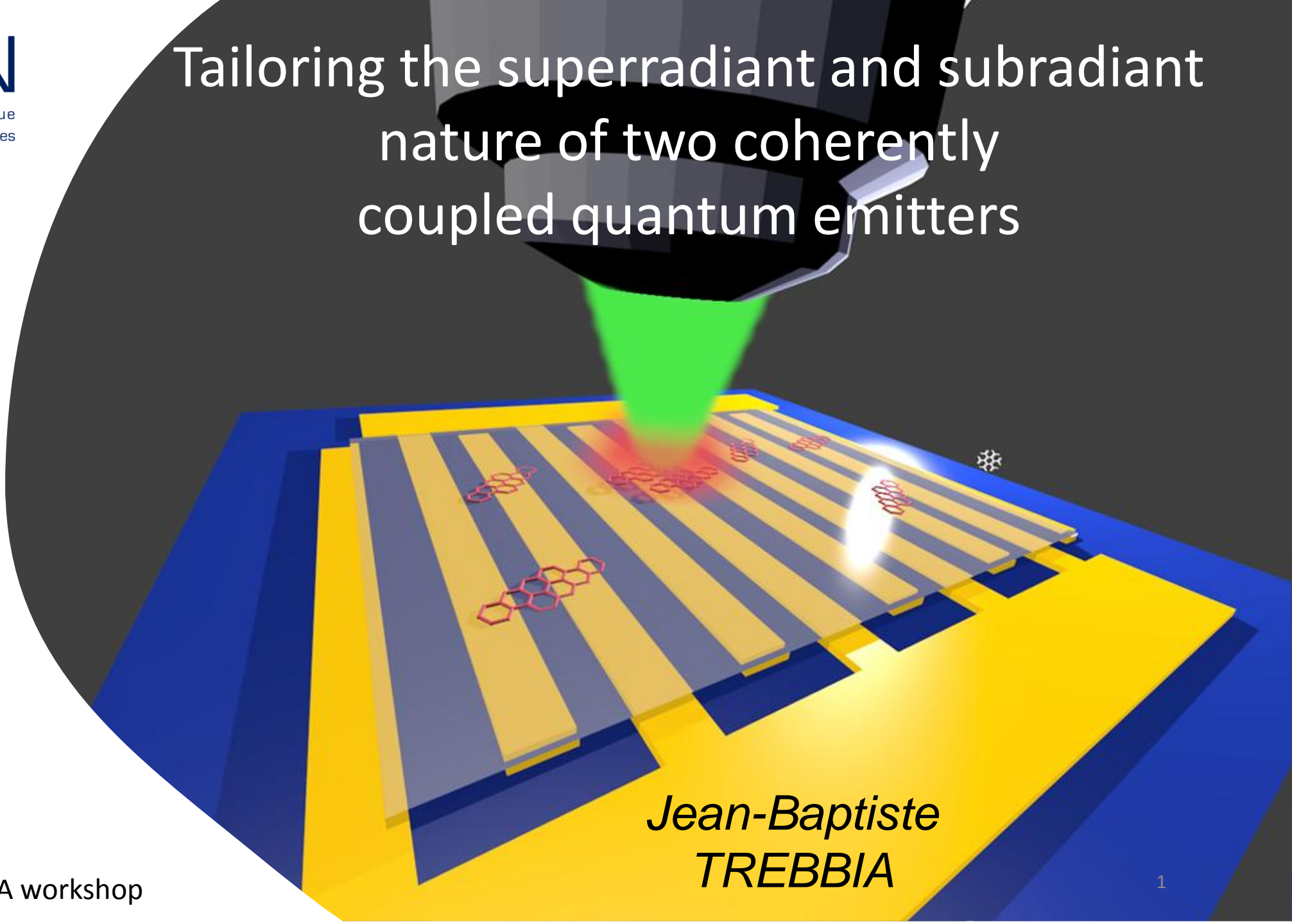


Tailoring the superradiant and subradiant nature of two coherently coupled quantum emitters



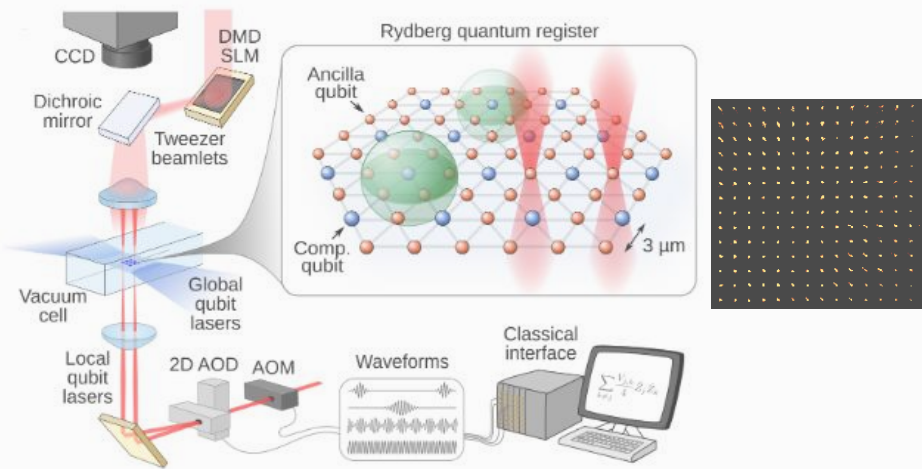
*Jean-Baptiste
TREBBIA*

Motivations

Controlled, coherent manipulation of delocalized quantum systems

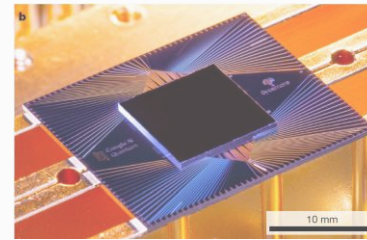
- An important challenge in modern science
- Significant applications in quantum technologies

Rydberg Atoms/ions

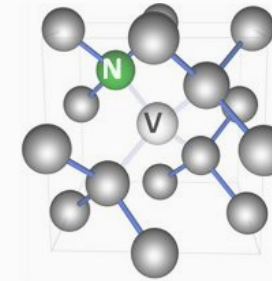


Scalable solid-state quantum devices

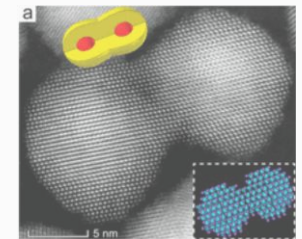
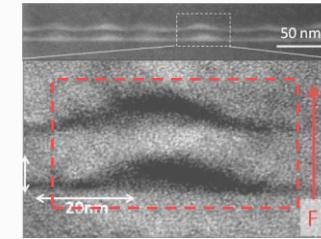
networks of strongly interacting qubits



superconducting circuits

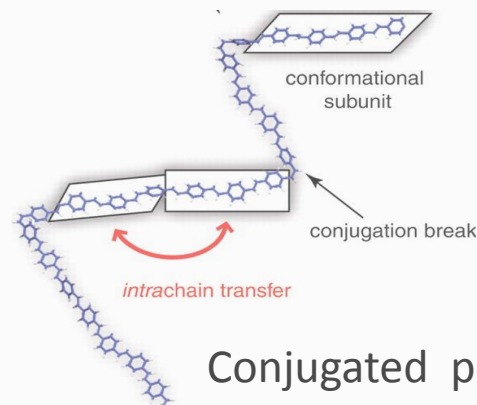


NV centers

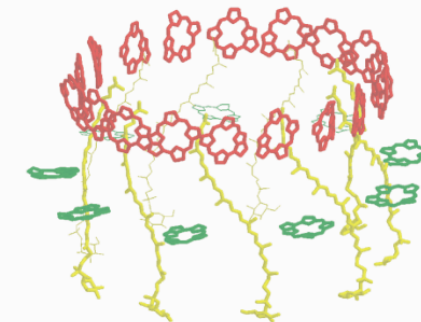


Quantum dot molecules

Collective quantum states:
at the heart of many
important phenomena



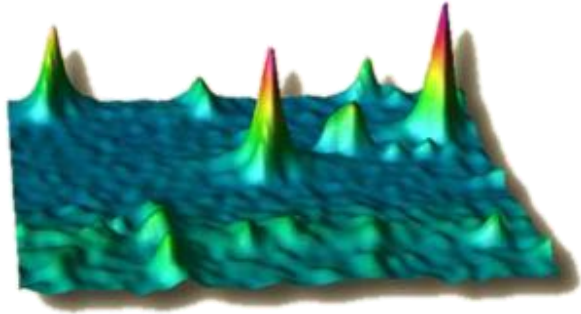
Conjugated polymers



Light harvesting complex

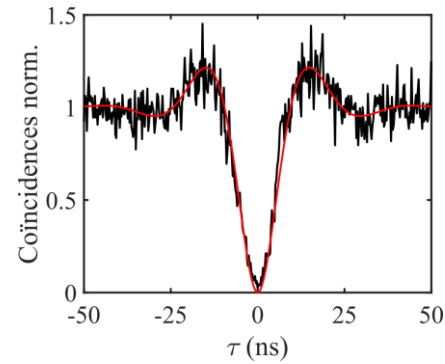
Why study individual molecules?

Photons :



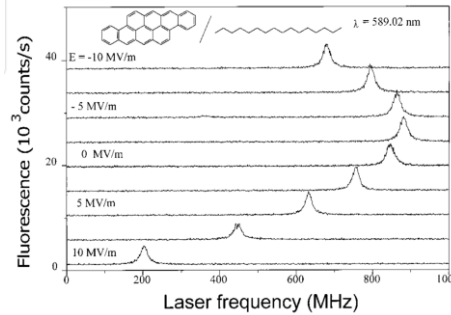
Moerner, 1989
Orrit et Bernard, 1990

single



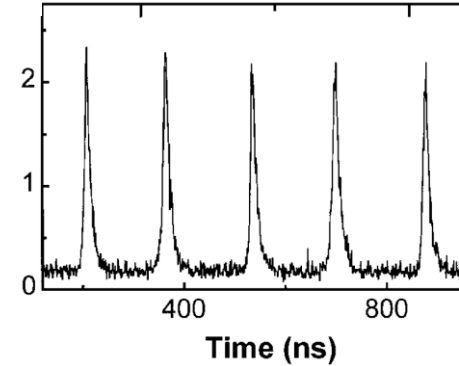
Brunel et al., 1998

tunable



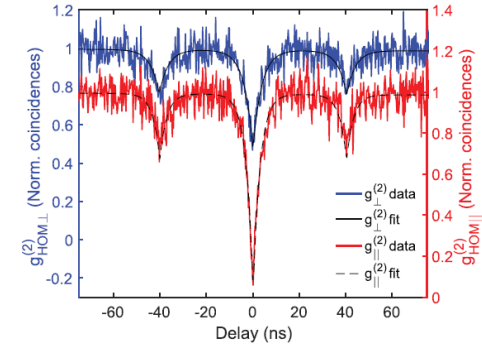
Tamarat *et al.*, 1999

On demand



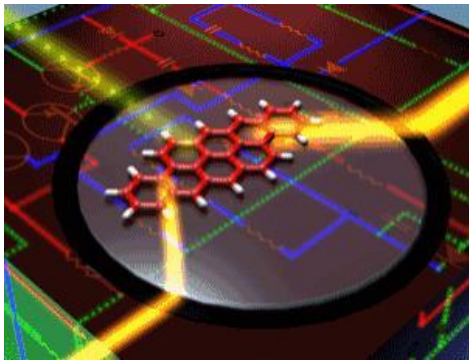
Brunel *et al.*, 1999

indistinguishable



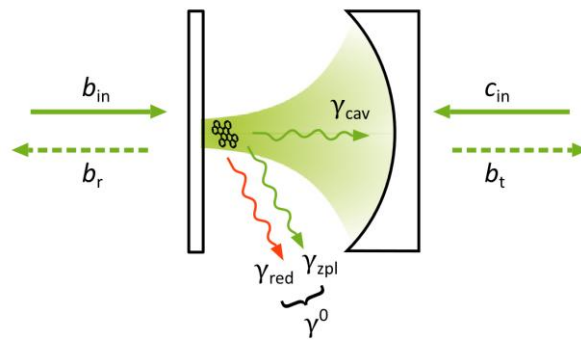
Athee *et al.*, 2008
Trebba *et al.*, 2009
Lombardi *et al.*, 2021

