



What's New in Nonlinear Optics

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University of Glasgow

The visuals from my talk are posted on my website at BoydNLO.ca/presentations/

Presented at the University of Maryland, March 13, 2018.



Canada Excellence Research Chair (CERC) in Nonlinear Quantum Optics

Research interest:

Nonlinear optics, quantum optics,
integrated photonics, meta-materials, etc.

What's New in Nonlinear Optics?

1. Introduction to nonlinear optics
2. Influence of nonlinearity on rogue waves
(studied in an optical system)
3. Nonlinear optical properties of epsilon-near-zero materials

Brief Introduction to Nonlinear Optics



Why Study Nonlinear Optics?

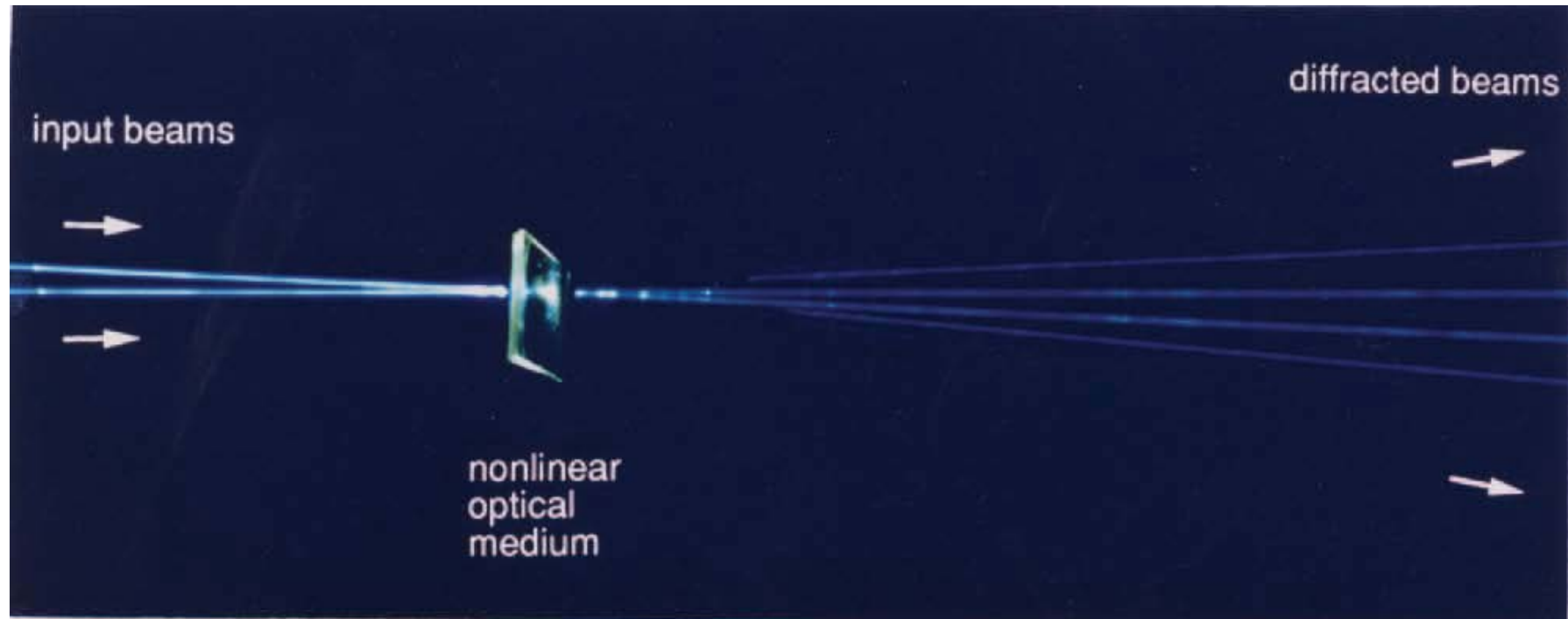
It is good fundamental physics.

It leads to important applications.

It is a lot of fun.

Demonstrate these features with examples in remainder of talk.

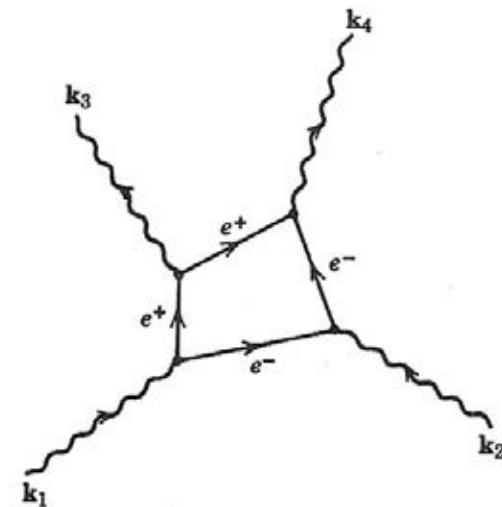
Nonlinear Optics and Light-by-Light Scattering



The elementary process of light-by-light scattering has never been observed in vacuum, but is readily observed using the nonlinear response of material systems.

Nonlinear material is fluorescein-doped boric acid glass (FBAG)

$$n_2(\text{FBAG}) \approx 10^{14} n_2(\text{silica}) \quad [\text{But very slow response!}]$$




M. A. Kramer, W. R. Tompkin, and R. W. Boyd, Phys. Rev. A, 34, 2026, 1986.

W. R. Tompkin, M. S. Malcuit, and R. W. Boyd, Applied Optics 29, 3921, 1990.

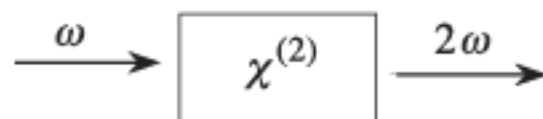
Simple Formulation of the Theory of Nonlinear Optics

$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots$$

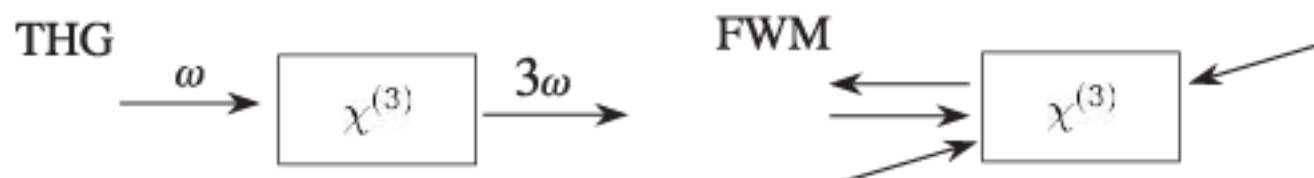
Here P is the induced dipole moment per unit volume and E is the field amplitude

$\chi^{(1)}$ describes linear optics, e.g., how lenses work: 

$\chi^{(2)}$ describes second-order effects, e.g., second-harmonic generation (SHG)



$\chi^{(3)}$ describes third-order effects such as third-harmonic generation, four-wave mixing, and the intensity dependence of the index of refraction.

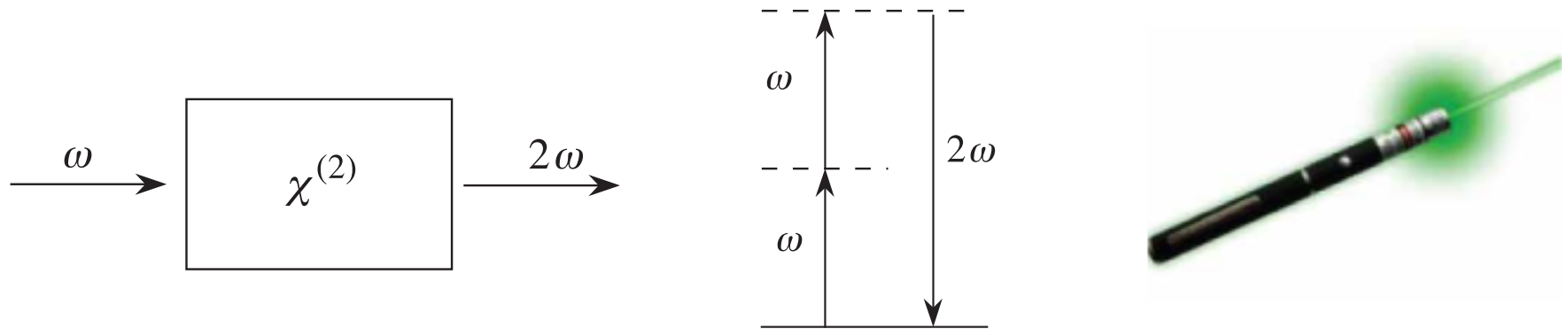


NL index

$$n = n_0 + n_2 I \quad \text{where} \quad n_2 = \frac{3}{4n_0^2 \epsilon_0 c} \chi^{(3)}$$



Second-Harmonic Generation: The Prototypical Nonlinear Optical Process



VOLUME 7, NUMBER 4

PHYSICAL REVIEW LETTERS

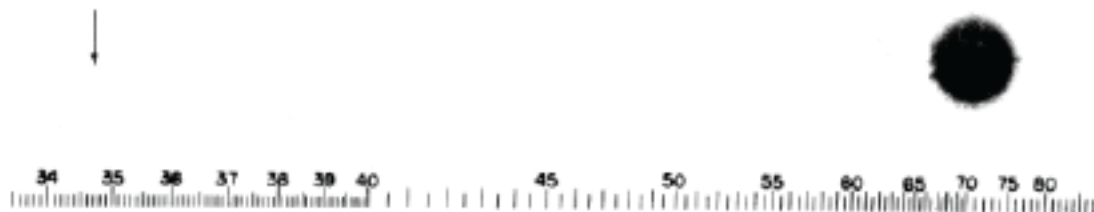
AUGUST 15, 1961

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich

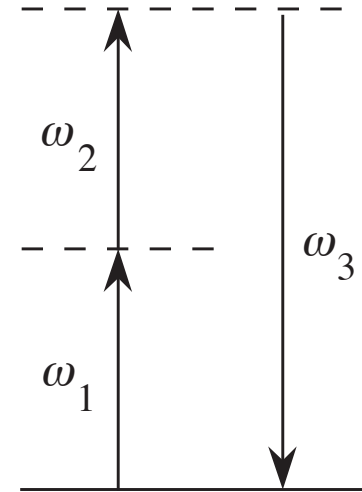
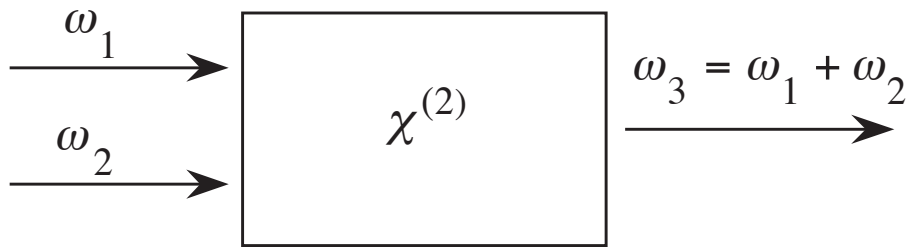
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan

(Received July 21, 1961)



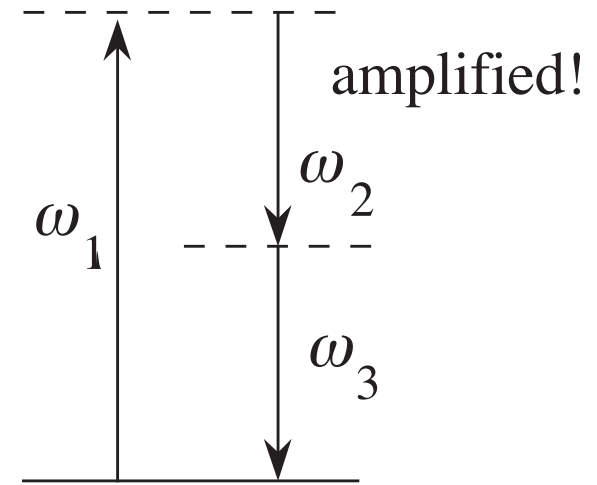
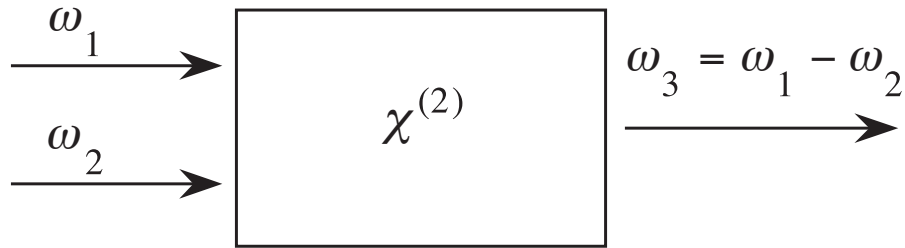
Some Fundamental Nonlinear Optical Processes: II

Sum-Frequency Generation



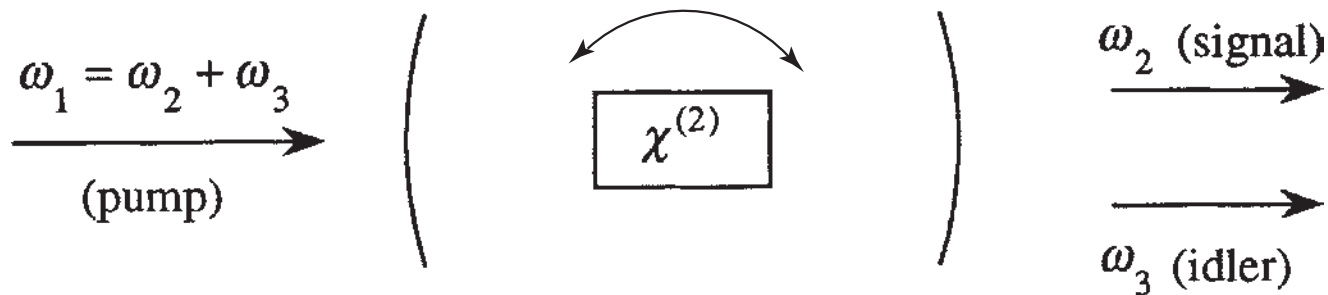
Difference-Frequency Generation and Optical Parametric Amplification

Difference-Frequency Generation



Optical Parametric Amplification

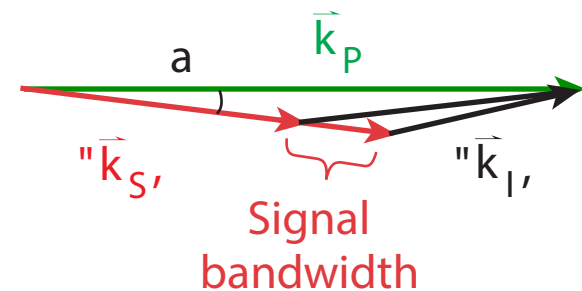
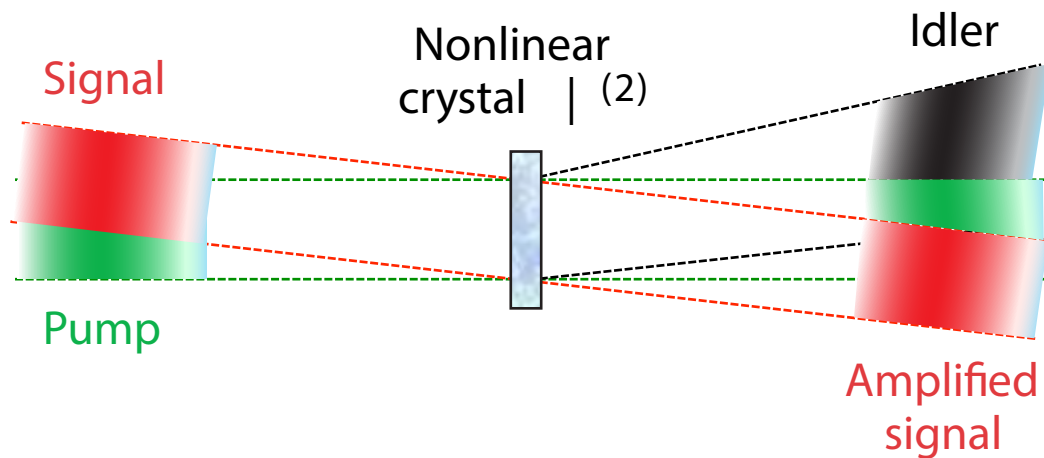
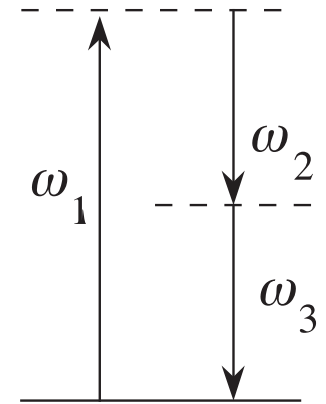
Optical Parametric Oscillator (very broadly tunable)



Optical Parametric Amplification Can Amplify Extremely Broadband Pulses

Can amplify extremely short laser pulses or broadband chirped pulses.

