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FRIEDRICH-ALEXANDER
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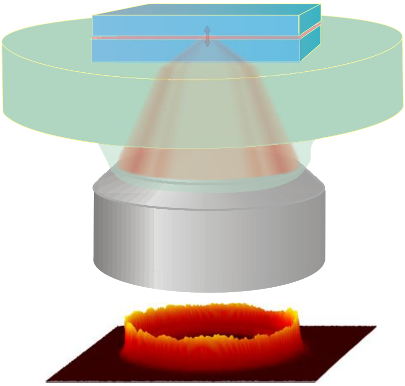
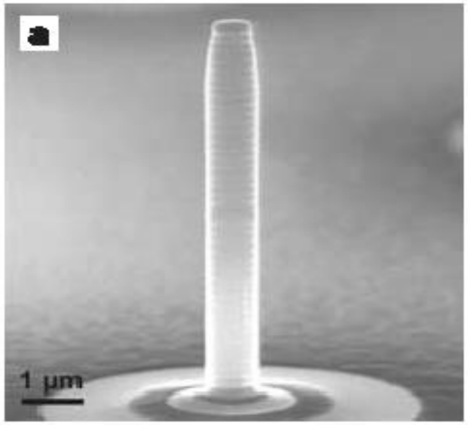
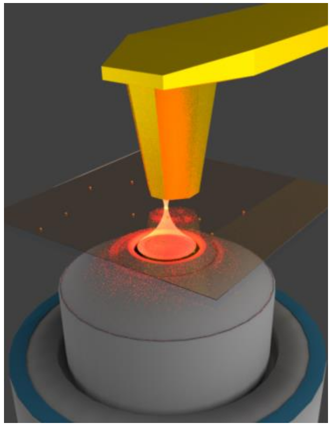
Quantum optics in nanosystems







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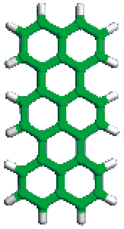
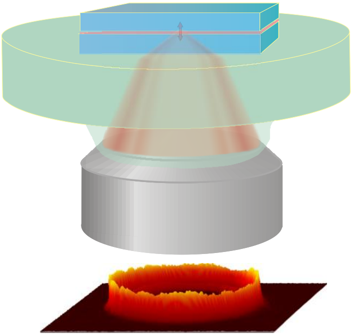
Outline

1. **Optical detection of a single solid state emitter (single-molecule microscopy)**

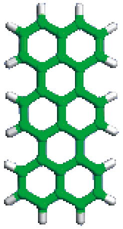


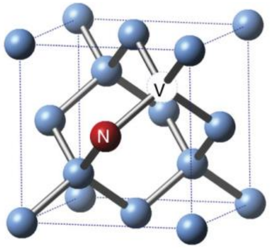
2. **Highly efficient single-photon sources: photon collection strategies**

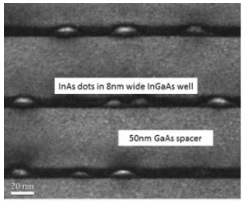


Part 1

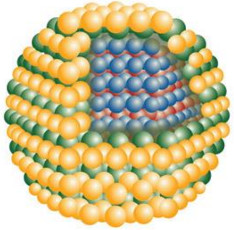
Optical detection of a single solid state emitter (single-molecule microscopy)

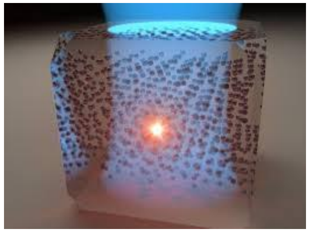






TEM image courtesy of Richard Beanland





molecule defect center quantum dots ion

Historical remark on single emitter experiments

E. Schrödinger: „Are there quantum jumps?“
Brit. J. Phil. Sci. 3, 109 (1952)

„… we *never* experiment with just *one* electron, atom or (small) molecule. In thought-experiments we sometimes assume that we do; this invariably entails ridiculous consequences…“

„In the first place it is fair to state that we are not experimenting with single particles, any more than we can raise Ichthyosauria in the zoo.“

A brief history of single emitter detection

1977: **antibunching** measured from single atom

1980s: scattered efforts to detect single molecules

1989: first single molecule detection: (detected in absorption)
liquid helium temp., high resolution spectroscopy (by W.E. Moerner)

1990: first single molecule detection in fluorescence (by M. Orrit)

1993: first spatially resolved single molecule detection
room temp., SNOM

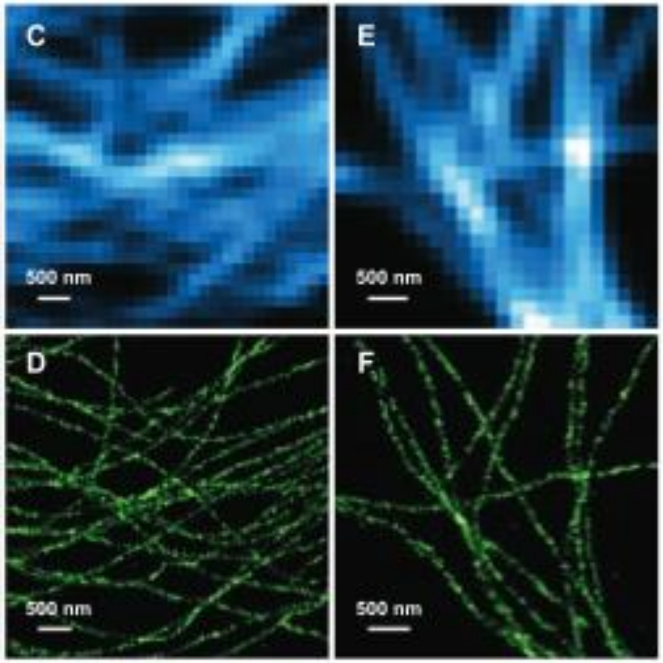
1994: first confocal room temperature detection of single molecules

1996: first single pair FRET

2000: Antibunching measured from single quantum dot and NV center

2006: Large area high resolution imaging based on colocalization of single
molecules (PALM, STORM, etc)

Super resolution microscopy



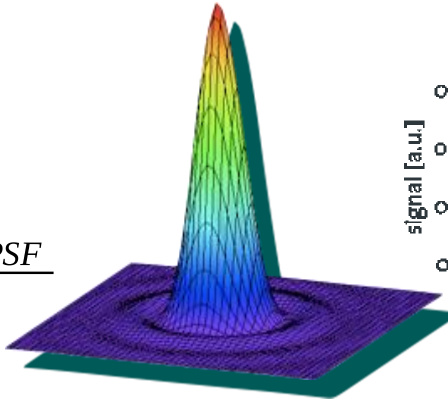
Multicolor Super-Resolution Imaging with Photo-Switchable Fluorescent Probes

