Quantum Nonlinear Optics: New Materials and Interactions

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The visuals of this talk will be posted at boydnlo.ca/presentations

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Canada Excellence Research
Chair (CERC) in Nonlinear Quantum Optics

Research interest:
Nonlinear optics, quantum optics,
integrated photonics, meta-materials, etc.

Some New Results in Nonlinear Optics

1. **Nonlinear optical properties of epsilon-near-zero materials**

2. How to prevent laser-beam filamentation

3. Influence of nonlinearity on optical rogue waves

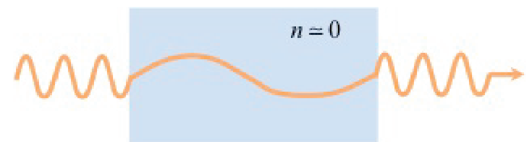
Physics of Epsilon-Near-Zero (ENZ) Materials

- ENZ materials possess exotic electromagnetic properties

Recall that $n = \sqrt{\epsilon\mu}$ where ϵ is the permittivity and μ is the magnetic permeability

Many opportunities in photonics are afforded by ENZ materials and ZIM (zero-index materials)

$$\lambda = \lambda_{\mathrm{vac}}/n \qquad v = c/n$$



For $n = 0$ the wavelength is stretched and the phase velocity becomes infinite

Light oscillates in time but not in space; oscillations are in phase everywhere

Silveirinha and Engheta, Phys. Rev. Lett. 97, 157403 (2006).

- Radiative processes are modified in ENZ materials

Einstein A coefficient (spontaneous emission lifetime = $1/A$)

$A = n\, A_{\mathrm{vac}}$

We can control (inhibit!) spontaneous emission!

Einstein B coefficient (for $\mu = 1$)

Stimulated emission rate = B times EM field energy density

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

Optical gain is very large!

Einstein, Physikalische Zeitschrift 18, 121 (1917).
Milonni, Journal of Modern Optics 42, 1991 (1995).

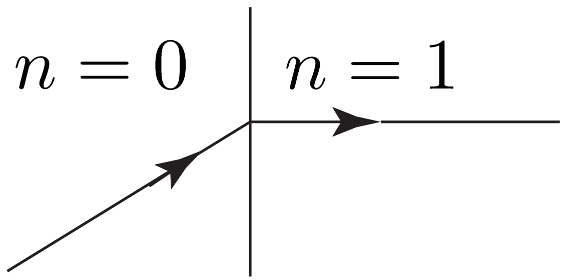
Terminology: ZIM= zero-index material; ENZ=epsilon near zero; EMNZ= epsilon and mu near zero

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

- Snell's law leads to intriguing predictions

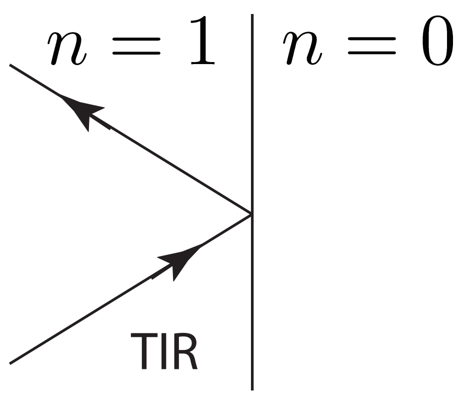
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

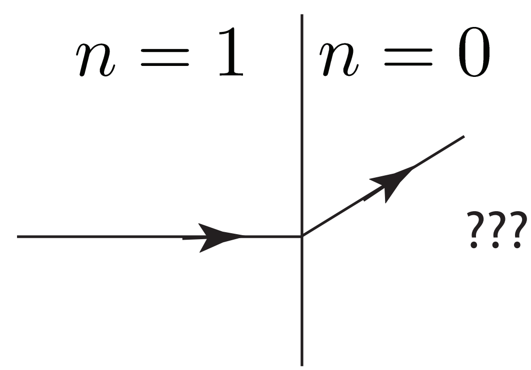
- Light always leaves perpendicular to surface of ENZ material!



Y. Li, et al., Nat. Photonics 9, 738, 2015; D. I. Vulis, et al., Opt. Express 25, 12381, 2017.

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

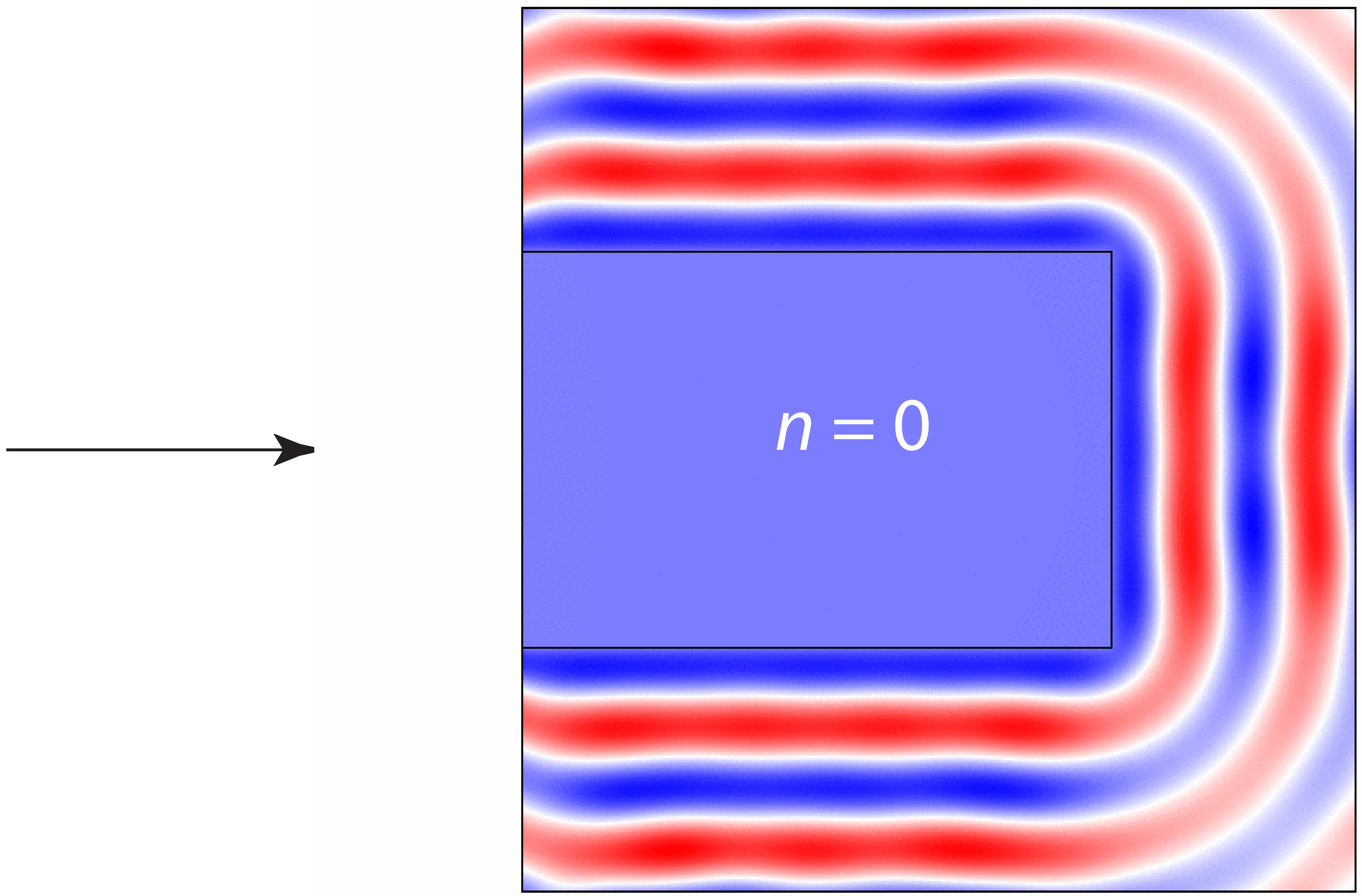




Y. Li, et al., Nat. Photonics 9, 738, 2015.

Maxwell Equations Prediction

- light enters slab at normal incidence

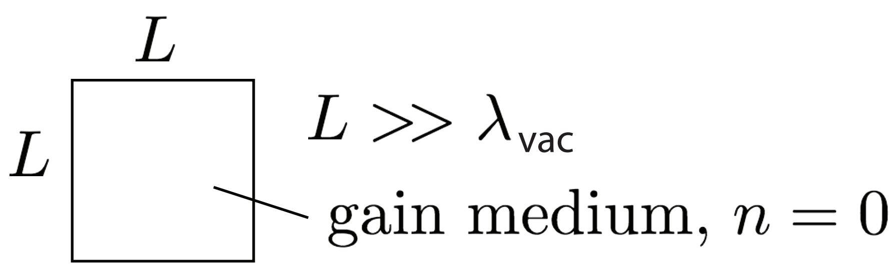


Some Consequences of ENZ Behaviour - 1

- Funny lenses

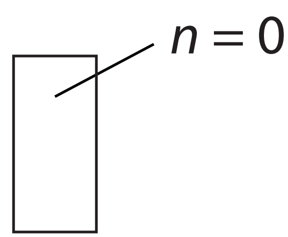
A. Alù et al., Phys. Rev. B 75, 155410, 2007; X.-T. He, ACS Photonics, 3, 2262, 2016.

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



J. Bravo-Abad et al., Proc. Natl. Acad. Sci. USA 109, 976, 2012.

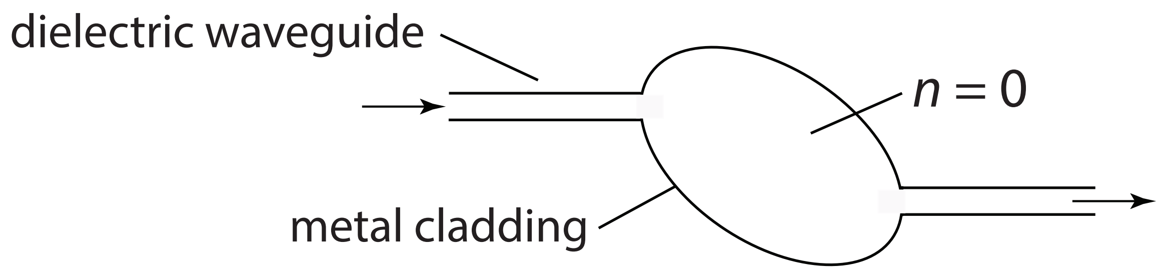
- No Fabry-Perot interference



O. Reshef et al., ACS Photonics 4, 2385, 2017.

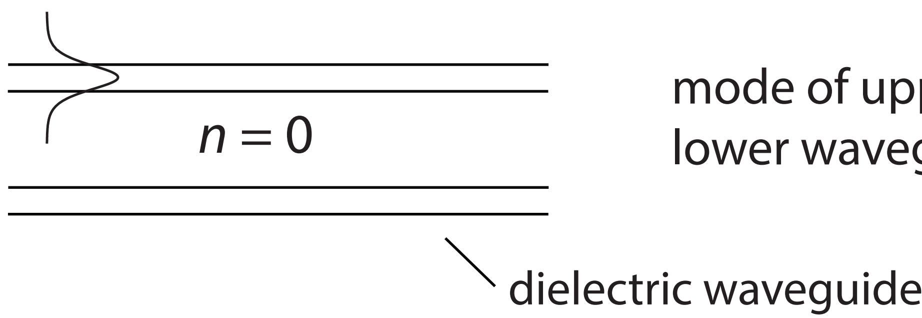
Some Consequences of ENZ Behavior - 2

- Super-coupling (of waveguides)



M. G. Silveirinha and N. Engheta, Phys. Rev. B 76, 245109, 2007; B. Edwards et al., Phys. Rev. Lett. 100, 033903, 2008.

- Large evanescent tails for waveguide coupling

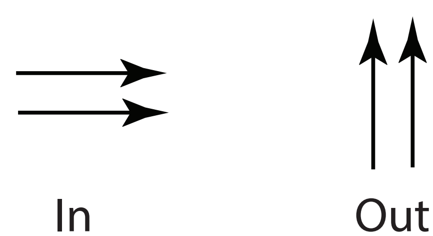


mode of upper waveguide extends to lower waveguide for any separation

- Automatic phase matching of NLO processes

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

For example, the following 4WM proces is allowed



H. Suchowski et al., Science 342, 1223, 2013.

Some Implications of Near-Zero Index

- Wavelength is stretched ($\lambda = \lambda_0 / n$).

- Phase velocity becomes very large ($v = c / n$).
  Frequency is unchanged (for static fields)

- The field becomes static in space, yet remains dynamic in time.
  Space and time decouples.

- k-vector is undefined (has zero length) in the medium.

- Light exits a zero-index medium with k-vector perpendicular to the interface.

- Phase matching conditions of nonlinear optics is relaxed.

- Propagation nearly always nonparaxial ($\lambda >>$ beam diameter).

- E field is enhanced due to the boundary condition.

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

- Challenge: Obtain low-loss ENZ materials
  Want Im ε as small as possible at the frequency where Re $\varepsilon = 0$.

- 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).

Implications of ENZ Behavior for Nonlinear Optics

Here is the intuition for why the ENZ condition is 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 under ENZ conditions the denominator becomes very small, leading to a very large value of n_2

Footnote:

Standard notation for perturbative NLO

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

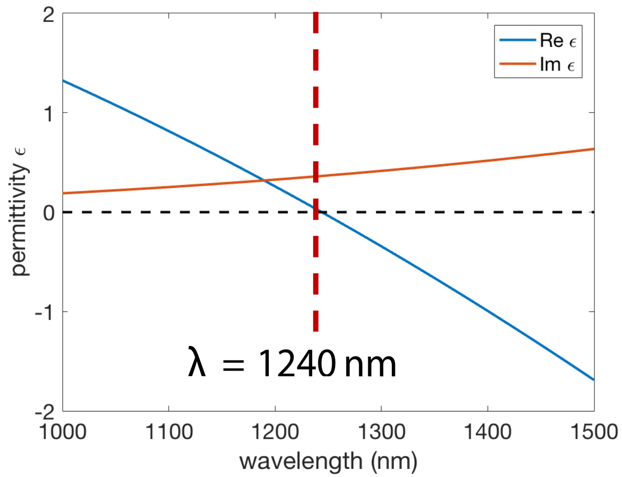
P is the induced dipole moment per unit volume and E is the field amplitude.

