Quantum Imaging

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Please feel free to photograph my visuals if you wish.

Presented at the New Frontiers in Quantum Imaging Symposium, Glasgow, 25-26th of September 2017.

Boyd Name Origin

(Road outside Glasgow)

Quantum Imaging Outline

Introduction to Quantum Imaging

Examples of Quantum Imaging
- Two-color ghost imaging
- Interaction-free ghost imaging
- Imaging with photon-added states
- Imaging using weak values
- Imaging with "undetected photons"

Structured Light Fields for Quantum Information
- Dense coding of information using orbital angular momentum of light
- Secure Communication transmitting more than one bit per photon

Materials and Structures for Quantum Imaging
- Epsilon-near-zero materials
- Single-photon sources
- Chip-scale photonic devices for quantum information

Quantum Imaging

- Goal of quantum imaging is to produce "better" images using quantum methods
  - image with a smaller number of photons
  - achieve better spatial resolution
  - achieve better signal-to-noise ratio

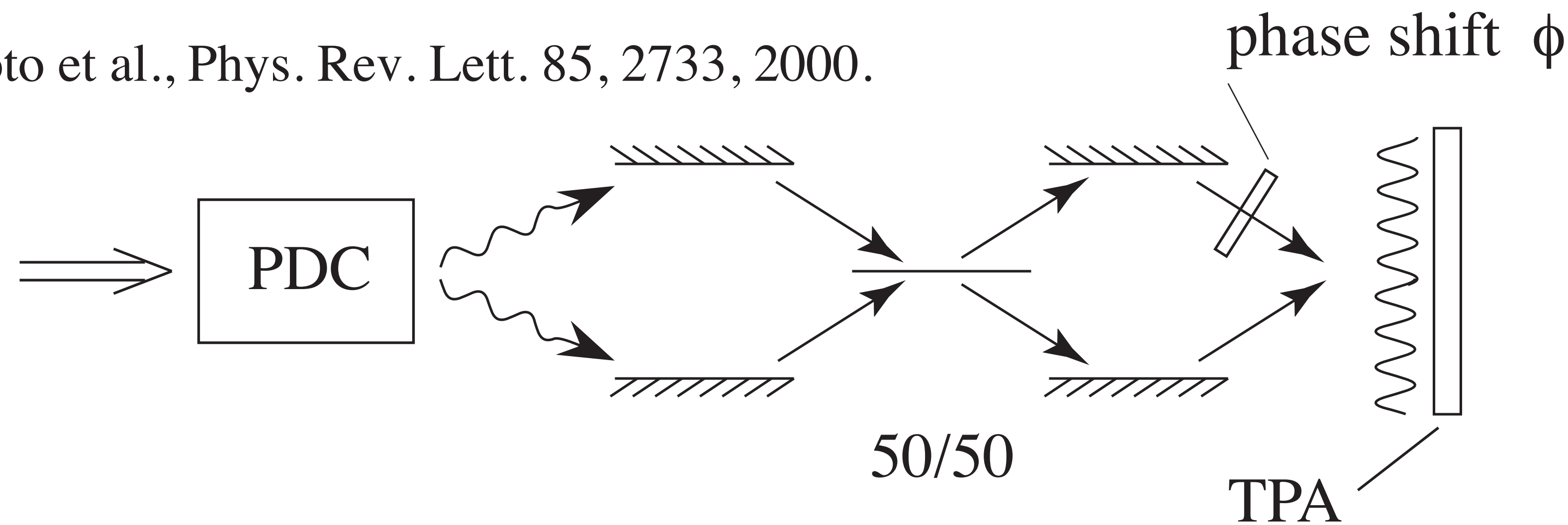
- Alternatively, quantum imaging exploits the quantum properties of the transverse structure of light fields

SHARPER IMAGE

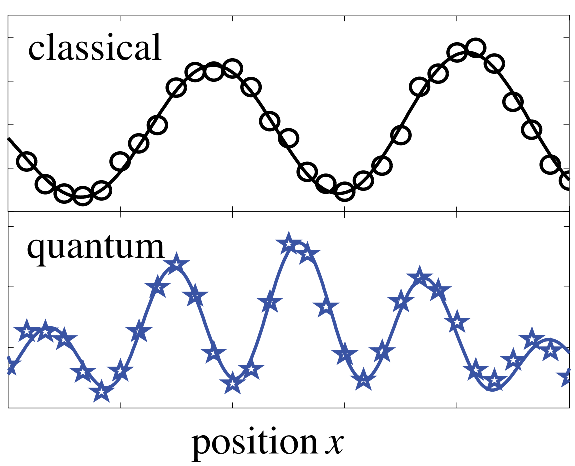
Quantum Lithography: Concept of Jonathan Dowling

- Entangled photons can be used to form an interference pattern with detail finer than the Rayleigh limit
- Resolution ≈ $\lambda/2N$, where N = number of entangled photons

Boto et al., Phys. Rev. Lett. 85, 2733, 2000.



- No practical implementation to date, but some laboratory results

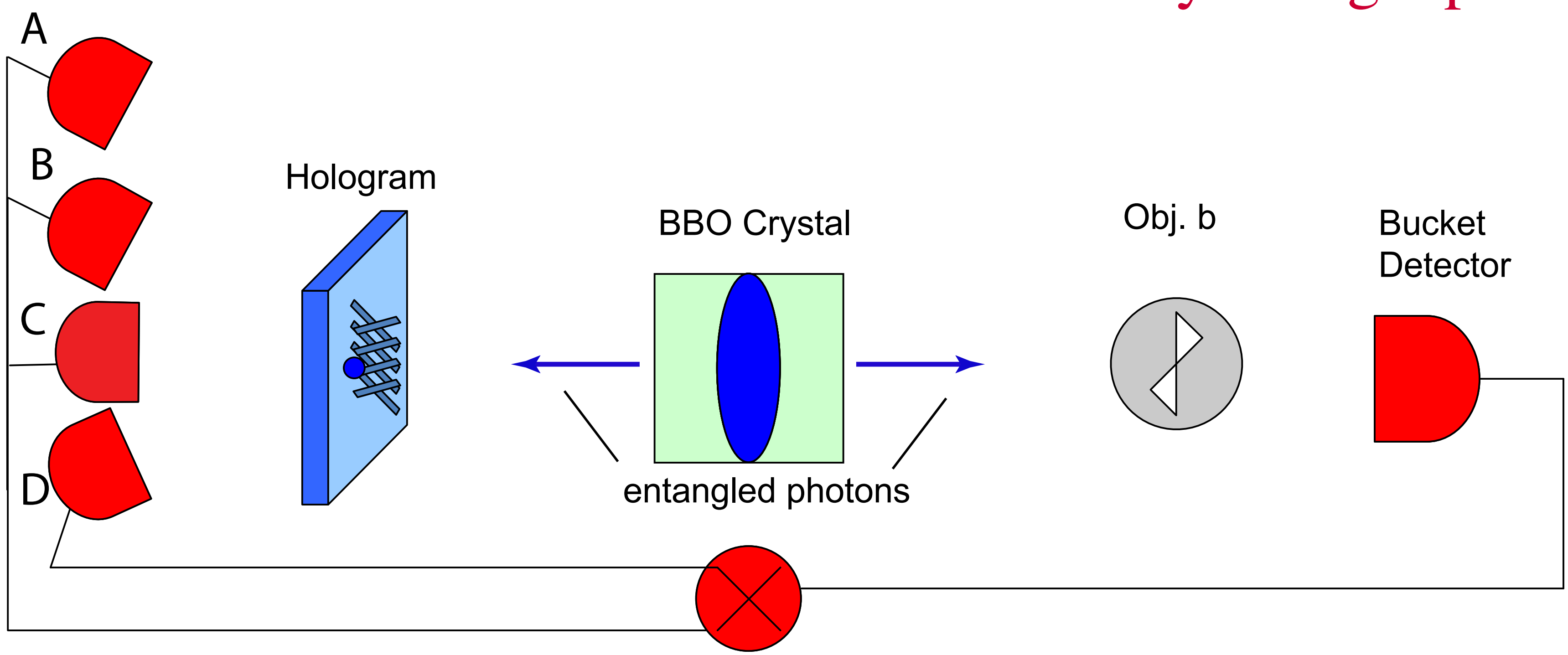


Quantum spatial superresolution by optical centroid measurements, Shin, Chan, Chang, and Boyd, Phys. Rev. Lett. 107, 083603 (2011).

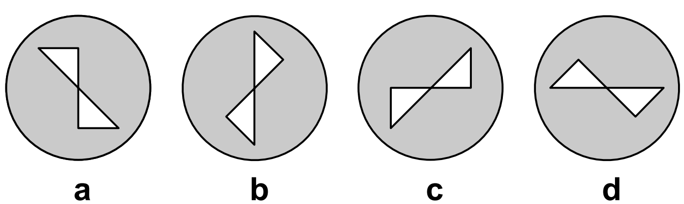
See also, Quantum Lithography: Status of the Field, R.W. Boyd and J.P. Dowling, Quantum Information Processing, 11:891–901 (2012).

Single-Photon Coincidence Imaging

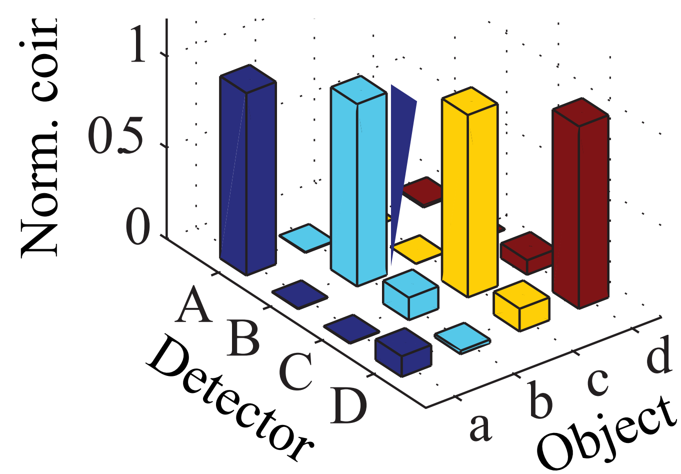
How much information can be carried by a single photon?



We discriminate among four orthogonal images using single-photon interrogation in a coincidence imaging configuration.



coincidence count rate



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

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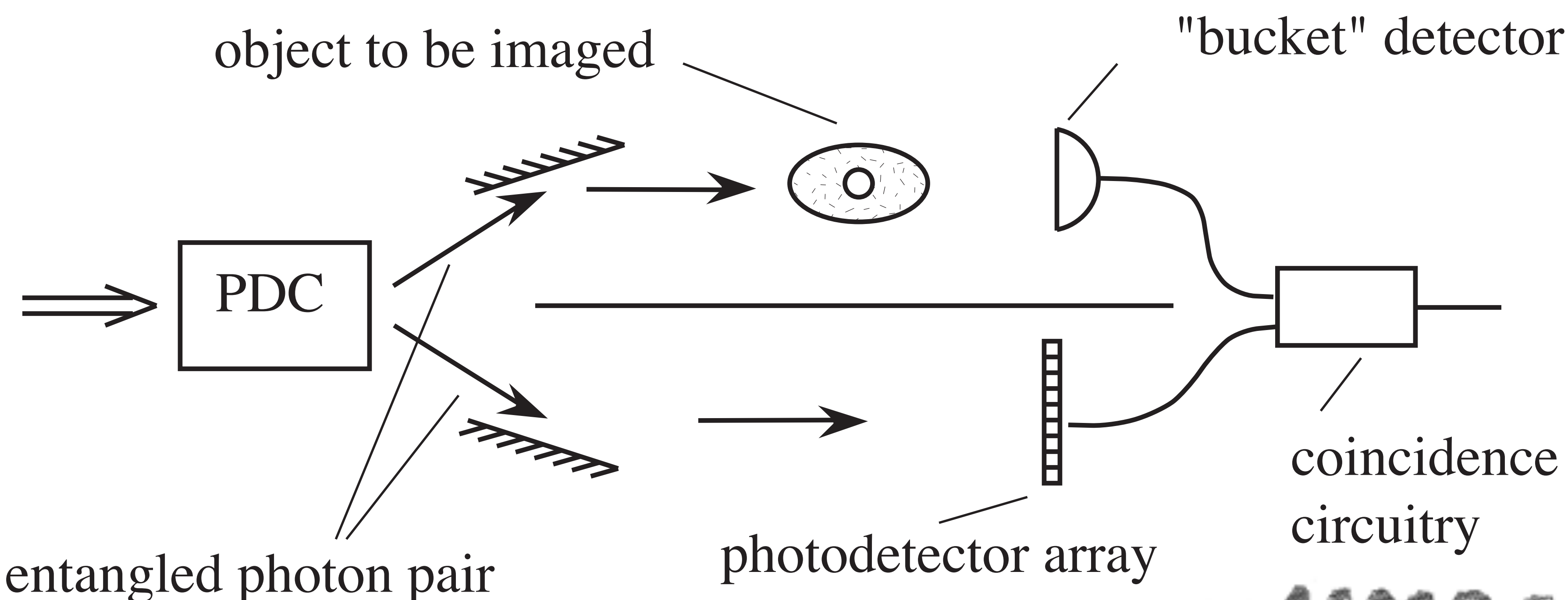
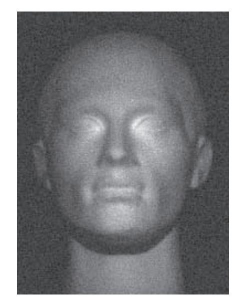
Ghost (Coincidence) Imaging



- Obvious applicability to remote sensing!
  (imaging under adverse situations, bio, two-color, etc.)
- Is this a purely quantum mechanical process? (No)
- Can Brown-Twiss intensity correlations lead to ghost imaging? (Yes)



Padgett Group

Strekalov et al., Phys. Rev. Lett. 74, 3600 (1995).
Pittman et al., Phys. Rev. A 52 R3429 (1995).
Abouraddy et al., Phys. Rev. Lett. 87, 123602 (2001).
Bennink, Bentley, and Boyd, Phys. Rev. Lett. 89 113601 (2002).
Bennink, Bentley, Boyd, and Howell, PRL 92 033601 (2004)
Gatti, Brambilla, and Lugiato, PRL 90 133603 (2003)
Gatti, Brambilla, Bache, and Lugiato, PRL 93 093602 (2003)

Is Ghost Imaging a Quantum Phenomenon?

VOLUME 90, NUMBER 13 PHYSICAL REVIEW LETTERS week ending 4 APRIL 2003

Entangled Imaging and Wave-Particle Duality: From the Microscopic to the Macroscopic Realm

A. Gatti, E. Brambilla, and L. A. Lugiato

INFM, Dipartimento di Scienze CC.FF.MM., Università dellíInsubria, Via Valleggio 11, 22100 Como, Italy

(Received 11 October 2002; published 3 April 2003)

We formulate a theory for entangled imaging, which includes also the case of a large number of photons in the two entangled beams. We show that the results for imaging and for the wave-particle duality features, which have been demonstrated in the microscopic case, persist in the macroscopic domain. We show that the quantum character of the imaging phenomena is guaranteed by the simultaneous spatial entanglement in the near and in the far field.

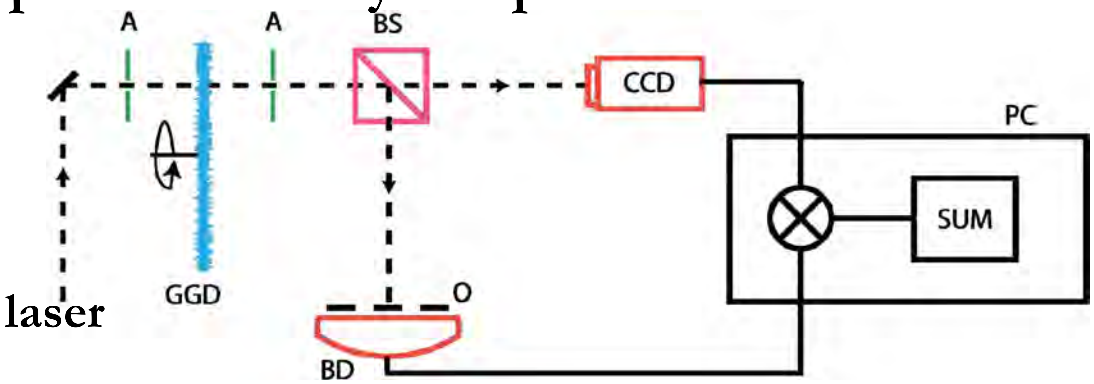
Experimental verification by Bennink, Bentley, Boyd, and Howell, Phys. Rev. Lett., 92, 033601, 2004.

