Workshop on Quantum Light Generation, Detection, and Applications

Program & Abstracts

Organizers: Ralph Jimenez and Shuo Sun

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Date: July 17 - 19, 2024

Workshop on Quantum Light Generation, Application, and Detection

All events are in JILA X317/X325 unless otherwise noted.

Day 1: July 17, 2024 (Wednesday)

8:00 am – 9:00 am	Breakfast & Welcome

Session 1: Chip-integrated generation of quantum light 1

9:00 am – 9:30 am	Squeezed quantum microcombs with integrated photonics, Xu Yi, University of Virginia
9:30 am – 10:00 am	Quantum Frequency Conversion and N-Way Frequency Splitters, Alexander Gaeta, Columbia University
10:00 am – 10:30 am	Quantum light sources on silicon nitride PICs: Bulk nonlinearity, heterogeneously integrated quantum dots, and vapor-phase atoms, Kartik Srinivasan, NIST Gaithersburg & University of Maryland College Park
10:30 am – 11:00 am	Coffee Break

Session 2: Generation of squeezed light via Four Wave Mixing

11:00 am – 11:30 am	Beatitudes and woes of atom-based quantum light, Irina Novikova, William and Mary
11:30 am – 12:00 pm	Generation, engineering, and storage of broadband photonic quantum states, Virginia O. Lorenz, University of Illinois at Urbana-Champaign
12:00 pm – 1:30 pm	Group Photo & Lunch

Session 3: Quantum metrology with light

1:30 pm – 2:00 pm	Superconducting detectors for quantum light sensing, Martin J. Stevens, NIST Boulder
2:00 pm – 2:30 pm	Quantum sensing: from Applications to Scientific Discovery, Alberto M. Marino, Oak Ridge National Laboratory & University of Oklahoma
2:30 pm – 3:00 pm	Time reversed quantum metrology, G. S. Agarwal, Texas A&M University
3:00 pm – 3:30 pm	Coffee Break

Poster Sessions

3:30 pm – 5:30 pm	Poster sessions (the list of poster titles is listed at the end of this document)

Day 2: July 18, 2024 (Thursday)

8:00 am – 9:00 am	Breakfast

Session 1: Chip-integrated generation of quantum light 2

9:00 am – 9:30 am	Ultrafast quantum nanophotonic circuits, Alireza Marandi, Caltech
9:30 am – 9:50 am	Single-mode squeezed-light generation and tomography with an integrated optical parametric oscillator, Taewon Park, Stanford University
9:50am – 10:20 am	Integrated photonics for quantum sensing and manipulating light, Scott Papp, NIST
10:20 am – 10:50 am	Coffee Break

Session 2: Generation of squeezed light in a frequency comb

10:50 am – 11:20 am	Generation, control, and measurement of squeezed light across >400 frequency modes, Peter McMahon, Cornell University
11:20 am – 11:50 am	Photonic quadrature lattices, Eran Lustig, Stanford University
11:50 am – 12:20 pm	The Fellowship of the Ring, John E. Sipe, University of Toronto
12:20 pm – 1:30 pm	Lunch

Session 3: Quantum spectroscopy of complex systems

1:30 pm – 2:00 pm	The 3DQ Microscope: A novel system using entangled photons to generate volumetric fluorescence and scattering images for bioenergy applications, Audrey Eshun, Lawrence Livermore National Lab
2:00 pm – 2:30 pm	Photon number measurements for bio-applications, Sergey Polyakov, NIST Gaithersburg
2:30 pm – 3:00 pm	Scattered biphotonic state by a two-photon absorbing medium, Samuel Corona-Aquino, Universidad Nacional Autonoma de Mexico
3:00 pm – 3:30 pm	Coffee Break

Free Discussion & Lab Tours

3:30 pm – 5:30 pm	Free discussions & JILA Lab Tours

Workshop Reception

6:00 pm – 9:00 pm	Reception

Day 3: July 19, 2024 (Friday)

8:00 am – 9:00 am	Breakfast

Session 1: Entangled two-photon absorption and spectroscopy 1

9:00 am – 9:30 am	Role of time-frequency correlations in entangled-two-photon resonance energy transfer, Roberto de J. Leon-Montiel, Universidad Nacional Autonoma de Mexico
9:30 am – 10:00 am	Limitations in fluorescence-detected entangled two-photon-absorption experiments: exploring the low- to high-gain squeezing regimes, Michael G. Raymer, University of Oregon
10:00 am – 10:30 am	Coffee Break

Session 2: Entangled two-photon absorption and spectroscopy 2

10:30 am – 10:50 am	Quantifying E2PA: ultracold rubidium in a magneto-optical trap, Alan Mclean, JILA/NIST Boulder
10:50 am – 11:10 am	Fluorescence by nonclassical light: getting it right, C. Drago, University of Toronto
11:10 am – 12:10 pm	Roundtable discussions
12:10 pm	Departure

Poster Presentations:

Keegan Finger, JILA and University of Colorado Boulder, Pulse characterization via two-photon auto- and cross-correlation signals

Daniel I. Herman, University of Colorado Boulder, Soliton squeezing in polarization-maintaining nonlinear fiber with a 1 GHz laser comb

Zhenfei Jiang, Texas A&M University, Experimental demonstration of recovering quantum mutual information from scattering

Gautam A. Kavuri, University of Colorado Boulder, Publicly certified random numbers using entangled photons

Katherine Koch, Wake Forest University, Exciton-lattice interactions in two-dimensional Ruddlesden Popper metal halides and the role of organic cation substitution

Evan Kumar, Wake Forest University, Estimation of spectral mode distribution of SPDC photons for applications in quantum spectroscopy

Neelesh Kumar Vij, University of Maryland College Park, Inverse design of photonic crystal cavities

Miles San Soucie, University of Colorado Boulder, Inducing two-photon absorption in fluorescent molecles with intensity-difference squeezed light

Nanako Shitara, University of Colorado Boulder, Decoherence of a qubit interacting with a complex spin bath

Zach Wiethorn, University of Colorado Boulder, Symmetry breaking fluctuations split the porphyrin Q bands

Ziqi Niu, William and Mary, Bi-chromatic intensity squeezing at telecom wavelength using four-wave mixing in Rb vapor

Xu Yi, University of Virginia

Title: Squeezed quantum microcombs with integrated photonics

Squeezed light is having broad applications in science and technology, ranging from quantum-enhanced sensing to demonstrating quantum advantage. Recently, there has been increasing interest in photonic continuous-variable-based quantum computing (CVQC), which demands a large number of squeezed quantum modes (qumodes) generated with scalable optical platforms. While scalable generation of squeezing is actively pursued with integrated photonic circuits (IPCs), further combination of squeezing and microresonator-based frequency combs (microcombs) introduces an additional dimension for scalability through frequency multiplexing.

