Beyond the perturbative description of the nonlinear optical response of highly nonlinear, epsilon-near-zero materials

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OUTLINE

1. Why do epsilon-near-zero (ENZ) materials give a huge nonlinear response?

2. Does the usual perturbative assumption of NLO (power series expansion of P^{NL}) fail for ENZ materials?

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 $\chi^{(3)}$

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

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 μm.

Recall the Drude formula

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

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

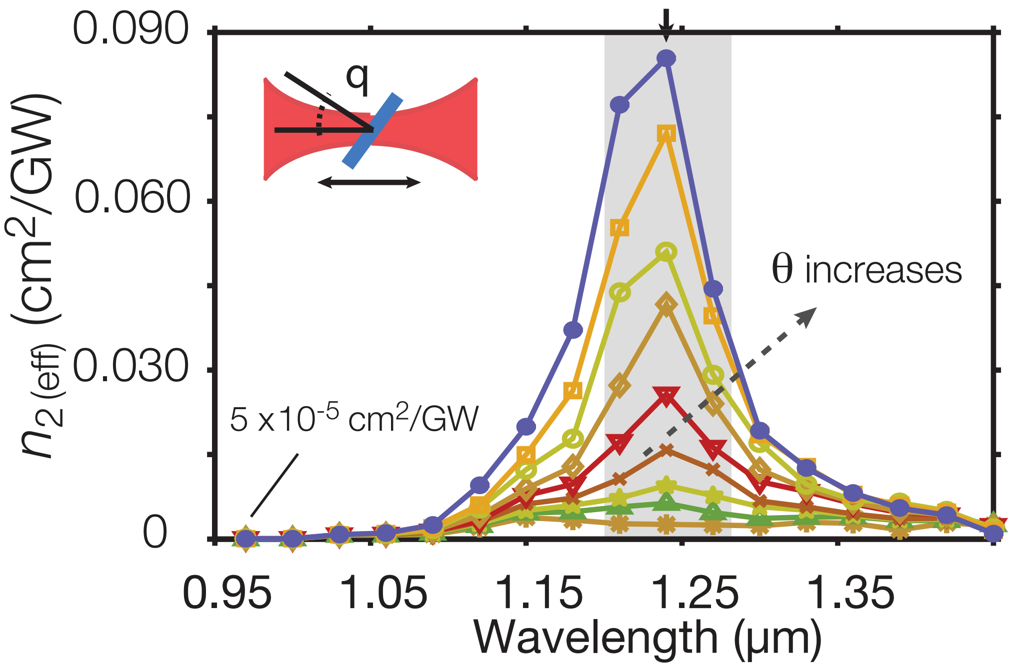
The region near ω_0 is known as the epsilon-near-zero (ENZ) region.

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

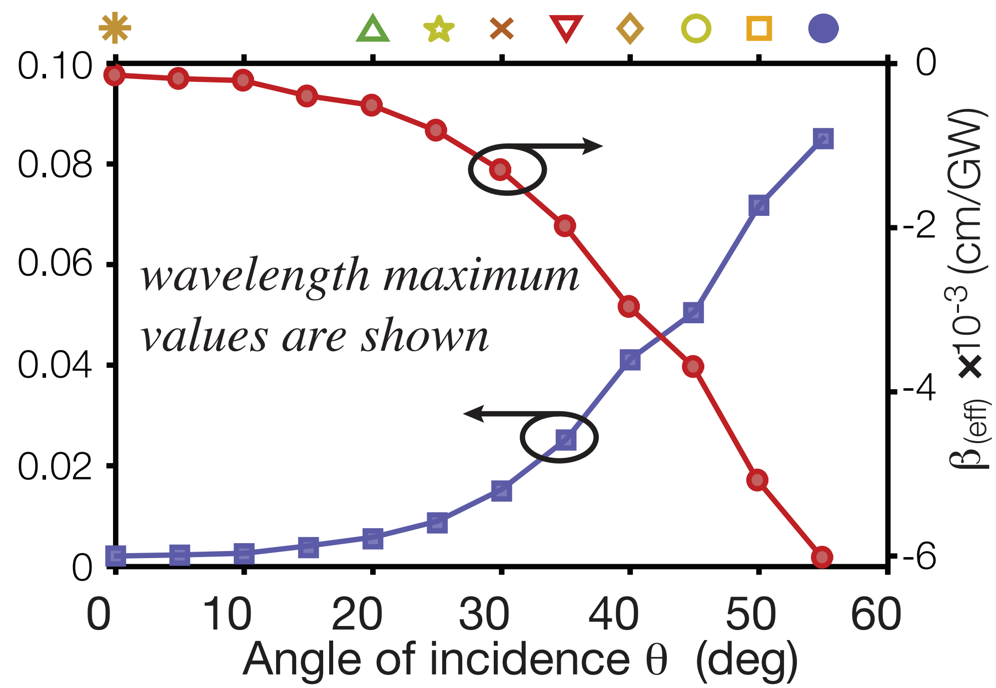
H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, Science 342, 1223 (2013).
C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, Phys. Rev. B 85, 045129 (2012).
S. Campione, D. de Ceglia, M. A. Vincenti, M. Scalora, and F. Capolino, Phys. Rev. B 87, 035120 (2013).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

