

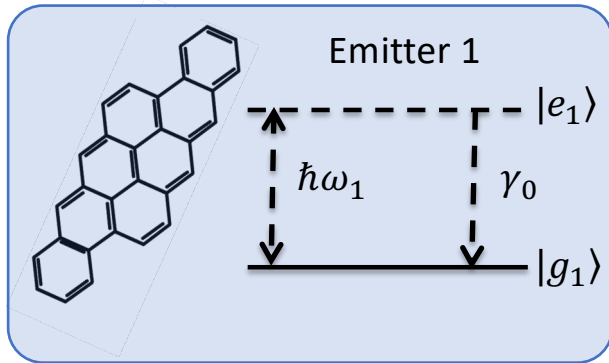
# Tailoring the superradiant and subradiant nature of two coherently coupled quantum emitters

Jean-Baptiste Trebbia

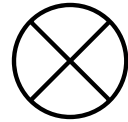
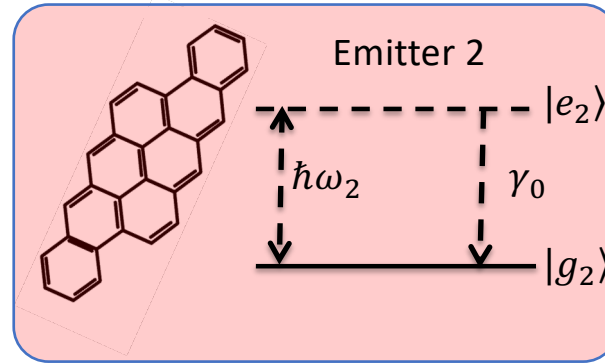
Laboratoire Photonique, Numérique et Nanosciences - LP2N  
Université Bordeaux - IOGS - CNRS : UMR 5298

# Entanglement

Hilbert space 1



Hilbert space 2



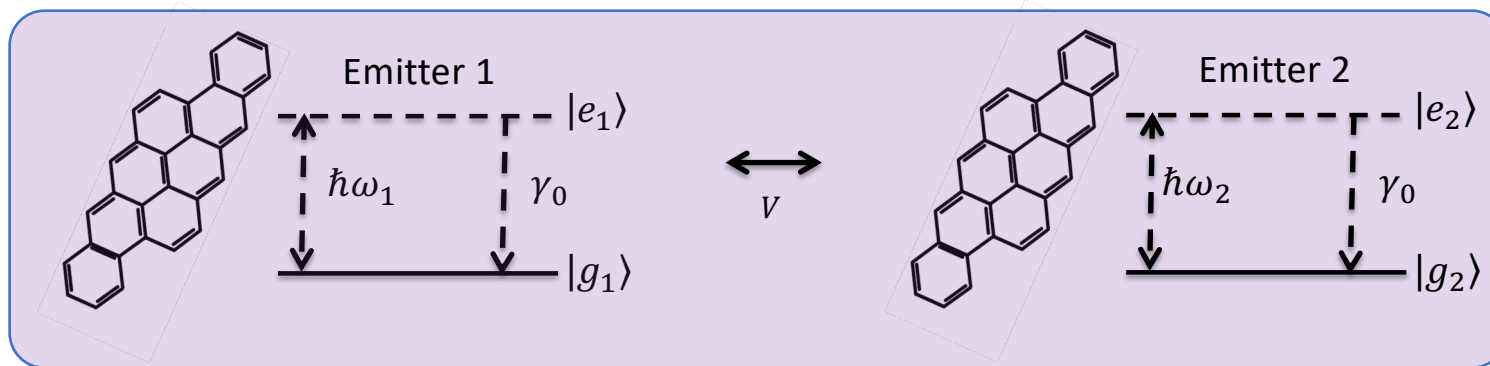
**Eigen States :**

- $|e_1\rangle|e_2\rangle$
- $|e_1\rangle|g_2\rangle$
- $|g_1\rangle|e_2\rangle$
- $|g_1\rangle|g_2\rangle$

**Four separable states**

**Coherently coupled emitters :**

Hilbert space



**Eigen States :**

- $|e_1\rangle|e_2\rangle$
- $1/\sqrt{2}(|e_1\rangle|g_2\rangle + |g_1\rangle|e_2\rangle)$
- $1/\sqrt{2}(|e_1\rangle|g_2\rangle - |g_1\rangle|e_2\rangle)$
- $|g_1\rangle|g_2\rangle$

**Two entangled states**

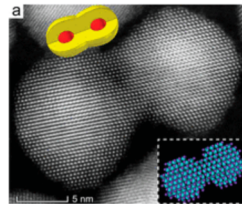
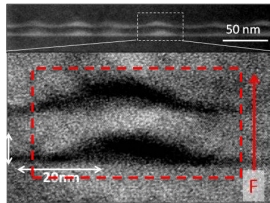
# Motivations

Controlled, coherent manipulation of delocalized quantum systems

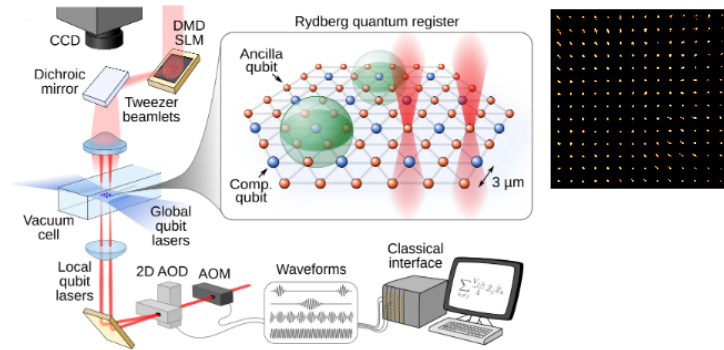
- An important challenge in modern science
- Significant applications in quantum technologies

## Scalable solid-state quantum devices

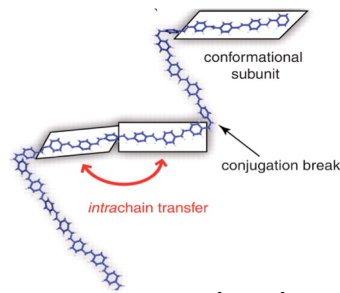
networks of strongly interacting qubits



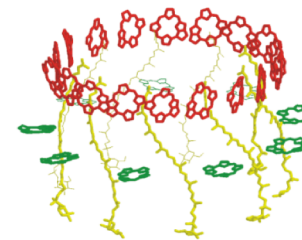
## Rydberg Atoms



Collective quantum states:  
at the heart of many  
important phenomena



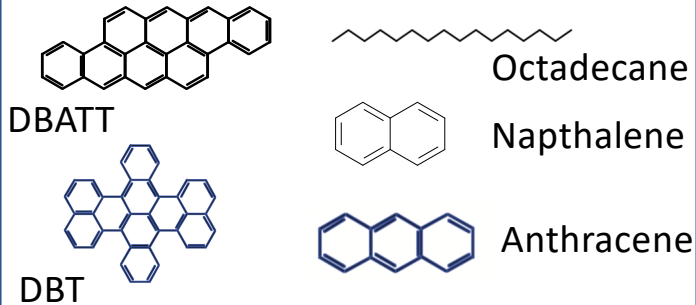
Conjugated polymers



Light harvesting complex

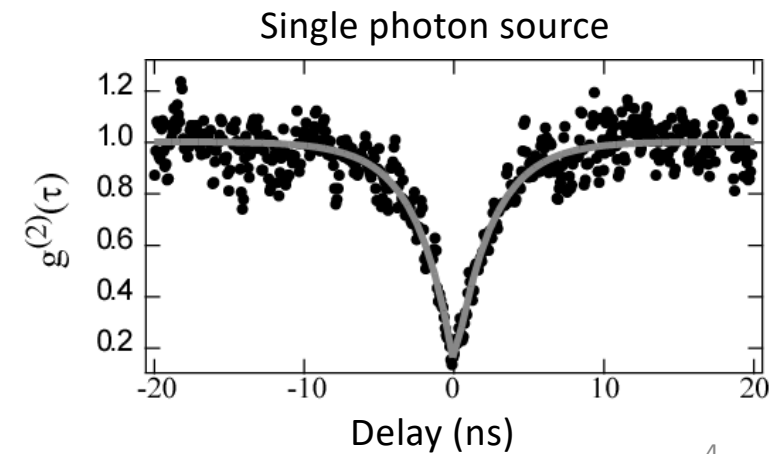
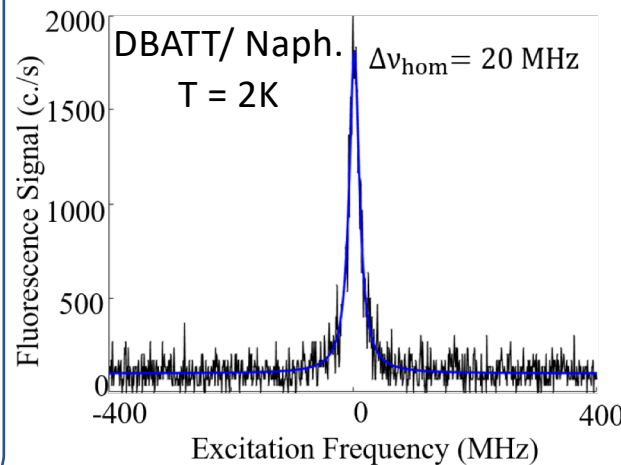
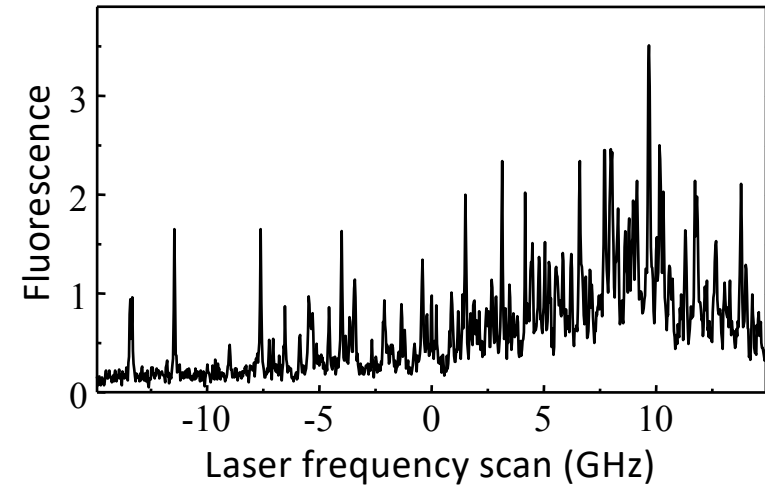
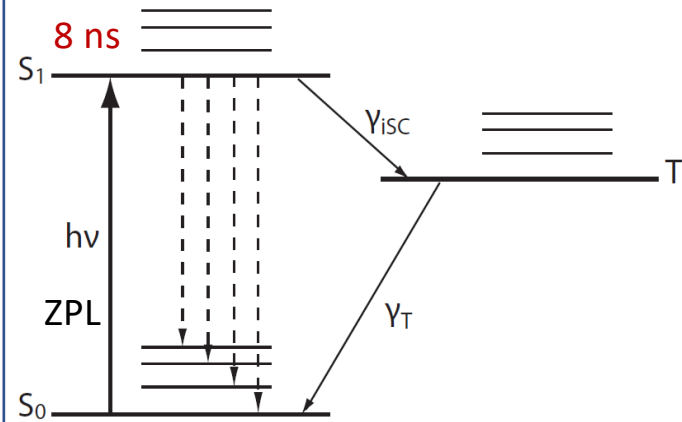
# Single molecule at low temperature (2K)