Thermal Ghost Imaging

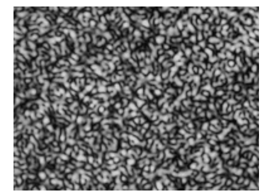
Instead of using entangled photons, one can perform ghost imaging using the (HBT) correlations of thermal (or quasithermal) light.

(Gatti et al., Phys. Rev. Lett. 93, 093602, 2004).

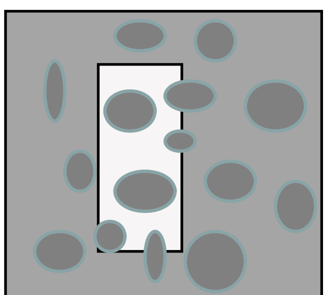
- **Typical laboratory setup**



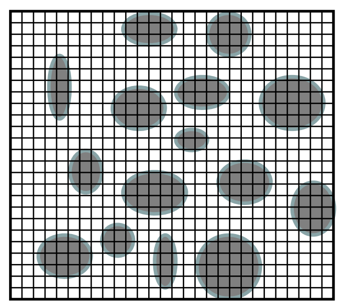
identical speckle patterns in each arm



- **How does this work? (Consider the image of a slit.)**

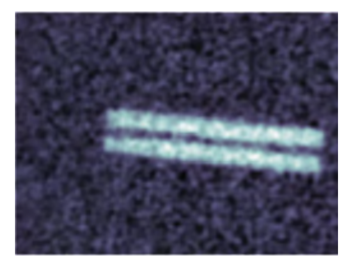


Object arm, bucket detector



Reference arm, CCD

Example ghost image



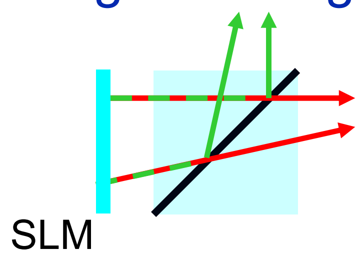
Zerom et al., A 86, 063817 (2012)

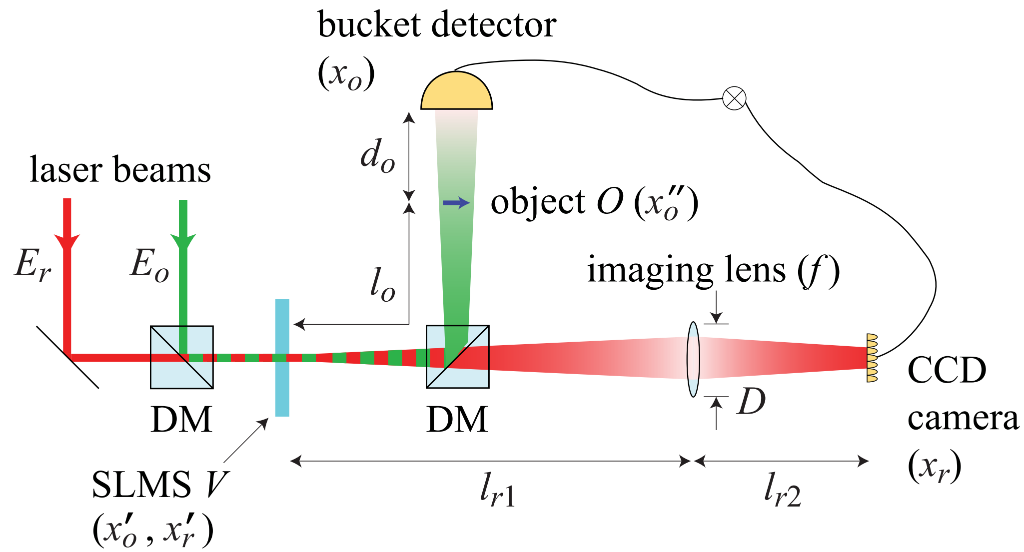
Calculate (total transmitted power) x (intensity at each pixel) and average over many speckle patterns.

Two-Color Ghost Imaging

New possibilities afforded by using different colors in object and reference arms

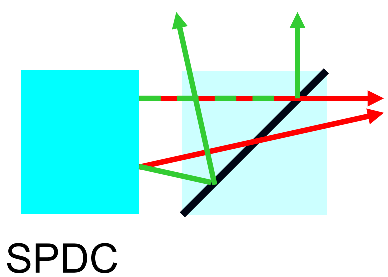
Thermal ghost imaging

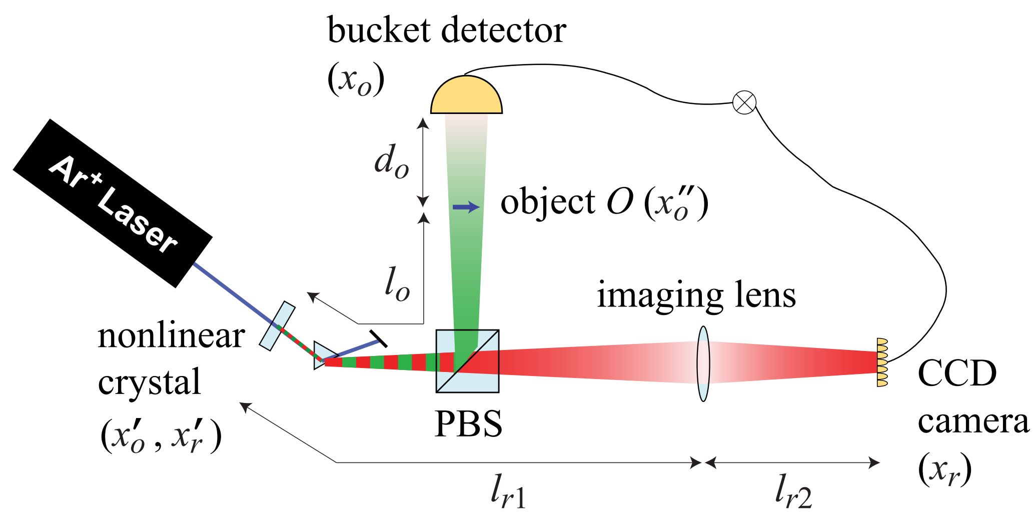




But no obvious way to make identical speckle patterns at two wavelengths

Quantum ghost imaging





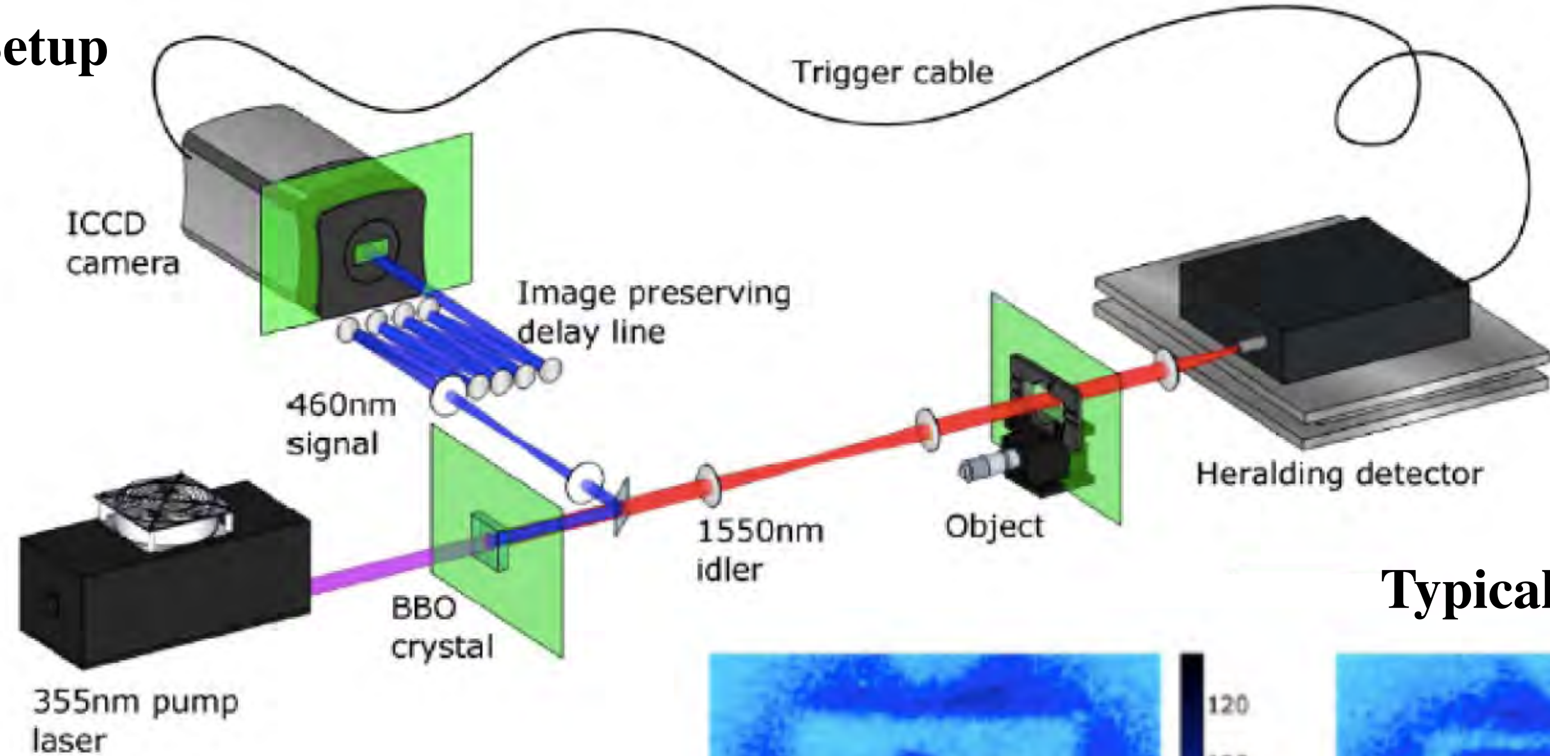
Spatial resolution depends on wavelength used to illuminate object.

Two-Color Ghost Imaging, K.W.C. Chan, M.N. O'Sullivan, and R.W. Boyd, Phys. Rev. A 79, 033808 (2009).

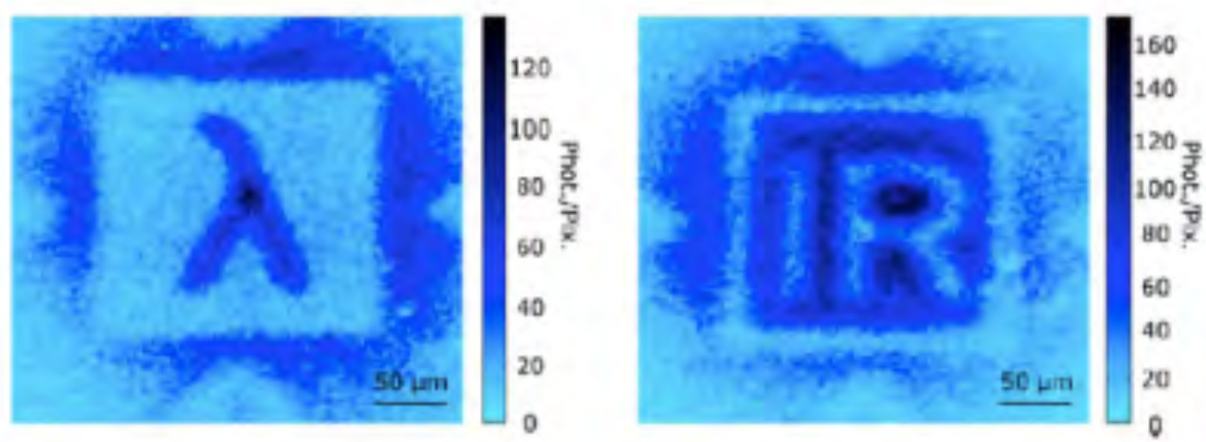
Wavelength-Shifted (Two-Color) Ghost Microscopy

- Pump at 355 nm produces signal at 460 nm and idler at 1550 nm
- Object is illuminated at 1550 nm, but image is formed (in coincidence) at 460 nm
- Wavelength ratio of 3.4 is the largest yet reported.

Setup



Typical images



Photon-sparse microscopy: visible light imaging using infrared illumination, R.S. Aspden, N. R. Gemmell, P.A. Morris, D.S. Tasca, L. Mertens, M.G. Tanner, R. A. Kirkwood, A. Ruggeri, A. Tosi, R. W. Boyd, G.S. Buller, R.H. Hadfield, and M.J. Padgett, Optica 2, 1049 (2015).

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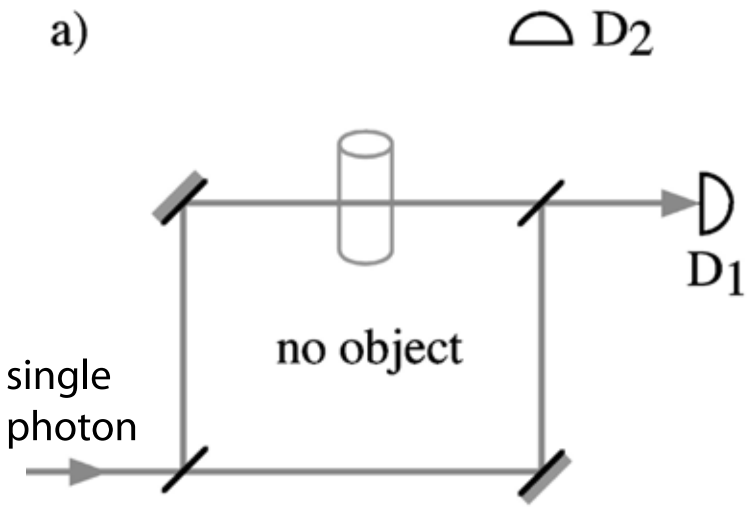
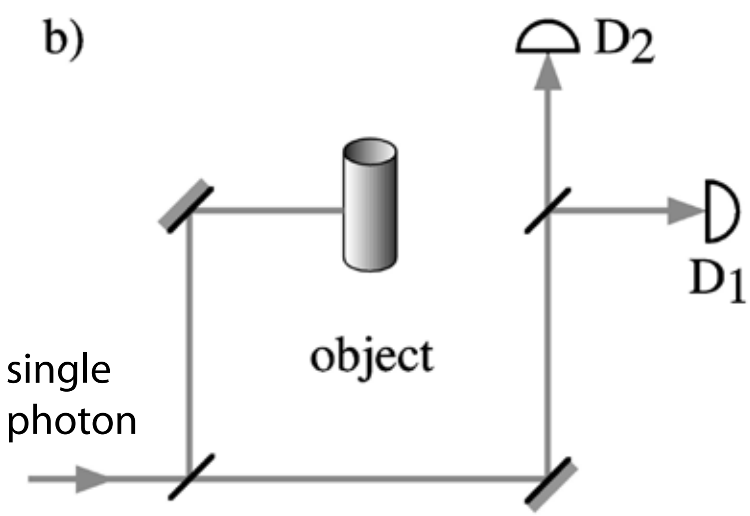
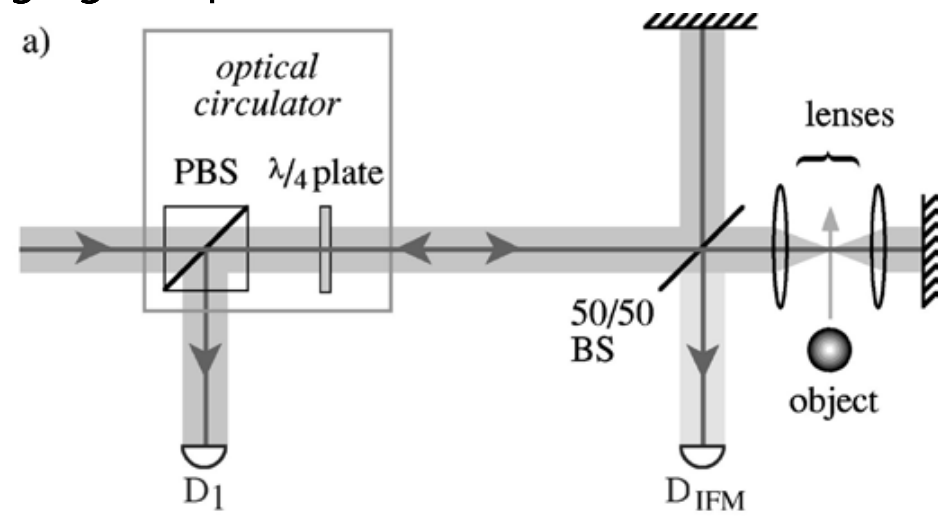
Quantum Imaging by Interaction-Free Measurement