Huge Nonlinear Optical Response Measured by Z-scan

Wavelength dependence of n_2

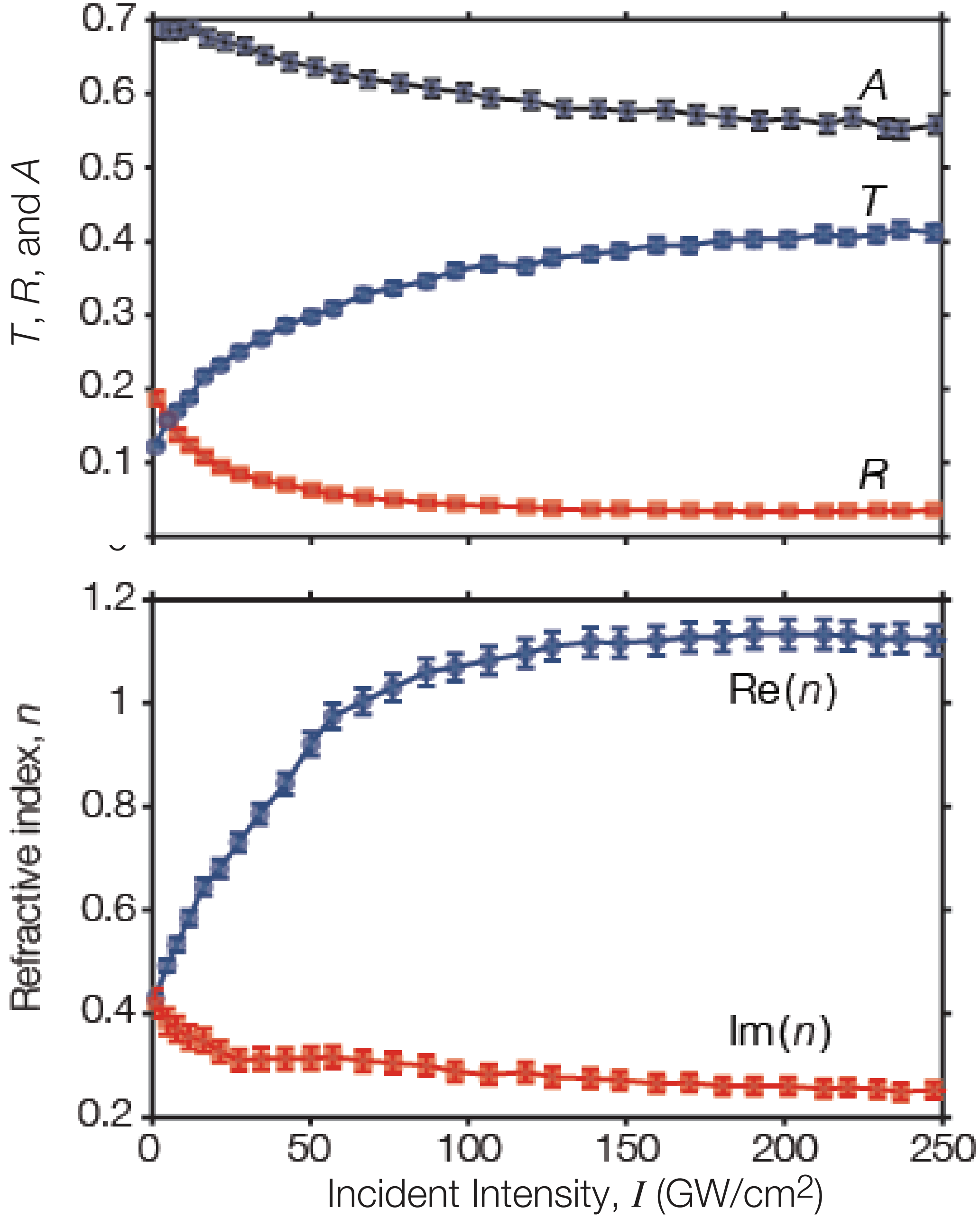


Variation with incidence angle



- 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 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.
- 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

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.

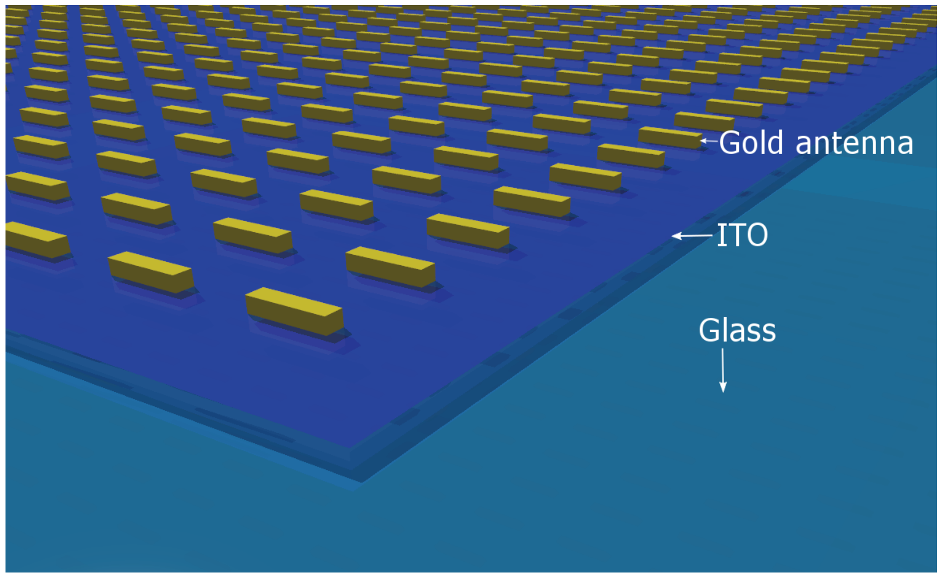
Note that the total nonlinear change in refractive index is $\Delta n = 0.8$.

