Quantum Optics with single Nano-Objects

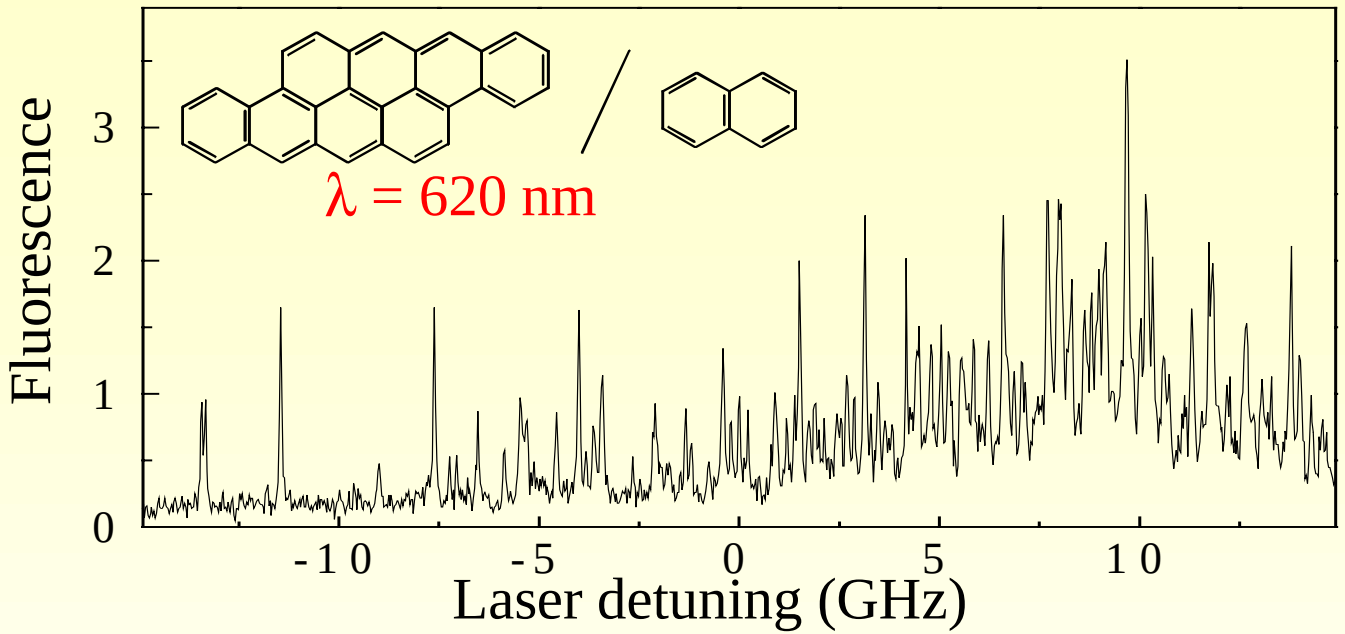
Outline:

- Introduction : nonlinear optics with single molecule
- Single Photon sources
- Photon antibunching in single quantum dot fluorescence
- Conclusion

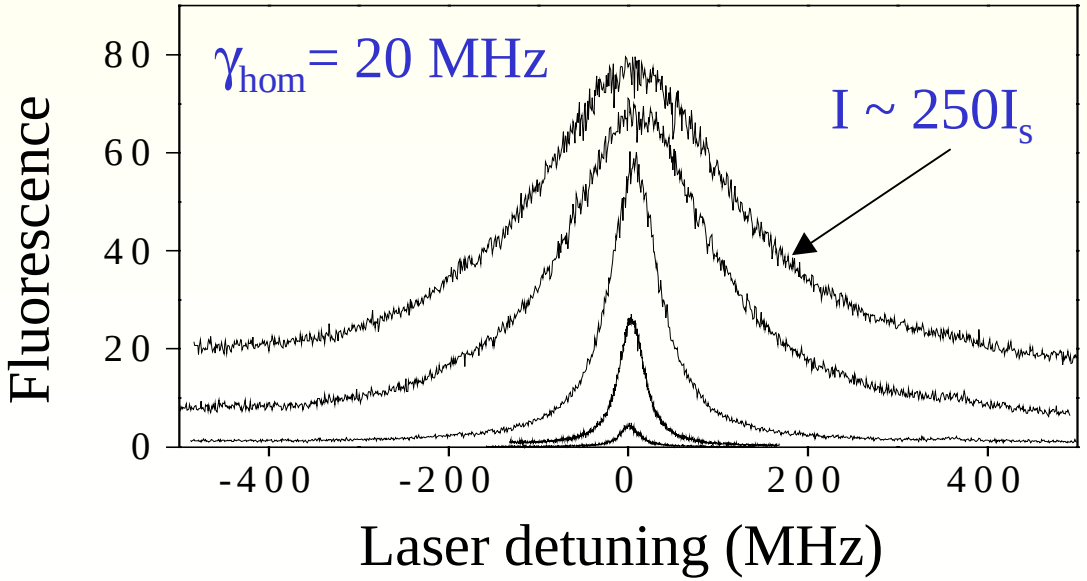
ADVANTAGES

- No ensemble averaging
- Statistical correlations
- Extreme sensitivity to immediate local nanoenvironment
- No synchronisation needed, time evolution
- Single quantum system

Inhomogenous spectrum at 2 K



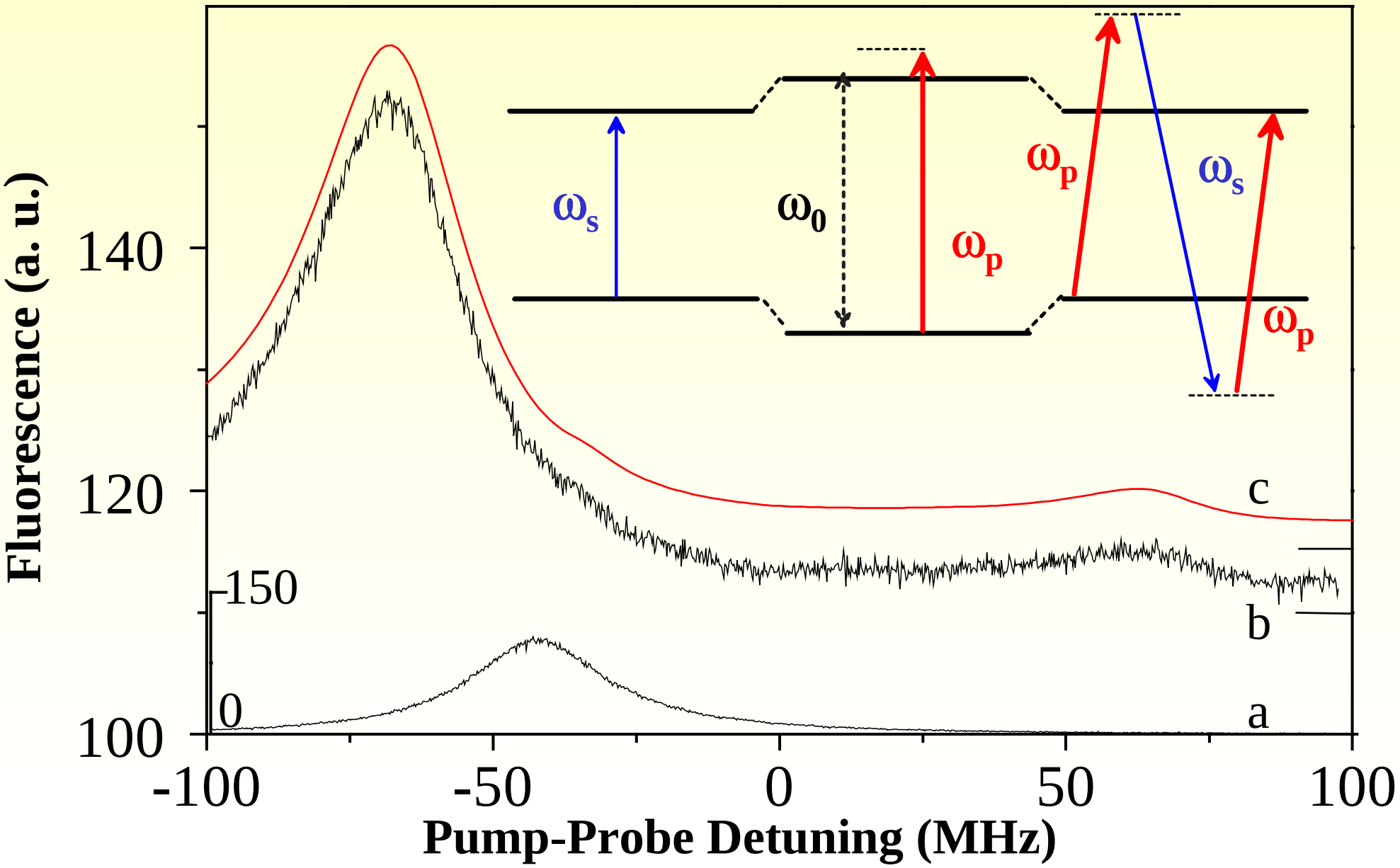
Saturation of a SM line

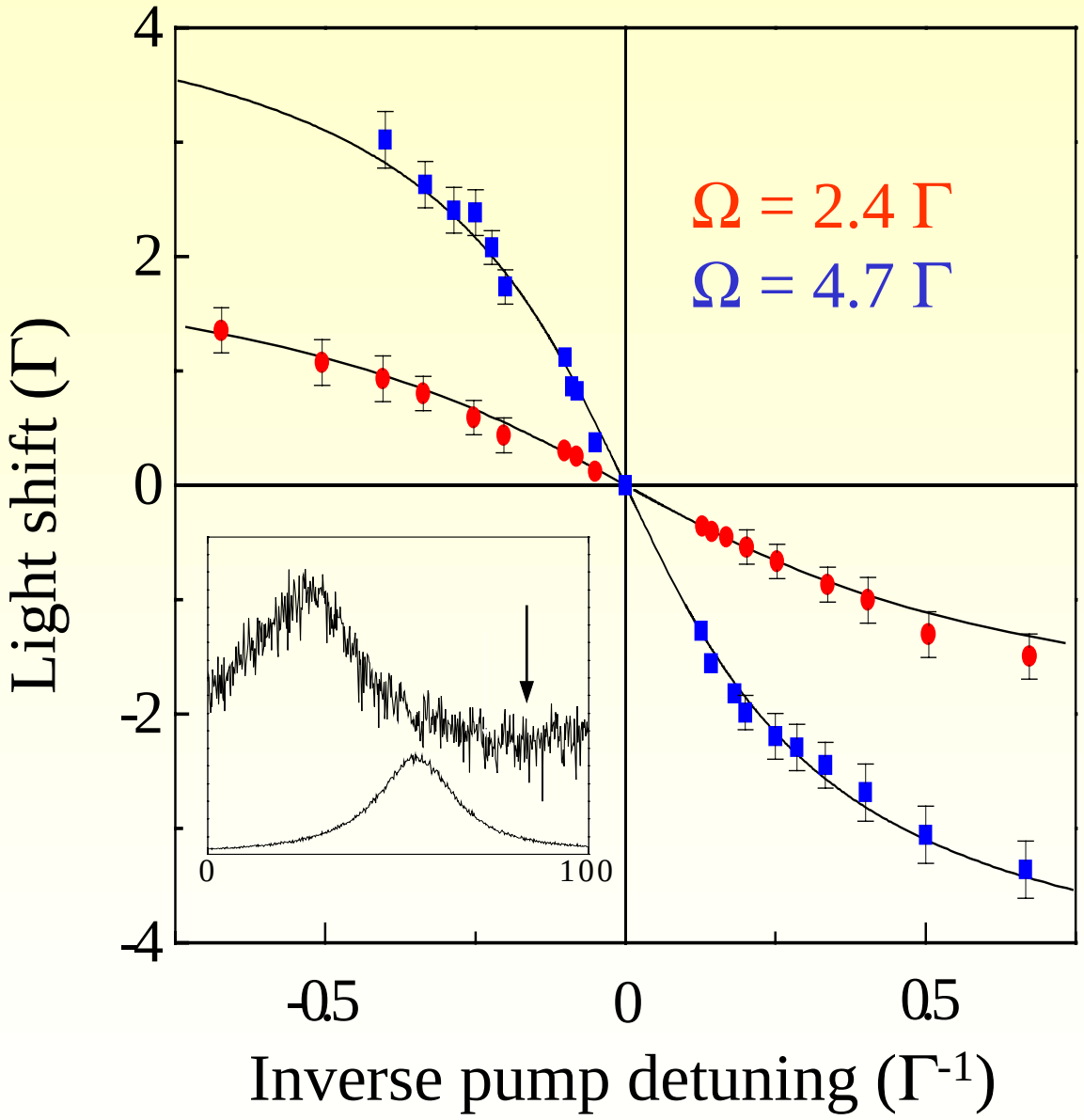


$$S = S_{\infty} \frac{I/I_s}{1 + I/I_s}$$

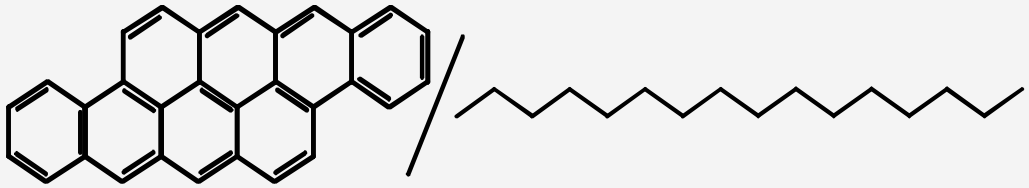
Pump-probe experiments:

