



Nonlinear Optical Metastructures

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

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Canada Excellence Research Chair (CERC) in Nonlinear Quantum Optics

Research interest:

Nonlinear optics, quantum optics,
integrated photonics, meta-materials, etc.

Epsilon-Near-Zero (ENZ) Materials

- Physics of Epsilon-Near-Zero (ENZ) Materials
- Huge NLO Response of ENZ Materials and Metastructures
- Non-perturbative nature of the NLO Response (usual power series do not converge)

With Special Thanks To:

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Physics of Epsilon-Near-Zero (ENZ) Materials

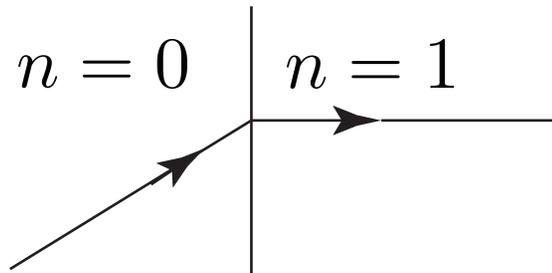
- ENZ materials possess exotic electromagnetic properties
Silveirinha, Engheta, Phys. Rev. Lett. 97, 157403, 2006.
- If the dielectric permittivity ϵ is nearly zero, then refractive index $n = \sqrt{\epsilon}$ is nearly zero.
Thus $v_{\text{phase}} = c/n$ is nearly infinite
 $\lambda = \lambda_{\text{vac}} / n$ 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

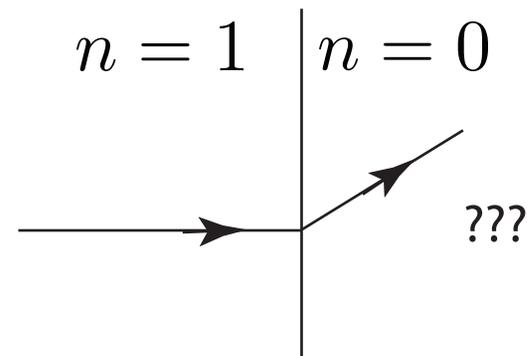
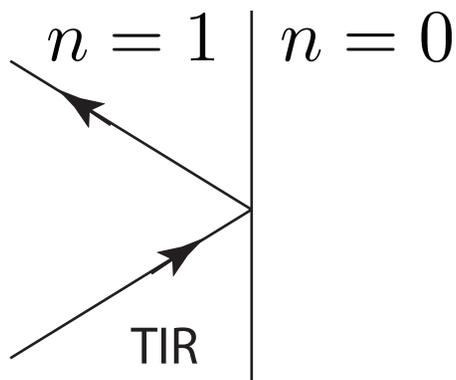
$$n_1 \sin \theta_1 = n_2 \sin \theta_2$$

- 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.

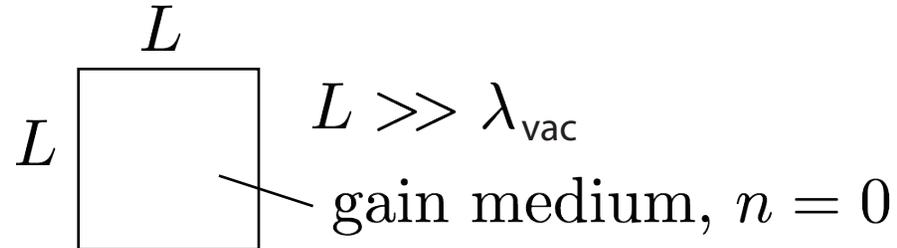
Some Consequences of ENZ Behaviour - 1

- Funny lenses



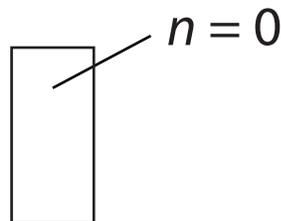
A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

