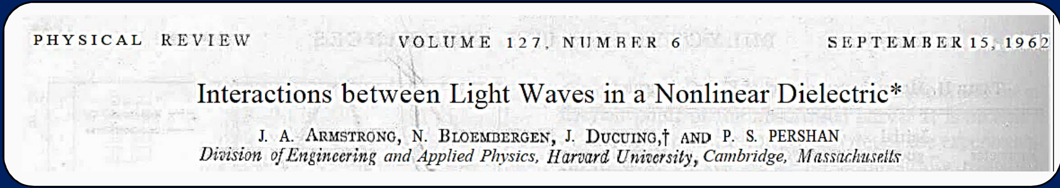
FI255 - Tópicos de Óptica e Fotônica II

Óptica Não-Linear

15ª. aula

Prof. Cid B. de Araújo
UNICAMP - 29 de junho de 2018

General theoretical approach


PHYSICAL REVIEW VOLUME 127, NUMBER 6 SEPTEMBER 15, 1962

Interactions between Light Waves in a Nonlinear Dielectric*

J. A. ARMSTRONG, N. BLOEMBERGEN, J. DUCUING,† AND P. S. PERSHAN

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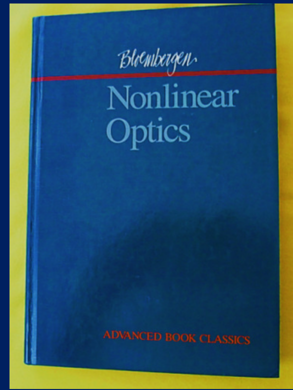


When there is inversion symmetry:

$$\chi^{(j)} \equiv 0$$

j = even

$$P_L + P_{NL} = \epsilon_0 \sum_{N=0}^{\infty} \chi^{(2N+1)} E^{(2N+1)}$$

$$n_N \propto Re\, \chi^{(2N+1)}$$

Nonlinear refractive index

$$\alpha_N \propto Im\, \chi^{(2N+1)}$$

Nonlinear absorption coefficient

linear + nonlinear

$$n = n_0 + n_2 I + n_4 I^2 + n_6 I^3 + \cdots$$

$$\alpha = \alpha_0 + \alpha_2 I + \alpha_4 I^2 + \alpha_6 I^3 + \cdots$$

High-order nonlinearities

- Generation of higher harmonics
- Multiphoton excitation processes
- Multi-wave mixing

Studies of multiphoton ionization: 60´s; Many studies in the 70´s, 80´s.

Multiphoton dissociation and ionization processes applied to isotope separation (70´s and 80´s).

More recently:
Solitons, filamentation, extreme events, generation of VUV and soft X-rays – 400th harmonic, proposals for attosecond X-ray pulse generation ....

Como proceder para aumentar a resposta não linear de materiais?

- Usar lasers com frequências próximas de ressonância com transições ópticas do material
- Fabricação de compósitos com nano-inclusões que tenham maior resposta não linear que o material hospedeiro
- Fabricação de hetero-estruturas com componentes de diferentes índices de refração
- Usar micro-cavidades de alto Q
- Cristais fotônicos com propagação de luz lenta

VANTAGENS E DESVANTAGENS DE CADA ABORDAGEM

Uma (recente) abordagem alternativa

Investigar o comportamento de sistemas que apresentem função dielétrica próxima de zero

ENZ materials – epsilon near zero materials

Para entender a motivação, considere um meio transparente. Temos que

$$n^2 = \epsilon$$

A mudança no índice de refração induzida por um laser será

$$\Delta n = \frac{\Delta\epsilon}{2\sqrt{\epsilon}}$$

Quanto menor for épsilon, maior será a variação induzida no índice de refração

Artigos que ilustram esta abordagem

1500 OPTICS LETTERS / Vol. 40, No. 7 / April 1, 2015

Enhanced third-harmonic generation in Si-compatible epsilon-near-zero indium tin oxide nanolayers

Antonio Capretti,[1,2] Yu Wang,[1] Nader Engheta,[3] and Luca Dal Negro[1]

We experimentally demonstrate enhanced third-harmonic generation from indium tin oxide nanolayers at telecommunication wavelengths with an efficiency that is approximately 600 times larger than crystalline silicon (Si). The increased optical nonlinearity of the fabricated nanolayers is driven by their epsilon-near-zero response, which can be tailored on-demand in the near-infrared region. The present material platform is obtained without any specialized nanofabrication process and is fully compatible with the standard Si-planar technology. The proposed approach can lead to largely scalable and highly integrated optical nonlinearities in Si-integrated devices for information processing and optical sensing applications.