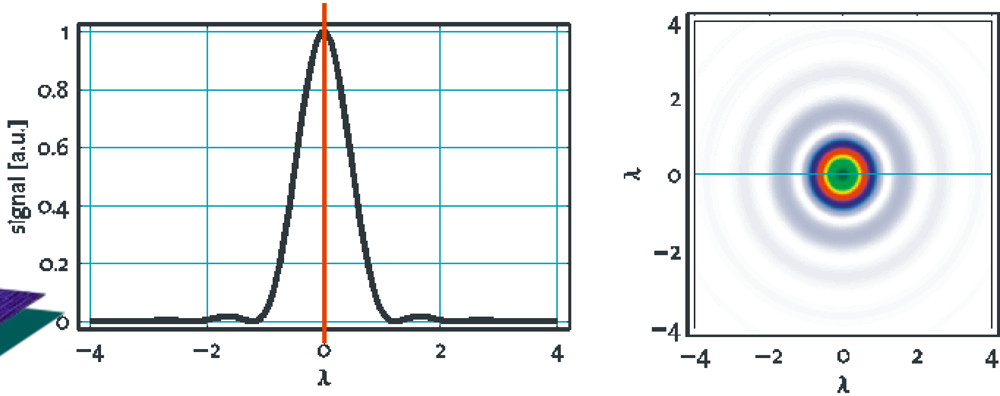
Mark Bates,[1] Bo Huang,[2,3] Graham T. Dempsey,[4] Xiaowei Zhuang[2,3,5]*

Localization and Colocalization

Localization:

“Resolution“: $\dfrac{FWHM_{PSF}}{\sqrt{N}}$

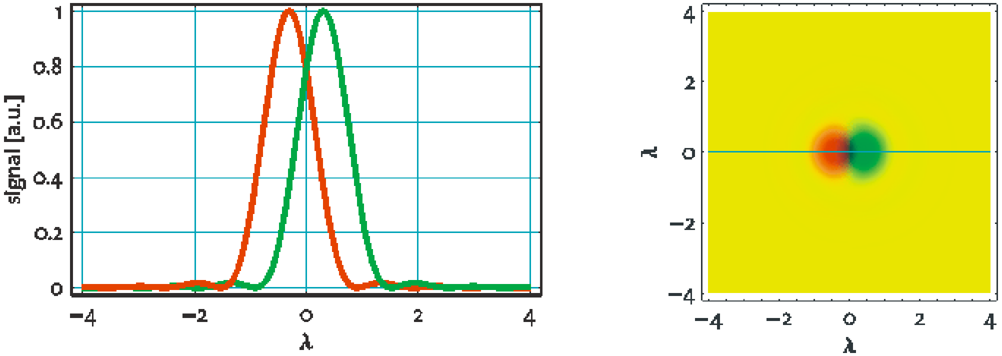




N: number of collected photons

Colocalization:

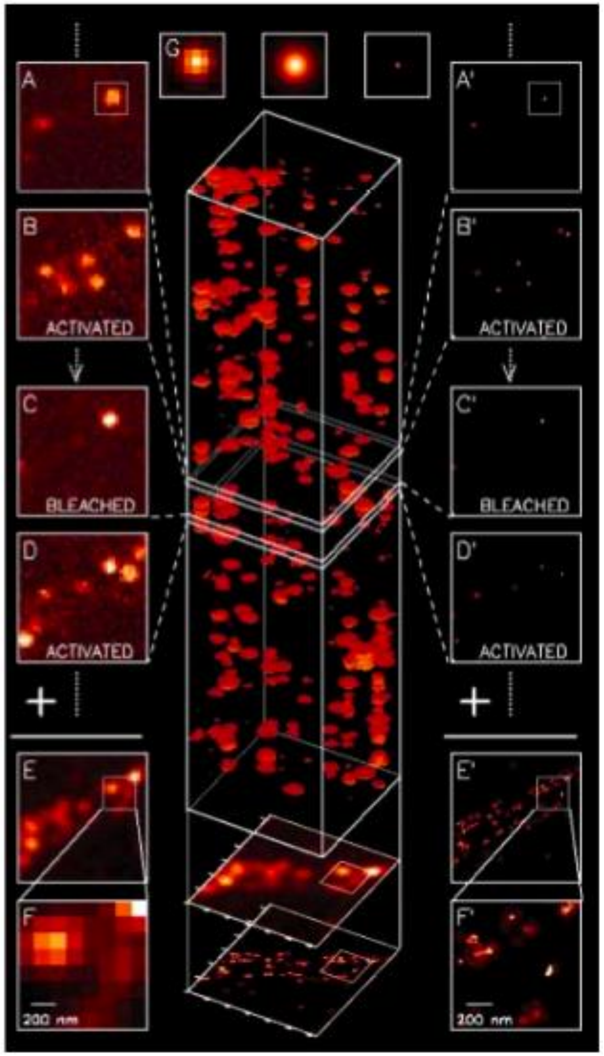
- spectral difference
- photobleaching

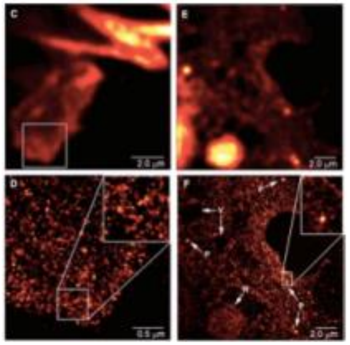


Basic idea: Determine center of point spread function as good as possible

PALM (photo-activated localization microscopy)

Betzig et al, Science. ***313****, 1642 (2006)*





chromophore:

