



# Physics and Applications of Epsilon-Near-Zero Materials

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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

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- 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

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- ENZ materials possess exotic electromagnetic properties  
Silveirinha, Engheta, Phys. Rev. Lett. 97, 157403, 2006.
- If the dielectric permittivity  $\epsilon$  is nearly zero, then refractive index  $n = \sqrt{\epsilon}$  is nearly zero.  
Thus  $v_{\text{phase}} = c/n$  is nearly infinite  
 $\lambda = \lambda_{\text{vac}} / n$  is nearly infinite  
Light oscillates in time but not in space; everything is in phase  
Light “oscillates” but does not “propagate.”
- 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  
 $B = B_{\text{vac}} / n^2$   
Optical gain is very large!  
Einstein, Physikalische Zeitschrift 18, 121 (1917).  
Milonni, Journal of Modern Optics 42, 1991 (1995).

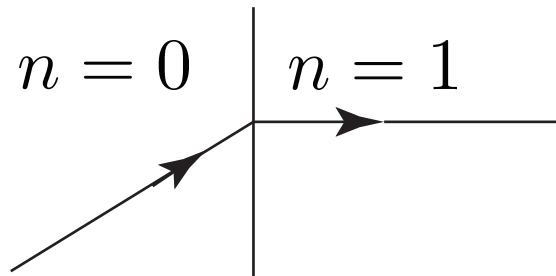


# Physics of Epsilon-Near-Zero (ENZ) Materials -- More

- Snell's law leads to intriguing predictions

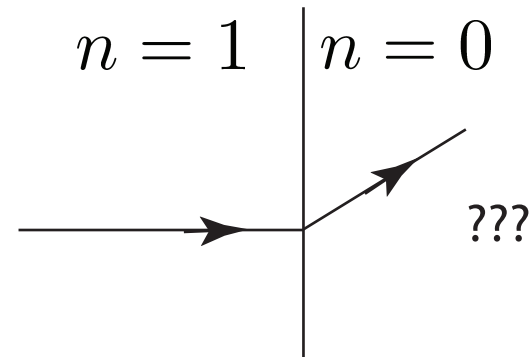
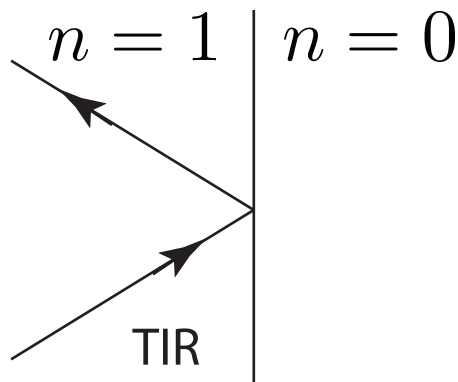
$$n_1 \sin \theta_1 = n_2 \sin \theta_2$$

- 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.

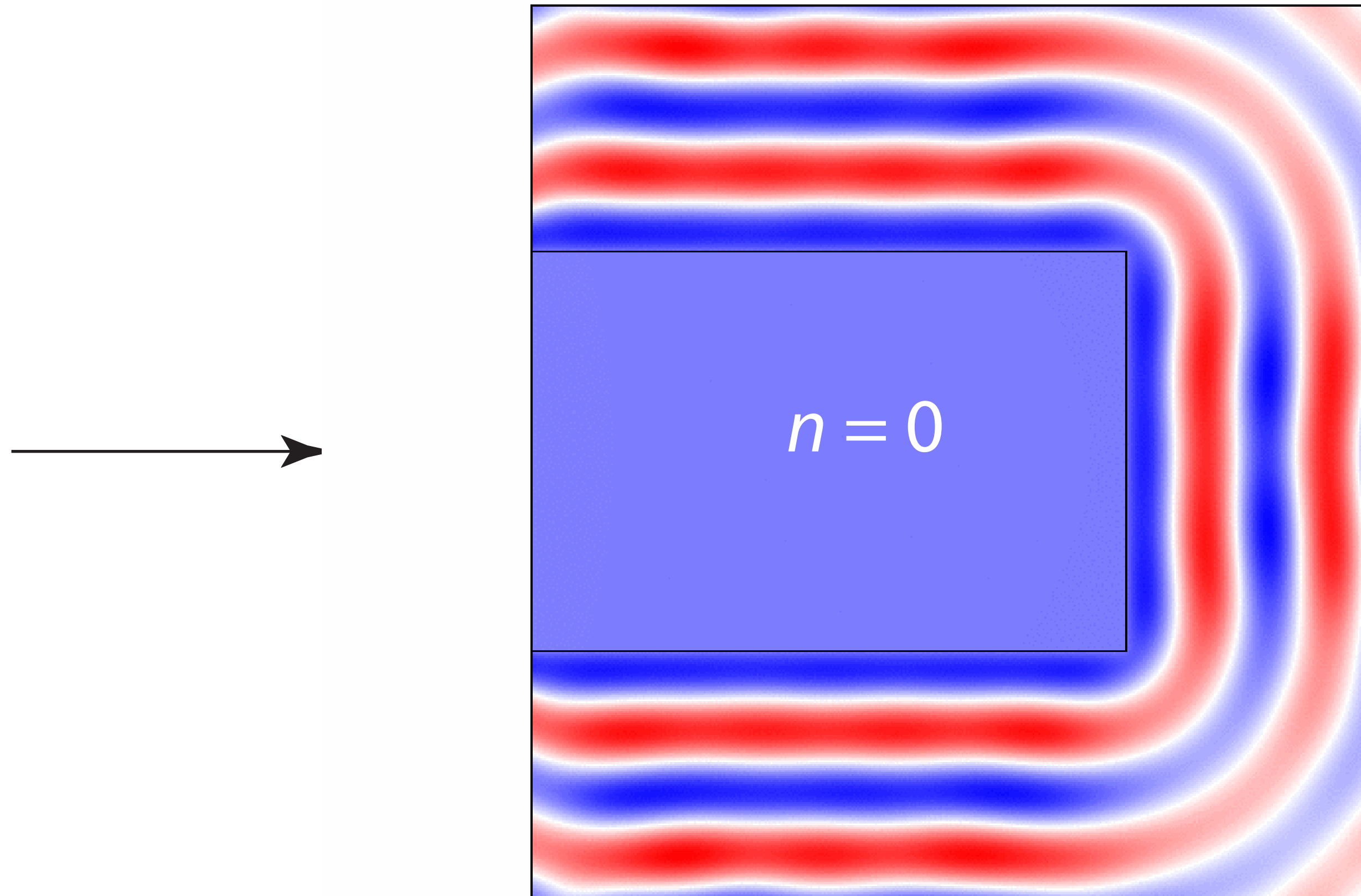
- Thus light can enter an ENZ material only at normal incidence!