Optical transistor



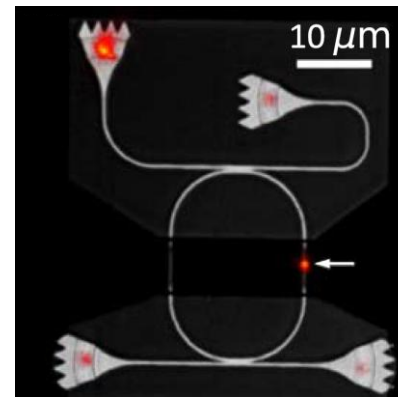
Hwang *et al.*, 2009

Molecules in cavities



Wang *et al.*, 2019

Integrated resonators

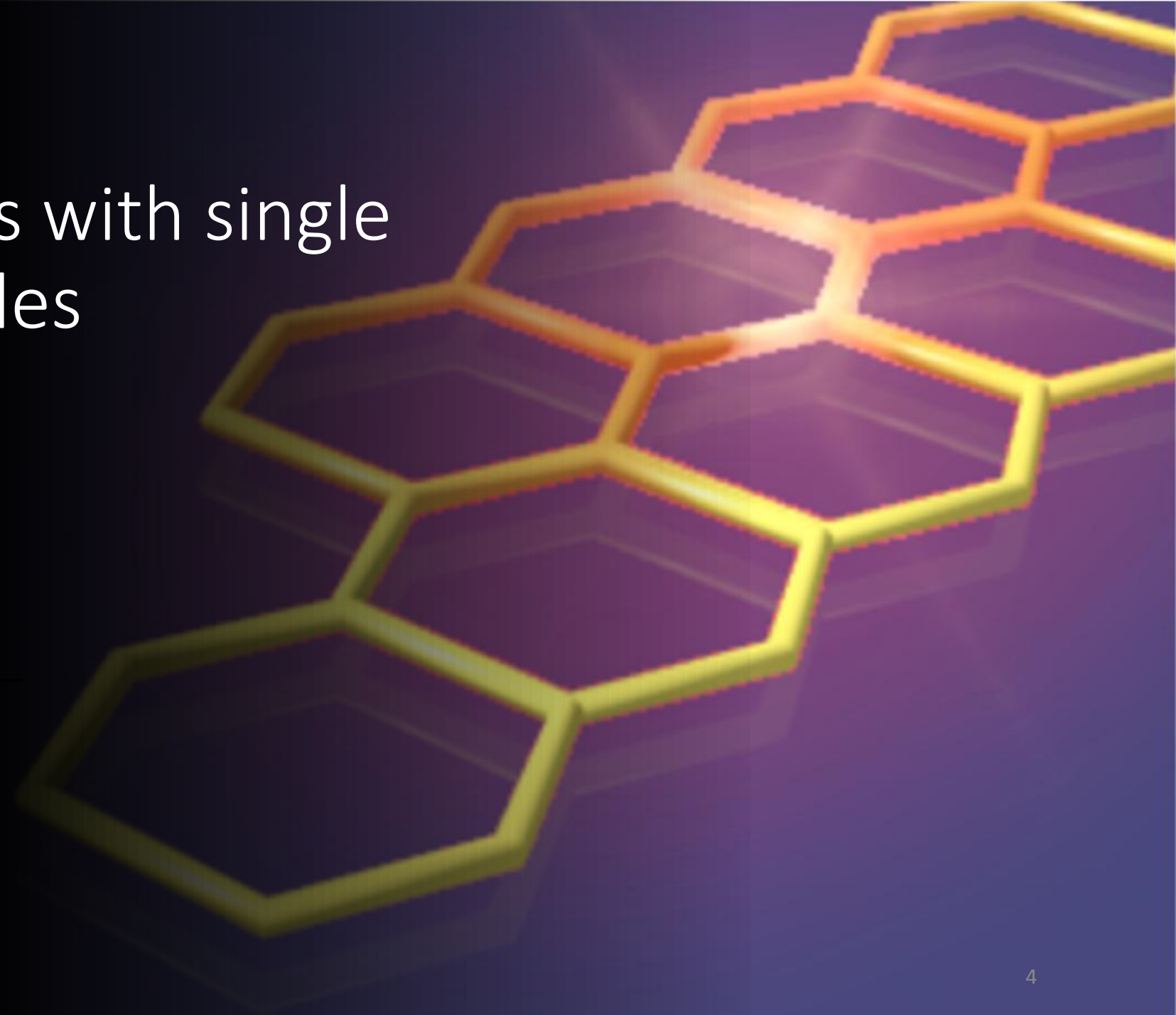


Rattenbacher *et al.*, 2019

Few studies on
molecular qubits



I. Quantum optics with single molecules

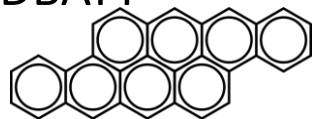


Single molecules at low temperature (3K)

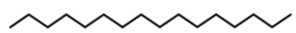
Molecules

Matrices

DBATT



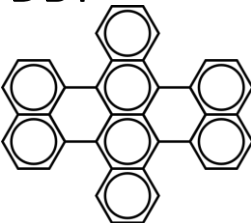
Octadecane



Naphtalene



DBT

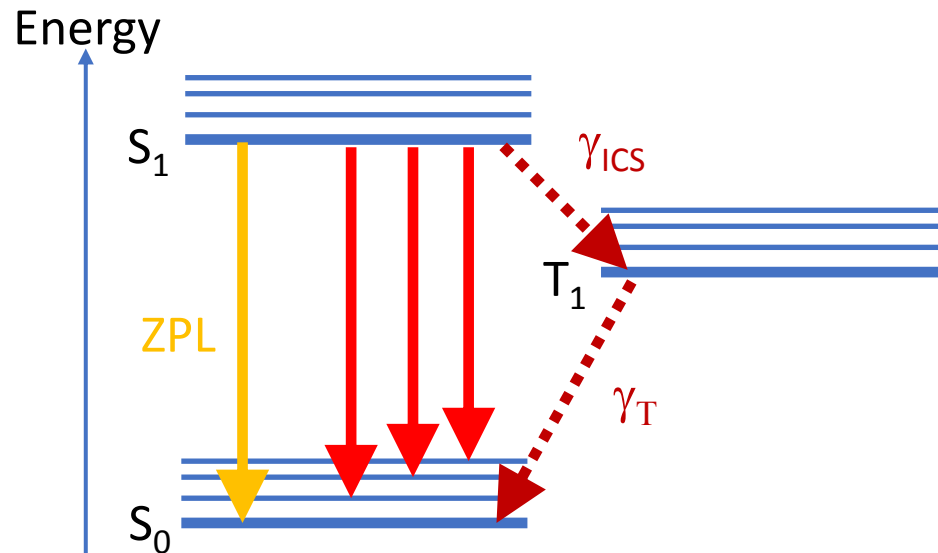


Anthracene

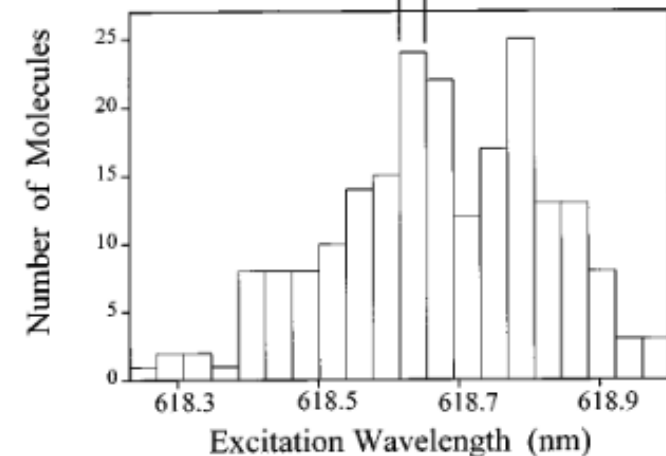
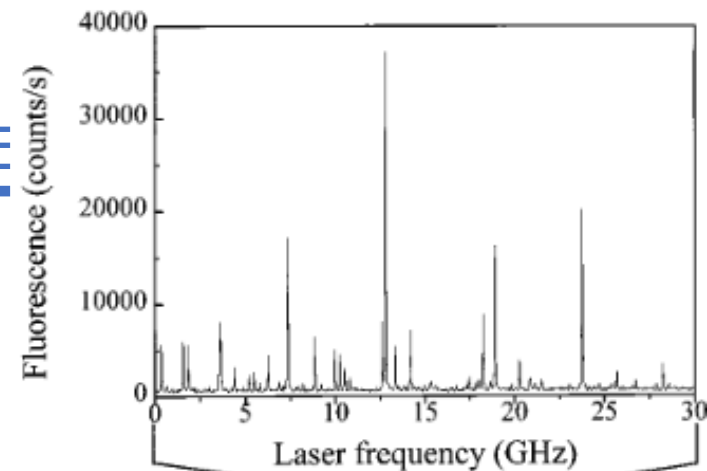
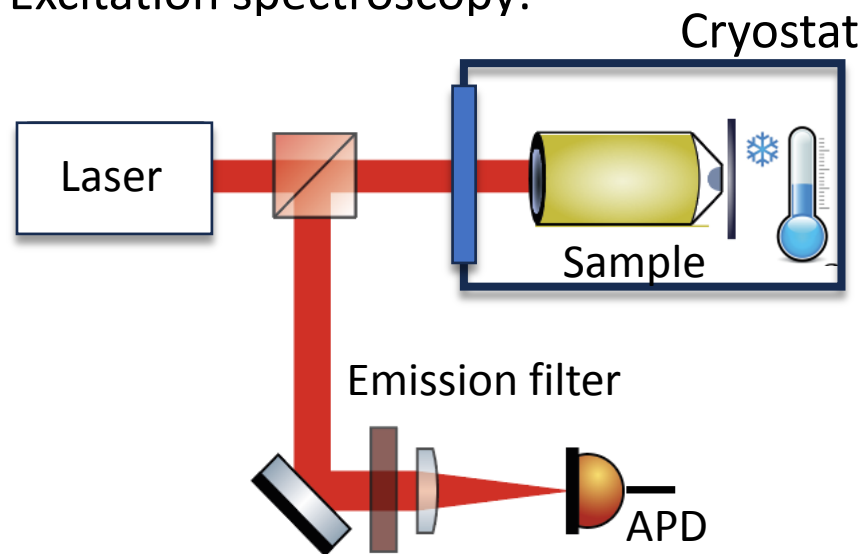


- High quality factors (10^8)
- No spectral diffusion

$$\sigma_{ZPL} \approx \frac{3\lambda^2}{2\pi} \approx 6 \cdot 10^{-10} \text{ cm}^2$$

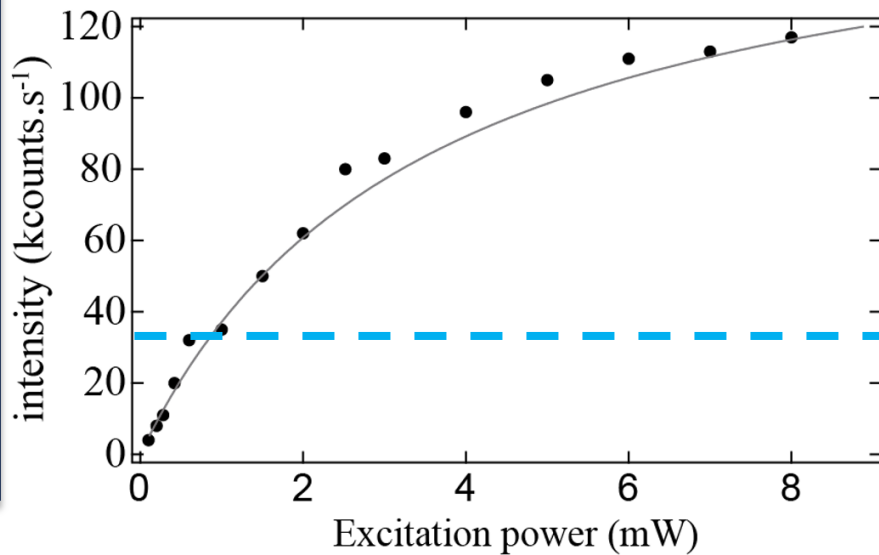
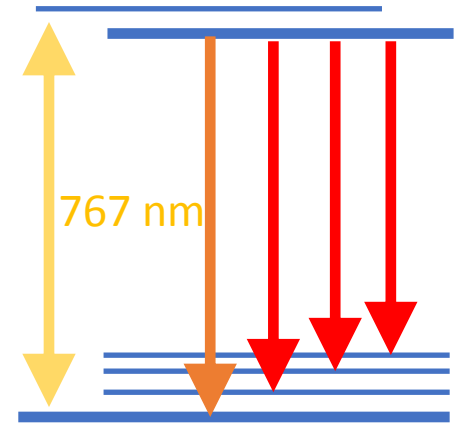
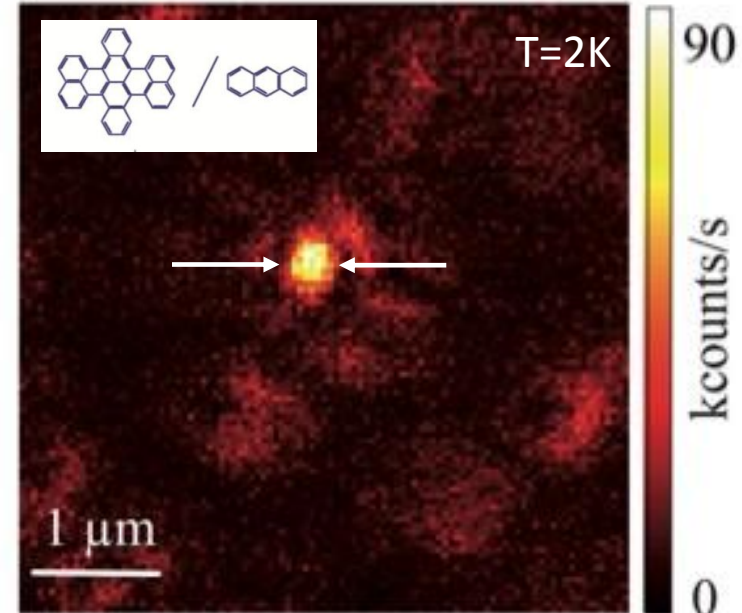
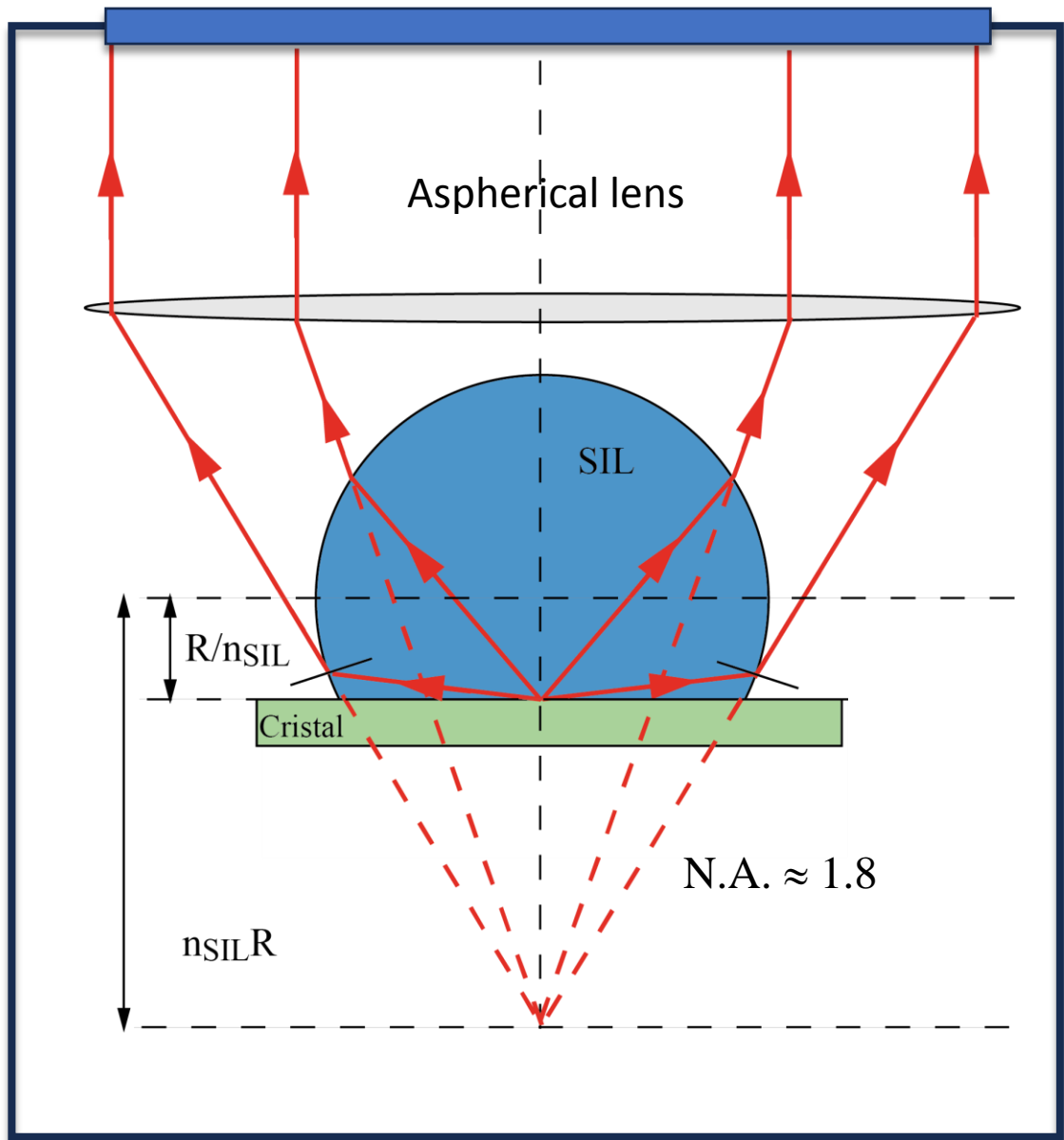