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

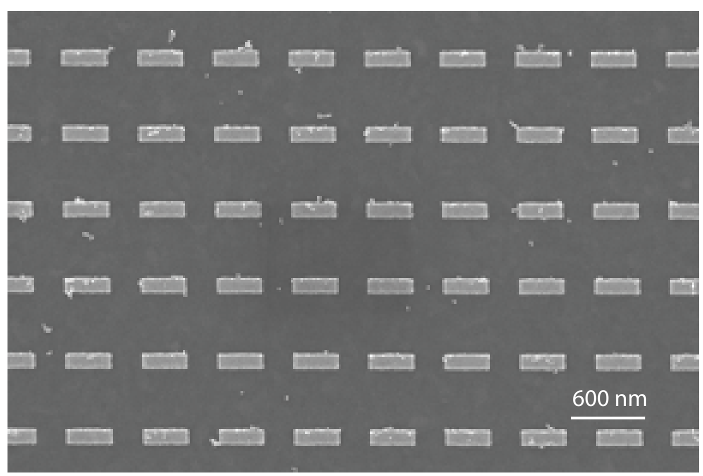
An ENZ Metasurface

- Can we obtain an even larger NLO response by placing a gold antenna array on top of ITO?
- Lightning rod effect: antennas concetrate the field within the ITO

Concept:



SEM:



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

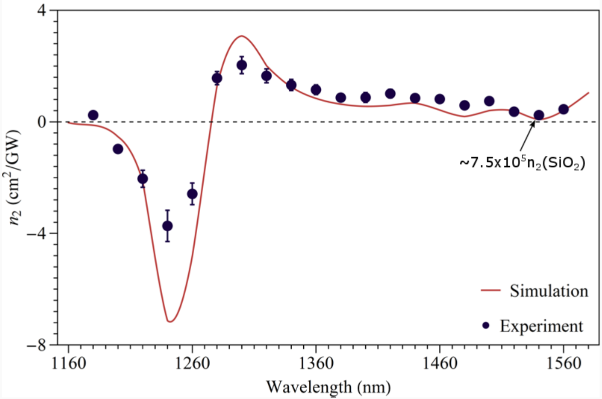


Figure 5: The material exhibits extremely large n_2 for the entire spectral range. The magnitude of the on-resonance value is 7 orders of magnitude larger than that of SiO_2.

OUTLINE

1. Why do epsilon-near-zero (ENZ) materials give a huge nonlinear response?

2. Does the usual perturbative assumption of NLO (power series expansion of P^{NL}) fail for ENZ materials?

Beyond the perturbative description of the nonlinear optical response of low-index materials

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(To appear in Optics Letters)

(Standard derivation)

To lowest nonlinear order, the polarization of a material illuminated by a monochromatic laser field is described as:

$$P^{\mathrm{TOT}} = P + P^{\mathrm{NL}} = \epsilon_0 E \left[\chi^{(1)} + 3\chi^{(3)}|E|^2\right]. \tag{1}$$

Here, E is the complex amplitude of the applied electric field and $\chi^{(1)} \equiv \epsilon^{(1)} - 1$ corresponds to the linear response of the material, with $\epsilon^{(1)}$ being the linear relative permittivity. The relative permittivity including only the $\chi^{(3)}$ nonlinearity is thus

$$\epsilon = \epsilon^{(1)} + 3\chi^{(3)}|E|^2. \tag{2}$$

Since all of these quantities may be complex, we define the complex relative permittivity as $\epsilon = \epsilon' + i\epsilon''$ and the complex refractive index as $n = n' + in''$, where a single prime denotes the real part, and the double prime the imaginary part, respectively. These two quantities are related by [20]

$$n = \sqrt{\epsilon} = \sqrt{\epsilon^{(1)} + 3\chi^{(3)}|E|^2}. \tag{3}$$

Now we introduce n_2

Together, these equations can be used to obtain the complex, intensity-dependent index of refraction n due to third-order contributions. We find that

$$n = \sqrt{n_0^2 + 2n_0 n_2 I}, \tag{4}$$

This form is valid, but weird

where we take $n_0 = \sqrt{\epsilon^{(1)}}$ to be the linear refractive index, I to be the optical field intensity

$$I = 2\mathrm{Re}(n_0)\epsilon_0 c|E|^2, \tag{5}$$

Note that intensity vanishes under ENZ conditions!