## Possible molecules (PAH) and host systems :



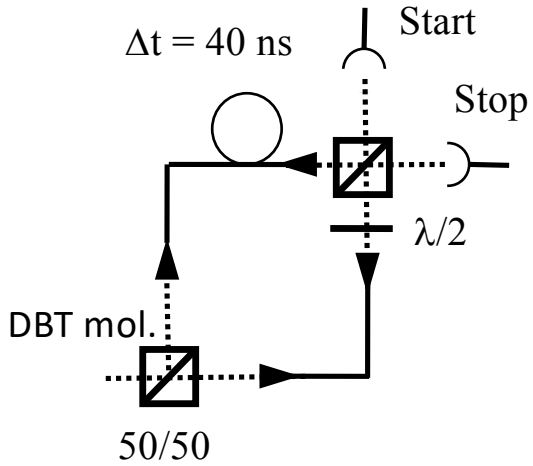
## Why Molecules are interesting?

- ✓ Photostability
- ✓ Behaves like a two level system
- ✓ Resonances with high quality factors (up to  $\sim 10^8$ )
- ✓ Narrow Zero Phonon Line
- ✓ Single photon source

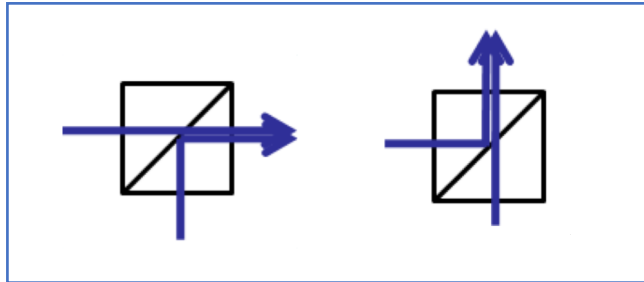


# Two photon interferences (ZPL)

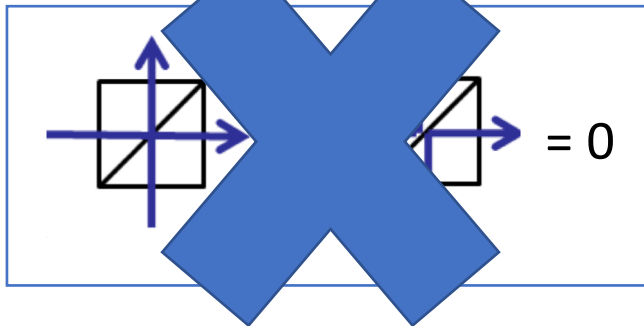
## Hong Ou Mandel Experiment :



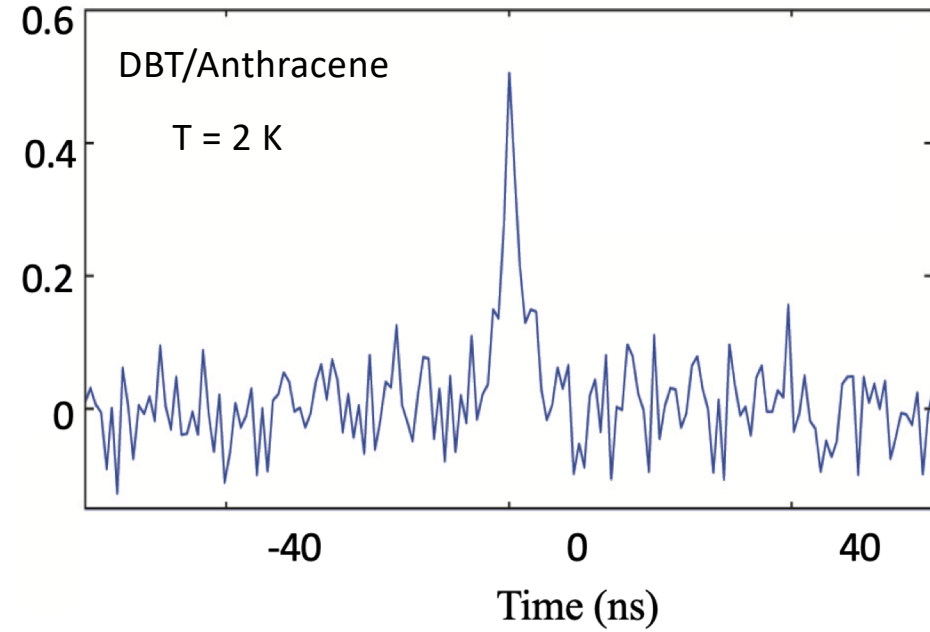
## Distinguishable events :



## Indistinguishable events :

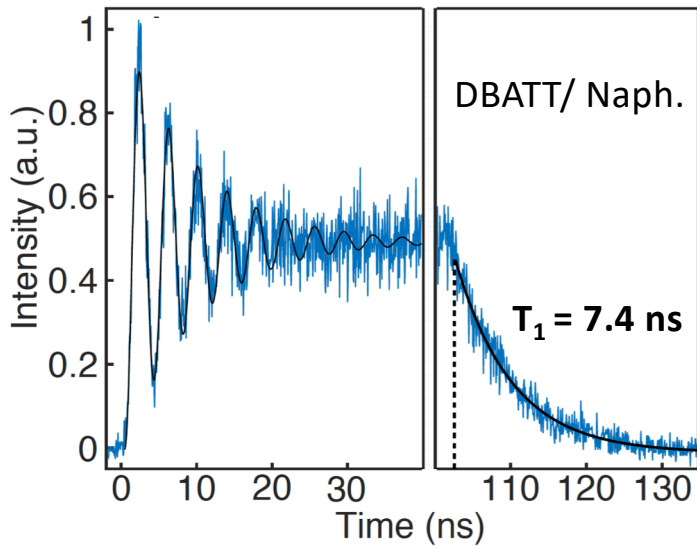
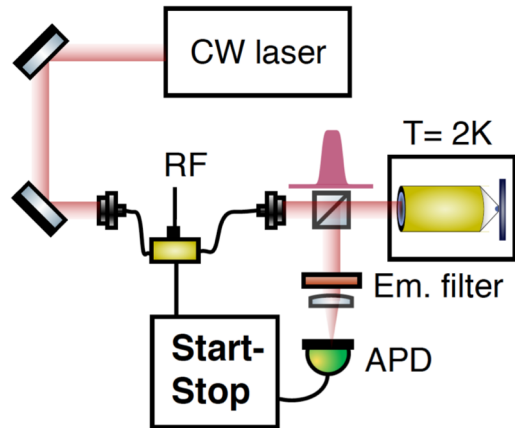


