Quantum Nonlinear Optics:
New Materials and Interactions

Robert W. Boyd

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

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

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

Presented at the Photonics for Quantum Workshop, Rochester Institute of Technology
Prospectus

• Some timing issues

• Photonics with epsilon-near-zero (ENZ) materials
  - Huge nonlinear optical response
  - Unexpected linear optical properties

• Quantum Radiometry
  - Quantum calibration of a laboratory spectrophotometer
New Nonlinear Optical Material for Quantum Photonics
(Why we care about Epsilon-Near-Zero Materials)

- We want all-optical switches that work at the single-photon level
- We need photonic materials with a much larger NLO response
- We recently reported a new NLO material with an $n_2$ value 100 times larger than those previously reported (but with some background absorption).
- Material makes use of strong enhancement that occurs in the epsilon-near-zero (ENZ) spectral region.
- A potential game changer for the field of photonics

• ENZ materials possess exotic electromagnetic properties

Recall that \( n = \sqrt{\varepsilon \mu} \) where \( \varepsilon \) is the permittivity and \( \mu \) is the magnetic permeability

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

\[
\lambda = \frac{\lambda_{\text{vac}}}{n} \quad v = \frac{c}{n}
\]

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


• 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!

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
Physics and Applications of Epsilon-Near-Zero Materials

- Physics of ENZ Materials

- Huge NLO Response of ENZ Materials and Metastructures

- Some Applications of ENZ Materials
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 $\chi^{(3)}$

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

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

$$n = n_0 + n_2I + n_4I^2 + \ldots$$
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 $\Re \epsilon = 0$ 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 of ITO at its ENZ wavelength

Indium tin oxide (ITO)

\[ \lambda = 1240 \text{ nm} \]

Fast, ultra-large response of ITO at its ENZ wavelength

- overall change in refractive index of 0.8
- sub picosecond reponse time

*M. Z. Alam et al., Science 352, 795-797 (2016)*
Some Nonlinear Optical Materials

Nonlinearity of traditional nonlinear materials:

- SiO\textsubscript{2}  \( n_2 = 3.2 \times 10^{-20} \text{ m}^2/\text{W} \)
- SiN  \( n_2 = 2.5 \times 10^{-19} \text{ m}^2/\text{W} \)  \( 10 \times \text{SiO}_2 \)
- Si  \( n_2 = 2.7 \times 10^{-18} \text{ m}^2/\text{W} \)  \( 100 \times \text{SiO}_2 \)
- Chalcogenide glasses  \( n_2 = 2.0 \times 10^{-17} \text{ m}^2/\text{W} \)  \( 600 \times \text{SiO}_2 \)

A new class of materials known as **epsilon-near-zero** materials have demonstrated incredible nonlinear properties

- Indium tin oxide (ITO)  \( n_2 = 1.1 \times 10^{-14} \text{ m}^2/\text{W} \)  \( 600 \times \text{ChG} \)
  - \( \Delta n = n_2 I = 0.7 \)

- Al-doped zinc oxide (AZO)  \( n_2 = 3.5 \times 10^{-17} \text{ m}^2/\text{W} \)  \( 2 \times \text{ChG} \)
  - \( \Delta n/n = 4.4 \)

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

Coupled resonators: ENZ resonance and nano-antennas

Concept:

SEM:

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$_2$.

Physics and Applications of Epsilon-Near-Zero Materials

• Physics of ENZ Materials

• Huge NLO Response of ENZ Materials and Metastructures

• Some Applications of ENZ Materials
Some Potential Applications of ENZ Behavior

(a) Non-magnetic isolation
- Forward direction
  - Input beam
  - Output beam
- Backward direction
  - No output beam
  - Intense input beam
- Geometry mismatch.
- Non-uniform power distribution.
- Breaking reciprocity.

(b) Full-band shifting and conjugation
- Pump
- Wideband input beam + pump
- Output beam: shifted band (forward FWM)
- Output beam: shifted band (backward FWM)

(c) High-speed tunable interferometers
- Input beam
- Pump
- NLO-ENZ
- Interference
- Output beam
- Mach-Zehnder interferometer

(d) On-demand quantum emitter
- Short pulse laser
- Quantum emitter embedded in LNO-ENZ
- Output photons
- Single photon detector
• Extremely interesting physical processes occur in ENZ materials

• ENZ materials, metamaterials, and metastructures display extremely large NLO response

• The huge, ultrafast NLO response of ENZ materials lend themselves to many important applications

The visuals of this talk are posted at boydnlo.ca/presentations
Quantum Radiometry: 
Vacuum fluctuations as a primary standard for radiometry

Samuel Lemieux, Enno Giese, Robert Fickler, 
Maria V. Chekhova, and Robert W. Boyd
Two of my research interests are radiometry and nonlinear optics. Why not combine them both into a single project?
Earlier Work on Quantum Radiometry

- Absolute measurement of detector quantum efficiency (Klyshko, Sergienko, Migdall, Polyakov, etc.)

- Earlier work (Klyshko) established that the light produced by spontaneous parametric downconversion (SPDC) can be characterized in terms of the radiometric property known as brightness (or radiance).
Goal of Our Research

• How to perform absolute calibration of a spectrophotometer?

