Room temperature single photon sources with colloidal semiconductor nanocrystals

Collaborations

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Physique quantique et applications

Collaborations

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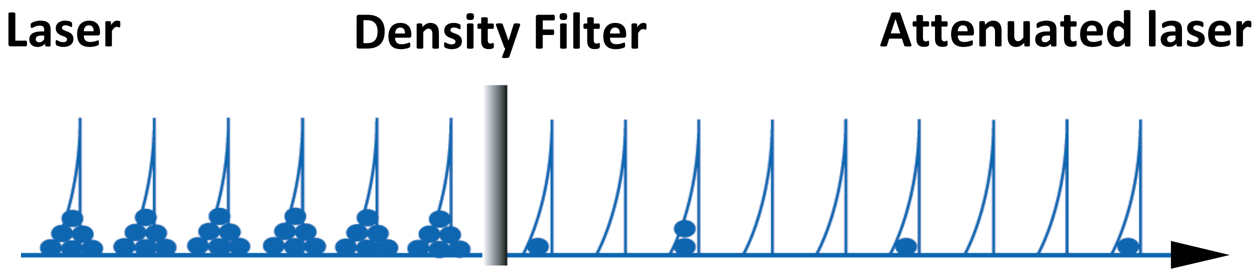
UPMC, INSP, Paris — A Maître

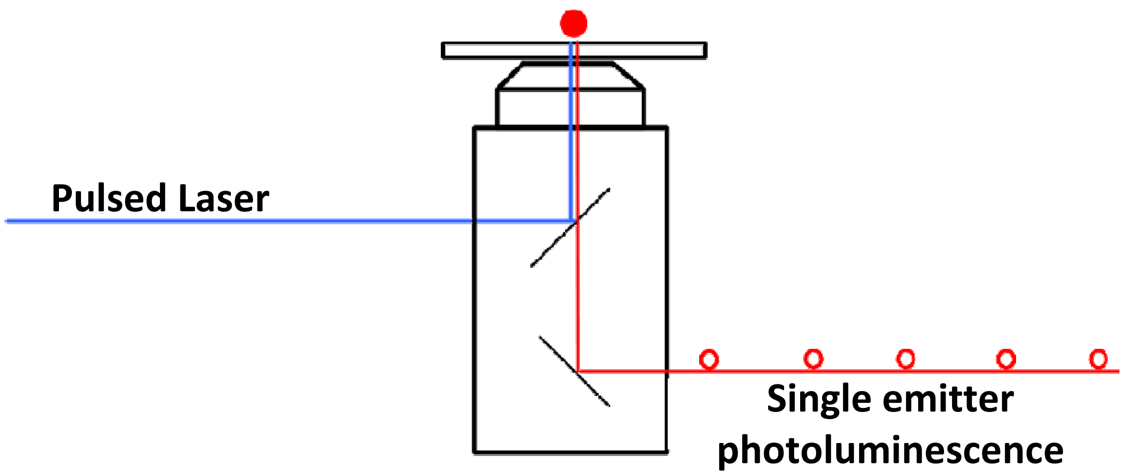
NNL Lab., Lecce — M. DeVittorio

Outline

- Introduction
- Standard nanocrystals
- Blinking free-nanocrystals
- Polarized Single Photon Emission
- Coupling with nanocavities

Why single emitters as single-photon sources?

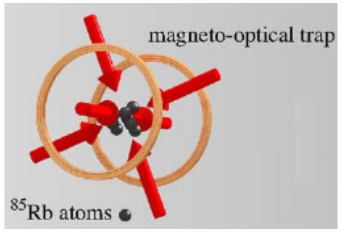


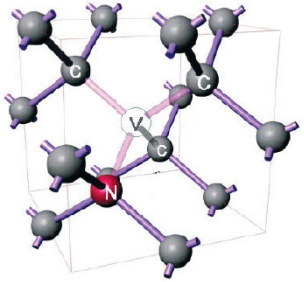


Quantum emitters for single photon generation

Single atom

Kimble et al. PRL (1977)
Hennrich et al New Journal of Physics (2004)



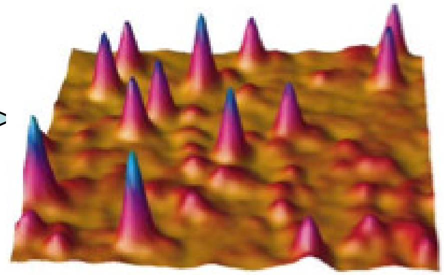


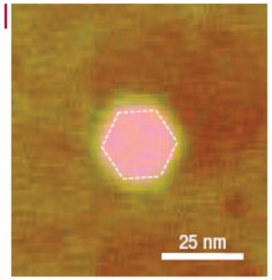
Single nitrogen vacancy in diamond

H. Weinfurter et al. PRL (2000)
P. Grangier et al. Optics Letters (2000)

Single Molecule at room temperature

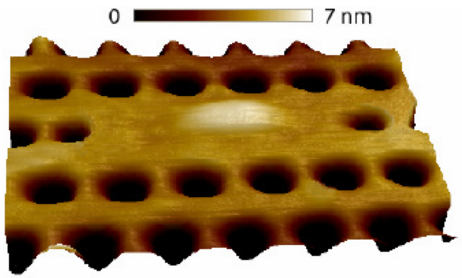
B. Lounis and W.E. Moerner, Nature (2000)





Single Quantum Dot

Arakawa et al. Nature Materials 5, 887 (2006)



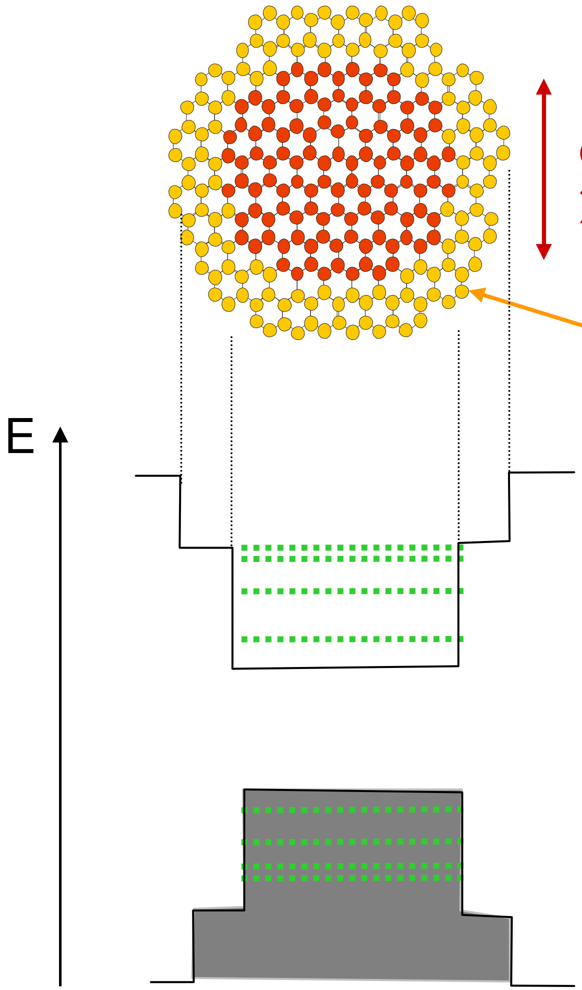
Single Quantum Dot in microcavity

Imamoglu et al. Nature 445, 896 (2007)

NNL

LKB

Core/Shell Colloidal Semiconductor Nanocrystals



core CdSe
2.R = 3,5 nm (~ 1000 atoms)

Passivation shell ZnS (some nm)