EosFP switch from green (516 nm) to red (581 nm) upon UV (390 nm) irradiation

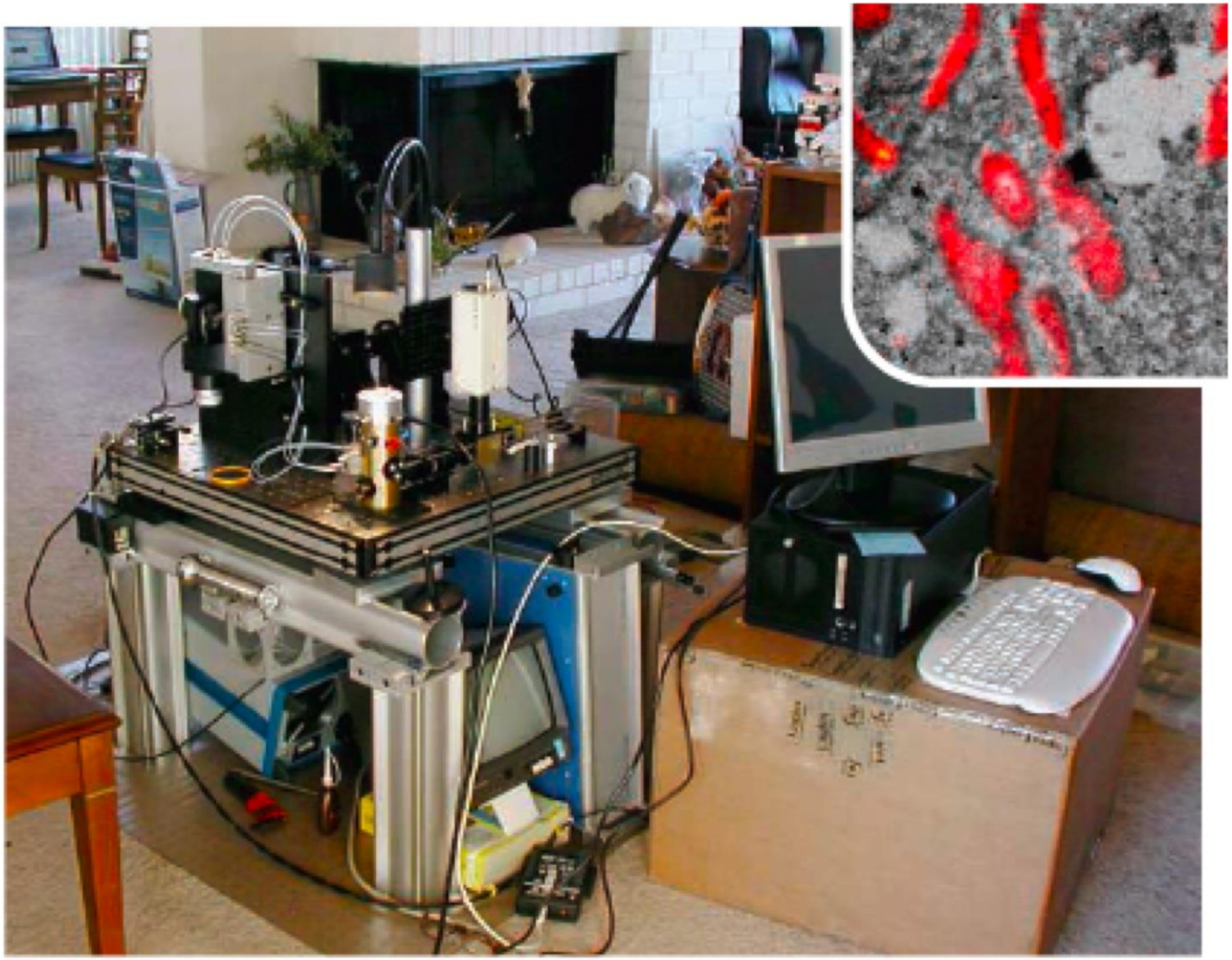
accuracy:

2-20 nm, depending on the number of collected photons

disadvantages:

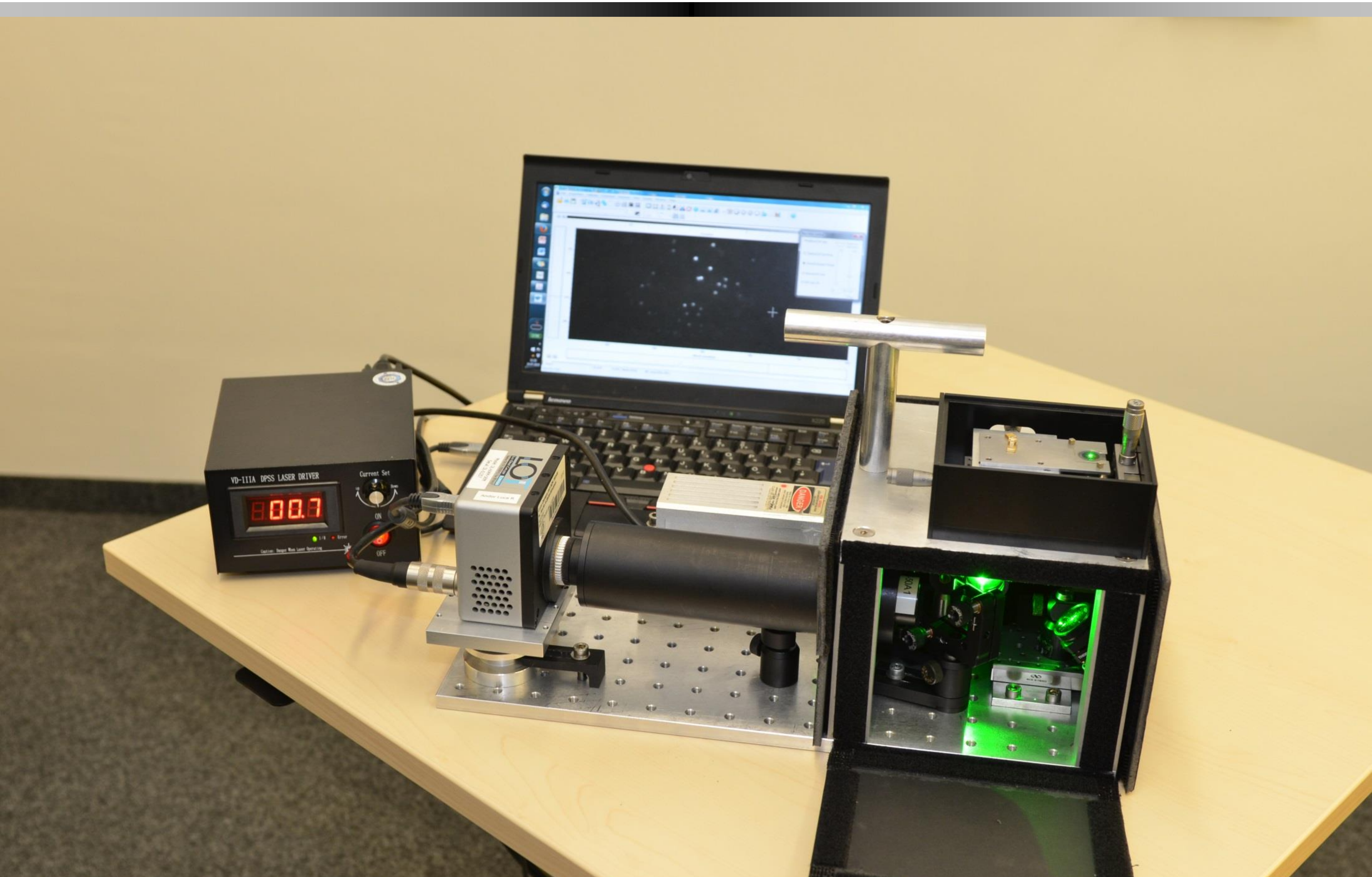
- **2 lasers needed**
- **slow acquisition** (2-12 h for 10^6 molecules)

E. Betzig`s nobel prize setup



Up close. A high-tech microscope, assembled in a living room (*above*), revealed molecules (red, *inset*) nanometers apart inside a cell's mitochondria.

