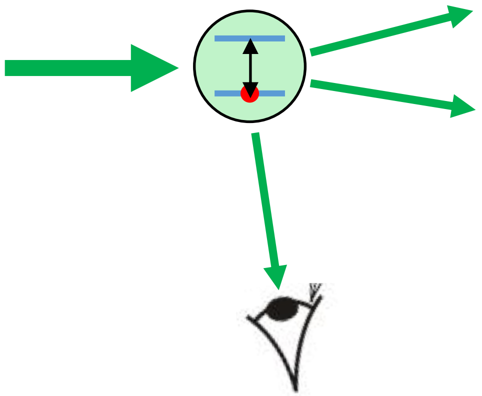
Rethinking Resonance Fluorescence:
Fundamental Insights and Emerging Quantum Technologies

100 Years of Quantum Physics
Quy Nhon, Vietnam
October 6–9, 2025

Arno Rauschenbeutel
Humboldt-Universität zu Berlin, Germany

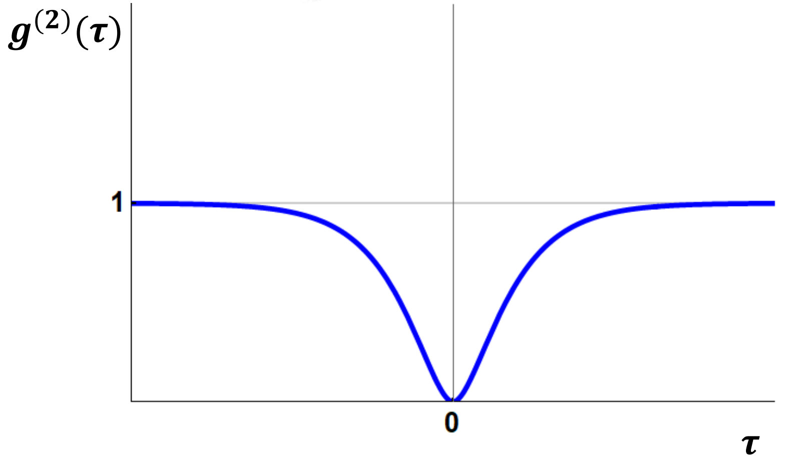
Antibunching in Resonance Fluorescence

- **Single two-level emitter**
  Atom in an optical tweezer, *single ion, quantum dot, single molecule, …*



- **Near resonant, weak driving**
  $S = I/I_{sat} << 1$

- **Scattered field features coherent and incoherent component**
  $n_{coh} \propto S$ and $n_{incoh} \propto S^2$



- **Probability for detecting two photons simultaneously = 0**

Standing on the Shoulders of Giants

Credit: Caltech

Jeff Kimble (1949–2024)

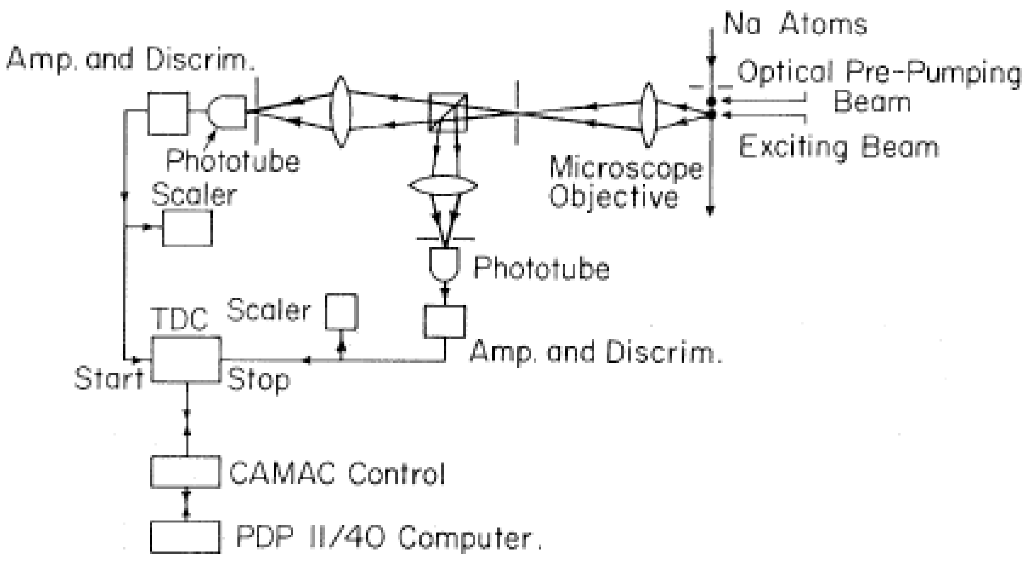
Antibunching in Resonance Fluorescence

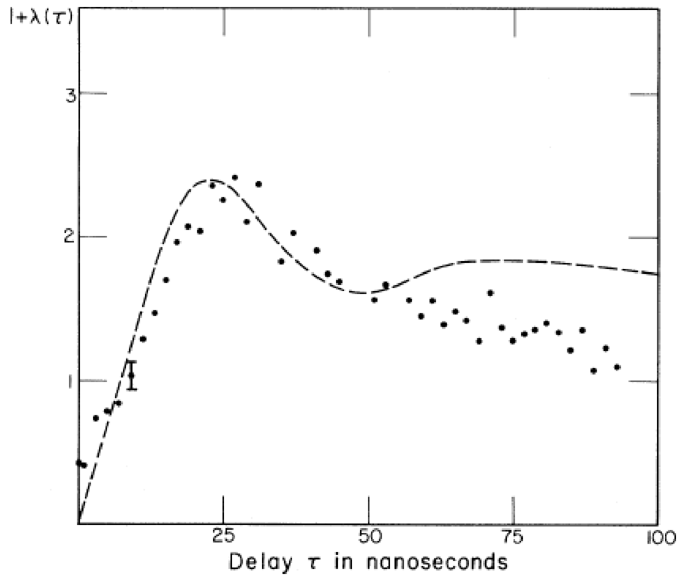
VOLUME 39, NUMBER 11 PHYSICAL REVIEW LETTERS 12 SEPTEMBER 1977

Photon Antibunching in Resonance Fluorescence

H. J. Kimble,[(a)] M. Dagenais, and L. Mandel
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627

(Received 22 July 1977)





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Photon Antibunching in Resonance Fluorescence

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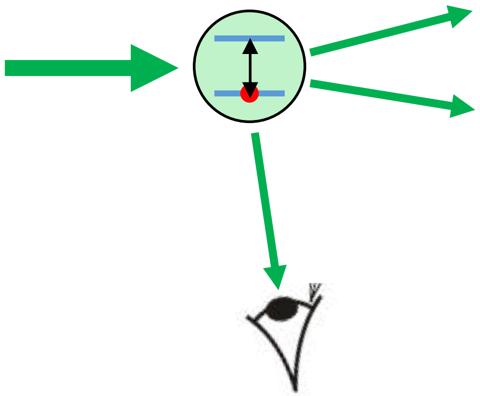
photon is detected. That $P_2(t, t+\tau)$ vanishes when $\tau=0$ for a single atom may be regarded as a reflection of the fact that the atom, having emitted a photon at time t, is unable to radiate again immediately after having made a quantum jump back to the lower state. The quantum nature of the radiation field and the quantum jump in emission, which are of course inextricably connected, are therefore both manifest in these photoelectric correlation measurements.

Antibunching in Resonance Fluorescence

- **Single two-level emitter**
  Atom in an optical tweezer, *single ion, quantum dot, single molecule, …*



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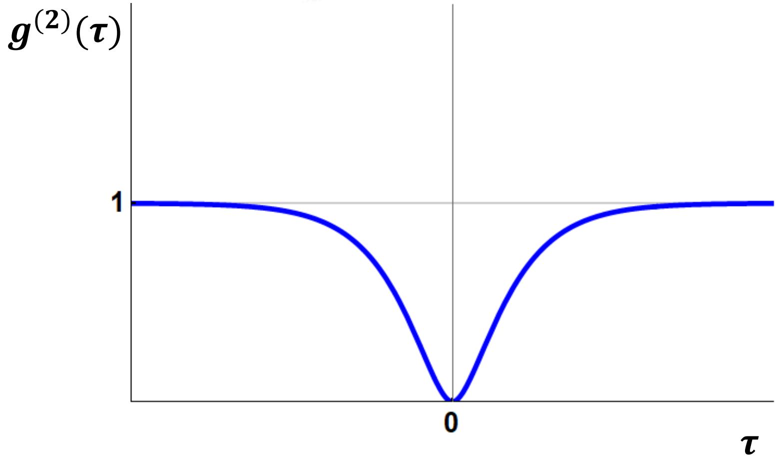


- **Scattered field features coherent and incoherent component**
  $n_{coh} \propto S$ and $n_{incoh} \propto S^2$

- **Probability for detecting two photons simultaneously = 0**
  A single two-level atom will not simultaneouly scatter two photons! Really?

Outline

Will a single two-level emitter scatter 2 photons simultaneously?

How to harness resonance fluorescence as a highly efficient source of time-bin entangled photon pairs.

Standing on the Shoulders of Giants

Jean Dalibard

Serge Reynaud

Antibunching through Quantum Interference

J. Physique **44** (1983) 1337-1343 DÉCEMBRE **1983,** PAGE 1337

Correlation signals in resonance fluorescence : interpretation *via* photon scattering amplitudes

J. Dalibard and S. Reynaud

Laboratoire de Spectroscopie Hertzienne de l'E.N.S., 24, rue Lhomond, 75231 Paris Cedex 05, France

(*Reçu le 20 juin 1983, accepté le 29 août 1983*)

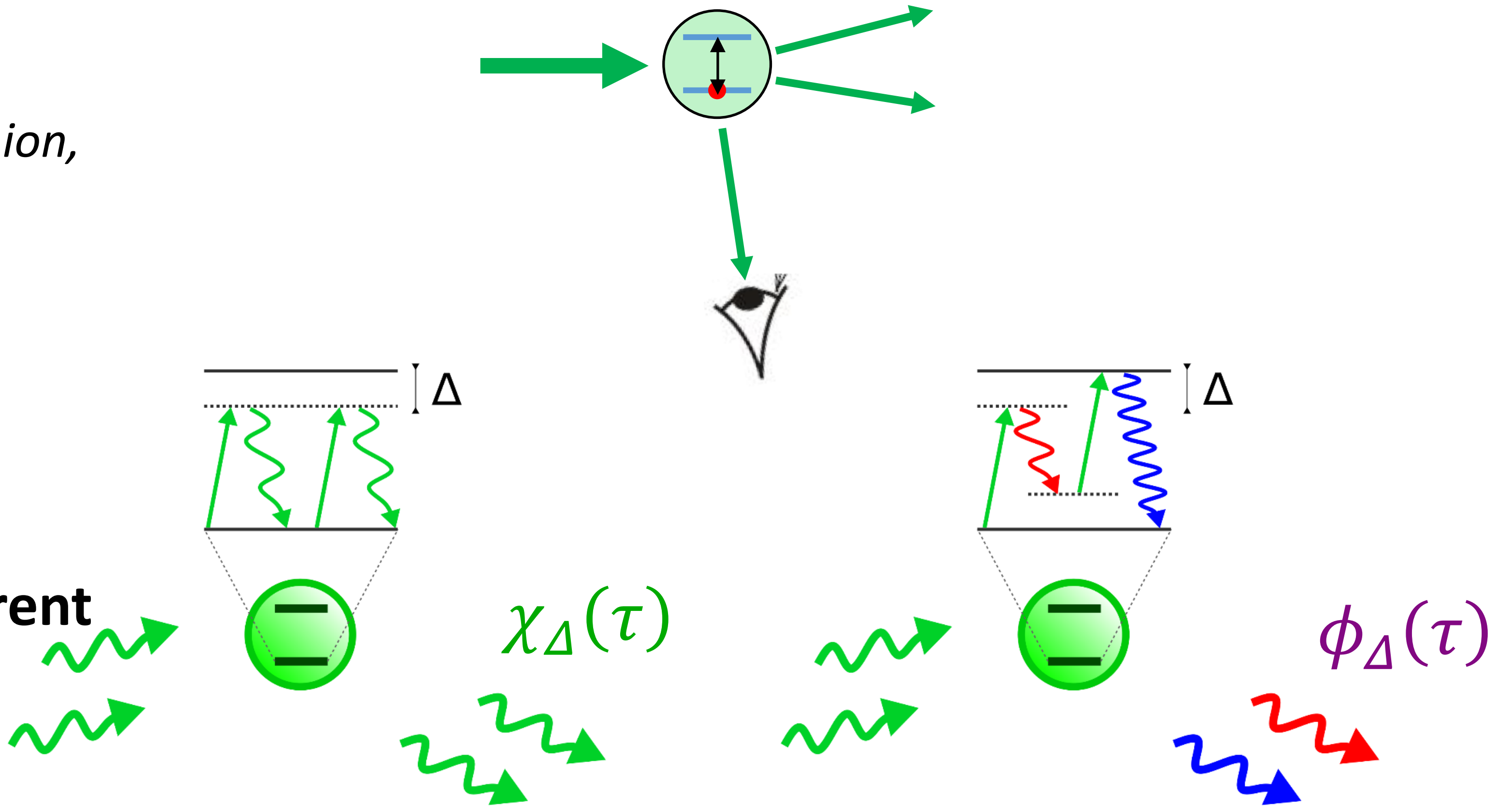
Abstract. — Resonance fluorescence is treated as a collision process where incident laser photons are scattered by an atom. Correlation signals are extracted from an expansion to the second order of the post collision field state. Photon antibunching effect appears as a quantum interference between all the possible scattering amplitudes. When Rayleigh photons are rejected, some amplitudes vanish, leading to a bunching behaviour.