In this talk, I would introduce our recent results in generating squeezed quantum microcombs on photonic chips. Unconditional entanglement can be created by the broadband Kerr parametric gain in the microresonator and verified through two-mode squeezing measurements. In the first experiment, we generated 40 continuous variable quantum modes, in the form of 20 simultaneously two-mode squeezed comb pairs, in a silica wedge resonator on a silicon chip. Raw squeezing of 2.1 dB is measured [1,2]. In the second experiment, we generate a squeezed quantum microcomb with 70 qumodes from CMOS-compatible silicon nitride (SiN) [3]. Our demonstration offers the possibility to leverage deterministically generated, frequency multiplexed quantum states and integrated photonics to open up new avenues in fields of spectroscopy, quantum metrology, and scalable, continuous-variable-based quantum information processing.

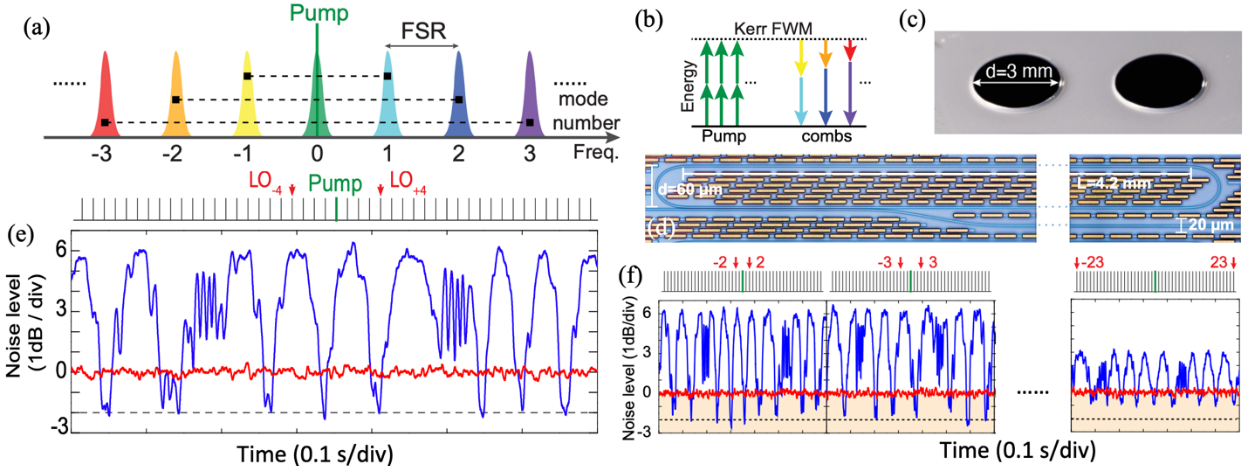

Figure 1 (a) Conceptual illustration of squeezed quantum microcombs. Dash lines represents EPR entanglement. (b) Illustration of Kerr four-wave mixing (FWM). (c) Picture of silica resonator on a silicon chip. (d) Integrated SiN resonator. (e) Measured 2.1 dB two-mode squeezing of comb pair (-4,4). (f) Measured two-mode squeezing of comb pair (-2,2), (-3,3) and (-23,23).

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Quantum light sources on silicon nitride PICs: bulk nonlinearity, heterogeneously-integrated quantum dots, and vapor-phase atoms

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Silicon nitride, with its wide optical transparency window, relatively high refractive index, low optical loss, and silicon photonics compatibility, is one of the most heavily investigated photonic integrated circuit (PIC) platforms. In this talk, I will describe three approaches for quantum light generation in this platform and will contrast their relative merits and challenges. The first approach utilizes the bulk third-order nonlinearity of the material, which we harness within dispersion-engineered microresonators to realize entangled photon-pair sources [1] whose output colors are highly non-degenerate and can be widely and accurately controlled [2], but are fundamentally probabilistic in nature. The second approach uses heterogeneously-integrated III-V epitaxial quantum dots [3], [4], whose short radiative lifetime and on-demand nature are advantageous, but whose decoherence mechanisms and inhomogeneous broadening necessitate special consideration. The final approach integrates alkali atomic vapors with silicon nitride photonics [5] and provides a path towards streams of controllable and indistinguishable photons, though important challenges associated with the atoms' limited transit time must be overcome.

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Beatitudes and woes of atom-based quantum light

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Atomic platforms occupy an interesting position in the rapidly developing landscape of quantum technologies. On one hand, decades of their applications for precision measurements provide scientists exquisite understanding of light-atom interactions and quantum control. On the other hand, compare to solid-state nonlinear optics and integrated photonics, atoms bring a slew of challenges with, e.g., miniaturization, speed and operation bandwidth. Here we will discuss two experiments using Rb vapor for generation of squeezed light, and discuss their pros and cons.

Thanks to strong resonant enhancement, nonlinear optical processes in cold and how atoms have been extensively explored for generation of non-classical light fields. For example, Four-Wave Mixing (FWM) in ERb vapor has been used for various applications such as quantum imaging and sensing [1]. However, the necessity to operate near atomic transition frequency can cause problems. For example, for any optical signal that has to travel through optical fibers, it is advantageous to be in the telecom wavelength range (1300-1500 nm). This is far fro near-IR optical frequencies, required to efficiently couple to atoms in the ground state for both atoic sensor and quantum information realizations. A FWM in a double-ladder scheme in Rb atoms aimed to generate either correlated photon pairs [3–5] or, potentially, intensity squeezed light [2] may offer a solution. Here we report our recent results for such two-mode two-color intensity squeezing generation. We use two strong pump lasers couple to the transitions $5S_{1/2}F = 3 \rightarrow 5P_{3/2}$ (wavelength: 780nm) and $5P_{3/2} \rightarrow 6S_{1/2}$ (wavelength: 1367nm). A weak probe field applied to $5S_{1/2}F = 3 \rightarrow 5P_{1/2}$ (wavelength: 795nm) serves as the probe seed, that gets amplified by the FWM process and produces a Stokes field at 1324 nm. By optimizing the experimental parameters, we achieved bi-chromatic FWM amplification gain exceeding 2 with relatively low resonant absorption for the probe field. We also observe reduction in differental intensity noise for stokes and probe signals below the shot-noise level by the amount consistent with FWM gain.

In parallel, we investigating the spatial structure of atom-generated quantum fields. The lack of cavity allows for multimode operation, vital for efficient quantum imaging. On the other hand, presence of multiple spatial modes can be detrimental for realization of quantum advantage. To get better understanding of the generated quantum noise profiles, we are developing a protocol for spatial mode analysis for optical fields with near-zero mean photon counts, such as squeezed vacuum. The approach is the extension of the quadrature noise shadow imaging approach [6, 7].

This work was supported by the Department of Energy (Grant No. DE-SC0022069).

