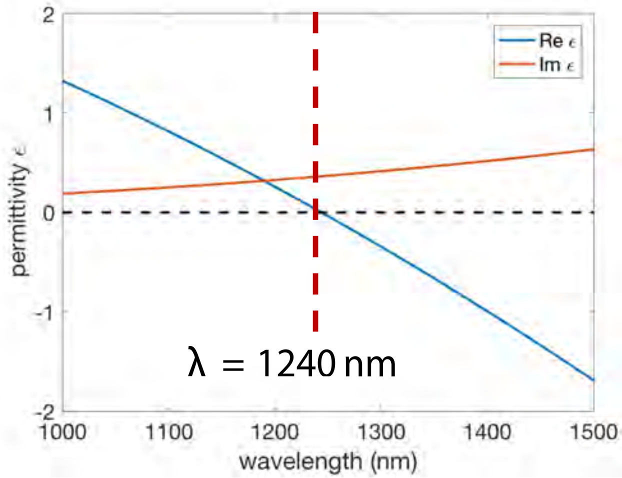
$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

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

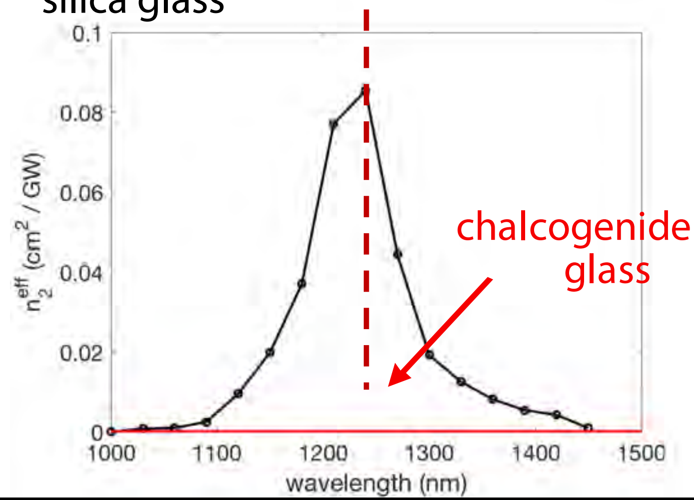
1. Alam, De Leon and Boyd, Science 352, 795–797 (2016)
2. Caspani, Shalaev, Boltasseva, Faccio et al., Phys. Rev. Lett. 116, 233901 (2016).

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

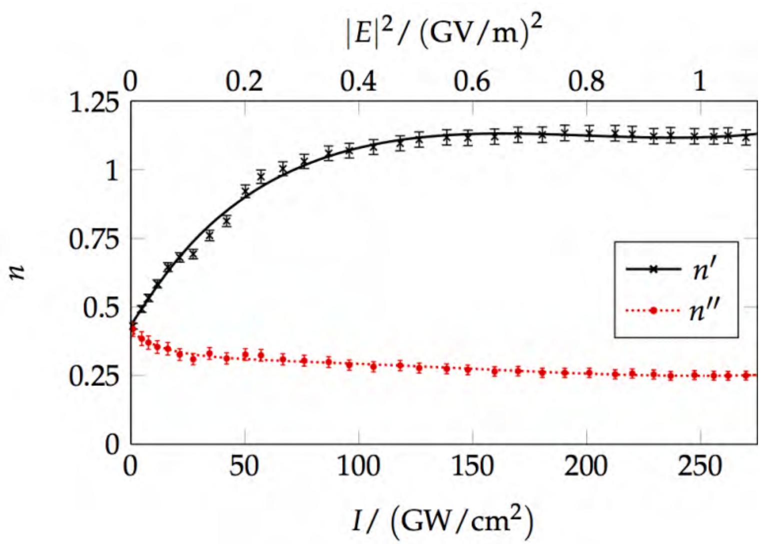
- ellipsometry



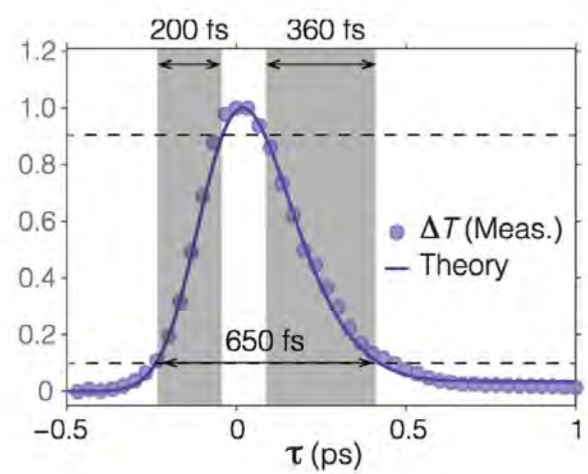
- n2 can be 3.4 x10^5 times larger than that of silica glass



- overall change in refractive index of 0.8



- sub picosecond reponse time

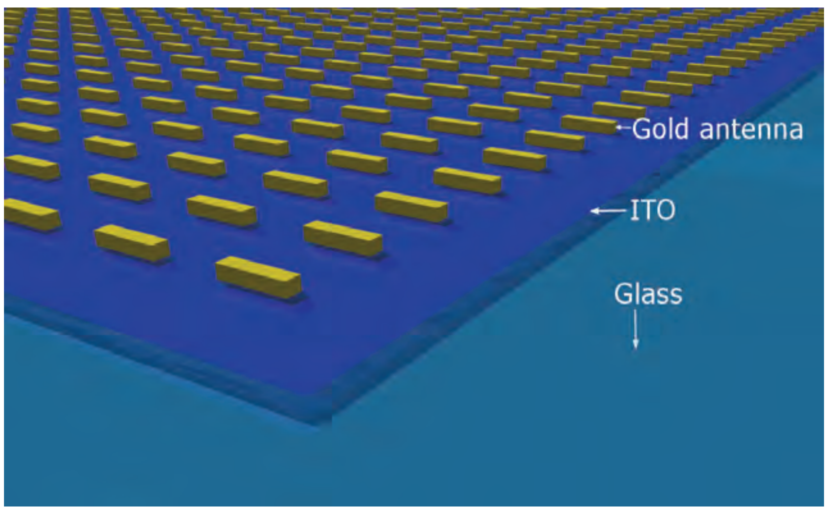


M. Z. Alam et al., Science 352, 795-797 (2016)

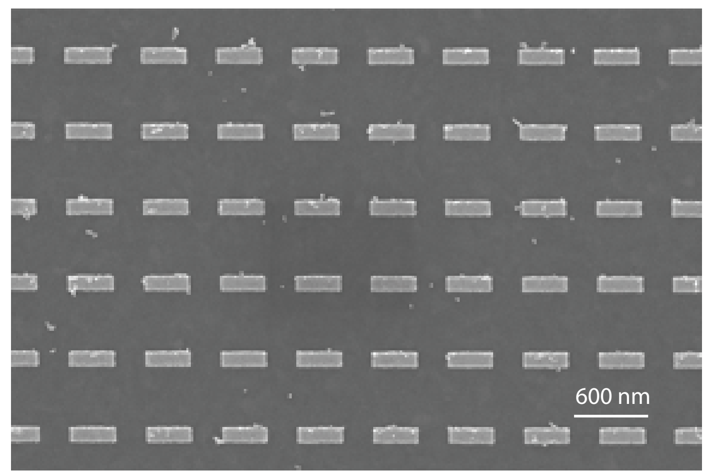
An ENZ Metasurface

- 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:

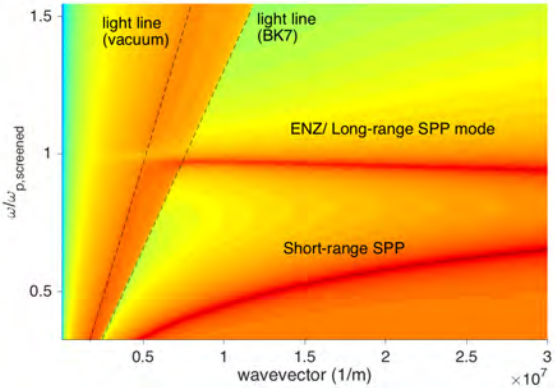


SEM:



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

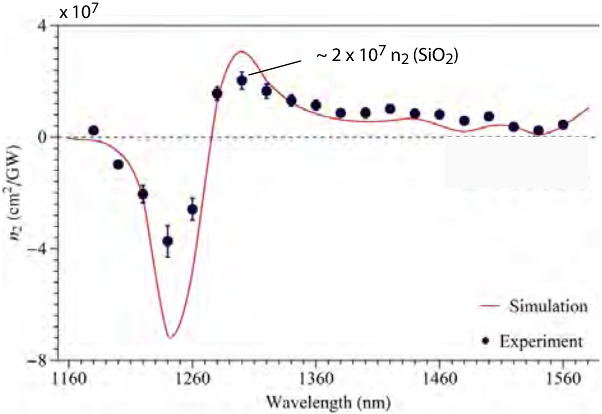
A thin ENZ medium supports a bulk plasma mode.