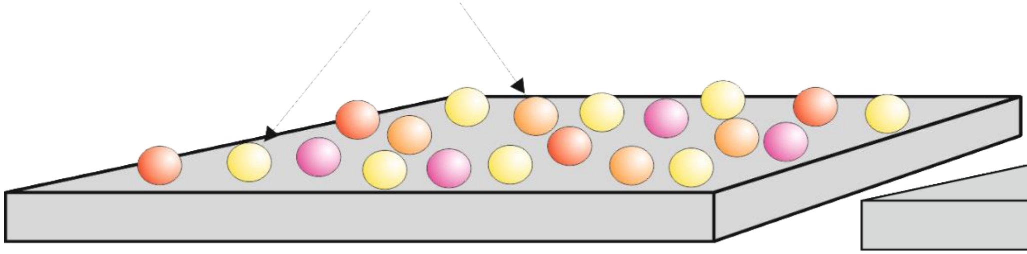
Historical remark on single emitter experiments



Single emitter microscopy (molecules as an example)

Sample requirements: molecules need to be distinguishable!

molecules



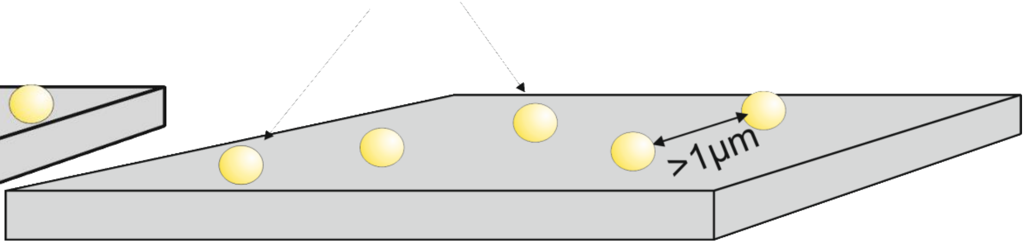
cover glass

spectral selection

or

temporal selection, only a few molecules fluoresce
at a time(see e.g. PALM)

molecules

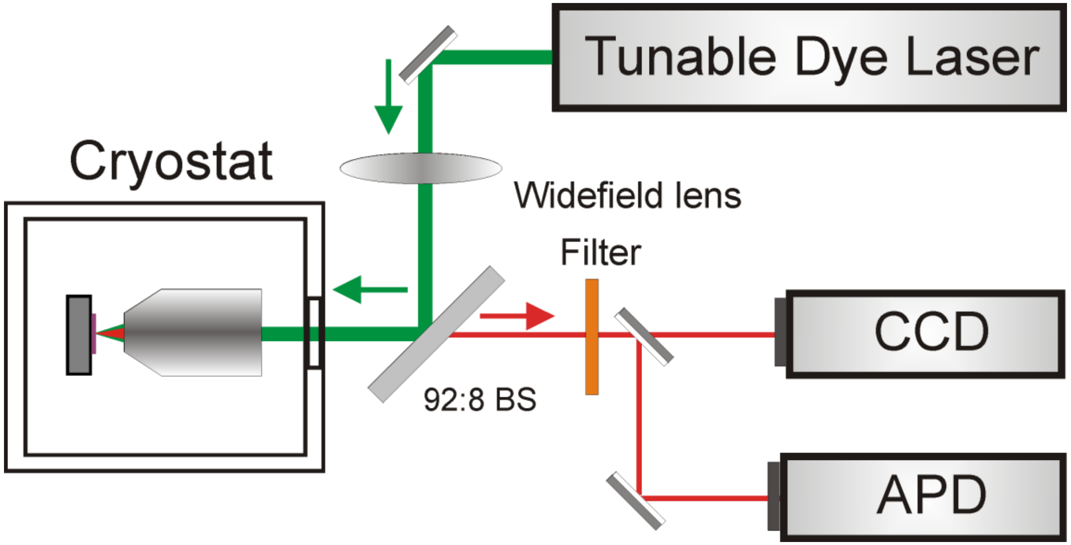


cover glass

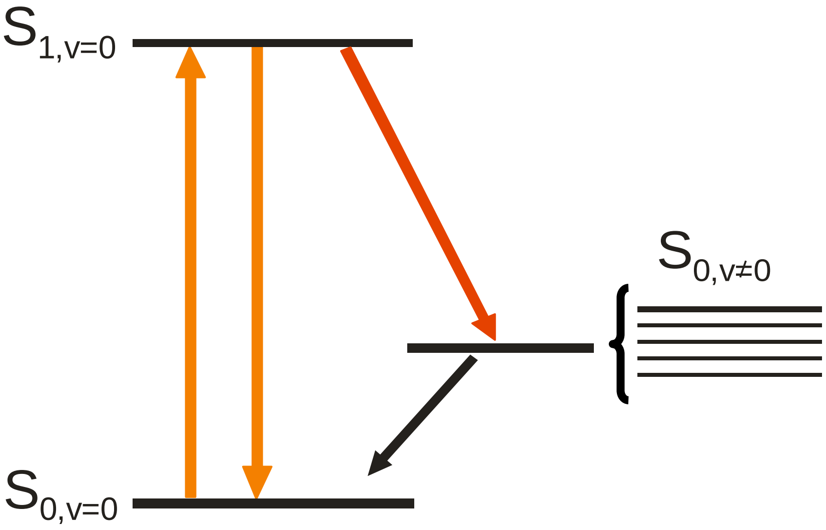
spatial selection

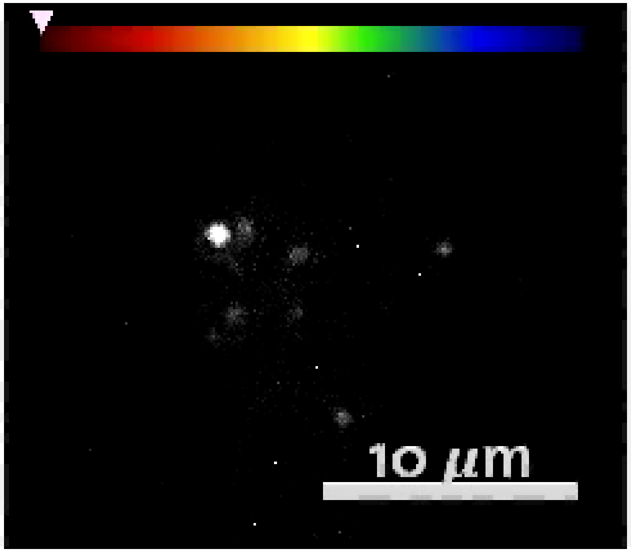
spatial selection can be achieved via a simple spin-coating process using a nanomolar solution

