Quantum Nonlinear Optics:
New Materials and Interactions

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

Presented at the Photonics for Quantum Workshop, Rochester Institute of Technology
January 23, 2019.

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

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).

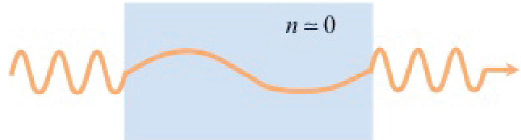
Physics of Epsilon-Near-Zero (ENZ) Materials

- ENZ materials possess exotic electromagnetic properties

Recall that $n = \sqrt{\epsilon\mu}$ where ϵ is the permittivity and μ is the magnetic permeability

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

$$\lambda = \lambda_{\mathrm{vac}}/n \qquad v = c/n$$



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

Silveirinha and Engheta, Phys. Rev. Lett. 97, 157403 (2006).

- Radiative processes are modified in ENZ materials

Einstein *A* coefficient (spontaneous emission lifetime = 1/*A*)

$A = n\, A_{\mathrm{vac}}$

We can control (inhibit!) spontaneous emission!

Einstein *B* coefficient

Stimulated emission rate = *B* times EM field energy density

$B = B_{\mathrm{vac}}\ / n^2$

Optical gain is very large!

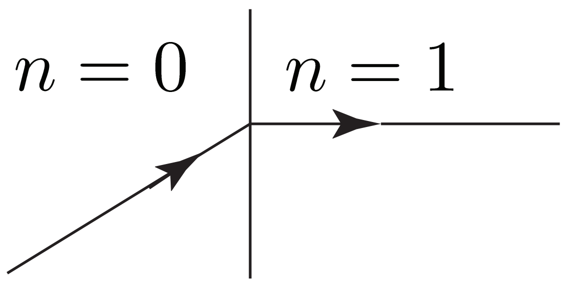
Einstein, Physikalische Zeitschrift 18, 121 (1917).
Milonni, Journal of Modern Optics 42, 1991 (1995).

Physics of Epsilon-Near-Zero (ENZ) Materials -- More

- Snell's law leads to intriguing predictions

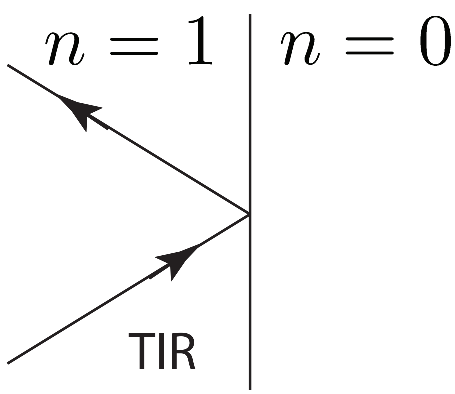
$$n_1 \sin\theta_1 = n_2 \sin\theta_2$$

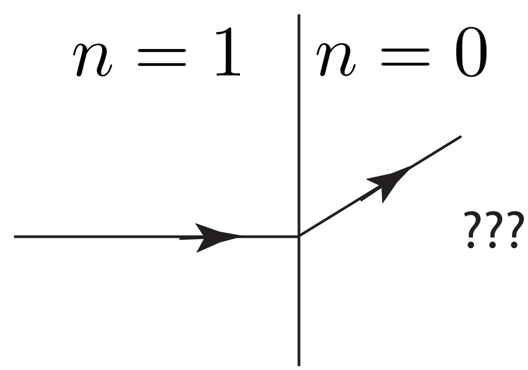
- Light always leaves perpendicular to surface of ENZ material!



Y. Li, et al., Nat. Photonics 9, 738, 2015; D. I. Vulis, et al., Opt. Express 25, 12381, 2017.

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





Y. Li, et al., Nat. Photonics 9, 738, 2015.

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

- Challenge: Obtain low-loss ENZ materials
  Want Im ε as small as possible at the frequency where Re $\varepsilon = 0$.

- 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 $\chi^{(3)}$

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

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

$$n = n_0 + n_2 I + n_4 I^2 + \ldots$$

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

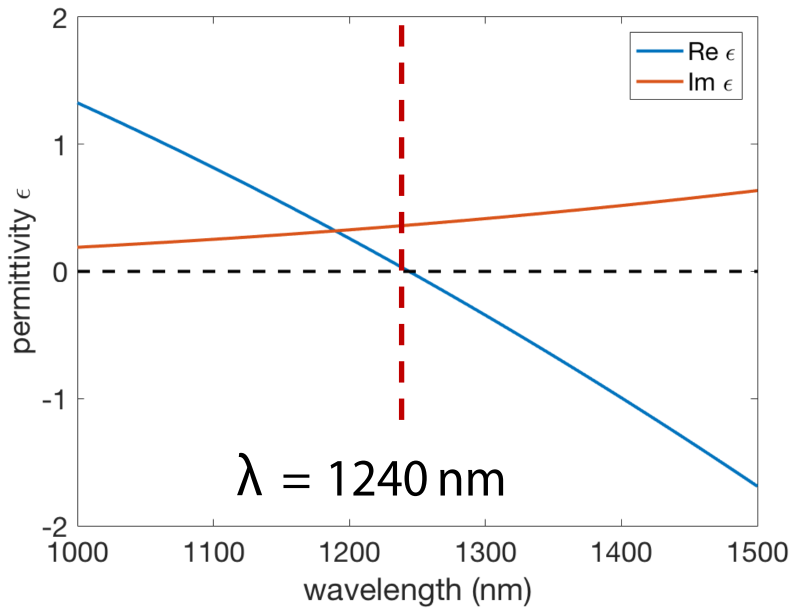
The region near ω_0 is known as the epsilon-near-zero (ENZ) region.