Excitation spectroscopy:



Tamarat *et al.*, 1999

Low-Temperature Imaging: Solid Immersion Lens (SIL)

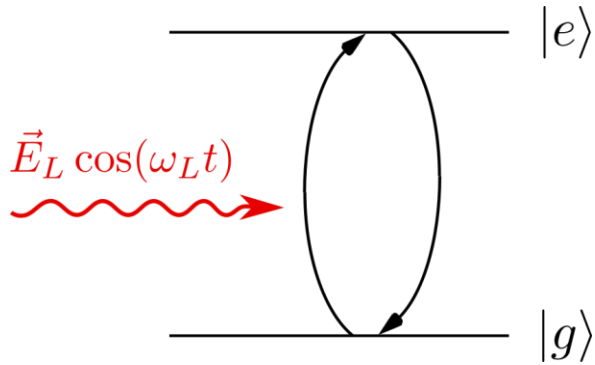


Optical resolution :
300 nm (0.4λ)

Increased collection
efficiency (x4)

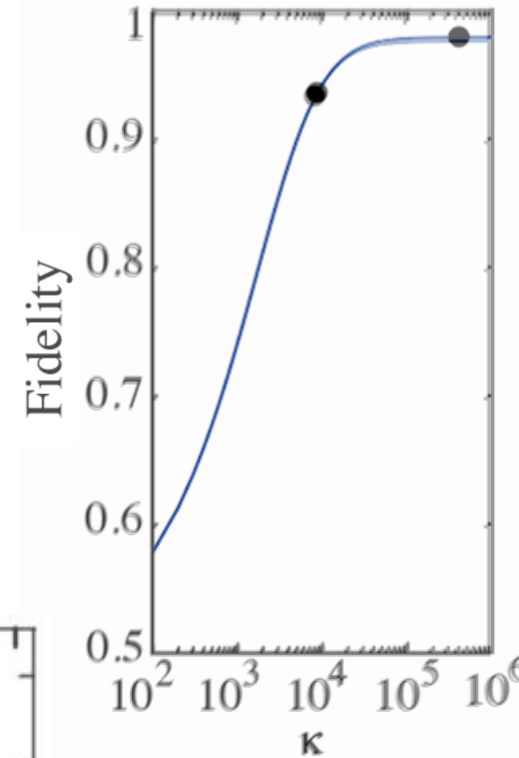
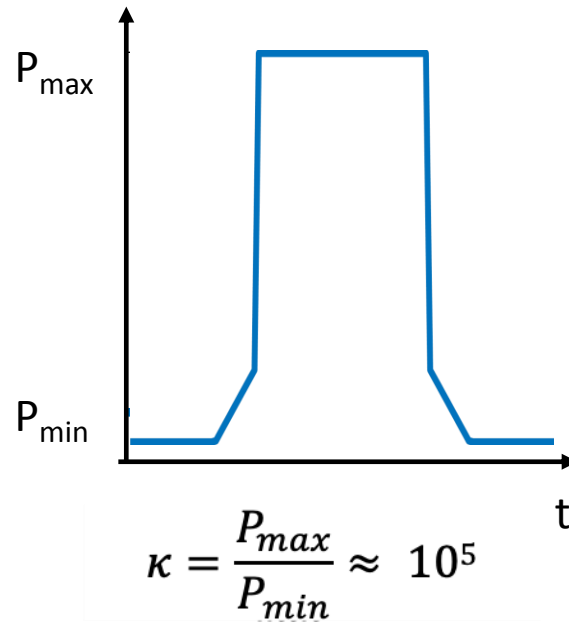
Diffraction-limited field
($35 \times 35 \mu\text{m}^2$)

Sub-nanosecond Rabi oscillations

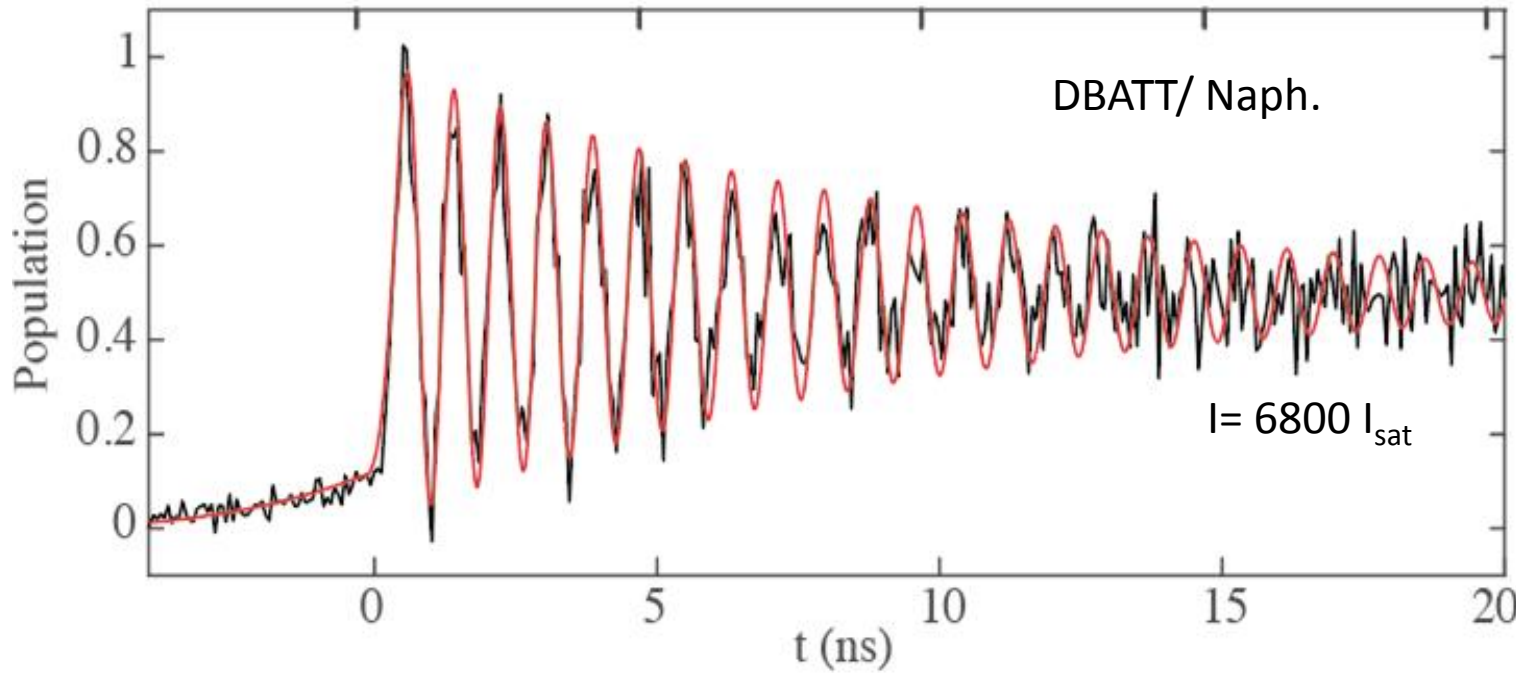


Rabi Frequency

$$\Omega = \frac{-\vec{d} \cdot \vec{E}_L}{\hbar}$$



- AOM + EOM
- Laser frequency locking
- Power control of pulses

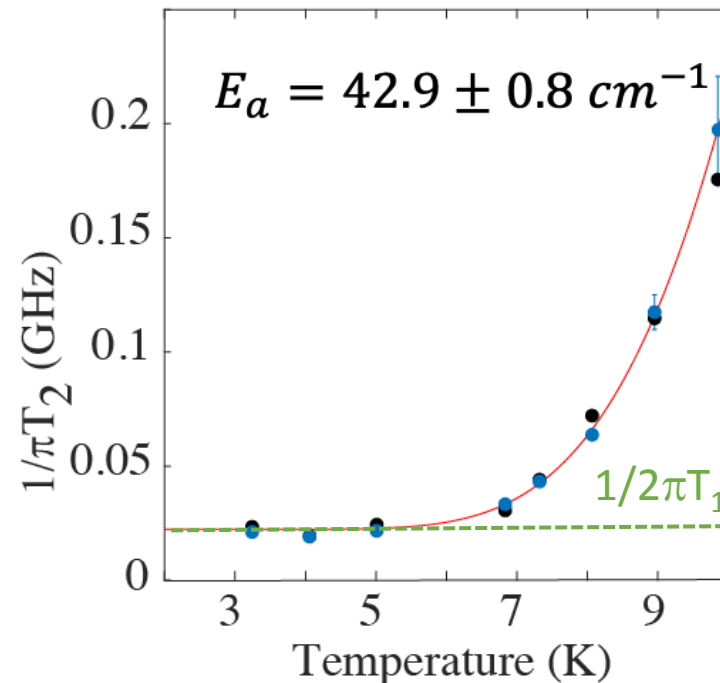
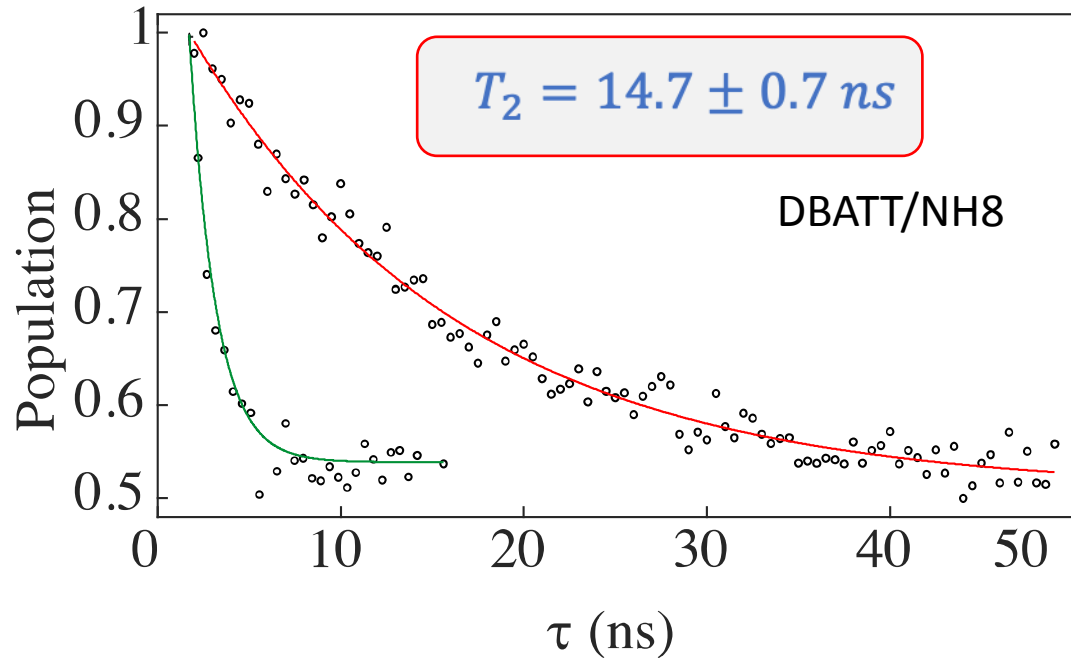
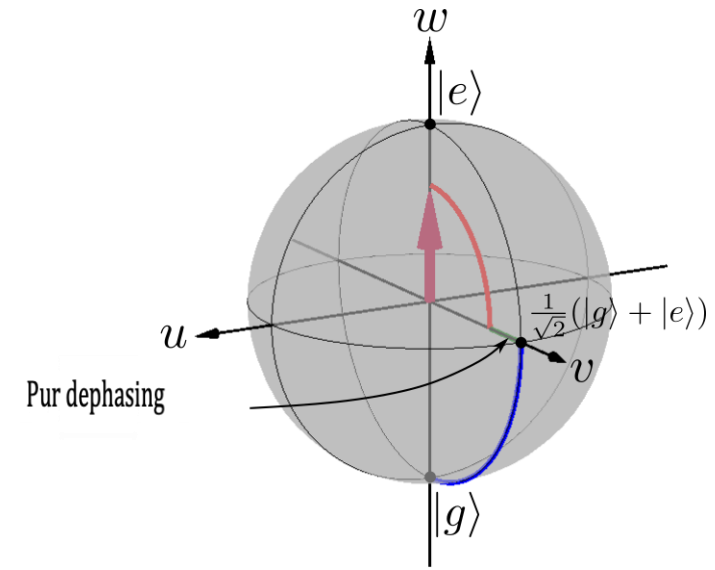
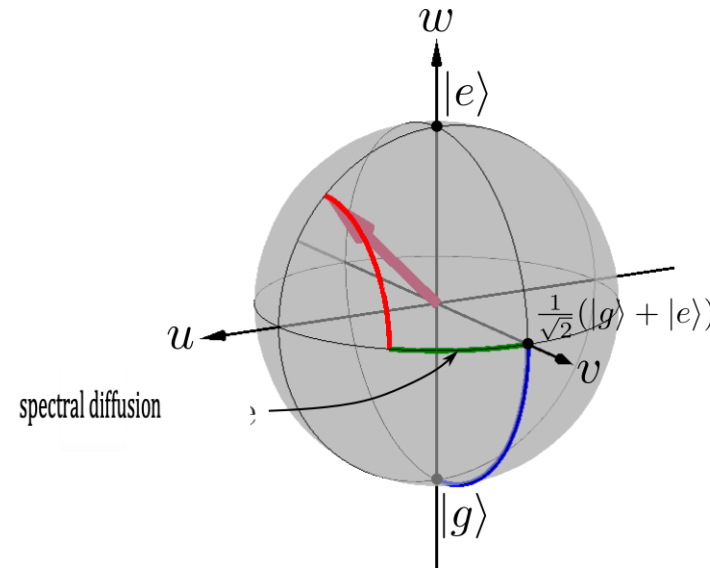
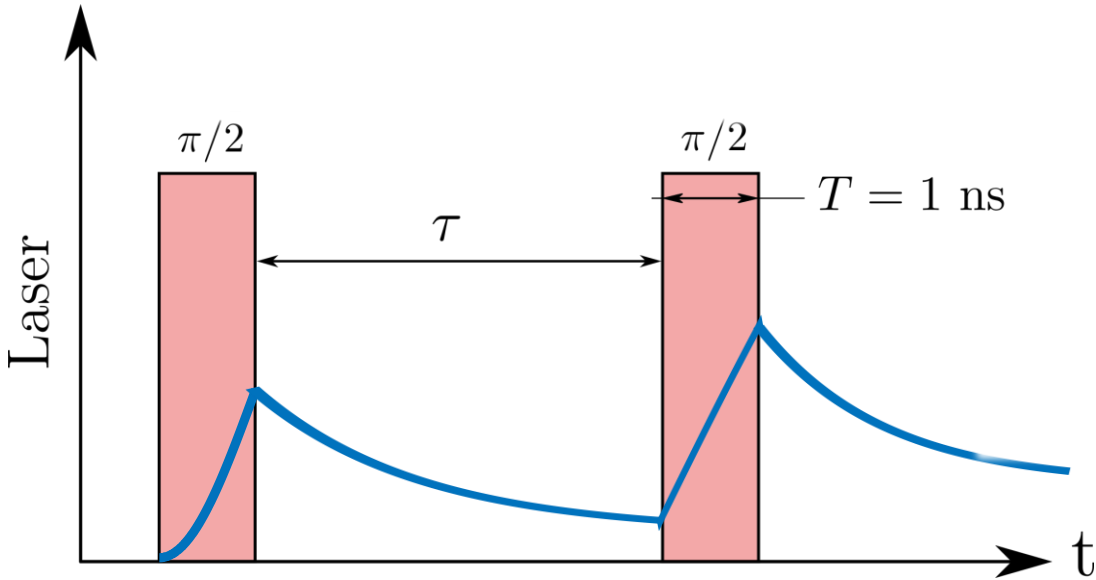