Single molecule spectroscopy (spectral selection)

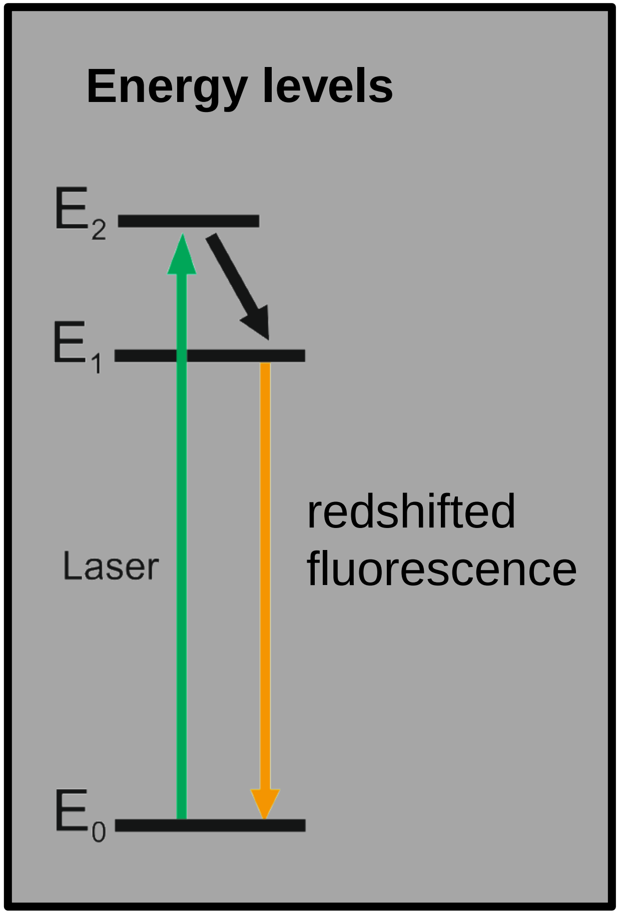


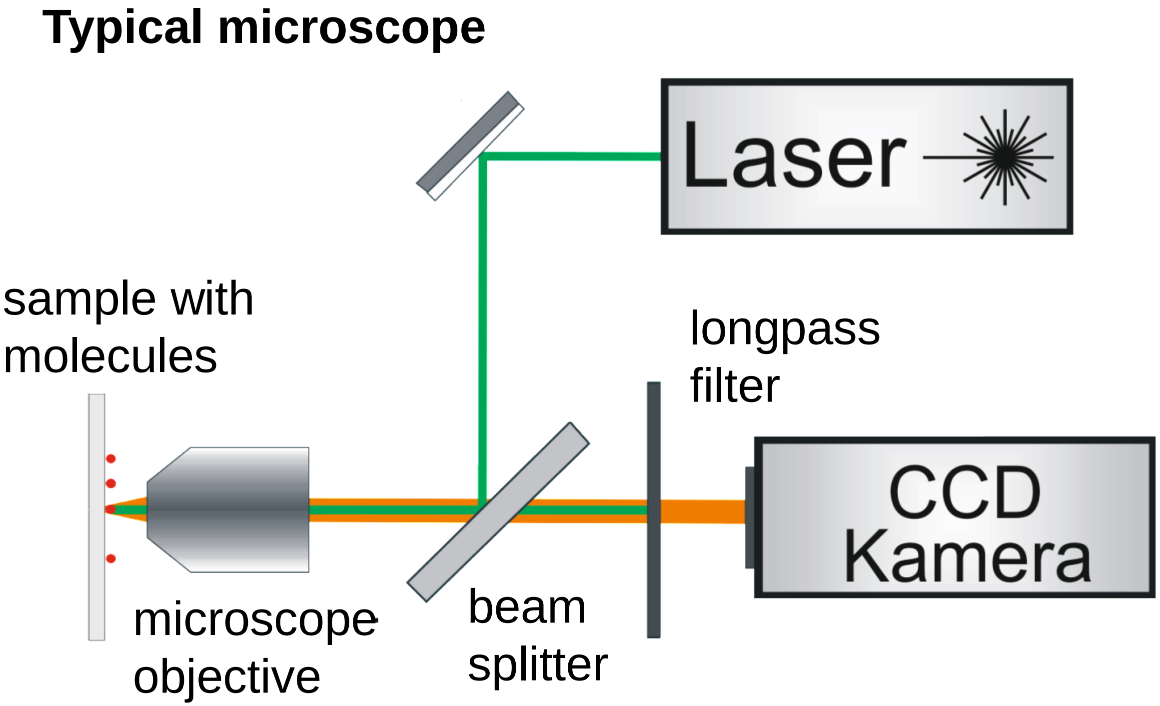
Emitter in a matrix show inhomogeneous broadening!





