



---

# Correlation of cascaded photons: Two-photon processes in the Mollow Regime

---

**Alexander Carmele**, Julian Schleibner, and Andreas Knorr  
Technische Universität Berlin, Institut für Theoretische Physik

Samir Bounouar, Max Strauß, and Stephan Reitzenstein  
Technische Universität Berlin, Institut für Festkörperphysik

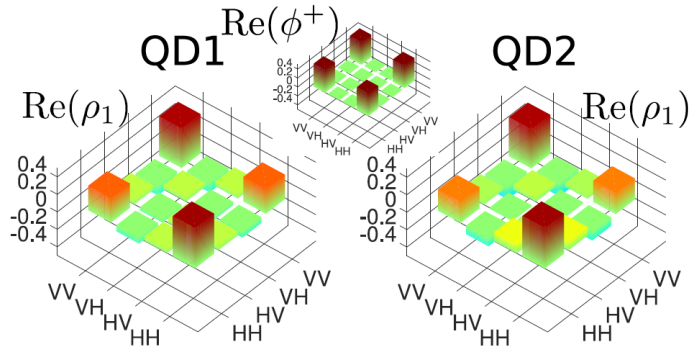
---

# Recent successes in the field of semiconductor quantum optics

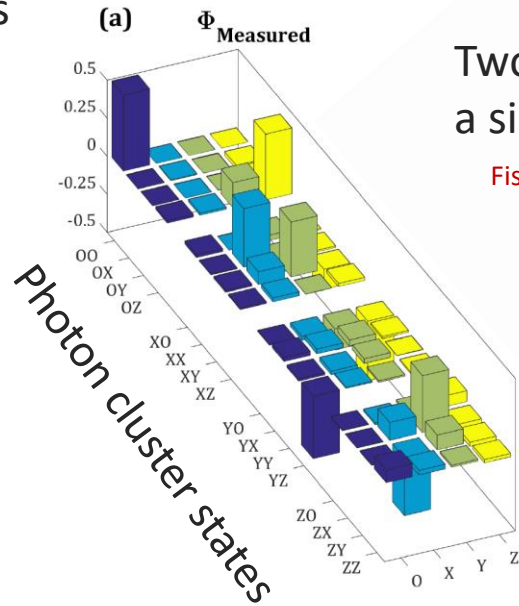
---

# Recent highlights in Semiconductor Quantum Optics

## Generation of highly entangled photon pairs

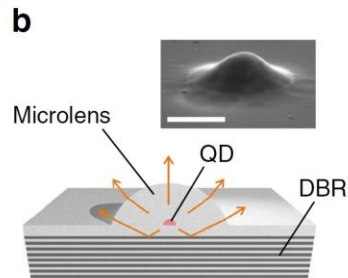
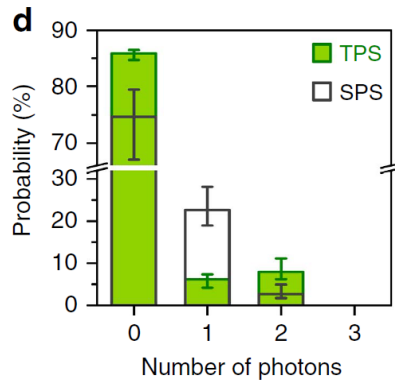
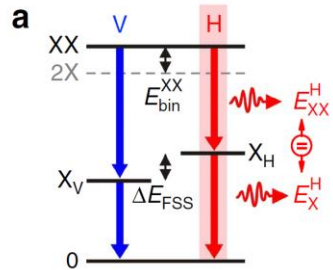
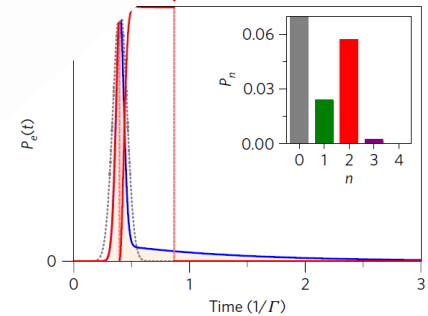


Bounouar et al, *Appl.Phys.Lett.* 112, 153107 (2018)  
Winik et al, *Phys. Rev. B* 95, 235435 (2017).



## Two-photon processes of a single Quantum dot

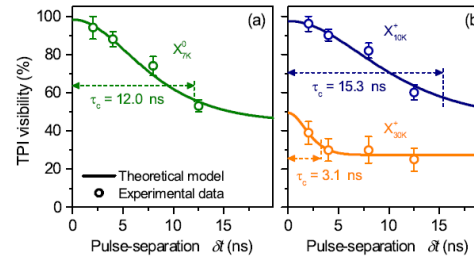
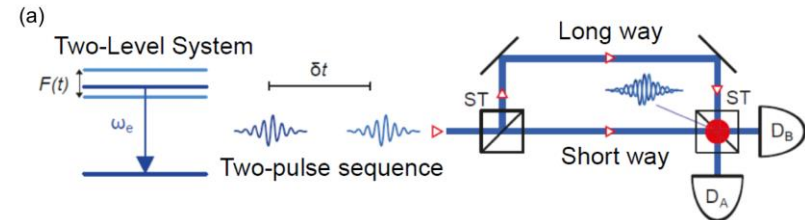
Fisher et al, *Nat.Physics* 13, 649 (2017)