How do we perform an absolute measurement of the vertical axis. Units of Watts per nm of spectral bandwidth.
Traditional Approach to Calibration

- Use a black body source, or a lamp calibrated to a black body source
- Theory of black body radiation is very well understood

(1) Density of field modes (number of modes per unit volume per unit frequency interval) is given by

$$\rho_\nu = \frac{8\pi\nu^2}{(c/\bar{n})^3}$$

(2) Energy per field mode is $h\nu\bar{n}$ where $\bar{n}$ is the mean number of photons per mode:

$$\bar{n} = \frac{1}{e^{(h\nu/k_B T)} - 1} \quad \text{Planck distribution}$$

(3) Energy density of black body radiation (energy per unit volume) give by

$$u_\nu = 2r_\nu h\nu \bar{n} = \frac{8\pi h\nu^3}{(c/\bar{n})^3(e^{h\nu/k_B T} - 1)} \quad \text{Planck radiation law}$$

(4) Brightness (radiance) of black body radiation (power per unit area per unit solid angle) is given by

$$B_\nu = \frac{(c/\bar{n})}{4\pi} u_\nu = \frac{2h\nu^3}{(c/\bar{n})^2(e^{h\nu/k_B T} - 1)} \quad \text{Planck radiation law}$$
Problem with Traditional Approach to Calibration

Need to use a non-optical means to determine the temperature of a black body source

This step is not easy and is prone to error.
Our New Approach to Absolute Calibration

We make use of black body radiation, but at very low temperature.*

At low temperature the emission vanishes, but the field fluctuations remain. We use these fluctuations to seed the process of spontaneous parametric down conversion (SPDC).

We calibrate our spectrophotometer with this radiation, whose strength can be traced back to Planck’s constant $h$.

Our approach builds upon the work of Klyshko, Sergienko, Migdall, Polyakov, etc., but is distinct from it

* For SPDC from 400 nm to 800 nm, room temperature is sufficiently low.
The theory of SPDC is very well developed (see, for instance, D. N. Klyshko, *Photons and Nonlinear Optics*, Gordon and Breach, 1989). Here we convey only a few key elements.

Macroscopic Hamiltonian

\[ \hat{H} \sim E_p \chi^{(2)} \hat{E}_s \hat{E}_i^\dagger + \text{h.c.} \]

Quantization with

\[ \hat{E}_j \sim \int d^3k_j \sqrt{\frac{\omega_j}{n_j}} \hat{a}_j e^{i(k_j \cdot r - \omega_j t)} \]

Vacuum amplitude

We then determine the number of downconverted signal photons per mode

\[
N(k_s) \sim \left( \frac{E_p \chi^{(2)} L}{\sqrt{n_s n_i}} \right)^2 \omega_s \omega_i \text{sinc}^2 \frac{\Delta \kappa L}{2} \]

\[ g(\lambda_s) \quad Q(\lambda_s) \quad S(\lambda_s) \]
Our Experimental Setup

We rotate the crystal to vary the phase matched wavelength. We end up with data that looks like this.

Note that

$$\omega_s \omega_i = \omega_s (\omega_p - \omega_s)$$

which give the form of the parabola shown in the figure.
Determining the Response Function

Laboratory measurements of the response function

There is no discernable difference between the results obtained with a calibrated lamp and those obtained with the new methods based on SPDC
Relative and Absolute Calibration

The results from the last slide demonstrate that SPDC can provide good *relative* calibration.

We also want to determine the absolute calibration, that is to determine exactly how many watts (per spectral bandwidth) are leaving the spectrometer.

The theory and data presented up till now have been taken in the limit of a weak pump wave, that is for *spontaneous* parametric downconversion. As we will show when this procedure is repeated for a strong pump wave, the process becomes one of *stimulated* parametric down conversion.

For this situation, additional information becomes available and it is possible to perform an absolute calibration.
Absolute Calibration

• The predicted signal with an intense pump beam under phase-matched conditions is given by

\[ \mathcal{N}_{PM} = \sinh^2 \left( G \sqrt{\omega (\omega_p - \omega)} \right) \]

where we have introduced the gain parameter \( G = c \frac{L \chi^{(2)} E_p}{\sqrt{n n_i}} \),

• Note that the shape of the curve of \( \mathcal{N}_{PM} \) versus omega changes with pump intensity. By measuring the shape of this curve, we can determine \( G \) and thus perform an absolute calibration.
Conclusions

We have proposed and demonstrated a new protocol for calibrating a spectrophometer.

The method is based on parametric downconversion (PDC).

For a weak pump beam, the emission is induced only by vacuum fluctuation and is known as spontaneous PDC. The emission can provide a relative calibration of a spectrophometer.

For a strong pump beam, the emission is initiated by vacuum fluctuation, although the emission is then amplified by parametric gain processes. The process is then stimulated PDC, and when combined with spontaneous PDC can provide an absolute calibration of a spectrophometer.

The advantage of this method is that it relies only on basic quantum physics to provide the calibration.
Max Planck Centre for Extreme and Quantum Photonics

Research Interests:
Nonlinear optics, quantum optics integrated photonics, metamaterials, etc.
Special Thanks To My Students and Postdocs!

Ottawa Group

Rochester Group