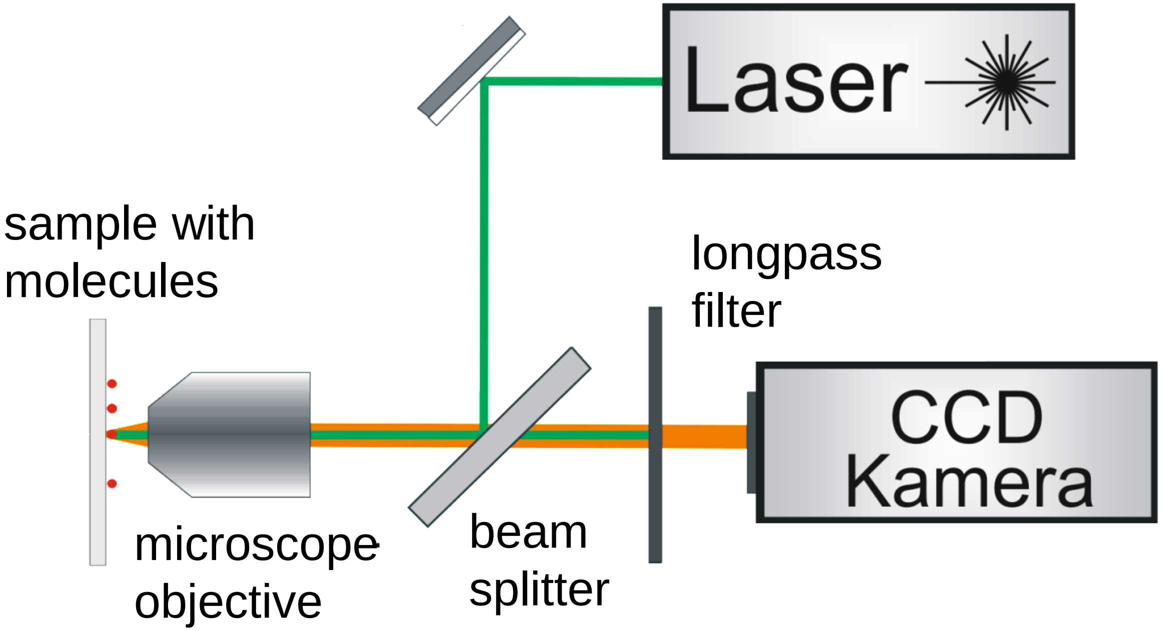
Single molecule microscopy (spatial selection)


