Some new results in nonlinear optics: epsilon-near-zero materials, preventing beam filamentation, and the nature of rogue waves

Robert W. Boyd

Department of Physics and
Max-Planck Centre for Extreme and Quantum Photonics
University of Ottawa

The Institute of Optics and
Department of Physics and Astronomy
University of Rochester

Department of Physics and Astronomy
University of Glasgow

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

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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
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)

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Physics of Epsilon-Near-Zero (ENZ) Materials

- ENZ materials possess exotic electromagnetic properties

- If the dielectric permittivity $\varepsilon$ is nearly zero, then refractive index $n = \sqrt{\varepsilon}$ is 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).

- 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!
  \[ n = 0 \quad \quad n = 1 \]


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

Some Consequences of ENZ Behaviour - 1

- Funny lenses

  \[ n = 0 \]


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

  \[ L \]

  \[ L \gg \lambda_{\text{vac}} \]

  \[ \text{gain medium, } n = 0 \]


- No Fabry-Perot interference

  \[ n = 0 \]

  O. Reshef et al., ACS Photonics 4, 2385, 2017.
**Some Consequences of ENZ Behaviour - 2**

- **Super-coupling (of waveguides)**
  
  ![Diagram of dielectric waveguide and metal cladding with n = 0]


- **Large evanescent tails for waveguide coupling**

  \[
  n = 0 \\
  \]

  transverse profile of upper waveguide extends to lower waveguide for any distance

  dielectric waveguide

- **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

  ![Diagram of light propagation in and out]

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.

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 \( \chi^{(3)} \)

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

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 Re \( \epsilon = 0 \) for \( \omega = \omega_p/\sqrt{\epsilon_\infty} \equiv \omega_0 \).

The region near \( \omega_0 \) is known as the epsilon-near-zero (ENZ) region.

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

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 $\beta$ 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 $1.25 \mu\text{m}$ and $60$ deg. This value is $2000$ times larger than that away from ENZ region.
- $n_2$ is $3.4 \times 10^5$ times larger than that of fused silica
- $n_2$ is $200$ times larger than that of chalcogenide glass
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

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

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\]

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:

Figure 5: The material exhibits extremely large $n_2$ over a broad spectral range. The magnitude of the on-resonance value is 7 orders of magnitude larger than that of SiO$_2$. 

Figure 5: The material exhibits extremely large $n_2$ over a broad spectral range. The magnitude of the on-resonance value is 7 orders of magnitude larger than that of SiO$_2$. 

~ $2 \times 10^7 n_2$ (SiO$_2$)
• A broadband nonlinear material with $n_2$ values up to 7 orders 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.
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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 ELEKTRONS 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

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.
SELF-TRAPPING OF OPTICAL BEAMS*

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

\[ n = n_0 \]
\[ n = n_0 + \delta n \]
\[ P_{cr} = \frac{\pi (0.61)^2 \lambda_0^2}{8n_0n_2} \]

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

![Graph showing exponential growth rate vs. transverse wavevector](image)
Optical Solitons

Field distributions that propagate without change of form

Temporal solitons (nonlinearity balances gvd)

\[
\frac{\partial \tilde{A}_s}{\partial z} + \frac{1}{2} ik_2 \frac{\partial^2 \tilde{A}_s}{\partial \tau^2} = i \gamma |\tilde{A}_s|^2 \tilde{A}_s.
\]

1973: Hasegawa & Tappert
1980: Mollenauer, Stolen, Gordon

Spatial solitons (nonlinearity balances diffraction)

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

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**

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

- **Loss of spatial coherence**
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 means 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?
• 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 $2\ell$ filaments.
  (But aberrated OAM beams tend to break into $2\ell + 1$ filaments.)

Space-Varying Polarized Light Beams

- Vector Vortex Beams

\[ \frac{1}{\sqrt{2}} \left( \begin{array}{c} \ell = -1 \\ \ell = 1 \end{array} \right) + \begin{array}{c} \ell = -1 \\ \ell = 1 \end{array} \right) = \text{Radial} \\
\frac{1}{\sqrt{2}} \left( \begin{array}{c} \ell = -1 \\ \ell = 1 \end{array} \right) + i \begin{array}{c} \ell = -1 \\ \ell = 1 \end{array} \right) = \text{Spiral} \\

- Poincare Beams

\[ \frac{1}{\sqrt{2}} \left( \begin{array}{c} \ell = 0 \\ \ell = 1 \end{array} \right) + \begin{array}{c} \ell = 0 \\ \ell = 1 \end{array} \right) = \text{Lemon} \\
\frac{1}{\sqrt{2}} \left( \begin{array}{c} \ell = 0 \\ \ell = -1 \end{array} \right) + \begin{array}{c} \ell = 0 \\ \ell = -1 \end{array} \right) = \text{Star} \\

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

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

\[
\frac{\partial E_L}{\partial \zeta} - \frac{i}{2} \nabla^2 E_L = i\gamma \frac{|E_L|^2 + \nu |E_R|^2}{1 + \sigma \left( |E_L|^2 + \nu |E_R|^2 \right)} E_L \\
\frac{\partial E_R}{\partial \zeta} - \frac{i}{2} \nabla^2 E_R = i\gamma \frac{|E_R|^2 + \nu |E_L|^2}{1 + \sigma \left( |E_R|^2 + \nu |E_L|^2 \right)} E_R
\]

- Comparison

\[
\begin{array}{ccccc}
\gamma = 0 & \gamma = \pi/8 & \gamma = \pi/4 & \gamma = 3\pi/8 & \gamma = \pi/2 \\
\text{Simulation} & \text{Simulation} & \text{Simulation} & \text{Simulation} & \text{Simulation} \\
\text{Experimental} & \text{Experimental} & \text{Experimental} & \text{Experimental} & \text{Experimental}
\end{array}
\]
Conclusions: stability of vector OAM beams

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

Summary

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

- If you want to learn more:
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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?

Oceanic rogue waves

Before 1995

Sailors: we see gigantic waves.
Scientists: it is a fairy tale!

Ocean waves follow Gaussian distribution.

\[ P \propto e^{-A^2} \]
Oceanic rogue waves

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

- 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.

\[
\frac{\partial A}{\partial x} + \frac{1}{2} ik_2 \frac{\partial^2 A}{\partial t^2} = i\gamma |A|^2 A
\]


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
Statistics of caustics

Caustics exhibit long-tailed probability distribution

1000 different patterns for each $\Delta$

Intensity distributions with fit to $A \exp(-BI^C)$

Nonlinear focusing

Self focusing:
Refractive index depends on intensity:

\[ n = n_0 + n_2 I \]

Rubidium vapors show large nonlinear effects
Effect of nonlinearity on caustics

Phase variations:

After linear propagation:

After nonlinear propagation:
Intensity distributions with fit to $A \exp(-BI^C)$

Linear propagation:

After nonlinear propagation:
Linear propagation was simulated by FFT beam propagation
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:

- Caustics are rogue waves!

- Generation of caustics by linear propagation requires large phase fluctuations

- Nonlinear effects can enhance the generation of caustics.
Special Thanks To My Students and Postdocs!

Ottawa Group

Rochester Group
Caustics in ocean waves

Caustic of tsunami focused by an underwater island lens

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