Rabi oscillations shorter than a nanosecond
 Period : **850 ps**

- Without any additional decoherence effect (**limited by $2T_1$**)
- Excited state preparation fidelity **$(97 \pm 3) \%$**

Deplano et al,
AVS Quantum Science (2023)

Ramsey spectroscopy and measurement of the optical coherence time



Arrhenius Law :

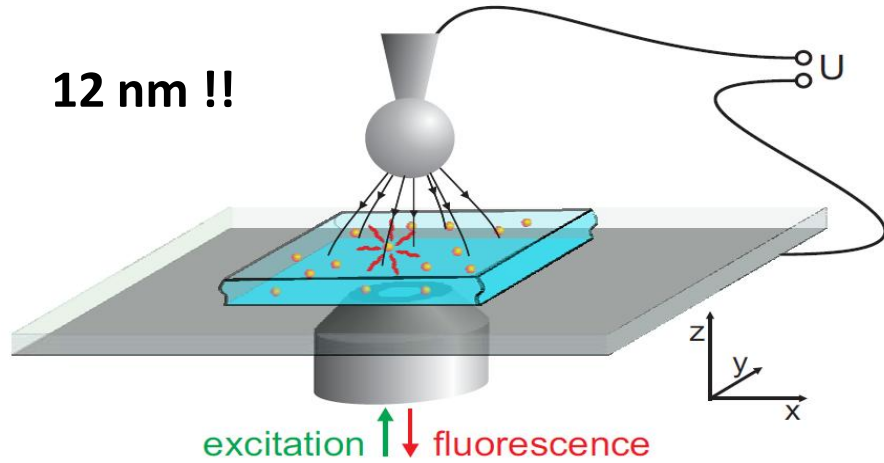
$$\gamma(T) = A \cdot e^{-\frac{E_a}{k_B T}} + \gamma_0$$

Good agreement
between T_2 and the
homogeneous linewidth

**Deplano et al.,
AVS Quantum Science (2023)**

From single to multiple interacting emitters

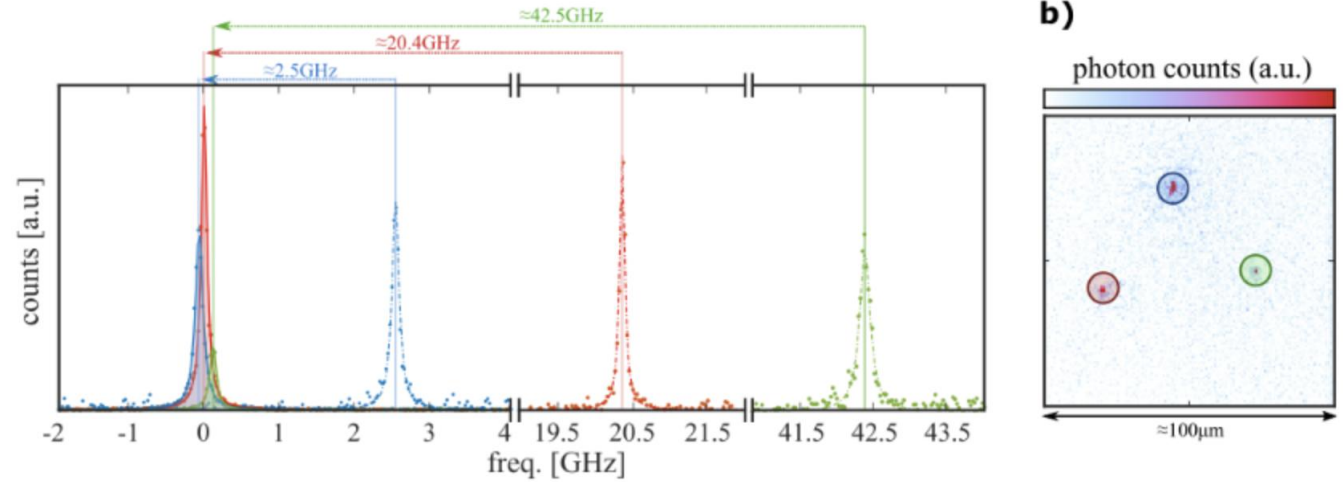
First observation of two coupled molecules



12 nm !!

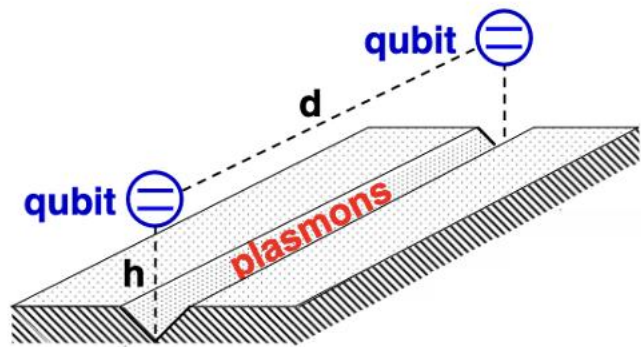
C. Hettich *et al.*, (2002)

Light-assisted resonance tuning



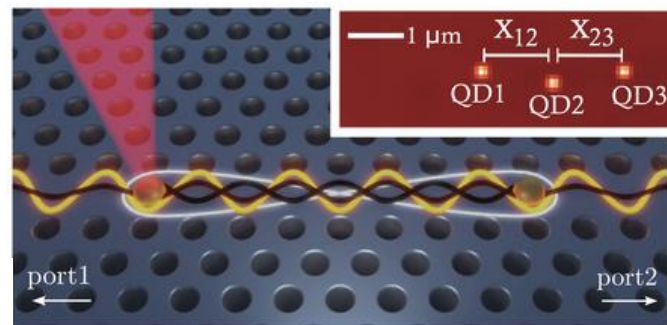
M. Colauti *et al.*, (2020)

Long-distance coupling via surface plasmons



A. Gonzalez-Tudela *et al.*, (2011)

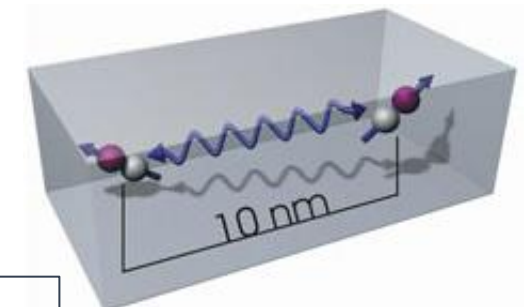
Coupled quantum dots



A. Tiranov *et al.*, (2023)

Optical nanoscopy :

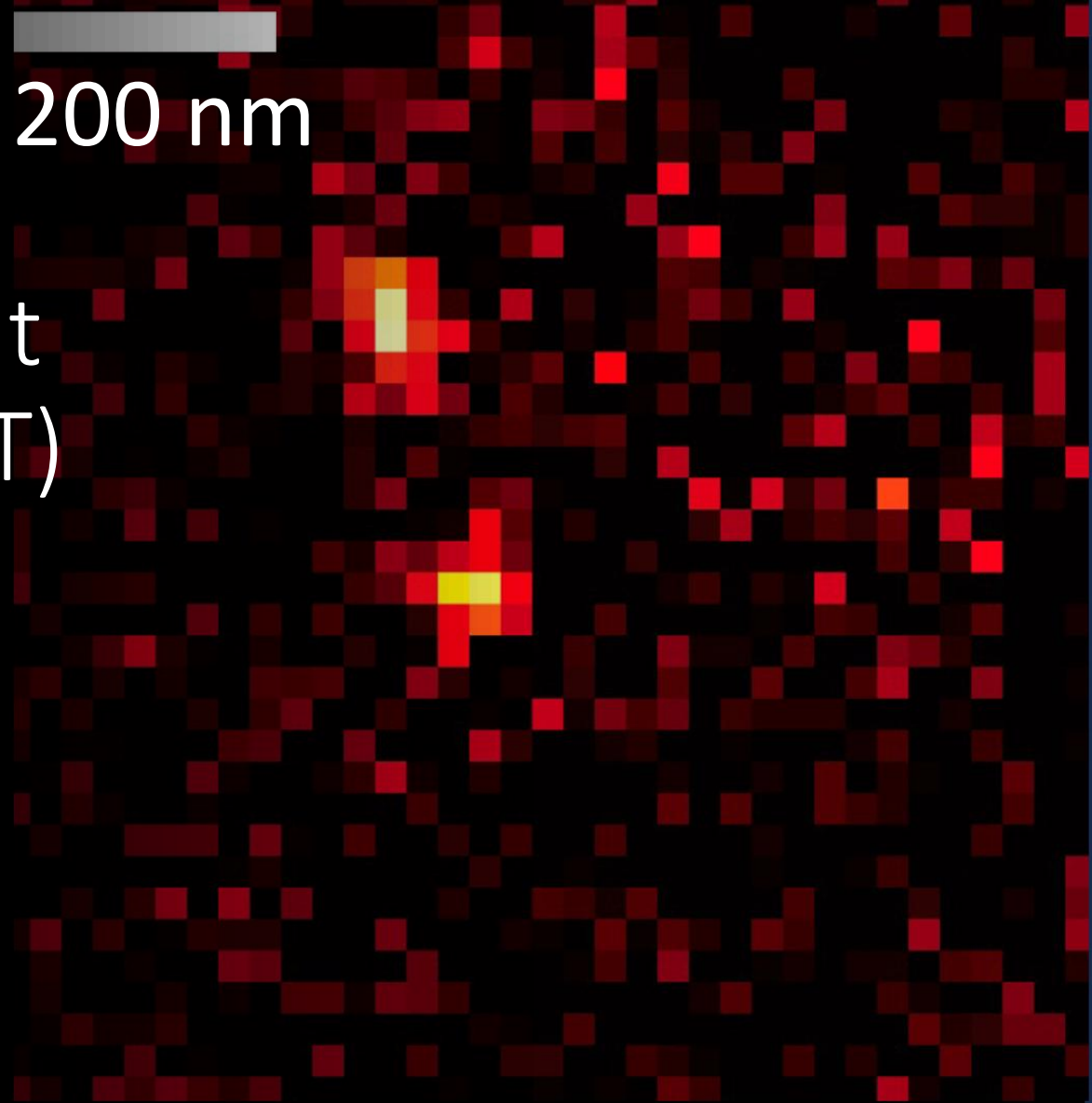
A challenging step towards the control of delocalized states formed by strongly and coherently interacting emitters