Theory: *Two-Photon Scattering*

- **Single two-level emitter**
  Atom in an optical tweezer*, single ion, quantum dot, single molecule, …*

- **Near resonant, weak driving**
  $S = I/I_{sat} << 1$

- **Scattered field features coherent and incoherent component**
  $n_{coh} \propto S$ and $n_{incoh} \propto S^2$



Coherent two-photon scattering

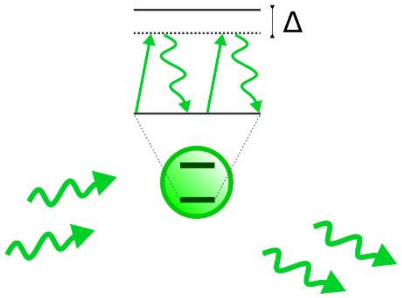
Two separable output photons

Incoherent two-photon scattering

Entangled photon-pair

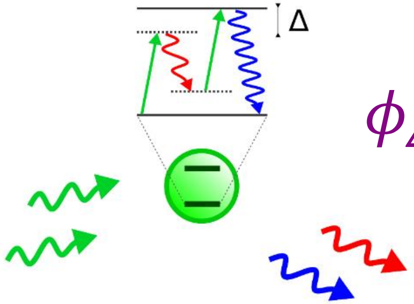
Theory: *Two-Photon Scattering*

Coherent



$$\chi_\Delta(\tau) = \frac{n_{coh}}{2}$$

Incoherent



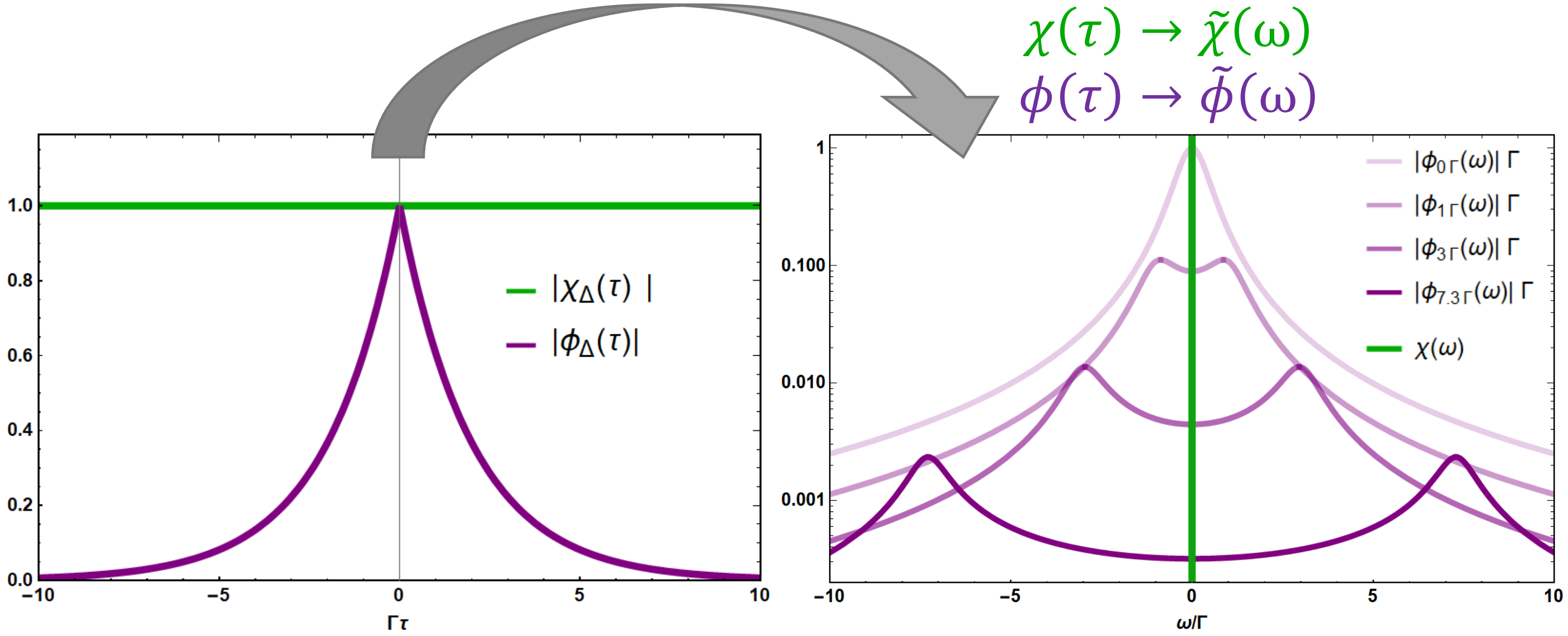
$$\phi_\Delta(\tau) = -\frac{n_{coh}}{2} e^{-\frac{\Gamma}{2}|\tau|} e^{i\Delta|\tau|}$$

nonlinear phase

Fourier Transform:

$\chi(\tau) \rightarrow \tilde{\chi}(\omega)$

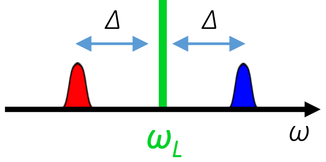
$\phi(\tau) \rightarrow \tilde{\phi}(\omega)$



Output two-photon wavefunction:

$$\Psi_2(\tau) = \chi(\tau) + \phi(\tau)$$

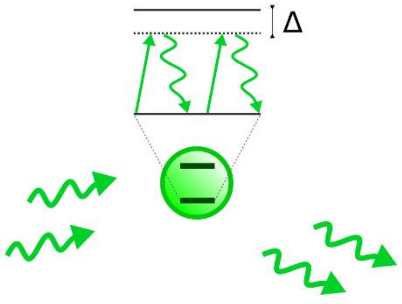
$|\tilde{\Psi}_2(\omega)|^2$



➢ **Superposition** of coherently and incoherently scattered two-photon amplitudes

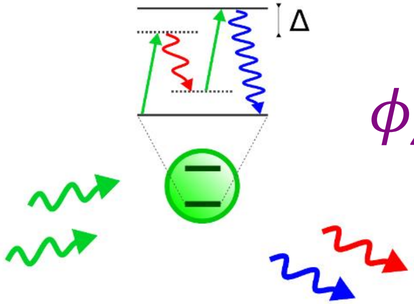
Theory: *Two-Photon Scattering*

Coherent

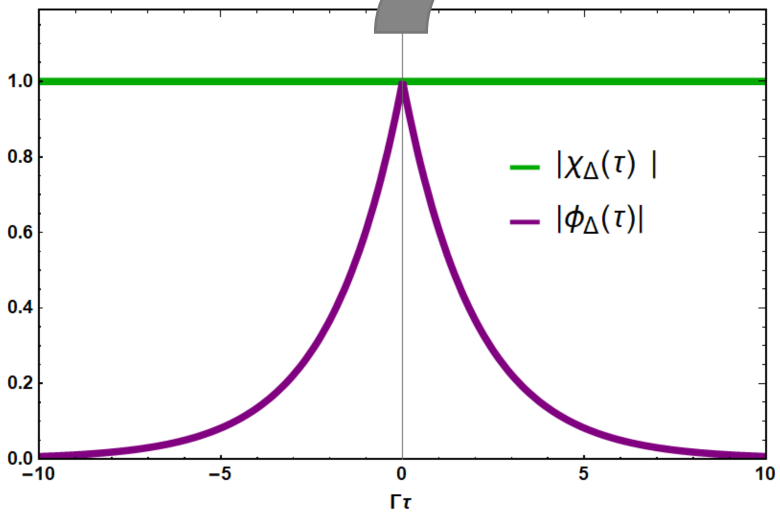


$$\chi_\Delta(\tau) = \frac{n_{coh}}{2}$$

Incoherent



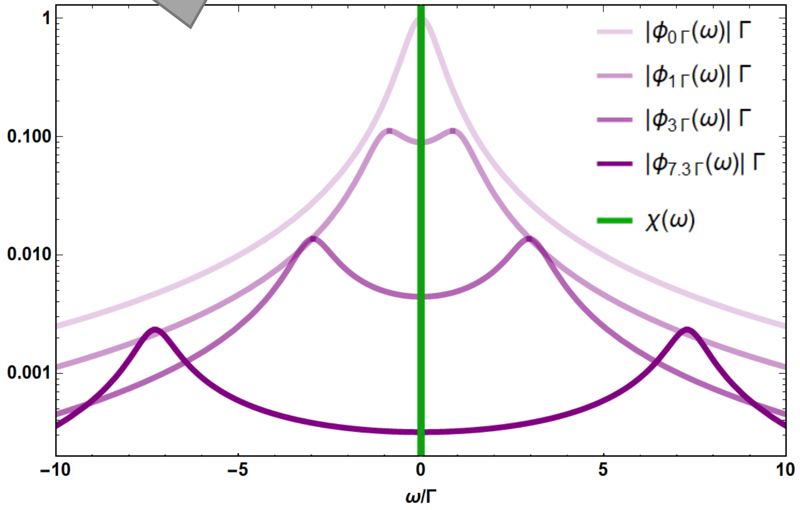
$$\phi_\Delta(\tau = 0) = -\chi_\Delta(\tau = 0)$$



Fourier Transform:

$\chi(\tau) \to \tilde{\chi}(\omega)$

$\phi(\tau) \to \tilde{\phi}(\omega)$



Output two-photon wavefunction:

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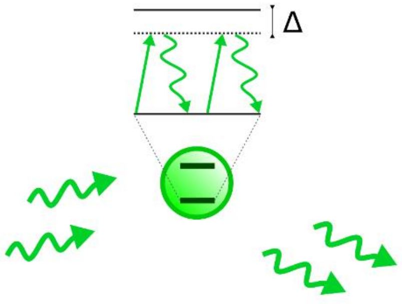
➢ **Superposition** of coherently and incoherently scattered two-photon amplitudes

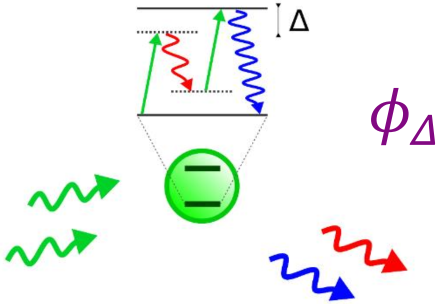
Theory: *Two-Photon Scattering*

Coherent



$$\chi_\Delta(\tau) = \frac{n_{coh}}{2}$$

Incoherent



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Fourier Transform:

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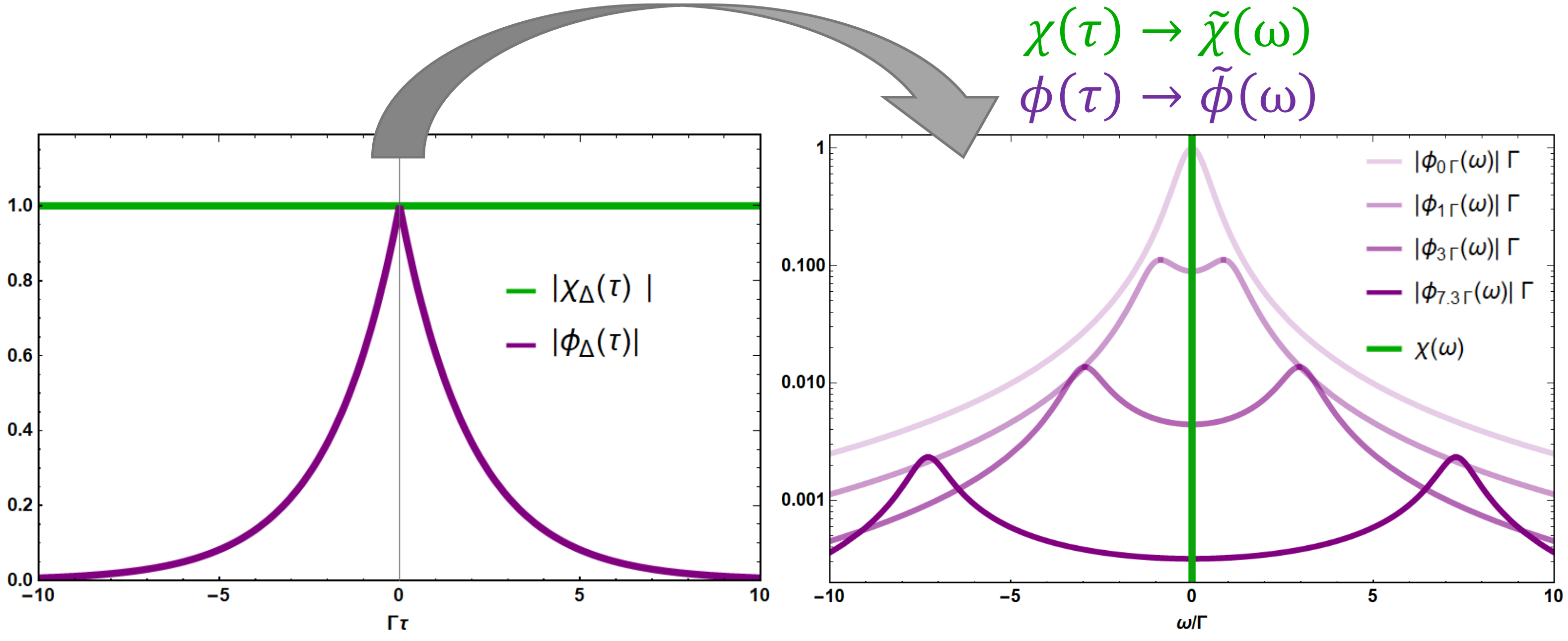


Output two-photon wavefunction:

$$\Psi_2(\tau = 0) = 0$$

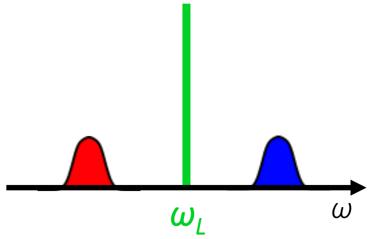
- **Destructive interference** of coherently and incoherently scattered two-photon amplitudes

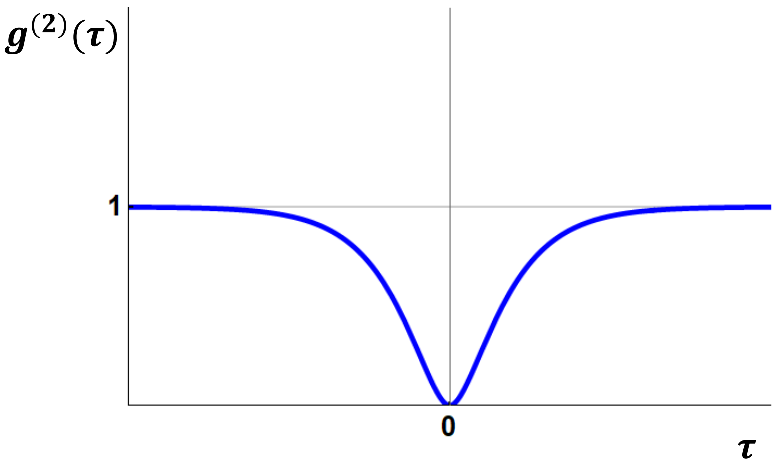
Theory: 2^{nd} *Order Quantum Coherence Function*

$$g^{(2)}(\tau) \propto |\Psi_2(\tau)|^2 = |\chi(\tau) + \phi(\tau)|^2$$

***Note**: *for clarity, Δ ≠ 0 for 2-photon spectra, but Δ = 0 for $g^{(2)}(\tau)$ plots*

- **Interference** between coherently and incoherently scattered photons





Theory: *2nd Order Quantum Coherence Function*

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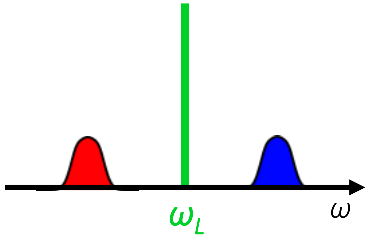
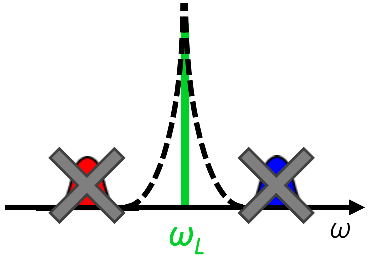
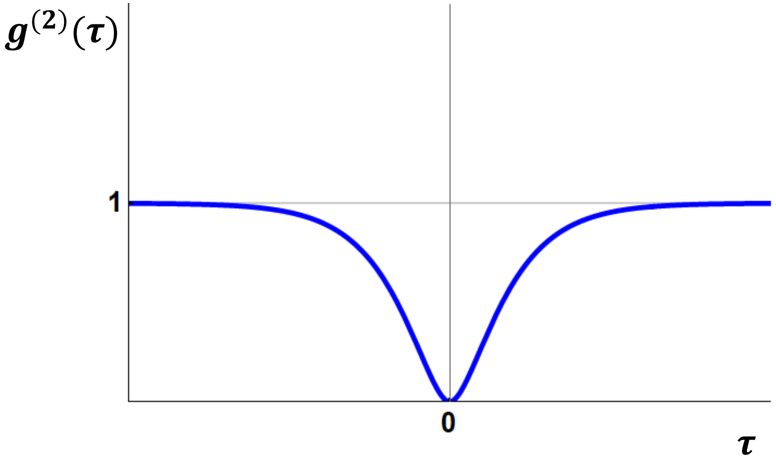
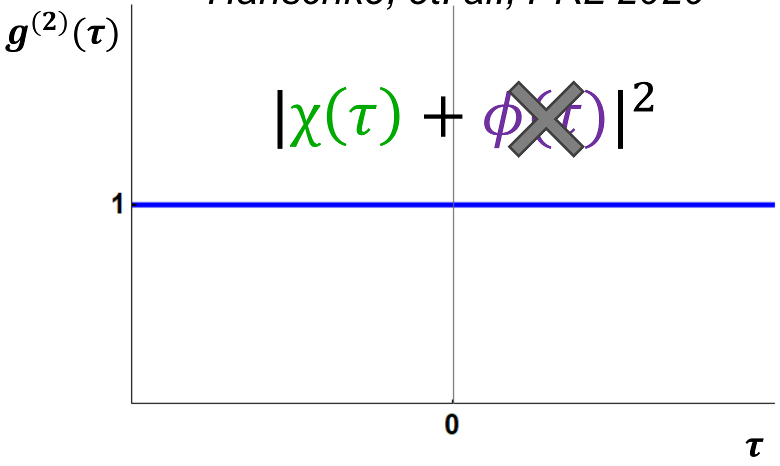




