Structured Materials and Structured Light for Quantum Photonics

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Structured Materials and Structured Light for Quantum Photonics

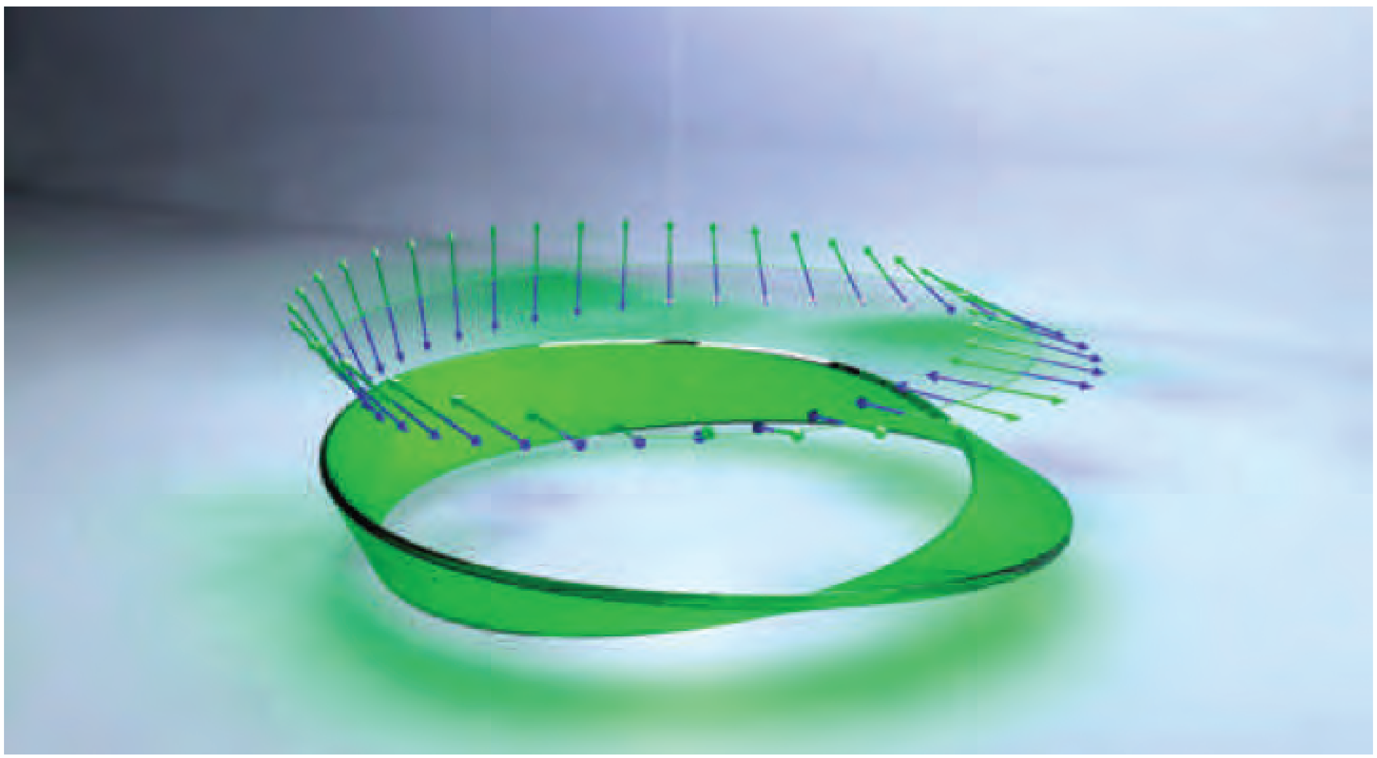
- Not all materials are uniform dielectrics
- Not all light waves are plane waves

Example of Structured Light

Optical Möbius strips!

Observation of Optical Polarization Möbius Strips

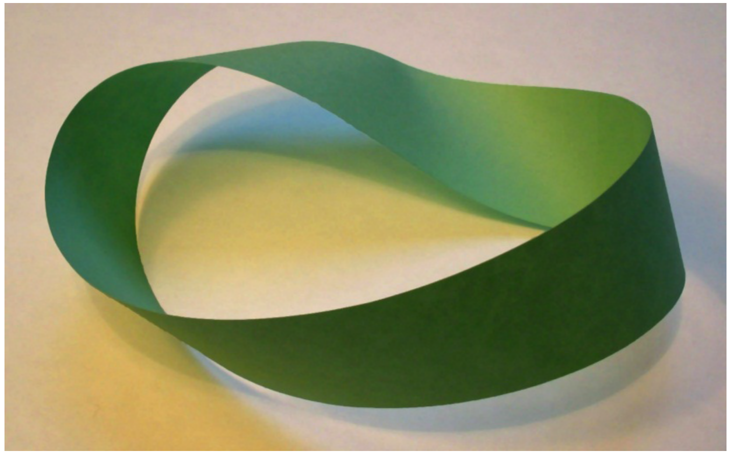
- Möbius strips are familiar geometrical structures, but their occurrence in nature is extremely rare.
- We generate such structures in the nanoscale in tightly focused vector light beams and confirm experimentally their Möbius topology.



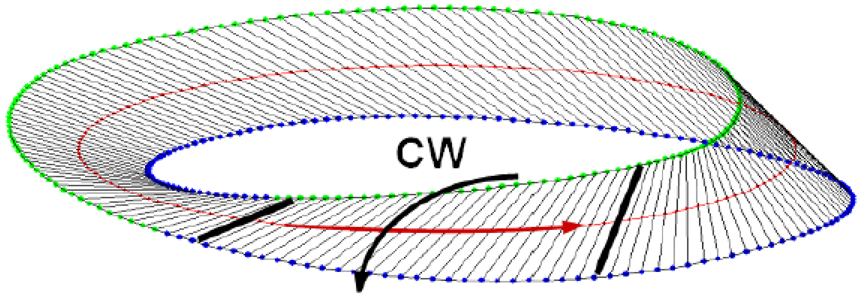
Bauer, Banzer, Karimi, Orlov, Rubano, Marrucci, Santamato, Boyd and Leuchs, Science, 347, 964 (2015)

Prediction of Optical Möbius Strips

An “ordinary” Möbius strip



A polarization Möbius strip
(introduced by Isaac Freund)



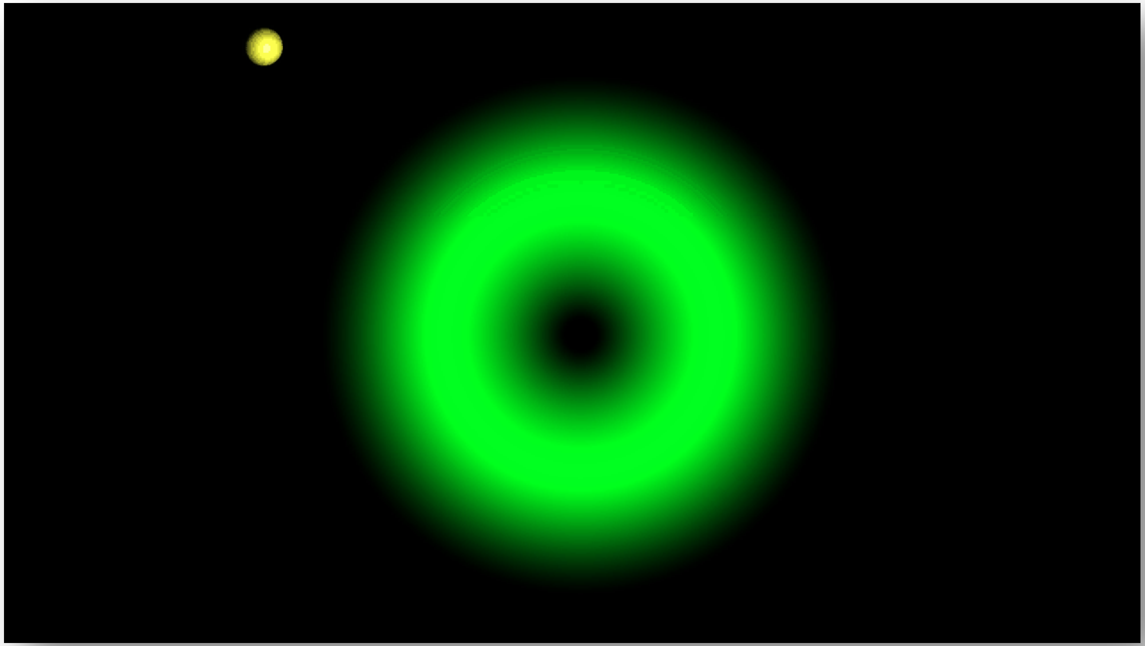
- Isaac Freund discovered, described, and investigated these unusual structures
- To observe these structures, one needs to create a very special field distribution (e.g., a Poincaré beam)
- One also needs to observe the field distribution in a very special way (measure polarization as a function of position around a very tightly focused light beam)

[1] Wikipedia
[2] Isaac Freund, Bar-Ilan Univ., Talk: *Optical Moebius Strips and Twisted Ribbons*, Conf. on Singular Optics, ICTP Trieste, Part II, 30 May 2011
Isaac Freund, Opt. Commun. 242, 65-78 (2004)
Isaac Freund, Opt. Commun. 249, 7-22 (2005)
Isaac Freund, Opt. Commun. 256, 220-241 (2005)
Isaac Freund, Opt. Commun. 283, 1-15 (2010)
Isaac Freund, Opt. Commun. 283, 16-28 (2010)
Isaac Freund, Opt. Lett. 35, 148-150 (2010)
Isaac Freund, Opt. Commun. 284, 3816-3845 (2011)

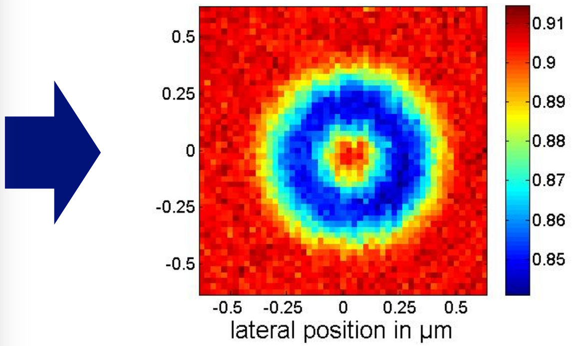
Full vectorial beam measurement on the nanoscale

Nanoparticle-based probing technique for vector beam reconstruction

1. A dipole-like spherical nanoparticle (90 nm diameter) is scanned through the beam
2. The forward- and backward-scattered light for each position of the nanoparticle relative to the beam in the focal plane is measured