Goal: Design laser source capable of reaching focused intensities as large as 10^{24} W/cm².



Work of Jake Bromage and others at U. Rochester LLE.

See also Lozhkarev et al. Laser Phys. Lett. 4, 421 (2007) and Y. Tang et al. Opt. Lett. 33, 2386 (2008).



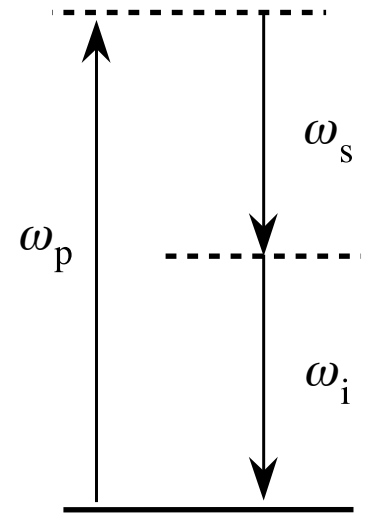
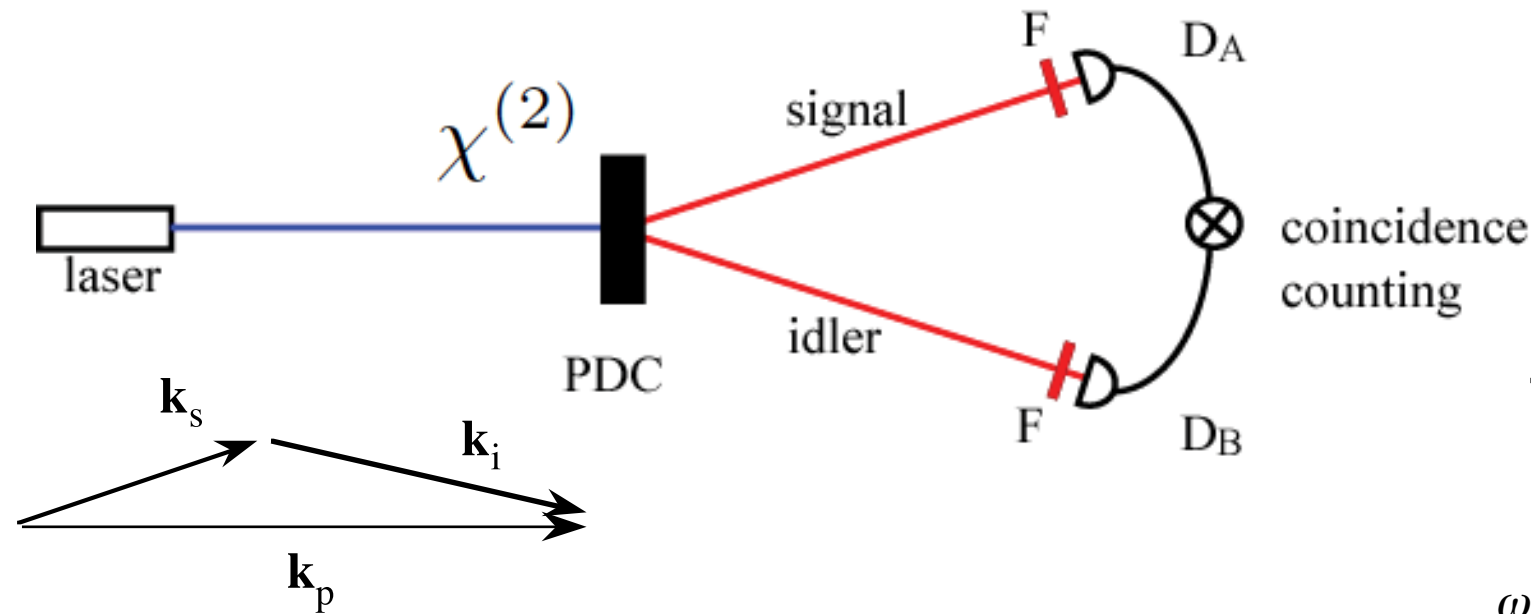
Why Interest in Quantum Nonlinear Optics?

Explore the relation between traditional nonlinear optics (NLO) and phenomena in quantum information science (QIS).

QIS holds great promise for secure communication, quantum logic, quantum computing, etc.

Many processes in QIS rely on nonlinear optical interactions.

Parametric Downconversion: A Source of Entangled Photons



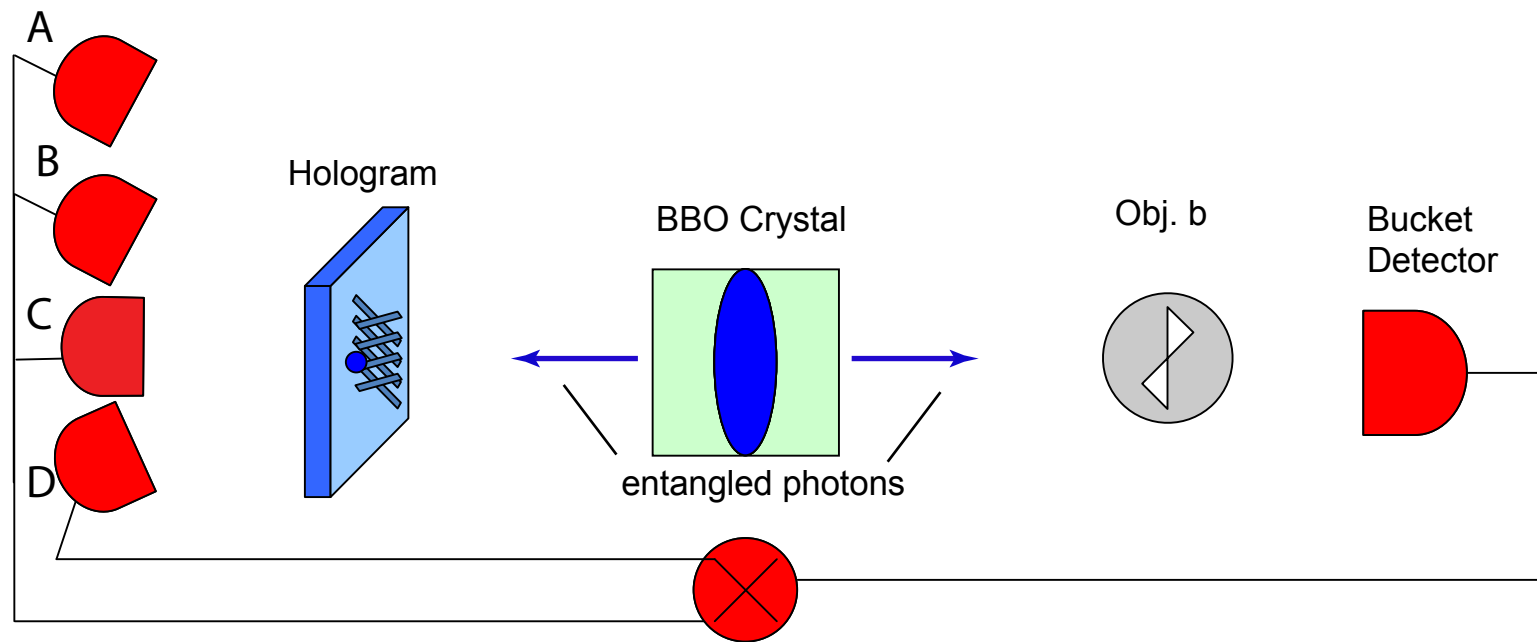
The signal and idler photons are entangled in:

- (a) polarization
- (b) time and energy
- (c) position and transverse momentum
- (d) angular position and orbital angular momentum

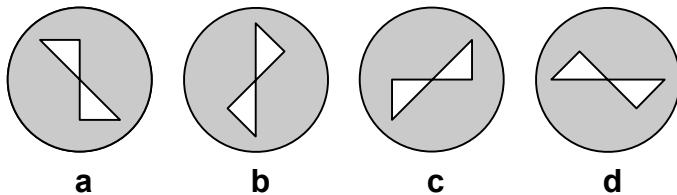
Entanglement is important for:

- (a) Fundamental tests of QM (e.g., nonlocality, Bell tests)
- (a) Quantum technologies (e.g., secure communications, Q teleportation)

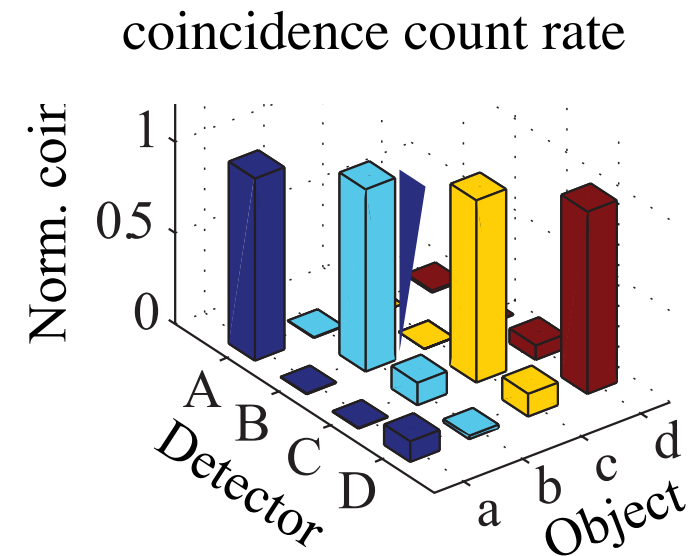
Single-Photon Coincidence Imaging



- We discriminate among four orthogonal images using single-photon interrogation in a coincidence imaging configuration.

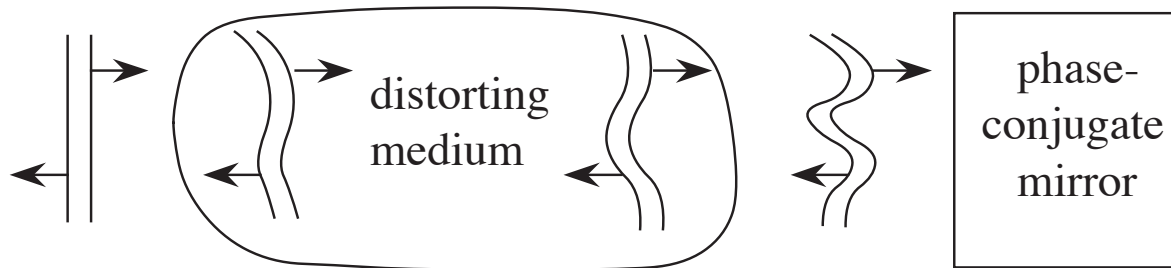


- Note that a single photon can carry more than one bit of information.



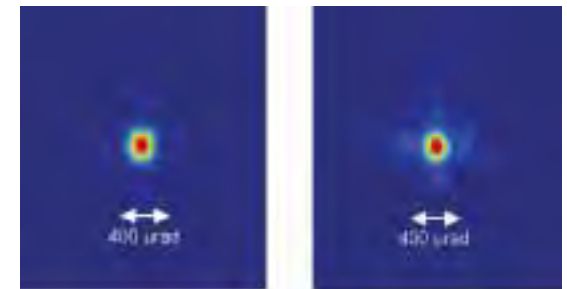
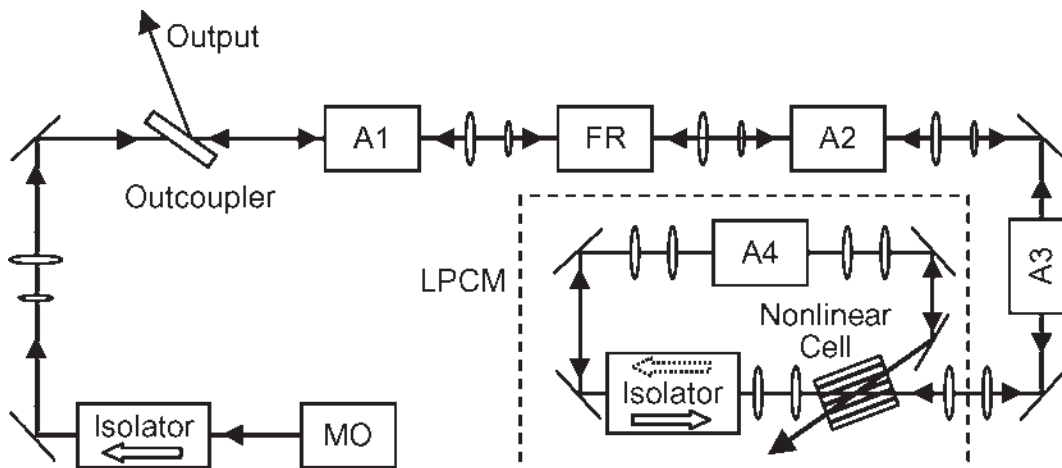
Optical Phase Conjugation: A Nonlinear Optics Success Story

- A phase conjugate mirror (a nonlinear optical device) can remove the influence of aberrations in double pass.



(Zeldovich, Pilipetsky, Shkunov, Yariv, Hellwarth, Fisher, 1980s).

- Phase conjugation is extremely useful in high power laser systems
2-kW average power phase-conjugate master oscillator power amplifier



near-diffraction-limited output

Zakharenkov, Clatterbuck, Shkunov, Betin, Filgas, Ostby, Strohkendl, Rockwell, and Baltimore, IEEE JSTQE (2007).

Theory of nonlinear optics

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

Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts

(Received April 16, 1962)

THE interaction between electromagnetic waves and atomic matter was carried out to higher orders of perturbation theory in the early years of modern quantum mechanics.¹⁻³ The interest in the absorption of two or more light quanta and scattering processes, in which three or more light quanta are involved, has recently been revived,⁴⁻⁷ because intense light fluxes available from laser sources have made possible the experimental observation of such higher order processes in the laboratory.

