Interaction of ultrashort laser pulses with epsilon-near-zero materials

Robert W. Boyd

Department of Physics and
Max-Planck Centre for Extreme and Quantum Photonics
University of Ottawa

The Institute of Optics and
Department of Physics and Astronomy
University of Rochester

Department of Physics and Astronomy
University of Glasgow

Presented at the SPIE Defense + Commercial Sensing Conference, Anaheim, CA, USA, April 11, 2017.

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.

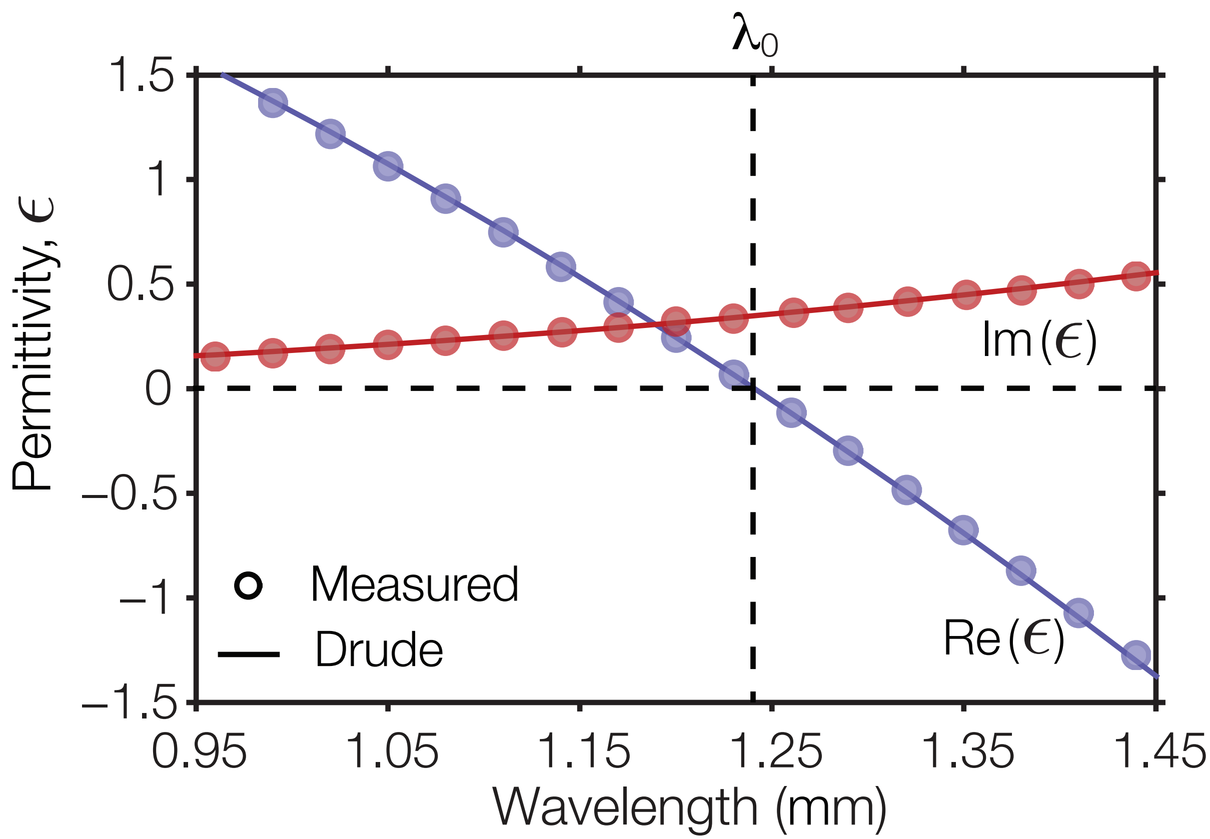
There has been great recent interest in studies of ENZ phenomena:

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

$\epsilon_\infty = 3.77$

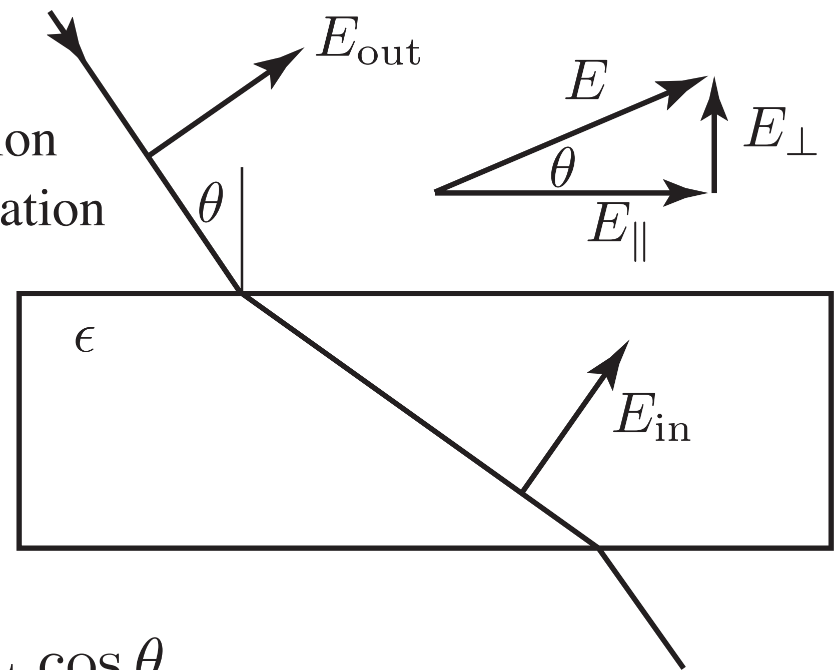
$\gamma = 0.0468\ \omega_p$

$\omega_p/2\pi = 473$ THz

Note that $\mathrm{Re}(\epsilon)$ vanishes at 1.24 mm, but that the loss-part $\mathrm{Im}(\epsilon)$ is non-zero.

The NLO Response Is Even Larger at Oblique Incidence

p polarization
TM polarization



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

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

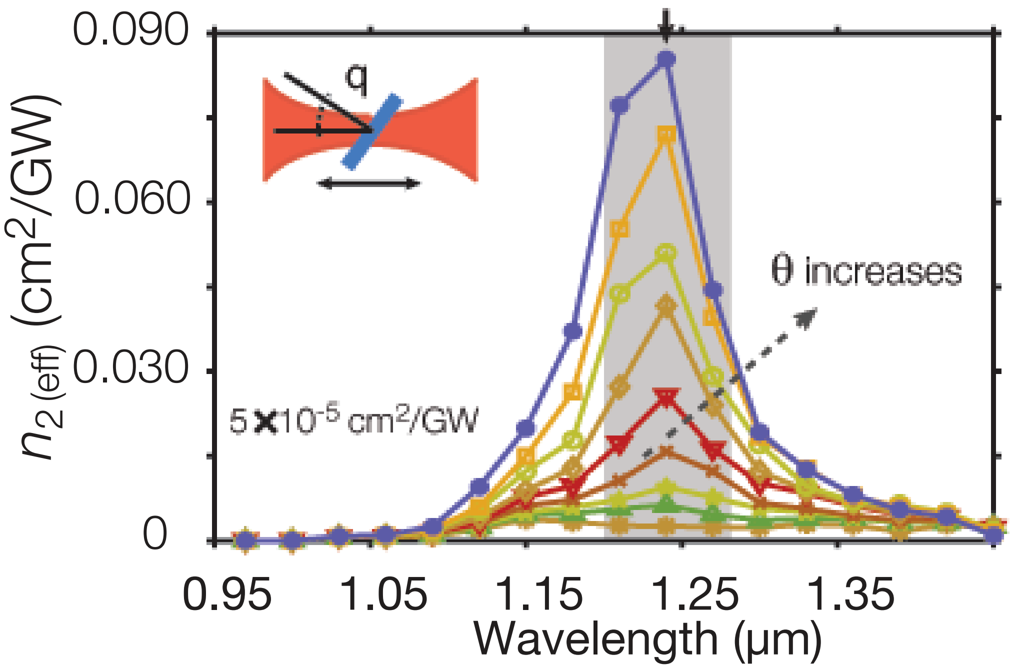
Note that, for $\epsilon < 1$, E_{in} exceeds E_{out} for $\theta \neq 0$.