- *Hanschke, et. al., PRL 2020*





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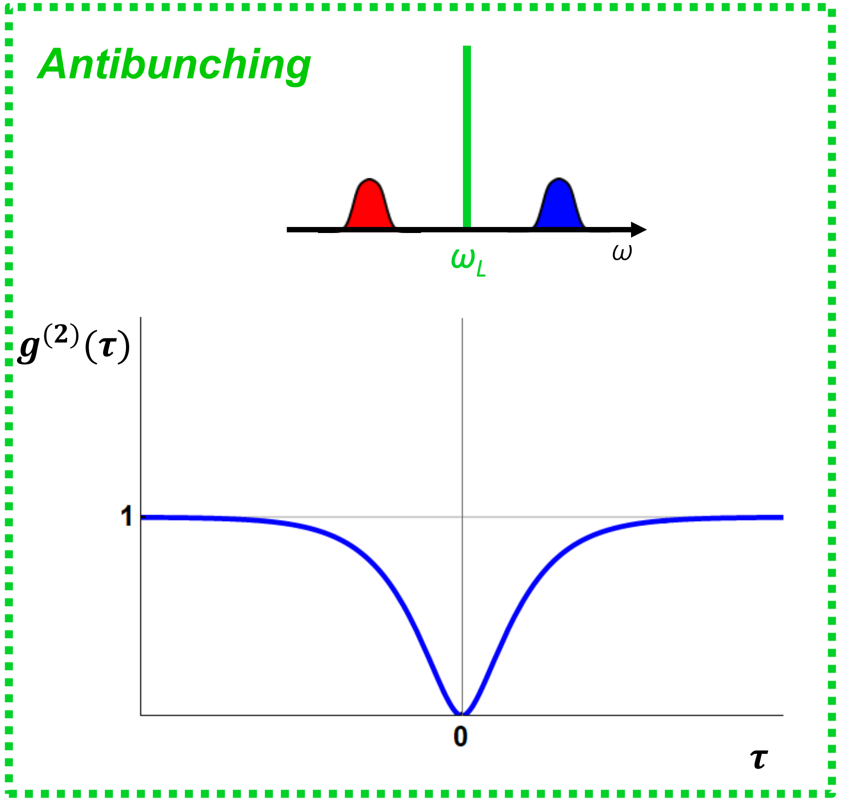
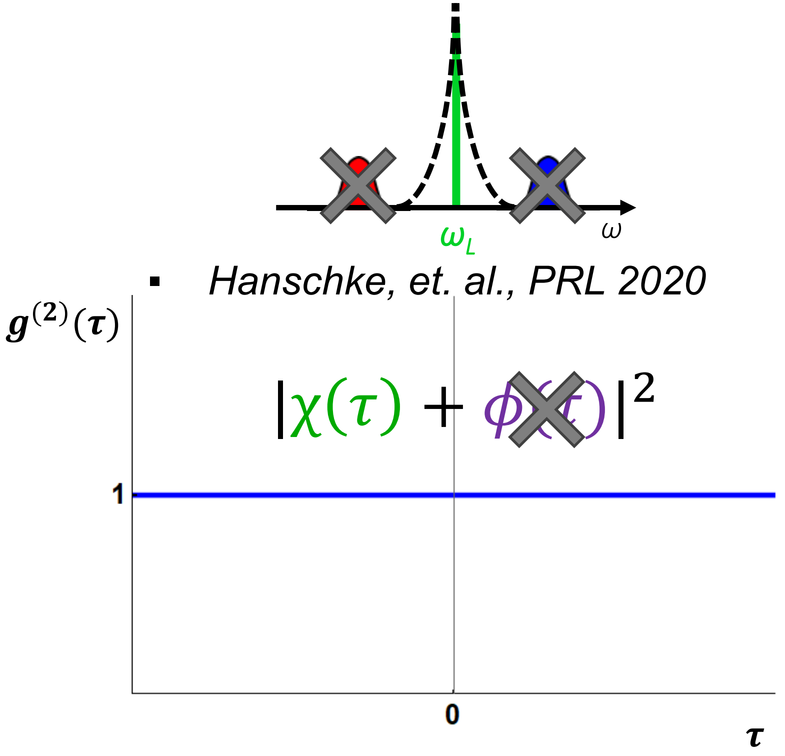
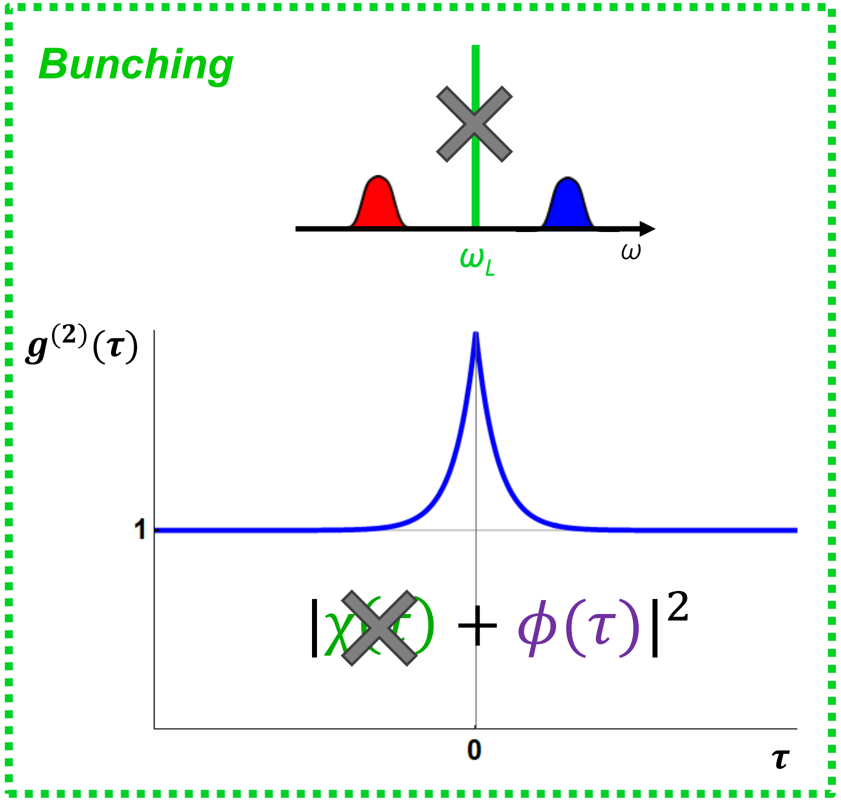
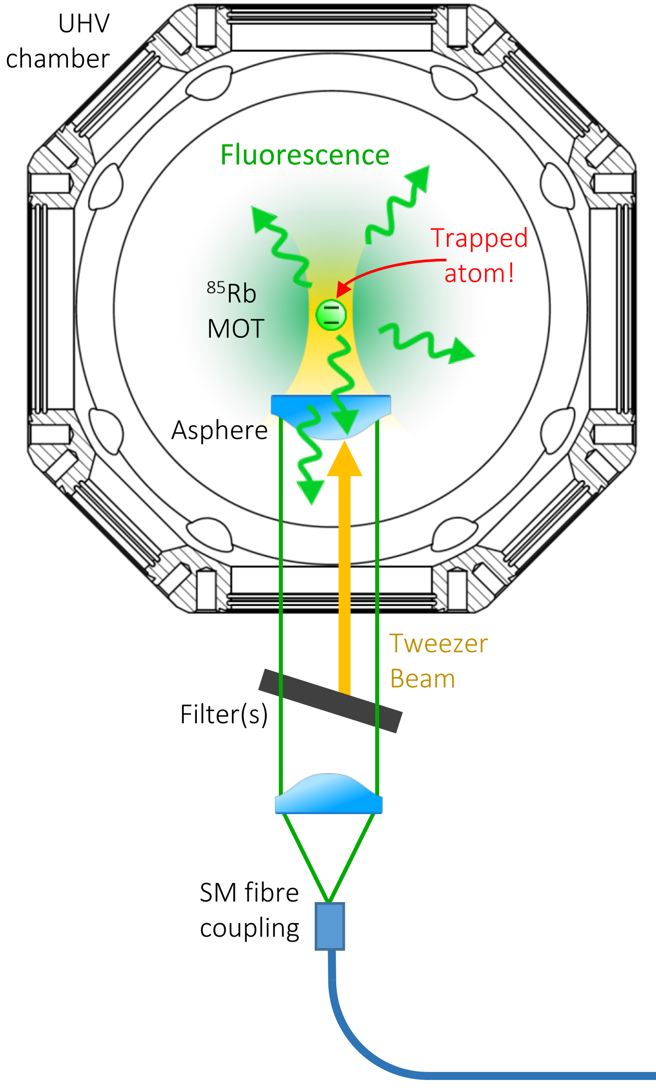
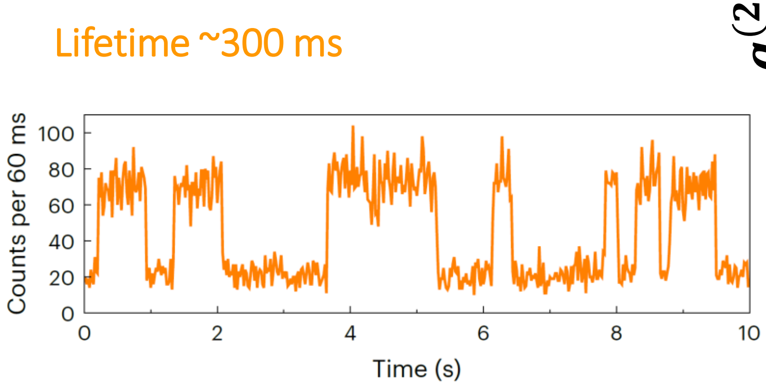


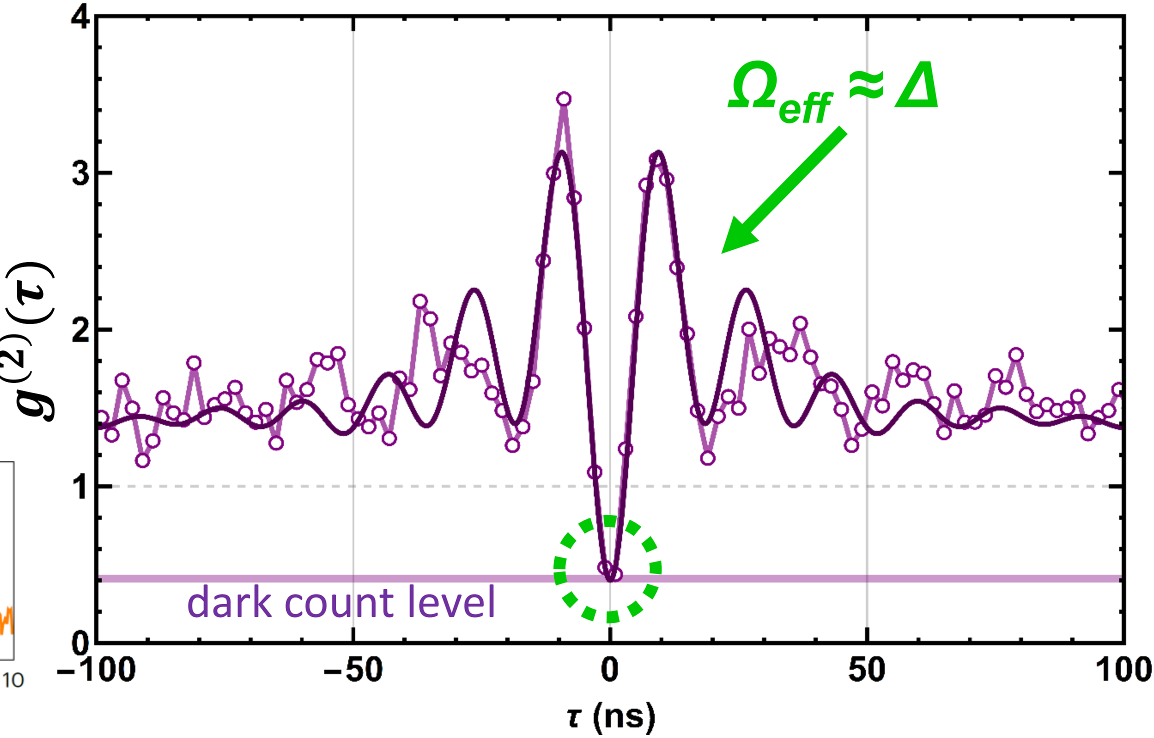


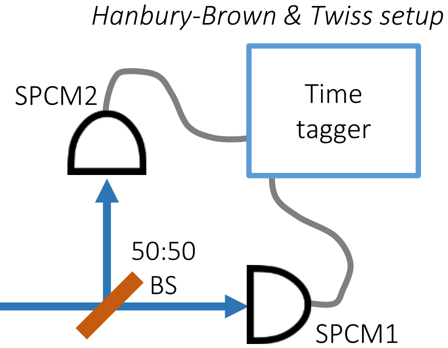


Experiment: *Antibunching*







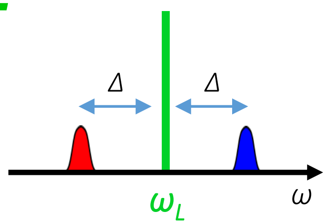


- ➢ $g^{(2)}(0) \approx 0.4$
- ➢ $\Delta \approx 60$ MHz

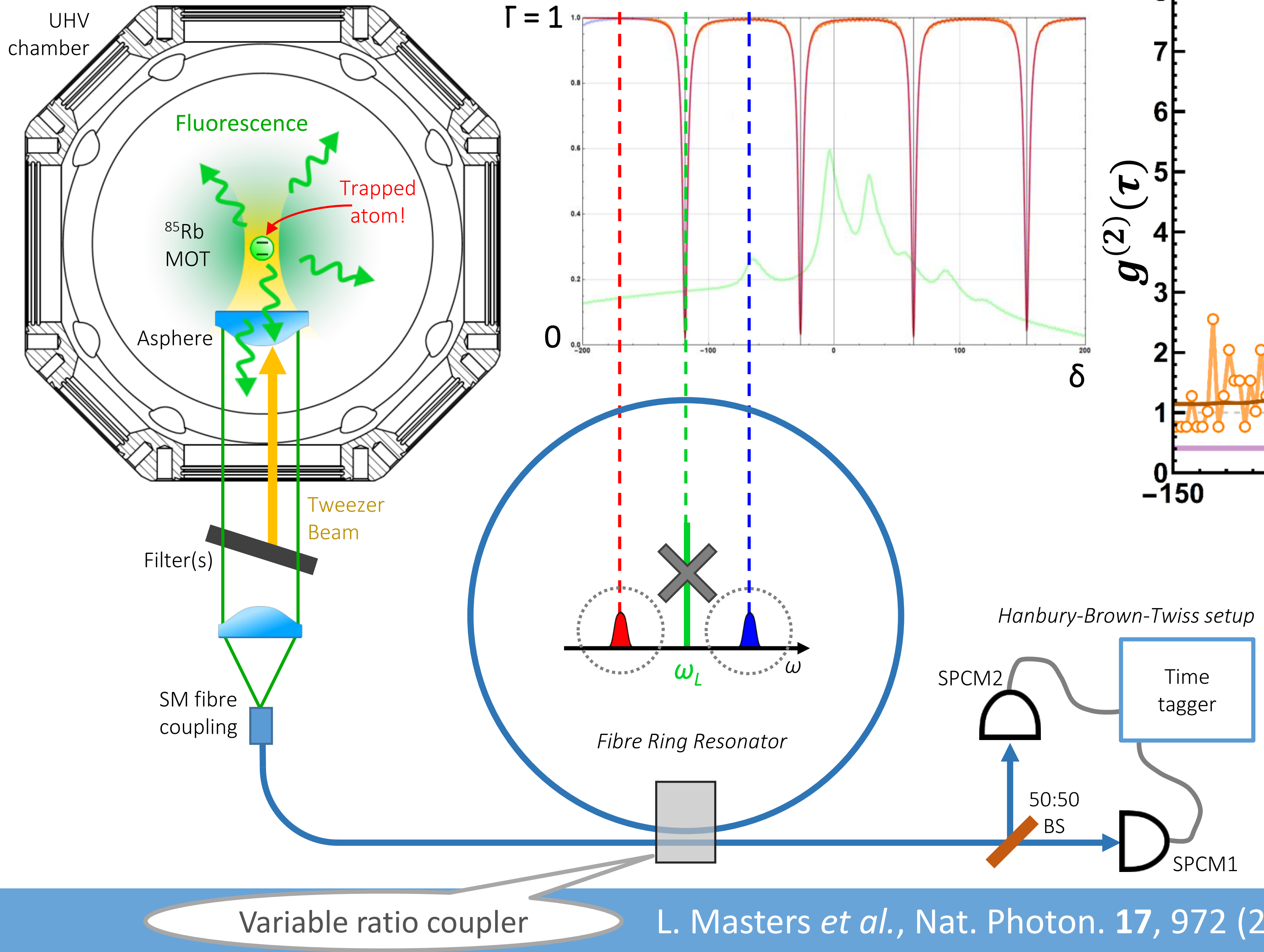
$\Delta \approx \delta_{MOT} + \delta_{STARK}$

