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Coherently coupled molecules at cryogenic temperatures : from super/sub radiance to optical selection rules

Q. Deplano, J.-B. Trebbia, Ph. Tamarat & B. Lounis

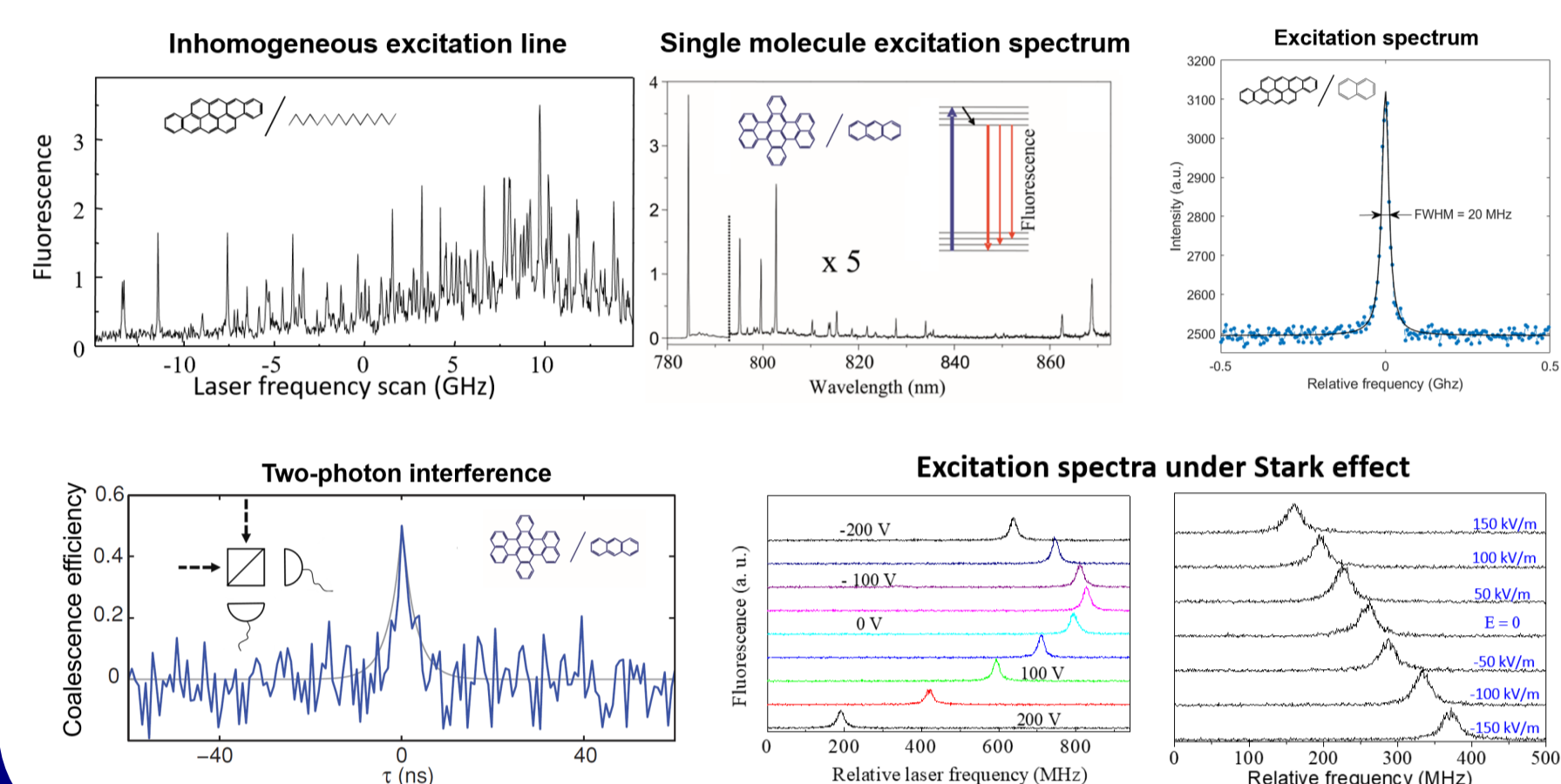
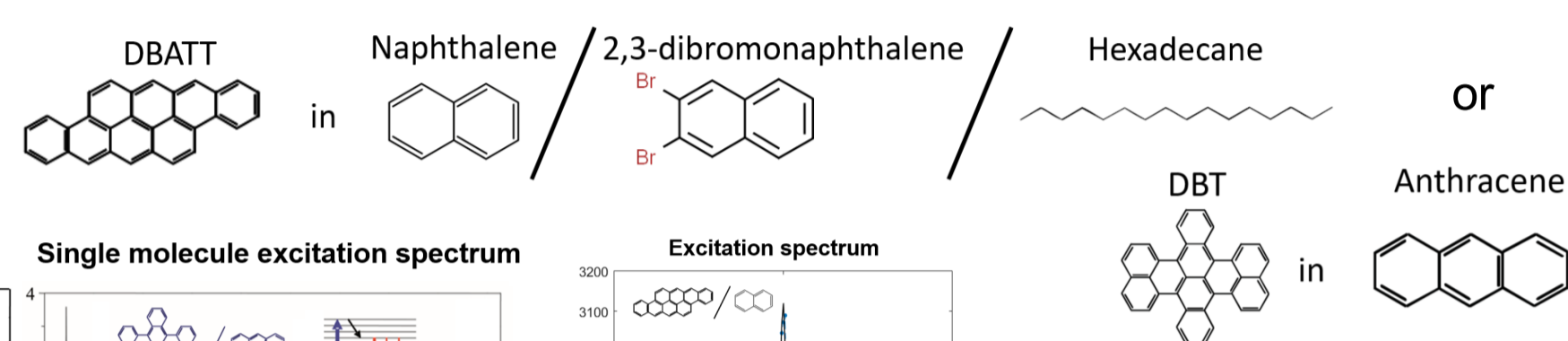
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Abstract