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

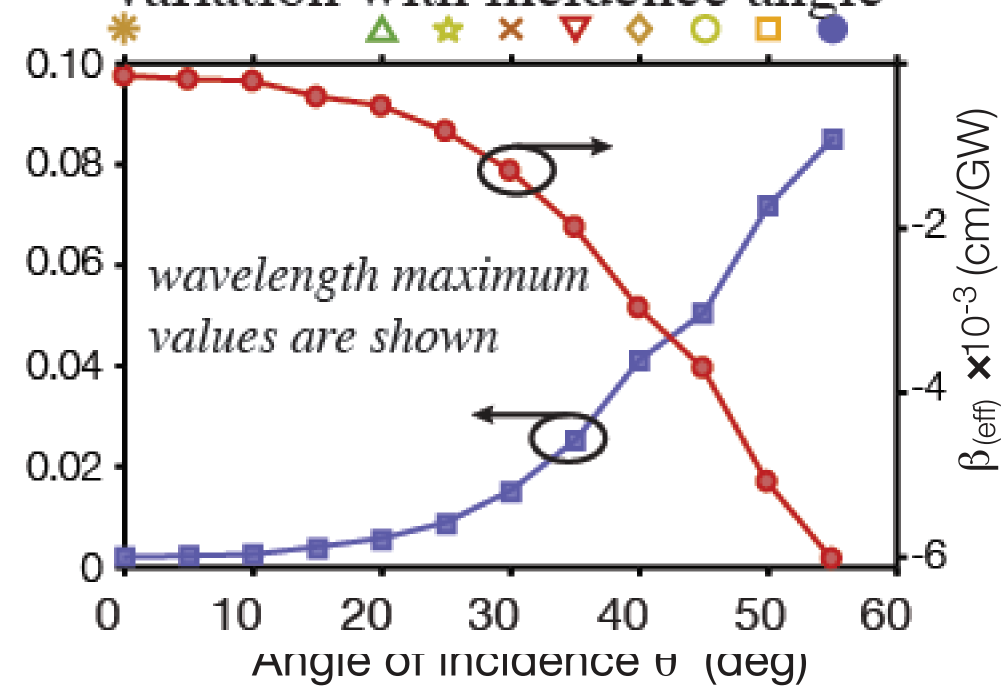
Huge Nonlinear Optical Response of ITO

- Z-scan measurements for various angles of incidence

Wavelength dependence of n_2



Variation with incidence angle

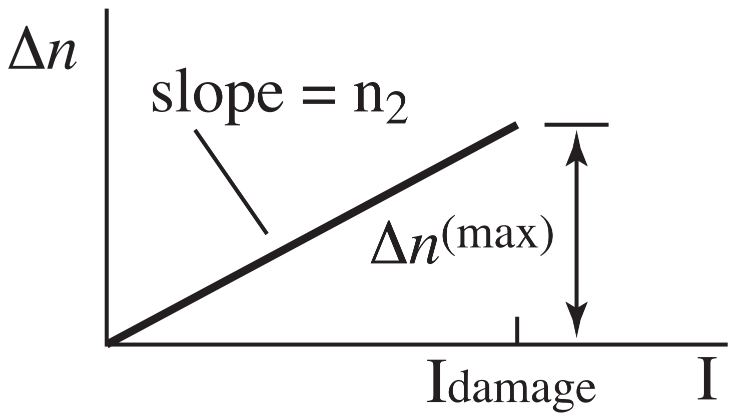


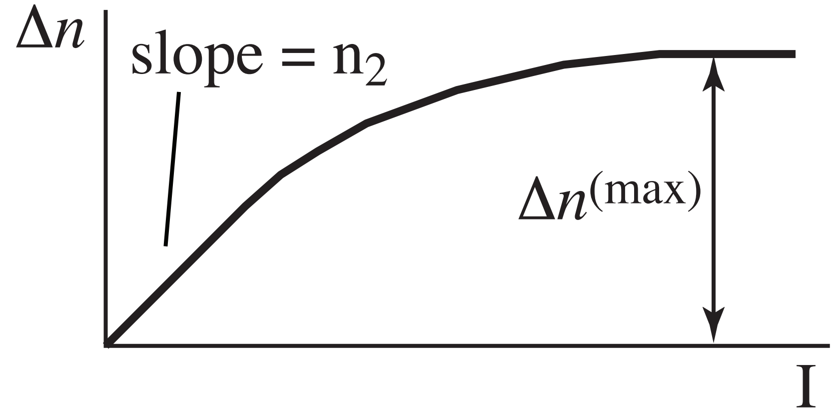
peak laser intensity was 50 GW cm-2

- 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 × 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 $\Delta n^{(\max)}$





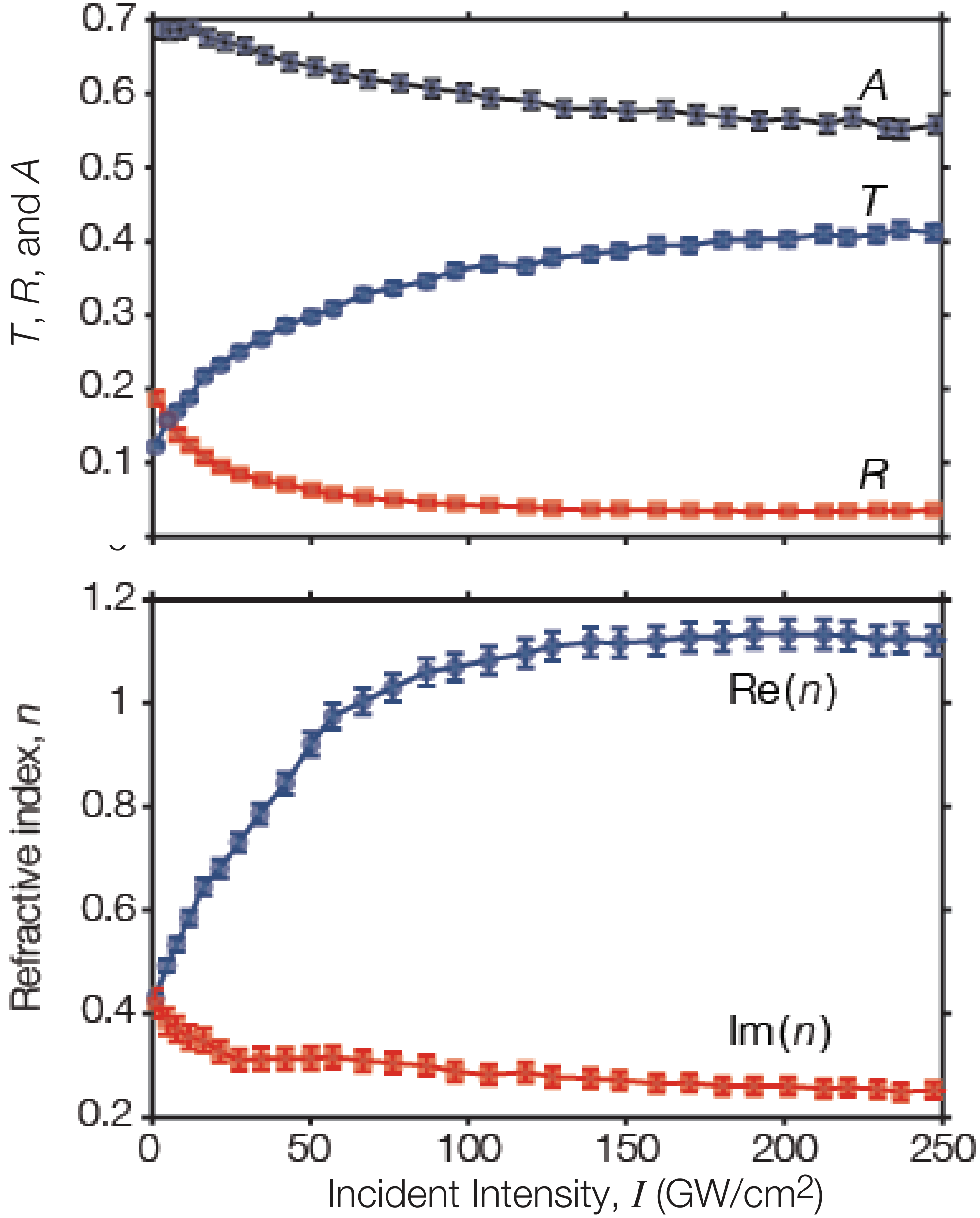
- For ITO at ENZ wavelength, both n_2 and $\Delta n^{(\max)}$ are extremely large
  ($n_2 = 1.1 \times 10^{-10}$ cm^2/W and $\Delta n^{(\max)} = 0.8$)

- 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.