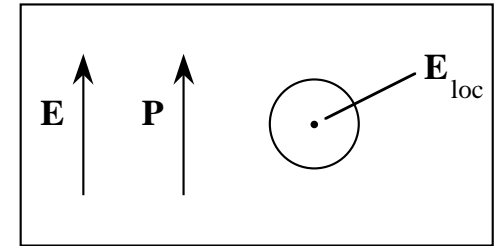


The Nobel Prize in Physics 1981

Nicolaas Bloembergen, Arthur L. Schawlow, Kai M. Siegbahn

Local Field Effects in Nonlinear Optics

Bloembergen (1962, 1965) showed that



$$\chi^{(3)}(\omega = \omega + \omega - \omega) = N\gamma^{(3)}|L(\omega)|^2[L(\omega)]^2.$$

where $\gamma^{(3)}$ is the second hyperpolarizability and where

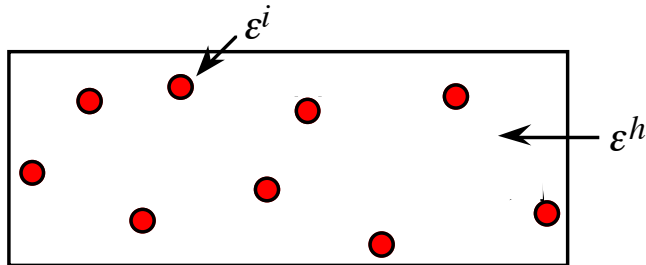
$$L(\omega) = \frac{\epsilon(\omega) + 2}{3}$$

For the typical value $n = 2$, $L = 2$, and $L^4 = 16$. Local field effects can be very large in nonlinear optics! But can we tailor them for our benefit?

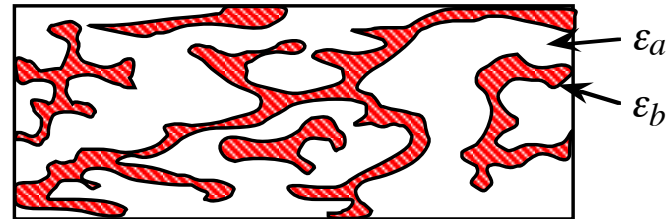
We have been developing new photonic materials with enhanced NLO response by using composite structures that exploit local field effects.

Metamaterials and Nanocomposite Materials for Nonlinear Optics

- Maxwell Garnett



- Bruggeman (interdispersed)



- Fractal Structure



- Layered



- In each case, scale size of inhomogeneity \ll optical wavelength
- Thus all optical properties, such as n and $\chi^{(3)}$, can be described by effective (volume averaged) values

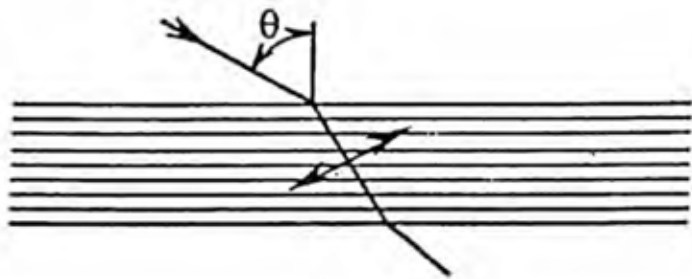
V. M. Shalaev and M. I. Stockman, Z. Phys. D 10, 71 (1988);

J. E. Sipe and R. W. Boyd, Phys. Rev. A 46, 1614 (1992).

Enhanced NLO Response from Layered Composite Materials

A composite material can display a larger NL response than its constituents!

Alternating layers of TiO_2 and the conjugated polymer PBZT.

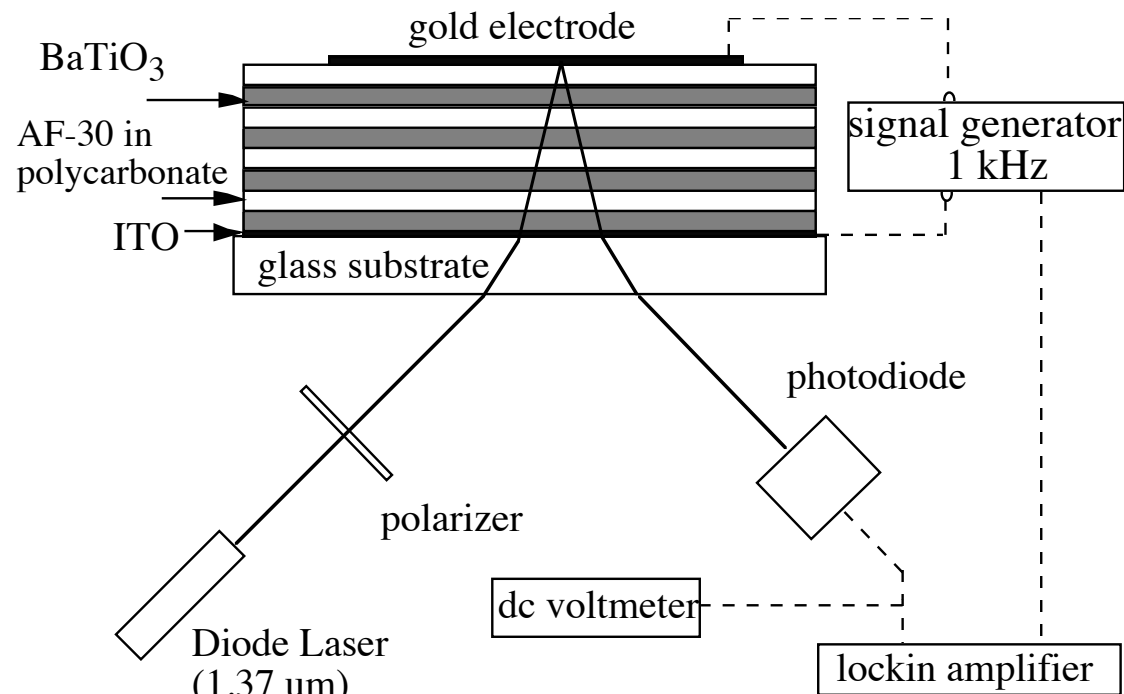


$\nabla \cdot \mathbf{D} = 0$ implies that $(\epsilon \mathbf{E})_{\perp}$ is continuous.

Measure NL phase shift as a function of angle of incidence.

35% enhancement in $\chi^{(3)}$

Quadratic EO effect



3.2 times enhancement!

Intense Field and Attosecond Physics

PHYSICAL REVIEW LETTERS

PHYSICAL REVIEW LETTERS

13 MARCH

Above Threshold Ionization Beyond the High Harmonic Cutoff

K. J. Schafer,⁽¹⁾ Baorui Yang,⁽²⁾ L. F. DiMauro,⁽²⁾ and K. C. Kulander⁽¹⁾

⁽¹⁾Lawrence Livermore National Laboratory, Livermore, California 94550

⁽²⁾Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973

(Received 2 December 1992)

VOLUME 71, NUMBER 13

PHYSICAL REVIEW LETTERS

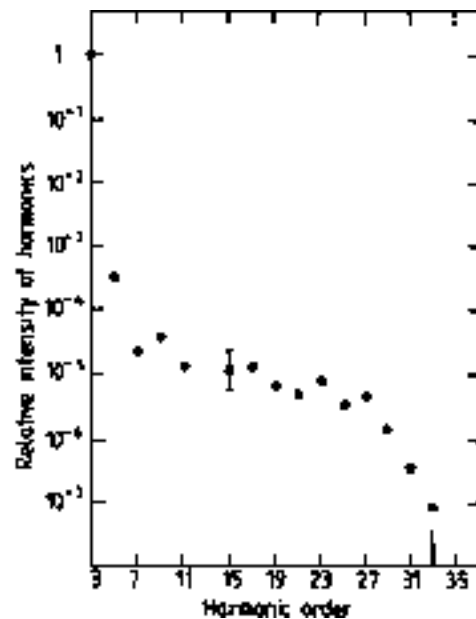
27 SEPTEMBER 1993

Plasma Perspective on Strong-Field Multiphoton Ionization

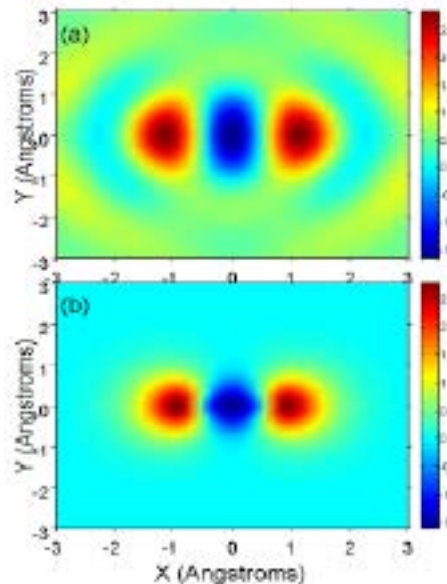
P. B. Corkum

National Research Council of Canada, Ottawa, Ontario, Canada K1A 0R6

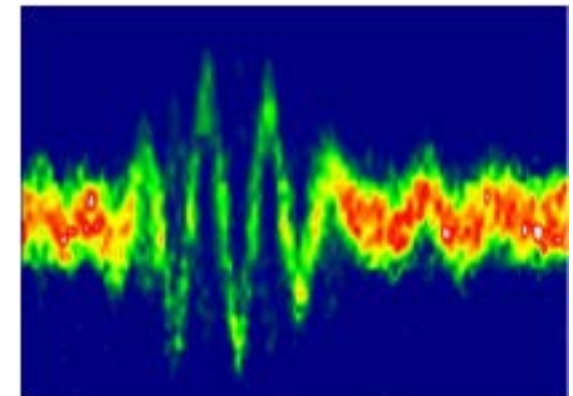
(Received 9 February 1993)



High-harmonic generation



Measuring the molecular nitrogen wavefunction



Attosecond pulses to sample a visible E-field; F. Krausz

Influence of Nonlinearity on the Development of Rogue Waves

Study in a well-characterized optical system

Is nonlinearity necessary? Required? Or does it inhibit rogue waves?

A. Safari, R. Fickler, M. J. Padgett and R. W. Boyd, Phys. Rev. Lett. 119, 203901 (2017).

Oceanic rogue waves



uOttawa

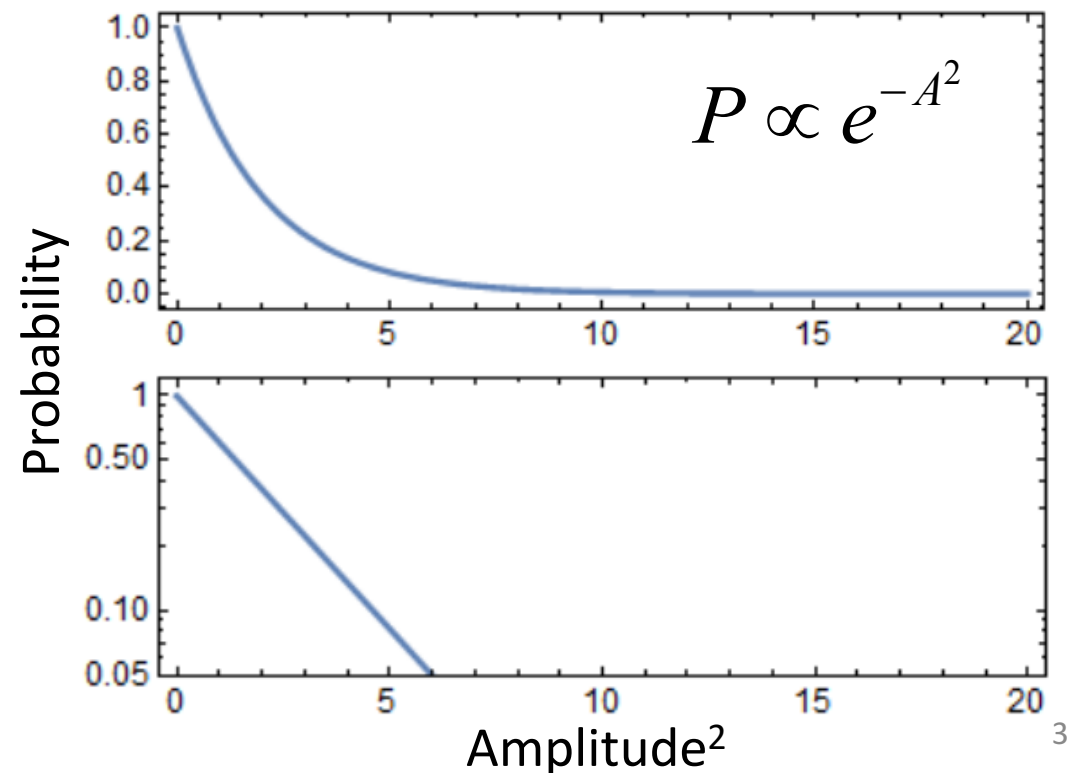


Before 1995

Sailors: we see gigantic waves.

Scientists: it is a fairy tail!

Ocean waves follow
Gaussian distribution.