## Deterministic twin-photon source

Heindel et al, *Nat. Comm.* 8, 14870 (2017)

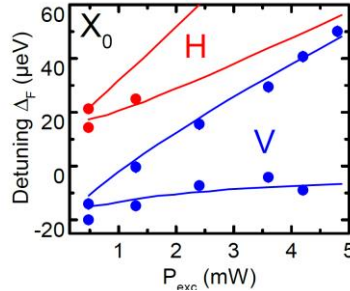
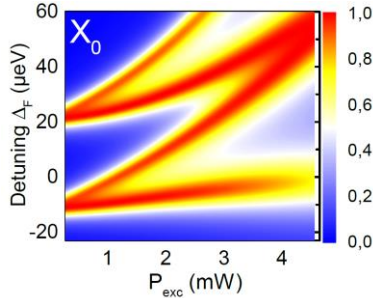
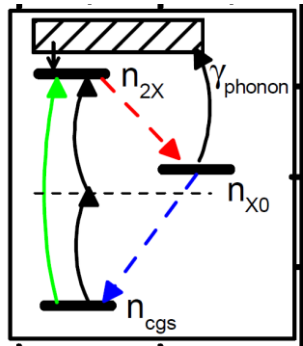
Schwartz et al, *Science* 354, 434 (2016)



## Generation of indistinguishable photons (up to 99% visibility)

Thoma et al, *PRL* 116, 033601 (2016)  
Ding et al, *PRL* 116, 020401 (2016)  
Kim et al, *Optica* 3, 577 (2016)  
Wang et al, *PRL* 116, 213601 (2016)

# N-Photon bundles in cascade processes: Two-Photon resonance fluorescence

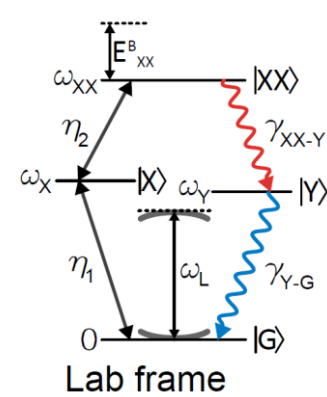
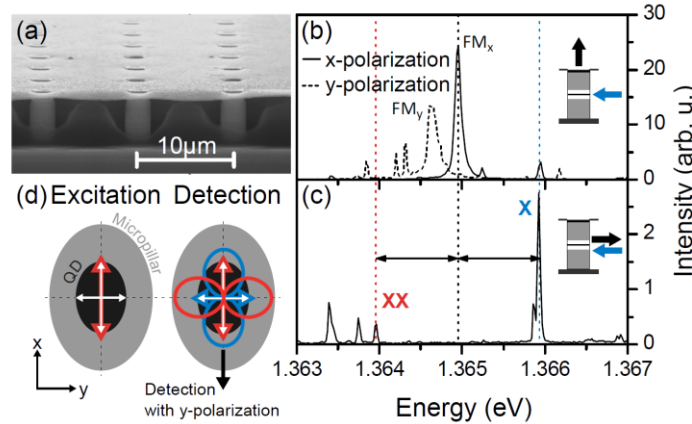
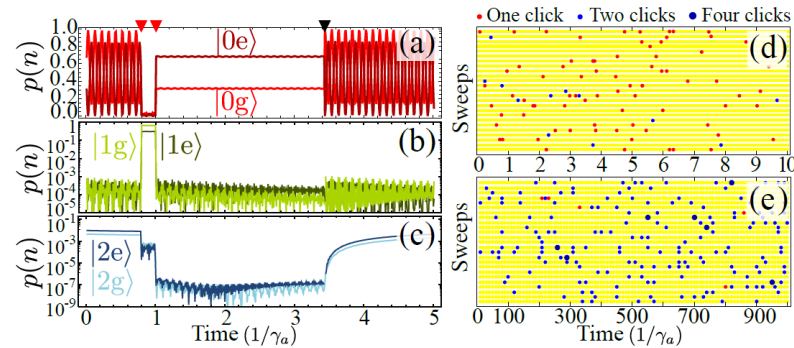


Cavity-assisted two-photon excitation with directionality in the nonlinear excitation regime

Hargart et al, Phys. Rev. B 93, 115308 (2016)

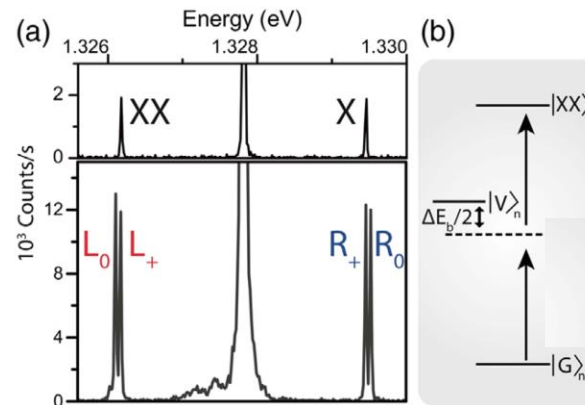
Ardelt et al, Phys. Rev. B 90, 241404 (R) (2014)

Phonon-assisted emission processes and comparison between single-photon and two-photon excitations



