Nonlinear Optics of Epsilon-Near-Zero Materials

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New Nonlinear Optical Material for Quantum Photonics

- We want all-optical switches that work at the single-photon level
- We need photonic materials with a much larger NLO response
- We recently reported a new NLO material with an n_2 value 100 times larger than those previously reported (but with some background absorption).
- Material makes use of strong enhancement that occurs in the epsilon-near zero (ENZ) spectral region.
- A potential game changer for the field of photonics

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 $\chi^{(3)}$

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

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

Nonlinear 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 μm.

Recall the Drude formula

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

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

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

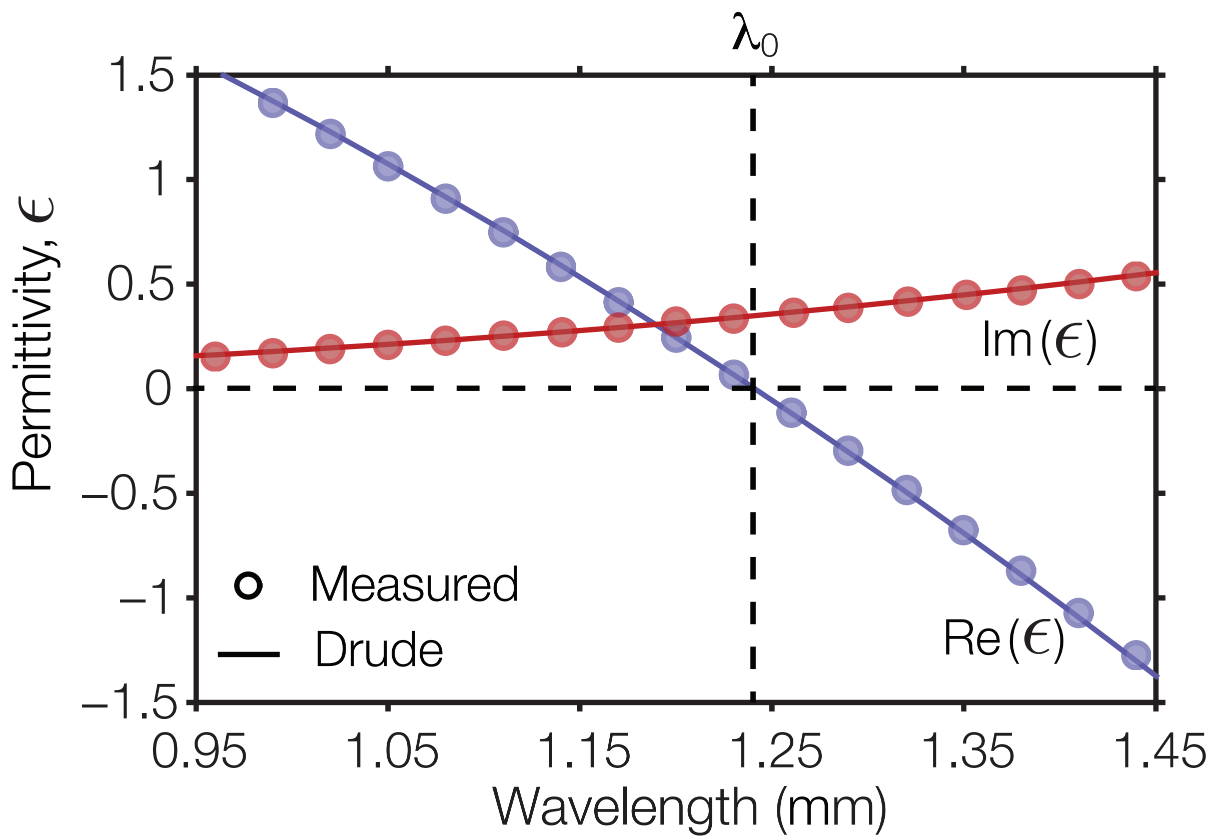
There has been great recent interest in studies of ENZ phenomena:

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).

The Epsilon-Near-Zero (ENZ) region of Indium Tin Oxide (ITO)

Measured real and imaginary parts of the dielectric permittivity.

