Physics and Applications of Epsilon-Near-Zero Materials

How Light Behaves when the Refractive Index Vanishes

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Physics and Applications of Epsilon-Near-Zero Materials

- Physics of ENZ Materials
  - Huge NLO Response of ENZ Materials and Metastructures
  - Non-perturbative Nature of the NLO Response
  - Some Applications of ENZ Materials
Physics of Epsilon-Near-Zero (ENZ) Materials

• 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)

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

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


• 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!

Thus light can enter an ENZ material only at normal incidence!
Maxwell Equations Prediction

• light enters slab at normal incidence
Some Consequences of ENZ Behaviour - 1

- Funny lenses

\[ n = 0 \]


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

\[ L \]

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

gain medium, \( n = 0 \)


- No Fabry-Perot interference

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

• Super-coupling (of waveguides)


• Large evanescent tails for waveguide coupling

mode of upper waveguide extends to lower waveguide for any separation

dielectric waveguide

• Automatic phase matching of NLO processes

Recall that $k = n \frac{\omega}{c}$ vanishes in an ENZ medium.
For example, the following 4WM process is allowed

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 \( n_0 = 0 \)?
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- Some Applications of ENZ Materials
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 Re $\epsilon = 0$ for $\omega = \omega_p/\sqrt{\epsilon_\infty} \equiv \omega_0$.

- **Challenge:** Obtain low-loss ENZ materials
  Want Im $\epsilon$ as small as possible at the frequency where 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.

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

$$n = n_0 + n_2 I + n_4 I^2 + \ldots$$
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 $1.24 \, \mu m$.

Recall the Drude formula

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

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

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)

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

\[ n^2 \text{ (cm}^2 / \text{GW)} \]

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

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

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

Nonlinearity of traditional nonlinear materials:

- SiO$_2$  \( n_2 = 3.2 \times 10^{-20} \text{ m}^2/\text{W} \)
- 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)  \( n_2 = 3.5 \times 10^{-17} \text{ m}^2/\text{W} \) \( 2 \times \text{ChG} \)
  - \( \Delta n/n = 4.4 \)

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
- 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$_2$.

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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 problem is that $n_2$ is not a reliable metric under ENZ conditions. But $n_2$ is the standard way for quantifying intensity-dependent index changes. What can we do?

3. 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

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

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

$$n = \sqrt{\epsilon^{(1)} + 3\chi^{(3)} |E|^2 + 10\chi^{(5)} |E|^4 + \cdots}.$$
Nonlinear Response of ITO is Nonperturbative
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Giant Nonlinear Response of ENZ Metastructures

**Boyd** (Rochester), **Engheta** (UPenn), **Mazur** (Harvard), **Willner** (USC)

### Concept

- **Nonlinear optical phenomena** currently require high-intensity sources, which are generally incompatible with nanophotonics.
- Photonics could have an enormous impact on many new fields if nonlinearity is fundamentally enhanced.
- **ENZ metastructures** provide giant tailorable nonlinearity.
- Enhanced nonlinearity will open door to manipulating light on the nanoscale.

### Impact

- **New fundamental understanding** of light-matter interaction in ENZ/EMNZ materials.
- **Complete dynamic control** of beam parameters: amplitude, phase, polarization, wavelength, and propagation direction.
- **Low-threshold NLO devices** that are integrated, fast, tunable, broadband, power-efficient.

### Approach

- Re-formulate nonlinear optics for non-perturbative regime.
- Explore experimental characteristics of ENZ/EMNZ platforms.
- Fabricate devices to exploit novel features.

### Context

- Nonlinear optical phenomena require high-intensity sources, which are generally incompatible with nanophotonics.
- Photonics could have an enormous impact on many new fields if nonlinearity is fundamentally enhanced.
- **ENZ metastructures** provide giant tailorable nonlinearity.
- Enhanced nonlinearity will open door to manipulating light on the nanoscale.

**Source Selection Information** – see FAR 2.101 & 3.104
Some Potential Applications of ENZ Behavior

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

(b) Full-band shifting and conjugation
- Pump: $\Delta \geq 100$ nm bandwidth
- NLO-ENZ
- Output beam with shifted band: (backward FWM)
- Output beam with shifted band: (forward FWM)

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

(d) On-demand quantum emitter
- NLO-ENZ
- Short pulse laser
- Output photons
- Single photon detector
Ultrafast Holography and Beam Copying

- Real-time holography with sub-picosecond response time

- Schematic of beam-copying procedure

- Laboratory results

![Schematic of beam-copying procedure](image1)

![Laboratory results](image2)

- Signal conjugate (x5)
- Signal reference beam
- Signal (x5)
- Reference beam
- Object beam
• 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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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