Unity-Order Intensity-Dependent Change in Refractive Index in Indium-Tin Oxide at its Epsilon-Near-ZeroWavelength

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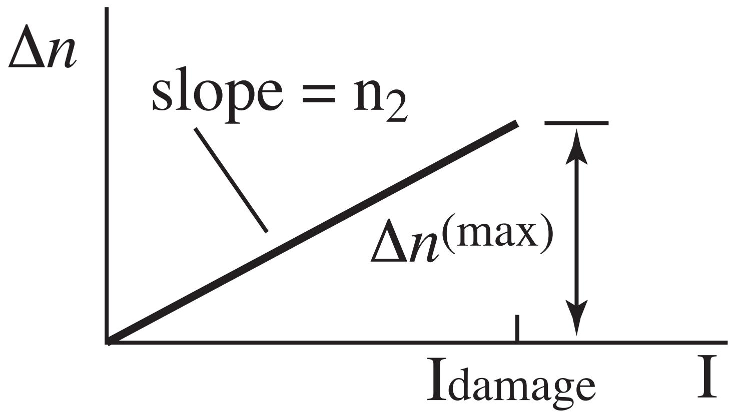
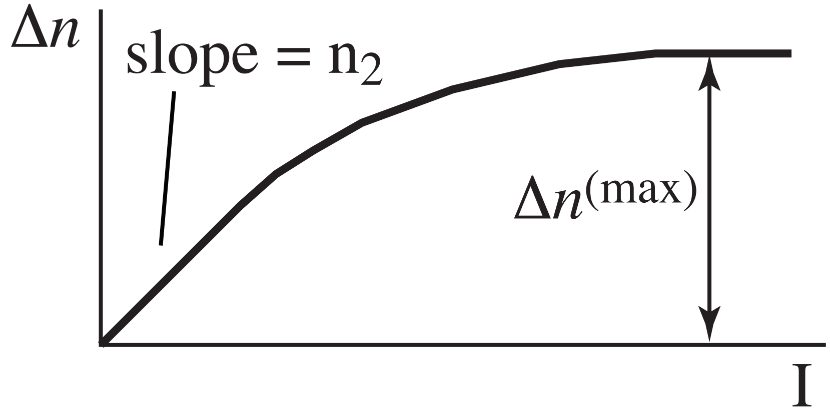
What Makes a Good (Kerr-Effect) Nonlinear Optical Material?

Want n_2 large; and 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, $n_2 = 8 \times 10^{-11}$ 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 $\Delta n^{(\max)} = 3 \times 10^{-4}$)

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.