and we introduce the standard definition for the nonlinear index of refraction [4, 20]

$$n_2 = \frac{3\chi^{(3)}}{4n_0 \mathrm{Re}(n_0)\epsilon_0 c}. \tag{6}$$

In order to obtain a simpler relation for n, Eq. (4) is usually expanded in a power series under the assumption that $|2n_2 I/n_0| \ll 1$ [4], yielding

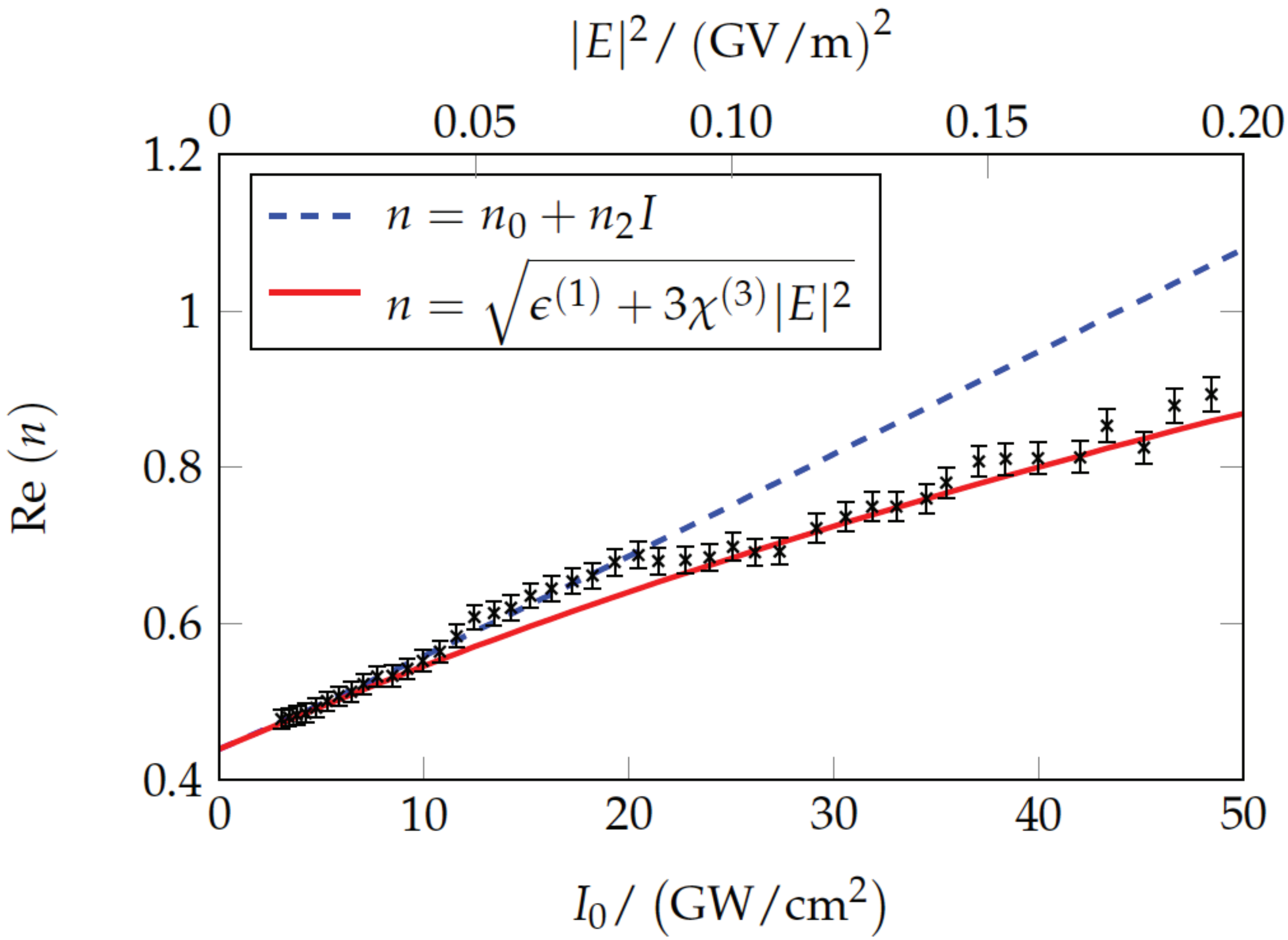
$$n = n_0\sqrt{1 + 2\frac{n_2 I}{n_0}} \approx n_0\left[1 + \frac{1}{2}\left(2\frac{n_2 I}{n_0}\right) + \ldots\right]. \tag{7}$$

In most materials, $|2n_2 I/n_0|$ is very small so that only the lowest order correction term is kept, resulting in the intensity-dependent refractive index being widely defined as

$$n = n_0 + n_2 I. \tag{8}$$

This form is invalid for ENZ materials (because Series 7 does not converge)