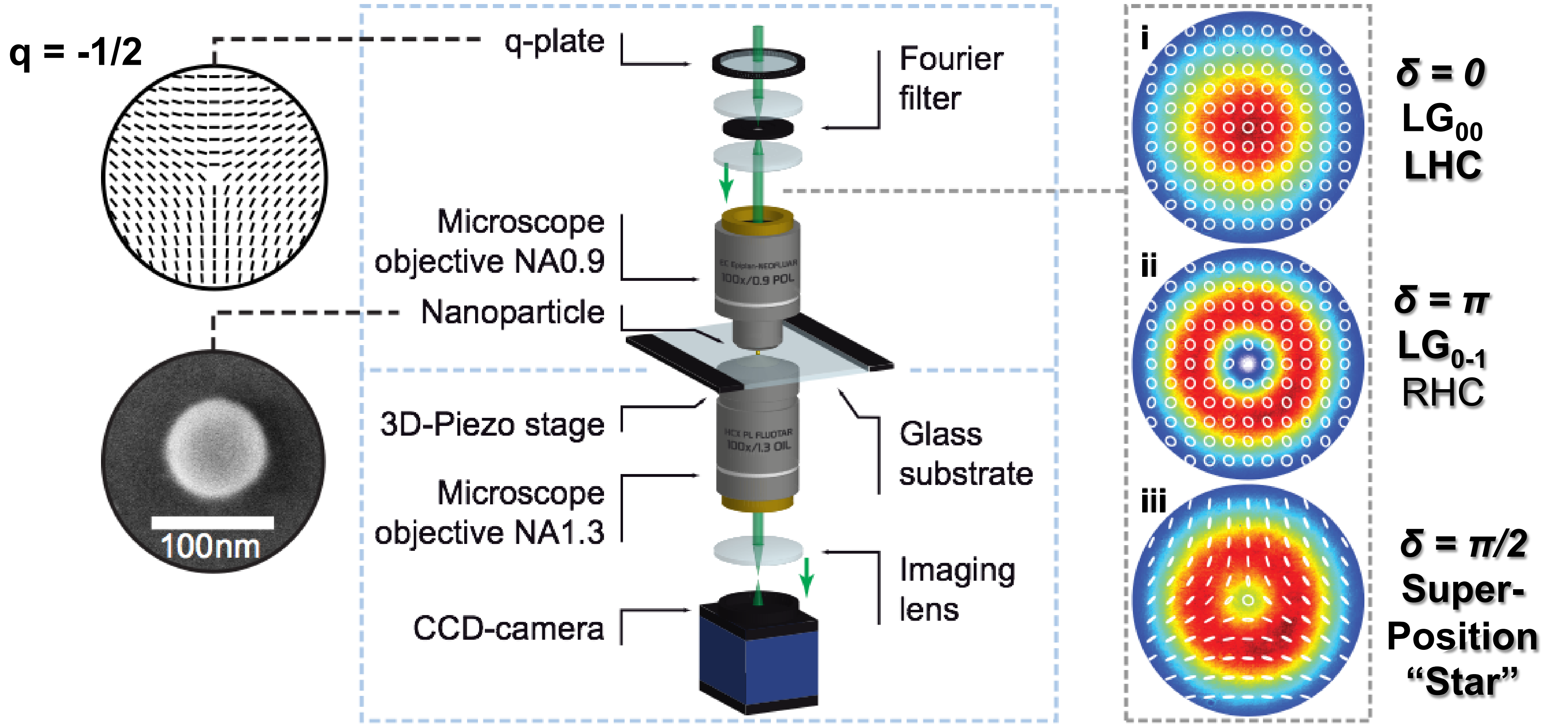


measured intensity
(can also measure
polarization and phase)



Full amplitude and phase reconstruction scheme:
T. Bauer, S. Orlov, U. Peschel, P. B. and G. Leuchs, "Nanointerferometric Amplitude and Phase Reconstruction of Tightly Focused Vector Beams", Nat. Photon 8, 23 - 27 (2014).

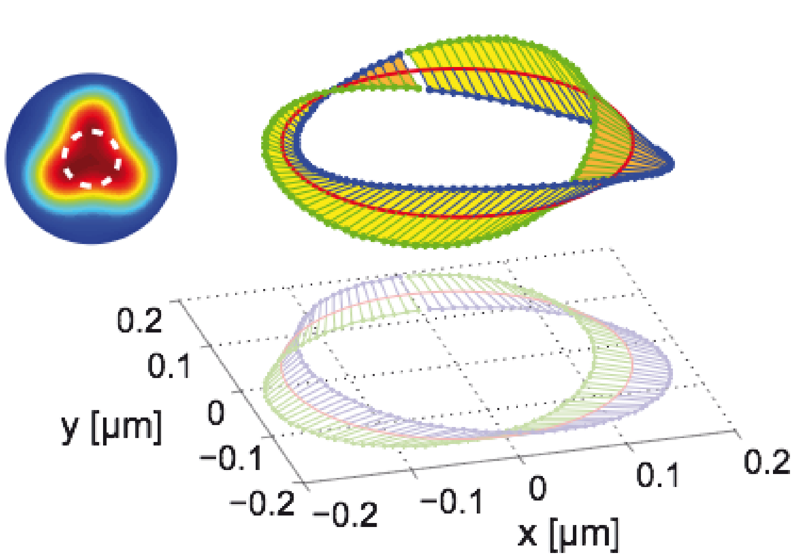
Lab Setup to Observe a Polarization Möbius Strip

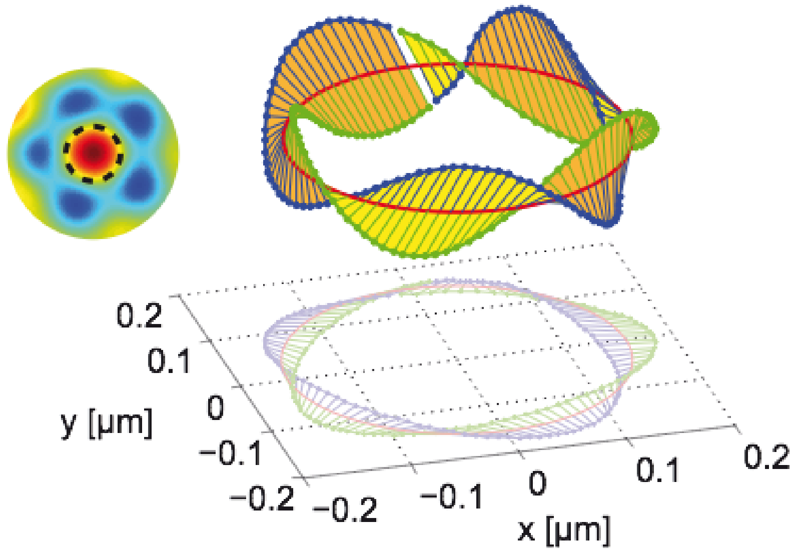


- q-plate: waveplate with a spatially varying orientation (q is the topological charge)
- output beam has a spatially varying state of polarization (vector beam, Poincaré beam, etc.)

Tight focusing enhances the Möbius effect,which depends on the z component of the field

Observation of Polarization Möbius Strips





structure possesses $|q|+1$ half twists

Remarks

- First observation of a polarization Möbius strip
- Light fields can possess rich spatial structure on subwavelength scales
- Current technology is capable of controllably creating beams with such structures and measuring it at subwavelength distances.

Bauer T, Banzer P, Karimi E, Orlovas S, Rubano A, Marrucci L, Santamato E, Boyd RW, and Leuchs G. Science, 2015.

Why structured materials?

To tailor the linear response and nonlinear optical (NLO) response of optical materials.

This could be useful!

The Promise of Nonlinear Optics

Nonlinear optical techniques hold great promise for applications including:

- Photonic Devices
- Quantum Imaging
- Quantum Computing/Communications
- Optical Switching
- Buffers and Routers Based on “Slow Light”

But the lack of high-quality photonic material is often the chief limitation in implementing these ideas.

Our approach to developing improved photonic materials is to use composite and structured materials.

Outline: Structured Nonlinear Optical Materials

1. Nanocomposite materials
   Nanocomposite materials display enhanced NLO response
   Enhancement occurs through local field effects
   NLO response of metal/dielectric nanocomposite materials
   Optical properties of semicontinuous metal films

2. Enhanced nonlinear response of 1-D photonic crystals

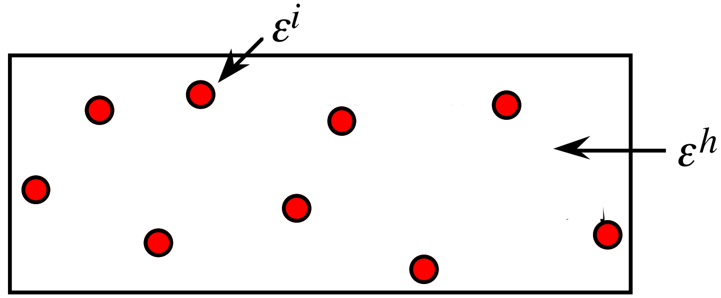
3. NLO response of surface plasmon polaritons

4. Optical properties of structured surfaces

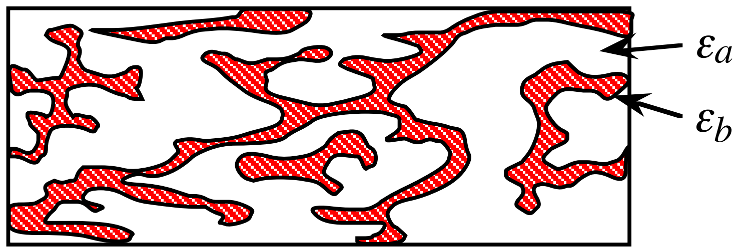
5. Optical properties of guest-host systems
   FBAG: A highly nonlinear material
   Microscopic cascading and enhanced nonlinearity
   Consequences: Image rotation. backwards light, momentum

Nanocomposite Materials for Nonlinear Optics

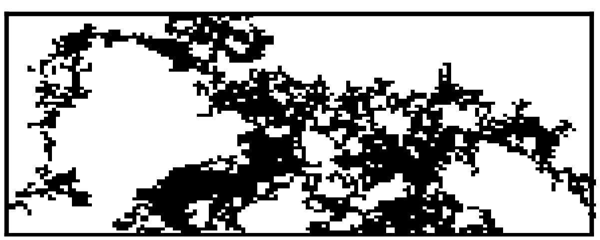
• Maxwell Garnett



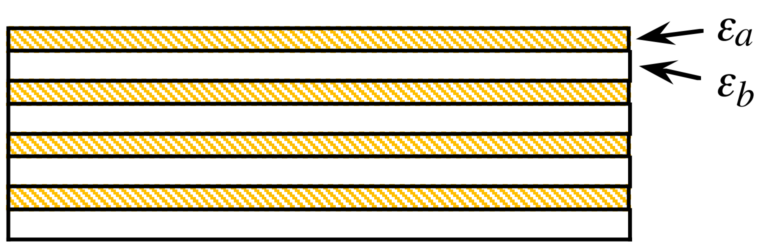
• Bruggeman (interdispersed)



• Fractal Structure



• Layered



- In each case, scale size of inhomogeneity << optical wavelength

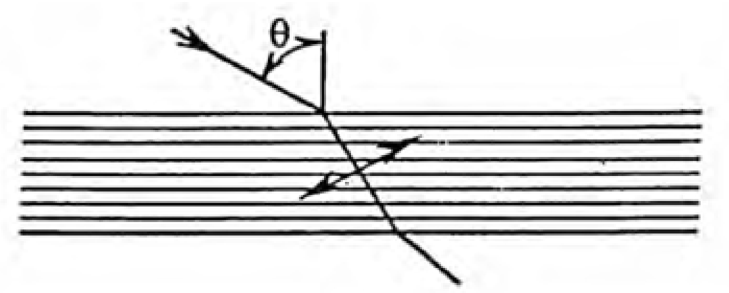
- Thus all optical properties, such as n and $\chi^{(3)}$, can be described by effective (volume averaged) values

Recent review: Dolgaleva and Boyd, Advances in Optics and Photonics 4, 1–77 (2012).