- Light Shift
- Hyper-Raman Resonance

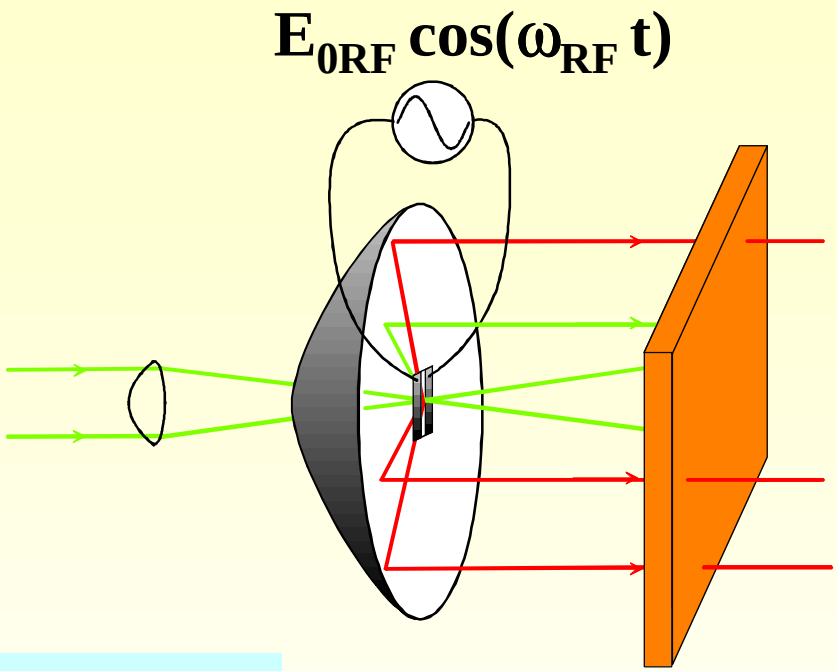



Ω = 2.4 Γ
Ω = 4.7 Γ
Light shift (Γ)
Inverse pump detuning (Γ-1)
4
2
0
-2
-4
-0.5
0
0.5
0
100

Electro-Optical Effects



Linear Stark Shift: $\Delta\mu \sim 0.3$ D



Linear Coupling with the RF field

$$\hbar\Omega_{RF} = \Delta\mu \cdot E_{0\,RF}$$

- 1st case: High RF field, low laser intensity

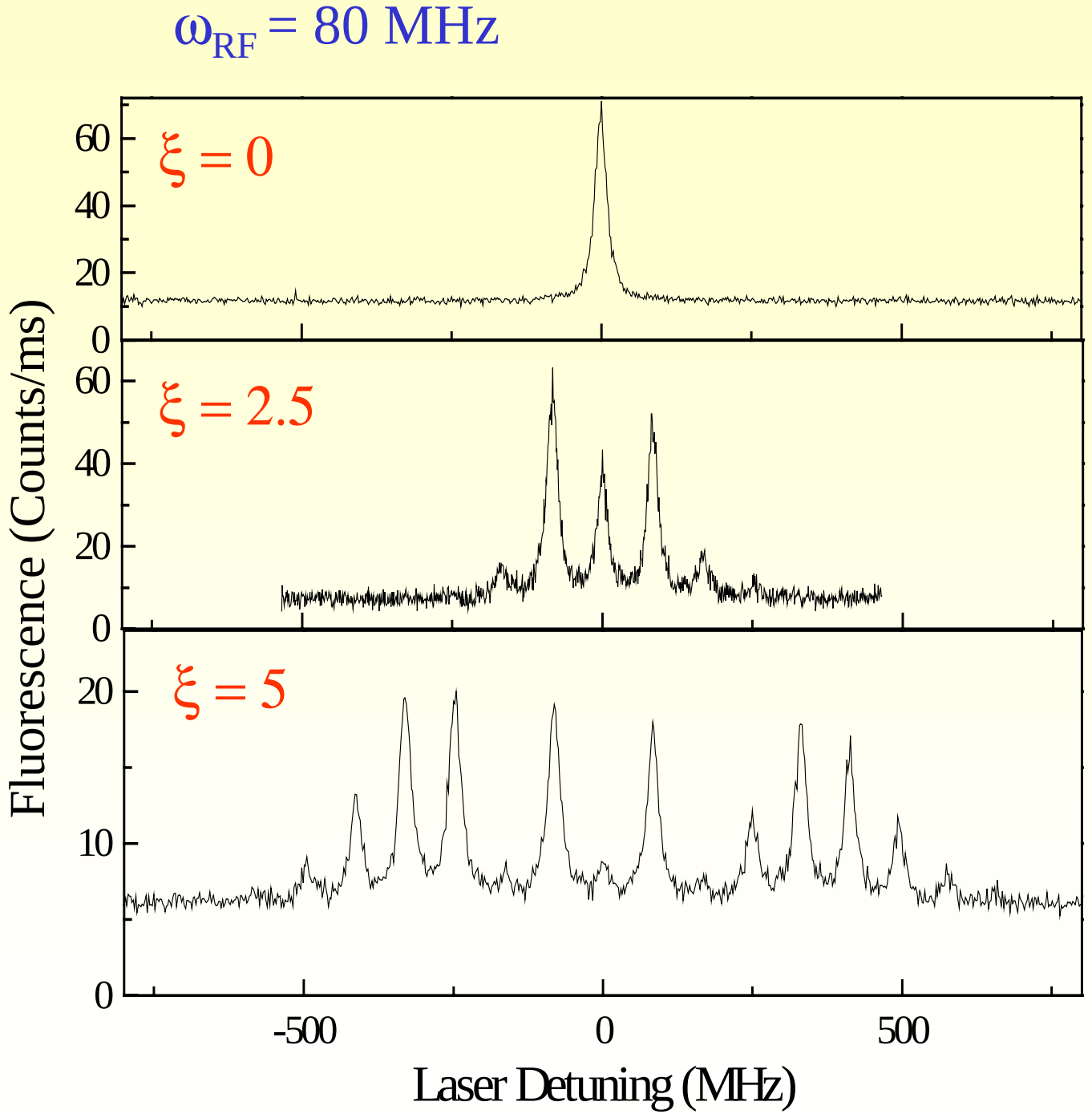
Modulation of the molecular transition frequency

⇕

Modulation of the laser frequency

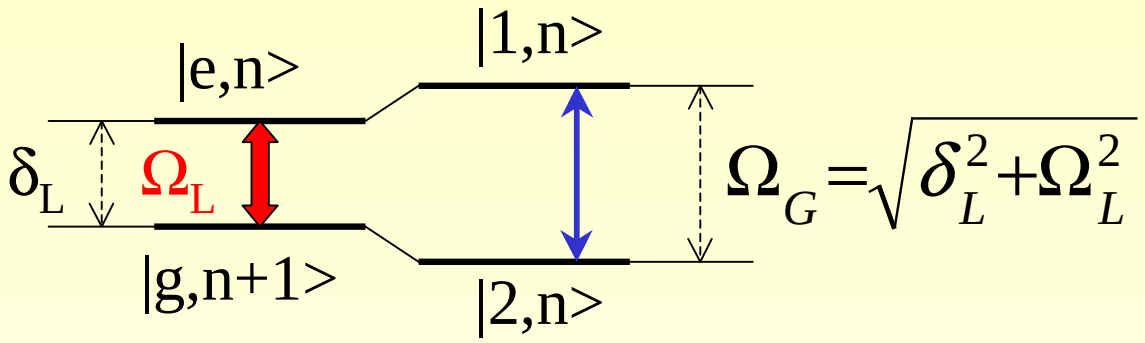
Side Bands

$$J_n\left(\xi = \frac{\Omega_{RF}}{\omega_{RF}}\right)$$


ω_{RF} = 80 MHz
ξ = 0
ξ = 2.5
ξ = 5
Fluorescence (Counts/ms)
Laser Detuning (MHz)
60
40
20
0
60
40
20
0
20
10
0
-500
0
500

2nd case: High laser intensity, low RF field

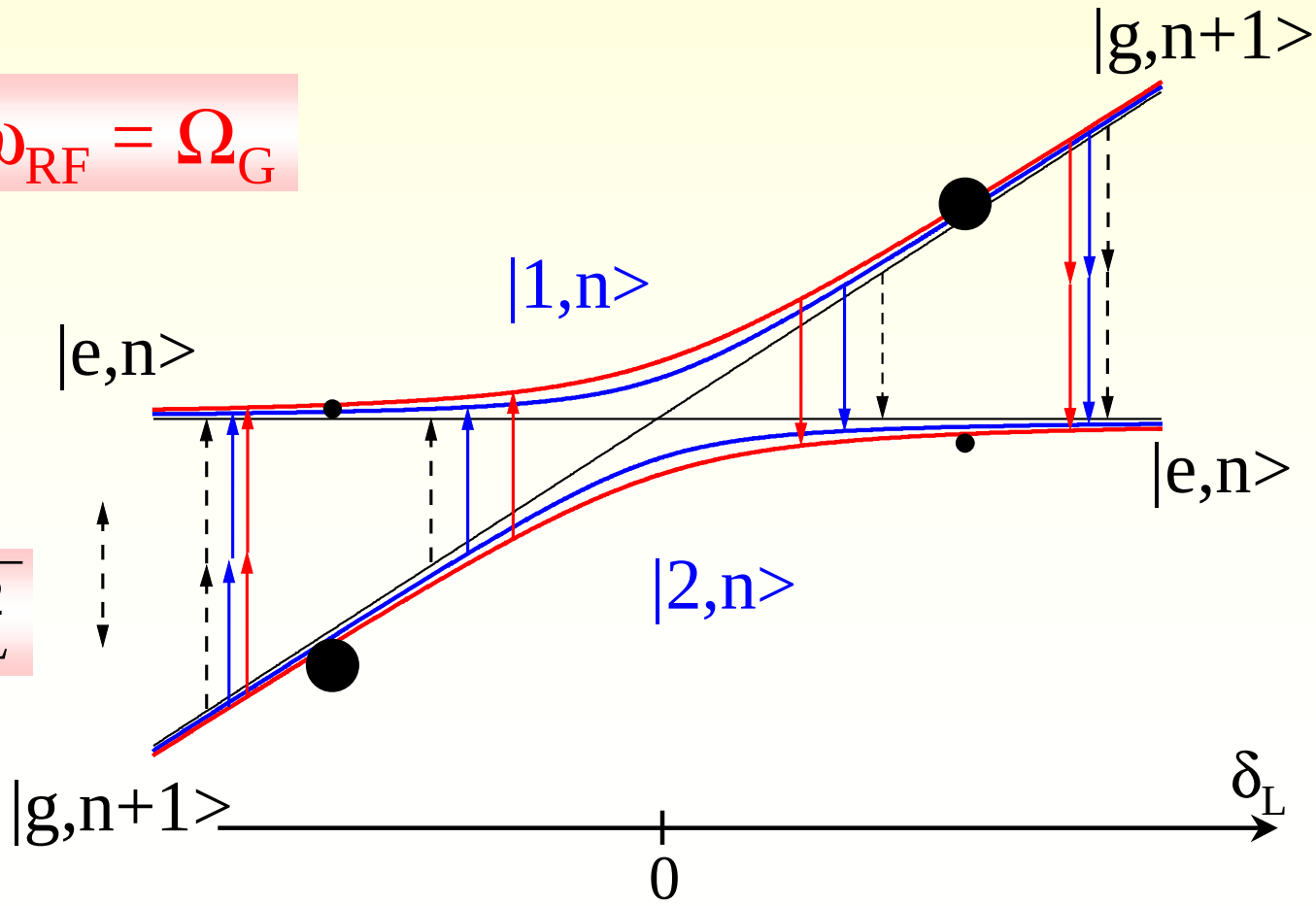
- Molecule dressed by the laser field
- RF coupling between 1,n> and |2,n> states