Commercial ITO sample, 310 nm thick on a glass substrate



Drude fit

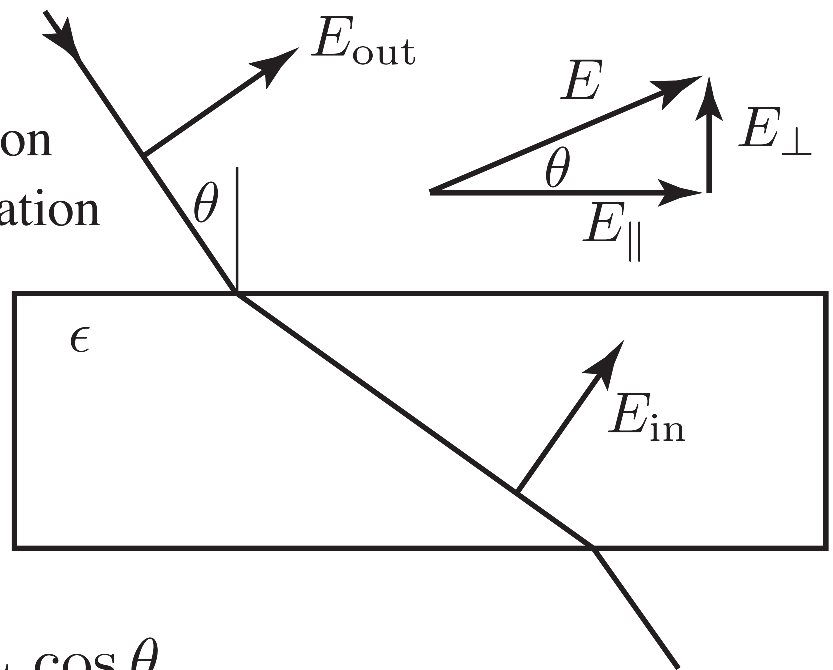
$\epsilon_\infty = 3.77$

$\gamma = 0.0468\ \omega_p$

$\omega_p/2\pi = 473$ THz

Note that Re(ϵ) vanishes at 1.24 mm, but that the loss-part Im(ϵ) is non-zero.

The NLO Response Is Even Larger at Oblique Incidence



Standard boundary conditions show that:

$$E_{\text{in},\parallel} = E_{\text{out},\parallel} = E_{\text{out}} \cos\theta$$

$$D_{\text{in},\perp} = D_{\text{out},\perp} \quad \Rightarrow \quad E_{\text{in},\perp} = E_{\text{out},\perp}/\epsilon = E_{\text{out}} \cos\theta/\epsilon$$

Thus the total field inside of the medium is given by

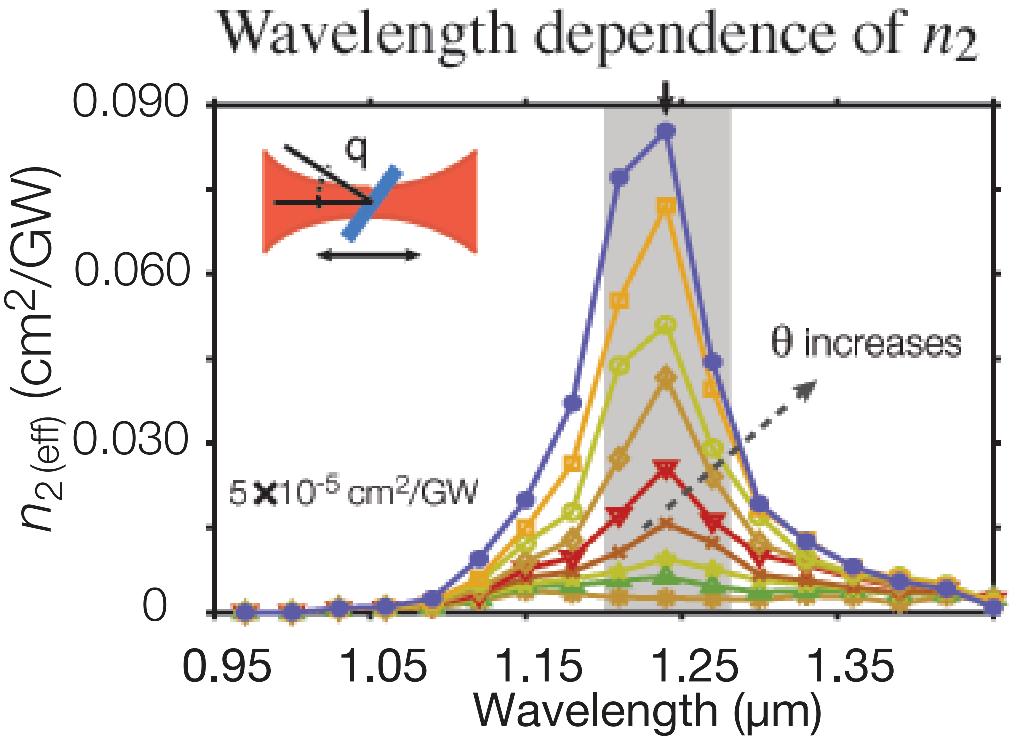
$$E_{\text{in}} = E_{\text{out}} \sqrt{\cos^2\theta + \frac{\sin^2\theta}{\epsilon}}$$