Also, the refractive index changes according to

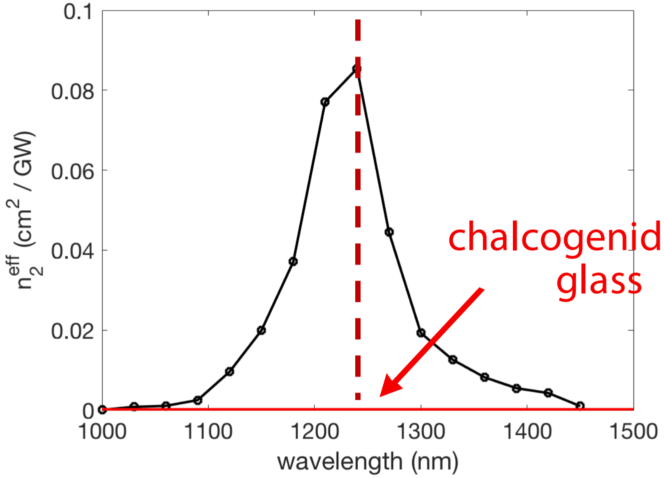
$$n = n_0 + n_2 I + n_4 I^2 + \ldots$$

Huge, Fast NLO Response of Indium Tin Oxide at its ENZ Wavelength

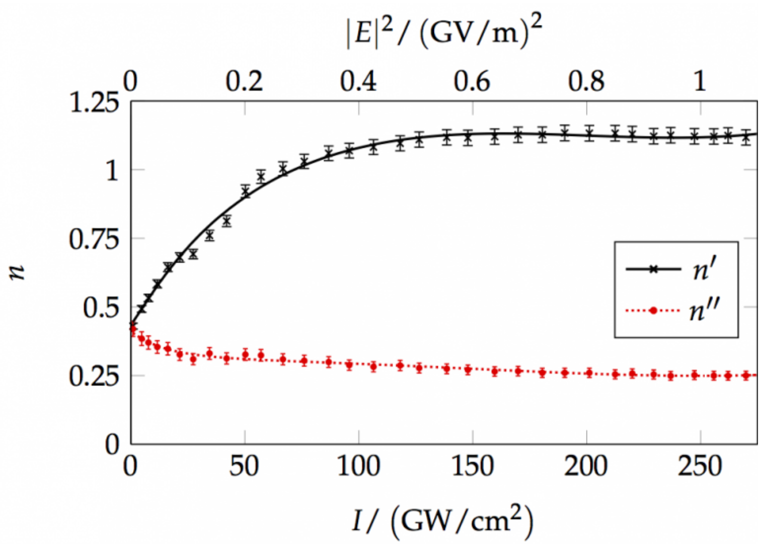
- ellipsometry



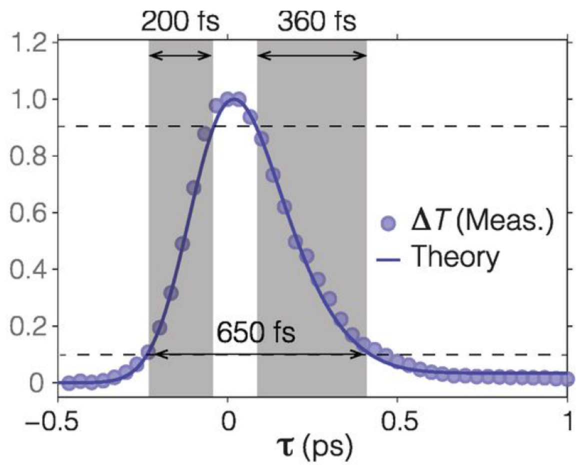
- n_2 is 3.4 x10^5 times larger than that of silica glass



- overall change in refractive index of 0.8



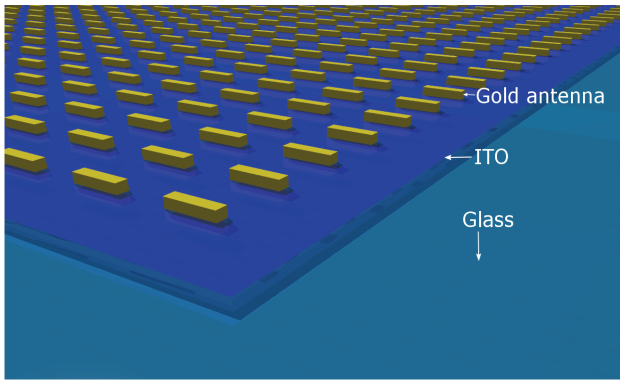
- sub picosecond reponse time

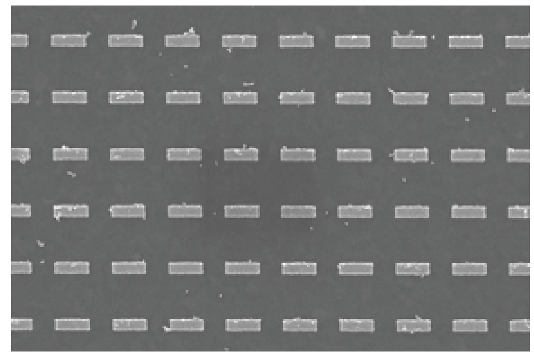


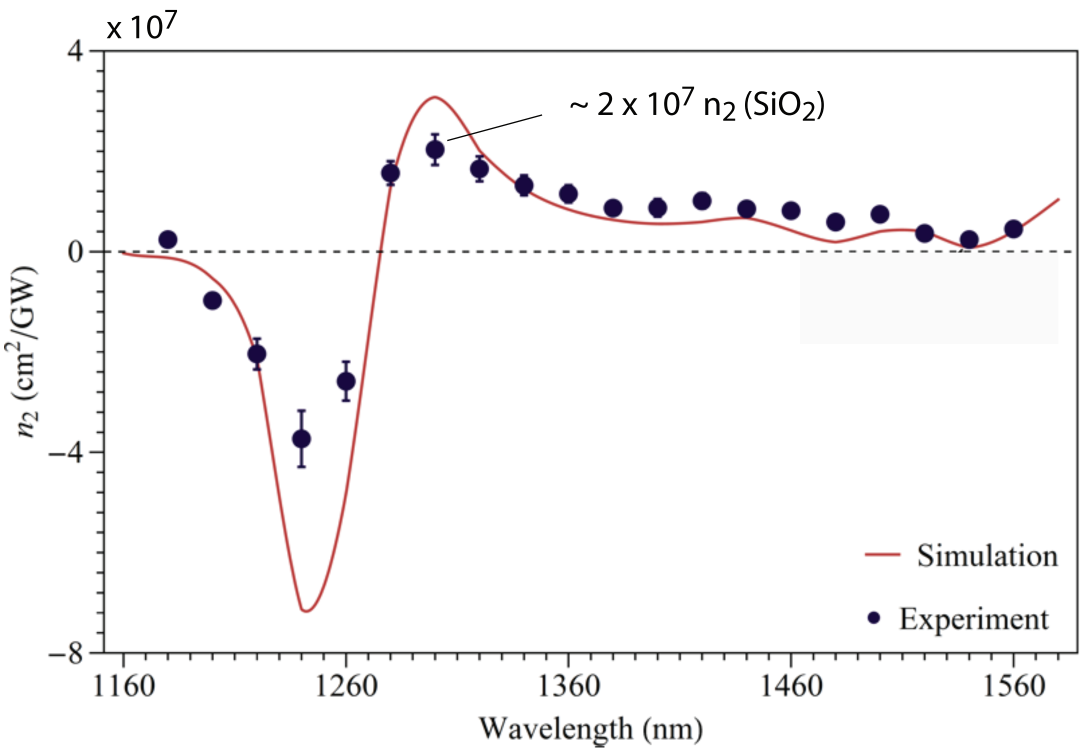
M. Z. Alam et al., Science 352, 795-797 (2016)

ENZ Metasurface: Gold Nanoantennas on ITO

- Even larger NLO response by placing a gold antenna array on top of ITO?
- Lightning rod effect: antennas concetrate the field within the ITO
- Coupled resonators: ENZ resonance and nano-antennas



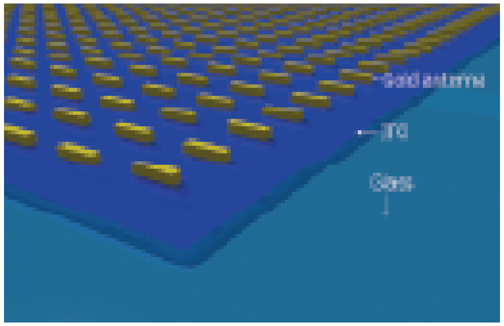




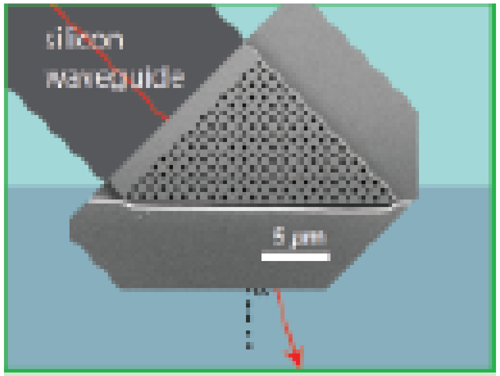
Alam, Schulz, Upham, De Leon and Boyd, Nature Photonics 12, 79-83 (2018).

Three Material Platforms Under Investigation

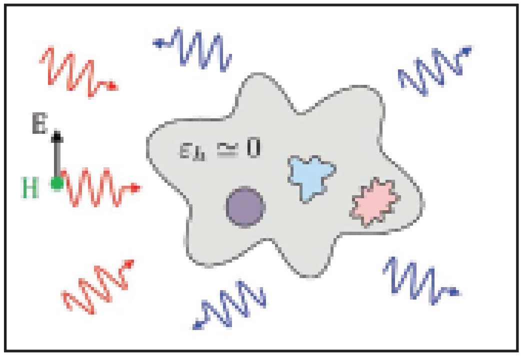
- Nanoantennas coupled to ENZ substrate
  (out of plane; free-space coupling)
  (Rochester)



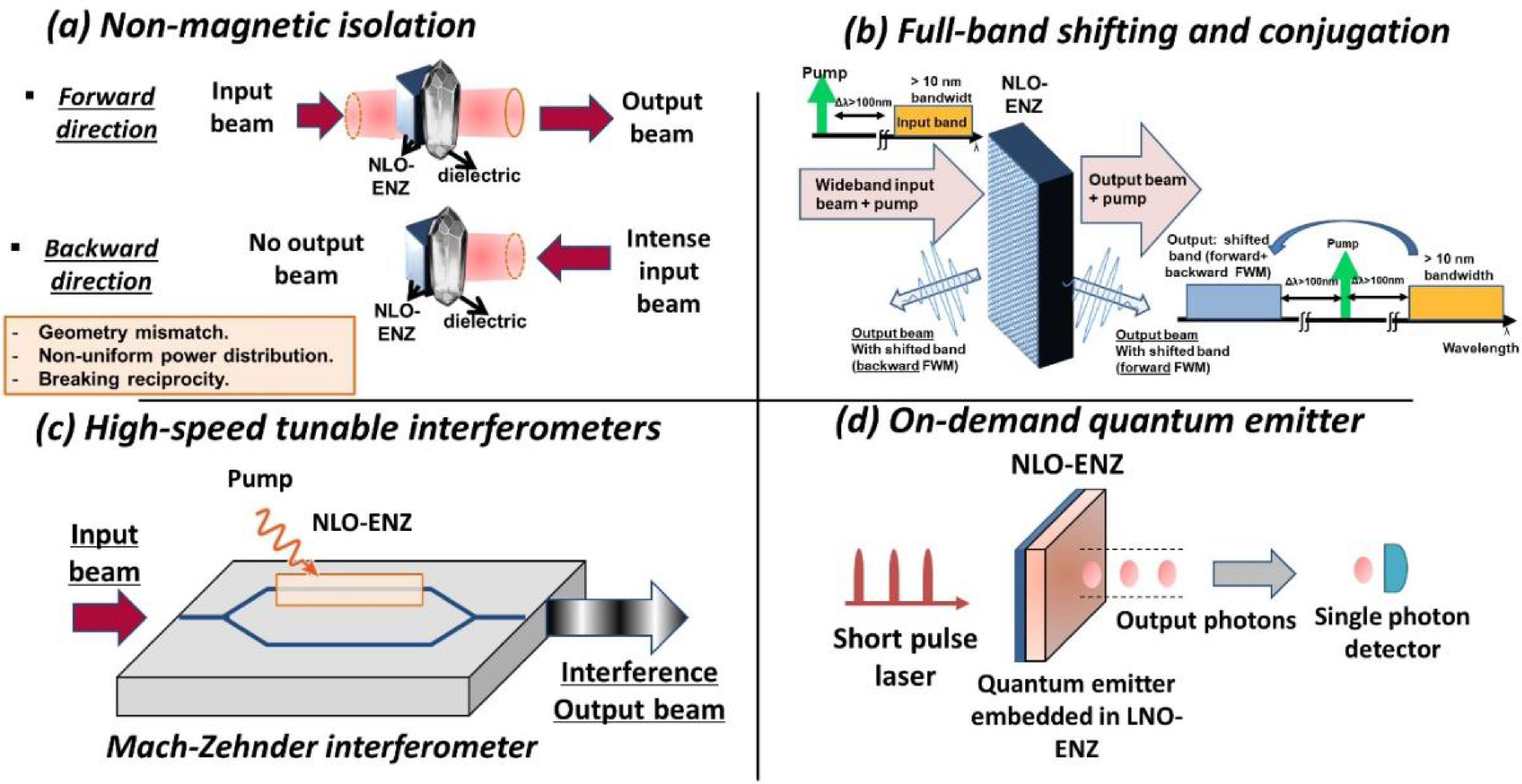
- Dirac cone metamaterials
  (in plane; compatible with integrated optics)
  (Harvard)



- Photonically doped metamaterials
  (out of plane; free-space coupling)
  (Penn)



Some Potential Applications of ENZ Behavior



Some New Results in Nonlinear Optics

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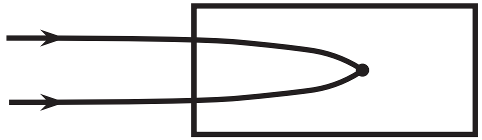
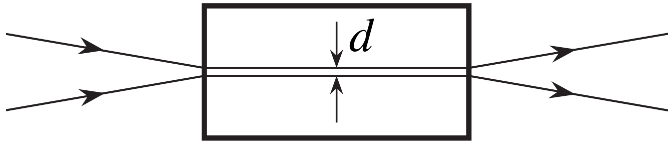
Self Action Effects in Nonlinear Optics

Self-action effects: light beam modifies its own propagation

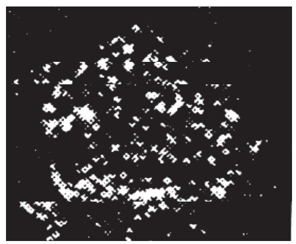
self focusing



self trapping



small-scale filamentation



Why Care About Self-Focusing and Filamentation?

