

Rydberg mediated interactions: atom-by-atom and photon-by-photon

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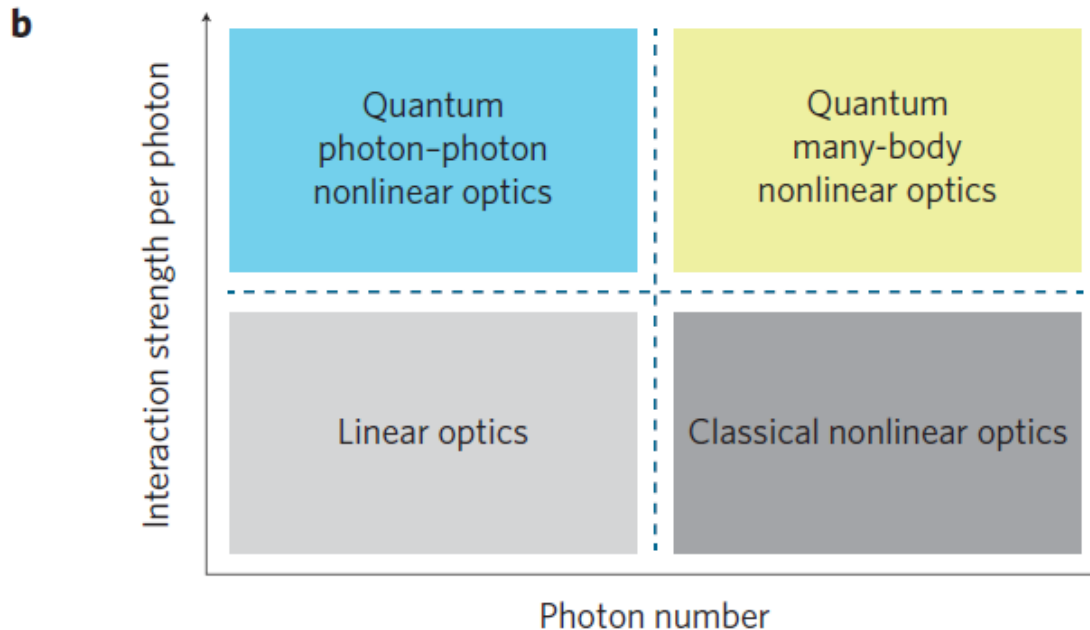
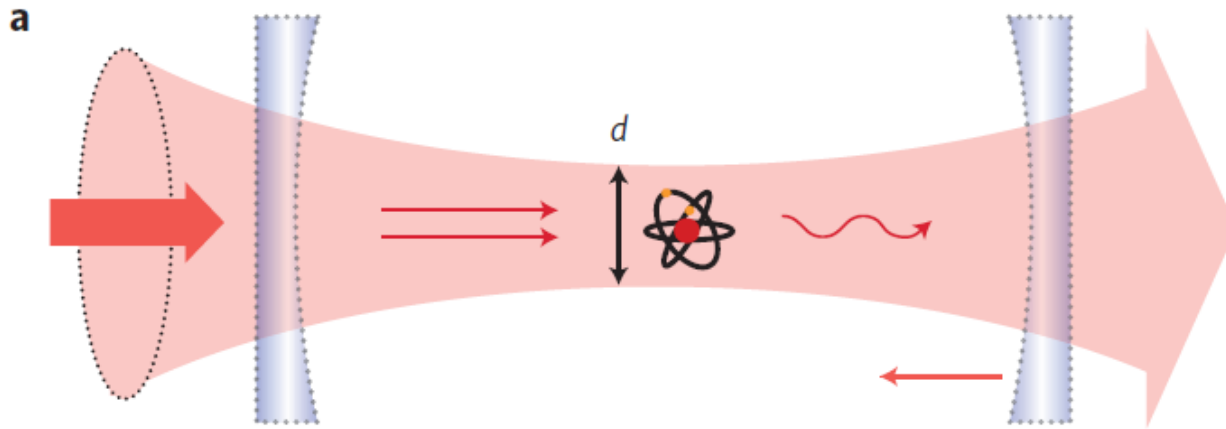
Rydberg mediated interactions: atom-by-atom and photon-by-photon

- **Photon by Photon:** Use Rydberg interaction in combination with electromagnetically induced transparency to implement quantum nonlinear optics
- **Atom by Atom:** Use Rydberg interactions in array of individually trapped atoms to realize strongly interacting spin models

What is quantum nonlinear optics?

- Optical nonlinearities are very interesting and useful: sum frequency generation, frequency doubling, photon frequency conversion, ...
- Even highly nonlinear standard optical media (nonlinear crystals) require very large numbers of photons.
- Can we imagine having optical nonlinearities at the level of individual photons?
- Would be of value for fundamental understanding and useful for quantum network, small quantum information processors, ...

Quantum nonlinear optics

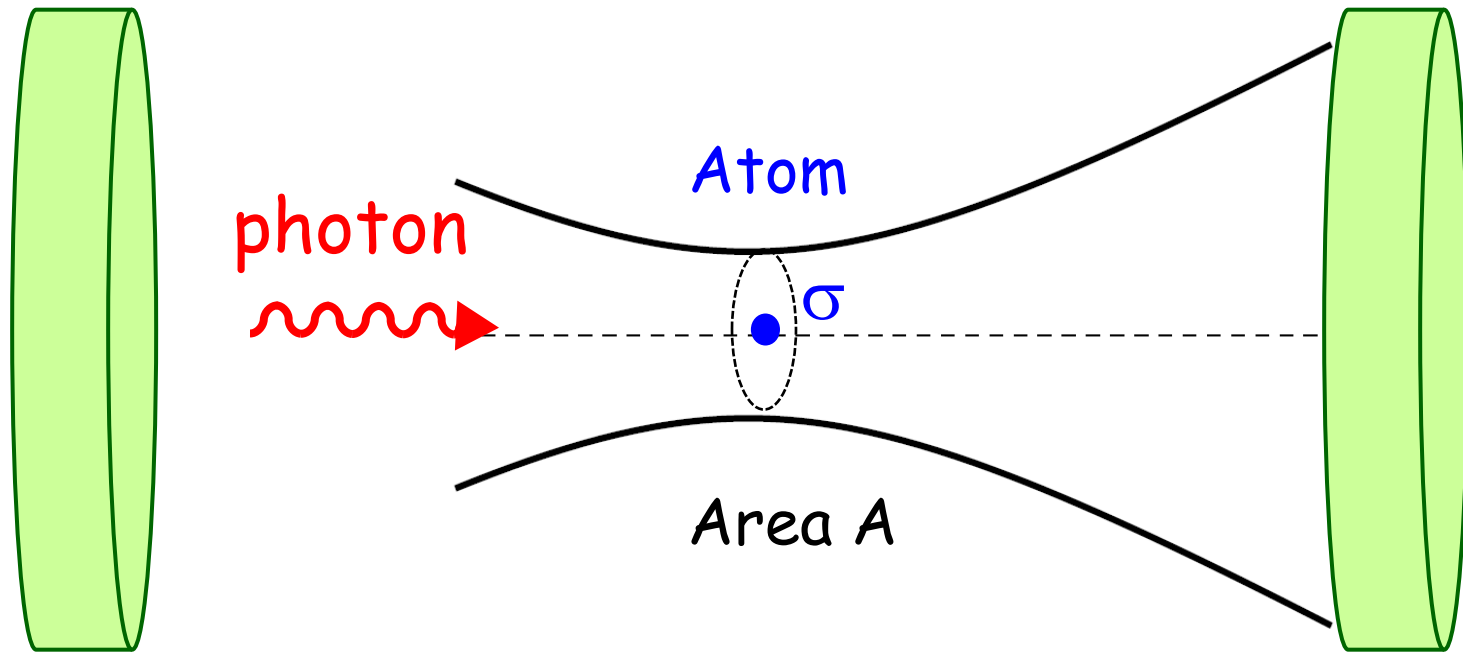


Quantum Nonlinear Optics – Photon by Photon. D. Chang, V. Vuletić, and M.D. Lukin, *Nature Photonics* **8**, 685–694 (2014).

The problem of quantum nonlinear optics

- Photons in free space just pass through one another (almost).
- Photon-photon interaction must be mediated by matter.
- If photon-photon interaction is to be coherent (reversible), then photon-matter interaction must be coherent.
- If interaction is to be nonlinear at level of one photon, it must involve single atoms or single excitations.
- But single atoms or single excitations interact only weakly with single photons.

Absorption requirement for atom-photon interaction



Absorption probability σ/A needs to exceed 1

Problem:

$$\sigma_{\max} = 3\lambda^2/2\pi < \lambda^2$$

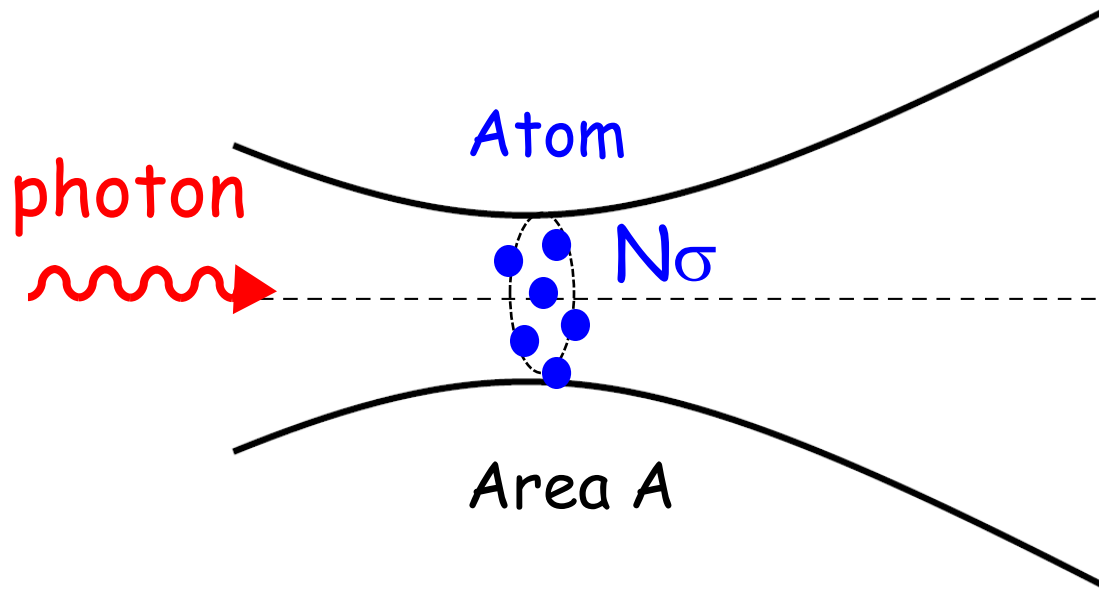
$$A > \lambda^2$$

(resonant atomic cross section)

(diffraction limit for focusing)

Need M multiple passes for $M\sigma/A > 1$ (cavity)

Absorption requirement for coherent atom-photon interaction



Absorption probability σ/A needs to exceed 1

Problem:

$$\sigma_{\max} = 3\lambda^2/2\pi < \lambda^2$$

$$A > \lambda^2$$

(resonant atomic cross section)

(diffraction limit for focusing)

N atoms for $N\sigma/A > 1$

But with N atoms the nonlinearity is only weak.

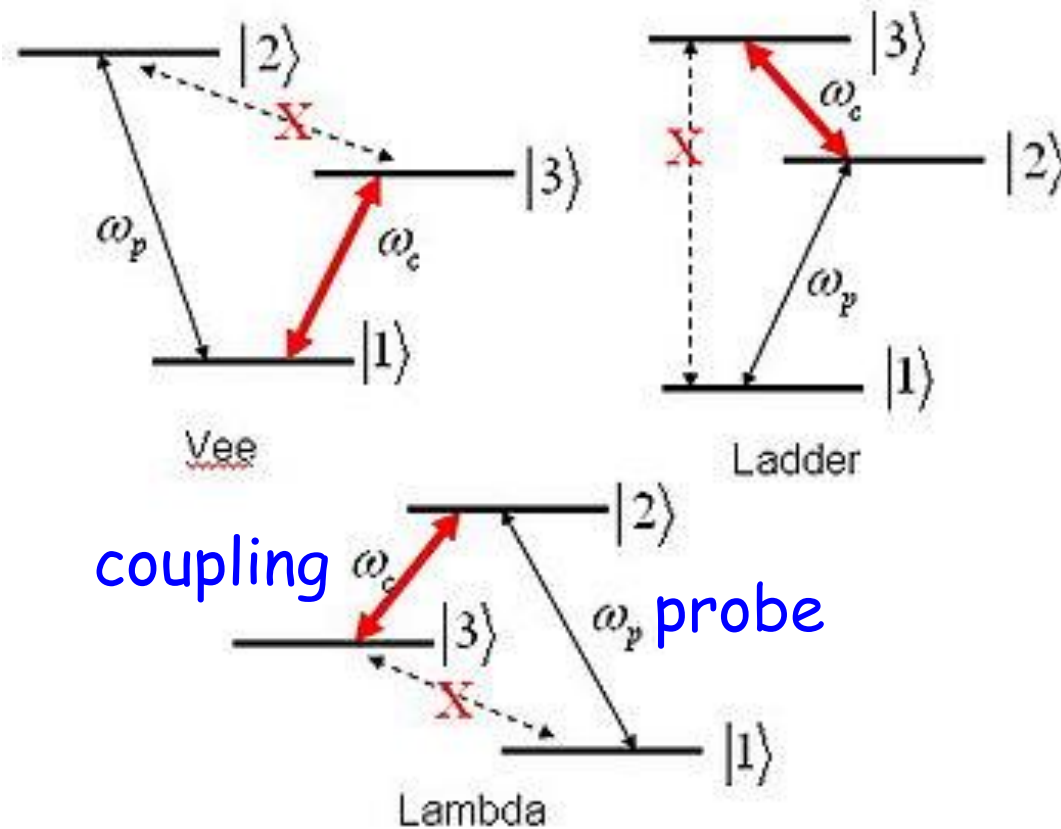
Outline

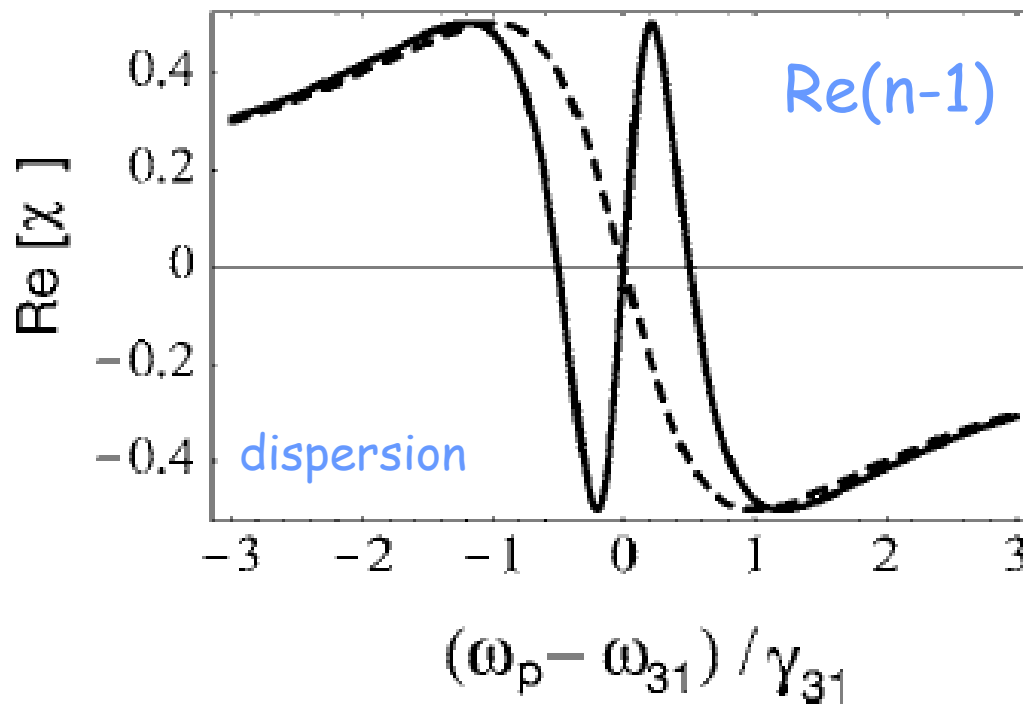
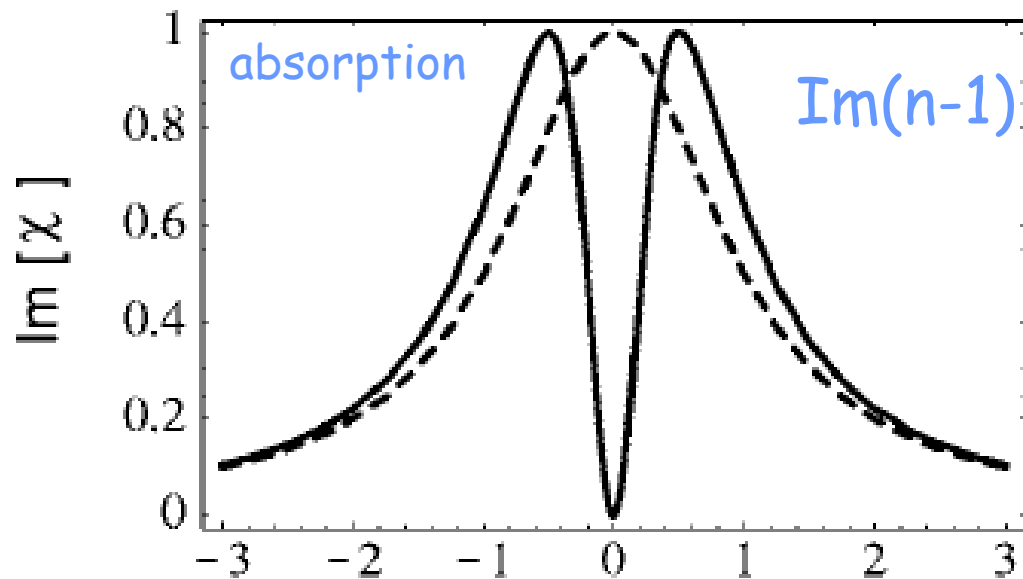
- Electromagnetically induced transparency: mapping photons coherently onto collective excitations
- Rydberg mediated photon-photon interactions
 - Dissipative interactions: a two-photon absorber
 - Dispersive interactions: bound state of two and three photons
 - Dispersive interactions: repulsive interactions between photons

Electromagnetically induced transparency (EIT)

M. Fleischhauer, A. Imamoglu, J. Marangos, Rev. Mod. Phys. **77**, 633 (2005).

Level schemes in EIT





Sharp variation of transmission and dispersion with frequency in an otherwise opaque medium