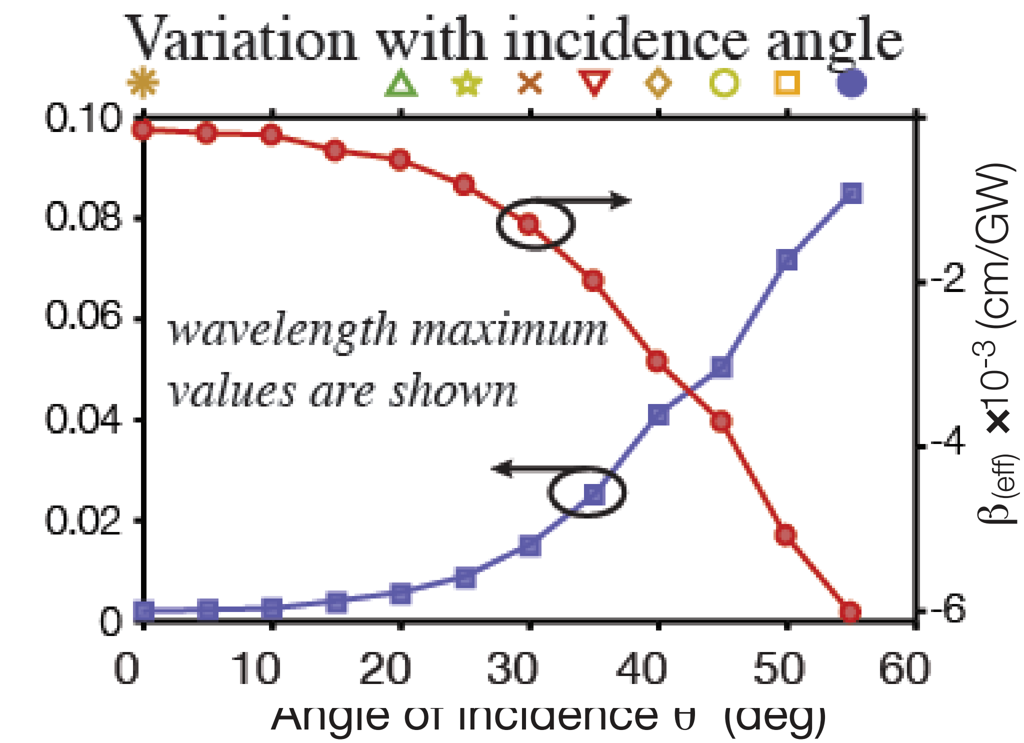
Note that, for $\epsilon < 1$, E_{in} exceeds E_{out} for $\theta \neq 0$.

Note also that, for $\epsilon < 1$, E_{in} increases as θ increases.

