Designer Materials for Photonics

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The visuals of this talk are posted at BoydNLO.ca/presentations

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Implications of Low-Index Behavior for Nonlinear Optics

Here is the intuition for why the low-index 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 low-index 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

$$n = n_0 + n_2 I + n_4 I^2 + \ldots$$
How Light Behaves when the Refractive Index Vanishes

- Physics of Near-Zero Index (NZI) and Epsilon-Near Zero (ENZ) Materials
- Nonlinear Optical Properties of NZI and ENZ Materials
- Metamaterials for NZI and ENZ Studies
- Applications of NZI and ENZ Materials
Giant Nonlinear Response of ENZ Metastructures

- Nonlinear Optics is important for a variety of reasons:
  
  Photonic Devices
  - All-optical switching, buffers and routers based on slow light
  Used to create quantum states of light for
  - Quantum Computing/Communications/Imaging
  Fundamental understanding of light-matter interactions
  - Not “just” Lorentz oscillator formalism
  - Understand rogue waves
  - Control filamentation process

- However, the nonlinear response is usually much weaker than the linear response

- Means to enhance the nonlinear response
  - Resonance interactions (atomic vapors)
  - Plasmonic systems
  - Electromagnetically induced transparency (EIT)
  - Metamaterials (composite materials)

- Our approach: Use epsilon-near-zero (ENZ) materials and metamaterials
• The wavelength of light is given by

\[ \lambda = \lambda_{\text{vac}} / n \]

and is significantly lengthened in a NZI material. The wavelength approaches infinity as \( n \) approaches zero.

• The phase velocity of light is given by

\[ v = c / n \]

and also approaches infinity as \( n \) approaches zero.

• For \( n \) approaching zero, the field oscillates in time but not in space; oscillations are in phase everywhere

Some Details from Electromagnetic Theory

- The linear response of any material to electromagnetic radiation can be described by

  - The dielectric permittivity (dielectric constant) $\varepsilon$ define through the relation
    \[ D = \varepsilon_0 \varepsilon E \]
    where $D$, known as the dielectric displacement, and $E$, known as the electric field, are the two fields that describe the material response to an electric field.

  - The magnetic permeability $\mu$ define through the relation
    \[ B = \mu_0 \mu H \]
    where $B$, known as the magnetic field, and $H$, known as the magnetic intensity, are the two fields that describe the magnet response of a material to an applied field.

- It is straightforward to shown from the equations of electromagnetism that
  \[ n = \sqrt{\varepsilon \mu} \]
  Thus, $n=0$ when either $\varepsilon = 0$ or $\mu=0$ (or both $\varepsilon$ and $\mu$ equal zero).

- Terminology:
  ENZ: epsilon near zero
  MNZ: mu near zero
  EMNZ: epsilon and mu near zero
There is a problem getting light into a zero-index material.

There is always reflection from the boundary between two materials.

The impedance and surface reflectivity are given by

\[ Z = \sqrt{\mu / \varepsilon} \quad R = \left| \frac{Z - 1}{Z + 1} \right|^2 \]

Thus the reflectivity will be 100% if \( \varepsilon = 0 \) unless \( \mu = 0 \) as well (with \( \varepsilon/\mu \) finite).

This is one reason for the interest in developing EMNZ materials (epsilon and mu near zero materials).
Physics of Epsilon-Near-Zero (ENZ) Materials

• 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
  \[ B = B_{\text{vac}} / (n \, n_g) \]
  Optical gain is very large!

  Einstein, Physikalische Zeitschrift 18, 121 (1917).

  Equations are shown for nonmagnetic ($\mu = 1$) materials

• Implications:
  - If we can inhibit spontaneous emission, we can build thresholdless lasers.
  - Expect superradiance effects to be pronounced in ENZ materials.
Optics of Zero-Index Materials

- 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!

\[ n = 0 \quad \text{and} \quad n = 1 \]


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

Light enters at normal incidence but leaves in all directions.


(wave-optics simulation - O. Reshef)
Some Consequences of ENZ Behavior - 1

- “Funny” lenses

\[ n = 0 \]


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

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

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


- No Fabry-Perot interference

\[ n = 0 \]

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

- Super-coupling (of waveguides)
  
  ![Diagram](image)


- Coupling between two distant waveguides
  
  ![Diagram](image)

  Mode of upper waveguide beams into the lower waveguide even for large separation

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

- Automatic phase matching of NLO processes
  
  - Recall that we need $\Delta k = 0$, but when $n=0$ $k = n \omega / c$ vanishes and so does $\Delta k$.