Using quantum emitters (such as quantum dots, atoms, NV defects in diamond ...) in order to create quantum logic gates, is a widely studied topic in modern physics. Among all of those objects, organic molecules trapped in condensed matter are nearly ideal quantum emitters dispensing single indistinguishable photons. Moreover, at low temperature ($\approx 2\text{K}$), molecules are very well modeled by a two-level system. At nanometric distances, two molecules can be coupled by the dipole-dipole interaction leading to the apparition of two delocalized states respectively sub- and super-radiant. The observation of two coupled dipoles has been reported only once in the literature in 2002 [1], despite of numerous interesting applications for quantum computing. The major limitation is the low probability to find an entangled pair in a sample. For this purpose, we lately developed an optical nanoscopy method based on the saturation of the excited state by a scanned donut shaped beam, and operating at cryogenic temperature, allowing us to find some pairs of coupled dipoles. We obtained a sub 10-nm resolution for a single dipole imaging. When the donut is scanned over an entangled pair, the Rabi frequencies seen by the two emitters varies which modifies the four energy levels of the pair by AC lightshift and leads to the apparition of fluorescence dips in the final image, when two levels become degenerated. With this technique we succeeded in doing specific images of an entangled system delocalized over 20 nm. The goal of the research activity presented here is to manipulate, on demand, the degree of entanglement of such a system. The latter could only be changed by applying an external electric field on the pair, which tunes the energy difference of the two emitters by Stark effect. We have been able to put at resonance the two emitters and observed a broadening and shrinking of the symmetric and antisymmetric transitions compatible with a modification of their relative lifetimes.

Fluorescent organic molecules at 2K

Different host-guest systems are possible



- Single molecules dispense indistinguishable photons
- DBATT in naphthalene has been chosen for its high photostability
- The Stark shift depends on the states polarizability α and the permanent dipole moment $\langle d \rangle$:

$$\Delta\omega(E) = -\frac{1}{2}\Delta\alpha E^2 - \langle d \rangle \cdot E$$

Tailoring the degree of entanglement

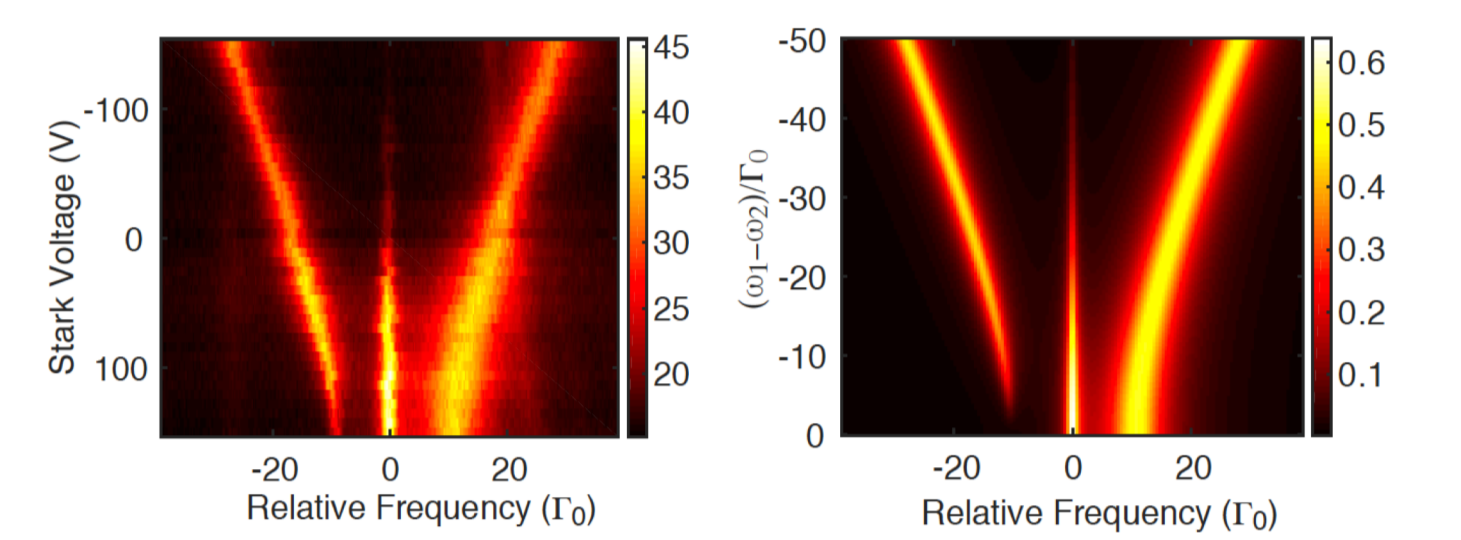
Applying a DC external electric field :

- Microfabricated gold electrodes :

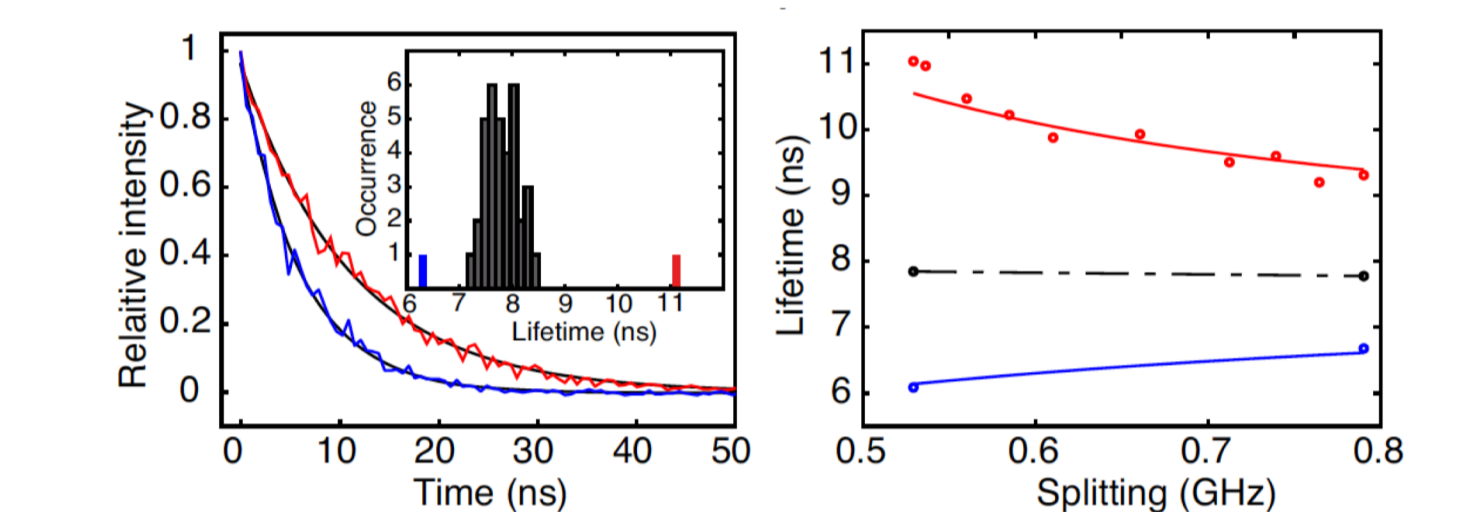
$$E_{max} \approx 5 \cdot 10^6 \text{ V} \cdot \text{m}^{-1}$$

- Close to resonance, the width and the amplitude of the subradiant transition decrease while the supradiant ones and the two photon amplitude increase.
- Clear difference between the sub- and the super-radiant lifetimes close to resonance.
- Experimental data follow the theoretical evolution determined by V , δ and the Debye Waller factor.
- Electron-phonon coupling \Leftrightarrow Debye-Waller factor that limits the sub- and super-radiance

$$\alpha_{DW} = 1 - \frac{\sum \gamma_{ov}}{\Gamma_0} \approx 0.3 \Rightarrow \Gamma_{00} = \alpha_{DW} \Gamma_0$$



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



Delocalized states coupling to laser fields :