Oceanic rogue waves



uOttawa

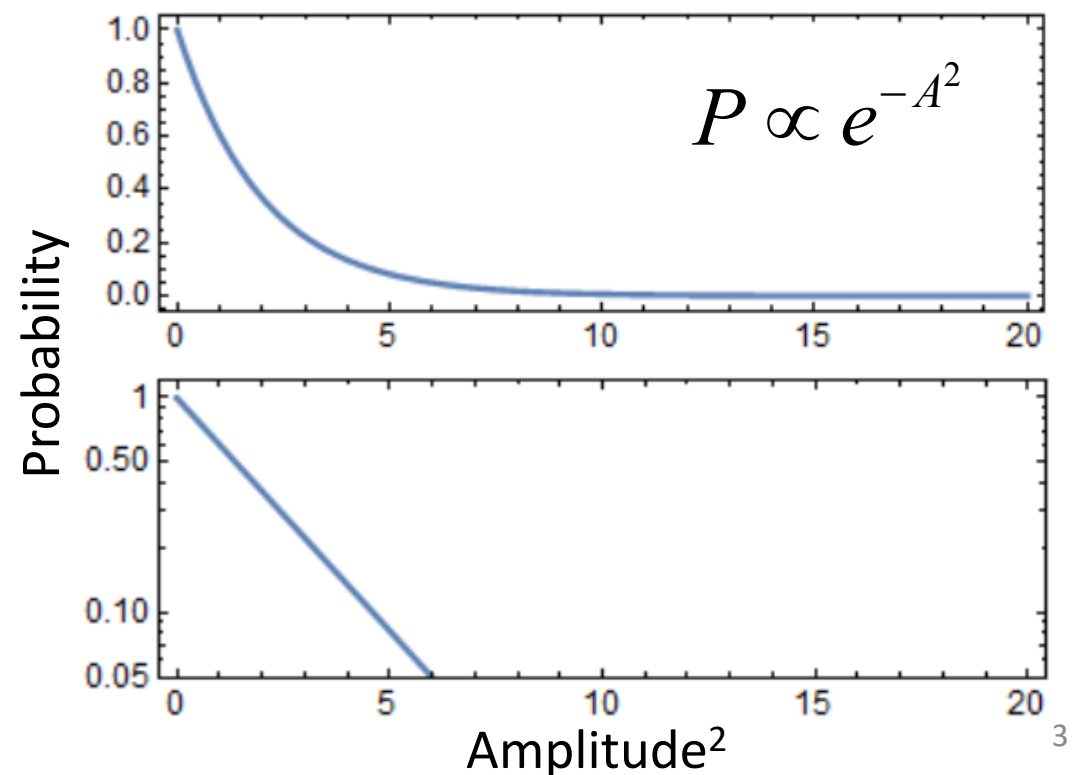


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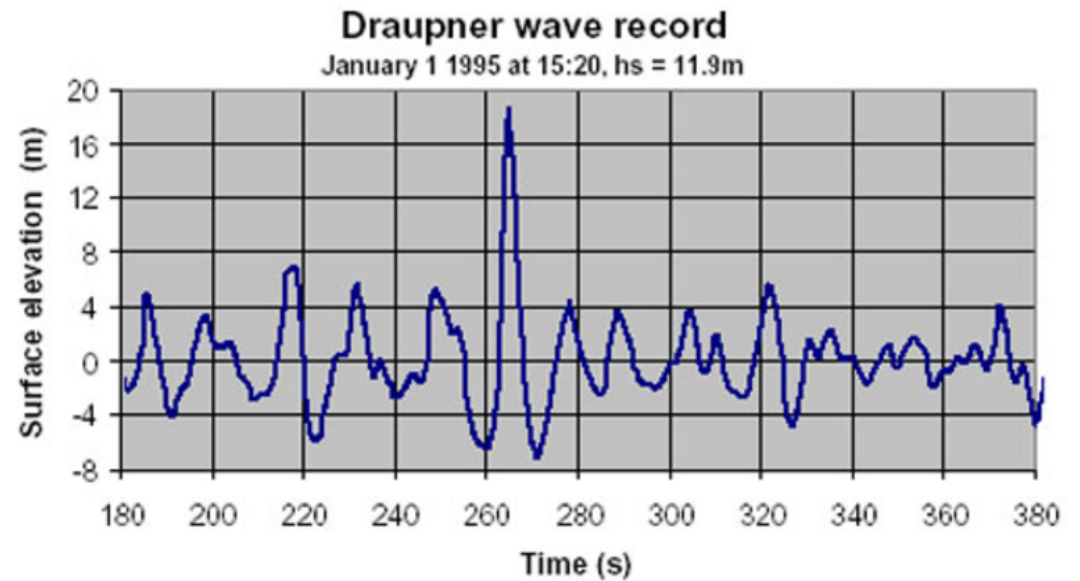
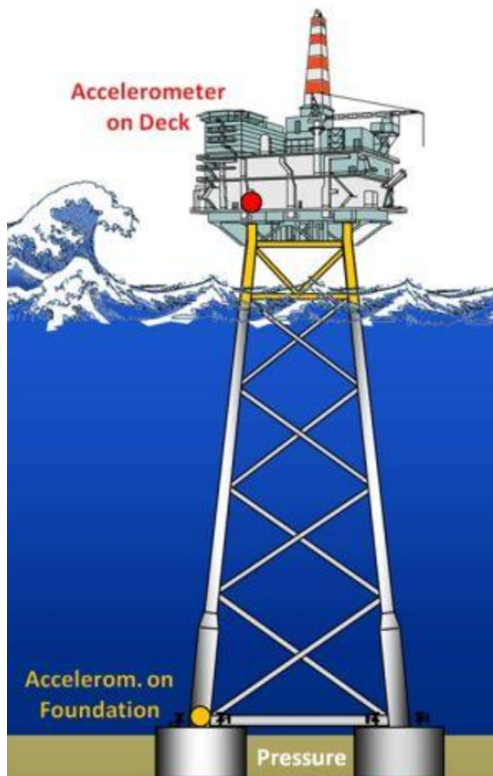


Oceanic rogue waves



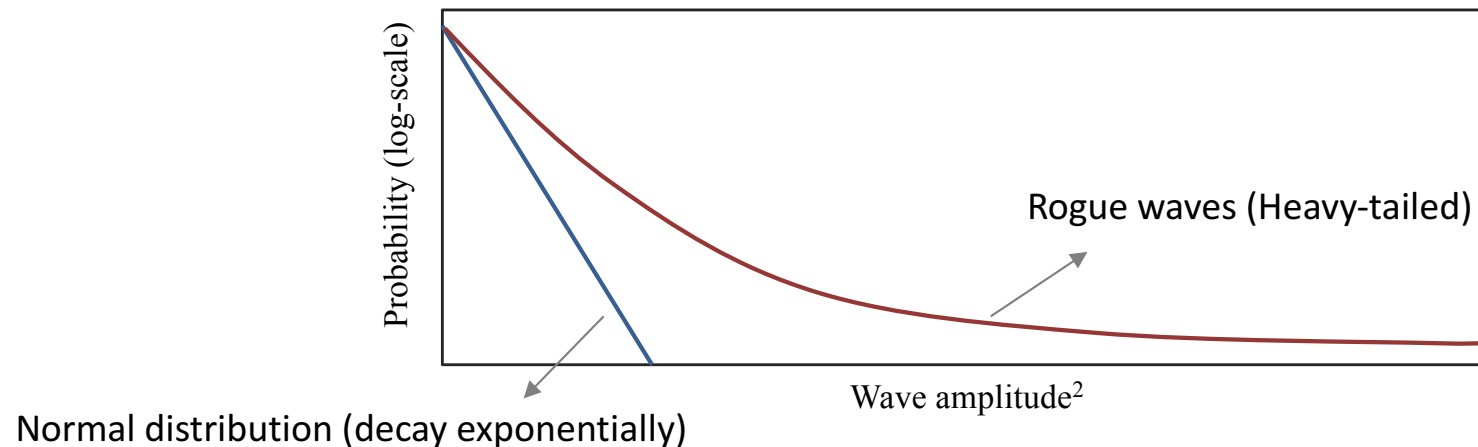
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First scientific observation
of rogue waves in Draupner
oil platform (1995):



- Rogue waves appear from nowhere and disappear without a trace.
- Rogue waves \neq accidental constructive interference
- They occur much more frequently than expected in ordinary wave statistics.

Probability distribution in rogue systems:



- Not limited to ocean: Observed in many other wave systems including **optics**.

“Nonlinear Schrödinger equation” explains the wave dynamics in the ocean as well as in optics.

Rogue events studied extensively in 1D systems, such as optical fibers.

$$\frac{\partial A}{\partial x} + \frac{1}{2} i k_2 \frac{\partial^2 A}{\partial t^2} = i \gamma |A|^2 A$$

D. R. Solli, C. Ropers, P. Koonath & B. Jalali, Nature 450, 1054 (2007).

J.M. Dudley et al, Nat. Photon, 8, 755 (2014)

Water waves are not 1D.

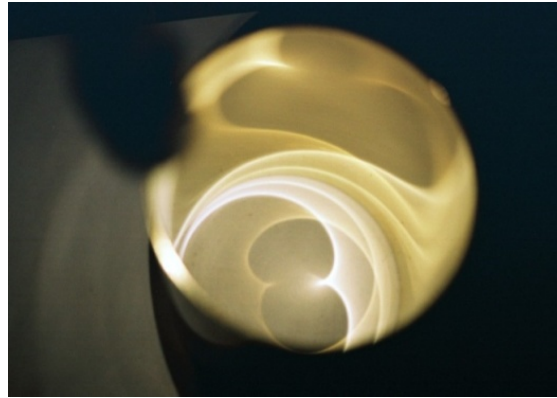
$$2ik \frac{\partial A}{\partial x} + \nabla_{\perp}^2 A = i \gamma |A|^2 A$$

Two focusing effects in 2D systems:

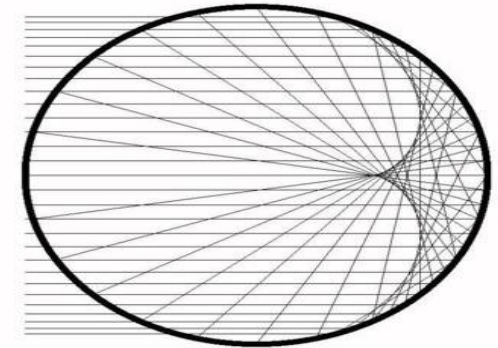
- **Linear:** Spatial (geometrical) focusing
- **Nonlinear:** Self focusing



Swimming pool



Coffee cup



Ray picture

- Caustics are defined as envelope of a family of rays
- Singularities in ray optics
- Catastrophe theory is required to remove singularity

Books:

J.F. Nye, *Natural Focusing and Fine Structure of Light*.

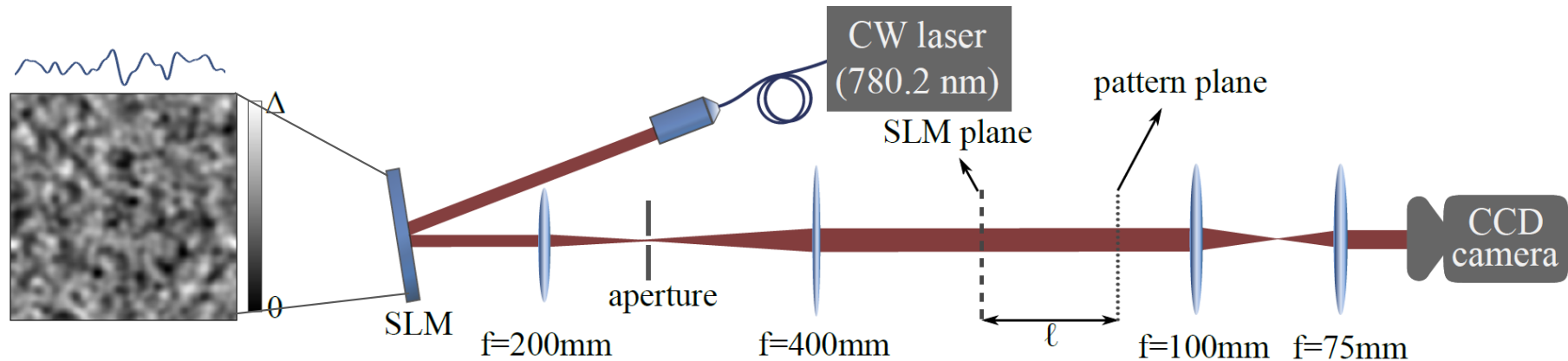
Y.A. Kravtsov, *Caustics, Catastrophes and Wave Fields*.

O.N. Stavroudis, *The Optics of Rays, Wavefronts, and Caustics*.

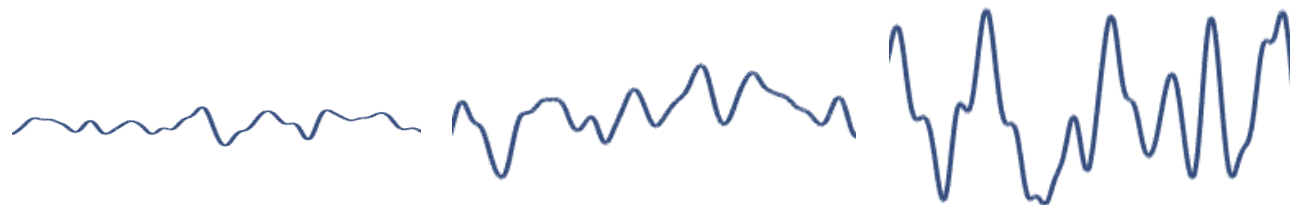
Generation of optical caustics



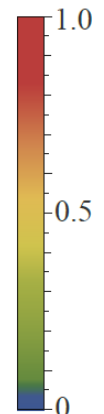
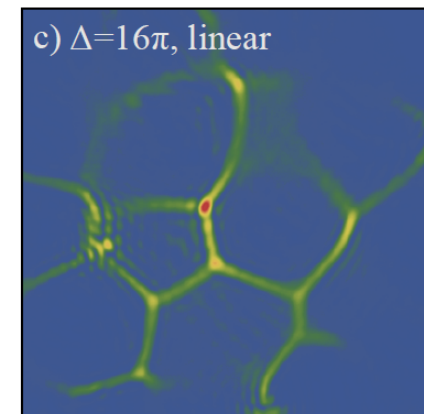
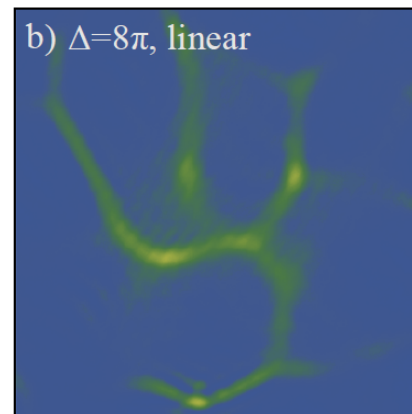
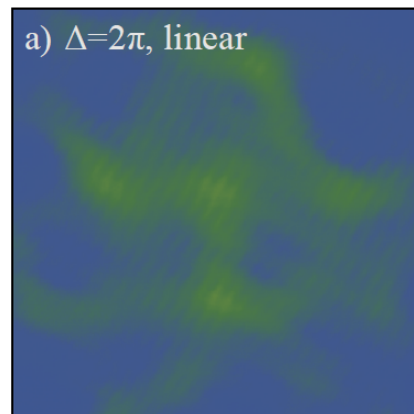
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Phase variations:



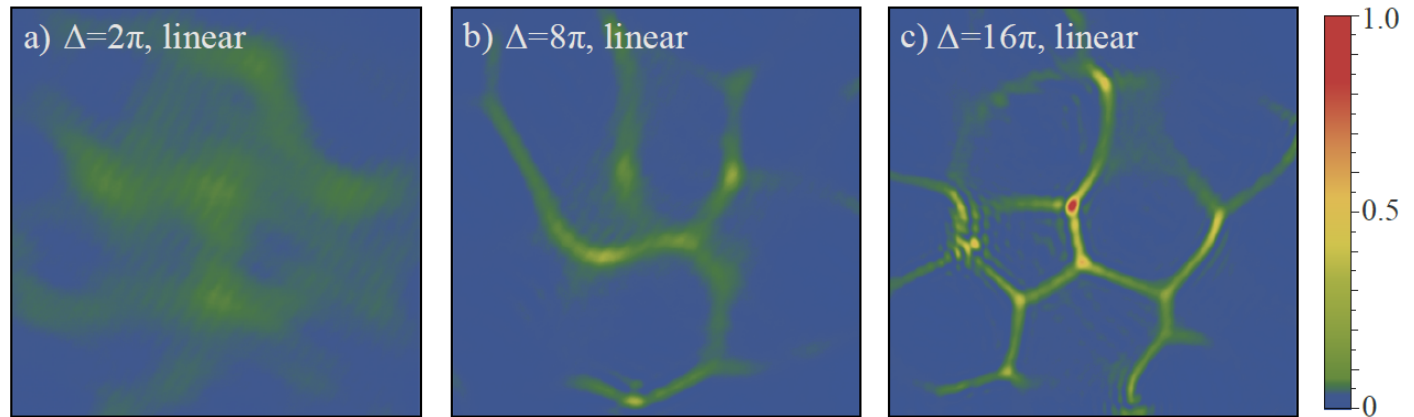
Corresponding
intensity
variations:



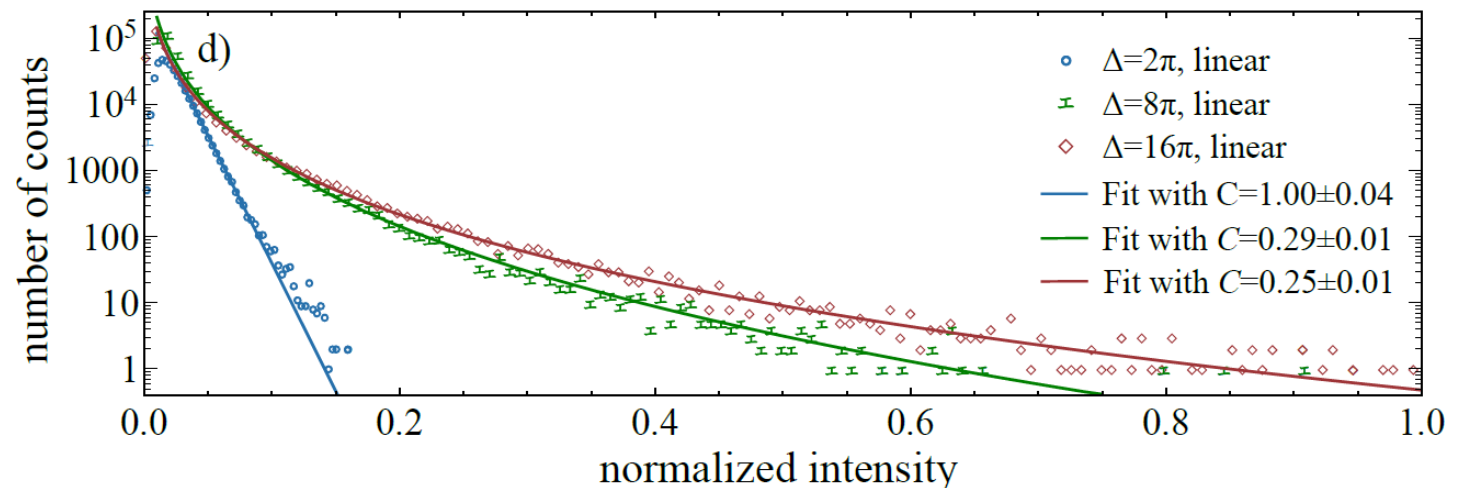
A sharp caustic is formed only if the phase variations are large