Comparison To Our Experimental Data

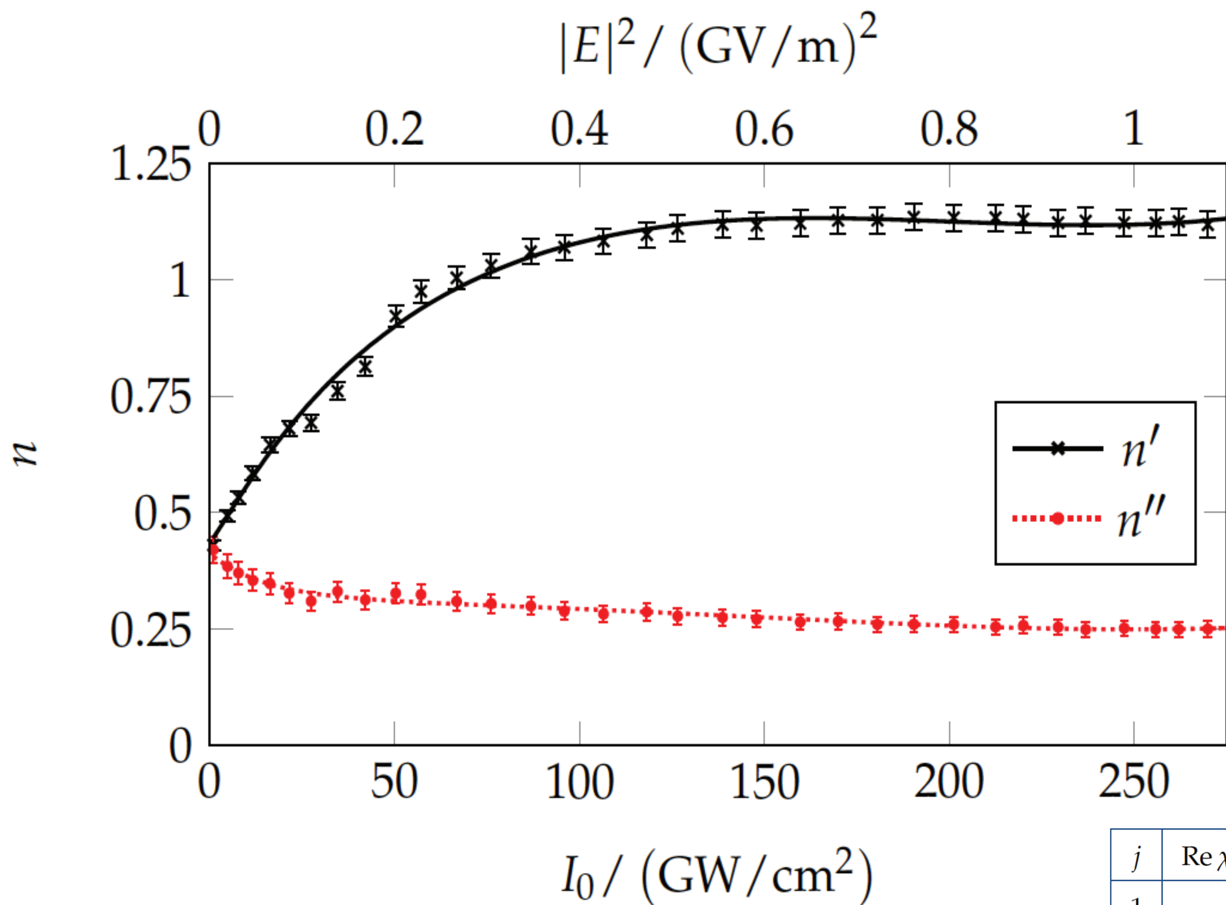


The "square-root" model fits the data much better than the $n_2 I$ model.

I_0 is the incident intensity (measured outside the material);
E is the electric field measured inside the material

For higher intensities, we need to include $\chi^{(5)}$ and $\chi^{(7)}$ contributions