Hamiltonian governing the coupled with a driving laser of frequency ω_L :

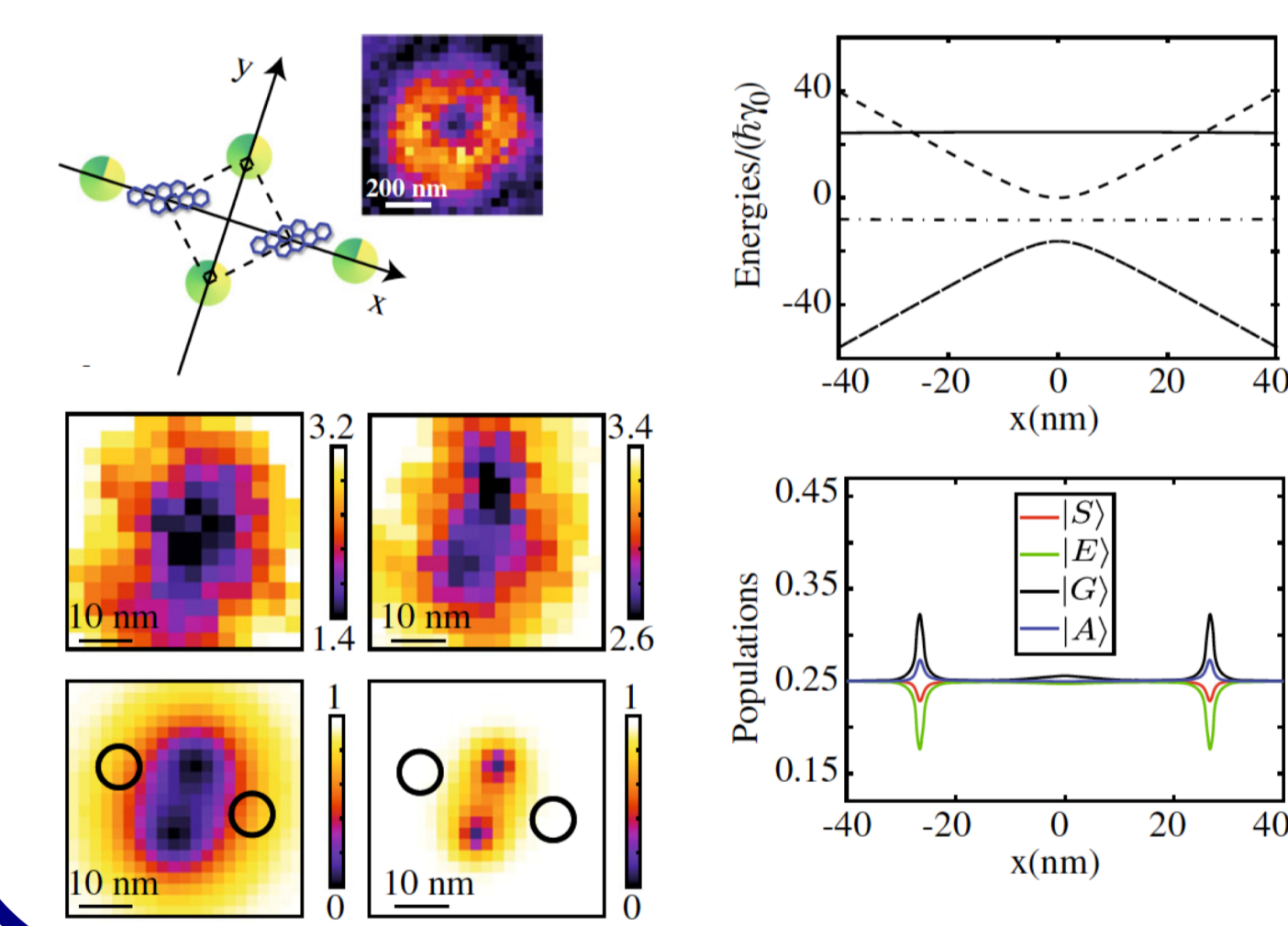
$$\begin{aligned}
 H = & \hbar\omega_0(|E\rangle\langle E| + |G\rangle\langle G|) + \hbar\lambda(|+\rangle\langle +| - |-\rangle\langle -|) \\
 & - \frac{\hbar}{2} \{ (\alpha\Omega_1 + \beta\Omega_2)|E\rangle\langle +| + (\beta\Omega_1 + \alpha\Omega_2)|E\rangle\langle -| \} e^{i\omega_L t} + \\
 & \{ (\alpha\Omega_2 - \beta\Omega_1)|E\rangle\langle -| - (\beta\Omega_2 - \alpha\Omega_1)|E\rangle\langle +| \} e^{i\omega_L t} + h.c.
 \end{aligned}$$

Gaussian laser spot centered between the molecules:
 - Generation of in-phase dipole oscillations with $\Omega_1 = \Omega_2$.
 When Δ 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.