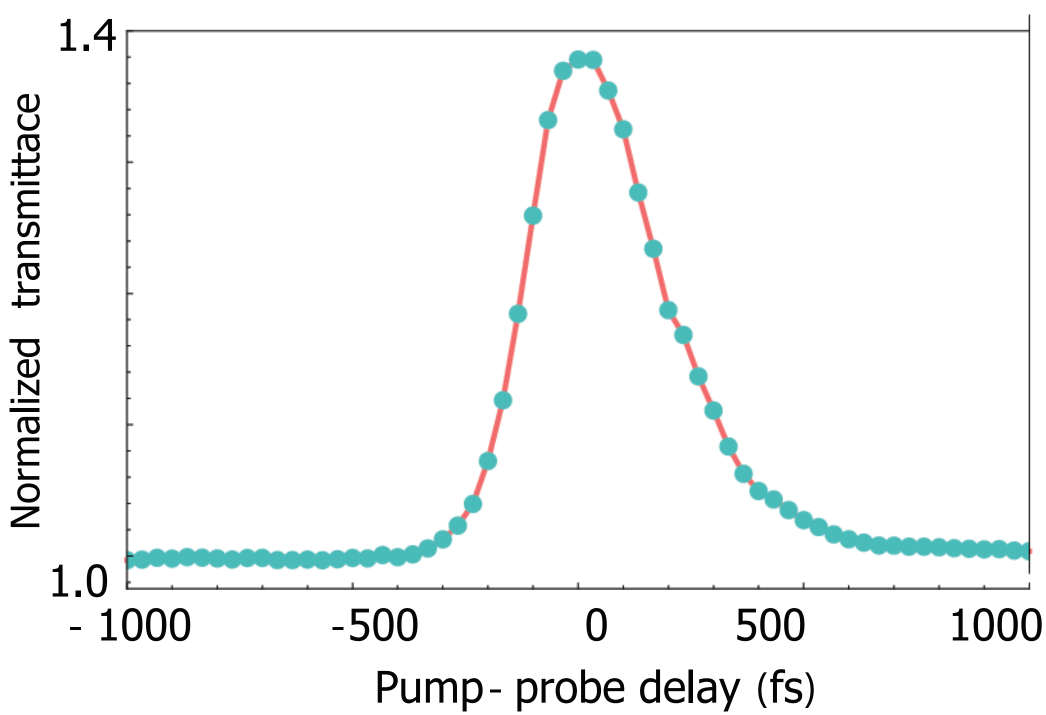
Here is the refractive index extracted from the above data.

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 x 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

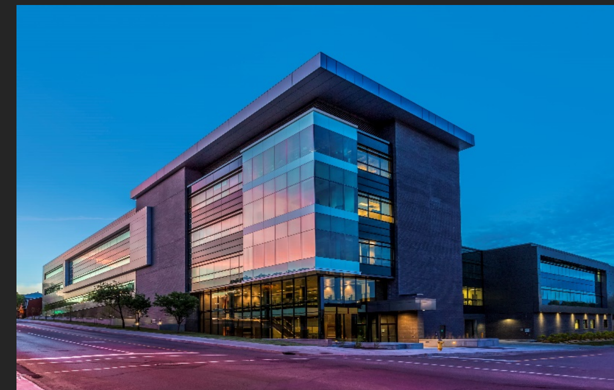
Other Reports of Highly Nonlinear Response in ENZ Material and Metamaterialc

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

M. Zahirul Alam[1], S. A. Schulz[1], J. Upham[1], I. De Leon[1,2] and R. W. Boyd[1,3]

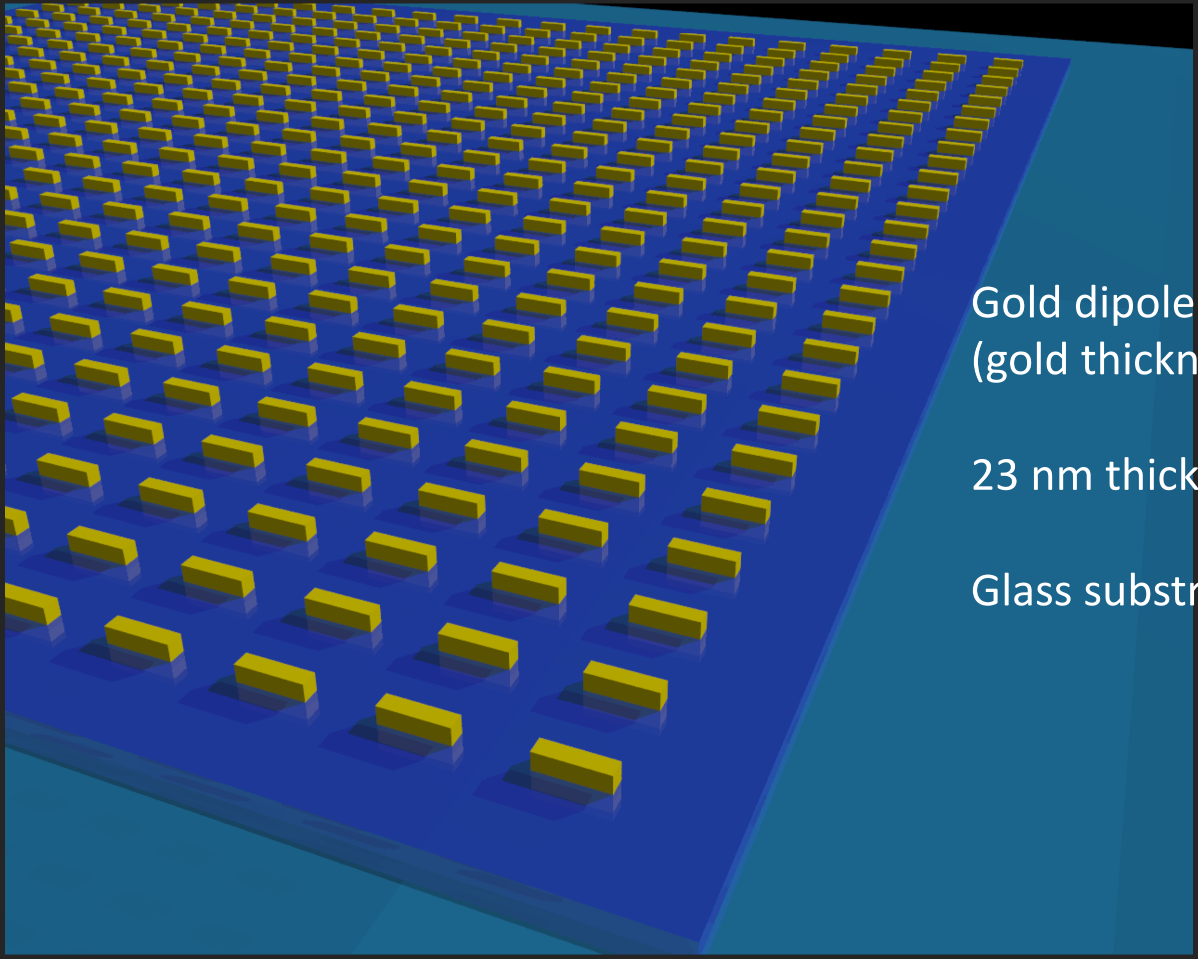
[1]Department of Physics and Max Planck Centre for Extreme and Quantum Optics, University of Ottawa, Canada.

[2]School of Engineering and Sciences, Tecnológico de Monterrey, Mexico.

[3]Institute of Optics, University of Rochester.