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



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

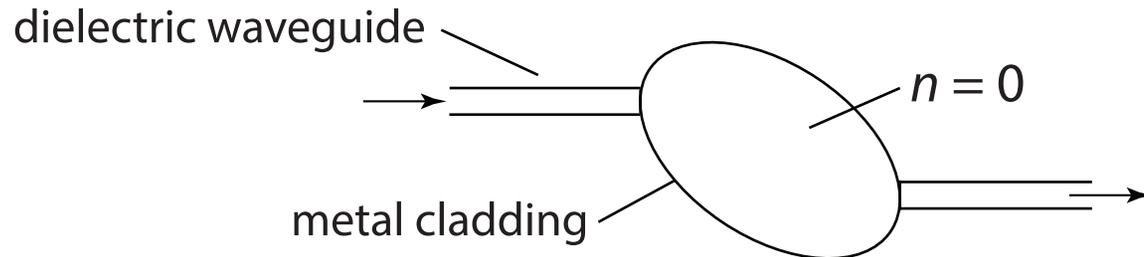
- No Fabry-Perot interference



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

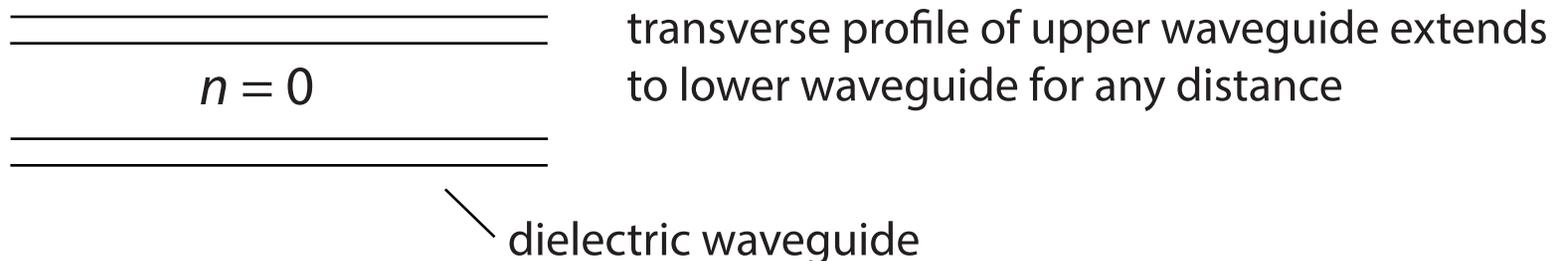
Some Consequences of ENZ Behaviour - 2

- 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.

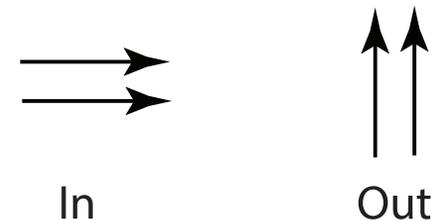
- Large evanescent tails for waveguide coupling



- 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

- How is the theory of self-focusing modified?
- Does the theory of Z-scan need to be modified?
- How is the theory of blackbody radiation modified?
- Do we expect very strong superradiance effects?
- More generally, how is any NLO process modified when $n_0 = 0$?

Epsilon-Near-Zero Materials

- Metamaterials

Materials tailor-made to display ENZ behaviour

- Homogeneous materials

All materials display ENZ behaviour at their (reduced) plasma frequency

Recall the Drude formula

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

Note that $\text{Re } \epsilon = 0$ 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

Epsilon-Near-Zero Materials for Nonlinear Optics

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

Large optical nonlinearity of indium tin oxide in its epsilon-near-zero region, M. Zahirul Alam, I. De Leon, R. W. Boyd, Science 352, 795 (2016).

Implications of ENZ Behavior for Nonlinear Optics

Here is the intuition for why the ENZ 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)

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 $\omega = \omega_p / \sqrt{\epsilon_{\infty}} \equiv \omega_0$.