Doughnut-shaped laser spot centered between the molecules:
 - Opposite-phase dipole oscillations, $\Omega_1 = -\Omega_2$.
 When Δ approaches zero ($\alpha \approx \beta \approx 1/\sqrt{2}$):
 - Excitation of the subradiant state is enhanced, while that of the superradiant state is forbidden.

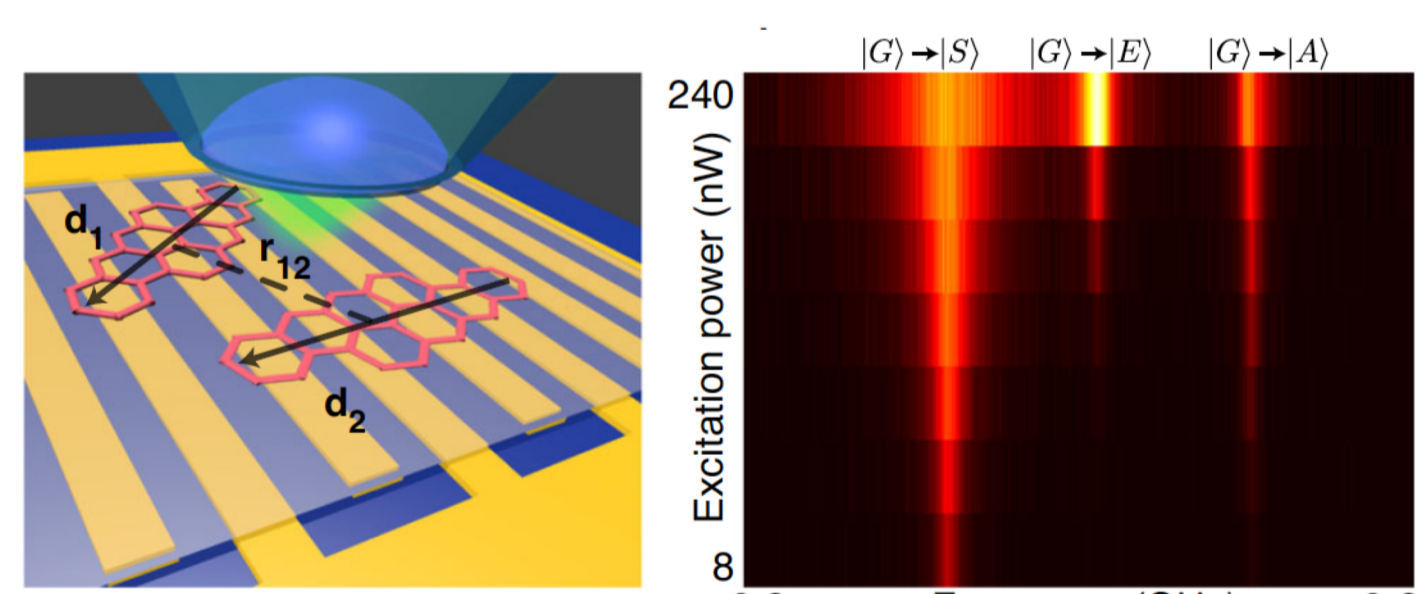
ESSAT images of coupled molecules :

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- The ESSAT images of coupled molecules do not show fluorescence dips at the molecules positions
- When the donut is scanned, the two molecules experience a differential lightshift modifying their energy levels.
- The fluorescence dips appear where two energy levels become degenerated. This causes a destructive interference between two excitation paths leading to the fluorescence dips
- By comparing the experimental images with the simulation, we can get access to the positions of the two molecules (circles)

Two coherently coupled molecules



dipole-dipole interaction :

$$V_{12} = \frac{3\sqrt{\Gamma_1\Gamma_2}}{4(k_0r_{12})^3} [(\vec{d}_1 \cdot \vec{d}_2) - 3(\vec{d}_1 \cdot \vec{r}_{12})(\vec{d}_2 \cdot \vec{r}_{12})]$$

In the rotating frame :

$$H_0 = \begin{pmatrix} \Delta/2 & 0 & 0 & 0 \\ 0 & \delta/2 & V_{12} & 0 \\ 0 & V_{12}^* & -\delta/2 & 0 \\ 0 & 0 & 0 & -\Delta/2 \end{pmatrix}$$