Huge Nonlinear Optical Response of ITO

- Z-scan measurements for various angles of incidence



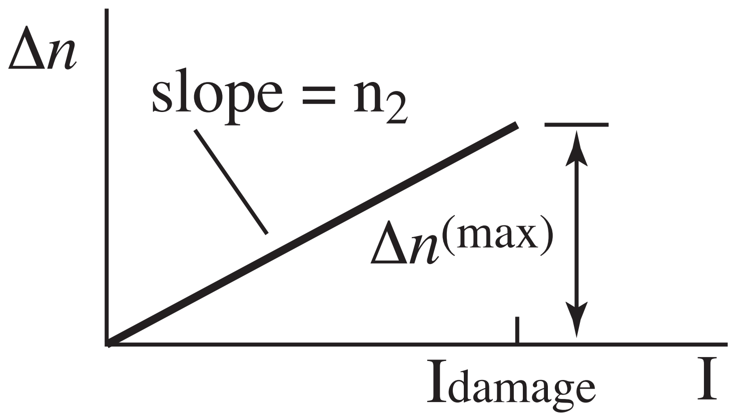


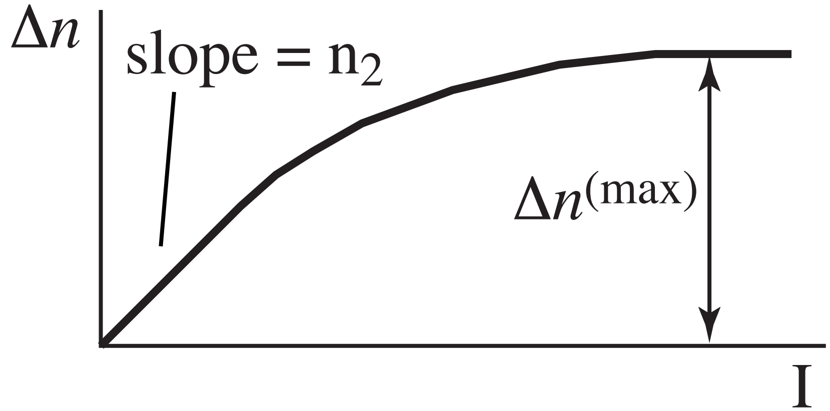
peak laser intensity was 50 GW cm-2

- 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 cm^2/GW = 1.1 x 10^{-10} cm^2/W at 1.25 μm and 60 deg. This value is 2000 times larger than that away from ENZ region.

What Makes a Good (Kerr-Effect) Nonlinear Optical Material?

- We want n_2 large ($\Delta n = n_2 I$). We also want $\Delta n^{(\max)}$ large.
  These are distinct concepts! Damage and saturation can limit $\Delta n^{(\max)}$



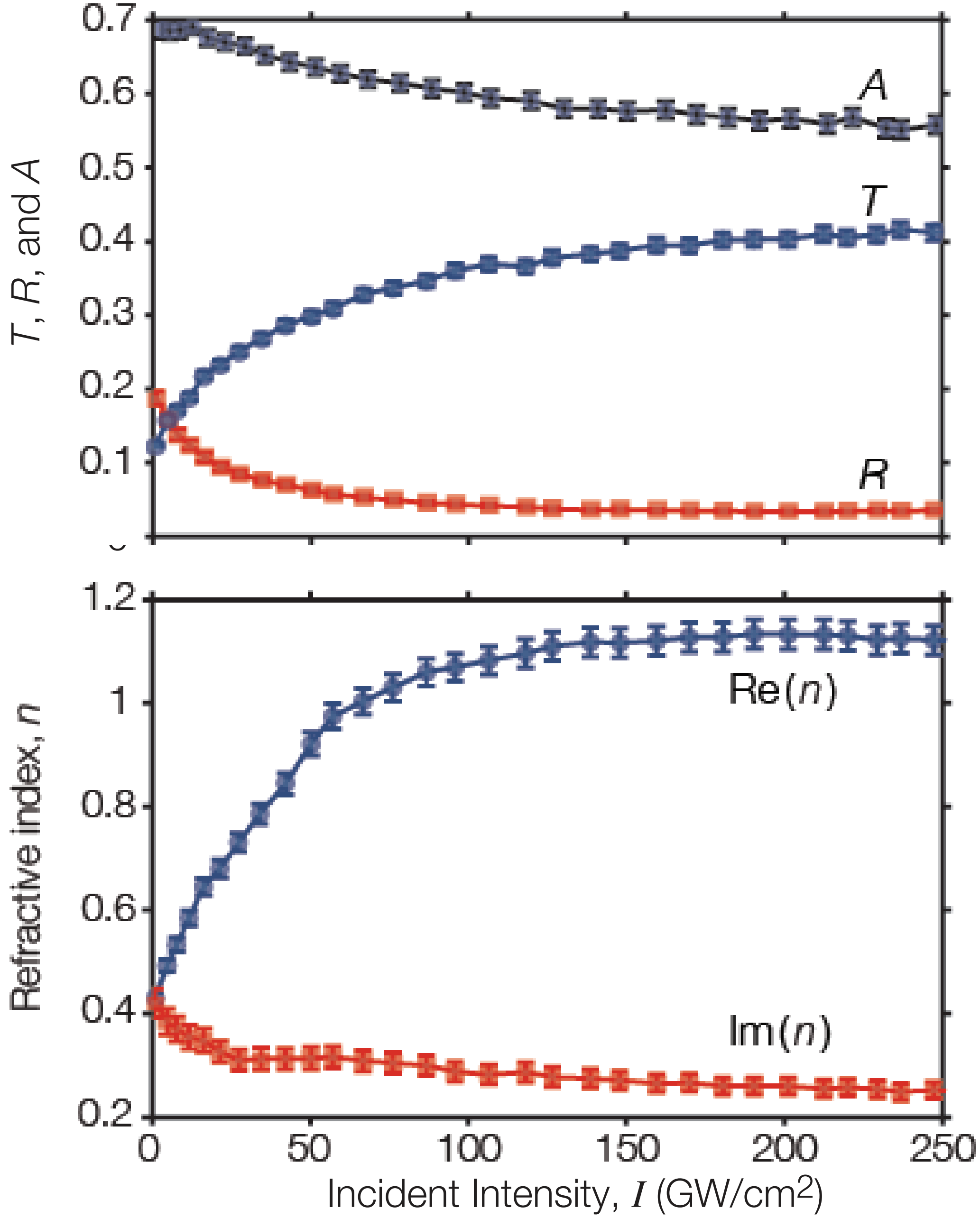


- For ITO at ENZ wavelength, both n_2 and $\Delta n^{(\max)}$ are extremely large
  ($n_2 = 1.1 \times 10^{-10}$ cm^2/W and $\Delta n^{(\max)} = 0.8$)