## Coalescence efficiency

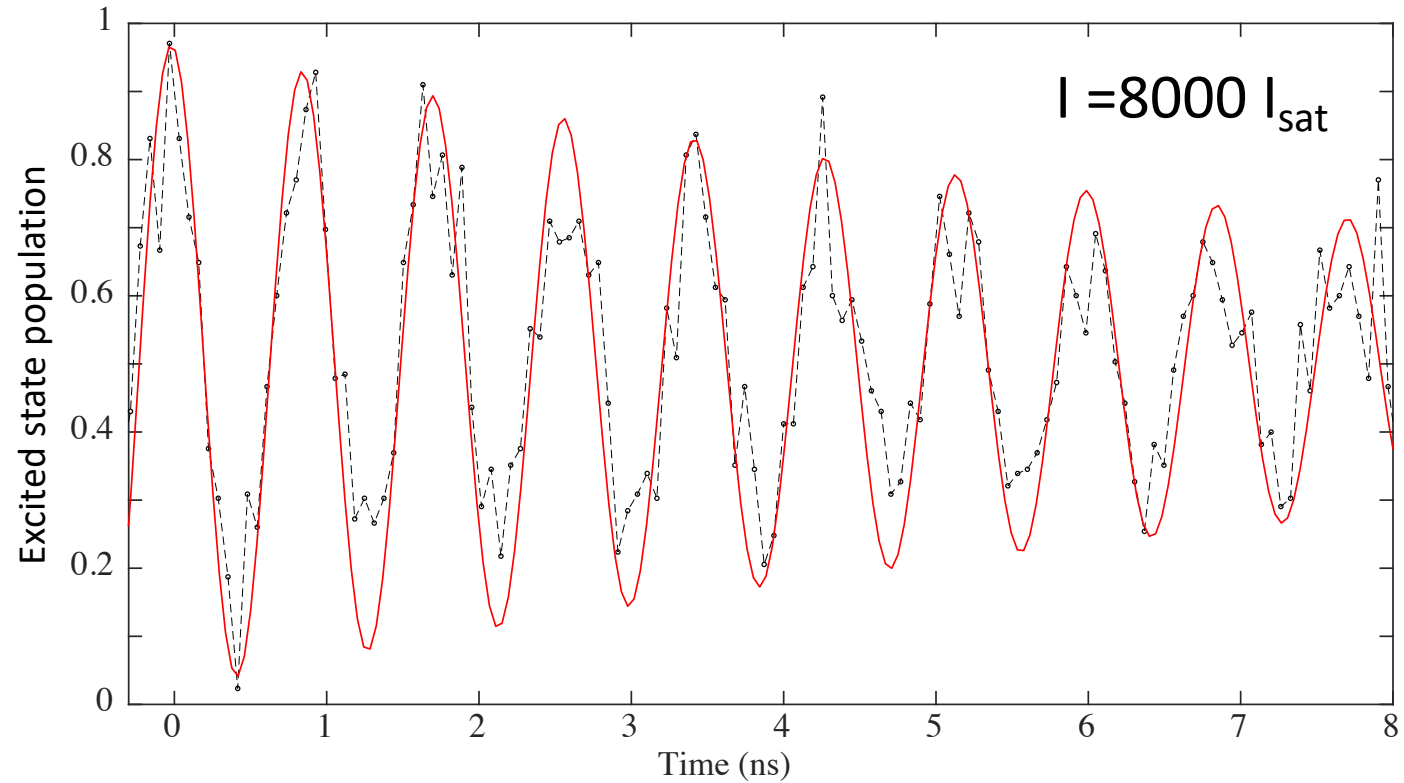


# Ultra fast Rabi oscillations :

## Experimental Setup :



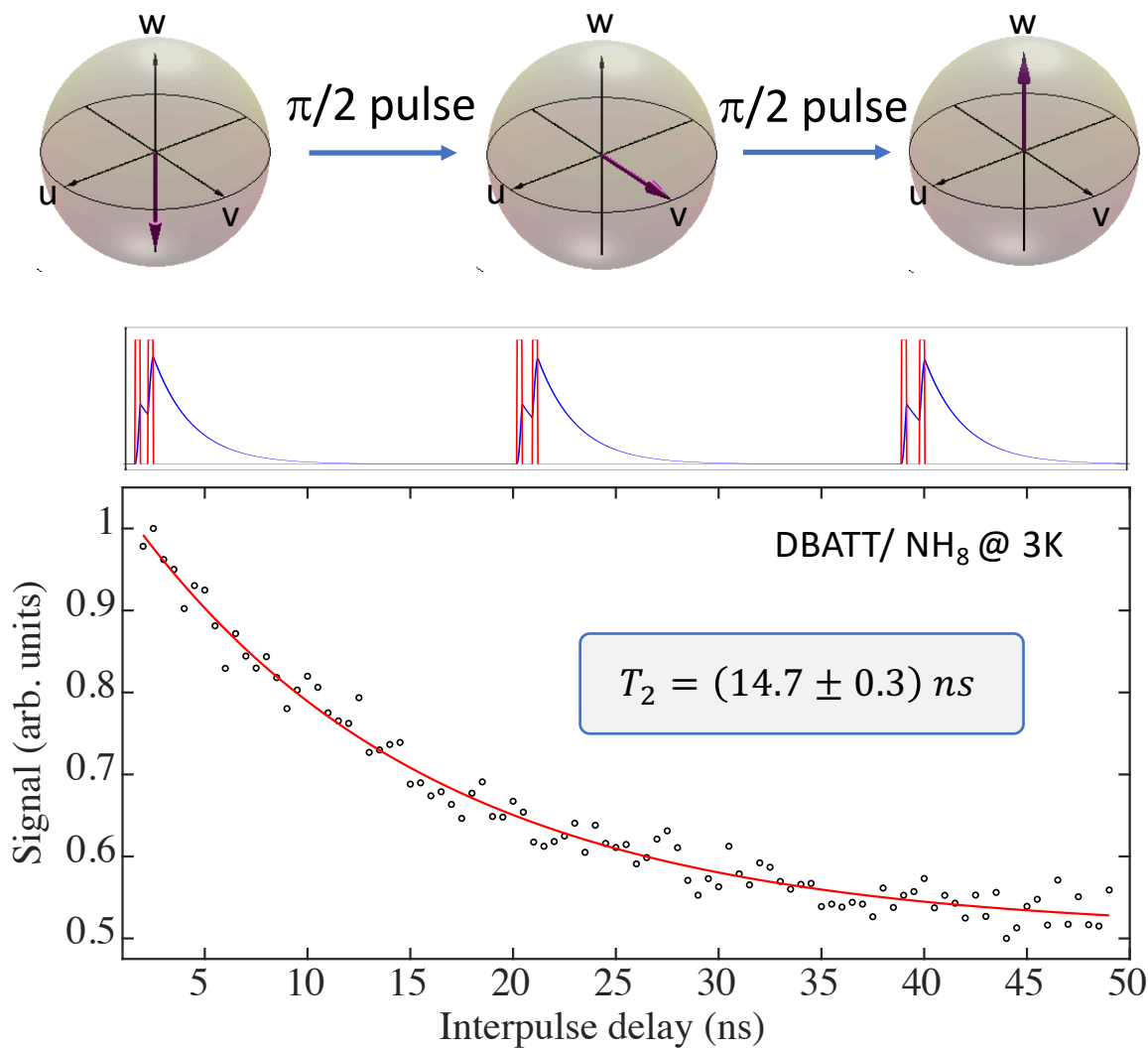
Decay measurements give :  $T_1$  and  $T_2$  times



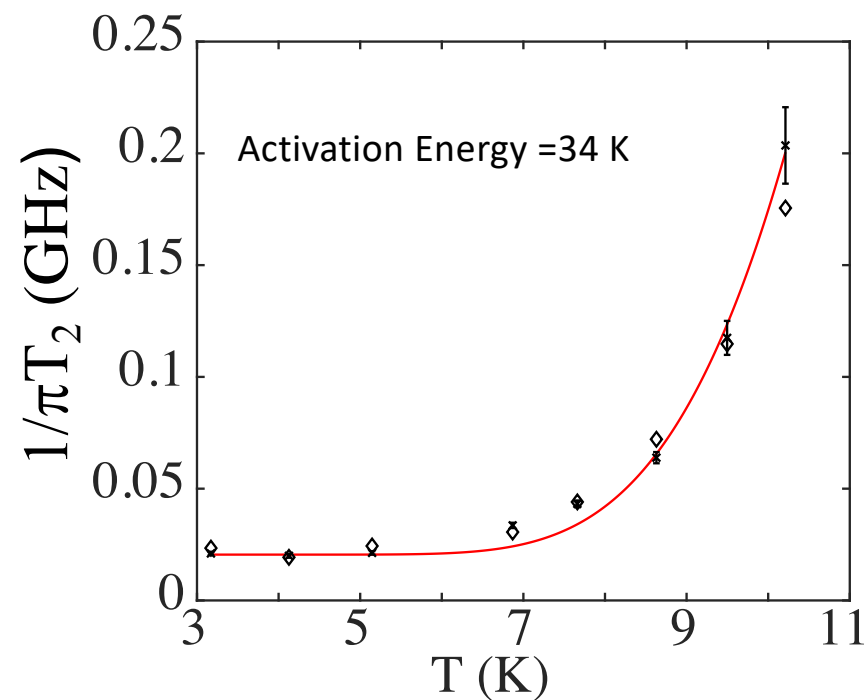
## Sub nanosecond Rabi oscillations :

- Period : **850 ps**
- No additional decoherence (**limited by  $2T_1$** )
- Excited state preparation fidelity :  **$(96 \pm 3) \%$**

# T<sub>2</sub> determination with Ramsey measurement



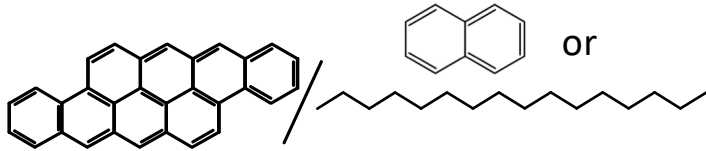
Deplano *et al.* In preparation  
(2022)



Follows the Arrhenius law :  $\gamma(T) = \gamma(0)e^{\frac{E_a}{RT}}$   
In agreement with the homogeneous FWHM evolution

# Stark Effect

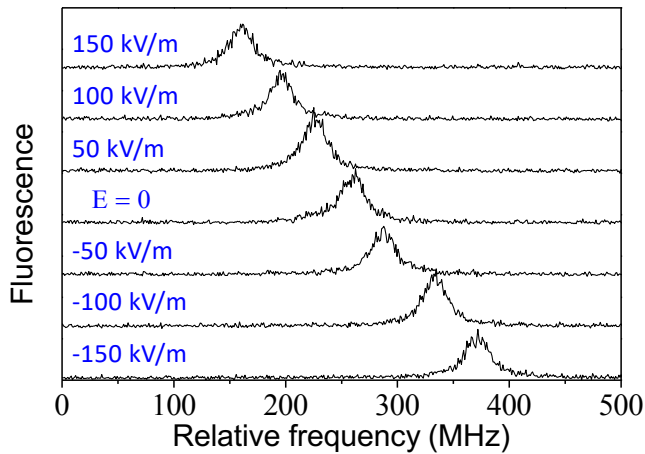
Molecular transition frequency shifts with electric fields :



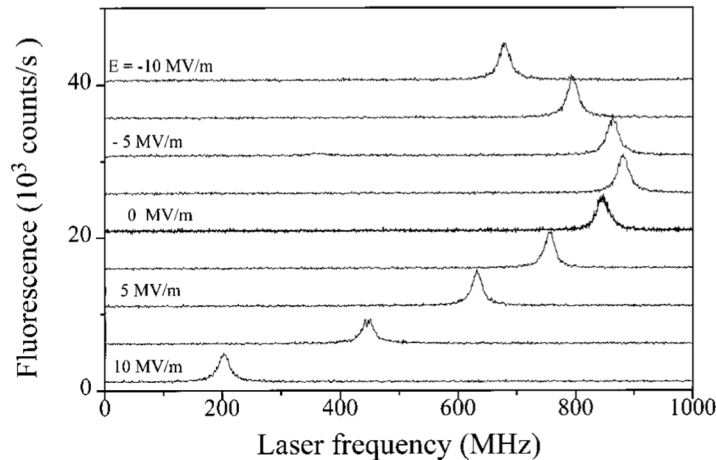
$$\hbar\omega(E) - \hbar\omega_0 = -\Delta\mu E - \frac{1}{2} \Delta\alpha E^2$$

Extremely sensitive probe of local electric fields

## Linear Stark Shift

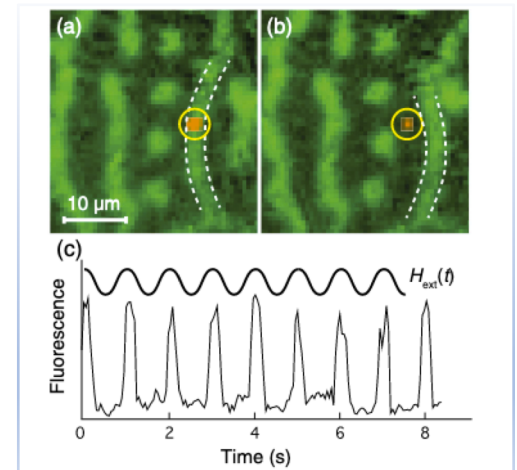


## Quadratic Stark Shift



Permanent dipole moment:  
 $\Delta\mu \sim 0.3 \text{ D}$ , ( $\sim 1 \text{ GHz/MV/m}$ )

J. Phys. Chem. A 103, 2429-2434 (1999)



Evidence of the electric field induced by a magnetization inhomogeneity in an iron garnet film.