Max Planck – University of Ottawa Centre for Extreme and Quantum Photonics

The Structure

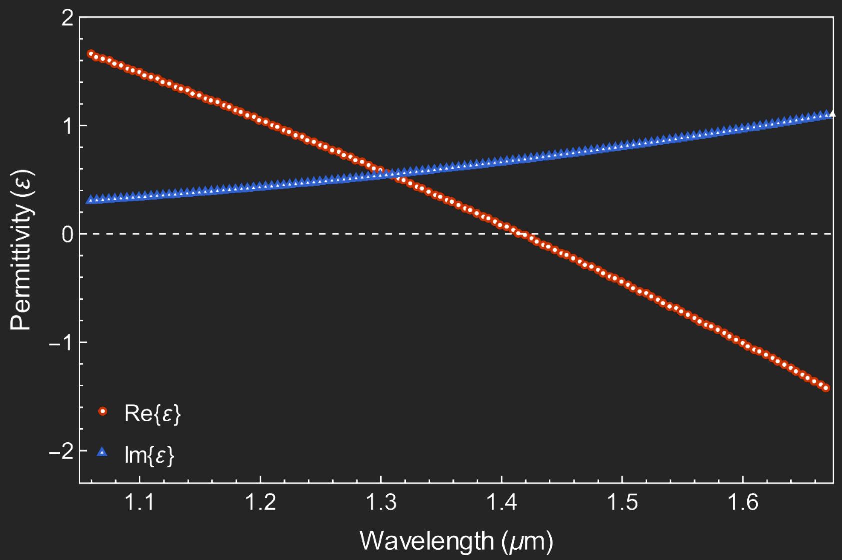


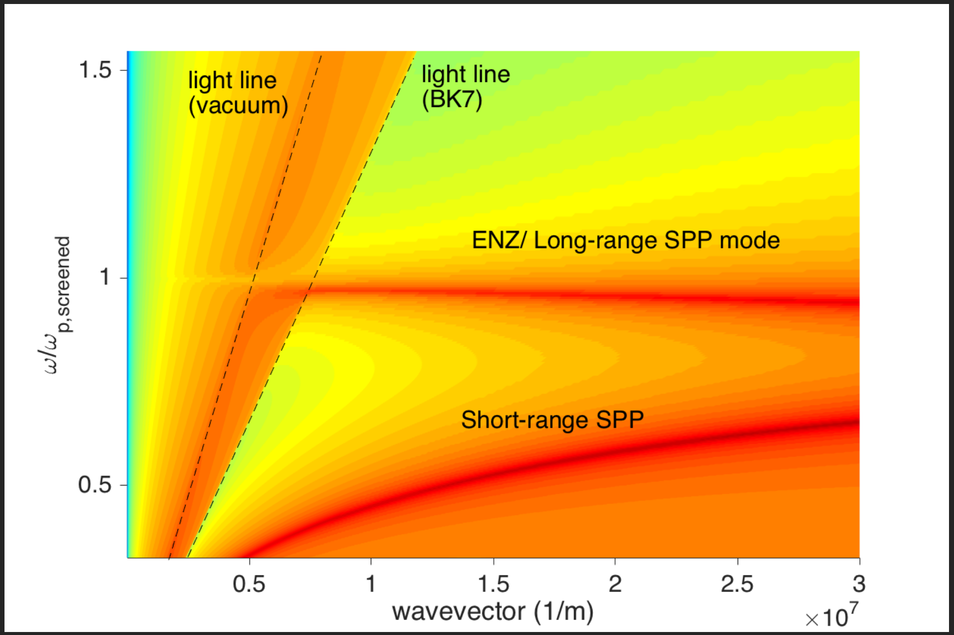
Gold dipole antenna array
(gold thickness 27 nm)

23 nm thick ITO

Glass substrate

Our 23-nm-thick ITO layer has zero real permittivity at $\lambda = 1420$ nm.



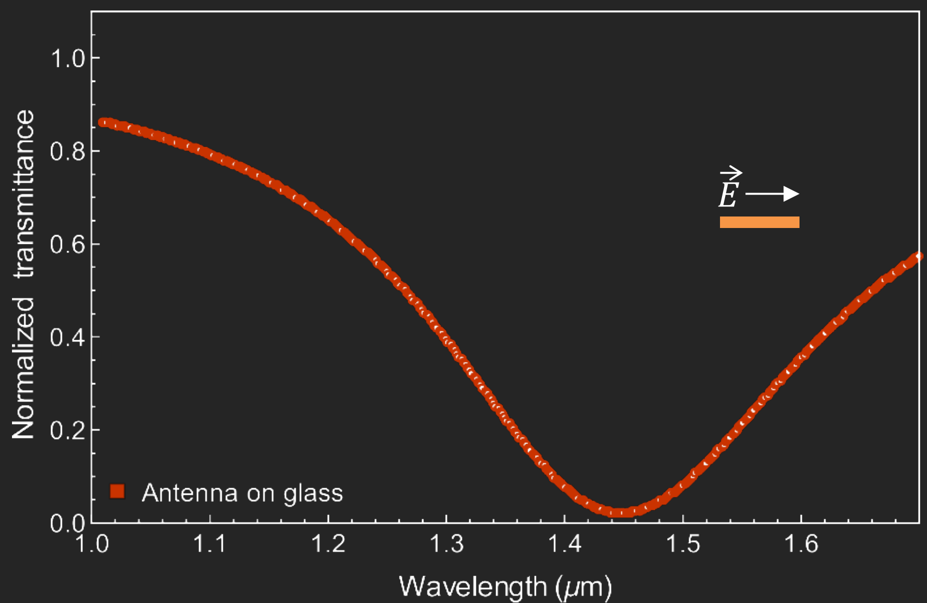


- The ENZ wavelength is tailorable

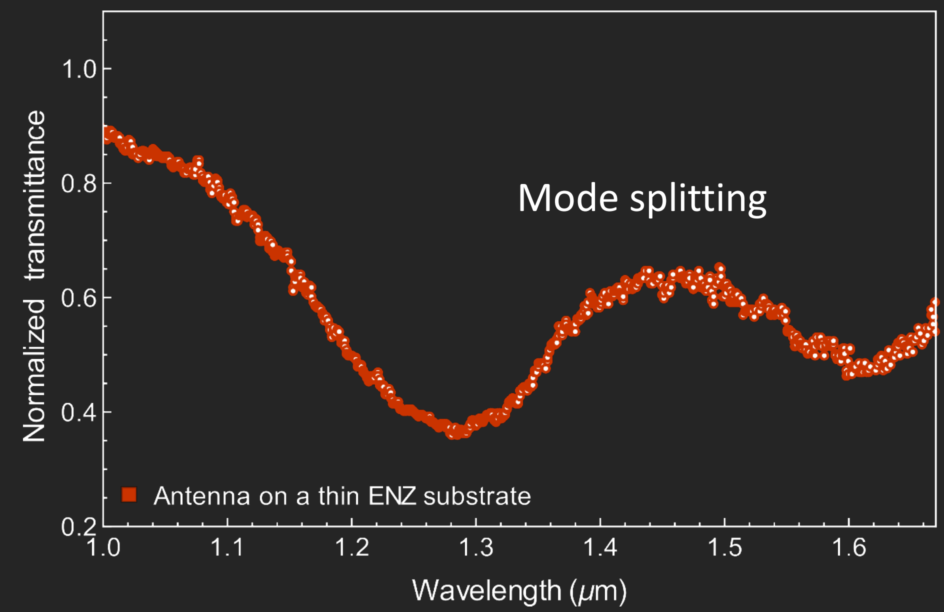
See also works by Engheta, Alù, Boltasseva, Shalaev, Muskens,Atwater, Zheludev, Zayats, etc.