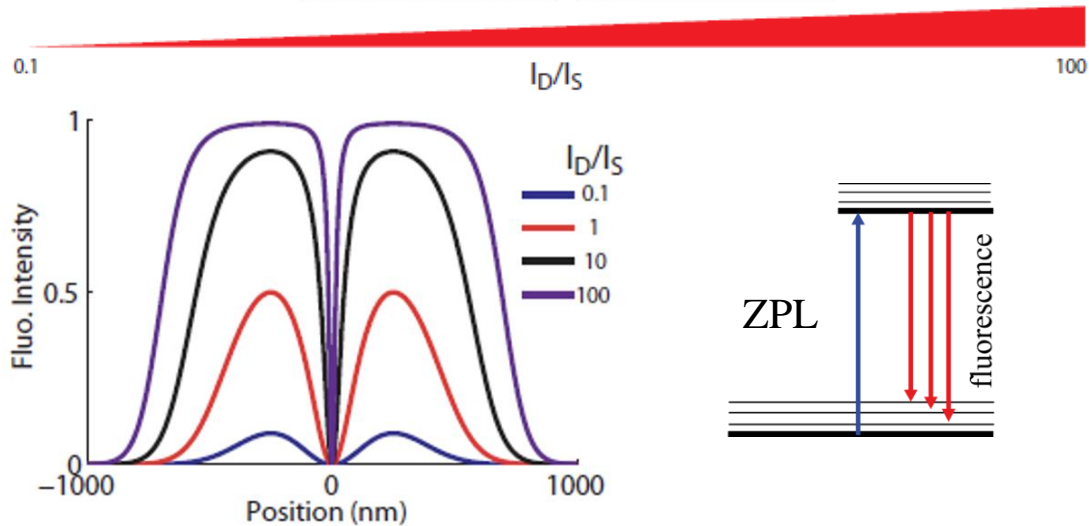
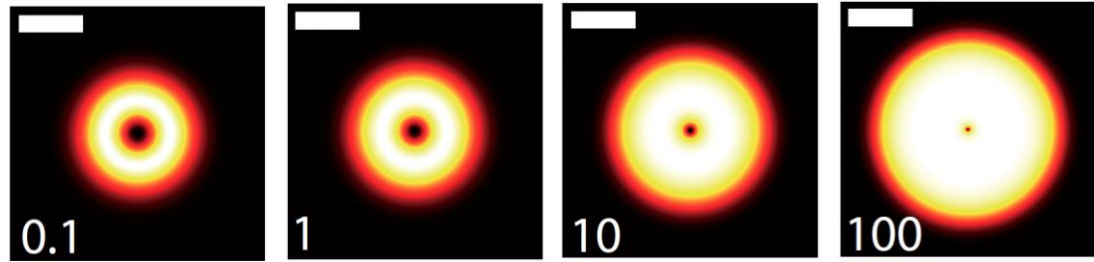
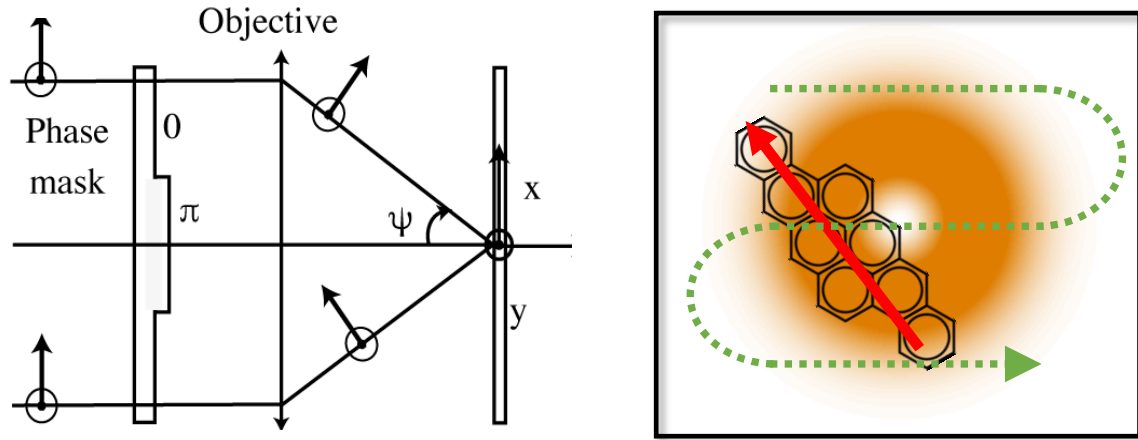
Need for an optical method to image interacting emitters

II . Super-resolution imaging at cryogenic temperatures (ESSAT)

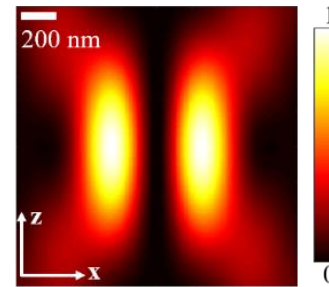
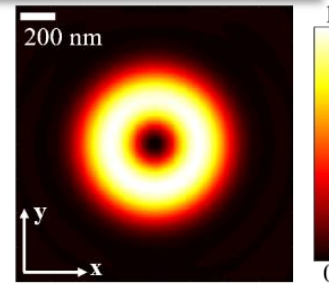
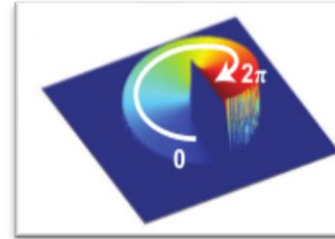
200 nm



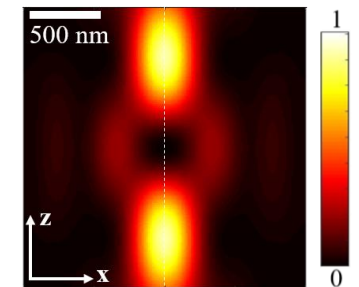
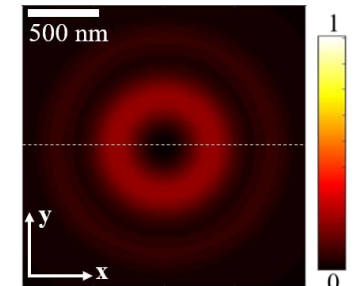
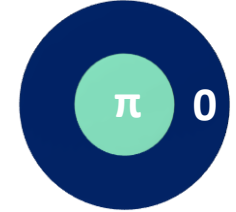
Excited State Saturation Nanoscopy (ESSat)



Helical phase plate :
2D Minimum
Unchanged axial resolution



0- π phase plate :
3D intensity gradient around the center

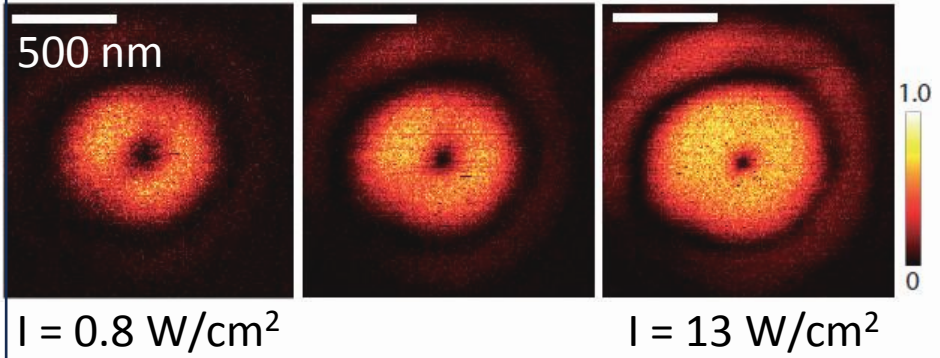


➔ Nanoscopic Essat 2D

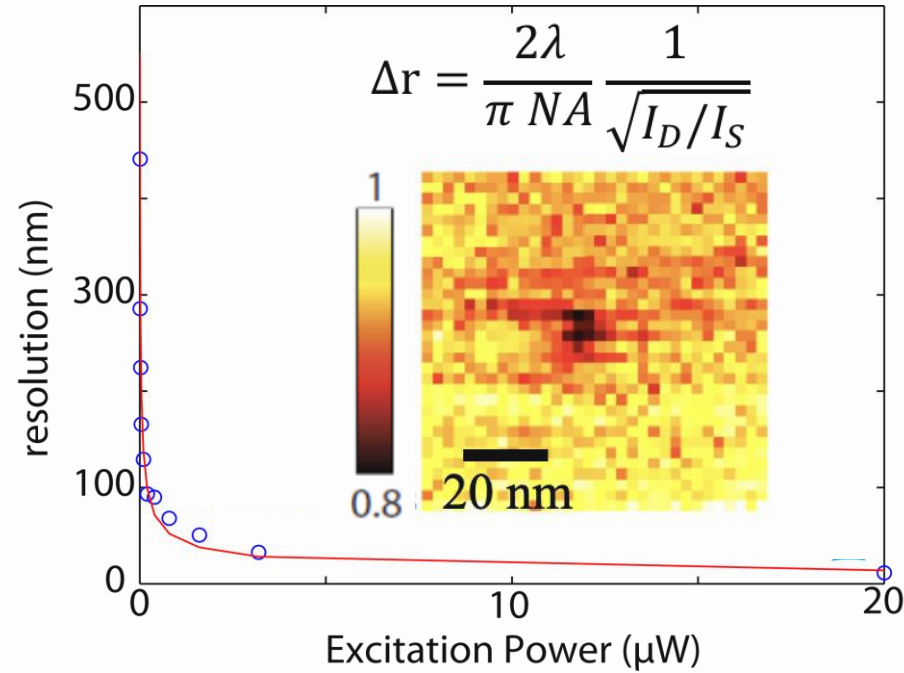
➔ Nanoscopic Essat 3D

Excited State Saturation Nanoscopy (ESSat)

2D :



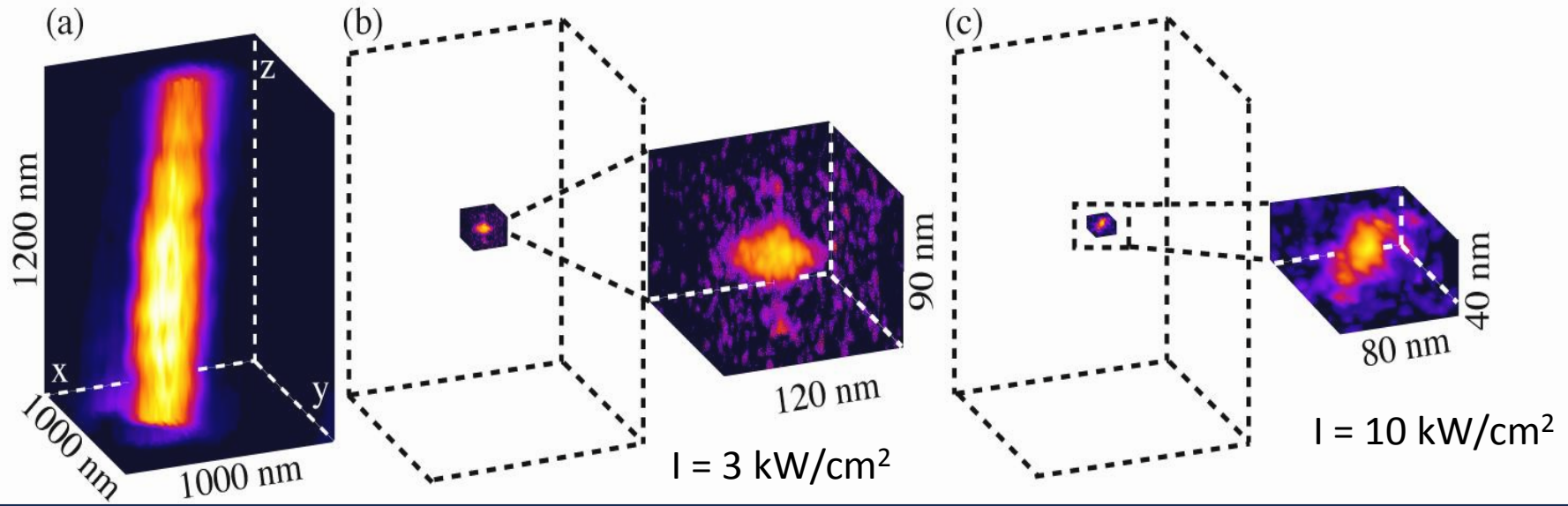
Aberration compensations :
Zernike polynomial



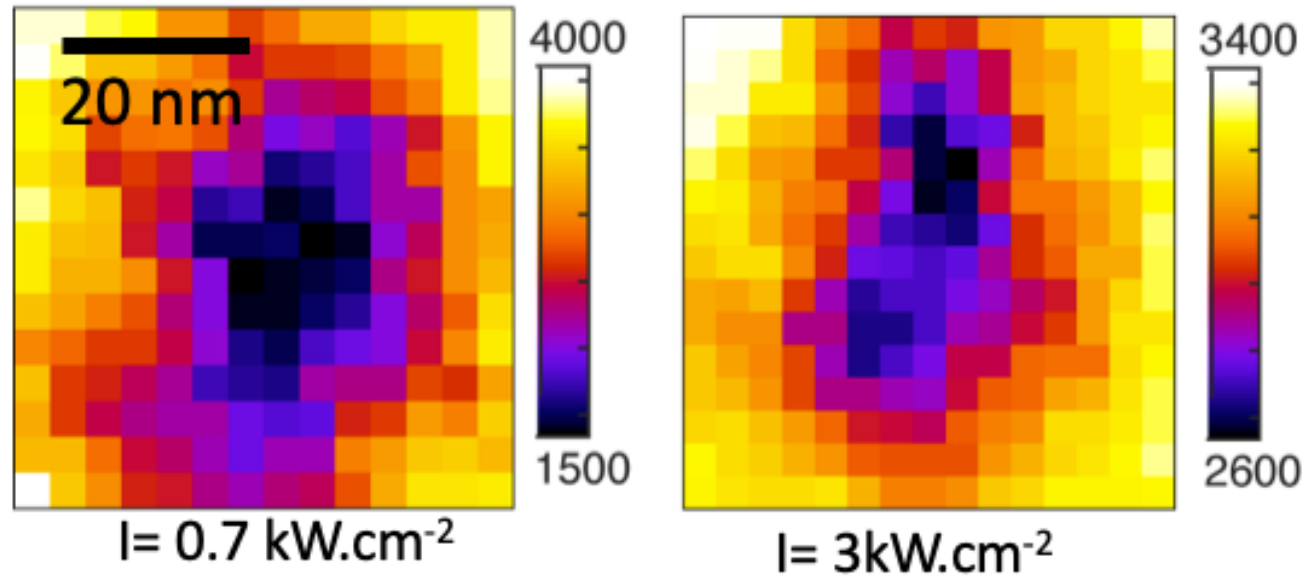
- Ultimate optical resolution :
 $\Delta r = 5 \text{ nm}$
- limited by mechanical vibrations

**Yang et al.,
Nature Photonics (2015)**

3D:



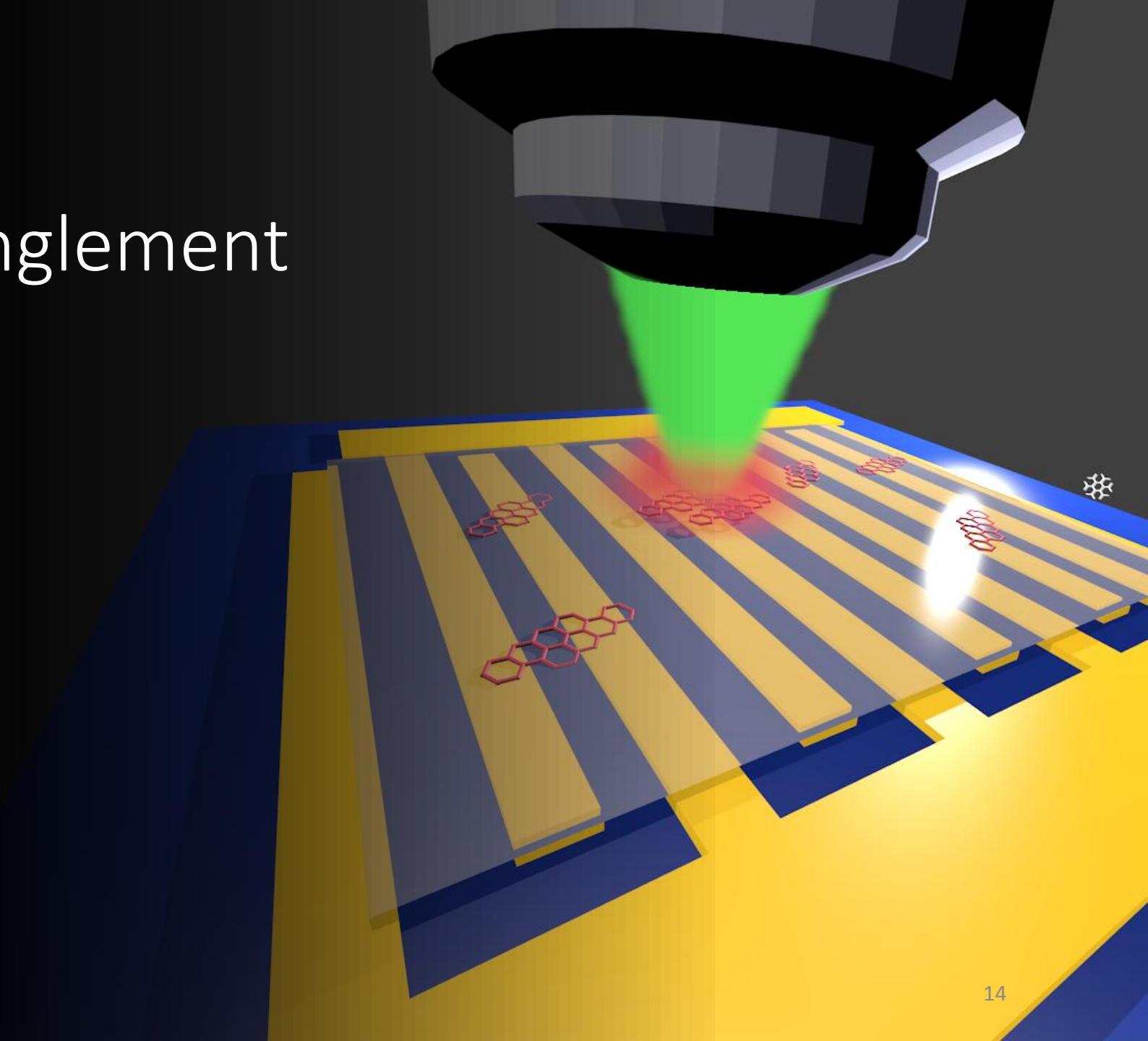
Finding pairs ?



**Coupled and/or entangled
molecular system?**

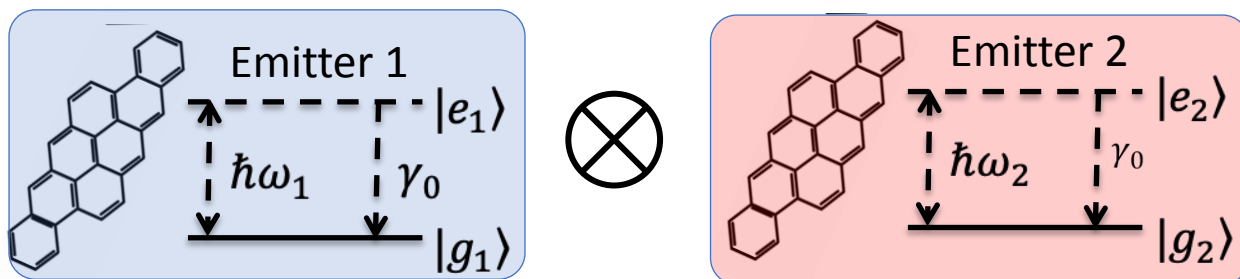


III. Molecular entanglement

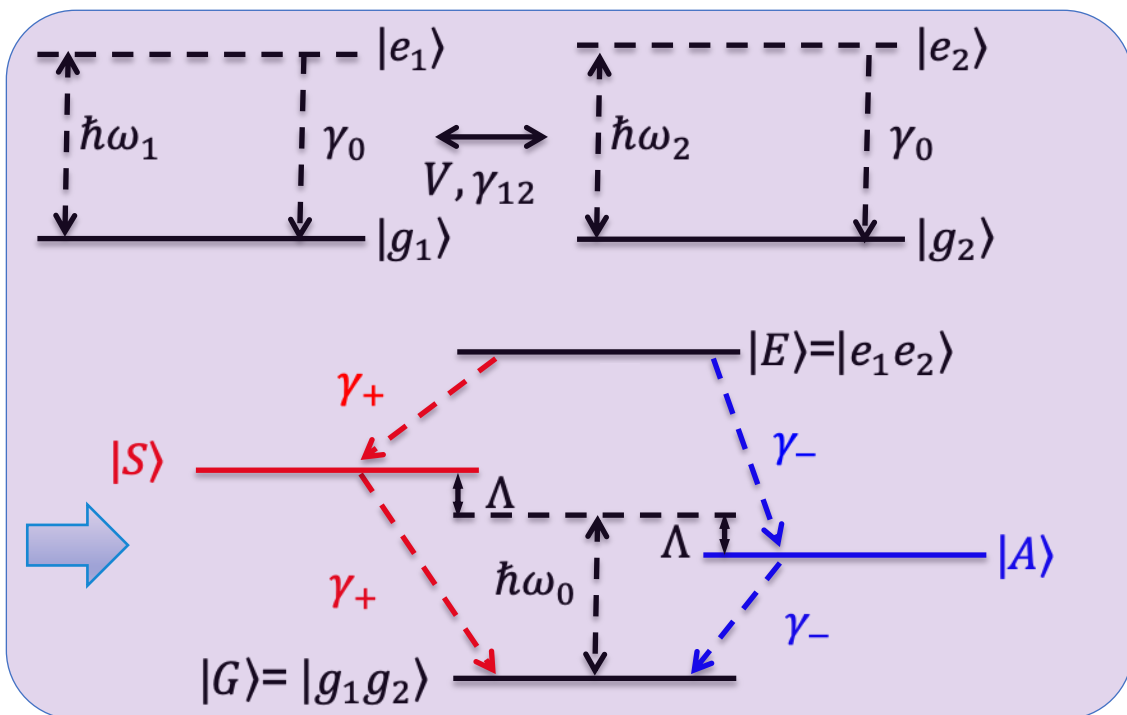


Coherent coupling: dipole-dipole interaction

Non interacting emitters :



interacting emitters :



$$H = \underbrace{H_0}_{\hbar\omega_1 S_1^+ S_1^- + \hbar\omega_2 S_2^+ S_2^-} + \underbrace{H_{dd}}_{\hbar V S_1^- S_2^+ + h.c.}$$

Resonant dipole dipole coupling ($d \ll \lambda$) :

$$V = \frac{3\hbar\gamma_0}{4(kd)^3} [(\hat{\mu}_1 \cdot \hat{\mu}_2) - 3(\hat{\mu}_1 \cdot \hat{e}_{12})(\hat{\mu}_2 \cdot \hat{e}_{12})]$$

2 entangled states :

Symmetric & Antisymmetric

$$\begin{cases} |S\rangle = \alpha|e_1, g_2\rangle + \beta|g_1, e_2\rangle \\ |A\rangle = \beta|e_1, g_2\rangle - \alpha|g_1, e_2\rangle \end{cases} \Rightarrow \text{Maximal entanglement}$$

For $V \neq 0$
and $\delta = \omega_2 - \omega_1 \rightarrow 0$

Cross damping rate :

$$\gamma_{12} = \gamma_0 (\hat{\mu}_1 \cdot \hat{\mu}_2)$$

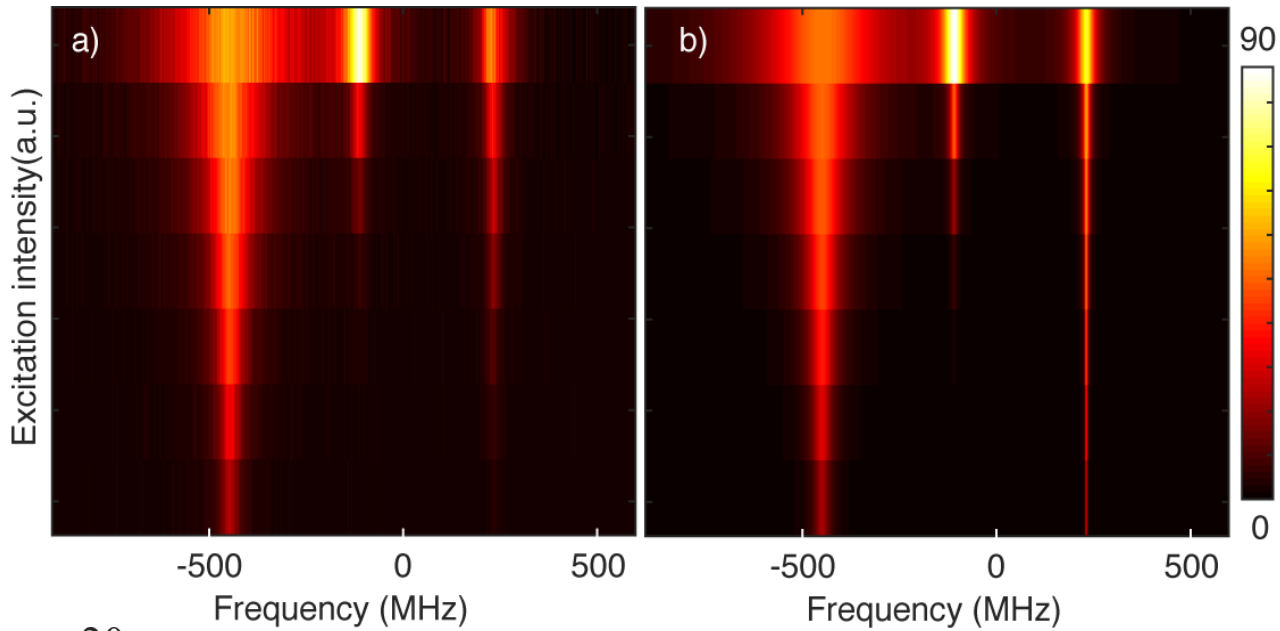
For Bell states and $\mu_1 // \mu_2$

$$\gamma_+ \rightarrow 2\gamma_0, \gamma_- \rightarrow 0$$

Signatures of the dipole dipole interaction

Experimental

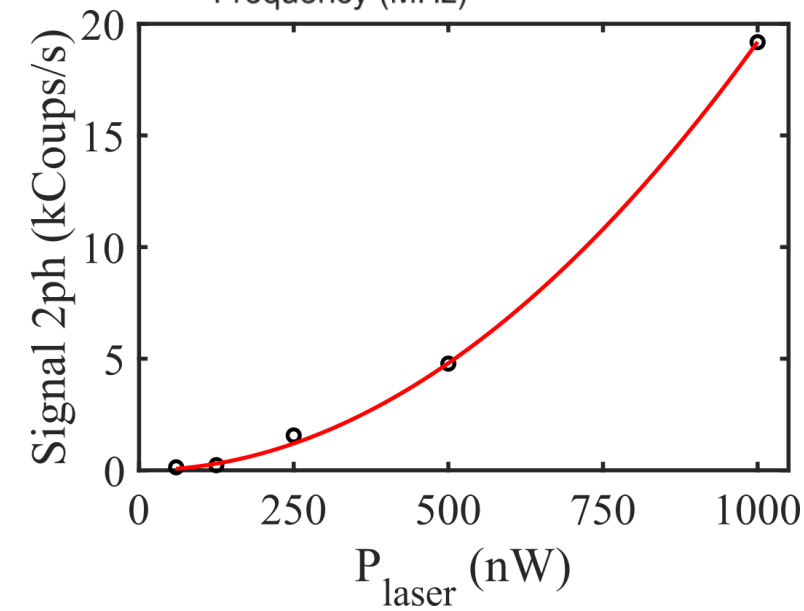
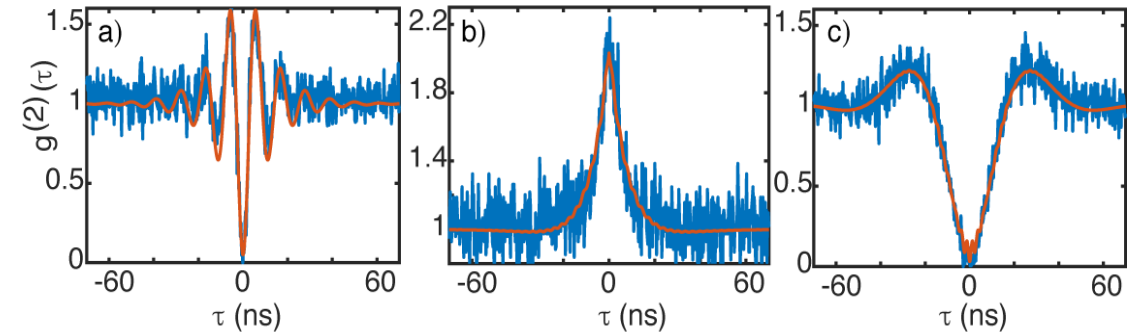
Fit