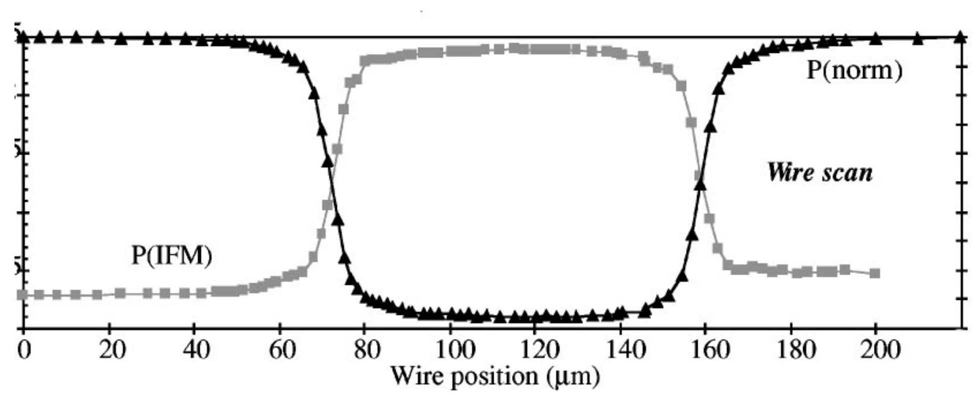




imaging setup

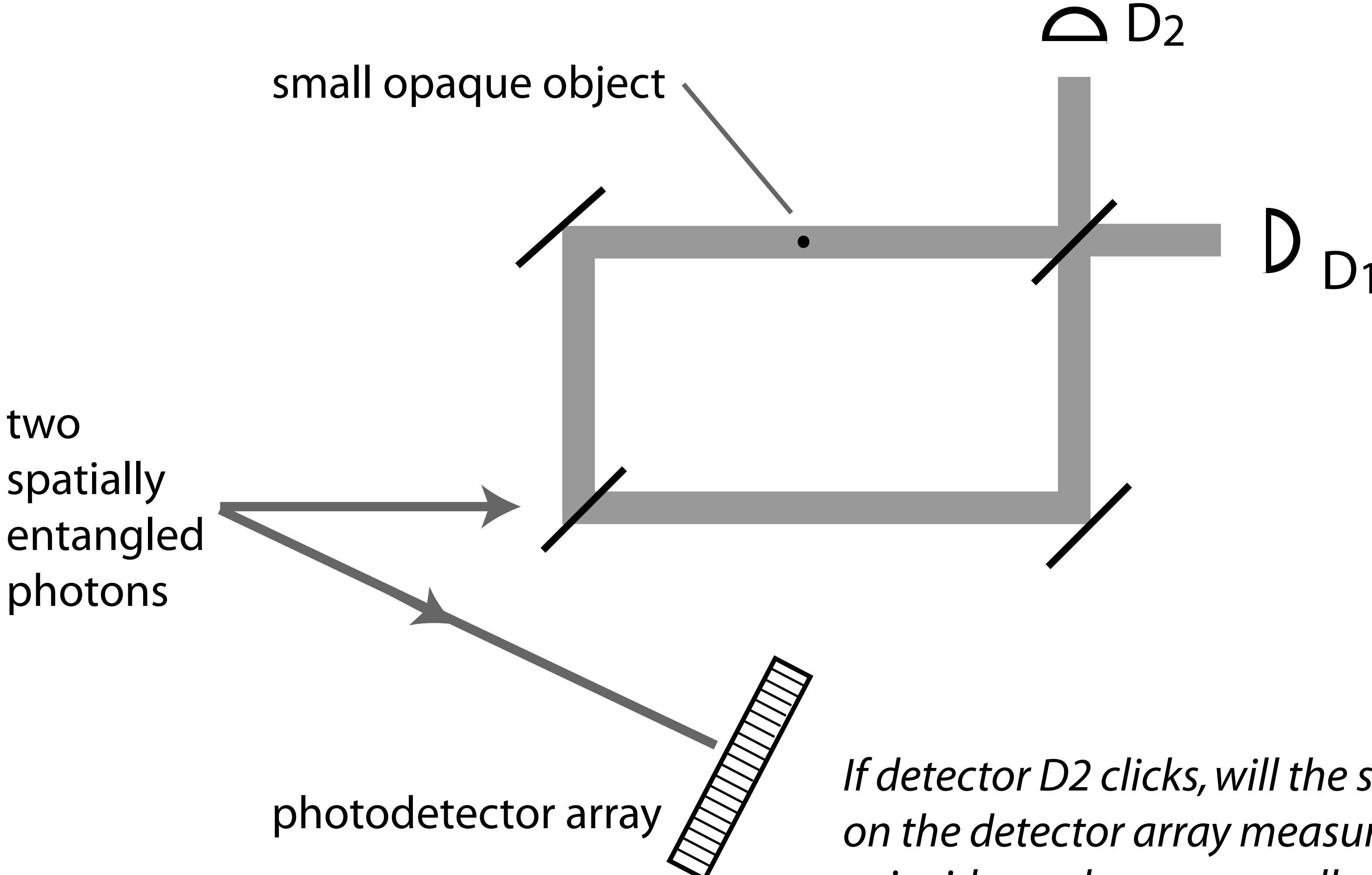


results



M. Renninger, Z. Phys. 15S, 417 (1960).
R. H. Dicke, Am. J. Phys. 49, 925 (1981).
A. Elitzur and L. Vaidman, Found. Phys. 23, 987 (1993).
L. Vaidman, Quant. Opt. 6, 119 (1994).
P. Kwiat, H. Weinfurter, T. Herzog, A. Zeilinger, and M. A. Kasevich, Phys. Rev. Lett. 74, 4763 (1995)
A. G. White, J. R. Mitchell, O. Nairz, and P. G. Kwiat, Phys. Rev. A 58, 605 (1998).

Interaction-Free Measurements and Entangled Photons



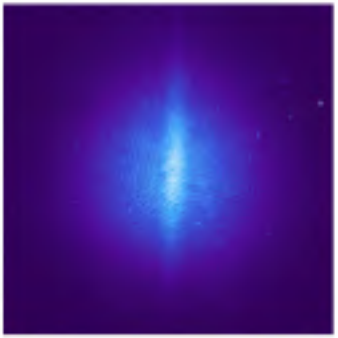
If detector D2 clicks, will the spot size on the detector array measured in coincidence become smaller?

- Does an interaction-free measurement constitute a "real" measurement?
- Does it lead to the collapse of the wavefunction of its entangled partner?
- More precisely, does the entire two-photon wavefunction collapse?

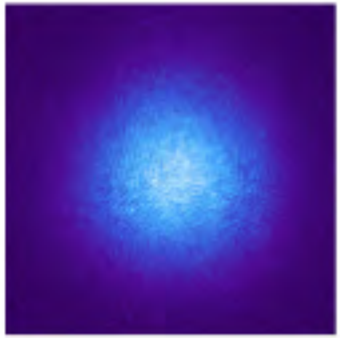
Experimental Results

Interaction-free ghost image of a straight wire

coincidence counts



singles counts



- Note that the interaction-free ghost image is about five times narrower than full spot size on the ICCD camera

- This result shows that interaction-free measurements lead to wavefunction collapse, just like standard measurements.

With Frédéric Bouchard, Harjaspreet Mand, and Ebrahim Karimi,

Is interaction-free imaging useful?

Interaction-free imaging allows us to see what something looks like *in the dark*!

Could be extremely useful for biophysics. What does the retina look like when light does not hit it?

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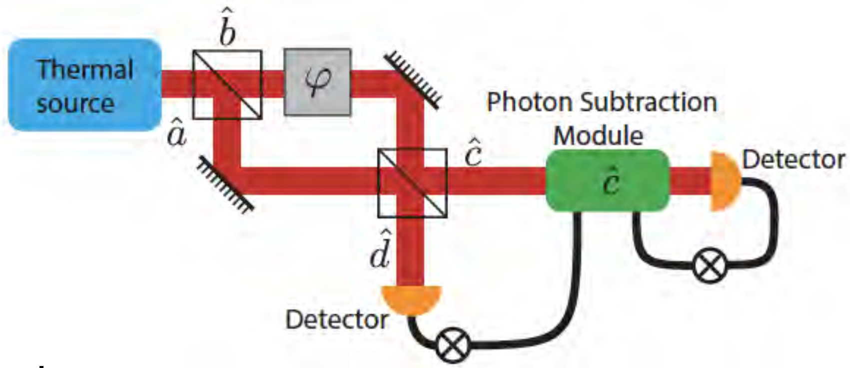
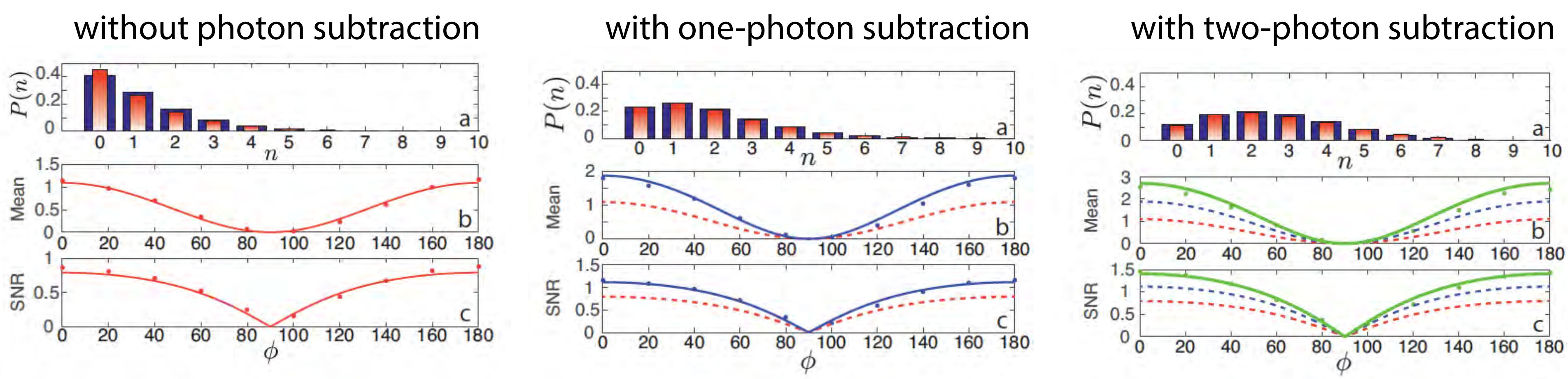
Enhanced Interferometry with Photon-Subtracted Thermal Light



Can we measure the phase ϕ more accurately by using photon-subtracted states?

- Results



- We find that the signal-to-noise ratio (SNR) is increased through use of photon-subtracted states!
- However, in the present setup, photon-subtraction occurs probabilistically and only a small fraction of the time
- Is there a means to obtain photon-addition and photon-subtraction deterministically?
- Can we use this method to perform quantum imaging with improved SNR?

Rochester, Boeing, LSU, Lehman collaboration

Photon-Added and Photon-Subtracted States

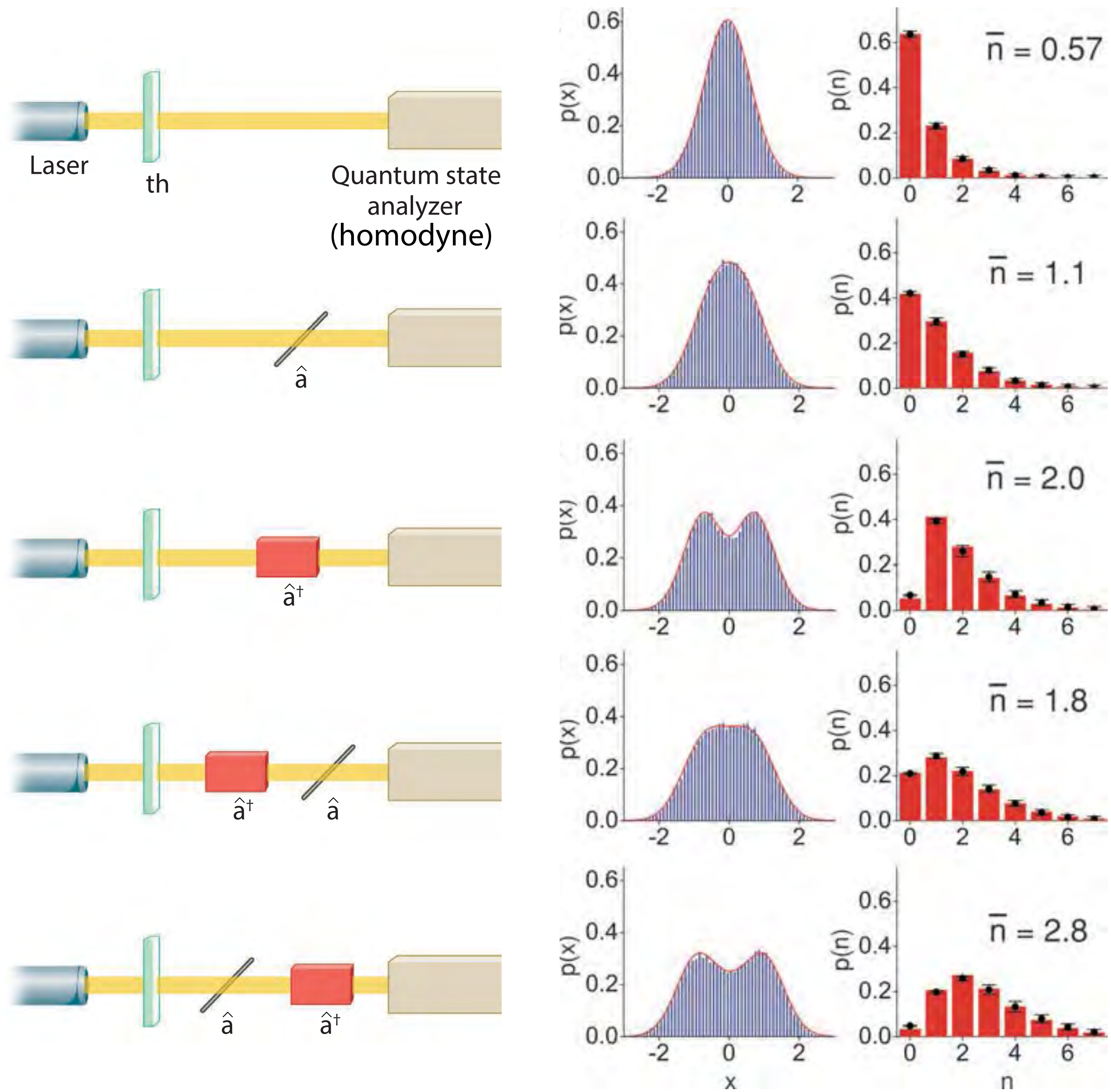
original thermal state

photon-subtracted

photon-added

photon-added and then subtracted

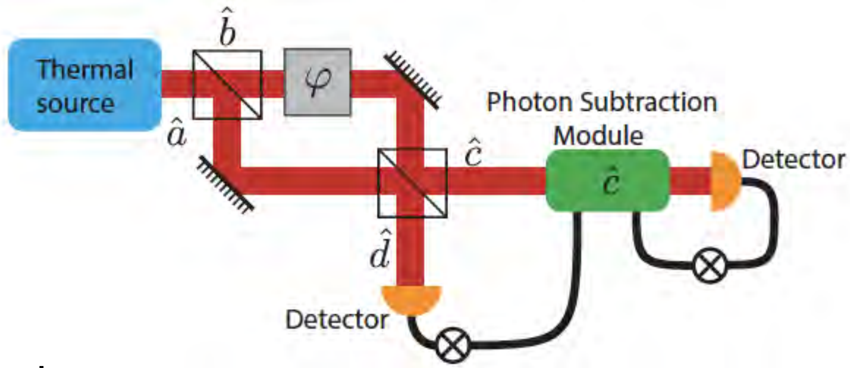
photon-subtracted and then added



Note!

