







## What's New in Nonlinear Optics

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The visuals from my talk are posted on my website 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.

### 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



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)  $n_2(FBAG) \approx 10^{14} n_2(silica)$  [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.





#### 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



#### Difference-Frequency Generation and Optical Parametric Amplification



Optical Parametric Oscillator (very broadly tunable)



### Optical Parametric Amplication 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/cm<sup>2</sup>.





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).



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



W

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 interogation in a coincidence imaging configuration.



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

Malik, Shin, O'Sullivan. Zerom, and Boyd, Phys. Rev. Lett. 104, 163602 (2010).





### 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



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,<sup>†</sup> AND P. S. PERSHAN Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts (Received April 16, 1962)

HE interaction between electromagnetic waves and atomic matter was carried out to higher orders of perturbation theory in the early years of modem quantum mechanics.<sup>1-3</sup> The interest in the absorpton of two or more light quanta and scattering processes, in which three or more light quanta are involved, has recently been revived,<sup>4-7</sup> 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 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



- In each case, scale size of inhomogeneity << 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<sub>2</sub> and the conjugated polymer PBZT.



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

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

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

Fischer, Boyd, Gehr, Jenekhe, Osaheni, Sipe, and Weller-Brophy, Phys. Rev. Lett. 74, 1871 (1995).



#### Quadratic EO effect

#### 3.2 times enhancement!

Nelson and Boyd, APL 74 2417 (1999)

# Intense Field and Attosecond Physics



nitrogen wavefunction

a visible E-field; F. Krausz

atomic core

### 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).



#### Before 1995

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





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## 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.

u Ottawa

### Rogue waves in 1D vs 2D systems

uOttawa

"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.

$$\frac{\partial A}{\partial x} + \frac{1}{2}ik_2\frac{\partial^2 A}{\partial t^2} = i\gamma \left|A\right|^2 A$$

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.

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

#### Two focusing effects in 2D systems:

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

### **Optical** caustics







Coffee cup



Ray picture

- Swimming pool
- 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





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

### Statistics of caustics



#### **Caustics exhibit long-tailed probability distribution**



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

### Caustics in ocean waves



H<sub>0</sub>



## 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:



## Statistics of caustics





### Simulation – Linear propagation



#### Linear propagation was simulated by FFT beam propagation



### Simulation – Rb model



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

Our Rb model includes:

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

85Rb

120.960 MHz

63.420 MHz

29.260 MHz

361.582 MHz

3.036 GHz

2

Optical pumping

5<sup>2</sup>P<sub>3/2</sub>

5°P<sub>1/2</sub>-

D<sub>1</sub>

377.11 THz

 $5^2 S_{1/2}$ 

 $D_2$ 

384.23 THz

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



### Simulation – Nonlinear propagation



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



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.





- 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 epsilon is nearly zero, then refractive index n nearly zero Thus v(phase) = c/n is nearly infinite lambda = lambda\_vac / n is nearly infinite Everything oscillates 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(vac)

We can control spontaneous emission!

Einstein B coefficient

Stimulated emission rate = B times EM field energy density

 $B = B(vac) / n^2$ 

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

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

• Light always leaves perpendicular to surface of ENZ material!



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



### 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).

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  $n_2$ 

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  $\operatorname{Re} \epsilon = 0$  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:

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



- 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 cm<sup>2</sup>/GW = 1.1 × 10<sup>-10</sup> cm<sup>2</sup>/W at 1.25 µm and 60 deg. This value is 2000 times larger than that away from ENZ region.
- $n_2$  is 3.4 x 10<sup>5</sup> 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.

- 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

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

and

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

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

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



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



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<sub>2</sub>.

- A broadband nonlinear material with *n*<sub>2</sub> values upto 7 order of magnitude larger than that of SiO<sub>2</sub>.
- Sub-picosecond response time.
- $\Delta n \approx \pm 2.5$  over very large bandwidth.
- One can tailor the sign of the nonlinearity by simply disigning the geometric parameters of the antenna appropriately.

### Special Thanks To My Students and Postdocs!

#### Ottawa Group



#### **Rochester Group**



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).