PRL 116, 233901 (2016) PHYSICAL REVIEW LETTERS week ending 10 JUNE 2016

Enhanced Nonlinear Refractive Index in ϵ-Near-Zero Materials

L. Caspani,[1] R. P. M. Kaipurath,[1] M. Clerici,[1,2] M. Ferrera,[1] T. Roger,[1] J. Kim,[3] N. Kinsey,[3] M. Pietrzyk,[4] A. Di Falco,[4] V. M. Shalaev,[3] A. Boltasseva,[3] and D. Faccio[1,*]

New propagation regimes for light arise from the ability to tune the dielectric permittivity to extremely low values. Here, we demonstrate a universal approach based on the low linear permittivity values attained in the ϵ-near-zero (ENZ) regime for enhancing the nonlinear refractive index, which enables remarkable light-induced changes of the material properties. Experiments performed on Al-doped ZnO (AZO) thin films show a sixfold increase of the Kerr nonlinear refractive index (n_2) at the ENZ wavelength, located in the 1300 nm region. This in turn leads to ultrafast light-induced refractive index changes of the order of unity, thus representing a new paradigm for nonlinear optics.

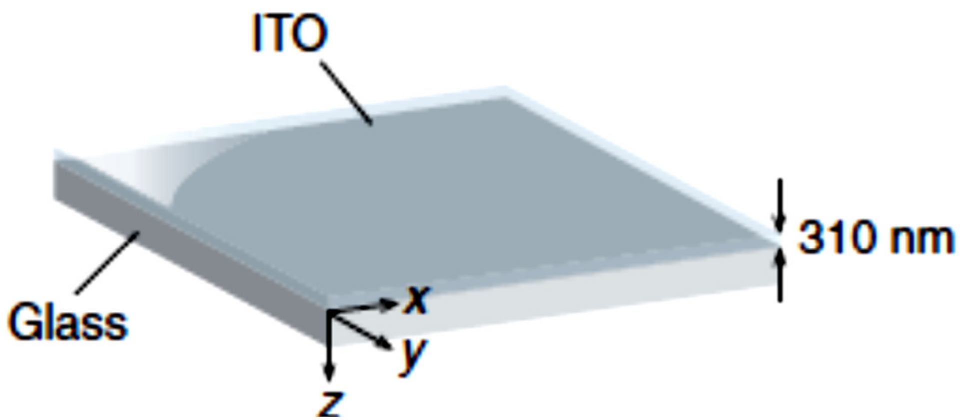
NONLINEAR OPTICS SCIENCE 13 MAY 2016 • VOL 352 ISSUE 6287 795

Large optical nonlinearity of indium tin oxide in its epsilon-near-zero region

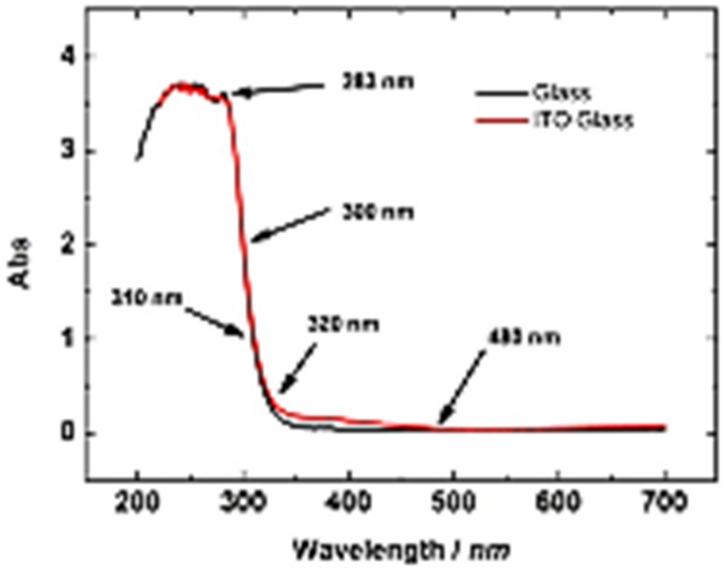
M. Zahirul Alam,[1] Israel De Leon,[1,3]* Robert W. Boyd[1,2]