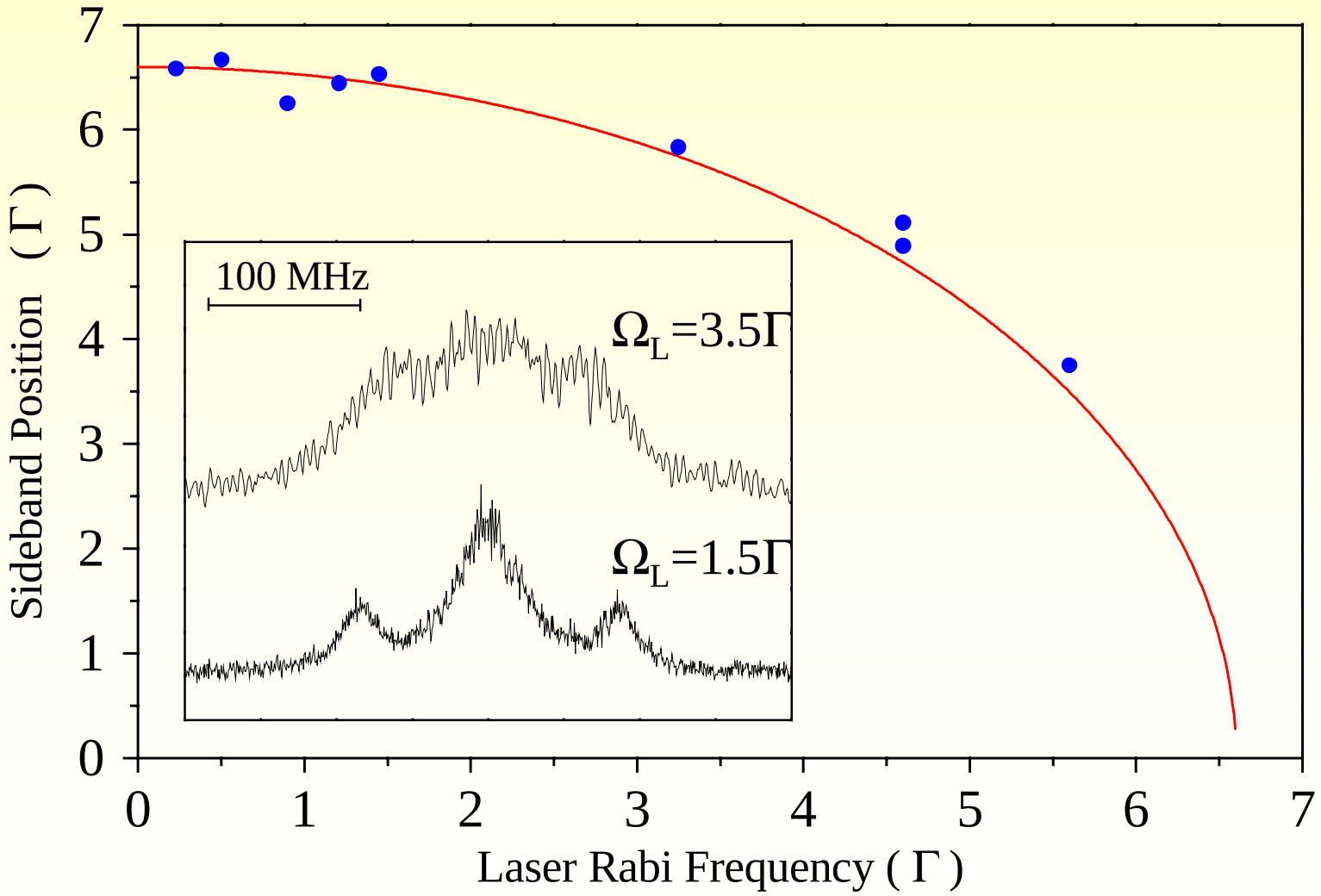
- Rabi Resonance: $\omega_{RF} = \Omega_G$

fixed ω_{RF}, tuned δ_L

$$\delta_{res} = \pm\sqrt{\omega_{RF}^2 - \Omega_L^2}$$



Shift of the Rabi resonance function of the laser field amplitude



Triggered Single Photon Sources

Practical need for Single Photons:

Quantum Cryptography:

* Quantum mechanics provide unconditional security for communication

* Encoding information on the polarization of single photons

Quantum computing:

Quantum logic gates based on single photons have been demonstrated

Present day sources of single photons

- Correlated photon pairs
  - * Atomic cascade
  - * Parametric down conversion
- Highly attenuated laser (or LED) pulses

Problems:
- Random time generation
- Average photon's number <<1

Present day sources of single photons

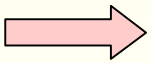
Use of a single quantum system

Yamamoto's group experiment:

- Coulomb Blockade in a mesoscopic double barrier p-n junction
- temperature 50 mK (dilution cryostat)
- low detection efficiency ~10^{-4}
- low e-h recombination rate

Photon antibunching in single molecule fluorescence

Controlled excitation of a single molecule



Deterministic generation of single photons

Excitation:
Rapid adiabatic passage

Two conditions:

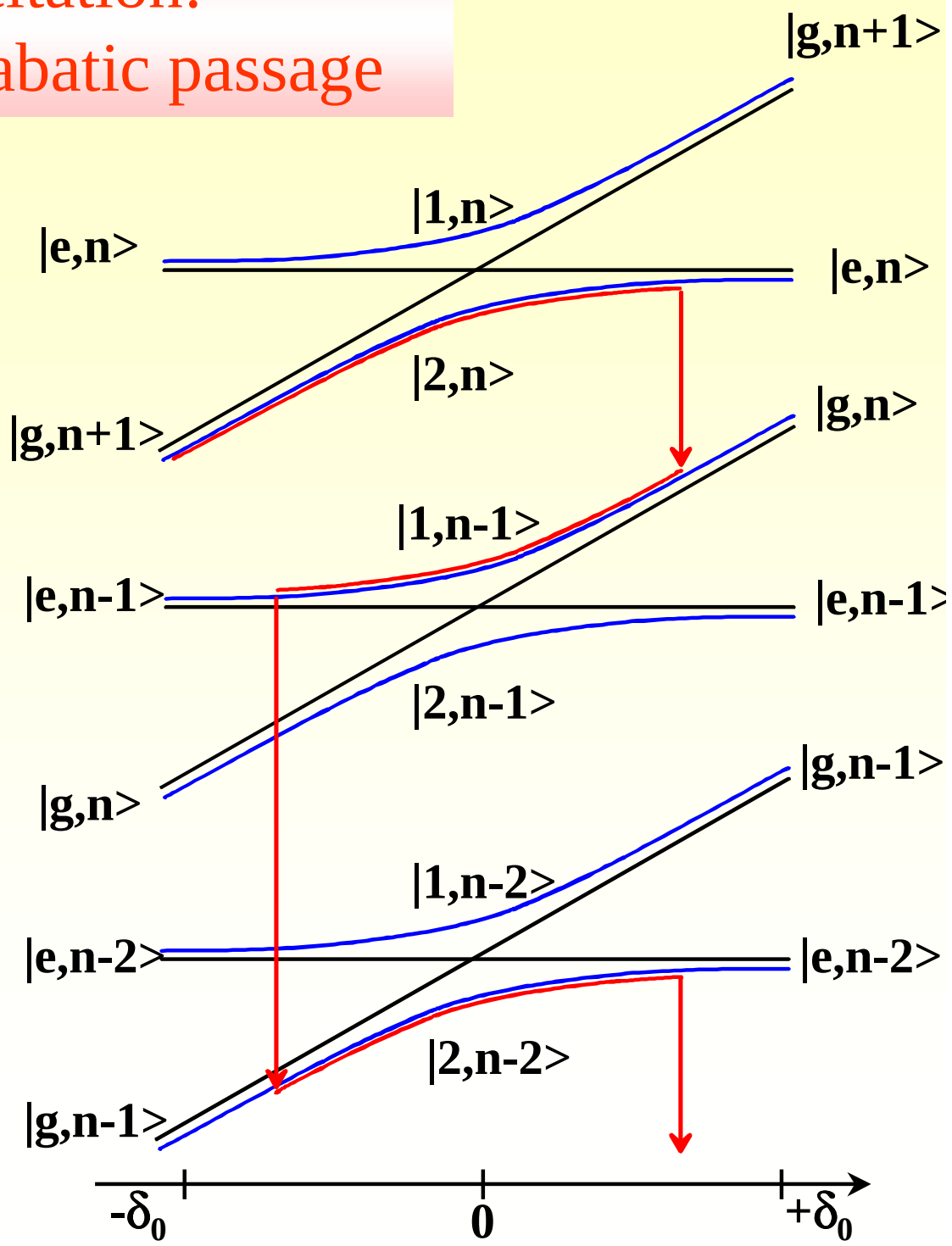
Adiabatic passage

$T_{pass} > T_{Rabi}$

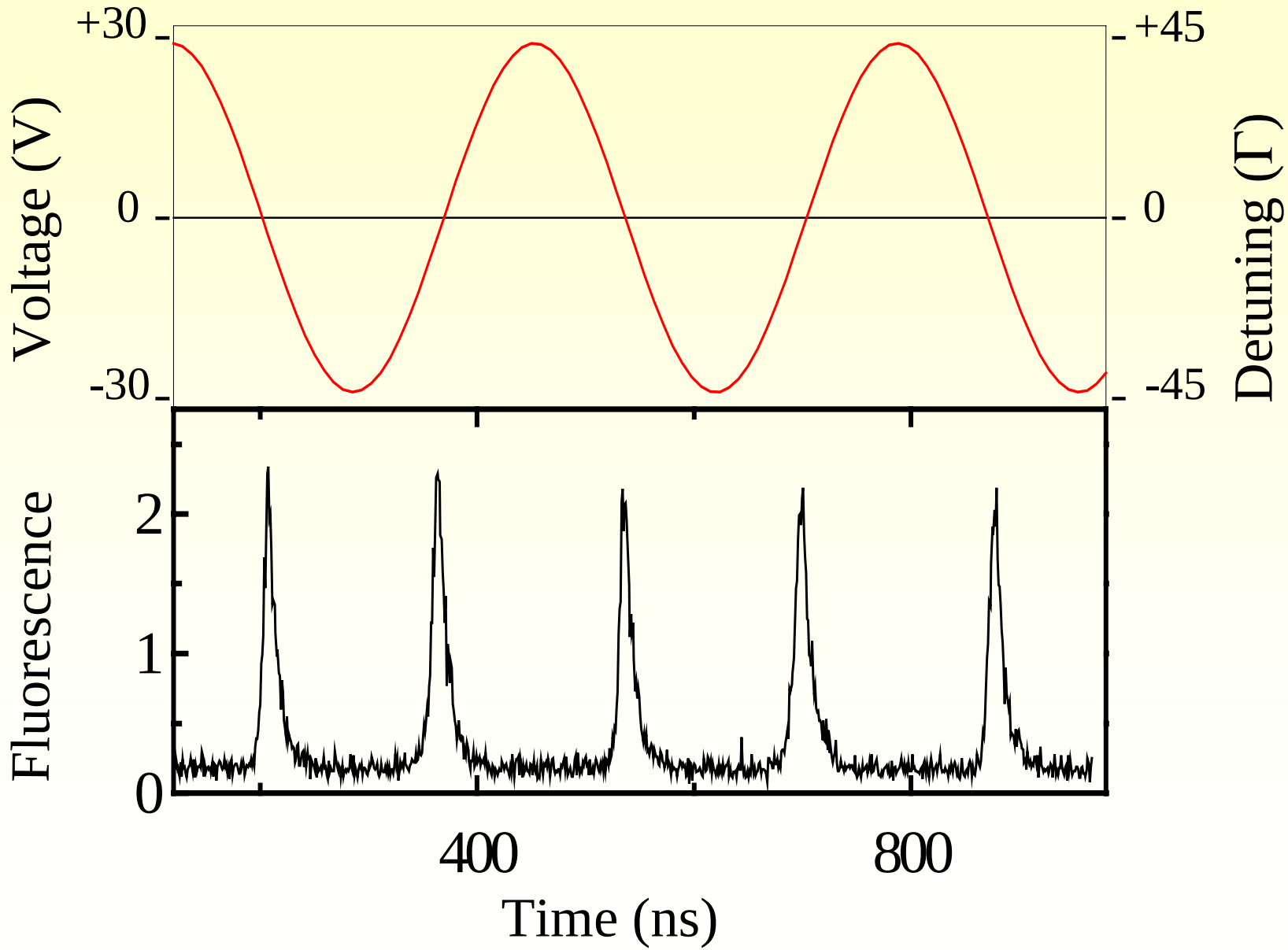
Rapid passage

$T_{pass} << \tau_f$

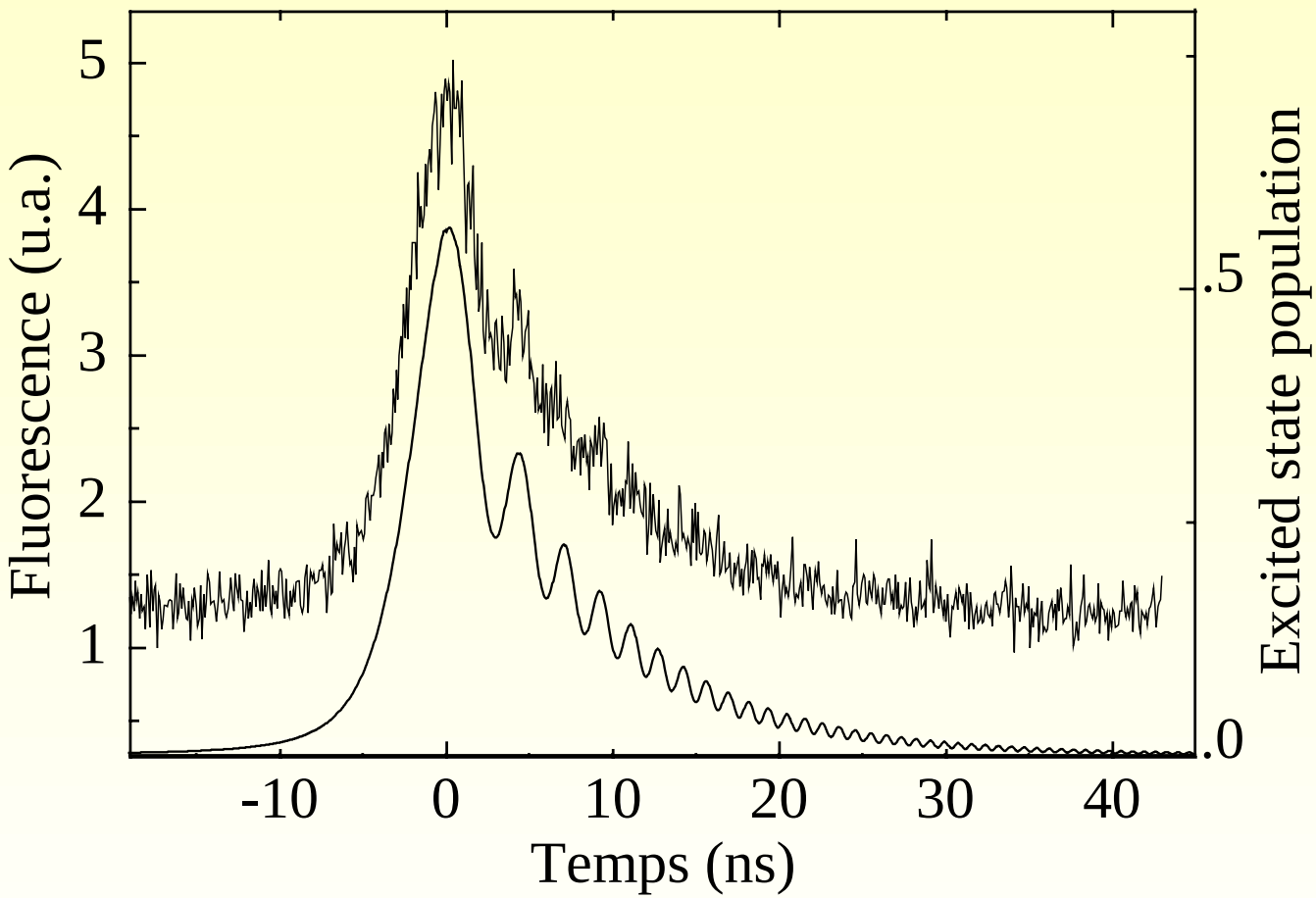
$\Omega_L >> \Gamma$



ν_{RF} = 3 MHz, Ω_L = 2.6 Γ

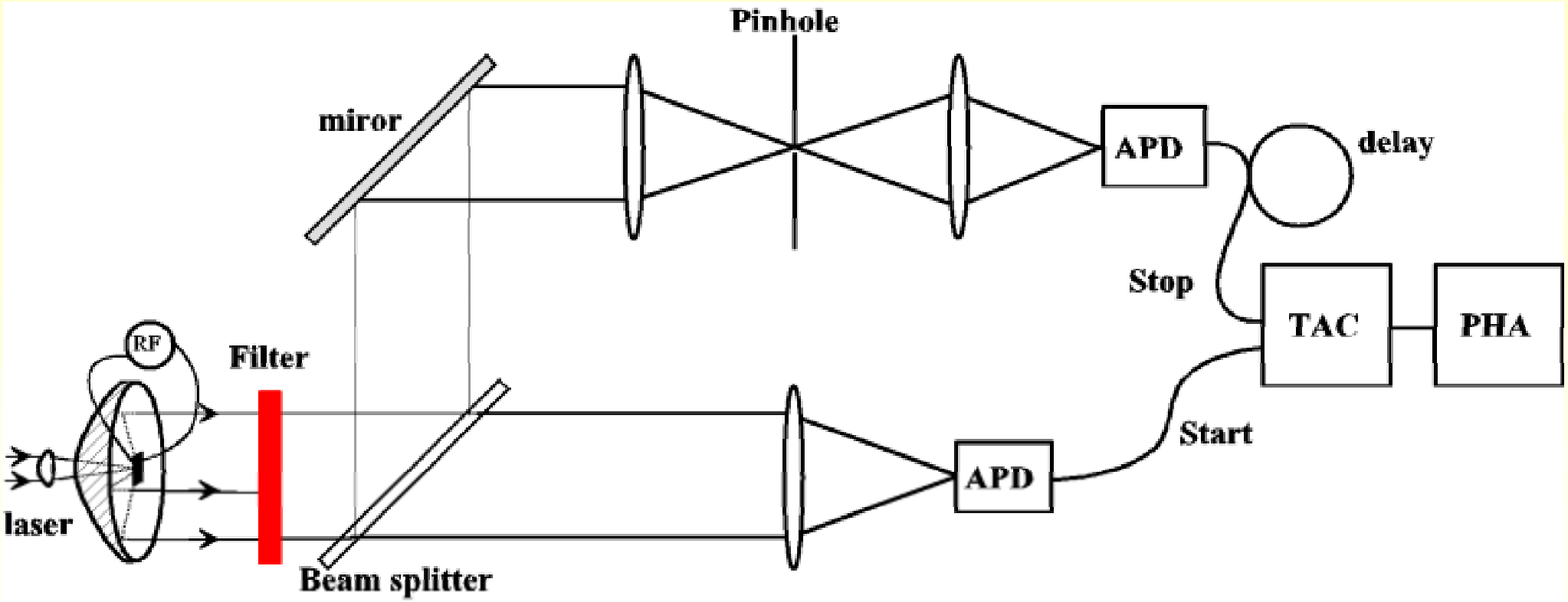


Detailed shape of a fluorescence burst



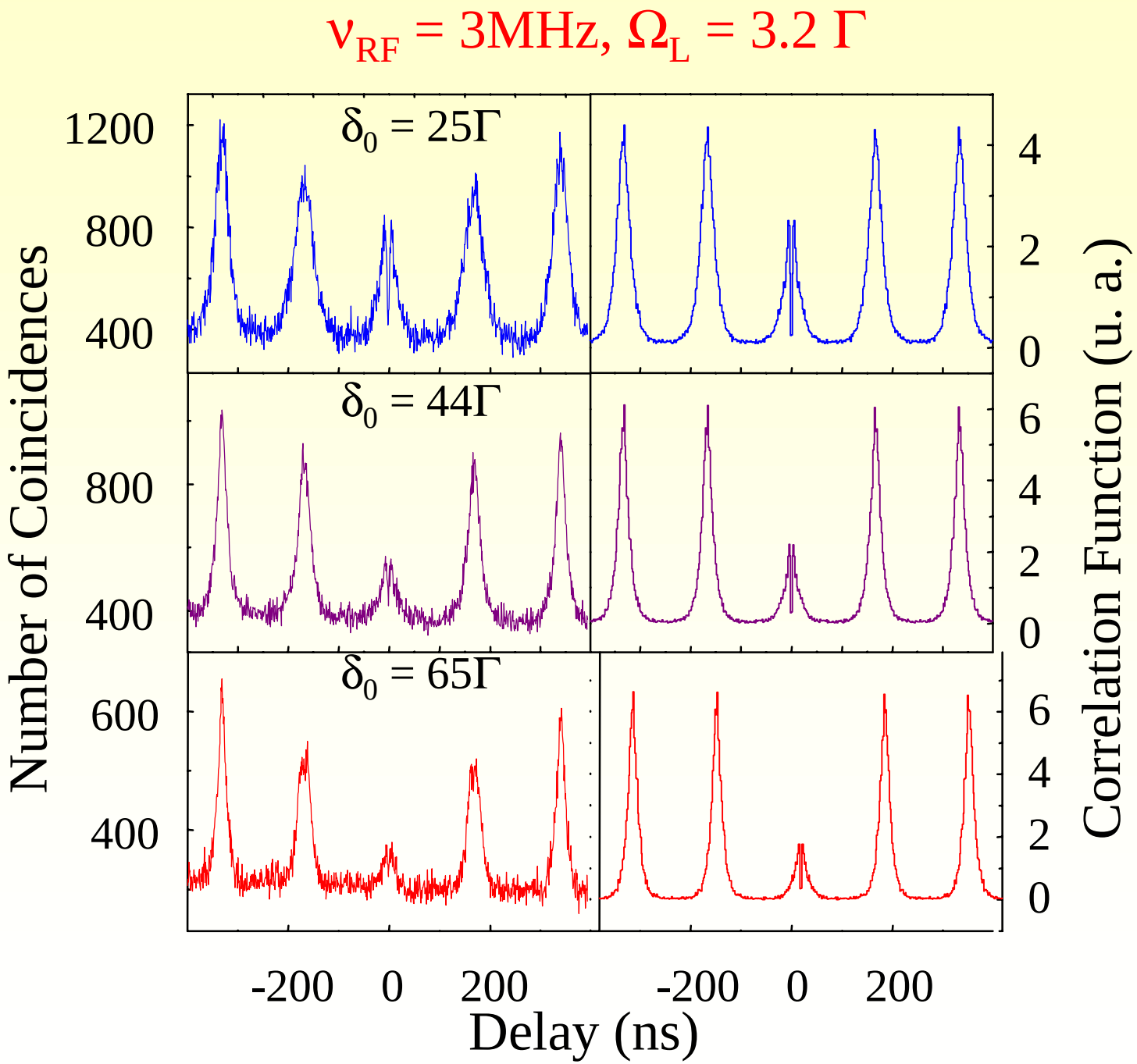
- **T = 250 ns, $\Omega = 3\,\Gamma$, $\delta_0 = 80\,\Gamma$**
- **Short rise time**
- **Relaxation time ($\Gamma^{-1} \approx 8$ ns)**
- **Oscillations**

How many emitted photons per sweep?