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Generation, engineering, and storage of broadband photonic quantum states

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This talk will cover the progress our group has been involved in regarding photonic quantum sources, photonic entanglement, and broadband photonic quantum state storage and retrieval. These efforts have opened up interesting new avenues for studies of coherent control, multidimensional entanglement, and optical and material engineering of quantum properties.

Optical fiber provides a means to generate photon pairs that are naturally mode-matched to optical fiber networks. The fiber core's third-order nonlinear optical susceptibility supports annihilation of two pump photons and creation of signal and idler photon pairs in spontaneous four-wave mixing (SFWM). The detection of one photon can be used to indicate the existence of the other, which can then be used in quantum protocols. Using two pumps instead of just one in the SFWM process opens up capabilities for optical engineering of joint correlations [1]. The tight confinement in the fiber core leads to high brightness, enabling demonstrations of tripartite entanglement [2]. The cylindrical geometry supports discrete transverse modes, which can be utilized to create scalable, multidimensional entanglement [3]. By applying stimulated emission tomography, the properties of the created quantum states can be efficiently characterized and engineered [4]. The broadband photons produced by SFWM and spontaneous parametric down-conversion lack correspondingly broadband, efficient, matter-based quantum memories. Experimentally, we implement near-off-resonant broadband quantum memory in an atomic barium vapor [5]. We show that collisional broadening can serve as a resource to achieve storage efficiencies over 90%. We also explore theoretically and experimentally new materials and methods for broadband photonic quantum state generation with built-in memory capability through Raman scattering, shedding light on photon-matter quantum correlations [6].

This work was supported in part by NSF Grant Nos. 1640968, 1806572, 1839177, 1936321, 2207822, and the U.S. Department of Energy, Office of Science, Biological and Environmental Research program, under Award Number DE-SC0023167.

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Superconducting detectors for quantum light sensing

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Over the past 20 years, superconducting single-photon detectors have emerged as the premier devices for measuring single photons and entangled photon pairs. I will review two devices: the superconducting nanowire single-photon detector (SNSPD) and the transition edge sensor (TES). The TES is the highest-performing photon number-resolving detector, capable of distinguishing photon number with unmatched fidelity [1]. SNSPDs offer the highest count rates (>1 Gcps [2]) and lowest timing jitter (<3 ps [3]) of any single-photon detector. Both detectors are sensitive over a broad wavelength range, spanning from the UV to the long-wave infrared. Although superconducting detectors must be operated in cryogenic environments, recent progress in cryogenics have made the task easier. This talk will cover the pros and cons of each superconducting detector, illustrated with examples where they have been used to detect quantum light.

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Quantum Sensing: from Applications to Scientific Discovery

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Quantum metrology takes advantage of quantum correlations to enhance the sensitivity of sensors and measurement techniques beyond what is possible with classical resources. For optical based sensors, the temporal quantum correlations present in quantum states of light make it possible to enhance the sensitivity of devices beyond their fundamental classical shot noise limit to enable the detection of signals that would otherwise be hidden in the noise. Such enhanced capabilities will be critical for applications that range from biochemistry to high-energy physics [1]. In addition to these temporal correlations, quantum states of light can also exhibit spatial quantum correlations, which in combination with the temporal correlations can enable novel parallel sensing configurations capable of simultaneously and independently probing an array of sensors while enabling a quantum enhancement for all sensors. Such a spatially resolved quantum sensing approach makes it possible to better take advantage of the available quantum resources to perform faster and more efficient measurements by simultaneously estimating multiple parameters or the same parameter multiple times.

To illustrate the capabilities of these novel sensing approaches, we extend our previous work on quantum-enhanced plasmonic sensing [4] to implement a parallel quantum-sensing configuration. To this end, we leverage two-mode squeezed states, or twin beams, generated through a four-wave mixing process based on a double-Λ configuration in a ^{85}Rb vapor cell, as they have been shown to contain quantum correlations in both the temporal and spatial degrees of freedom [2, 3]. We use these multi-spatial mode twin beams to simultaneously probe an array of four independent plasmonic sensors fabricated in a single plasmonic structure. We show that it is possible to independently and simultaneously measure local changes in refractive index for all four sensors beyond the shot noise limit [5].

A focus on plasmonic sensors points to the applicability of quantum sensing techniques for practical applications, including sub-shot-noise quantum biosensing and chemical detection, and their extension to quantum-enhanced parallel configurations. Beyond plasmonic sensing, these results provide a first step towards applications that require a network of quantum sensors or imaging capabilities, such as the spatial characterization of domain walls in quantum materials or the detection of dark matter through networked quantum sensors.

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Time Reversed Quantum Metrology-G. S. Agarwal*, Texas A&M University

It is well recognized that quantum physics can be used to build better sensors. Such sensors can be for parameters that correspond to the unitary evolution of the system, like phases, forces, fields or for parameters like absorption, scattering that require description in terms open system dynamics. The framework of the quantum Fisher information enables one to obtain best estimates of the parameters and then one can design experiments that can reach Cramer- Rao bounds. I would highlight the importance of the quantum states used as probes, and the importance of the quantum-ness of the measurement schemes. It turns out that in many cases the schemes based on time reversed metrology saturate Cramer-Rao bounds. I would discuss the importance of squeezed states of bosonic systems like photons, ions and cat states of qubits for metrological applications. I would present results on quantum advantage in the determination of phases, displacement and absorption and scattering parameters [1-3].

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Ultrafast Quantum Nanophotonic Circuits

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Despite tremendous progress on quantum optics using table-top experiments, in nanophotonics, it remains challenging to generate, manipulate, and measure quantum states with the performance required for a wide range of quantum information systems. Utilizing few-optical-cycle pulses in lithium niobate nanophotonics not only promises a path for addressing this challenge it also enables realization of ultrafast quantum information processors with THz clock rates. In this talk, I will present the experimental progress towards such processors including the recent results on generation and measurement of squeezed states on a single chip with about 5 dB of squeezing and spanning more than 25 THz of bandwidth [1]. I will also overview other important components in nanophotonic LN including intense optical parametric amplifiers [2], ultrafast ultra-low-energy all-optical switches [3], and ultrafast mode-locked lasers [4]. I will discuss the ongoing efforts and the outlook towards generation of non-Gaussian states and large-scale quantum nanophotonic circuits.

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Single-mode squeezed-light generation and tomography with an integrated optical parametric oscillator

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Quantum optical technologies promise advances in sensing, computing, and communication. A key resource is squeezed light, where quantum noise is redistributed between optical quadratures. We introduce a monolithic, chip-scale platform that exploits the $\chi^{(2)}$ nonlinearity of a thin-film lithium niobate (TFLN) resonator device to efficiently generate squeezed states of light. Our system integrates all essential components—except for the laser and two detectors—on a single chip with an area of one square centimeter, significantly reducing the size, operational complexity, and power consumption associated with conventional setups. Our work addresses challenges that have limited previous integrated nonlinear photonic implementations that rely on either $\chi^{(3)}$ nonlinear resonators or on integrated waveguide $\chi^{(2)}$ parametric amplifiers. Using the balanced homodyne measurement subsystem that we implemented on the same chip, we measure a squeezing of 0.55 dB and an anti-squeezing of 1.55 dB. We use 20 mW of input power to generate the parametric oscillator pump field by employing second harmonic generation on the same chip. Our work represents a substantial step toward compact and efficient quantum optical systems posed to leverage the rapid advances in integrated nonlinear and quantum photonics [1].