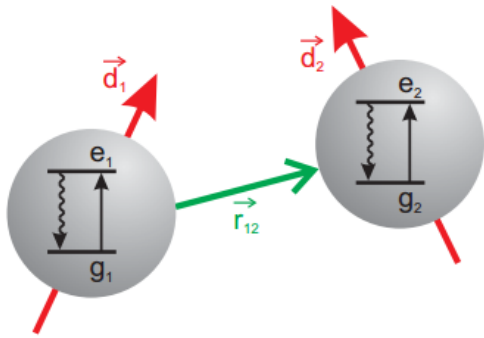
Phys. Rev. Lett. 115, 027601 (2015)



# Coherent optical dipole coupling of two emitters

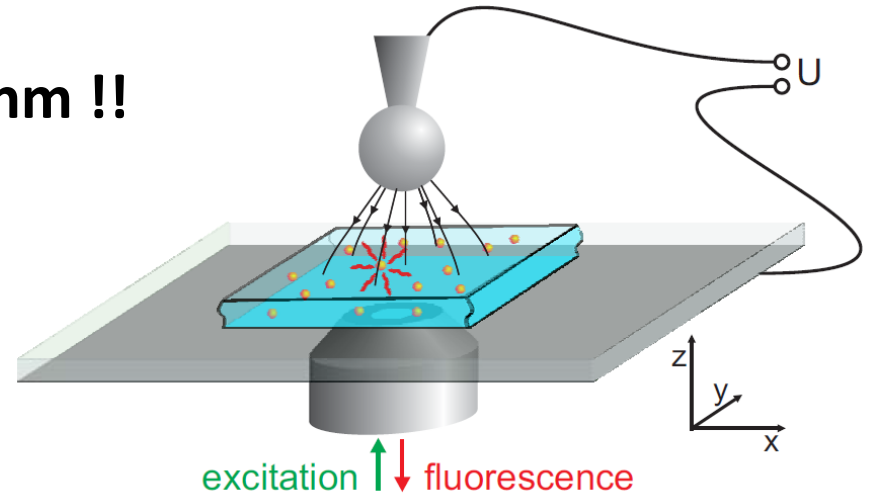
First experimental demonstration of the **Coherent Optical Dipole Coupling of Two Individual Molecules**

Hettich, C. et al. *Science*  
298, 385 (2002)



$$\text{Dipole interaction} \propto \frac{1}{(r_{12})^3}$$

12 nm !!



Need for an optical method to image the interacting emitters

# **Cryogenic super-resolution microscopy by Excited State Saturation (ESSat)**

# Super-resolution fluorescence microscopies

## Imaging with resolutions below the diffraction limit

- revolutionized imaging capabilities offered by far-field optical microscopy
- development mainly driven by the need for better resolutions to study biological processes in live samples
- nm optical resolutions give access to nanoscale molecular organizations



Photo: Matt Staley/HHMI  
**Eric Betzig**  
Prize share: 1/3



© Bernd Schuller, Max-Planck-Institut  
**Stefan W. Hell**  
Prize share: 1/3

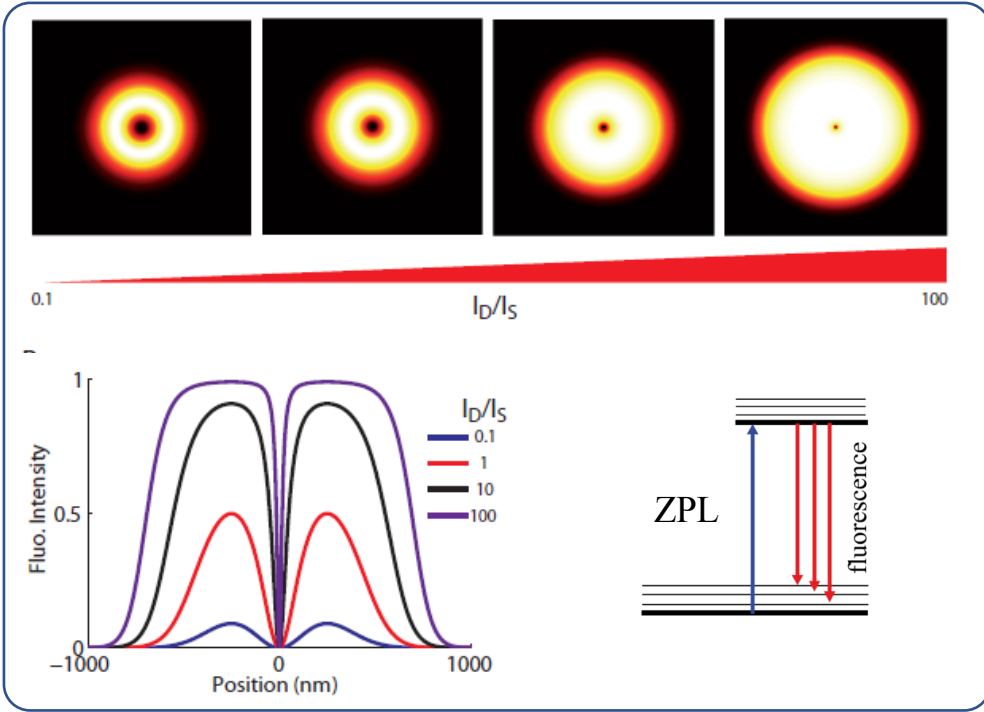


Photo: K. Lowder via Wikimedia Commons, CC-BY-SA-3.0  
**William E. Moerner**  
Prize share: 1/3

The Nobel Prize in Chemistry 2014 was awarded jointly to Eric Betzig, Stefan W. Hell and William E. Moerner *"for the development of super-resolved fluorescence microscopy"*.

Demonstrated @ Room temperatures and are based on: controlling fluorescence emission volumes to highly localized regions, using structured illumination schemes or stochastic detection of single emitters

# Principle of ESSat nanoscopy

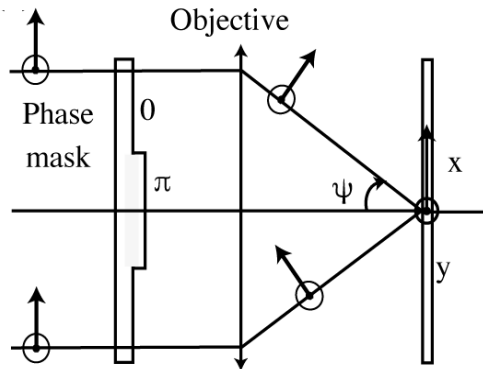


For a two-level system :

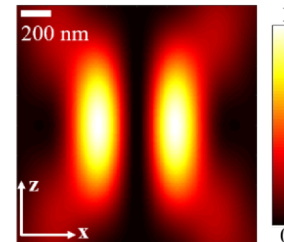
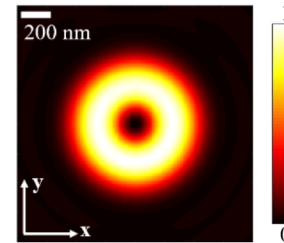
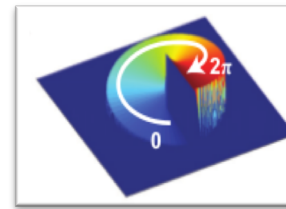
$$\sigma = \alpha_{DW} \frac{3\lambda^2 \Gamma_{rad}}{2\pi \gamma_{hom}}$$

L.T. :  $\sigma = 1.6 \times 10^{-9} \text{ cm}^2$

R.T. :  $\sigma \sim 10^{-16} \text{ cm}^2$

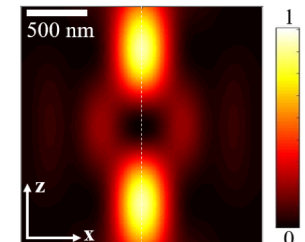
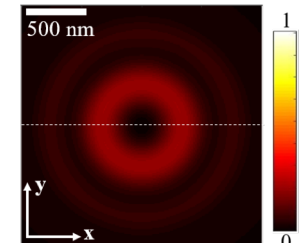
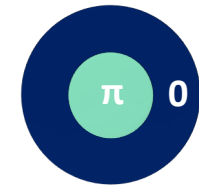