Electromagnetically induced transparency

$$v_g = \frac{c}{n + \omega \frac{\partial n}{\partial \omega}}$$

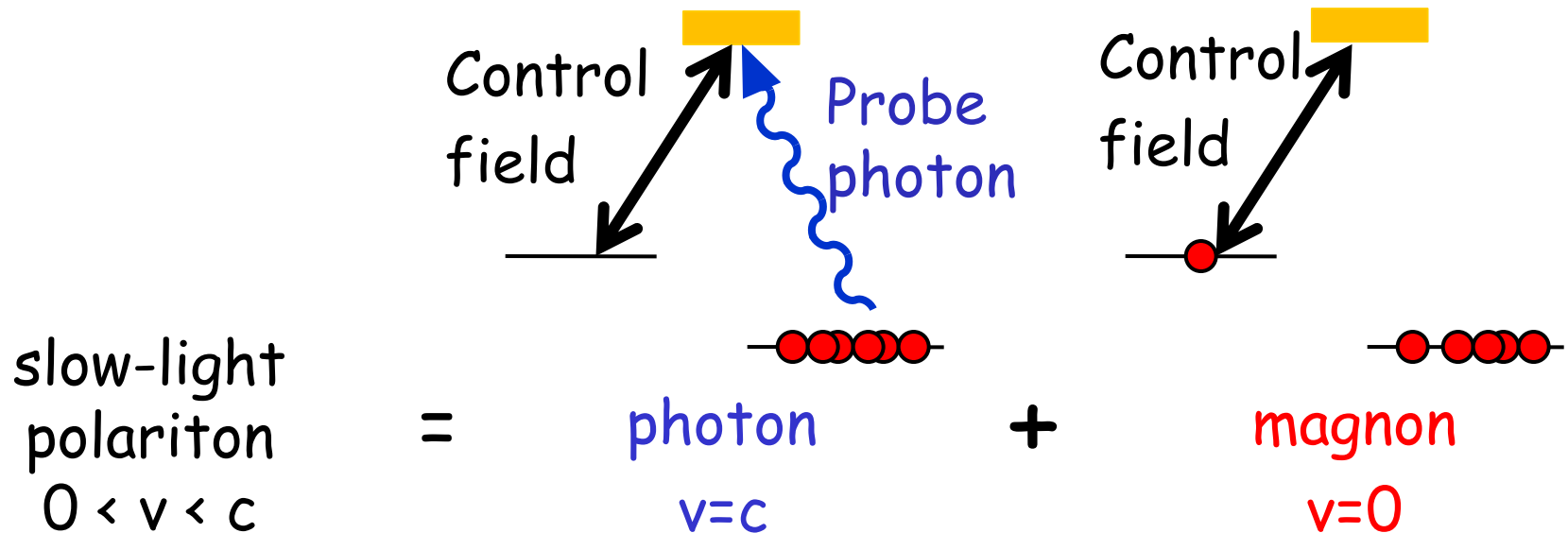


EIT for lasers without inversion
1989

Steve Harris

Electromagnetically Induced Transparency

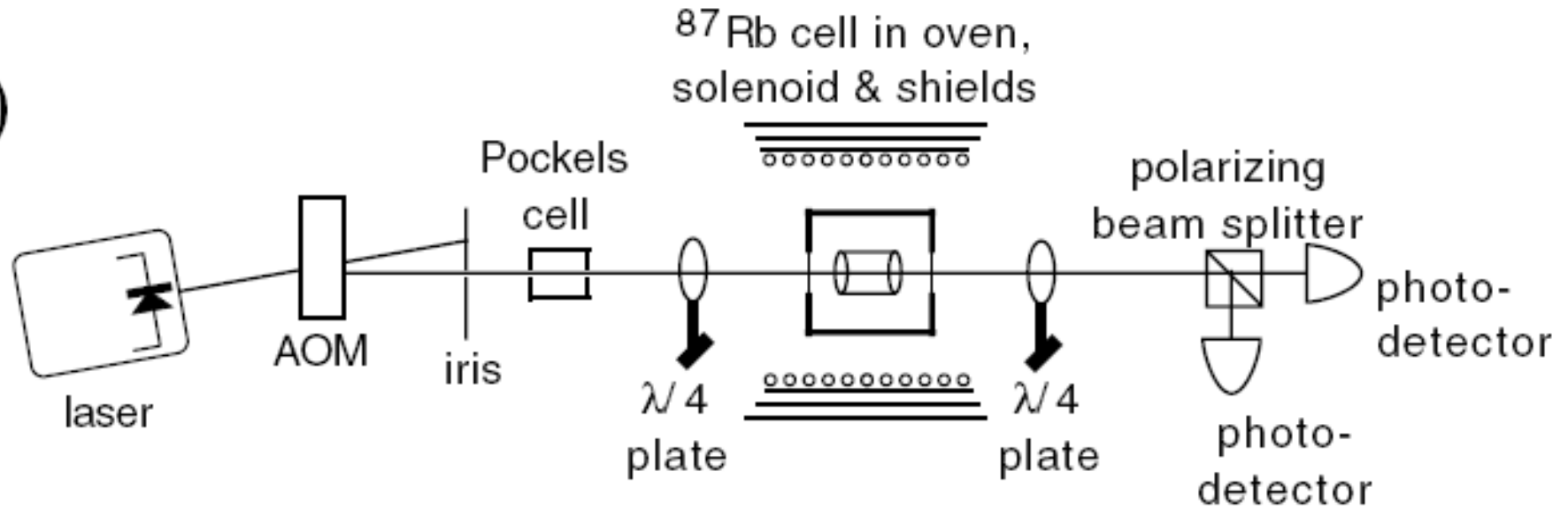
- EIT produces slow light by converting photons into collective atomic (spin) excitations



Collective (phase coherent excitation) \rightarrow preserves direction of light travel

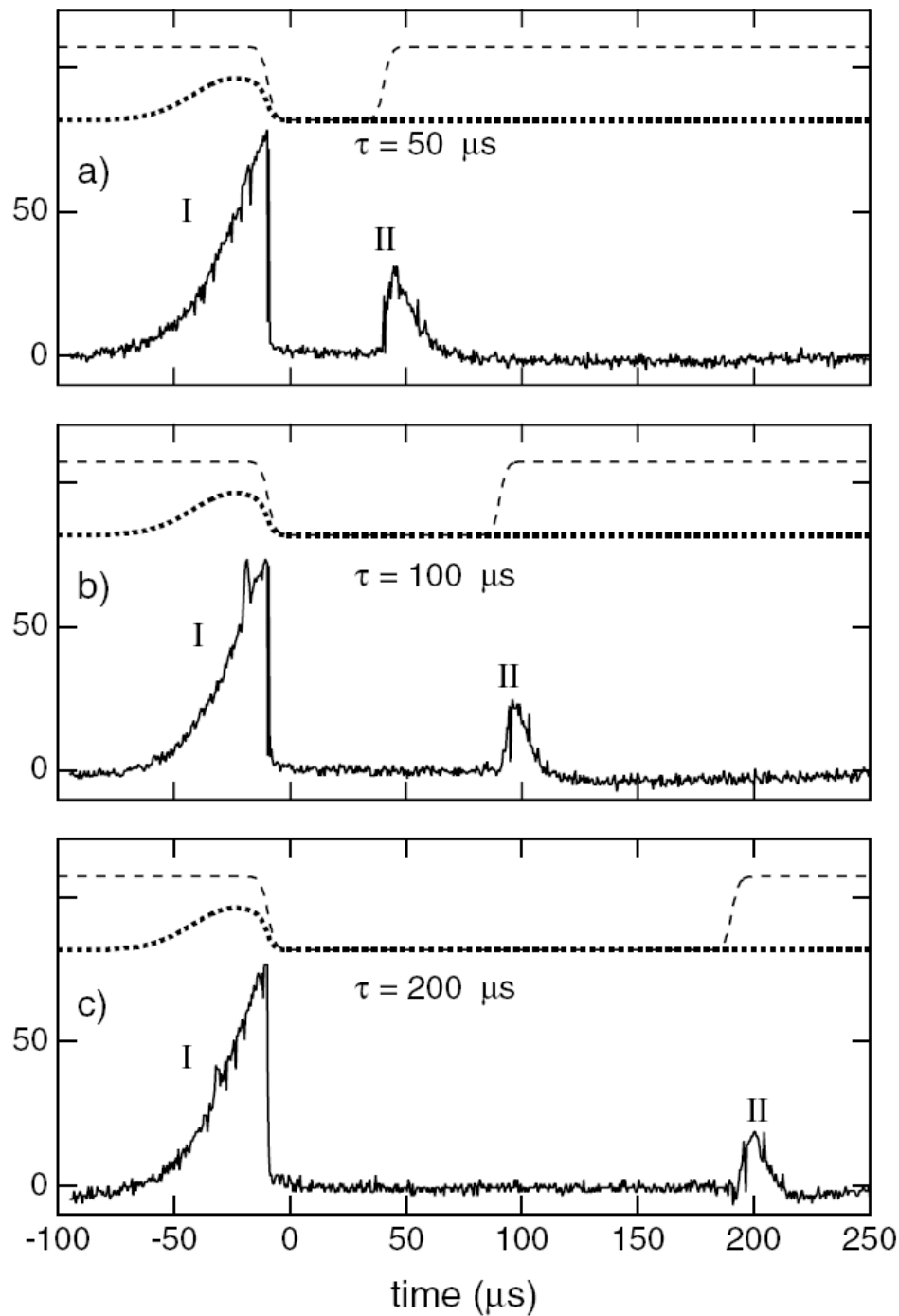
Light storage

c)



R. Walsworth et al.

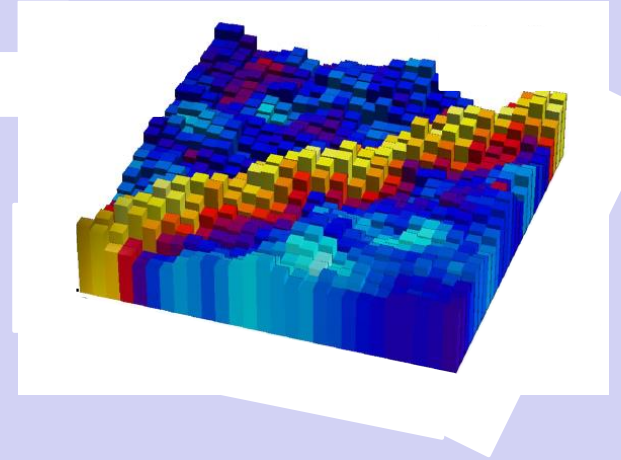
Stored light



Photon-photon interactions mediated by atomic Rydberg interactions

Very attractive photons

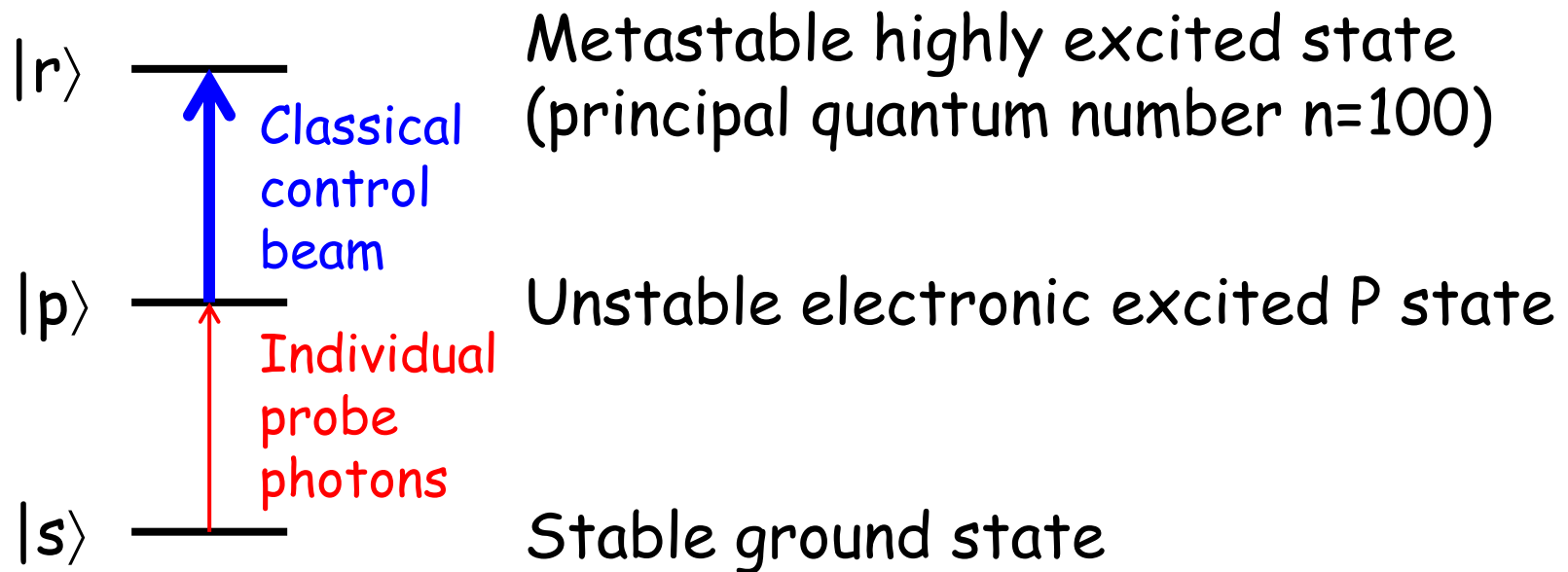
Joint experiment with Mikhail Lukin's group (Harvard)



T. Peyronel, O. Firstenberg, Q.-Y. Liang, S. Hofferberth, A.V. Gorshkov, T. Pohl, M.D. Lukin, and V. Vuletić, *Nature* **488**, 57-60 (2012).

O. Firstenberg, T. Peyronel, Q.-Y. Liang, A.V. Gorshkov, M.D. Lukin, and V. Vuletić, *Nature* **502**, 71-74 (2013).

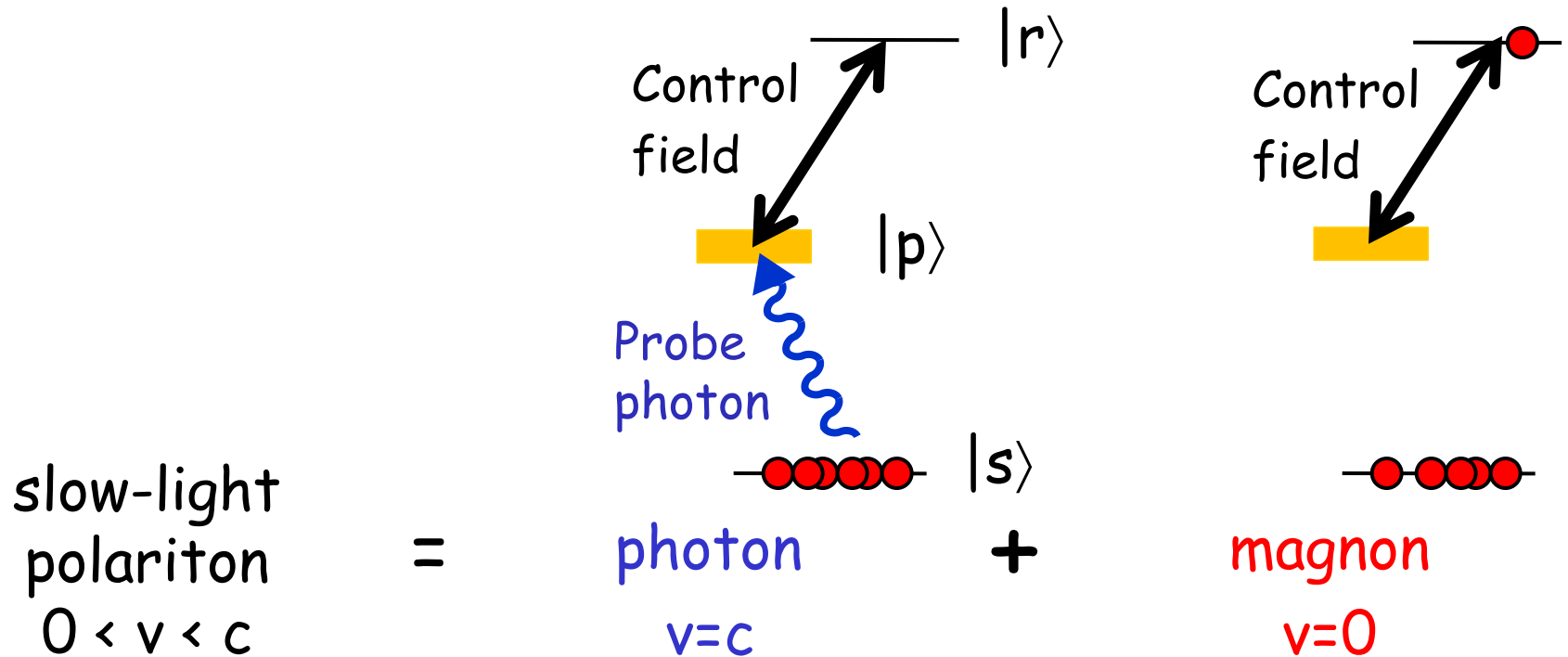
Electromagnetically induced transparency (EIT) with interacting Rydberg atoms



EIT to high-lying Rydberg state via unstable $|P\rangle$ level in dense cold ^{87}Rb gas

Electromagnetically Induced Transparency

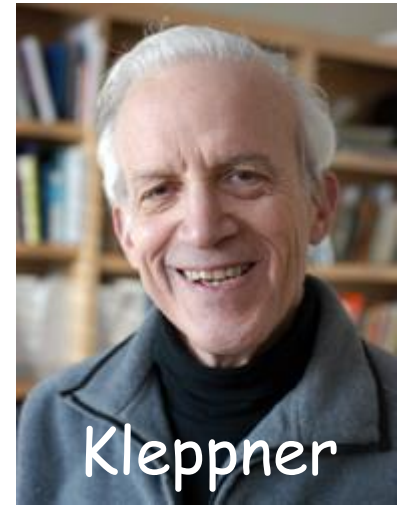
- EIT produces slow light by converting photons into collective atomic (spin) excitations



Collective (phase coherent excitation) \rightarrow preserves direction of light travel at $v=1 \text{ km/s}$ for our conditions.

Rydberg atoms

- Rubidium Rydberg atoms: hydrogen-like, very high principal excitation number ($n=100$ for our experiment);
- Extensively studied by Dan Kleppner. See also work by Pfau, Weidemueller, Adams, and lecture by T. Pfau.
- Electron very far from nucleus, very large dipole moment, strong interaction with electromagnetic fields.
- Serge Haroche – Nobel Prize 2012 for work in cavity quantum electrodynamics.
- Very large polarizability, strong Rydberg-Rydberg interactions.

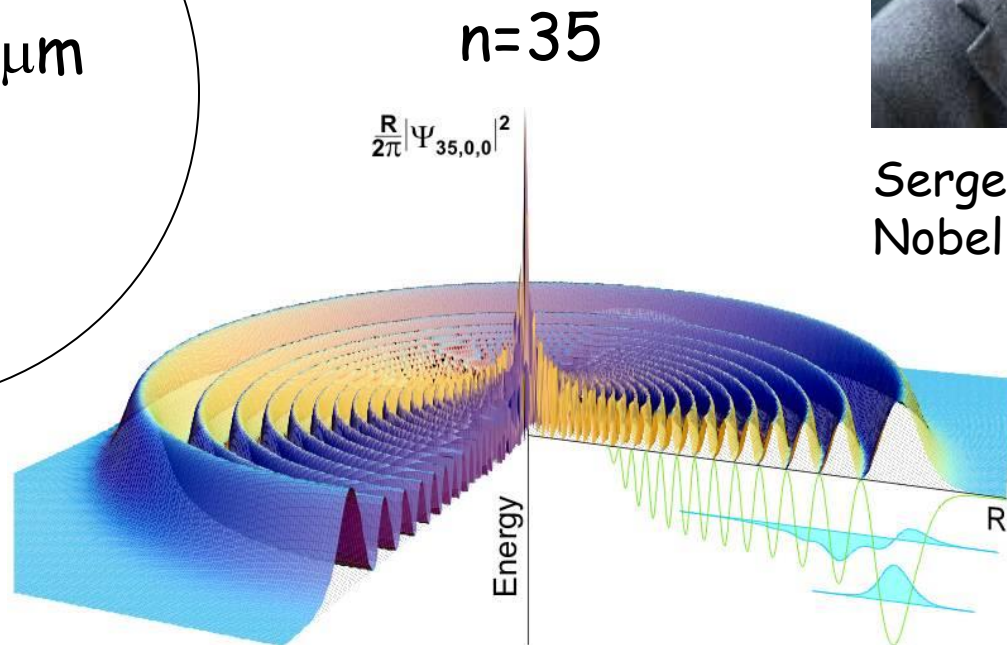
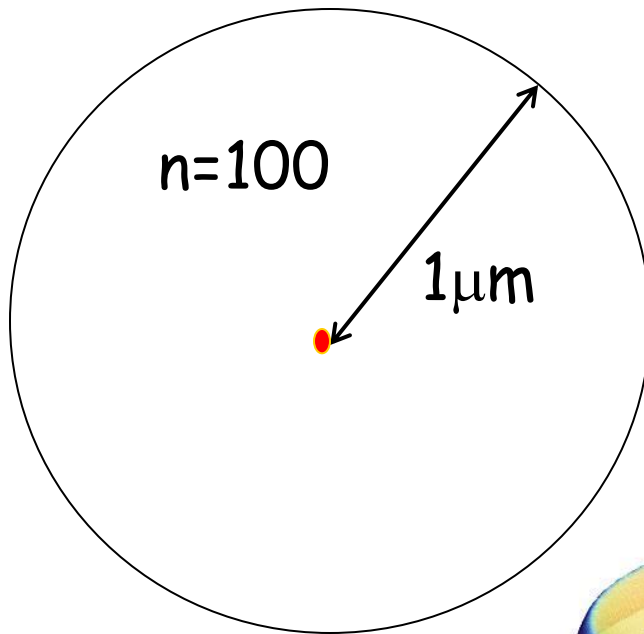