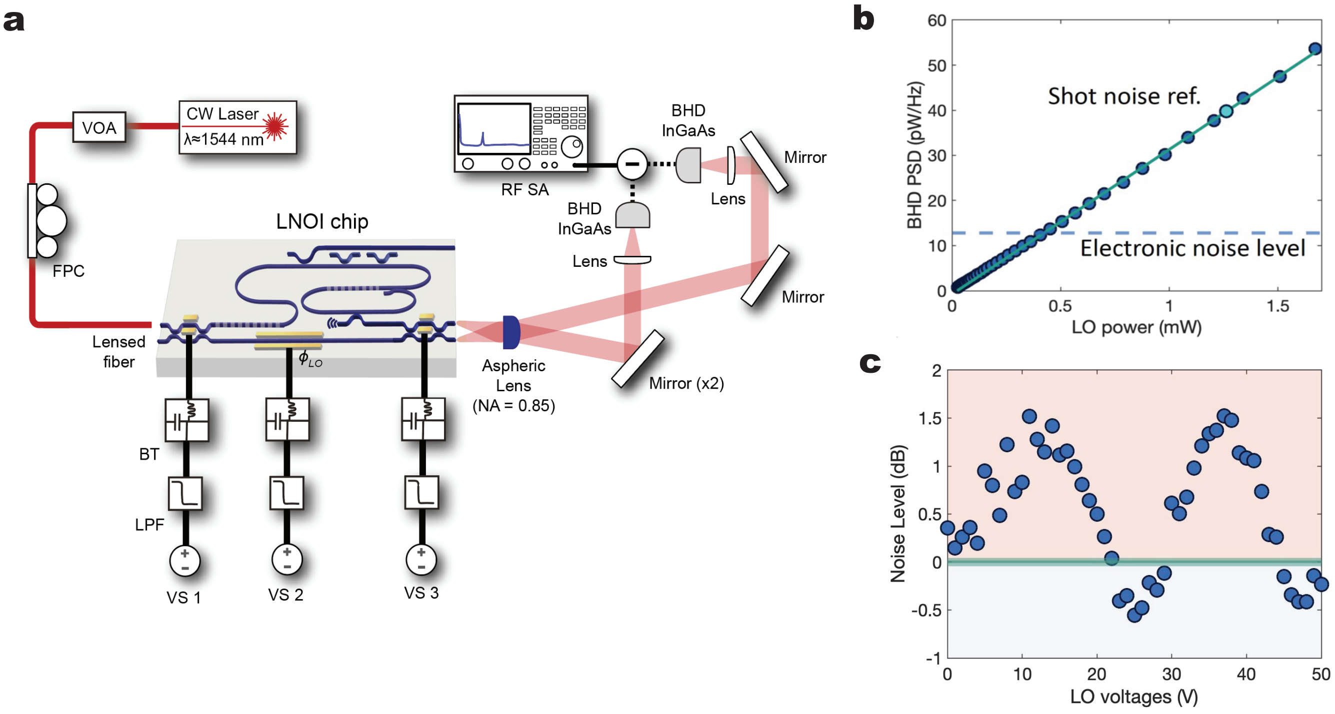

Fig. 1: **Squeezing measurement setup and results. (a)** Squeezing measurement setup. Continuous wave light at 1544 nm from the low-noise laser is coupled into the chip using a lensed fiber. We control the input power going into the chip using a variable optical attenuator (VOA) and adjust the polarization using a fiber polarization controller (FPC). Three voltage sources are employed to control the phase of the light where we use low pass filter (LPF) and bias tee (BT) to suppress non-DC signals going into the electrodes. We use high numerical aperture (NA) aspheric lens to efficiently collect light from the two output waveguides. The two beams are routed and focused onto the InGaAs photodetectors of the balanced homodyne detector respectively. We measure the power spectral density (PSD) of the difference between the two photocurrents using the radio frequency spectrum analyzer (RFSA). **(b)** Measured PSD on the RFSA (electronic noise subtracted) averaged from 58 MHz to 60 MHz vs. LO power. **(c)** Measured PSD on the RFSA (electronic noise subtracted) averaged from 58 MHz to 60 MHz vs. DC voltage applied to the local oscillator (LO) phase shifter at the TEC temperature of 43.4 °C. On-chip fundamental harmonic power going into the SHG section was 20 mW.

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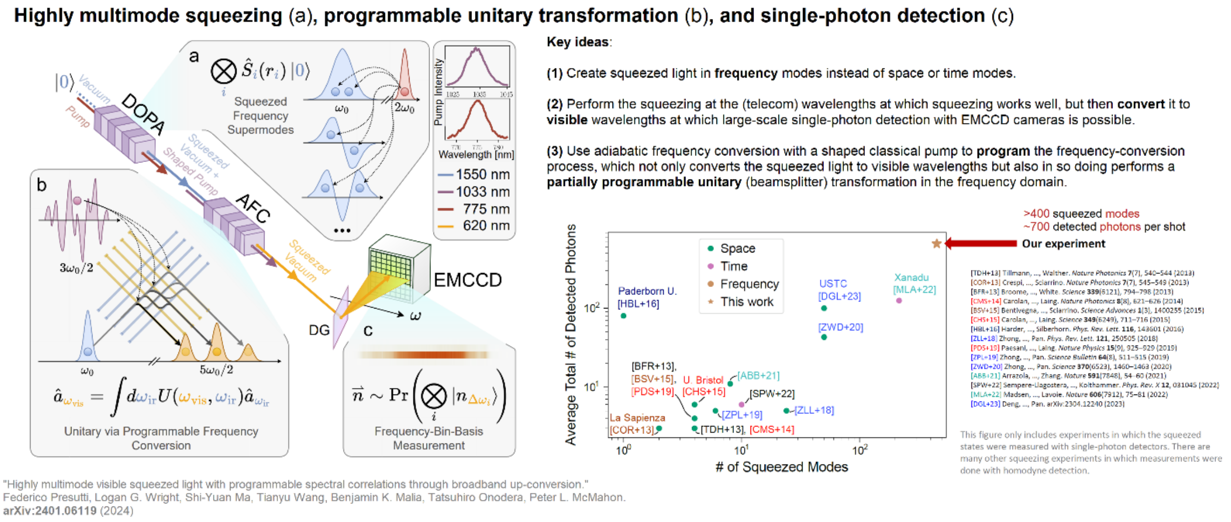
Generation, control, and measurement of squeezed light across >400 frequency modes

Peter McMahon
Cornell University

In this talk I will discuss our recent work [1] on making a controllable source of pulsed squeezed light that spans >400 frequency modes and produces an average of ~700 photons per pulse.