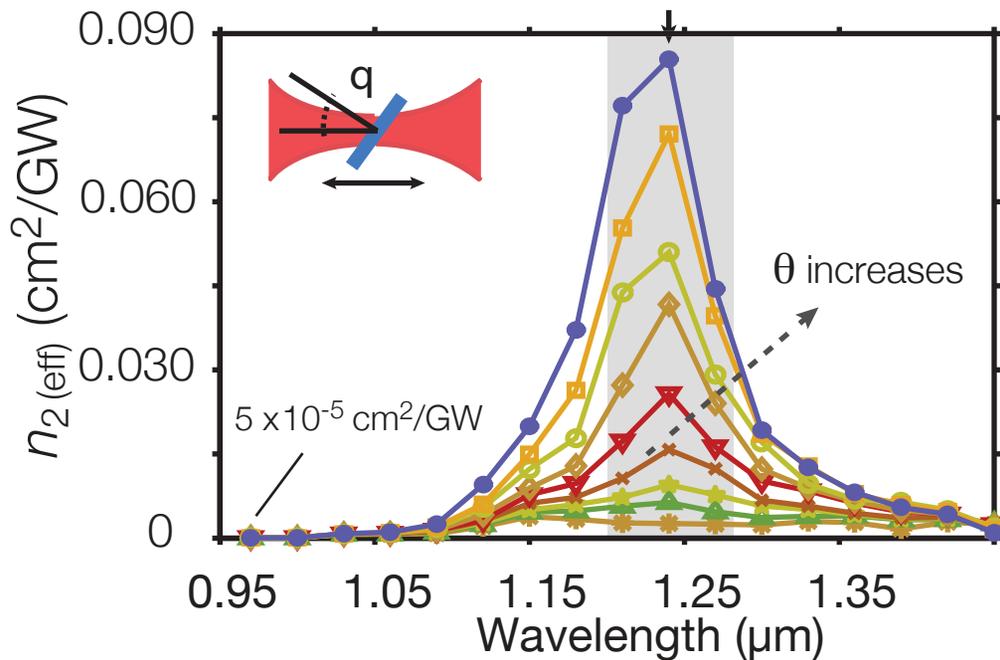
The region near ω_0 is known as the epsilon-near-zero (ENZ) region.

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

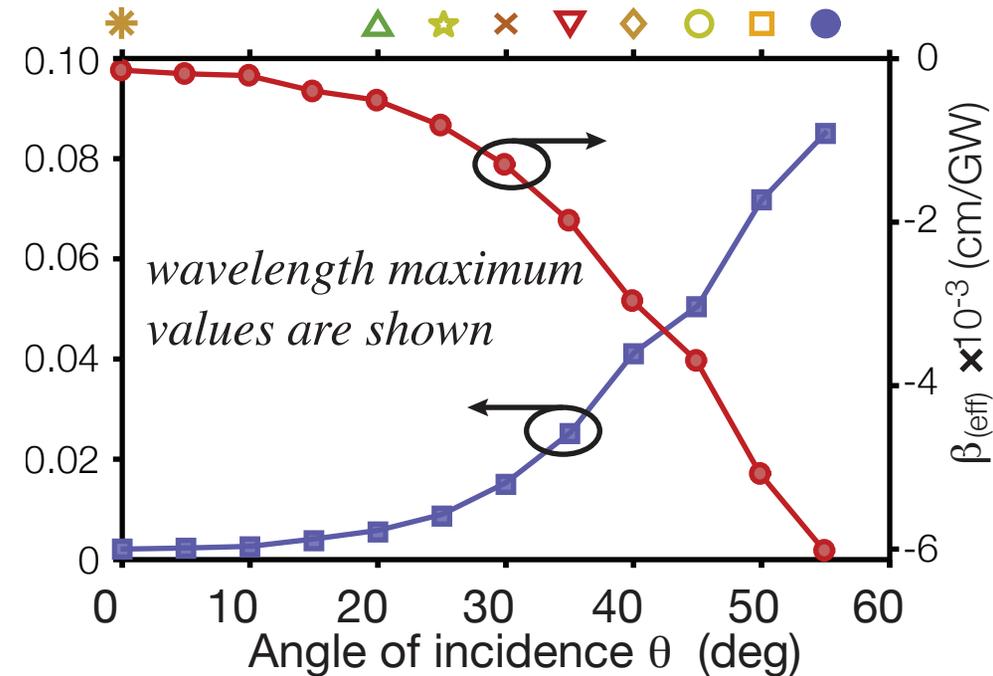
- H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, *Science* 342, 1223 (2013).
- C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, *Phys. Rev. B* 85, 045129 (2012).
- S. Campione, D. de Ceglia, M. A. Vincenti, M. Scalora, and F. Capolino, *Phys. Rev. B* 87,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