- Optical switching

- Laser modelocking

- Directed energy
  - prevent filamentation
  -controlled self focusing

EFFECTS OF THE GRADIENT OF A STRONG ELECTROMAGNETIC BEAM ON ELECTRONS AND ATOMS

G. A. ASKAR'YAN

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor December 22, 1961

J. Exptl. Theoret. Phys. (U.S.S.R.) **42**, 1567-1570 (June, 1962)

It is shown that the transverse inhomogeneity of a strong electromagnetic beam can exert a strong effect on the electrons and atoms of a medium. Thus, if the frequency exceeds the natural frequency of the electron oscillations (in a plasma or in atoms), then the electrons or atoms will be forced out of the beam field. At subresonance frequencies, the particles will be pulled in, the force being especially large at resonance. It is noted that this effect can create either a rarefaction or a compression in the beam and at the focus of the radiation, maintain a pressure gradient near an opening from an evacuated vessel to the atmosphere, and create a channel for the passage of charged particles in the medium.

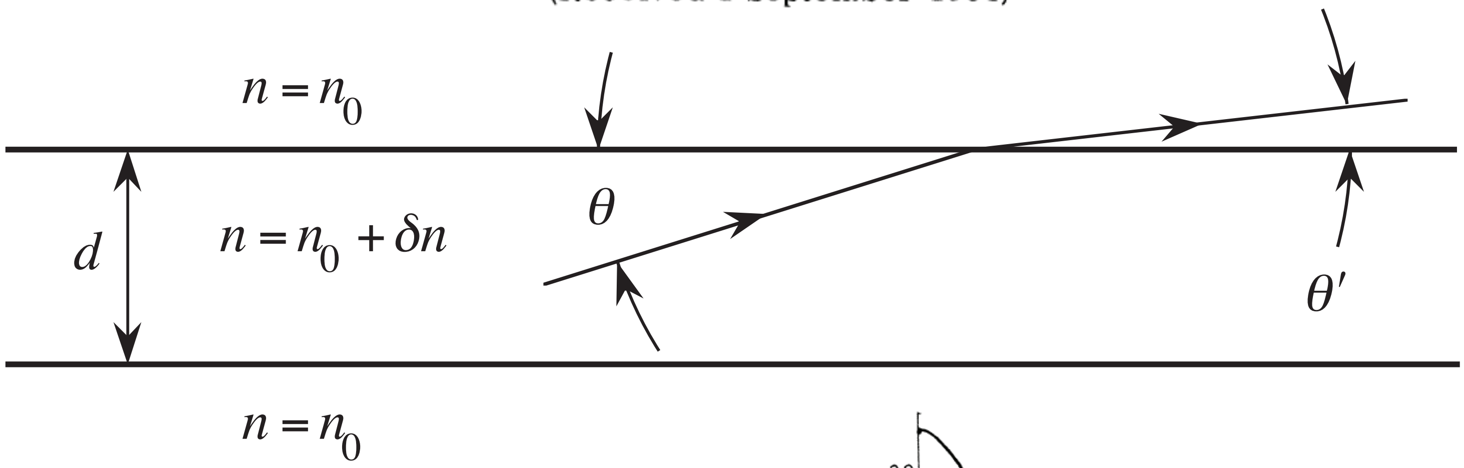
It is shown that the strong thermal ionizing and separating effects of the ray on the medium can be used to set up waveguide propagation conditions and to eliminate divergence of the beam (self-focusing). It is noted that hollow beams can give rise to directional flow and ejection of the plasma along the beam axis for plasma transport and creation of plasma current conductors. The possibilities of accelerating and heating plasma electrons by a modulated beam are indicated.

Prediction of Self Trapping

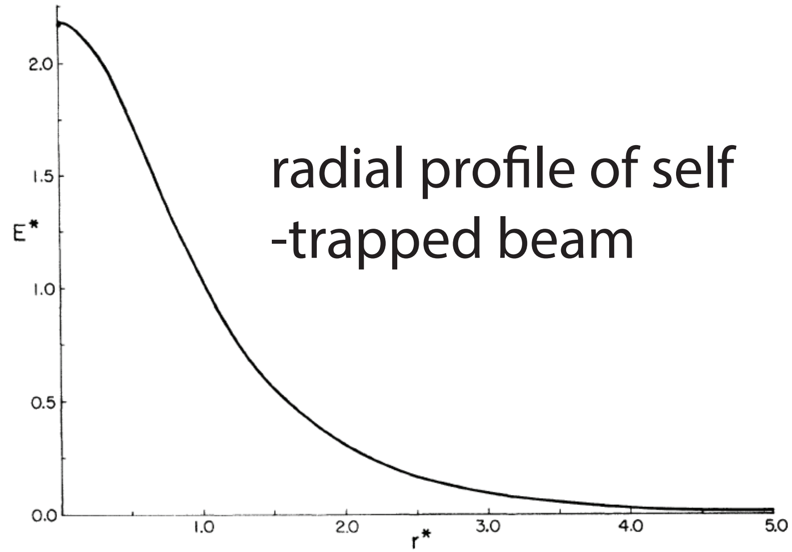
VOLUME 13, NUMBER 15 PHYSICAL REVIEW LETTERS 12 OCTOBER 1964

SELF-TRAPPING OF OPTICAL BEAMS*

R. Y. Chiao, E. Garmire, and C. H. Townes
Massachusetts Institute of Technology, Cambridge, Massachusetts
(Received 1 September 1964)



$$P_{\rm cr} = \frac{\pi(0.61)^2\lambda_0^2}{8n_0n_2}$$

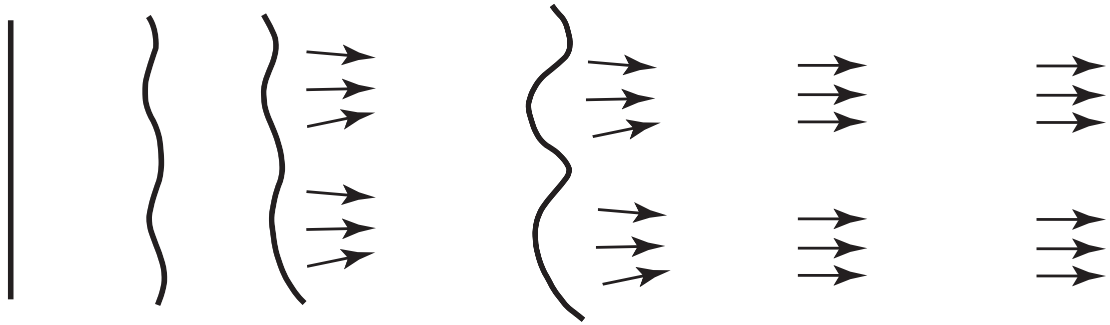


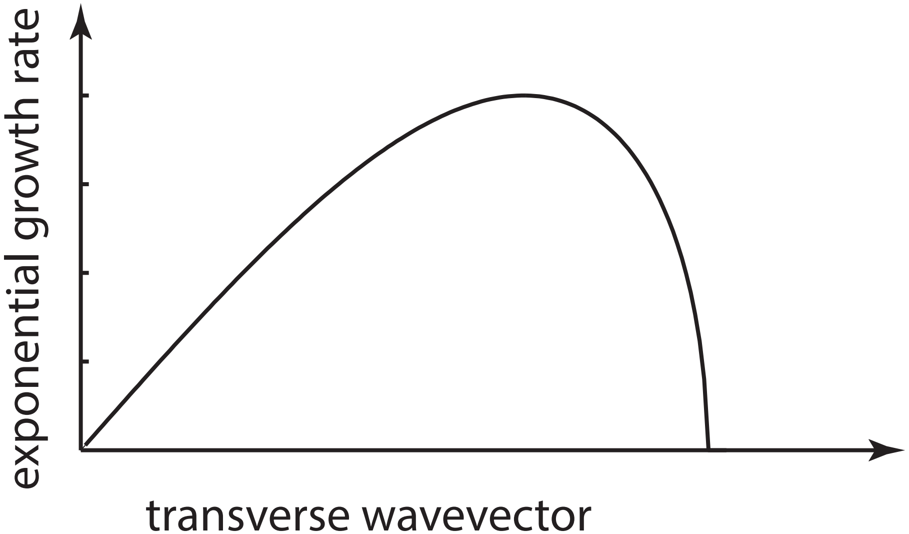
radial profile of self -trapped beam

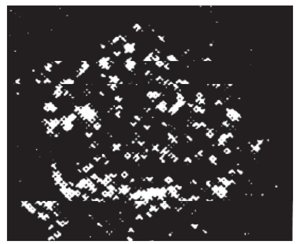
Beam Breakup by Small-Scale Filamentation

Predicted by Bespalov and Talanov (1966)

Exponential growth of wavefront imperfections by four-wave mixing processes





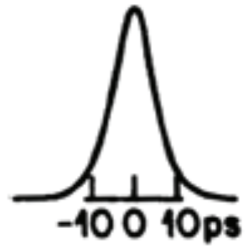


Optical Solitons

Field distributions that propagate without change of form

Temporal solitons (nonlinearity balances gvd)

$$\frac{\partial \tilde{A}_s}{\partial z} + \frac{1}{2} i k_2 \frac{\partial^2 \tilde{A}_s}{\partial \tau^2} = i\gamma |\tilde{A}_s|^2 \tilde{A}_s.$$

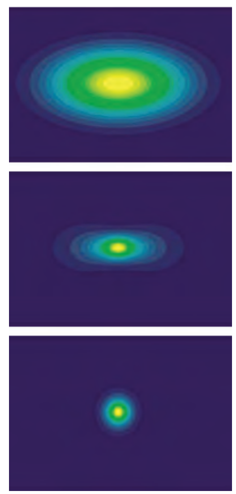


1973: Hasegawa & Tappert

1980: Mollenauer, Stolen, Gordon

Spatial solitons (nonlinearity balances diffraction)

$$2ik_0 \frac{\partial A}{\partial z} + \frac{\partial^2 A}{\partial x^2} = -3\chi^{(3)} \frac{\omega^2}{c^2} |A|^2 A$$



1964: Garmire, Chiao, Townes

1974: Ashkin and Bjorkholm (Na)

1985: Barthelemy, Froehly (CS2)

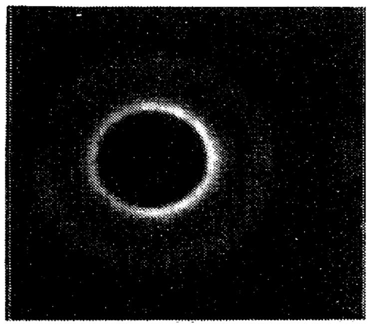
1991: Aitchison et al. (planar glass waveguide

1992: Segev, (photorefractive)

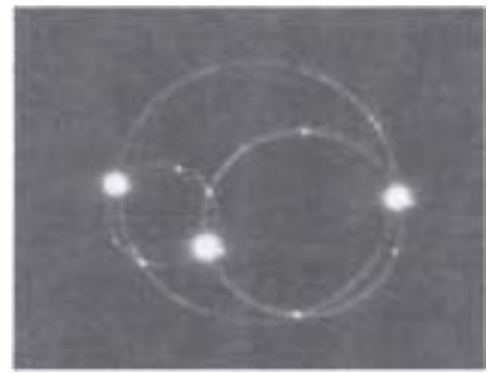
Self-Focusing Can Produce Unusual Beam Patterns

Pattern depends sensitively upon initial conditions

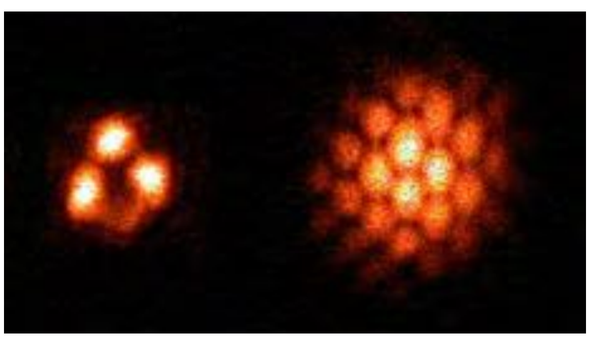
- Conical emission
  Harter et al., PRL 46, 1192 (1981)



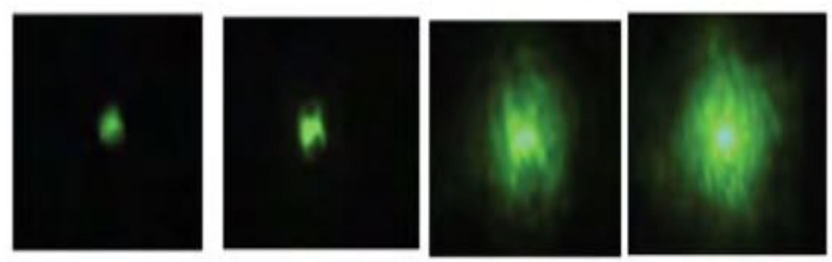
- Multiple ring patterns
  Kauranen et al, Opt. Lett. 16, 943, 1991;



- Honeycomb pattern formation
  Bennink et al., PRL 88, 113901 2002.



- Loss of spatial coherence
  Schweinsberg et al., Phys. Rev. A 84, 053837 (2011).