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

# Maxwell Equations Prediction

- light enters slab at normal incidence



# Some Consequences of ENZ Behaviour - 1

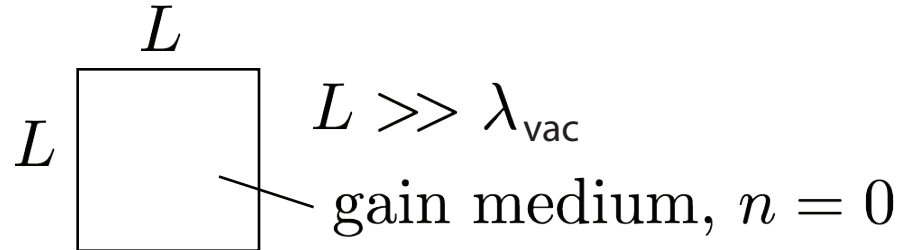
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- Funny lenses



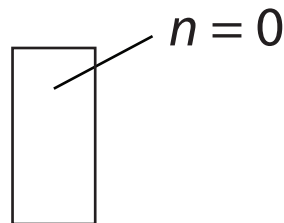
A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

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



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

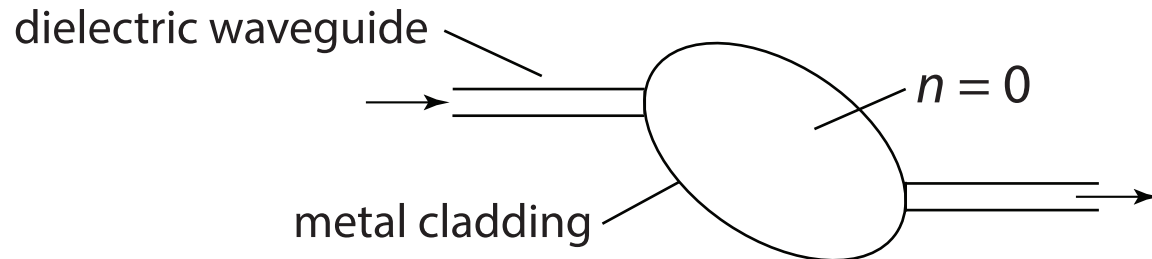
- No Fabry-Perot interference



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

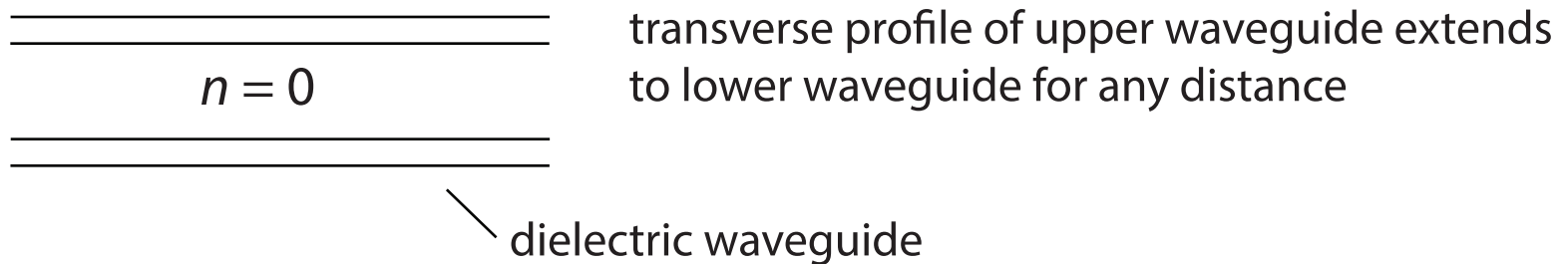
# Some Consequences of ENZ Behaviour - 2

- 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.

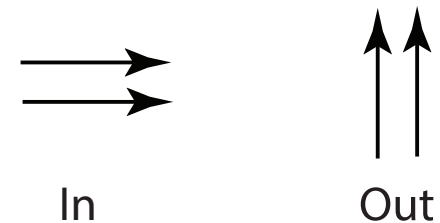
- Large evanescent tails for waveguide coupling



- Automatic phase matching of NLO processes

Recall that  $k = n \omega / c$  vanishes in an ENZ medium.