Rydberg states

Very highly excited hydrogen-like states

Extremely large size, dipole moment, polarizability

Strong Rydberg-Rydberg interactions $V(R)=C_6/R^6$

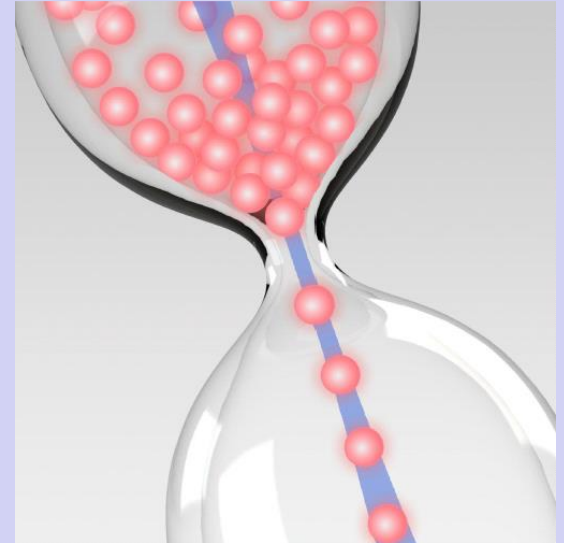


Serge Haroche,
Nobel Prize 2012

Rydberg atoms for quantum control

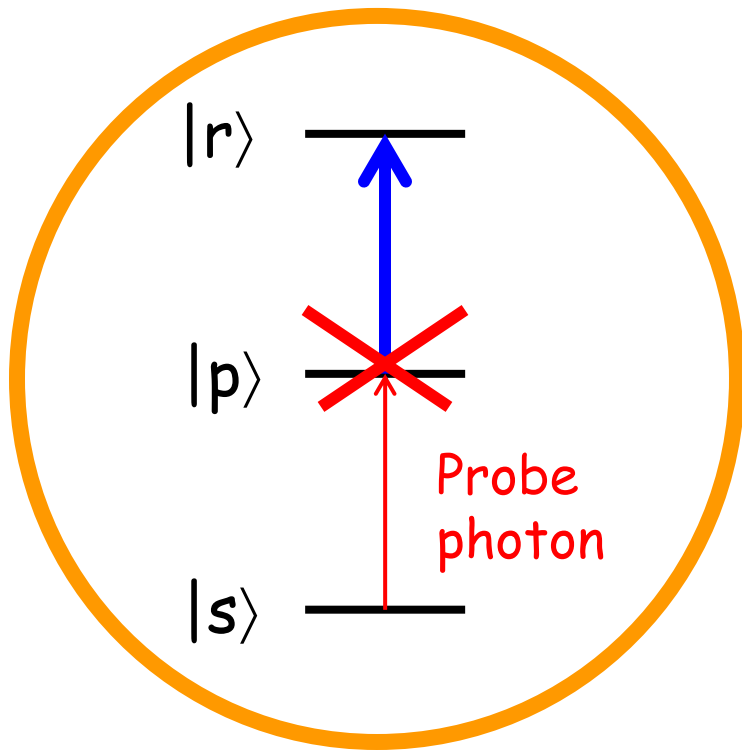
- Nonlinearities in Rydberg excitation
 - Tong, D. et al., PRL **93**, 063001 (2004); Singer et al., PRL **93**, 163001 (2004); Liebisch et al., PRL **95**, 253002 (2005); Heidemann et al., PRL **100**, 033601 (2008); Hoffmann et al., PRL **110**, 203601 (2013) (Weidemüller group).
- Photon switch, single-photon source
 - Dudin, Y. O. & Kuzmich, Science **336**, 887 (2012).; Baur et al., arXiv: 1307.3509 (Rempe group).
- Quantum gate between two Rydberg atoms
 - Urban et al., Nature Phys. **5**, 110-114 (2009); (Wisconsin)
 - Gaetan et al., Nature Phys. **5**, 115-118 (2009). (Paris)
- EIT with Rydberg atoms (classical regime, but same idea as this work)
 - Pritchard et al., PRL **105**, 193603 (2010). (Adams group)
- Theory work
 - Lukin et al., PRL **87**, 037901 (2001); Petrosyan, Otterbach, & Fleischhauer, PRL **107**, 213601 (2011); Gorshkov et al., PRL **107**, 133602 (2011); Muller, Lesanovsky, Weimer, Buechler, & Zoller, PRL **102**, 170502 (2009).

Dissipative Rydberg-induced interactions: A one-photon nonlinear filter

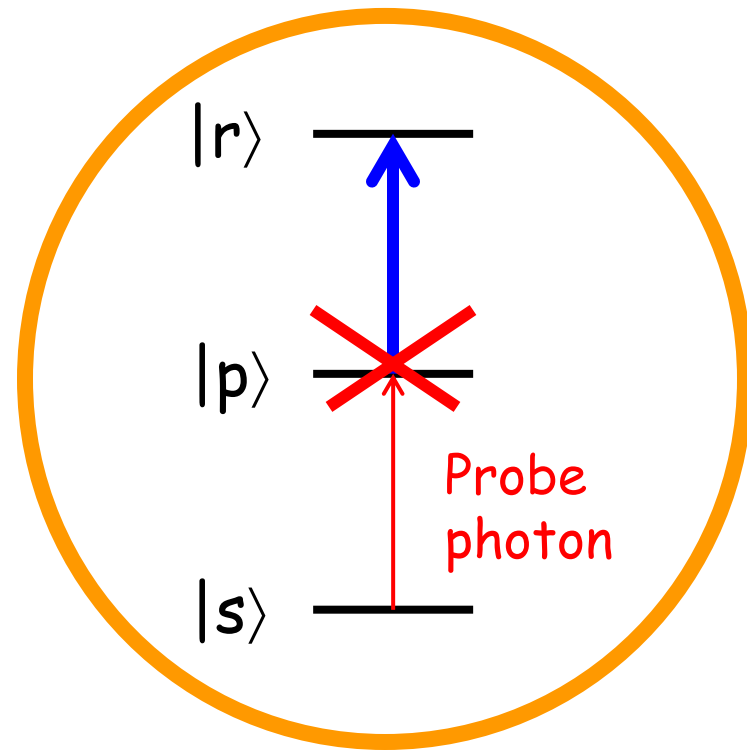


T. Peyronel, O. Firstenberg, Q.-Y. Liang, S. Hofferberth, A.V. Gorshkov, T. Pohl, M.D. Lukin, and V. Vuletic, *Nature* **488**, 57-60 (2012).

Rydberg Electromagnetically induced transparency

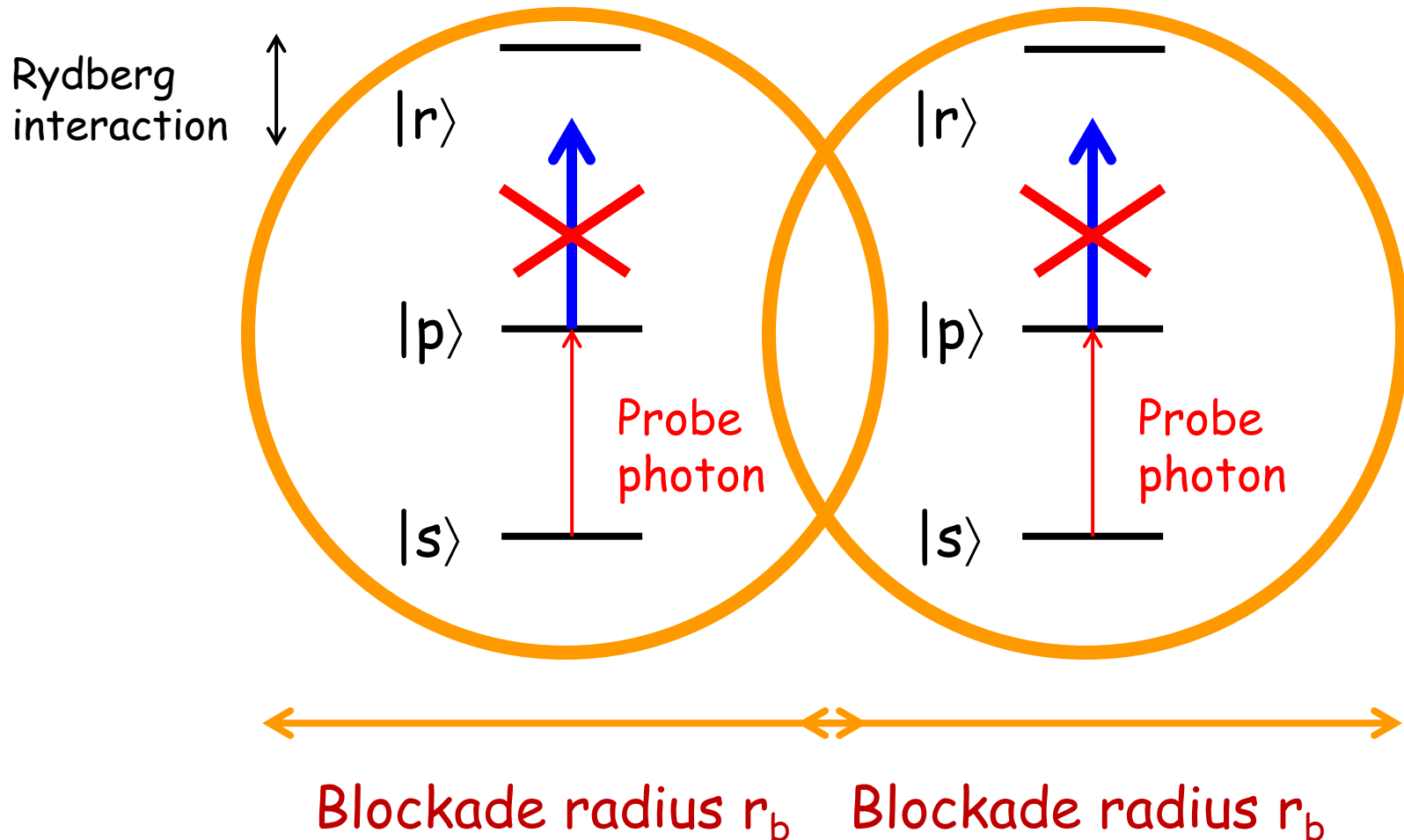


Blockade radius r_b

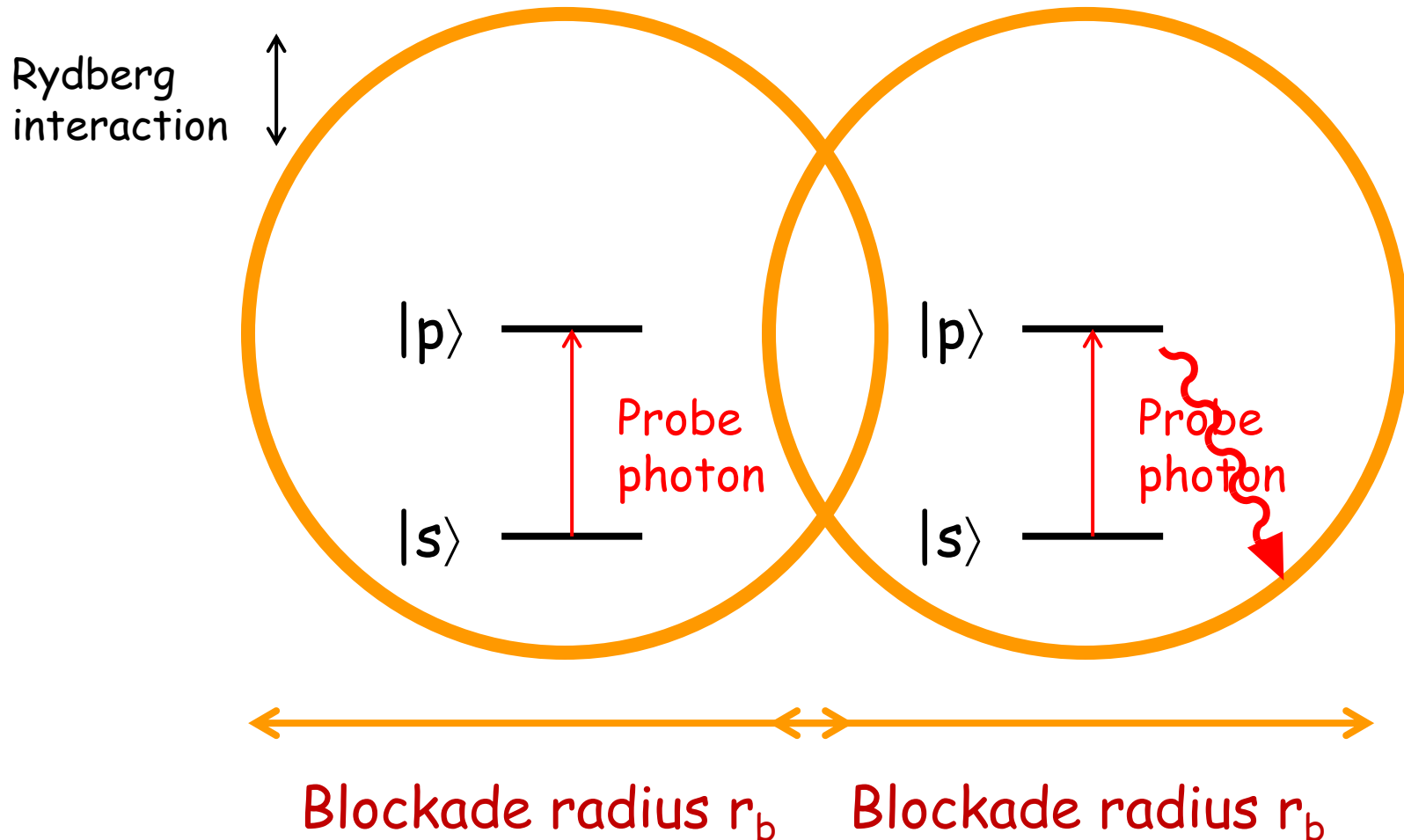


Blockade radius r_b

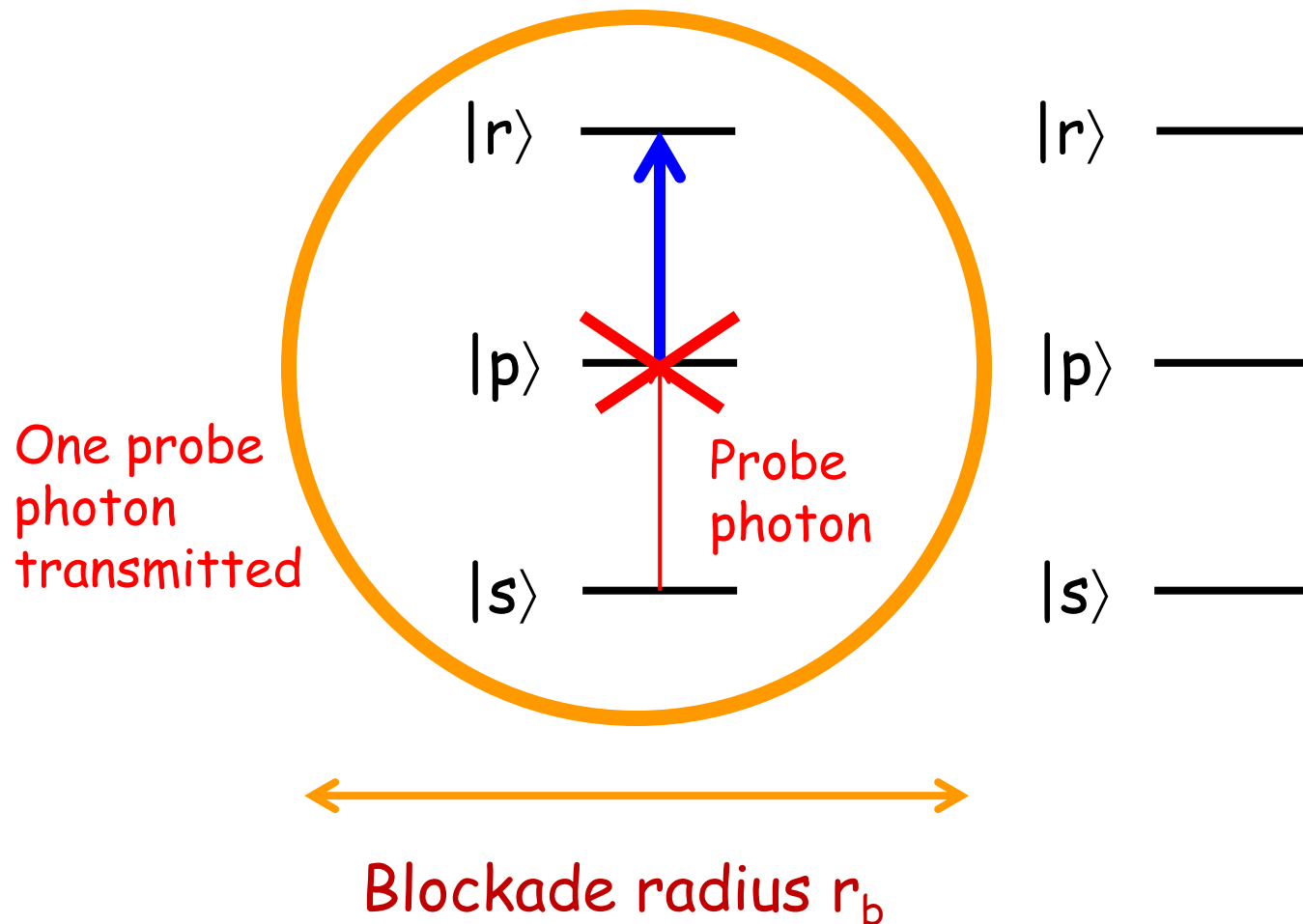
Rydberg Electromagnetically induced transparency



Rydberg Electromagnetically induced transparency



Rydberg Electromagnetically induced transparency



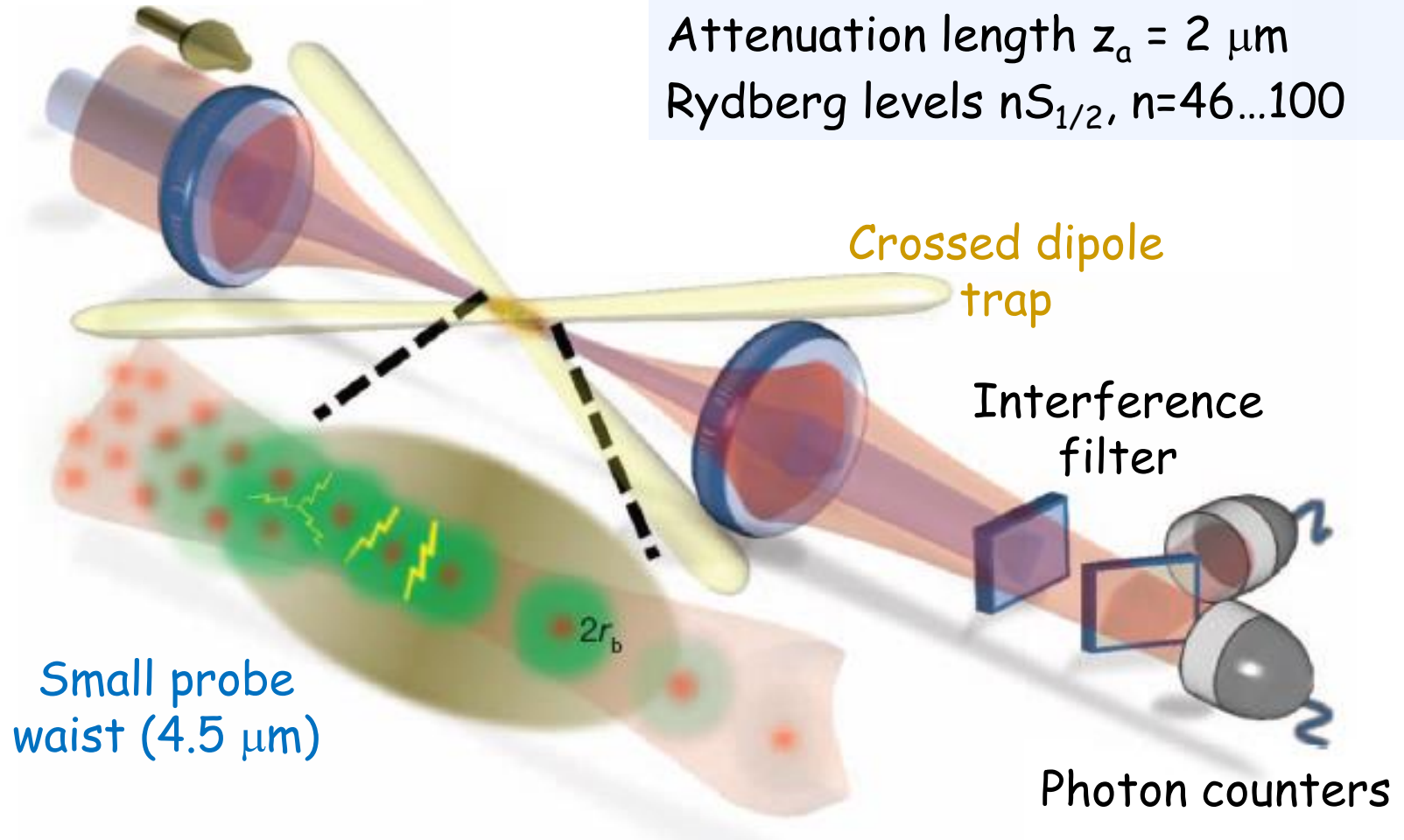
Experimental setup

Continuous probe
and control beams

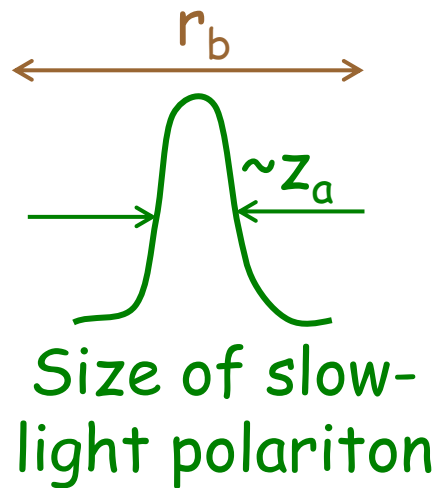
Ultracold high-density ^{87}Rb
ensemble (10^{12} cm^{-3})

Attenuation length $z_a = 2 \mu\text{m}$

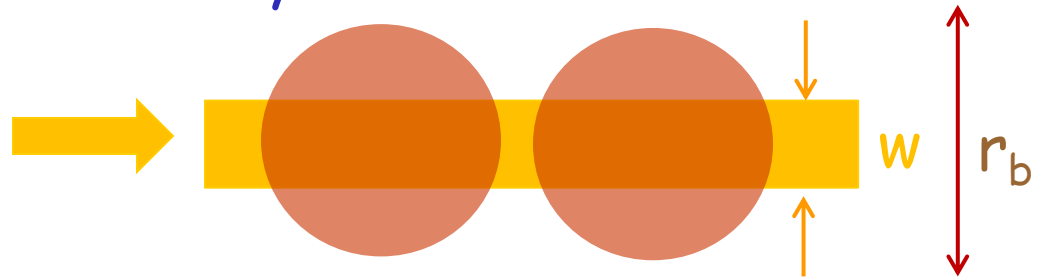
Rydberg levels $nS_{1/2}$, $n=46\ldots 100$



Rydberg EIT in 1D system



Single photon nonlinearity for $z_a < r_b$, i.e. at high atomic density.