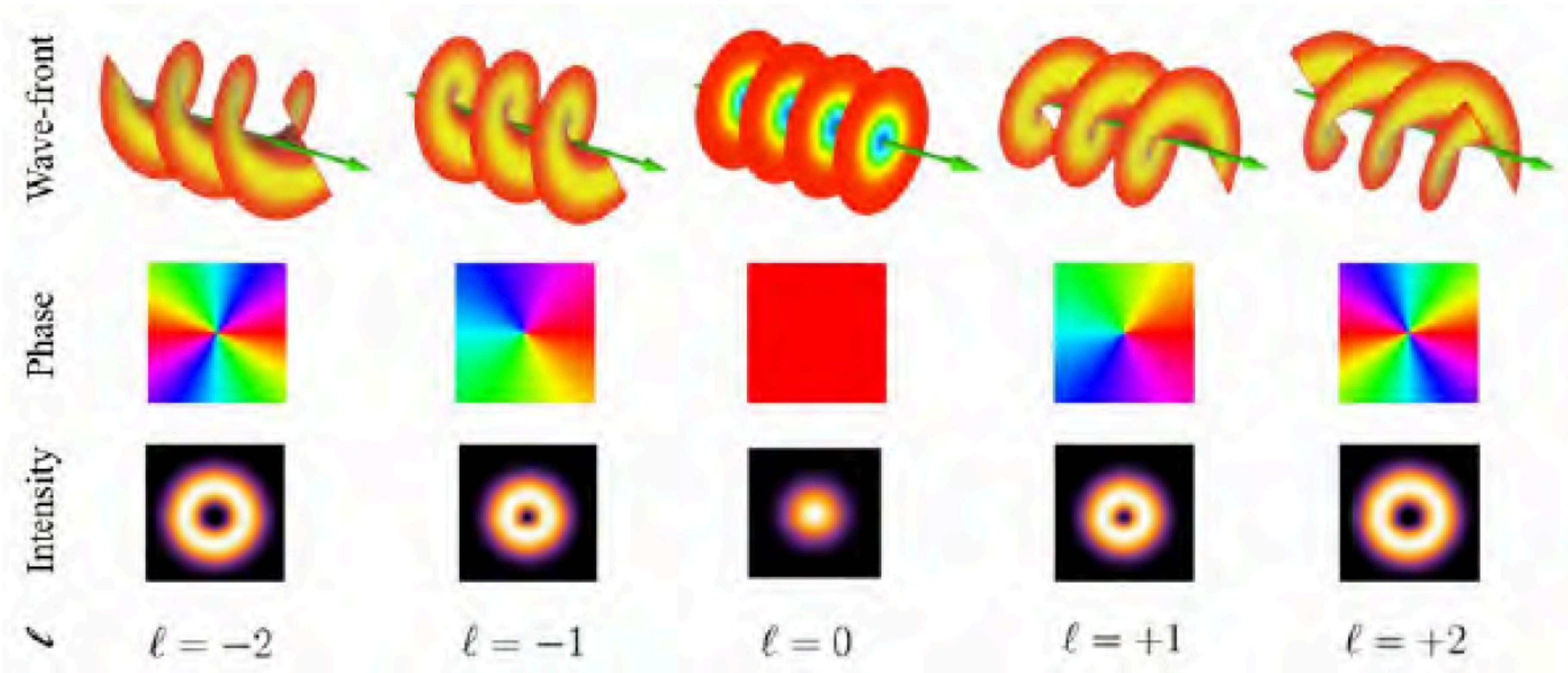


Self-Focusing of Structured Light: OAM States of Light

- Light can carry spin angular momentum by means of its circular polarization.

- Light can also carry orbital angular momentum by mean of a phase winding of the optical wavefront

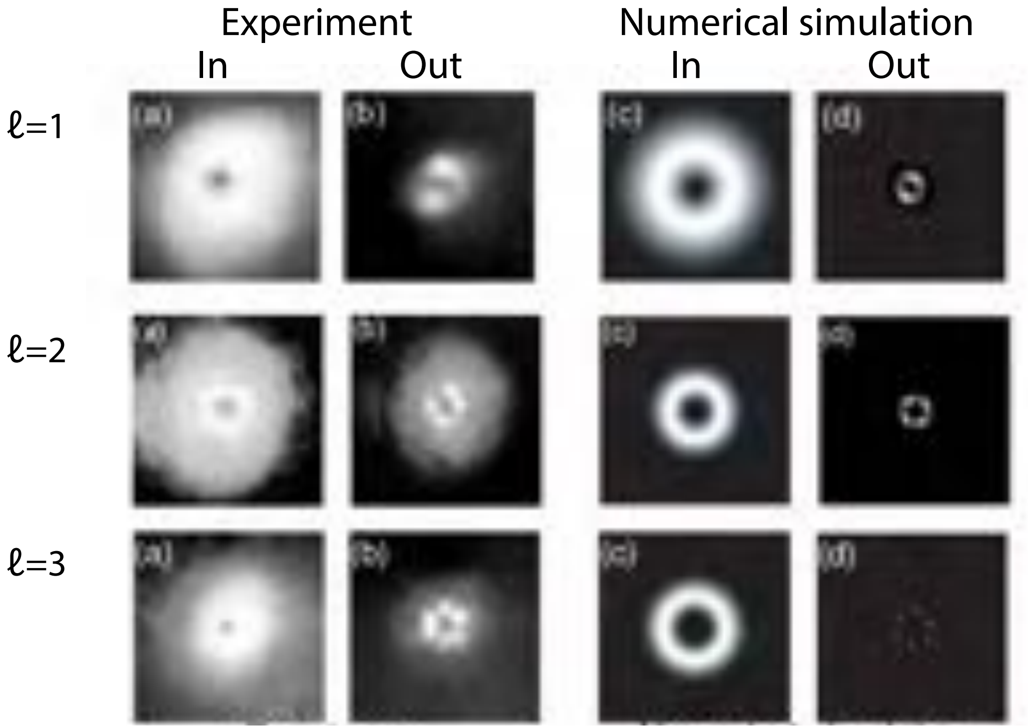
- A well-known example are the Laguerre-Gauss modes. These modes contain a phase factor of $\exp(i\ell\phi)$ and carry angular momentum of $\ell\hbar$ per photon



- How is self-focusing modified by the structuring of a light beam?

Breakup of Ring Beams Carrying Orbital Angular Momentum (OAM) in Sodium Vapor

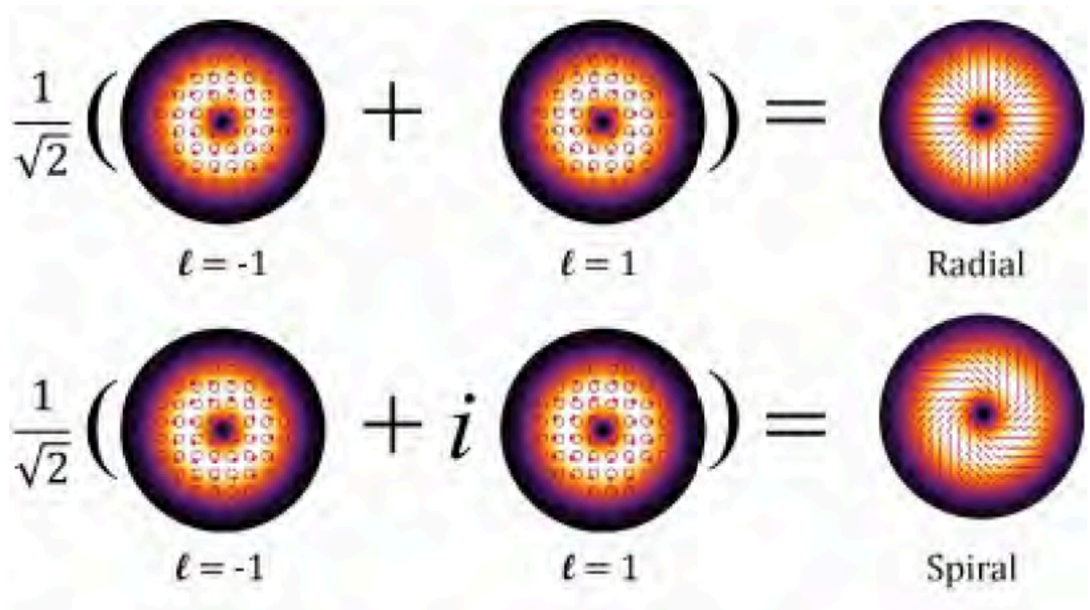
- Firth and Skryabin predicted that ring shaped beams in a saturable Kerr medium are unstable to azimuthal instabilities.
- Beams with OAM of $\ell\hbar$ tend to break into 2ℓ filaments.
  (But aberrated OAM beams tend to break into $2\ell + 1$ filaments.)



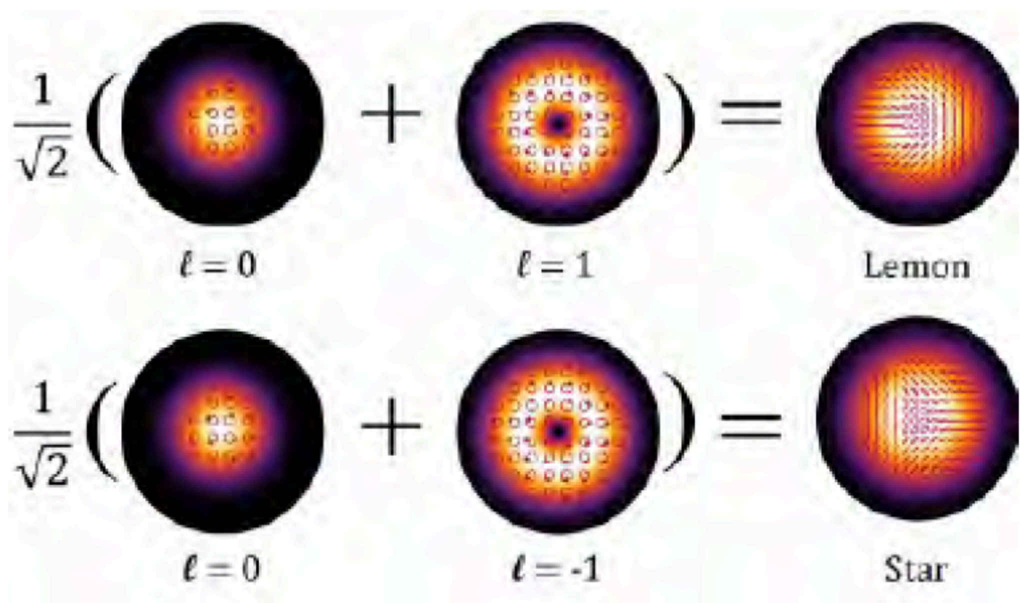
M.S. Bigelow, P. Zerom, and R.W. Boyd, Phys. Rev. Lett 92, 083902 (2004)

Space-Varying Polarized Light Beams

– Vector Vortex Beams



– Poincare Beams

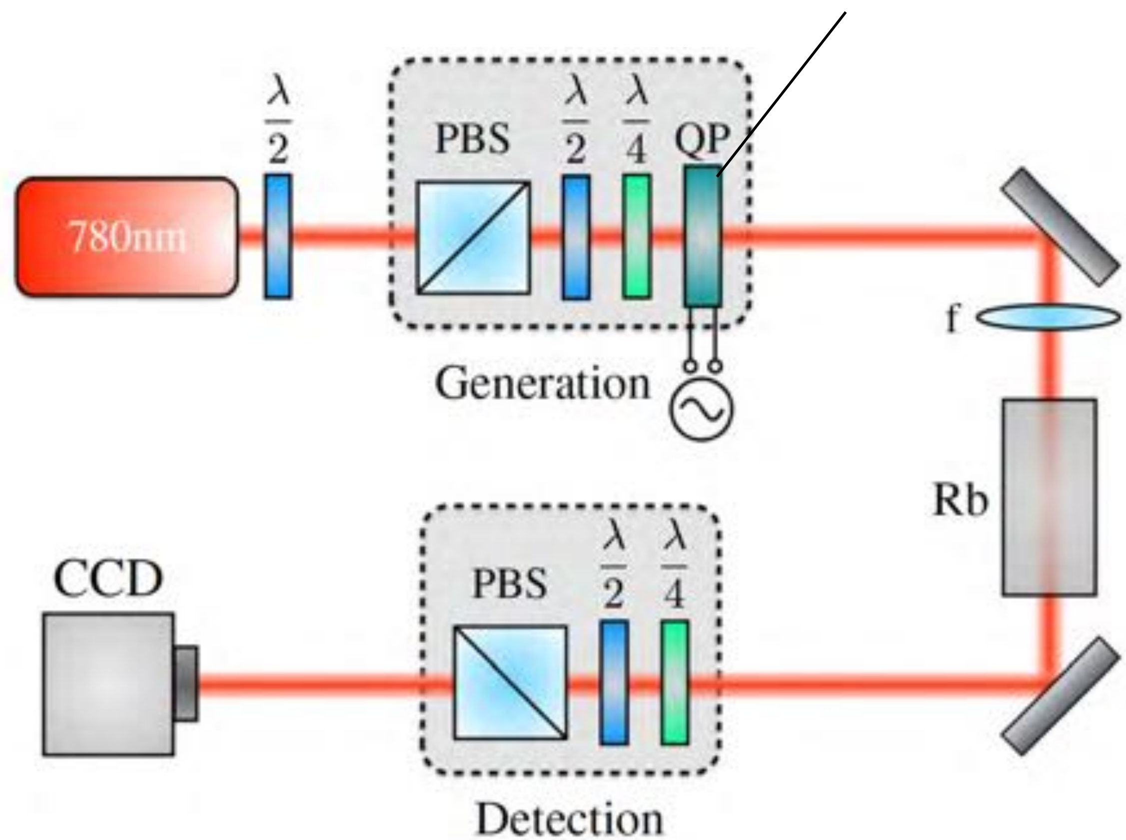


- How do these beams behave under conditions of self-focusing and filamentation?