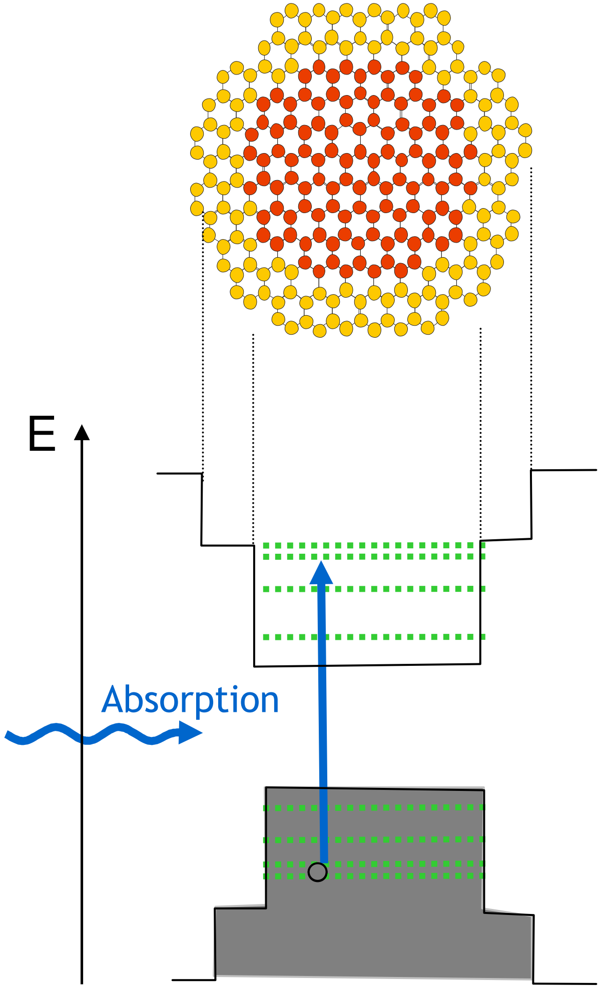
$R << a_{Bohr}, \lambda_{de\ Broglie}$

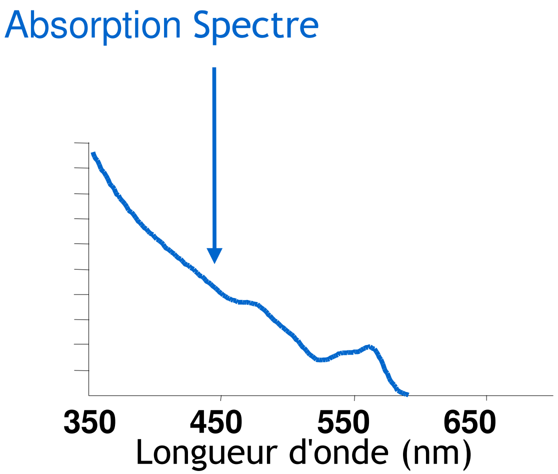
3D quantum confinement

Discrete energy levels

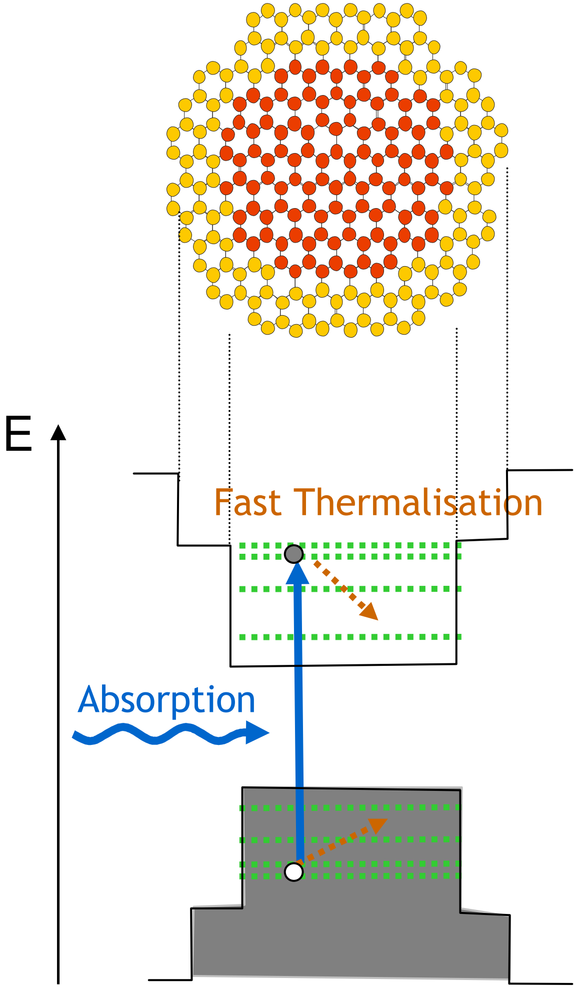
energy depending on R

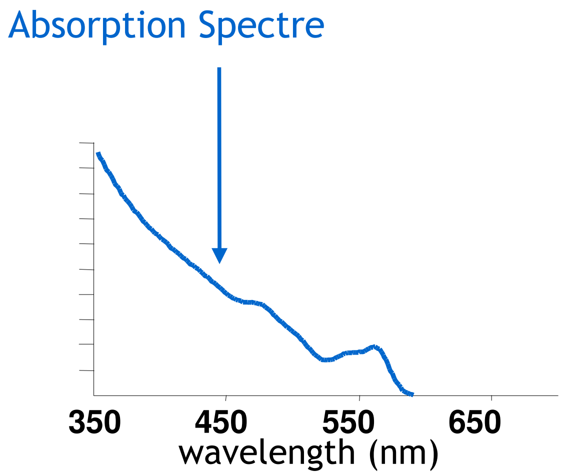
Optical properties



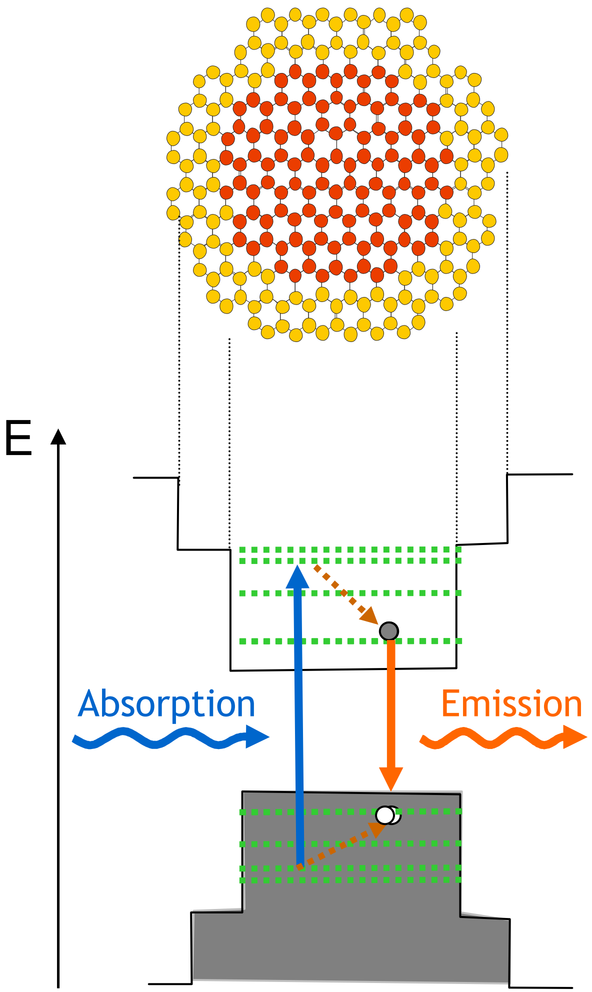


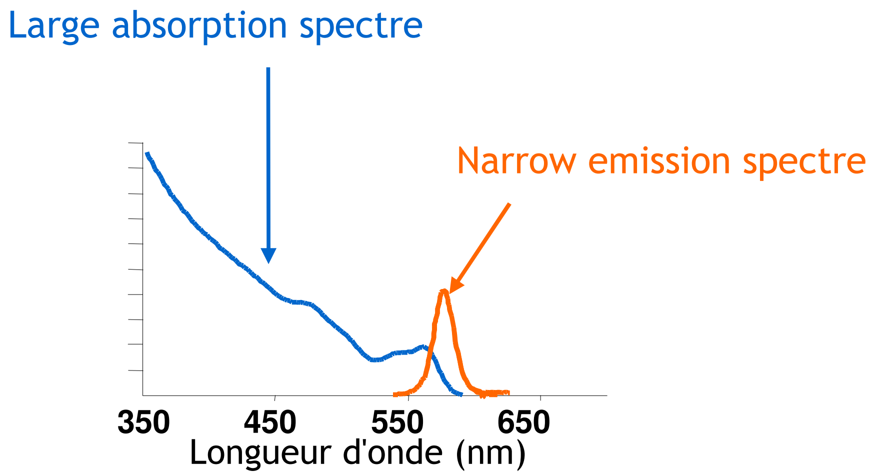
Optical properties





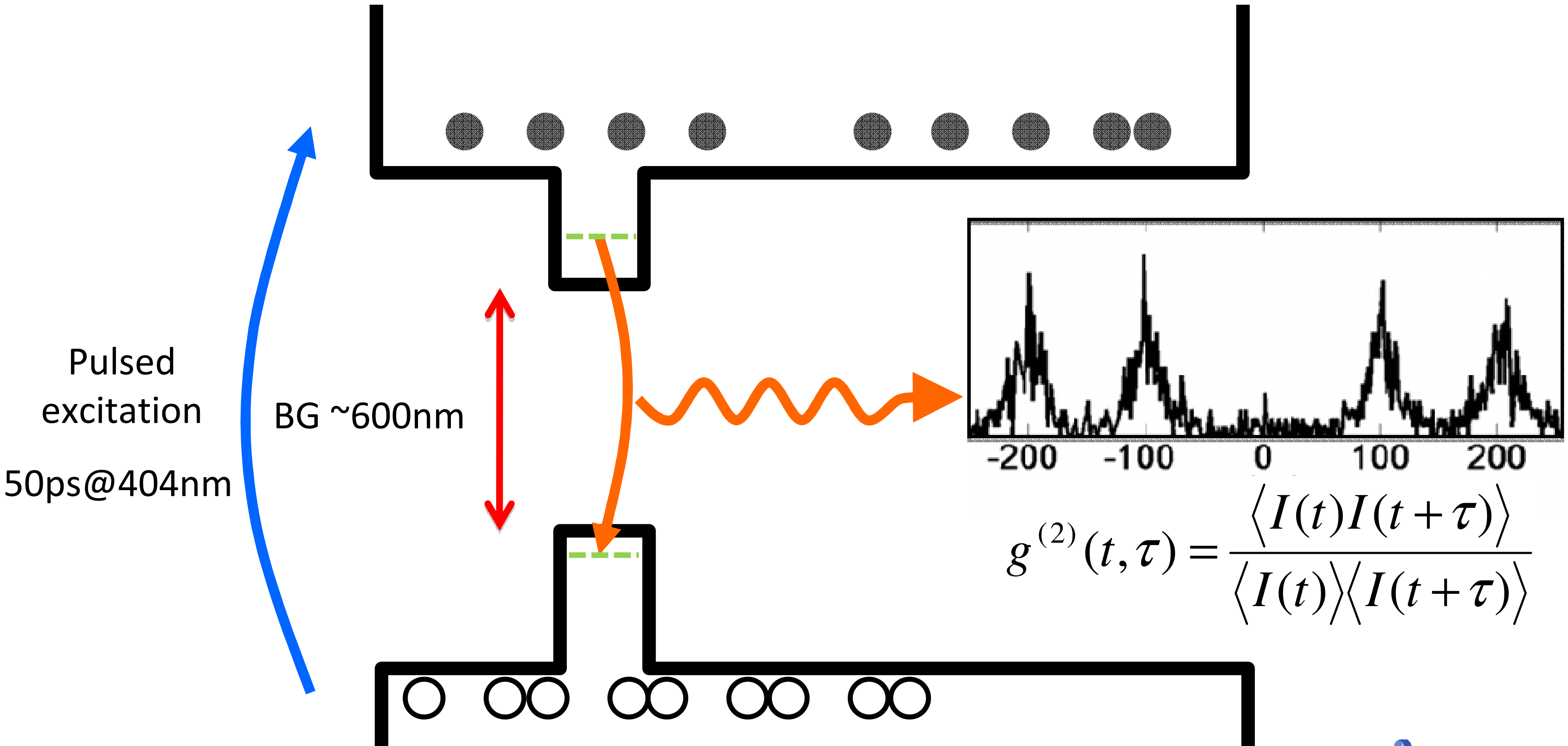
Optical properties





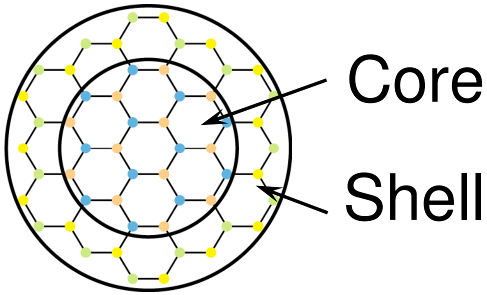
emission from lowest excited level
- narrow emission line
- tunability with R
- Two level system

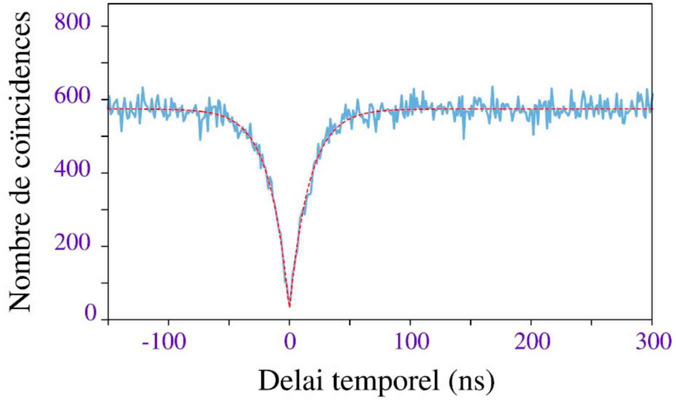
Efficient Auger Recombination



$$g^{(2)}(t,\tau) = \frac{\langle I(t)I(t+\tau)\rangle}{\langle I(t)\rangle\langle I(t+\tau)\rangle}$$

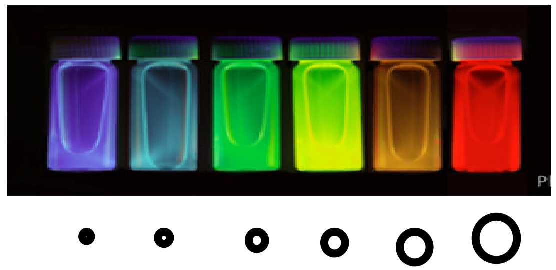
Colloidal semiconductor nanocrystals





Photon antibunching observed at room temperature

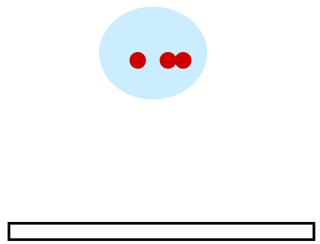
Wet- chemistry synthesis
well controlled → bright, stable,
with tunable wavelength



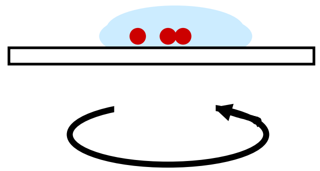
optoelectronics (LEDs, lasers, solar cells…)
biology (fluorescent markers)
quantum optics (single emitters)

Single nanocrystal Observation

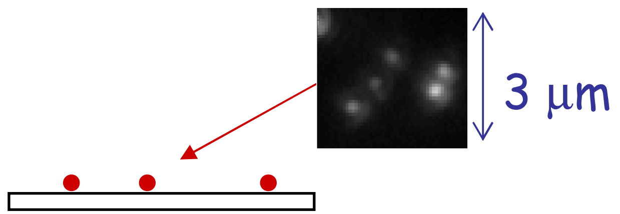
spin coating deposition :



Single nanocrystal Observation

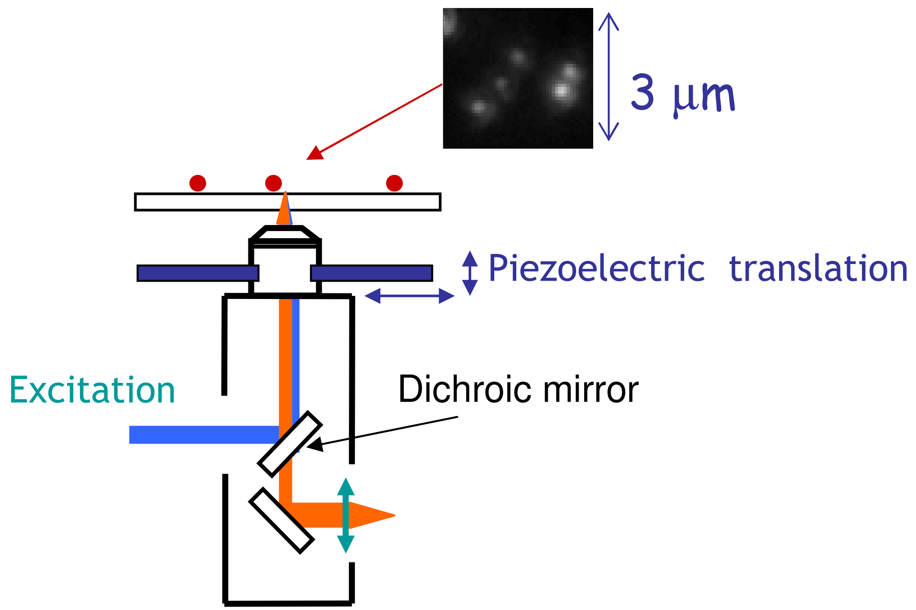


Single nanocrystal Observation



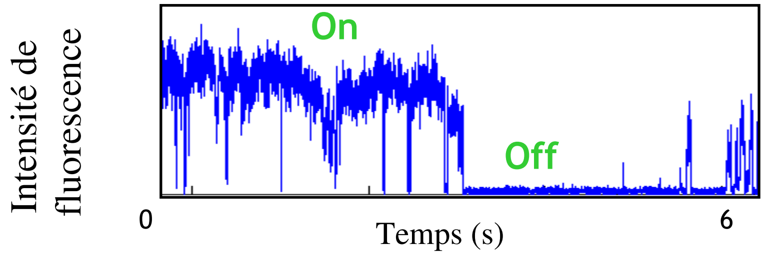
Single nanocrystal Observation

Confocal microscopy:

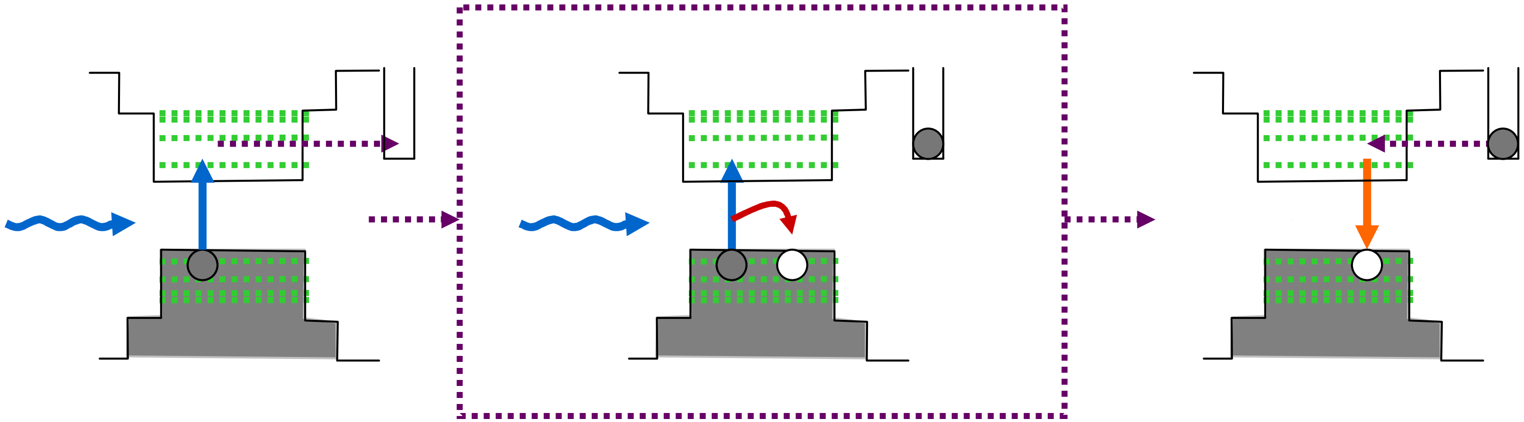


Single nanocrystal Observation

→ blinking



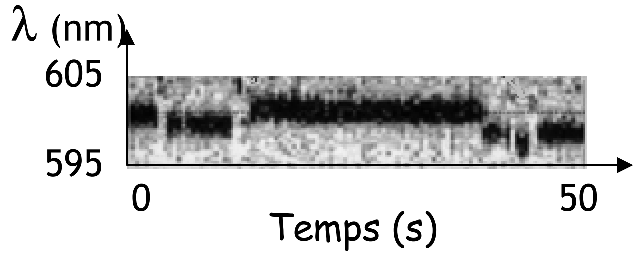
Model : ionisation of nanocrystal
(non radiative relaxation by effet Auger)



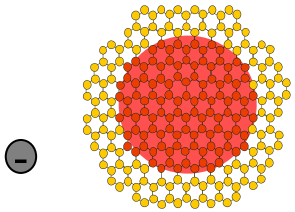
Single nanocrystal Observation

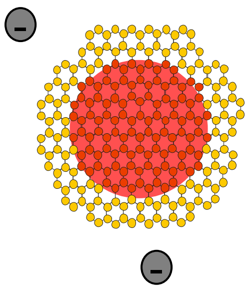
blinking

spectral diffusion



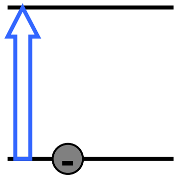
Fluctuations of the local electric field



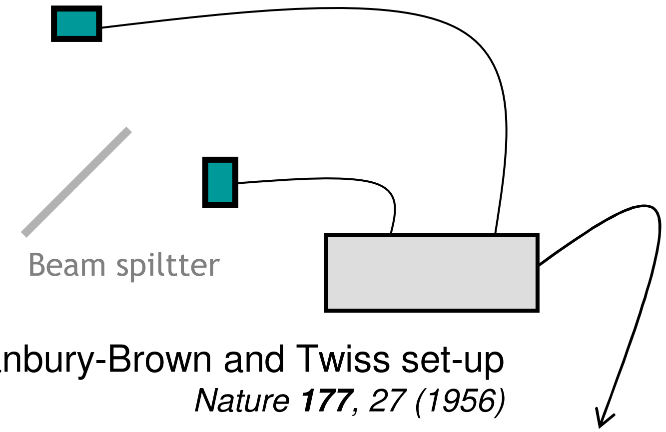


Single nanocrystal Observation: Antibunching

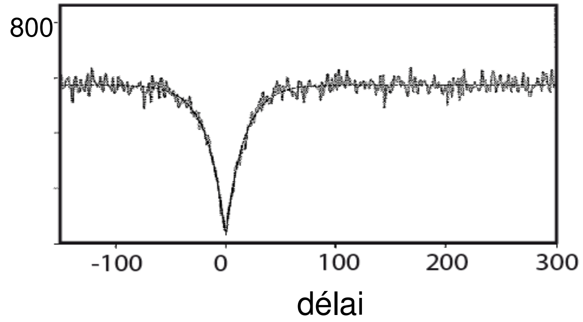
→ Demonstration of single photon emission



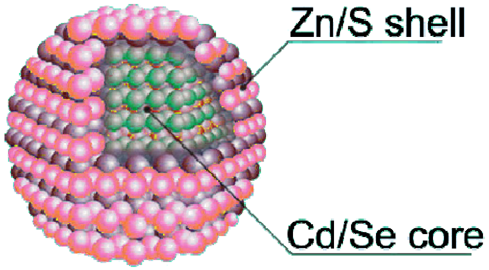
two level system



Hanbury-Brown and Twiss set-up
Nature ***177***, *27 (1956)*



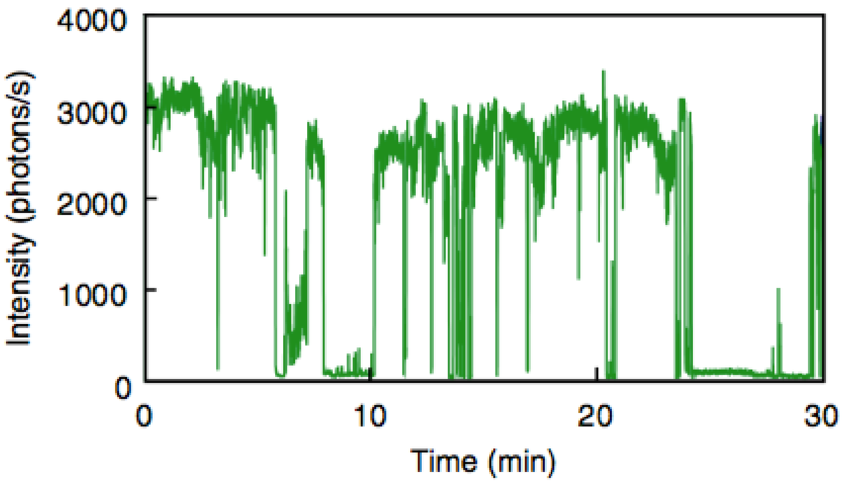
Nanocrystals drawbacks

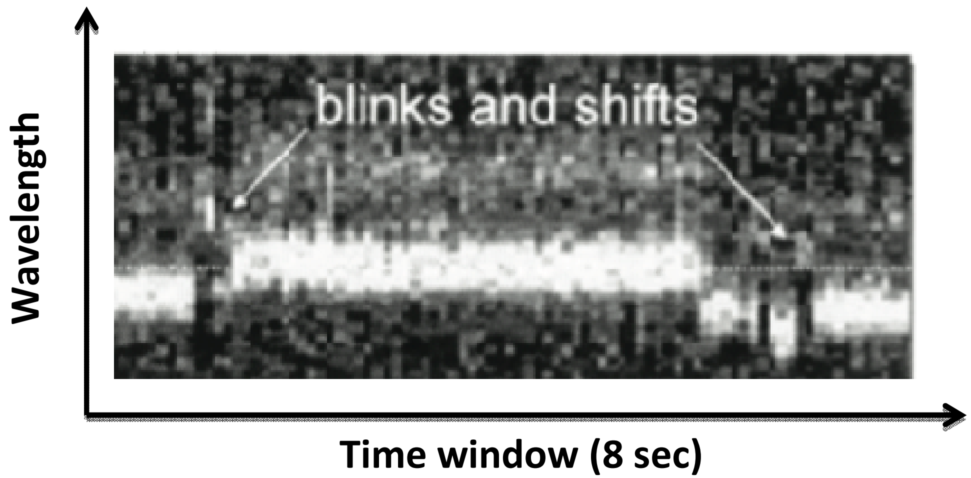


☹ Blinking

☹ Spectral Diffusion

☹ Not polarized emission





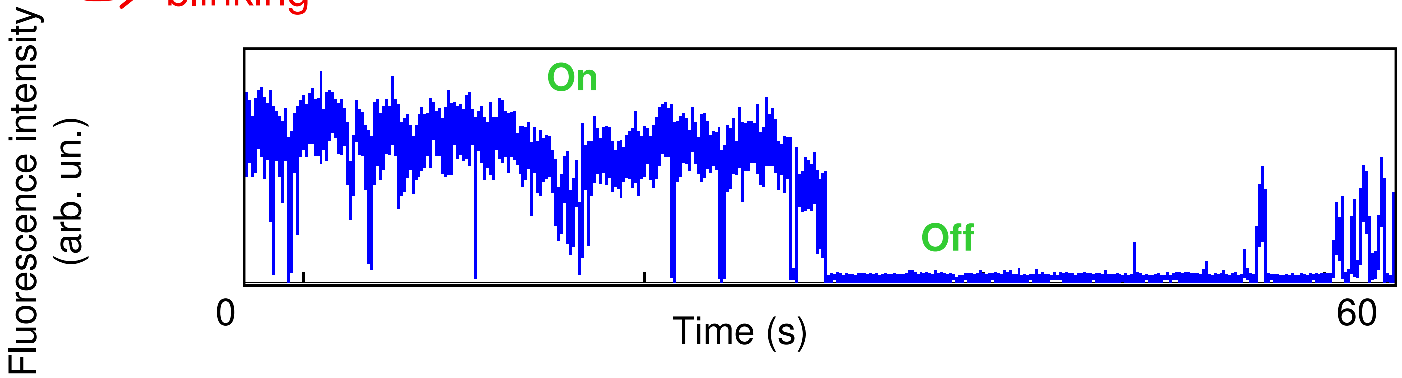
R. G. Neuhause et al., Phys. Rev. Lett. **85**, 3301 (2000).
X Brokmann et al., New journal of physics **6**, 99 (2004).

Non-blinking colloidal quantum dots

CdSe/ZnS Single nanocrystal observation

→ suppression of average effects

↳ blinking

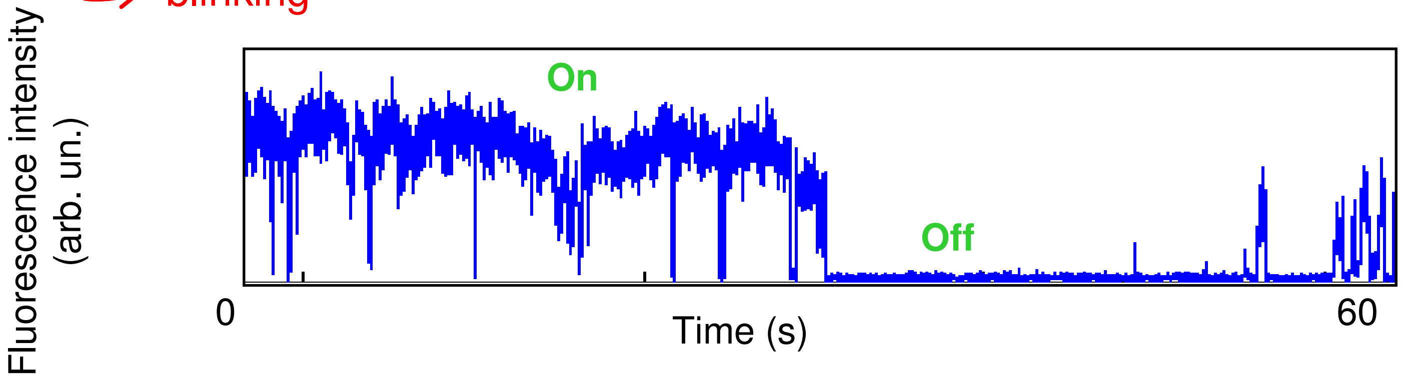


How does blinking work?

CdSe/ZnS Single nanocrystal observation

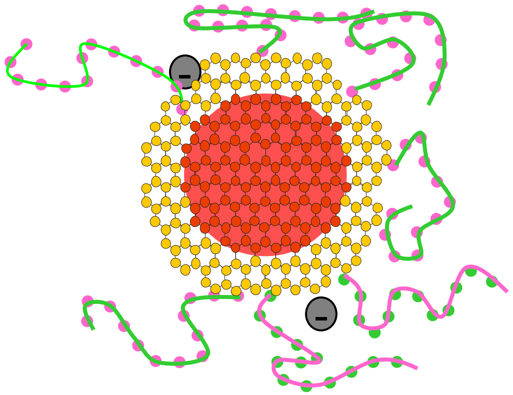
→ suppression of average effects

↳ blinking



How does blinking work?