Theoretical proposal via N-photon bundles in leapfrog processes

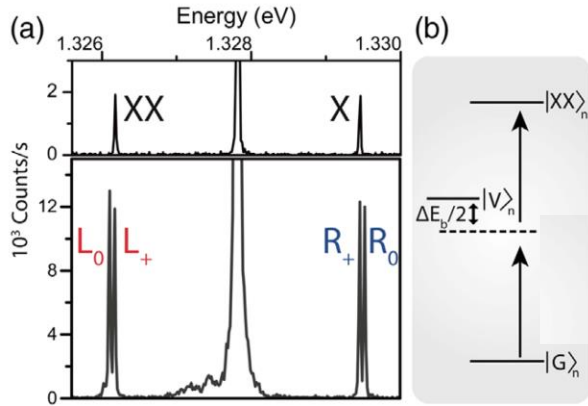
Munoz et al, Nat. Photonics 8, 550 (2014)



Mollow physics in cascade processes

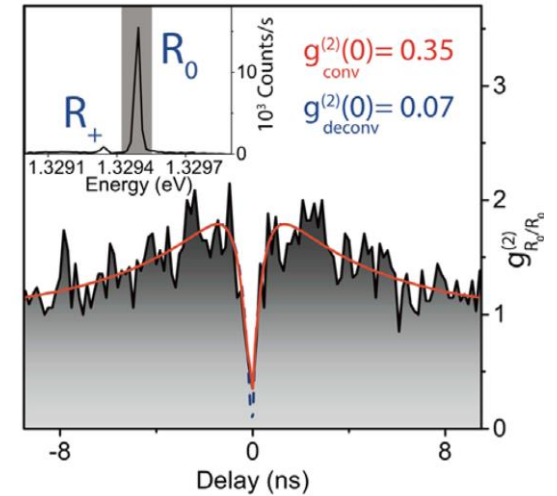
Bounouar et al, PRL 118, 233601 (2017)

# Mollow physics in the two-photon regime – polarization selective

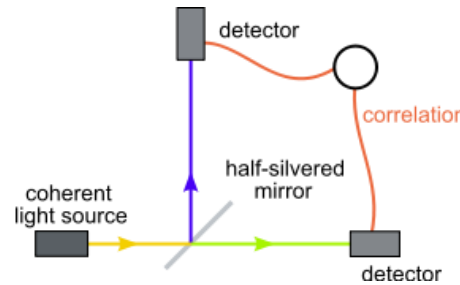
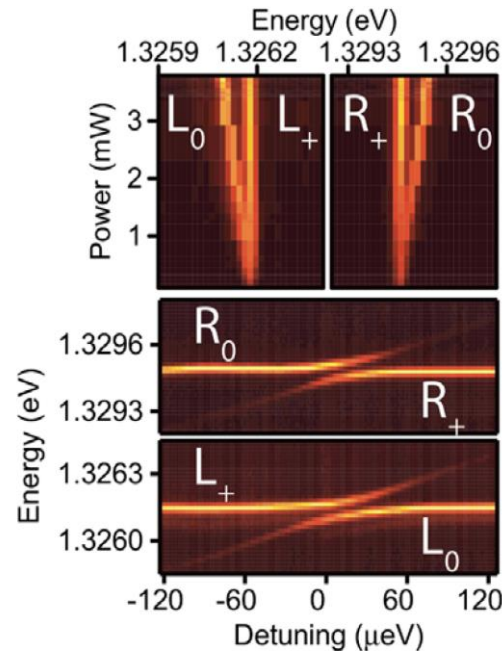


Antibunching for single lines confirm the non-classical character of the quantum light source

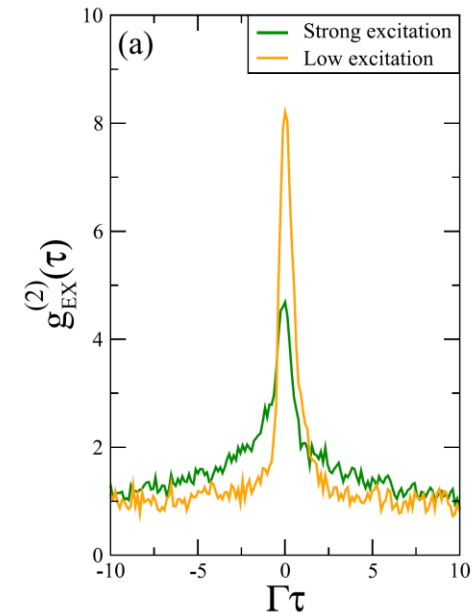
Bounouar et al, PRL 118, 233601 (2017)



Two-photon driving in resonance with biexciton and off-resonant with exciton



Unexpected behavior for combined biexciton and exciton photons – no antibunching is visible



---

# Theory of two-photon Mollow physics

---

Hamiltonian in two-photon resonance with exciting laser field and radiative decay and master equation approach:

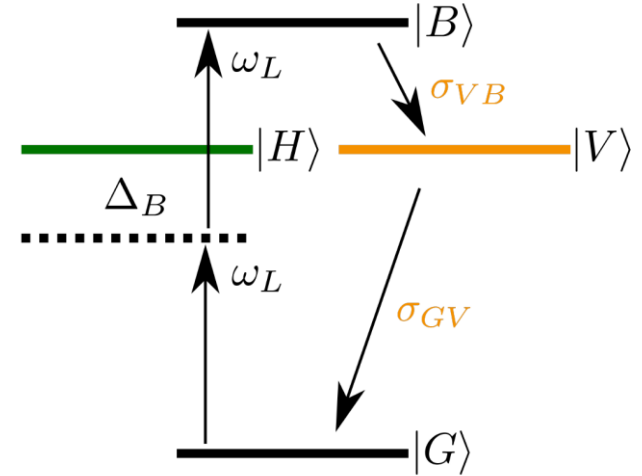
$$H = \Delta (\sigma_{HH} + \sigma_{VV}) + \Omega_L (\sigma_{GH} + \sigma_{HB} + \text{h.c.})$$

$$\dot{\rho} = -i [H, \rho] + \Gamma_X \sum_{i=H,V} (\mathcal{D}[\sigma_{Gi}] + \mathcal{D}[\sigma_{iB}]) \rho$$

Set of differential equation can be drastically reduced if symmetries and time-scales are taken into account

$$\left. \begin{aligned} \dot{\rho}_{GG} &= 2\Gamma_X (\rho_{HH} + \rho_{VV}) - i\Omega_L (\rho_{HG} - \rho_{GH}), \\ \dot{\rho}_{HG} &= -(\Gamma_X + i\Delta) \rho_{HG} - i\Omega_L (\rho_{BG} + \rho_{GG} - \rho_{HH}), \\ \dot{\rho}_{HH} &= -2\Gamma_X (\rho_{HH} - \rho_{BB}) - i\Omega_L (\rho_{GH} - \rho_{HG} + \rho_{BH} - \rho_{HB}), \\ \dot{\rho}_{BG} &= -2\Gamma_X \rho_{BG} - i\Omega_L (\rho_{HG} - \rho_{BH}), \\ \dot{\rho}_{BH} &= -(3\Gamma_X - i\Delta) \rho_{BH} - i\Omega_L (\rho_{HH} - \rho_{BB} - \rho_{BG}), \\ \dot{\rho}_{BB} &= -4\Gamma_X \rho_{BB} - i\Omega_L (\rho_{HB} - \rho_{BH}), \\ \dot{\rho}_{VV} &= -2\Gamma_X (\rho_{VV} - \rho_{BB}). \end{aligned} \right\}$$

$$\begin{aligned} \dot{D}(t) &= -\Gamma [D(t) + \Sigma_0] - i\Omega B(t) \\ \dot{B}(t) &= -\Gamma B(t) - i\Omega D(t). \end{aligned}$$



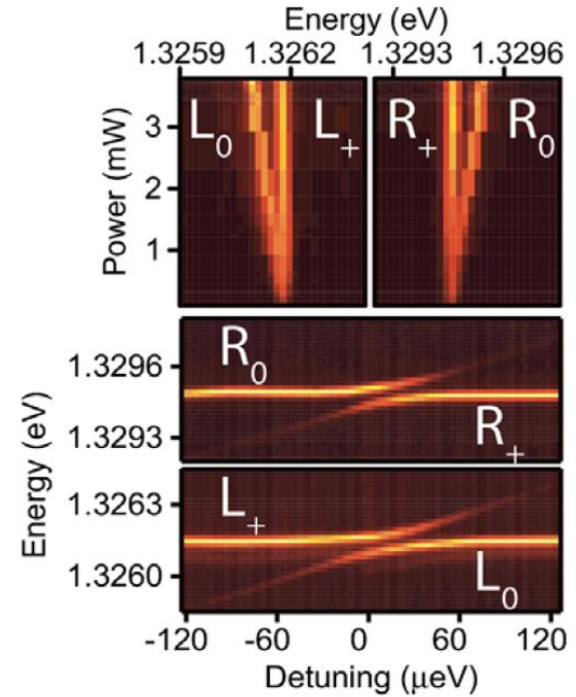
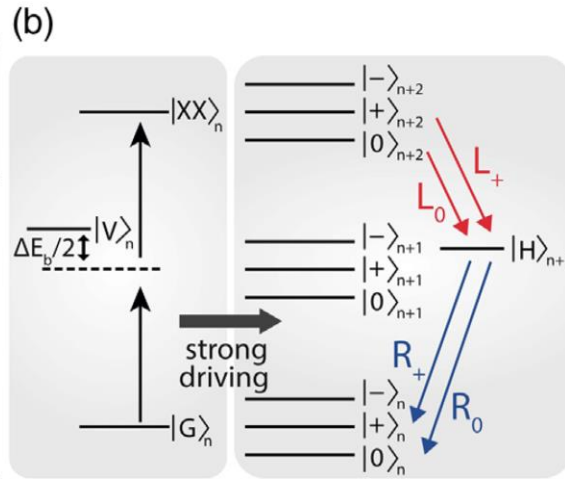
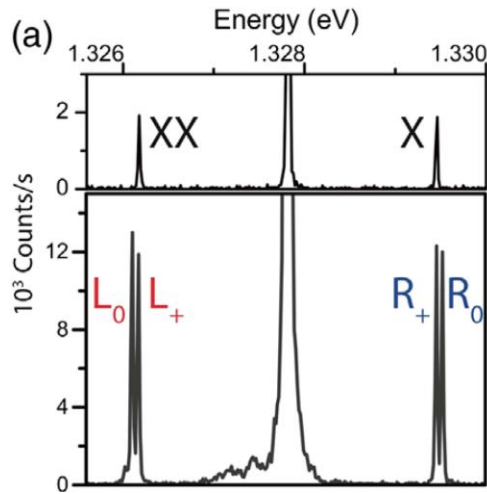
Schleibner, submitted, arXiv: 1710.03031