- n_2 is 3.4×10^5 times larger than that of silica glass
  $\Delta n^{(\max)}$ is 2700 times larger that that of silica glass
  (For silica glass $n_2 = 3.2 \times 10^{-16}$ cm^2/W, $I_{damage} = 1$ TW/cm^2, and thus $\Delta n_{(\max)} = 3 \times 10^{-4}$)

M. Z. Alam, I. De Leon, R. W. Boyd, Science 352, 795 (2016).

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.

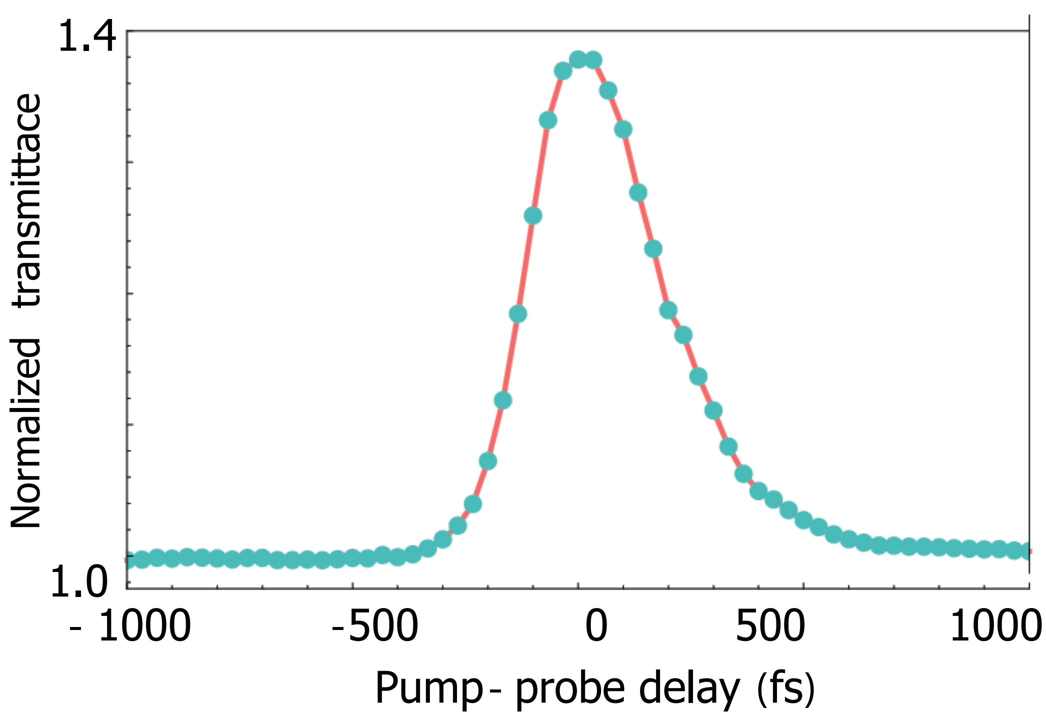
Here is the refractive index extracted from the above data.

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.

Measurement of Response Time of ITO

- We have performed a pump-probe measurement of the response time. Both pump and probe are 100 fs pulses at 1.2 μm.
- Data shows a rise time of no longer than 200 fs and a recover time of of 360 fs.
- Results suggest a hot-electron origin of the nonlinear response
- ITO will support switching speeds as large as 1.5 THz



Implications of the Large NLO Response of ITO

Indium Tin Oxide at its ENZ wavelength displays enormously strong NLO properties:

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
Nonlinear change in refractive index as large as 0.8

Note that the usual "power-series" description of NLO is not adequate for describing this material. (We can have fun reformulating the laws of NLO!)

Some possible new effects
Waveguiding outside the "weakly-guiding" regime
Efficient all-optical switching
No need for phase-matching
Control of radiative processes

Other Reports of Highly Nonlinear Response in ENZ Material and Metamaterialc

Enhanced Nonlinear Refractive Index in epsilon-Near-Zero Materials,
L. Caspani, R. P. M. Kaipurath, M. Clerici, M. Ferrera, T. Roger, J. Kim, N. Kinsey,
M. Pietrzyk, A. D. Falco, V. M. Shalaev, A. Boltasseva and D. Faccio,
Phys. Rev. Lett. 116, 233901, 2016.