V. Parigi, A. Zavatta, M. Kim, M. Bellini, Science 317,1890 (2007).

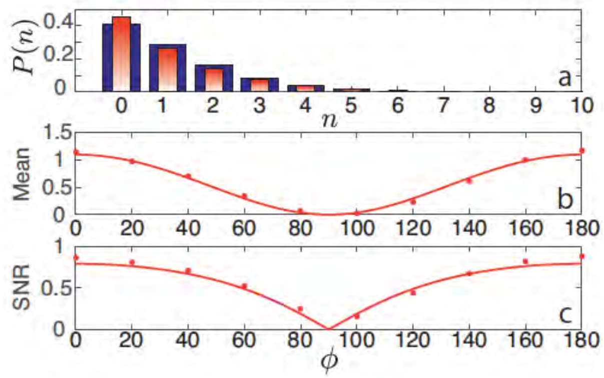
Enhanced Interferometry with Photon-Subtracted Thermal Light



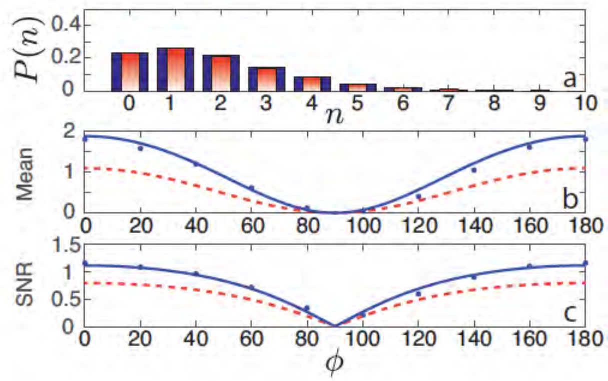
Can we measure the phase ϕ more accurately by using photon-subtracted states?

- Results

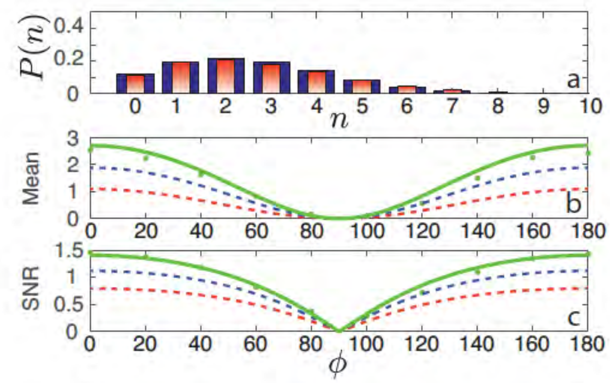
without photon subtraction



with one-photon subtraction



with two-photon subtraction



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How the Result of a Measurement of a Component of the Spin of a Spin-$\frac{1}{2}$ Particle Can Turn Out to be 100

Yakir Aharonov, David Z. Albert, and Lev Vaidman
Physics Department, University of South Carolina, Columbia, South Carolina 29208, and School of Physics and Astronomy, Tel-Aviv University, Ramat Aviv 69978, Israel
(Received 30 June 1987)

We have found that the usual measuring procedure for preselected and postselected ensembles of quantum systems gives unusual results. Under some natural conditions of weakness of the measurement, its result consistently defines a new kind of value for a quantum variable, which we call the weak value. A description of the measurement of the weak value of a component of a spin for an ensemble of preselected and postselected spin-$\frac{1}{2}$ particles is presented.

PACS numbers: 03.65.Bz

standard expectation value: $< A >=< \Psi|\hat{A}|\Psi >$

weak value: $A_w \equiv \langle \psi_f \mid A \mid \psi_{\text{in}} \rangle / \langle \psi_f \mid \psi_{\text{in}} \rangle.$

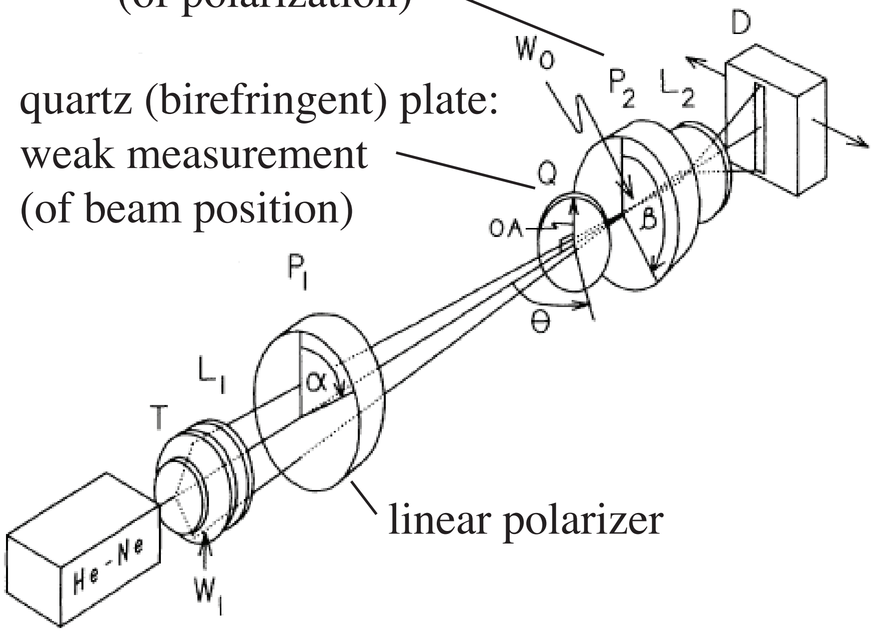
Why are weak values important?
- can lead to amplification of small signals
- can lead to direct measurement of the quantum wavefunction

Realization of a Measurement of a "Weak Value"

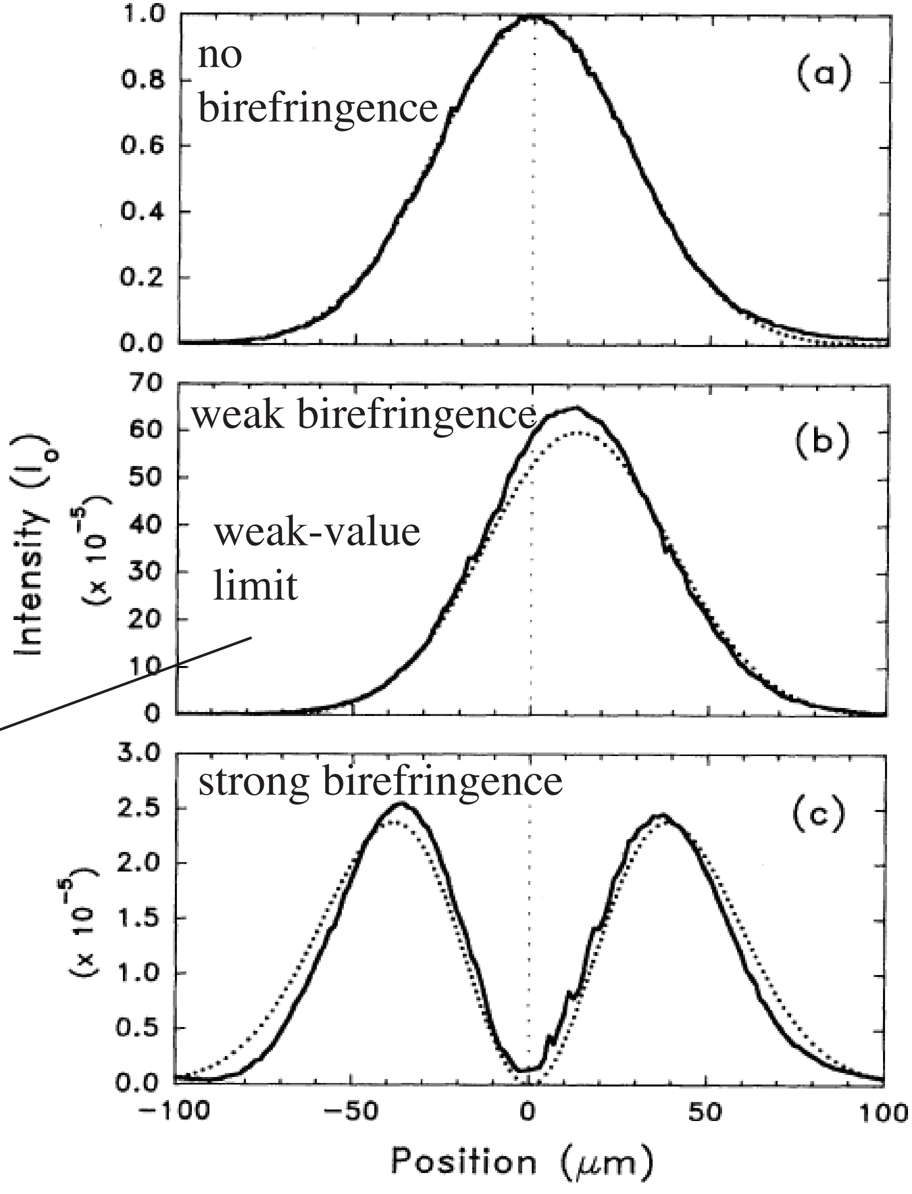
N. W. M. Ritchie, J. G. Story, and Randall G. Hulet
Department of Physics and Rice Quantum Institute, Rice University, Houston, Texas 77251-1892
(Received 7 December 1990)

linear polarizer:
strong measurement
(of polarization)

quartz (birefringent) plate:
weak measurement
(of beam position)



linear polarizer



Birefringence separates polarized beams by 0.64 μm, but gaussian in (b) is displaced by 12 μm.

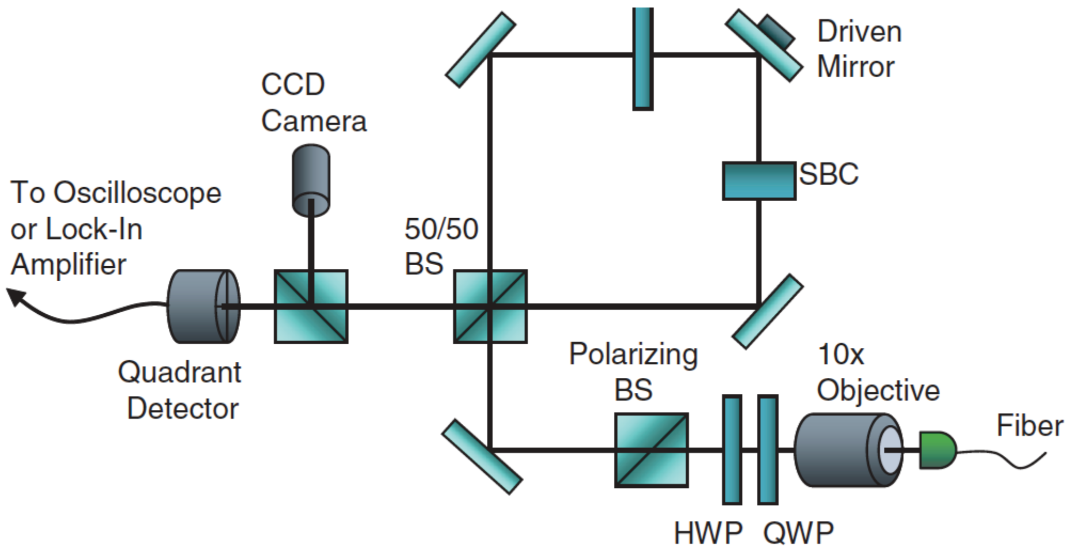
PRL 1991

Selected for a Viewpoint in *Physics*
PRL **102,** 173601 (2009) PHYSICAL REVIEW LETTERS week ending 1 MAY 2009

Ultrasensitive Beam Deflection Measurement via Interferometric Weak Value Amplification

P. Ben Dixon, David J. Starling, Andrew N. Jordan, and John C. Howell
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627, USA
(Received 12 January 2009; published 27 April 2009)

We report on the use of an interferometric weak value technique to amplify very small transverse deflections of an optical beam. By entangling the beam's transverse degrees of freedom with the which-path states of a Sagnac interferometer, it is possible to realize an optical amplifier for polarization independent deflections. The theory for the interferometric weak value amplification method is presented along with the experimental results, which are in good agreement. Of particular interest, we measured the angular deflection of a mirror down to 400 ± 200 frad and the linear travel of a piezo actuator down to 14 ± 7 fm.



LETTER

doi:10.1038/nature10120

Direct measurement of the quantum wavefunction

Jeff S. Lundeen[1], Brandon Sutherland[1], Aabid Patel[1], Corey Stewart[1] & Charles Bamber[1]

$$\langle A\rangle_W = \frac{\langle c|A|\Psi\rangle}{\langle c|\Psi\rangle}$$

Returning to our example of a single particle, consider the weak measurement of position ($A = \pi_x \equiv |x\rangle\langle x|$) followed by a strong measurement of momentum giving $P = p$. In this case, the weak value is:

$$\langle \pi_x\rangle_W = \frac{\langle p|x\rangle\ \langle x|\Psi\rangle}{\langle p|\Psi\rangle} \qquad (2)$$

$$= \frac{e^{ipx/\hbar}\Psi(x)}{\Phi(p)} \qquad (3)$$

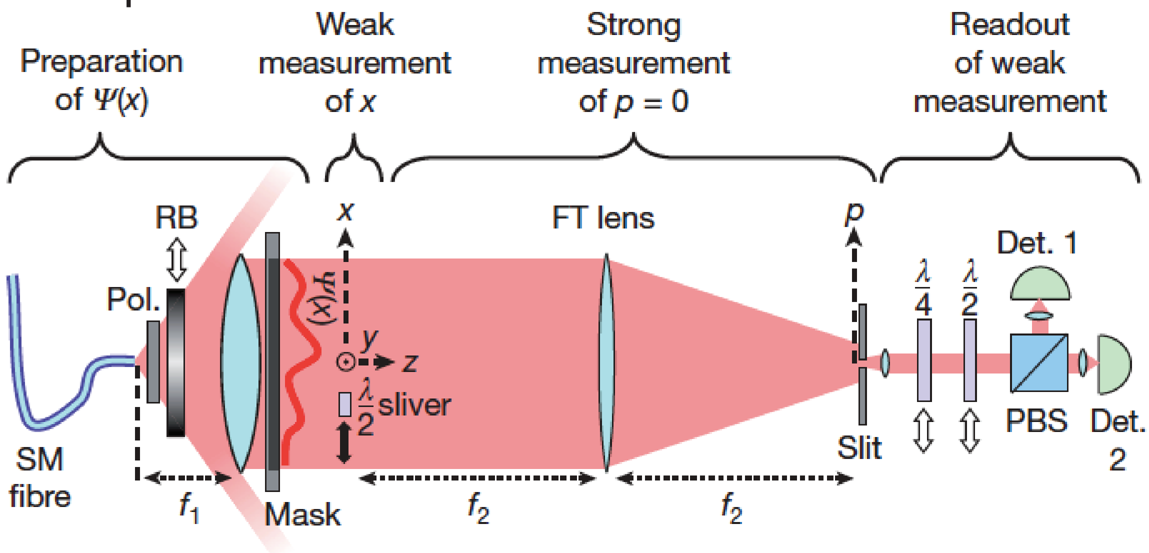
In the case $p = 0$, this simplifies to

$$\langle \pi_x\rangle_W = k\Psi(x) \qquad (4)$$

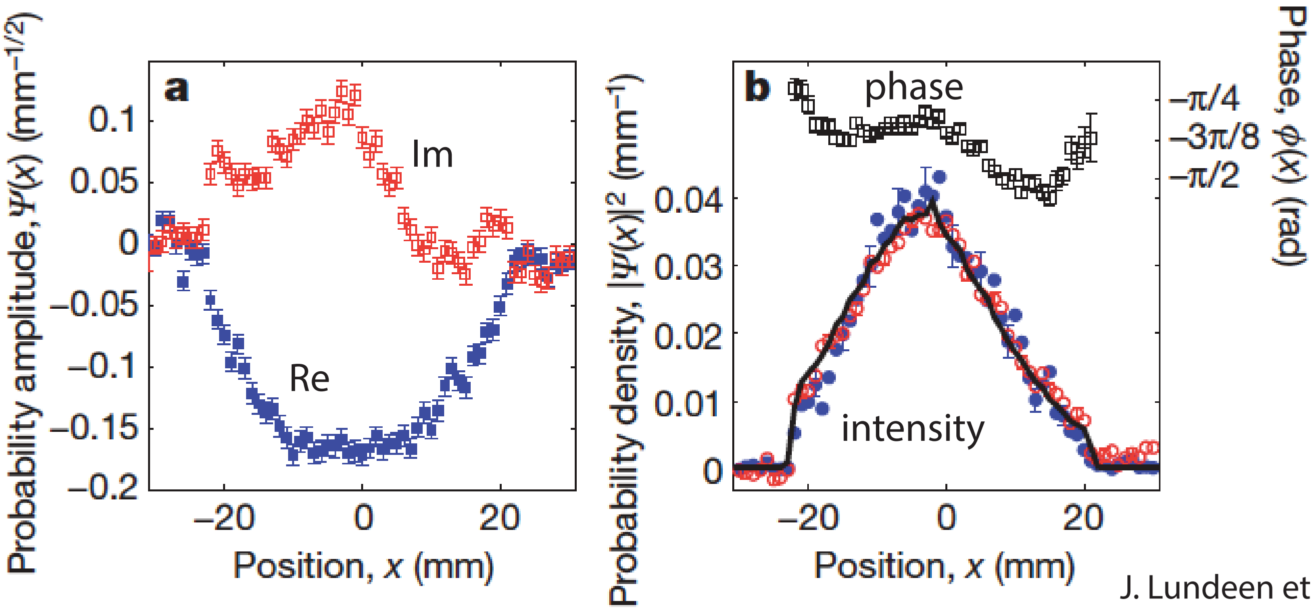
where $k = 1/\Phi(0)$ is a constant (which can be eliminated later by normalizing the wavefunction). The average result of the weak mea-