Huge Nonlinear Optical Response Measured by Z-scan

Wavelength dependence of n_2

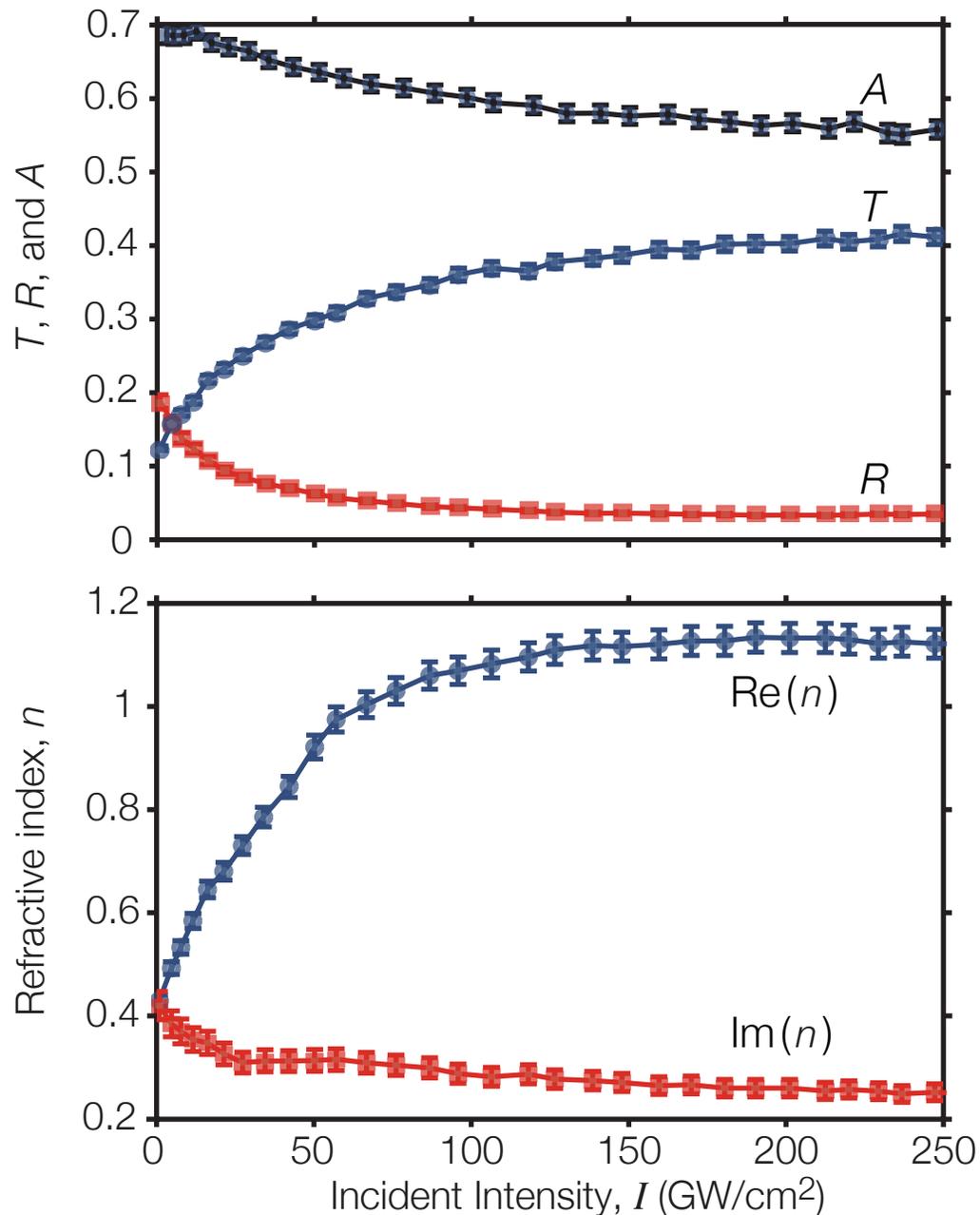


Variation with incidence angle



- Note that n_2 is positive (self focusing) and β is negative (saturable absorption)
- Both n_2 and nonlinear absorption increase with angle of incidence
- n_2 shows a maximum value of $0.11 \text{ cm}^2/\text{GW} = 1.1 \times 10^{-10} \text{ cm}^2/\text{W}$ at $1.25 \mu\text{m}$ and 60° . This value is 2000 times larger than that away from ENZ region.
- n_2 is 3.4×10^5 times larger than that of fused silica
- n_2 is 200 times larger than that of chalcogenide glass

Beyond the $\chi^{(3)}$ limit



The nonlinear change in refractive index is so large as to change the transmission, absorption, and reflection!

Note that transmission is increased at high intensity.

Here is the refractive index extracted from the above data.

Note that the total nonlinear change in refractive index is $\Delta n = 0.8$.

The absorption decreases at high intensity, allowing a predicted NL phase shift of 0.5 radians.