Highly multimode squeezing (a), **programmable unitary transformation** (b), **and single-photon detection** (c)

Key ideas:

(1) Create squeezed light in **frequency** modes instead of space or time modes.

(2) Perform the squeezing at the (telecom) wavelengths at which squeezing works well, but **then convert** it to **visible** wavelengths at which large-scale single-photon detection with EMCCD cameras is possible.

(3) Use adiabatic frequency conversion with a shaped classical pump to **program the** frequency-conversion process, which not only converts the squeezed light to visible wavelengths but also in so doing performs a **partially programmable unitary** (beamsplitter) transformation in the frequency domain.

>400 squeezed modes
~700 detected photons per shot
Our experiment

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[DGL+23] Deng, ..., Pan. arXiv:2304.12240 (2023)

This figure only includes experiments in which the squeezed states were measured with single-photon detectors. There are many other squeezing experiments in which measurements were done with homodyne detection.

"Highly multimode visible squeezed light with programmable spectral correlations through broadband up-conversion."
Federico Presutti, Logan G. Wright, Shi-Yuan Ma, Tianyu Wang, Benjamin K. Malia, Tatsuhiro Onodera, Peter L. McMahon.
arXiv:2401.06119 (2024)

[1] Federico Presutti, Logan G. Wright, Shi-Yuan Ma, Tianyu Wang, Benjamin K. Malia, Tatsuhiro Onodera, Peter L. McMahon. “Highly multimode visible squeezed light with programmable spectral correlations through broadband up-conversion.” arXiv:2401.06119 (2024)

Photonic Quadrature Lattices

Eran Lustig†, Melissa Guidry†, Daniil Lukin, Shanhui Fan and Jelena Vučković
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†These authors contributed equally to this work

Abstract: We experimentally study photonic quadrature lattices which offer new avenues in shaping the quantum properties of multi-mode light for the first time. We demonstrate the relation between lattice symmetries and anti-squeezing frequencies in subthreshold Kerr combs.

In recent years there is a growing interest in quadrature lattices which are coupled arrays of squeezed field quadratures **[1,2]**. Nonlinear resonators are suitable for studying these lattices due to the multimode pair-generation processes in $\boldsymbol{\chi^{(2)}}$ and $\boldsymbol{\chi^{(3)}}$ materials **[3–5]**. Coupling neighboring modes either by electro-optic modulation **[6]** or by the native Bragg scattering in $\boldsymbol{\chi^{(3)}}$ **[7]** near the optical parametric oscillation (OPO) threshold **[8]** generates these lattices. The quadrature lattice dynamics are described in the framework of non-Hermitian lattices relating point gap topology and non-Hermitian lattice symmetries to multimode squeezing and entanglement **[1,9]**.

Here we study theoretically and experimentally photonic quadrature lattices for the first time. In this work we focus on the anti-squeezing properties of Parity-Time(PT) Symmetric and broken PT lattice states generated by Kerr combs. Our combs are narrow-band 2-FSR Kerr combs in 4H-Silicon Carbide micro-ring resonators pumped by CW light. They form as 2-FSR narrow-band combs due to a mode splitting tuned by on-chip heaters. Each lattice element is composed of 4-quadratures $(\boldsymbol{q_\mu, p_\mu, q_{-\mu}, p_{-\mu}})$ where $\boldsymbol{\mu}$ is an odd number (pump is $\boldsymbol{\mu = 0}$). The elements are connected to form a lattice by Bragg scattering and acquire parametric gain and loss from the above threshold comb (Fig.1a). The lattice modes form robust PT symmetric states that are broken by violation of the parity symmetry around $\boldsymbol{\mu = 0}$ tuned by the mode crossing (Fig.1b). We utilize multimode balanced homodyne detection with controlled phase shifting to extract the quadrature's anti-squeezing directly, showing that both type of lattice modes simultaneously exist (Fig.1c). We furthermore study the threshold crossings of both types of lattice modes (Fig.1d-e), as well as their photon statistics properties and correlations (Fig.1f).

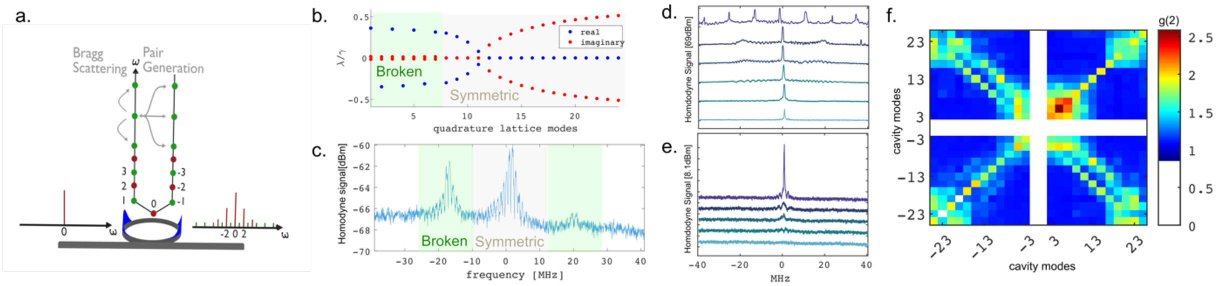

*Figure 1: **a.** Schematics of the quadrature lattice induced by a 2-FSR Kerr comb **b.** Multi-mode eigen-value (normalized with loss γ) spectrum with broken and symmetric lattice modes co-existing. **c.** Multimode (-,1,1) Homodyne reveling the antisqueezing of both broken PT and PT symmetric quadrature lattice modes, oscillations are due to controlled phase shifting. **d-e.** Threshold crossing of a PT symmetric quadrature lattice mode followed by a broken (detuned) threshold. **f.** g(2) of the below threshold lattice.*

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The Fellowship of the Ring

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Department of Physics
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Microring resonators and structures made of them have a host of applications in linear and nonlinear optics. They are also the focus of a body of work on nonlinear quantum optics, where the goal is the generation of pairs of entangled photons, squeezed states, and even more general quantum states. We review some of this work, with an emphasis on the careful treatment of both the properties of the structures and the proper calculation of the quantum states of light that can be generated.