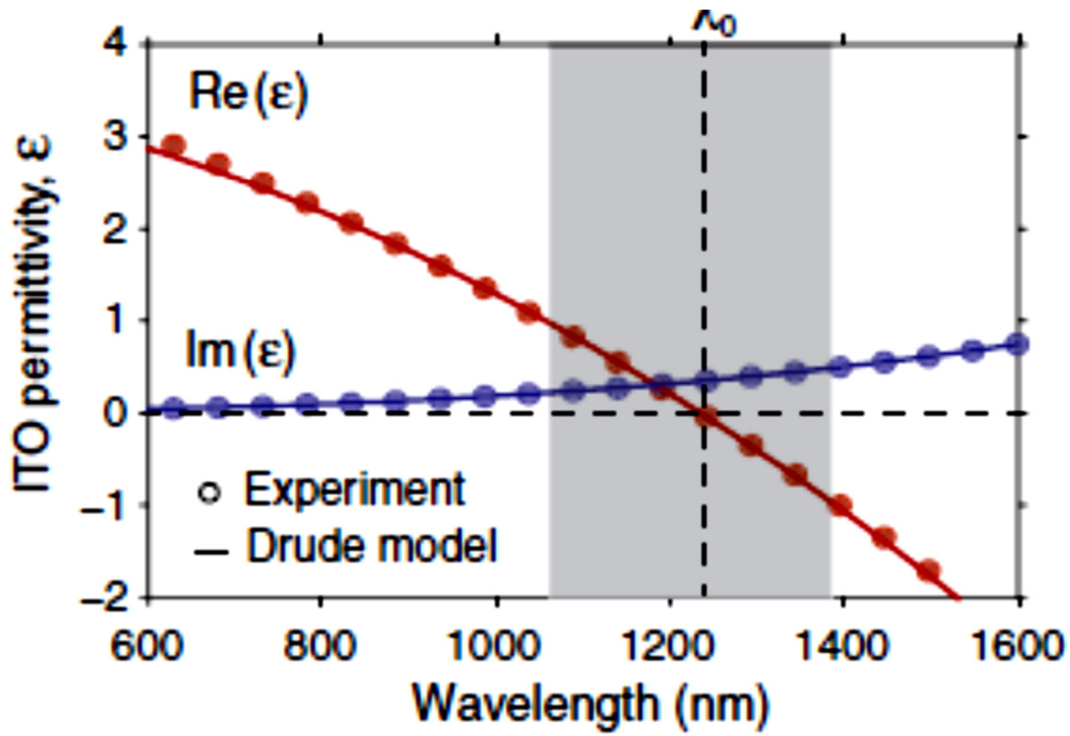
Nonlinear optical phenomena are crucial for a broad range of applications, such as microscopy, all-optical data processing, and quantum information. However, materials usually exhibit a weak optical nonlinearity even under intense coherent illumination. We report that indium tin oxide can acquire an ultrafast and large intensity-dependent refractive index in the region of the spectrum where the real part of its permittivity vanishes. We observe a change in the real part of the refractive index of 0.72 ± 0.025, corresponding to 170% of the linear refractive index. This change in refractive index is reversible with a recovery time of about 360 femtoseconds. Our results offer the possibility of designing material structures with large ultrafast nonlinearity for applications in nanophotonics.