Rydberg blockade: Rydberg slow-light polaritons cannot coexist within blockade radius r_b .

Size of Rydberg polariton \sim resonant **attenuation length** $z_a \times \sqrt{OD}$

Our system:

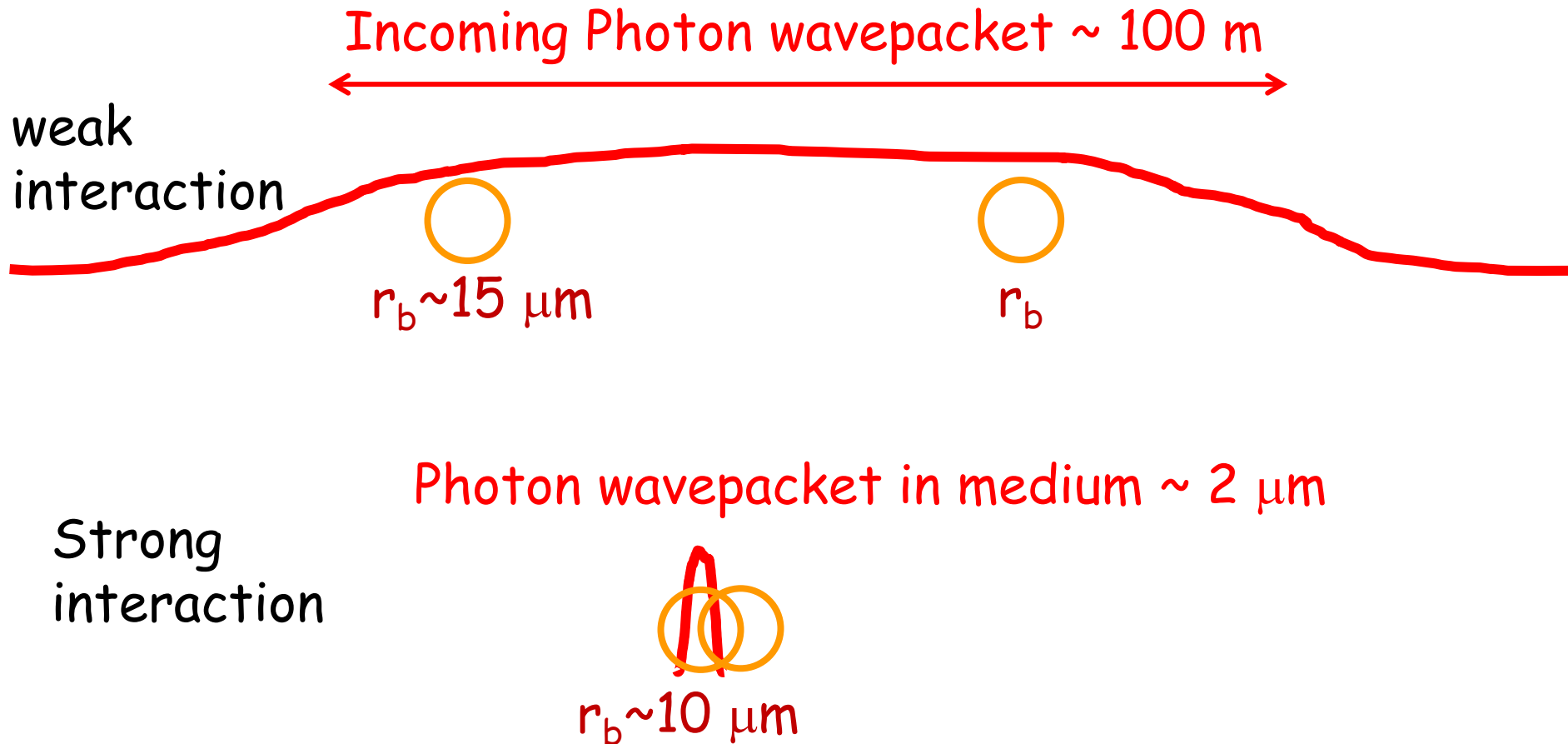
$$z_a \sim 2\mu\text{m}$$

$$r_b = 12-15\mu\text{m}$$

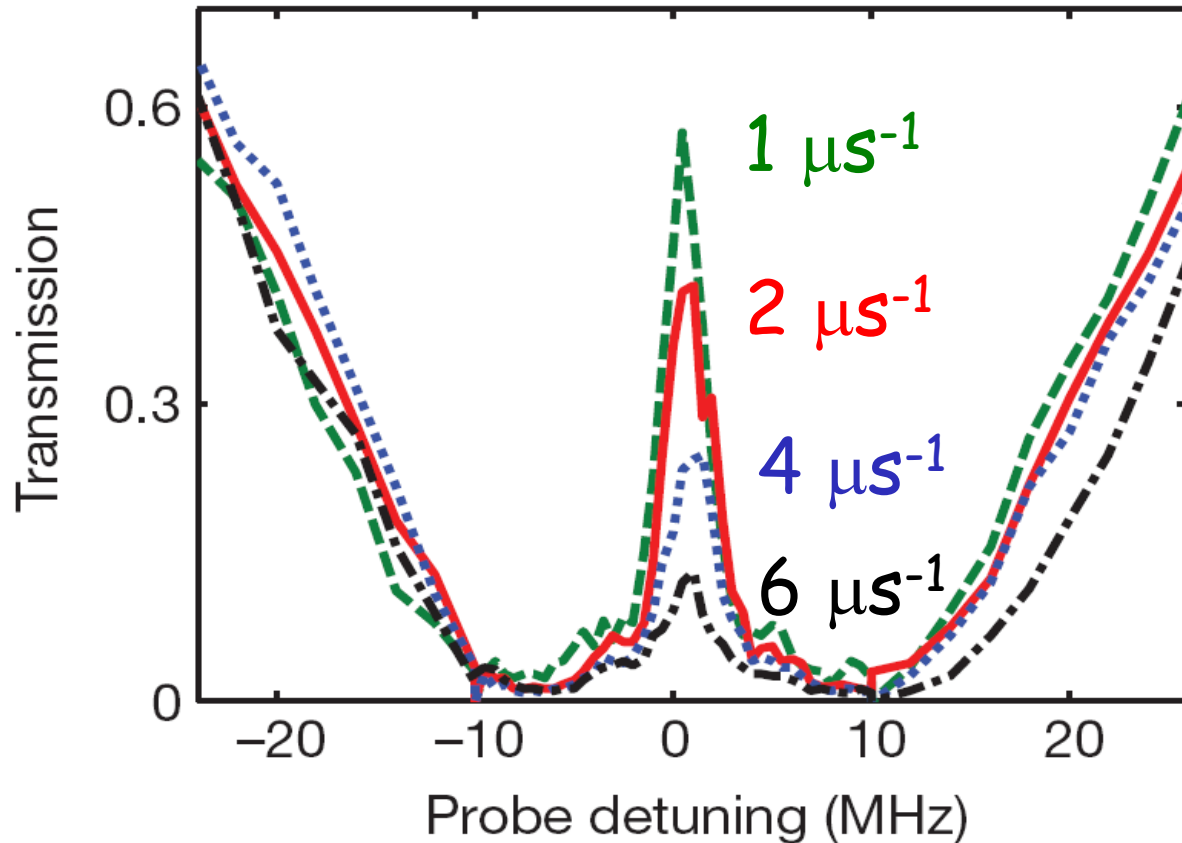
One-dimensional system:

$$w = 4\mu\text{m} < r_b$$

Compressed photon wavepacket



Rydberg EIT spectra vs. probe photon rate

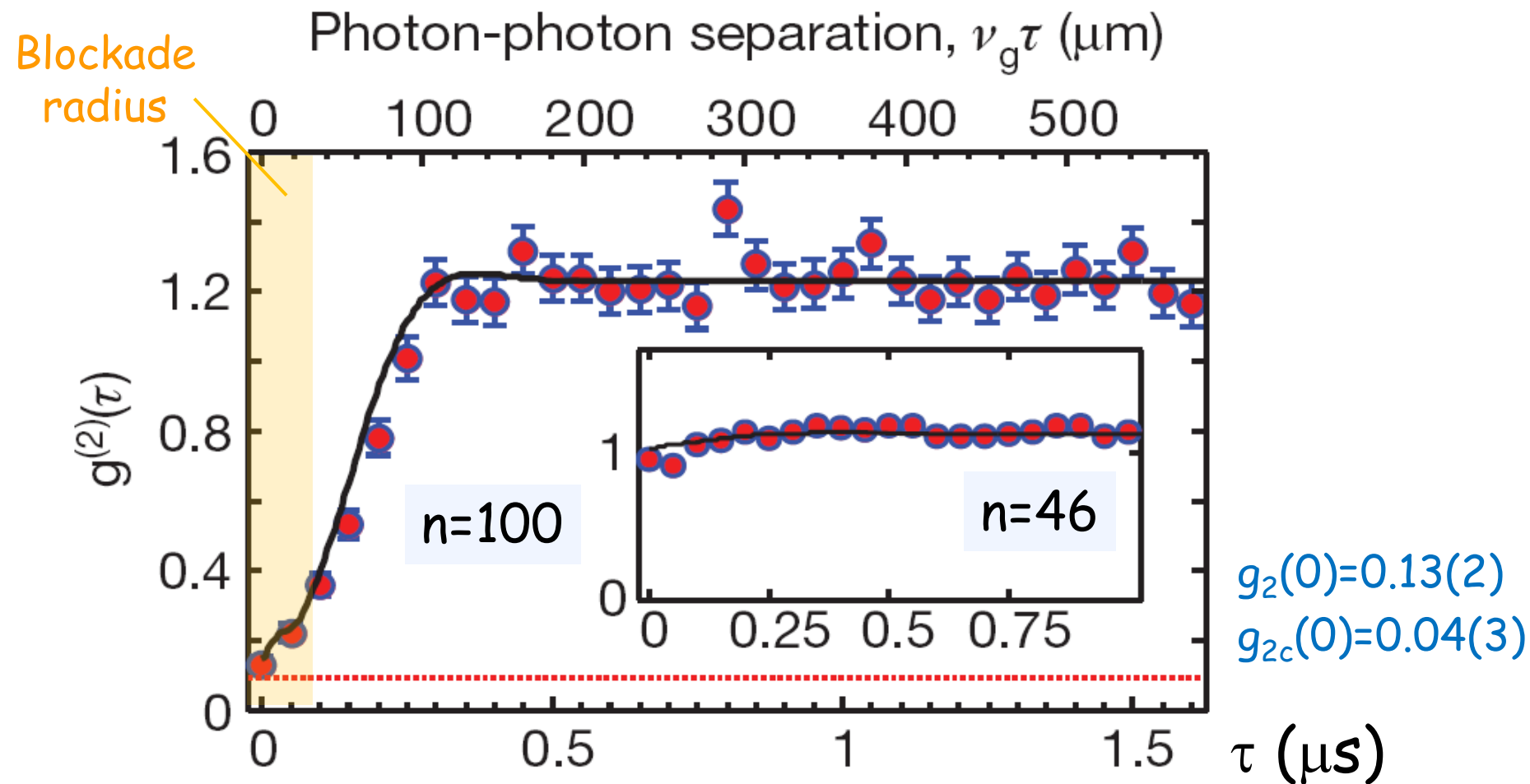


$|n=100 S_{1/2}\rangle$
Optical depth $OD=40$
Attenuation length $2\mu\text{m}$
Blockade radius $13\mu\text{m}$

Similar measurements of large optical nonlinearity (in classical regime attenuation length $>$ blockade radius):

Pritchard, Maxwell, Gauguet, Weatherill, Jones, and Adams, Phys. Rev. Lett. **105**, 193603 (2010).

Train of single-photon wavepackets

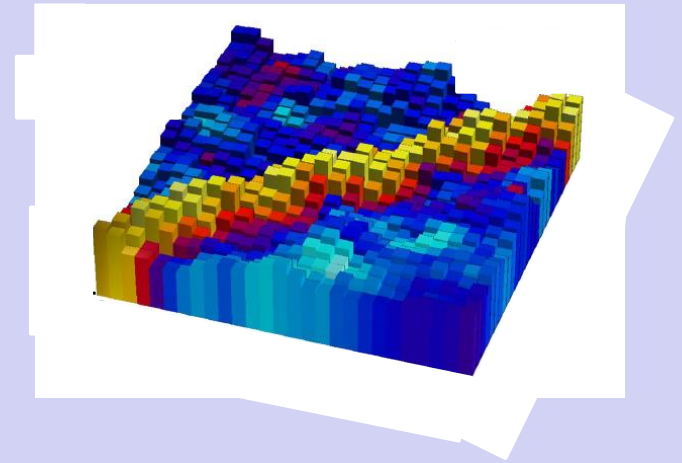


T. Peyronel, O. Firstenberg, Q.-Y. Liang, A. Gorshkov, T. Pohl, M. Lukin, and V. Vuletic, *Nature* **488**, 47 (2012).

Dissipative vs. dispersive effects

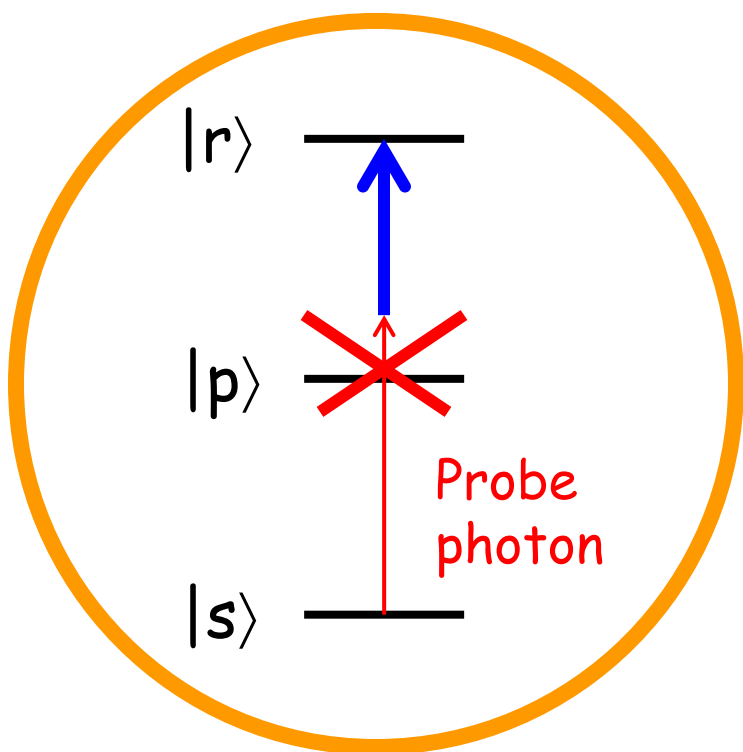
- The above establishes a dissipative and extremely nonlinear two-photon filter: one photon transmitted, second photon absorbed.
 - Train of single-photon pulses in transmission.
- Can one induce dispersive effects instead?

