Large Optical Nonlinearity of Indium Tin Oxide in its Epsilon-Near-Zero Spectral Region

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 47th Winter Colloquium on the Phyics of Quantum Electronics, Snowbird, Utah, USA January 8-13 2017

Large Nonlinear Optical Response of Intium Tin Oxide in its Epsilon-Near-Zero Spectral Region

OUTLINE

1. Highly nonlinear response of ITO
2. Radiative properties of ENZ materials
3. Breakout session

With special thanks to
Ksenia Dolgaleva, Peter Milonni, John Sipe, Zahirul Alam,
Israel De Leon, Orad Reshef, and Eric Mazur.

New Nonlinear Optical Material for Quantum Technologies

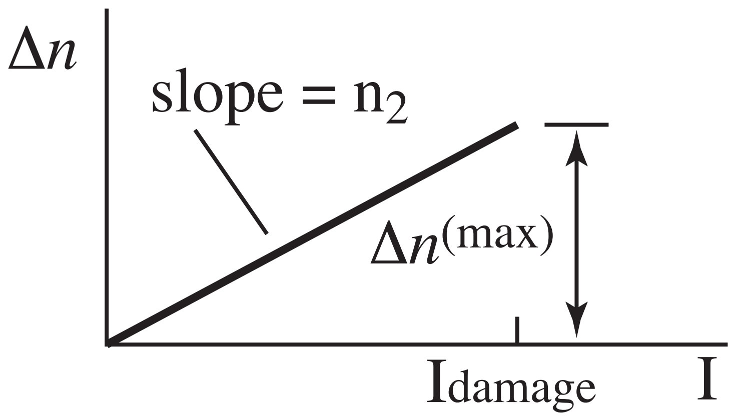
- We want all-optical switches that work at the single-photon level
- We need photonic materials with a much larger NLO response
- We recently reported a new NLO material with an n_2 value 100 times larger than any previously reported results (but with some background absorption).
- A potential game changer for the field of photonics

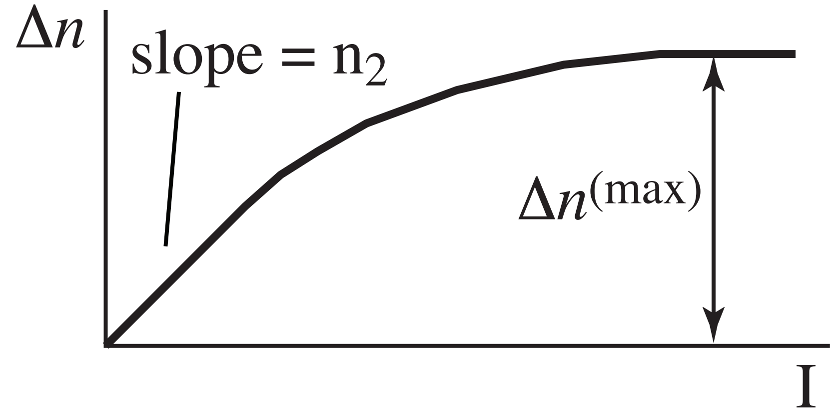
Large optical nonlinearity of indium tin oxide in its epsilon-near-zero region, M. Zahirul Alam, I. De Leon, R. W. Boyd, Science 352, 795 (2016).

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 $\Delta n^{(max)}$





- We report a material for which both n_2 and $\Delta n^{(max)}$ are extremely large

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

  (For silica glass $n_2 = 3.2 \times 10^{-16}$ cm^2/W, $I_{damage} = 1$ TW/cm^2, and thus $\Delta n^{(max)} = 3 \times 10^{-4}$)

- Thus 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

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

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.