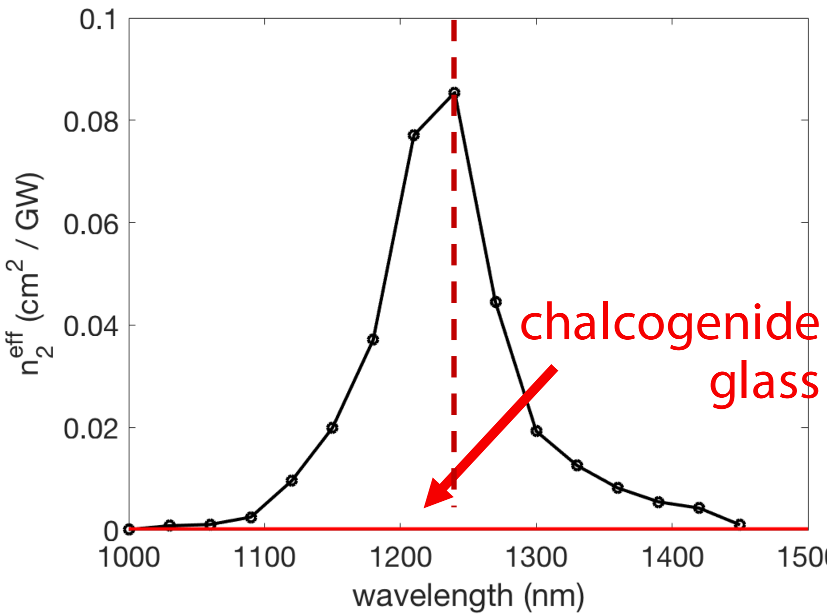
There has been great recent interest in studies of ENZ phenomena:

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 of ITO at its ENZ wavelength

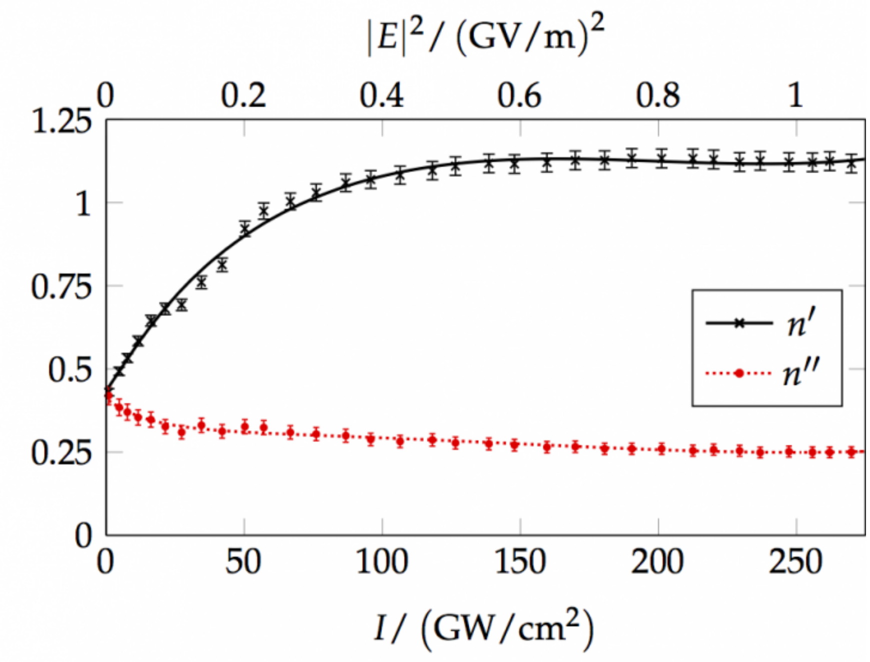
Indium tin oxide (ITO)