Enhanced NLO Response from Layered Composite Materials

A composite material can display a larger NL response than its constituents!

Alternating layers of TiO_2 and the conjugated polymer PBZT.



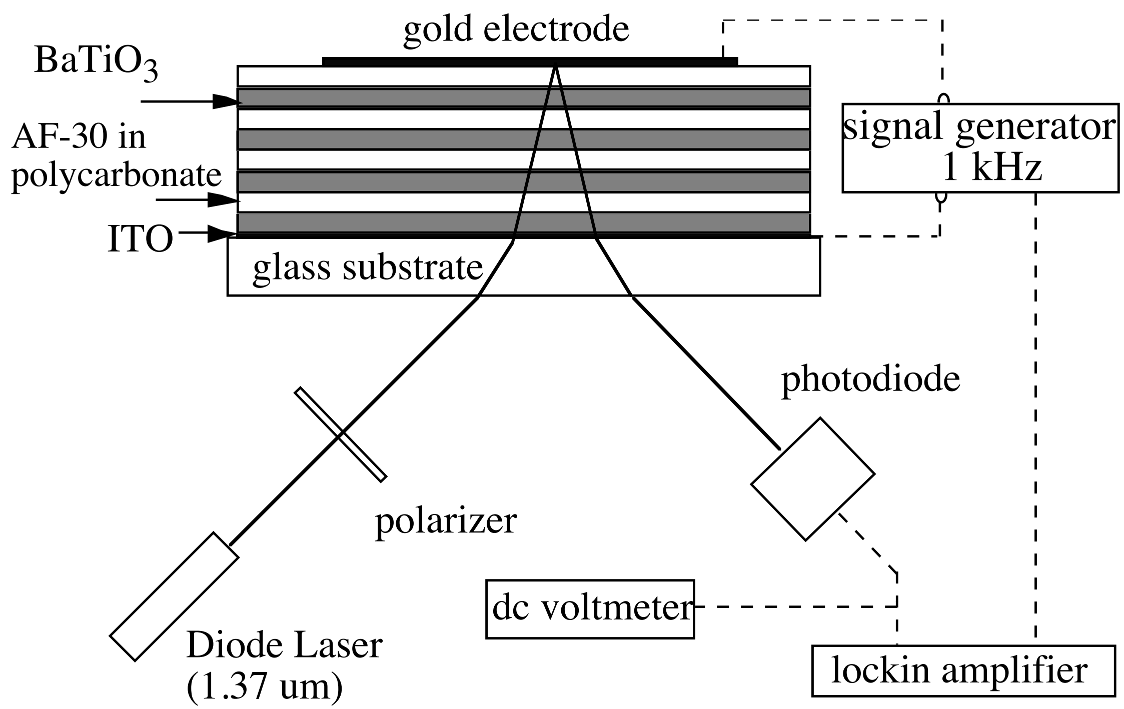
$\nabla \cdot \mathbf{D} = 0$ implies that $(\varepsilon \mathbf{E})_\perp$ is continuous.

Measure NL phase shift as a function of angle of incidence.

35% enhancement in $\chi^{(3)}$

Fischer, Boyd, Gehr, Jenekhe, Osaheni, Sipe, and Weller-Brophy, Phys. Rev. Lett. 74, 1871 (1995).

Quadratic EO effect



3.2 times enhancement!

Nelson and Boyd, APL 74 2417 (1999)

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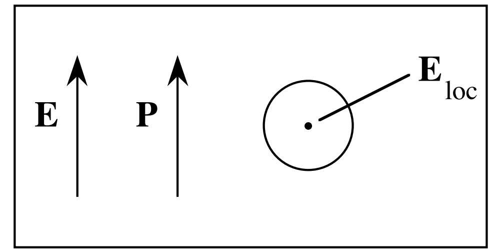
Local Field Effects in Nonlinear Optics – I

Recall the Lorentz-Lorenz Law
(linear optics)



$$\chi^{(1)} = \frac{N\alpha}{1 - \frac{4}{3}\pi N\alpha} \quad \text{or} \quad \frac{\epsilon^{(1)} - 1}{\epsilon^{(1)} + 2} = \frac{4}{3}\pi N\alpha.$$

This result follows from the assumption that the field that acts on a representative atom is not the macroscopic Maxwell field but rather the Lorentz local field given by

$$E_{\text{loc}} = E + \tfrac{4}{3}\pi P \qquad \text{where} \qquad P = \chi^{(1)} E$$

We can rewrite this result as

$$E_{\text{loc}} = LE \quad \text{where} \quad L = \frac{\epsilon^{(1)} + 2}{3} \quad \text{is the local field factor.}$$

Local Field Effects in Nonlinear Optics – II

For the case of nonlinear optics, Bloembergen (1962, 1965) showed that, for instance,

$$\chi^{(3)}(\omega = \omega + \omega - \omega) = N\gamma^{(3)}|L(\omega)|^2[L(\omega)]^2.$$

where $\gamma^{(3)}$ is the second hyperpolarizability and where

$$L(\omega) = \frac{\epsilon(\omega) + 2}{3}$$

For the typical value $n = 2$, $L = 2$, and $L^4 = 16$. Local field effects can be very large in nonlinear optics! But can we tailor them for our benefit?

We have been developing new photonic materials with enhanced NLO response by using composite structures that exploit local field effects.

Enhancement of the NLO Response

- Under very general conditions, we can express the NL response as

$$\chi_{\mathrm{eff}}^{(3)} = f\, L^2 |L|^2 \chi^{(3)}$$

where f is the volume fraction of nonlinear material and L is the local-field factor, which is different for each material geometry.

- Under appropriate conditions, the product $f\, L^2 |L|^2$ can exceed unity.

- For a homogeneous material $L = \dfrac{\varepsilon + 2}{3}$

- For a spherical particle of dielectric constant ε_m embedded in a host of dielectic constant ε_h

$$L = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h}$$

- For a layered geometry with the electric field perpendicular to the plane of the layers, the local field factor for component a is given by

$$L = \frac{\varepsilon_{\mathrm{eff}}}{\varepsilon_{\mathrm{a}}} \qquad \frac{1}{\varepsilon_{\mathrm{eff}}} = \frac{f_{\mathrm{a}}}{\varepsilon_{\mathrm{a}}} + \frac{f_{\mathrm{b}}}{\varepsilon_{\mathrm{b}}}$$

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Material Systems for Composite NLO Materials

All-dielectric composite materials

Minimum loss, but limited NL response

Metal-dielectric composite materials

Larger loss, but larger NL response

Note that $\chi^{(3)}$ of gold $\approx 10^6$ $\chi^{(3)}$ of silica glass!

Also, metal-dielectric composites possess surface plasmon resonances, which can further enhance the NL response.

Gold-Doped Glass: A Maxwell-Garnett Composite

Developmental Glass, Corning Inc.

Red Glass Caraffe
Nurenberg, ca. 1700

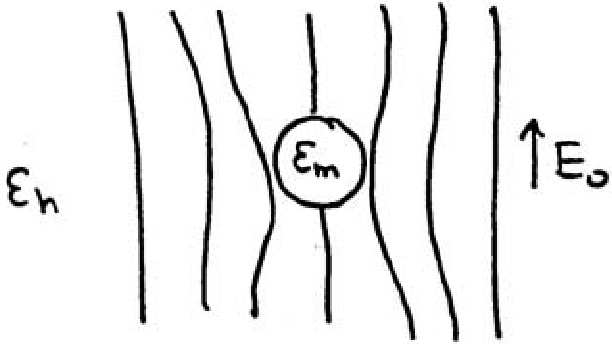
Huelsmann Museum, Bielefeld

gold volume fraction approximately 10^{-6}
gold particles approximately 10 nm diameter

- Composite materials can possess properties very different from those of their constituents.
- Red color is because the material absorbs very strong in the blue, at the surface plasmon frequency

Metal / Dielectric Composites

Very large local field effects



$$E_{in} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} E_0$$

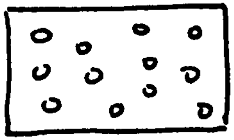
$$\equiv \mathcal{L} E_0$$

(ε_m is negative!)

At resonance

$$\mathcal{L} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} \longrightarrow \frac{3\varepsilon_h}{i\varepsilon_m''} \approx (3 \text{ to } 30)\, i$$

Counterintuitive Consequence of Local Field Effects



gold nanoparticles in a liquid dye solution (HITCI)

Both constituents are reverse saturable absorbers $\Rightarrow$ $\mathrm{Im}\,\chi^{(3)} > 0$

Effective NL susceptibility of composite

$$\chi^{(3)}_{eff} = f\, \mathcal{L}^2 |\mathcal{L}|^2 \chi^{(3)}_{Au} + (1-f)\, \chi^{(3)}_{\text{dye sol'n}}$$