Helix phase plate :  
Unaffected axial resolution



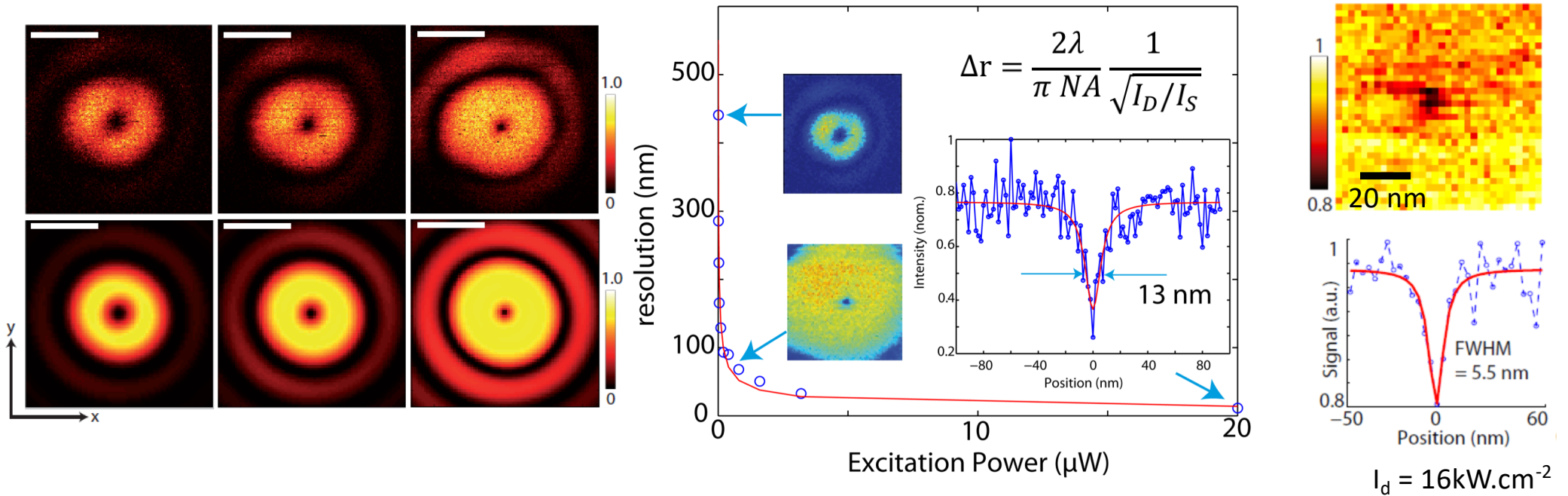
2D ESSat nanoscopy

0- $\pi$  phase plate :  
3D intensity gradient around the center

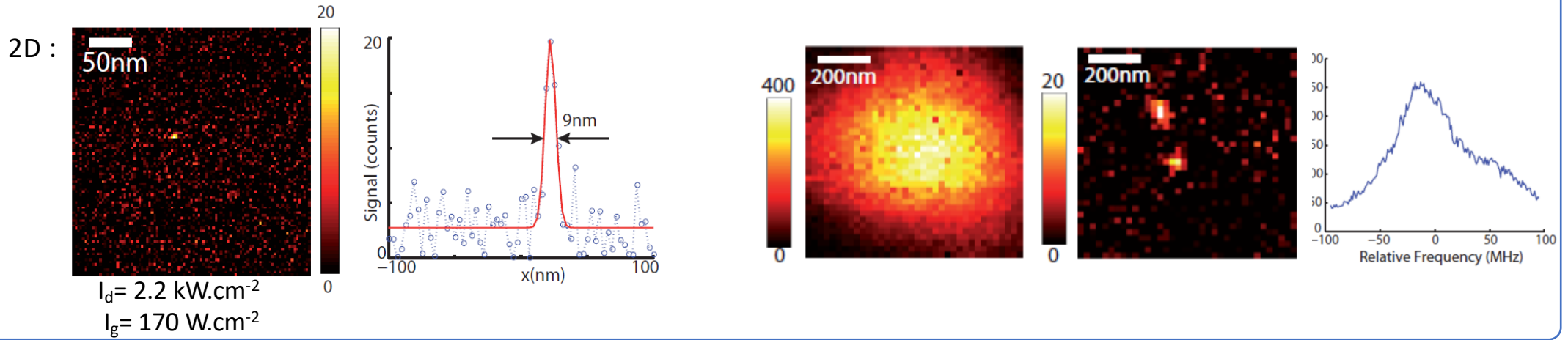


3D ESSat nanoscopy

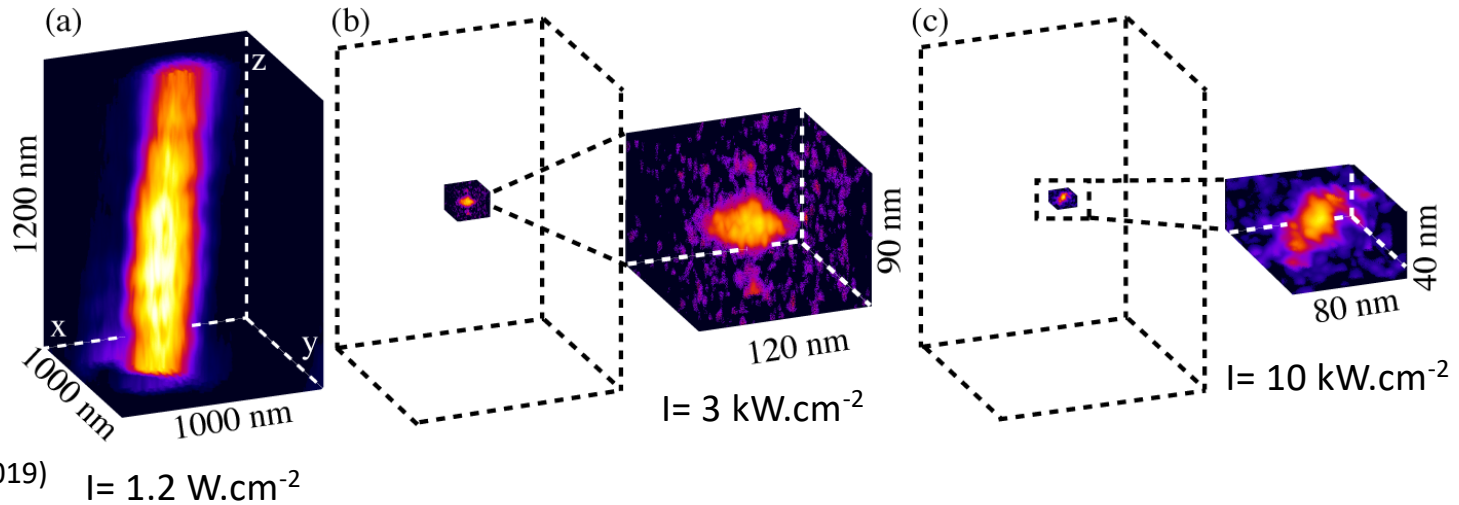
# 2D-Excited State Saturation Nanoscopy (ESSat)



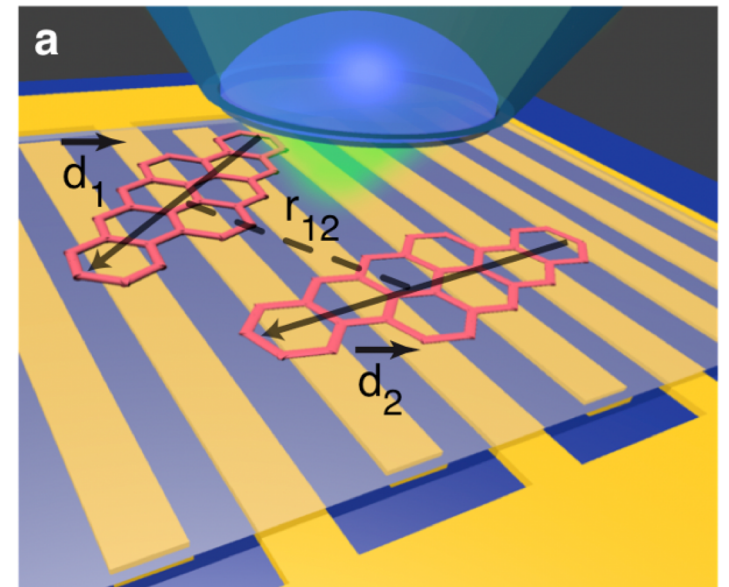
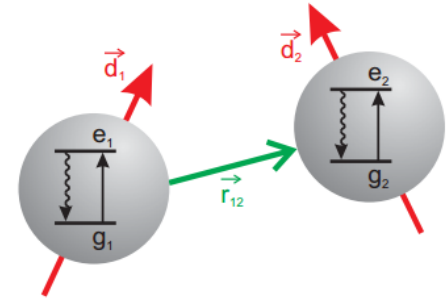
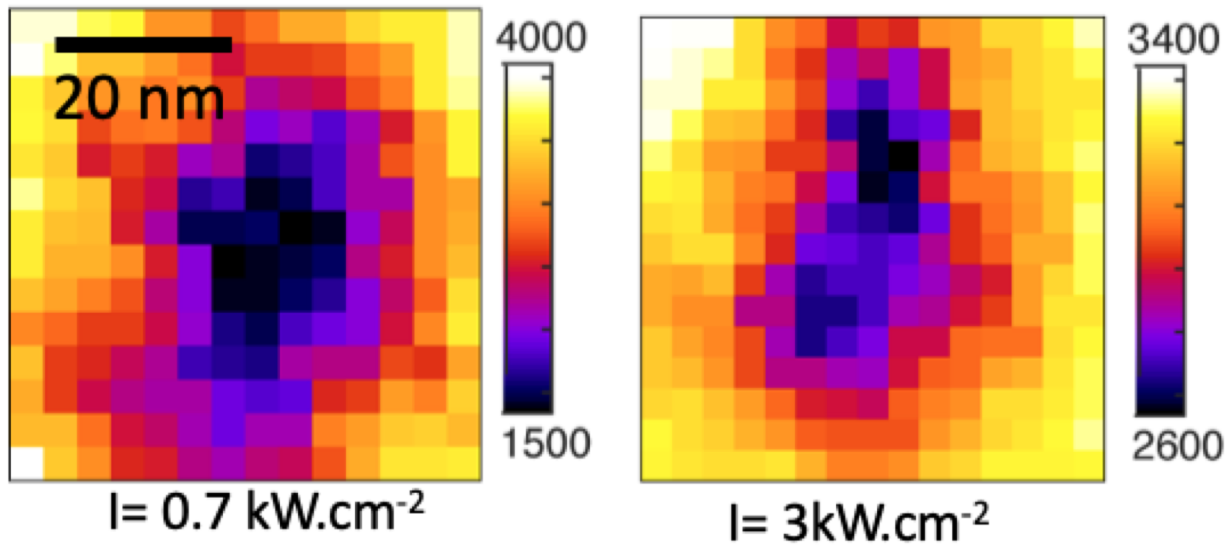
# Modulated Excited State Saturation at 2K :



3D :



# Finding close emitters at 2K :



Molecular entanglement ?

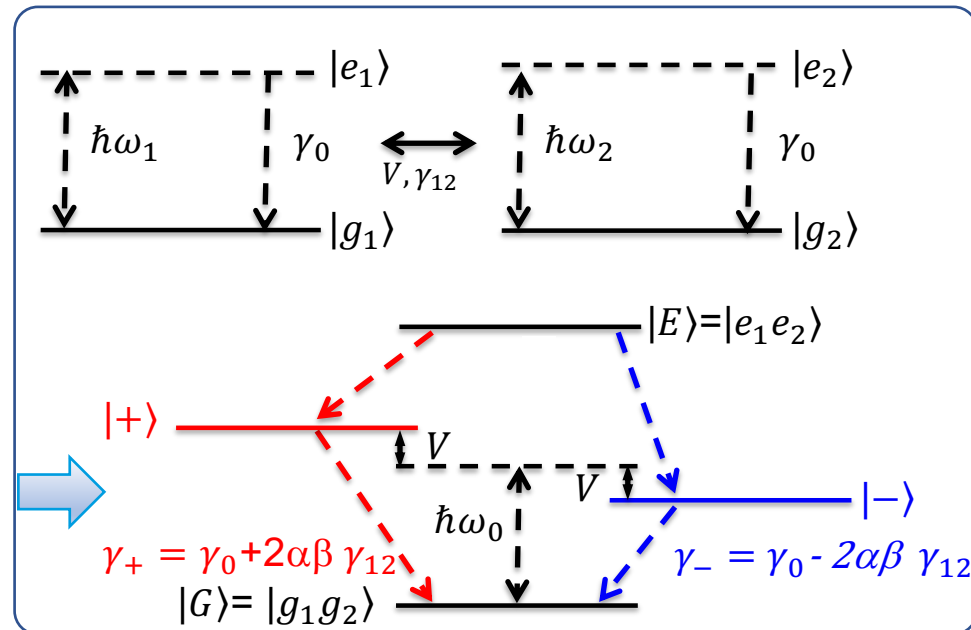
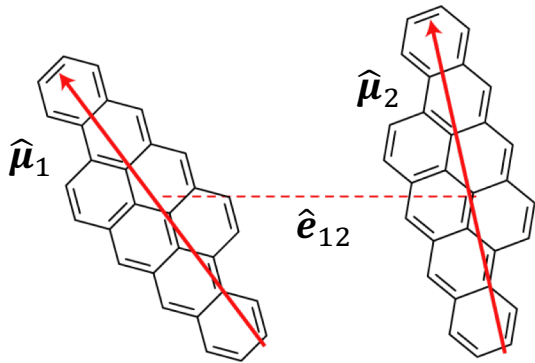
# Coherent Optical Dipole Coupling of Two Individual Molecules