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



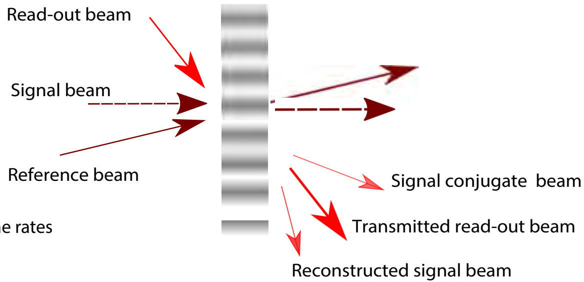
The structure exhibits and extremely large n2 value over a broad spectral range. The on-resonance n2 value is seven orders of magnitude larger than that of silica glass.

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

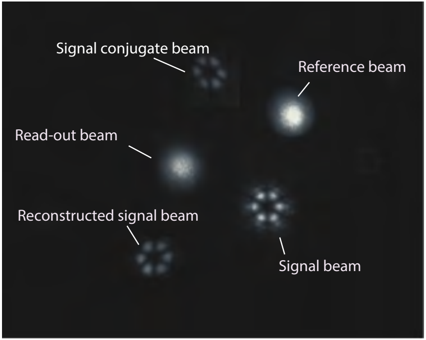
Applications of Time-Varying Epsilon-Near-Zero Materials

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)

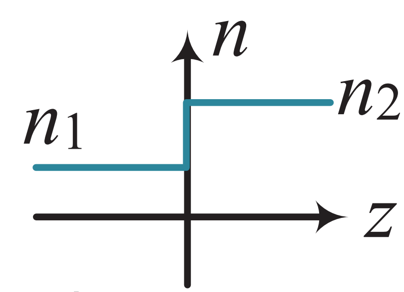


Alam, Fickler, Reshef, Giese, Upham, and Boyd

Adiabatic Wavelength Conversion through Time Refraction

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

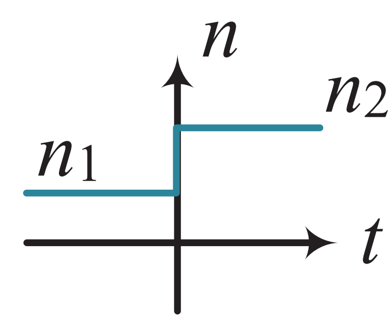
$$\frac{c}{f} = n \cdot \lambda \longrightarrow n_1\lambda_1 = n_2\lambda_2$$



index vs. position

- Time refraction (analog of Snell's law)

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



index vs. time

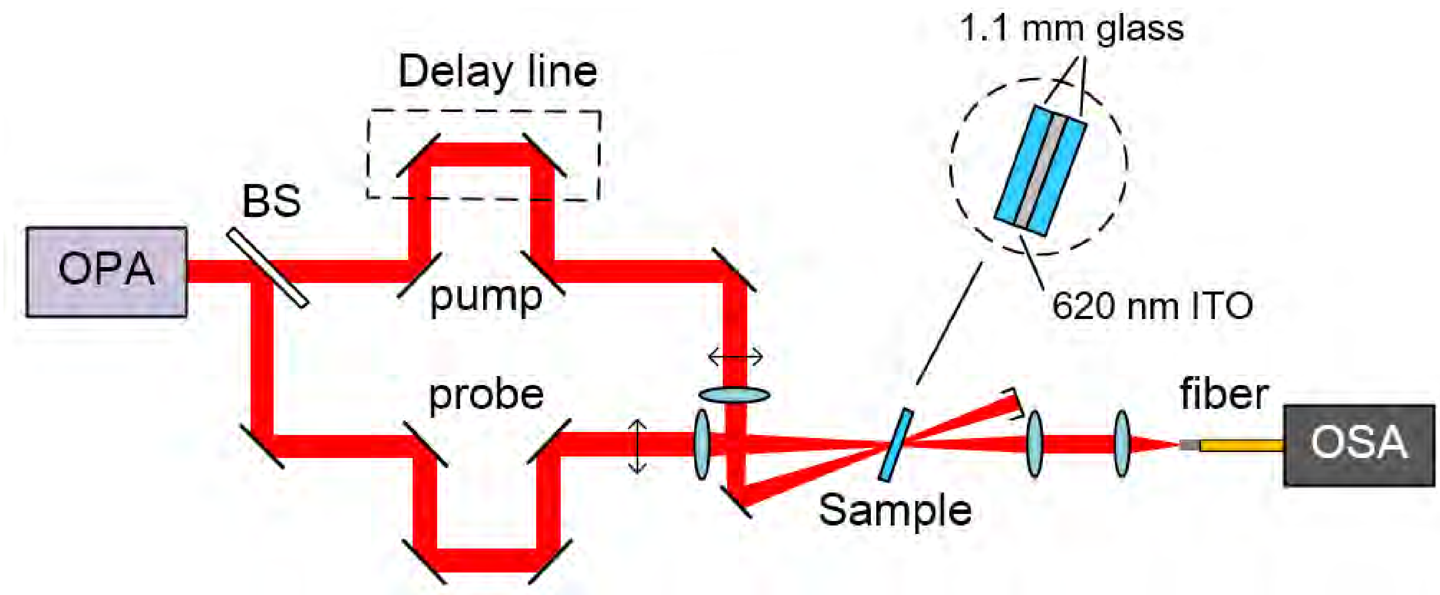
Photon frequency (energy) is changed because of the temporal change in index, but the wavelength (inverse of momentum) is conserved in the absence of any spatial asymmetry

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

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

Laboratory Study of Wavelength Conversion by Time Refraction

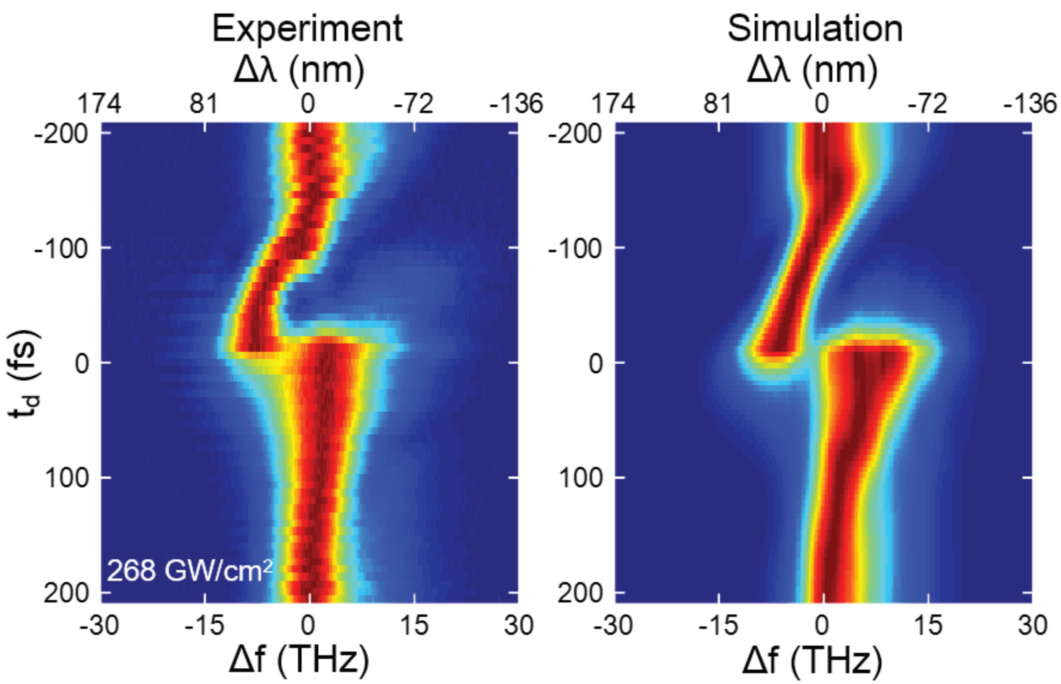
- Pump beam creates a time-varying refractive index in ITO sample
- Frequency of probe beam is thereby modified.