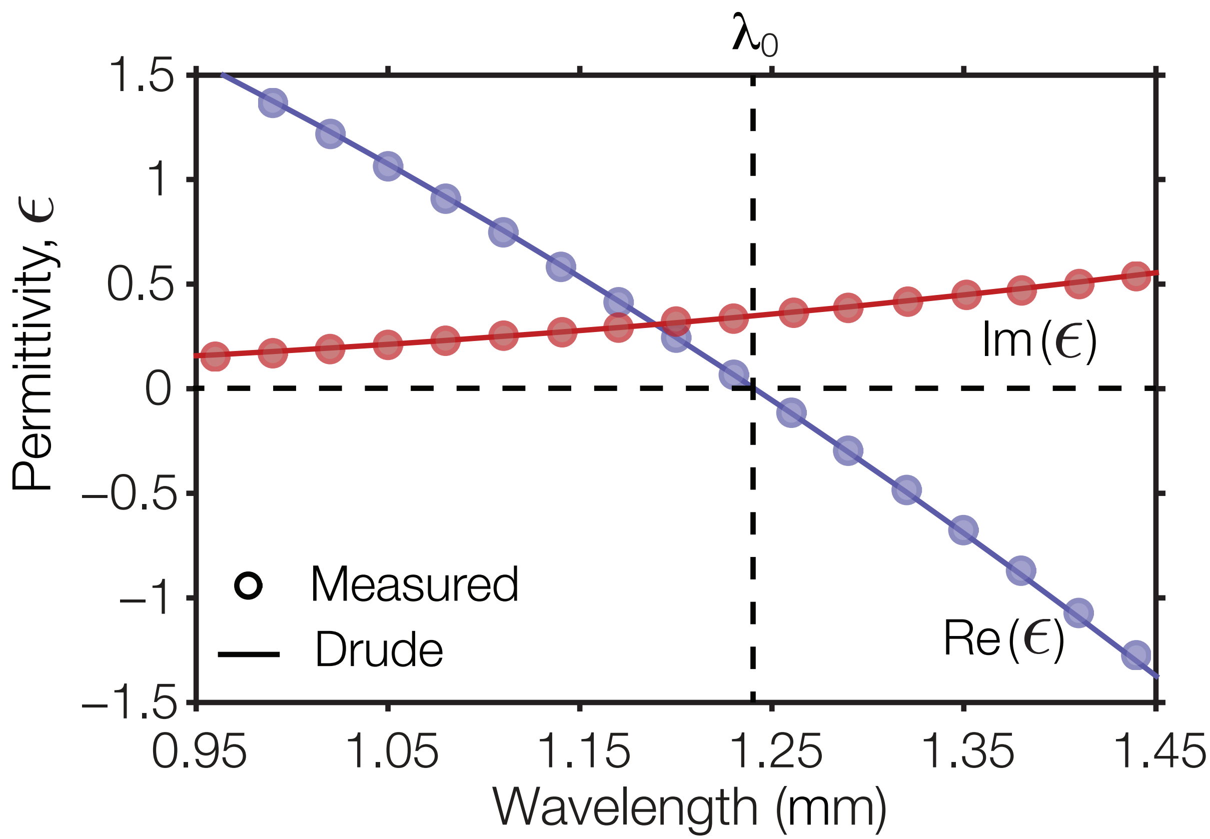
There has been great recent interest in studies of ENZ phenomena:

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.

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

Why Does ENZ Lead to Large NLO Response?

+

Simple Math:

$$\epsilon = \epsilon_b + \Delta\epsilon \quad (\text{b} = \text{“bulk”})$$
$$n = \sqrt{\epsilon} = \sqrt{\epsilon_b + \Delta\epsilon}$$

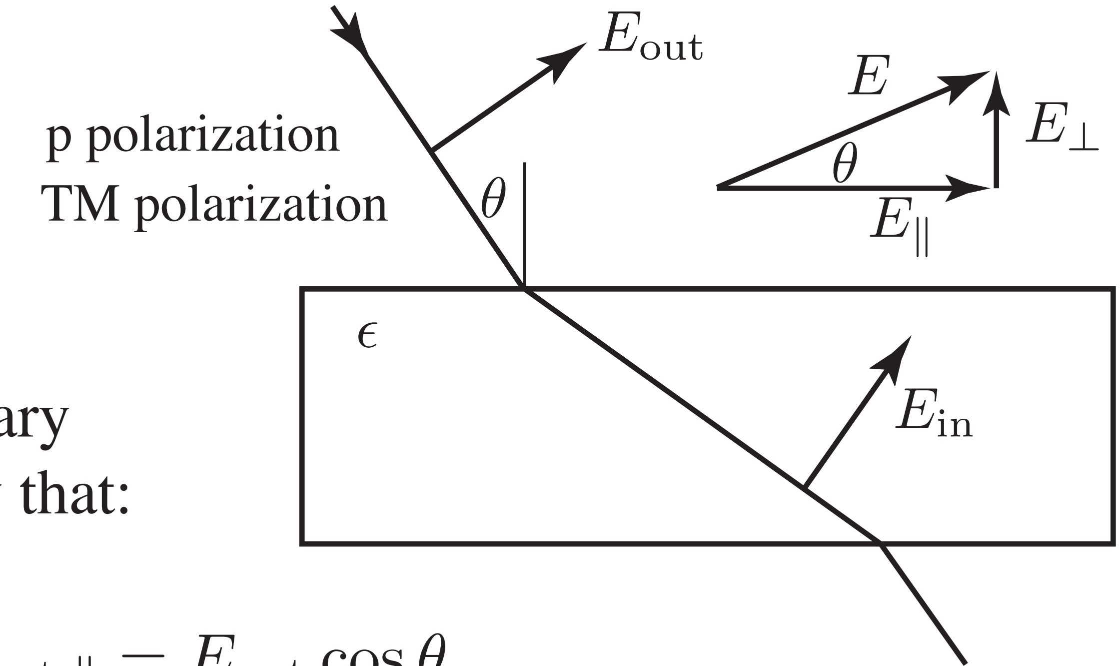
Assume $\Delta\epsilon \ll \epsilon_b$ (this assumption can be violated).