Direct Measurement of the Photon "Wavefunction"

Measurement setup



Typical results



J. Lundeen et al., Nature 474, 188 (2011)

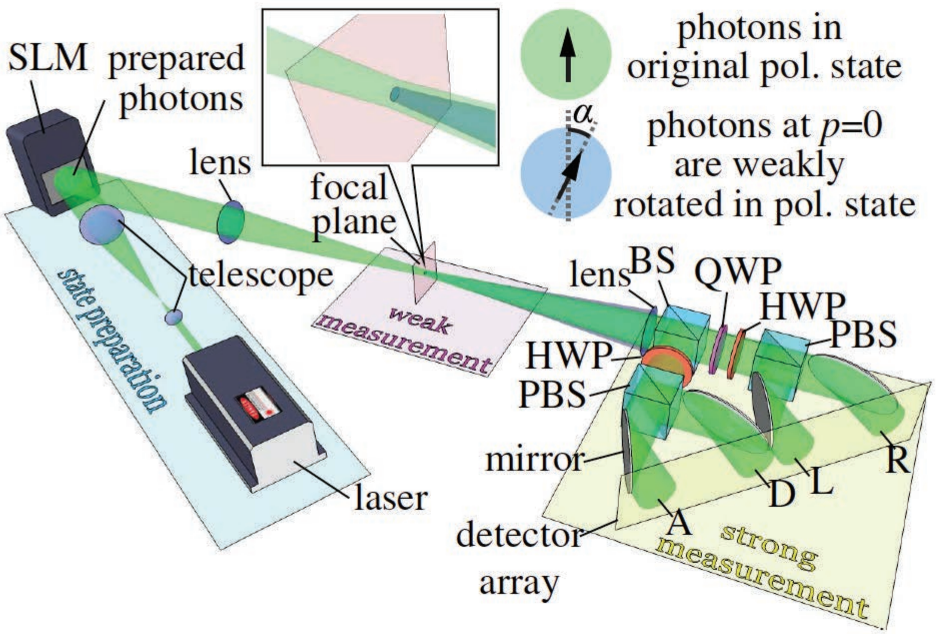
Research Article Vol. 2, No. 4 / April 2015 / Optica 388

optica

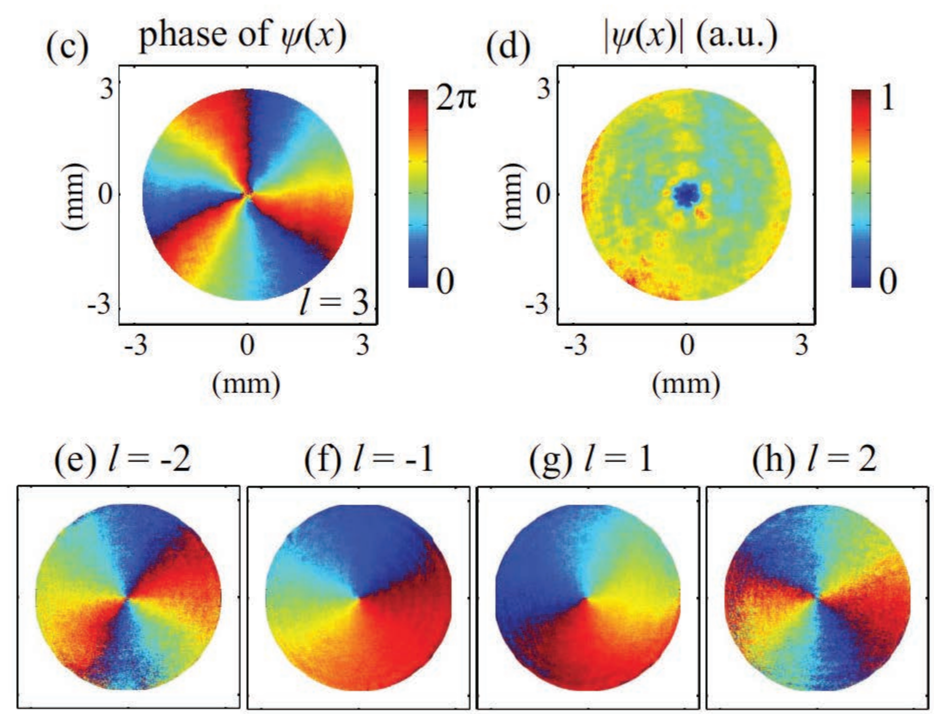
Scan-free direct measurement of an extremely high-dimensional photonic state

ZHIMIN SHI,[1,*] MOHAMMAD MIRHOSSEINI,[2] JESSICA MARGIEWICZ,[1] MEHUL MALIK,[2,3] FREIDA RIVERA,[1] ZIYI ZHU,[1] AND ROBERT W. BOYD[2,4]

Laboratory setup



Laboratory results for OAM beams



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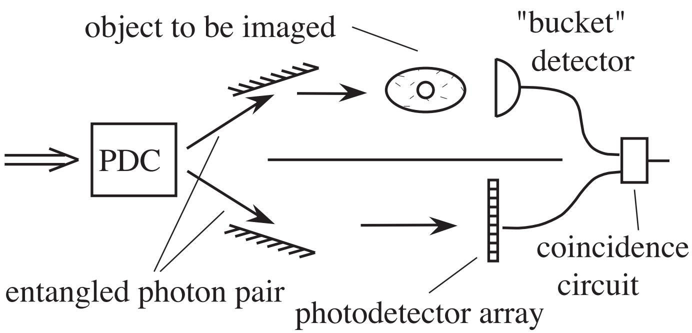
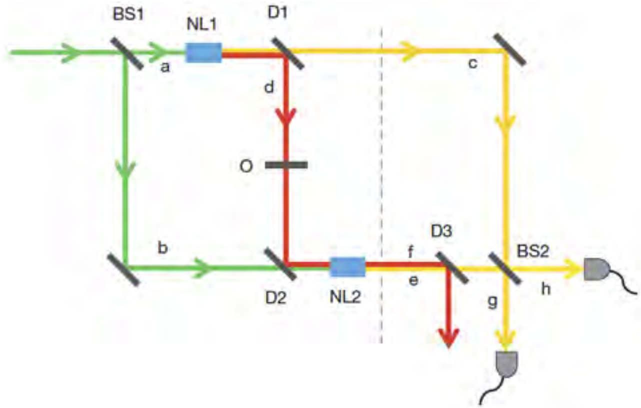
- Epsilon-near-zero materials
- Single-photon sources
- Chip-scale photonic devices for quantum information

Quantum Imaging Overview

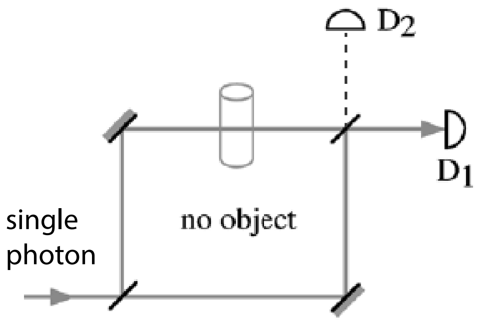
Ghost Imaging (Shih)



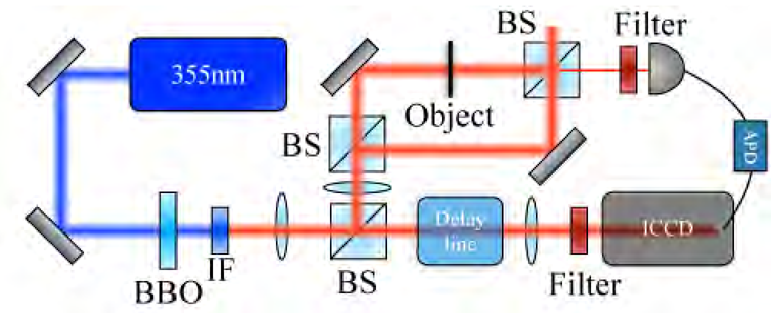
Imaging with Undetected Photons (Zeilinger)



Interaction-Free Imaging (White)



Interaction-Free Ghost Imaging (this talk)



Do We Study Quantum Imaging or Quantum Imogene?

Sid Caesar and
Imogene Coca in 1953

Quantum Imaging Outline

Introduction to Quantum Imaging

Examples of Quantum Imaging
- Two-color ghost imaging
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- Imaging using weak values
- Imaging with “undetected photons”

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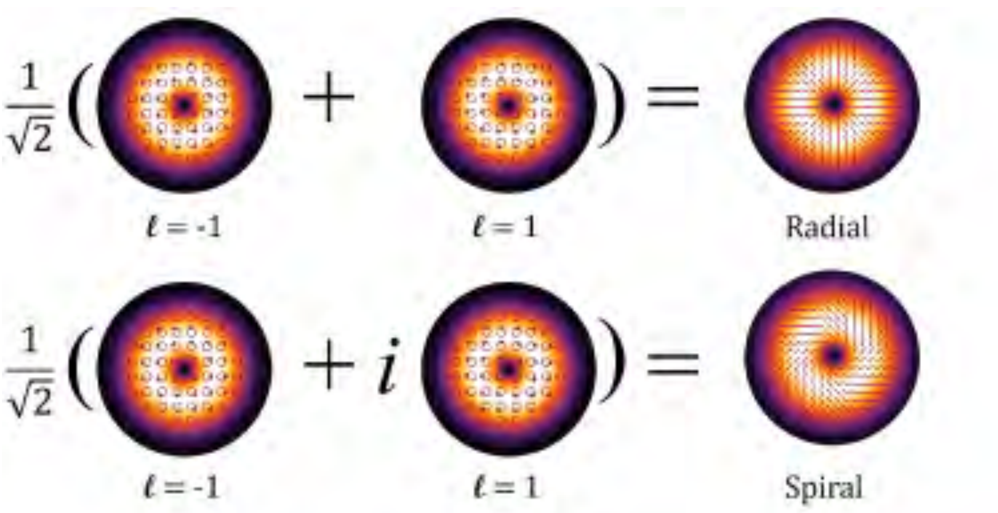
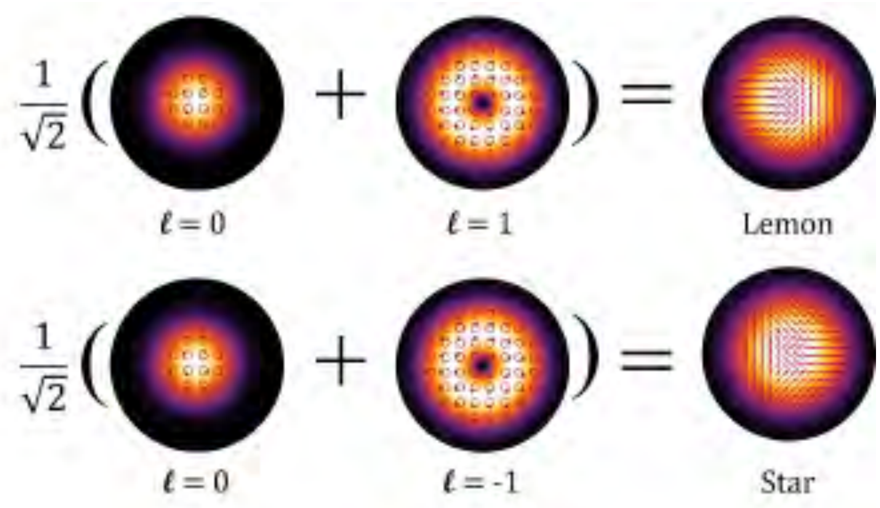
Structured Light Beams

- One can use the transverse degree of freedom of the light field to encode information.
- Not all light waves are infinite plane waves!
- Even a single photon in such a structured field can carry many bits of information
- Example: Space-Varying Polarized Light Beams

Vector Vortex Beams



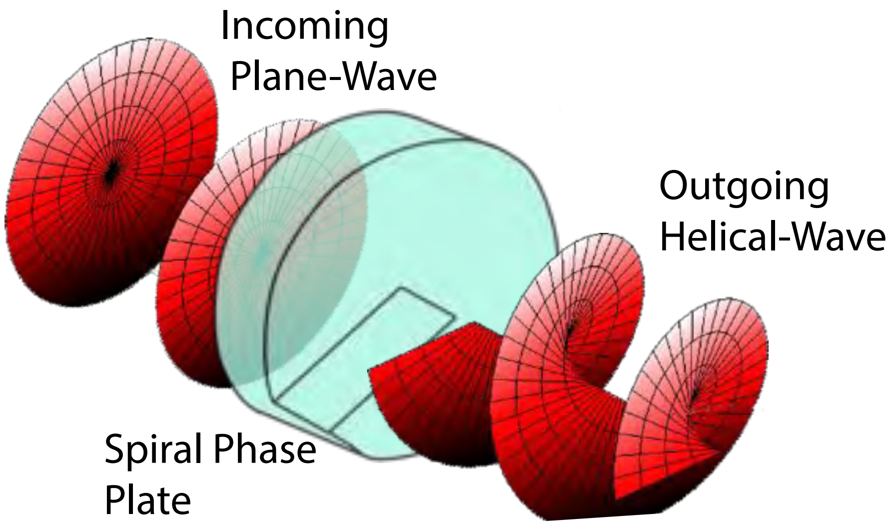
Poincaré Beams



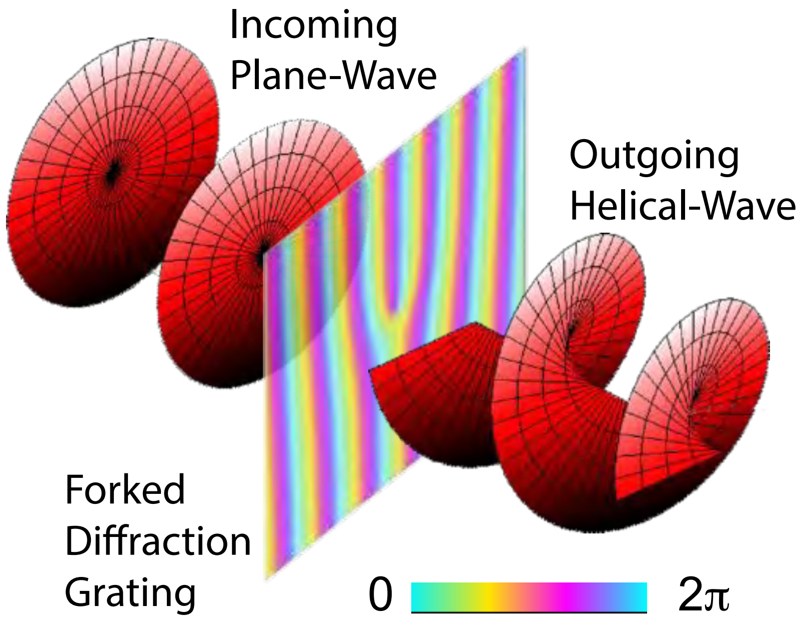
Larocque et al, PRL 2016 (in press)

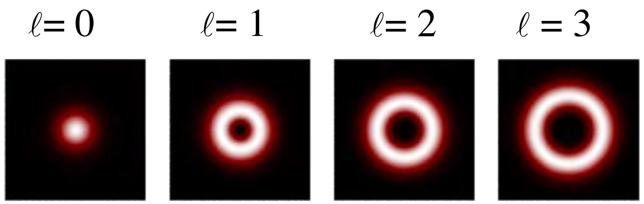
How to create a beam carrying orbital angular momentum?