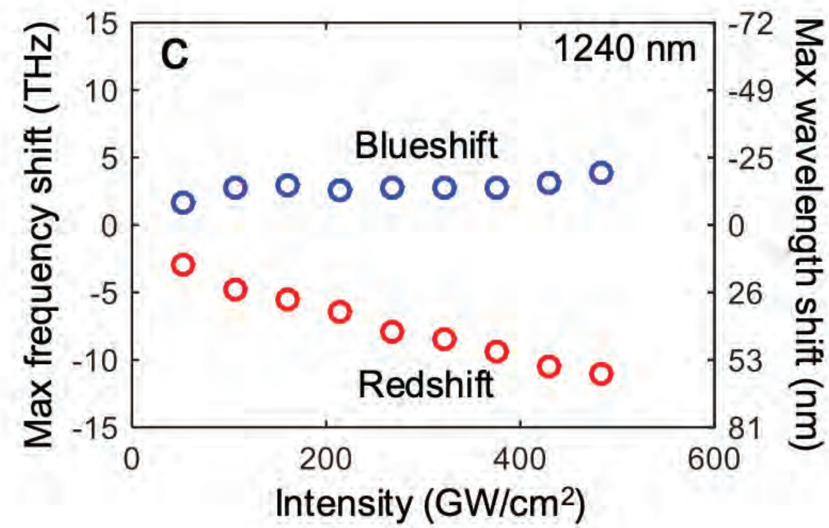


OPA = optical parametric amplifier
wavelength = 1240 nm
pulse duration = 120 fs
OSA = optical spectrum analyzer

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)

More Work on Adiabatic Wavelength Conversion

ACS Photonics
pubs.acs.org/journal/apchd5 Letter

Photon Acceleration Using a Time-Varying Epsilon-near-Zero Metasurface

Cong Liu,* M. Zahirul Alam, Kai Pang, Karapet Manukyan, Orad Reshef, Yiyu Zhou, Saumya Choudhary, Joel Patrow, Anuj Pennathurs, Hao Song, Zhe Zhao, Runzhou Zhang, Fatemeh Alishahi, Ahmad Fallahpour, Yinwen Cao, Ahmed Almaiman, Jahan M. Dawlaty, Moshe Tur, Robert W. Boyd, and Alan E. Willner

Cite This: https://dx.doi.org/10.1021/acsphotonics.0c01929 Read Online

Tunable Doppler Shift using a Time-Varying Epsilon-Near-Zero Thin Film near 1550 nm

Cong Liu[1,2,*], M. Zahirul Alam[3,4], Kai Pang[1], Karapet Manukyan[1], Joshua R. Hendrickson[5], Evan M. Smith[5,6], Yiyu Zhou[4], Orad Reshef[3], Hao Song[1], Runzhou Zhang[1], Haoqian Song[1], Fatemeh Alishahi[1], Ahmad Fallahpour[1], Ahmed Almaiman[7], Robert W. Boyd[3,4], Moshe Tur[8], and Alan E. Willner[1]

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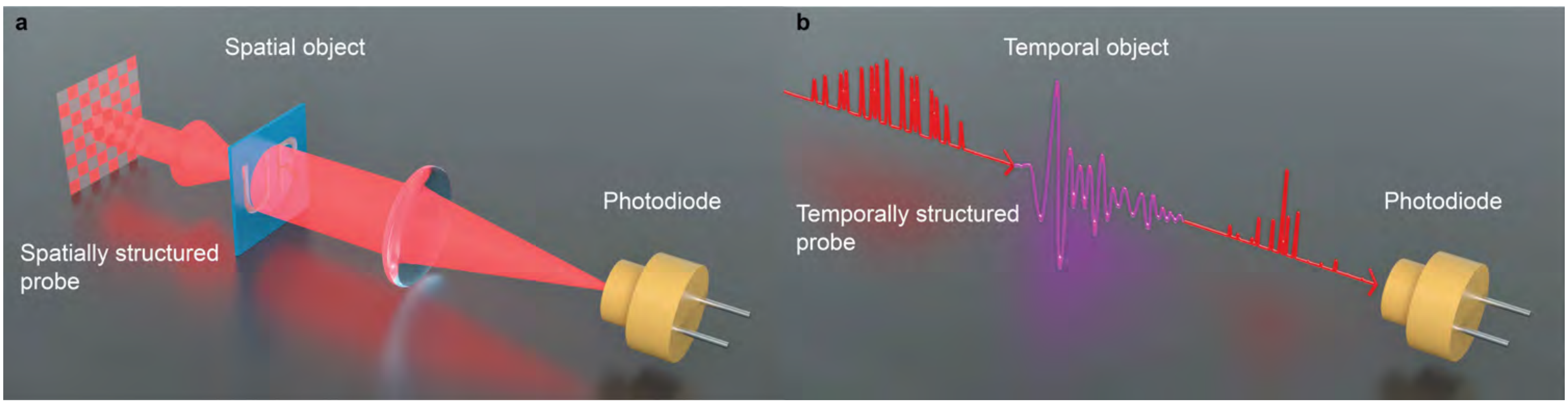
[*] Corresponding author: liucong@usc.edu

Ultrafast Pulse Measurements by Time-Domain Single-Pixel Imaging

Single-pixel imaging is now a standard method for image acquisition under certain circumstances.

Can we apply the same concepts to ultrafast pulse measurements?