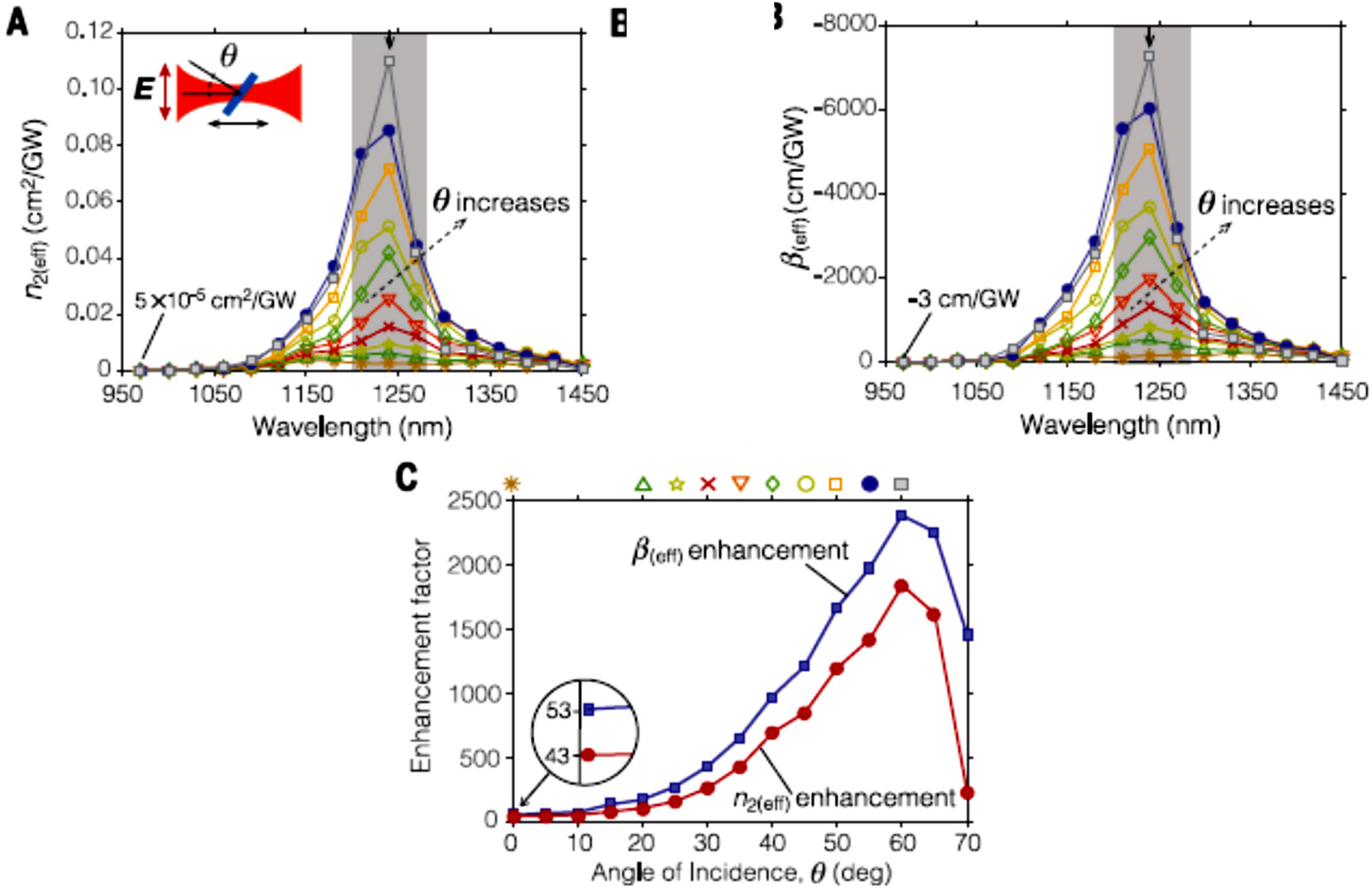
Indium tin oxide



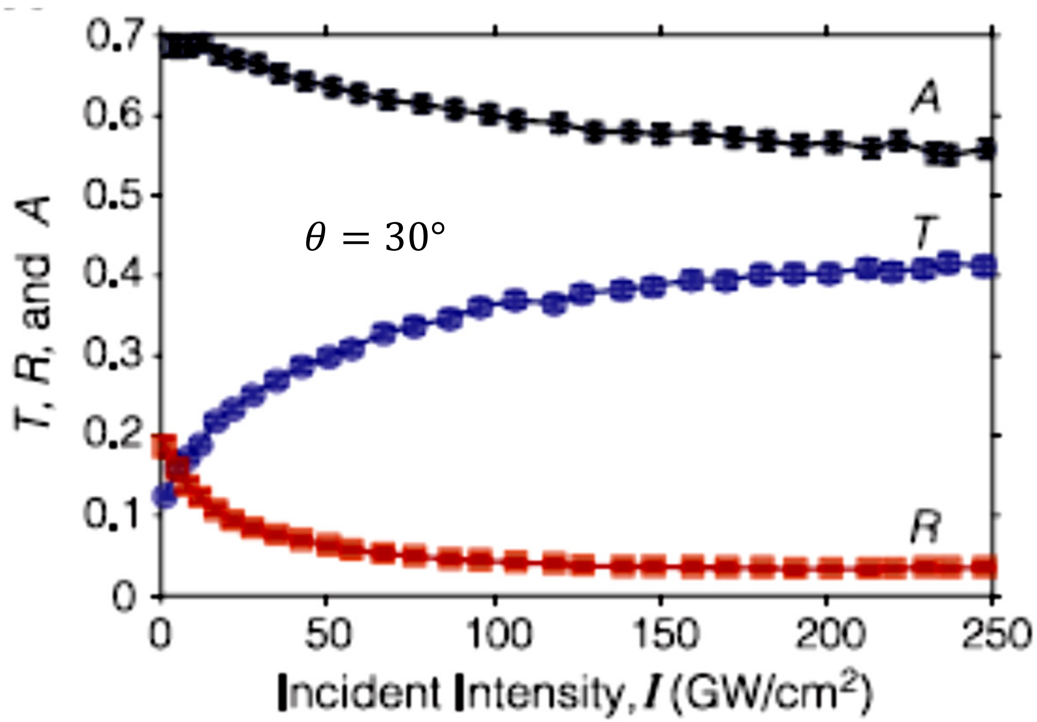
Uma das possíveis composições:
74% In; 18% O_2 ; 8% Sn