Coupling constant :

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

Cross-damping rate :

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



2 entangled « delocalized » states

$$\begin{cases} |+\rangle = (\alpha|e_1, g_2\rangle + \beta|g_1, e_2\rangle) \\ |-\rangle = (\beta|e_1, g_2\rangle - \alpha|g_1, e_2\rangle) \end{cases}$$

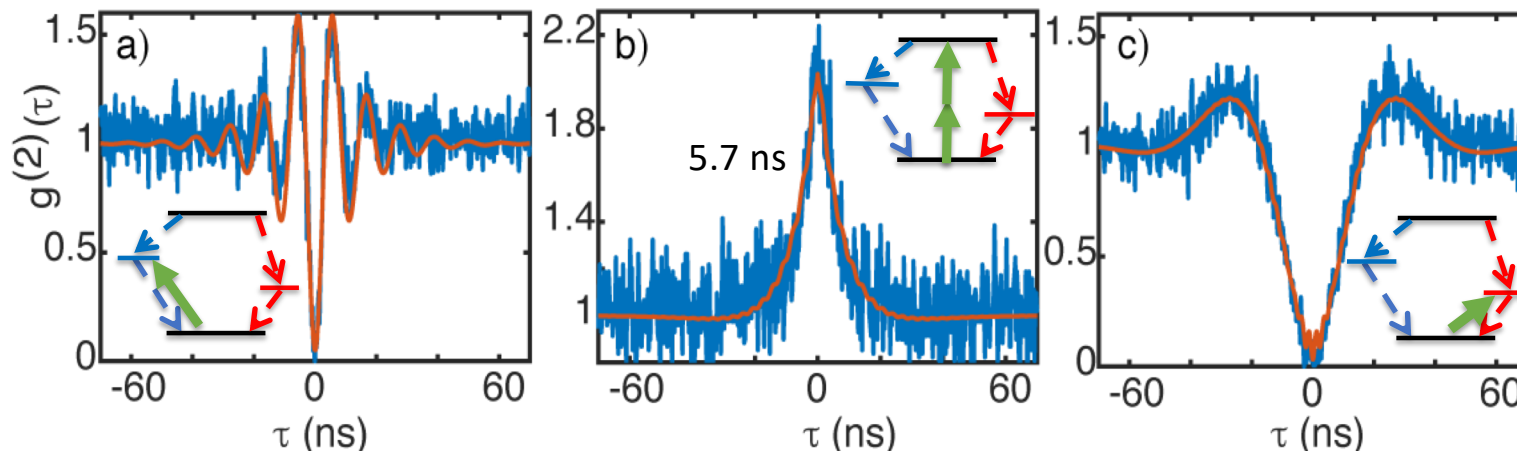
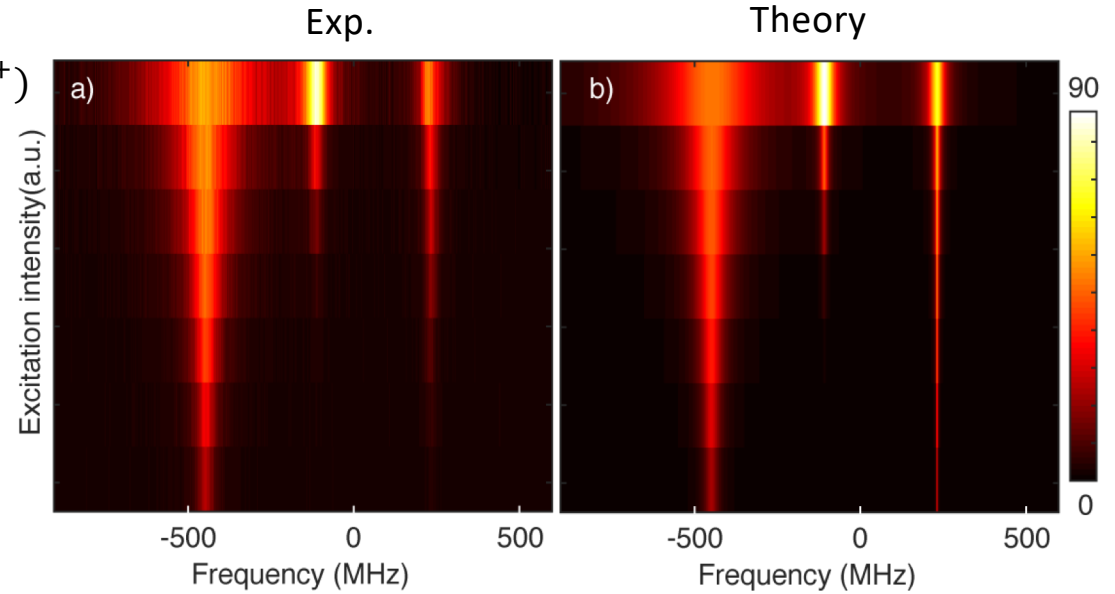
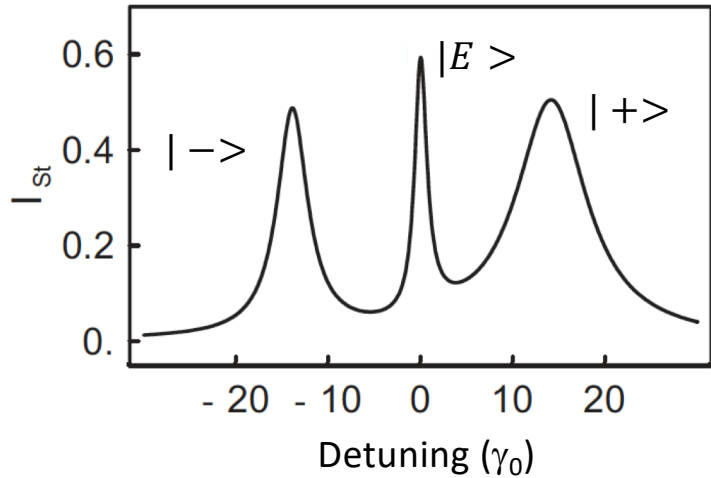
Hettich et al. Science (2002)



# Signatures of the coherent dipole dipole interaction

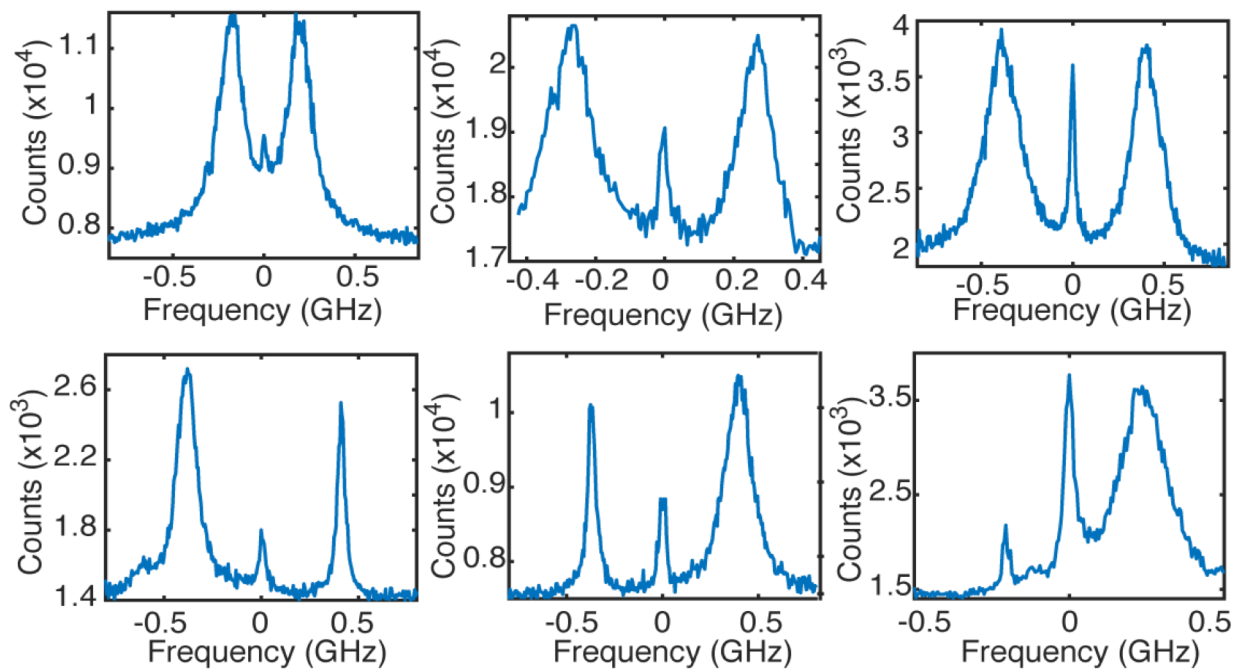
Excitation spectrum calculated using Master Equation :

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

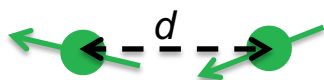


# Gallery of coupled molecules

Optical nanoscopy allows finding coherently coupled molecules with different degree of entanglement

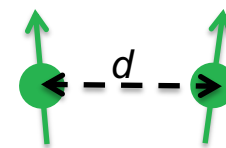


J Configuration

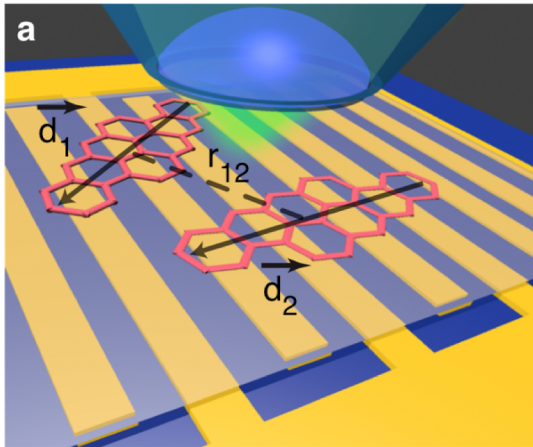


*Nat Commun* **13**, 2962 (2022)