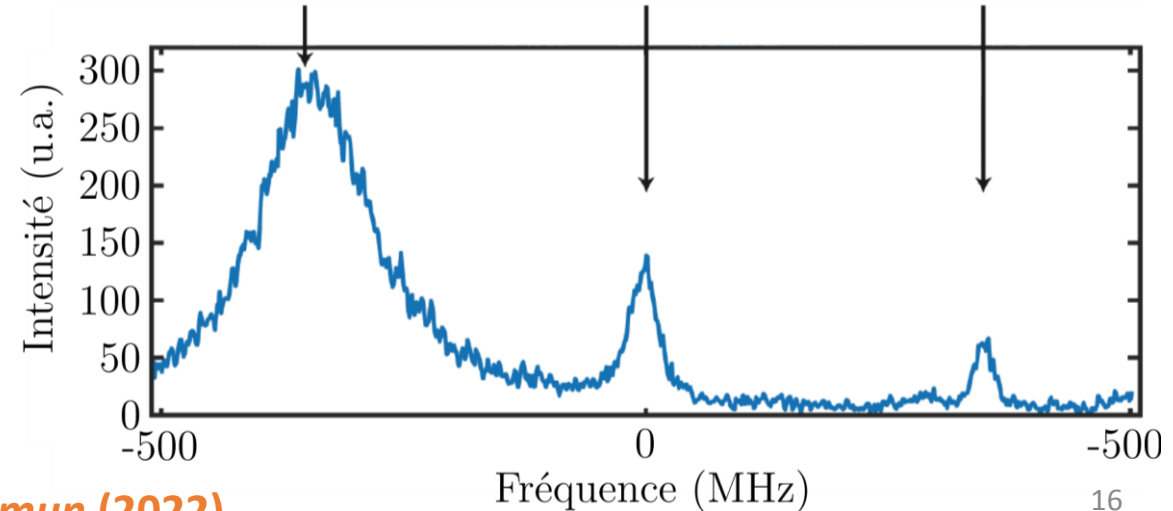
Excitation spectra calculated from the master equation :

$$\dot{\rho} = -\frac{i}{\hbar} [H', \rho] - \frac{1}{2} \sum_{i,j} \gamma_{ij} (S_i^+ S_j^- \rho + \rho S_i^+ S_j^- - 2S_j^- \rho S_i^+)$$

Second order correlation measurements
(red shifted photons) :



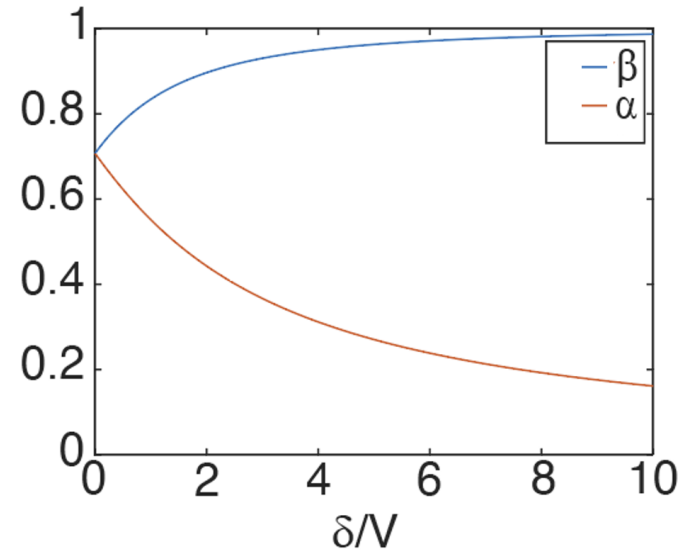
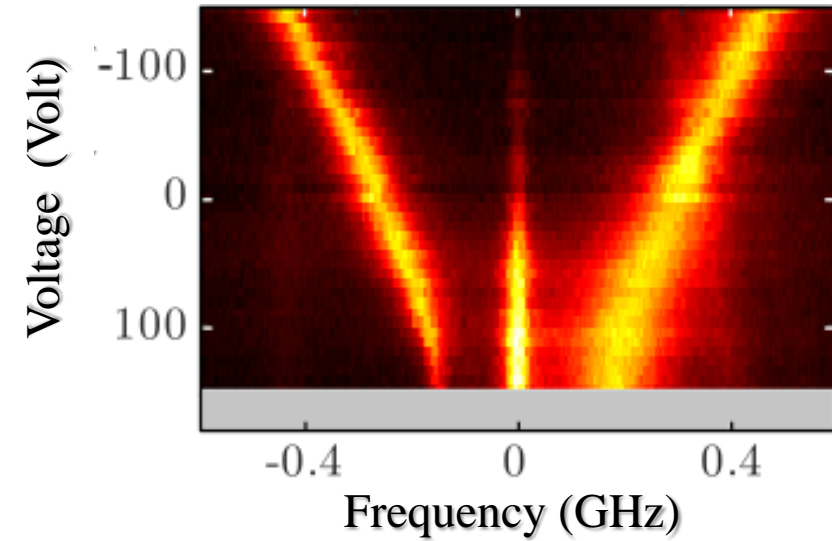
$V = -17 \gamma_0,$
 $\delta = 10 \gamma_0,$
 $\gamma_{12} = 0.3 \gamma_0$



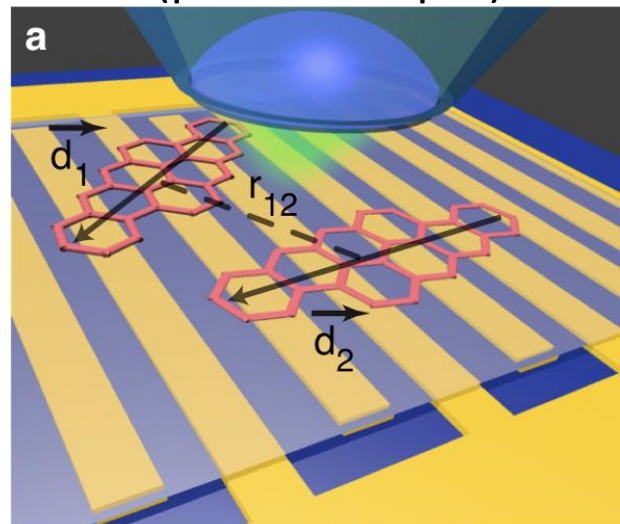
Trebbia et al., Nat Commun (2022)

Entanglement Manipulation

Nat Commun 13, 2962 (2022)



Interdigitated electrodes
(period : 20 μm)



➤ Electric field \approx MV/m

Quasi-separable states :

$$|S\rangle = \frac{0.43}{\sqrt{2}} |e_1, g_2\rangle + \frac{1.34}{\sqrt{2}} |g_1, e_2\rangle$$

$$|A\rangle = \frac{1.34}{\sqrt{2}} |e_1, g_2\rangle - \frac{0.43}{\sqrt{2}} |g_1, e_2\rangle$$



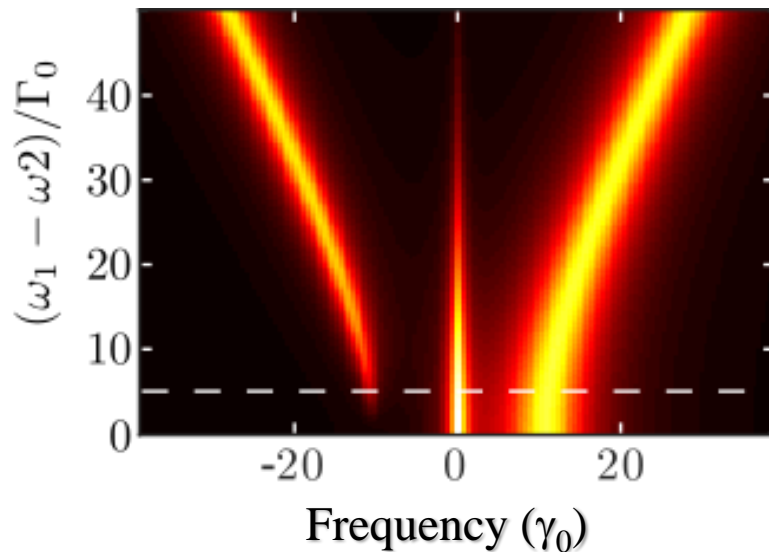
quasi Bell States :

$$|S\rangle = \frac{0.86}{\sqrt{2}} |e_1, g_2\rangle + \frac{1.12}{\sqrt{2}} |g_1, e_2\rangle$$

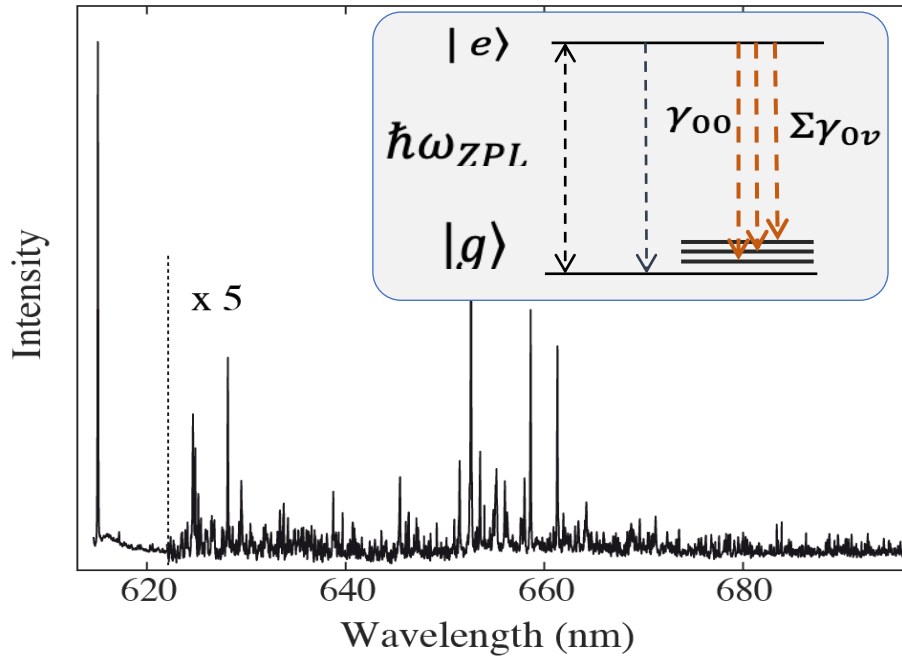
$$|A\rangle = \frac{1.12}{\sqrt{2}} |e_1, g_2\rangle - \frac{0.86}{\sqrt{2}} |g_1, e_2\rangle$$

$V/h \sim 10\gamma_0$,

Estimated distance : $d \sim 25\text{nm}$ 17



Demonstration of sub/super radiance effects for two coupled emitters

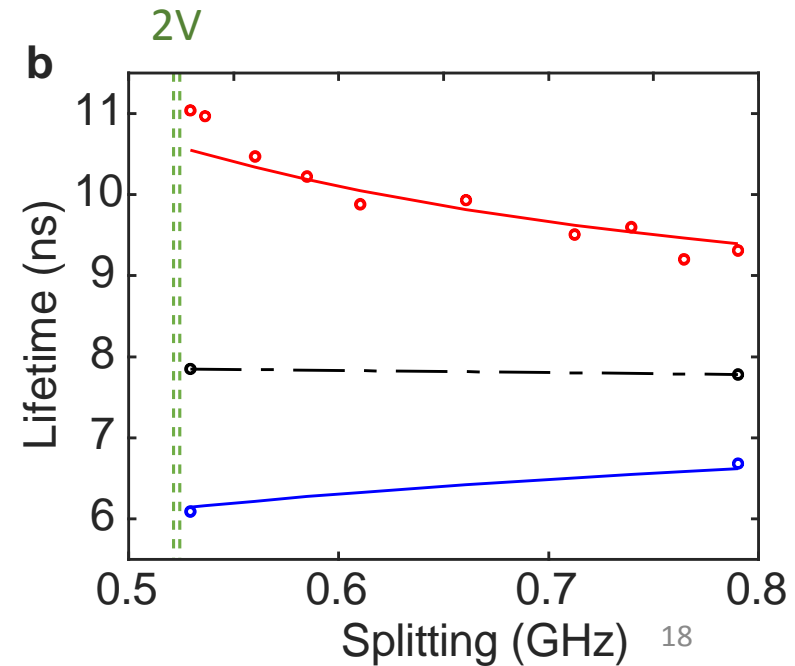
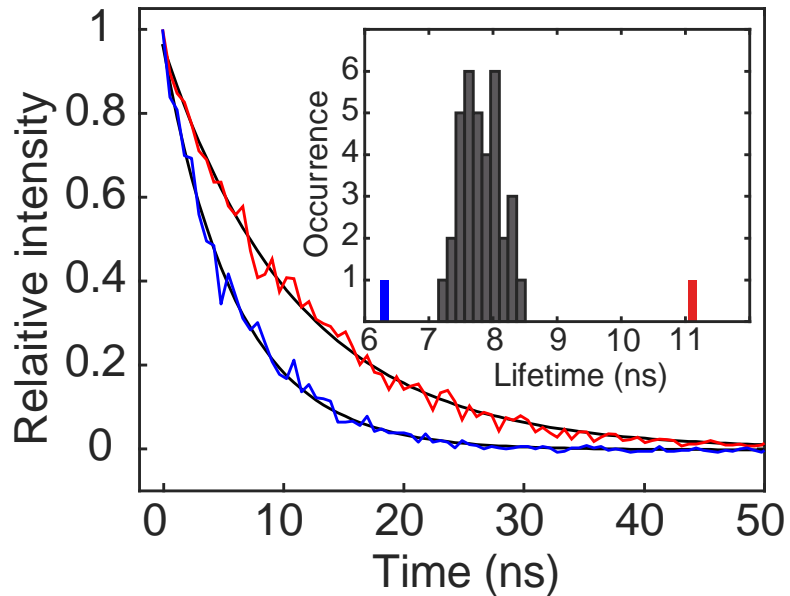


$$\alpha_{DW/FC} = 0.3$$

$$\gamma_S \rightarrow \gamma_0(1 + \alpha_{DW/FC})$$

$$\gamma_A \rightarrow \gamma_0(1 - \alpha_{DW/FC})$$

(for $\omega_1 = \omega_2$ and $\mu_1 // \mu_2$)