  - We have observed this effect in a Dirac-cone, zero-index metamaterial.

  - Usual four-wave mixing process

  ![Diagram](image)

  - With zero-index materials we can have

  ![Diagram](image)
How Light Behaves when the Refractive Index Vanishes

- Physics of Near-Zero Index (NZI) Materials
- Nonlinear Optical Properties of NZI Materials
- Meta-materials for NZI Studies
- Applications of NZI Materials
• An important application in photonic technologies is optical switching.

control (for on/off switching)

• One wants a switch with fast switching times and that operates with weak control fields.

• One needs a nonlinear interaction in order for one optical field to control another field.

• A strong nonlinear response is needed. How does one quantify the strength of a nonlinear response? Two standard methods:

\[ n = n_0 + n_2 I \]

\[ P^{NL} = 3 \chi^{(3)} |E|^2 E \]

• The nonlinear coefficients are \( n_2 \) and \( \chi^{(3)} \)
Implications of ENZ Behavior for Nonlinear Optics

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How to Choose an Epsilon-Near-Zero Materials

• Electrical conductors
  All conductors display ENZ behavior at their (reduced) plasma frequency
  Recall the Drude formula
  \[ \varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \]
  Note that \( \text{Re} \varepsilon = 0 \) for \( \omega = \omega_p/\sqrt{\varepsilon_\infty} \equiv \omega_0 \).
  ENZ wavelength restricted to a limited range in the visible.

• Electrical insulators (dielectrics)
  Dielectrics can show ENZ behavior at their (optical) phonon resonance.
  ENZ wavelength restricted to a limited range in the mid-IR.

• Metamaterials
  Can design the material so that the ENZ or EMNZ wavelengths are at any desired value.

• Challenge (for any material system). For low loss, we want \( \text{Im} \varepsilon \) as small as possible at the wavelength where \( \text{Re} \varepsilon = 0 \).
Nonlinear Optics of Indium Tin Oxide (ITO)

- We recently reported that, at its ENZ wavelength, ITO possesses a nonlinear coefficient $n_2$ that is 100 times larger than those of previously reported materials [1].
- ITO is a degenerate semiconductor (so highly doped as to be metal-like).
- ITO has a large density of free electrons, and a bulk plasma frequency corresponding to a wavelength of approximately 1.24 μm.
- Dielectric properties of ITO are well described by the Drude formula.

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

- Note that aluminum-doped zinc oxide (AZO), another transparent conducting oxide, also has strong nonlinear response at its ENZ wavelength [2].

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

- ellipsometry

![Graph of permittivity vs. wavelength](image)

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

- overall change in refractive index of 0.8

![Graph of refractive index vs. intensity](image)

- \( n^2 \) can be \( 3.4 \times 10^5 \) times larger than that of silica glass

![Graph of refractive index vs. wavelength](image)

- sub picosecond response time

![Graph of time response](image)

*M. Z. Alam et al., Science 352, 795-797 (2016)*
The NLO Response Is Larger For Oblique Incidence

Standard boundary conditions show that:

\[ E_{\text{in},||} = E_{\text{out},||} = E_{\text{out}} \cos \theta \]

\[ D_{\text{in},\perp} = D_{\text{out},\perp} \Rightarrow E_{\text{in},\perp} = E_{\text{out},\perp} / \epsilon = E_{\text{out}} \cos \theta / \epsilon \]

Thus the total field inside of the medium is given by

\[ E_{\text{in}} = E_{\text{out}} \sqrt{\cos^2 \theta + \frac{\sin^2 \theta}{\epsilon}} \]

Note that, for \( \epsilon < 1 \), \( E_{\text{in}} \) exceeds \( E_{\text{out}} \) for \( \theta \neq 0 \).

Note also that, for \( \epsilon < 1 \), \( E_{\text{in}} \) increases as \( \theta \) increases.
Huge Nonlinear Optical Response of ITO

- Z-scan measurements for various angles of incidence

<table>
<thead>
<tr>
<th>Angle of incidence (θ, deg)</th>
<th>β (eff) (10^{-3}) (cm/GW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>0.95</td>
<td>0.030</td>
</tr>
<tr>
<td>1.05</td>
<td>0.060</td>
</tr>
<tr>
<td>1.15</td>
<td>0.090</td>
</tr>
<tr>
<td>1.25</td>
<td>0.120</td>
</tr>
<tr>
<td>1.35</td>
<td>0.150</td>
</tr>
</tbody>
</table>

- Both \(n_2\) and nonlinear absorption increase with angle of incidence.