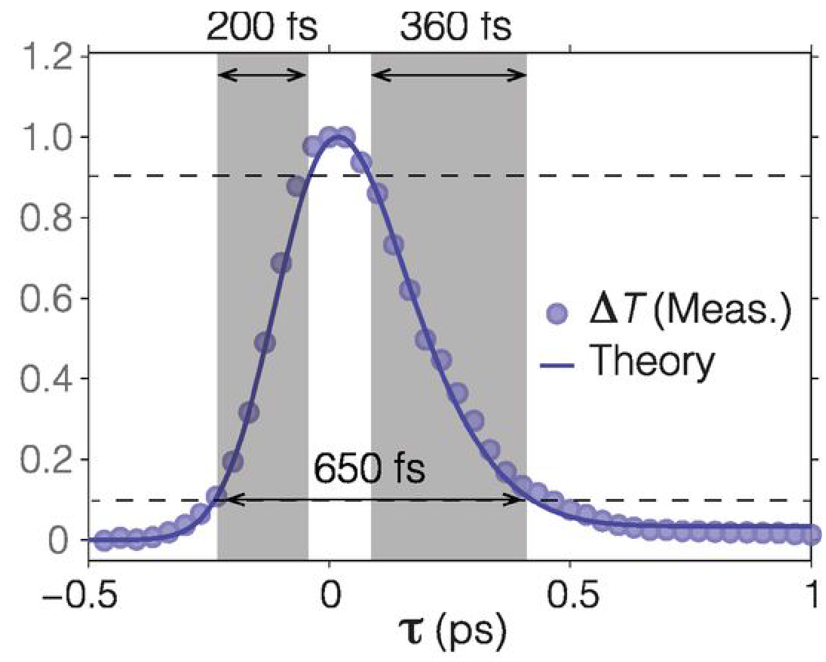


M. Z. Alam et al, Science 352, 795–797 (2016)

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_2	$n_2 = 3.2 \times 10^{-20}$ m^2/W	
• SiN	$n_2 = 2.5 \times 10^{-19}$ m^2/W	$10 \times SiO_2$
• Si	$n_2 = 2.7 \times 10^{-18}$ m^2/W	$100 \times SiO_2$
• Chalcogenide glasses	$n_2 = 2.0 \times 10^{-17}$ m^2/W	$600 \times SiO_2$

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

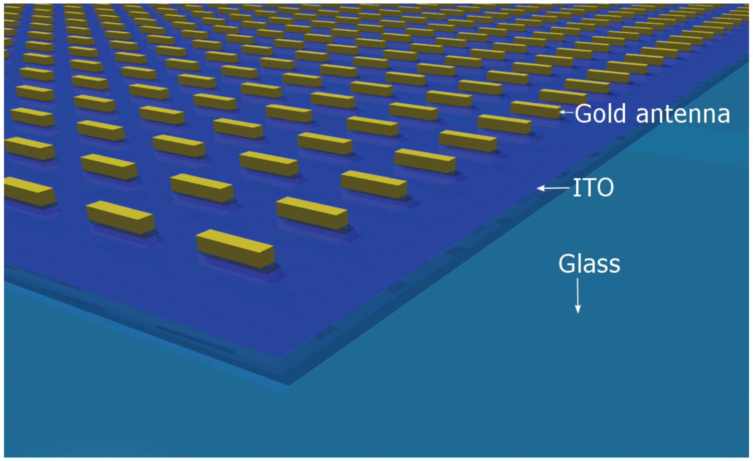
• Indium tin oxide (ITO)	$n_2 = 1.1 \times 10^{-14}$ m^2/W	$600 \times$ **ChG**
	- $\Delta n = n_2 I = 0.7$	
• Al-doped zinc oxide (AZO)	$n_2 = 3.5 \times 10^{-17}$ m^2/W	$2 \times$ **ChG**
	- $\Delta n/n = 4.4$	

R. W. Boyd, *Nonlinear Optics*, Third edition (2008), M. Z. Alam et al., *Science* 352, 795 (2016),
L. Caspani et al., *Physical Review Letters* 116, 233901(2016)

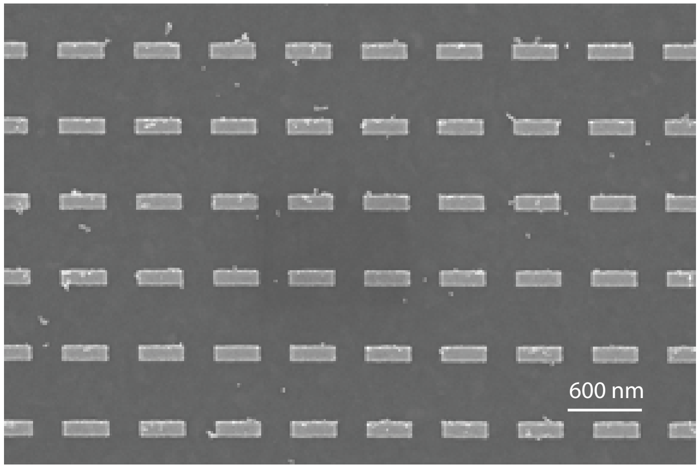
An ENZ Metasurface

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

Concept:

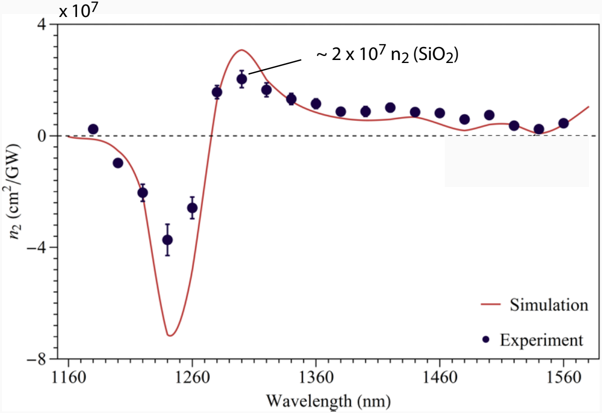


SEM:



Alam, Schulz, Upham, De Leon and Boyd, Nature Photonics 12, 79-83 (2018).