Giant nonlinearity in a superconducting sub-terahertz metamaterial,
V. Savinov, K. Delfanazari, V. A. Fedotov, and N. I. Zheludev
Applied Physics Letters 108, 101107 (2016); doi: 10.1063/1.4943649

Nano-optomechanical nonlinear dielectric metamaterials
Artemios Karvounis, Jun-Yu Ou, Weiping Wu, Kevin F. MacDonald, and Nikolay I. Zheludev
Applied Physics Letters 107, 191110 (2015); doi: 10.1063/1.4935795.

Nanostructured Plasmonic Medium for Terahertz Bandwidth All-Optical Switching
Mengxin Ren , Baohua Jia , Jun-Yu Ou , Eric Plum, Jianfa Zhang , Kevin F. MacDonald ,
Andrey E. Nikolaenko , Jingjun Xu, Min Gu, and Nikolay I. Zheludev *
Adv. Mater. 2011, 23, 5540–5544 (2011).

A Metasurface for Large Nonlinear Refraction

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The ultimate goal:

The computation speed and energy requirements of a classical computer are fundamentally limited by the RC-constant.

We want a solid-state system that can be a platform for future all-optical computation and signal processing devices using a few tens of photons.

Holy grail of all-optical computation:

- Energy expenditure: aJ per bit.
- Speed: Multi-terahertz switching speed.
- Contrast: Large on-off contrast.
- Size: less than 100 nm.
- A solid-state system.
- CMOS compatibility and other integration challenges.

There are also two fundamental (in technological sense) questions:

- How large can ultrafast nonlinearity be?
- How large can the total nonlinear contribution to the refractive index, Δn, be?

The structure under consideration: antenna array on ITO.

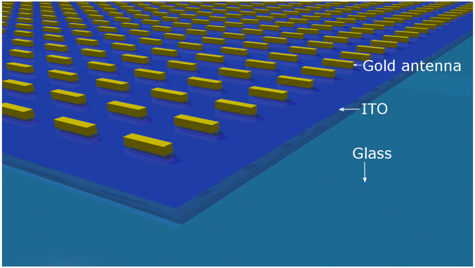


Figure 1: The artist view of the structure.

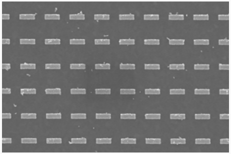
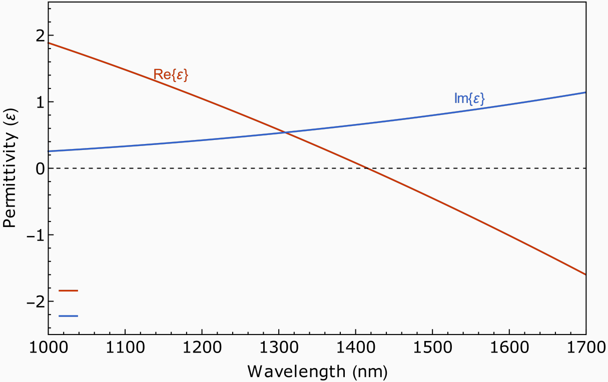


Figure 2: An SEM image of the fabricated sample.

A 23 nm thick ITO as the ENZ medium.



The real part of the permittivity crosses zero at 1420 nm.

A thin ENZ medium supports a bulk plasma mode.

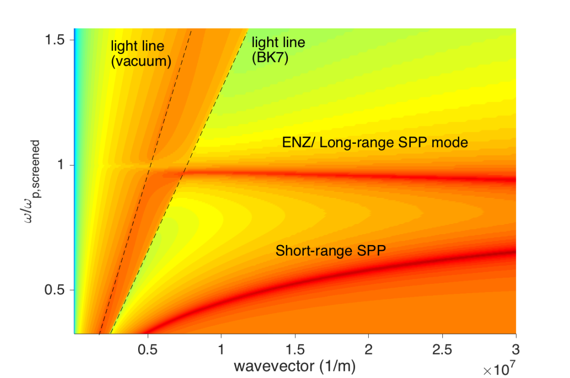


Figure 3: A thin layer of ITO supports two modes: bulk plasma, short range surface plasmon (SPP).

The fundamental mode of antenna couples strongly with the ENZ mode.