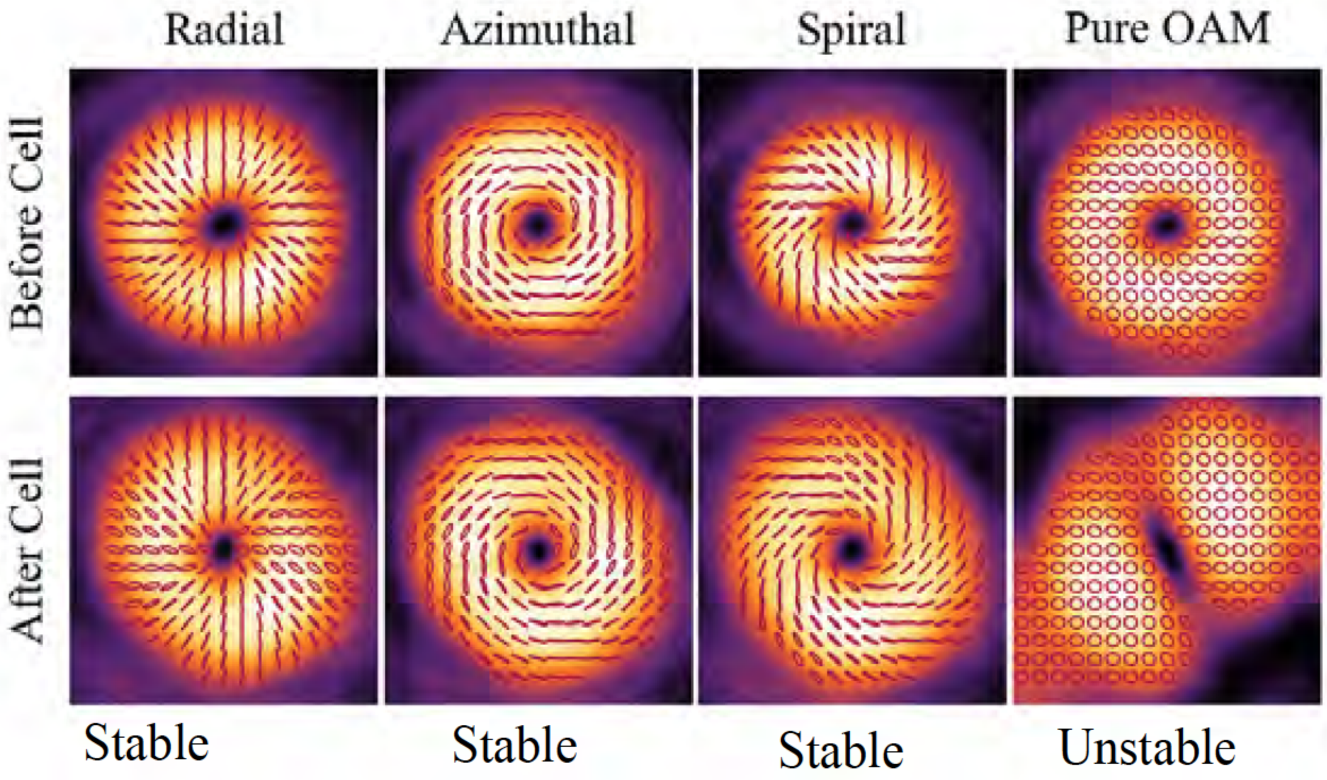
Bouchard et al, PRL 117, 233903 (2016).

Experimental Setup

Q-plate: SAM to OAM converter



Results – Vector Beams (Experimental Results)



Intensity and polarization distributions of vector and LG beams before and after propagating through the Rb atomic vapour.

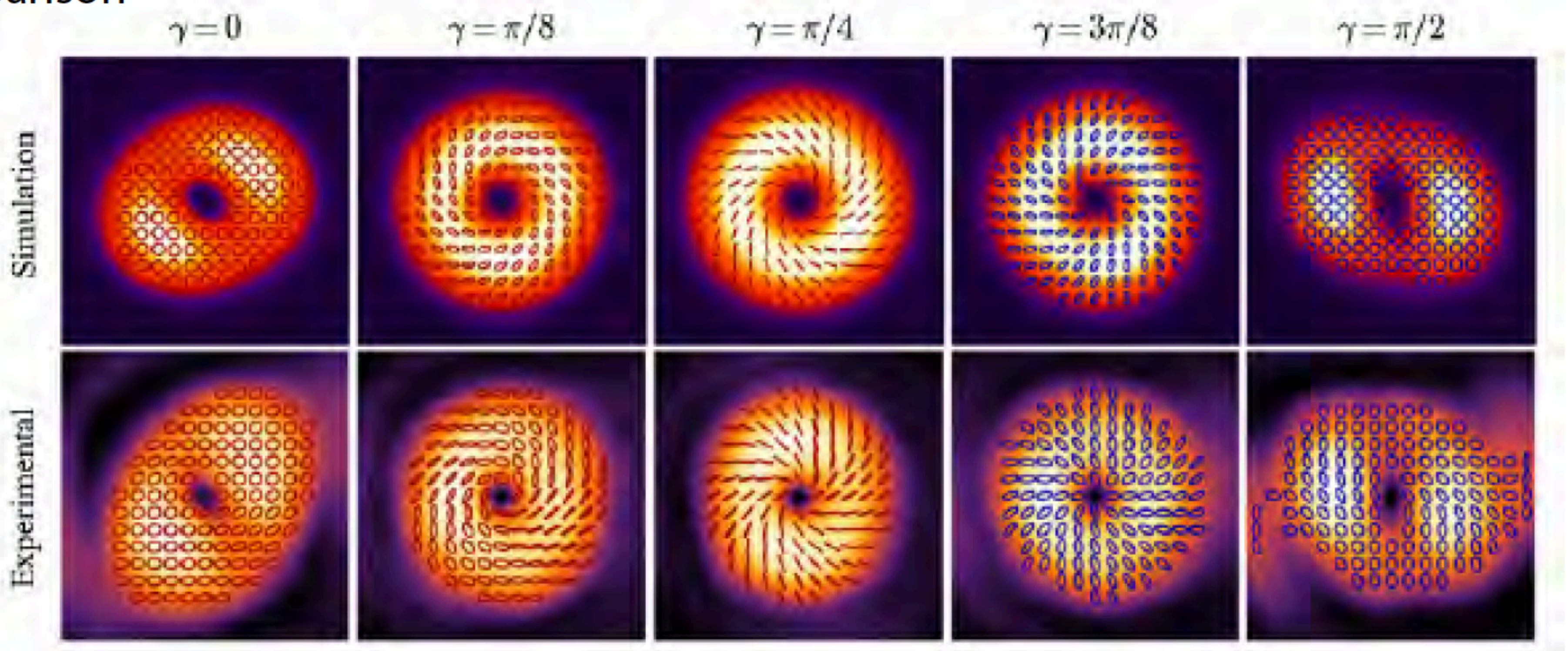
Numerical Modeling of the Experimental Results

- Coupled nonlinear propagation equations

$$\frac{\partial E_L}{\partial \zeta} - \frac{i}{2}\nabla_\perp^2 E_L = i\gamma \frac{|E_L|^2 + \nu |E_R|^2}{1+\sigma\left(|E_L|^2 + \nu |E_R|^2\right)} E_L$$

$$\frac{\partial E_R}{\partial \zeta} - \frac{i}{2}\nabla_\perp^2 E_R = i\gamma \frac{|E_R|^2 + \nu |E_L|^2}{1+\sigma\left(|E_R|^2 + \nu |E_L|^2\right)} E_R$$

- Comparison



Conclusions: stability of vector OAM beams

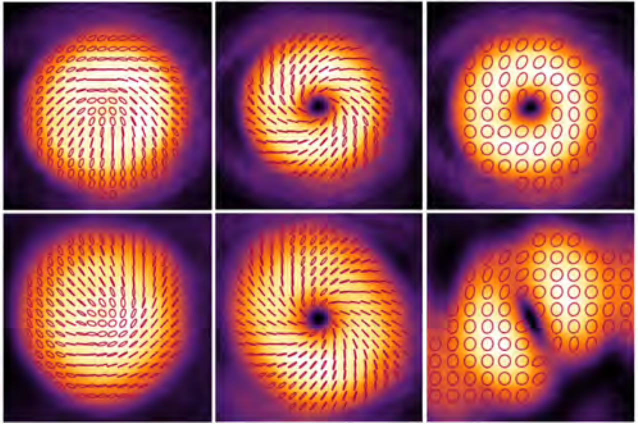
- Pure OAM beam:
  beam breakup
- Vector vortex beams:
  stable propagation
- Poincaré beams:
  stable propagation

Bouchard et al, PRL 117, 233903 (2016).

Poincaré beam | Vector vortex beam | Pure OAM beam

In

Out

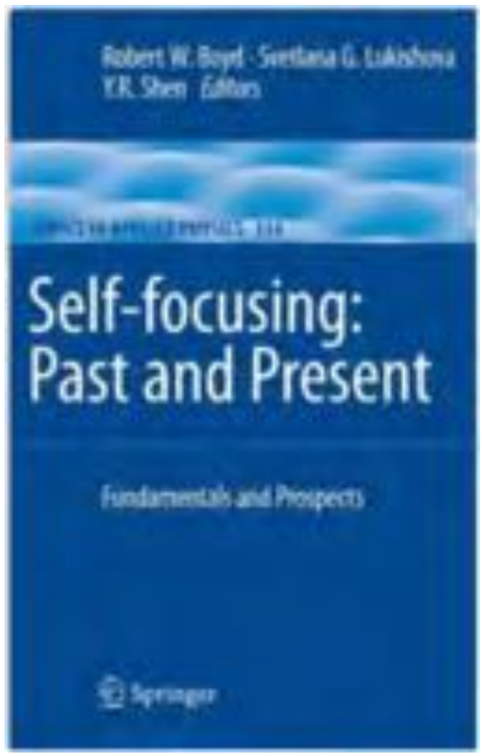


Stable | Stable | Unstable

Summary

- Even more than 50 years after their inceptions, self-focusing and filamentation remain fascinating topics for investigation.

- If you want to learn more:



Some New Results in Nonlinear Optics

1. Nonlinear optical properties of epsilon-near-zero materials

2. How to prevent laser-beam filamentation

3. **Influence of nonlinearity on optical rogue waves**

Influence of Nonlinearity on the Creation of Rogue Waves

- Study rogue-wave behavior in a well-characterized optical system
- Is nonlinearity important? Required? Or does it actually inhibit rogue-wave formation?

A. Mathis, L. Froehly, S. Toenger, F. Dias, G. Genty & J. Dudley. Scientific Reports 5, 1 (2015).

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

Oceanic rogue waves

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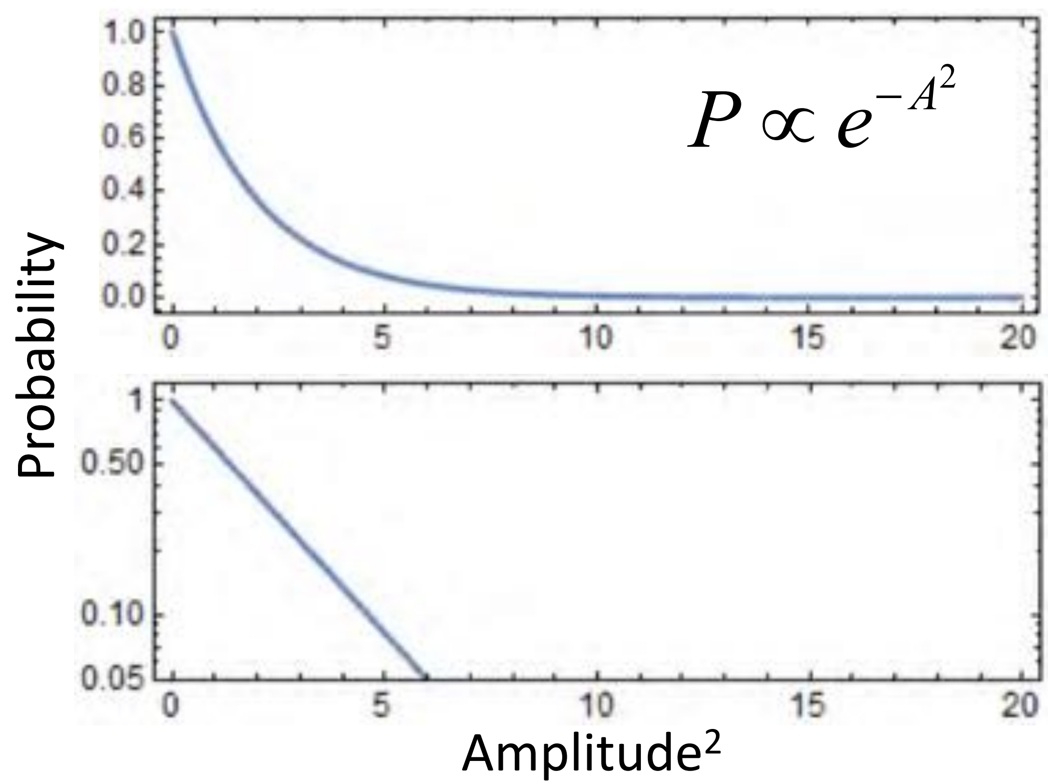
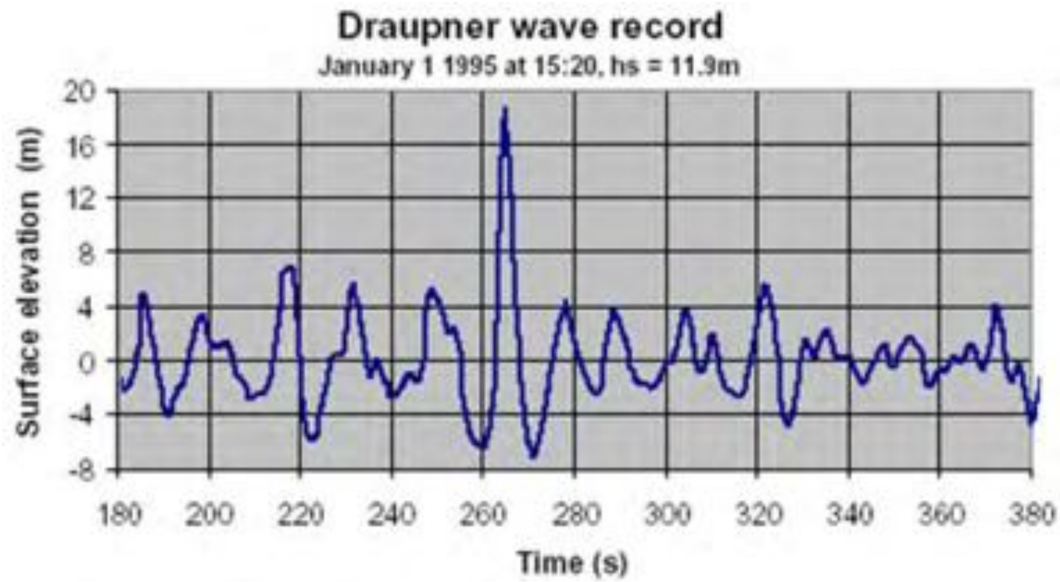
Before 1995

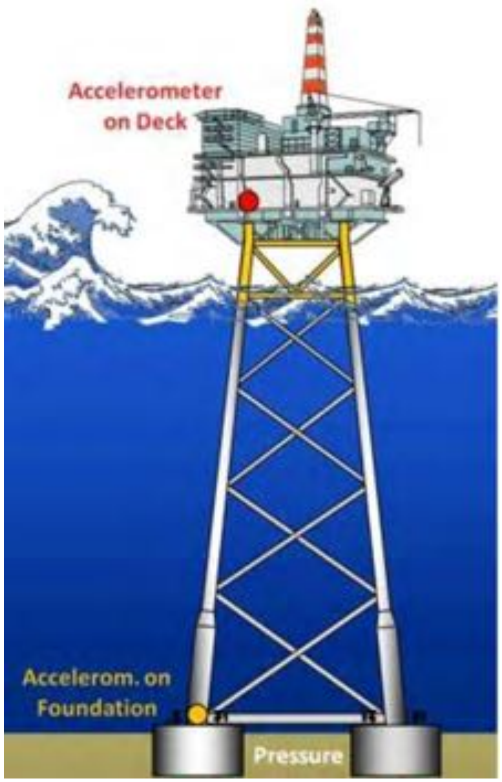
Sailors: we see gigantic waves.
Scientists: it is a fairy tale!
Ocean waves follow
Gaussian distribution.