$$n = \sqrt{\epsilon_b}\left(1 + \frac{\Delta\epsilon}{2\epsilon_b} + \cdots\right) = \sqrt{\epsilon_b} + \frac{\Delta\epsilon}{2\sqrt{\epsilon_b}}$$

or

$$n = n_b + \Delta n \quad \text{where} \quad \Delta n = \frac{\Delta\epsilon}{2n_b}$$

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

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

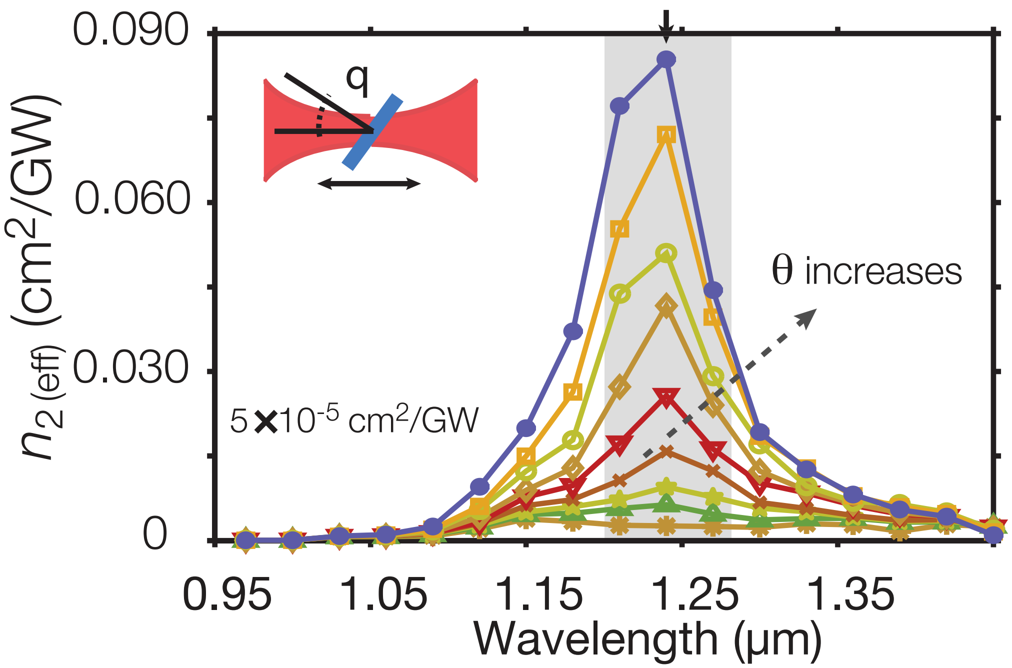
Note that, for $\epsilon < 1$, E_{in} exceeds E_{out} for $\theta \neq 0$.