NLO response of the coupled antenna-ENZ system



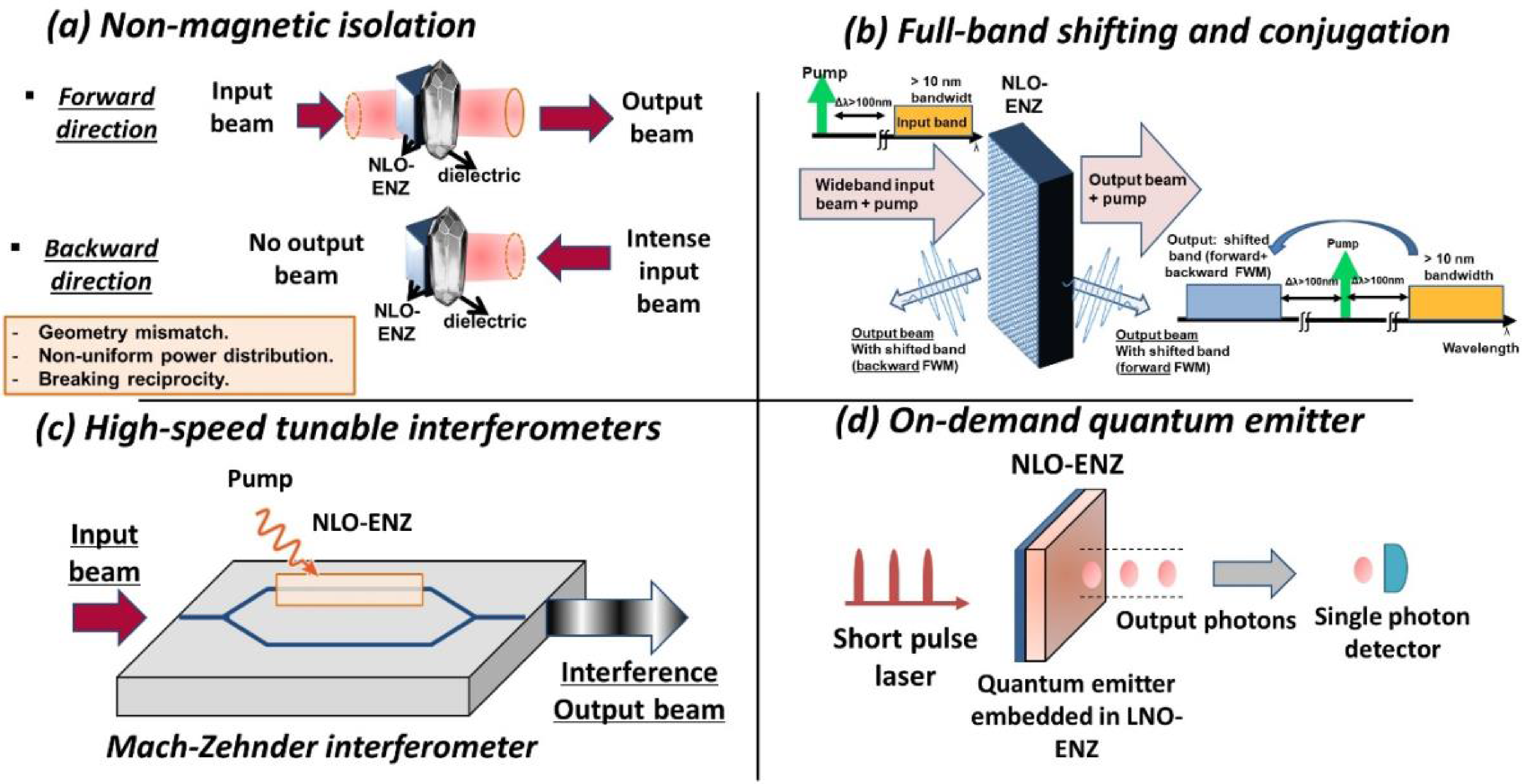
The material exhibits extremely large n_2 over a broad spectral range. The magnitude of the on-resonance value is 7 orders of magnitude larger than that of SiO_2.

Alam, Schulz, Upham, De Leon and Boyd, Nature Photonics 12, 79-83 (2018).

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



Summary: Physics and Applications of ENZ Materials

- 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

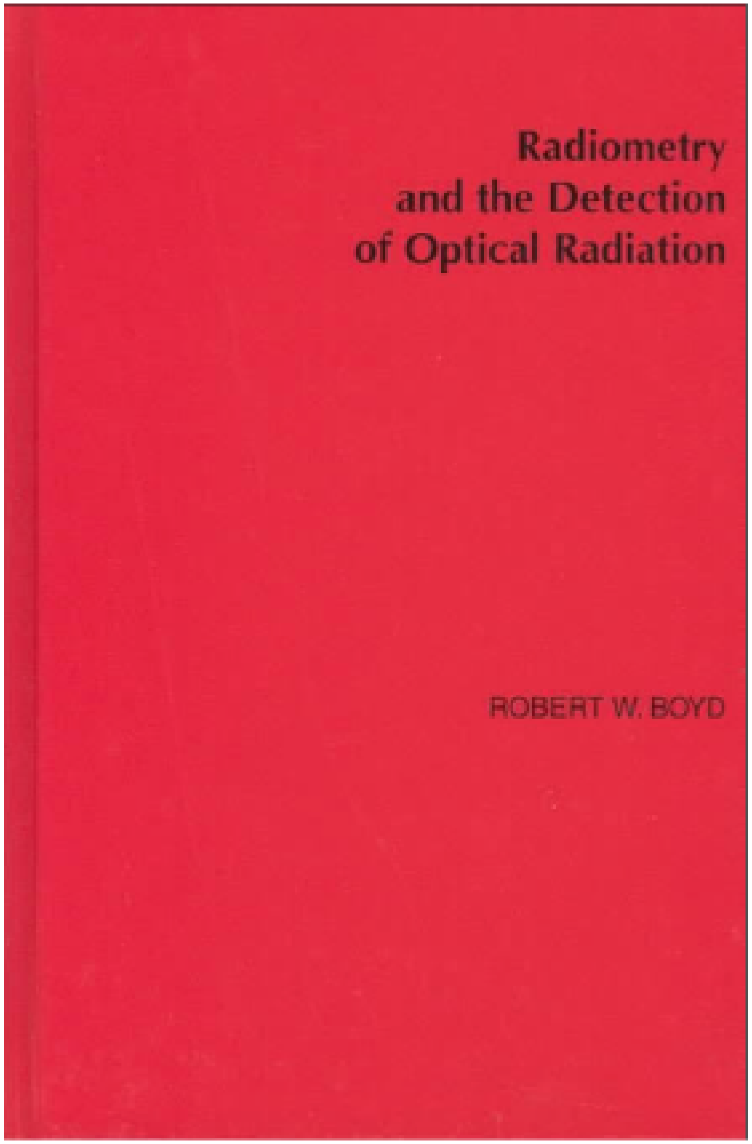
Quantum Radiometry:

Vacuum fluctuations as a primary standard for radiometry

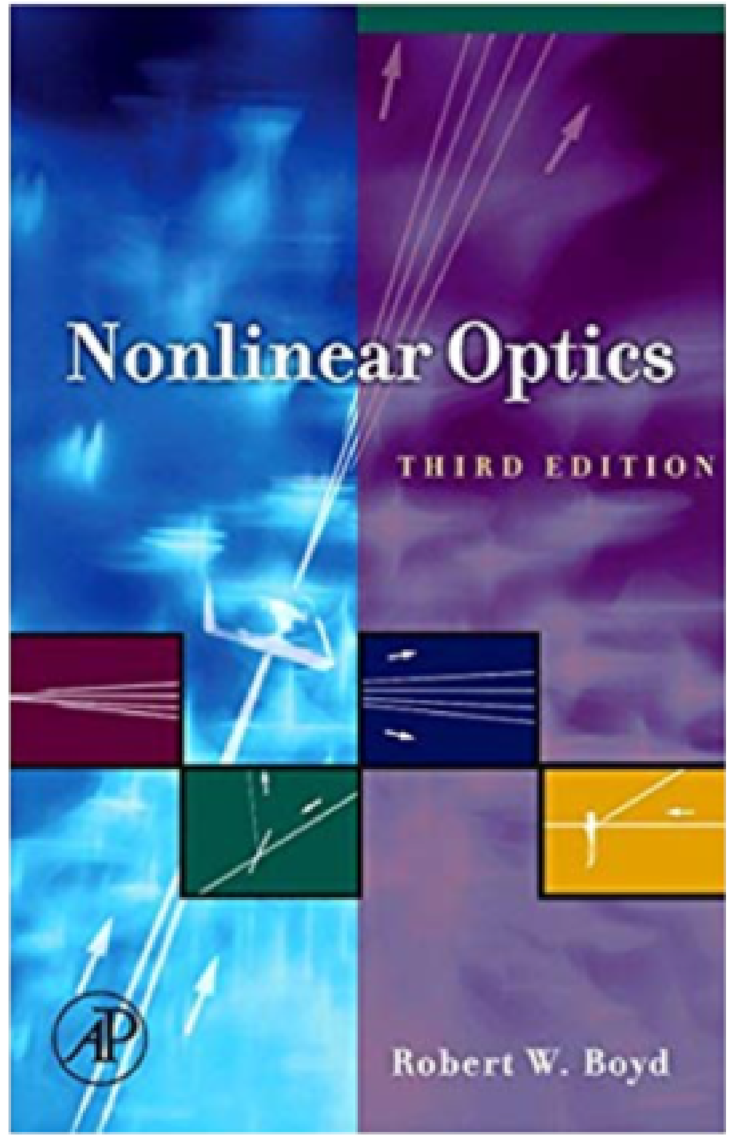
Samuel Lemieux, Enno Giese, Robert Fickler,
Maria V. Chekhova, and Robert W. Boyd

Quantum Radiometry

Two of my research interests are radiometry and nonlinear optics.
Why not combine them both into a single project?