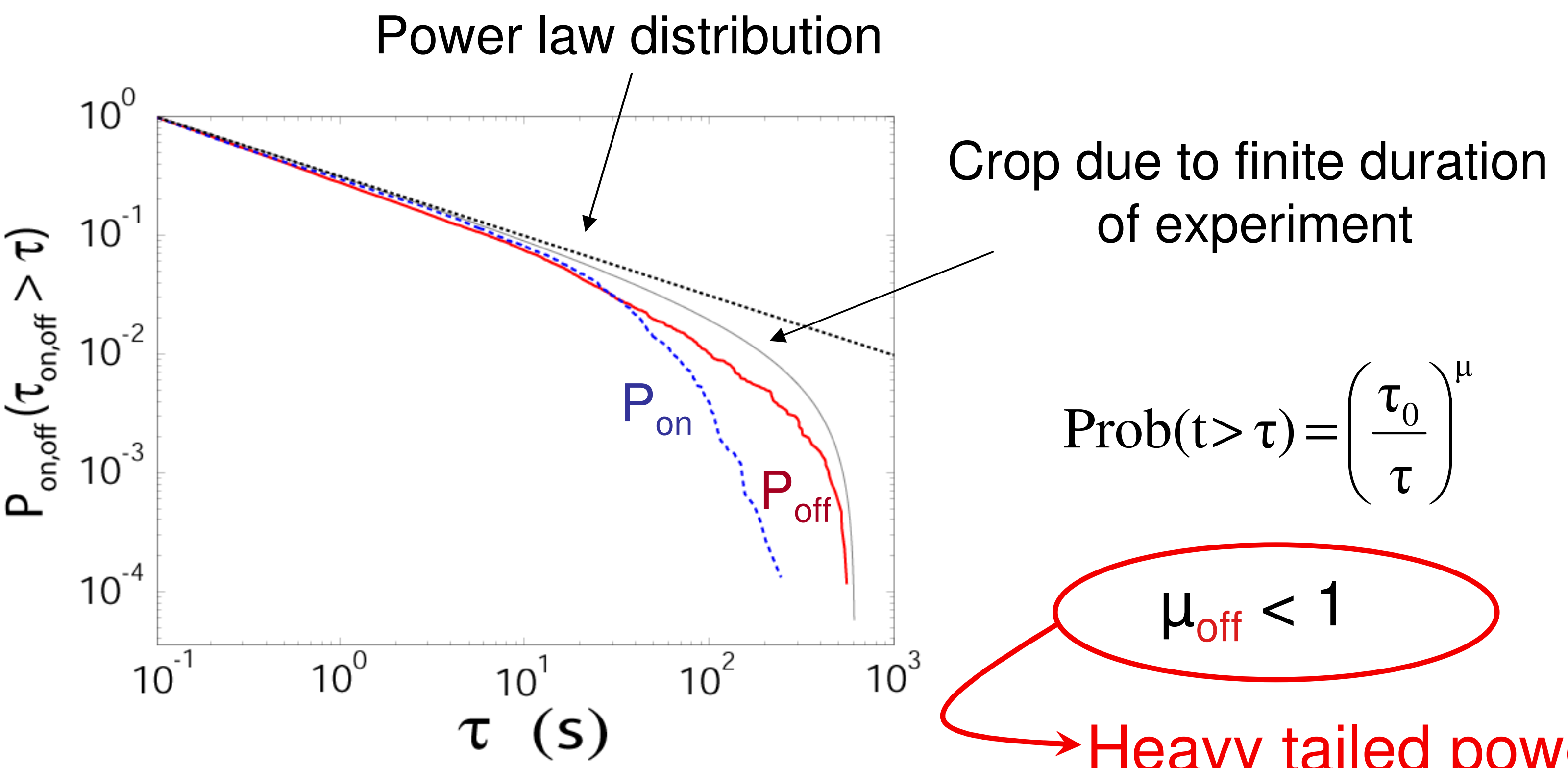
… ionisation processes.
(traps model)



Lattice mismatch between CdSe – ZnS > 10%

Verbek *et al.*, "*Simple model for the power-law of single semiconducotr nanocrystal.*", **PRB 66,** 233202-1-4 (2002)

CdSe/ZnS single nanocrystal statistics



$$\mathrm{Prob}(t > \tau) = \left(\frac{\tau_0}{\tau}\right)^{\mu}$$

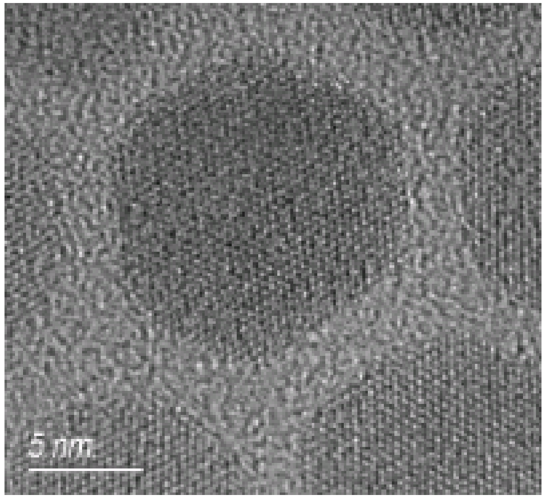
$\mu_{off} < 1$

Heavy tailed power law

- Is not possible to define statistical quantities $(\langle \tau_{off} \rangle, \sigma_{off}, ..)$
- τ_{off} comparable to total acquisition time

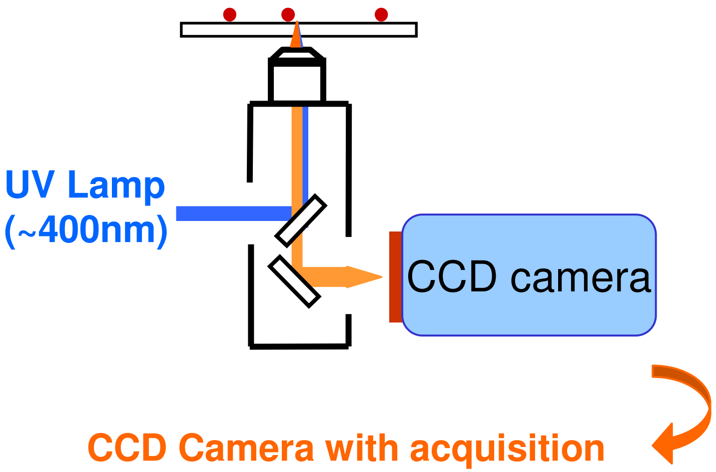
Brokmann *et al.*, "*Statistical Aging and Nonergodicity in the Fluorescence of Single Nanocrystals*", **PRL 90,** 120601 (2003)

CdSe/CdS single nanocrystal observation

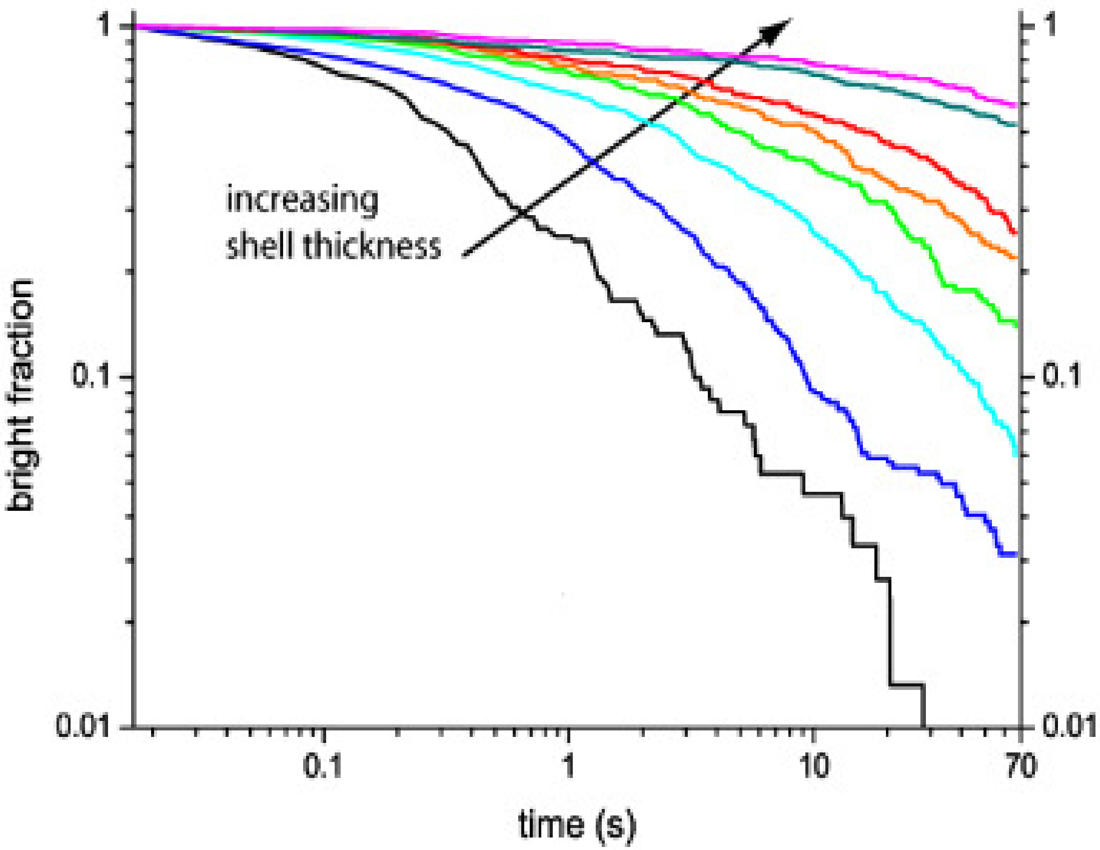


Lattice mismatch between CdSe - CdS ~ 4%

"Huge" high crystalline nanocrystals can be synthesized

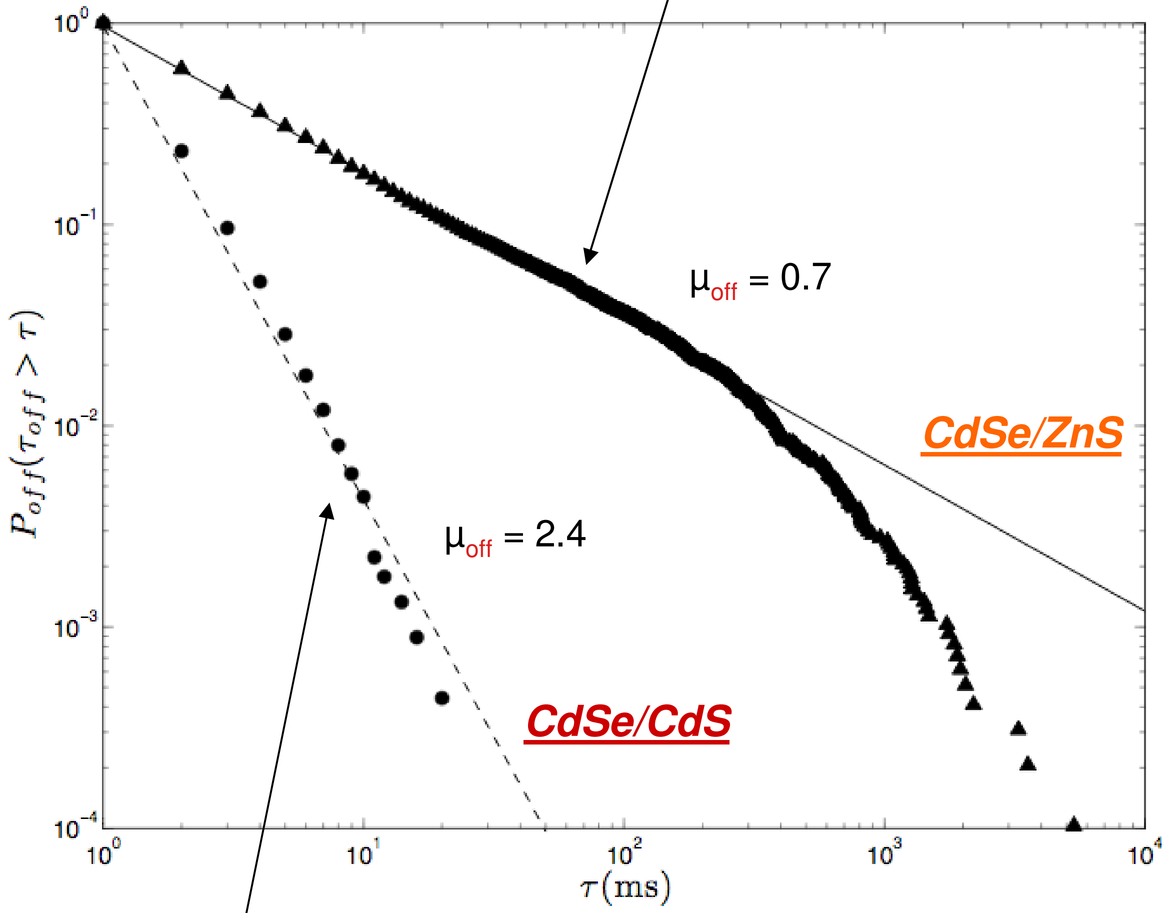


CCD Camera with acquisition rate of 33Hz



CdSe/CdS single nanocrystal statistics

Heavy tailed power law (ZnS)



Short tailed power law (CdS)

CdSe/CdS

$\mu_{off} >> 1$

Short tailed power law

No long off states!