Antenna array on a thin ENZ substrate

antenna array on a glass substrate (simulation)



antenna array on a 23 nm ITO-on-glass substrate (measured)



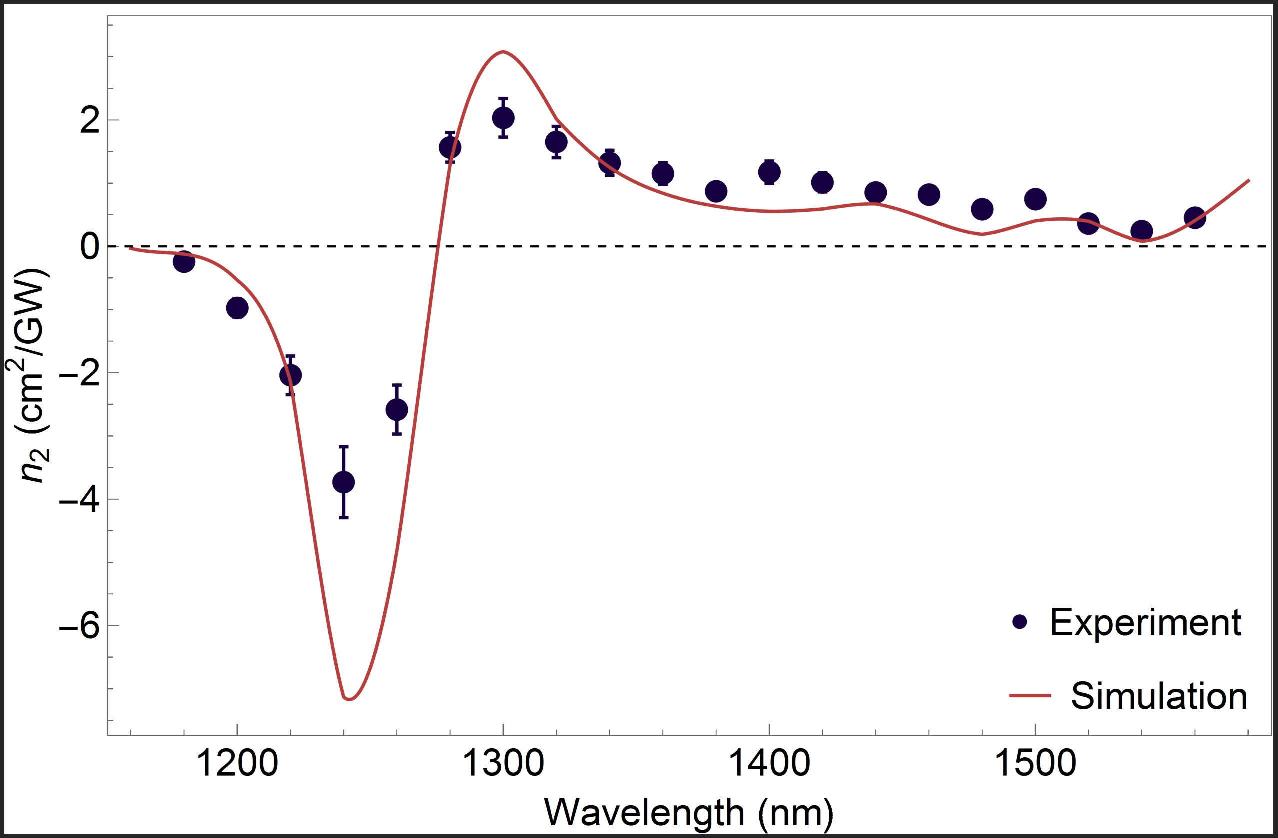
Gold dipole antenna on ITO

Dimensions of the antenna:

Length: 360 nm
Width : 110 nm
Thickness: 27 nm
Unit cell: 500 nm X 500 nm

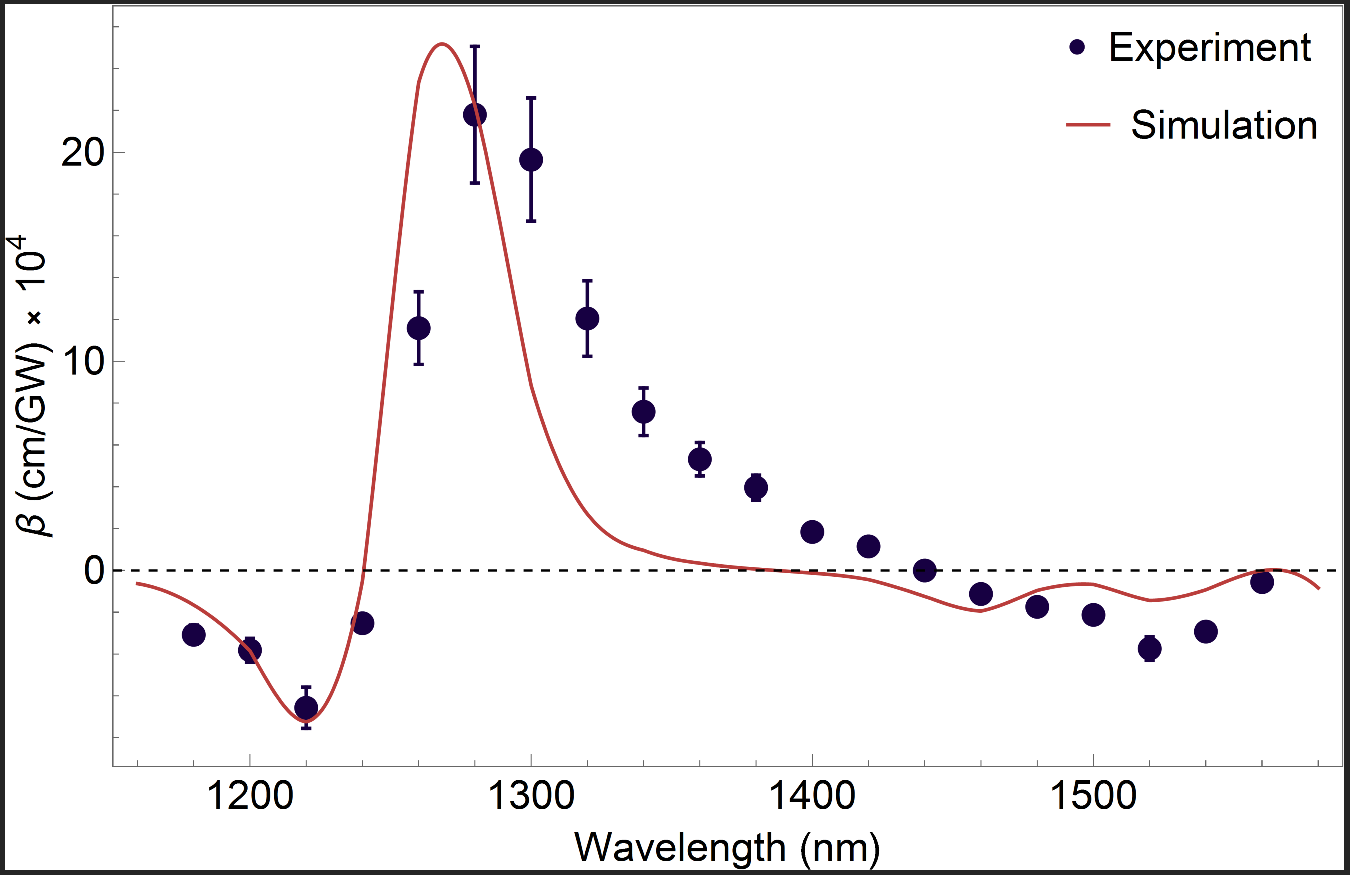
- Schulz et. al, Physical Review A. 93(6), 2016
- Campione et. al, ACS Photonics 3(2), 2016
- Jun et. al, Nano Lett. 13(11), 2013

We measured nonlinear refraction using the z-scan technique with ~ 140 fs pulse



Values are up to 7 orders of magnitude larger than that of silica glass and three orders of magnitude larger than the normal-incident ITO n_2 at ENZ ($2.58*10^{-3}$ cm^2/GW at normal incidence)