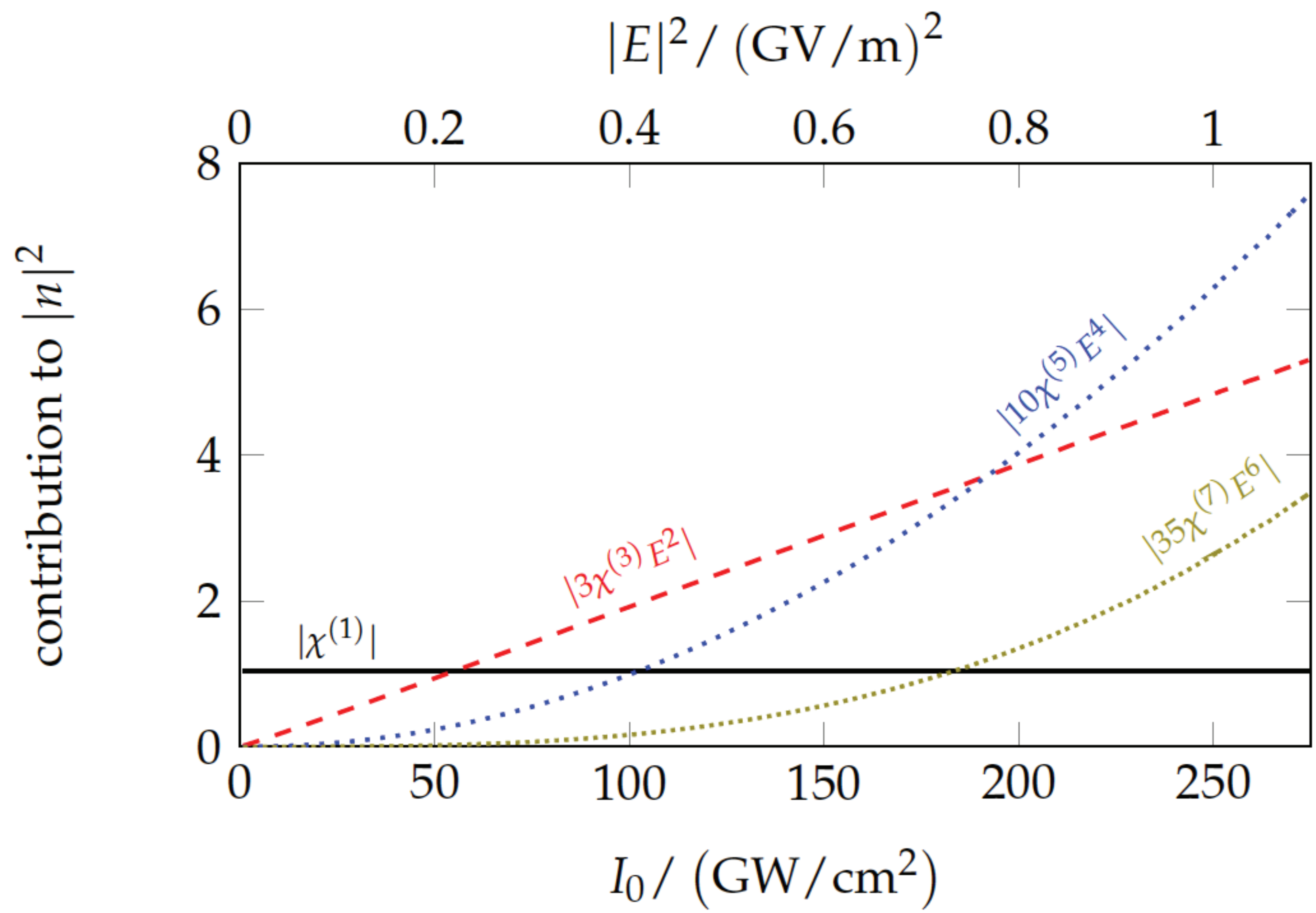
(but no need to include higher-order contributions, up to damage threshold)



j	$\mathrm{Re}\,\chi^{(j)} / (10^{-9}\mathrm{m/V})^{j-1}$	$\mathrm{Im}\,\chi^{(j)} / (10^{-9}\mathrm{m/V})^{j-1}$
1	-0.980 ± 0.008	0.36 ± 0.01
3	1.60 ± 0.03	0.50 ± 0.05
5	-0.63 ± 0.02	-0.25 ± 0.04
7	$(7.7 \pm 0.3) \times 10^{-2}$	$(3.5 \pm 0.8) \times 10^{-2}$

TABLE I. Values extracted from the fit to Eq. (9) with a third, fifth and seventh-order nonlinearity.

Nonlinear Response of ITO is Nonperturbative



Conclusions

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

2. The nonlinear response can be accurately modeled in the $\chi^{(3)}$ limit by

$$n = \sqrt{n_0^2 + 2n_0 n_2 I}$$

3. More generally, the intensity dependent refractive index can be described by

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

4. The nonlinear response of ITO is nonperturbative.


A Metasurface for Large Nonlinear Refraction

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[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

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

A thin ENZ medium supports a bulk plasma mode.

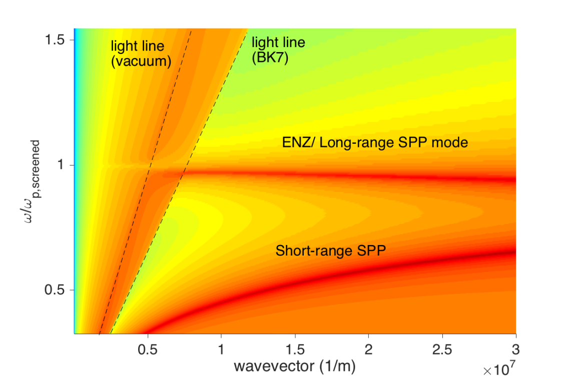


Figure 3: A thin layer of ITO supports two modes: bulk plasma, short range surface plasmon (SPP).