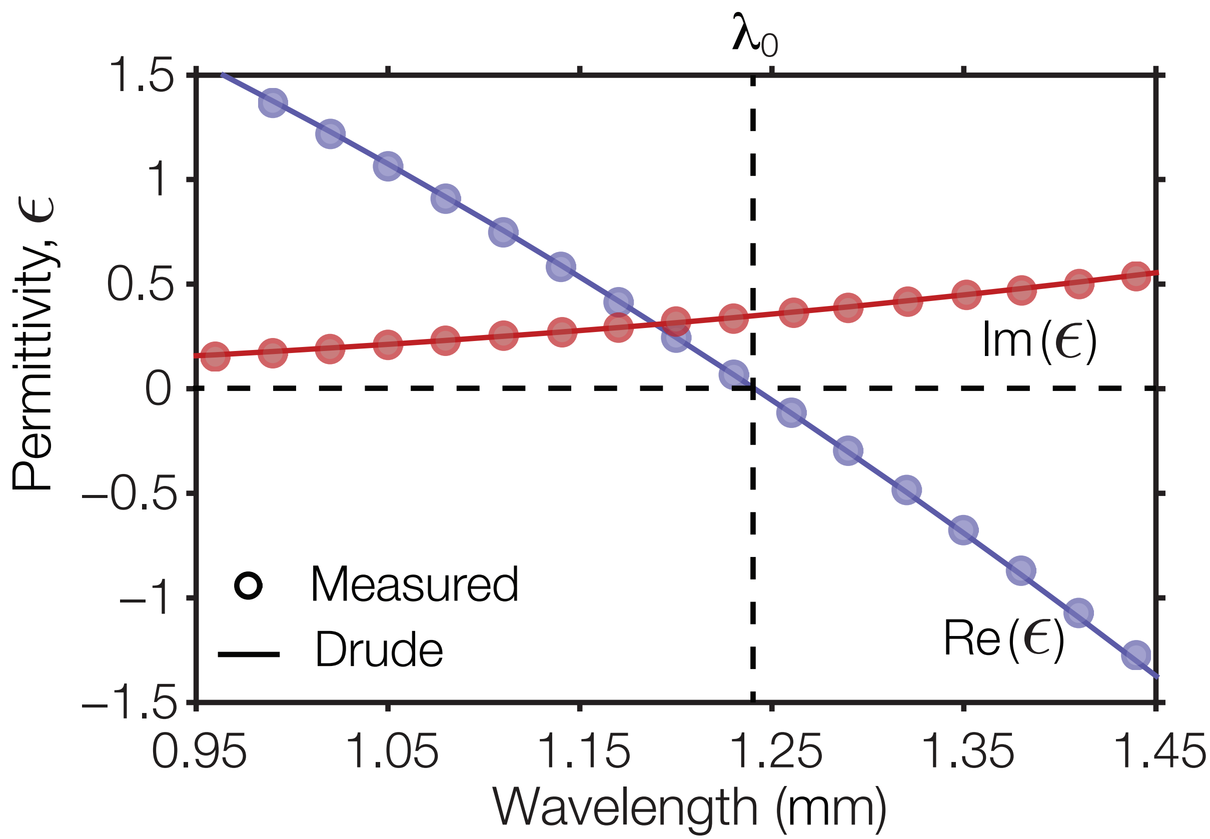
There has been great recent interest in studies of ENZ phenomena:

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.

Implications of ENZ Behavior for Nonlinear Optics

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

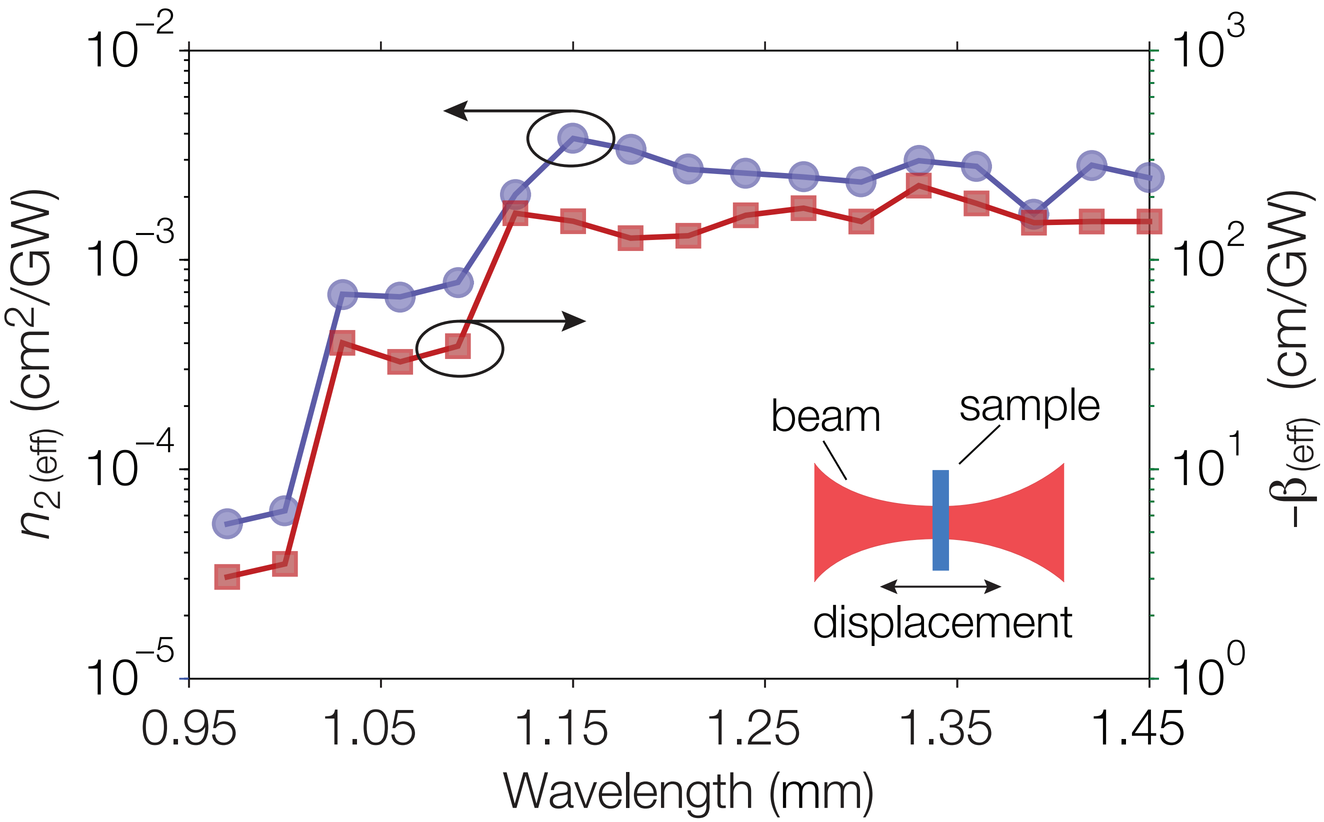
Recall the standard relation between n_2 and $\chi^{(3)}$