For example, the following 4WM process is allowed



H. Suchowski et al., Science 342, 1223, 2013.

# Some Consequences of ENZ Behaviour - 3

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- 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

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- 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  $\text{Re } \epsilon = 0$  for  $\omega = \omega_p / \sqrt{\epsilon_{\infty}} \equiv \omega_0$ .

- Challenge: Obtain low-loss ENZ materials

Want  $\text{Im } \epsilon$  as small as possible at the frequency where  $\text{Re } \epsilon = 0$ .

- We are examining a several materials

ITO: indium tin oxide

AZO: aluminum zinc oxide

FTO: fluorine tin oxide



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# Epsilon-Near-Zero Materials for Nonlinear Optics

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- We need materials with a much larger NLO response
- We recently reported a material (indium tin oxide, ITO) with an  $n_2$  value 100 times 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

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Here is the intuition for why the ENZ conditions are of interest in NLO

Recall the standard relation between  $n_2$  and  $\chi^{(3)}$

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

Note that for ENZ conditions the denominator becomes very small, leading to a very large value of  $n_2$

# Optical Properties of Indium Tin Oxide (ITO)

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ITO is a degenerate semiconductor (so highly doped as to be metal-like).

It has a very large density of free electrons, and a bulk plasma frequency corresponding to a wavelength of approximately 1.24  $\mu\text{m}$ .

Recall the Drude formula

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

Note that  $\text{Re } \epsilon = 0$  for  $\omega = \omega_p / \sqrt{\epsilon_{\infty}} \equiv \omega_0$ .

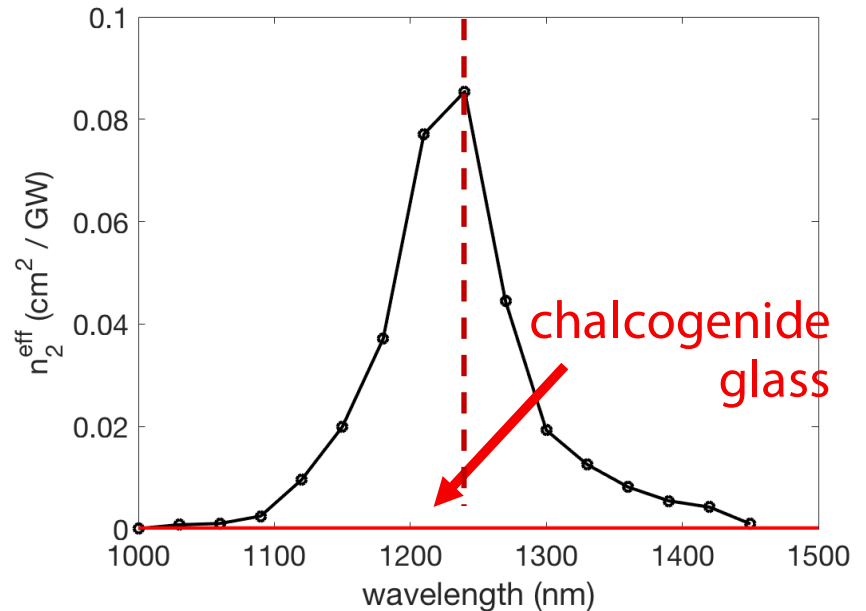
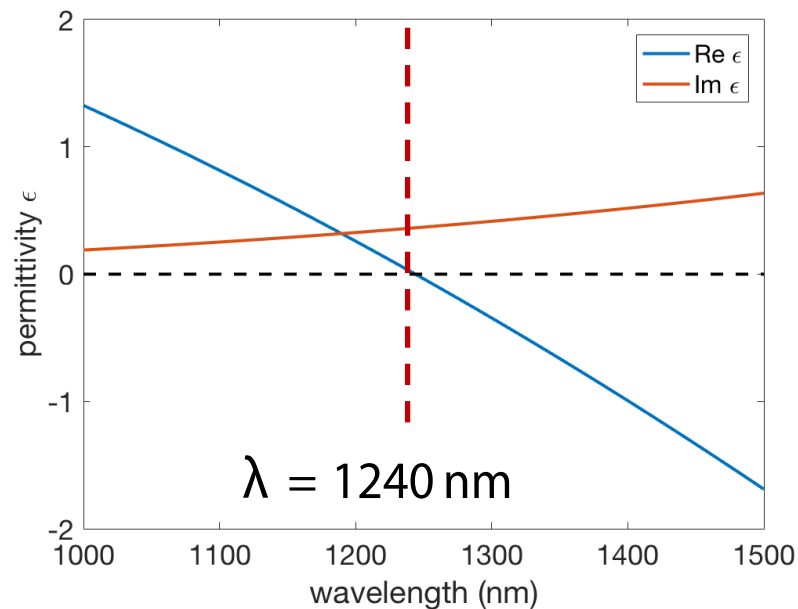
The region near  $\omega_0$  is known as the epsilon-near-zero (ENZ) region.