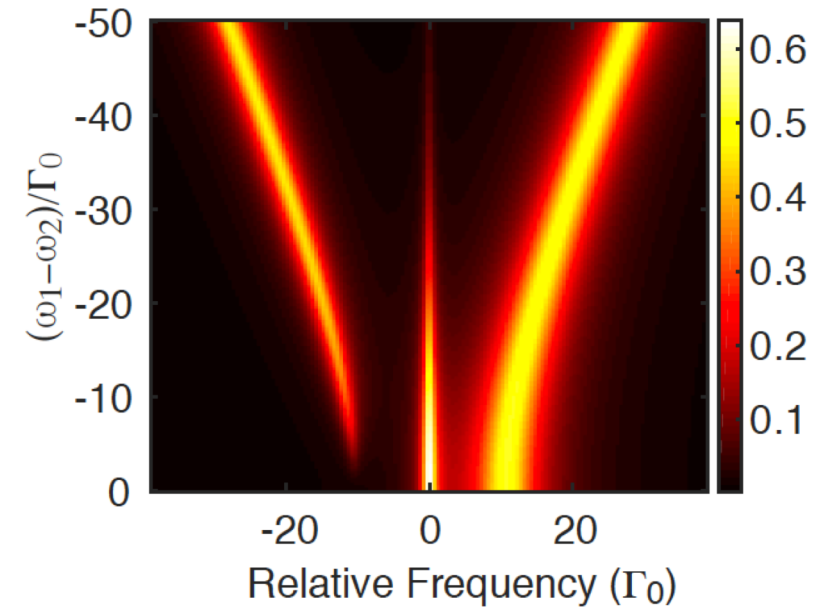
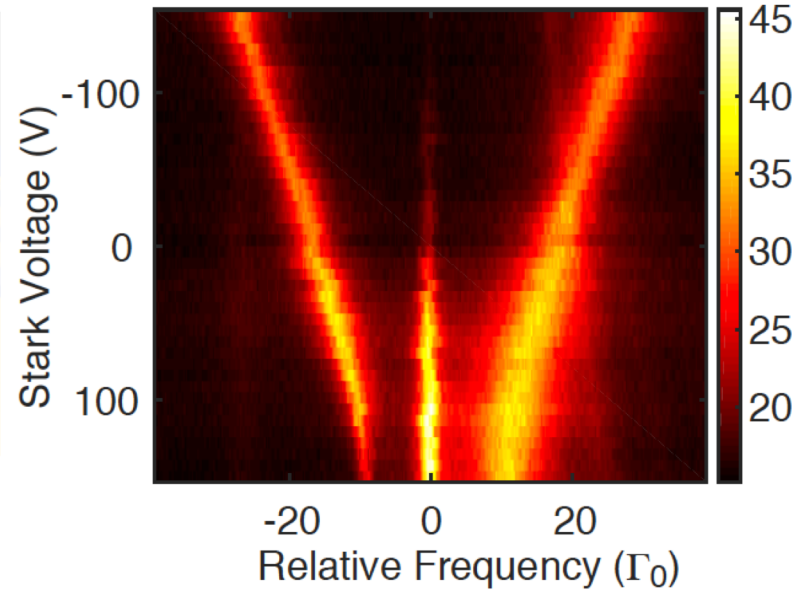
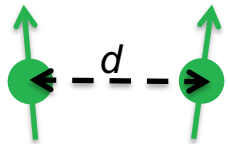
H Configuration



# Manipulating the molecular Entanglement



H Configuration



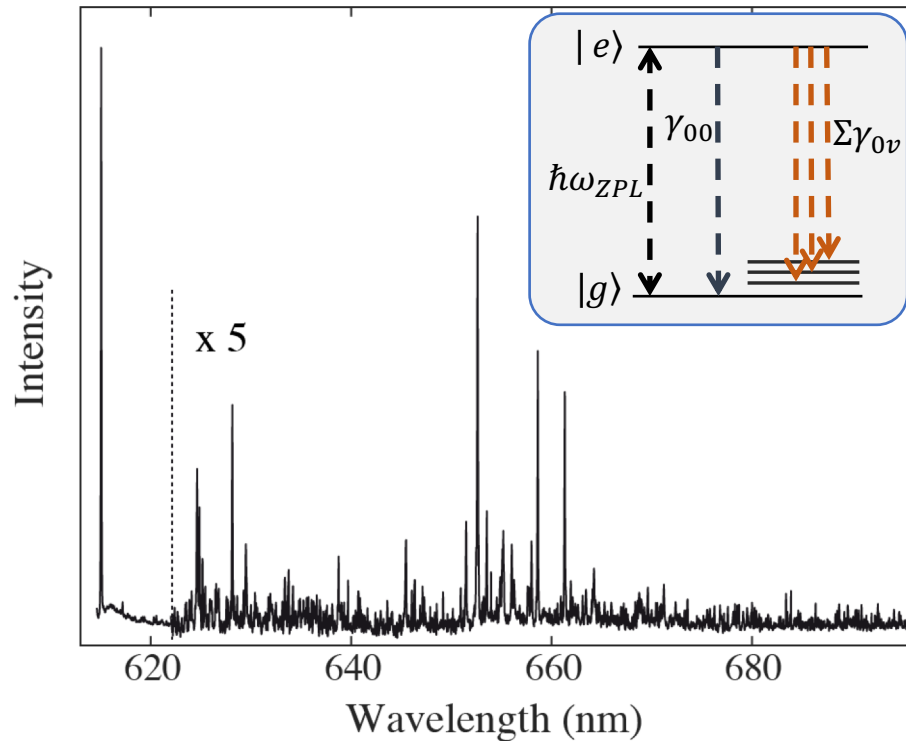
Maximum entanglement when  $\Delta \rightarrow 0$ :  
 $\alpha, \beta \rightarrow \frac{1}{\sqrt{2}}$ , and  $\gamma_+ \rightarrow 2\gamma_0, \gamma_- \rightarrow 0$   
 From  $V/h \sim 10\gamma_0$ , estimated distance:  
 $d \sim 25\text{nm}$

$|+\rangle$  and  $|-\rangle$  are close to  
 Bell States :  
 $|+\rangle = \frac{1}{\sqrt{2}}(1.13|eg\rangle - 0.84|ge\rangle)$   
 $|-\rangle = \frac{1}{\sqrt{2}}(0.84|eg\rangle - 1.13|ge\rangle)$

# Aromatic molecules in Molecular Crystals

## Emission Spectrum

- Narrow Zero Phonon Lines: purely radiative transition
- Red-shifted vibrational lines and phonon sidebands



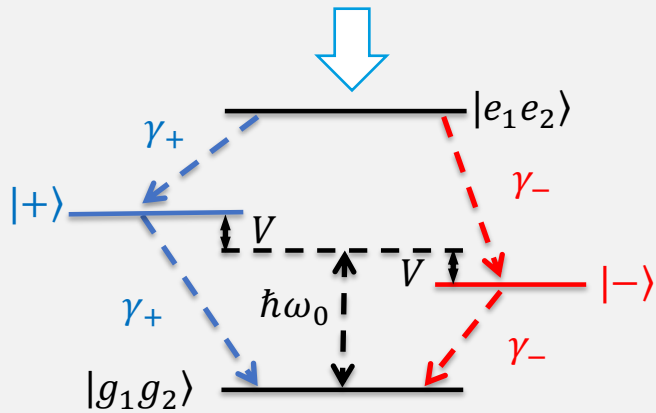
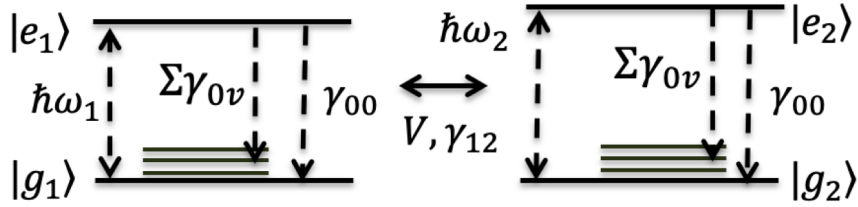
Excited state lifetime  $\gamma_0^{-1}$   
 $\gamma_0 = \gamma_{00} + \Sigma\gamma_{0v}$   
 with  $\gamma_{00} = \eta_{ZPL}\gamma_0$

Combined Debye-Waller /  
 Franck-Condon factor  

$$\eta_{ZPL} = \frac{\gamma_{00}}{\gamma_{00} + \Sigma\gamma_{0v}}$$

Low Temperature single molecule Emission Spectrum  $\rightarrow \eta_{ZPL} \sim 0.35$

# sub/super-radiance for two coherently coupled molecules :



For  $\omega_2 = \omega_1$  and  $\mu_1 // \mu_2$  :  
 $\gamma_+ \rightarrow \gamma_0(1 + \eta_{ZPL})$   
 $\gamma_- \rightarrow \gamma_0(1 - \eta_{ZPL})$

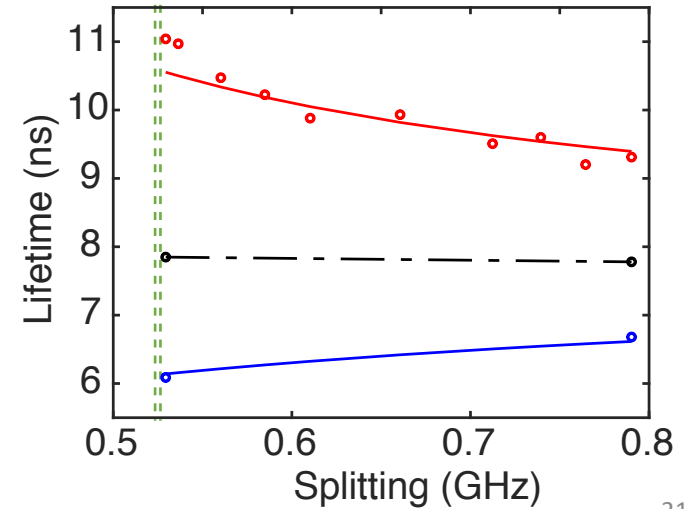
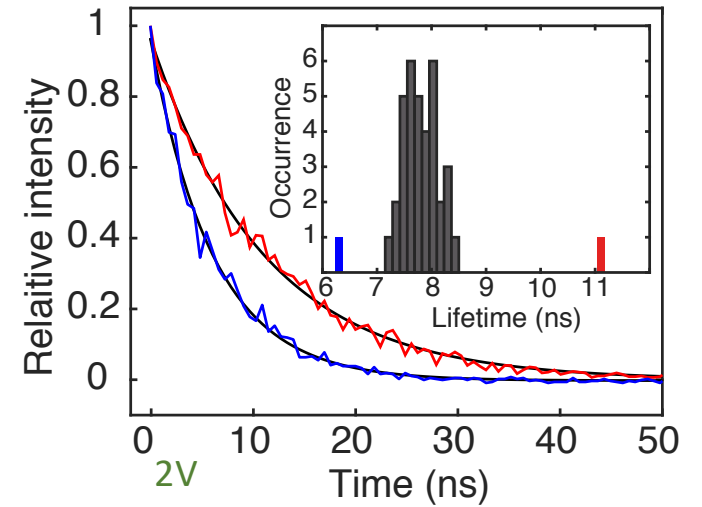
Coherent dipole-dipole coupling