DOTS IN ROD AS POLARIZED SINGLE PHOTON SOURCES

Nanocrystals drawbacks: solutions

Blinking suppression with giant shells

B. Malher et al., Nature Mat. 7, 659 (2008).

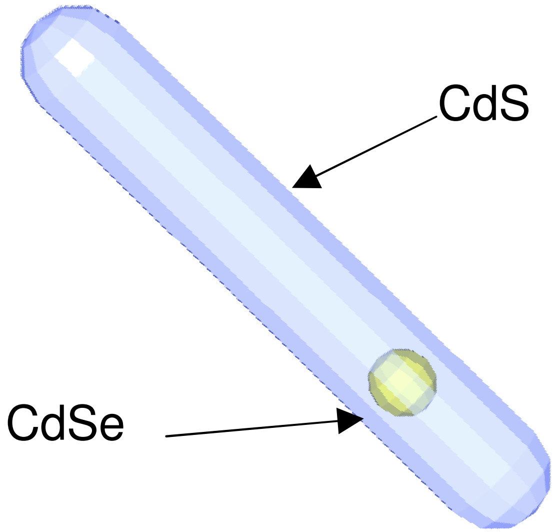
B. N. Pal *et al. Nano Lett.*, 2012, 12 (1)

Polarized emission with rods

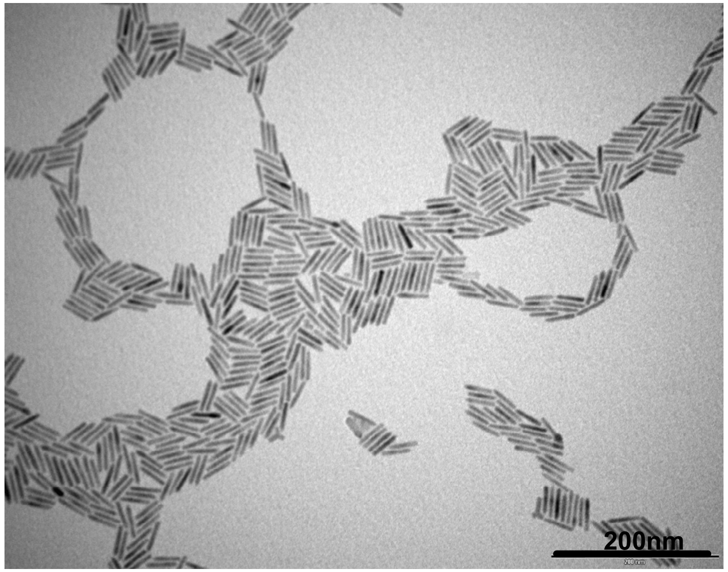
Jiangtao Hu *et al. Science* **292**, 2060 (2001)

Loss of the antibunching behavior

Polarization control: synthesis of new type of nanocrystals in rod CdSe/CdS

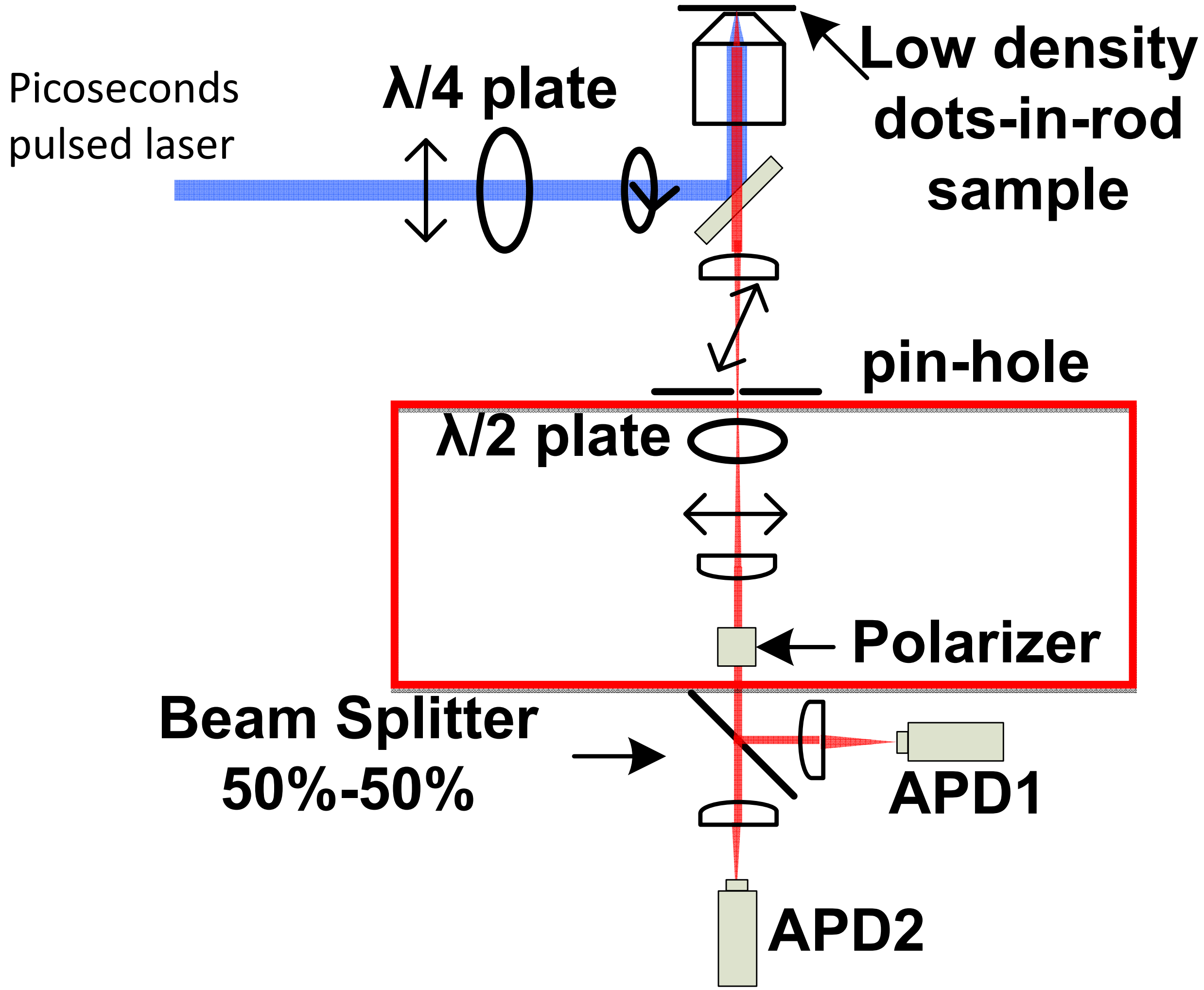


TEM image of a typical output of the synthesis

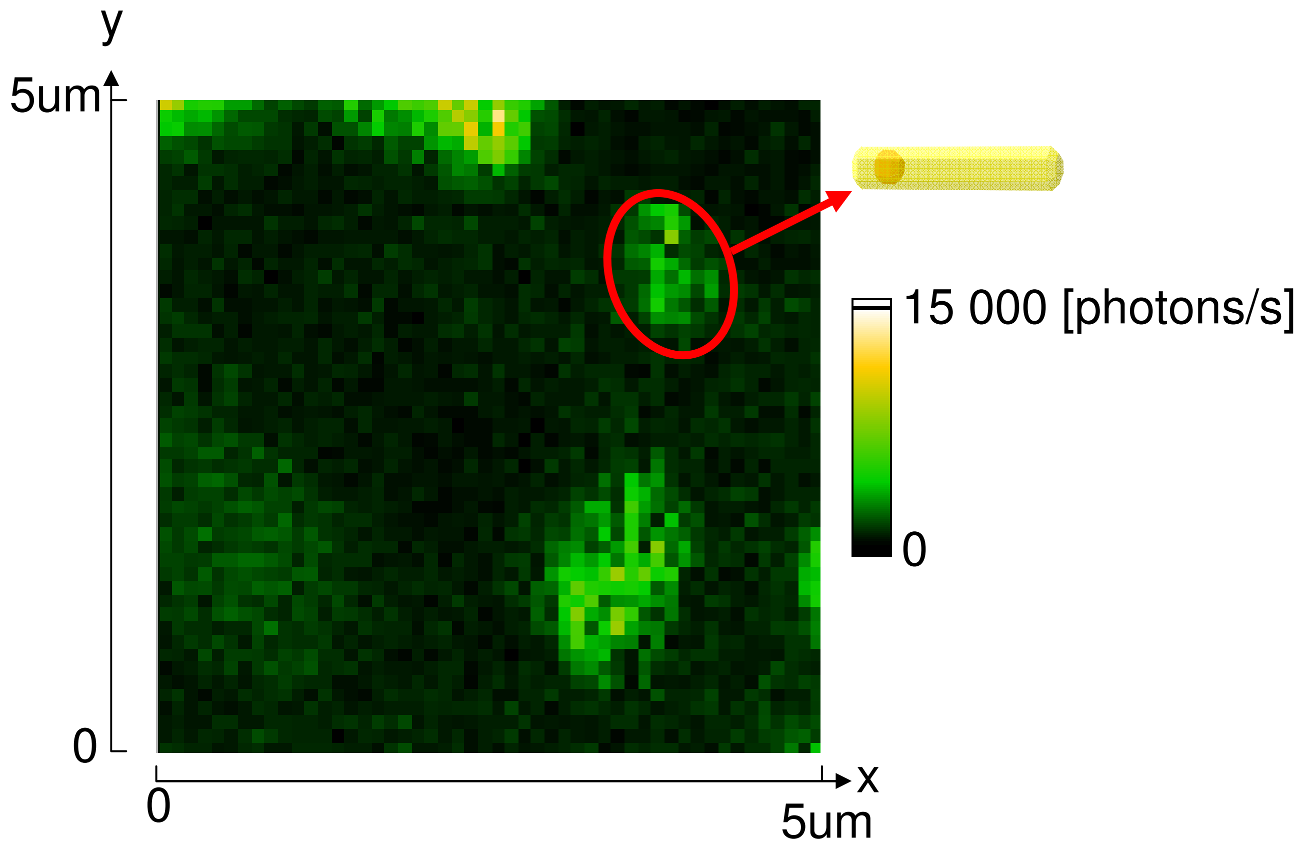


F. Pisanello *et al.*, Appl. Phys. Lett., *2010*

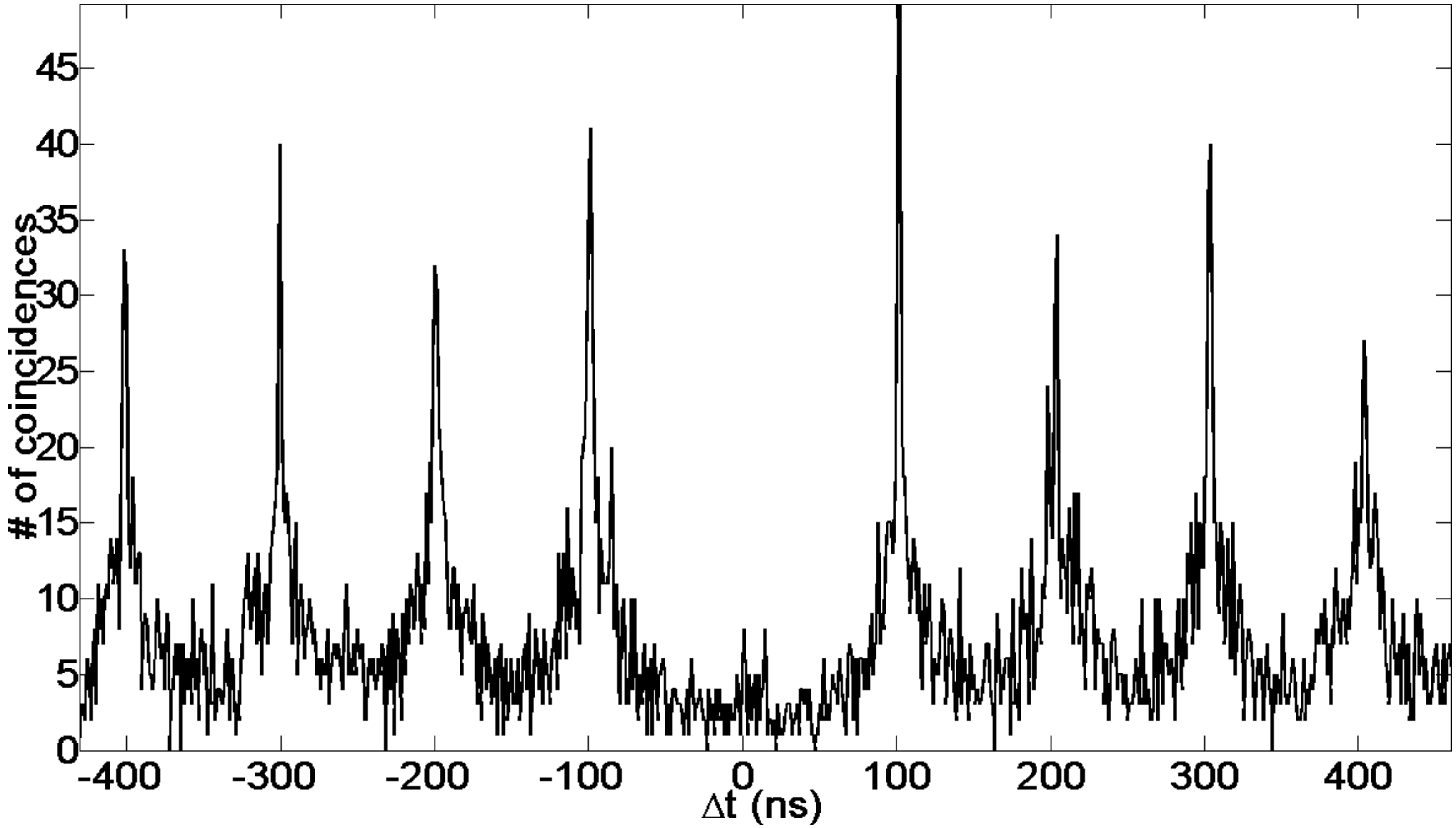
Characterization of a polarized single photon source