There has been great recent interest in studies of ENZ phenomena:

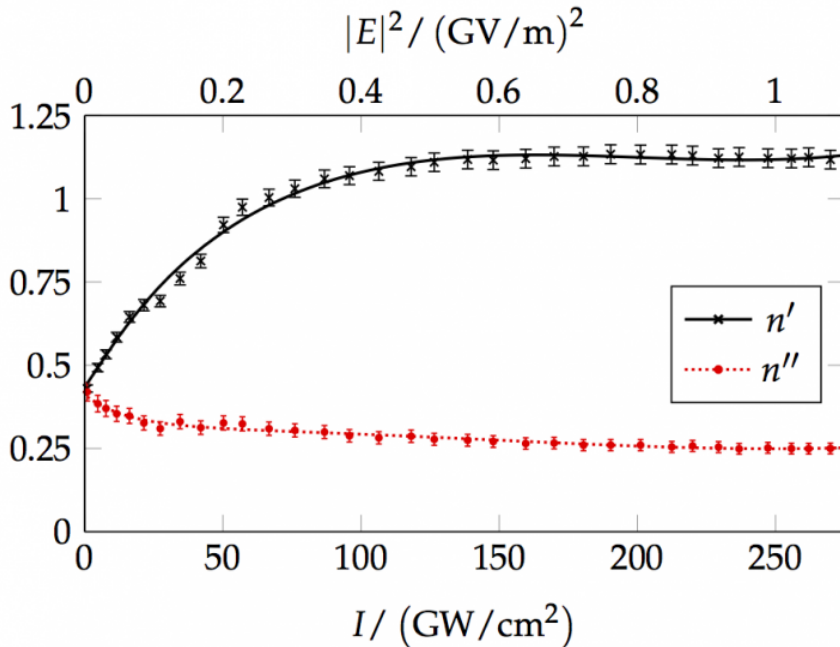
- H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, Science 342, 1223 (2013).
- C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, Phys. Rev. B 85, 045129 (2012).
- S. Campione, D. de Ceglia, M. A. Vincenti, M. Scalora, and F. Capolino, Phys. Rev. B 87, 035120 (2013).
- A. Ciattoni, C. Rizza, and E. Palange, Phys. Rev. A 81, 043839 (2010).

# Huge nonlinear optical response of ITO at its ENZ wavelength

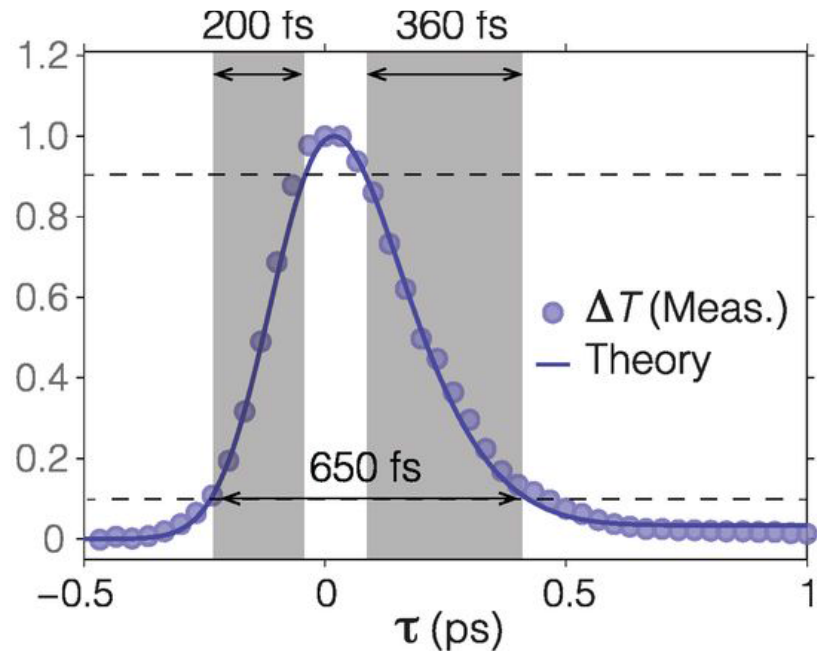
## Indium tin oxide (ITO)



# Fast, ultra-large response of ITO at its ENZ wavelength



- overall change in refractive index of 0.8



- sub picosecond response time



# Some Nonlinear Optical Materials

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Nonlinearity of traditional nonlinear materials:

• SiO <sub>2</sub>	$n_2 = 3.2 \times 10^{-20} \text{ m}^2/\text{W}$	
• SiN	$n_2 = 2.5 \times 10^{-19} \text{ m}^2/\text{W}$	10 × SiO <sub>2</sub>
• Si	$n_2 = 2.7 \times 10^{-18} \text{ m}^2/\text{W}$	100 × SiO <sub>2</sub>
• Chalcogenide glasses	$n_2 = 2.0 \times 10^{-17} \text{ m}^2/\text{W}$	600 × SiO <sub>2</sub>

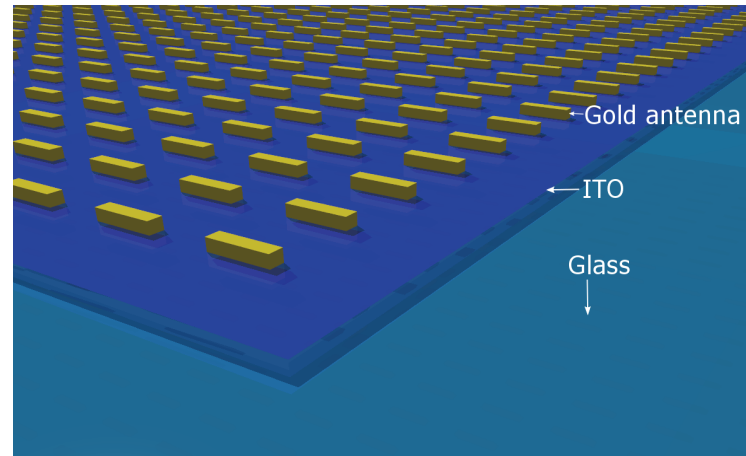
A new class of materials known as **epsilon-near-zero** materials have demonstrated incredible nonlinear properties