Detuned Rydberg EIT: Bound state of two photons

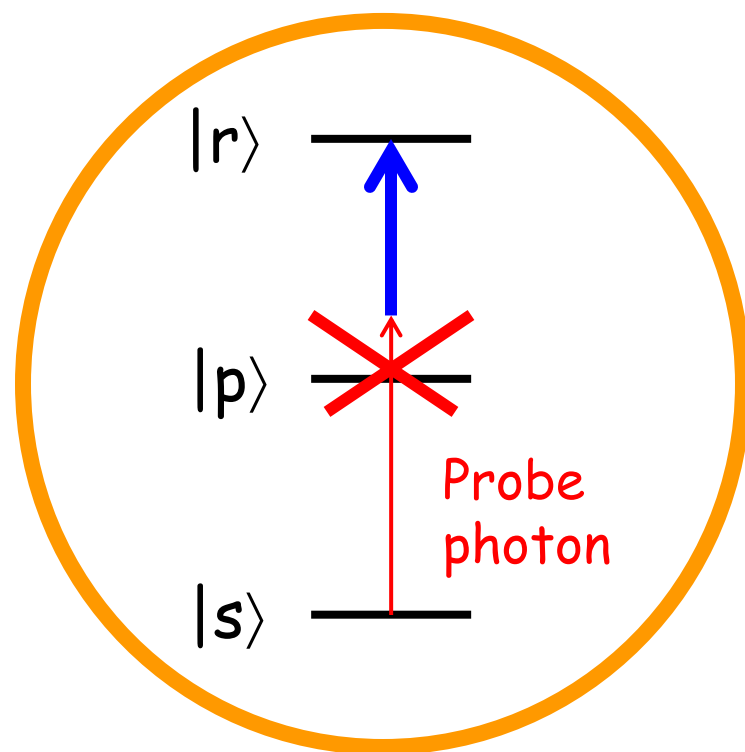


O. Firstenberg, T. Peyronel, Q.-Y. Liang, A.V. Gorshkov, M.D. Lukin, and V. Vuletic, *Nature* **502**, 71-74 (2013).

Rydberg EIT off resonance

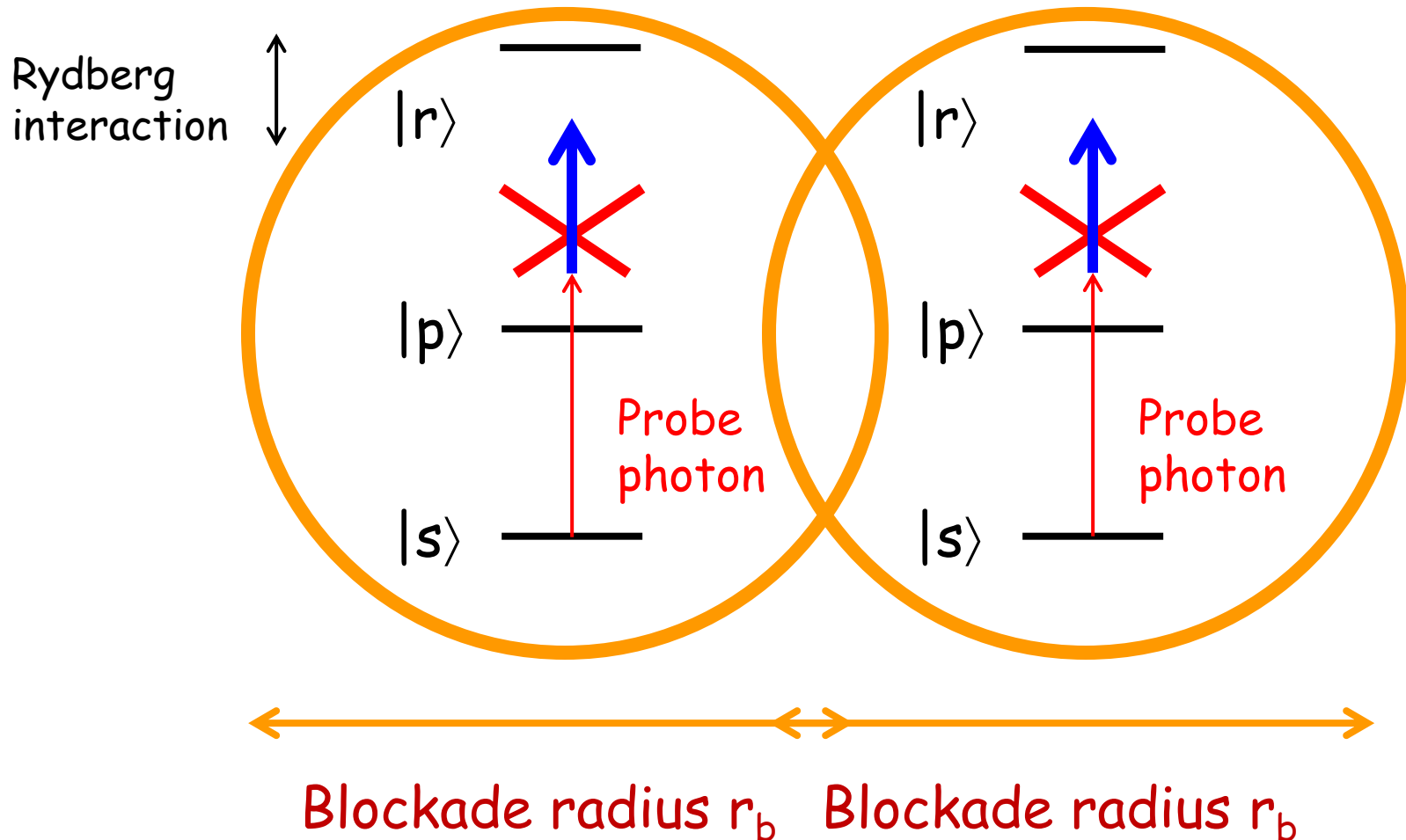


Blockade radius r_b



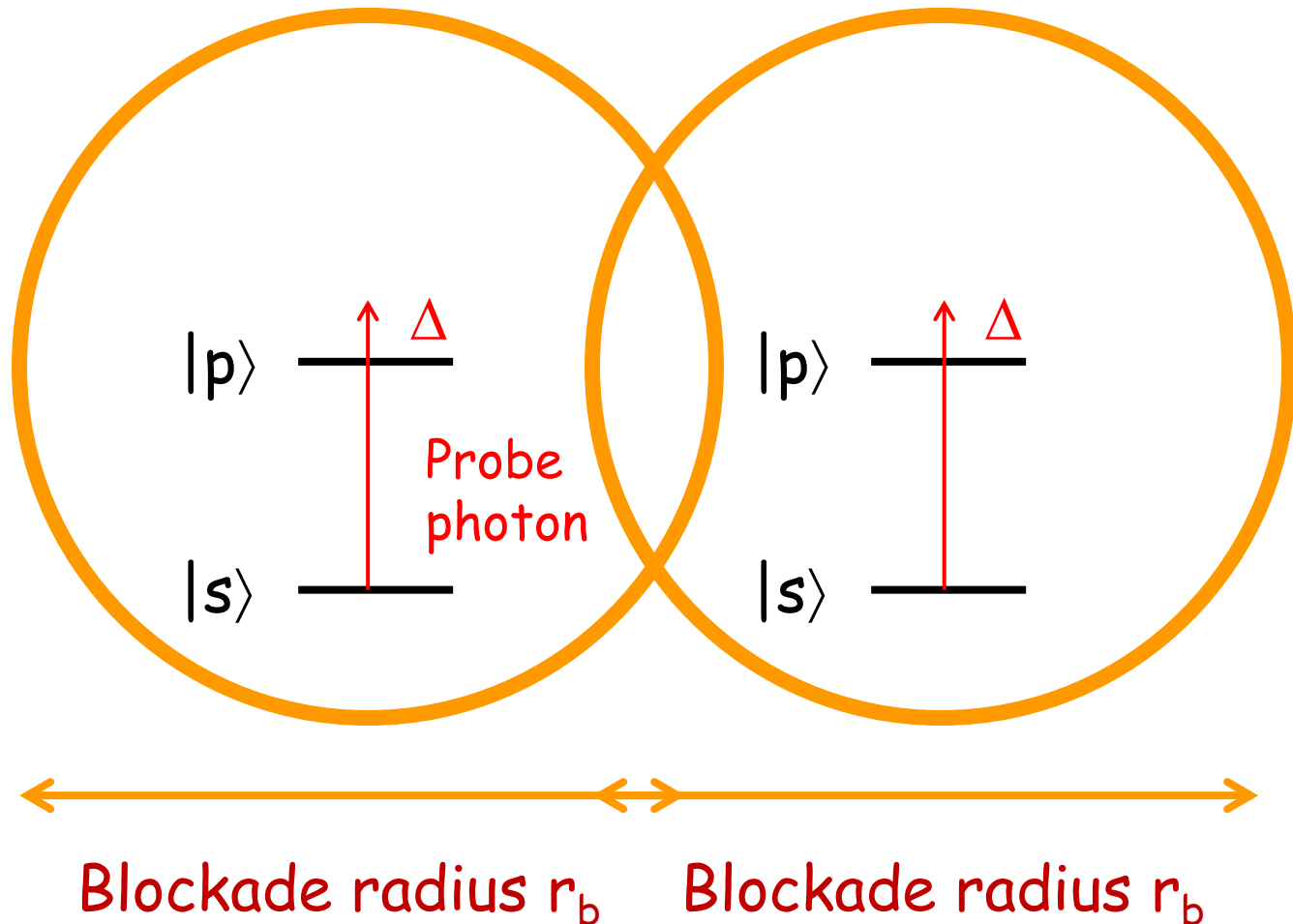
Blockade radius r_b

Rydberg EIT off resonance

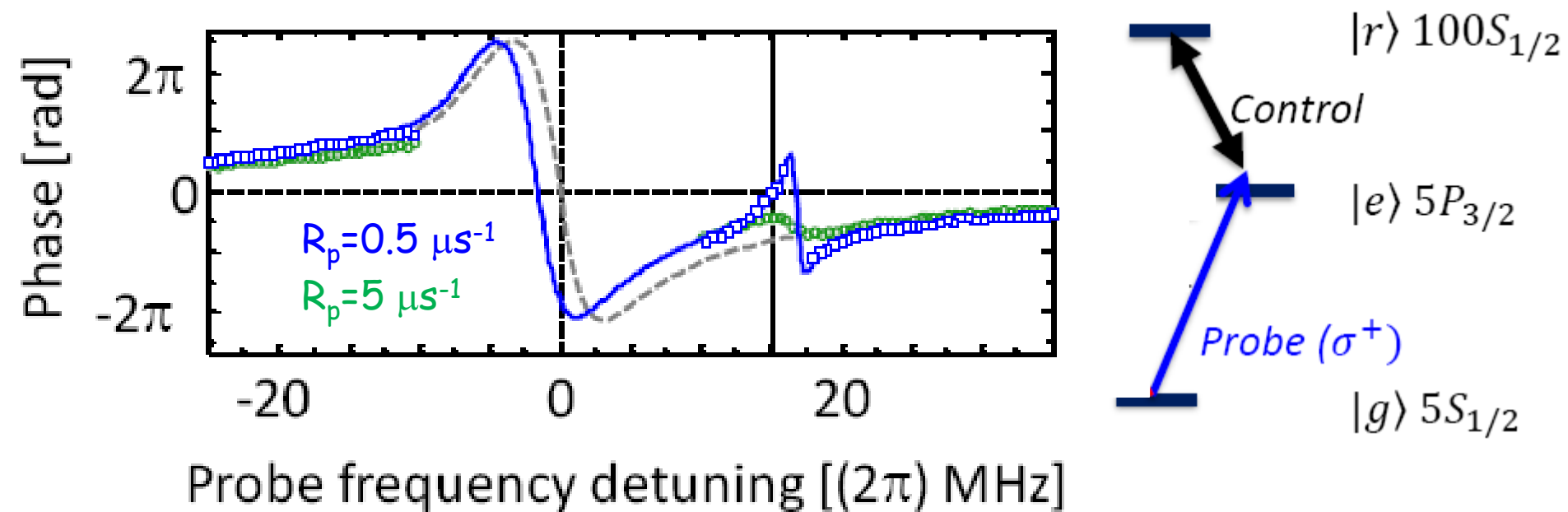


Rydberg EIT off resonance

Probe
photons
experience
index of
refraction
when close



Rydberg induced nonlinear phase shift



No EIT for photons within blockade radius \rightarrow light experiences no phase shift outside blockade radius, but some phase shift within blockade radius.

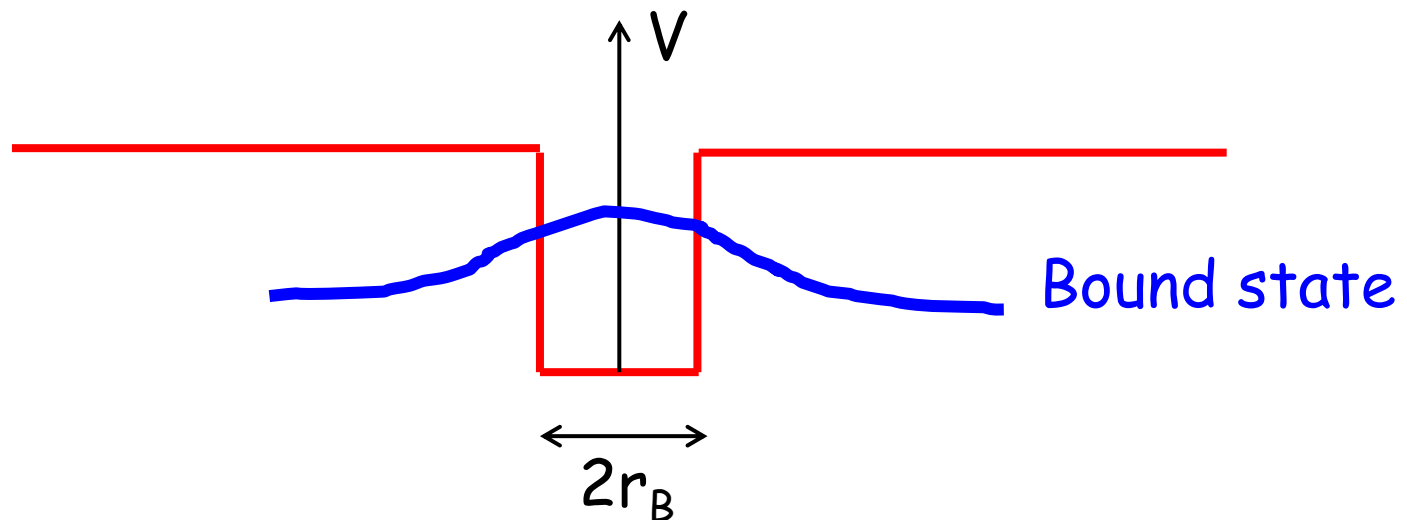
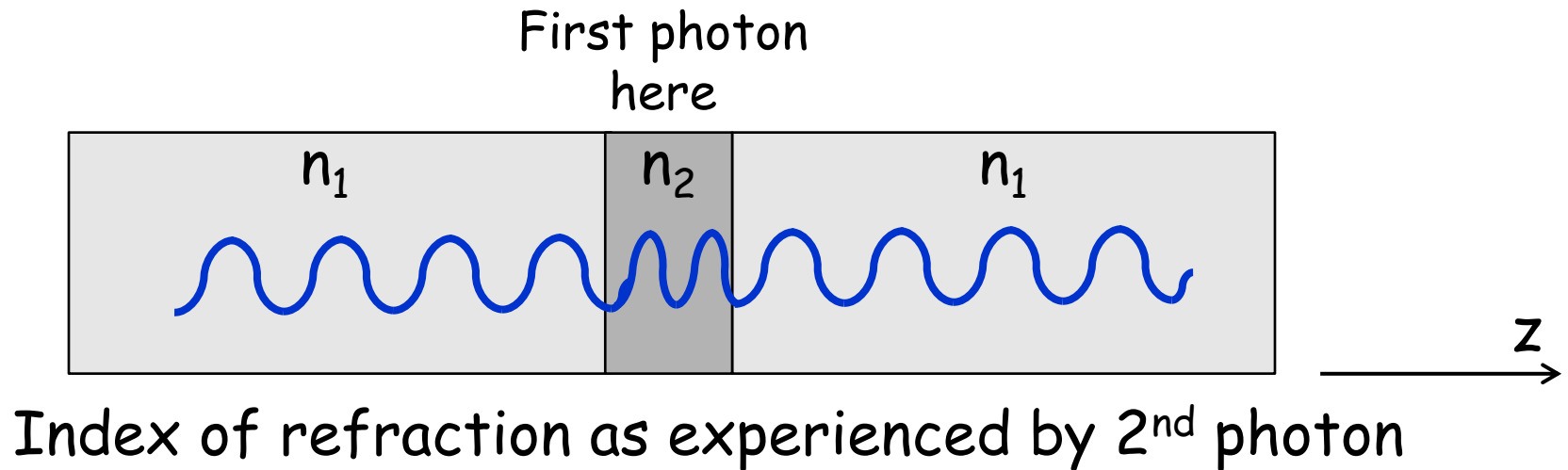
Curvature of dispersion relation:

Photons also acquire mass $\sim 1000 \hbar\omega/c^2$

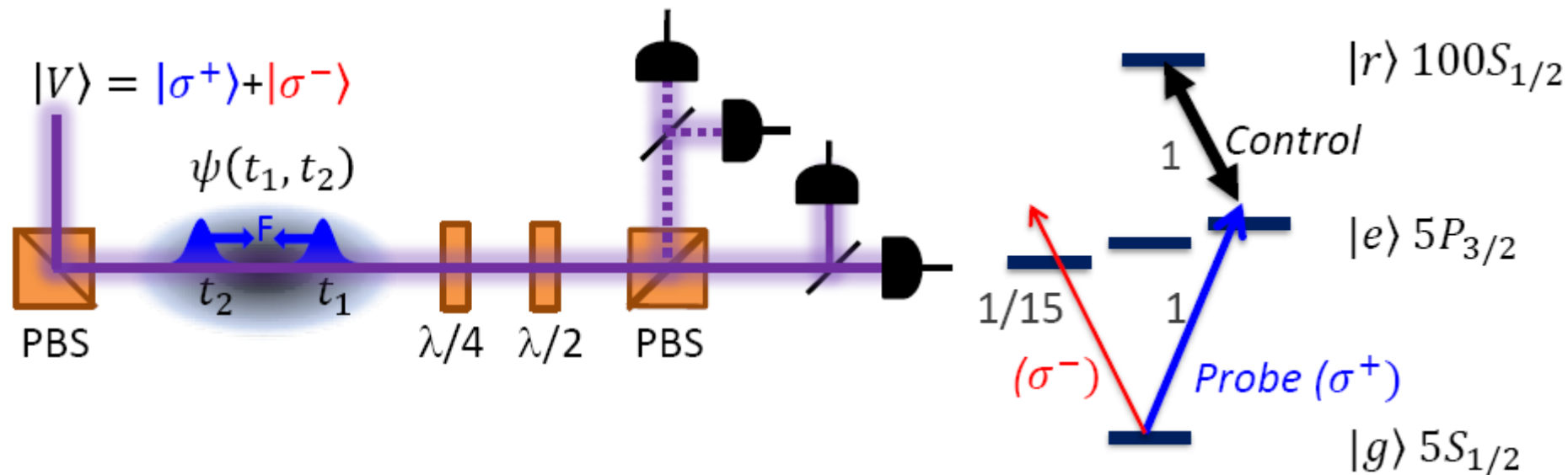
Typical group velocity $v_g = 1000$ m/s

Slow, massive photons

Rydberg blockade as square well potential



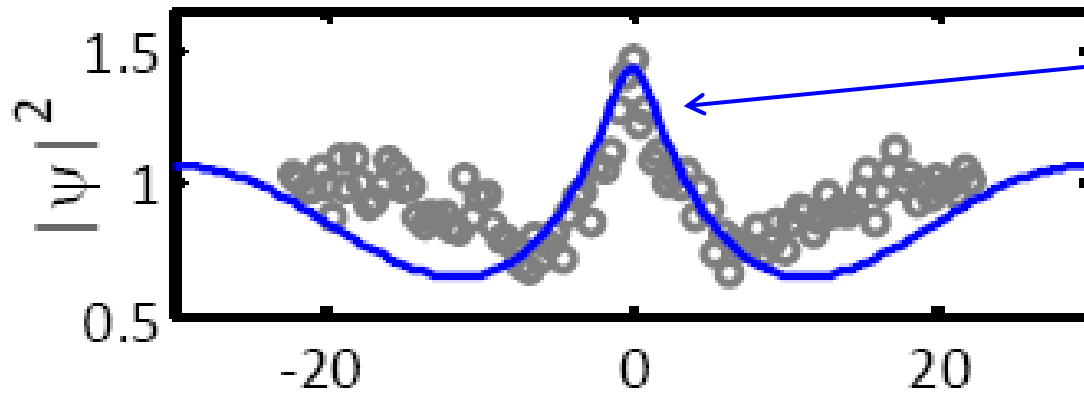
Measuring the two- and three-photon wavefunction



Non-interacting σ^- light serves as phase reference to measure two-photon wavefunction for interacting $\sigma^+ \sigma^+$ pairs.

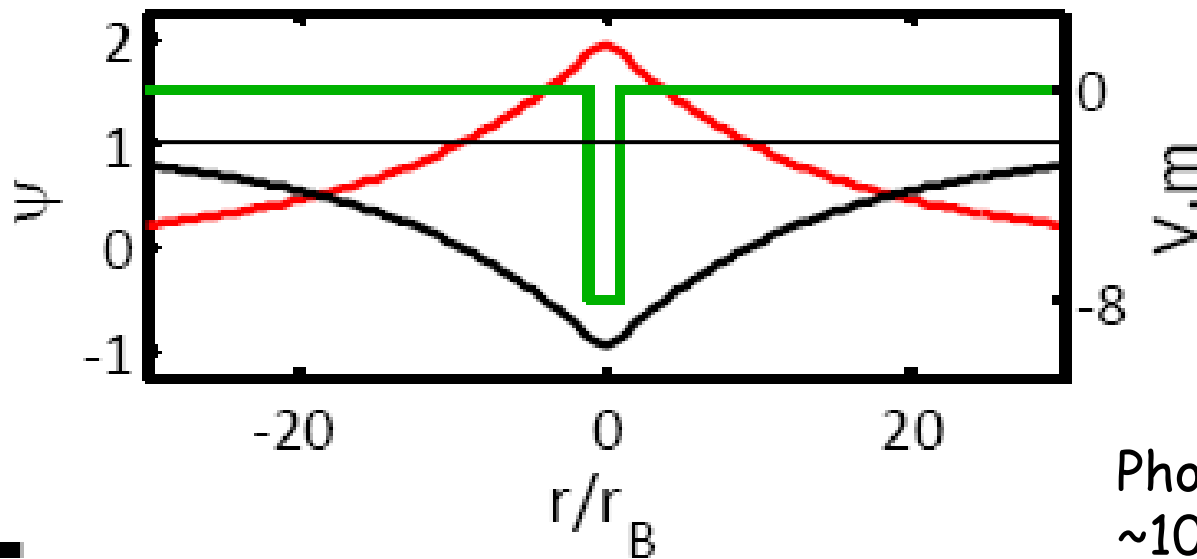
Measurement in different polarization bases: quantum tomographic reconstruction of time dependent two-photon wavefunction.