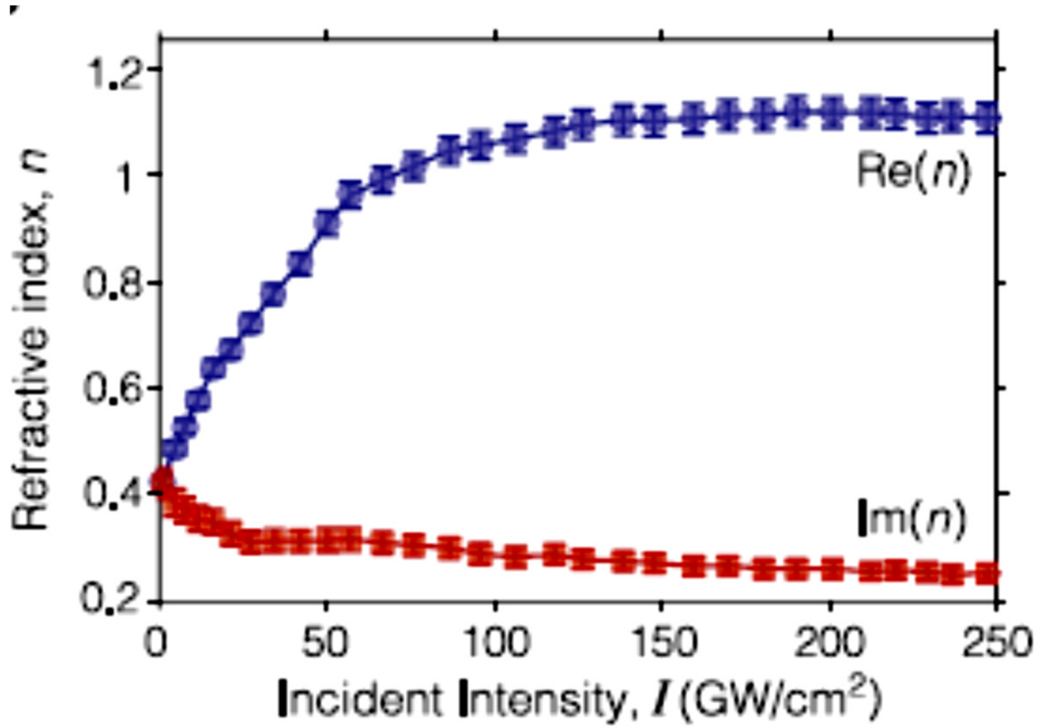






A
B
C
E
θ
θ increases
5×10⁻⁵ cm²/GW
n2(eff) (cm²/GW)
Wavelength (nm)
-3 cm/GW
β(eff) (cm/GW)
β(eff) enhancement
n2(eff) enhancement
53
43
Enhancement factor
Angle of Incidence, θ (deg)





Mudança gigantesca no valor da parte real do índice de refração

Em baixa intensidade: 0,42

Em 150 GW/cm^2: $1{,}14 \pm 0.025$

Letter
Vol. 42, No. 16 / August 15 2017 / *Optics Letters* 3225

Optics Letters

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

ORAD RESHEF,[1,*] ENNO GIESE,[1] M. ZAHIRUL ALAM,[1] ISRAEL DE LEON,[2] JEREMY UPHAM,[1] AND ROBERT W. BOYD[1,3]

We show that standard approximations in nonlinear optics are violated for situations involving a small value of the linear refractive index. Consequently, the conventional equation for the intensity-dependent refractive index, $n(I) = n_0 + n_2 I$, becomes inapplicable in epsilon-near-zero and low-index media, even in the presence of only third-order effects. For the particular case of indium tin oxide, we find that the $\chi^{(3)}$, $\chi^{(5)}$, and $\chi^{(7)}$ contributions to refraction eclipse the linear term; thus, the nonlinear response can no longer be interpreted as a perturbation in these materials. Although the response is non-perturbative, we find no evidence that the power series expansion of the material polarization diverges.