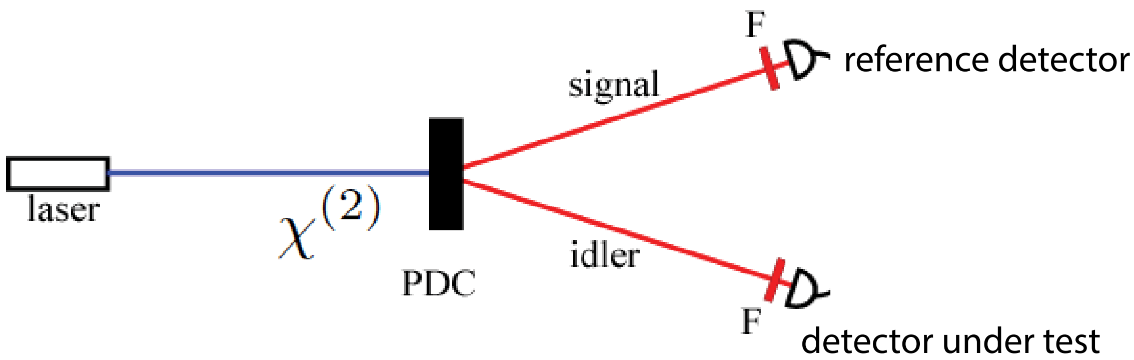
John Wiley, 1983

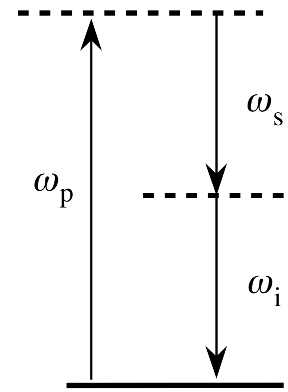


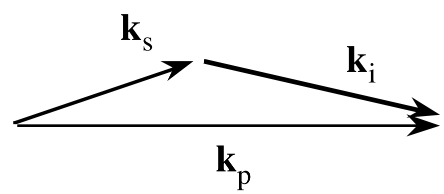
Academic Press, 2008

Earlier Work on Quantum Radiometry

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



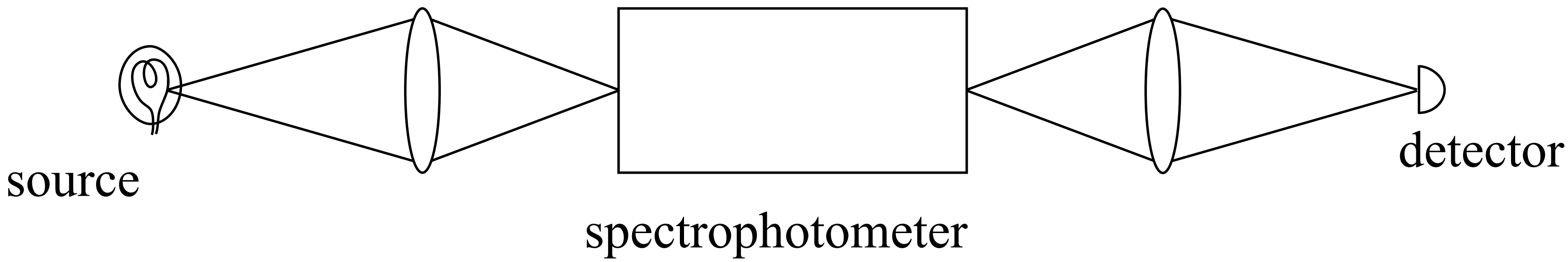


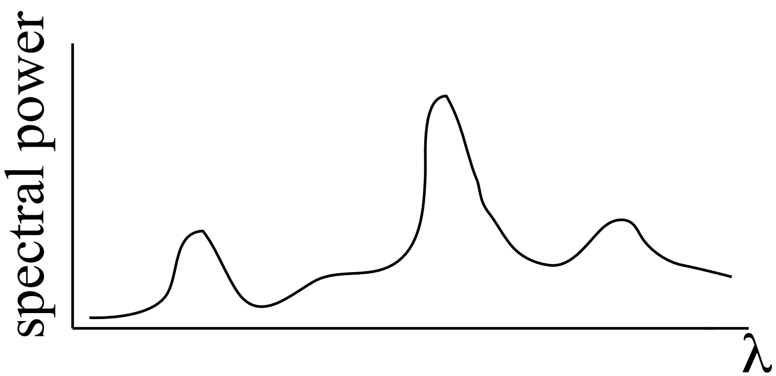


- Earlier work (Klyshko) established that the light produced by spontaneous parametric downconversion (SPDC) can be characterized in terms of the radiometric property known as brightness (or radiance).

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

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

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

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

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

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

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

$$B_\nu = \frac{(c/n)}{4\pi}u_\nu = \frac{2h\nu^3}{(c/n)^2(e^{h\nu/k_B T} - 1)}$$ 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

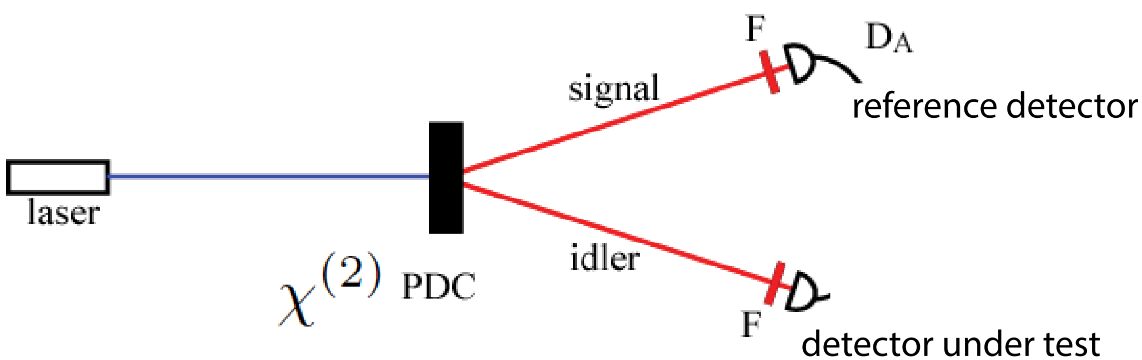
We make use of black body radiation, but at very low temperature.*

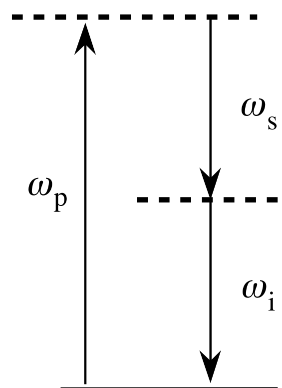
At low temperature the emission vanishes, but the field fluctuations remain.