Two-photon bound state



Two-photon
bound state

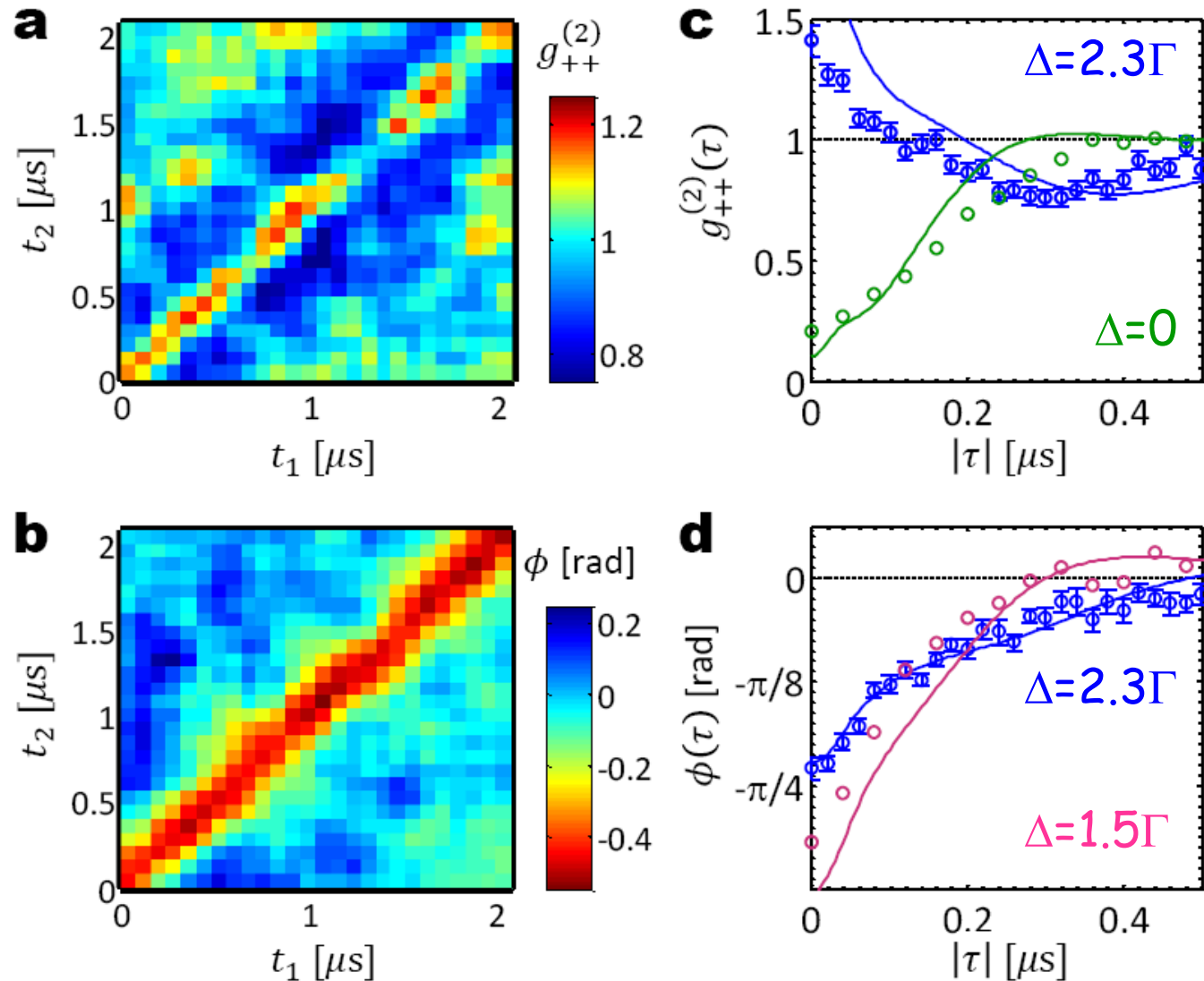
Experiment



Simple
theoretical
picture
(Schrödinger
equation)

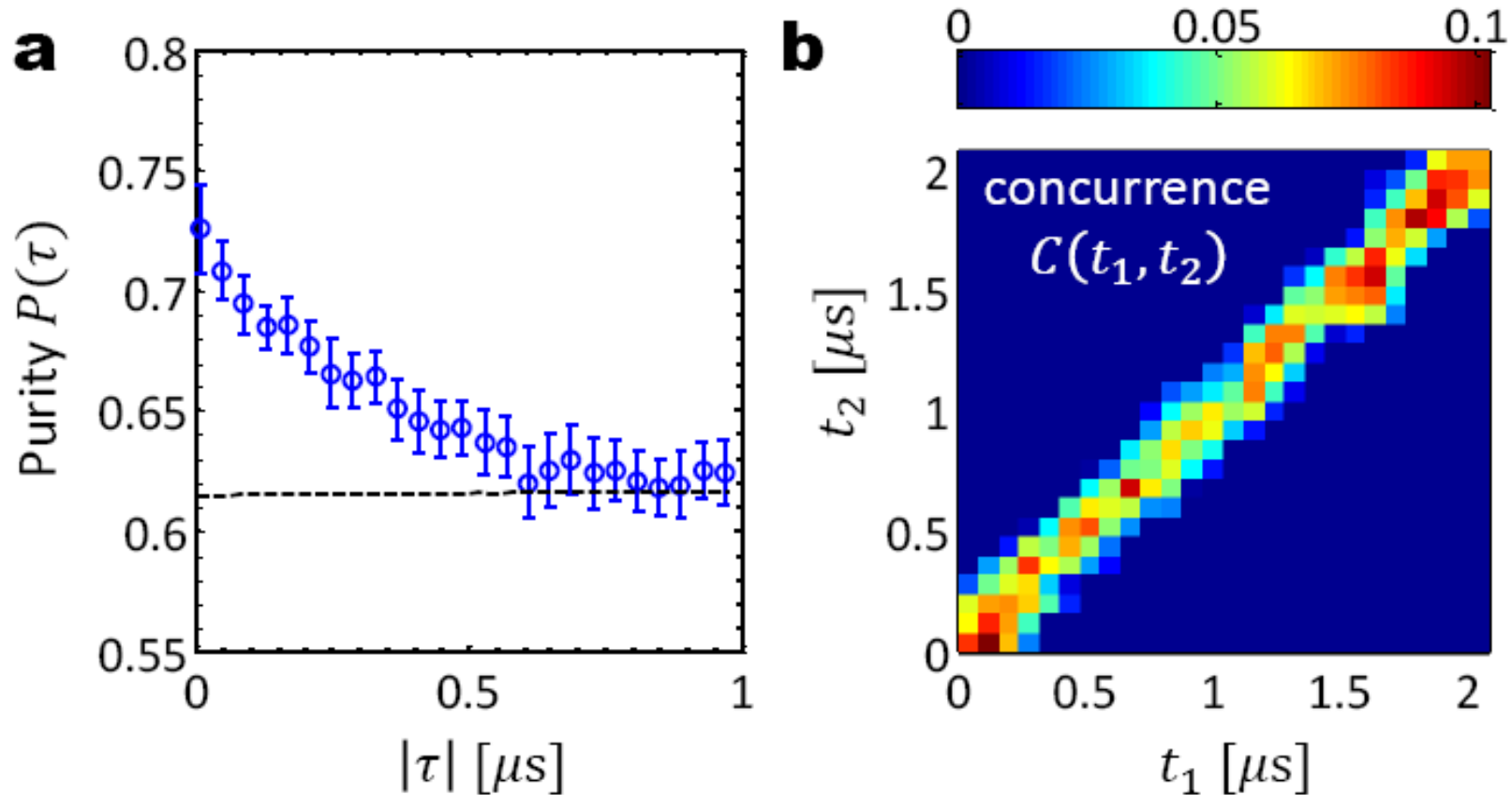
Photons also acquire mass
 $\sim 1000 \hbar\omega/c^2$ (curvature
of dispersion).

Reconstructed two-photon wavefunction



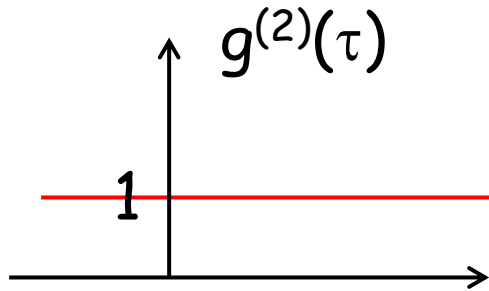
O. Firstenberg, T. Peyronel, Q.-Y. Liang, A.V. Gorshkov, M.D. Lukin, and V. Vuletic, *Nature* **502**, 71-74 (2013).

Interaction creates polarization entanglement



Concurrence >0 demonstrates deterministic polarization entanglement between photons.

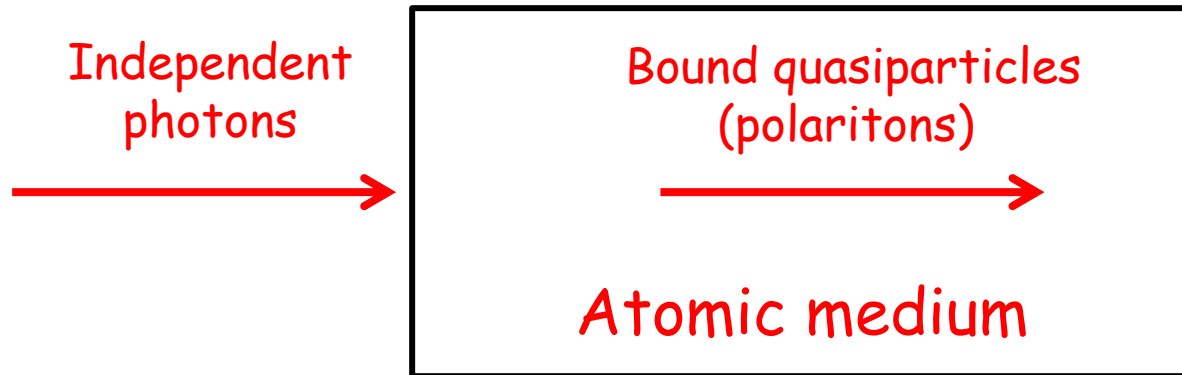
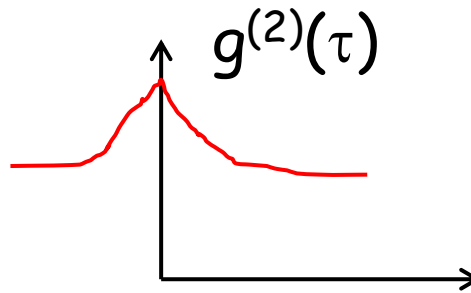
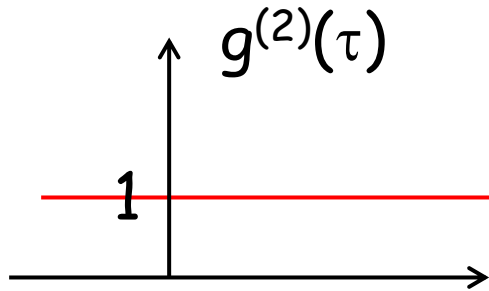
Photon bound state - disclaimer



Independent
photons

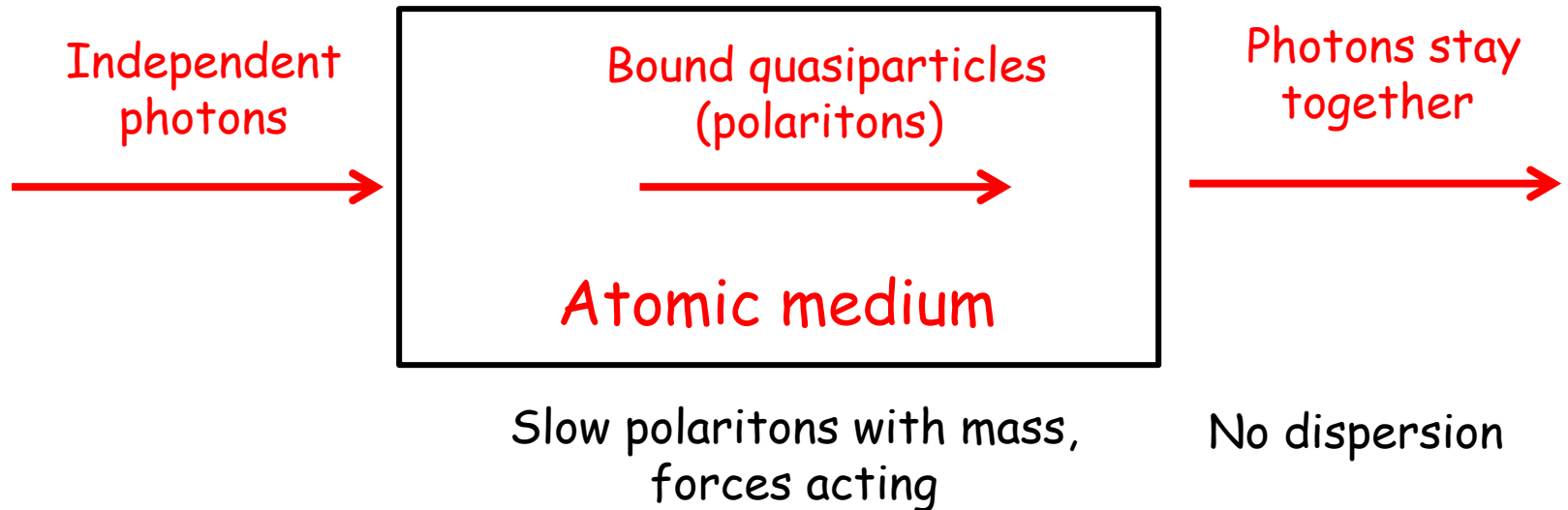
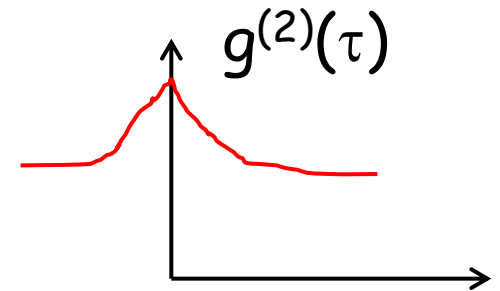
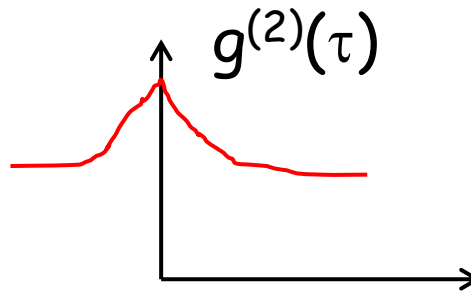
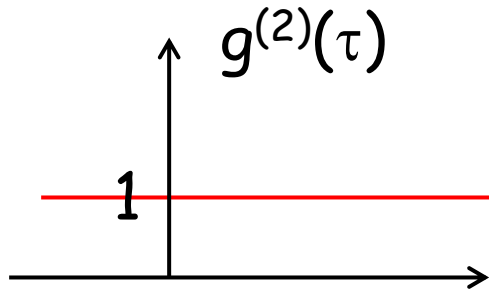


Photon bound state - disclaimer



Slow polaritons with mass,
forces acting

Photon bound state - disclaimer



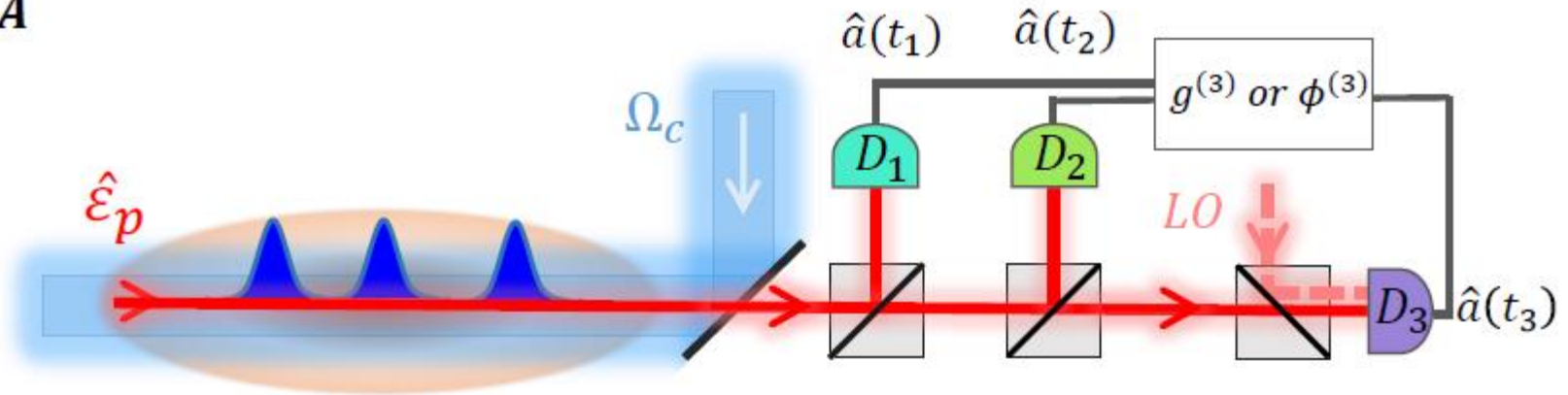
Three-photon bound states

Observation of three-photon bound states in a quantum nonlinear medium.

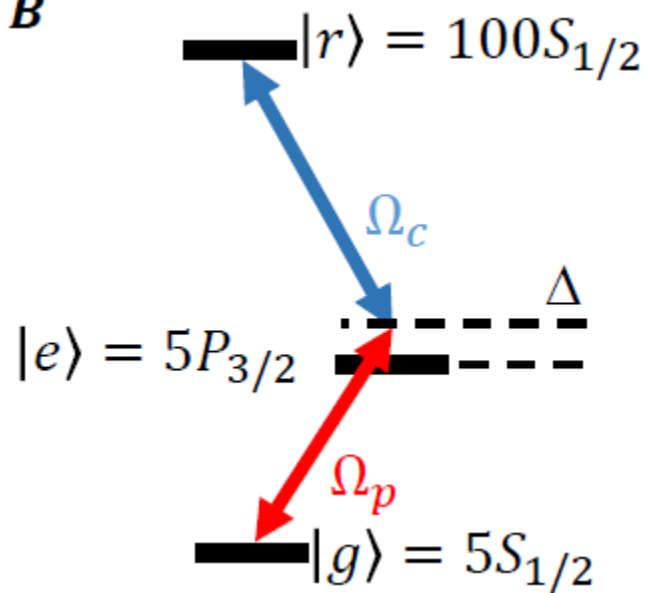
Q.-Y. Liang, A.V. Venkatramani, S.H. Cantu, T.L. Nicholson, M.J. Gullans, A.V. Gorshkov, J.D. Thompson, C. Chin, M.D. Lukin, and V. Vuletić, *Science* 359, 783-786 (2018).