• Indium tin oxide (ITO)	$n_2 = 1.1 \times 10^{-14} \text{ m}^2/\text{W}$	600 × <b>ChG</b>
	- $\Delta n = n_2 I = 0.7$	
• Al-doped zinc oxide (AZO)	$n_2 = 3.5 \times 10^{-17} \text{ m}^2/\text{W}$	2 × <b>ChG</b>
	- $\Delta n/n = 4.4$	

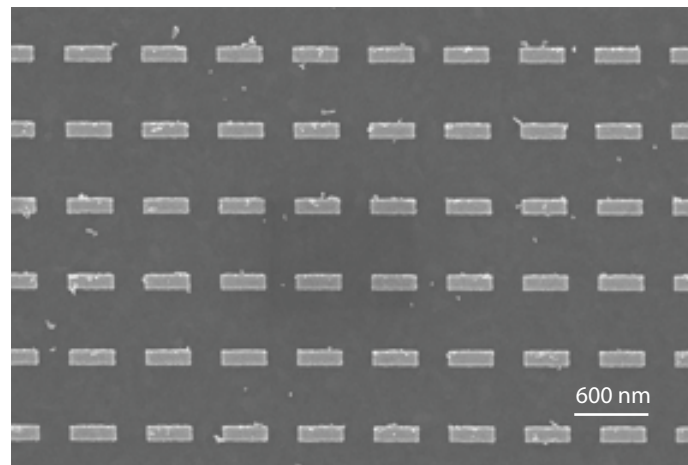
# An ENZ Metasurface

- Can 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

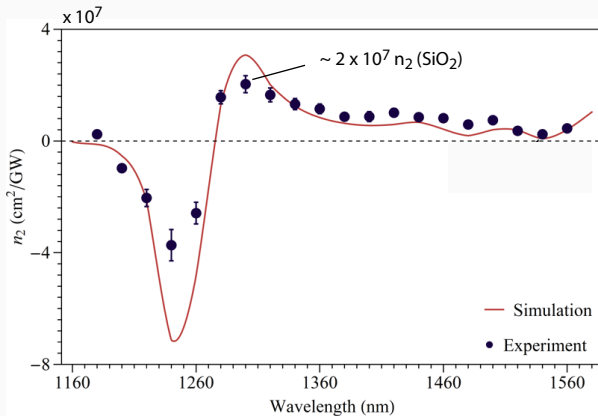
Concept:



SEM:



# NLO response of the coupled antenna-ENZ system



The material exhibits extremely large  $n_2$  over a broad spectral range. The magnitude of the on-resonance value is **7 orders of magnitude larger than that of  $\text{SiO}_2$** .

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

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# Nonperturbative Nature of the NLO Response

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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 nonlinear response can be accurately modeled in the  $\chi^{(3)}$  limit by