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.

Theory of Spontaneous Parametric Down Conversion (SPDC)

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 $\hat{H} \sim E_p \chi^{(2)} \hat{E}_s^\dagger \hat{E}_i^\dagger + \text{h.c.}$
- Quantization $\hat{E}_j \sim \int \mathrm{d}^3 k_j \sqrt{\frac{\omega_j}{n_j}} \hat{a}_j \, \mathrm{e}^{i(\boldsymbol{k}_j \boldsymbol{r} - \omega_j t)}$

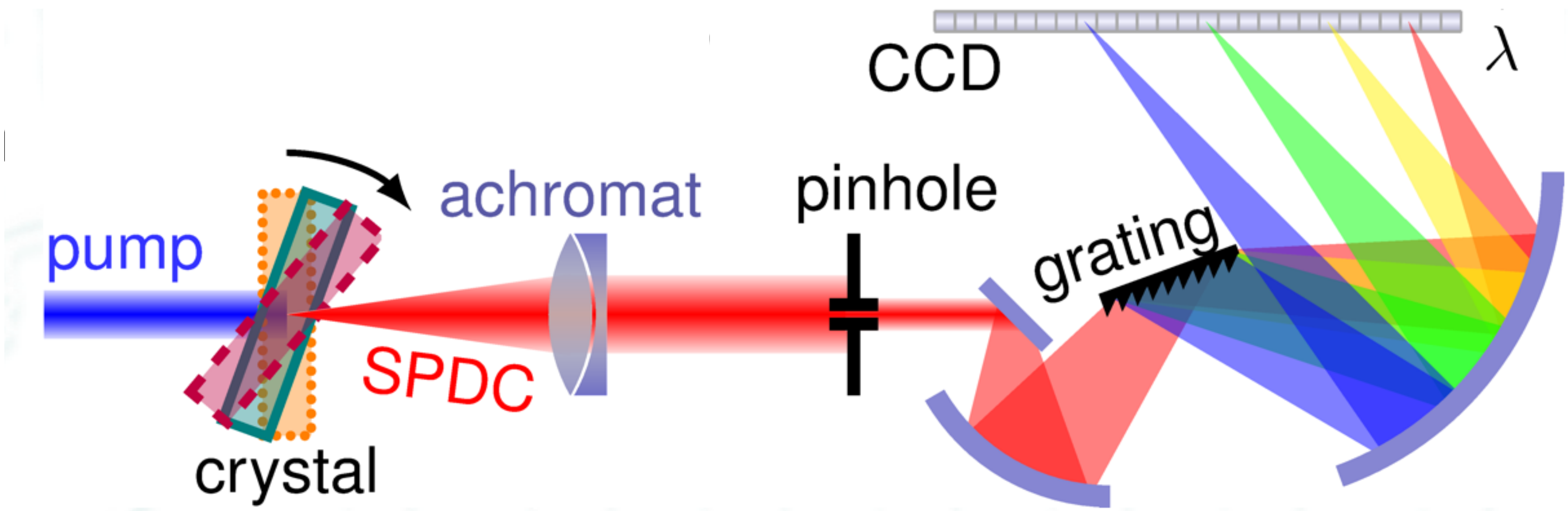
  ↑ Vacuum amplitude

  with
  $\boldsymbol{k}_j = (\boldsymbol{q}_j, \kappa_j)$

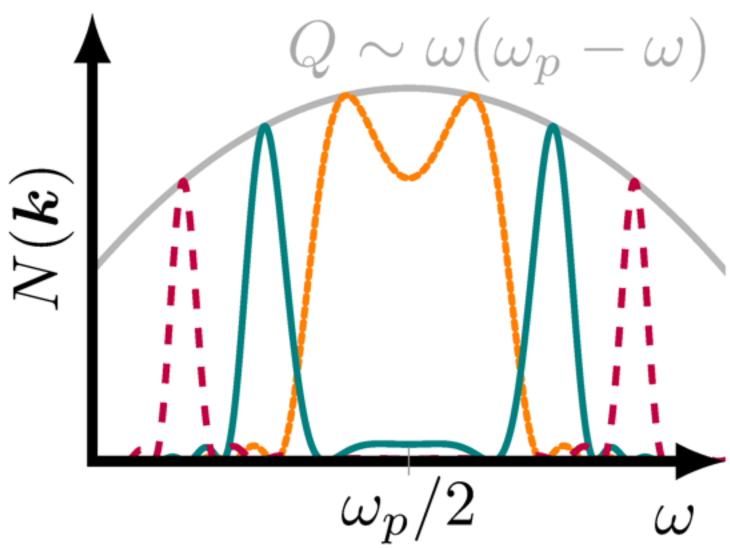
We then determine the number of downconverted signal photons per mode

$$N(\boldsymbol{k}_s) \sim \underbrace{\left(\frac{\mathcal{E}_p \chi^{(2)} L}{\sqrt{n_s n_i}}\right)^2}_{\mathcal{G}(\lambda_s)} \underbrace{\omega_s \omega_i}_{Q(\lambda_s)} \underbrace{\operatorname{sinc}^2 \frac{\Delta \kappa L}{2}}_{\mathcal{S}(\lambda_s)}$$