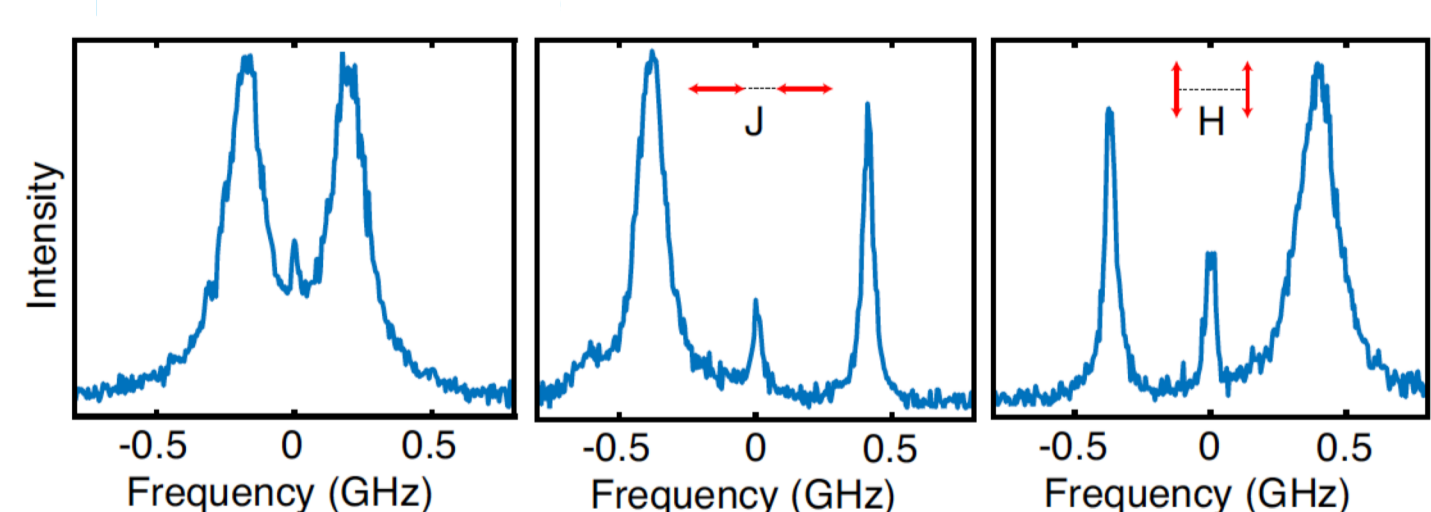
With $\Delta = \hbar(\omega_1 + \omega_2 - 2\omega_{laser})$

The diagonalization of the Hamiltonian gives :

$$\tan 2\theta = \frac{2V_{12}}{\delta}$$

and

$$\begin{aligned}
 |S\rangle &= \cos\theta |e_1g_2\rangle + \sin\theta |e_1e_2\rangle \\
 |A\rangle &= -\sin\theta |e_1g_2\rangle + \cos\theta |g_1e_2\rangle
 \end{aligned}$$



States population dynamics

The dynamics of the system is given by the master equation :

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [(H_0 + H_{laser}), \rho] - \frac{1}{2} \sum_{i,j=1,2} \Gamma_{ij} (S_i^+ S_j^- \rho + \rho S_i^+ S_j^- - 2S_j^- \rho S_i^+)$$