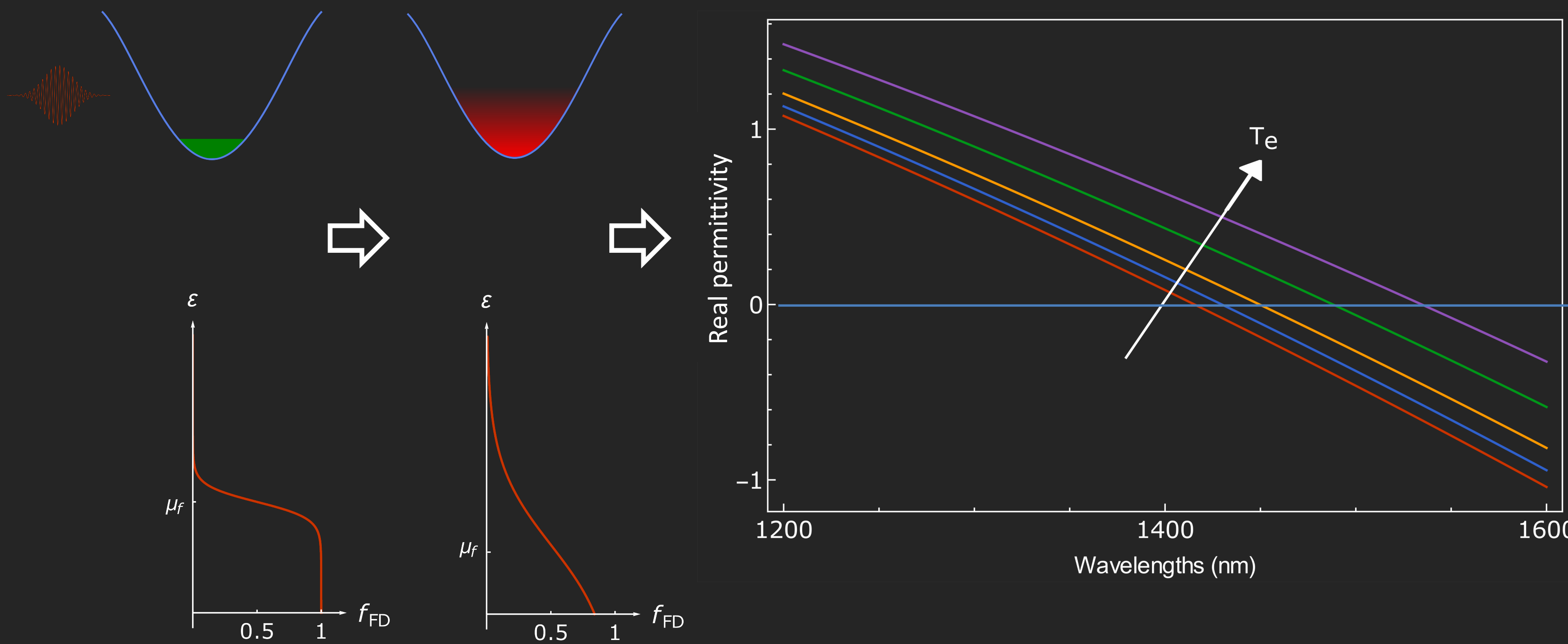
We measured nonlinear absorption using the z-scan technique with ~ 140 fs pulse



We measured wavelength dependent saturable and reverse-saturable absorption.

What is the origin of this large nonlinear response ?

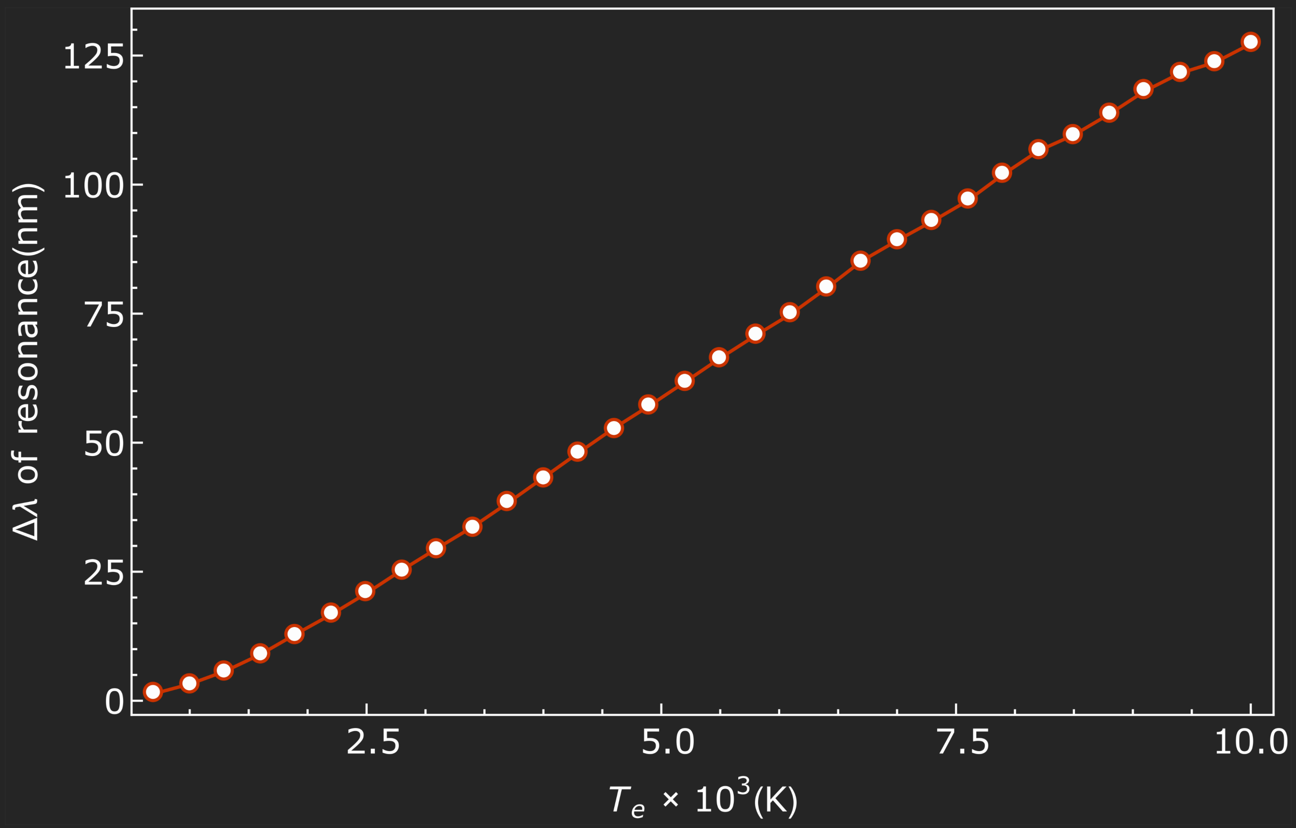
Mechanism of nonlinear response of ITO: a strong modification of the conduction band electron distribution results in ultrafast modification of the plasma frequency.



- Electronic heat capacity constant is an order of magnitude smaller than gold. Thus electrons can be heated more efficiently to a higher temperature.
- Pumping at ENZ results in highly non-equilibrium electron distribution and strong modification of the effective mass of the electrons due to conduction band nonparabolicity