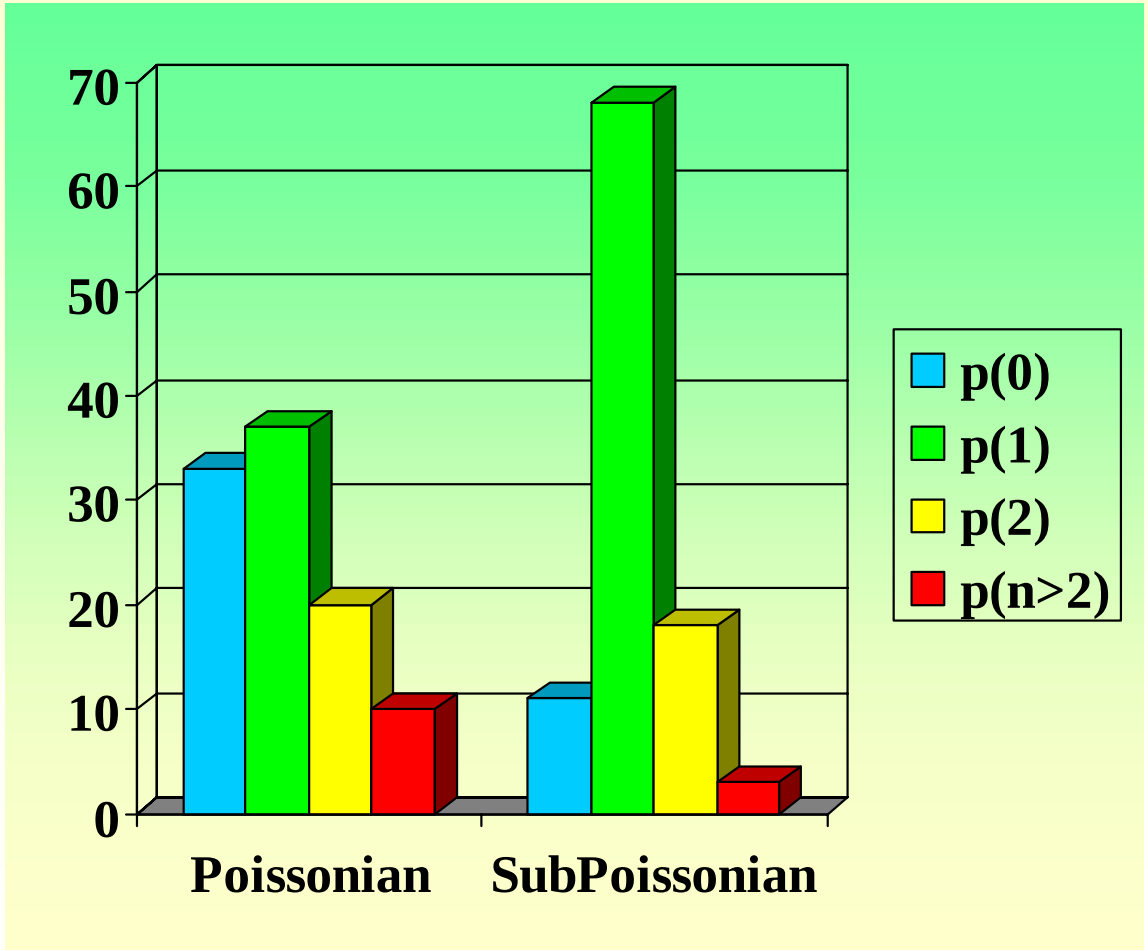


Measurement of the autocorrelation function $g^{(2)}(\tau)$
Comparaison with Q.M.C. simulations

Histogram of time delays:



Comparaison with a Coherent source



- ν = 6 MHz, $\delta_0 = 44\Gamma$
- $n_{av} = 1.12$
- p(1) = 0.68
- p(n>1) ~ 0.21
- Mandel Parameter

$$Q_{sour.} = -0.65$$

$$Q_{detc.} = -0.006$$

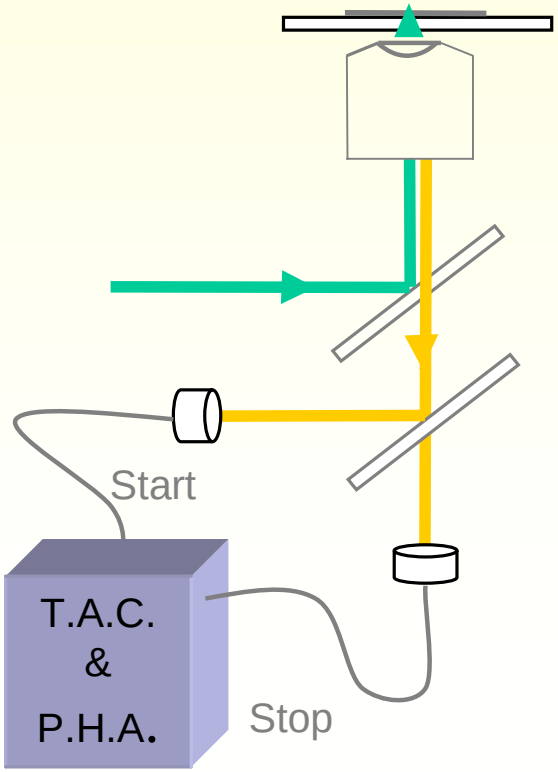
Room Temperature Single photon source

Principe de l'expérience

- Pulsed excitation to a vibrationnaly excite level
- Rapid Relaxation (ps) to the fluorescent state
- Emission of a single photon

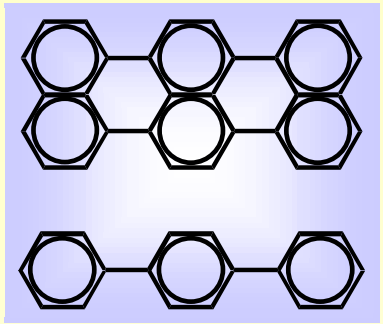
Experimental setup

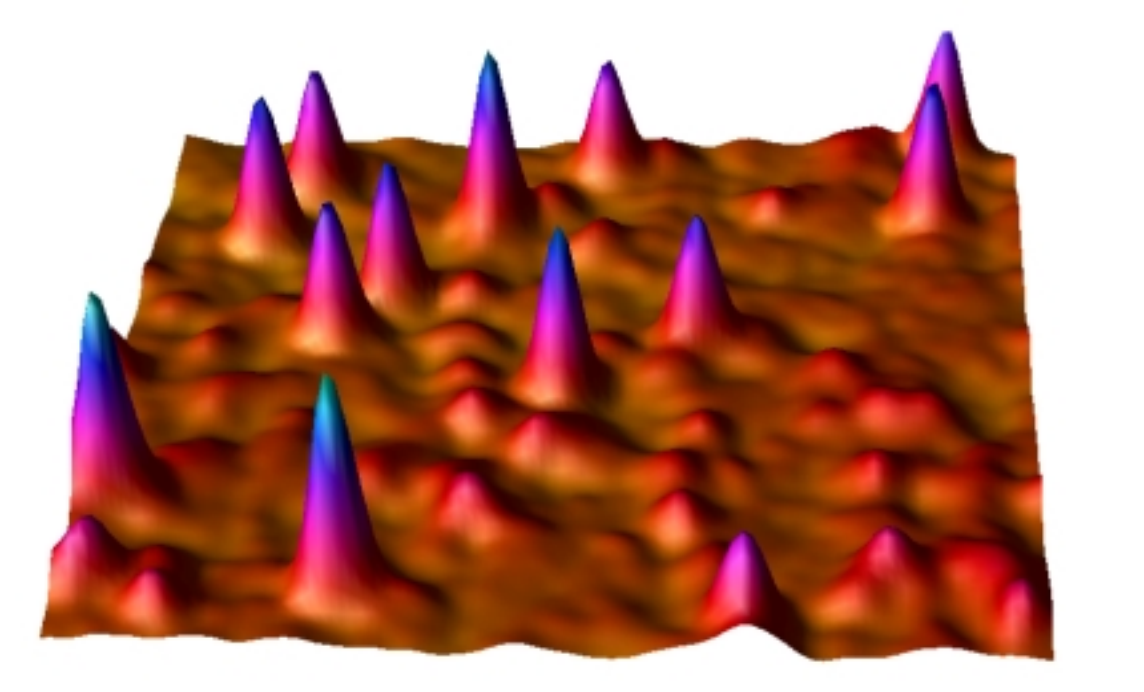
- Inverted Microscope
- Piezo-electric Scanner
- Coincidence Setup
- Detection effeciency 6%