- \(n_2\) shows a maximum value of 0.11 cm\(^2\)/GW = 1.1 \times 10^{-10} \text{ cm}^2/\text{W} at 1.25 \text{ μm} and 60 deg. This value is 2000 times larger than that away from ENZ region.

- Note that \(n_2\) is positive (self focusing) and \(β\) is negative (saturable absorption).

peak laser intensity was 50 GW cm\(^{-2}\)
Why is $n_2$ so large for ITO?

The short-wavelength (away from the ENZ resonance) value of $n_2$ of ITO is $5 \times 10^{-5}$ cm$^2$/GW, which is 150 times larger that of fused silica ($3.2 \times 10^{-7}$ cm$^2$/GW).

There is a $43 \times$ enhancement from working at the ENZ wavelength and an additional $43 \times$ enhancement from using non-normal incidence.

Thus $n_2 = 0.01$ cm$^2$/GW, which is $3.4 \times 10^5$ times that of fused silica.

Incidentally, for arsenic trisulfide glass, $n_2 = 2.4 \times 10^{-4}$ cm$^2$/GW, which is 800 times larger than that of fused silica.

Why Does ENZ Lead to Large NLO Response?

1. From the form of \( n_2 \)

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

2. From simple math: for \( n = n_b + \Delta n \) and \( \epsilon = \epsilon_b + \Delta \epsilon \)

\[
\Delta n = \frac{\Delta \epsilon}{2n_b}
\]

3. Note behavior of wave equation for \( \epsilon = 0 \)

\[
\nabla \times \nabla \times \mathbf{E} + \frac{\epsilon \mu}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E} = -\mu \frac{\partial^2 \mathbf{P}^{\text{NL}}}{\partial t^2}
\]

4. When a collimated laser beam enters a material its intensity remains constant. Recall that

\[
I = \frac{1}{2} n\epsilon_0 c |E|^2
\]

Thus for small \( n \) the electric field strength is increased, giving a large nonlinear response

5. Detailed numerical integration confirms this behavior.
• We solve the standard equations for second-harmonic generation

\[
\frac{dA_1}{dz} = i \eta_1 \omega_1 \chi^{(2)} \frac{1}{c} A_2(z) A_1^*(z) e^{-i \Delta k z},
\]

\[
\frac{dA_2}{dz} = i \eta_2 \omega_2 \chi^{(2)} \frac{1}{2c} A_1^2(z) e^{i \Delta k z},
\]

• We take \( \Delta k = 0 \) and plot the solution for various values of the permittivity \( \epsilon \).

• We find that the growth rate increases dramatically as the permittivity is decreased.

\[A_1(0) = 5 \times 10^8 \text{ [V/m]}\]

\[\chi^{(2)} = 5 \times 10^{-12} \text{ [m/V]}\]
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:

A thin ENZ medium supports a bulk plasma mode.

A thin layer of ITO supports two modes
- the bulk plasma mode, also called the ENZ or long-range SPP mode
- the short range surface plasmon polariton (SPP) mode
NLO response of the coupled antenna-ENZ system

The structure exhibits an extremely large $n^2$ value over a broad spectral range. The on-resonance $n^2$ value is seven orders of magnitude larger than that of silica glass.

$\sim 2 \times 10^7 n^2 (\text{SiO}_2)$


The structure exhibits an extremely large $n^2$ value over a broad spectral range. The on-resonance $n^2$ value is seven orders of magnitude larger than that of silica glass.
Physics and Applications of Epsilon-Near-Zero Materials

• Physics of ENZ Materials

• Huge NLO Response of ITO and ITO Metastructures

• Materials for ENZ

• Applications of ENZ Materials
Epsilon-Near-Zero (ENZ) and Near Zero-Index (NZI) Materials

Homogeneous materials

**TCO**

- Bi\(_{1.5}\)Sb\(_{0.5}\)Te\(_{1.8}\)Se\(_{1.2}\)

**SiC**

- ZnO, Al\(_2\)O\(_3\), Ga\(_2\)O\(_3\), Al\(_2\)ZnO, Ga\(_2\)ZnO

**Bi\(_{1.5}\)Sb\(_{0.5}\)Te\(_{1.8}\)Se\(_{1.2}\)**

Metamaterials

- Chan, Huang et al., Nat. Mater. (2011)
- Vesseur et al., PRL (2013)
- Re(\(\varepsilon\)) = 0

Wire SEM from: Zayat & Podolskiy
Pollard et al., PRL (2009)
StackSEM from: Polman & Engheta
Mass et al., Nat. Photon. (2013)
Giant Nonlinear Response of ENZ Metastructures: Our Team