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

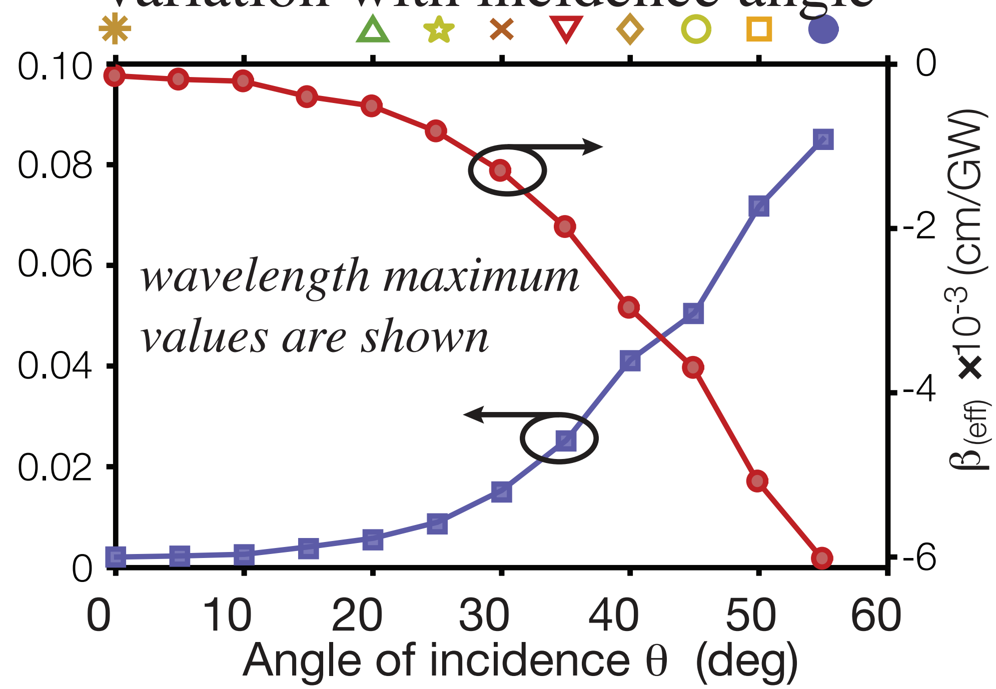
Huge Nonlinear Optical Response of ITO

- Z-scan measurements for various angles of incidence

Wavelength dependence of n_2



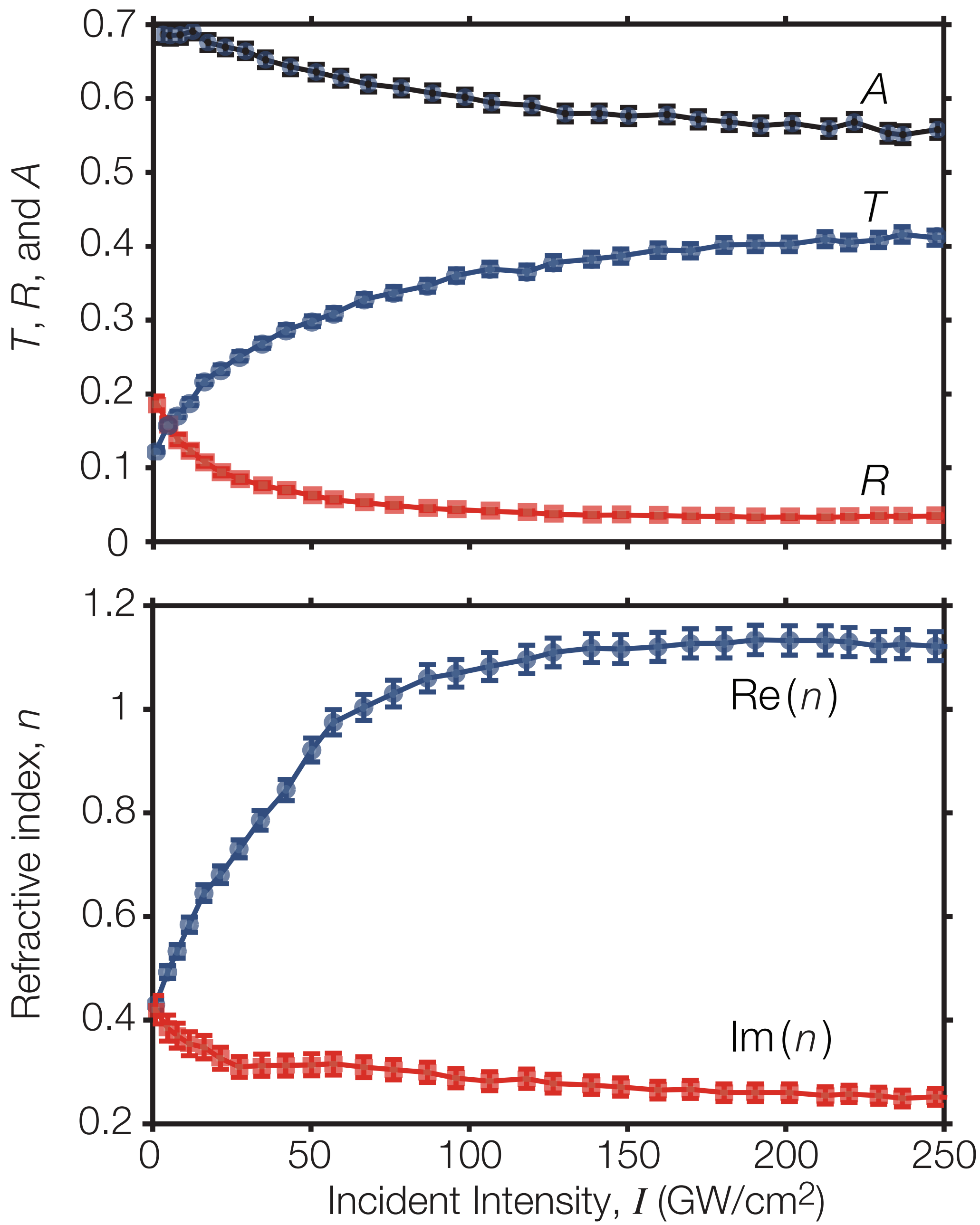
Variation with incidence angle



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.

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.