System: **Terrylene in p-terphenyl**

Favorable photophysical parameters and high photostability



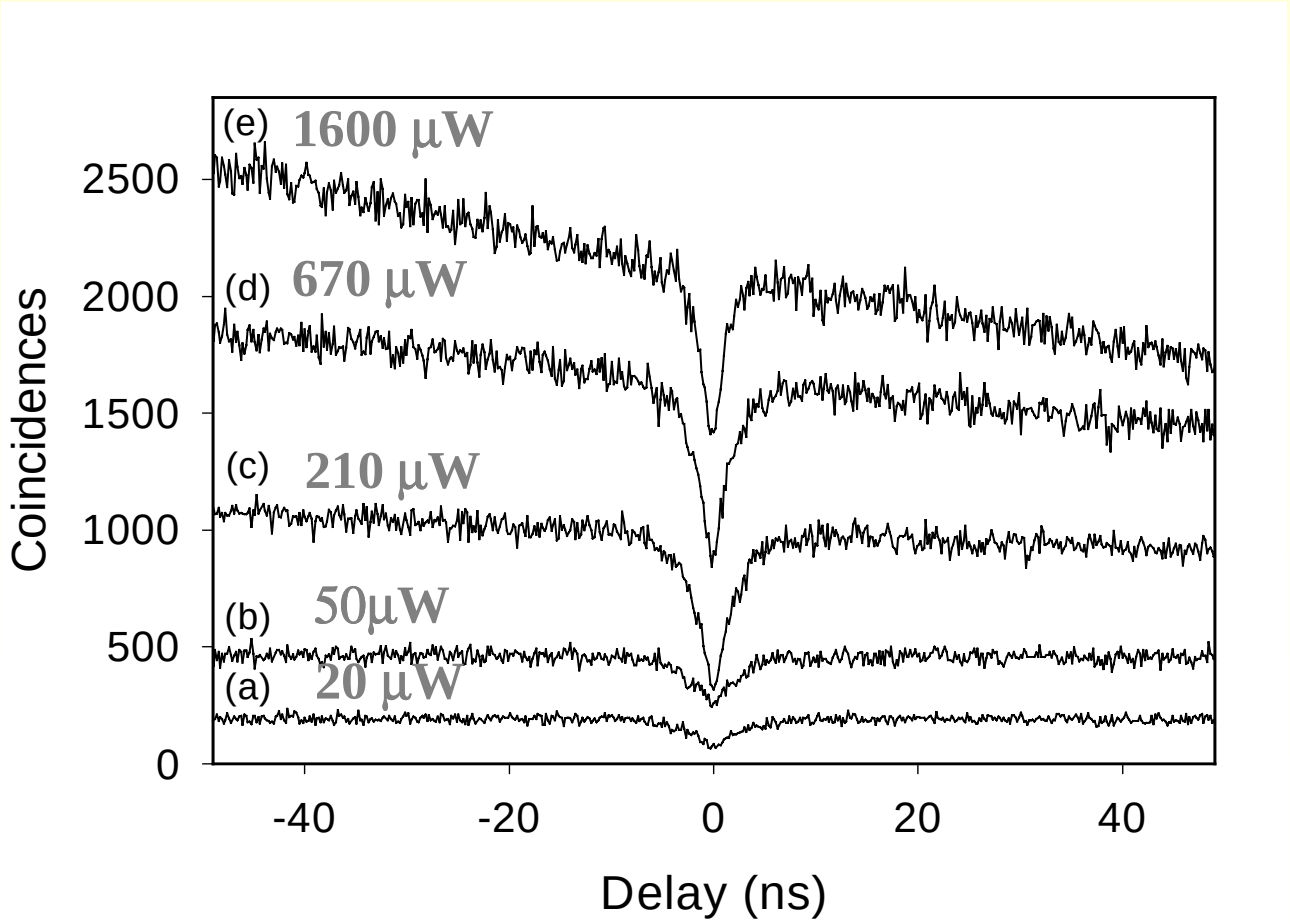


Confocal fluorescence image(10μm*10μm) of single Terrylene molecules

CW Excitation : Photon Antibunching

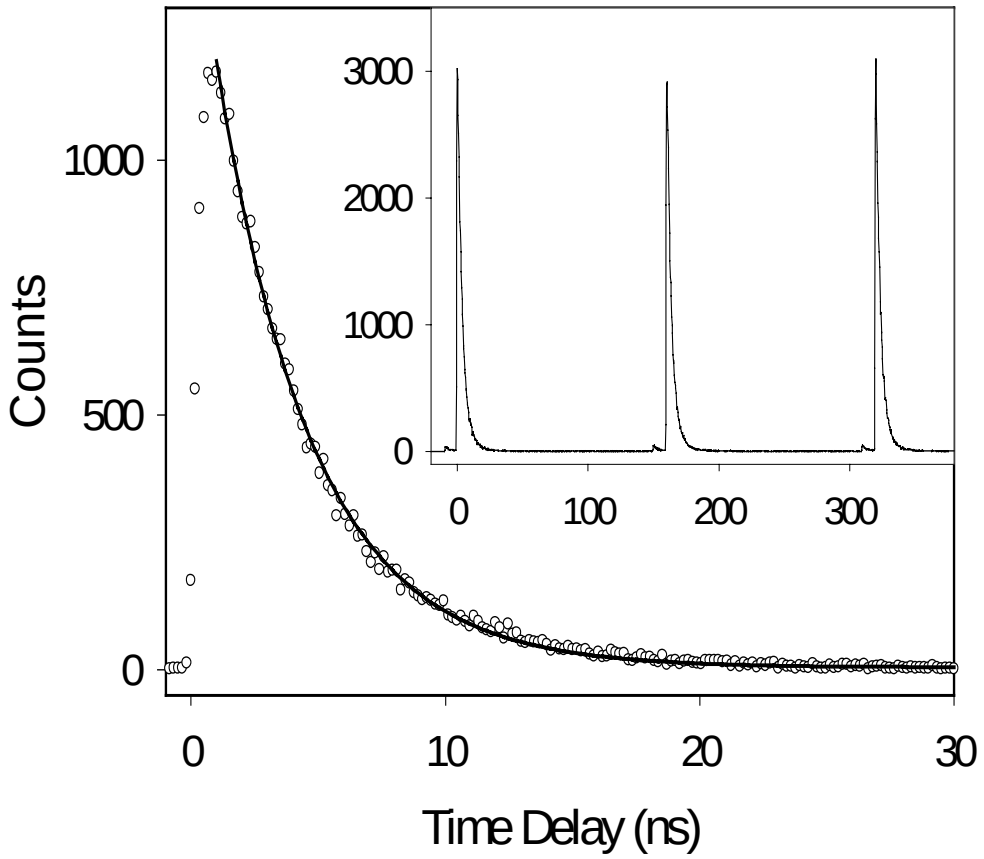
Fluorescence autocorrelation function $g^{(2)}(\tau)$ proportional to the excited state population, $\tau>0$:

$$\Pi(\tau) = \frac{I/I_s}{1 + I/I_s} \cdot \{1 - \exp[-(1 + I/I_s) \cdot \tau/\tau_f]\}$$



Signature of a single molecule emission

Pulsed excitation : Triggered single photon emission

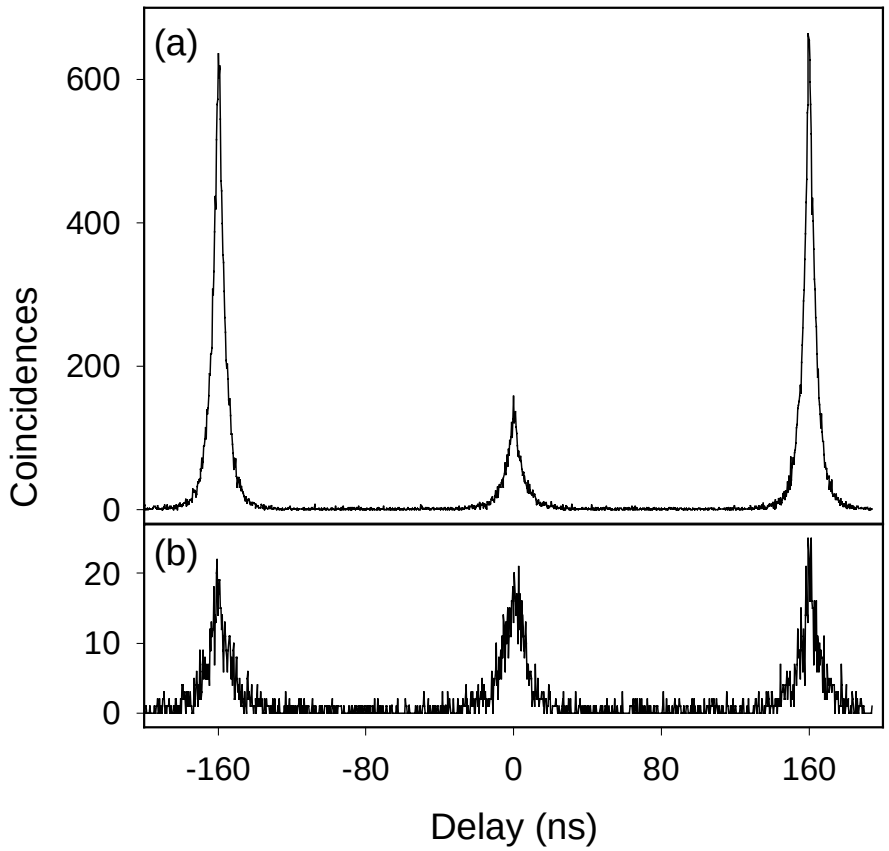


- **M.L. 532 nm laser:**
- **Pulse width: 35ps,**
- **Repetition rate: ν = 6.25 MHz**

- **Single exponential decay**
- **fluorescence lifetime:**

$$\tau_f = 3.8 \text{ ns}$$

Fluorescence autocorrelation Function



Laser spot positioned

- **on a single molecule (Signal/Background~ 6)**

(b) Away from any molecule (background coherent emission)