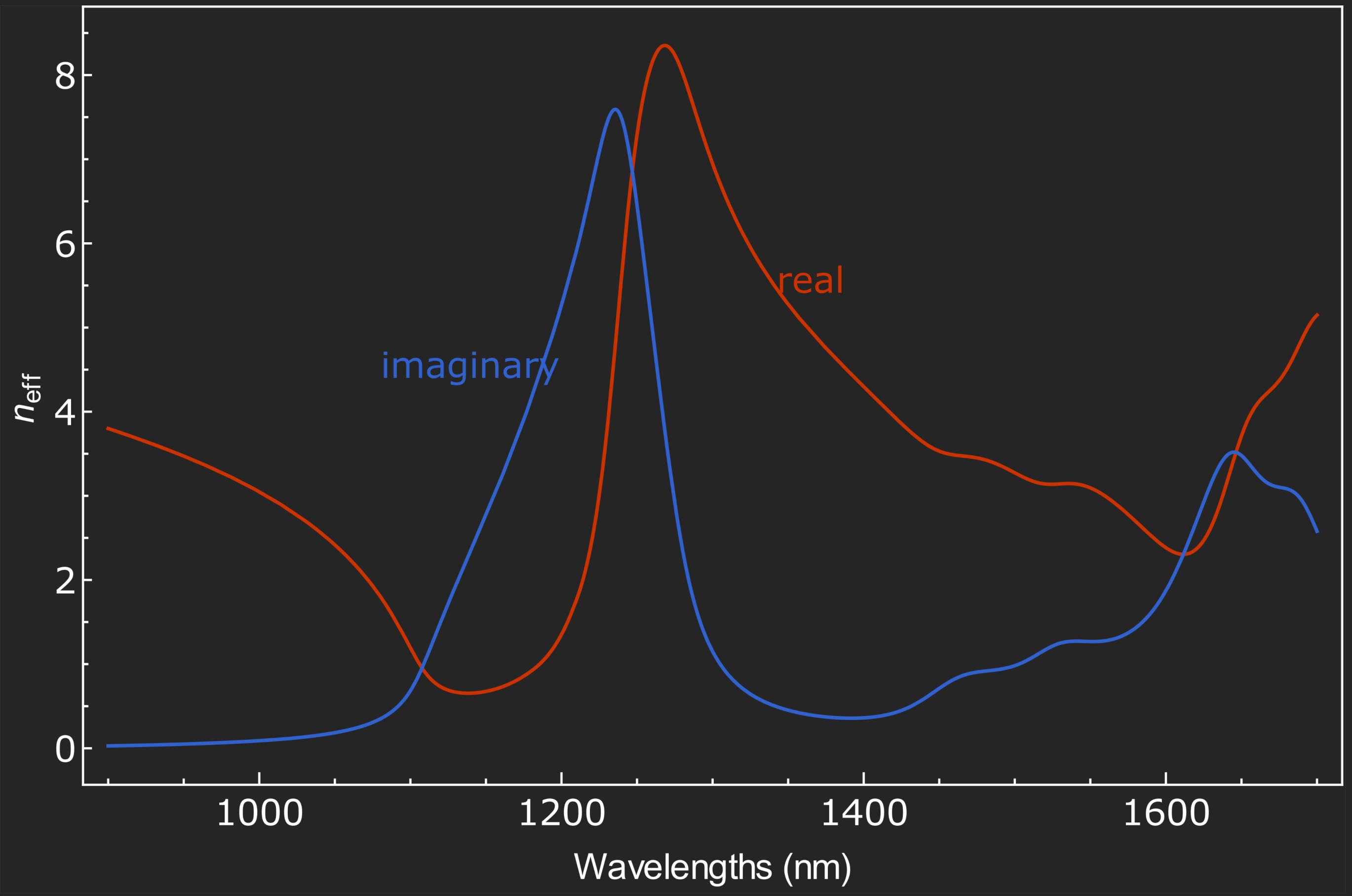
Mechanism of nonlinear response of ITO-antenna system:

a modification of the electron temperature in the ITO layer results in ultrafast red-shift of the resonance of the coupled system.



Mechanism of nonlinear response of ITO-antenna system:

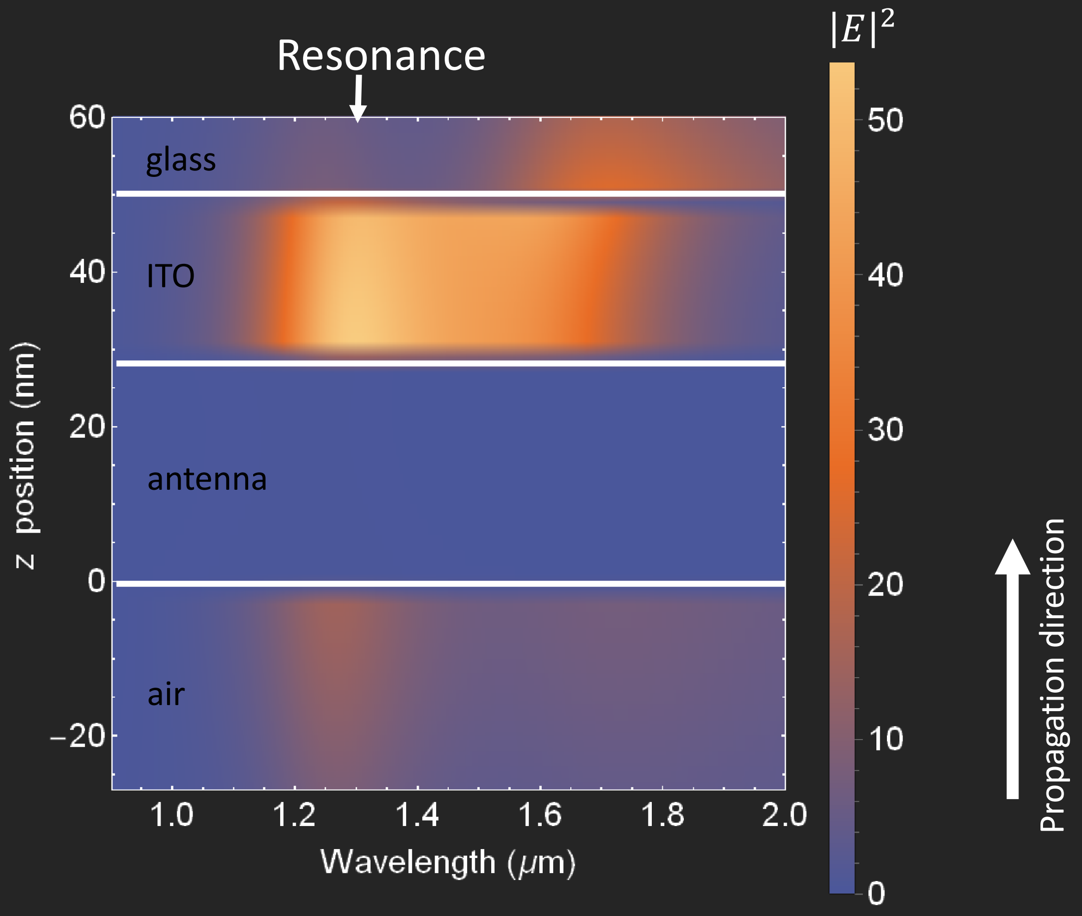
Time-dependent red-shift of the linear refractive index (due to the red-shift of the resonance wavelength) results in a large change in the refractive index.



Plot shows effective linear refractive index of the coupled ITO-antenna system.

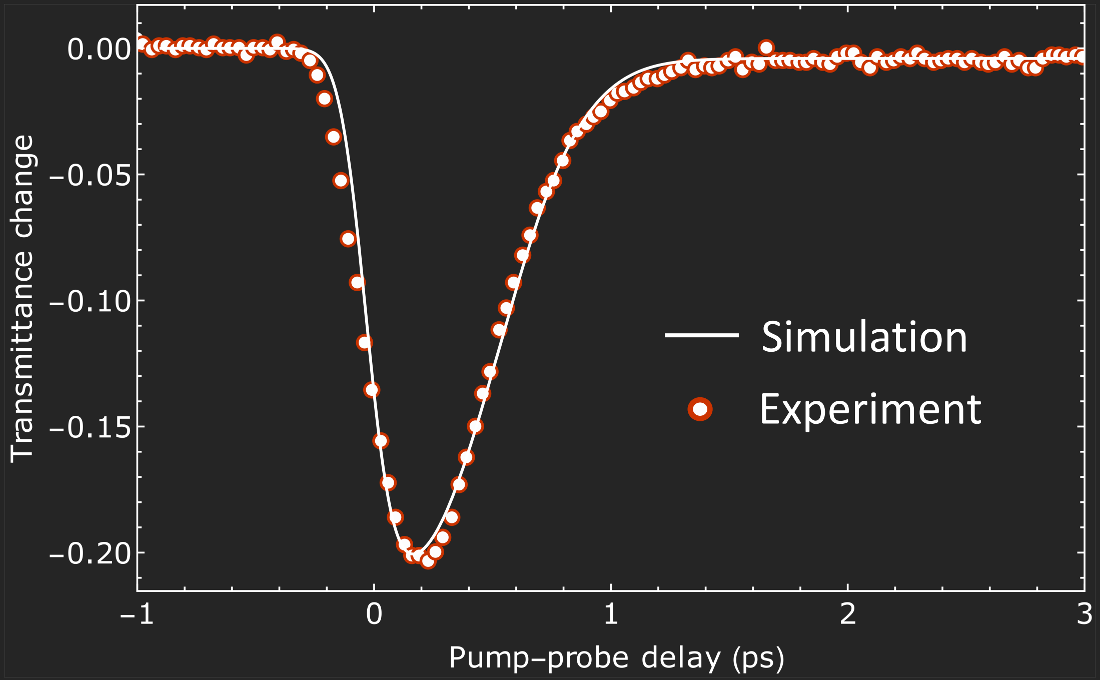
Refractive index homogenization procedure: Chen, Xudong, et al., PRE 70, 016608 (2004); Smith, D. R., et al., *PRB* 65, 195104 (2002)

Mechanism of nonlinear response of ITO-antenna system: antenna-mediated intensity enhancement in the ITO layer.