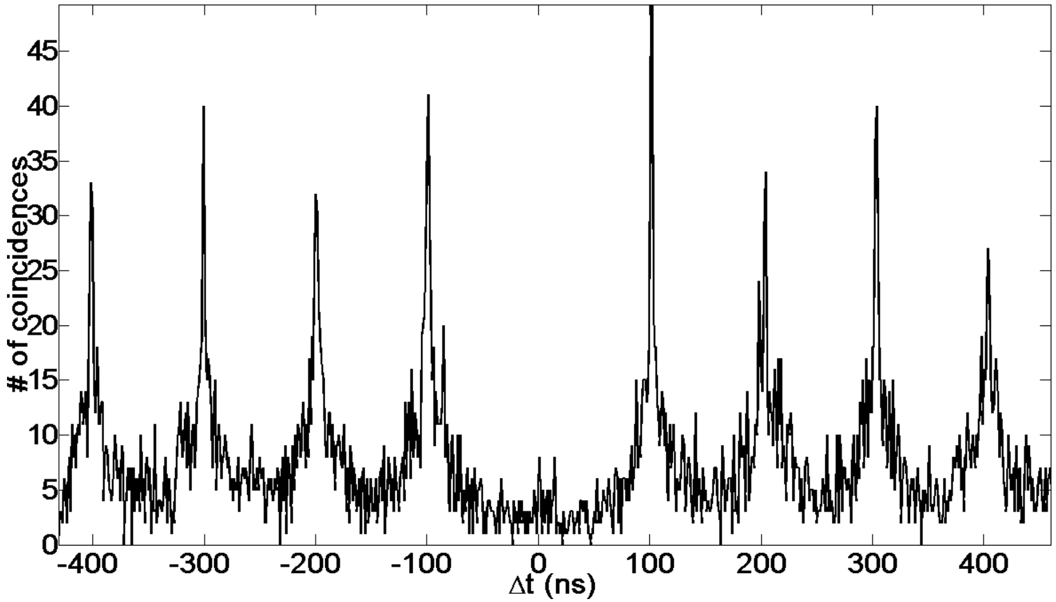
Luminescence topography



Antibunching from a single dot in a rod



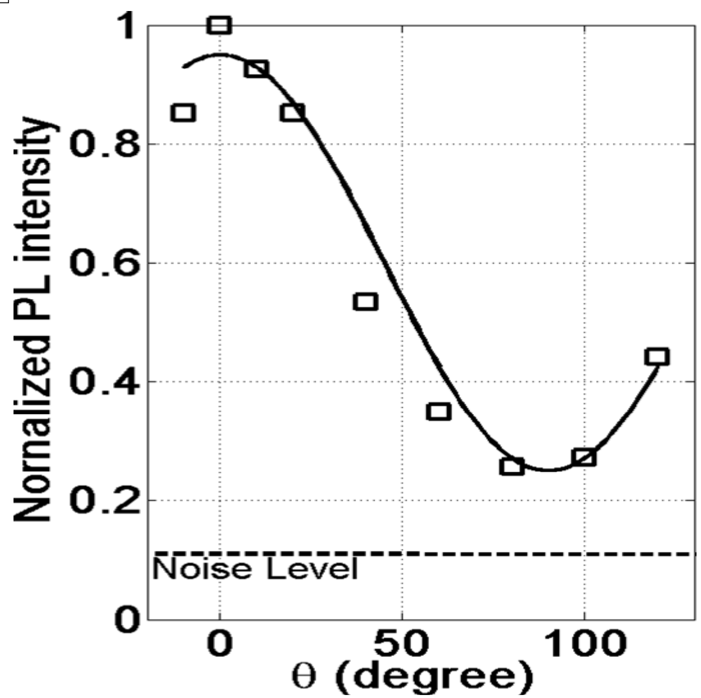
Polarization control: experimental results



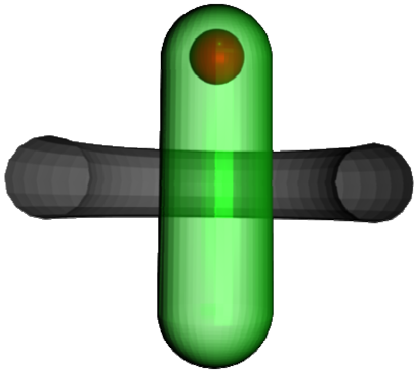
Good single photon sources

High Degree of linear polarization

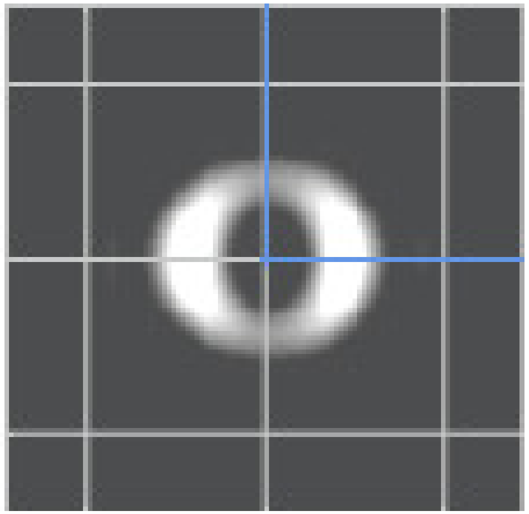
$$\frac{\left(I_{\parallel}-I_{\perp}\right)}{\left(I_{\parallel}+I_{\perp}\right)} \sim 85\%$$



Defocused microscopy

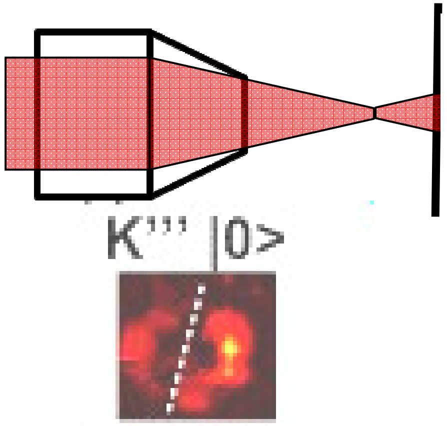


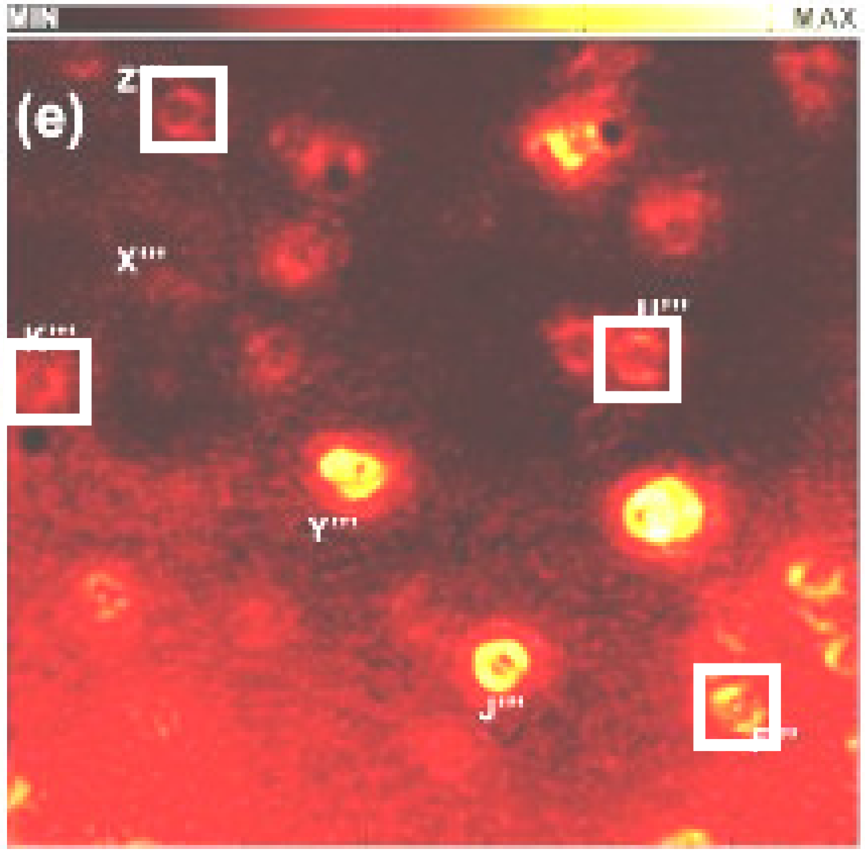
Defocused image for dipole
like emitter



X. Brokmann et al., Chem. Phys. Lett. **406**, 210 (2005).

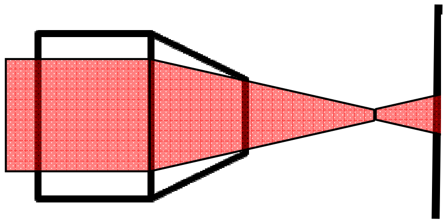
Orientation on the substrate and radiation diagram

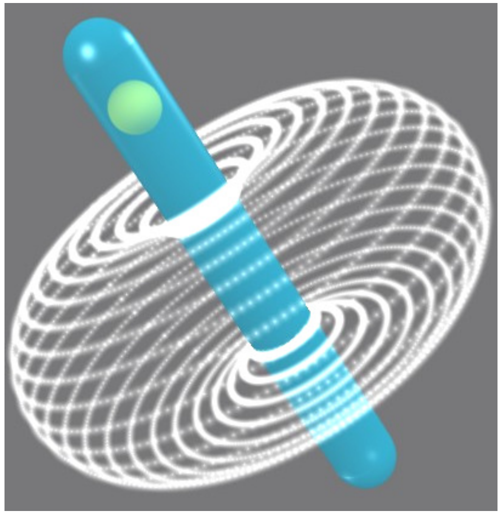




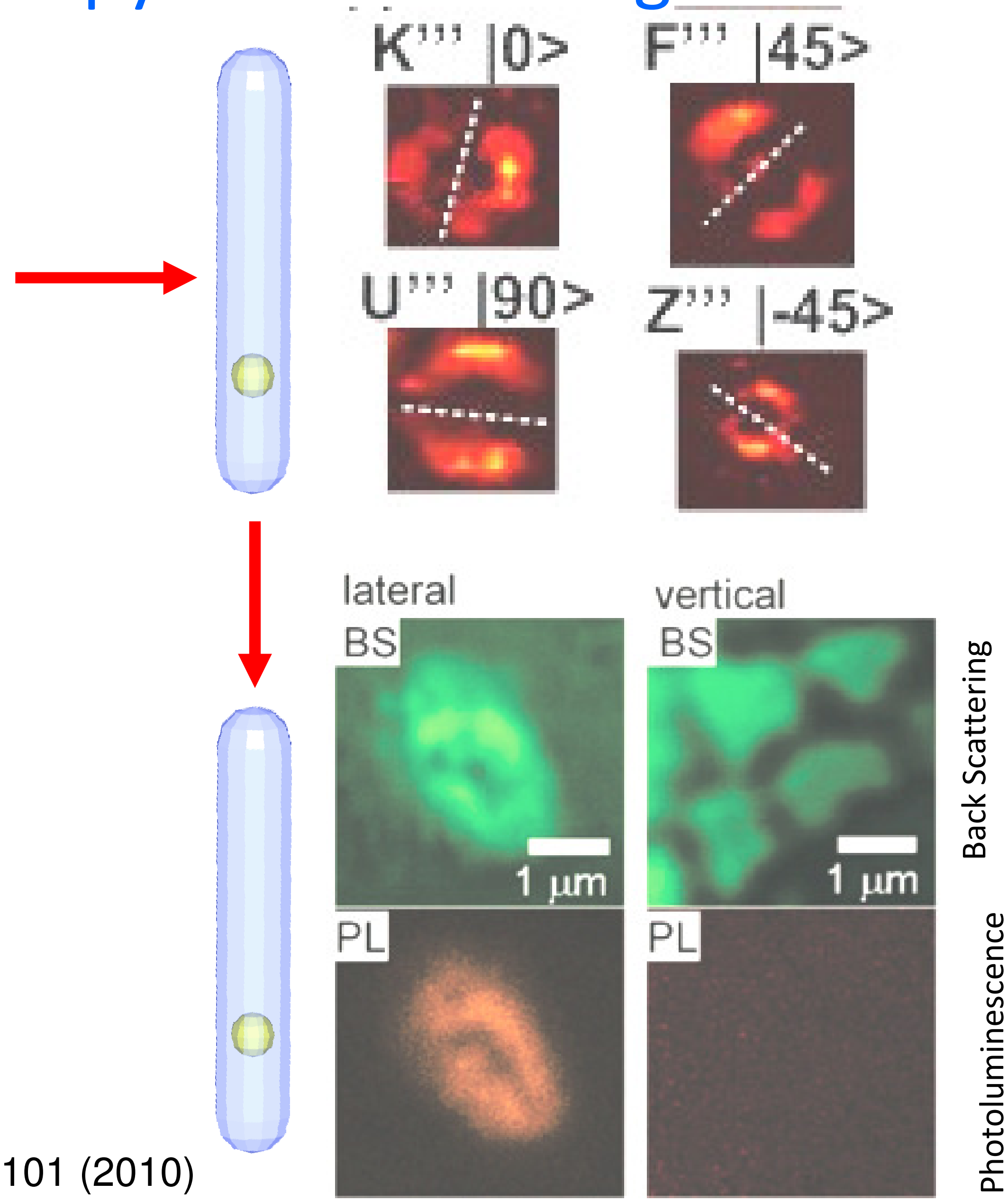
F. Pisanello et al., Appl. Phys. Let. **96**, 033101 (2010)