Nonperturbative Nature of the NLO Response

1. The conventional equation $n = n_0 + n_2 I$ is not applicable to ENZ and other low-index materials. The nonlinear response is nonperturbative.
2. The 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

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

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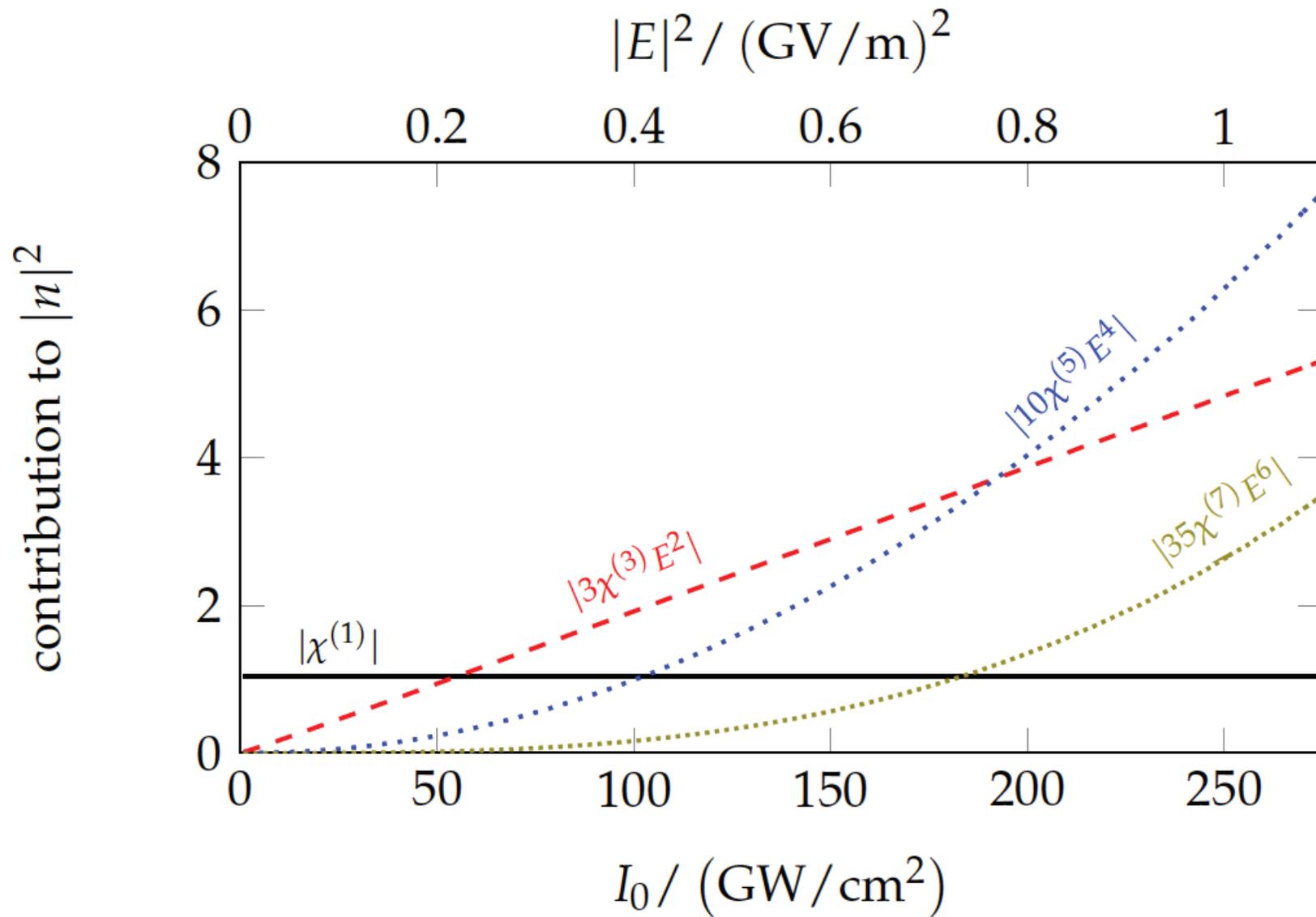
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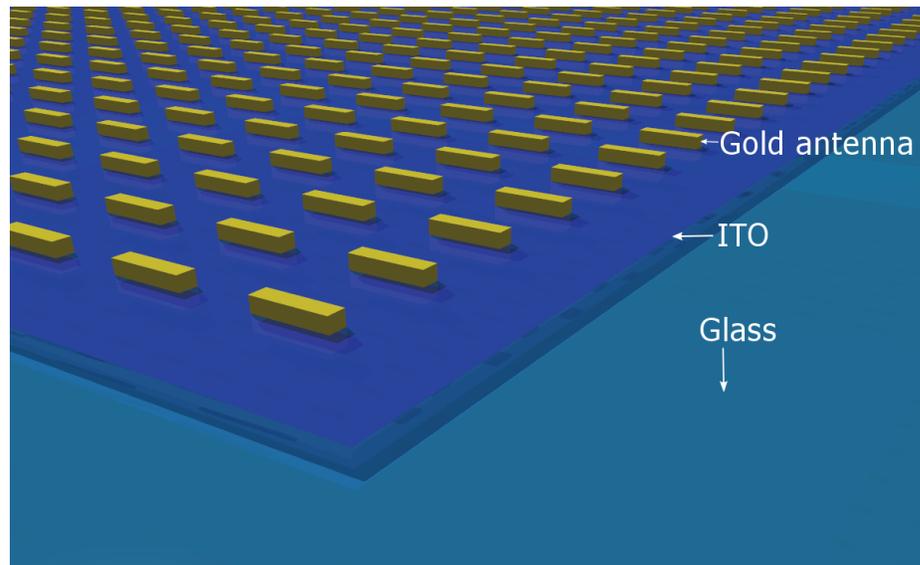
Nonlinear Response of ITO is Nonperturbative