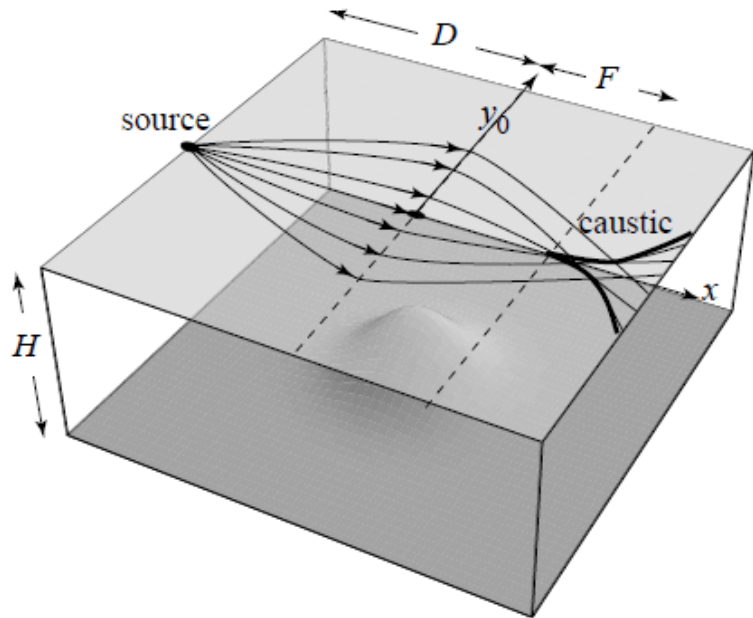
Caustics exhibit long-tailed probability distribution

1000 different patterns for each Δ



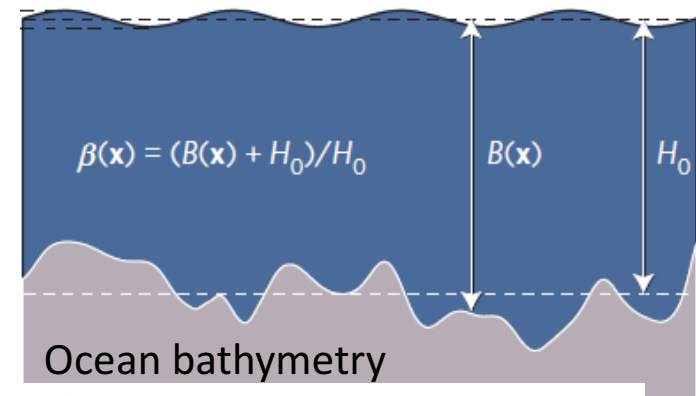
Intensity distributions with fit to $A \text{Exp}(-B I^C)$



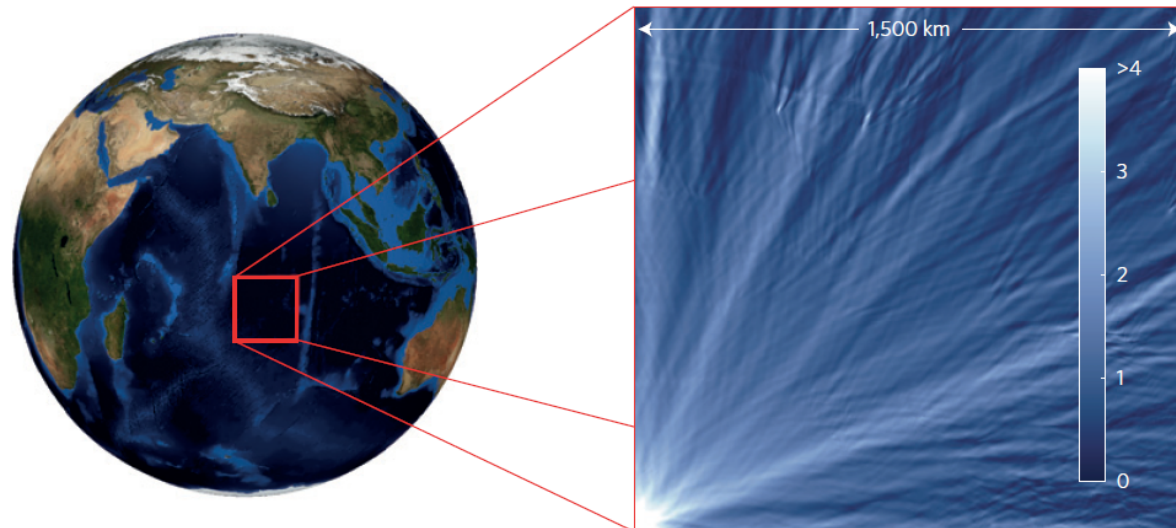


Caustic of tsunami focused by an underwater island lens

M. V. Berry, Focused tsunami waves, *Proc. R. Soc. A* (2007)



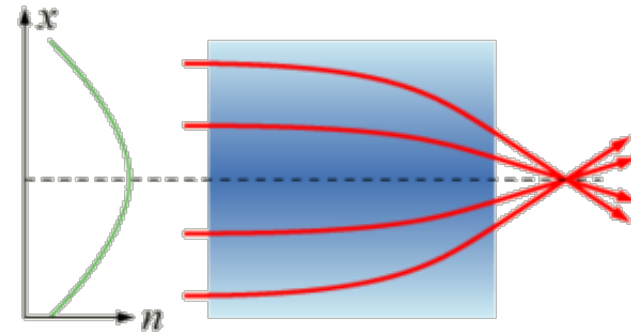
Simulated linear propagation of tsunami waves, using real ocean floor bathymetry:



Self focusing:

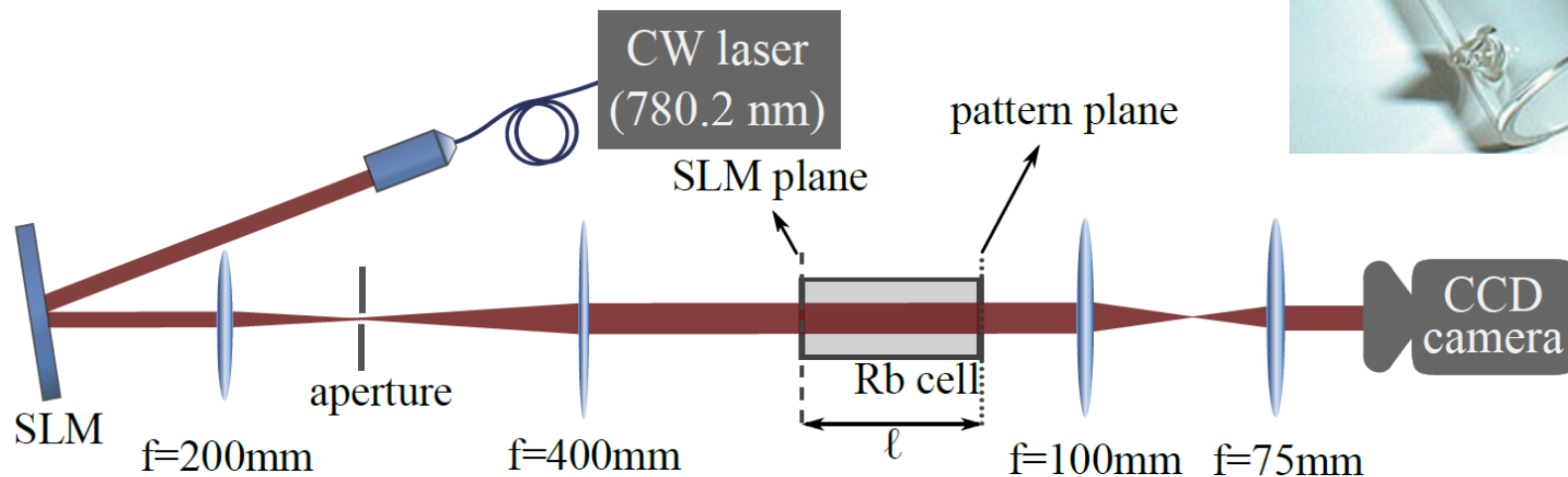
Refractive index depends on intensity:

$$n = n_0 + n_2 I$$



Rubidium vapors show large nonlinear effects

Rubidium cell



Effect of nonlinearity on caustics

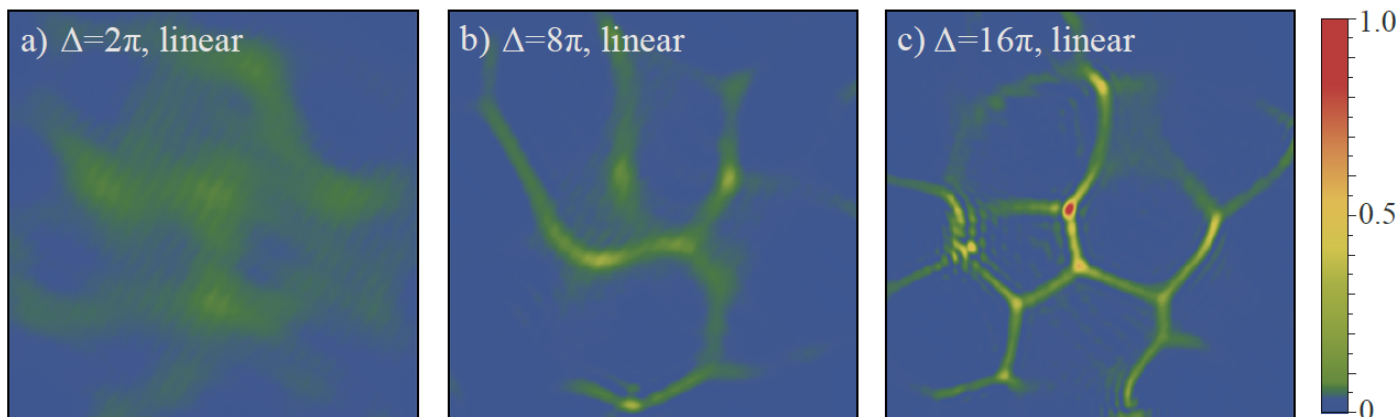


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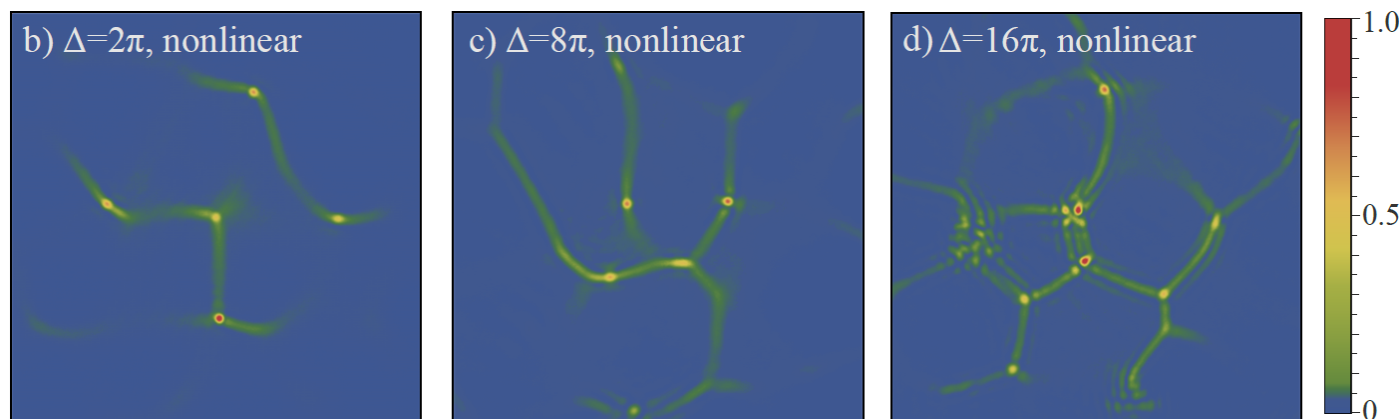
Phase variations:



After linear
propagation:

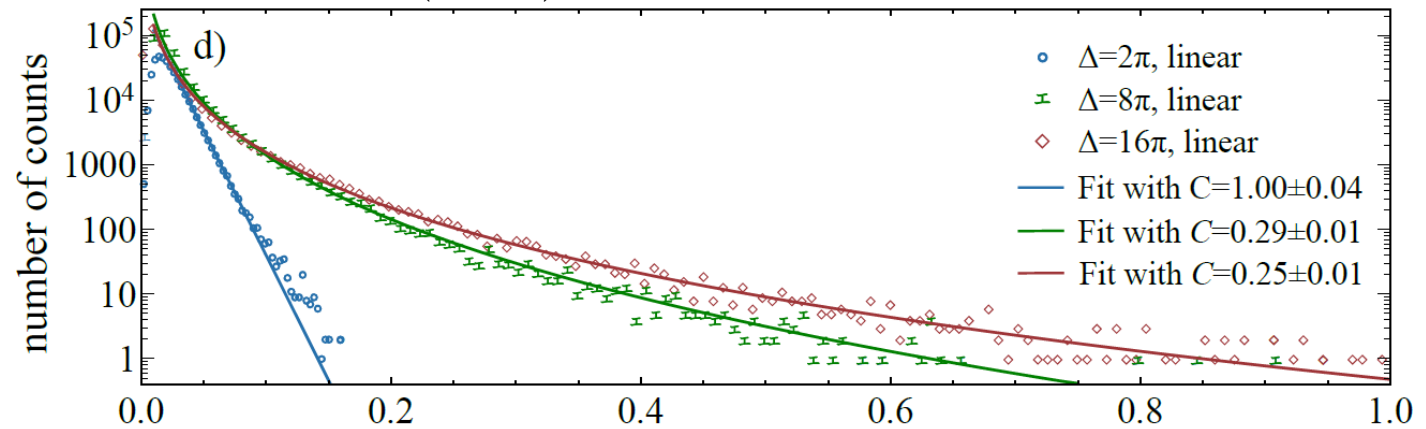


After nonlinear
propagation:

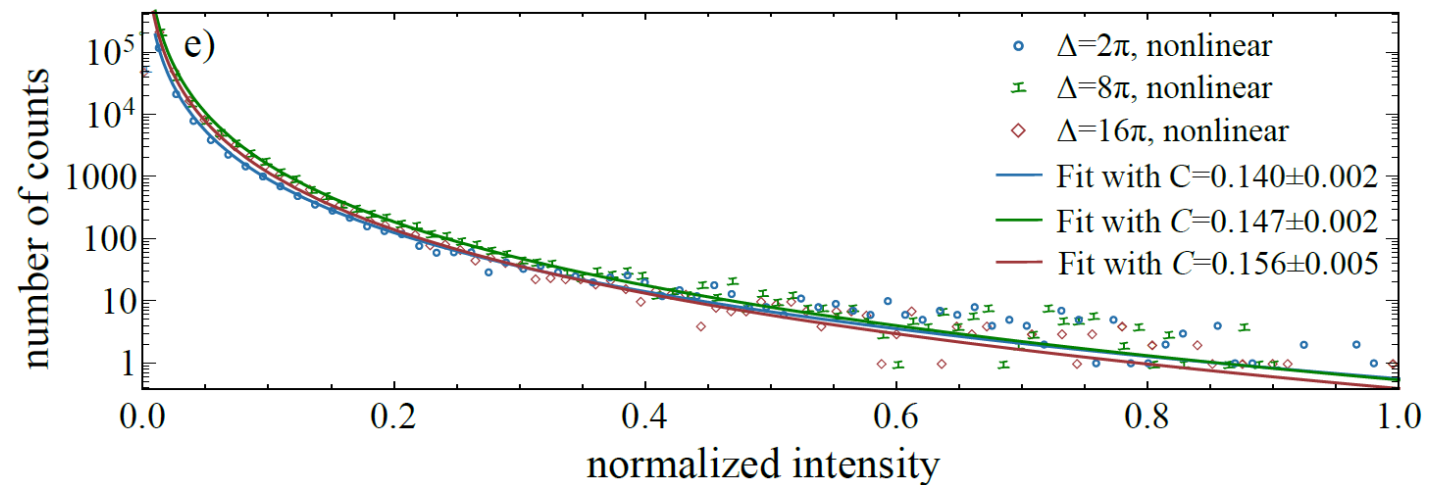


Intensity distributions with fit to $A \text{Exp}(-B I^C)$

Linear
propagation:

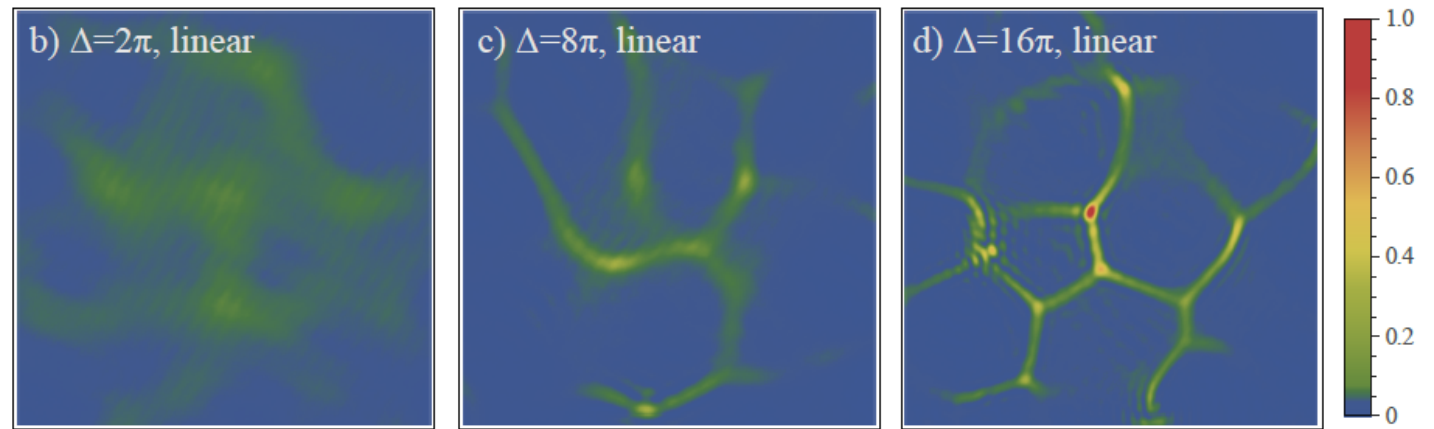


After nonlinear
propagation:

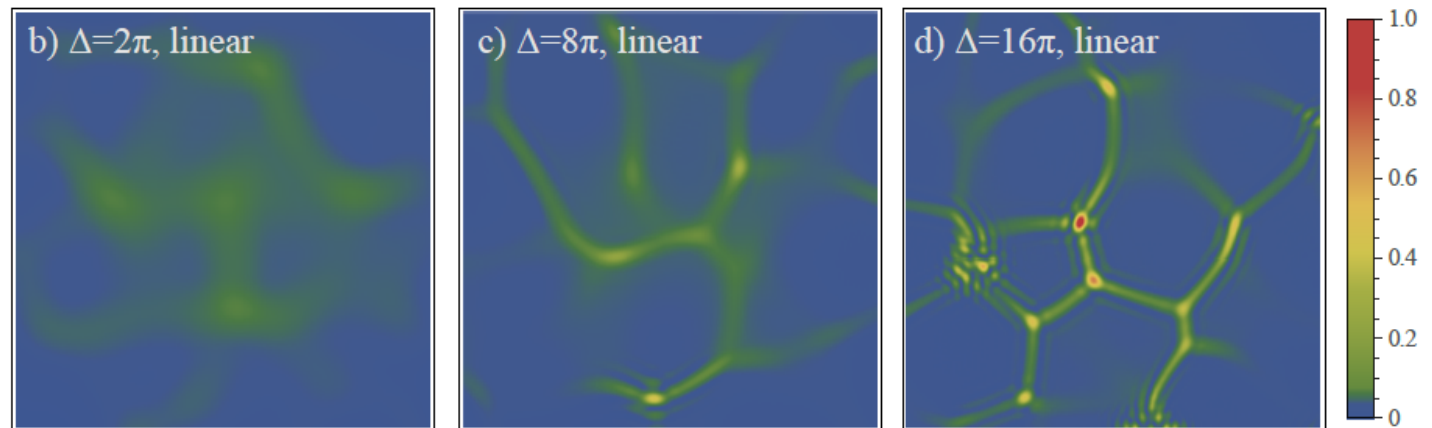


Linear propagation was simulated by FFT beam propagation

Experiment:



Simulation:

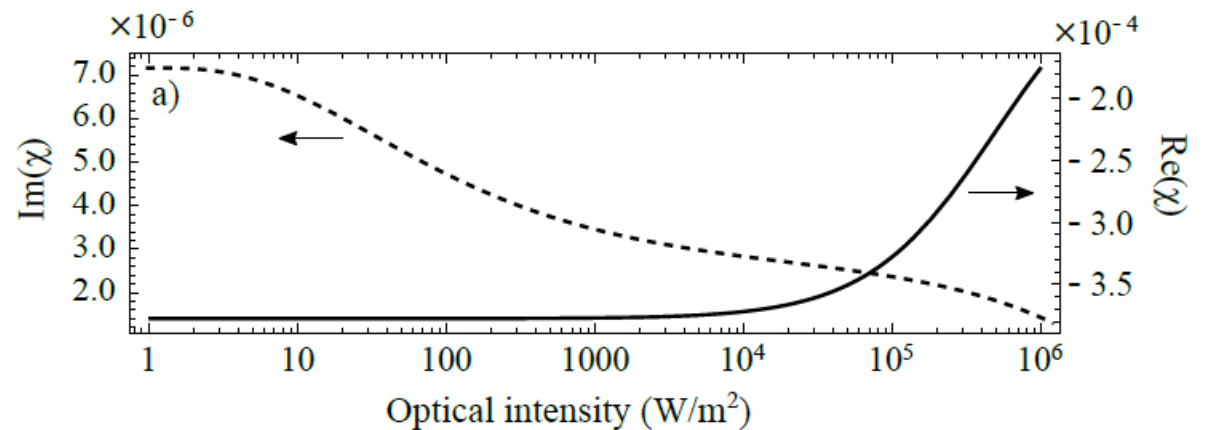
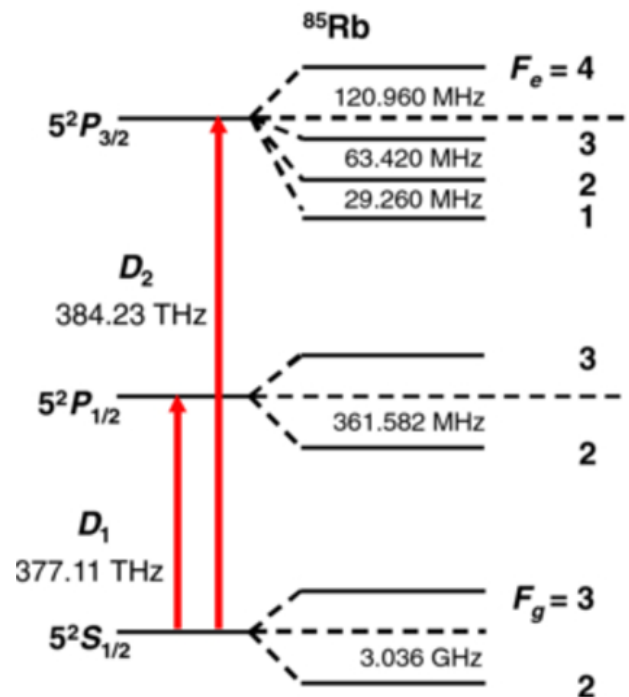
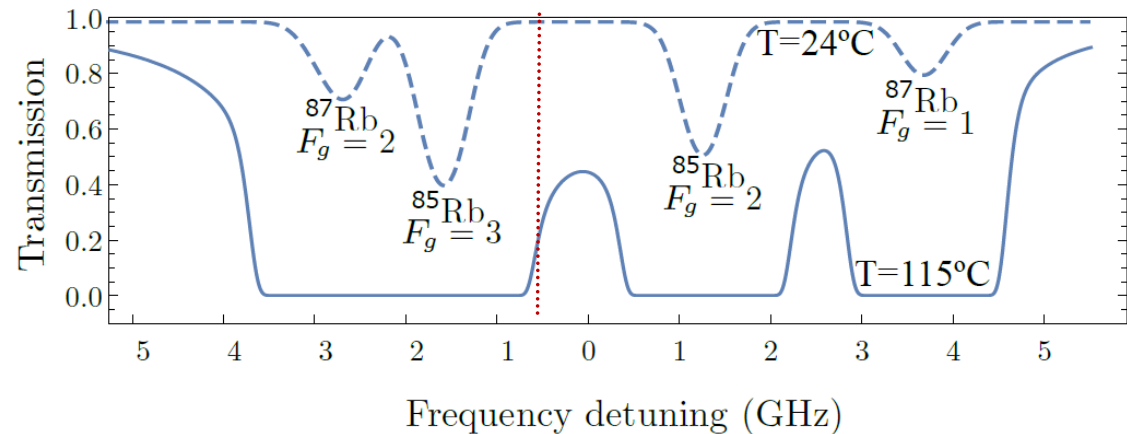


$$\text{NLSE: } \frac{\partial \mathcal{E}}{\partial z} - \frac{i}{2k} \nabla_{\perp}^2 \mathcal{E} = \frac{ik}{2\epsilon_0} P$$

$$\text{Atomic polarization: } P = \epsilon_0 \chi \mathcal{E}$$

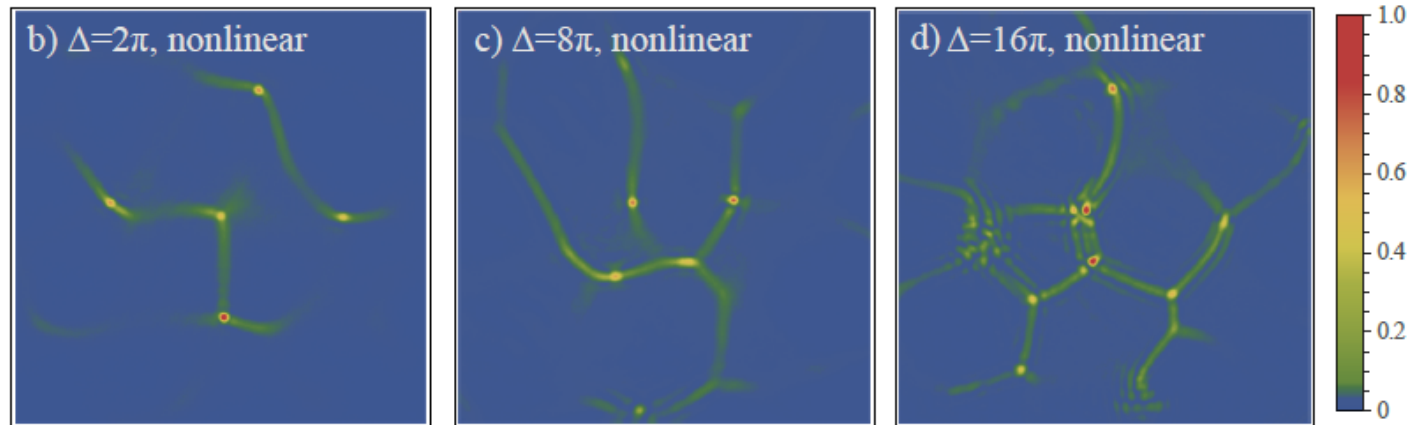
Our Rb model includes:

- All hyperfine transitions
- Doppler broadening
- Power broadening
- Collisional broadening
- Optical pumping

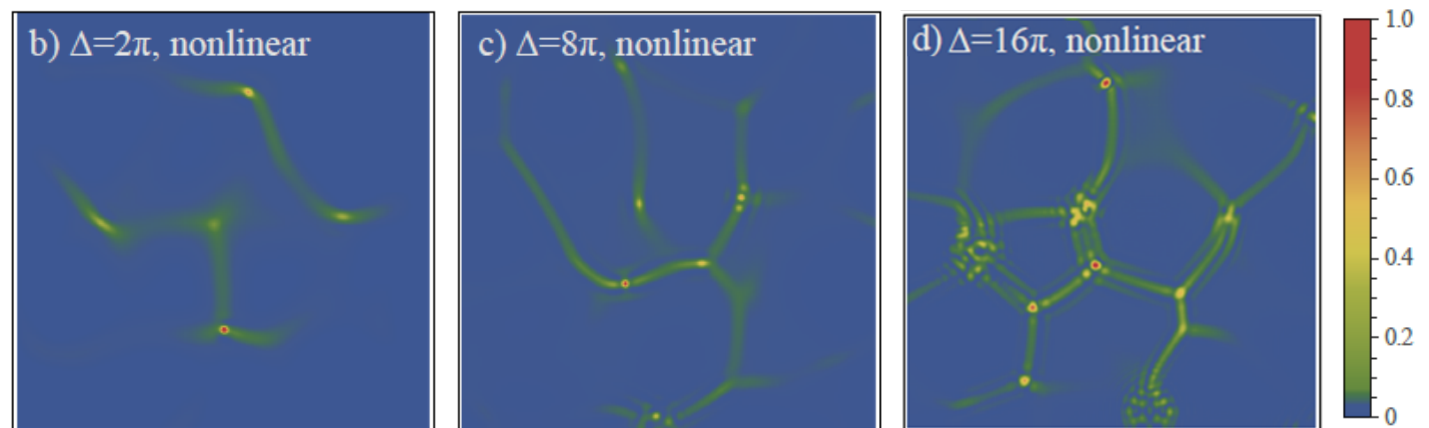


Nonlinear propagation was simulated by FFT beam propagation and split-step

Experiment:



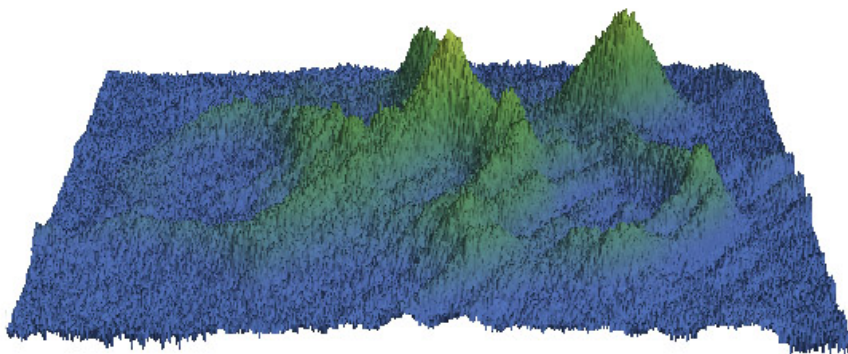
Simulation:



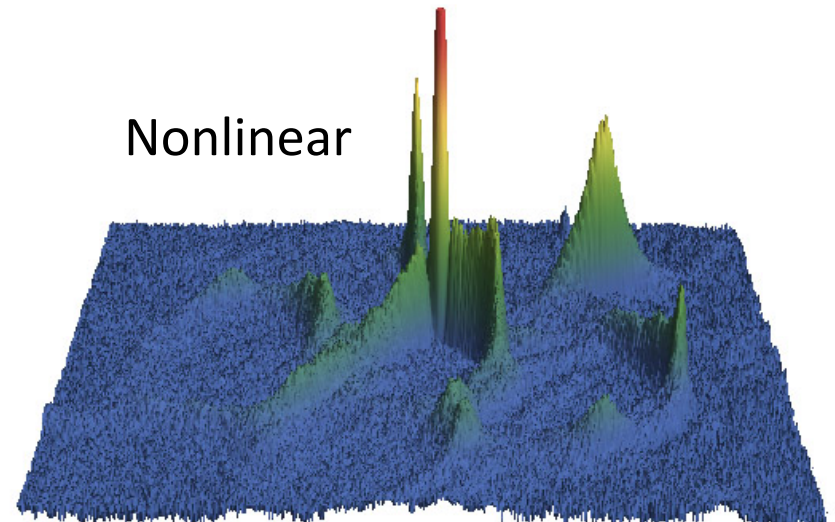
**A. Safari, R. Fickler, M. Padgett, R. Boyd ,
Physical Review Letters 119, 203901 (2017)**

- Caustics are rogue waves!
- Generation of caustics by linear propagation requires large phase fluctuations
- Nonlinear effects can enhance the generation of caustics.