Procedimento usual

$$P^{TOT} = P_L + P_{NL} = \epsilon_0 E \left[\chi^{(1)} + 3\chi^{(3)}\right]$$

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

$$n = \sqrt{n_0^2 + 2n_0 n_2 I} \qquad I = 2Re(n_0)\epsilon_0 c|E|^2 \qquad n_2 = \frac{3\chi^{(3)}}{4n_0 Re(n_0)\epsilon_0 c}$$

$$n = n_0\sqrt{1 + 2\frac{n_2}{n_0}I} \approx n_0\left\{1 + \frac{1}{2}\left(2\frac{n_2 I}{n_0}\right) + \cdots\right\} \qquad \left|2\frac{n_2 I}{n_0}\right| \ll 1$$

$$n = n_0 + n_2 I$$

Mechanism	n_2 (cm²/W)	$\chi^{(3)}_{1111}$ (esu)	Response time (sec)
Electronic Polarization	10^{-16}-10^{-13}	10^{-14}-10^{-11}	10^{-15}
Molecular Orientation	10^{-14}	10^{-12}	10^{-12}
Electrostriction	10^{-14}	10^{-12}	10^{-9}
Saturated Absorption	10^{-10}	10^{-8}	10^{-8}
Thermal effects	10^{-6}	10^{-4}	10^{-3}

$$\Delta n(r=0) = n_2 I_0$$

$$I_0 = 1\ \mathrm{GW/cm^2}$$

$$\Delta n = 10^{-7} - 10^{-4}$$

Electronic polarization

Nos materiais ENZ $\frac{\Delta n}{n_0} \gg 1$

A série não converge e a equação $n = n_0 + n_2 I$ não fornece uma aproximação válida para n_2

A partir da equação $n = \sqrt{n_0^2 + 2n_0 n_2 I}$ vemos que

Se $|n_0| \to 0 \Rightarrow n \to 0$; $I \to 0$ então $n_2 \to \infty \Rightarrow n_2 I$ $fica\ indefinido$

Note que n_0 só foi introduzido na equação para n para obter a equação $n = n_0 + n_2 I$

Se usarmos a expressão

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

não haverá problema mesmo se n_2 e I assumirem valores excepcionais

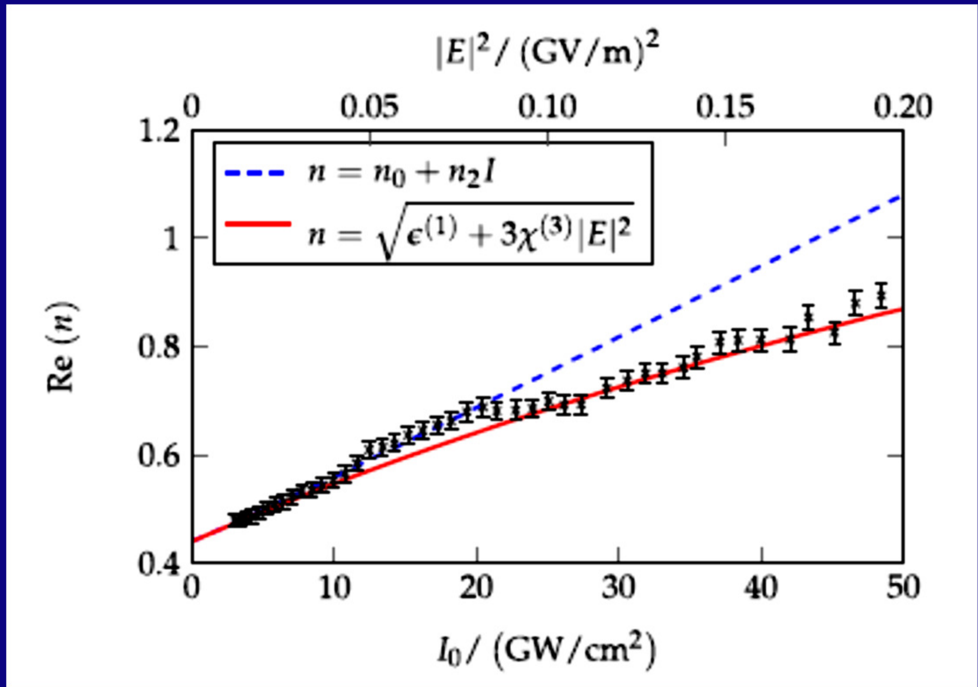
Conclusão: não devemos introduzir n_2 pela equação $n = n_0 + n_2 I$ no contexto dos materiais ENZ

Para demonstrar isto vamos usar os resultados experimentais para ITO em 1240 nm onde $Re\ \epsilon^{(1)} = 0$ e a não-linearidade é alta.