Benchmark the quantum optical power spectrum signal:

$$S_i(\omega) = \text{Re} \left[ \lim_{t \rightarrow \infty} \int_0^\infty \langle c_i^\dagger(t) c_i(t + \tau) \rangle e^{i(\omega - \omega_L)\tau} d\tau \right] \quad c_i(t) = \sigma_{iB}(t) + \sigma_{Gi}(t)$$

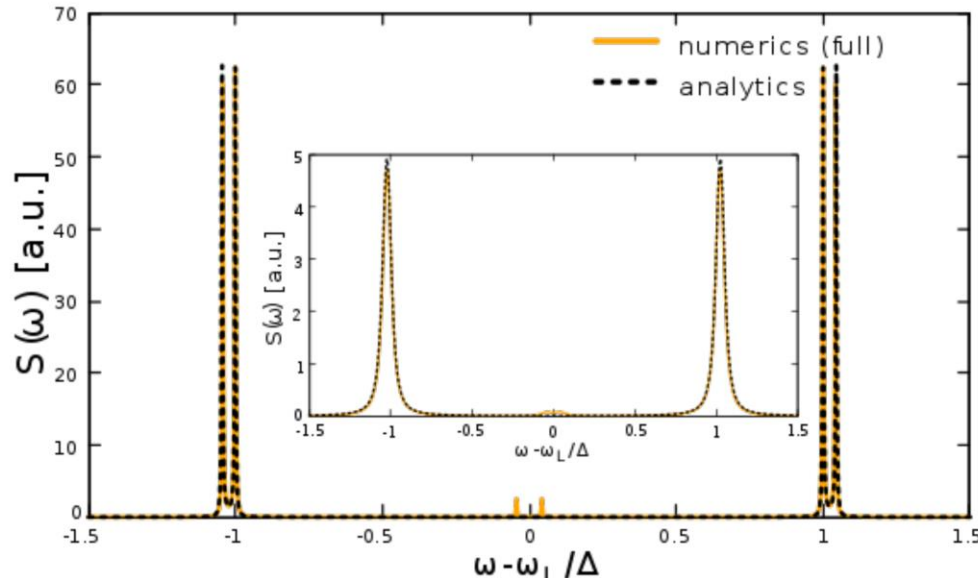


# Benchmark the experimental signal with theoretical modelling



Bounouar et al, PRL 118, 233601 (2017)

Splitting occurs for strong driving, but only one peak shifts



Two-photon spectrum solution:  
In the strong-driving limit, one peak does not shift, splitting proportional to the square of the Rabi energy

But what about the correlation function?

$$S_{BV}(\omega) = \alpha \left( \frac{\Omega_R \Gamma + \frac{\Gamma}{2}(\omega - \omega_L + \Delta + \frac{\Omega}{2} + \Omega_R)}{\Gamma^2 + [\omega - \omega_L - (-\Delta - \frac{\Omega}{2} - \Omega_R)]^2} + \frac{\Omega_R \Gamma - \frac{\Gamma}{2}(\omega - \omega_L + \Delta + \frac{\Omega}{2} - \Omega_R)}{\Gamma^2 + [\omega - \omega_L - (-\Delta - \frac{\Omega}{2} + \Omega_R)]^2} \right)$$

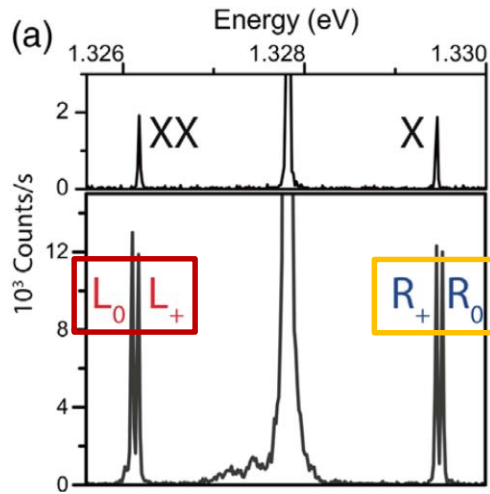
$$S_{VG}(\omega) = \alpha \left( \frac{\Omega_R \Gamma + \frac{\Gamma}{2}(\omega - \omega_L - \Delta - \frac{\Omega}{2} + \Omega_R)}{\Gamma^2 + [\omega - \omega_L - (\Delta + \frac{\Omega}{2} - \Omega_R)]^2} + \frac{\Omega_R \Gamma - \frac{\Gamma}{2}(\omega - \omega_L - \Delta - \frac{\Omega}{2} - \Omega_R)}{\Gamma^2 + [\omega - \omega_L - (\Delta + \frac{\Omega}{2} + \Omega_R)]^2} \right)$$