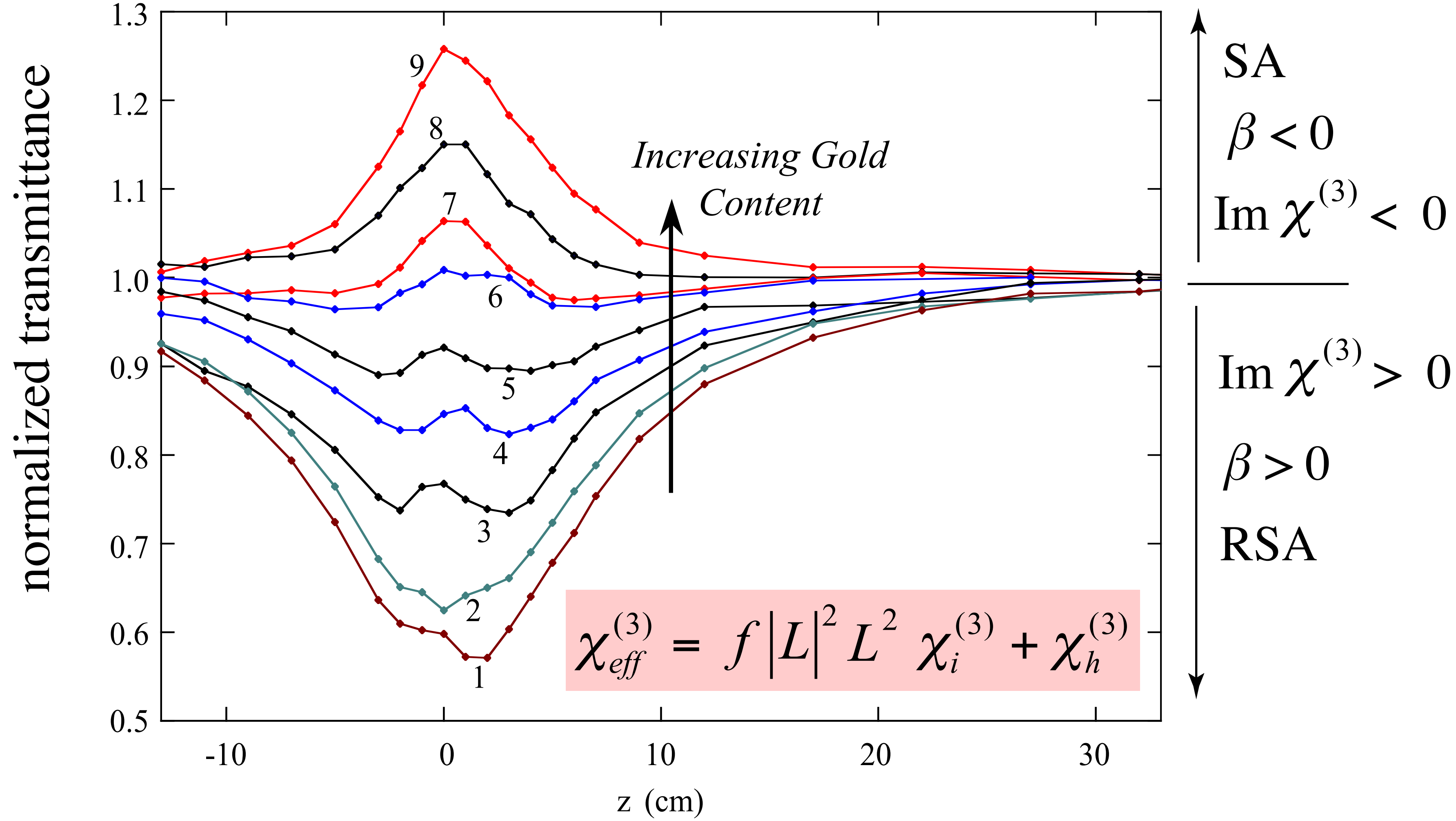
$$\mathcal{L} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} = \text{pure imaginary at resonance!}$$

A cancellation of the two contributions to $\chi^{(3)}$ can occur, even though they have same sign.

Counterintuitive Consequence of Local Field Effects

Cancellation of two contributions that have the same sign

Gold nanoparticles in a reverse saturable absorber dye solution (13 μM HITCI)



D.D. Smith, G. Fischer, R.W. Boyd, D.A.Gregory, *JOSA B* **14**, 1625, 1997.

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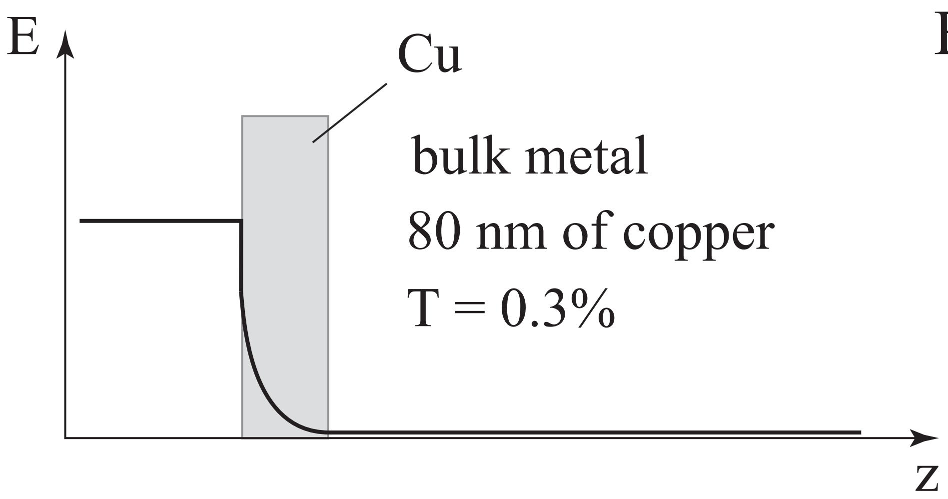
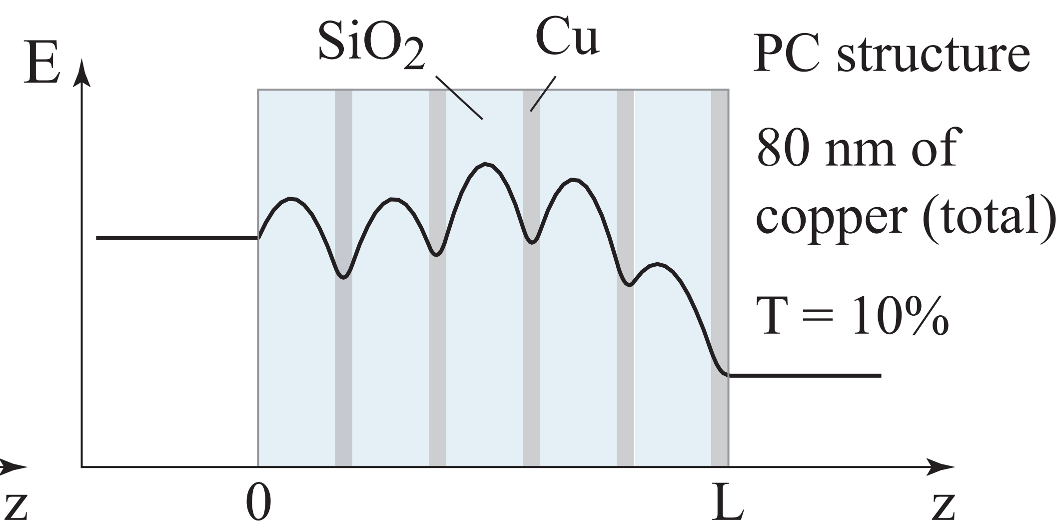
Accessing the Optical Nonlinearity of Metals with Metal-Dielectric Photonic Crystal Structures

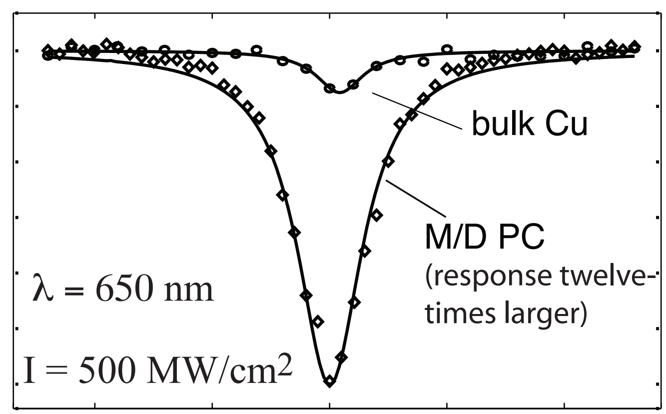
- Metals have very large optical nonlinearities but low transmission
- Low transmission because metals are highly reflecting (not because they are absorbing!)
- Solution: construct metal-dielectric photonic crystal structure (linear properties studied earlier by Bloemer and Scalora)





Bennink, Yoon, Boyd, and Sipe, Opt. Lett. 24, 1416 (1999).

Lepeshkin, Schweinsverg, Piredda, Bennink, and Boyd, Phys. Rev. Lett. 93, 123902 (2004).



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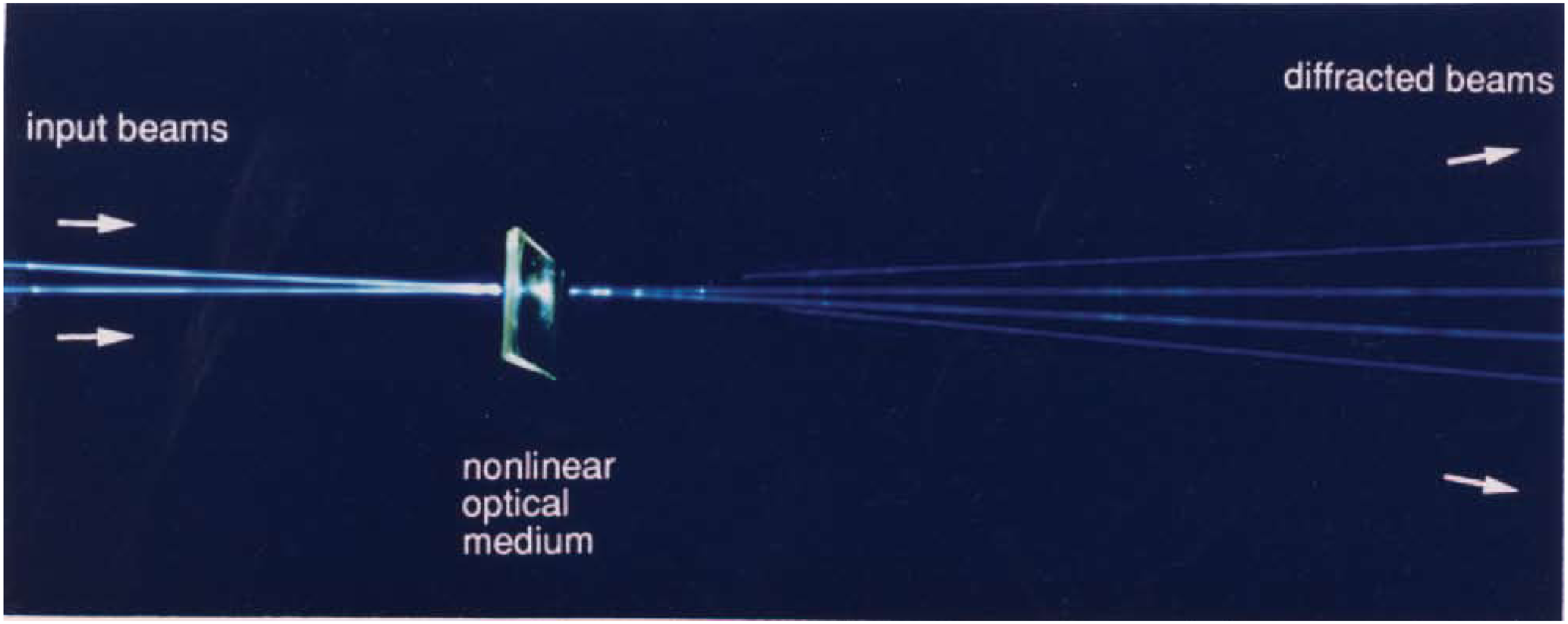
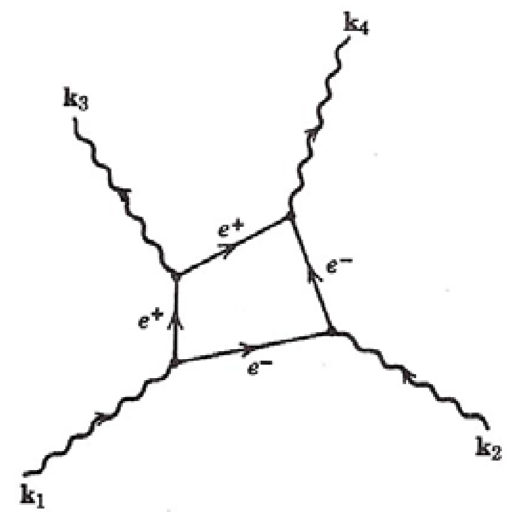
Nonlinear Optics and Light-by-Light Scattering



The elementary process of light-by-light scattering has never been observed in vacuum, but is readily observed using the nonlinear response of material systems.