- Pass beam through a spiral phase plate



- Use a spatial light modulator acting as a computer generated hologram (more versatile)





Exact solution to simultaneous intensity and phase masking with a single phase-only hologram, E. Bolduc, N. Bent, E. Santamato, E. Karimi, and R. W. Boyd, Optics Letters 38, 3546 (2013).

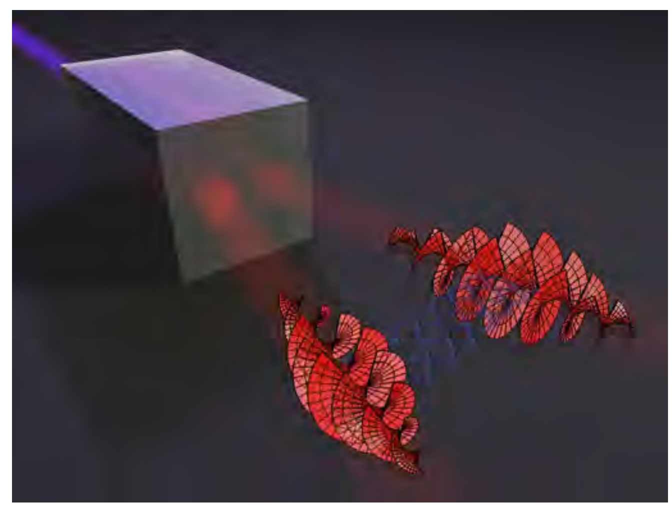
Use of Quantum States for Secure Optical Communication

- The celebrated BB84 protocol for quantum key distribution (QKD) transmits one bit of information per received photon

- We have built a QKD system that can carry more than one bit per photon.
  - Note that in traditional telecom, one uses many photons per bit!

- Our procedure is to encode using beams that carry orbital angular momentum (OAM), such as the Laguerre-Gauss states, which reside in an infinite dimensional Hilbert space.

Key collaborators: Karimi, Leuchs, Padgett, Willner.



QKD System Carrying Many Bits Per Photon

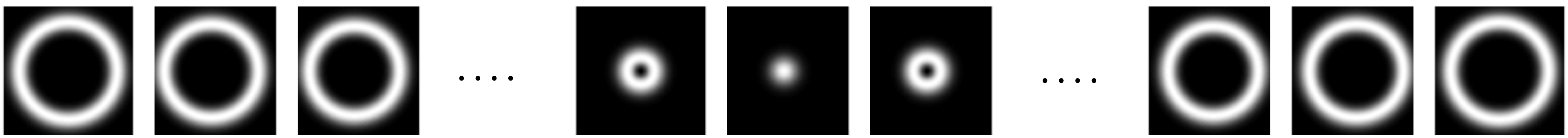
We are constructing a QKD system in which each photon carries many bits of information

We encode in states that carry OAM such as the Laguerre-Gauss states

We also need a second basis composed of linear combinations of these states

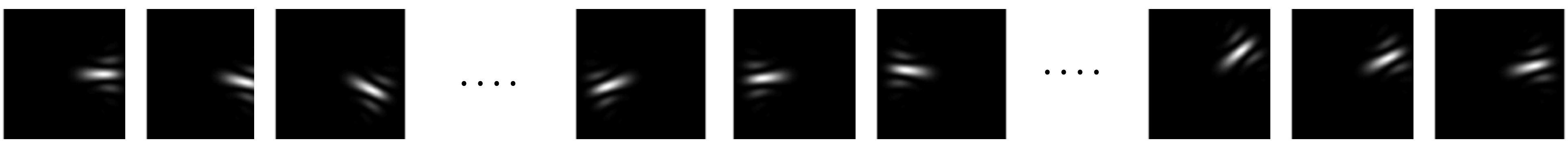
Single Photon States

Laguerre-Gaussian Basis $\ell = -13, \ldots, 13$



-13 -12 -11 .... -1 0 1 .... 11 12 13

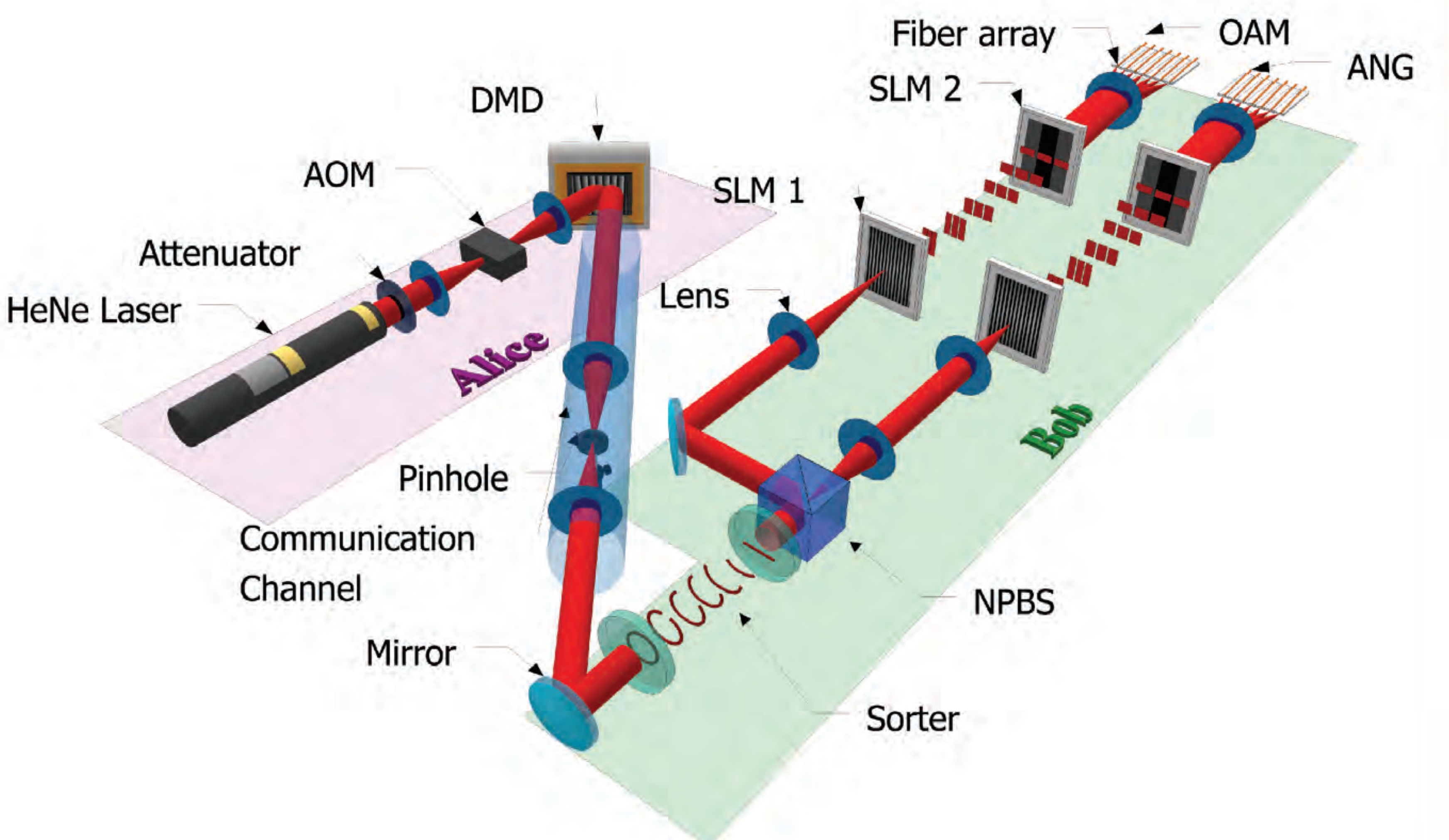
"Angular" Basis (mutually unbiased with respect to LG)

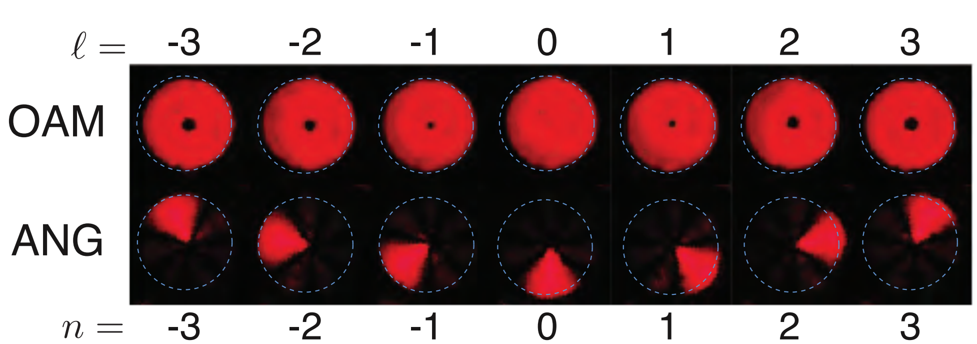


0 1 2 .... 12 13 14 .... 25 26 27

$$\Psi_{\mathrm{AB}}^{N} = \frac{1}{\sqrt{27}} \sum_{l=-13}^{13} \mathrm{LG}_{l,0} \exp\left(i 2\pi N l / 27\right)$$

Laboratory Demonstration of OAM-Based Secure Communication





We use a seven-dimensional state space.

We transfer 2.1 bits per detected photon

Mirhosseini et al., New Journal of Physics 17, 033033 (2015).

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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).

Implications of ENZ Behavior for Nonlinear Optics

Here is the intuition for why the ENZ conditions are of interest in NLO

Recall the standard relation between n_2 and $\chi^{(3)}$

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

Note that for ENZ conditions the denominator becomes very small, leading to a very large value of n_2

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 $\omega = \omega_p/\sqrt{\epsilon_\infty} \equiv \omega_0$.

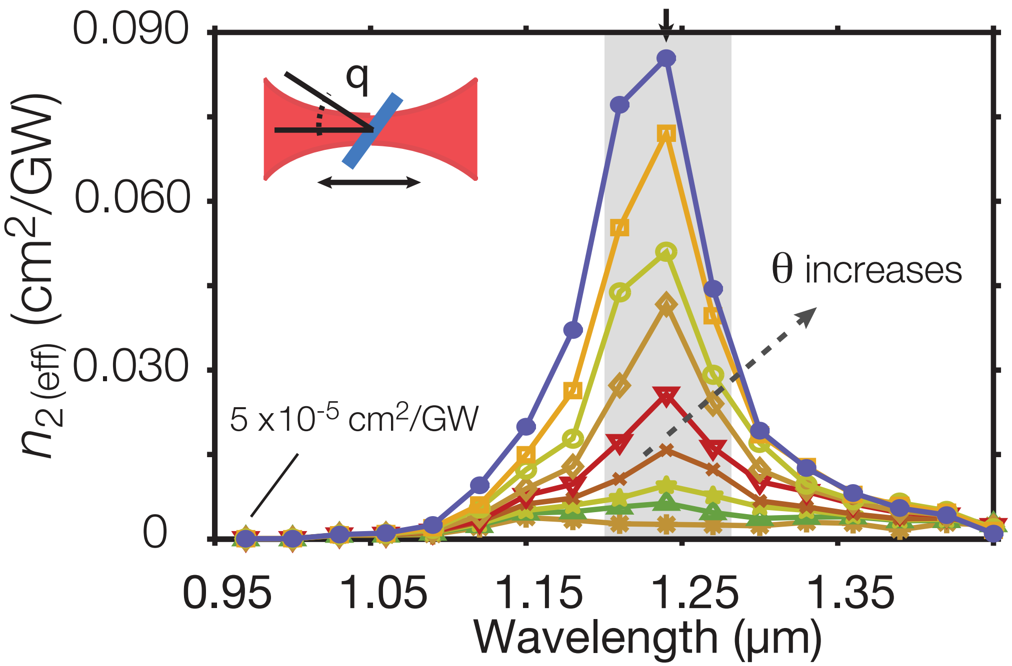
The region near ω_0 is known as the epsilon-near-zero (ENZ) region.

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

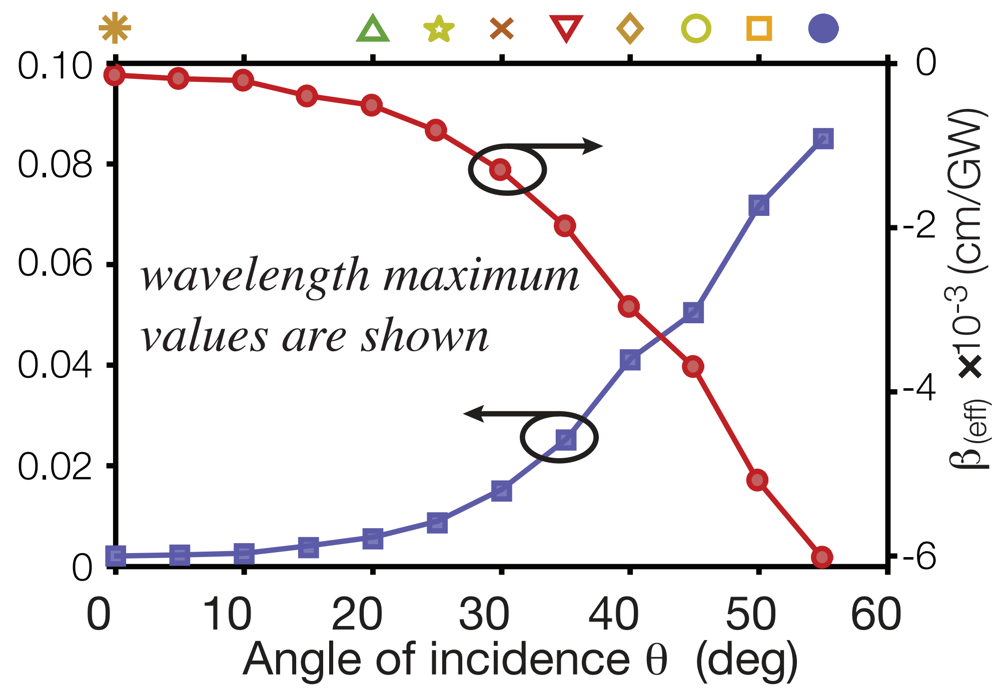
H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. Yin, and X. Zhang, Science 342, 1223 (2013).
C. Argyropoulos, P.-Y. Chen, G. D'Aguanno, N. Engheta, and A. Alu, Phys. Rev. B 85, 045129 (2012).
S. Campione, D. de Ceglia, M. A. Vincenti, M. Scalora, and F. Capolino, Phys. Rev. B 87, 035120 (2013).
A. Ciattoni, C. Rizza, and E. Palange, Phys. Rev. A 81,043839 (2010).