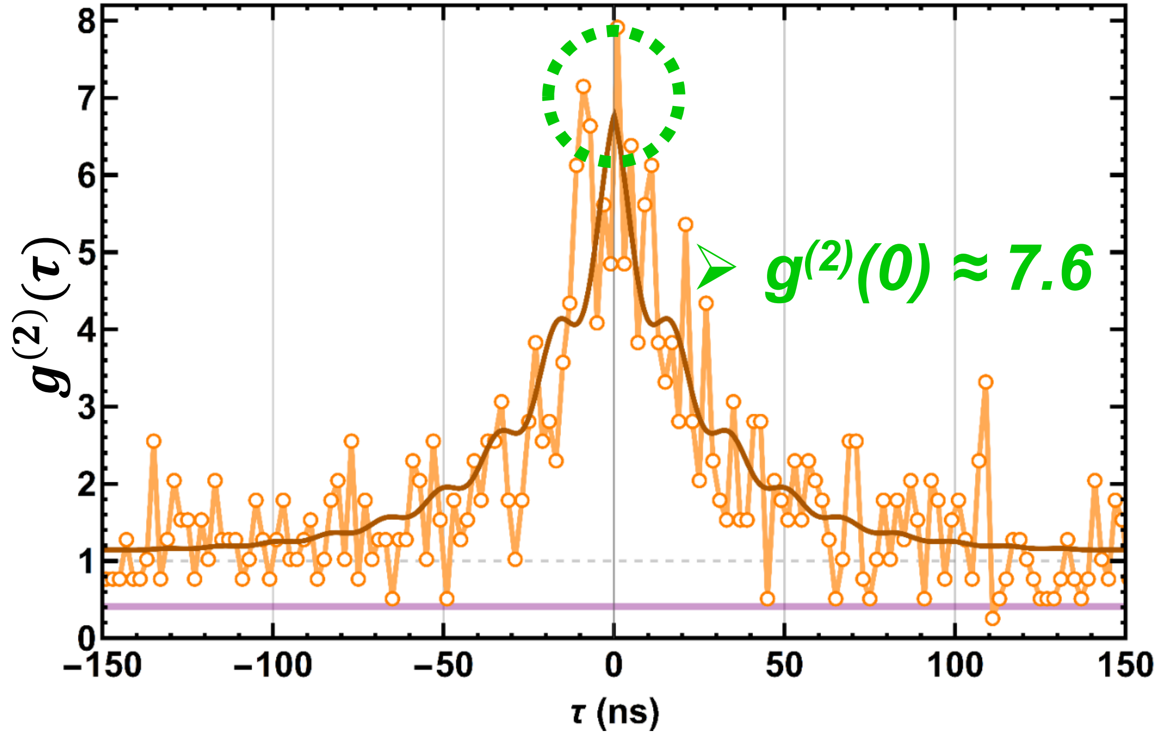
$\delta_{MOT} \approx 16$ MHz

$\delta_{STARK} \approx 44$ MHz



Experiment: *Bunching*

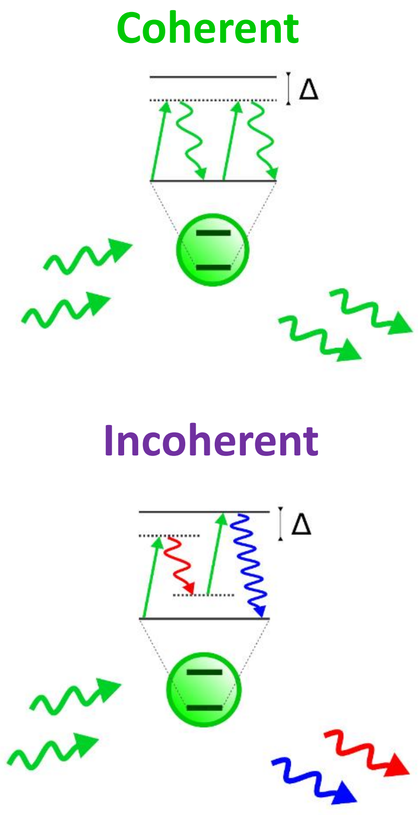


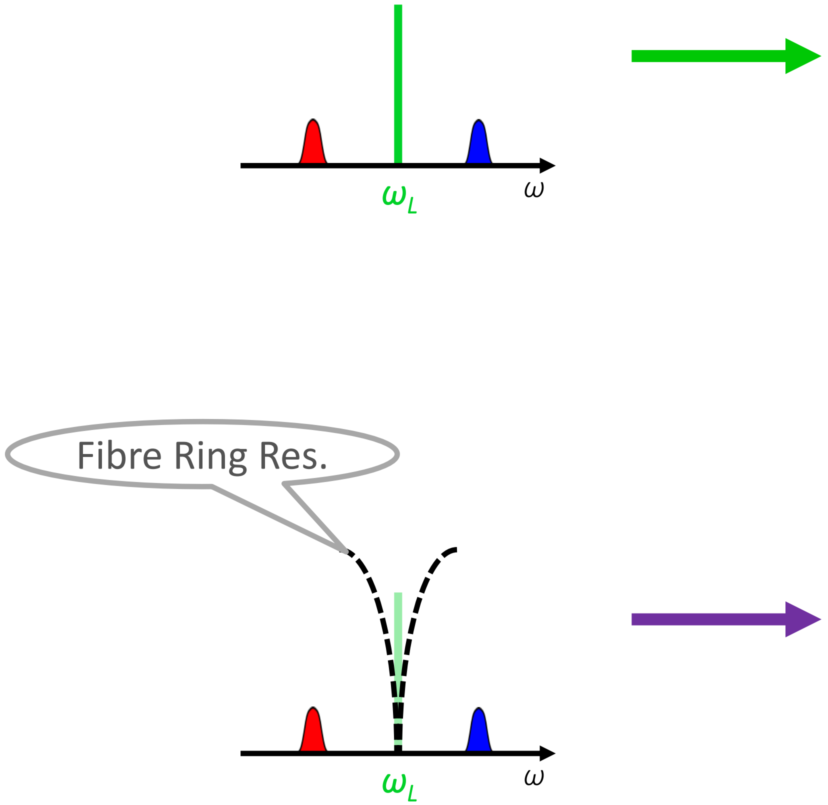


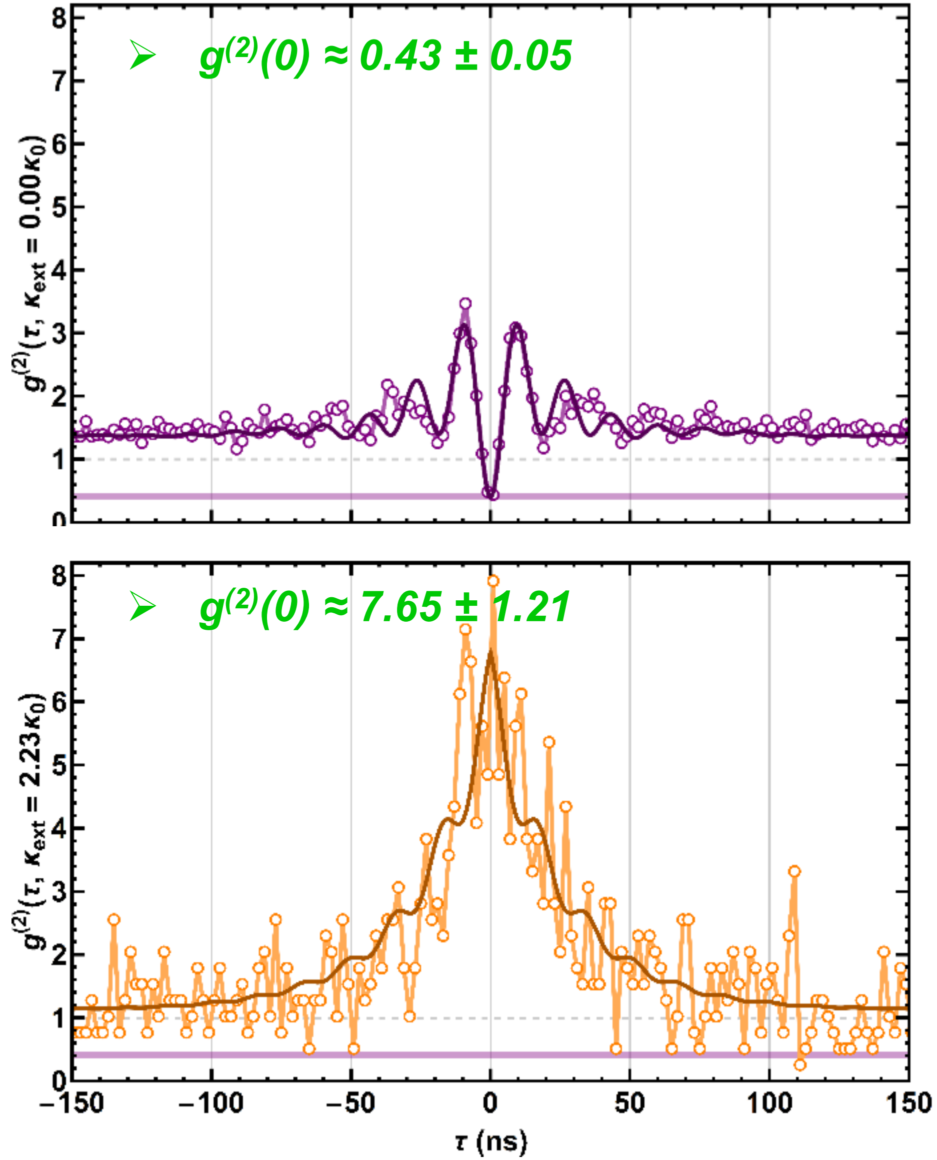
Length	~ 2.3 m
Linewidth	~ 4.4 MHz
FSR	~ 90 MHz
Δ	~ $^2/_3$FSR (previous slide)
(Finesse	~20)

In Summary...

- **Interference** between coherently and incoherently scattered two-photon component







Interpretation of Results

Will a single two-level emitter scatter 2 photons simultaneously?

→ Yes, it does so permanently, even in two different ways.

A classical linear notch filter can transform antibunched light into bunched light.

In other words: Selectively removing photons from antibunched light will cause those that pass through the filter to be bunched.

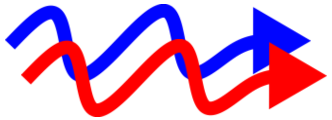
L. Masters *et al.*, Nat. Photon. **17**, 972 (2023)

Outline

Will a single two-level emitter scatter 2 photons simultaneously?

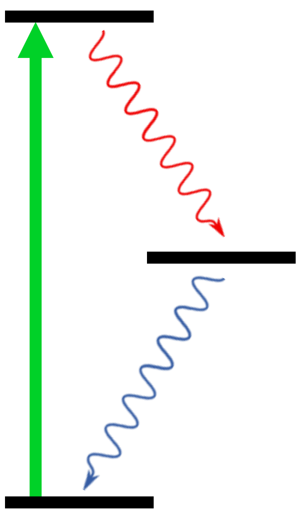
How to harness resonance fluorescence as a highly efficient source of time-bin entangled photon pairs.

Entangled Photon Pairs



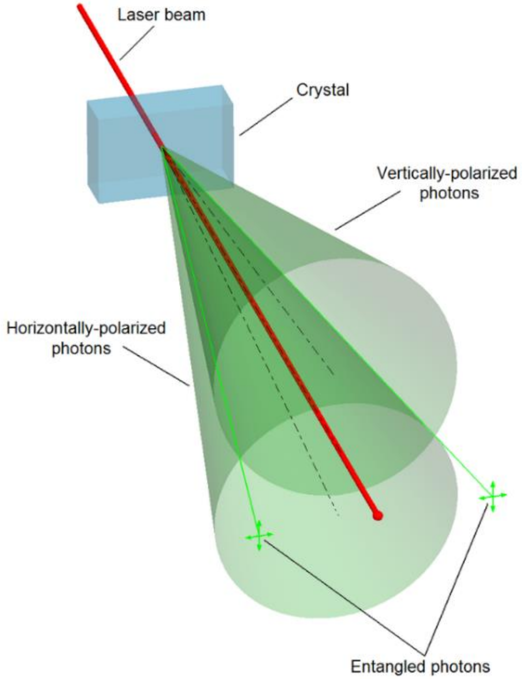
Usually generated by…

cascaded decay



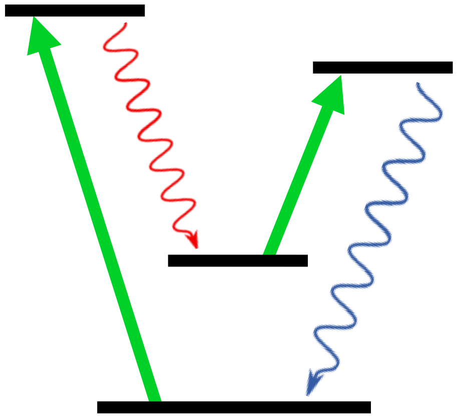
Phys. Rev. Lett. **28**, 938 (1972)
Phys. Rev. Lett. **47**, 460 (1981)

down conversion



Phys. Rev. Lett. **25**, 84 (1970)

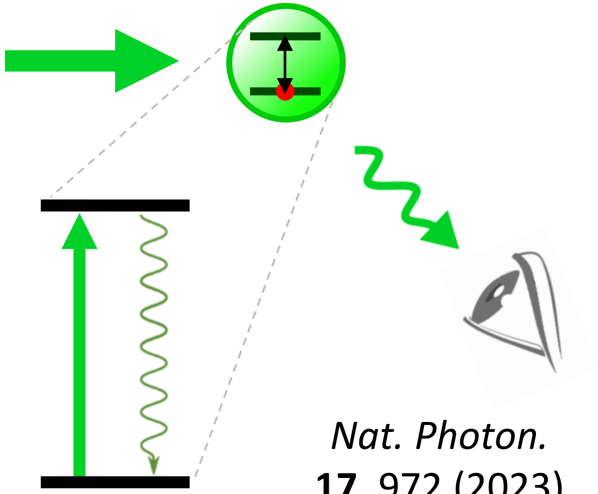
4-wave mixing
(e.g., in atomic gases)