Coupling of entangled states with the laser field

Hamiltonian of the system excited by a laser of frequency ω_L

$$H = \hbar\omega_0(|E\rangle\langle E| - |G\rangle\langle G|) + \hbar\Lambda(|S\rangle\langle S| - |A\rangle\langle A|) - \frac{\hbar}{2} \left\{ [(\alpha\Omega_1 + \beta\Omega_2)|E\rangle\langle S| + (\beta\Omega_1 + \alpha\Omega_2)|S\rangle\langle G|] e^{i\omega_L t} + [(\alpha\Omega_2 - \beta\Omega_1)|E\rangle\langle A| - (\beta\Omega_2 - \alpha\Omega_1)|A\rangle\langle G|] e^{i\omega_L t} + h.c. \right\}$$

Complex Rabi frequencies :

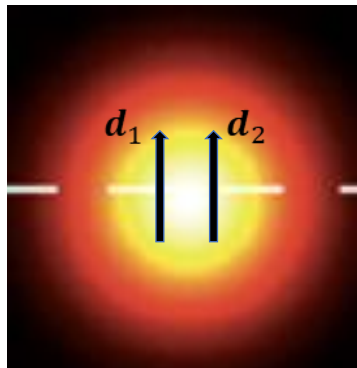
$$\Omega_i = -\vec{\mu}_i \cdot \vec{E}_L(\vec{r}_i - \vec{r}_c) / \hbar$$

Matrix elements describing laser coupling

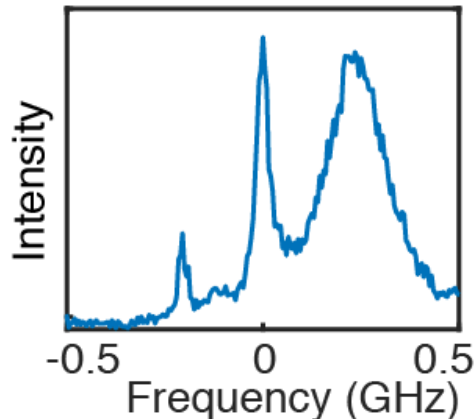
for $\alpha \approx \beta \approx 1/\sqrt{2}$:

$$\begin{cases} \langle A|H'|G \rangle \propto \Omega_1 - \Omega_2 \\ \langle S|H'|G \rangle \propto \Omega_1 + \Omega_2 \end{cases}$$

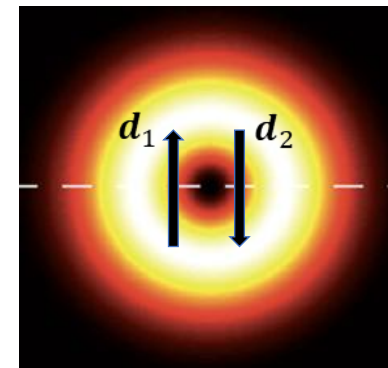
TEM_{00}



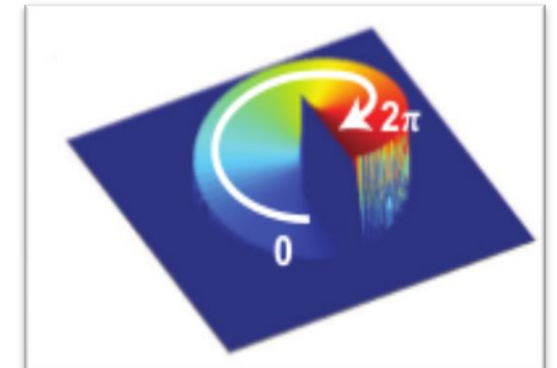
$$\Omega_1 = \Omega_2$$



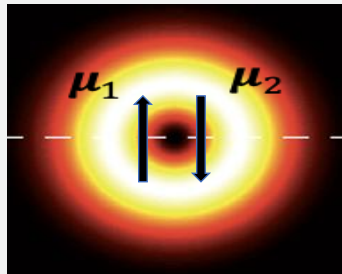
LG_{01}



$$\Omega_1 = -\Omega_2$$

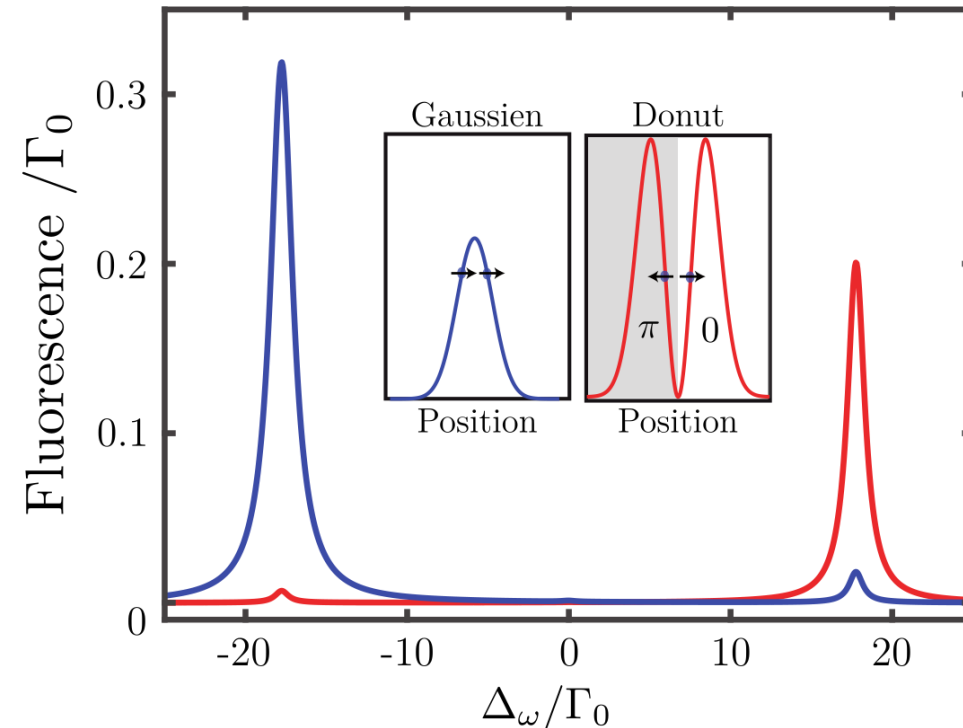
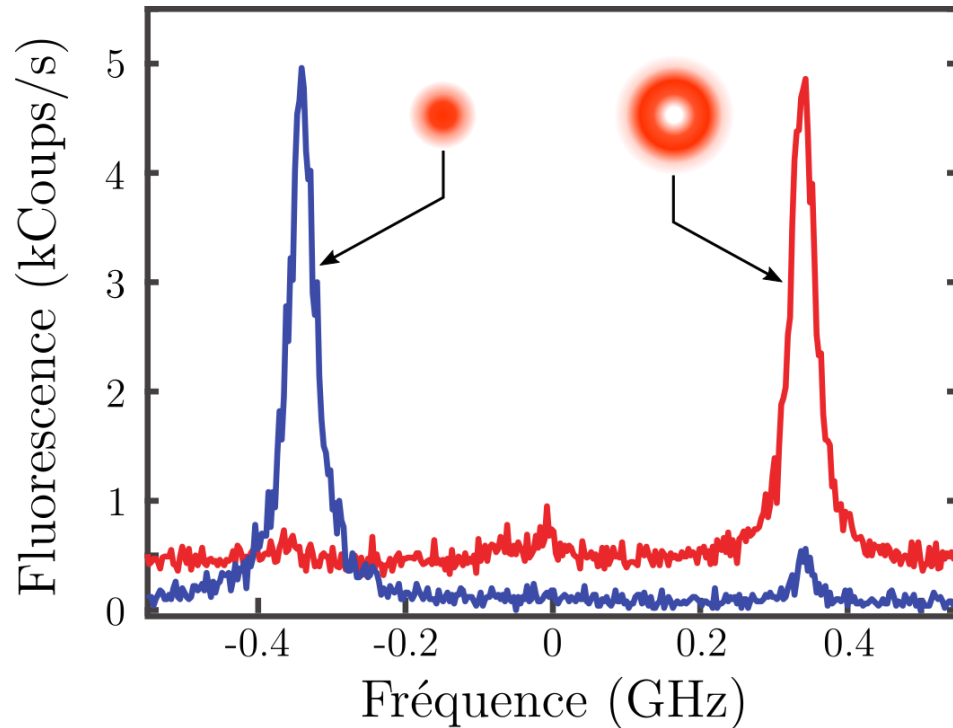
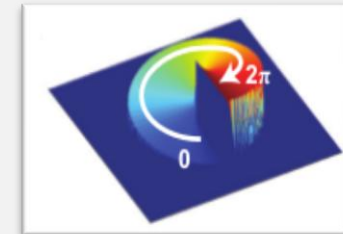


Optical preparation of the two entangled states



Donut centered between the molecules :

- Dipole oscillations in opposite phase $\Omega_1 = -\Omega_2$.
- Excitation of the subradiant state is enhanced, while that of the superradiant state is forbidden.

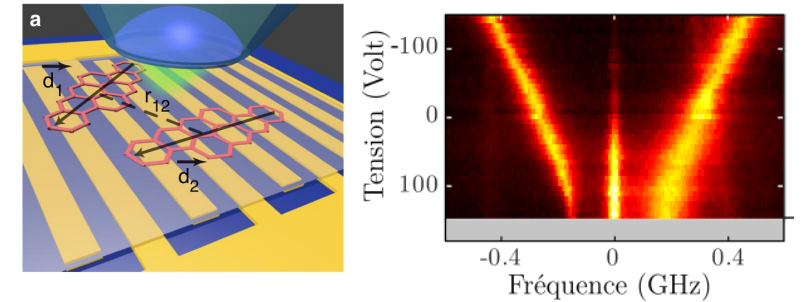
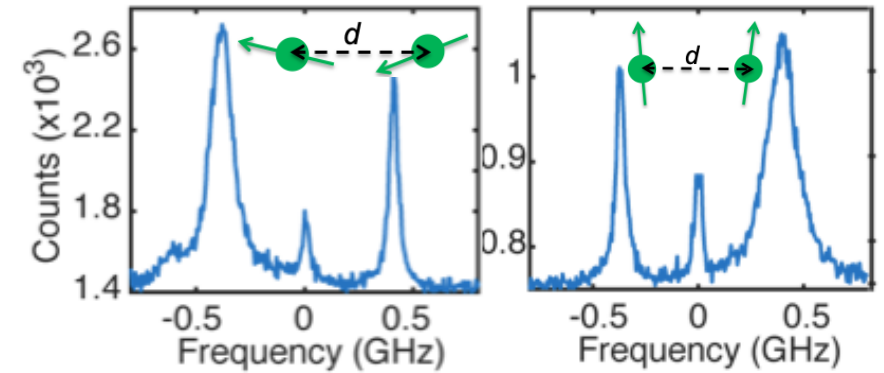


Method for preparing the long-lived subradiant state

Nat Commun **13**, 2962 (2022)

Conclusions

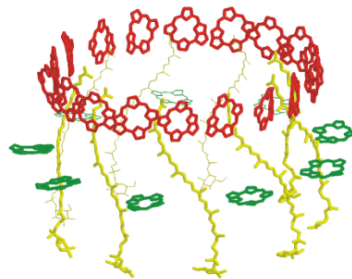
- Ultra-stable quantum systems: excitation lasting for several weeks without photobleaching or spectral diffusion
- Sub-nanosecond optical manipulation and coherence time measurement
- Control of the degree of entanglement via an electric field
- Selective addressing of entangled states using excitation beam shaping



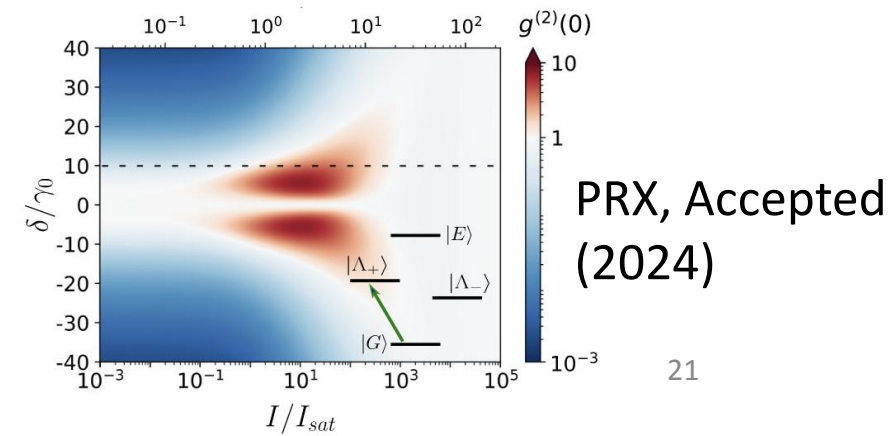
Perspectives:

Quantum logic gates

Decoherence effects with the temperature



Tailoring the statistics of light



PRX, Accepted (2024)

Acknowledgments

Phd Students :



B. Yang



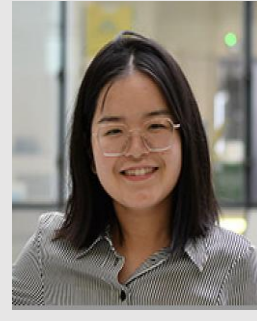
R. Baby



A. Jose



Q. Deplano

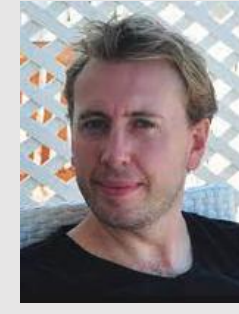


X. Zhou

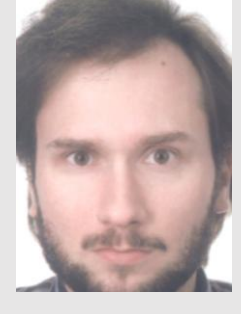
Post-Doctoral researchers :



F. Przybilla



M. Mestres



I. Veshchunov

Faculty members (Nanophotonics group) :



Prof
Philippe
Tamarat



Prof
Brahim
Lounis

Collaborators :

Gregory Giannone (IINS)
Dominique Mailly (C2N)
Javier Aizpurua (DIPC)

Thank you for your attention

Available Postdoctoral and PhD Positions, Please Contact Us!