central peak area / lateral peak area

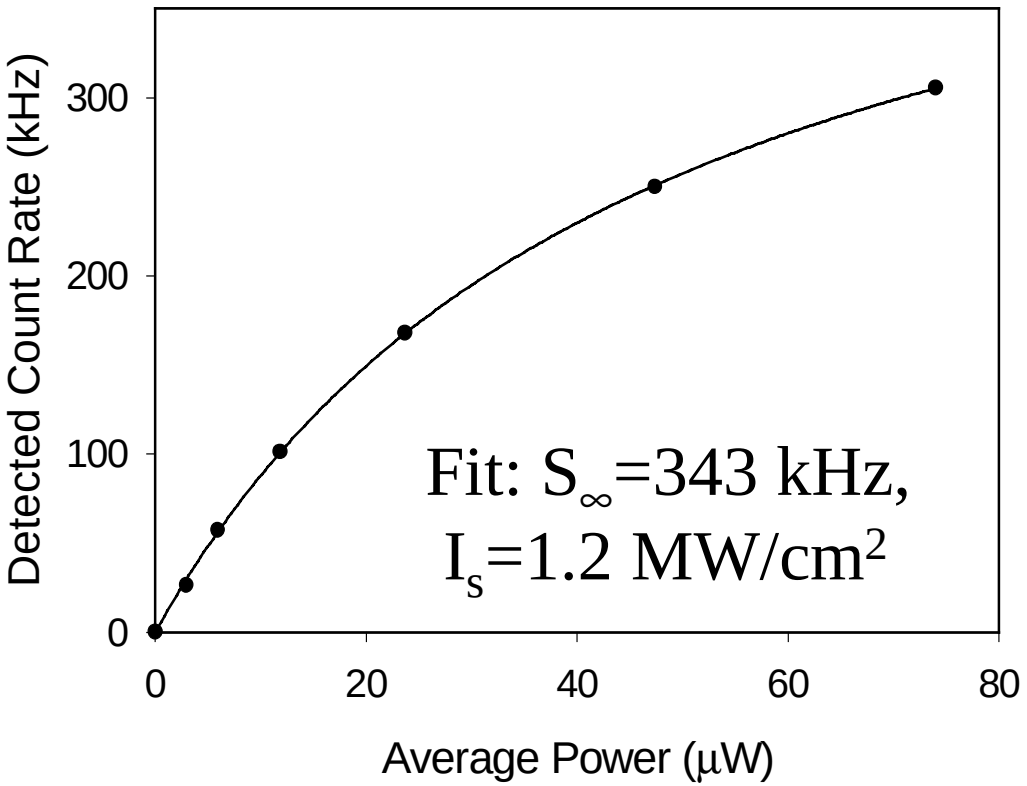
$= (B^2+2BS) / (B+S)^2 \sim 0.27$

Saturation du taux d'émission

Short laser pulse :
$\tau_p << \tau_f$

$p(2) \sim 0$
$p(1) = \Pi(\tau=\tau_p)$

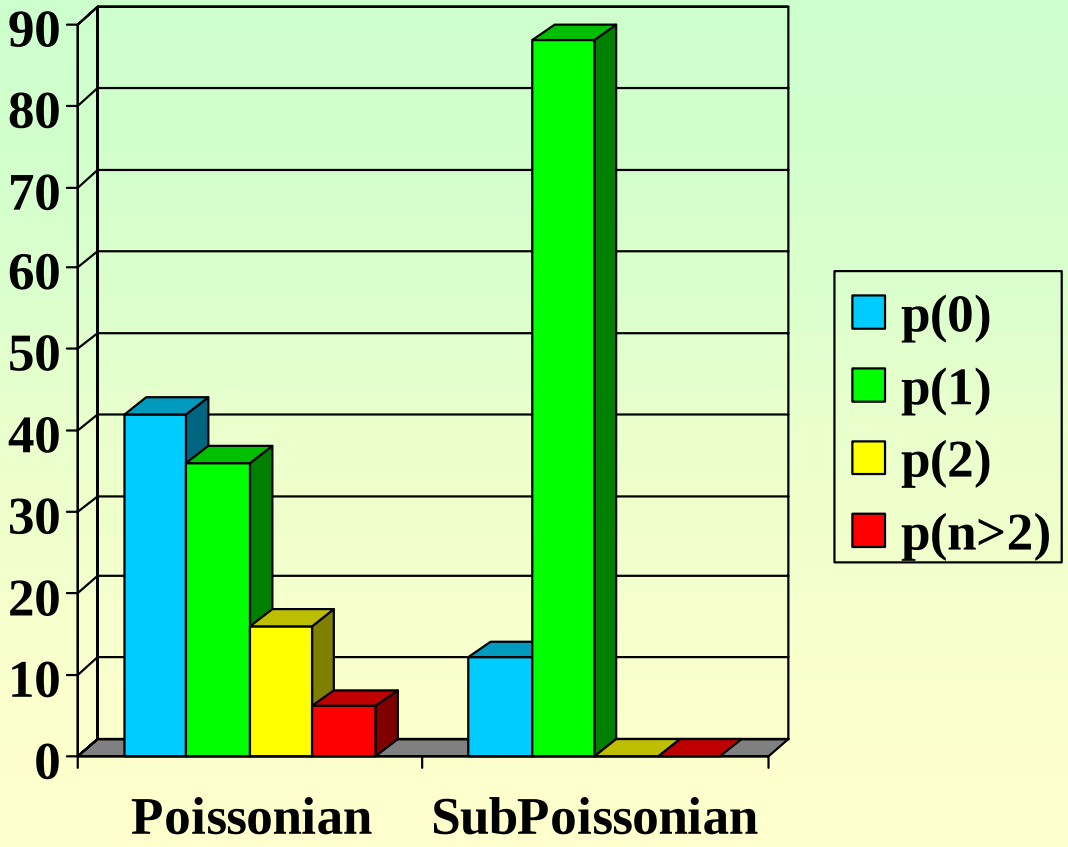
Emission rate:
$S = \eta \nu\, p(1) = S_\infty \Pi(\tau_p)$



At the maximum power :
S_{max}=310 kHz,
$p_{max}(1)$=0.86

86% of the pulses lead to a single photon emission

Comparaison with a coherent source



- $n_{av} = 0.86$
- $p(1) = 0.86$
- $\nu = 6.25$ MHz
- Mandel Parameter

$$Q_{sour.} = -0.86$$
$$Q_{detc.} = -0.03$$

- $P(n>1) < 8\ 10^{-4}$!

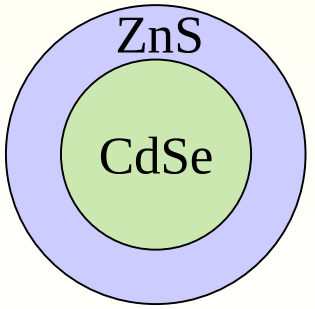
Photon statistics of single quantum dot fluorescence

- QDs bridge the gap between single molecules and bulk solid state
- Size-dependent optical properties
- Tunable absorbers and emitters
- Applications from labeling to nano-devices

- Colloidal CdSe/ZnS quantum dots

- 2 nm Radius, 575 peak emission

- fluorescence quantum yield ~50%, $\varepsilon \sim 10^5\ M^{-1}\ cm^{-1}$



Intermittence in single QD Fluorescence

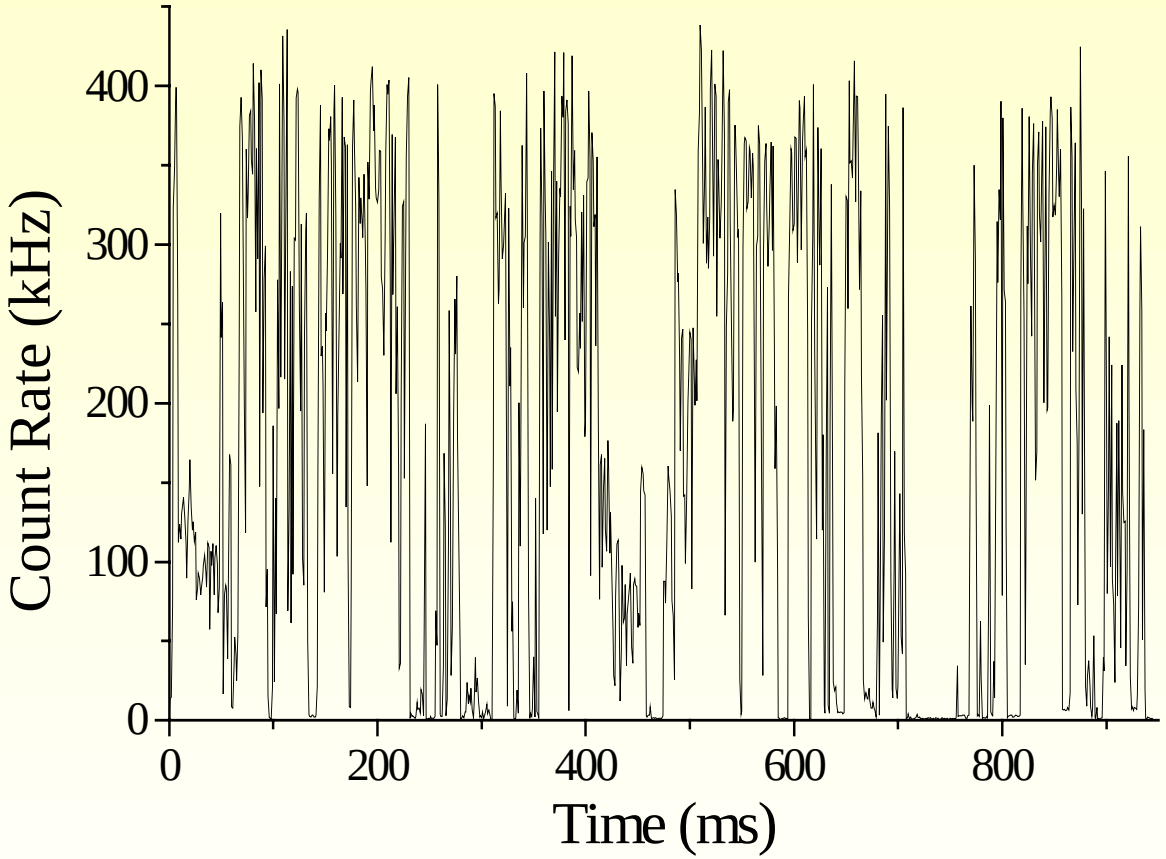
- High S/B ratio
- Low photobleaching rate

$\phi_{blea} < 10^{-8}$

Blinking:

t_{on} , Intensity dependence

t_{off} , no I dependence, inverse power law

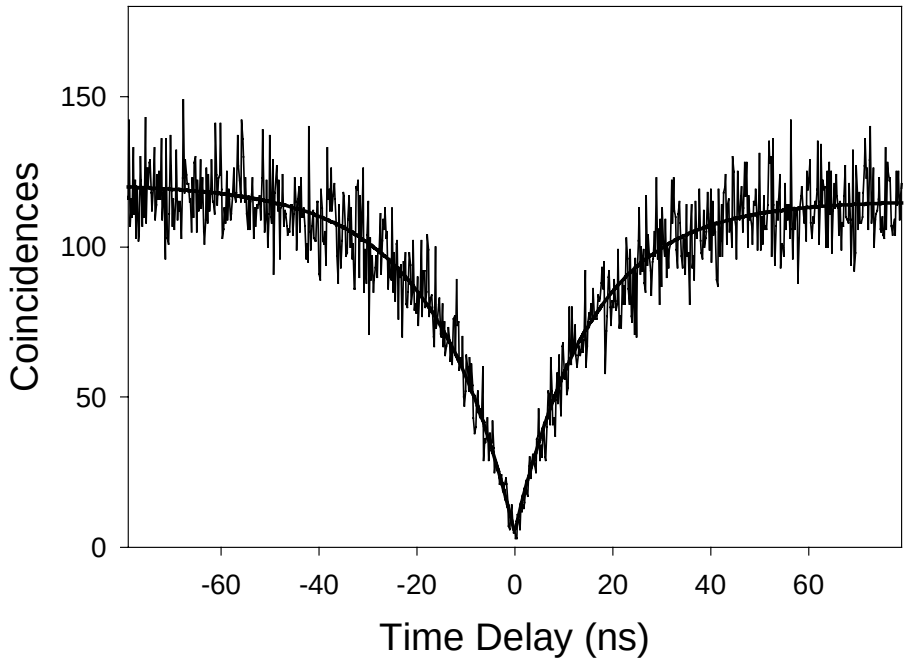


- Blinking attributed to Auger ionization

Photon antibunching in single QD fluorescence

- Start-Stop setup
- Coincidence histogram $C(\tau)$

(TAC time window t_{TAC} of 200 ns, bin width t_{bin} of 0.2 ns)

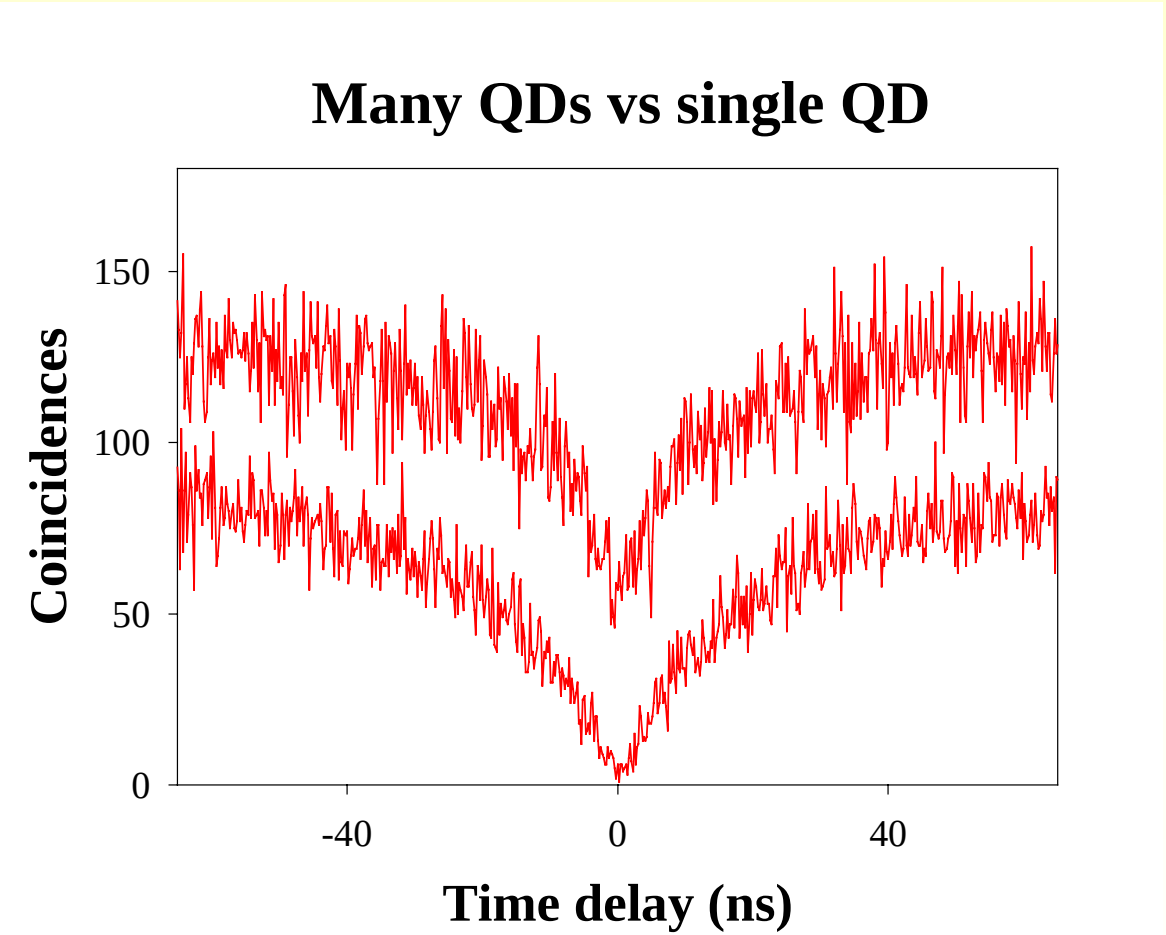


Dip at $\tau=0$, signature of a strong photon antibunching

$C(0) \sim 0$ for a large range of intensities (0.1 – 100 kW/cm^2)

High Auger ionization rates (~1/20 ps^{-1}, Klimov et al.)