A partir de experiências independentes temos:

Elipsometria: $Re\ (n_0) = 0.44$

Z-scan: $Re\ n_2 = 0.016\ cm^2/W$



Note que a aproximação de $\chi^{(3)}$ é razoável apenas para baixas intensidades usando ambas as formas de expressar n

$$P^{TOT} = \epsilon_0 E \sum_j^{\infty} c_j \chi^{(j)} |E|^{j-1}$$

fator de degenerescência: c_j $c_1 = 1; c_3 = 3; c_5 = 10; c_7 = 25$

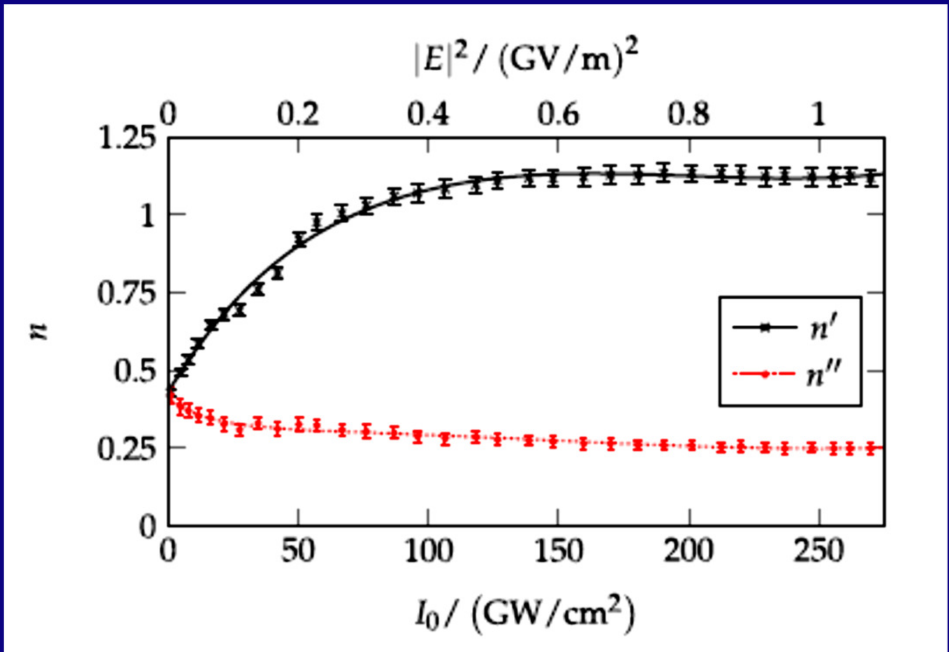
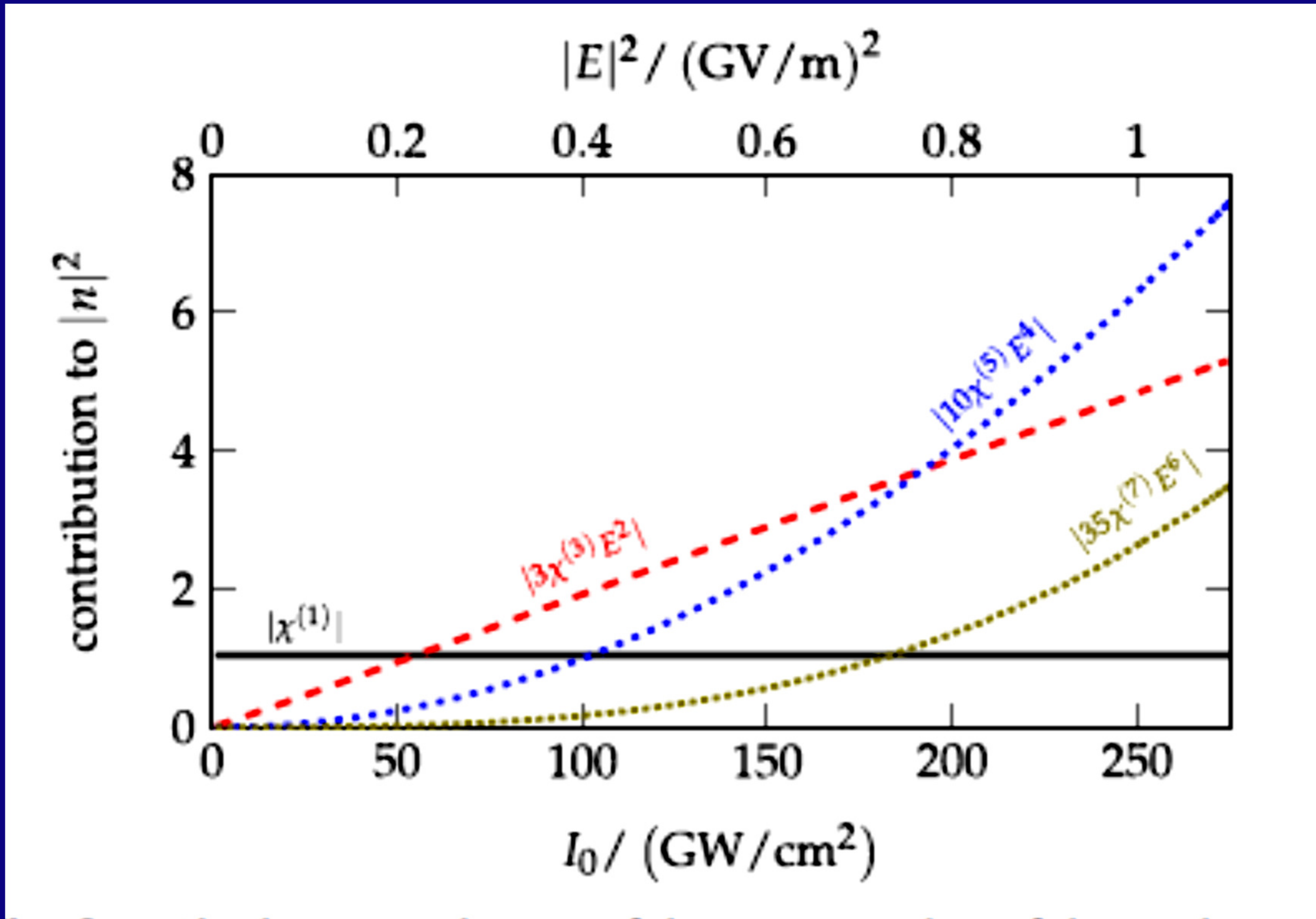