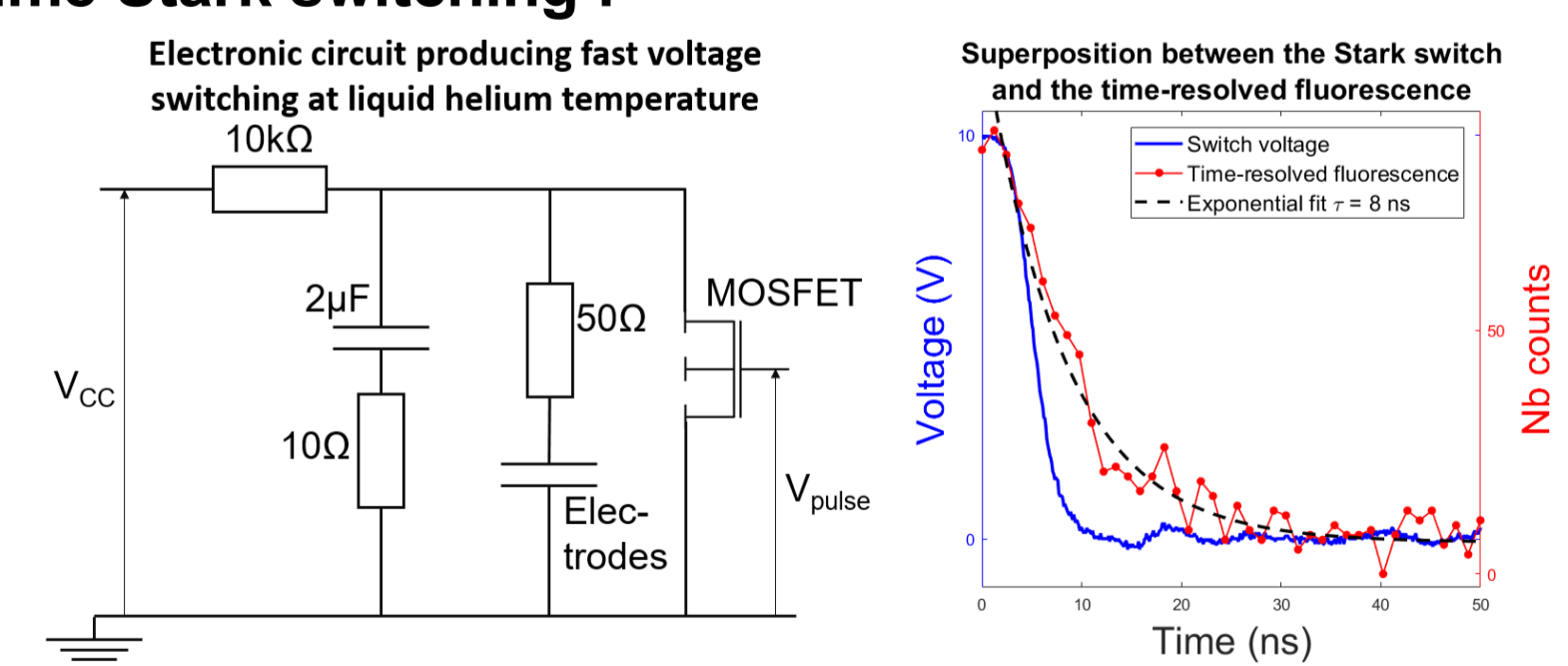
- For two identical molecules ($\Gamma_1 = \Gamma_2 = \Gamma_0$; $\vec{d}_1 // \vec{d}_2$ and $\omega_1 = \omega_2$), a maximal entanglement gives rise to a bright and a dark state :

$$\begin{aligned}
 \Gamma_+ &= \sin^2(\theta)\Gamma_1 + \cos^2(\theta)\Gamma_2 + \sin(2\theta)\Gamma_{12} \\
 \Gamma_- &= \cos^2(\theta)\Gamma_1 + \sin^2(\theta)\Gamma_2 - \sin(2\theta)\Gamma_{12} \\
 \Gamma_{12} &= \Gamma_{21} = \sqrt{\Gamma_1\Gamma_2}(\vec{d}_1 \cdot \vec{d}_2)
 \end{aligned}$$

Perspectives

Creation of quantum logic gates by sub-lifetime Stark switching :