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

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

Z-Scan Measurements of the NLO Response

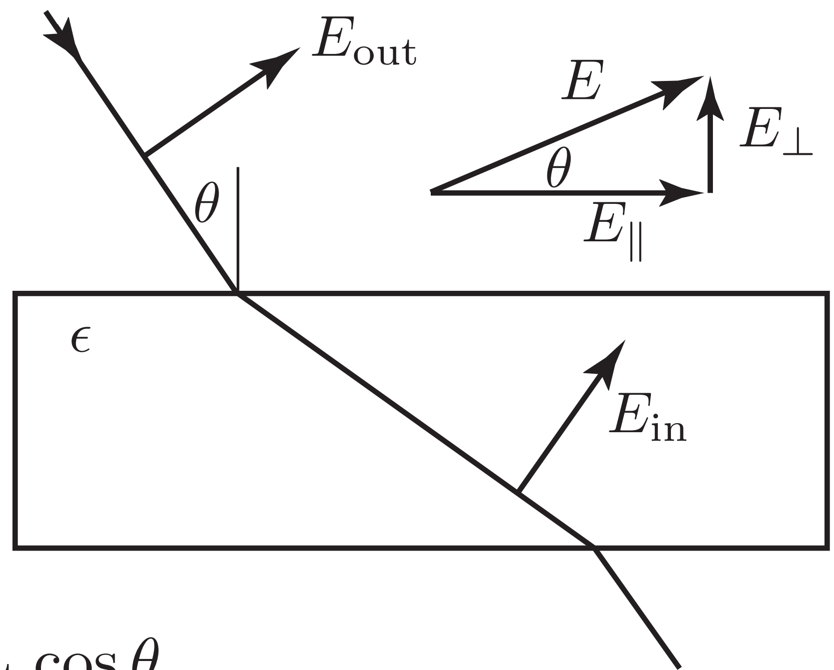
NLO response at *normal incidence*



Maximum measured value of n_2 is 3×10^{-3} cm^2/GW = 3×10^{-12} cm^2/W

Note that n_2 is positive (self focusing) and β is negative (saturable absorption).