Defocused microscopy: Radiation diagram



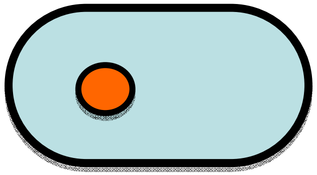


F. Pisanello et al., Appl. Phys. Lett. **96**, 033101 (2010)



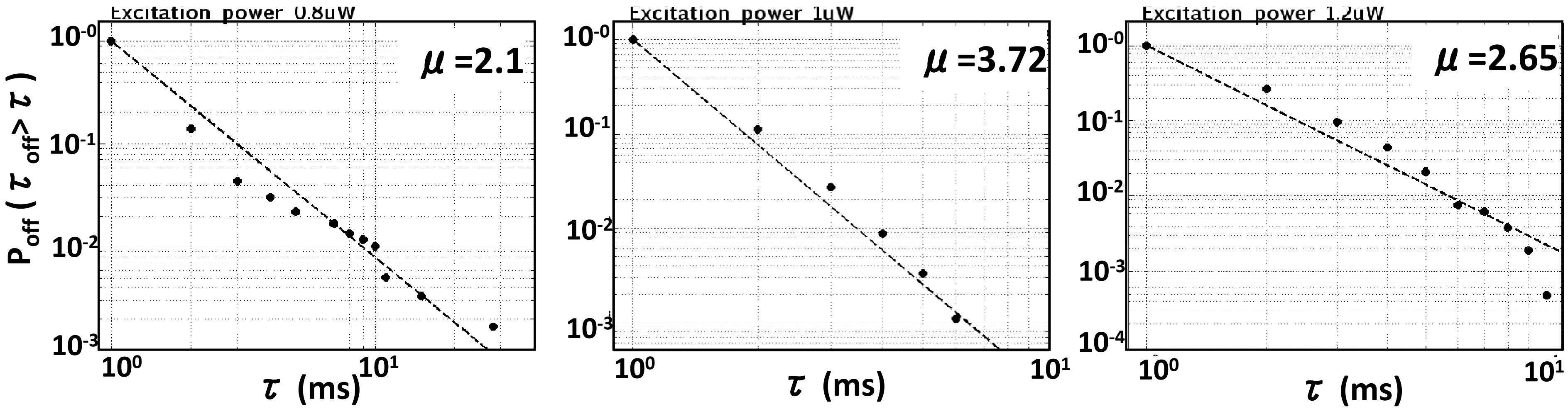
L. Carbone et al., Nano Lett. **7**, 2942 (2007)

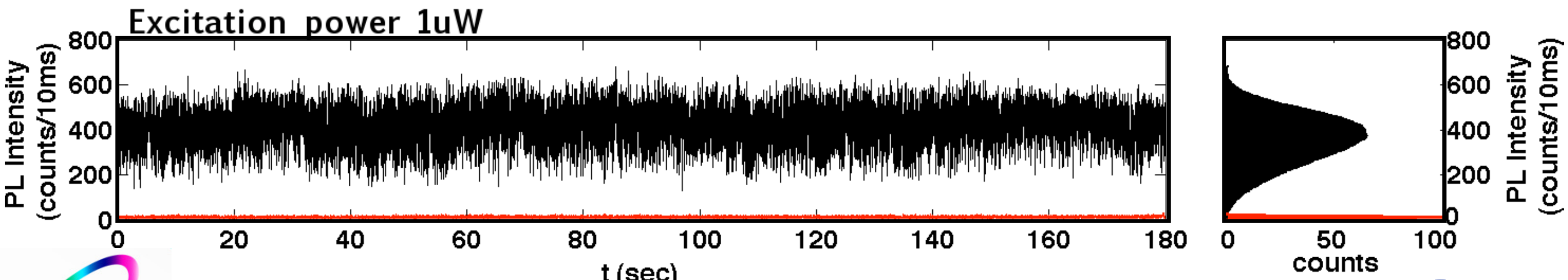
Thick shell: Blinking reduced



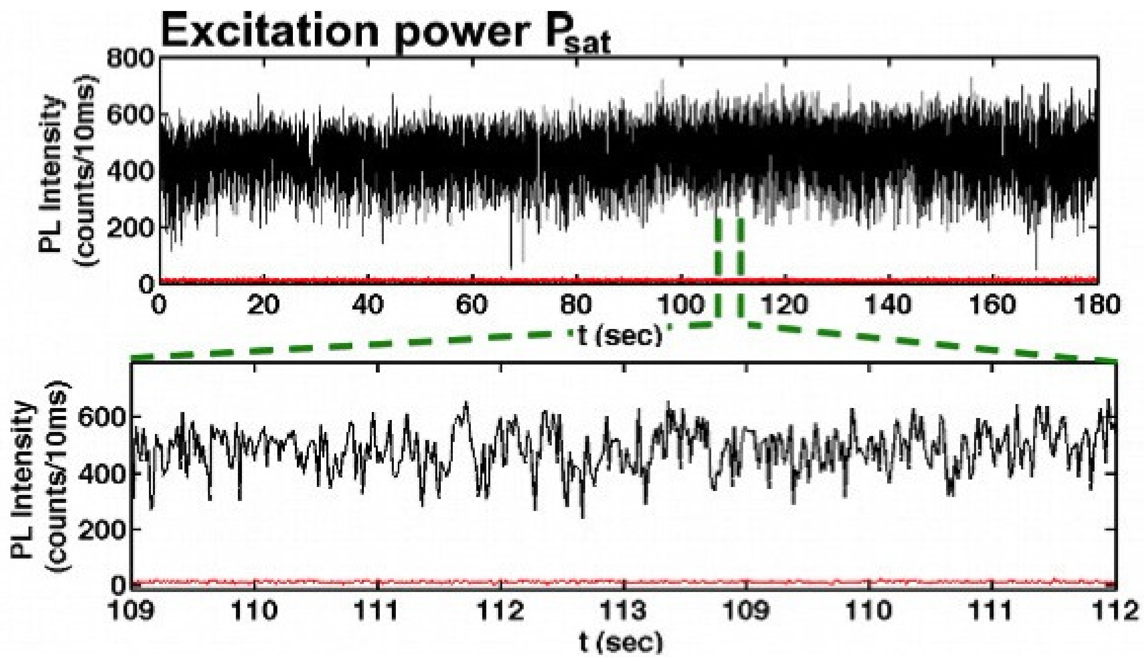
Core diameter d=2.7nm
Shell thickness t=7nm
Rod length l=22nm

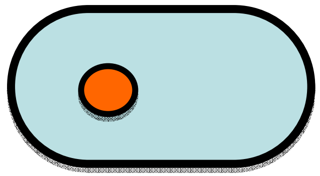
THE PROBABILITY TO HAVE BLINKING PERIODS LONGER THAN 50 ms IS AS LOW AS 0.001





Blinking-free DRs





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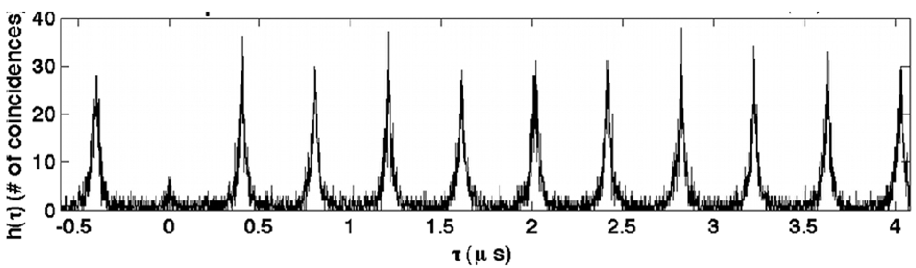
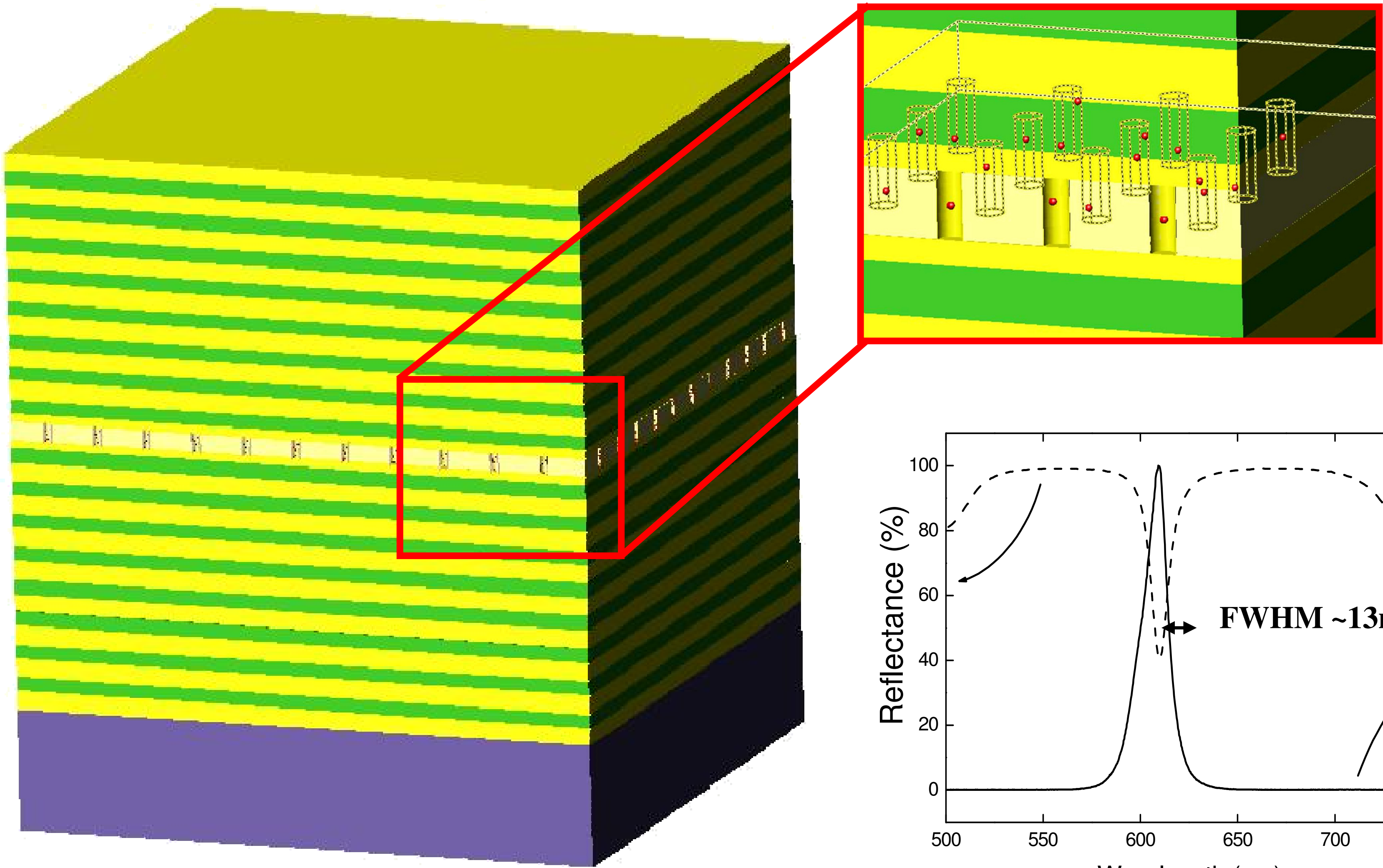
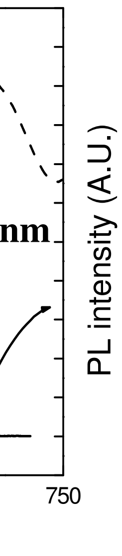