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

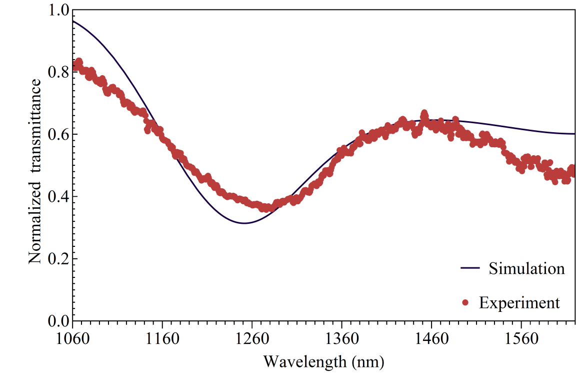
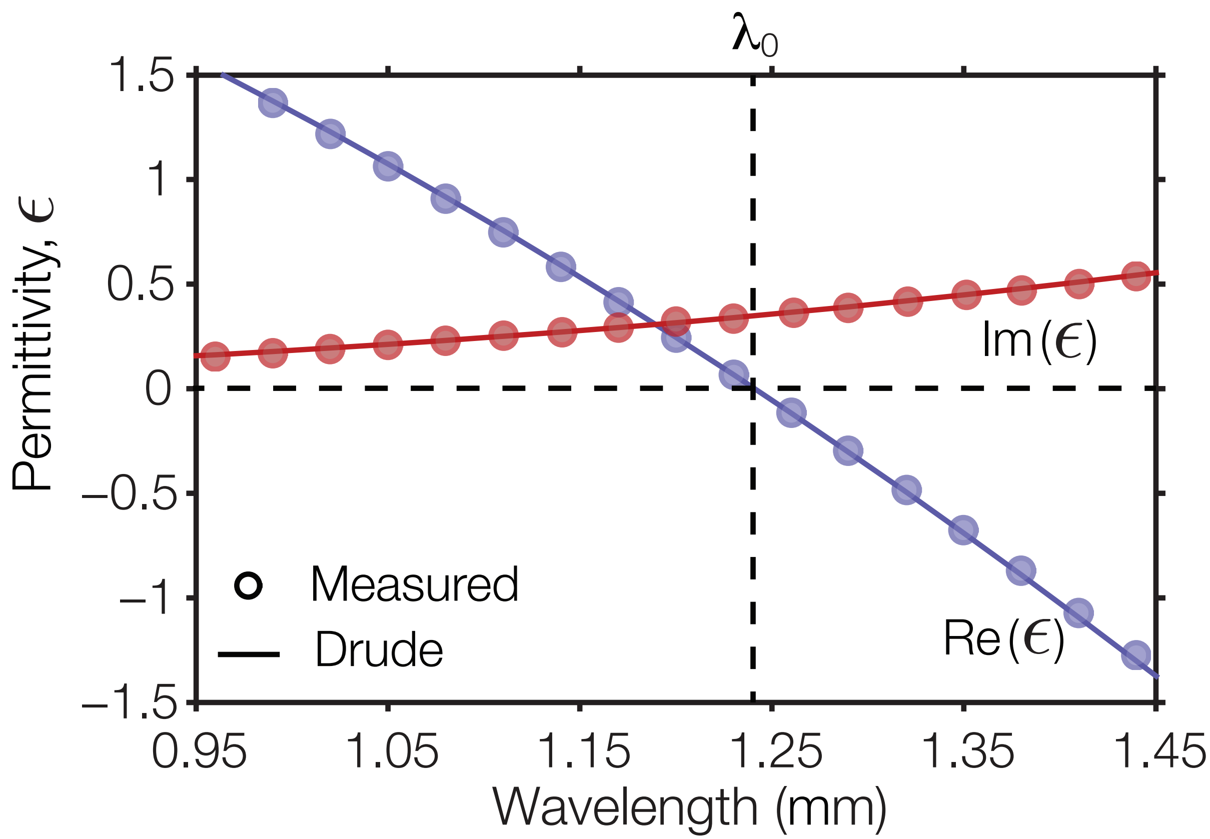


Figure 4: The coupling between the fundamental mode of the optical dipole antenna and the ENZ mode results in two distinct dips in the transmission spectrum.

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.

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

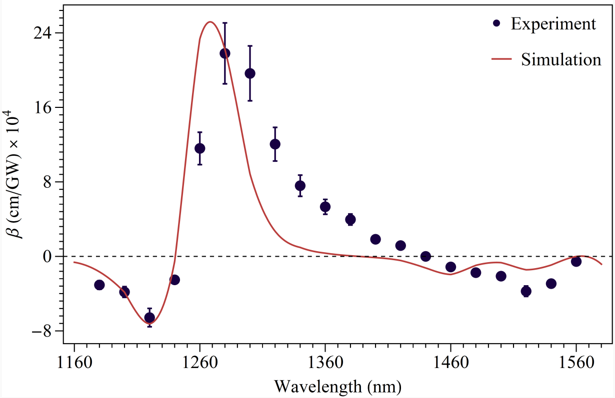


Figure 6: Nonlinear absorption shows both negative and positive signs indicating saturable and reverse saturable absorption respectively. Note: One can design to maximize the absorption for applications in power limiter, low power pulsed lasing, etc.

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.