Huge Nonlinear Optical Response Measured by Z-scan

Wavelength dependence of n_2

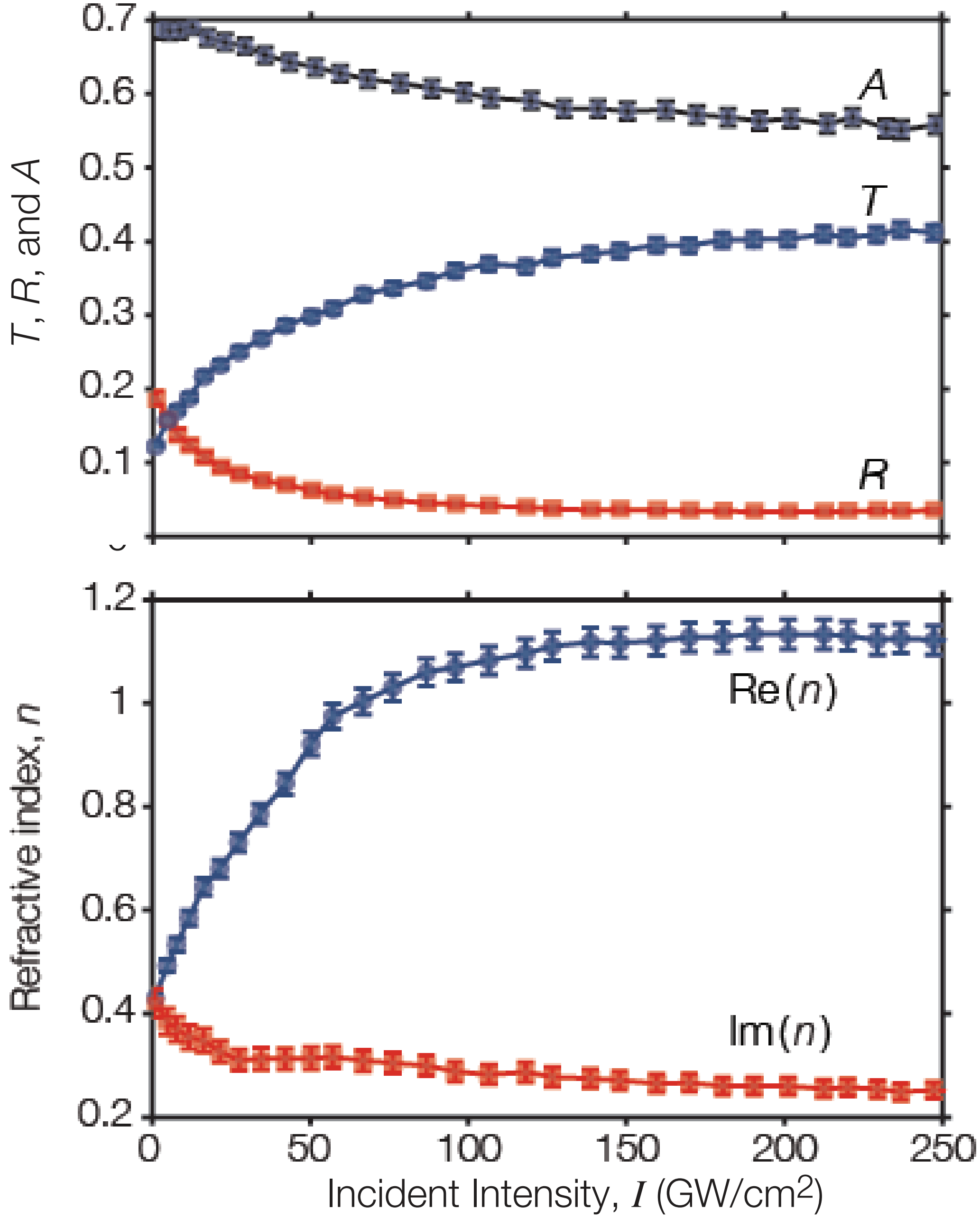


Variation with incidence angle



- Note that n_2 is positive (self focusing) and β 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^2/GW = 1.1 x 10^{-10} cm^2/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 10^5 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.

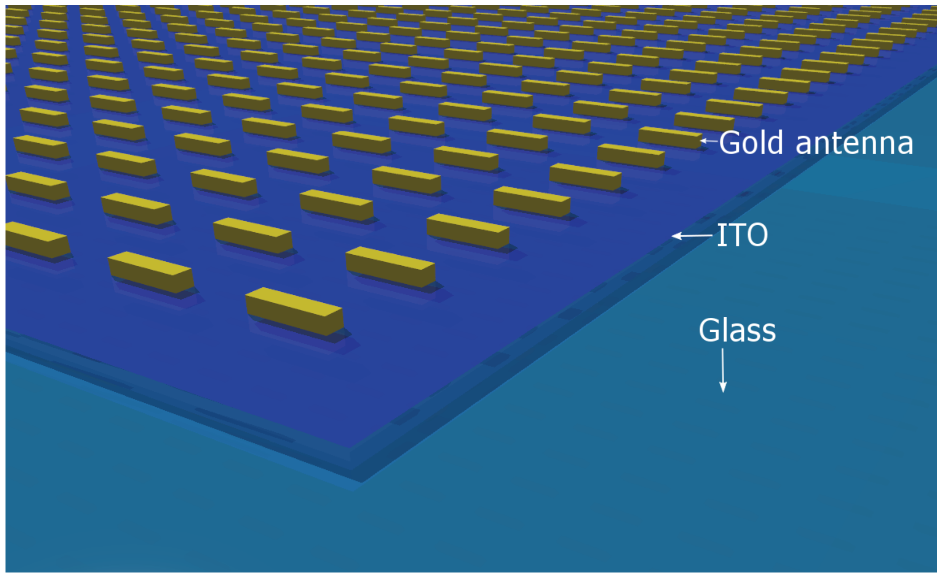
Note that the total nonlinear change in refractive index is $\Delta n = 0.8$.

The absorption decreases at high intensity, allowing a predicted NL phase shift of 0.5 radians.

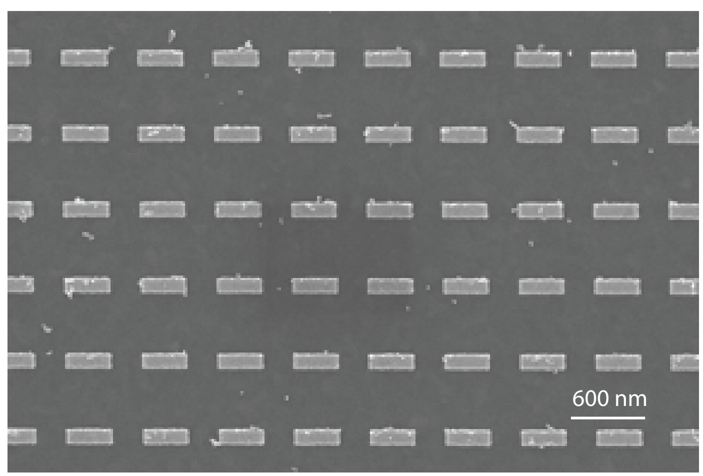
An ENZ Metasurface

- 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

Concept:



SEM:



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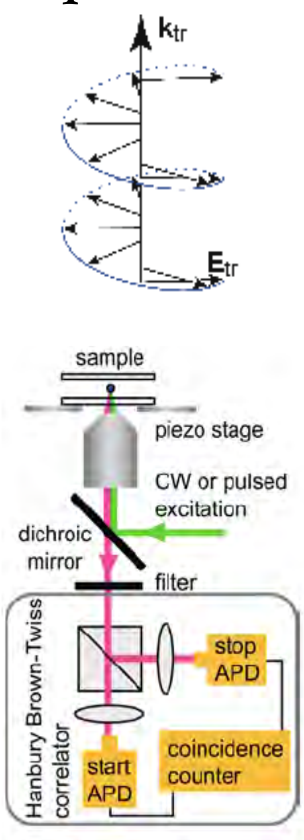
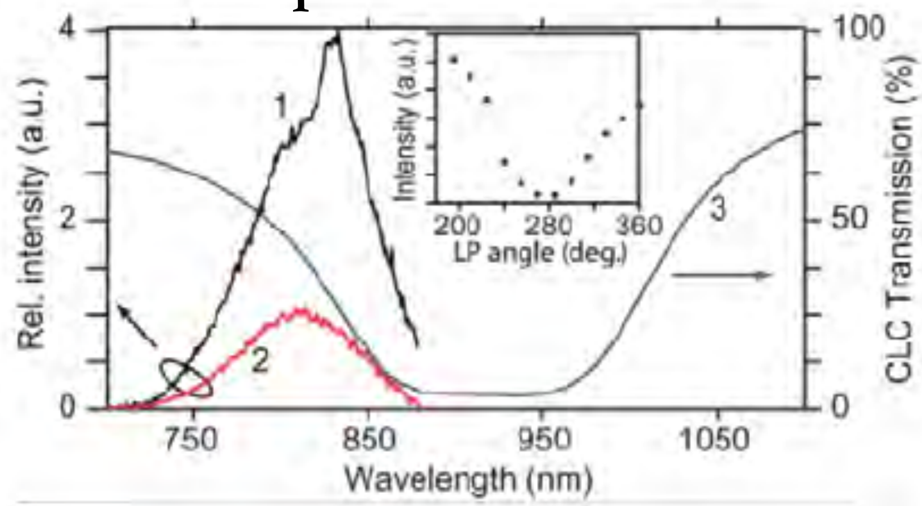
Single-Photon Sources

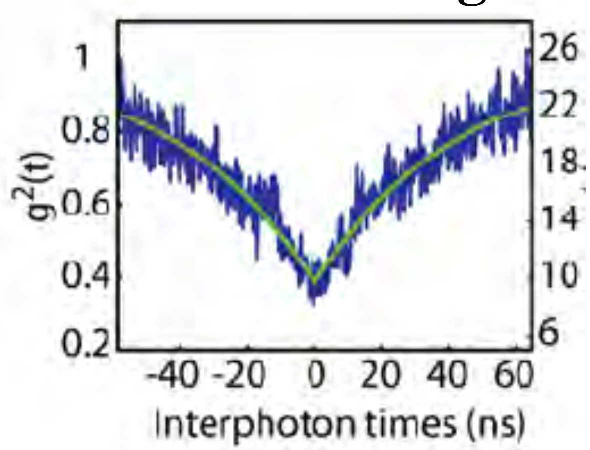
- Many protocols in quantum information require a single-photon source
- An example is the BB84 protocol of quantum key distribution
  - If by accident two photons were sent, one could be stolen by an eavesdropper
  - Even in a weak coherent state, there is a nonvanishing probability of two or more photons being sent
- Circularly polarized fluorescence and antibunching from a nanocrystal quantum dot doped into a glassy cholesteric liquid crystal microcavity



spectrum



anti-bunching

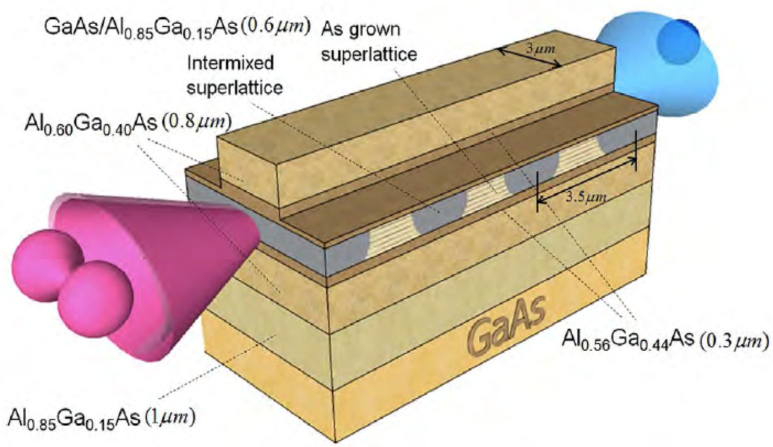


Lukishova et al., Journal of Physics: Conference Series 594 (2015) 012005

On-Chip Photonic Devices for Quantum Technologies

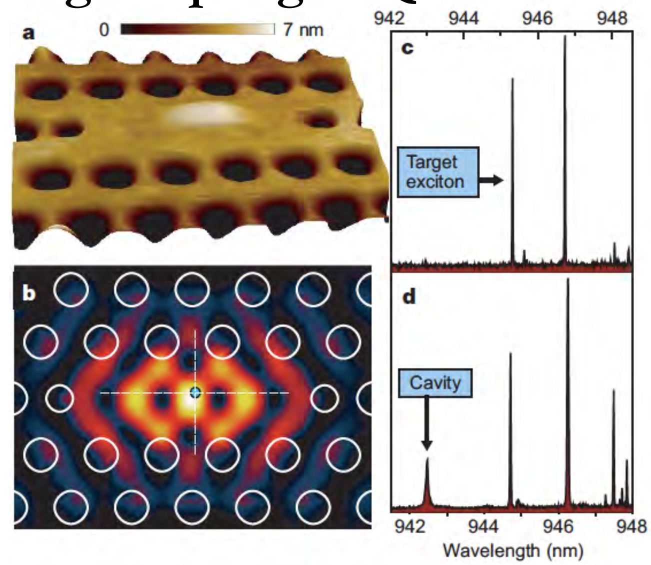
- To make quantum technolgies practical, we need to develop networks of quantum devices on a single chip

- Source of correlated photons



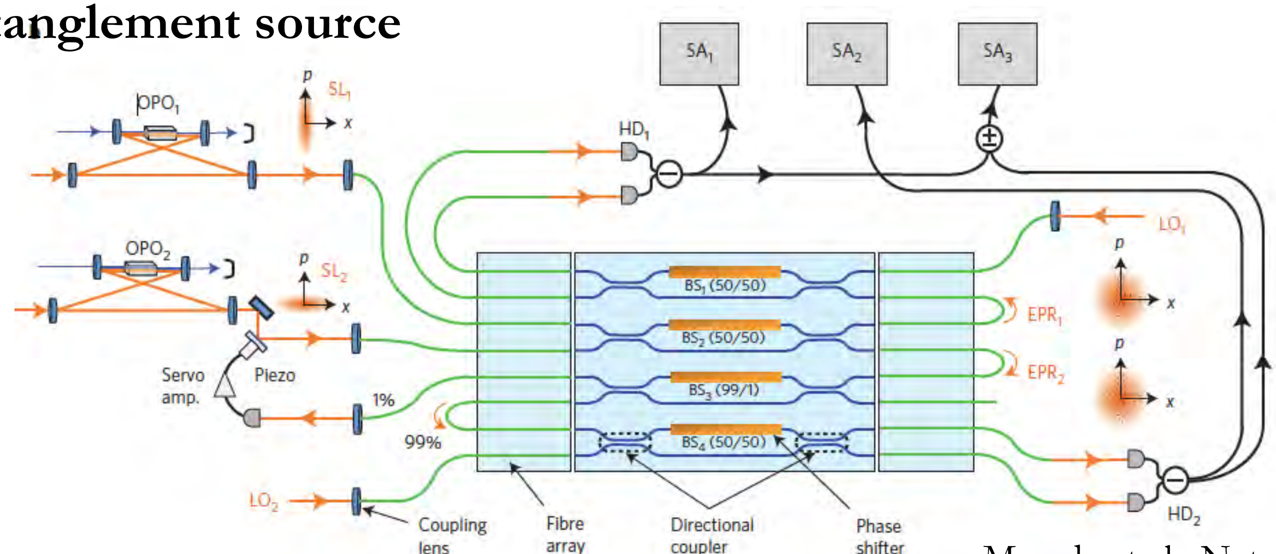
Sarrafi et al., Appl. Phys. Lett. 103, 251115 (2013).

- Strong coupling of QD to PhC resonator



Hennessy et al., Nature 445, 896 (2007)

- Entanglement source

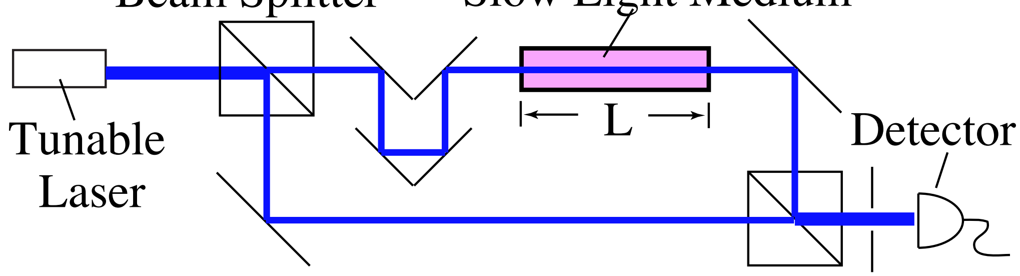


Masada et al., Nature Photonics 9, 316 (2015).

Related Project: Chip-Scale Slow-Light Spectrometer

- The spectral sensitivity of an interferometer is increased by a factor as large as the group index of a material placed within the interferometer.
- We want to exploit this effect to build chip-scale spectrometers with the same resoluation as large laboratory spectromters
- Here is why it works:

Slow-light interferometer:

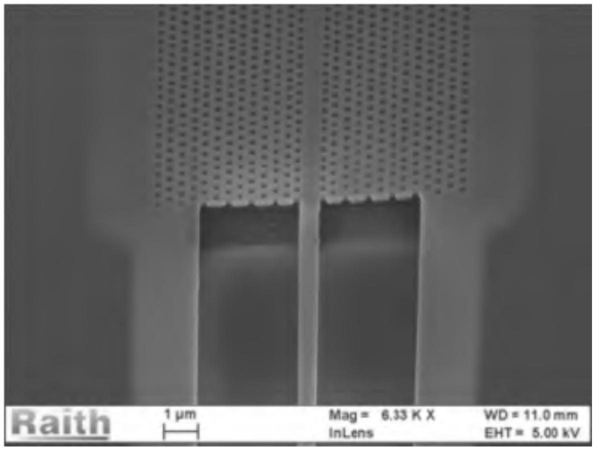


Simple analysis

$$\frac{d\,\Delta\phi}{d\omega} = \frac{d}{d\omega}\frac{\omega n L}{c} = \frac{L}{c}\left(n + \omega\frac{dn}{d\omega}\right) = \frac{L n_g}{c}$$

- We use line-defect waveguides in photonic crystals as our slow light mechanism

Slow-down factors of greater than 100 have been observed in such structures.