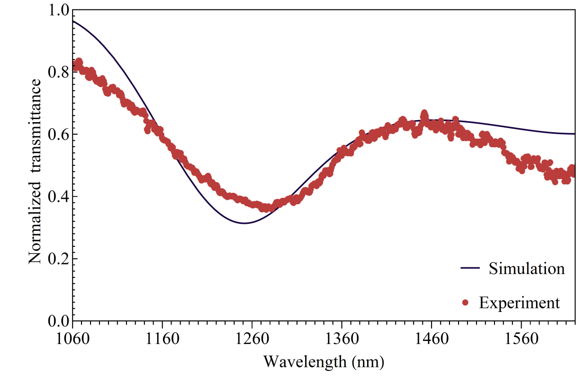


Figure 4: The coupling between the fundamental mode of the optical dipole antenna and the ENZ mode results in two distinct dips in the transmission spectrum.

We investigated the nonlinear response of the coupled system using a series of z-scan measurements.

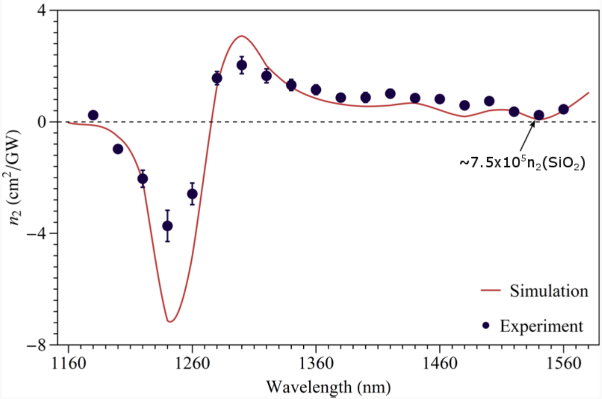


Figure 5: The material exhibits extremely large n_2 for the entire spectral range. The magnitude of the on-resonance value is 7 orders of magnitude larger than that of SiO_2.

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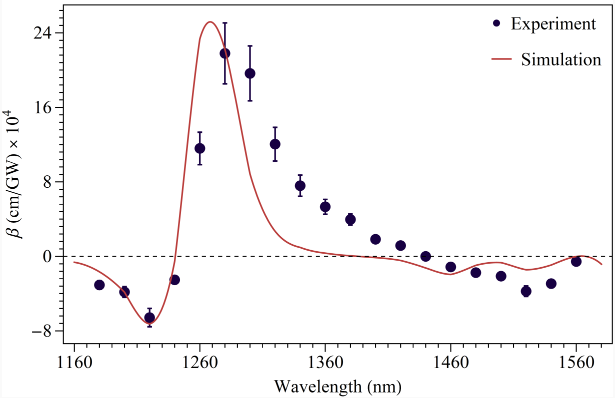


Figure 6: Nonlinear absorption shows both negative and positive signs indicating saturable and reverse saturable absorption respectively. Note: One can design to maximize the absorption for applications in power limiter, low power pulsed lasing, etc.

Temporal response using a pump-probe transmittance measurement at 1280 nm.

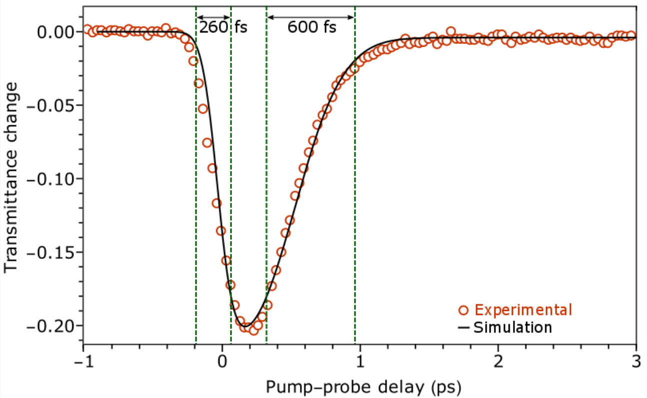
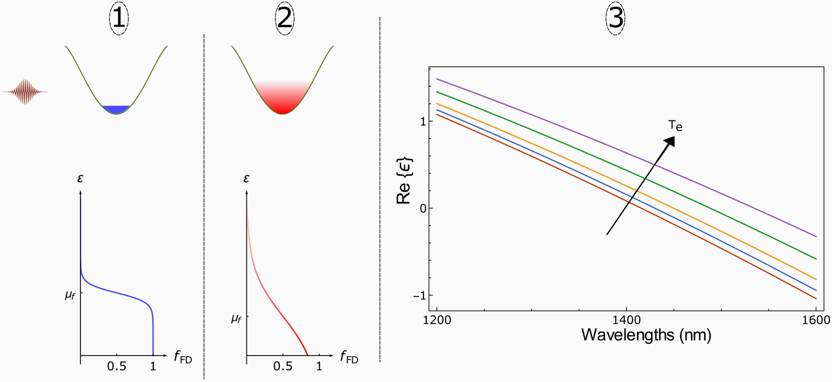


Figure 7: The total on-resonance response time 860 fs is 50% larger than that of a high conductivity bare ITO film.