Oceanic rogue waves

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





Characteristics of rogue waves

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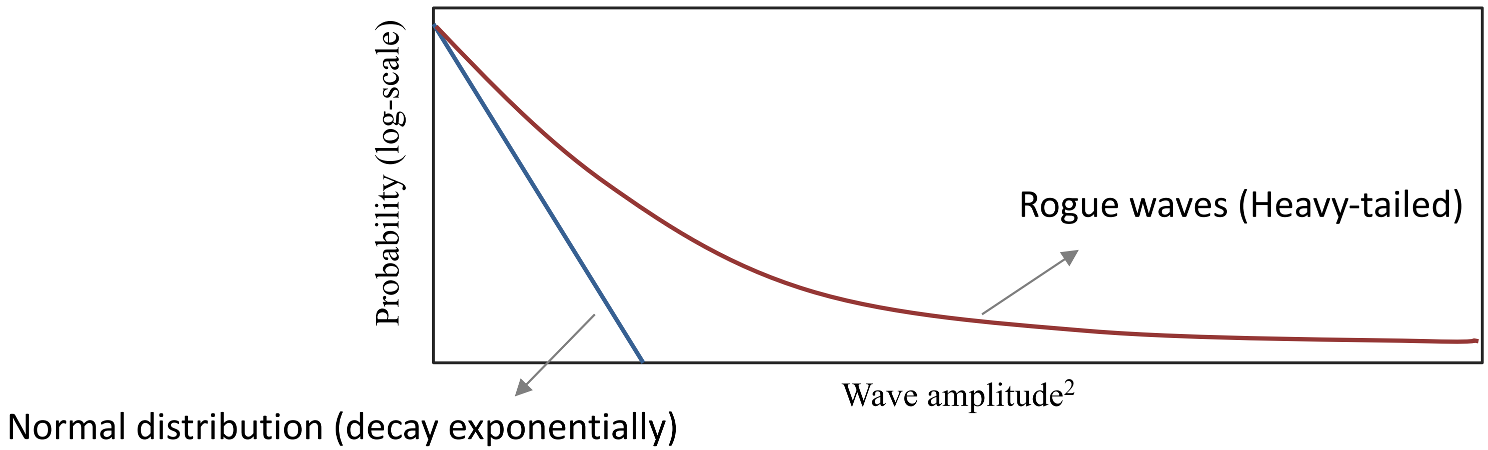
- Rogue waves appear from nowhere and disappear without a trace.
- Rogue waves ≠ accidental constructive interference
- They occur much more frequently than expected in ordinary wave statistics.

Probability distribution in rogue systems:



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

Rogue waves in 1D vs 2D systems

"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.

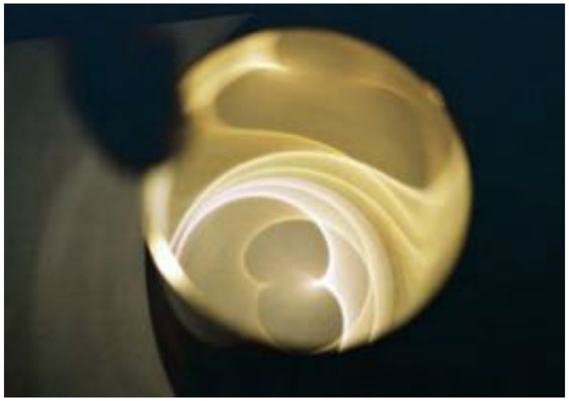
$$2ik \frac{\partial A}{\partial x} + \nabla_{\perp}^2 A = i\gamma |A|^2 A$$

Two focusing effects in 2D systems:

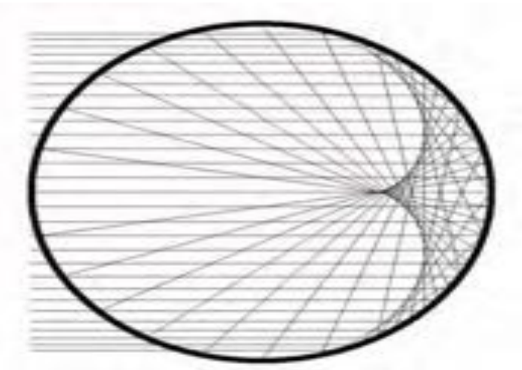
- **Linear**: Spatial (geometrical) focusing
- **Nonlinear**: Self focusing

Optical caustics

Swimming pool



Coffee cup

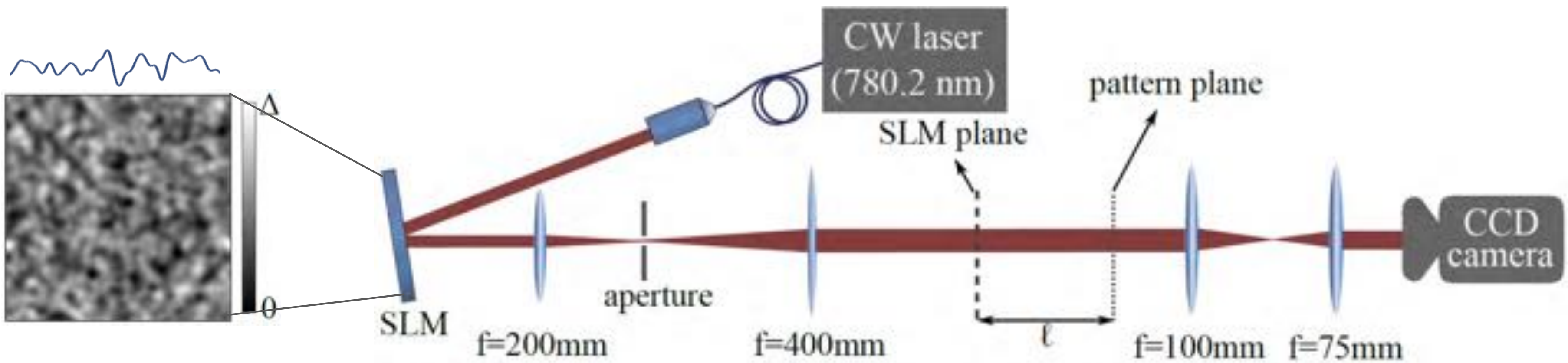


Ray picture

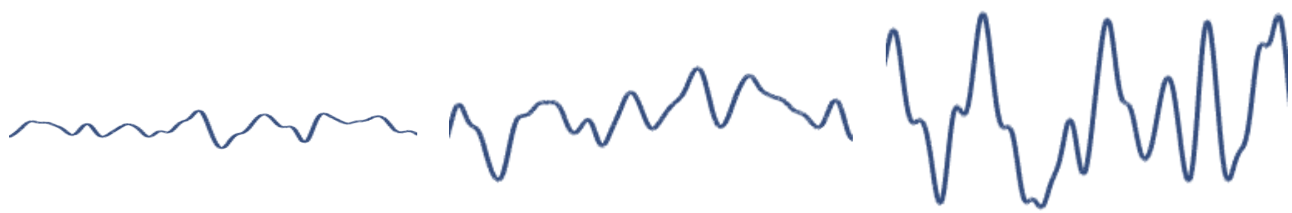
- Caustics are defined as envelope of a family of rays
- Singularities in ray optics
- Catastrophe theory is required to remove singularity

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*.

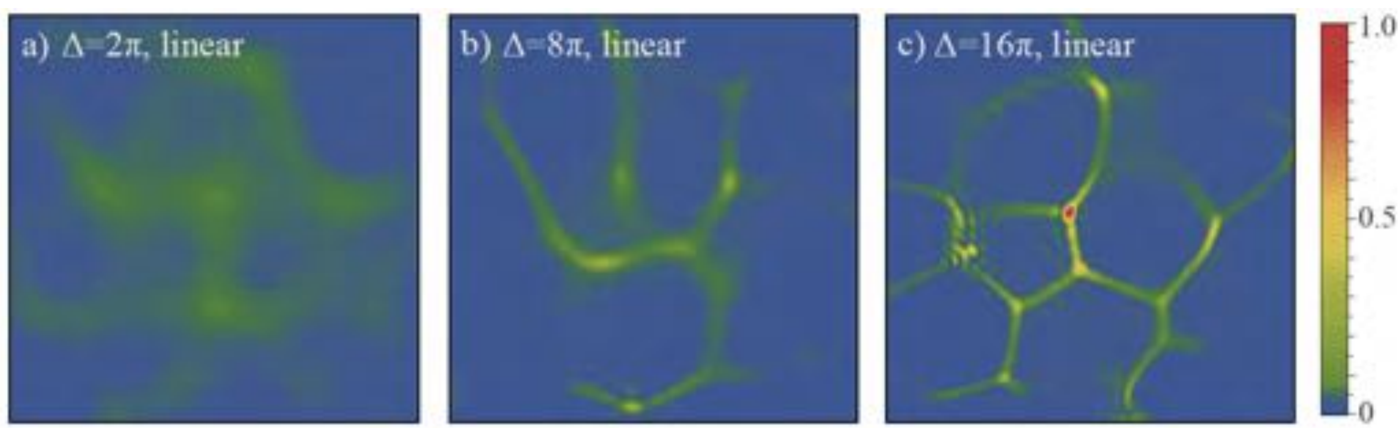
Generation of optical caustics



Phase variations:



Corresponding intensity variations:



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

Nonlinear focusing

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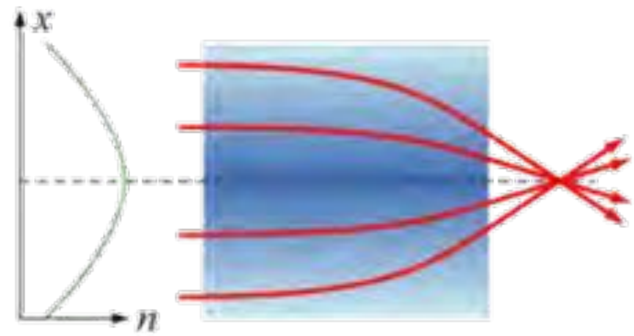
Self focusing:

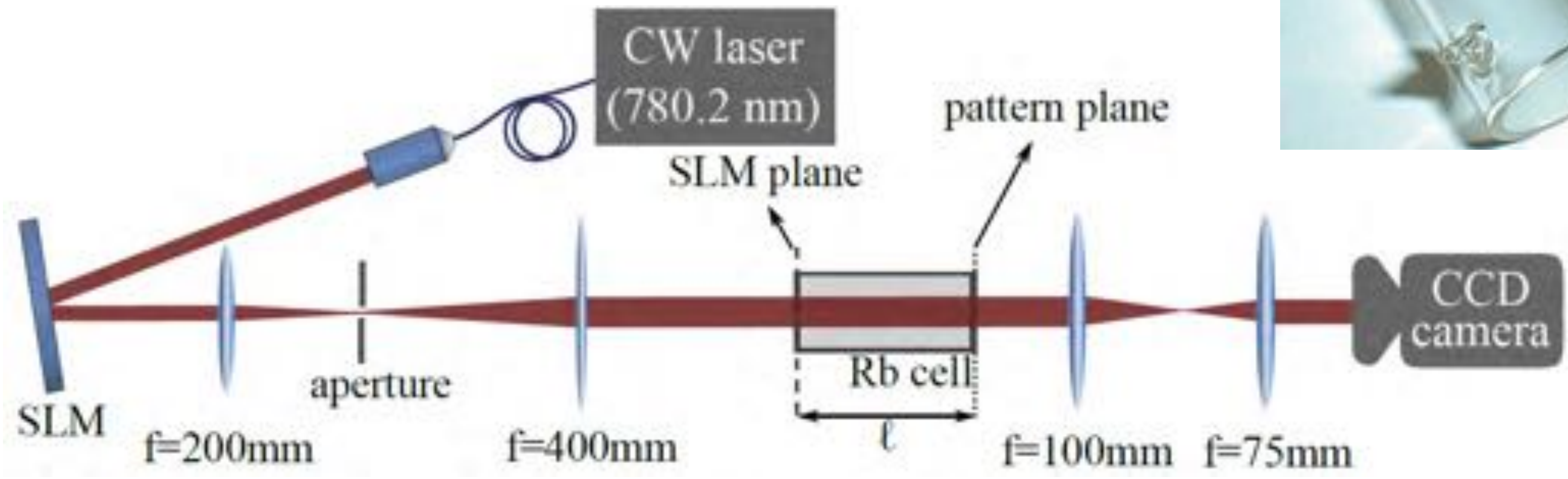
Refractive index depends on intensity:

$$n = n_0 + n_2 I$$



Rubidium cell

Rubidium vapors show large nonlinear effects

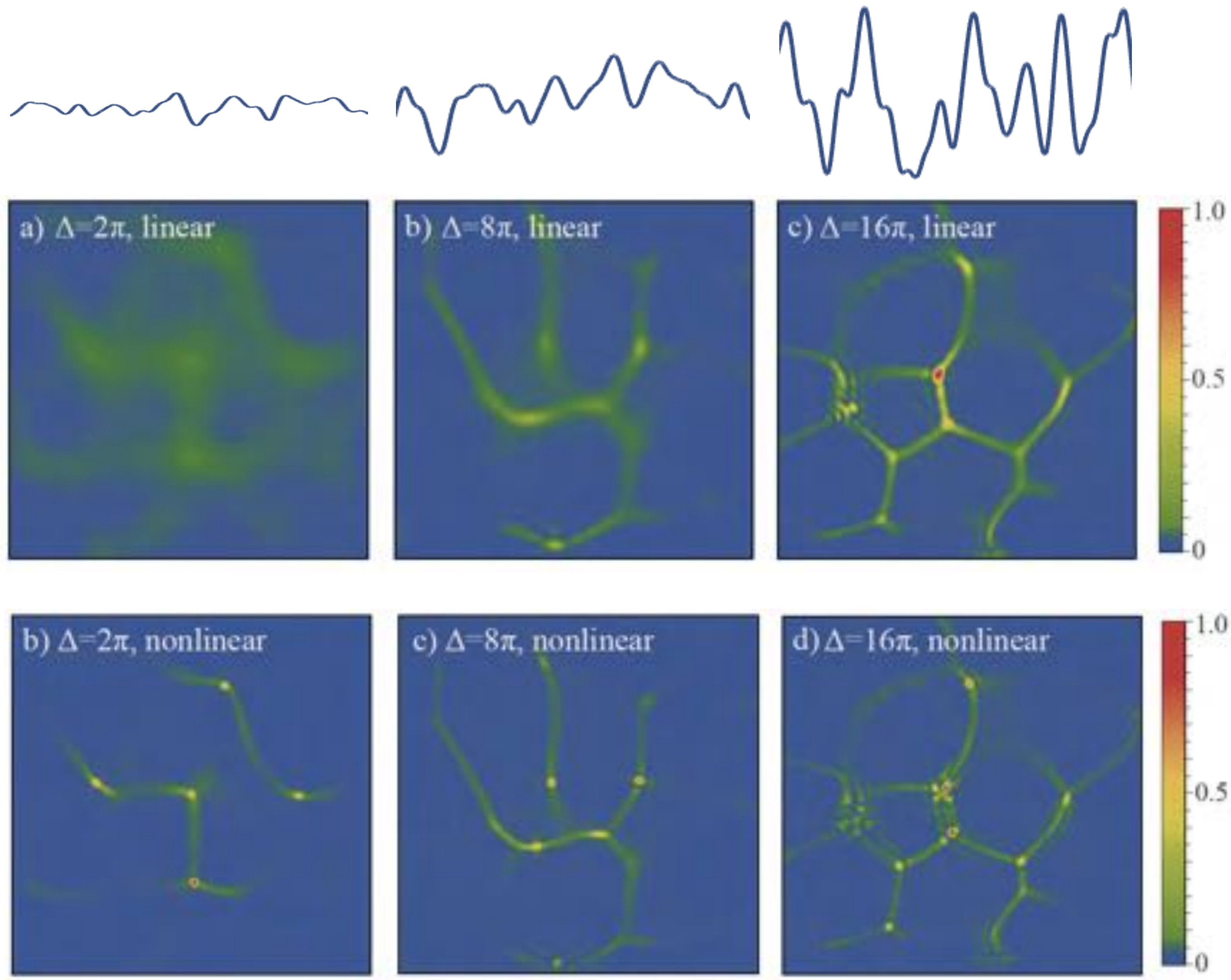


Effect of nonlinearity on caustics

Phase variations:

After linear propagation:

After nonlinear propagation:

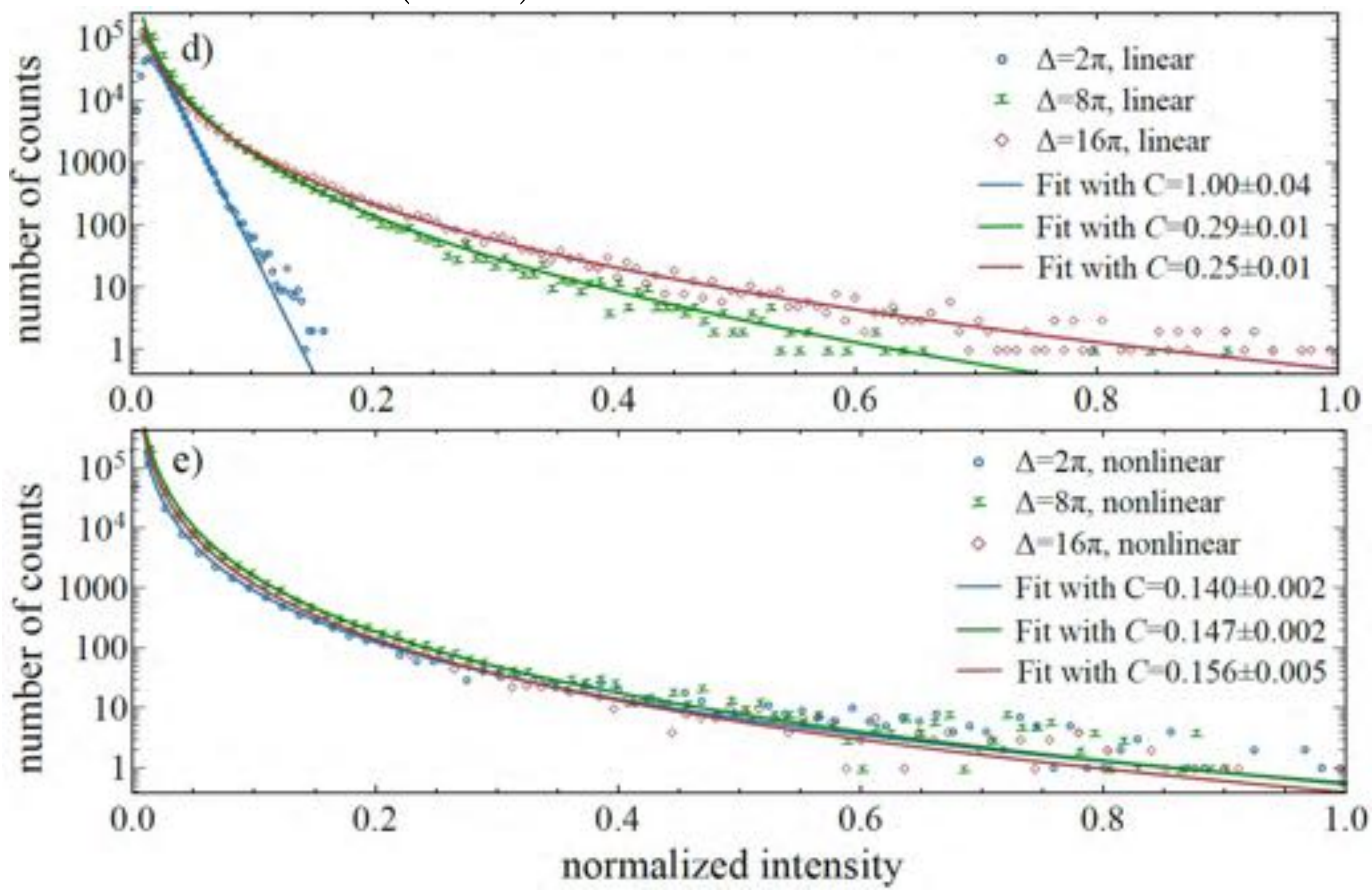


Statistics of caustics

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

Linear propagation:

After nonlinear propagation:

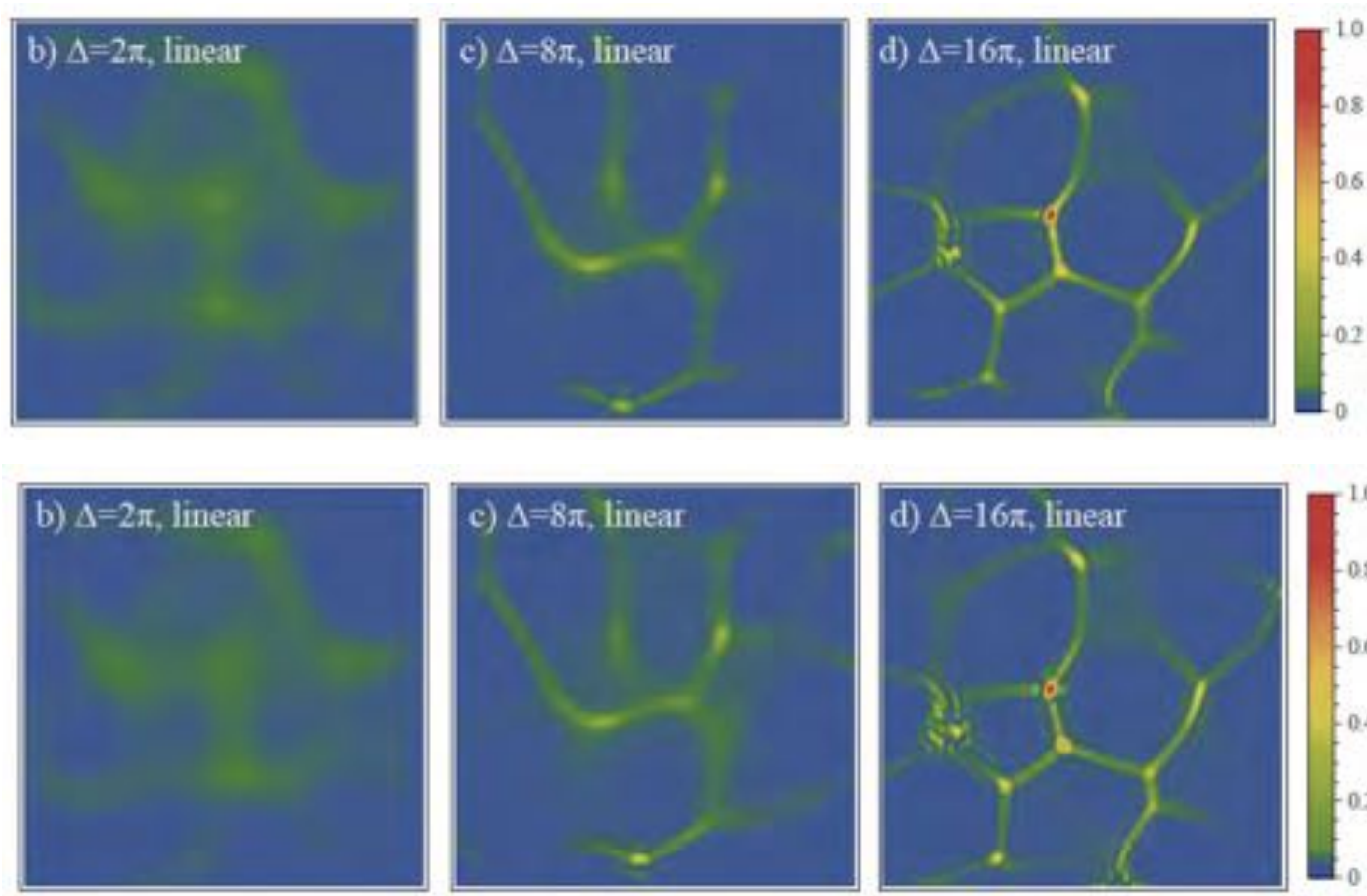


Simulation – Linear propagation

Linear propagation was simulated by FFT beam propagation

Experiment:

Simulation:



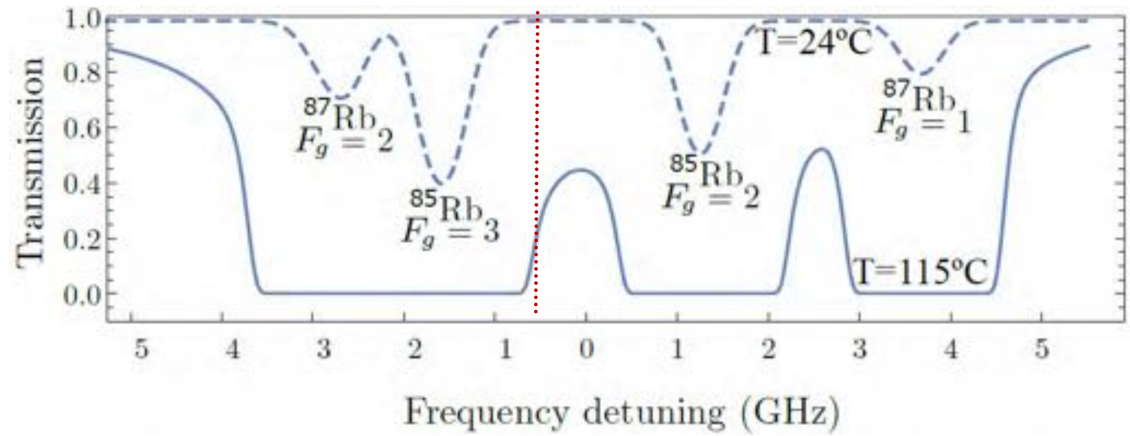
Simulation – Rb model

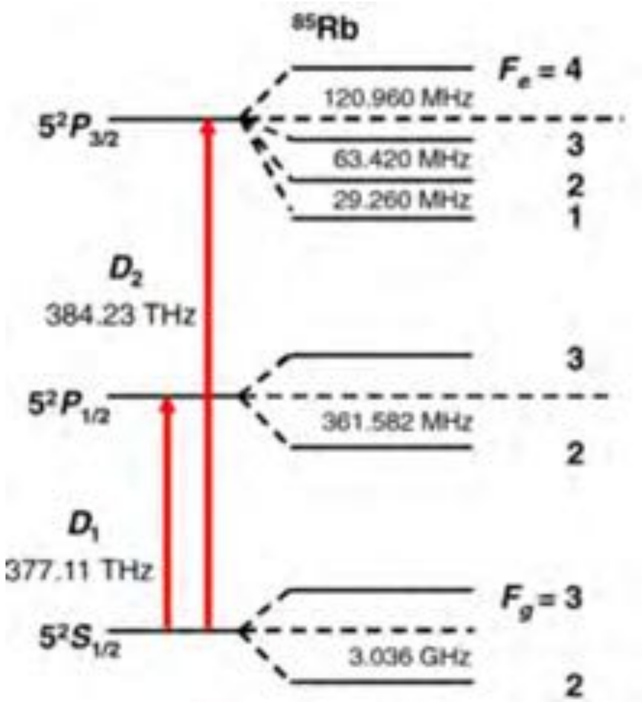
NLSE: $$\frac{\partial \mathcal{E}}{\partial z} - \frac{i}{2k}\nabla_\perp^2 \mathcal{E} = \frac{ik}{2\epsilon_0}P$$

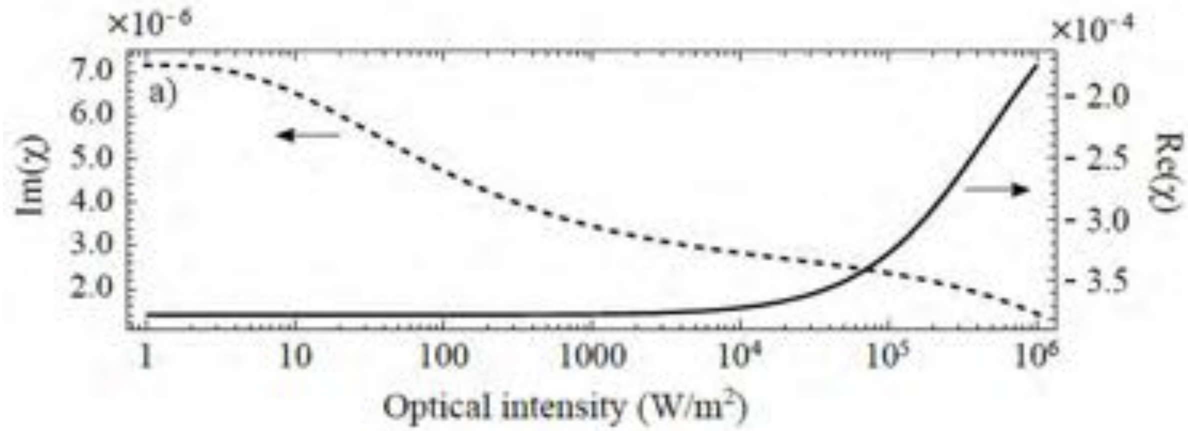
Atomic polarization: $$P = \epsilon_0 \chi \mathcal{E}$$