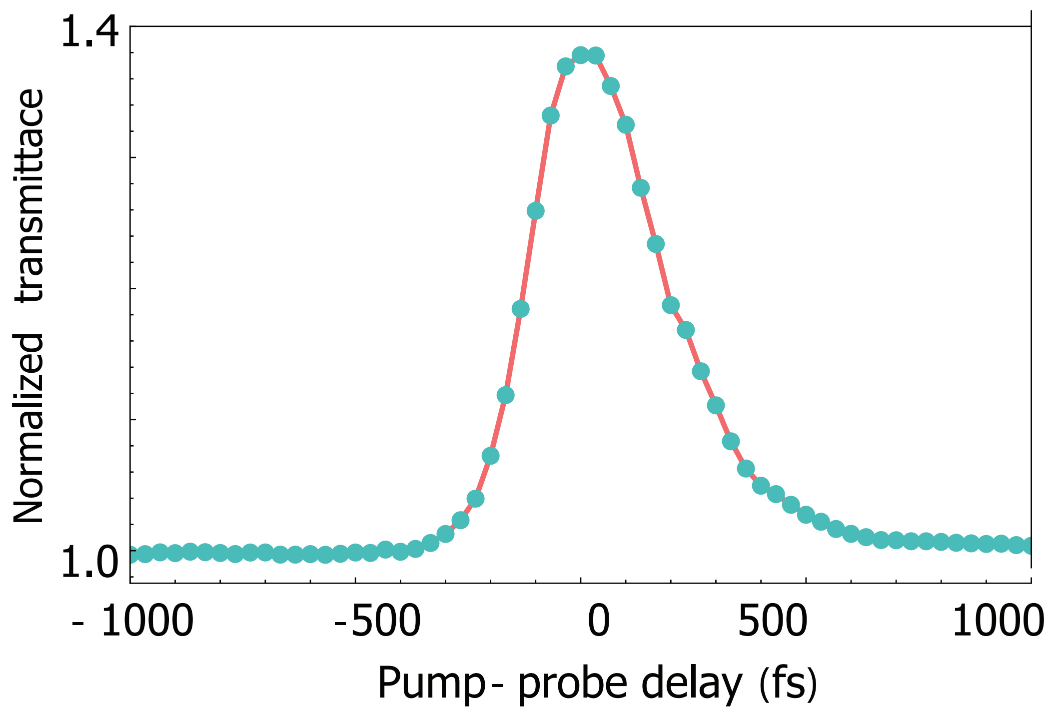
Here is the refractive index extracted from the above data.

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).

Radiative Properties of ENZ Materials

Recall that $n = \sqrt{\epsilon}$.
Thus $\epsilon = 0$ implies that $n = 0$

Since $\lambda = \lambda_{\text{vac}}/n$, we see that $\lambda = \infty$.
Thus the light field oscillates in time but is stationary in space.

Moreover, the phase velocity of light, $v = c/n$ becomes infinite.

Recall Poynting's Theorem

$$\langle \mathbf{S} \rangle = 2n\sqrt{\epsilon_0/\mu_0}\,|E_0|^2\hat{\mathbf{k}} = 2n\epsilon_0 c|E_0|^2\hat{\mathbf{k}},$$

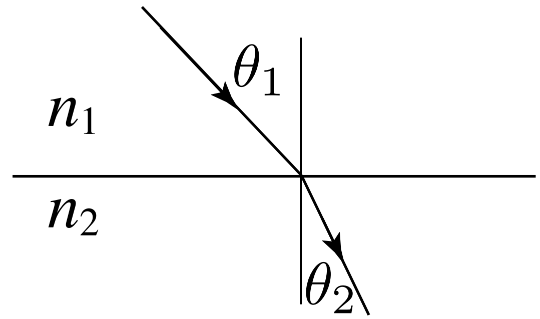
$$\langle u \rangle = 2n^2\epsilon_0|E_0|^2,$$

We see that the intensity (magnitude of Poynting vector) and the energy density vanish inside an ENZ material.

Snell's Law for ENZ Materials

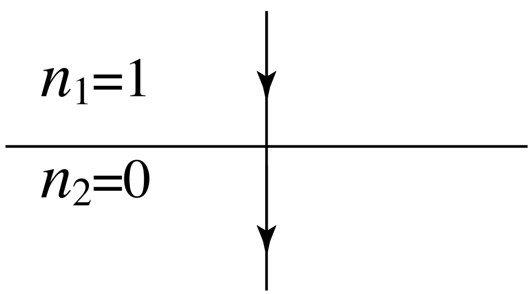
Snell's Law

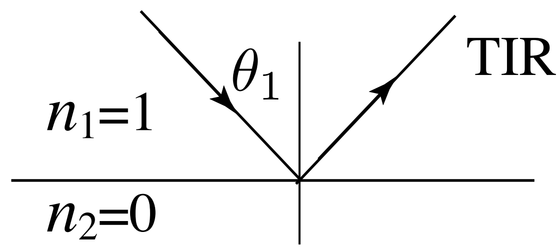
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$



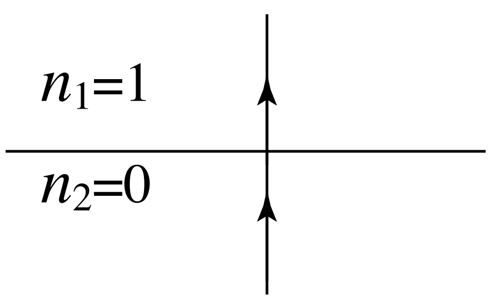
For an air-ENZ interface, n_1 =1 and n_2 =0.