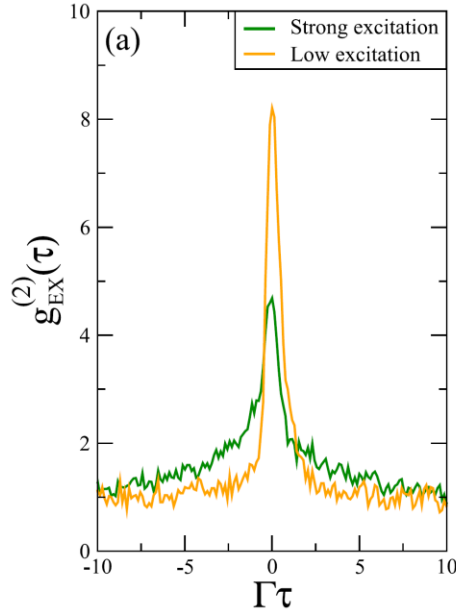
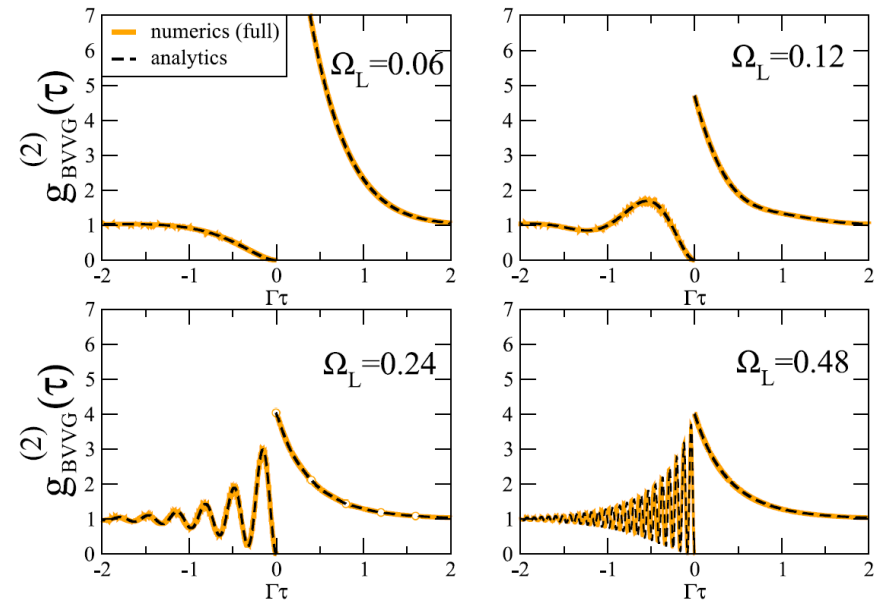
# Photon-photon correlation function in the bare-state basis

## Hanbury Brown-Twiss setup to probe photon-photon correlations

$$g_{ijkl}^{(2)}(\tau) = \frac{\rho_{ii}(\infty)\rho_{kk}^{jj}(\tau)}{\rho_{ii}(\infty)\rho_{kk}(\infty)} = \frac{\rho_{kk}^{jj}(\tau)}{\rho_{kk}(\infty)}$$



Biexciton and exciton photon correlation show in theory bunching and antibunching



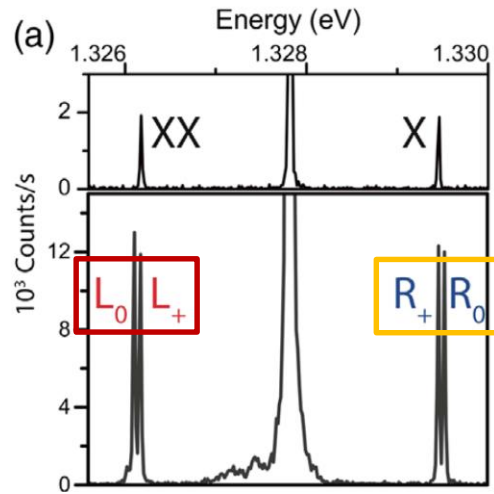
$$g_{BVVG}^{(2)}(\tau) = 2e^{-\Gamma\tau} (1 + \cosh(\Gamma\tau) + \alpha[1 + \cos(\Omega\tau)])$$

$$g_{VGBV}^{(2)}(\tau) = 1 + e^{-\Gamma\tau} (e^{-\Gamma\tau} - 2 \cos(\Omega\tau))$$

$$\alpha = \Gamma^2/\Omega^2$$

Not the full amount of biexciton and exciton photon emission is taken into account

# Photon-photon correlation in the dressed-state basis



$$|0\rangle = \frac{1}{\sqrt{2}} (|B\rangle - |G\rangle),$$

$$|+\rangle = \frac{\Omega}{\sqrt{2\Omega^2 + e_+^2}} (|G\rangle + \frac{e_+}{\Omega} |H\rangle + |B\rangle)$$

$$|-\rangle = \frac{\Omega}{\sqrt{2\Omega^2 + e_-^2}} (|G\rangle + \frac{e_-}{\Omega} |H\rangle + |B\rangle)$$