Nonlinear material is fluorescein-doped boric acid glass (FBAG)

$n_2(\text{FBAG}) \approx 10^{14}\, n_2(\text{silica})$ [But very slow response!]

M. A. Kramer, W. R. Tompkin, and R. W. Boyd, Phys. Rev. A, 34, 2026, 1986.
W. R. Tompkin, M. S. Malcuit, and R. W. Boyd, Applied Optics 29, 3921, 1990.

Why structured materials?

To tailor the linear response and nonlinear optical (NLO) response of optical materials.

This could be useful!

Large nonlinear optical response of polycrystalline $Bi_{3.25}La_{0.75}Ti_3O_{12}$ ferroelectric thin films on quartz substrates

Heedeuk Shin,[1] Hye Jeong Chang,[1,3,*] Robert W. Boyd,[1] M. R. Choi,[2] and W. Jo[2,4]

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[2]*Department of Physics, Ewha Womans University, Seoul 120-750, Korea*
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[4]*wmjo@ewha.ac.kr*
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posted July 17, 2007 (Doc. ID 83167); published August 10, 2007

We measure the nonlinear susceptibility of $Bi_{3.25}La_{0.75}Ti_3O_{12}$ (BLT) thin films grown on quartz substrates using the Z-scan technique with picosecond laser pulses at a wavelength of 532 nm. The third-order nonlinear refractive index coefficient γ and absorption coefficient β of the BLT thin film are 3.1×10^{-10} cm^2/W and 3×10^{-5} cm/W, respectively, which are much larger than those of most ferroelectric films. The results show that the BLT thin films on quartz substrates are good candidate materials for applications in nonlinear optical devices.

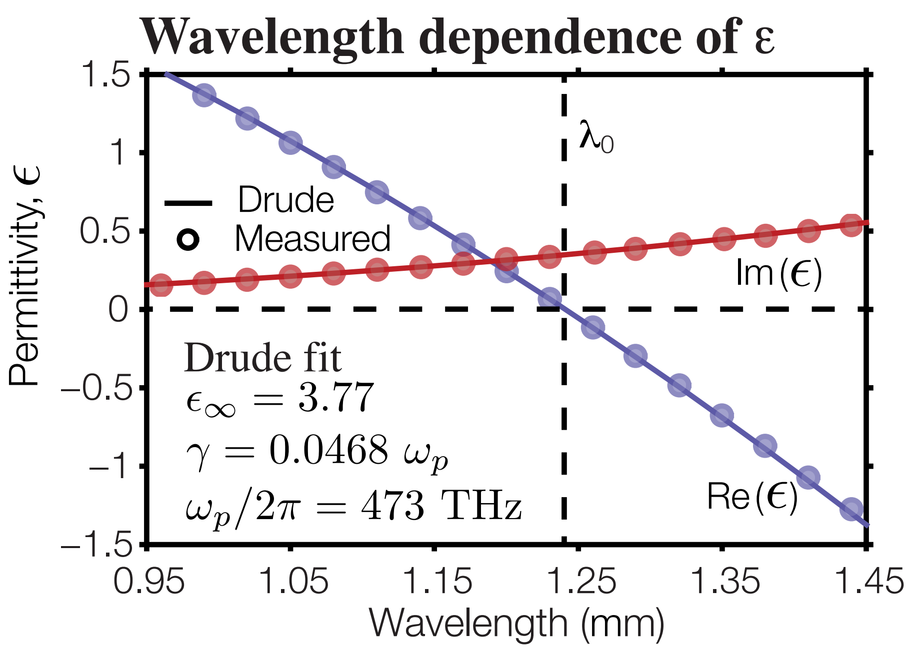
OCIS codes: 160.2260, 160.4330.

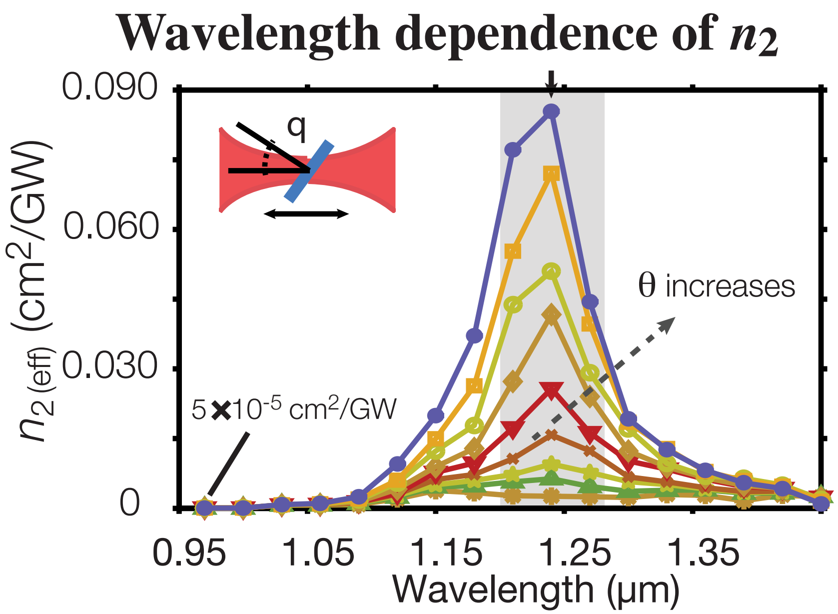
Huge Nonlinear Optical Response of ITO near its Epsilon-Near-Zero Wavelength

Indium Tin Oxide (ITO) displays enormously strong NLO properties:

- n_2 is 2.5 x 10^5 times that of fused silica
- nonlinear change in refractive index as large as 0.8
- response time of 270 fs

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





Some possible new effects

- Waveguiding outside the "weakly-guiding" regime
- Efficient all-optical switching
- No need for phase-matching

Alam, De Leon, Boyd

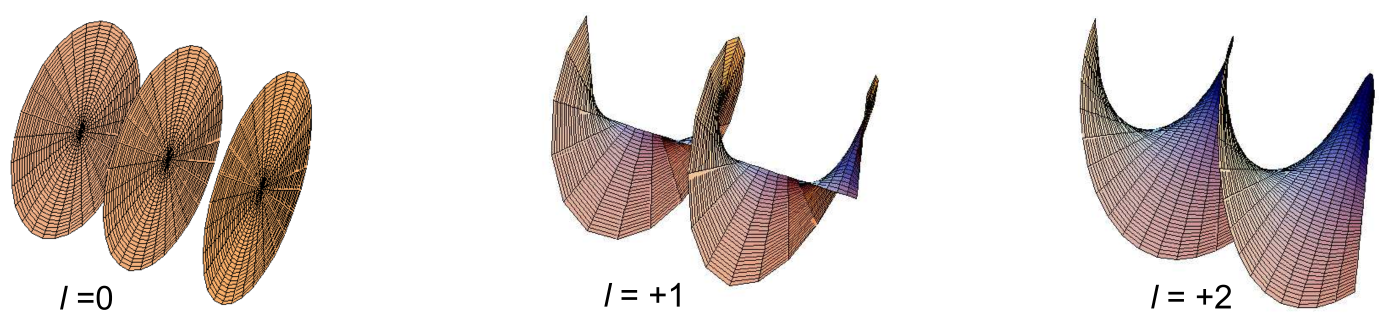
Another Example of Structured Light

Quantum Properties of Light Beams that Carry Orbital Angular Momentum (OAM)

What Are the OAM States of Light?

- Light can carry spin angular momentum (SAM) by means of its circular polarization.
- Light can also carry orbital angular momentum (OAM) by means of the phase winding of the optical wavefront.
- A well-known example are the Laguerre-Gauss modes. These modes contain a phase factor of $\exp(il\phi)$ and carry angular momentum of $l\hbar$ per photon. (Here ϕ is the azimuthal coordinate.)

Phase-front structure of some OAM states



See, for instance, A.M. Yao and M.J. Padgett, Advances in Photonics 3, 161 (2011)

Laguerre-Gauss Modes

The paraxial approximation to the Helmholtz equation $(\nabla^2 + k^2)E(\boldsymbol{k}) = 0$ gives the paraxial wave equation which is written in the cartesian coordinate system as

$$\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + 2ik\frac{\partial}{\partial z}\right) E(x, y, z) = 0. \tag{1}$$

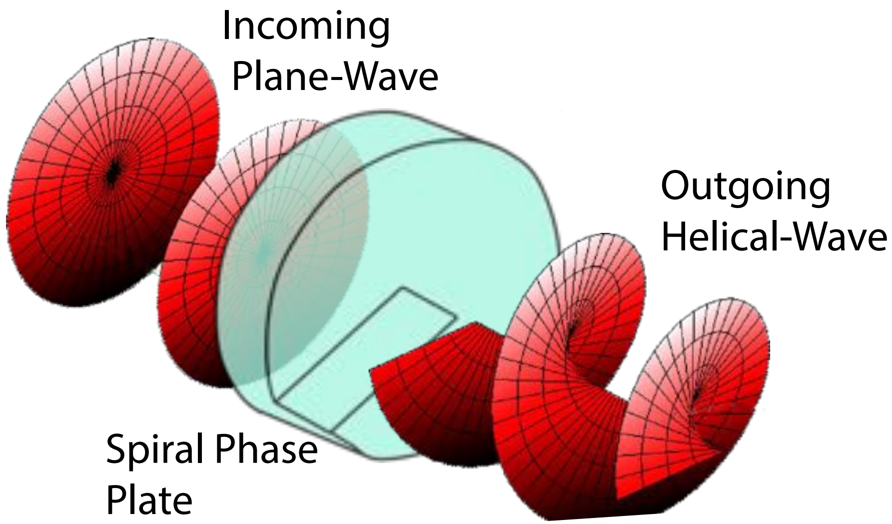
The paraxial wave equation is satisfied by the Laguerre-Gaussian modes, a family of orthogonal modes that have a well defined orbital angular momentum. The field amplitude $LG_p^l(\rho, \phi, z)$ of a normalized Laguerre-Gaussian modes is given by

$$\begin{aligned} LG_p^l(\rho, \phi, z) = &\sqrt{\frac{2p!}{\pi(|l|+p)!}}\frac{1}{w(z)}\left[\frac{\sqrt{2}\rho}{w(z)}\right]^{|l|} L_p^l\left[\frac{2\rho^2}{w^2(z)}\right] \\ &\times \exp\left[-\frac{\rho^2}{w^2(z)}\right]\exp\left[-\frac{ik^2\rho^2 z}{2(z^2+z_R^2)}\right]\exp\left[i(2p+|l|+1)\tan^{-1}\left(\frac{z}{z_R}\right)\right]e^{-il\phi}, \end{aligned} \tag{2}$$