Front. of Physics **7**, 494 (2012)

Novel approach

Resonance fluorescence of a single 2-level emitter



Nat. Photon. **17**, 972 (2023)

Entangled Photon Pairs

Novel approach

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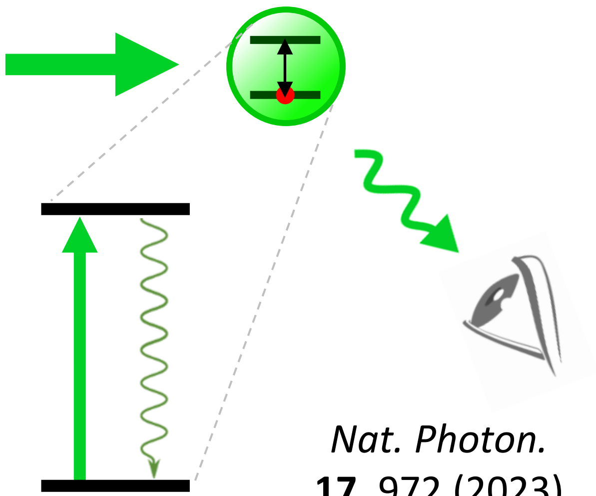
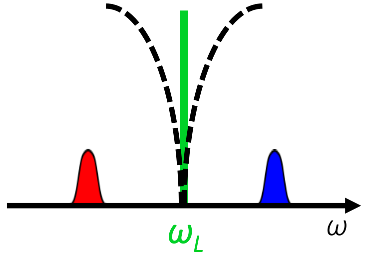


Nat. Photon. **17**, 972 (2023)

Spectral distribution

$$\tilde{\Psi}_2(\omega) = \cancel{\tilde{\chi}(\omega)} + \tilde{\phi}(\omega)$$

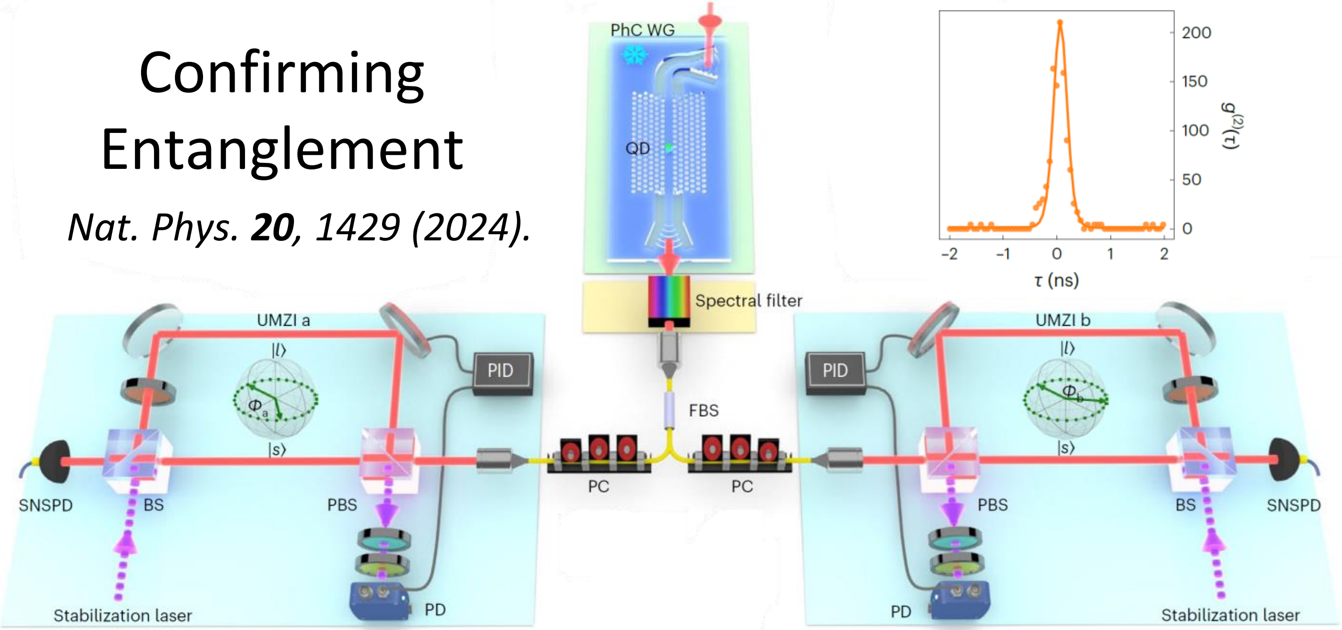


Incoherent scattering

$$\phi(\tau) = -\frac{n_{sc}}{2} e^{-(\gamma - i\Delta)|\tau|}$$

Confirming Entanglement

Nat. Phys. ***20****, 1429 (2024).*



Entangled Photon Pairs

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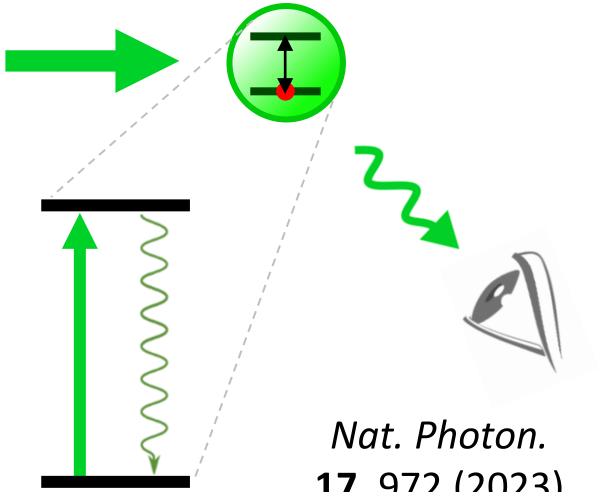

Nat. Photon. **17**, 972 (2023)

Spectral distribution

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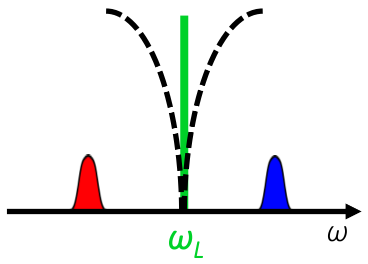
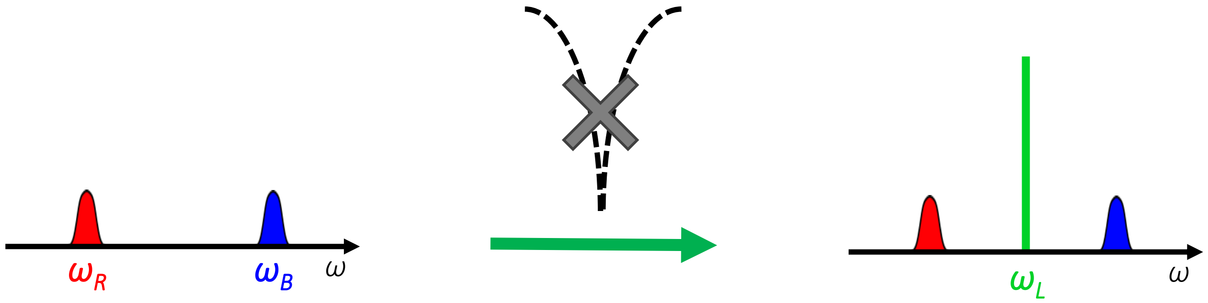


Incoherent scattering

$$\phi(\tau) = -\frac{n_{sc}}{2} e^{-(\gamma - i\Delta)|\tau|}$$

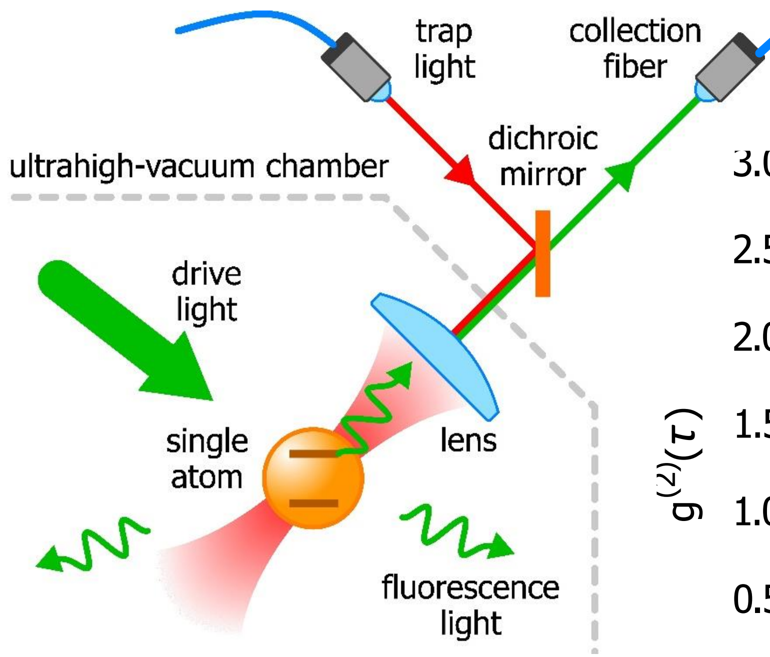
But, surprisingly, ...

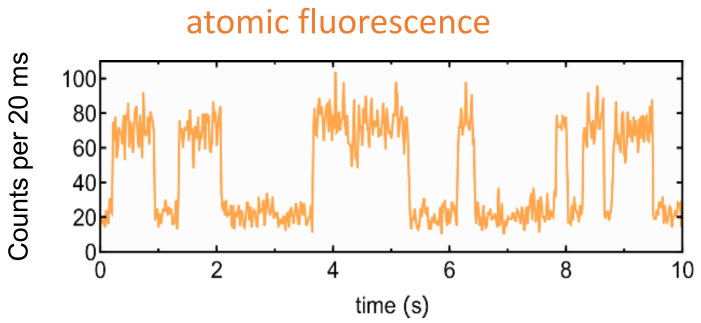


... a stream of time-bin entangled photon pairs can be generated from unfiltered resonance fluorescence!

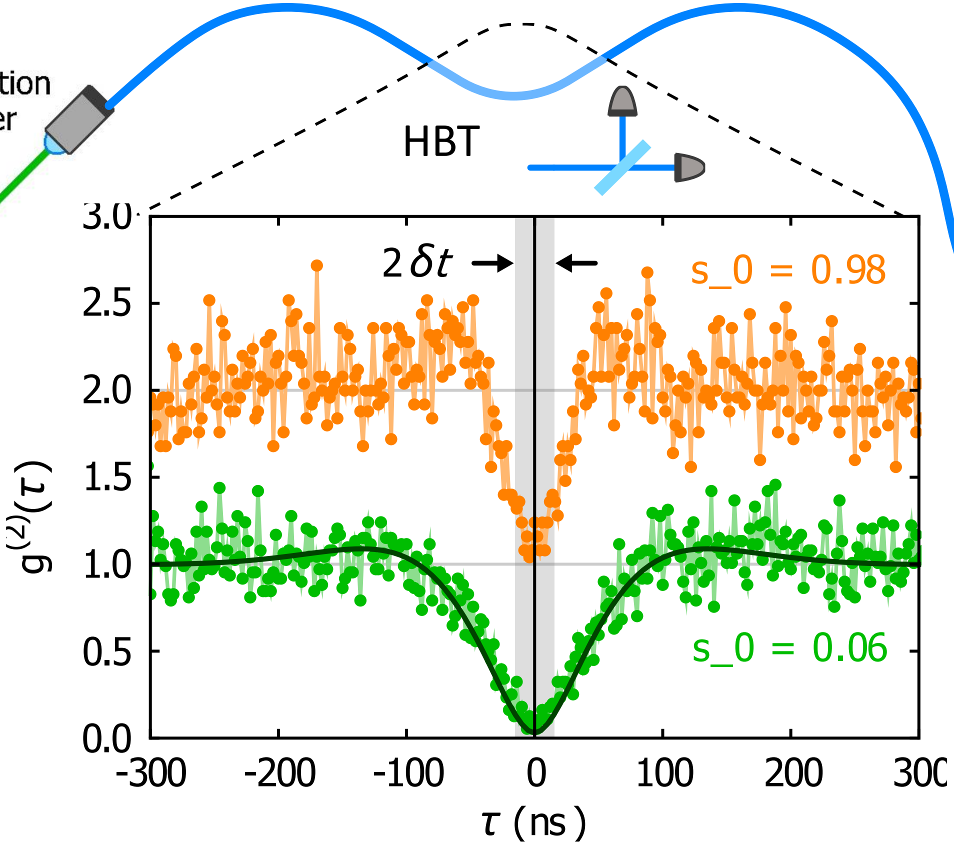
Setup

dipole trap for 85Rb





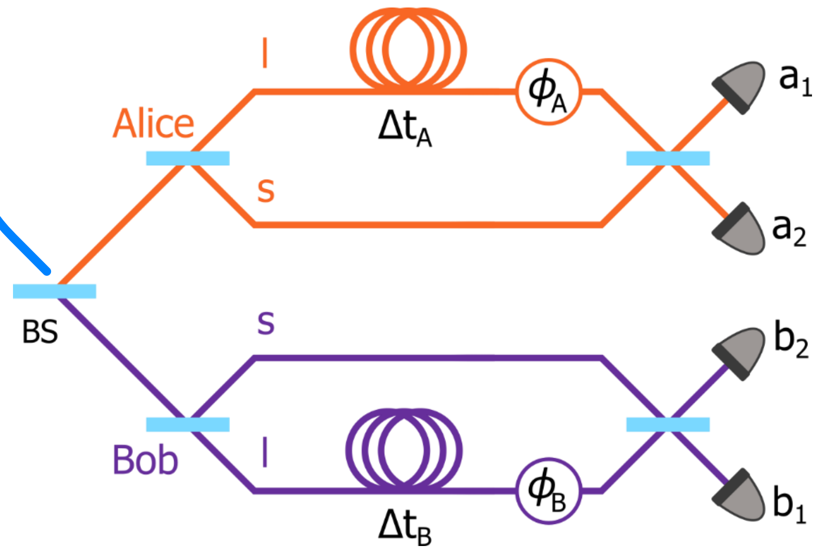
second-order correlations



low-saturation: $g^{(2)}(0) = 0.05 \pm 0.036$

high-saturation: $g^{(2)}(0) = 0.07 \pm 0.055$

Fiber-based Franson interferometer

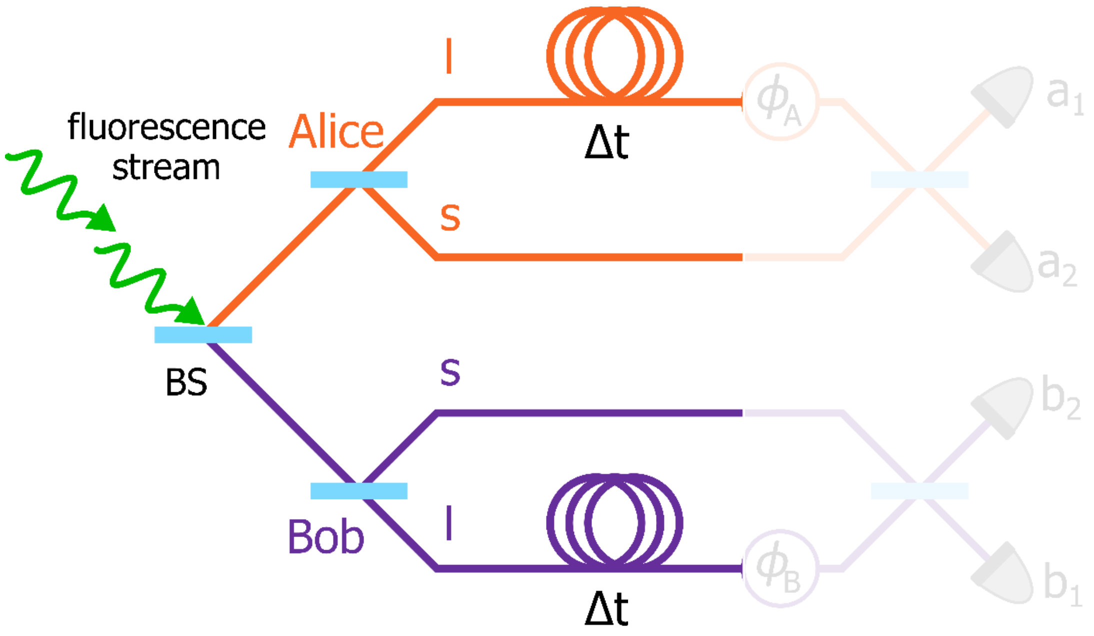


- ***Polarization compensated***
- ***Stabilized using phase shifter***
- ***Charaterized using drive laser***