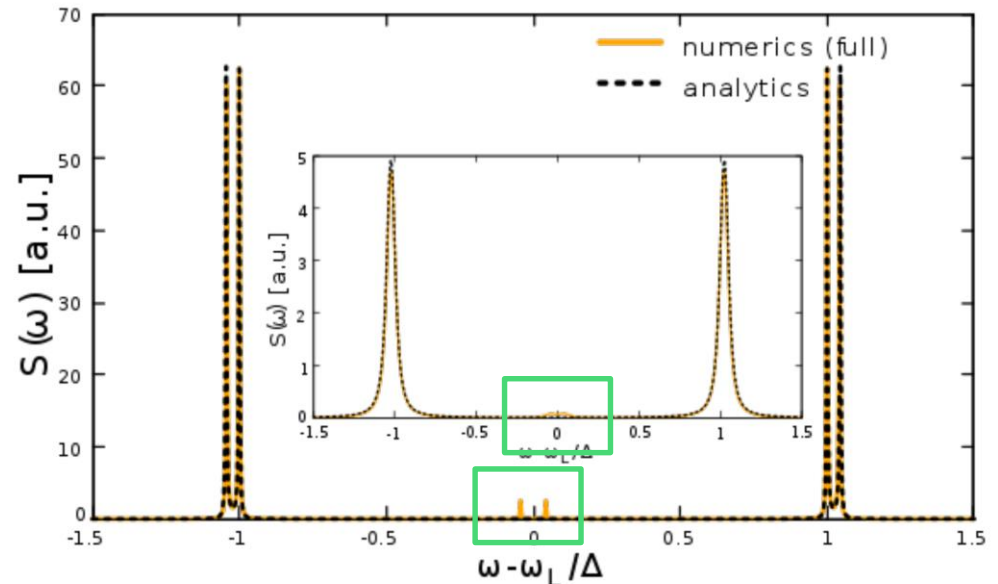
Only two out of three eigenstate enter into the measured signal (+,0)

Biexciton photons ( $\tau \leq 0$ )

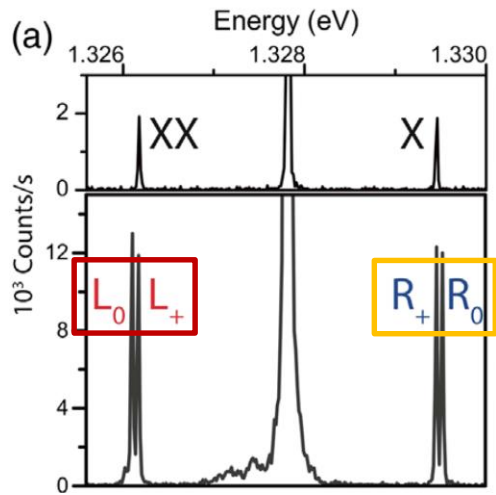
$$g_{\text{EX}}^{(2)}(\tau) = \frac{1}{4} \sum_{i,j=+,0} g_{iVjV}^{(2)}(\tau)$$

Exciton photons ( $\tau > 0$ )

$$g_{\text{EX}}^{(2)}(\tau) = \frac{1}{4} \sum_{i,j=+,0} g_{VijV}^{(2)}(\tau)$$



# Photon-photon correlation in the dressed-state basis



$$|0\rangle = \frac{1}{\sqrt{2}} (|B\rangle - |G\rangle),$$

$$|+\rangle = \frac{\Omega}{\sqrt{2\Omega^2 + e_+^2}} (|G\rangle + \frac{e_+}{\Omega} |H\rangle + |B\rangle)$$

$$|-\rangle = \frac{\Omega}{\sqrt{2\Omega^2 + e_-^2}} (|G\rangle + \frac{e_-}{\Omega} |H\rangle + |B\rangle)$$

Only two out of three eigenstate enter into the measured signal (+,0) and superponing this signal leads to very good agreement between theory and experiment

## Biexciton photons ( $\tau \leq 0$ )

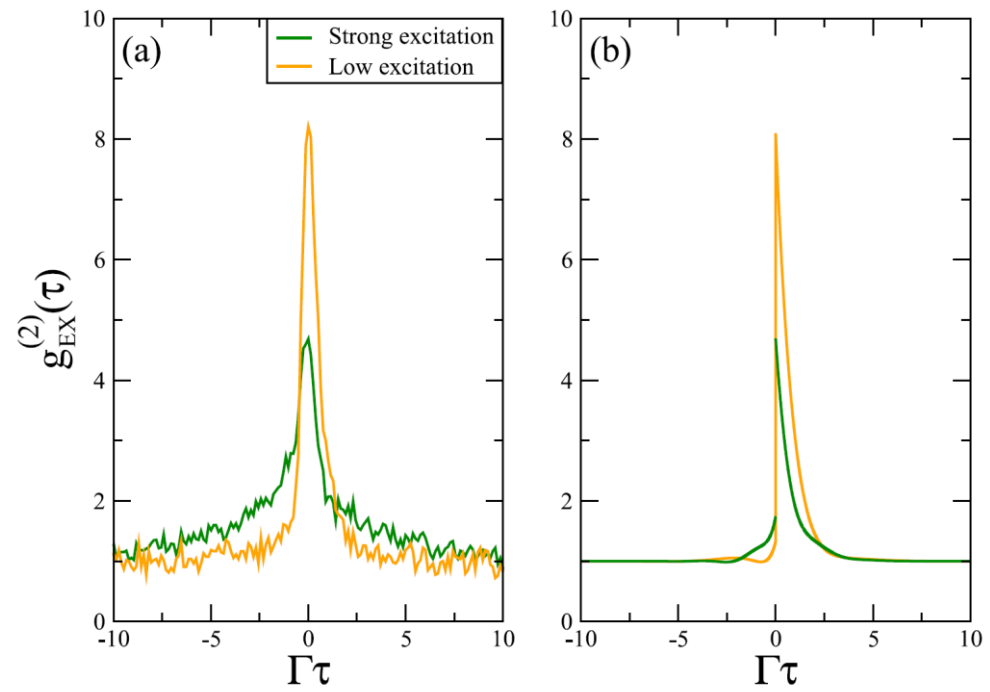
$$g_{EX}^{(2)}(\tau) = \frac{1}{4} \sum_{i,j=+,0} g_{iVV_j}^{(2)}(\tau) \quad \alpha = \Gamma^2 / \Omega^2$$