Our Experimental Setup



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



Note that

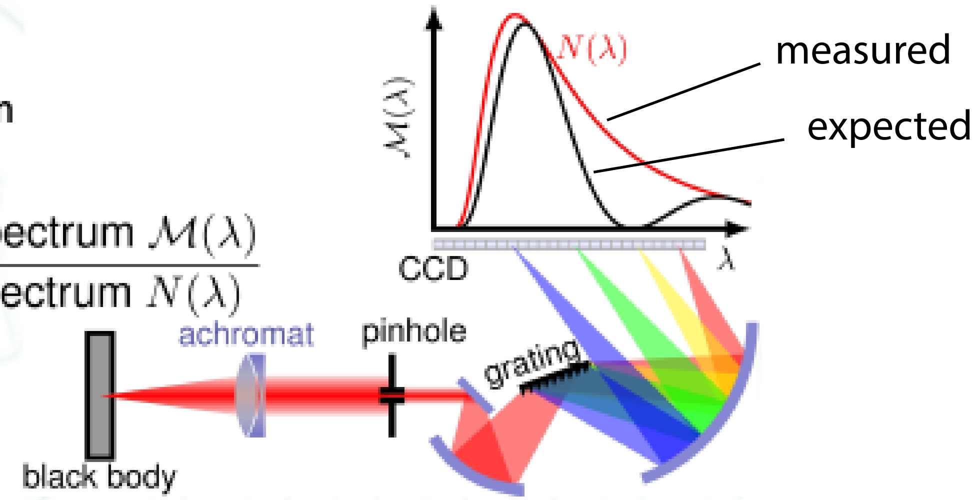
$$\omega_s \omega_i = \omega_s(\omega_p - \omega_s)$$

which give the form of the parabola shown in the figure.

Determining the Response Function

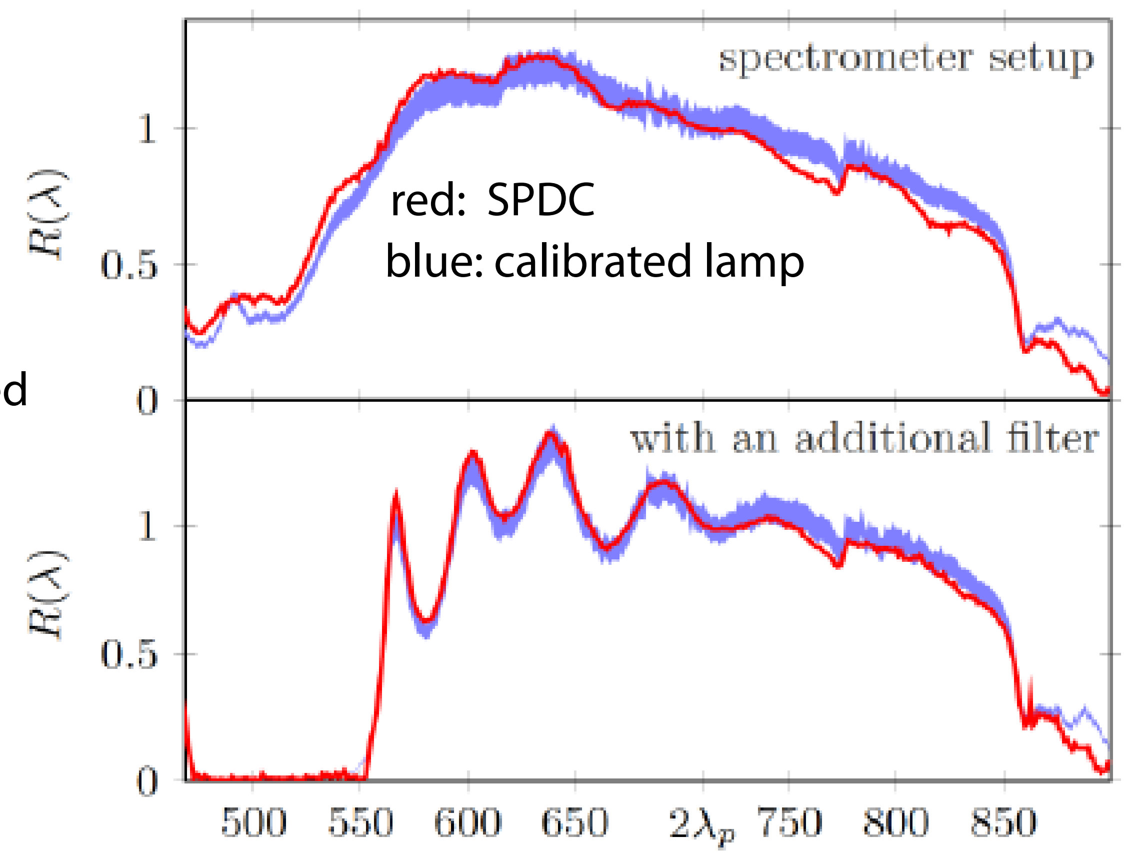
- Spectrometer might not show expected spectrum
- Response function:

$$\mathcal{R}(\lambda) = \frac{\text{measured spectrum } \mathcal{M}(\lambda)}{\text{expected spectrum } N(\lambda)}$$



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 give by

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

where we have introduced the gain parameter $\mathcal{G} = c^{-1}\, L\, \chi^{(2)} E_p/\sqrt{n\, n_i}$,

- Note that the shape of the curve of $\mathcal{N}_{\mathrm{PM}}$ versus omega changes with pump intensity. By measuring the shape of this curve, we can determine $\mathcal{G}$ 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