Energy levels


Typical microscope

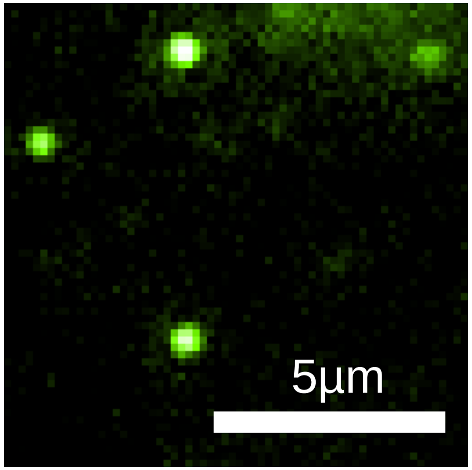
Single molecule microscopy

Typical microscope

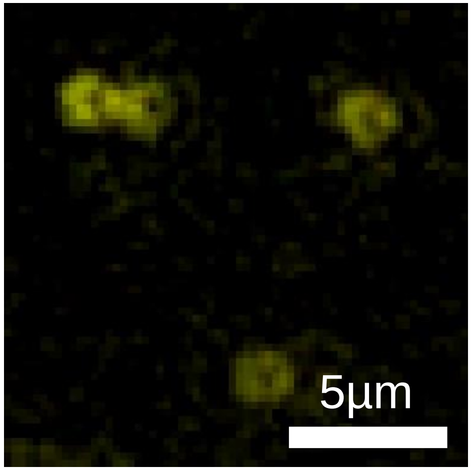


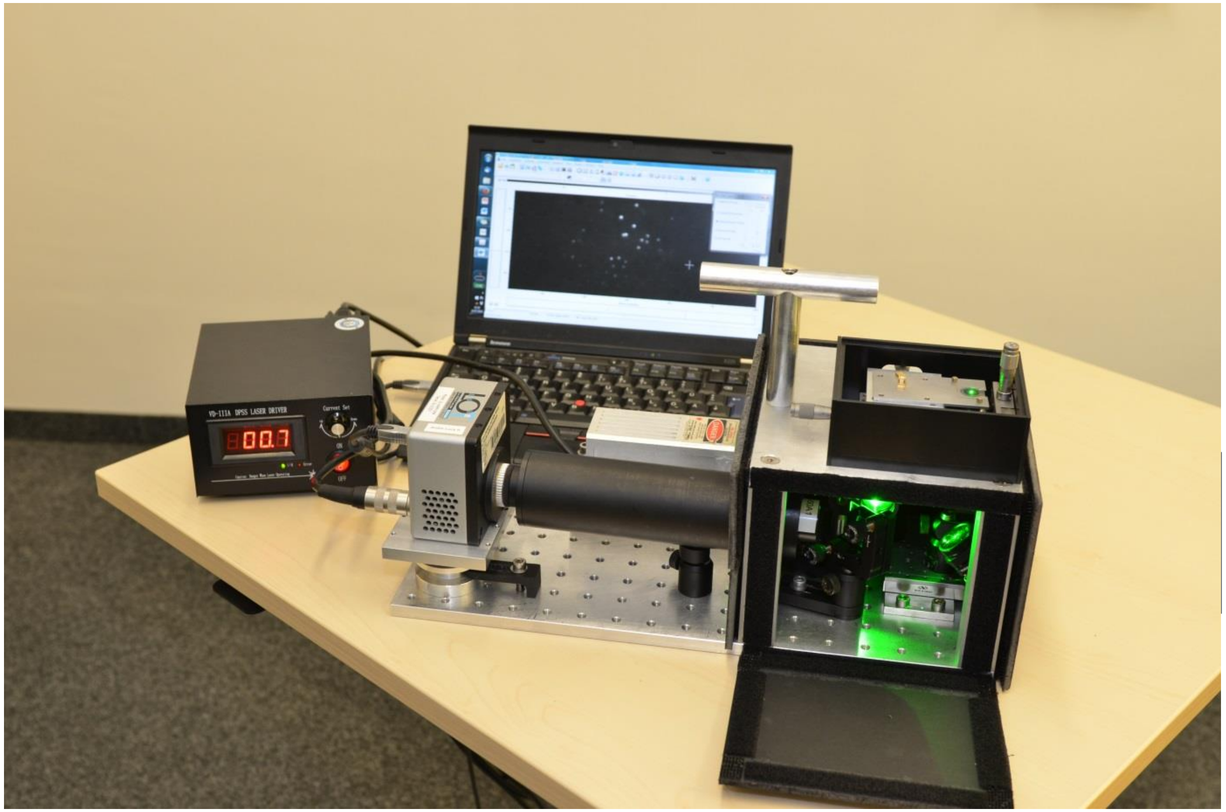
Single molecule microscopy

Alexa 532



Terrylene in p-Terphenyl



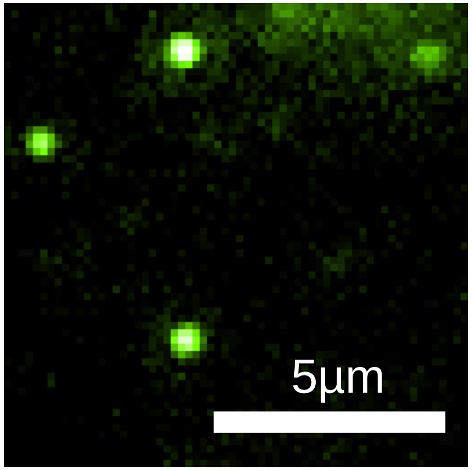


Size of the molecule: 1nm

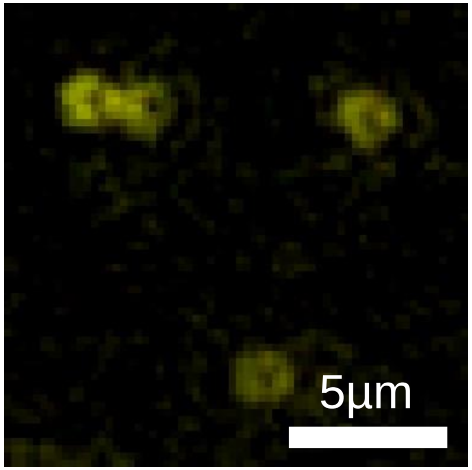
Camera image reveals orientation of the molecule!

Single molecule microscopy

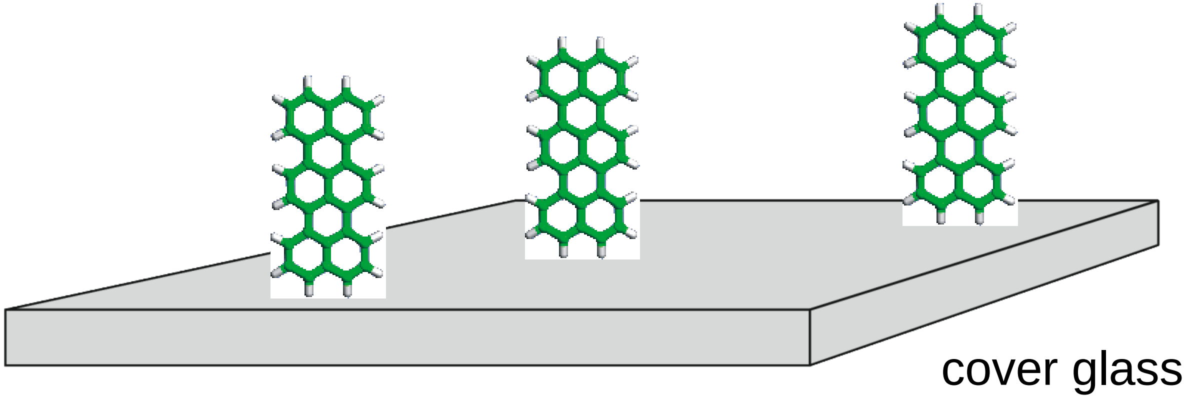
Alexa 532



Terrylene in p-Terphenyl



How can we be sure that it is a single molecule we are seeing?



Size of the molecule: 1nm

Camera image reveals orientation of the molecule!

Single-photon sources: Generating single photons

Theory:

$$a^{\dagger}|0\rangle = |1\rangle$$

Creation operator

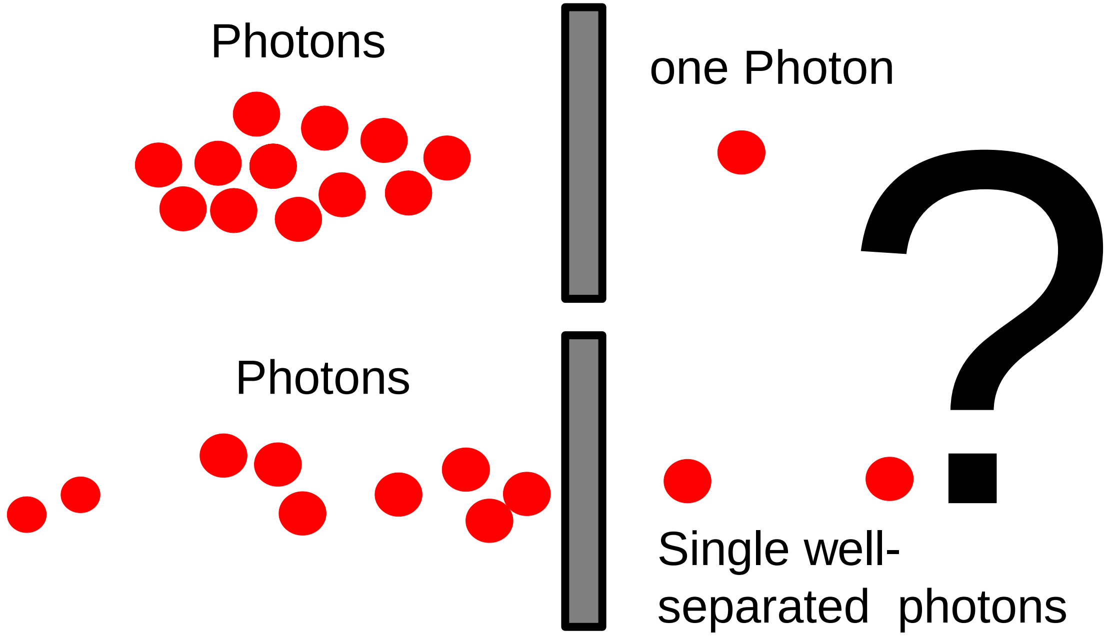
Vacuum

one photon (Fock state)

Experimental Realization:

Flash light (thermal source)

Laser



attenuator (grey filter)

Photons

one Photon

Photons

Single well-separated photons

Photon statistics of a laser beam

Lasers can be described by coherent states (Glauber states):

$$|\alpha\rangle = \sum_{n=0}^{\infty} e^{\frac{-|\alpha|^2}{2}} \frac{\alpha^n}{\sqrt{n!}} |n\rangle$$

$|n\rangle$: photon number state (or Fock state)

Therefore probability to find *n* photons in the mode obeys a Poisson distribution:

$$P_n = |\langle n|\alpha\rangle|^2 = e^{-|\alpha|^2} \frac{\alpha^{2n}}{n!} = e^{-\langle n\rangle} \frac{\langle n\rangle^n}{n!}$$