$$g^2(0) < 0.2$$

Nanocrystals in a microcavity

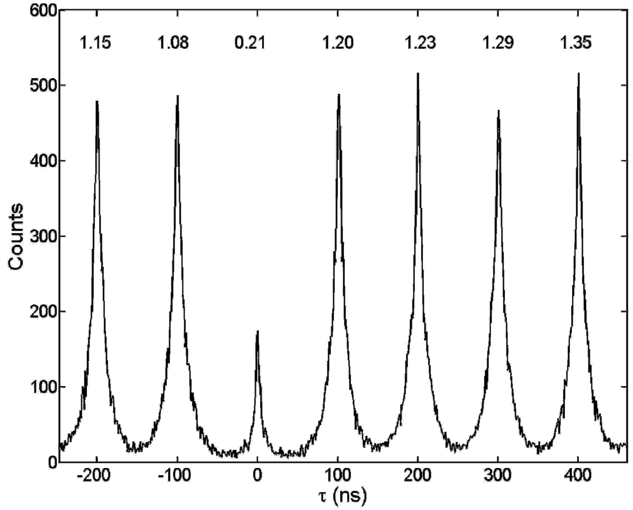
Nanocrystals in DBR cavity



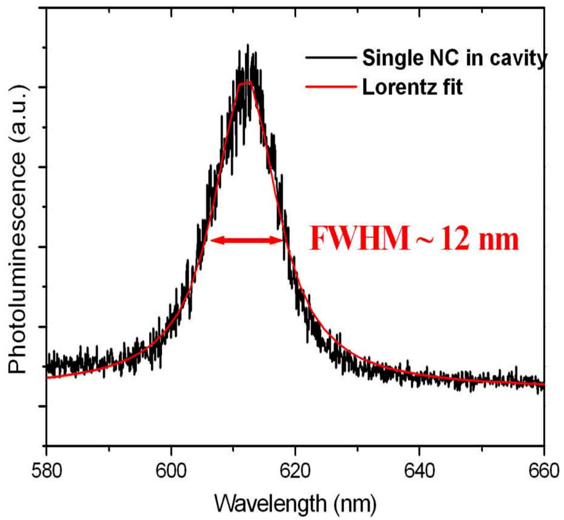


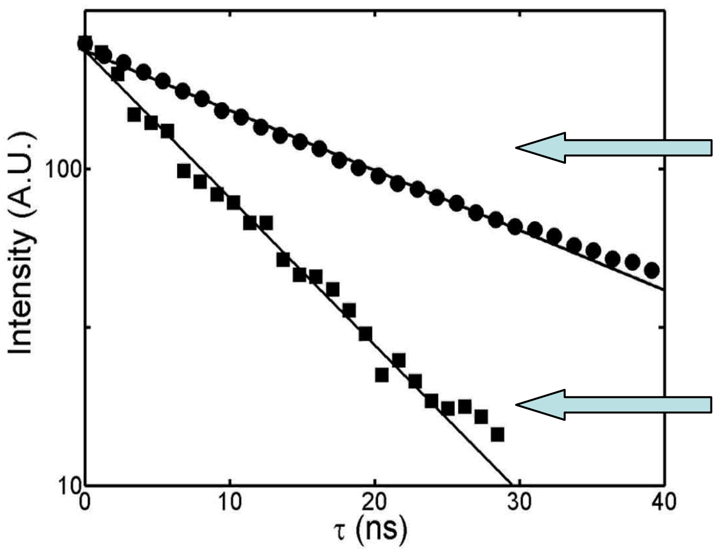
Nanocrystal in DBR cavity

<u>Antibunching</u>



<u>PL spectrum</u>: line narrowing



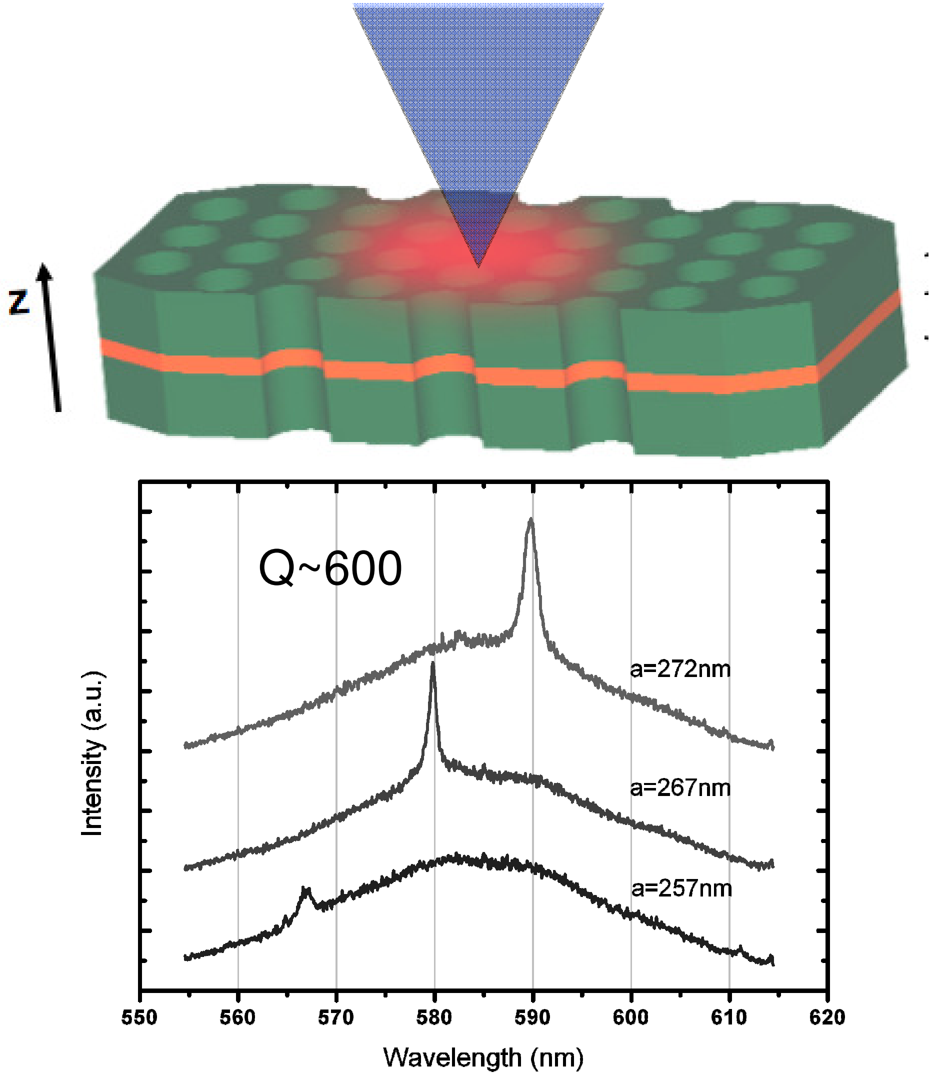


PL decay of free-space nanocrystals τ_{fs} ~ 23 ns

PL decay of a single nanocrystal in cavity: τ_{cav} ~ 9 ns

A. Qualtieri *et al., New J. Phys* 11, 033025 (2009).

Colloidal nanocrystals in a PhC cavity

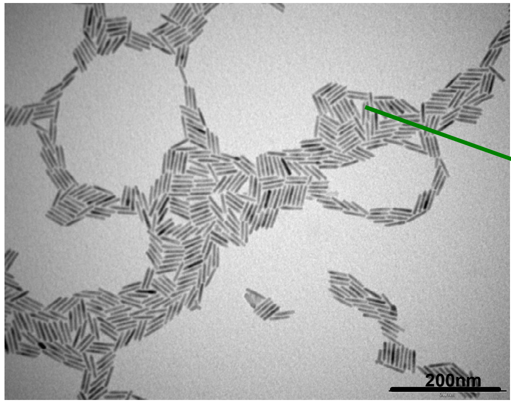


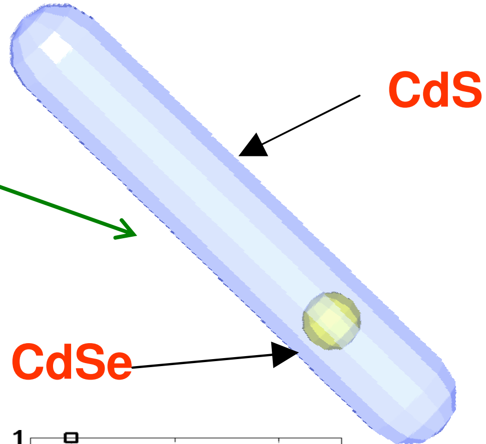
A. Qualtieri, F. Pisanello et al., Microel. Eng. **87**, 1435 (2010)

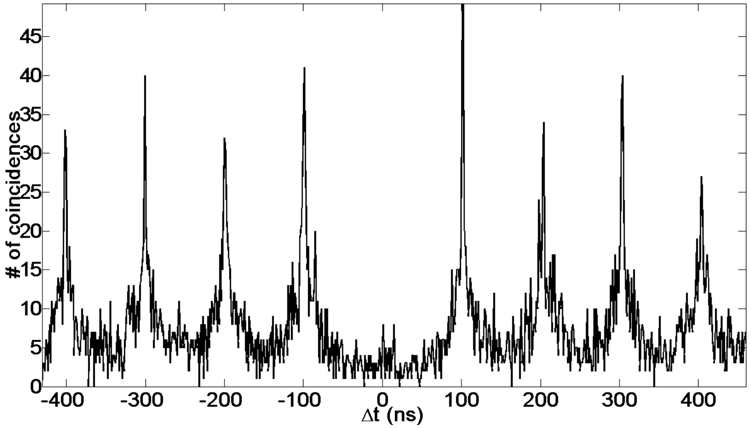
Conclusions

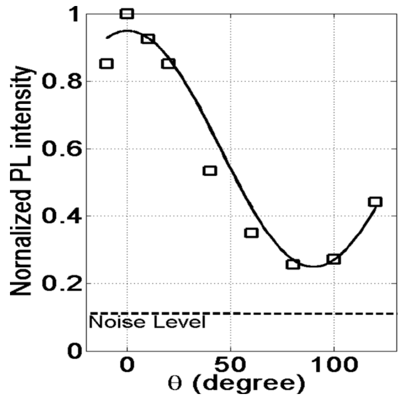
Single photon sources with semiconductor nanocrystals

-Colloidal dot in rod-
A novel and very promising structure for using NC as single-photon emitters

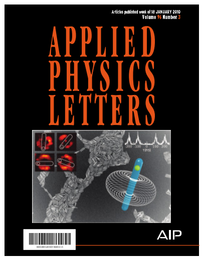






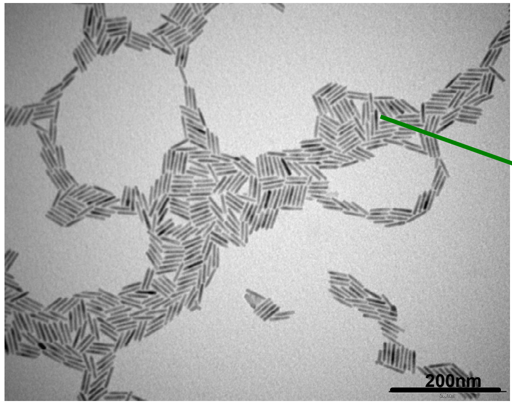


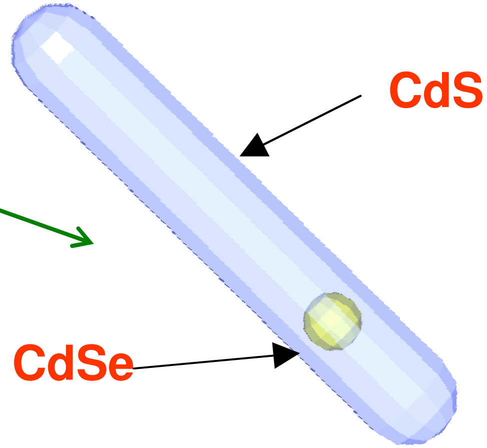
Very good single-photon sources with high degree of linear polarization

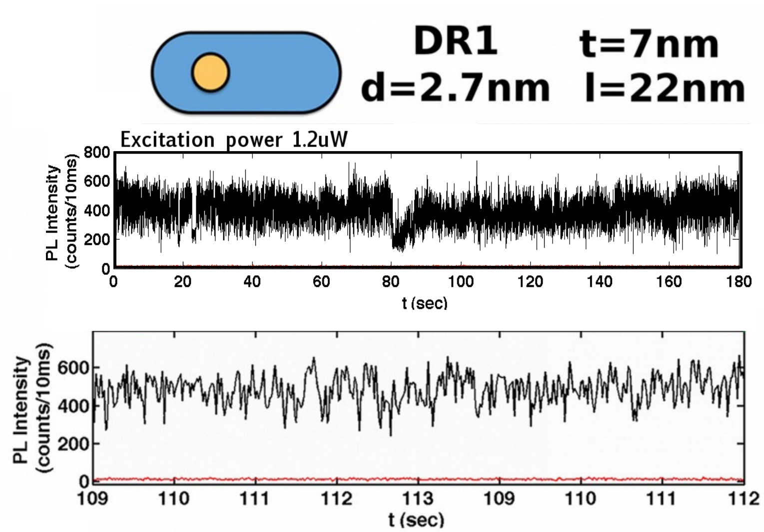


(APL 2010, Opt. Lett. 2010)

-Colloidal dot in rod-
A novel and very promising structure for using NC as single-photon emitters

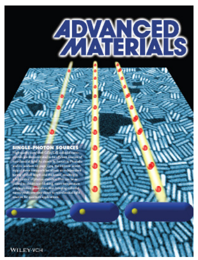






Another step: Thick shell

Demonstrate a strong reduction of blinking, while preserving a very low g2



Adv Material 2013, Opt. Comm. 2013