Space-domain **Time-domain**

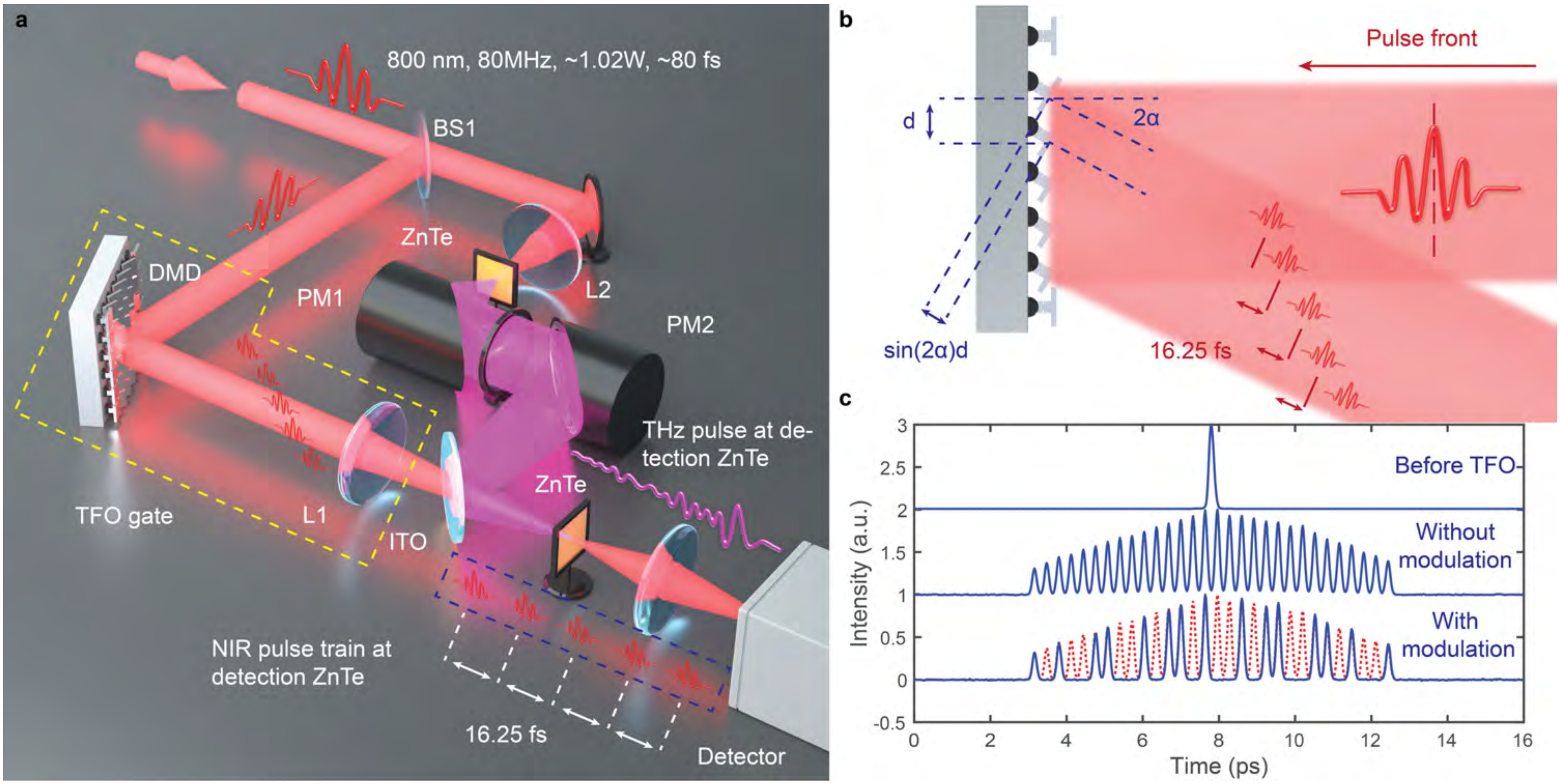


Zhao, Dai, Braverman, Zhang, and Boyd

arXiv 2009.13693

Ultrafast Pulse Measurements by Time-Domain Single-Pixel Imaging

Our laboratory implementation

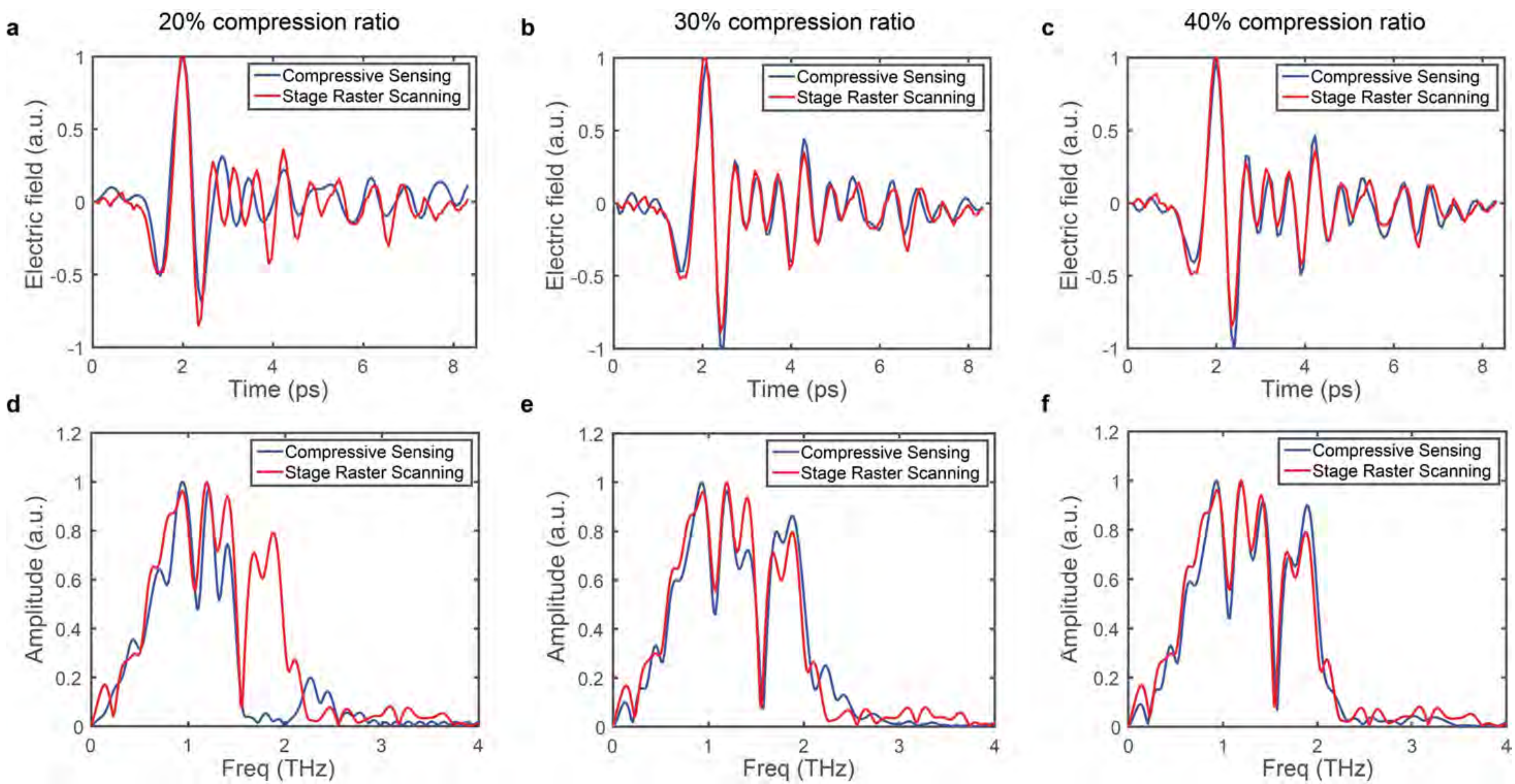


Zhao, Dai, Braverman, Zhang, and Boyd, arXiv 2009.13693

Ultrafast Pulse Measurements by Time-Domain Single-Pixel Imaging

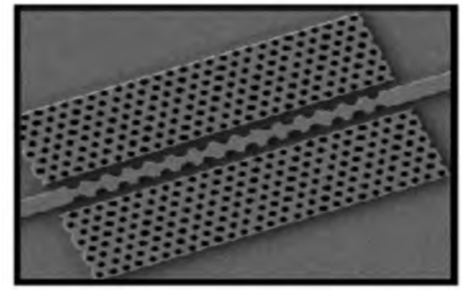
Demonstration of recovery of a THz pulse with sub-ps features

We use compressive sampling to speed up pulse measurements



Zhao, Dai, Braverman, Zhang, and Boyd, arXiv 2009.13693

Relaxed Phase-Matching Requirements in ENZ Media



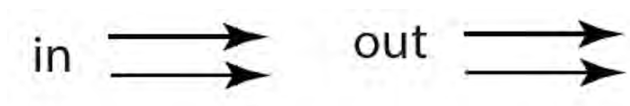
- We study four-wave mixing in a zero-index waveguide

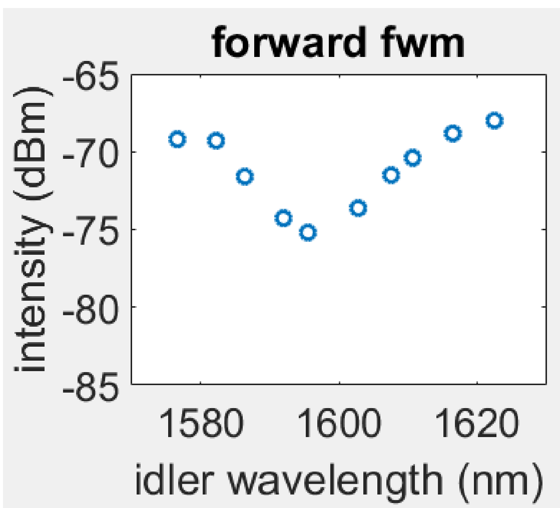
$$2\omega_p = \omega_s + \omega_i$$

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

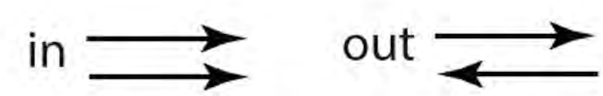
- Recall that we need $\Delta k = 0$, but when $n = 0$, $k = n\,\omega/c$ vanishes for each of the interacting waves and thus so does Δk.