Postselected Time-Bin Entangled State

Coincidence between Alice and Bob at time $t \pm \delta t$ projects onto state

$$|\Psi\rangle = \frac{1}{2}\psi(\Delta t)\left[a_s^\dagger(t)b_l^\dagger(\mathrm{t}) + a_l^\dagger(t)b_s^\dagger(\mathrm{t})\right]|0\rangle + \frac{1}{2}\psi(\delta t)\left[a_s^\dagger(t)b_s^\dagger(\mathrm{t}) + a_l^\dagger(t)b_l^\dagger(\mathrm{t})\right]|0\rangle$$



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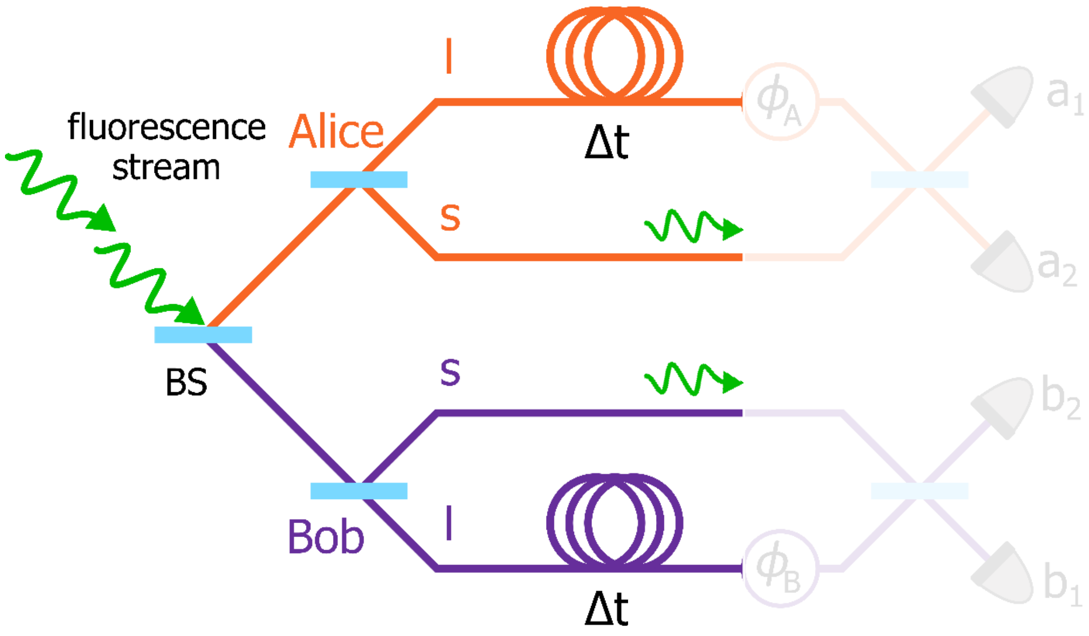

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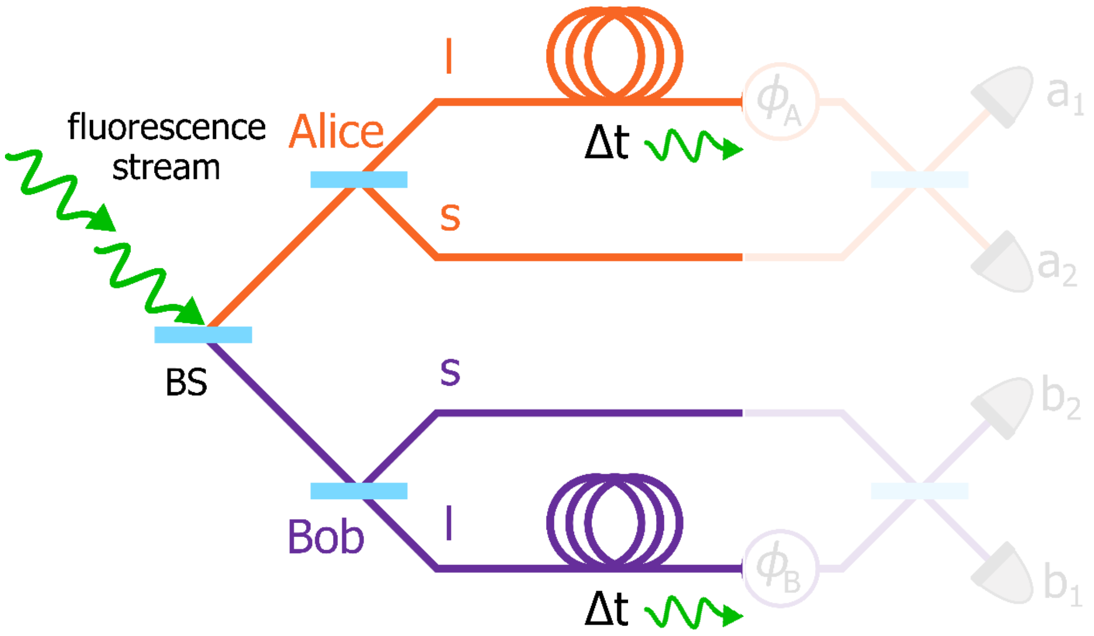

Postselected Time-Bin Entangled State

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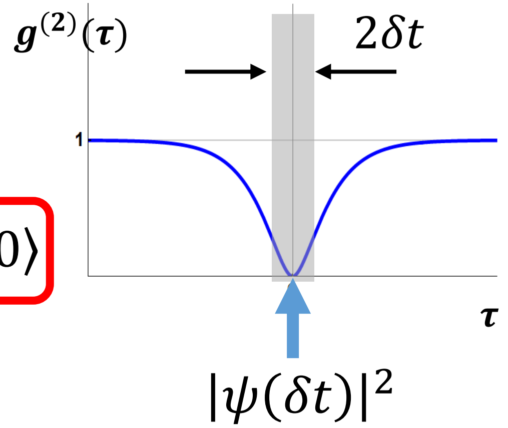
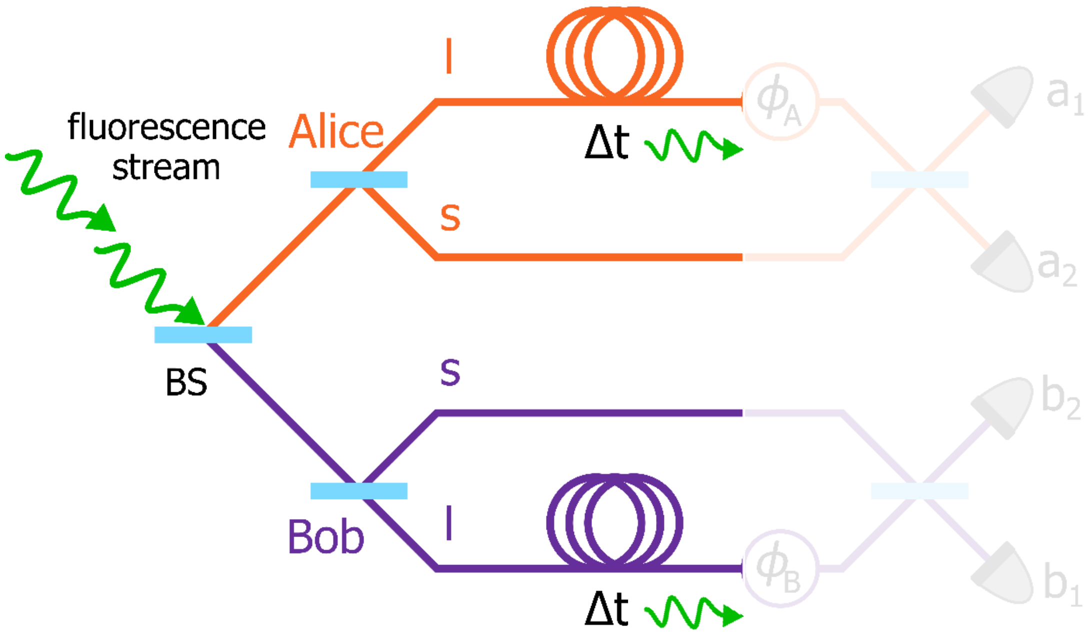


$$|\Psi\rangle = \frac{1}{2}\psi(\Delta t)\left[a_s^\dagger(t)b_l^\dagger(\mathrm{t}) + a_l^\dagger(t)b_s^\dagger(\mathrm{t})\right]|0\rangle + \boxed{\frac{1}{2}\psi(\delta t)\left[a_s^\dagger(t)b_s^\dagger(\mathrm{t}) + a_l^\dagger(t)b_l^\dagger(\mathrm{t})\right]|0\rangle}$$

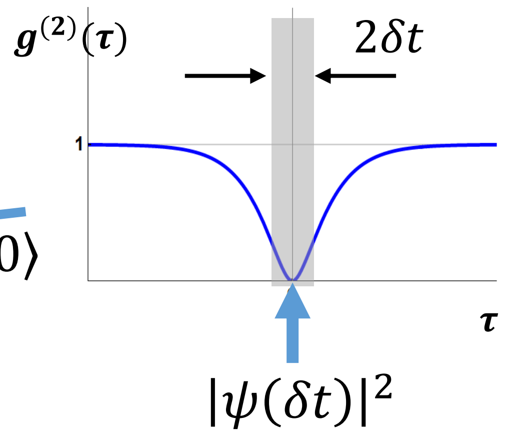


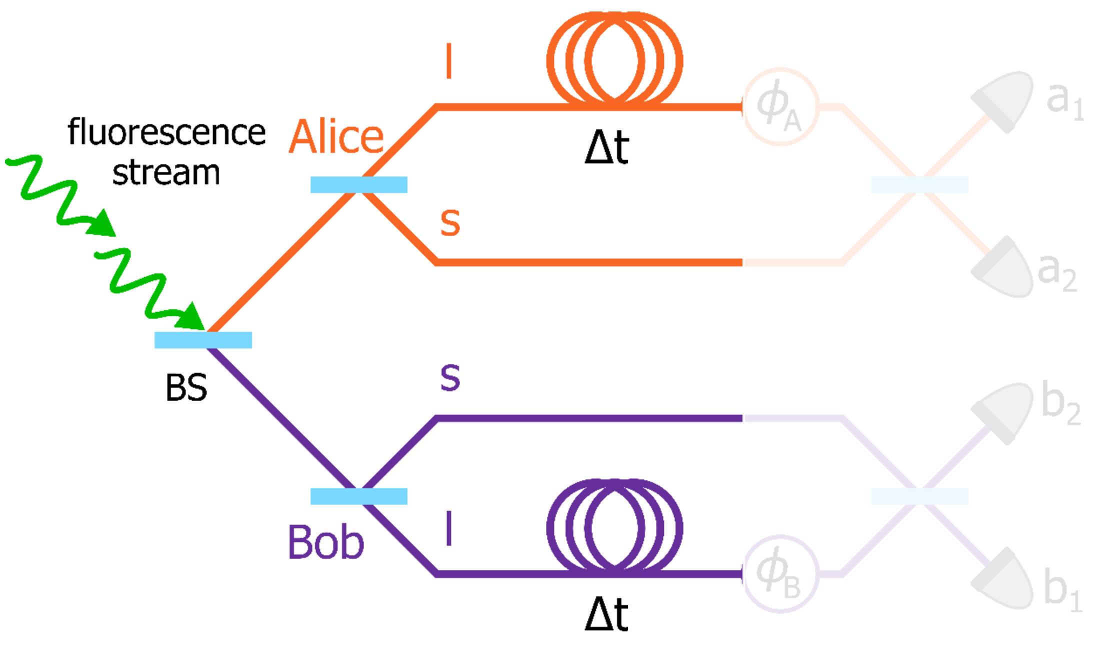
Postselected Time-Bin Entangled State

Coincidence between Alice and Bob at time $t \pm \delta t$ projects onto state

for $\delta t \ll 1/2\gamma$

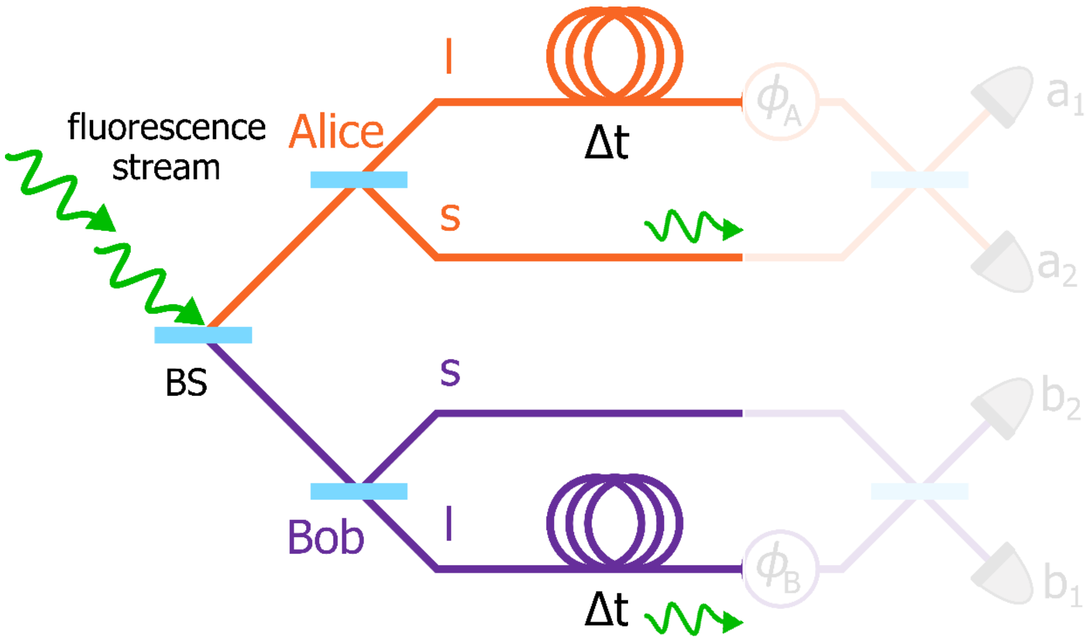
$$|\Psi\rangle = \frac{1}{2}\psi(\Delta t)\left[a_s^\dagger(t)b_l^\dagger(\mathrm{t}) + a_l^\dagger(t)b_s^\dagger(\mathrm{t})\right]|0\rangle + \frac{1}{2}\psi(\delta t)\left[a_s^\dagger(t)b_s^\dagger(t) + a_l^\dagger(t)b_l^\dagger(\mathrm{t})\right]|0\rangle$$





Postselected Time-Bin Entangled State

$$|\Psi\rangle = \frac{1}{2}\psi(\Delta t)\left[a_s^\dagger(t)b_l^\dagger(\mathrm{t}) + a_l^\dagger(t)b_s^\dagger(\mathrm{t})\right]|0\rangle$$