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

where

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

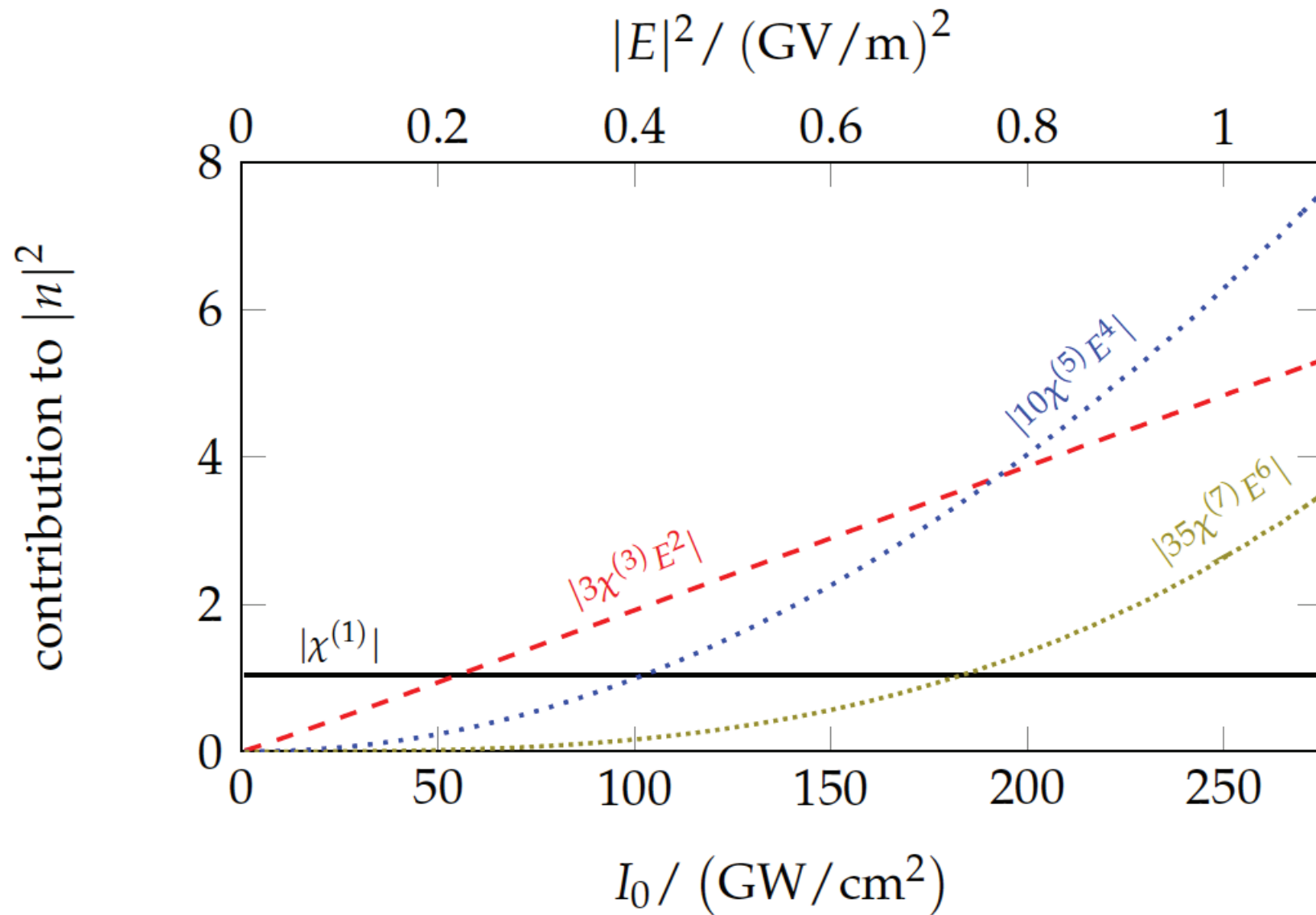
and

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

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

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

# Nonlinear Response of ITO is Nonperturbative





# Physics and Applications of Epsilon-Near-Zero Materials

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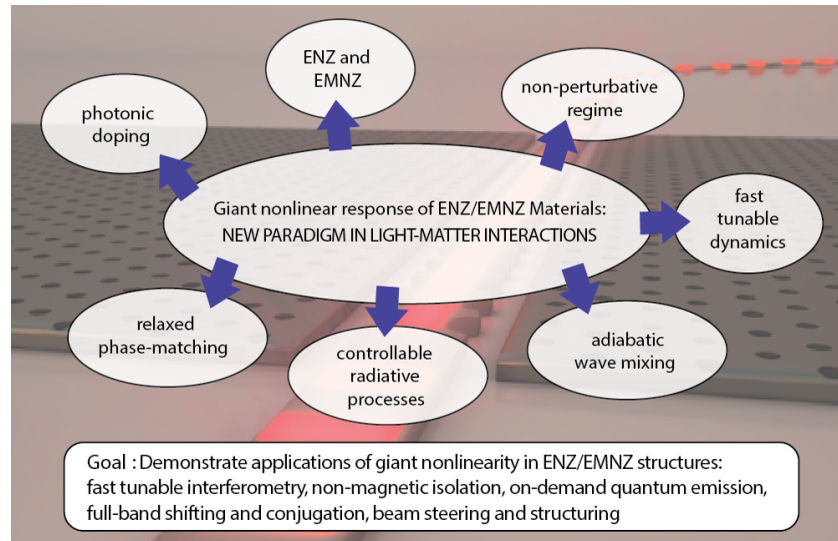
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# Giant Nonlinear Response of ENZ Metastructures

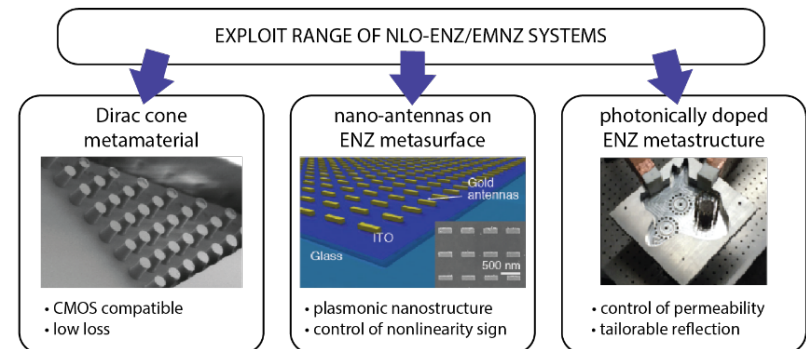
**Boyd** (Rochester), **Engheta** (UPenn), **Mazur** (Harvard), **Willner** (USC)

HR001118S0014 NLM BAA  
Attachment 1

## CONCEPT

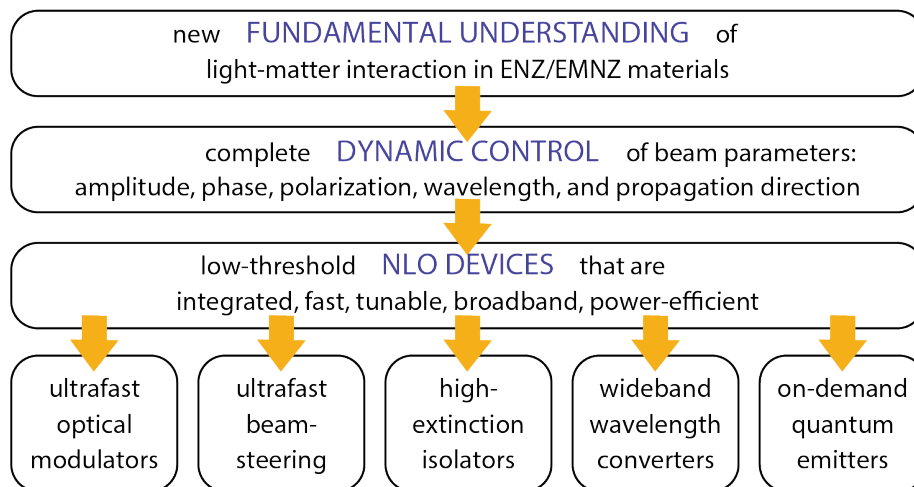


## APPROACH



- Re-formulate nonlinear optics for non-perturbative regime
- Explore experimental characteristics of ENZ/EMNZ platforms
- Fabricate devices to exploit novel features