The NLO Response Is Even Larger at Oblique Incidence



Standard boundary conditions show that:

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

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

Thus the total field inside of the medium is given by

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

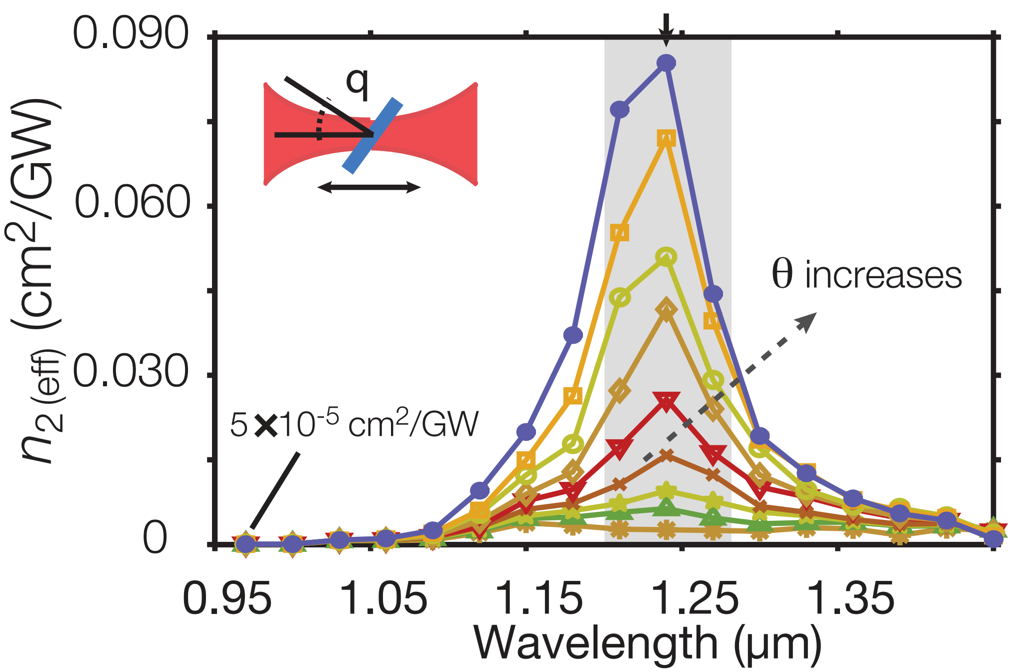
Note that, for $\epsilon < 1$, E_{in} exceeds E_{out} for $\theta \neq 0$.

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

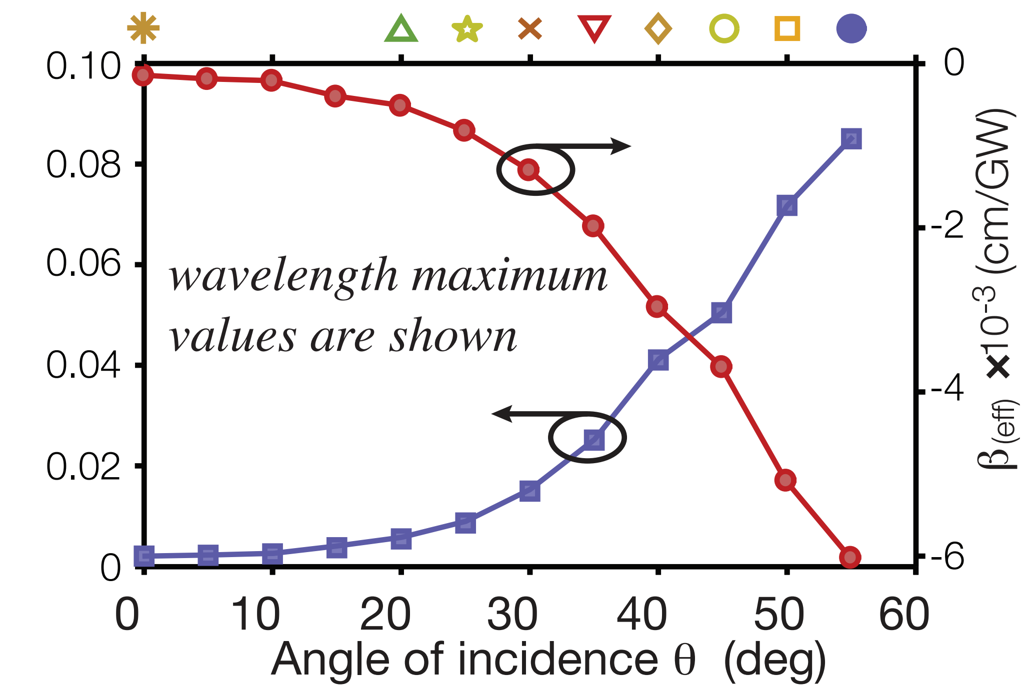
Nonlinear Optical Response at Oblique Incidence