$$g_{EX}^{(2)}(\tau) = 1 + e^{-2\Gamma\tau} - \frac{2\alpha \cos(\Omega\tau)}{1 + 2\alpha} e^{-\Gamma\tau}$$

## Exciton photons ( $\tau > 0$ )

$$g_{EX}^{(2)}(\tau) = \frac{1}{4} \sum_{i,j=+,0} g_{VijV}^{(2)}(\tau)$$

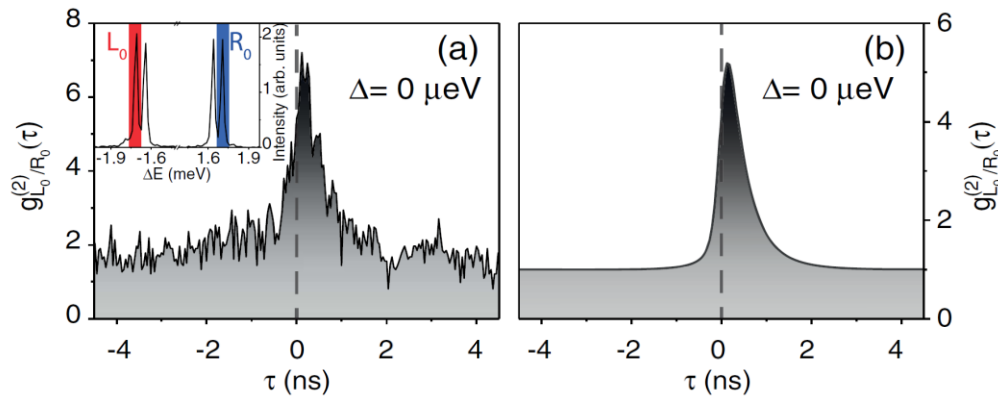
$$g_{EX}^{(2)}(\tau) = \frac{\rho_{VV}^{VV}(\tau)}{\rho_{VV}^{VV}(\infty)} = 3 + e^{-2\Gamma\tau} + 2\alpha e^{-\Gamma\tau} [1 + \cos(\Omega\tau)]$$



Deep antibunching only visible in this setup with in phase superposition of all possible photon paths

# Path-controlled vanishing and reestablished time-ordering

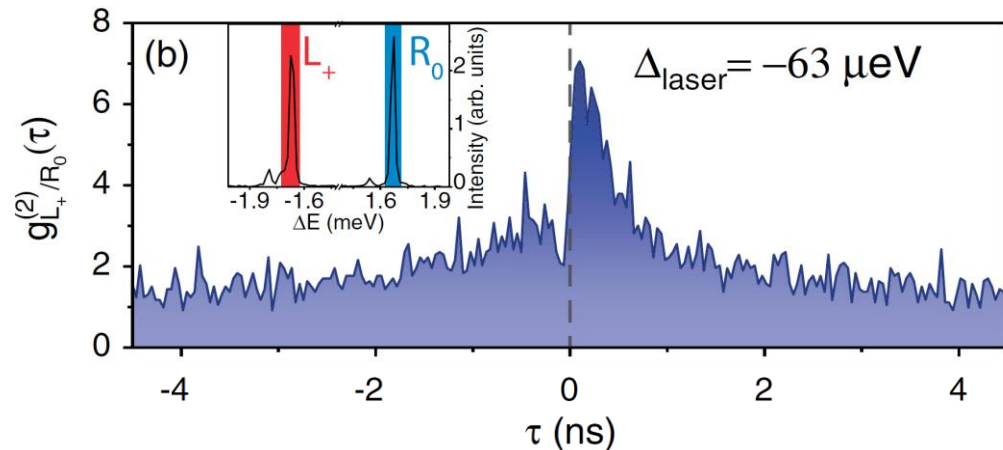
In resonance, vanishing time-order  
 in the strong driving limit



The correlation function loses information which photon has been emitted first (biexciton or exciton photon) – interesting for time-bin application and heralding

Bounouar et al, PRL 118, 233601 (2017)

Detuning shifts the weight in between the Eigenvalues to path-control the cascade process



Of resonance, time-order is reestablished – theory becomes due to detuning hard, even numerically due to transcendental Eigenvalue equations

---

Thank you for the attention!

---



**Technische Universität Berlin**