Three-photon correlations - setup

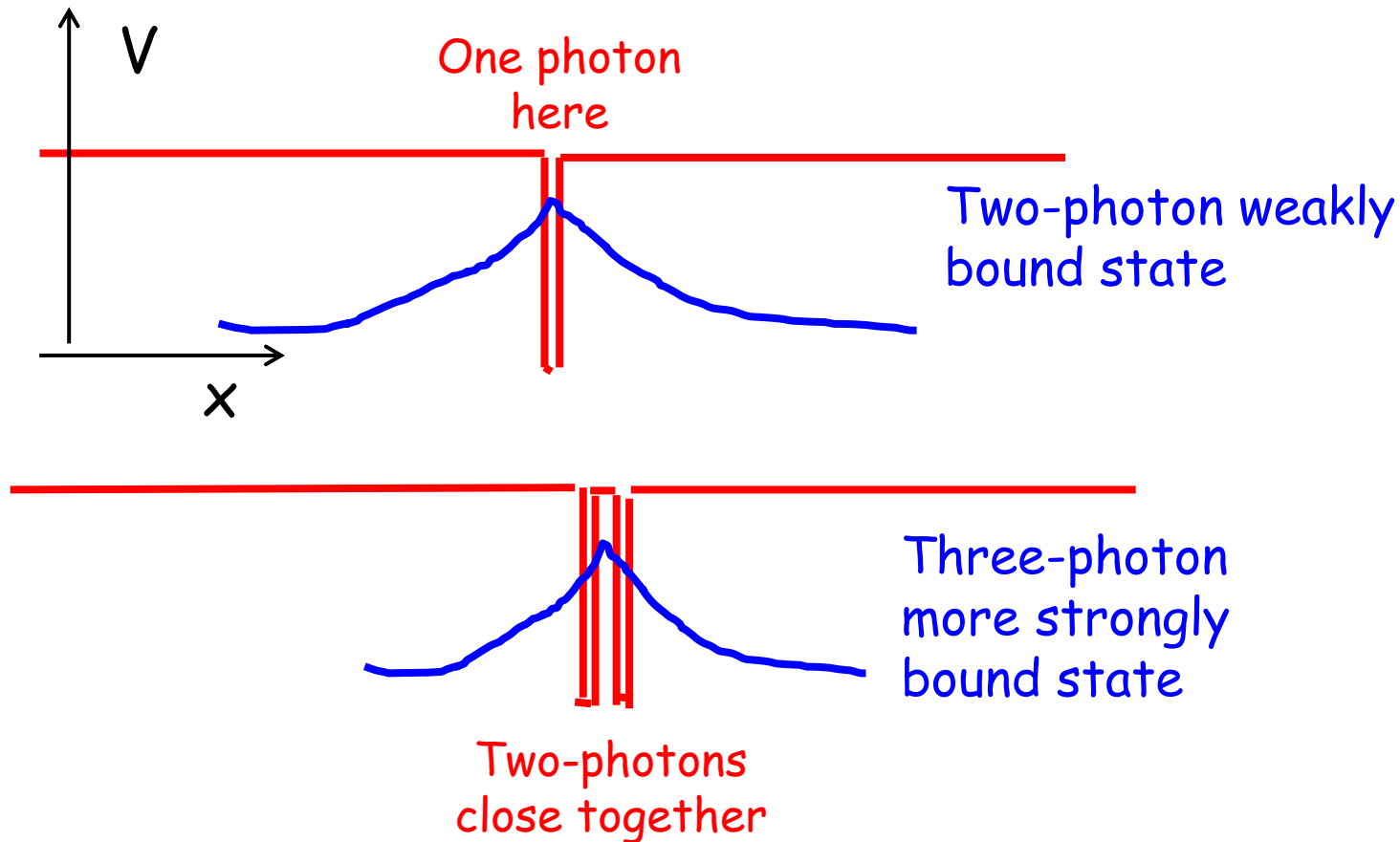
A



B

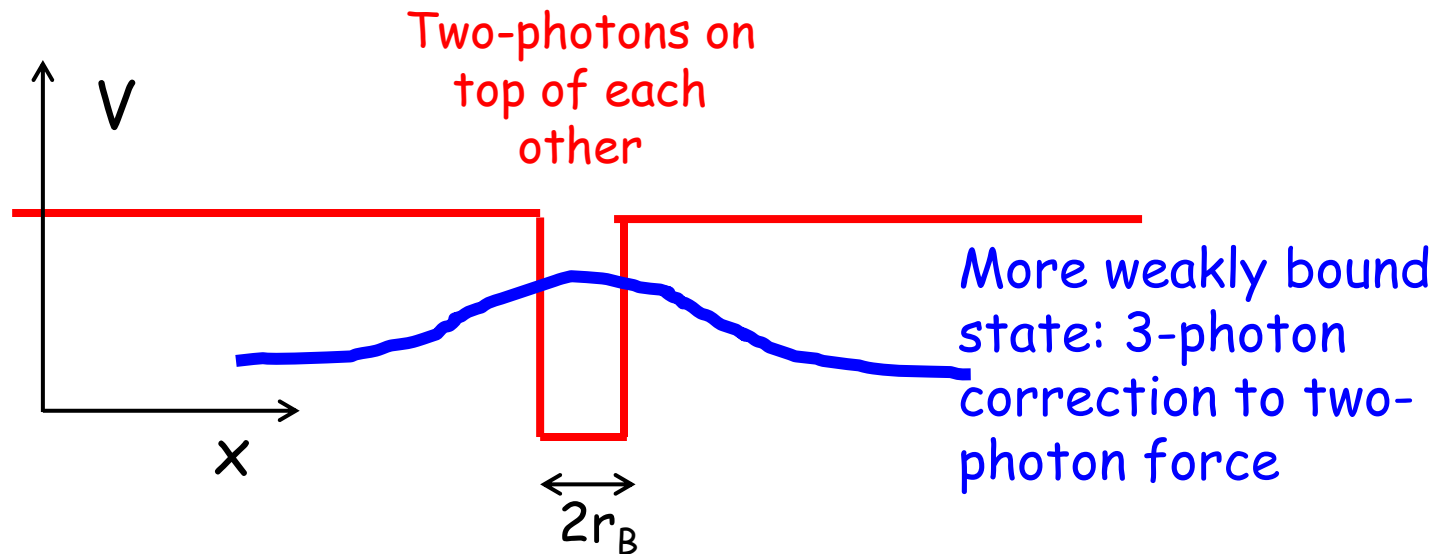
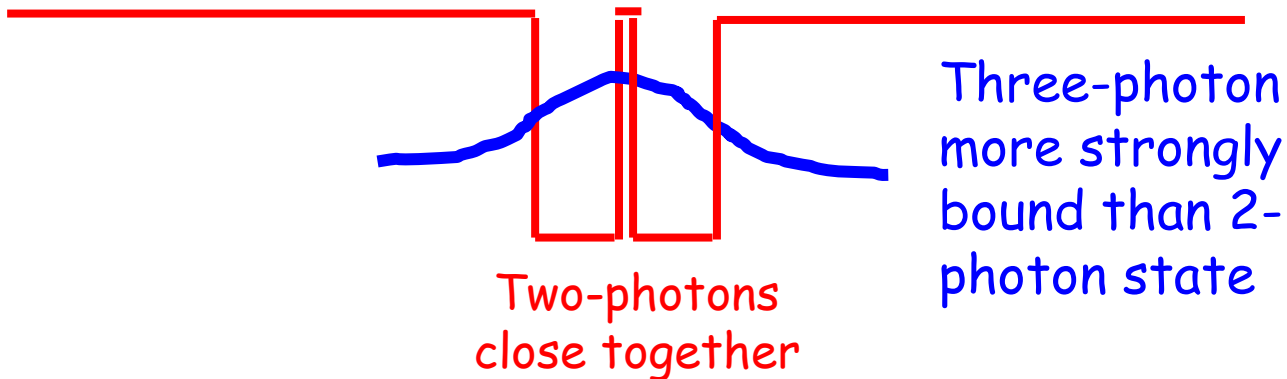


Two- vs. three-photon binding

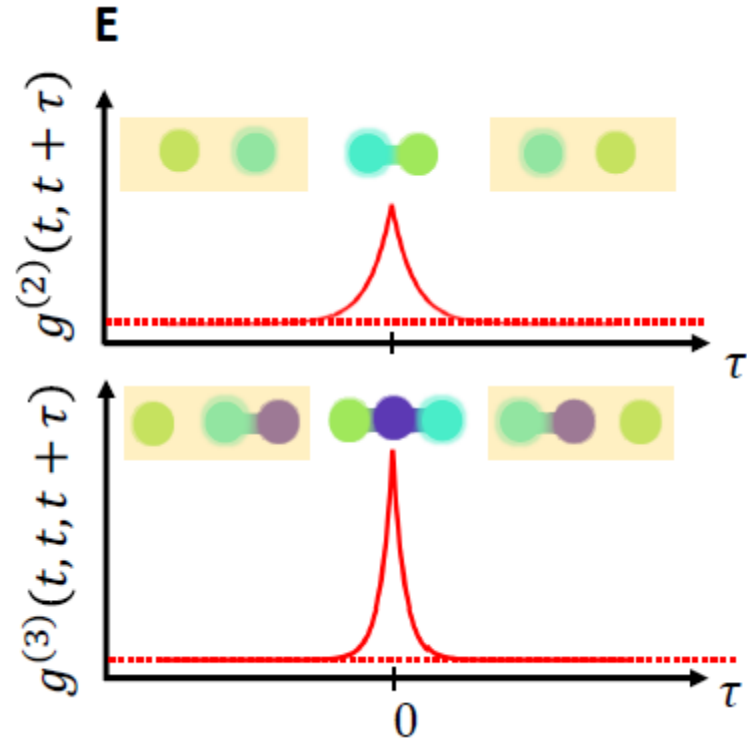
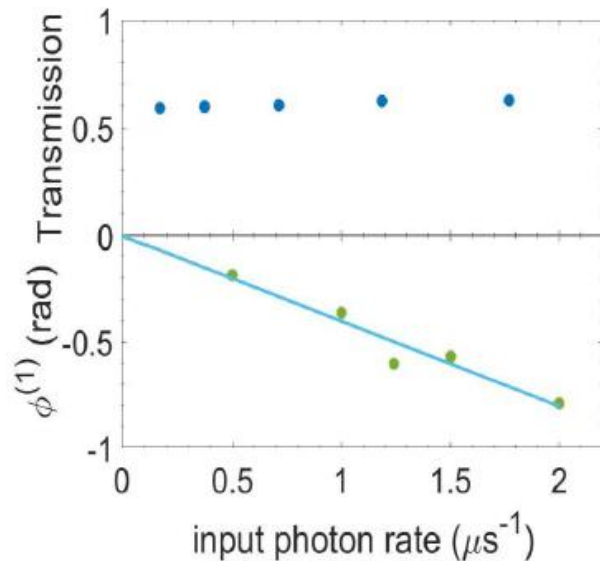
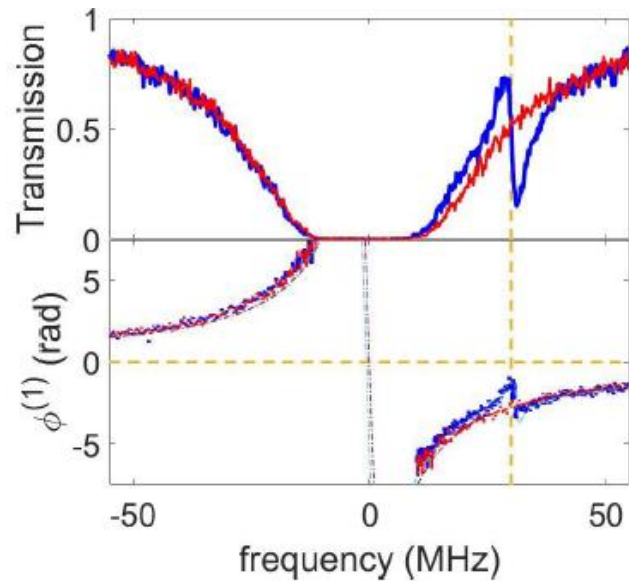


2 δ functions: decay constant doubles, so binding energy quadruples

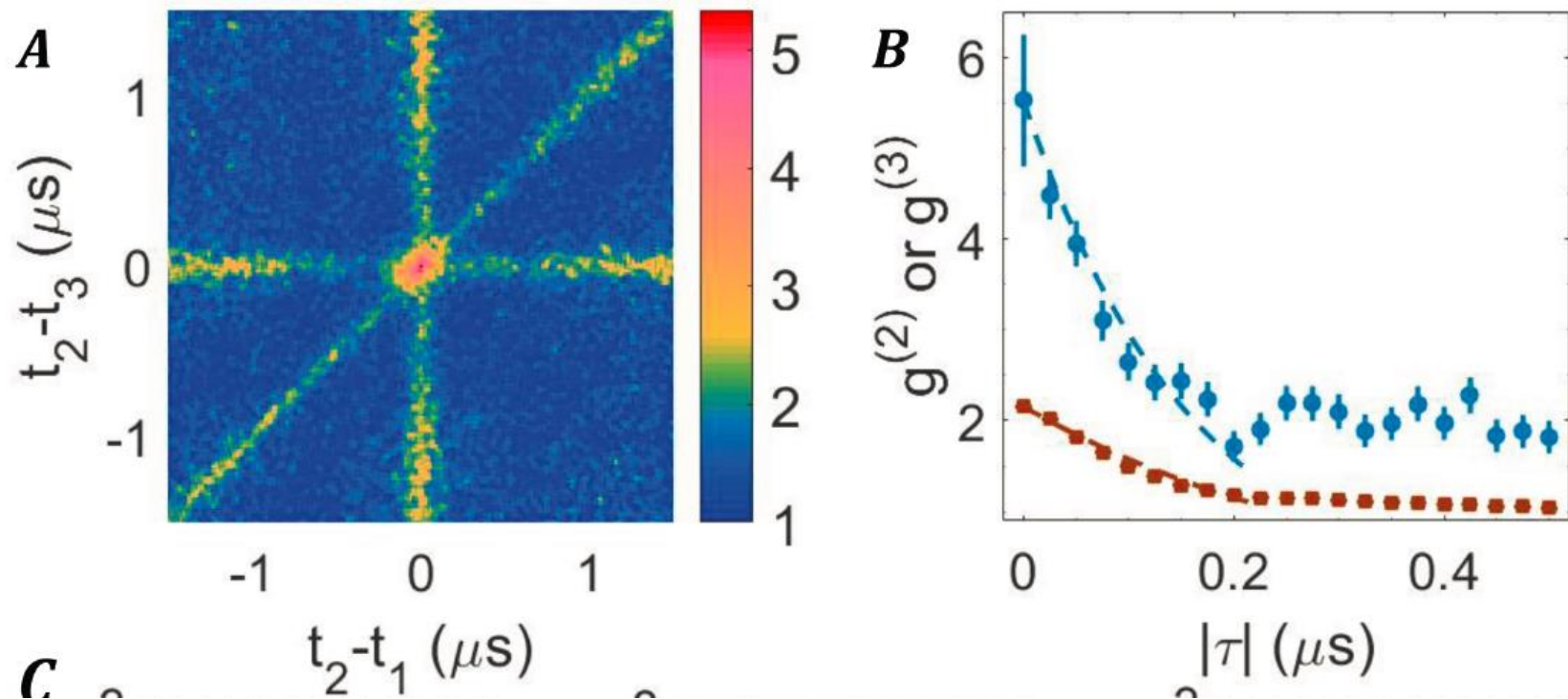
Three-photon binding



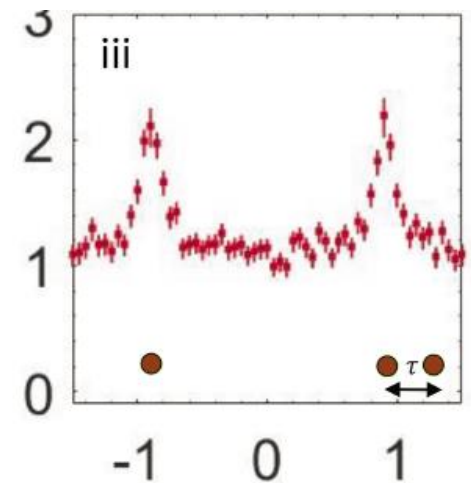
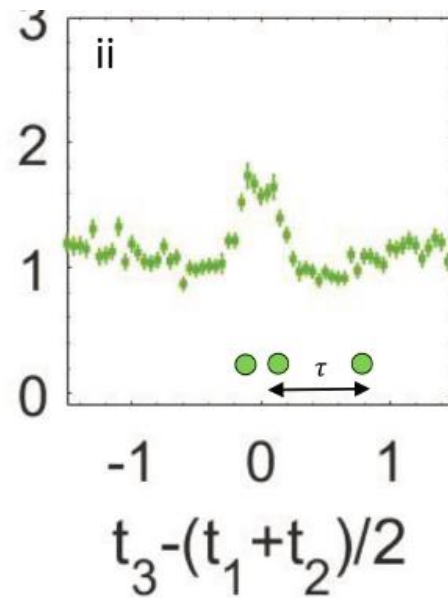
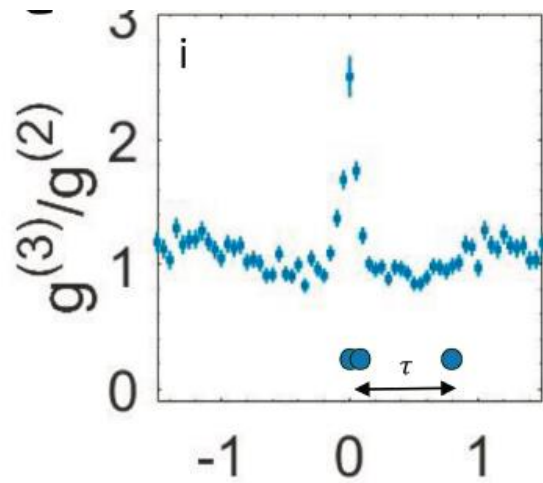
Three-photon correlations - setup



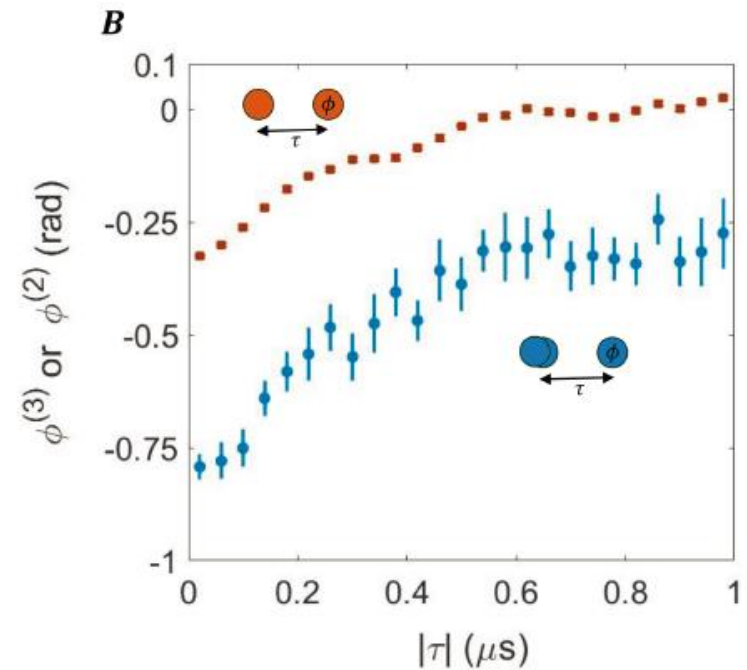
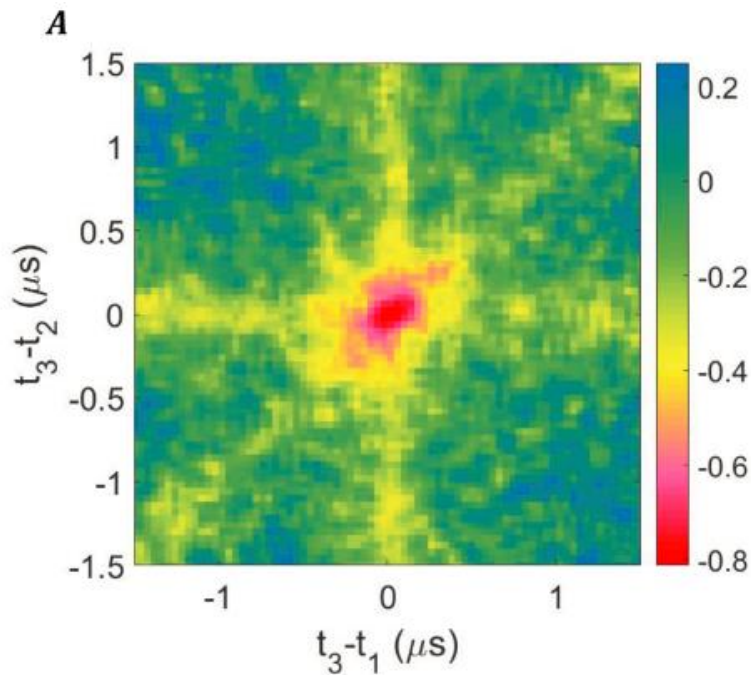
Three-photon correlation measurements



Three-photon correlation measurements



Conditional three-photon phase

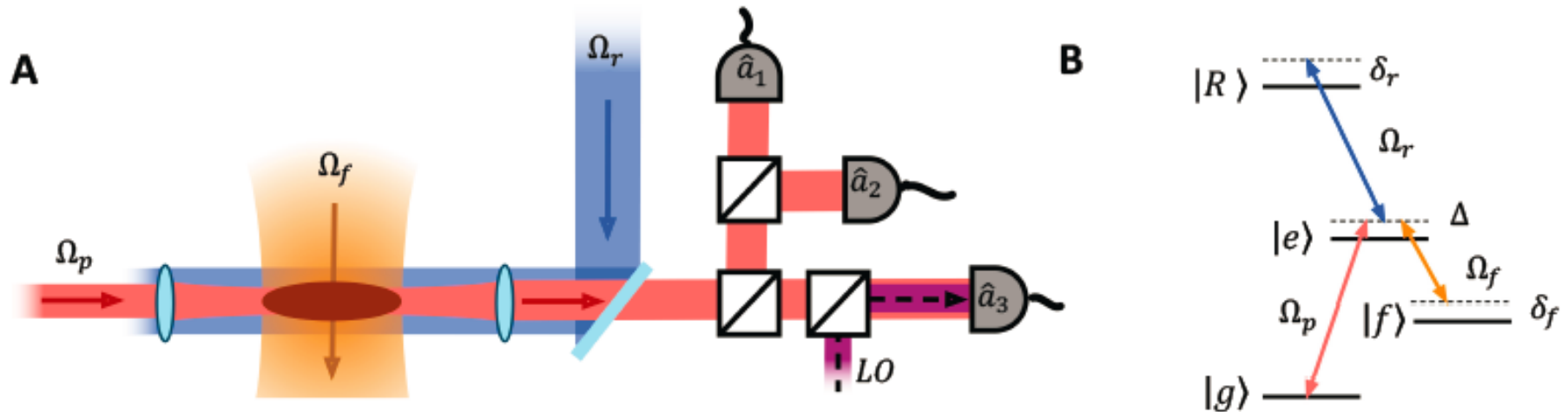


Repulsive interactions between photons

Repulsive photons in a quantum nonlinear medium.