Thus $\sin\theta_1 = 0$, and the only solution is $\theta_1 = 0$.

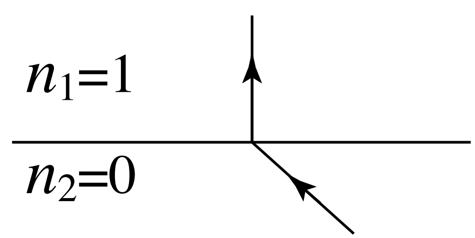




Light can exit an ENZ material only perpendicular to a boundary



or



Einstein A and B Coefficients in an ENZ Material

- The free-space atomic decay rate (Einstein A coefficent) is given by

$$\Gamma_0 = \frac{\mu^2 \omega_A^3}{3\pi c^3 \hbar \varepsilon_0},$$

In a dielectric material, not necessarily lossless, the decay rate becomes

$$\Gamma^{rad} = Re\{n(\omega_A)\}\Gamma_0.$$

(We have ignored local-field effects.)

Note that spontaneous emission cannot occur for a material with n=0.

- The Einstein B coefficient in a dispersionless material is given by

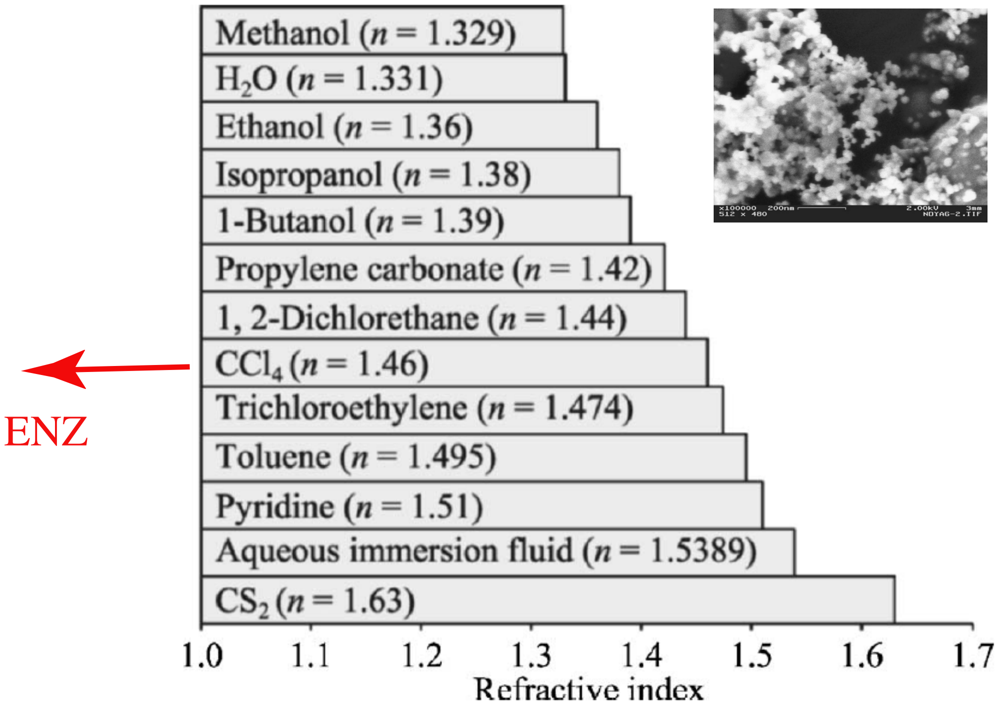
$$B = \frac{1}{n^2} B_{\mathrm{vac}}$$

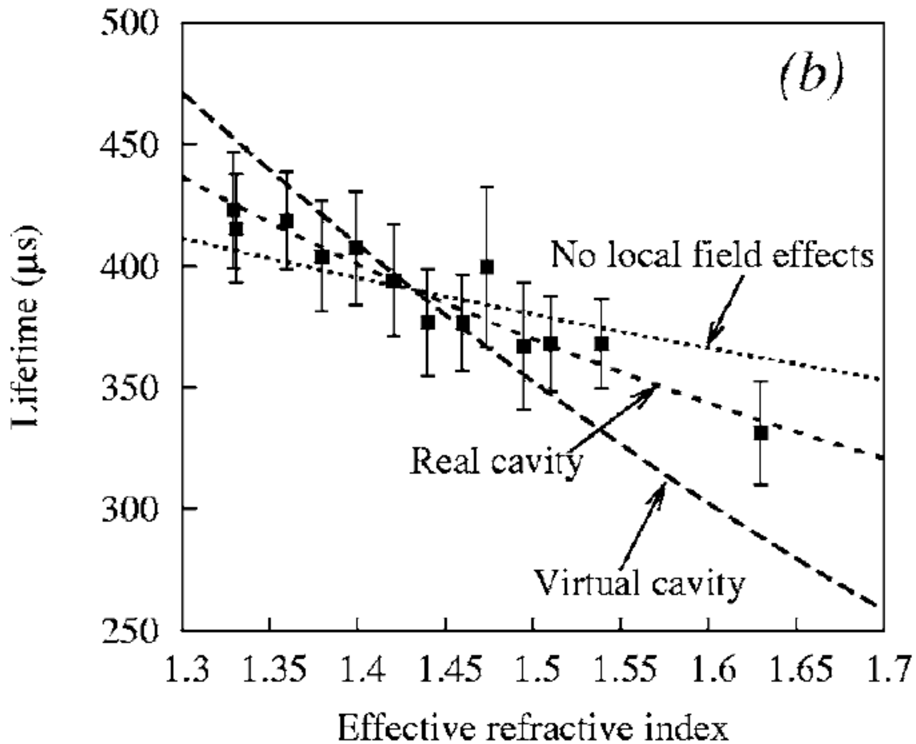