**Nader Engheta**
- H. Nedwill Ramsey Professor at the University of Pennsylvania
- B.S. degree from the University of Tehran and his M.S and Ph.D. from Caltech.
- Activities include ENZ, photonics, metamaterials, nano-optics, graphene optics, electrodynamics, microwave and optical antennas, studies of fields and waves.
- Many awards including the Streifer Award of IEEE and the Gold Medal from SPIE

**Eric Mazur**
- Balkanski Professor of Physics and Applied Physics at Harvard University
- Ph.D. University of Leiden.
- Activities include light-matter interactions with ultrashort laser pulses, nonlinear optics at the nanoscale, and zero-index dielectric metamaterials.
- Awards include the Beller Award of OSA and the Millikan Medal of the AAPT

**Alan Willner**
- Steven & Kathryn Sample Chair in Engineering at the University of Southern California.
- Ph.D. Columbia University
- Honors include Member of US National Academy of Engineering; Int’l Fellow of UK Royal Academy of Engineering; President of OSA and of IEEE Photonics Society.
- Activities include using nonlinearity for signal processing and wave manipulation.
Three Metamaterial 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)
Do layered metamaterials also show enhanced NLO response at ENZ wavelength?

- By controlling the metallic fill fraction $\rho$, we can set the ENZ wavelength to be anywhere from 300 to 700 nm. We use $\rho = 0.2$, which corresponds to 500 nm. We deposit five layer pairs

- Note that the real part of epsilon vanishes at 508 nm, close to the design wavelength. The SEM shows our structure. Ag thickness = 16 nm; silica thickness = 65 nm

- We perform Z-scan measurements on the sample. Note the enhanced response of the composite as compared to a single layer of silver.

- Note the pronounced peak in the value of $n_2$ around the ENZ wavelength. We find a good but not perfect agreement with a simple effective medium theory.

\[ \text{Suresh, Reshef, Alam, Upham, Karimi and Boyd} \]
Physics and Applications of Epsilon-Near-Zero Materials

• Physics of ENZ Materials

• Huge NLO Response of ITO and ITO Metastructures

• Materials for ENZ

• Applications of ENZ Materials
All-Optical, Nanoscale, Sub-Picosecond Beam Steering

• Concept
  Tune output direction by +/- 20 degrees under optical or electrical control
  Beam steerer can be made of one or many cells
  Sub-picosecond response time

• Design
  Top View of Cell
  nanoantennas
  Nonlinear response depends on antenna length
  phase ramp on reflected beam

• First results
  One application: Mode-division multiplexing for telecommunications

- Concept
  - 20 deg
  - ITO 200 nm
  - Glass
  - Gold, 30 nm
  - Side View of Cell

- Design
  - Top View of Cell
  - 600 nm
  - 1400 nm
  - Incident power = 7 μW
  - Incident power = 20 μW
  - Incident power = 50 μW

- First results
Adiabatic Wavelength Conversion by Time Refraction

Experimental setup

Experimental results at 1240 nm

- The observed effect is 100 times larger with almost 100 times smaller propagation distance than previous reports of AWC.
- Application: wavelength-division multiplexing for telecom

Relaxed Phase-Matching Requirements in ENZ Media

• We study four-wave mixing in a zero-index waveguide

\[ 2\omega_p = \omega_s + \omega_i \]

• We find that an idler field is generated in both the forward and backward directions!

• Recall that we need \( \Delta k = 0 \), but when \( n = 0 \), \( k = n \frac{\omega}{c} \) vanishes for each of the interacting waves and thus so does \( \Delta k \).

• Significance: Nonlinear optical processes that were previously believed to be too weak to be useful can be excited through use 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

- Wideband input beam + pump
- Output beam: shifted band (forward FWM)
- Output beam: shifted band (backward FWM)

(c) High-speed tunable interferometers

- Mach-Zehnder interferometer
- Interference Output beam

(d) On-demand quantum emitter

- Short pulse laser
- Output photons
- Single photon detector
Real-Time Holography with THz Refresh Rates

- Goal: Real-time holography with video or much faster refresh rates.
- The ultrafast response of ITO permits THz refresh rates.
- Important applications involve image processing and signal processing.
- Current real-time holographic materials cannot even support video frame rates.

- Demonstration of image processing (edge enhancement)

Alam, Fickler, Reshef, Giese, Upham, and Boyd
Special Thanks To My Students and Postdocs!

Ottawa Group

Rochester Group
• 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

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