Audrey Eshun, Lawrence Livermore National Lab

The 3DQ Microscope: A Novel System Using Entangled Photons to Generate Volumetric Fluorescence and Scattering Images for Bioenergy Applications

In the study of biological systems, real time 3D microscopy is an important tool in understanding how live cells move and interact with external elements. While these dynamics can currently be studied with confocal, light sheet, deconvolution processes, etc, these approaches require scanning of either the beam or the sample, exposing the sample to higher excitation energies and limiting time resolution of the imaging process. An alternative approach which is ideal for dynamic information is to simultaneously capture the scene from two perspectives. This limits the time resolution only by the acquisition rate of each sensor. The 3D scene could ideally be recreated by utilizing a shared spatial axis between the two sensors. However, correlating the information over a single axis can pose challenges at higher densities. We propose that quantum entangled light can be used to provide additional information as well as new detection architectures, while keeping peak excitation intensity low to preserve the integrity of the sample and avoiding biases that can be caused by scanning. Quantum entangled light can be spatially separated, while details of the momenta and temporal relations of entangled photons are preserved. This concept allows for the acquisition of quantum ghost images; which is traditionally achieved by separating entangled light into two paths and using one light path to interact with a specimen and the other light path to image the specimen. Utilizing quantum entangled light generated by a BBO crystal and the concepts of quantum ghost imaging, we present a microscope which can be used to view microscopic specimen in 3D from a single snapshot. This is achieved by utilizing two event-based 2D sensors in which information is relayed, through correlations, to generate two perspectives from a single scene. The quantum entangled light source allows us to correlate signal and idler, both spatially and temporally. We characterize the microscope by utilizing resolution targets at various depths and attempt to understand the achievable depth-of-field with the specific light source. After characterization, we image gold nanoclusters with our ghost imaging microscope. Additionally, we present our first use of utilizing scattering correlations with the 3DQ microscope for the imaging of nanoparticles. We foresee large potential for the 3DQ microscope in various areas, including longer wavelength and spectroscopy applications requiring higher resolution and sensitivity.

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Photon number measurements for bio-applications

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Abstract: Photon counting and photon number statistics measurements are an enabling technique that roots deep in fundamental quantum science. Here we present a quick overview of experimental methods that take advantage of quantum properties of single photon emission by a broad range of biomarkers. We also experimentally demonstrate the single photon character of bioluminescence, a nature's own single photon source that enables making quantum measurements with no external excitations, such as an auxiliary light source.

Within just a few decades, quantum measurements and quantum properties of light have found successful applications in various real-life scenarios. In biology, most of the light-emitting biomarkers are quantum sources of light, i.e. single photon sources. Indeed, single molecules of fluorophore, quantum dots, and color centers, typically used to label certain compounds can only emit one photon at a time. This property enables quite a few practical applications. First, by measuring second or higher-order autocorrelation of the light field emitted by a target, it is possible to learn exactly how many single photon sources contribute to the readout. To do so, photon number statistics should be measured. Such a measurement provides an absolute scale, that is distinctively different from the classical intensity scale. Moreover, A hallmark of observing fewer than N single photon sources is that the N-th order correlation function is equal to zero, an effect known as antibunching.

Beyond metrology, this antibunching can be used for other applications, including true super-resolution. For instance, most super-resolution techniques are based on reducing the number of active single photon emitters (in most cases, fluorophores) in the filed of view, such that there is only one active emitter per diffraction limited area (Airy disk). Then, super-resolving a point-source is merely finding a center of the Airy disk image. Therefore, to resolve many points within the diffraction limit one needs to cause point sources to turn on and off, and make certain that a maximum of one point source is active at a time, an exponentially difficult task if sources are probabilistically controlled. In addition, classical images do not provide information about the number of sources that contribute to an image. We use photon counting and take advantage of photon number distributions is available for each pixel. We have numerically established that a photon number resolving camera can be used to count the number of emitters per Airy disk and resolve the positions of overlapping single-photon sources. Photon counting commercial cameras that are available now enable experimental demonstrations of quantum super-resolution with thermal light modes simulating single photon sources. In experiment, more than two orders of magnitude super-resolution enhancement for five strongly overlapping thermal sources with unbalanced intensities was obtained. Beyond super-resolution, the mode reconstruction allows the extraction of information about the number of sources and intensity of each source even if sources are collocated (if the distance between sources is smaller than the super-resolution enhancement).

Many quantum photonics measurements in biology rely on single fluorophores as biomarkers, which is not ideal, because fluorophores require strong external light to function, leading to photobleaching and phototoxicity. We demonstrate that a natural biological effect, namely bioluminescence, leads to biocompatible optical single photon sources. Therefore, very long, practically unlimited, exposure times can be enabled and the quantum photonics methods can be consistently applied. In this work we experimentally show for the first time that each bioluminescent enzyme can only emit a single photon at the time by measuring the second-order correlation function. The second-order correlation function gives information about the source's multi-photon emission probability from first principles, and the reduced number of many-photon states is a hallmark of quantum light. Here we also report for the first time on the quantum measurement of the single, as opposed to ensemble-averaged, enzyme kinetic rates. We use measured temporal profile of the photon correlation function to extract molecular kinetic rates of the enzyme reaction.

Scattered biphotonic state by a two-photon absorbing medium

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Abstract: We present a theoretical model describing how a two-photon state, generated by spontaneous parametric down conversion (SPDC), is scattered after its interaction with a two-photon absorbing medium, in particular we study the $6S_{1/2}$ → $8S_{1/2}$ transition for Cesium atoms.

One of the most common ways to look for the entangled two-photon absorption signal (eTPA) has been through transmission schemes where coincidences or singles counts are monitored following transmission through the sample under study. However, it has been shown that this method can generate a signal screened by single photon losses that can mimic the expected behavior of biphoton absorption [1,2]. In order to correctly discriminate the signal originating from eTPA when using transmission methods, one proposal is to carry out spectrally-resolved measurements, since the process may act as a filter that removes certain frequencies from the joint spectral intensity (JSI) [3]. Here we present a theoretical development that predicts how spectral correlations are affected after interaction with the system. To achieve this, we use a four-wave interaction Hamiltonian mediated by third-order electrical susceptibility, χ(3), which contains the information of the $6S_{1/2}$ → $8S_{1/2}$ transition in Cesium atoms. The results show that the frequency distribution of the pairs that contribute to biphoton absorption – when excitation is reached both simultaneously and through intermediate states – exhibit an anti-diagonal behavior, which is characteristic of a two-photon resonance process.