Other Reports of Highly Nonlinear Response in ENZ Material and Metamaterialc

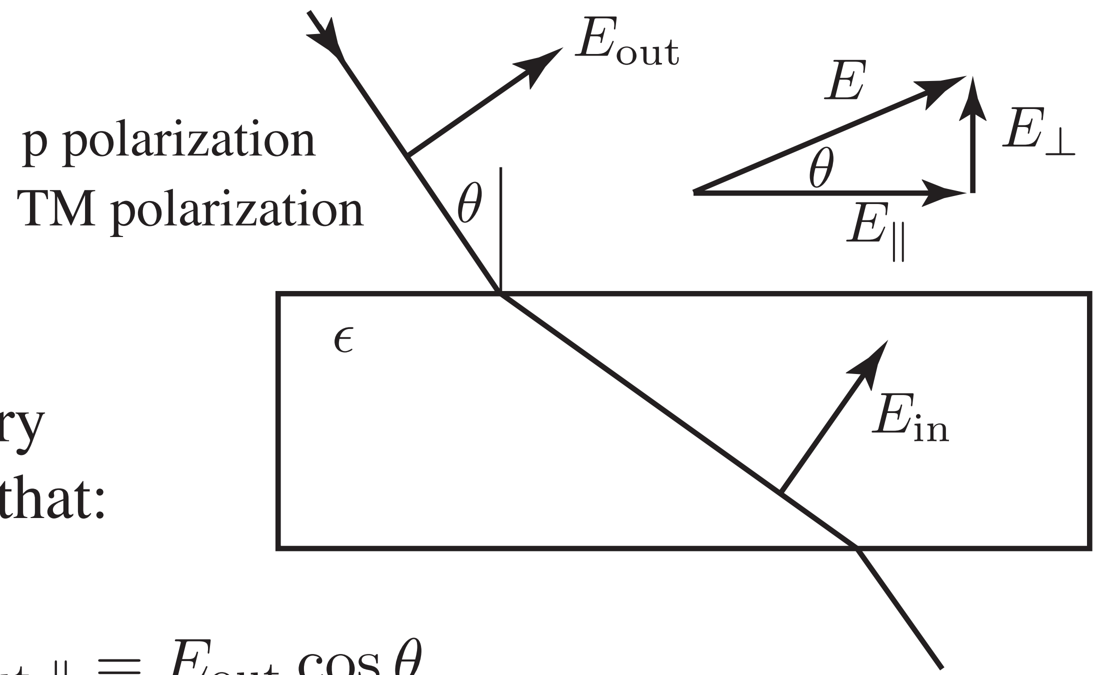
Enhanced Nonlinear Refractive Index in epsilon-Near-Zero Materials,
L. Caspani, R. P. M. Kaipurath, M. Clerici, M. Ferrera, T. Roger, J. Kim, N. Kinsey,
M. Pietrzyk, A. D. Falco, V. M. Shalaev, A. Boltasseva and D. Faccio,
Phys. Rev. Lett. 116, 233901, 2016.

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).

NLO Response Increases with 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

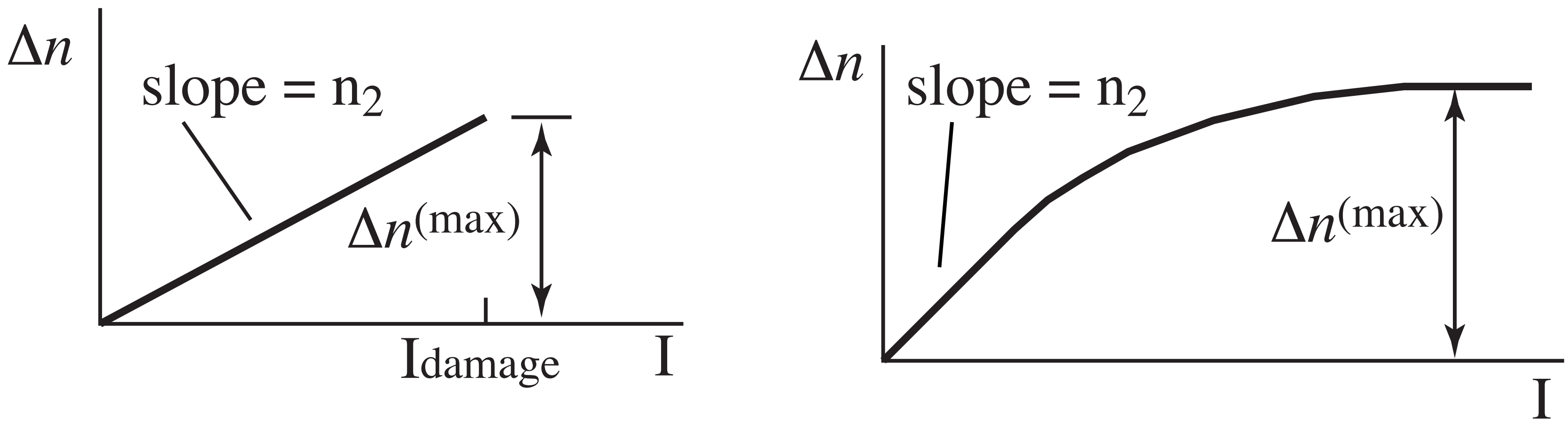
$$E_{\text{in}} = E_{\text{out}} \sqrt{\cos^2\theta + \frac{\sin^2\theta}{\epsilon}}$$

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.

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).