Linear



Nonlinear



Epsilon-Near-Zero (ENZ) Materials

- Physics of Epsilon-Near-Zero (ENZ) Materials
- Huge NLO Response of ENZ Materials and Metastructures
- Non-perturbative nature of the NLO Response (usual power series do not converge)

With Special Thanks To:

M. Zahirul Alam, Orad Reshef, Enno Giese, and Jeremy Upham, University of Ottawa
Israel De Leon, Tecnologico de Monterrey, Mexico
Sebastian Schulz, Cork Institute of Technology, Ireland

Physics of Epsilon-Near-Zero (ENZ) Materials

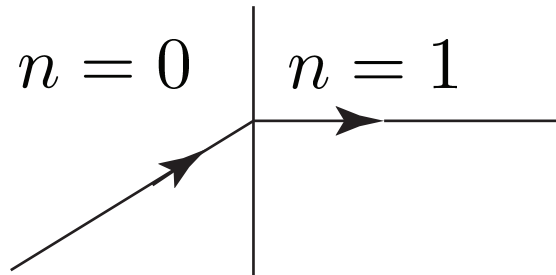
- ENZ materials possess exotic electromagnetic properties
Silveirinha, Engheta, Phys. Rev. Lett. 97, 157403, 2006.
- If the dielectric permittivity ϵ is nearly zero, then refractive index n nearly zero
Thus $v_{\text{phase}} = c/n$ is nearly infinite
 $\lambda = \lambda_{\text{vac}} / n$ is nearly infinite
Light oscillates in time but not in space; everything is in phase
Light “oscillates” but does not “propagate.”
- Radiative processes are modified in ENZ materials
Einstein A coefficient (spontaneous emission lifetime = $1/A$)
 $A = n A_{\text{vac}}$
We can control (inhibit!) spontaneous emission!
Einstein B coefficient
Stimulated emission rate = B times EM field energy density
 $B = B_{\text{vac}} / n^2$
Optical gain is very large!
Einstein, Physikalische Zeitschrift 18, 121 (1917).
Milonni, Journal of Modern Optics 42, 1991 (1995).

Physics of Epsilon-Near-Zero (ENZ) Materials -- More

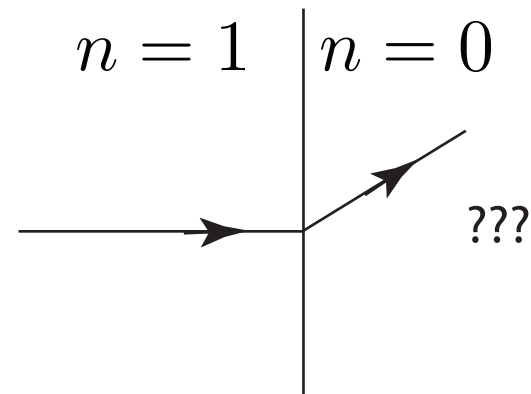
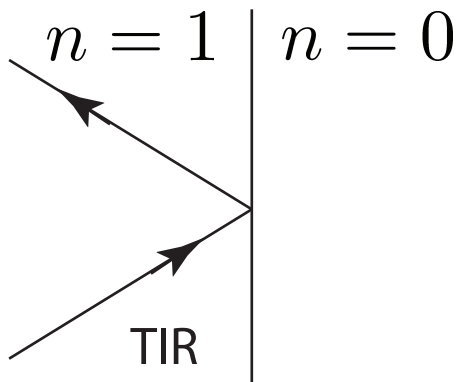
- Snell's law leads to intriguing predictions

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

- Light always leaves perpendicular to surface of ENZ material!

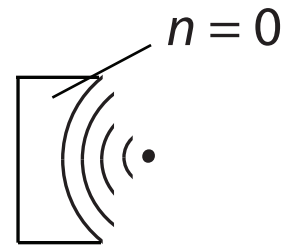
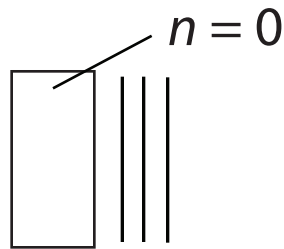


- Thus light can enter an ENZ material only at normal incidence!

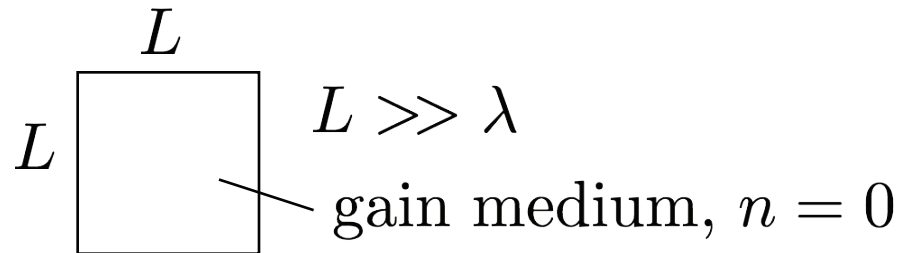


Some Consequences of ENZ Behaviour - 1

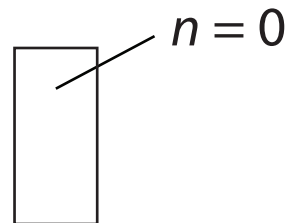
- Funny lenses



- Large-area single-transverse-mode surface-emitting lasers

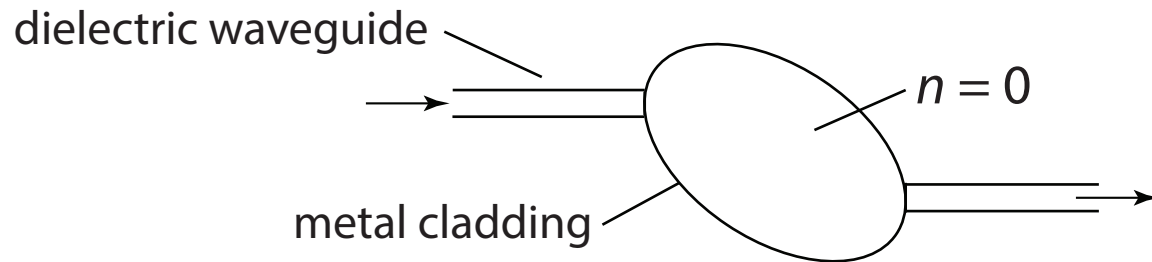


- No Fabry-Perot interference

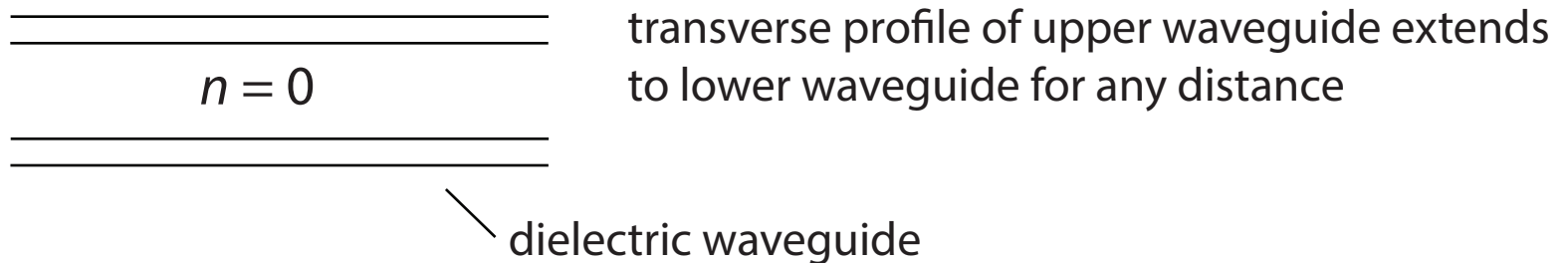


Some Consequences of ENZ Behaviour - 2

- Super-coupling (of waveguides)



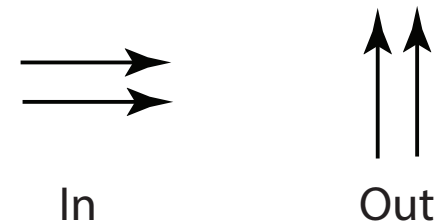
- Large evanescent tails for waveguide coupling



- Automatic phase matching of NLO processes

Recall that $k = n \omega / c$ vanishes in an ENZ medium.

For example, the following 4WM process is allowed



Some Consequences of ENZ Behaviour - 3

- How is the theory of self-focusing modified?
- Does the theory of Z-scan need to be modified?
- How is the theory of blackbody radiation modified?
- Do we expect very strong superradiance effects?
- More generally, how is any NLO process modified when $n_0 = 0$?

Epsilon-Near-Zero Materials

- Metamaterials

Materials tailor-made to display ENZ behaviour

- Homogeneous materials

All materials display ENZ behaviour at their (reduced) plasma frequency

Recall the Drude formula

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

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

- Challenge: Obtain low-loss ENZ materials

Want $\text{Im } \epsilon$ as small as possible at the frequency where $\text{Re } \epsilon = 0$.

- We are examining a several materials

ITO: indium tin oxide

AZO: aluminum zinc oxide

FTO: fluorine tin oxide

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).

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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 $\chi^{(3)}$

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

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 $1.24 \mu\text{m}$.

Recall the Drude formula

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

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

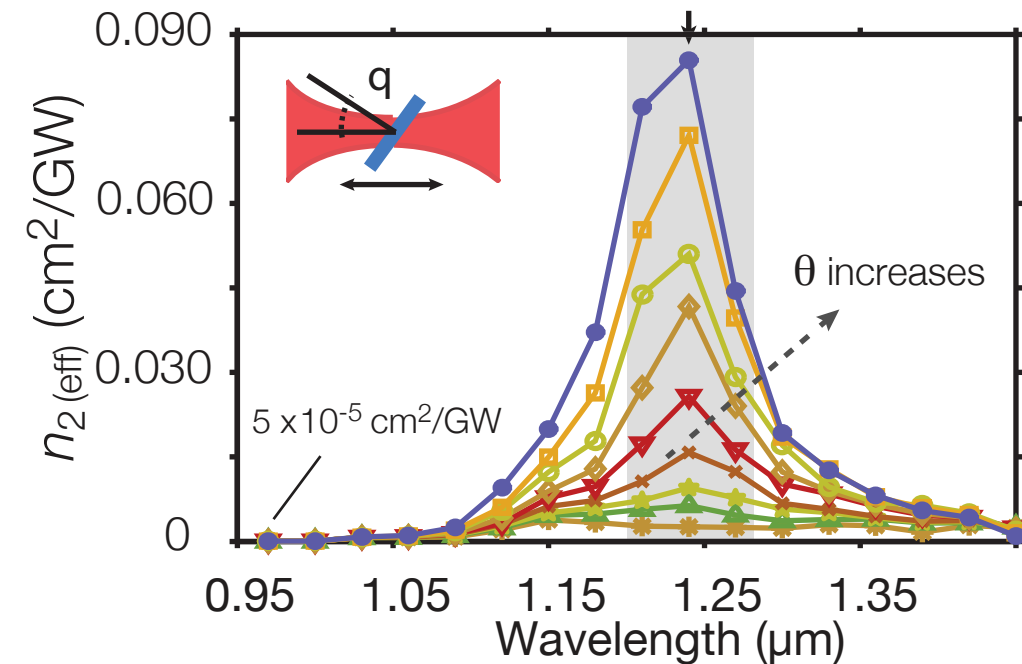
The region near ω_0 is known as the epsilon-near-zero (ENZ) region.