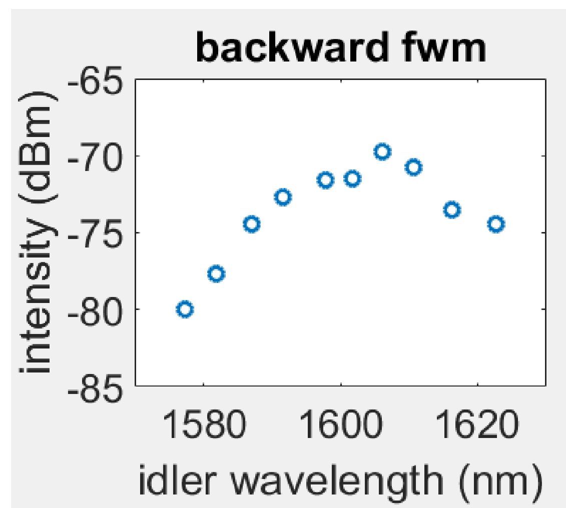
"forward" process





"backward" process



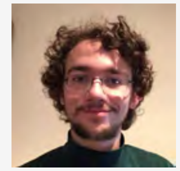


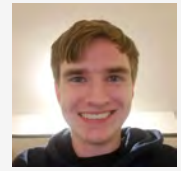
- Significance: Nonlinear optical processes that were previously believed to be too weak to be useful can be excited through use of ENZ materials.

Université d'Ottawa | University of Ottawa


Boris Braverman


Xialin Liu


Alexander Skerjanc


Nicholas Sullivan


Robert W. Boyd

Acousto-Optic Modulators for Arbitrary Spatial Mode Control

B. Braverman, X. Liu, A. Skerjanc, N. Sullivan, and R.W. Boyd

July 7, 2020

boydnlo.ca

Bourses postdoctorales Banting Postdoctoral Fellowships

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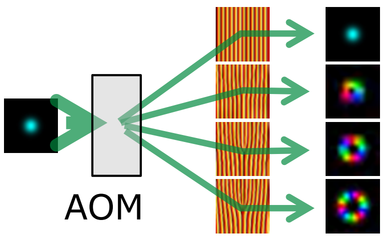
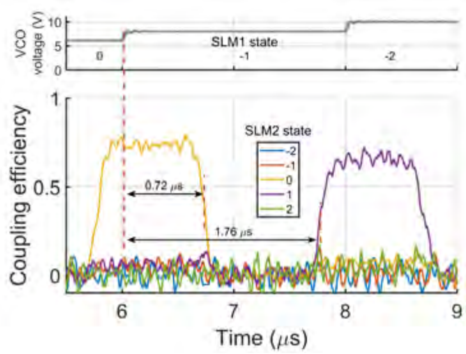
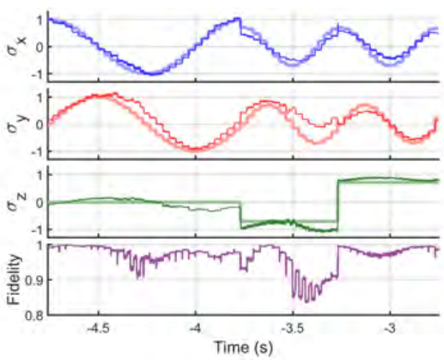
Acousto-Optic Modulators for Arbitrary Spatial Mode Control

An AOM can be used to **quickly select** one of several static holograms on an SLM

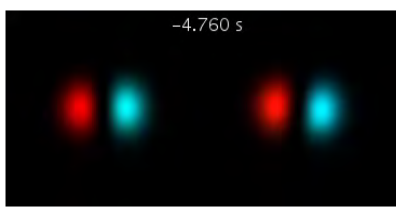




500 kHz switching among any 6 spatial modes

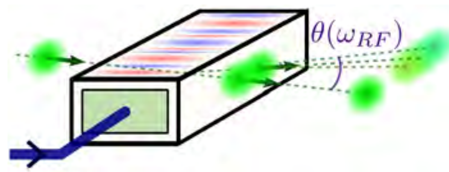


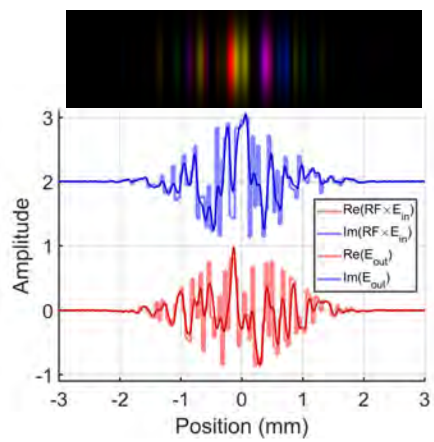
>1 kHz state tomography in 2-D Hilbert space, 97% fidelity



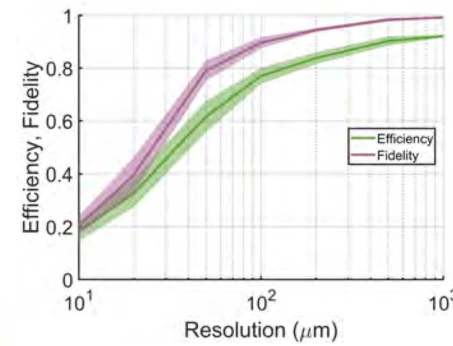
Predicted and reconstructed spatial modes

Using an appropriately modulated RF input, the **AOM itself** can generate spatial modes for pulsed light

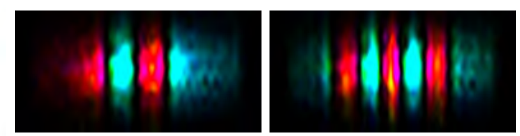




Highly complex modes can be produced



High fidelity, efficiency at 0.1 mm resolution with RF optimization



Experimentally generated HG3 and HG6 modes

ARTICLE

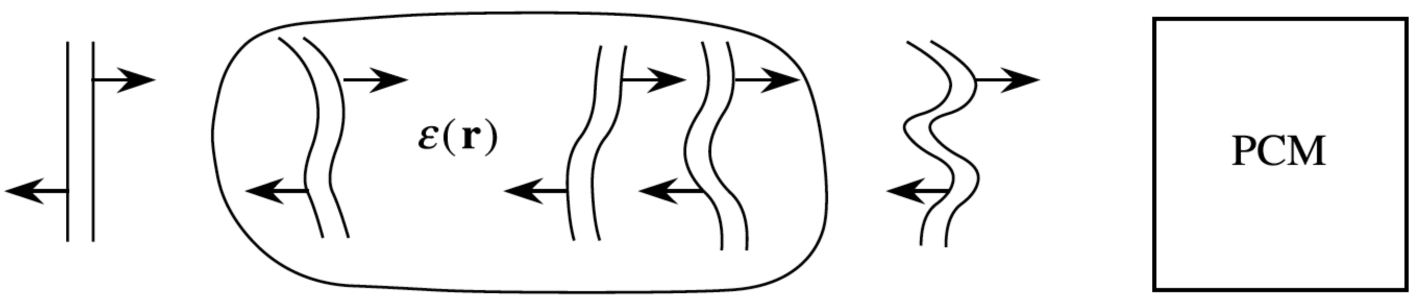
https://doi.org/10.1038/s41467-021-22071-w OPEN