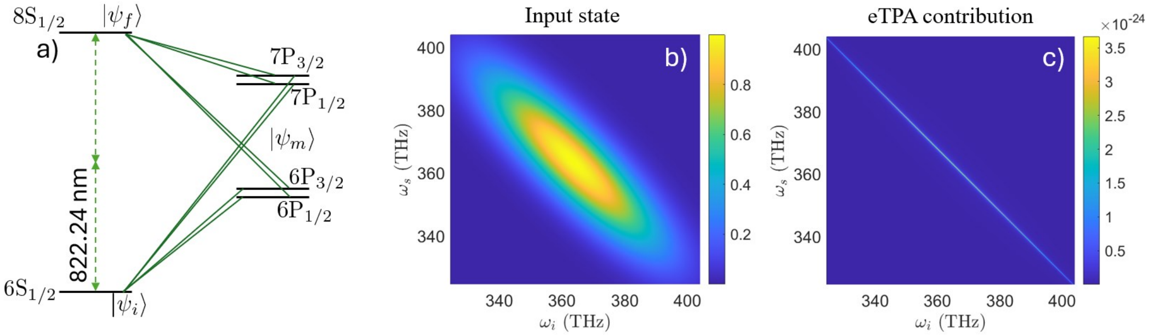

Fig. 1: a) Energy levels of the $6S_{1/2}$ → $8S_{1/2}$ transition in Cesium atoms used to study the eTPA process. (b) Joint spectral intensity of the incident state in the sample; photon pairs generated by SPDC have a central wavelength of 822 nm. c) Frequencies that contribute to the two-photon absorption, the transmitted state, thus, turns out to be the difference between Figure (b) and (c).

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Role of time-frequency correlations in entangled-two-photon resonance energy transfer

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Since 2004, there exists a long standing discussion on the role that frequency entanglement, as well as time-ordered excitation pathways, plays in the energy transfer from a two-photon donor to an acceptor comprising two non-interacting two-level particles. More specifically, the study of entangled-two-photon resonance energy transfer (E2P-RET) started with the work of A. Muthukrishnan *et al.,* [1], who demonstrated that two non-interacting two-level atoms could be jointly excited by time-frequency correlated photons. In their work, the authors claimed that joint excitation of atoms was possible because of the suppression of time-ordered excitation pathways. This suppression was a consequence of how time-frequency entangled photon pairs were produced in their theoretical model, namely a cascade decay of an atomic three-level system. In 2019, K. Nasiri Avanaki and G. C. Schatz [2] came to the same conclusion when describing the three orders of magnitude enhancement (compared to frequency-uncorrelated excitation) of E2P-RET using time-frequency entangled photons. Interestingly, in 2013, Z. Zheng *et al.,* [3] showed that the simultaneous excitation of non-interacting two-level atoms was a result of strong frequency anti-correlation between photons, and neither time-correlation nor time-frequency entanglement were required. The authors' conclusion applied to both cascade photon sources as well as Gaussian-spectral-shaped spontaneous parametric down-conversion (SPDC) photon pairs.

In this talk, we will discuss a general model for the joint excitation of two non-interacting two-level particles by SPDC photons. We will see that while strong frequency anti-correlation between photons guarantees a large two-photon absorption (TPA) probability, photons bearing a sine cardinal spectral shape (the joint photon spectrum that is naturally produced during the SPDC process) exhibit a ~3.8 times larger TPA signal than photons with a Gaussian spectrum. More importantly, and in stark contrast to previous authors, we find that suppression of time-ordered excitation pathways does not substantially modify the TPA probability for two-photon states with a Gaussian spectral shape; whereas photons with a sine cardinal spectrum exhibit the strongest TPA signal of all when two-photon excitation pathways are not suppressed. Our results not only help elucidating the role of time-frequency correlations in resonance energy transfer with SPDC photons, but also provide valuable information regarding the optimal source to be used in its experimental verification.

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Limitations in Fluorescence-Detected Entangled Two-Photon-Absorption Experiments: Exploring the Low- to High-Gain Squeezing Regimes

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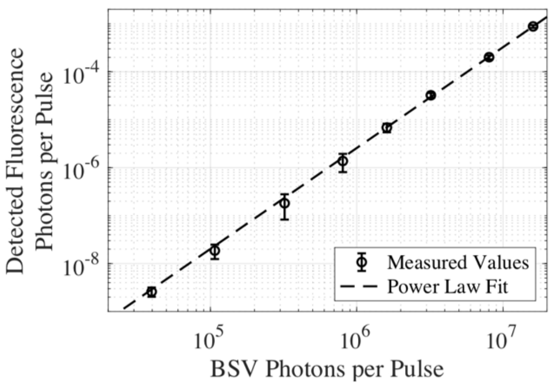
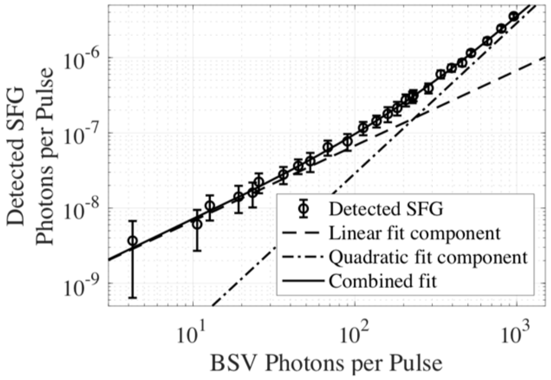
and Quantum Science, University of Oregon, Eugene, Oregon 97403, USA

I will review our theoretical and experimental work demonstrating that Entangled Two-Photon-Absorption (ETPA) is likely undetectable in solvated molecules using state-of-the-art techniques. In our latest experiment (https://arxiv.org/abs/2404.16342), we closely replicated and extended a recent experiment ("Spatial properties of entangled two- photon absorption," Phys. Rev. Lett. 129, 183601, 2022) that reportedly observed enhancement of ETPA in molecular samples, and we found that in the low-flux regime, where such enhancement is theoretically predicted in-principle, the two-photon fluorescence signal remains below detection threshold.

Using an optical parametric down- conversion photon-pair source that can be varied from the low-gain spontaneous regime to the high-gain squeezing regime (bright squeezed vacuum, BSV), we observed two-photon-induced fluorescence in the high-gain regime but in the low-gain regime any fluorescence was below detection threshold. We supplemented the molecular fluorescence experiments with a study of nonlinear-optical sum-frequency generation, for which we are able to observe the low-to-high-gain crossover, thereby verifying our theoretical models and experimental techniques. The observed rates (or lack thereof) in both experiments are consistent with our theoretical predictions and previous experiments.

	
Measurements of TPA-induced fluorescence from Rhodamine 6G (R6G) dye driven by pulsed BSV at an average power of 300 nW . Quadratic scaling dependence (dashed line) is observed over the entire measurement (power law fit coefficient = 2.1). Measurement durations were adjusted to ensure 5σ threshold was met for all measurements.	The crossover between linear and quadratic scaling regimes for SFG was measured to occur at approximately 250 photons per pulse, in good agreement with predictions of 1 photon per temporal mode. The measured result is higher by a factor of 2-3 than estimates of the number of modes for the source, a difference attributed to spatial correlations present in the bulk SPDC process, which were not considered in the theoretical description.