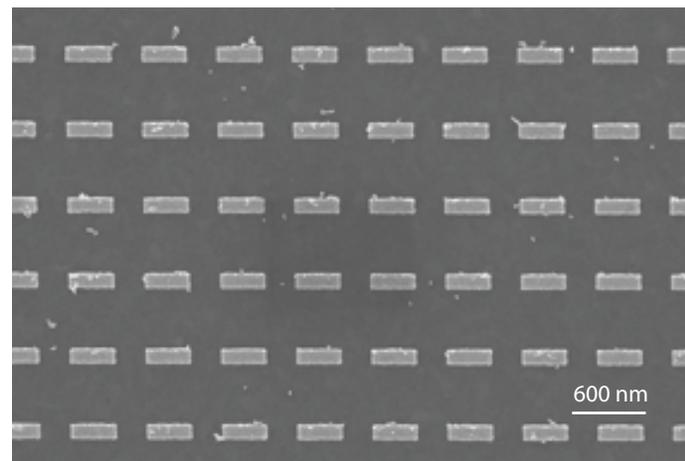
An ENZ Metasurface

- 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

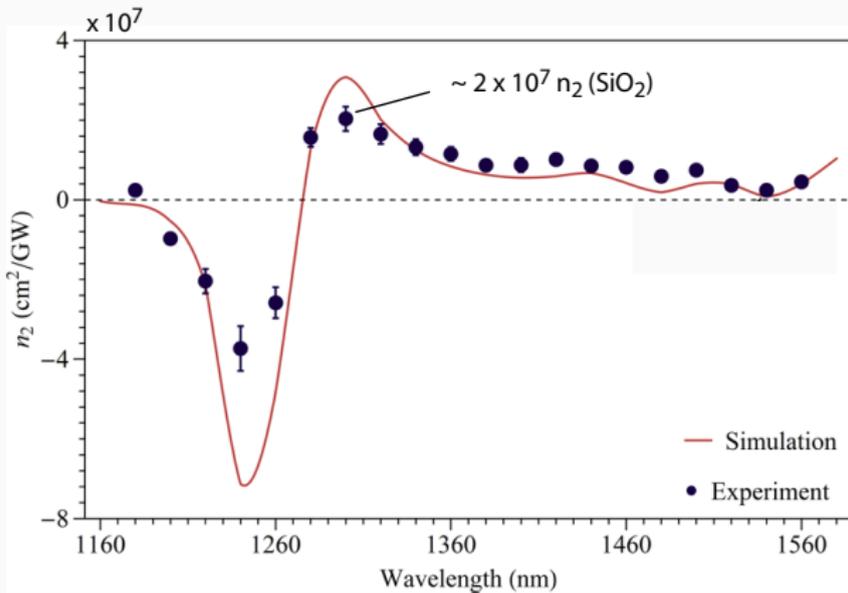
Concept:



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 SiO_2** .

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

Summary: ITO/ENZ - gold nanoantenna metasurface

- A broadband nonlinear material with n_2 values up to 7 orders of magnitude larger than that of SiO_2 .
- Sub-picosecond response time.
- $\Delta n \approx \pm 2.5$ over very large bandwidth.
- One can tailor the sign of the nonlinearity by simply designing the geometric parameters of the antenna appropriately.

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Special Thanks To My Students and Postdocs!

Ottawa Group



Rochester Group