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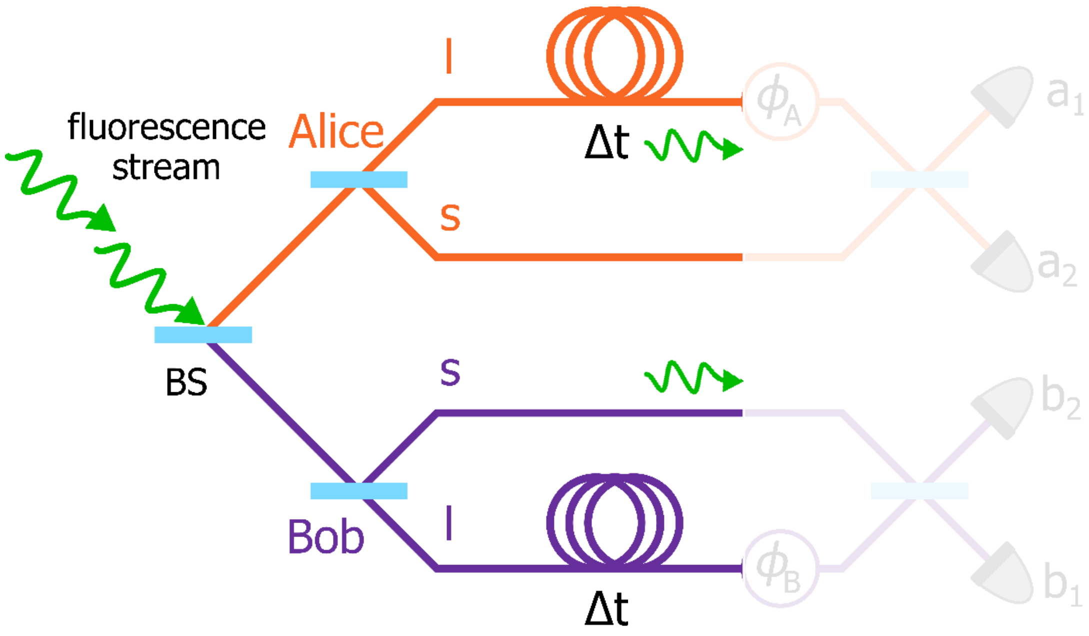

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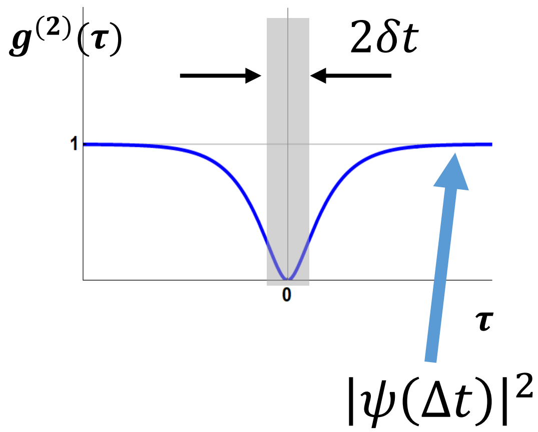

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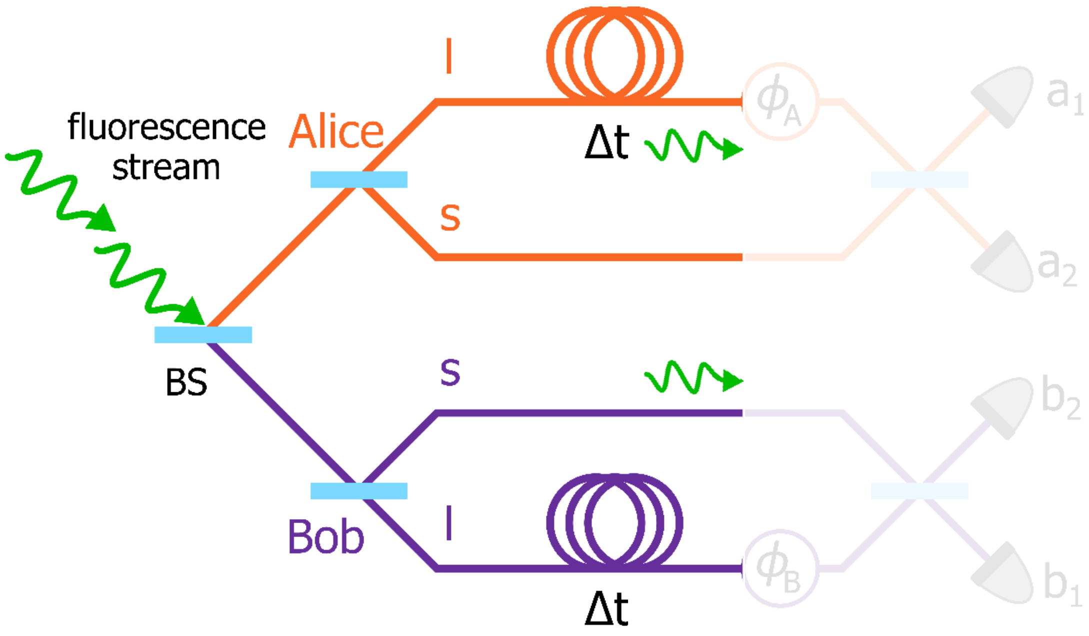
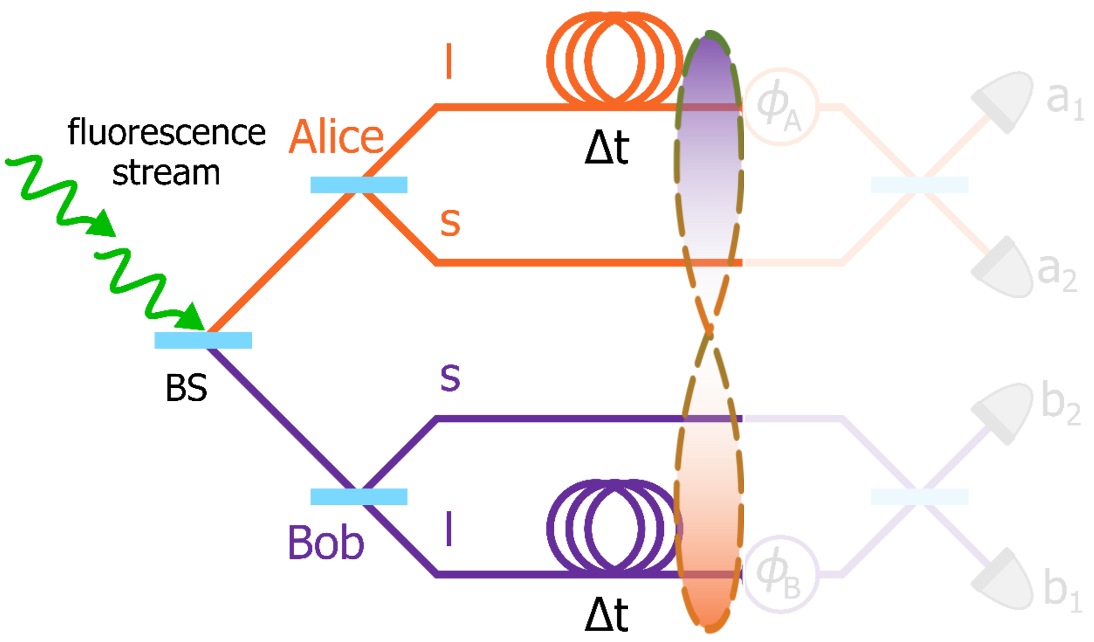
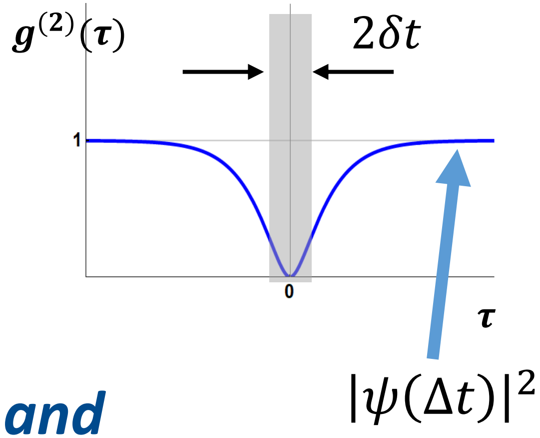


Postselected Time-Bin Entangled State

Bell state

$$|\Psi\rangle = \frac{1}{2}\psi(\Delta t)\left[a_s^\dagger(t)b_l^\dagger(\mathrm{t}) + a_l^\dagger(t)b_s^\dagger(\mathrm{t})\right]|0\rangle$$





A coincidence between Alice and Bob projects onto the Bell state

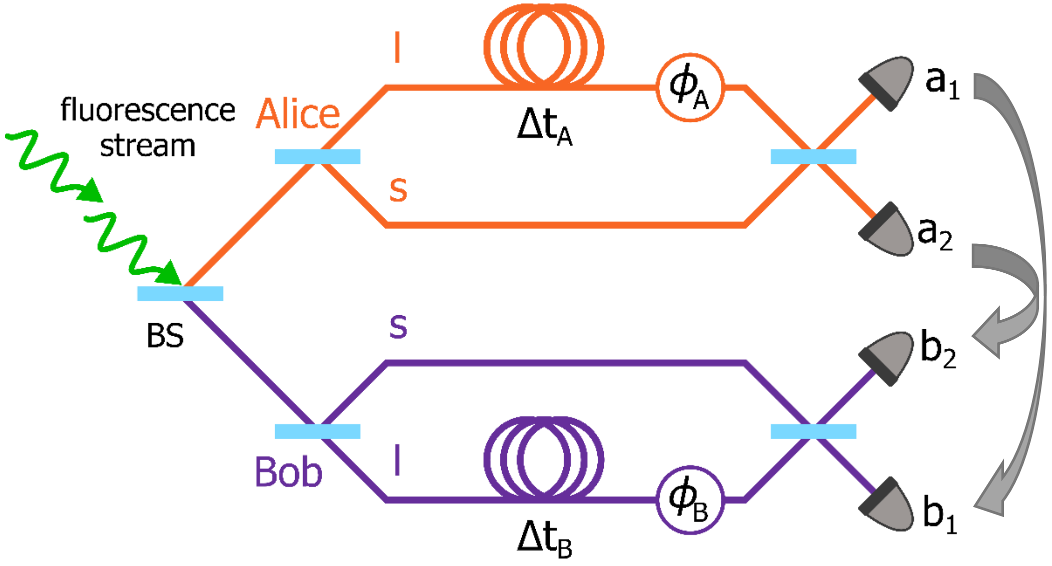
$$|\Psi_{\mathrm{Bell}}\rangle = \frac{1}{\sqrt{2}}(|s,l\rangle + |l,s\rangle)$$

The state is entangled for low- and high-saturation driving strength

CHSH Bell Inequality Test

- Measure S-parameter with proper phase settings:

$$S = \left|\langle\sigma_{\phi_A}\sigma_{\phi_B}\rangle\right| + \left|\langle\sigma_{\phi'_A}\sigma_{\phi_B}\rangle\right| + \left|\langle\sigma_{\phi_A}\sigma_{\phi'_B}\rangle\right| + \left|\langle\sigma_{\phi'_A}\sigma_{\phi'_B}\rangle\right|$$



$$\langle\sigma_{\phi_A}\sigma_{\phi_B}\rangle = \frac{N_{a_1,b_1} + N_{a_2,b_2} - N_{a_1,b_2} - N_{a_2,b_1}}{N_{a_1,b_1} + N_{a_2,b_2} + N_{a_1,b_2} + N_{a_2,b_1}}$$

CHSH Bell Inequality Test

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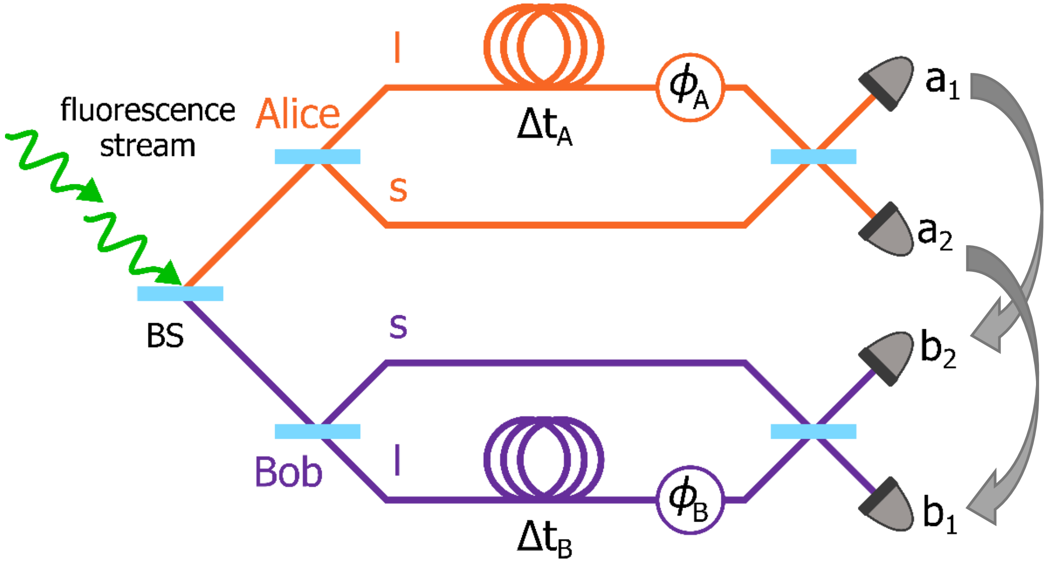
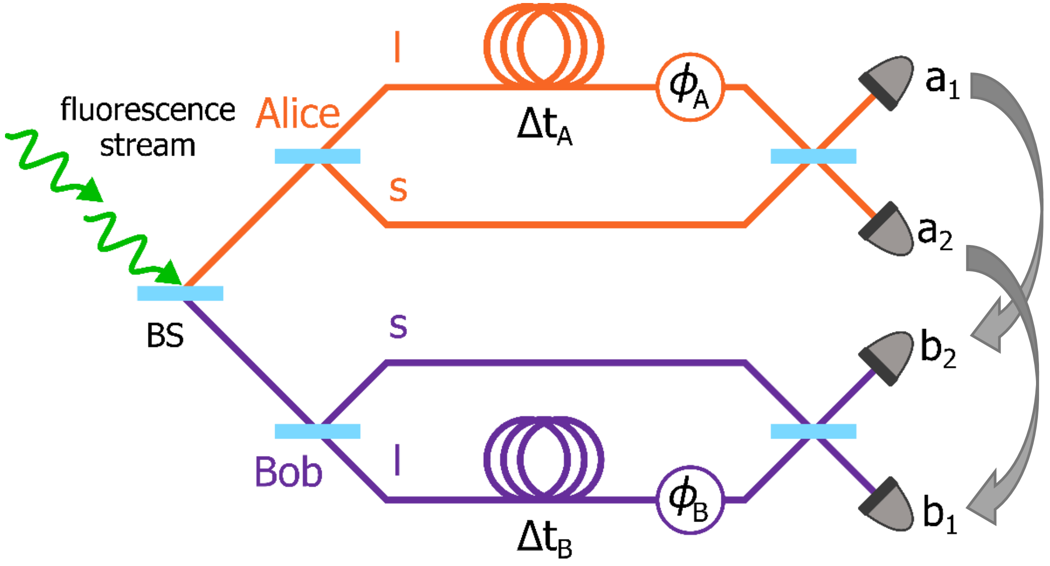


$$\langle\sigma_{\phi_A}\sigma_{\phi_B}\rangle = \frac{N_{a_1,b_1} + N_{a_2,b_2} - N_{a_1,b_2} - N_{a_2,b_1}}{N_{a_1,b_1} + N_{a_2,b_2} + N_{a_1,b_2} + N_{a_2,b_1}}$$