## IMPACT

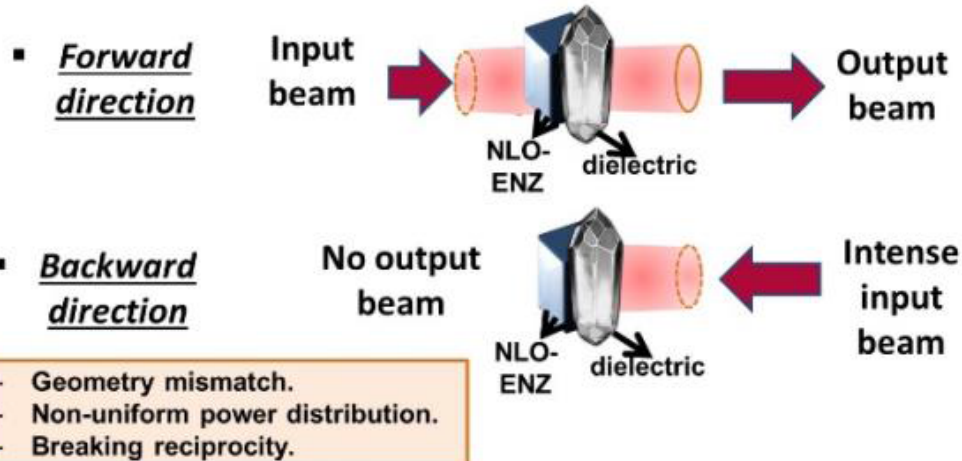


## CONTEXT

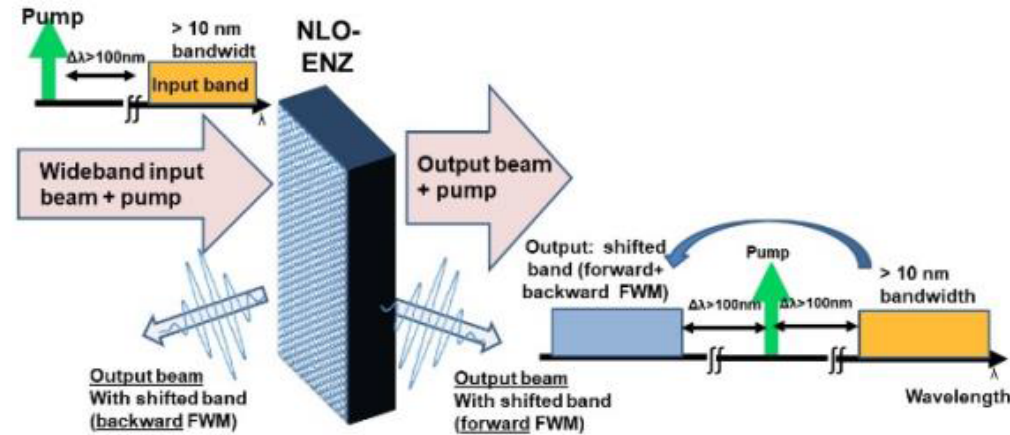
- Nonlinear optical phenomena currently require high-intensity sources, which are generally incompatible with nanophotonics
- Photonics could have an enormous impact on many new fields if nonlinearity is fundamentally enhanced
- ENZ metastructures provide giant tailorable nonlinearity
- Enhanced nonlinearity will open door to manipulating light on the nanoscale

# Some Potential Applications of ENZ Behavior

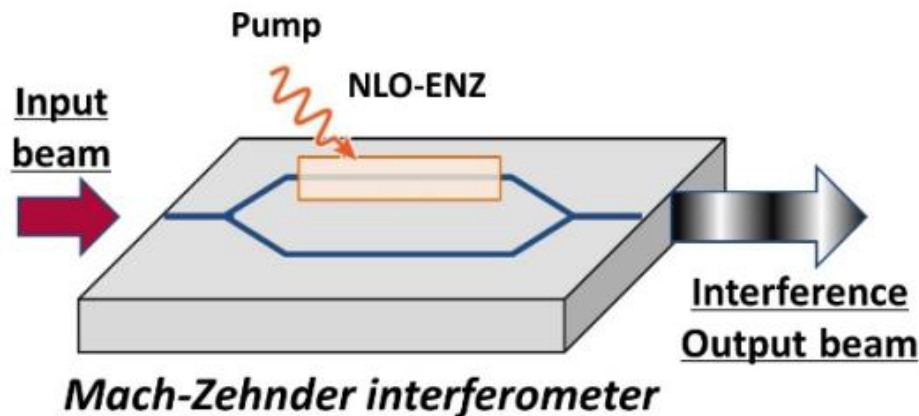
## (a) Non-magnetic isolation



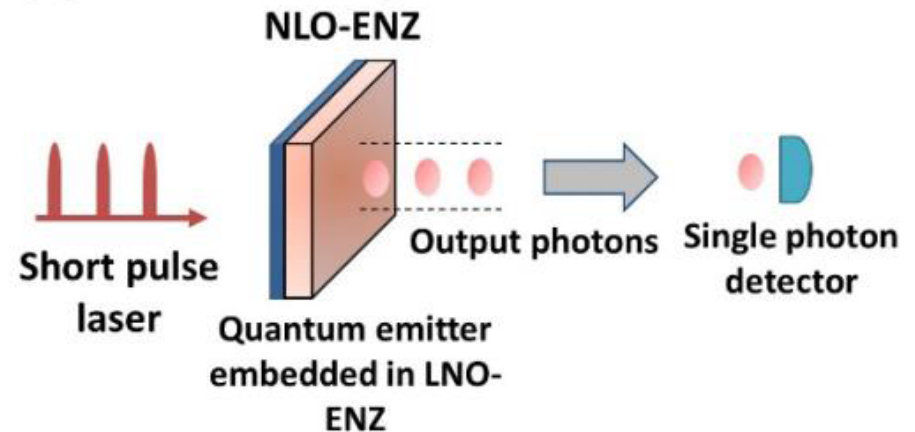
## (b) Full-band shifting and conjugation