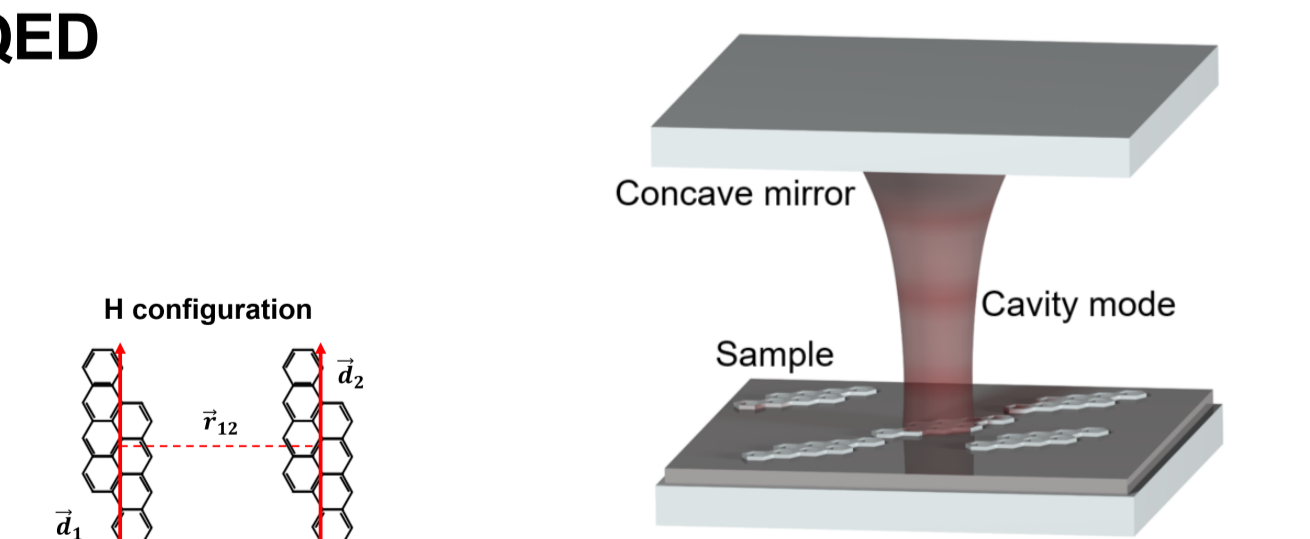
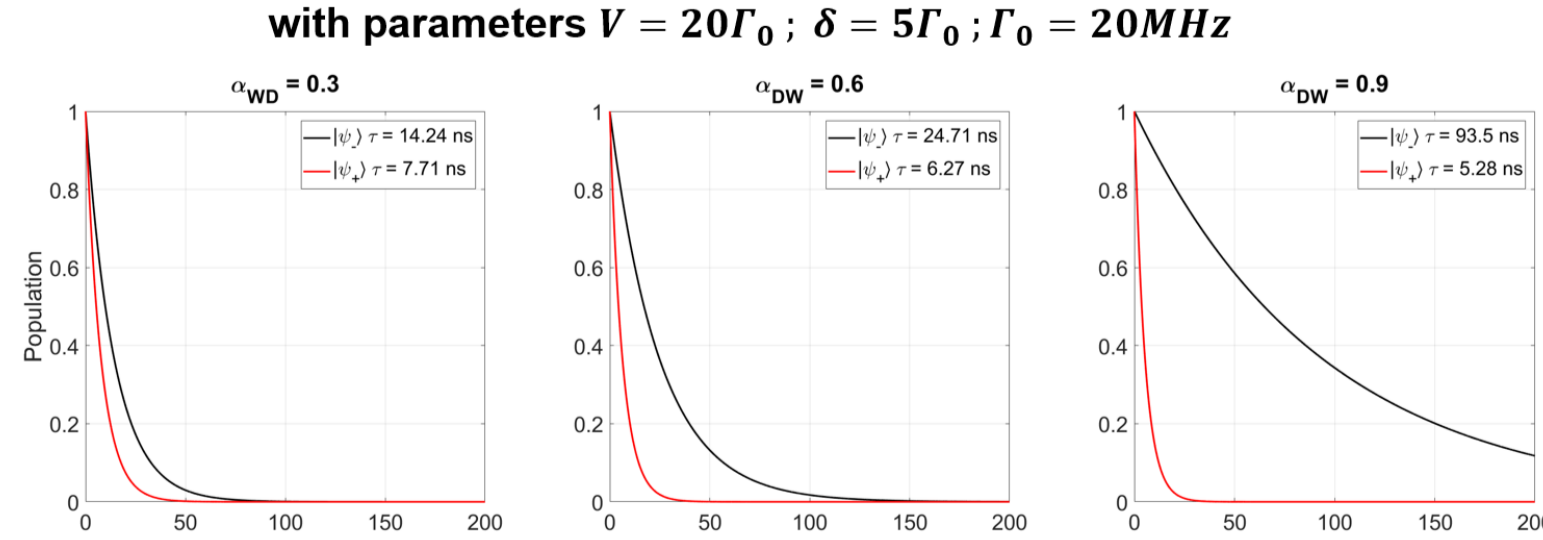
- Controlling the entanglement in the sub-lifetime range is a step for the creation of a quantum logic gate
- Fast switching on a DBATT molecule in 2,3-dibromonaphthalene (enhanced Stark effect $300 \text{ MHz} \cdot \text{V}^{-1}$). It is at resonance with laser excitation for $V=10$ Volts, out of resonance for $V=0$ Volts :
 \rightarrow Fluorescence decay slower than the voltage's one
 \rightarrow Validates the feasibility of sub-lifetime Stark switching



- A characteristic time of 8 ns for the decay is consistent with the lifetime of the molecule trapped in this matrix

Increasing the sub- and super-radiance with cavity QED

Lifetime simulations for coupled dipoles in H configuration with parameters $V = 20\Gamma_0$; $\delta = 5\Gamma_0$; $\Gamma_0 = 20\text{MHz}$



- Coupling the Zero Phonon Line to a cavity allows us to increase the Debye-Waller factor

References

[1] Hettich, C. « Nanometer Resolution and Coherent Optical Dipole Coupling of Two Individual Molecules ». *Science* 298, n° 5592 (October 11th 2002): 385-89.

[2] Yang, B., J.-B. Trebbia, R. Baby, Ph. Tamarat, et B. Lounis. « Optical Nanoscopy with Excited State Saturation at Liquid Helium Temperatures ». *Nature Photonics* 9, n° 10 (octobre 2015): 658-62.