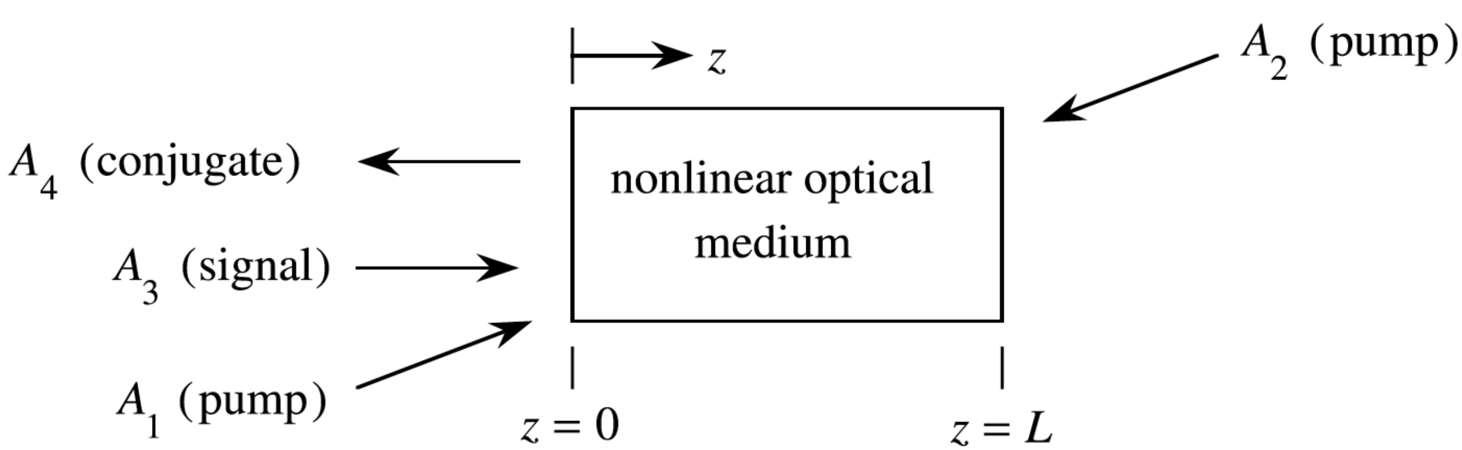
High-fidelity spatial mode transmission through a 1-km-long multimode fiber via vectorial time reversal

Yiyu Zhou[1]✉, Boris Braverman[2], Alexander Fyffe[3], Runzhou Zhang[4], Jiapeng Zhao[1], Alan E. Willner[4], Zhimin Shi[3] & Robert W. Boyd[1,2]

Phase conjugation (generation of time reversed wavefronts) has historically been implemented through use of a nonlinear optical interaction, and was inefficient.

We instead are implementing a digital form of phase conjugation.





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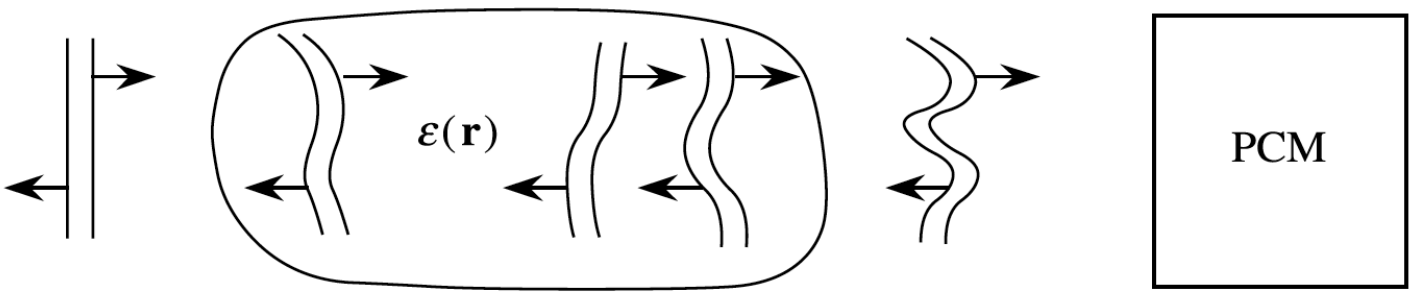
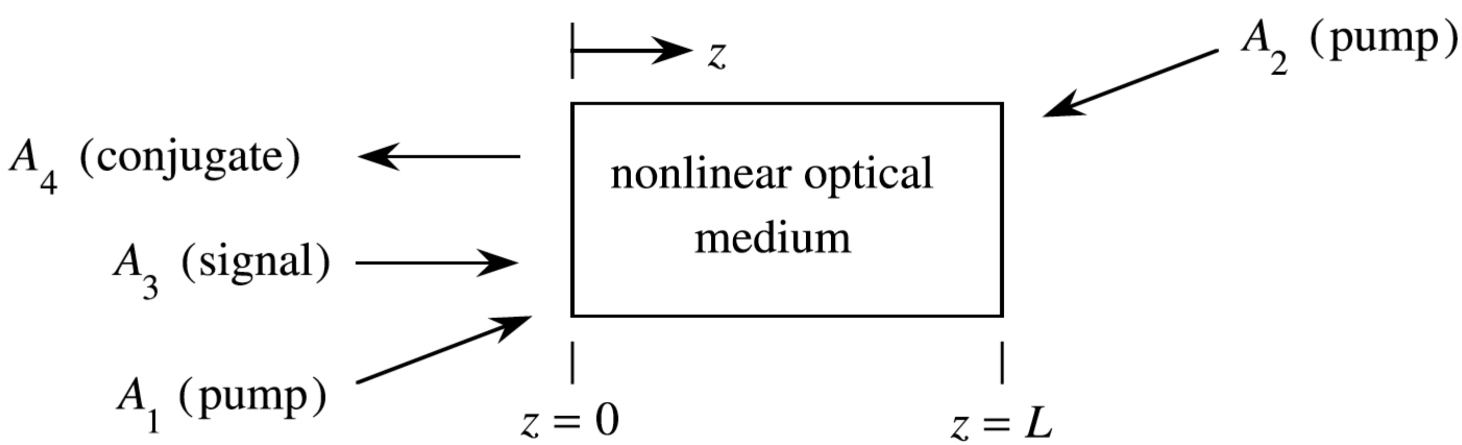
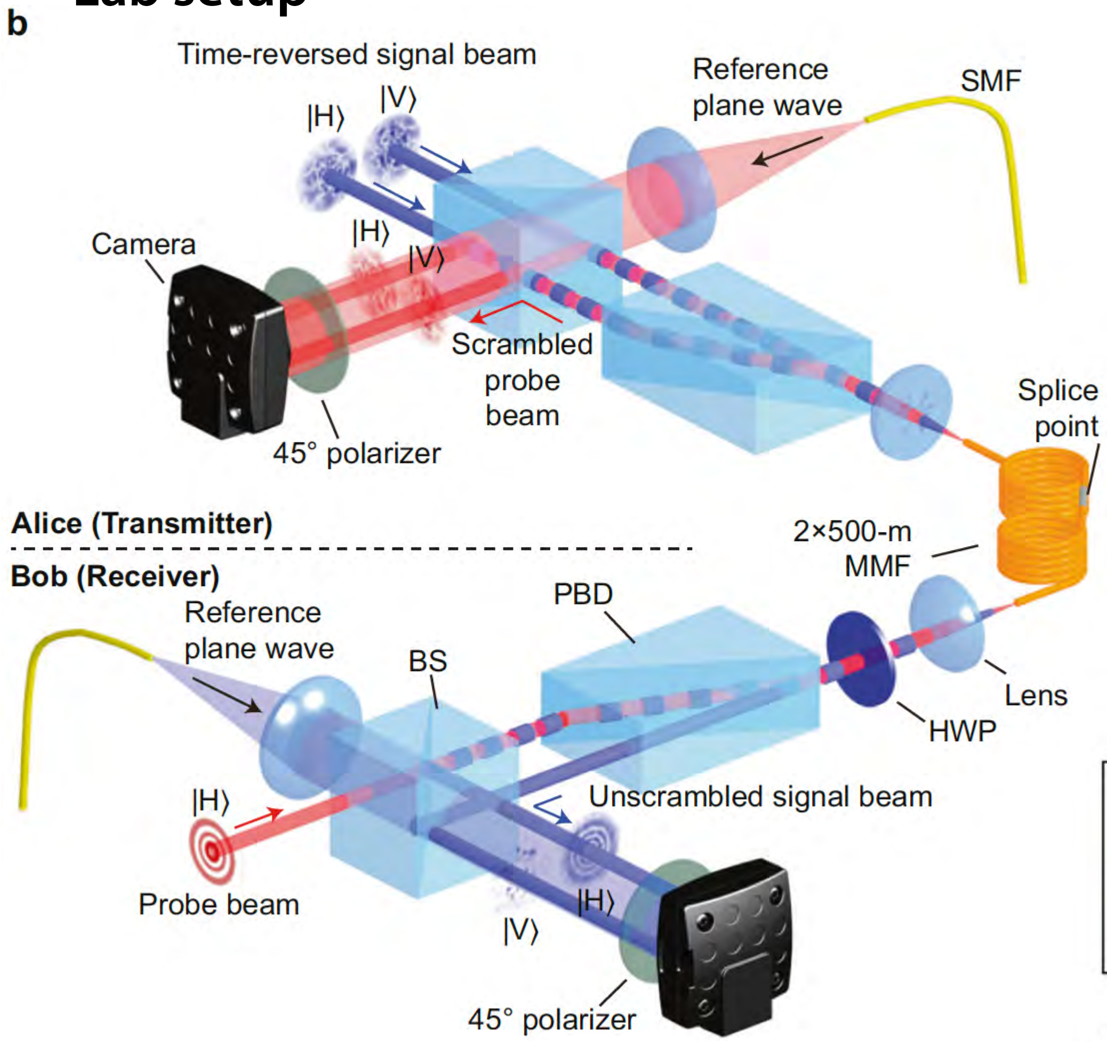