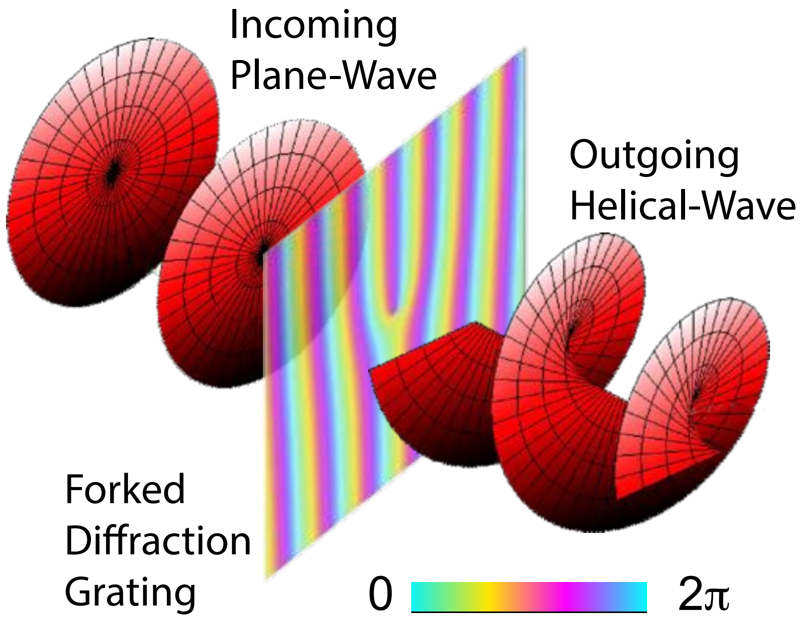
where k is the wave-vector magnitude of the field, z_R the Rayleigh range, $w(z)$ the radius of the beam at z, l is the azimuthal quantum number, and p is the radial quantum number. L_p^l is the associated Laguerre polynomial.

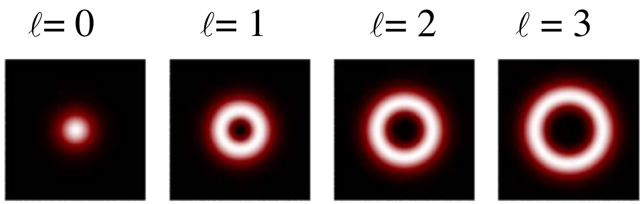
How to create a beam carrying orbital angular momentum?

- Pass beam through a spiral phase plate



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





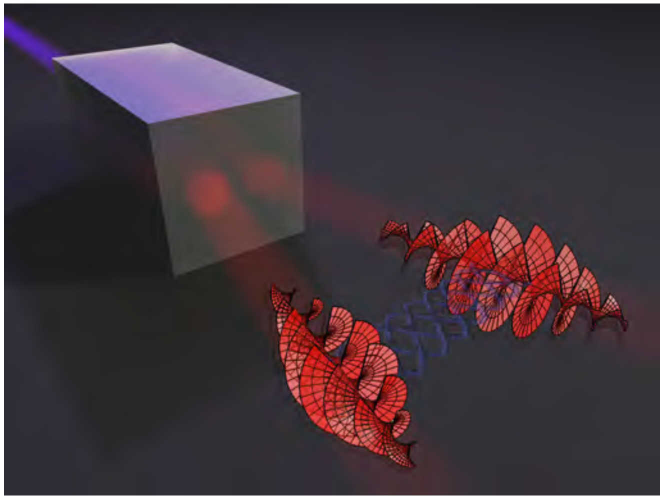
Exact solution to simultaneous intensity and phase masking with a single phase-only hologram, E. Bolduc, N. Bent, E. Santamato, E. Karimi, and R. W. Boyd, Optics Letters 38, 3546 (2013).

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.



QKD System Carrying Many Bits Per Photon

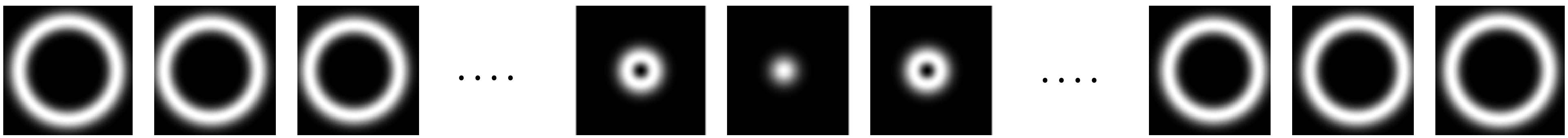
We are constructing a QKD system in which each photon carries many bits of information

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

As a diagnostic, we need to be able to measure the statevector of OAM states

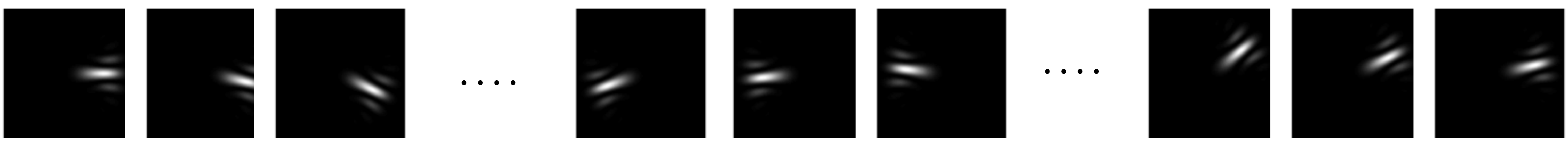
<u>Single Photon States</u>

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



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

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

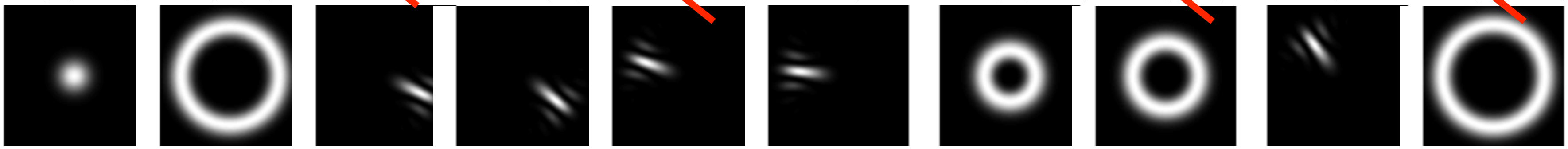


0 1 2 12 13 14 25 26 27

$$\Psi_{\mathrm{AB}}^{N} = \frac{1}{\sqrt{27}} \sum_{l=-13}^{13} \mathrm{LG}_{l,0} \exp\left(i2\pi N l/27\right)$$

Protocol

Alice	LG:13	LG:3	~~AB:2~~	AB:3	~~AB:15~~	AB:14	LG:16	~~LG:8~~	AB:24	~~LG:26~~



Bob	LG	LG	~~LG~~	AB	~~LG~~	AB	LG	~~AB~~	AB	~~AB~~
Result	13	3	~~15~~	3	~~15~~	14	16	~~17~~	24	~~10~~

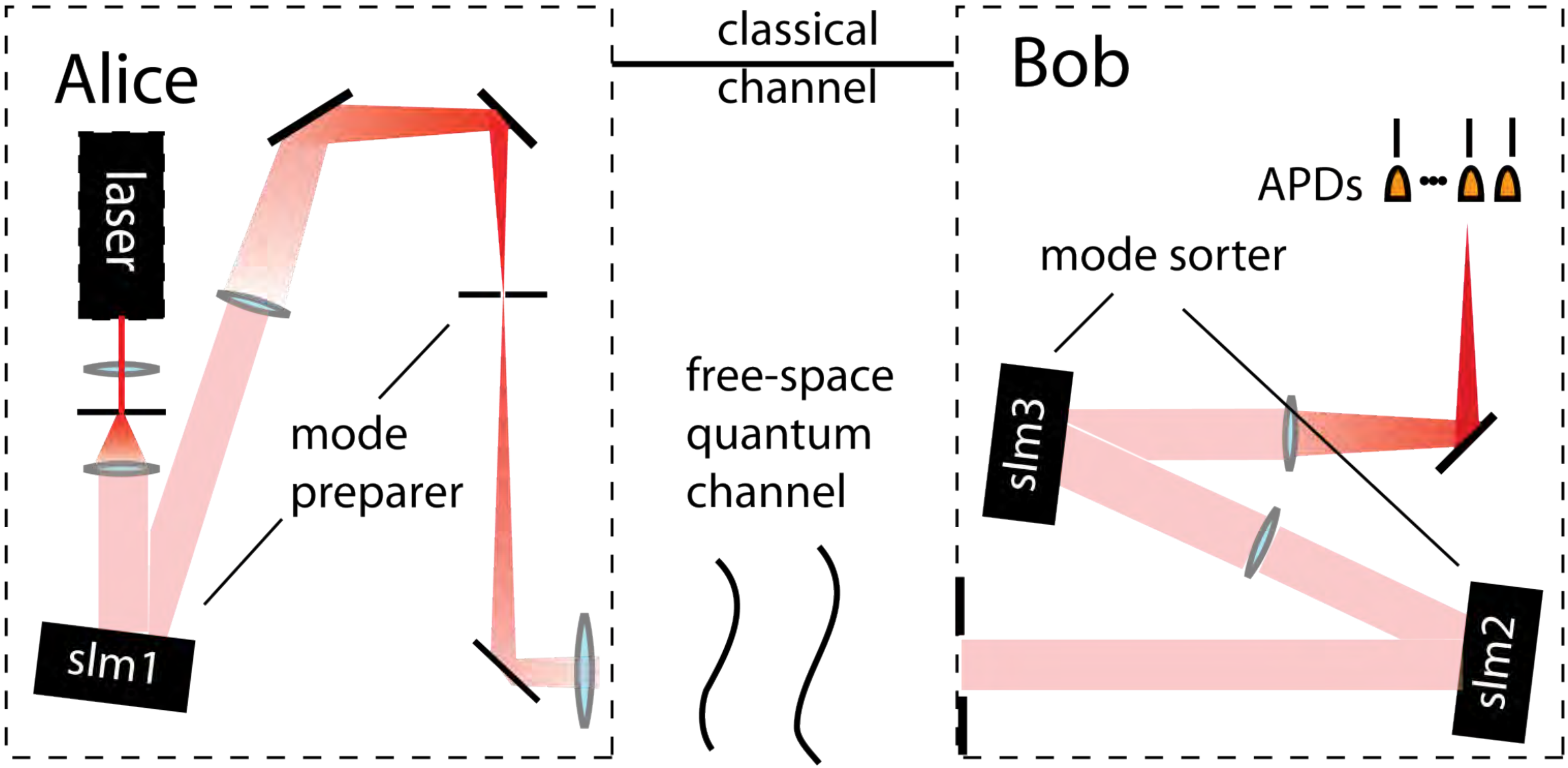
Sifted Key 13 3 3 14 16 24 … ← in principle contains no errors unless eavesdropper is present.