Workshop on Quantum Light Generation, Detection, and Applications

Name: Alan McLean, NIST/JILA, Jimenez Group (NRC Postdoc)

Full Author List:

Alan McLean, Christian Drago, Daniel Podos, John Sipe, Ralph Jimenez

Title:

Quantifying E2PA: Ultracold rubidium in a magneto-optical trap

Abstract:

The magnitude and mechanism of entangled two-photon absorption (E2PA) remains an outstanding question. Studies involving small molecules in solution have not yet produced observable E2PA. In this talk we discuss using ultracold rubidium to observe E2PA. Rubidium, and other atomic platforms generally, have several key advantages over solution-phase systems, a.) They can possess ultra-high two-photon cross-sections (>1000x bigger than molecules in solution), b.) Their electronic spectra are simple (with well-defined real states) and free from pitfalls of solution-phase spectra (such as solvent-induced line-broadenings), c.) Given their well-defined spectra, classical and E2PA effects can be accurately modeled using theory. Rubidium also has several advantages that set it apart from other atomic platforms, a.) It possesses a real-state very close (approx. 2 nm) to the two-photon virtual state transition, which greatly influences E2PA rates, b.) Rubidium can be optically cooled to ultralow temperatures. Regarding the latter point, when modeling E2PA in atomic rubidium at room-temperature, we find that the magnitude E2PA is far below the magnitude of incoherent (non-quantum) effects. However, when cooled to ultracold temperatures, we find that E2PA effects dominate by over a factor of 1000x as compared to incoherent effects. To realize this platform, we construct a magneto-optical trap to optically cool and trap rubidium atoms. From the ultracold rubidium MOT, we observe two-photon fluorescence from mW powers down to low powers of excitation. We compare the experimental classical two-photon count rates with expected count rates as calculated using second-order perturbation theory. Experiments with entangled photons in the ultracold rubidium MOT are discussed.

Fluorescence by nonclassical light: Getting it right

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Abstract: We calculate the fluorescence of cold cesium atoms excited by low intensity, non-degenerate squeezed light in the CW limit. The fluorescence rate is enhanced over what would be expected for excitation by classical light, and the signal is predicted to be experimentally detectable.

Introduction: Sources of bright squeezed light are becoming ubiquitous, and soon one can expect the same for sources of quantum light with more exotic properties. It is then natural to consider the interaction of quantum states of light with other quantum systems. One interesting process is the two-photon transition of an atom or molecule driven by squeezed light, which is said to enhance the excitation rate over the use of classical light in the low intensity limit [1]. However, given that the "quantum advantage" of squeezed light is largest when the detection signals are weak, there has been significant controversy about the interpretation of experiments [2]. This demonstrates the importance of carefully modeling quantum systems and their interactions in a way that can be used to predict real experiments.

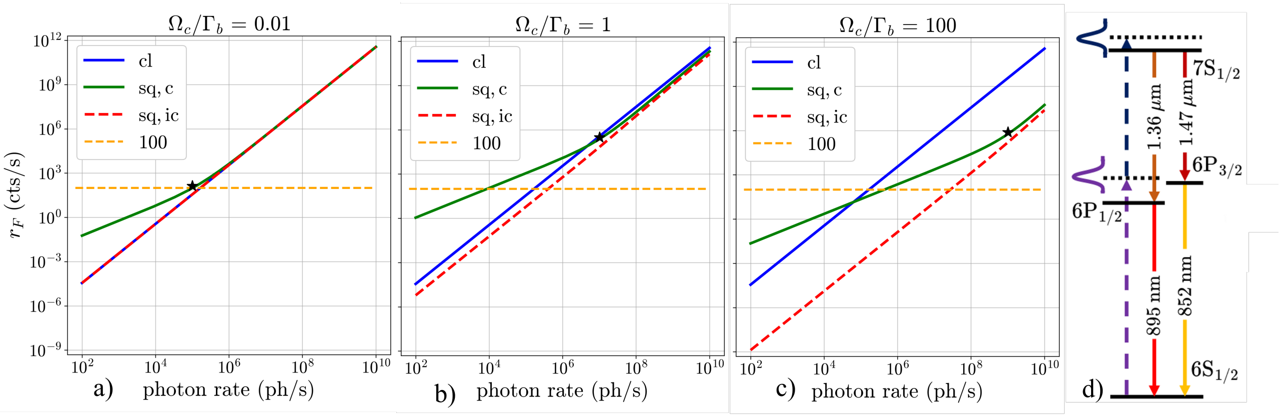

FIG. 1. a,b,c) Fluorescence rate as a function of incident photon rate for increasing bandwidth $\Omega_c/\Gamma_b = 0.01, 1, 100$; horizontal orange line is what we deem to be the minimum detectable threshold before experimental losses. d) Energy level diagram of cesium where downward (upward) arrows represent radiative and nonradiative decays (pumping transitions). For our calculations we use typical MOT parameters for the atom number and distribution.

Results: As an example calculation, we consider the non-degenerate two-photon excitation of atomic cesium (see Fig. 1d) and calculate the subsequent fluorescence. In our analysis we consider a full squeezed state and calculate both the "coherent" (due to anti-correlated frequencies) and "incoherent" (due to uncorrelated frequencies) contributions to the two-photon transition. For CW excitation, the expected fluorescence count rate at 852 nm is calculated for classical and squeezed light and plotted in Fig. 1a,b,c as a function of the incident photon rate, for three values of the squeezed bandwidth set by $\Omega_c/\Gamma_b = 0.01, 1, 100$; Γ_b is the intermediate state linewidth. In each plot we set the orange horizontal line to 100 counts/s, which we deem to be the minimum detectable threshold before experimental losses. The star point corresponds to the cross-over point, where the scaling changes from linear to quadratic. This point depends on the spectral-temporal correlations, and so moves towards the right in each plot.

In the narrowband regime ($\Omega_c/\Gamma_b = 0.01$), even with the enhancement that the squeezed light provides, the signal is below the detectable threshold in the low photon rate limit; in the high photon rate limit the classical and squeezed contributions are all identical because they are all narrow band and centered at the same center frequency. For the intermediate regime ($\Omega_c/\Gamma_b = 1$) the situation is more interesting. Due to the increased spectral-temporal correlations on the order of the system linewidth, a region opens where the squeezed light provides an enhancement that takes the emission rate over the detectable limit. In the broadband limit ($\Omega_c/\Gamma_b = 100$) there is no longer a detectable enhancement, and the "coherent" and "incoherent" contributions are significantly suppressed in the high photon rate limit due to the increased bandwidth. It is interesting to note that in the broadband limit the "incoherent" contribution remains relatively significant compared to the "coherent" contribution, despite the usual intuition. This is a result of the intermediate and final state linewidths (Γ_b, Γ_c) being comparable for cesium.

Conclusion: We find narrow regions of parameter space where squeezed light should provide an enhancement of the fluorescence rate from two-photon excited atomic cesium that takes it above the detectable limit. These regions, and the relation between the "coherent" and "incoherent" contributions, are very system dependent, and it should not be assumed that the "incoherent" contribution is always negligible.

[1] N Ph Georgiades et al. PRL, 75(19):3426, 1995. [2] KM Parzuchowski et al. PRApp, 15(4):044012, 2021.