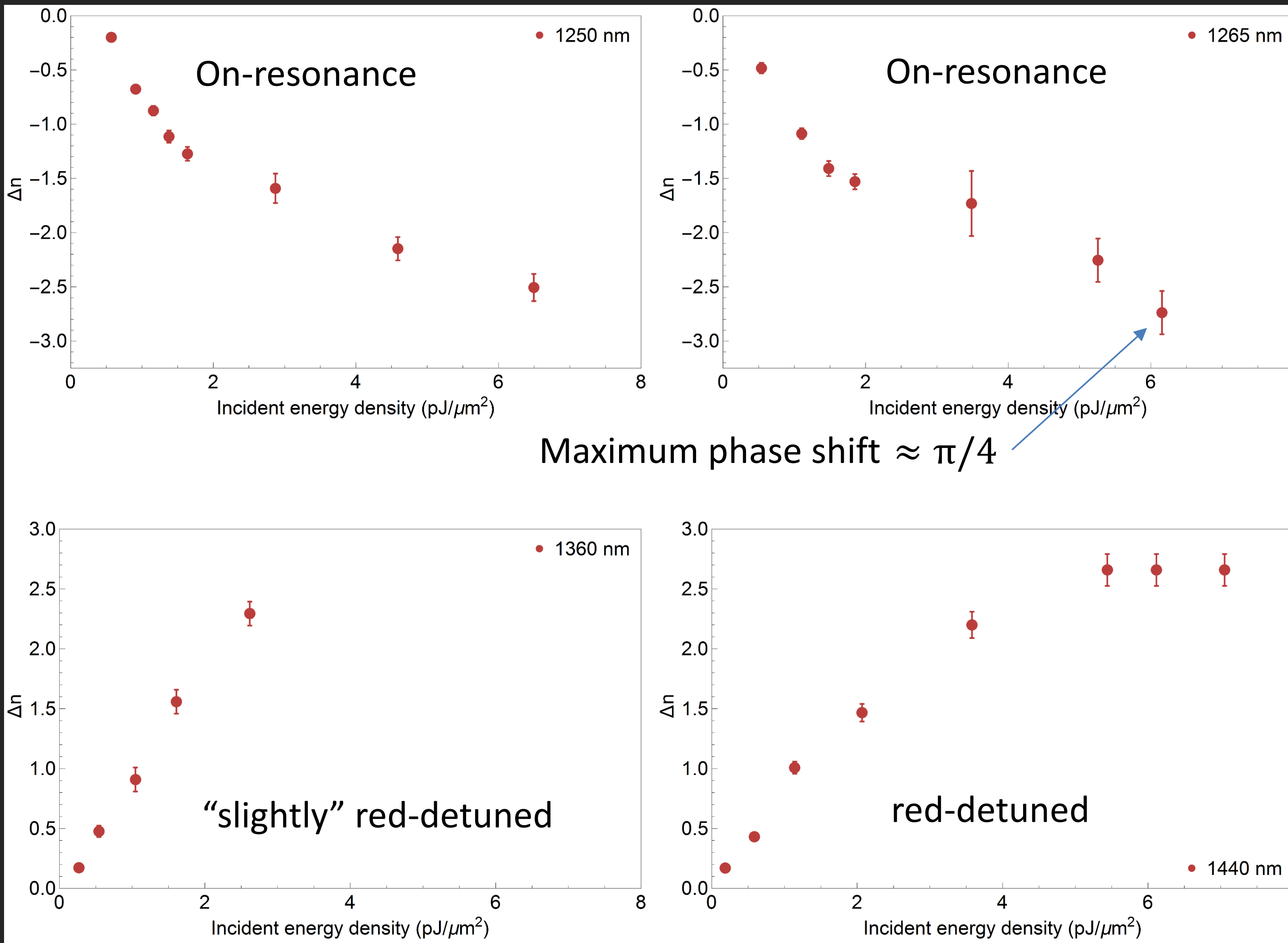
Pump-probe response at the resonance wavelength

- Recovery time: $\sim 1\text{ps}$ on-resonance (slower recovery time compared to ITO)
- Antennas' intrinsic nonlinearity is negligible.
- We use a two-temperature model to calculate the temporal response.



Nonlinear addition to the refractive index: experimental data

Material thickness is 50 nm



Improving the performance: how to achieve a π-radian phase shift with femtojoules of energy using a sub-100-nm-thick material.

- Increase the thickness.
- Substrate with larger electronic mobility and lower damping.
- Maximize the overlap of the antenna near-field with the substrate.
- Improve the antenna design to tailor the linear dispersion.
- CMOS compatible design using all-dielectric materials.

What is the source of the large nonlinear response?

- Large intrinsic nonlinearity of ITO at ENZ wavelengths
- Strong interaction between antenna and the ITO substrate
- Field enhancement
- Ultrafast nonlinear modification of the plasma frequency

Take home messages:

- We have introduced new design principles to engineer materials with large nonlinear response.
- $|\Delta n| > 2.5 \quad n_2 > 10^7 \, n_{2,glass}$
- Phase shift of $\pi/4$ using a 50 nm thick structure.
- Implications in nanophotonics in general and active metasurface design.

Thank You !

Sebastian Schulz

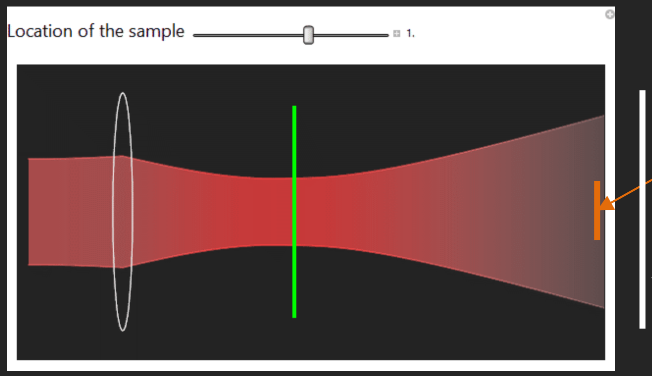
Jer Upham

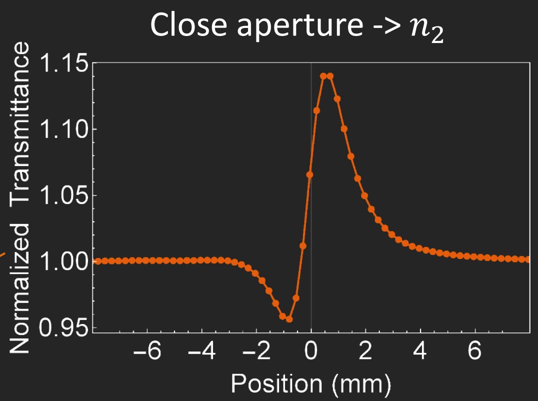
Israel de Leon

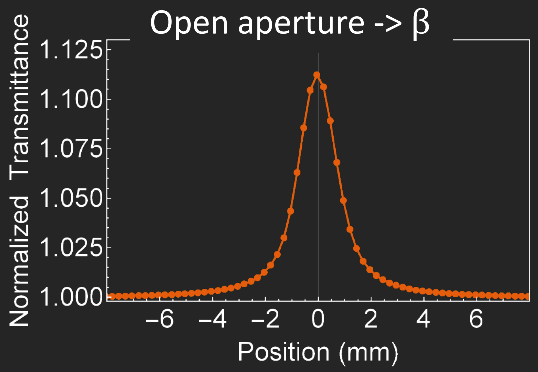
M. Zahirul Alam

Max Planck - University of Ottawa Centre
for Extreme and Quantum Photonics

Z-scan technique maps intensity dependent phase to far-field intensity variation







150 fs pulse; rep rate 1KHz

See works by Sheik-Bahae and van Stryland

Mechanism of nonlinear response of ITO : total nonlinear Contribution to refractive index for electronic temperature of 10^4K