Our Rb model includes:

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





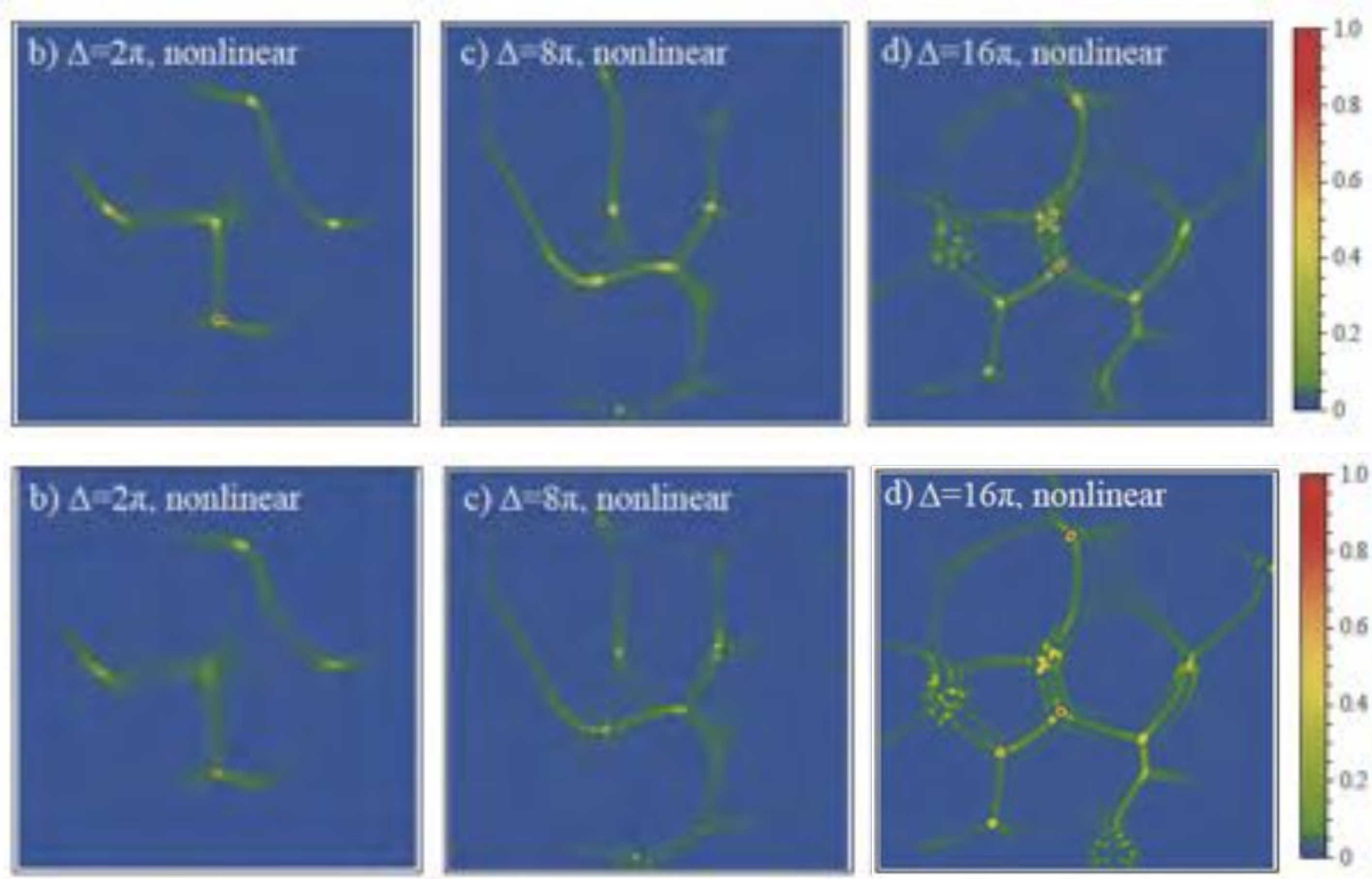


Simulation – Nonlinear propagation

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)

Conclusions

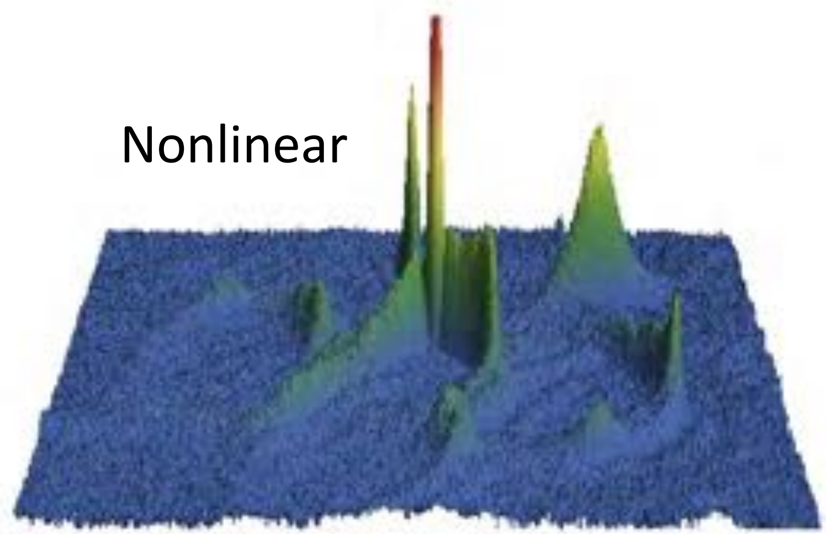
- Caustics are rogue waves!

- Generation of caustics by linear propagation requires large phase fluctuations

- Nonlinear effects can enhance the generation of caustics.

Linear

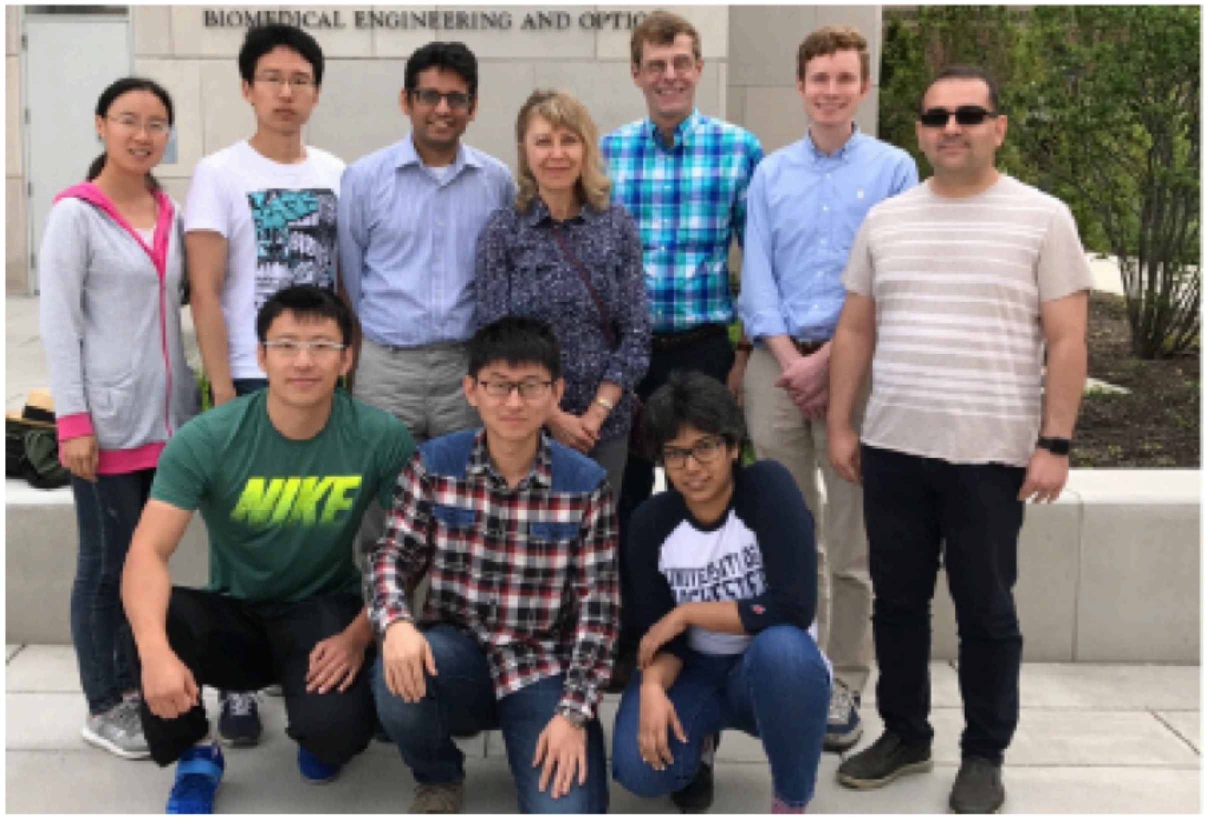
Nonlinear



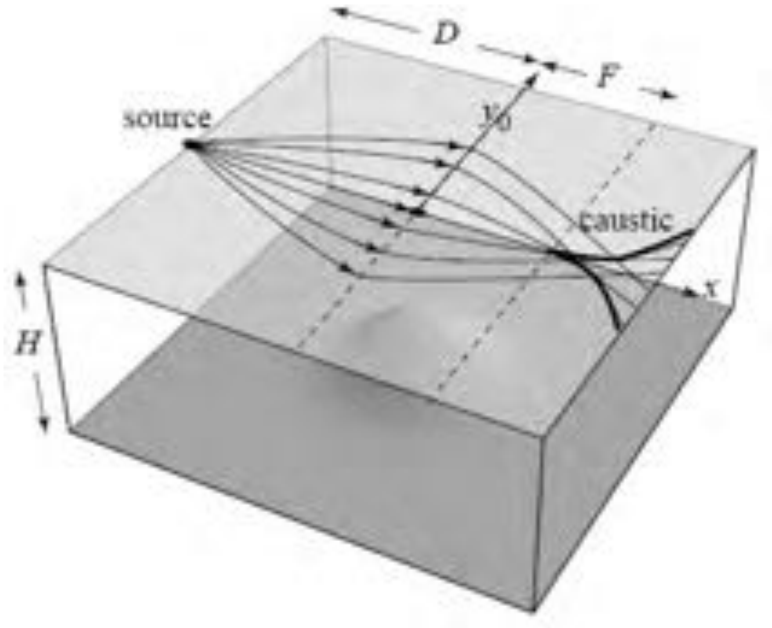
Special Thanks To My Students and Postdocs!

Ottawa Group

Rochester Group

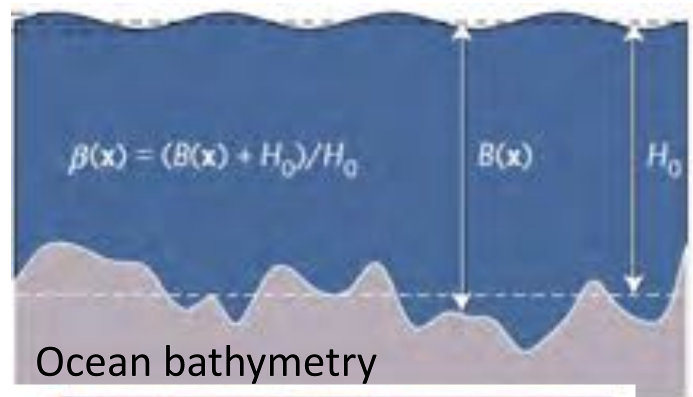


Caustics in ocean waves



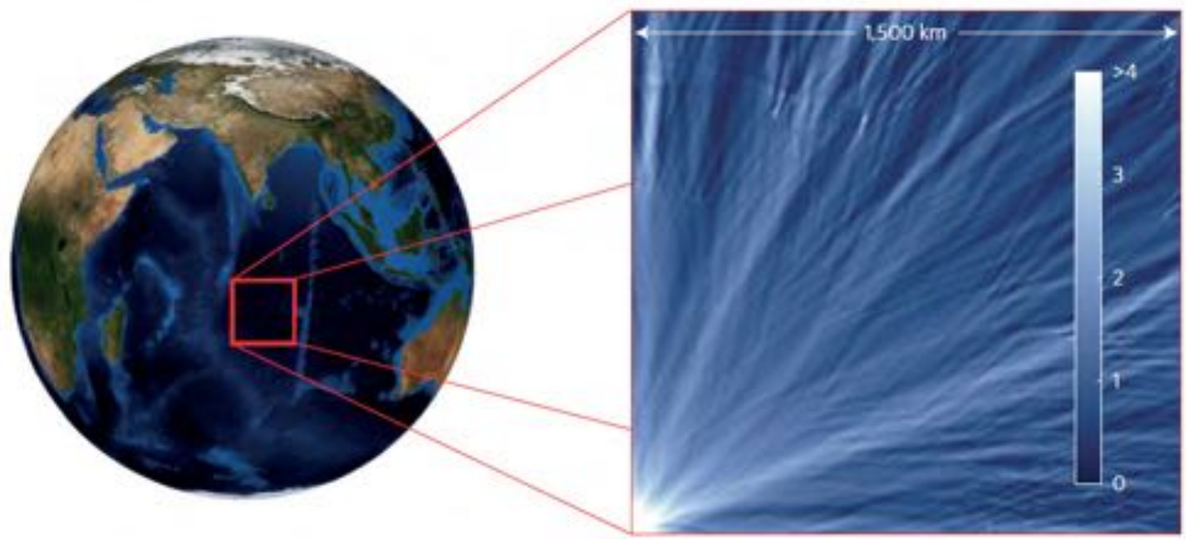
Caustic of tsunami focused by an underwater island lens

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



Ocean bathymetry

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



H. Degueldre, J. Metzger, T. Geisel and R. Fleischmann, *Nature Physics* **12, 259** (2016).