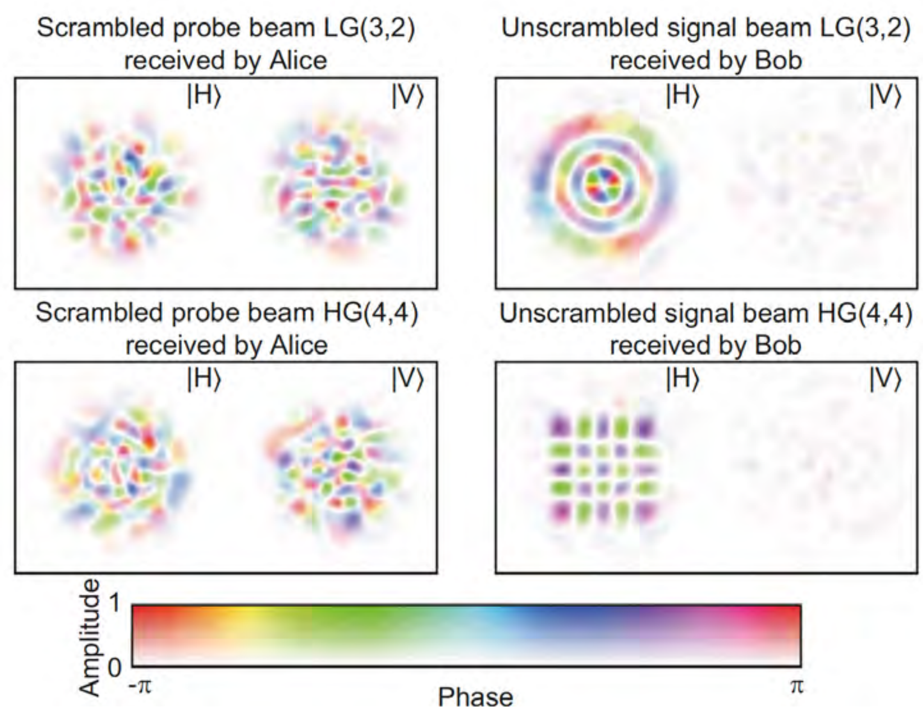
Implementation of Digital Phase Conjugation

Lab setup



Bob sends a probe beam to Alice which becomes distorted. Alice measures the wavefront distortion and send to Bob a wave that is the phase conjugate of the wave she received.

Results:



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

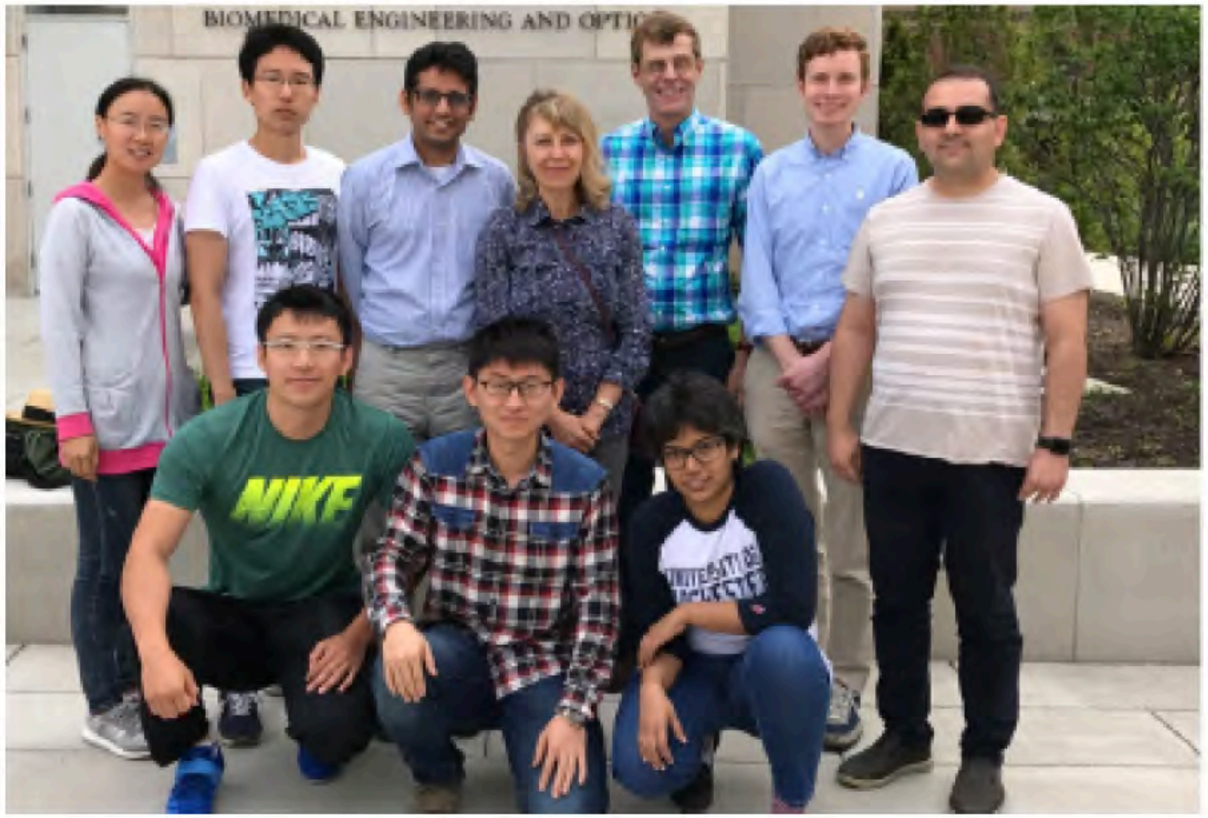
- The huge, ultrafast NLO response of ENZ materials lend themselves to many important applications

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

Special Thanks To My Students and Postdocs!

Ottawa Group

Rochester Group



Summary: Physics and Applications of ENZ Materials

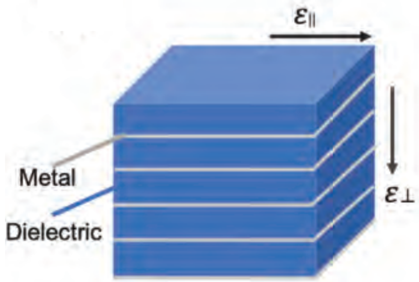
- Extremely interesting physical processes occur in ENZ materials
- ENZ materials, metamaterials, and metastructures display extremely large NLO response
- The huge, ultrafast NLO response of ENZ materials lend themselves to many important applications that require ultrafast time-varying media.