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

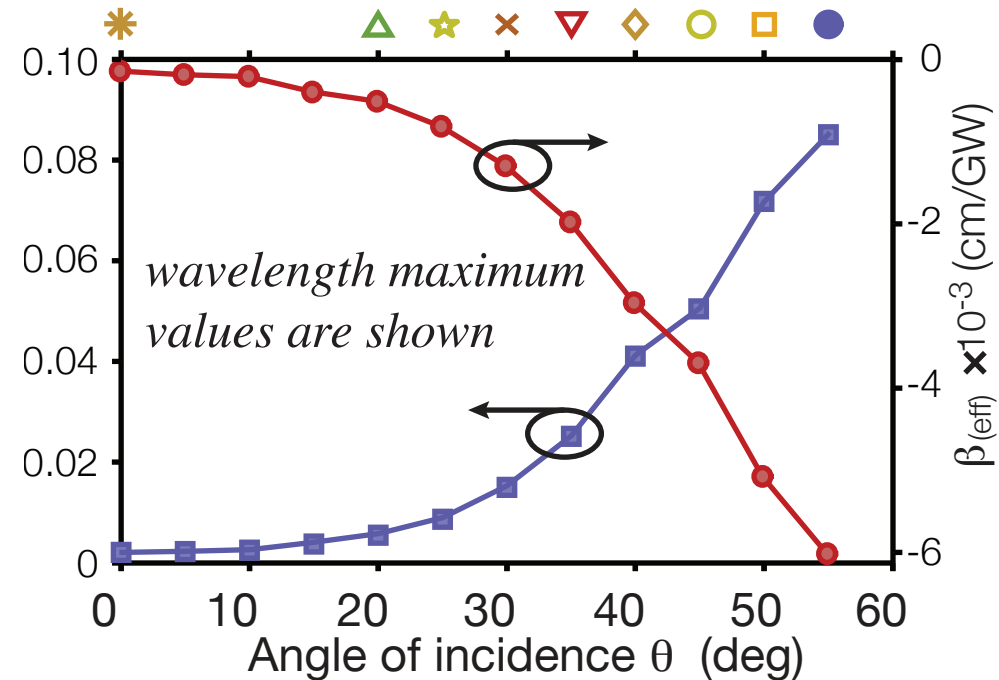
- H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, Science 342, 1223 (2013).
- C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, Phys. Rev. B 85, 045129 (2012).
- S. Campione, D. de Ceglia, M. A. Vincenti, M. Scalora, and F. Capolino, Phys. Rev. B 87, 035120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

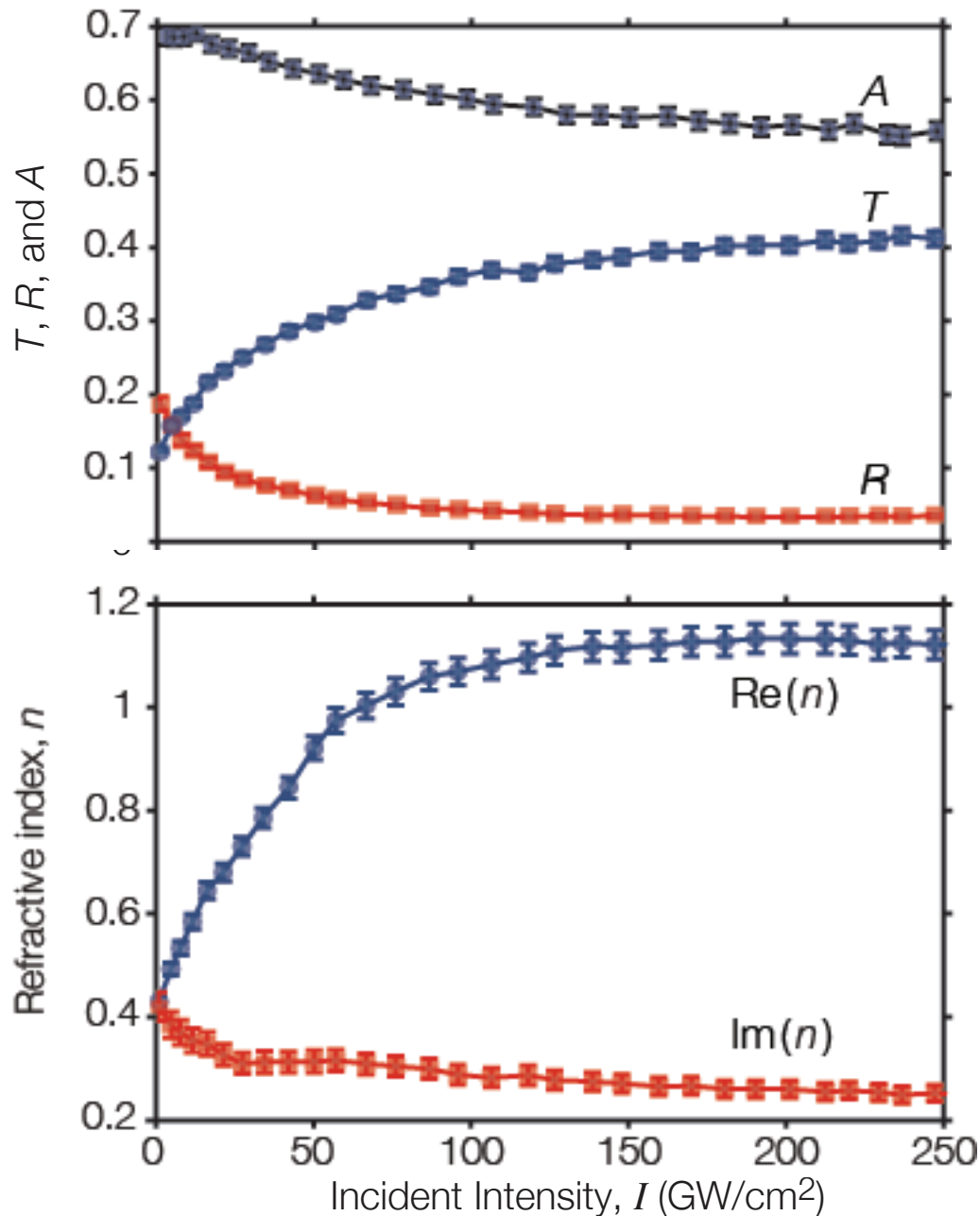


Variation with incidence angle



- 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 $0.11 \text{ cm}^2/\text{GW} = 1.1 \times 10^{-10} \text{ cm}^2/\text{W}$ at 1.25 μm and 60 deg . This value is 2000 times larger than that away from ENZ region.
- n_2 is 3.4×10^5 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.

Nonperturbative Nature of the NLO Response

1. The conventional equation $n = n_0 + n_2 I$ is not applicable to ENZ and other low-index materials. The nonlinear response is nonperturbative.
2. The nonlinear response can be accurately modeled in the $\chi^{(3)}$ limit by

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

where

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

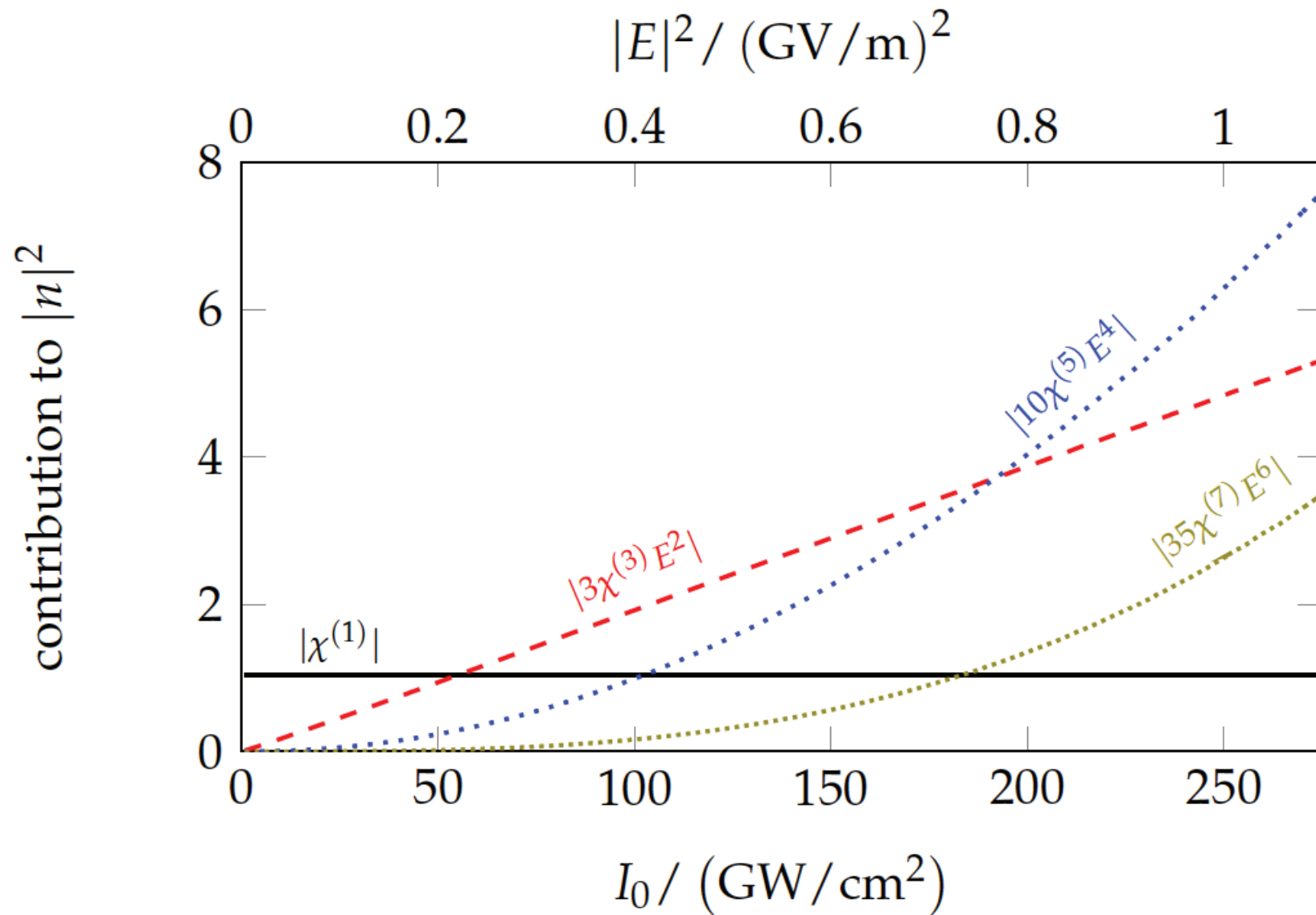
and

$$I = 2\text{Re}(n_0) \epsilon_0 c |E|^2.$$

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

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

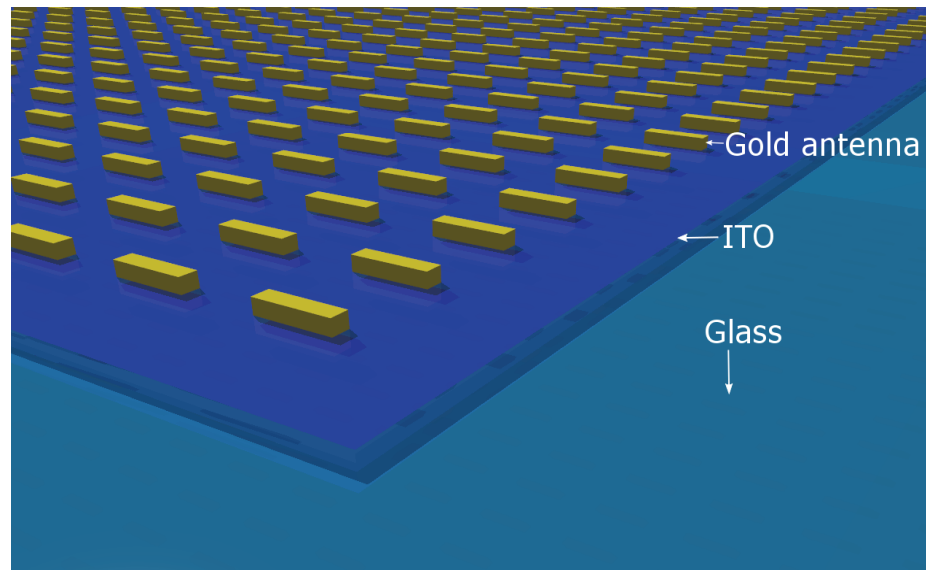
Nonlinear Response of ITO is Nonperturbative



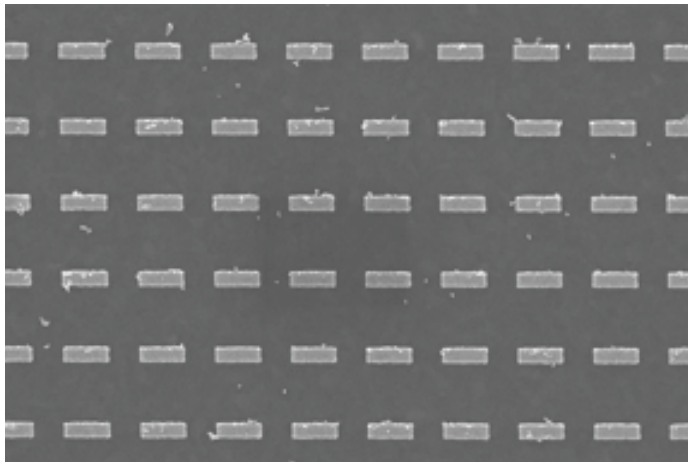
An ENZ Metasurface

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

Concept:



SEM:



Alam, Schulz, Upham, De Leon and Boyd,
Nature Photonics 12, 79-83 (2018).

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

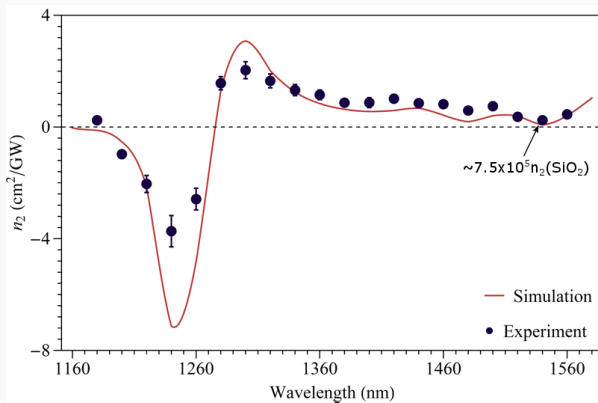


Figure 5: The material exhibits extremely large n_2 for the entire spectral range. The magnitude of the on-resonance value is 7 orders of magnitude larger than that of SiO_2 .

Summary

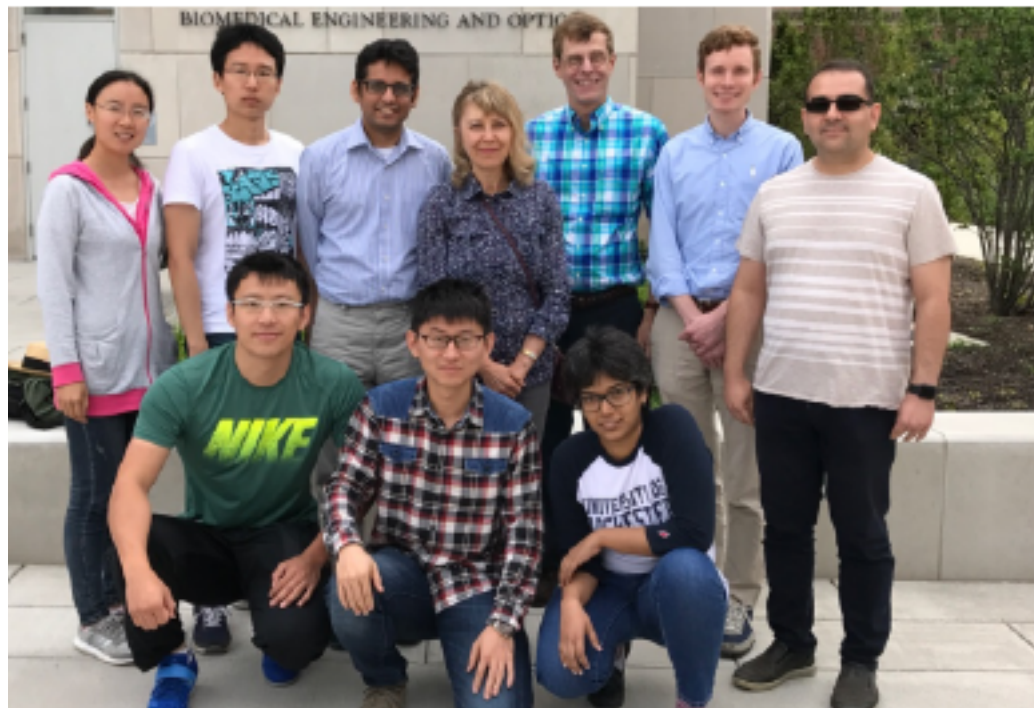
- A broadband nonlinear material with n_2 values upto 7 order of magnitude larger than that of SiO_2 .
- Sub-picosecond response time.
- $\Delta n \approx \pm 2.5$ over very large bandwidth.
- One can tailor the sign of the nonlinearity by simply designing the geometric parameters of the antenna appropriately.

Special Thanks To My Students and Postdocs!

Ottawa Group



Rochester Group



Other Reports of Highly Nonlinear Response in ENZ Material and Metamaterials

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).