## (c) High-speed tunable interferometers

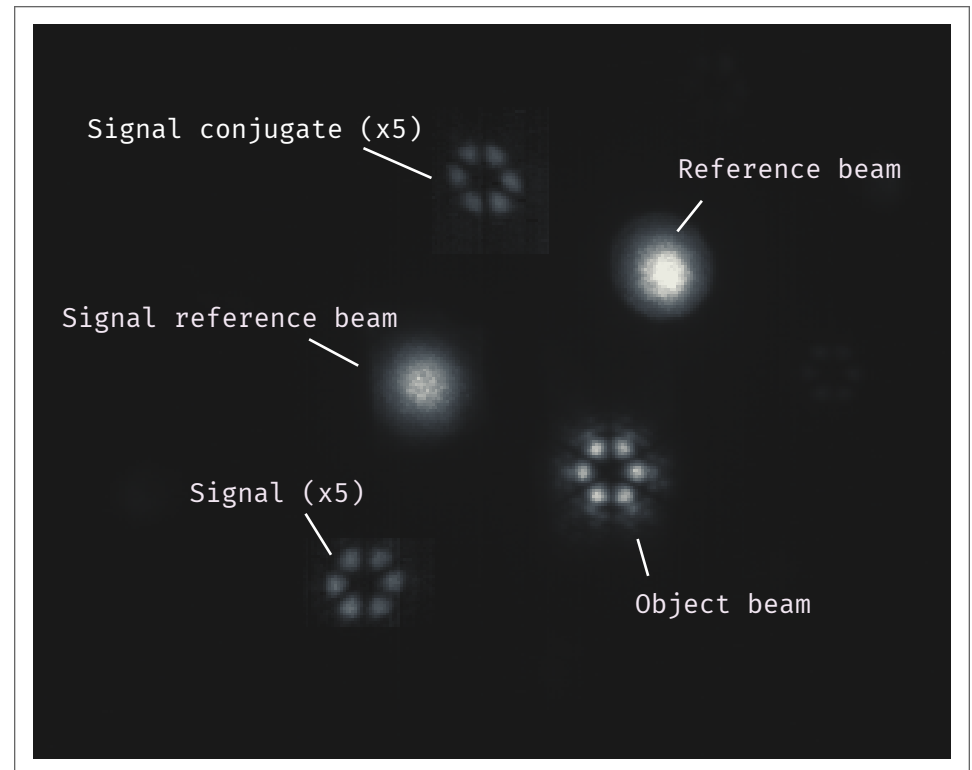
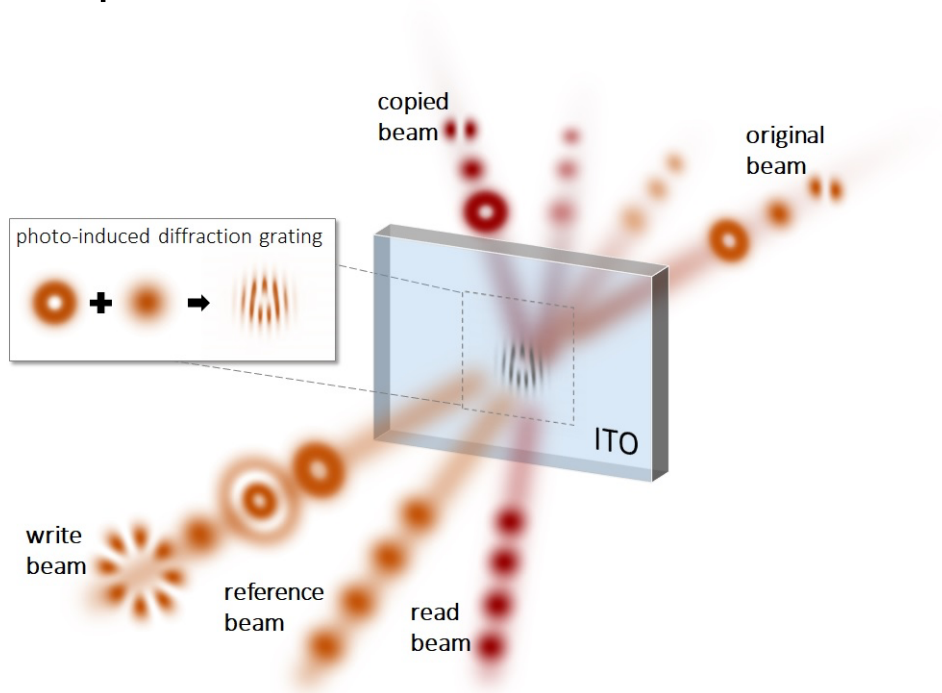


## (d) On-demand quantum emitter



# Ultrafast Holography and Beam Copying

- Real-time holography with sub-picosecond response time
- Schematic of beam-copying procedure
- Laboratory 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](http://boydnlo.ca/presentations)



# Special Thanks To My Students and Postdocs!

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## Ottawa Group



## Rochester Group