In any real system, Bob's key will have errors due to system imperfections.

1. Error Correction (Cascade Protocol)
2. Privacy Amplification

Under many conditions, these protocols can be successfully implemented if Alice/Bob share more bits of information than Alice and Eve.

Spatially Based QKD System



<u>Source</u>

Weak Coherent Light

Heralded Single Photon

<u>Protocol</u>

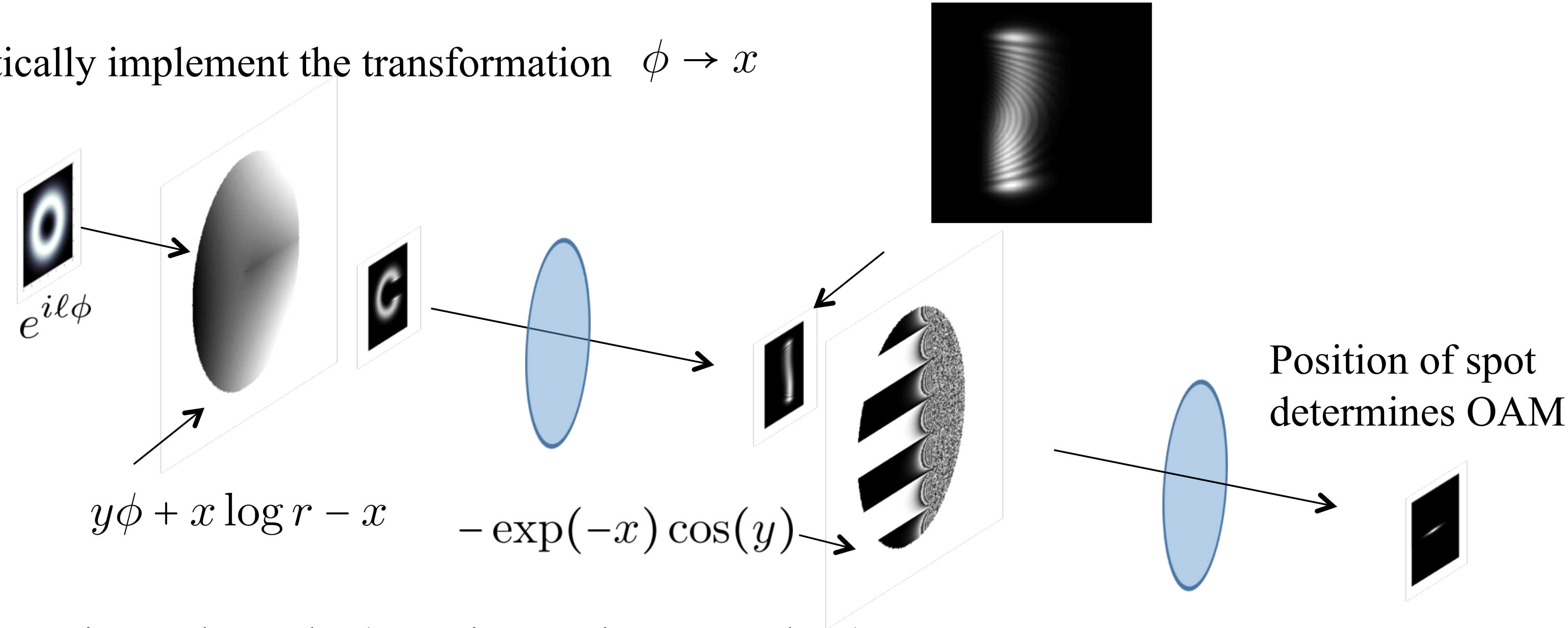
Modified BB84 as discussed

<u>Challenges</u>

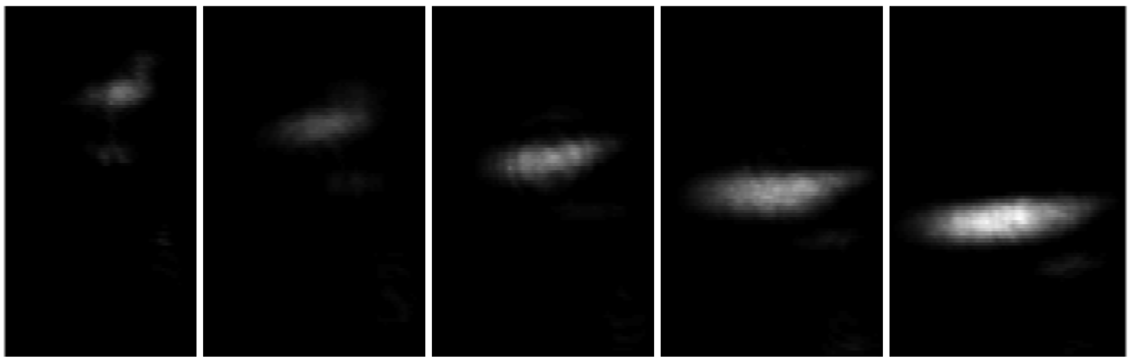
1. State Preparation
2. State Detection
3. Turbulence

Sorting OAM using Phase Unwrapping

Optically implement the transformation $\phi \rightarrow x$



Experimental Results (CCD images in output plane)



$\ell = 1$ $\ell = 2$ $\ell = 3$ $\ell = 4$ $\ell = 5$

-Can also sort angular position states.

-Limited by the overlap of neighboring states.

*Berkhout *et al.* *PRL* **105,** 153601 (2010).
O. Bryngdahl, *J. Opt. Soc. Am.* **64**, 1092 (1974).

UNIVERSITY of ROCHESTER

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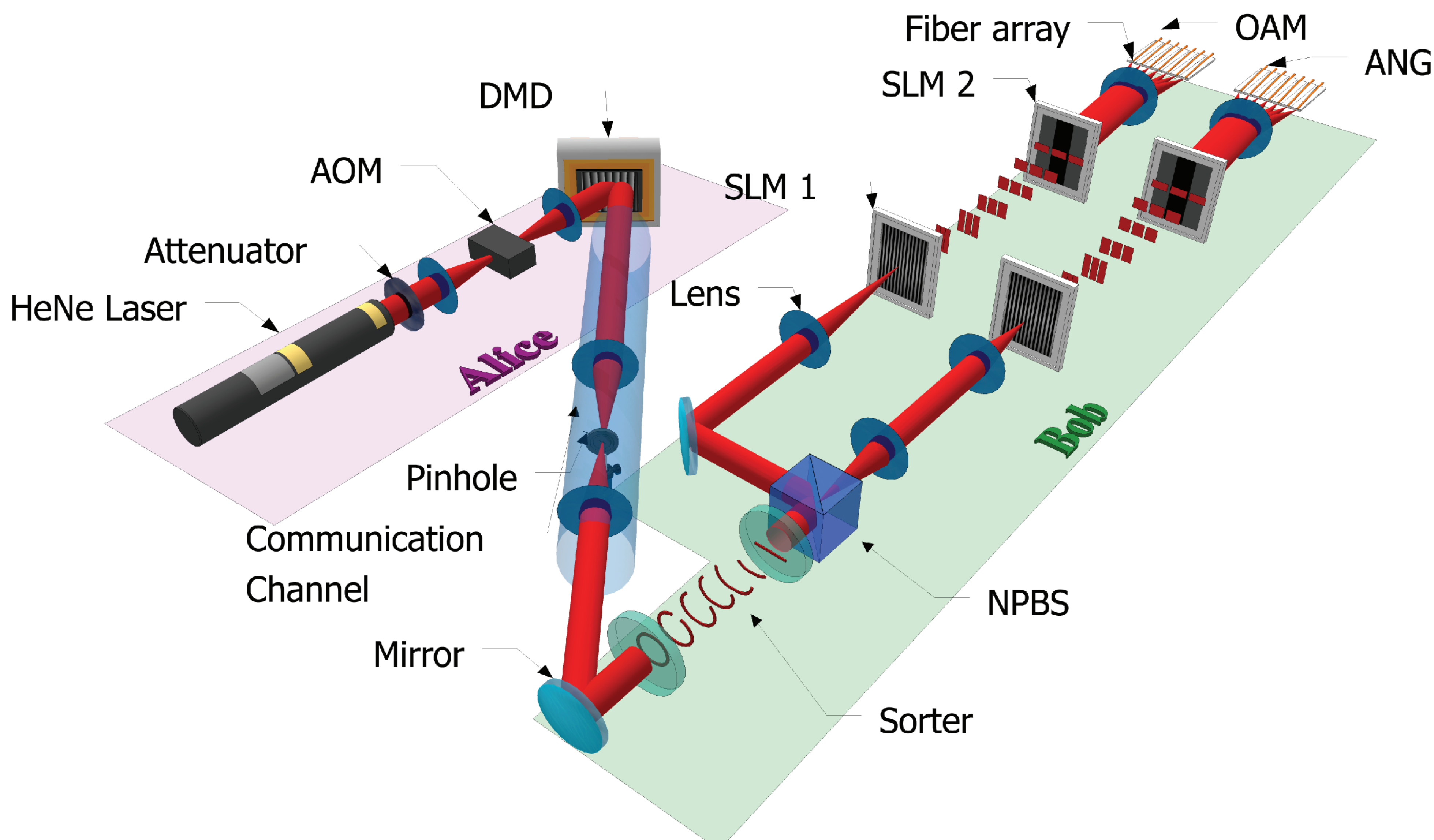
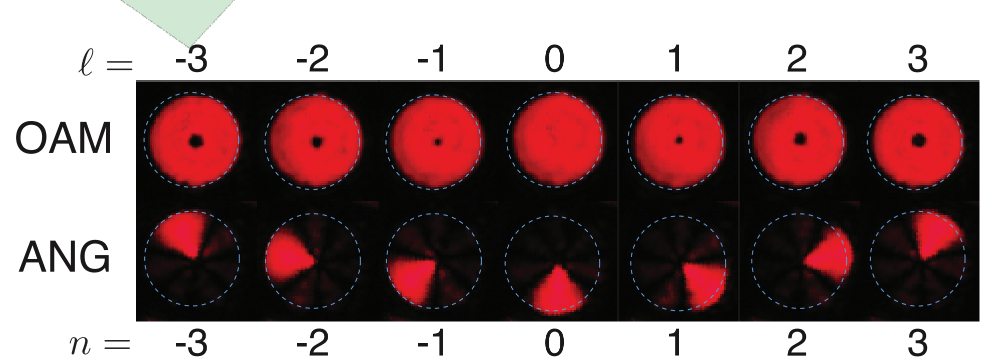
Our Laboratory Setup



We use a seven-dimensional state space



Mirhosseini, Magaña-Loaiza, O'Sullivan, Rodenburg, Malik, Lavery, Padgett, Gauthier and Boyd, New J. Phys. 17, 033033 (2015).