This expression diverges for $n = 0$, but we should recall that the energy density vanishes under ENZ conditions.

P. Milonni, J. Modern Optics 42, 1991 (1995).
S.M. Barnett, B. Huttner, and R. Loudon Phys. Rev. Lett. 68, 3698 (1992).

Refractive Index Dependence of the Radiative Lifetime

Influence of local-field effects on the radiative lifetime of liquid suspensions of Nd:YAG nanoparticles, K. Dolgaleva and R.W. Boyd and P.W. Milonni, J. Opt. Soc. Am. B 24, (2007).





virtual cavity model = Lorentz model

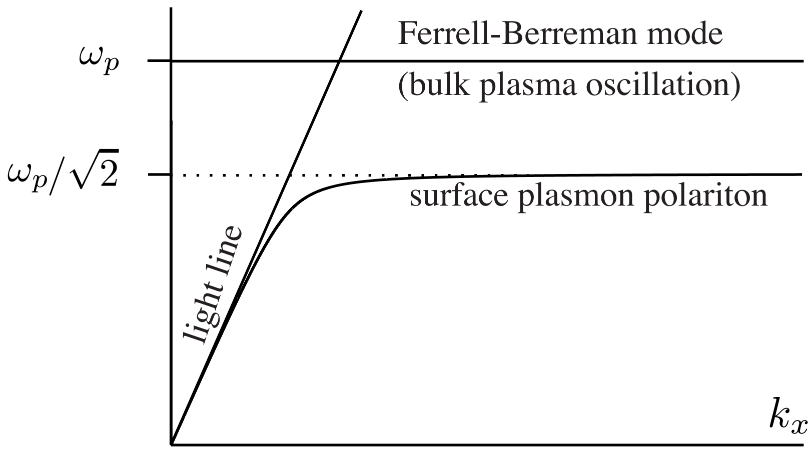
$$\tau_{\text{rad}} = \frac{\tau_{\text{rad}}^{(\text{vac})}}{n_{\text{eff}}\left(\dfrac{n_{\text{eff}}^2+2}{3}\right)^2}$$

real cavity model = Glauber-Lewenstein model

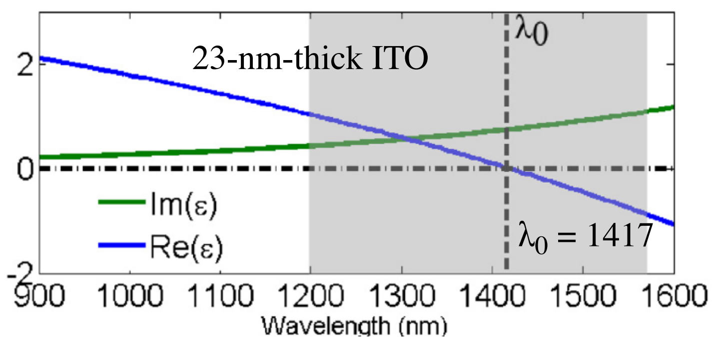
$$\tau_{\text{rad}} = \frac{\tau_{\text{rad}}^{(\text{vac})}}{n_{\text{eff}}\left(\dfrac{3n_{\text{eff}}^2}{2n_{\text{eff}}^2+1}\right)^2}$$