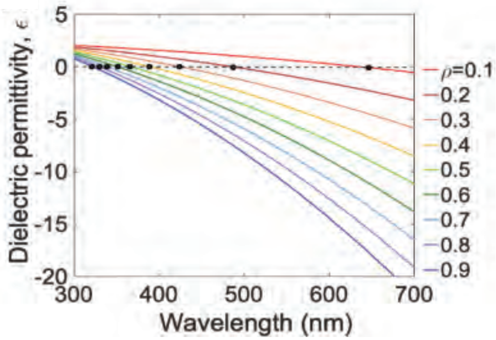
The visuals of this talk are posted at boydnlo.ca/presentations

Nonlinear Optical Properties of a Layered Metamaterial in its ENZ Region

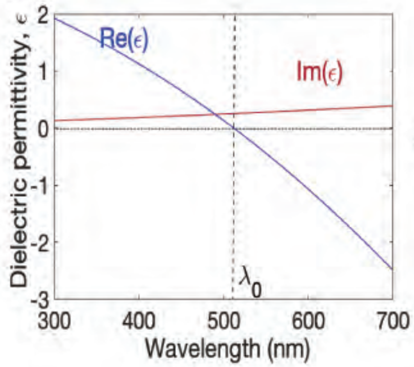
Do layered metamaterials also show enhanced NLO response at ENZ wavelength?

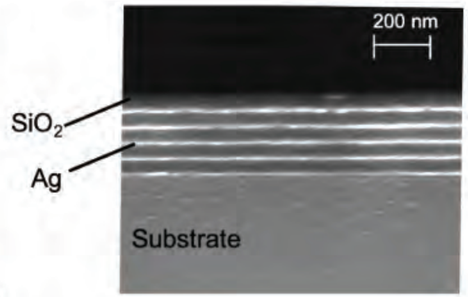
- By controlling the metallic fill fraction ρ, we can set the ENZ wavelength to be anywhere from 300 to 700 nm. We use $\rho = 0.2$, which corresponds to 500 nm. We deposit five layer pairs



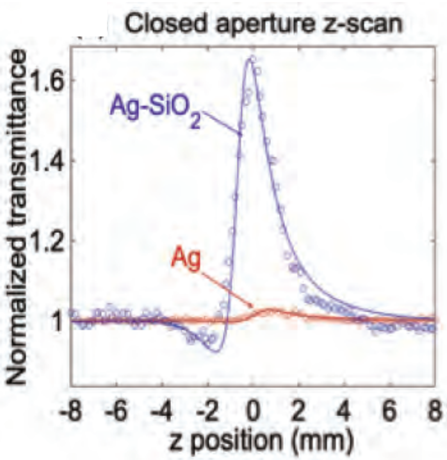


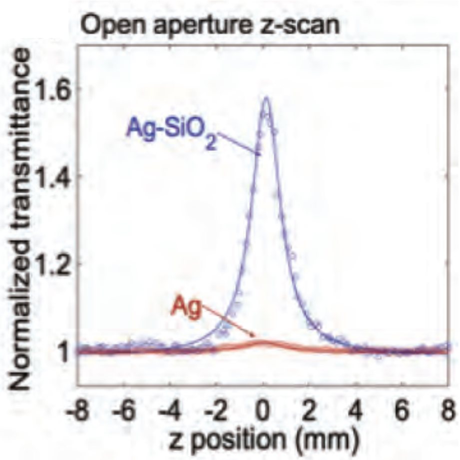
- Note that the real part of epsilon vanishes at 508 nm, close to the design wavelength. The SEM shows our structure. Ag thickness = 16 nm; silica thicness = 65 nm



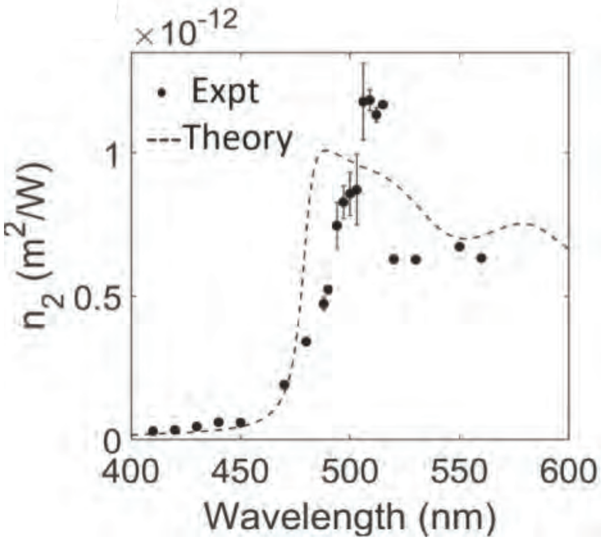


- We perform Z-scan measurements on the sample. Note the enhanced response of the composite as compared to a single layer of silver.





- 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

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

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

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.

Dependence of Second-Harmonic Generation on the Linear Dielectric Permittivity

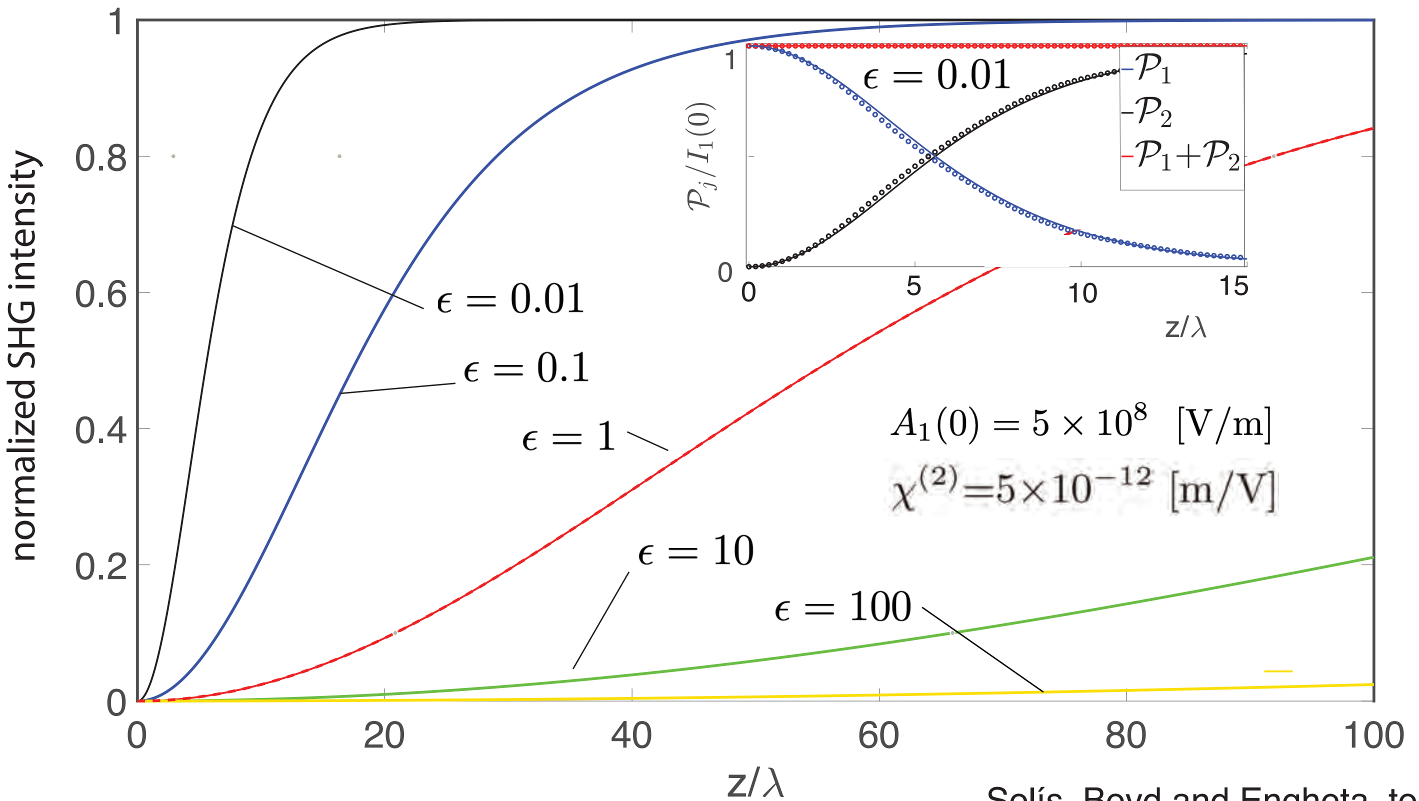
- We solve the standard equations for second-harmonic generation

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

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

- 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