Table 1. Values Extracted from the Fit to Eq. (9) with a Third-, Fifth-, and Seventh-Order Nonlinearity

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	$(7.7 \pm 0.3) \times 10^{-2}$	$(3.5 \pm 0.8) \times 10^{-2}$

Contribuições das várias ordens da não-linearidade



Convergência da série de potencias

$$\lim_{j\to\infty}\left|\frac{c_{j+2}\chi^{(j+2)}|E|^{j+1}}{c_j\chi^{(j)}|E|^{j-1}}\right| < 1$$

Contribuição de 1ª. Ordem para a refração: (1.043 ± 0.004)

Contribuição de 5ª. Ordem para a refração: (7.6 ± 0.3)

Contribuição de 7ª. Ordem para a refração: (5.30 ± 0.09)

Contudo a razão entre $\chi^{(7)}$ e $\chi^{(5)}$ satisfaz à desigualdade

Conclusão principal

Quando estudando materiais ENZ não devemos usar a relação $n = n_0 + n_2 I$
mesmo quando só levamos em conta efeitos de $\chi^{(3)}$

$$n = \sqrt{\epsilon} = \sqrt{\epsilon^{(1)} + \sum_{j}^{\infty} c_j \chi^{(j)} |E|^{j-1}}$$

Esta expressão preserva $$n_2 \equiv \lim_{I \to 0} \frac{\partial n}{\partial I}$$

Outras referências para consulta

Iñigo Liberal and Nader Engheta

28 OPTICS & PHOTONICS NEWS JULY/AUGUST 2016
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Zero-Index Platforms

Where Light Defies Geometry

Geometry-invariant effects in structures with near-zero permittivity or permeability could lead to deformable optical devices, and to new insights into unique light-matter interactions.

nature photonics

REVIEW ARTICLE

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Near-zero refractive index photonics

Iñigo Liberal and Nader Engheta*

nature photonics

LETTERS

https://doi.org/10.1038/s41566-017-0089-9

Large optical nonlinearity of nanoantennas coupled to an epsilon-near-zero material

M. Zahirul Alam[1], Sebastian A. Schulz[1,2,3], Jeremy Upham[1], Israel De Leon[4*] and Robert W. Boyd[1,5]

Epsilon-Near-Zero Photonics: A New Platform for Integrated Devices

Xinxiang Niu, Xiaoyong Hu, Saisai Chu, and Qihuang Gong*

Adv. Optical Mater. **2018**, 6, 1701292