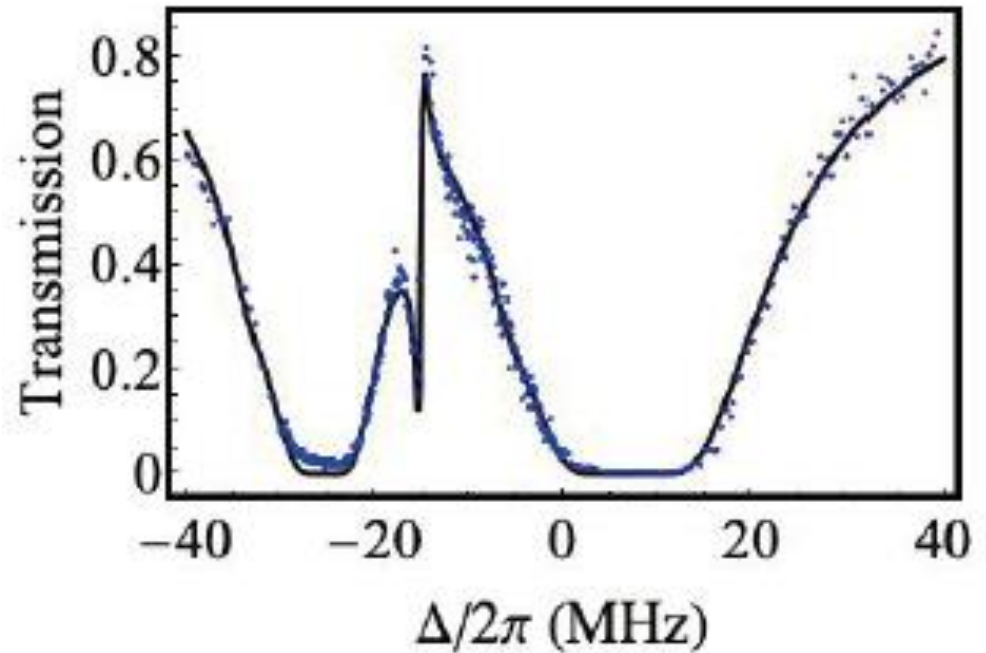
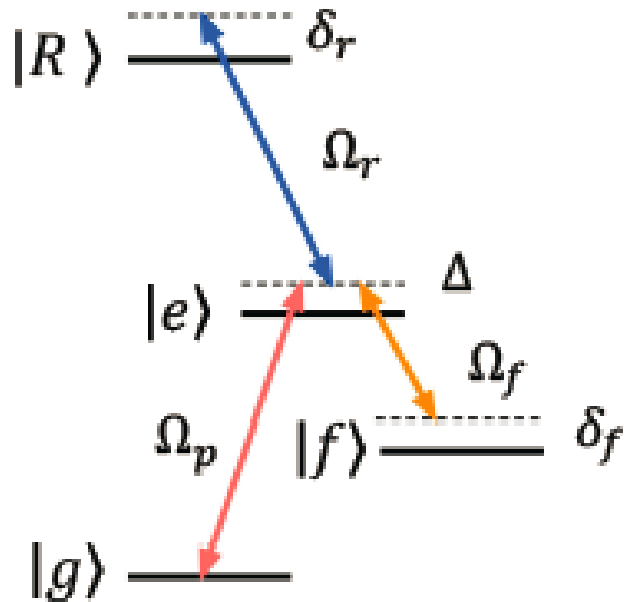
S.H. Cantu, A.V. Venkatramani, W. Xu, L. Zhou, B. Jelenković, M.D. Lukin, and V. Vuletić, Nature Physics 16, 921–925 (2020);

Star EIT level scheme for engineering photon-photon interactions



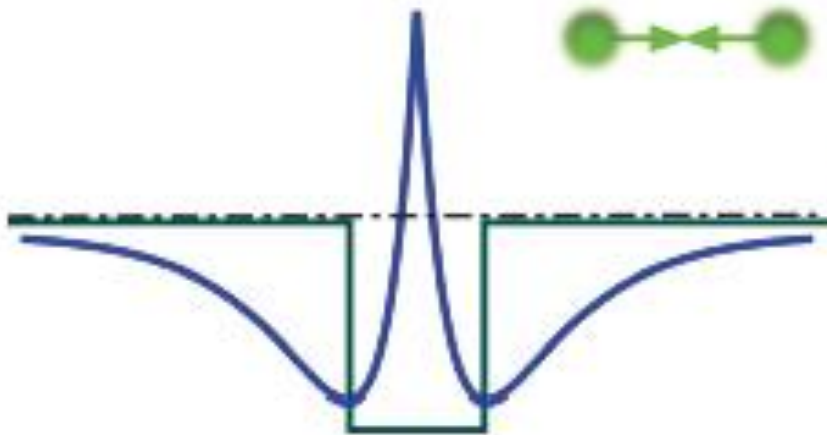
Double EIT scheme: more controllable parameters;
Control interactions separately from mass or group velocity.

Star EIT level scheme for engineering photon-photon interactions

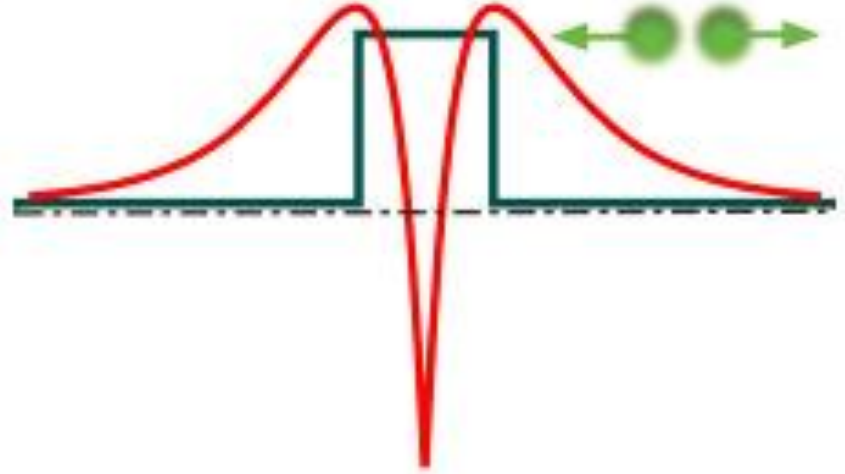


Double EIT, one with interacting Rydberg state, one with non-interacting state.

Expectation for correlation functions for positive mass

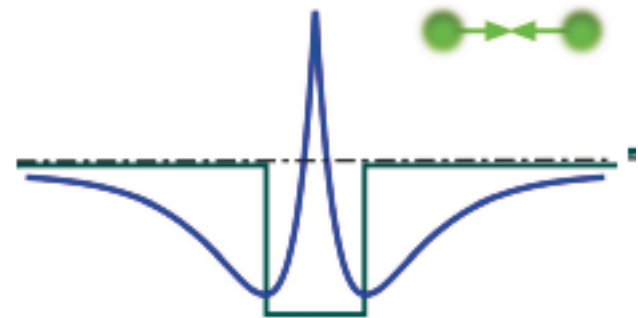
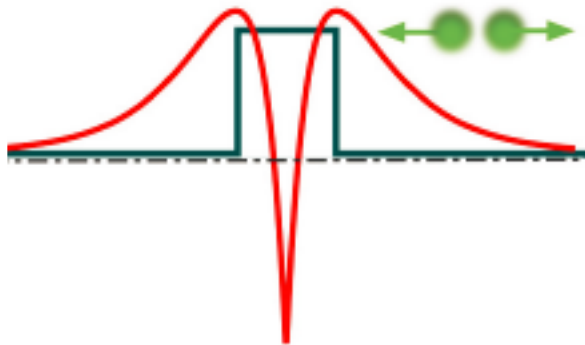
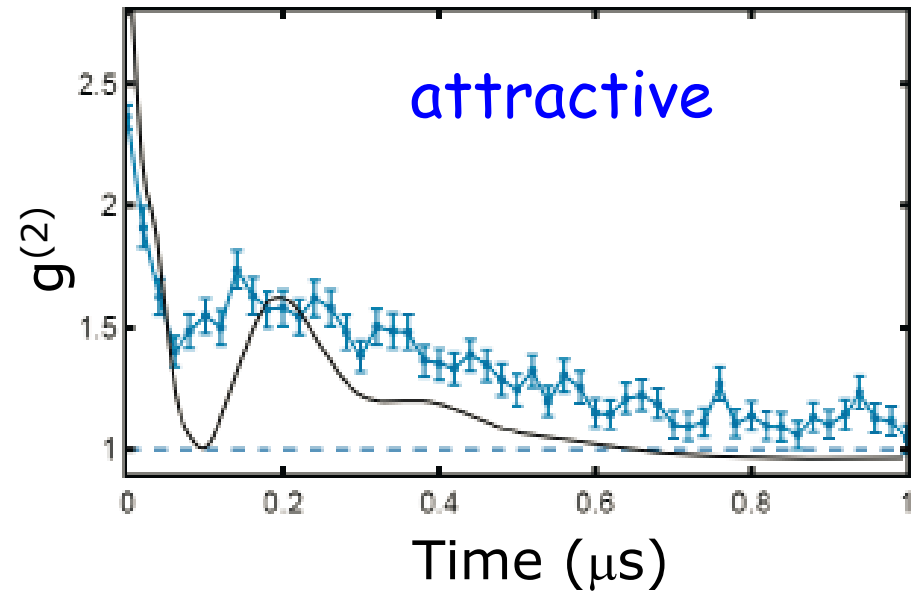
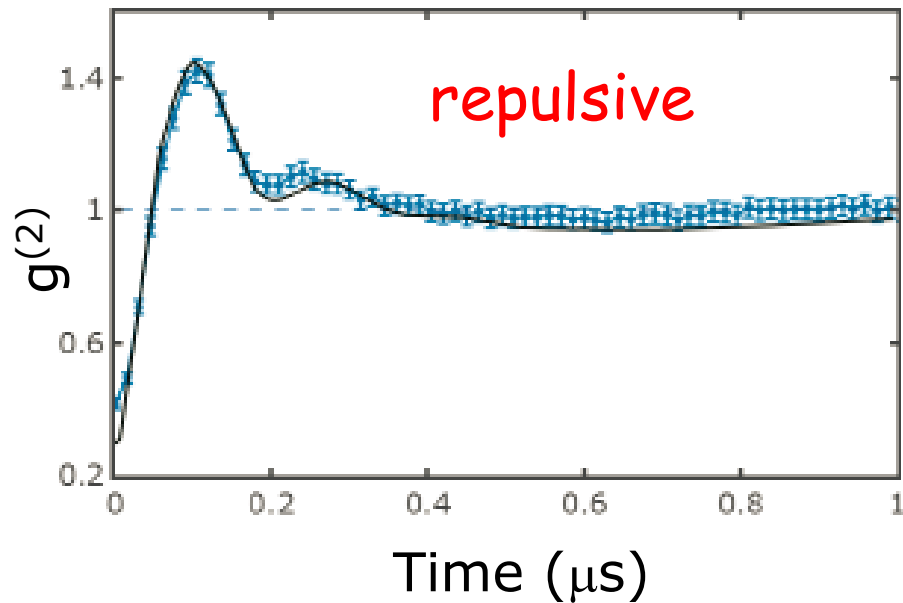


Attractive interactions:
bound state



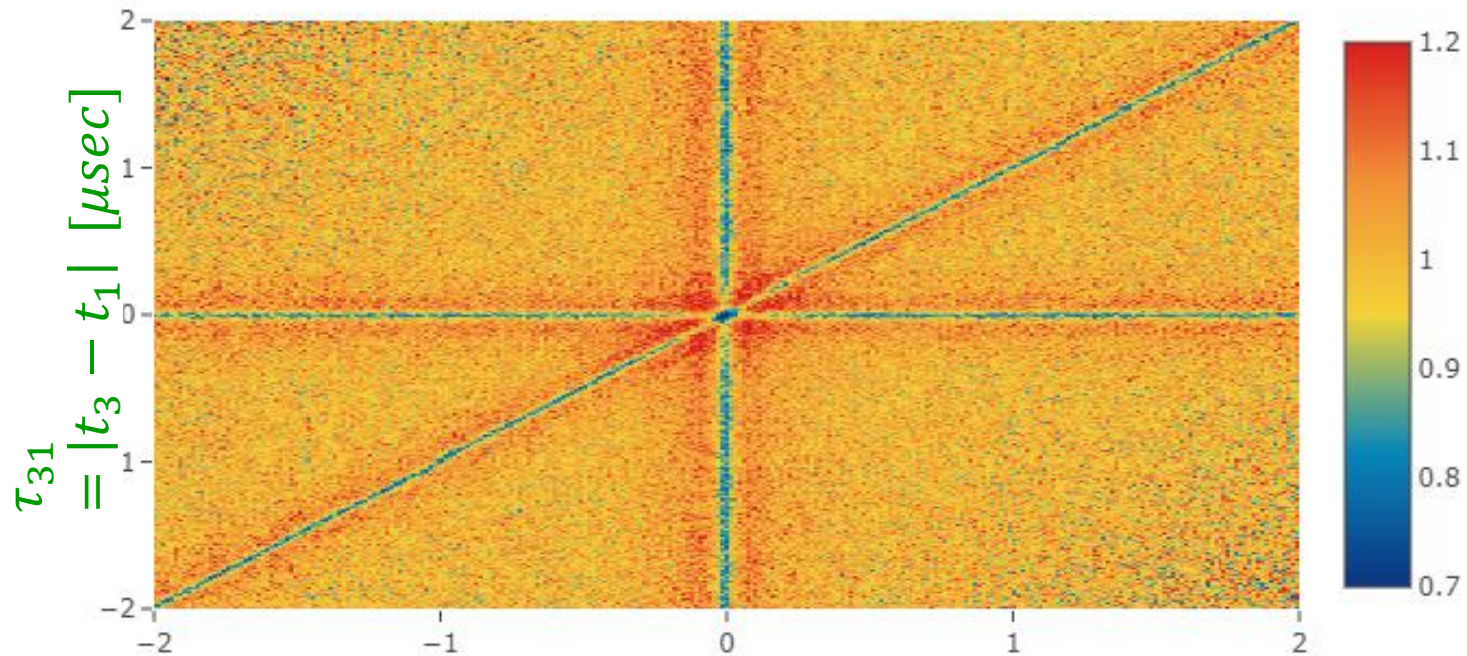
Repulsive interactions:
Antibunching of photons

Controlling the sign of the interactions



Two- and three-photon repulsive interactions

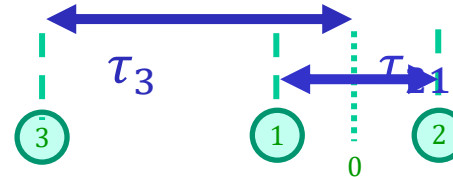
$$g^3(\tau_{31}, \tau_{21})$$



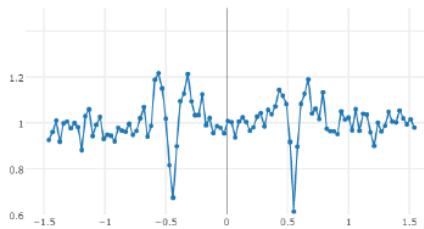
$$\tau_{21} = |t_2 - t_1| [\mu\text{sec}]$$

Towards crystals of photons

$$g^3\left(-\frac{\tau_{21}}{2}, \frac{\tau_{21}}{2}, \tau_3\right)$$

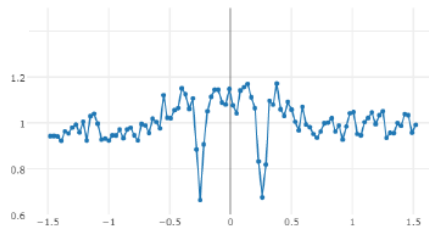


$\tau_{21} = 1.0 \mu\text{sec}$



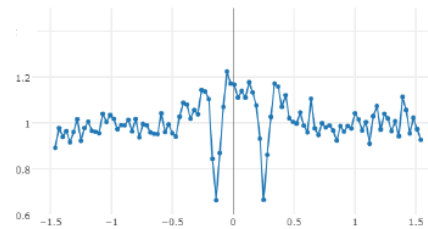
$\tau_3 [\mu\text{sec}]$

$\tau_{21} = 0.5 \mu\text{sec}$



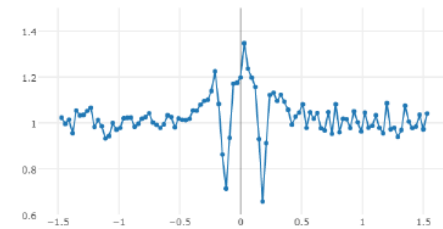
$\tau_3 [\mu\text{sec}]$

$\tau_{21} = 0.4 \mu\text{sec}$



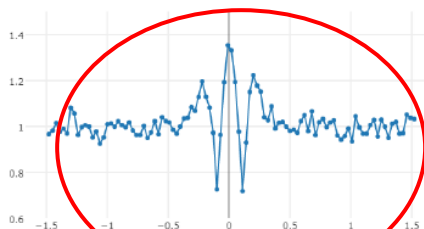
$\tau_3 [\mu\text{sec}]$

$\tau_{21} = 0.3 \mu\text{sec}$



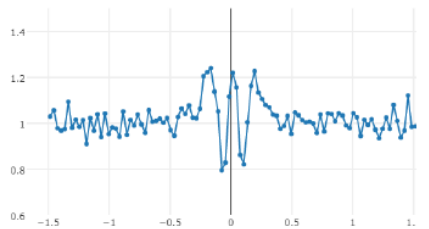
$\tau_3 [\mu\text{sec}]$

$\tau_{21} = 0.2 \mu\text{sec}$



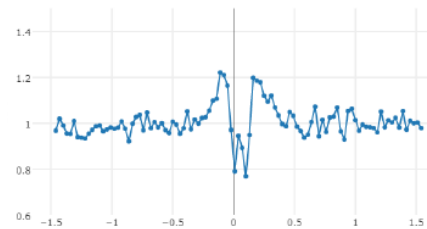
$\tau_3 [\mu\text{sec}]$

$\tau_{21} = 0.15 \mu\text{sec}$



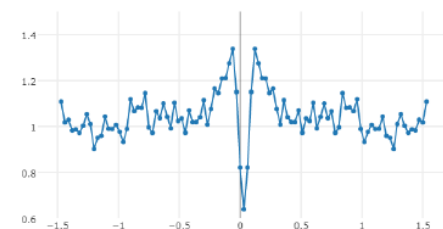
$\tau_3 [\mu\text{sec}]$

$\tau_{21} = 0.1 \mu\text{sec}$



$\tau_3 [\mu\text{sec}]$

$\tau_{21} = 0 \mu\text{sec}$



$\tau_3 [\mu\text{sec}]$

Summary

- One can use strong Rydberg interactions + EIT to generate highly non-classical optical devices, such as a two-photon absorptive filter, or two-photon (polariton) bound states.
- Can one apply the same principle in solid-state system where one could build “real” devices that you can buy some day to use in your experiments?
- Applications to quantum networks.