Z-scan measurements for non-normal incidence

Wavelength dependence of n_2



Variation with incidence angle



- Both n_2 and nonlinear absorption increase with angle of incidence
- n_2 achieves a maximum value of 0.08 cm^2/GW = 8 x 10^{-11} cm^2/W at 1.25 μm and 60 deg.

Why is n_2 so large?

The short-wavelength value of n_2 of ITO is 5×10^{-5} cm^2/GW, which is 150 times larger that of fused silica (3.2×10^{-7} cm^2/GW).

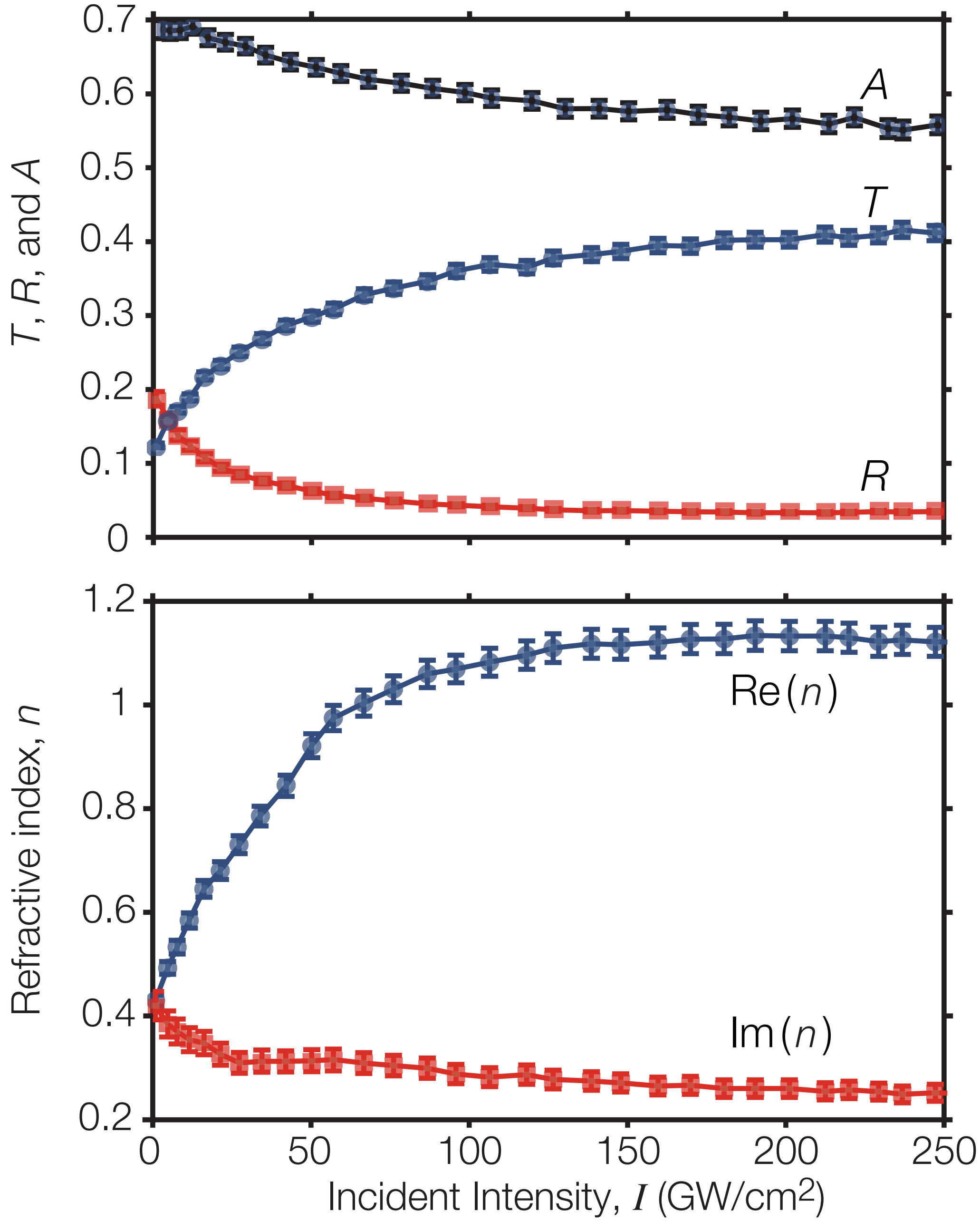
There is a 50x enhancement from working at the ENZ wavelength and an additional 30x enhancement from using non-normal incidence.