Optical Response of Dipole Antennas on an Epsilon-Near-Zero Substrate

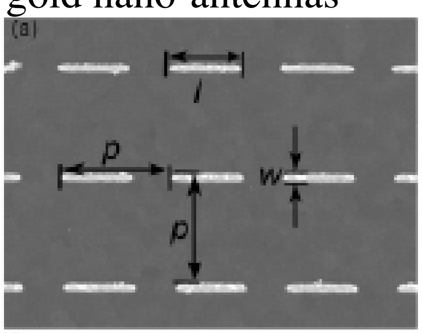
- Dispersion relations for the modes of a thin film



- Ellipsometry results for our ITO sample

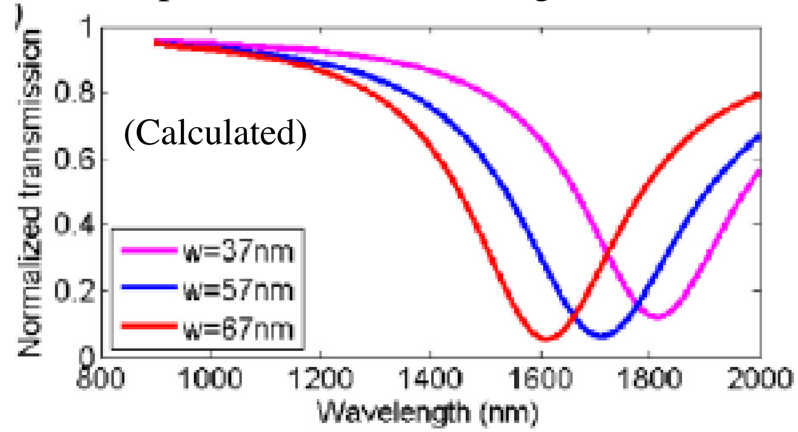


- Substrate (glass or ITO) with a periodic array of gold nano-antennas



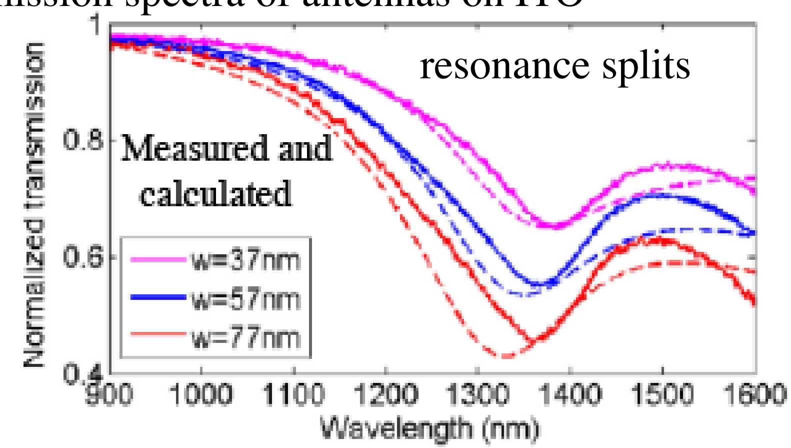
l = antenna length = 404 nm
p = period = 600 nm
w = width = 37, 57, and 77 nm

- Transmission spectra of antennas on glass (no ITO)

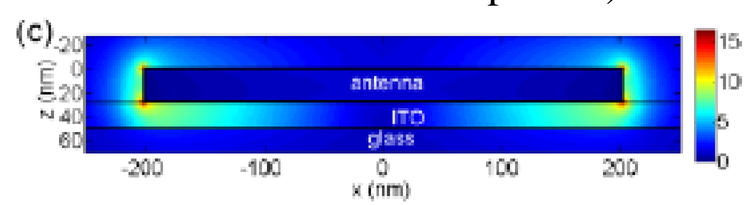


- What happends if the dipole responance coincides with the ENZ resonance?

- Transmission spectra of antennas on ITO



- Strong field enhancement at ends of dipole (further enhancement of NLO response?)



Schulz, Tahir, Alam, Upham, De Leon, and Boyd, PRA 93, 063846 (2016).

Breakout Session
Nonlinear Optics of Epsilon-Near-Zero and Plasmonic Materials

Israel De Leon,
Monterrey Institute of Technology
Radiative Processes of Dipole Emitters in Active Plasmonics and Epsilon-Near-Zero Materials

Orad Reshef,
Harvard University and University of Ottawa
Nonlinear optical response of highly nonlinear low-index materials

M. Zahirul Alam,
University of Ottawa
A Metasurface for Large Nonlinear Refraction

Daryl Vulis
Harvard University
Monolithic CMOS-compatible zero-index metamaterials

"Michal Lipson"
Columbia University
Extreme Nonlinear Optics in Nano-Photonics

Thank you for your attention!