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

Incoherent cross-damping rate

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

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# Delocalized states coupling to laser fields

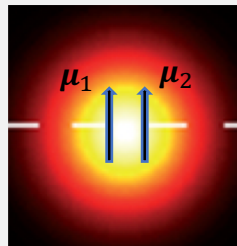
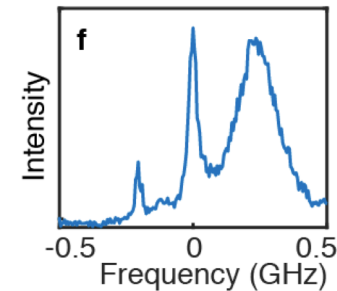
Hamiltonian governing the coupled with a driving laser of frequency  $\omega_L$

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

Complex Rabi frequencies:  $\Omega_i = -\vec{\mu}_i \cdot \vec{\mathcal{E}}(\vec{r}_i - \vec{r}_c) / \hbar$

Matrix elements that govern the laser coupling strengths:

$$\left. \begin{array}{l} \langle -|H|G\rangle \propto \alpha\Omega_1 - \beta\Omega_2 \\ \langle +|H|G\rangle \propto \beta\Omega_1 + \alpha\Omega_2 \end{array} \right\} \rightarrow \text{depend on } \alpha, \beta \text{ and } \Omega_i$$



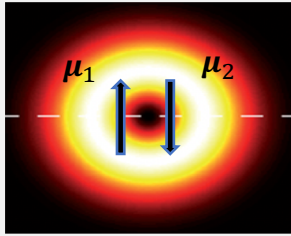
Gaussian laser spot centered between the molecules:

- Generation of in-phase dipole oscillations with  $\Omega_1 = \Omega_2$ .

When  $\Delta$  approaches zero ( $\alpha \approx \beta \approx 1/\sqrt{2}$ ):

- Excitation of the superradiant state is enhanced (by up to  $\sqrt{2}$ ), while that of the subradiant state becomes forbidden.

# Enhanced laser coupling to the subradiant state

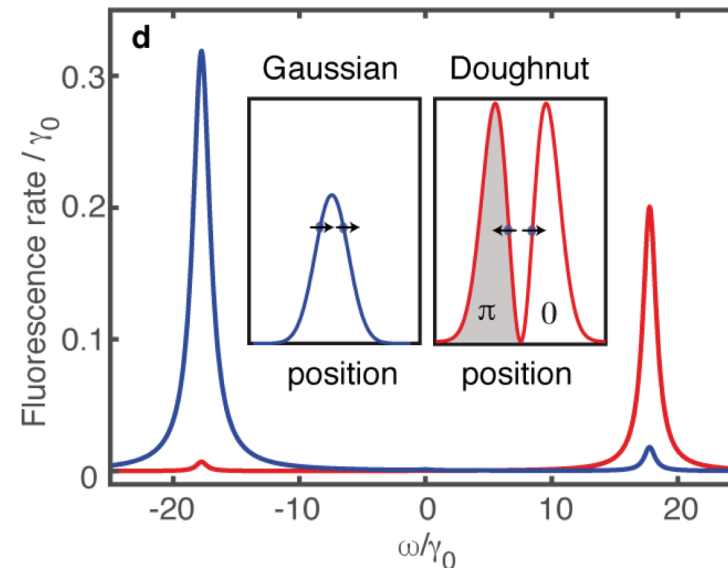
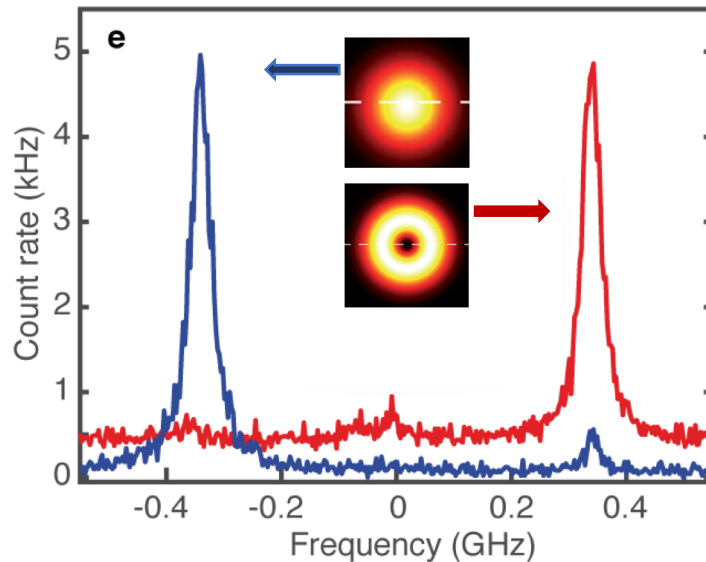
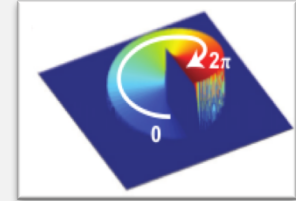


Doughnut-shaped laser spot centered between the molecules:

- Opposite-phase dipoles oscillations,  $\Omega_1 = -\Omega_2$ .

When  $\Delta$  approaches zero ( $\alpha \approx \beta \approx 1/\sqrt{2}$ ):

- Excitation of the subradiant state is enhanced, while that of the superradiant state is forbidden.



A method for selective preparation of long-lived subradiant states

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

# Conclusions and Perspectives

## Conclusions:

Ultra stable quantum system : excitation during weeks without any photobleaching,

Optical nanoscopy to find and super-resolve coupled pairs in various configurations,

Possibility to entangle and disentangle two molecules with an electric field.

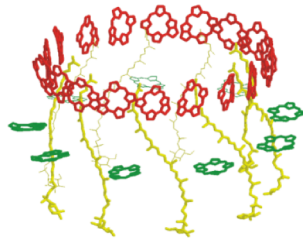
## Perspectives:

Quantum logic gates

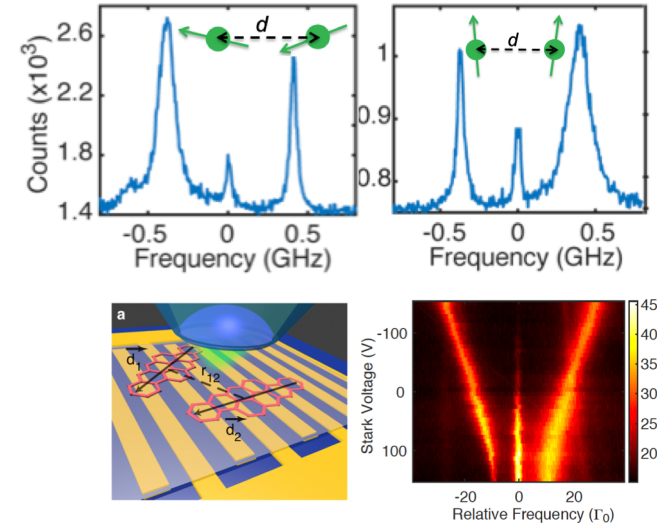


Phys. Rev. Lett. 104, 010503 (2010)

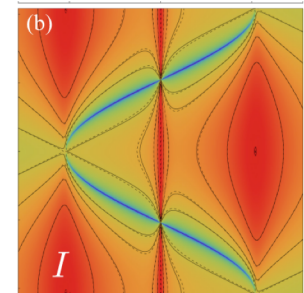
Decoherence effects with the temperature



Nature, 2007, 446, 782-786 (2007)



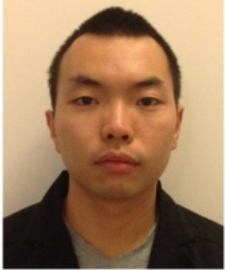
Resonant fluorescent excitation spectra



Phys. Rev. Research 3, 033136 (2021)



# Acknowledgements



Bin  
Yang



Reenu  
Baby



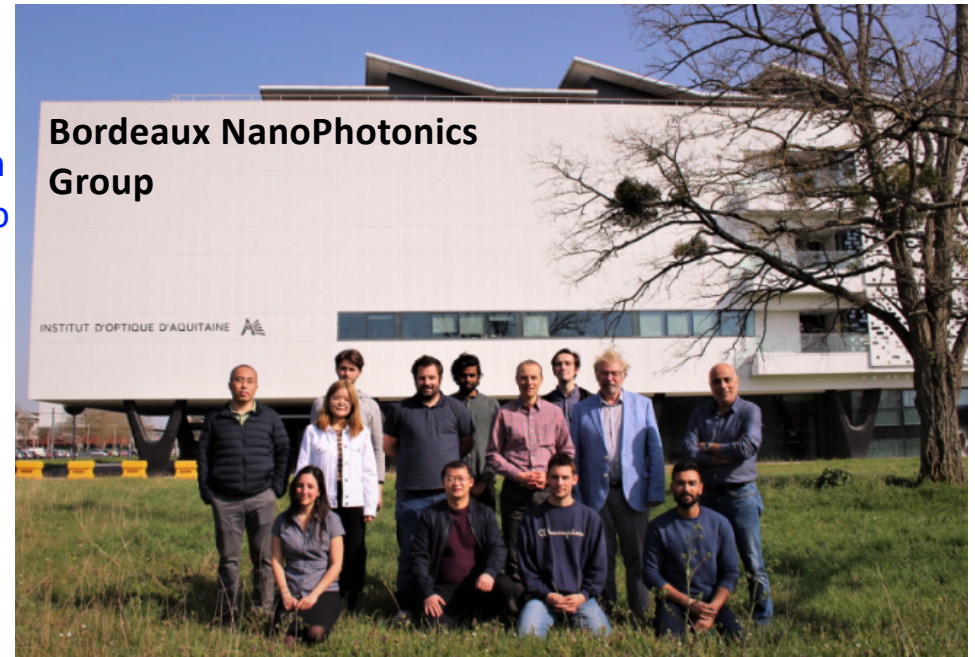
Quentin  
Deplano



Philippe  
Tamarat



Brahim  
Lounis



Thank you for your attention