Thus $n_2 = 8 \times 10^{-2}$ cm^2/GW, which is 2.5×10^5 times that of fused silica.

Incidentally, for arsenic trisulfide glass, $n_2 = 2.4 \times 10^{-4}$ cm^2/GW. which is 800 times larger than that of fused silica.

R.E. Slusher et al., J. Opt. Soc. Am. B 21, 1146 (2004).

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.

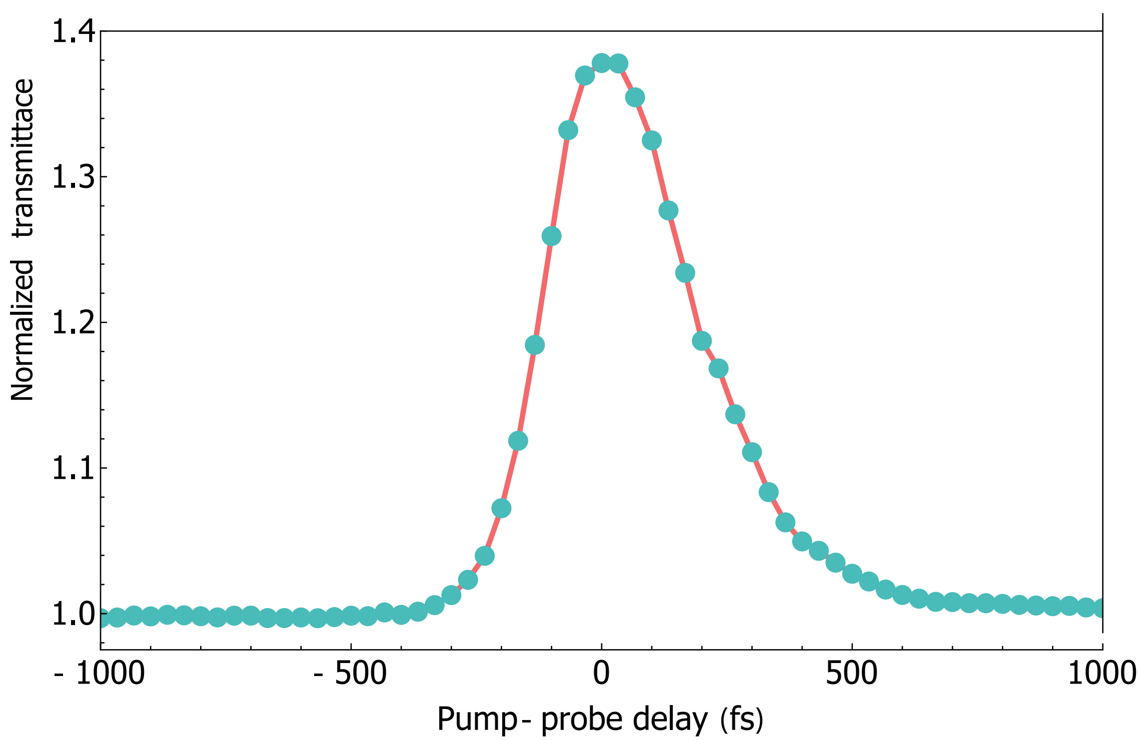
Note that the total nonlinear change in refractive index is $\Delta n = 0.8$.