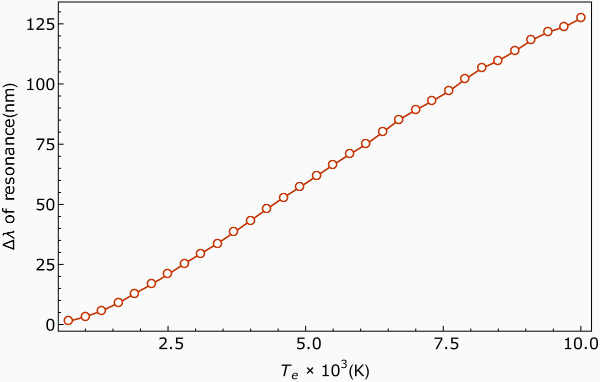
Mechanisms of the nonlinear response.

ITO has non-parabolic conduction band.



The plasma frequency ($\omega_p = \sqrt{\frac{n_e e^2}{m^*(T_e)\epsilon_0}}$) - hence, the permittivity - is a function of intensity due to the ultrafast modulation of the effective mass of the conduction band electrons.

The resonance of the coupled system is dictated by the zero crossing wavelength of the ITO.



The resonance wavelength of the coupled system red-shifts as the electron temperature rises (i.e. plasma frequency shifts to a longer wavelength).

Field enhancement increases the efficiency of the nonlinear response.

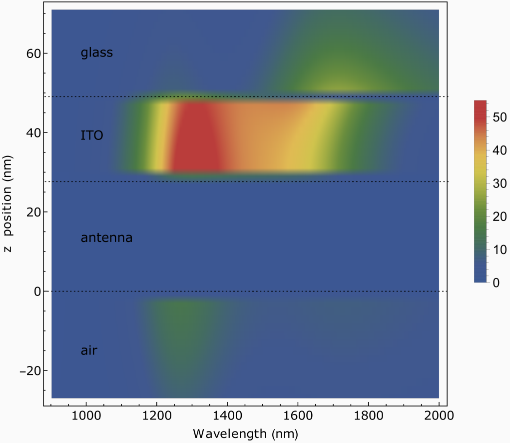


Figure 8: The coupled system exhibits intensity enhancement up to a factor of ~ 50 inside the ITO. Note: light is incident from air (k-vector points up).

Simplified description of the mechanism:

- The resonance wavelength of a dipole antenna depends on the refractive index of the surrounding medium.
- An ultrafast increase in the refractive index of the surrounding medium necessarily results in the red-shift of the resonance wavelength of the antenna.
- An optical beam that is nearly resonant to the system experiences large linear refractive index.
- Thus, a small change in the refractive index of the ENZ medium results in a much larger change in the refractive index of the coupled system due to the ultrafast red-shift of the resonance of the coupled system.

What about the maximum change in the index?

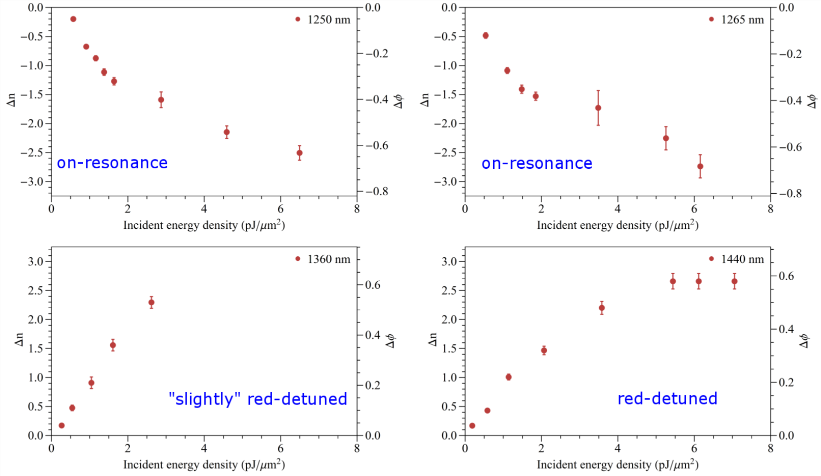


Figure 9: The change in the effective refractive index can be as larger than 2.5 over a broad spectral range.

Summary

- A broadband nonlinear material with n_2 values upto 7 order 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 disigning the geometric parameters of the antenna appropriately.

Thank You! Questions?