No multi-excitonic radiative recombination

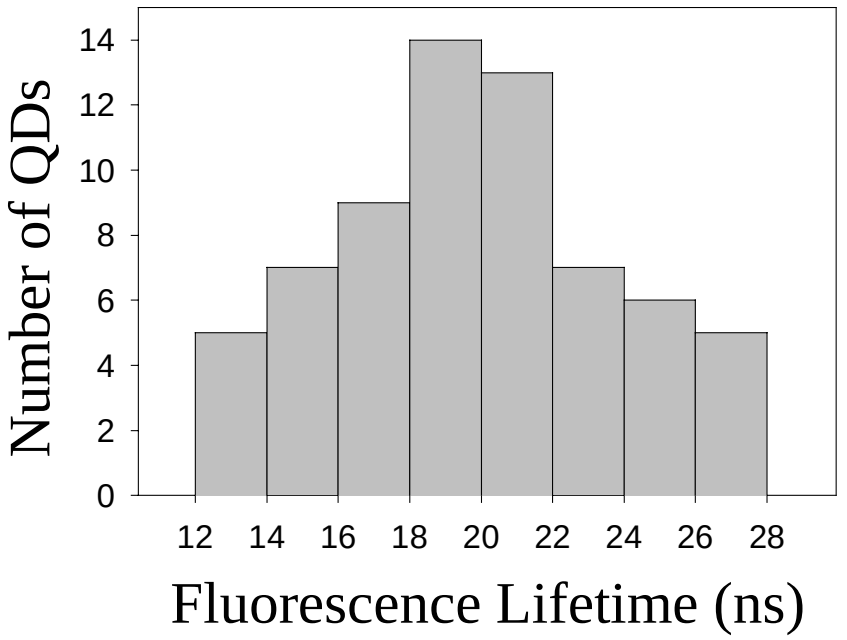

Many QDs vs single QD
Coincidences
150
100
50
0
-40
0
40
Time delay (ns)

Quantum dot lifetimes measurements

Single QDs measurements at low intensities

$$C(\tau) = G^{(2)} \propto 1 - \exp\left(-\tau/\tau_f\right)$$

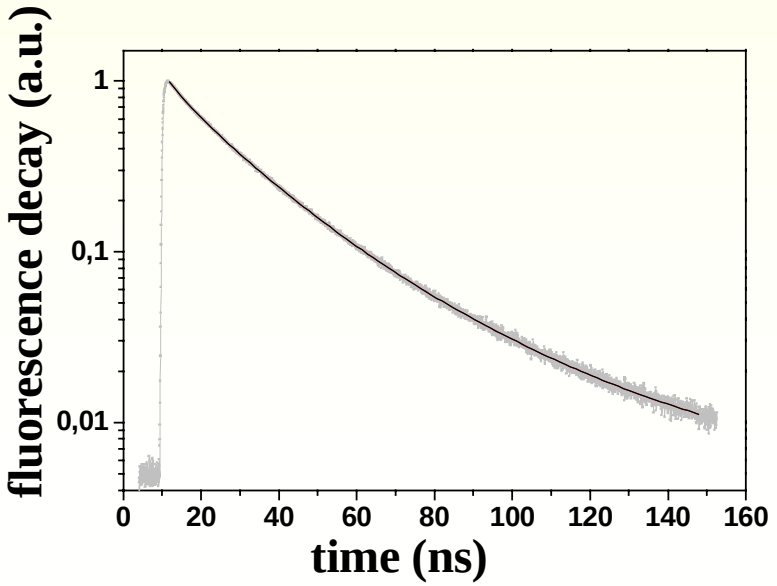
- Experimental accuracy ~ ns
- Width of τ_f histogram : heterogenety in the QDs structure !?



Bulk measurement with TCSPC

(M. Dahan et al., 2000)

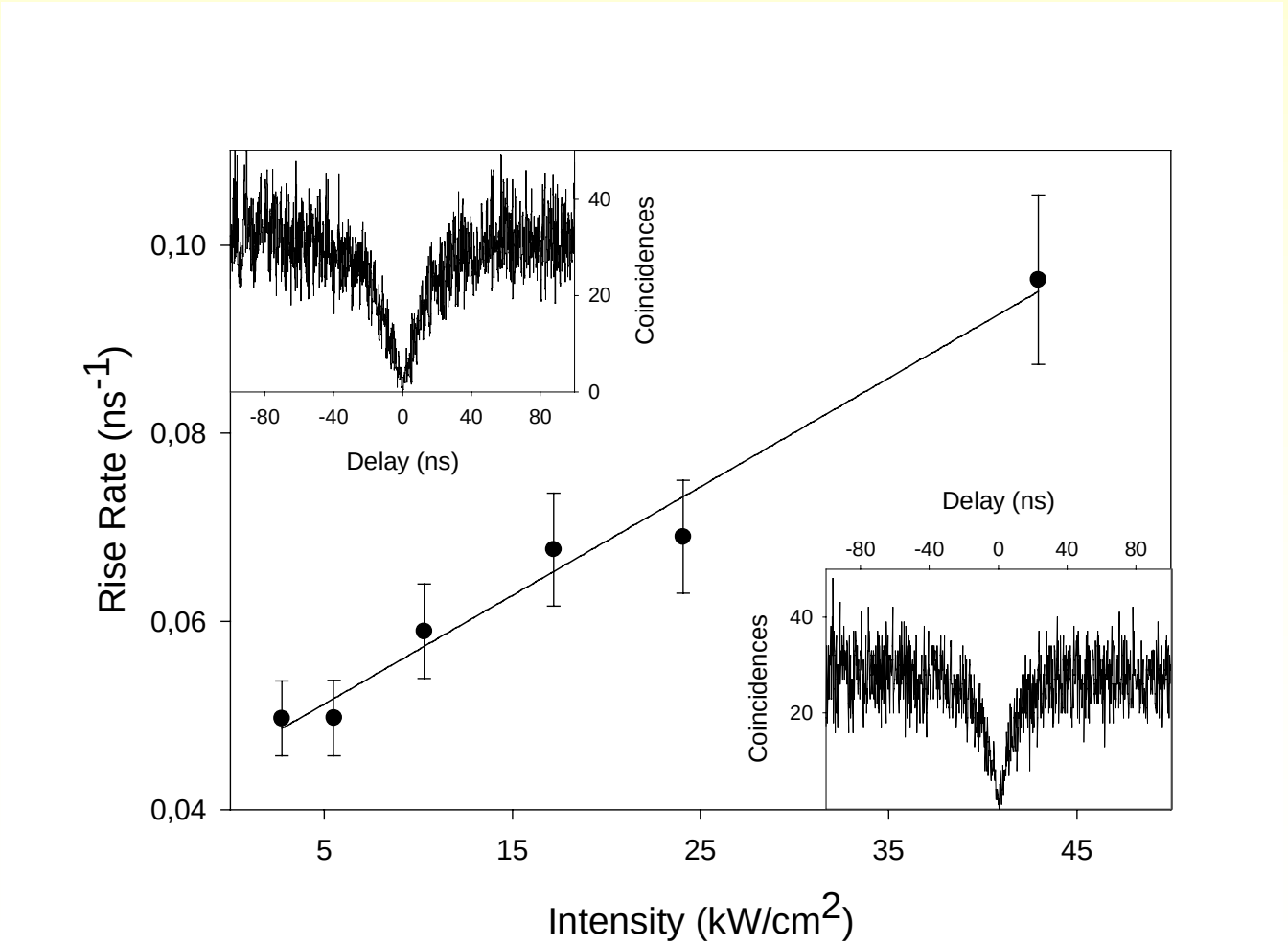
Multi-exponential decay



From the QD state filling

$$C(\tau) \propto 1 - \exp[-(1 + I/I_{sat}) \cdot \tau/\tau_f]$$

Saturation intensities:
I_{sat} ~ 10-80 kW/cm^2
Cross-section :
σ_{abs} ~ 2 – 16 10^{-16} cm^2

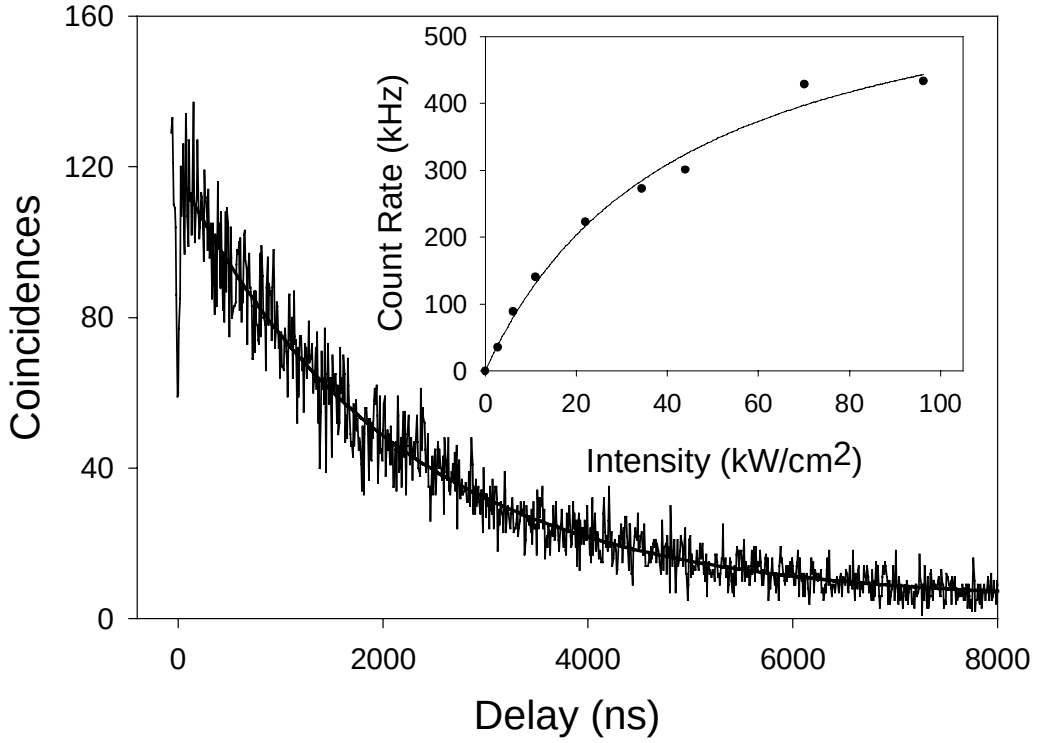


Count rate saturation

- At high intensities, very short t_{on} , average count rate R_{av} skewed
- Use coincidence histogram for accurate value R_{av} in the On state
- With large t_{TAC} and high R_{av} , $\tau \sim$ interphoton mean time ($1/ R_{av}$)

$$C(\tau) = G^{(2)}(\tau) \exp(-R_{av}\tau)$$

Good agreemeny
For the measured I_{sat}



Conclusion

- Demonstration of a single photon source based on controlled fluorescence from single molecule
- Room temperature operation
- Improve the collection efficiency de collection (cavity…)
- Other systems:
  * NV centers (antibunching observed)
  * Quantum dots : at low T (<5K) antibunching in spectrally selected fluorescence from InAs QDs
- Photon antibunching in colloidal CdSe QDs

  Efficient Auger ionization effect