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 suggests a response time of 270 fs.

ITO will support THz switching speeds



Implications of the Large NLO Response of ITO

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

n_2 is 2.5×10^5 times that of fused silica
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!)

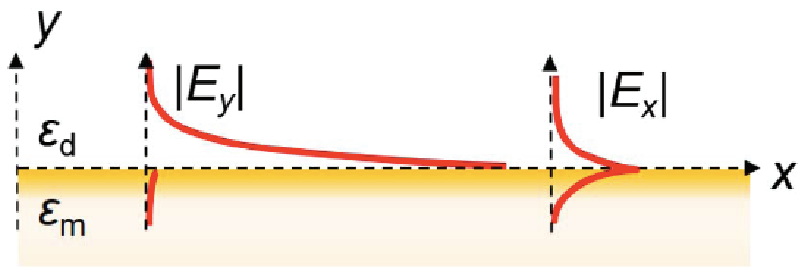
Some possible new effects
Waveguiding outside the "weakly-guiding" regime
Efficient all-optical switching
No need for phase-matching

An Additional Topic (If Time Permits)

What are the nonlinear optical properties of surface plasmon polaritons?

What are the NLO properties of Surface Plasmon Polaritons?

- Surface plasmon polaritons (SPPs) are surface excitations at the interface between a metal and a dielectric.

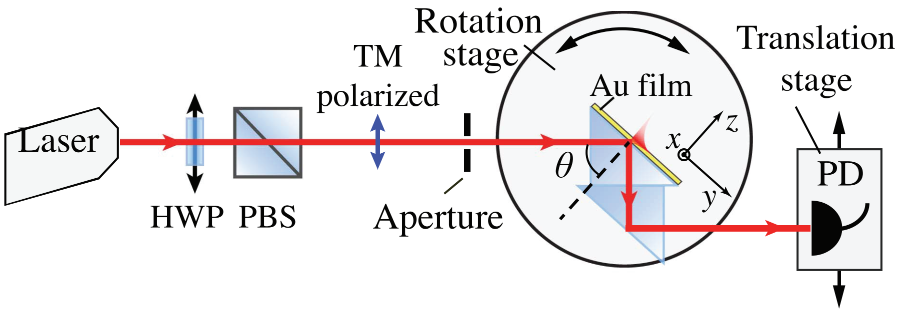


Baron et al., JOSA B (2014)