Shi, Boyd, Gauthier, and Dudley, Opt. Lett. 32, 915 (2007)
Shi, Boyd, Camacho, Vudyasetu, and Howell, PRL. 99, 240801 (2007)
Shi and Boyd, J. Opt. Soc. Am. B 25, C136 (2008).

Where are we?

Mar Hall

Mar Lodge, Cairngorms National Park

Beyond the perturbative description of the nonlinear optical response of low-index materials

ORAD RESHEF[1,*], ENNO GIESE[1], M. ZAHIRUL ALAM[1], ISRAEL DE LEON[2], JEREMY UPHAM[1], AND ROBERT W. BOYD[1,3]

[1]Department of Physics, University of Ottawa, 25 Templeton Street, Ottawa, Ontario K1N 6N5, Canada
[2]School of Engineering and Sciences, Tecnológico de Monterrey, Monterrey, Nuevo León 64849, Mexico
[3]Institute of Optics and Department of Physics and Astronomy, University of Rochester, Rochester, NY 14627, USA
[*]e-mail: orad@reshef.ca

(To appear in Optics Letters)

(Standard derivation)

To lowest nonlinear order, the polarization of a material illuminated by a monochromatic laser field is described as:

$$P^{\mathrm{TOT}} = P + P^{\mathrm{NL}} = \epsilon_0 E \left[\chi^{(1)} + 3\chi^{(3)}|E|^2\right]. \quad \textbf{(1)}$$

Here, E is the complex amplitude of the applied electric field and $\chi^{(1)} \equiv \epsilon^{(1)} - 1$ corresponds to the linear response of the material, with $\epsilon^{(1)}$ being the linear relative permittivity. The relative permittivity including only the $\chi^{(3)}$ nonlinearity is thus

$$\epsilon = \epsilon^{(1)} + 3\chi^{(3)}|E|^2. \quad \textbf{(2)}$$

Since all of these quantities may be complex, we define the complex relative permittivity as $\epsilon = \epsilon' + i\epsilon''$ and the complex refractive index as $n = n' + in''$, where a single prime denotes the real part, and the double prime the imaginary part, respectively. These two quantities are related by [20]

$$n = \sqrt{\epsilon} = \sqrt{\epsilon^{(1)} + 3\chi^{(3)}|E|^2}. \quad \textbf{(3)}$$

Now we introduce n_2

Together, these equations can be used to obtain the complex, intensity-dependent index of refraction n due to third-order contributions. We find that

$$n = \sqrt{n_0^2 + 2n_0 n_2 I}, \tag{4}$$

This form is valid, but weird

where we take $n_0 = \sqrt{\epsilon^{(1)}}$ to be the linear refractive index, I to be the optical field intensity

$$I = 2\mathrm{Re}(n_0)\epsilon_0 c|E|^2, \tag{5}$$

Note that intensity vanishes under ENZ conditions!

and we introduce the standard definition for the nonlinear index of refraction [4, 20]

$$n_2 = \frac{3\chi^{(3)}}{4n_0 \mathrm{Re}(n_0)\epsilon_0 c}. \tag{6}$$

In order to obtain a simpler relation for n, Eq. (4) is usually expanded in a power series under the assumption that $|2n_2 I/n_0| \ll 1$ [4], yielding

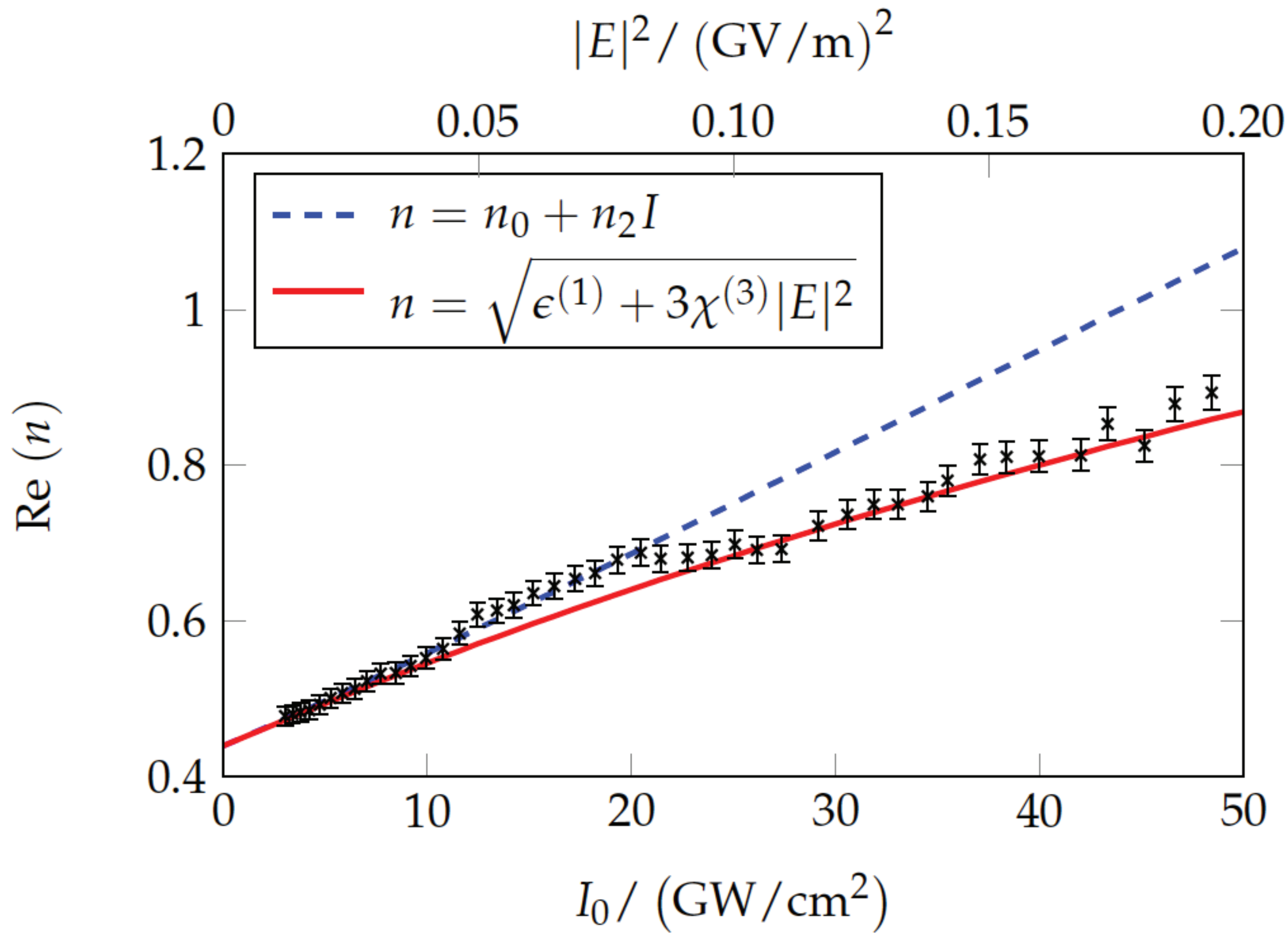
$$n = n_0\sqrt{1 + 2\frac{n_2 I}{n_0}} \approx n_0\left[1 + \frac{1}{2}\left(2\frac{n_2 I}{n_0}\right) + \ldots\right]. \tag{7}$$

In most materials, $|2n_2 I/n_0|$ is very small so that only the lowest order correction term is kept, resulting in the intensity-dependent refractive index being widely defined as

$$n = n_0 + n_2 I. \tag{8}$$

This form is invalid for ENZ materials (because Series 7 does not converge)

Comparison To Our Experimental Data

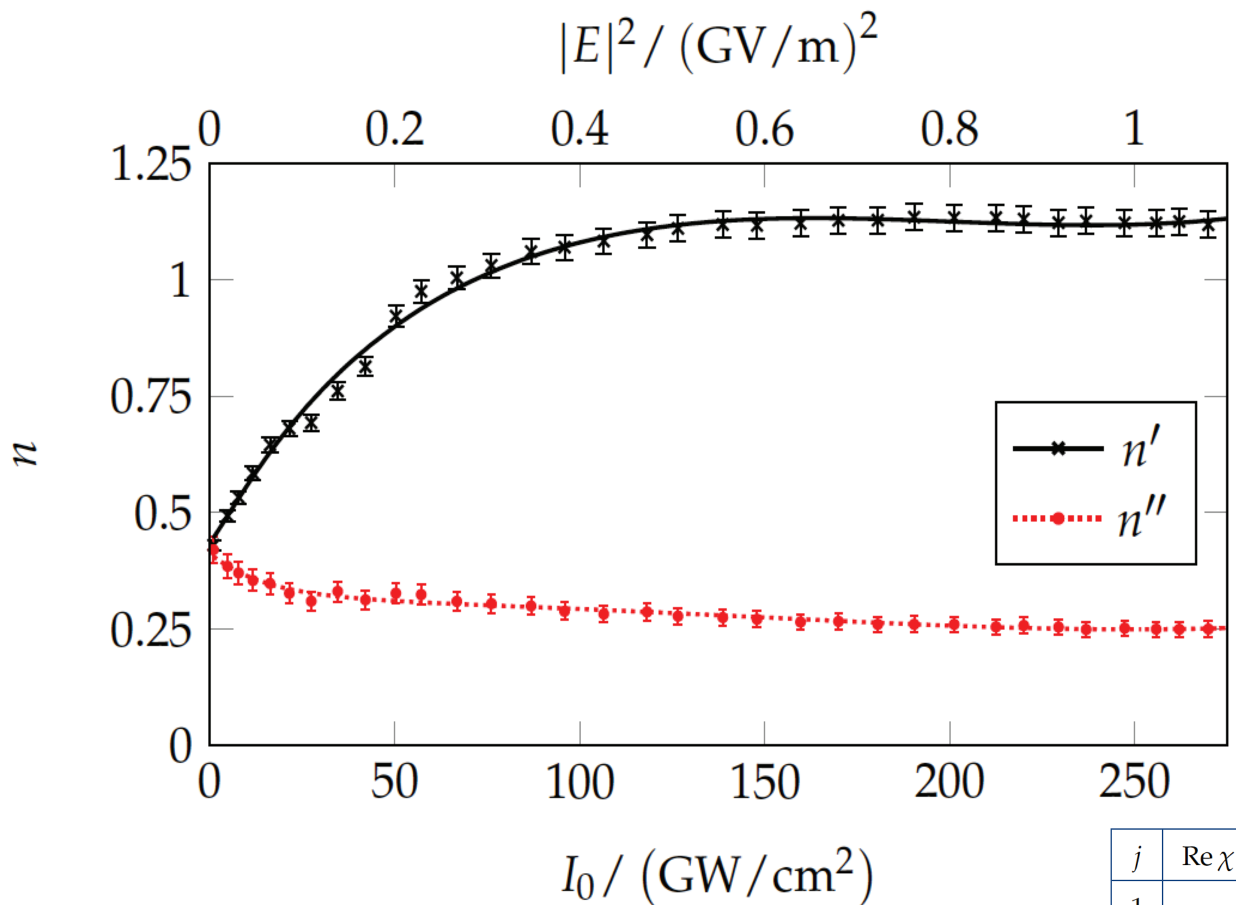


The "square-root" model fits the data much better than the $n_2 I$ model.

I_0 is the incident intensity (measured outside the material);
E is the electric field measured inside the material

For higher intensities, we need to include $\chi^{(5)}$ and $\chi^{(7)}$ contributions

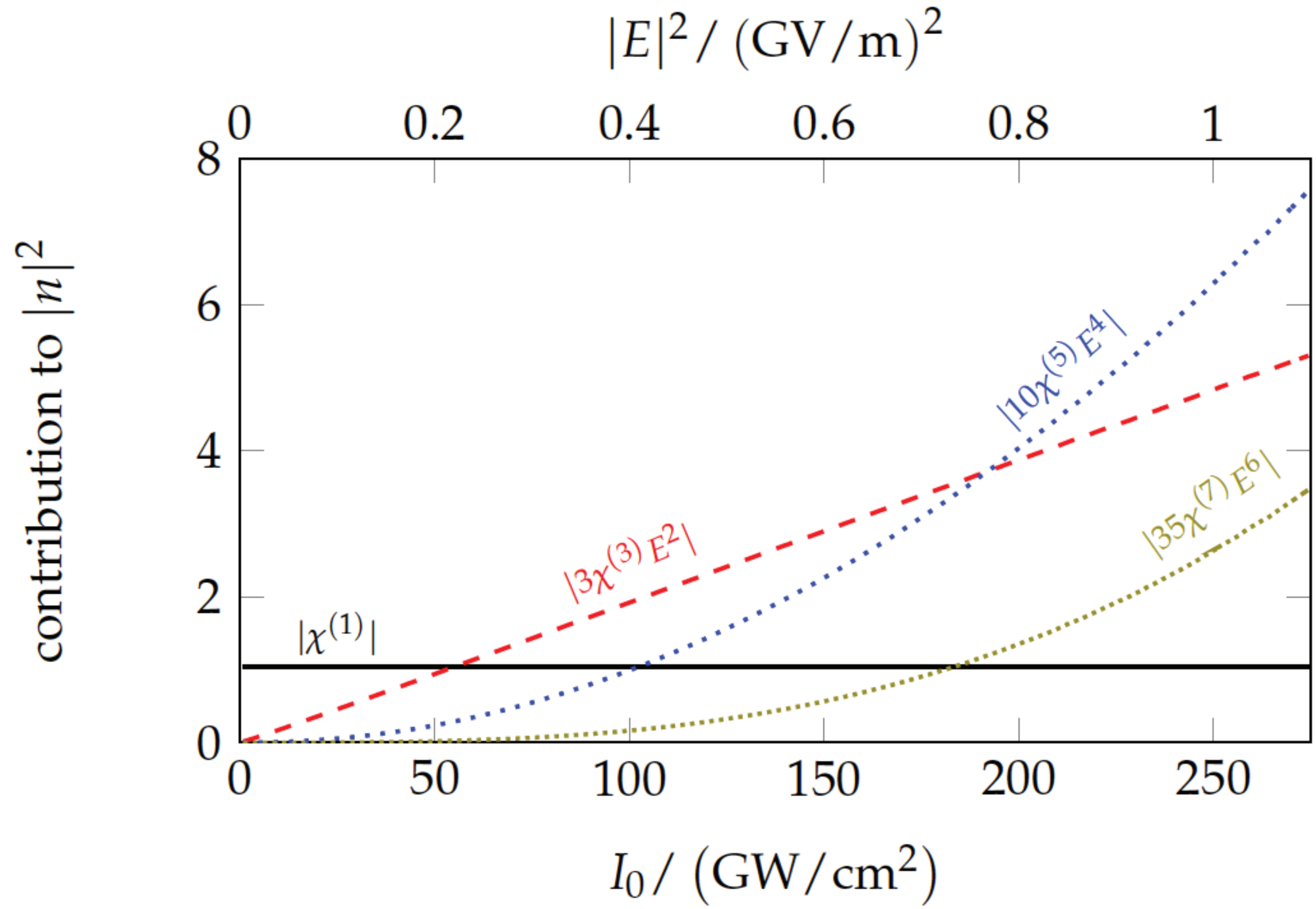
(but no need to include higher-order contributions, up to damage threshold)



j	$\mathrm{Re}\,\chi^{(j)} / (10^{-9}\mathrm{m/V})^{j-1}$	$\mathrm{Im}\,\chi^{(j)} / (10^{-9}\mathrm{m/V})^{j-1}$
1	-0.980 ± 0.008	0.36 ± 0.01
3	1.60 ± 0.03	0.50 ± 0.05
5	-0.63 ± 0.02	-0.25 ± 0.04
7	$(7.7 \pm 0.3) \times 10^{-2}$	$(3.5 \pm 0.8) \times 10^{-2}$

TABLE I. Values extracted from the fit to Eq. (9) with a third, fifth and seventh-order nonlinearity.

Nonlinear Response of ITO is Nonperturbative



Conclusions

1. The conventional equation $n = n_0 + n_2 I$ is not applicable to ENZ and other low-index materials.

2. The nonlinear response can be accurately modeled in the $\chi^{(3)}$ limit by

$$n = \sqrt{n_0^2 + 2n_0 n_2 I}$$

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

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

4. The nonlinear response of ITO is nonperturbative.