X.-X. Hu *et al.*, arXiv:2504.11294

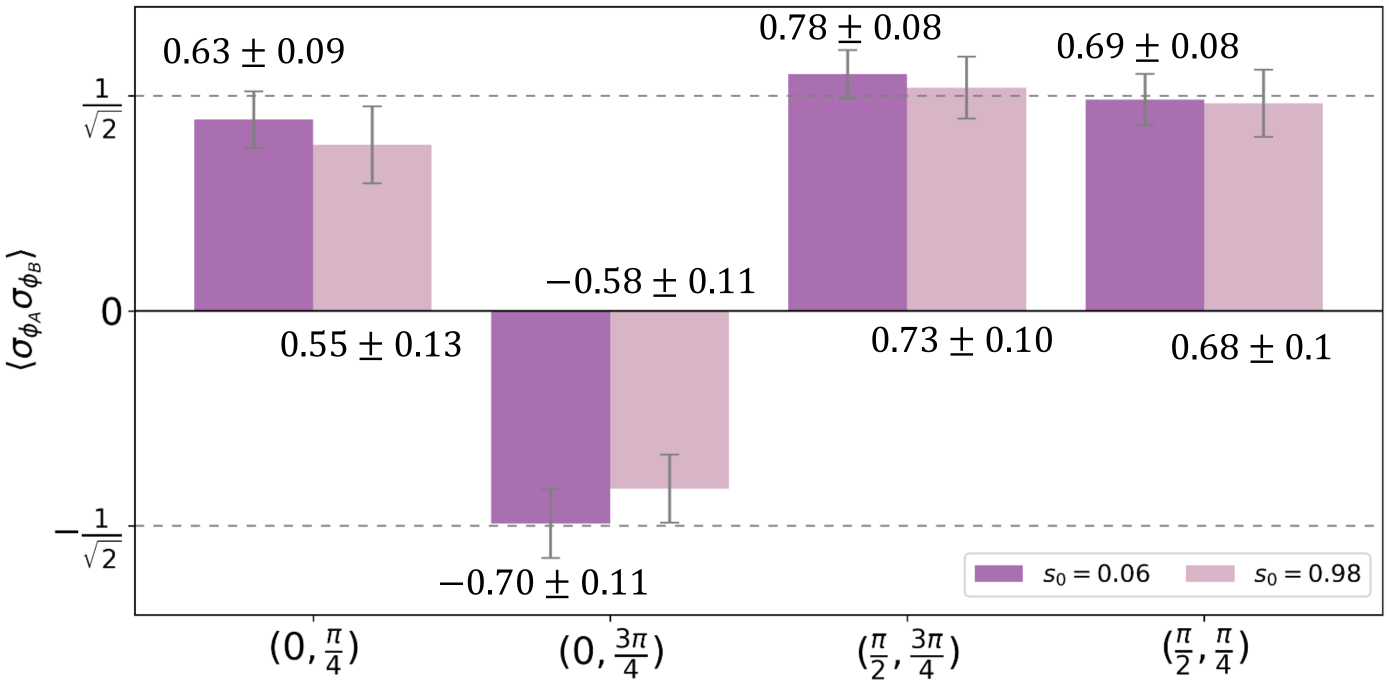
CHSH Bell Inequality Test



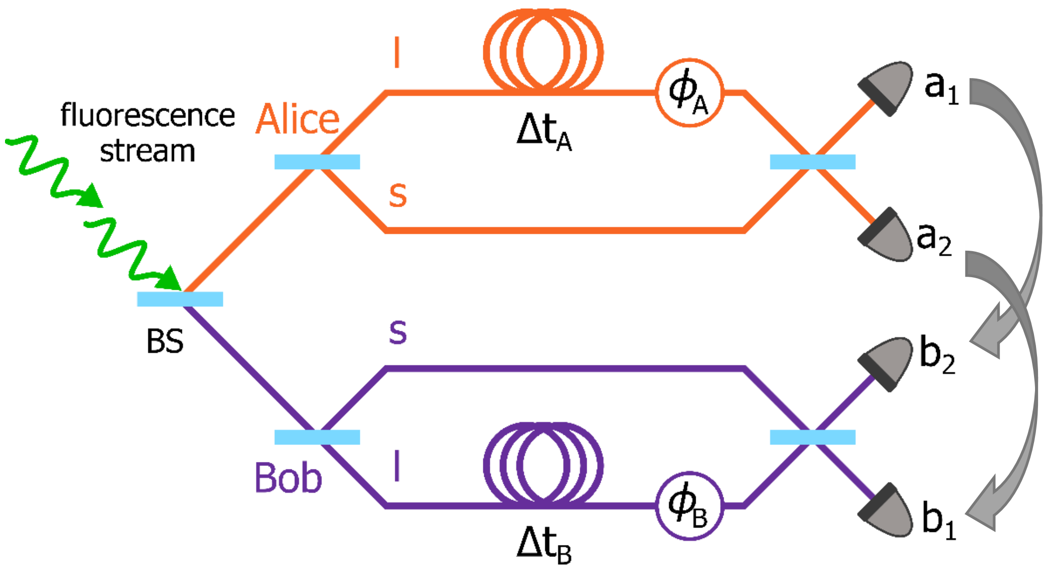
$$\langle \sigma_{\phi_A} \sigma_{\phi_B} \rangle = \frac{N_{a_1,b_1} + N_{a_2,b_2} - N_{a_1,b_2} - N_{a_2,b_1}}{N_{a_1,b_1} + N_{a_2,b_2} + N_{a_1,b_2} + N_{a_2,b_1}}$$

- Measure S-parameter with proper phase settings:

$$S = |\langle \sigma_{\phi_A} \sigma_{\phi_B} \rangle| + |\langle \sigma_{\phi'_A} \sigma_{\phi_B} \rangle| + |\langle \sigma_{\phi_A} \sigma_{\phi'_B} \rangle| + |\langle \sigma_{\phi'_A} \sigma_{\phi'_B} \rangle|$$



CHSH Bell Inequality Test



$$\langle \sigma_{\phi_A} \sigma_{\phi_B} \rangle = \frac{N_{a_1,b_1} + N_{a_2,b_2} - N_{a_1,b_2} - N_{a_2,b_1}}{N_{a_1,b_1} + N_{a_2,b_2} + N_{a_1,b_2} + N_{a_2,b_1}}$$

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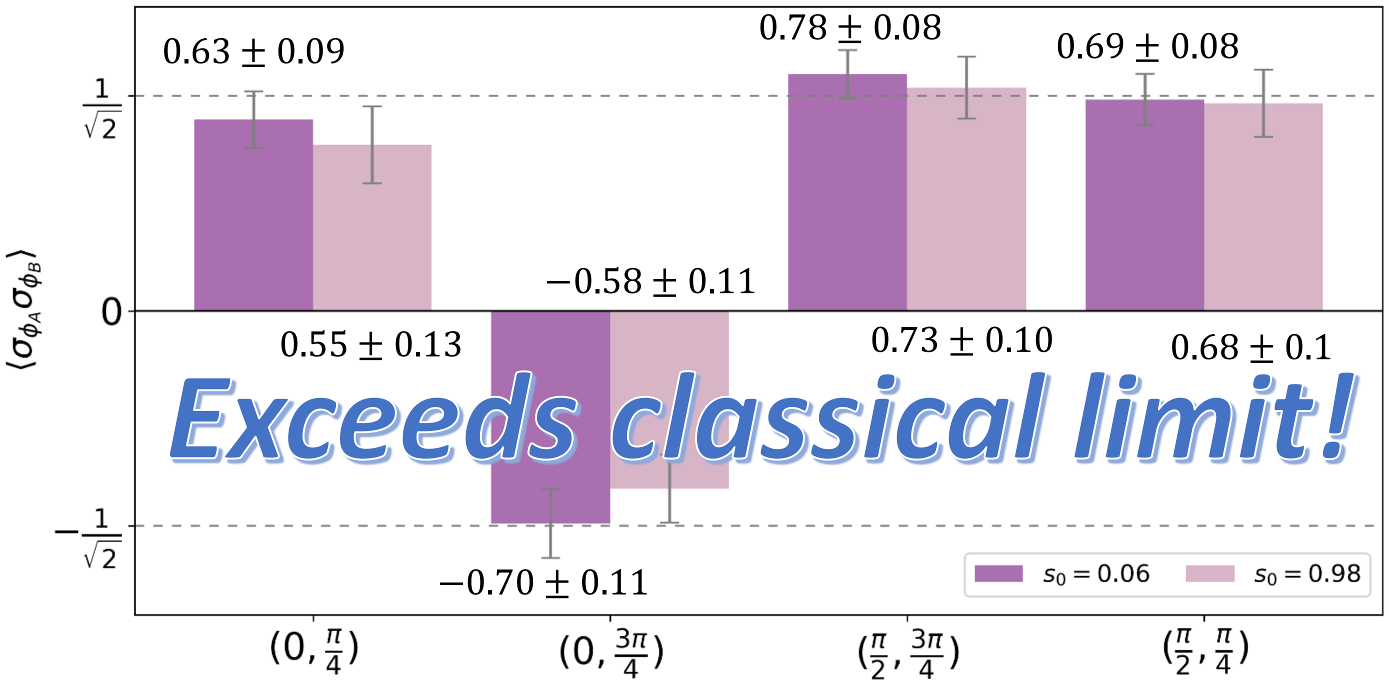


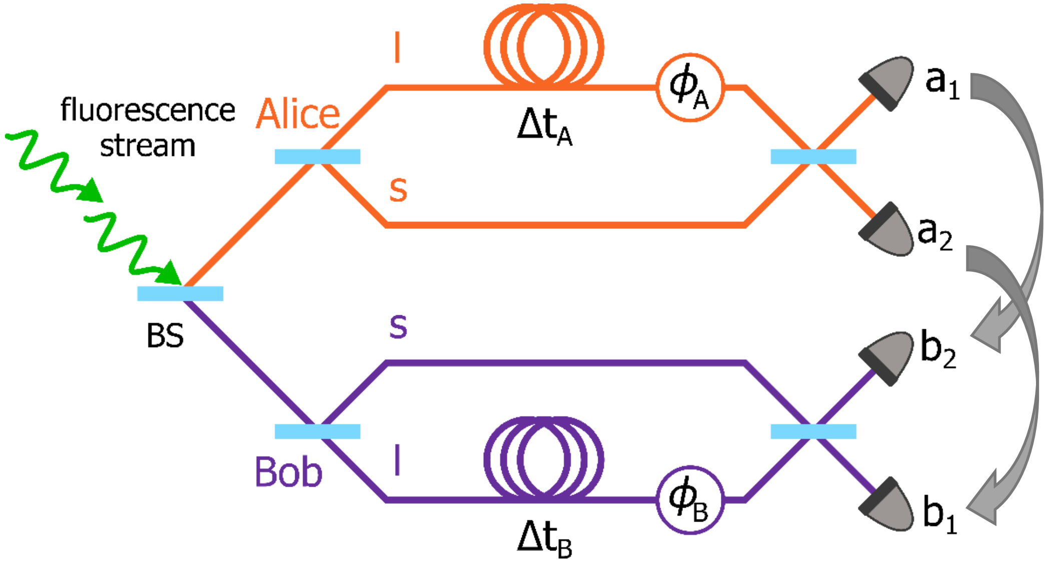
➡ For low-saturation regime: $S = 2.80 \pm 0.19$

➡ For high-saturation regime: $S = 2.55 \pm 0.22$

X.-X. Hu *et al.*, arXiv:2504.11294

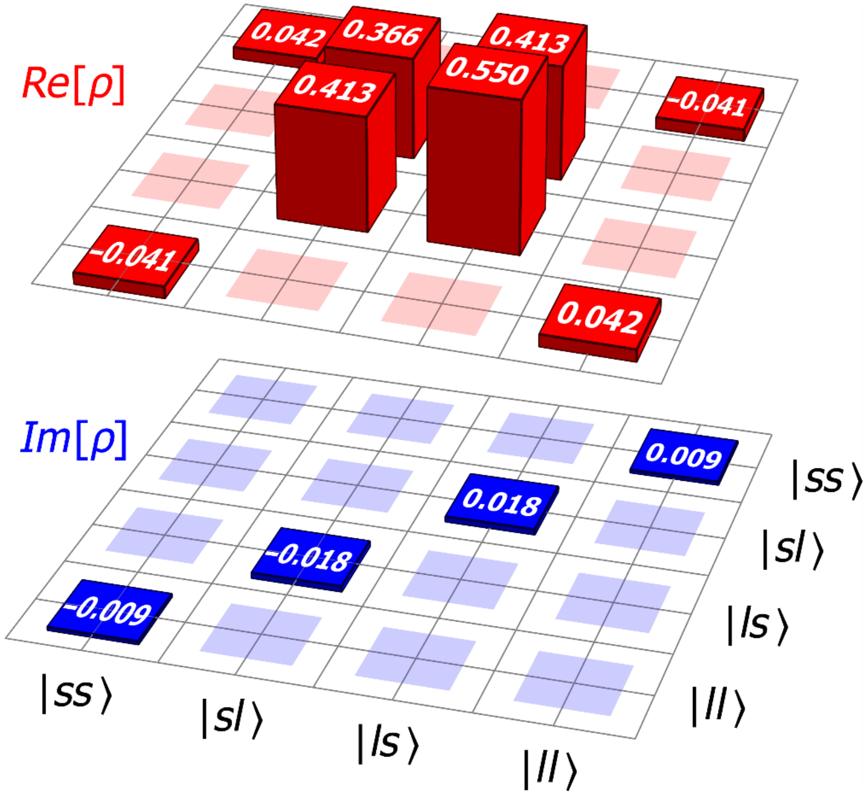
Quantum State Tomography

- Reconstruct density matrix using maximum likelihood estimation



- Set phase to: $(0,0), (0,\frac{\pi}{2}), (\frac{\pi}{2},0), (\frac{\pi}{2},\frac{\pi}{2})$

For measuring $\{\sigma_i \otimes \sigma_j\}, (i,j = x,y)$



Bell state $|\Psi_{\text{Bell}}\rangle = \frac{1}{\sqrt{2}}(|s,l\rangle + |l,s\rangle)$

Fidelity $F = \langle \Psi_{\text{Bell}}|\rho|\Psi_{\text{Bell}}\rangle = 0.87 \pm 0.02$

Conclusions

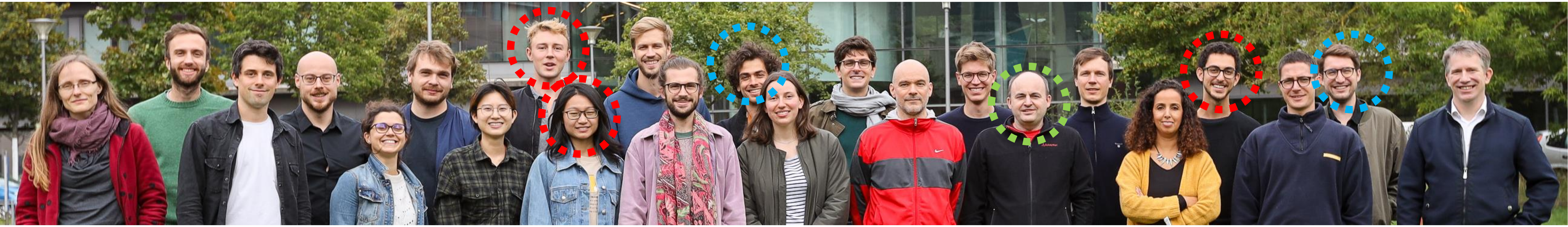
- Antibunching in resonance fluorescence originates from quantum interference between coherent & incoherent 2-photon component.
- Time-bin entangled photons can be postselected from resonance fluorescence using only beam splitters, delay lines, and coincidence detection.
- Entanglement and antibunching are typically considered to be distinct quantum phenomena but are closely related.

Perspectives

- Generated photon pairs are spectrally narrowband and thus naturally compatible with atomic quantum memories.
- Photon pair rates can reach Fourier limit without suffering from increasing multi-photon events at higher drive strengths.
- Scheme also applicable to other first-order coherent antibunched light sources. (Patent pending)

Thanks!

Fundamentals of Optics and Photonics



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UNTERSTÜTZT VON / SUPPORTED BY

Alexander von HUMBOLDT STIFTUNG

DAALI

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