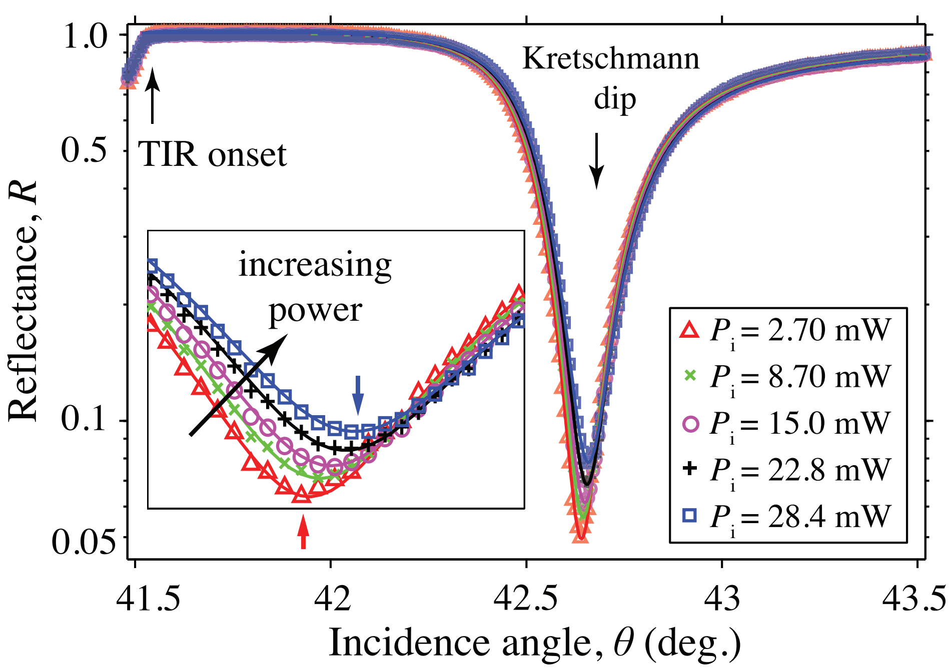
- Light is tightly confined in transverse direction.
- Permits high packing density of photonic circuits.
- Also leads to strong NLO response.
  (Metals are highly nonlinear and the light is tightly confined).
- But SPPs are highly lossy
- How much nonlinear phase shift does an SPP acquire (in one absorption length)? (Can it be as large as π radians?)

What are the NLO properties of Surface Plasmon Polaritons?

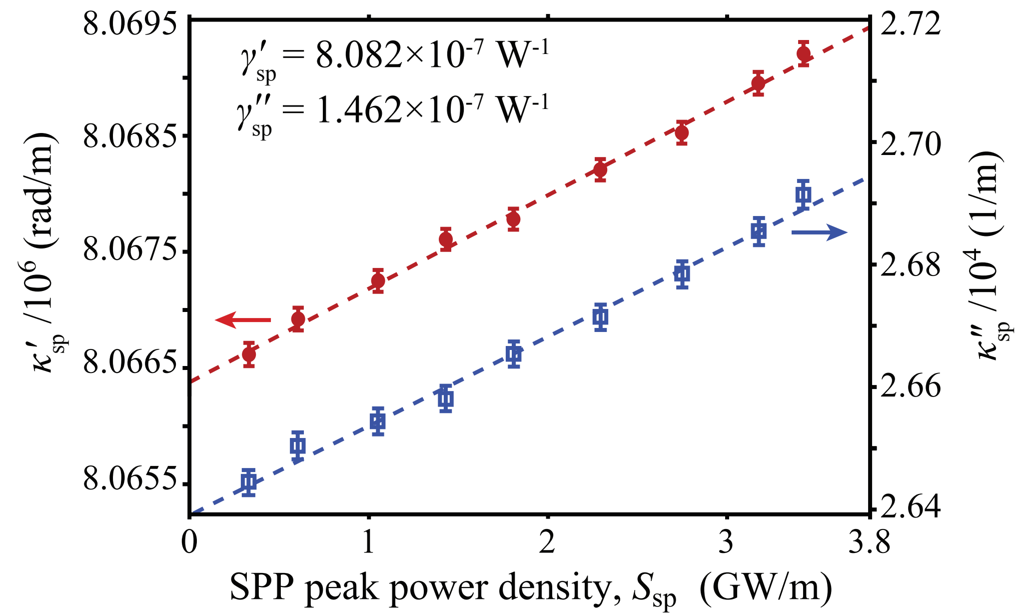
- Kretschmann setup



- Measure intensity dependence of the Kretschmann angle
  power varies from 2.7 to 28 mW
  intensity varies from 2 to 22 GW/cm
  laser wavelength is 796.5 nm



- We can extract the value of $\chi^{(3)}$ of gold



$$\tilde{\chi}^{(3)}_{\mathrm{Au}} = (4.67 + i3.03) \times 10^{-19}\ \mathrm{m}^2/\mathrm{V}^2$$

- But the predicted maximum NL phase shift in one absorption length is only π/60

I. De Leon, Z. Shi, A. Liapis and R.W.Boyd, Optics Letters 39, 2274 (2014)

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