Laboratory Results - OAM-Based QKD

Alice	Bob
063133602132045444456141026645 545050363603025261643215524164 230146602513401613222451551026	063033601132021444456141026645 545022353603025261643215524164 230146602515403613422451551026

Error Correction

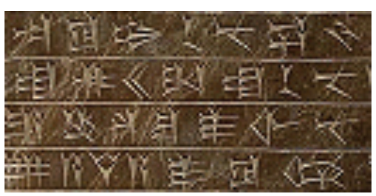
0000 0100 0111 0100 0101 0001 1100 0010 1100 1001 0010 0100 1101 0001 0000
0001 0011 0100 0000 0100 0010 0001 0001 0011 0000 0101 0010 0111 1000 0101
1101 0011 0001 1001 1011 0101 0000 0100 0111 0100 0011 0110 0101 0000 1101
0101 0010 1111 1010 1111 0010 1000

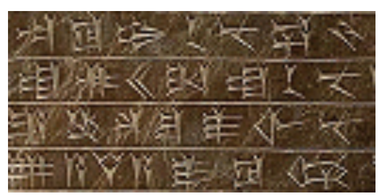
Privacy Amplification

0001 0100 1011 1111 1111 1010 1010 0111 1010 1011 1111 0001 0000 1101 1111
1110 1110 1101 1110 0001 0011 1000 0101 1101 1100 0011 0111 1101 0001 1001
0100 0110 1011 0000 0110 1010 1

Alice Encrypts

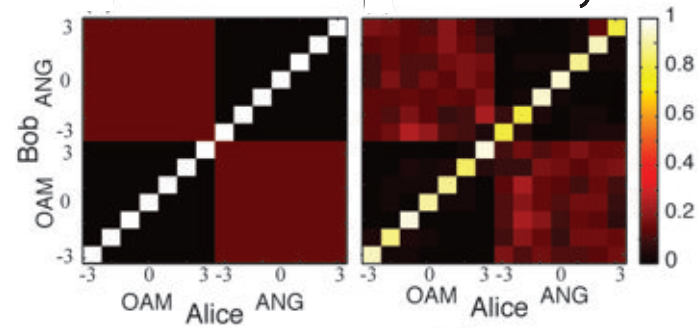
Bob Decrypts

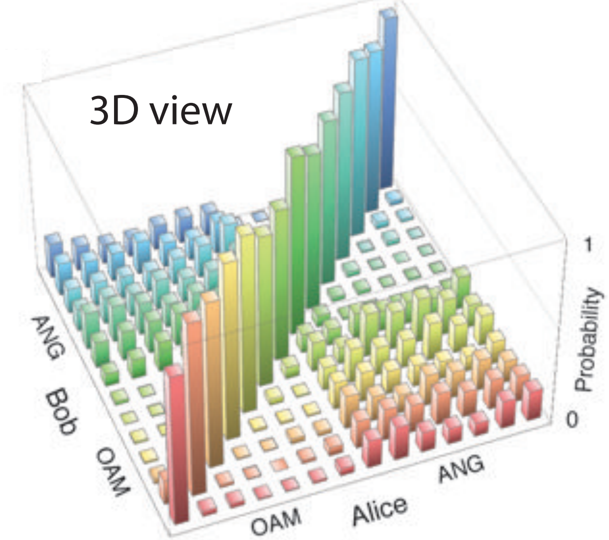




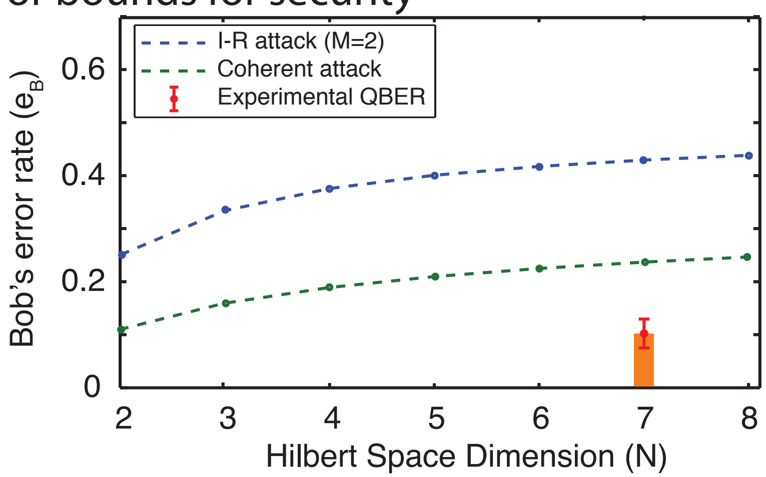
cross-talk matrices

ideal laboratory





- error bounds for security



We use a 7-letter alphabet, and achieve a channel capacity of 2.1 bits per sifted photon.

We do not reach the full 2.8 bits per photon for a variety of reasons, including dark counts in our detectors and cross-talk among channels resulting from imperfections in our sorter.

Nonetheless, our error rate is adequately low to provide full security,

ARTICLES
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nature photonics

Terabit free-space data transmission employing orbital angular momentum multiplexing

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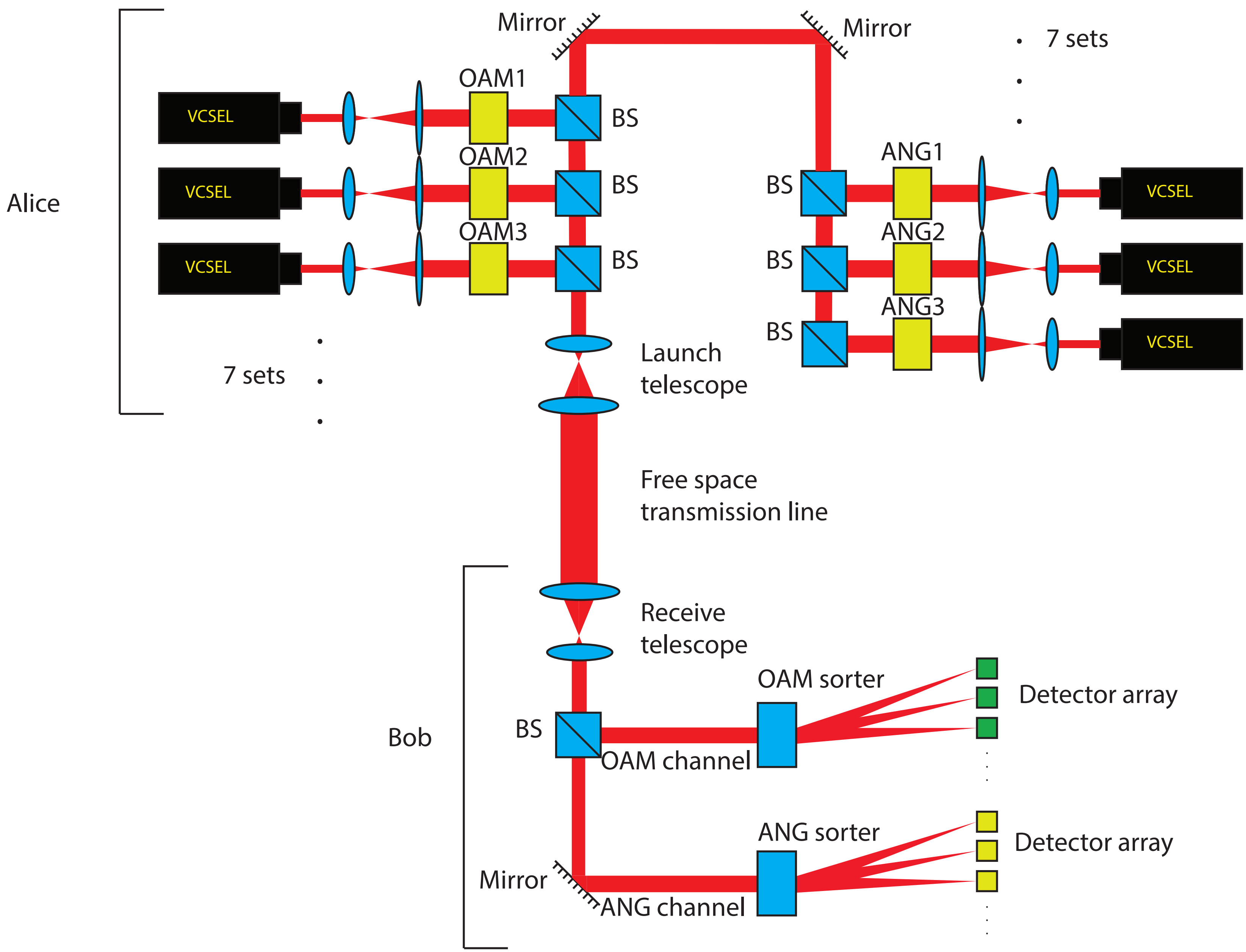
The recognition in the 1990s that light beams with a helical phase front have orbital angular momentum has benefited applications ranging from optical manipulation to quantum information processing. Recently, attention has been directed towards the opportunities for harnessing such beams in communications. Here, we demonstrate that four light beams with different values of orbital angular momentum and encoded with 42.8×4 Gbit s^{-1} quadrature amplitude modulation (16-QAM) signals can be multiplexed and demultiplexed, allowing a 1.37 Tbit s^{-1} aggregated rate and 25.6 bit s^{-1} Hz^{-1} spectral efficiency when combined with polarization multiplexing. Moreover, we show scalability in the spatial domain using two groups of concentric rings of eight polarization-multiplexed 20×4 Gbit s^{-1} 16-QAM-carrying orbital angular momentum beams, achieving a capacity of 2.56 Tbit s^{-1} and spectral efficiency of 95.7 bit s^{-1} Hz^{-1}. We also report data exchange between orbital angular momentum beams encoded with 100 Gbit s^{-1} differential quadrature phase-shift keying signals. These demonstrations suggest that orbital angular momentum could be a useful degree of freedom for increasing the capacity of free-space communications.

Next Step: gigabit-per-second OAM-based QKD system

- Use direct modulation of laser diode to encode at gigabits per sec.



Thank you for your attention!