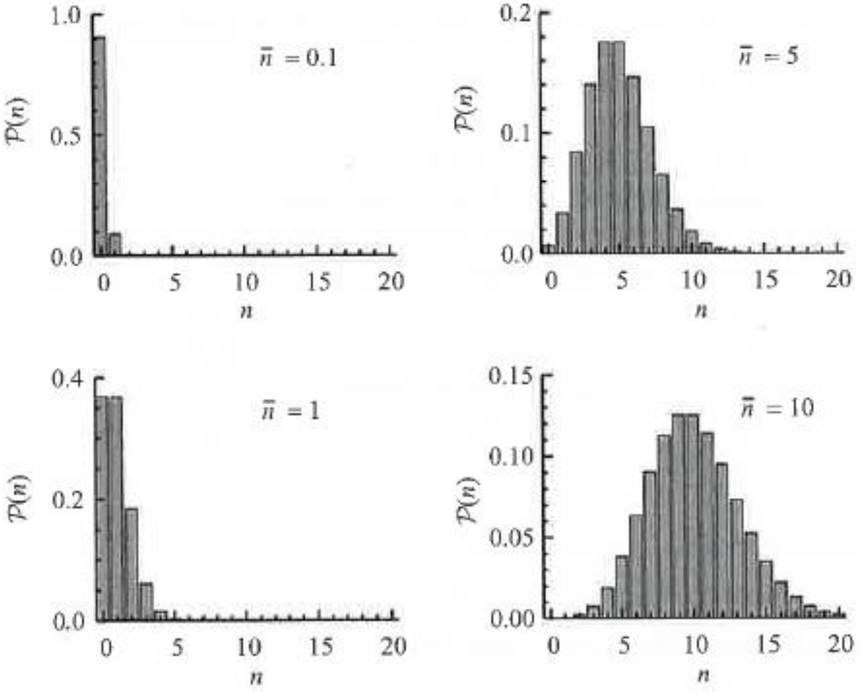
With mean photon number

$\langle n\rangle = \langle\alpha|\hat{n}|\alpha\rangle = \left\langle\alpha\middle|\hat{a}^\dagger\hat{a}\middle|\alpha\right\rangle = |\alpha|^2$

The variance is then also $\langle n\rangle$, Standard deviation $\sqrt{\langle n\rangle}$ (remember shot-noise)

Photon statistics of a laser beam

Poisson photon-number distribution of coherent states



M. Fox: Quantum optics: an introduction, Oxford

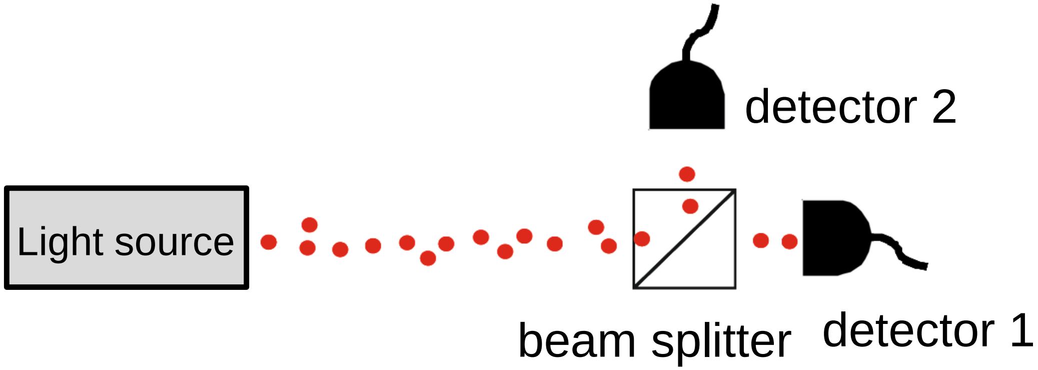
Impossible to generate exactly one photon!

Photon statistics:
How to identify a single-photon source

Method 1: Investigate noise of the source
(difficult, because losses and inefficiencies lead to noise)

Method 2:

Hanbury-Brown and Twiss Correlator



Intensity correlation

$$g^{2}(\tau)=\frac{\left\langle :a^{\dagger}(t)a^{\dagger}(t+\tau)a(t+\tau)a(t):\right\rangle}{\left\langle a^{\dagger}(t)a(t)\right\rangle^{2}}$$

Photon statistics of a laser beam

Second-order coherence function of a coherent state at τ=0

$$g^2\,(0) = \frac{\left\langle \hat{a}^\dagger \hat{a}^\dagger \hat{a}\hat{a} \right\rangle}{\langle \hat{a}^\dagger \hat{a} \rangle \langle \hat{a}^\dagger \hat{a} \rangle}$$

$$g^2\,(0) = \frac{\left\langle \alpha \middle| \hat{a}^\dagger \hat{a}^\dagger \hat{a}\hat{a} \middle| \alpha \right\rangle}{\left\langle \alpha \middle| \hat{a}^\dagger \hat{a} \middle| \alpha \right\rangle \left\langle \alpha \middle| \hat{a}^\dagger \hat{a} \middle| \alpha \right\rangle}$$

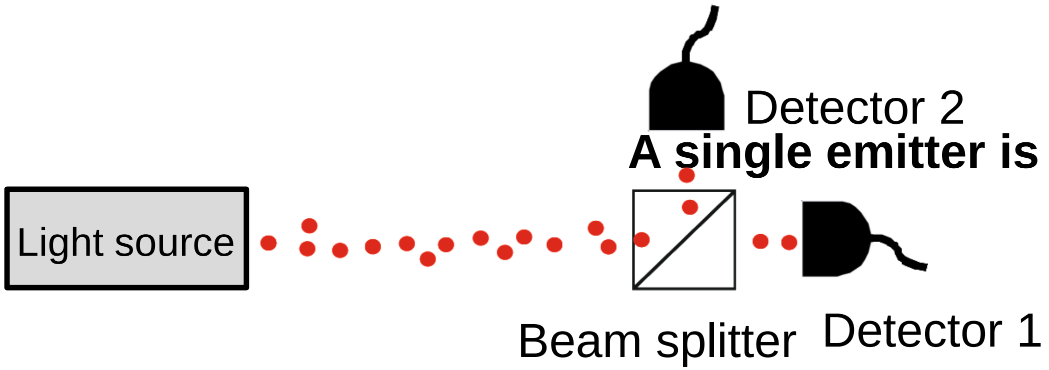
with: $\hat{a}|\alpha\rangle = \alpha|\alpha\rangle$

$\langle\alpha|\hat{a}^\dagger = \langle\alpha|\alpha^*$

$$g^2\,(0) = \frac{|\alpha|^2|\alpha|^2}{|\alpha|^2|\alpha|^2} = 1$$

Photon statistics:
How to identify a single-photon source

Hanbury-Brown and Twiss Correlator



A single emitter is needed

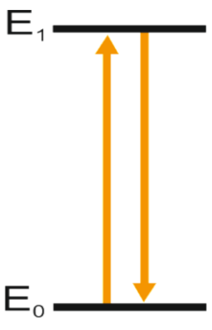
Intensity correlation

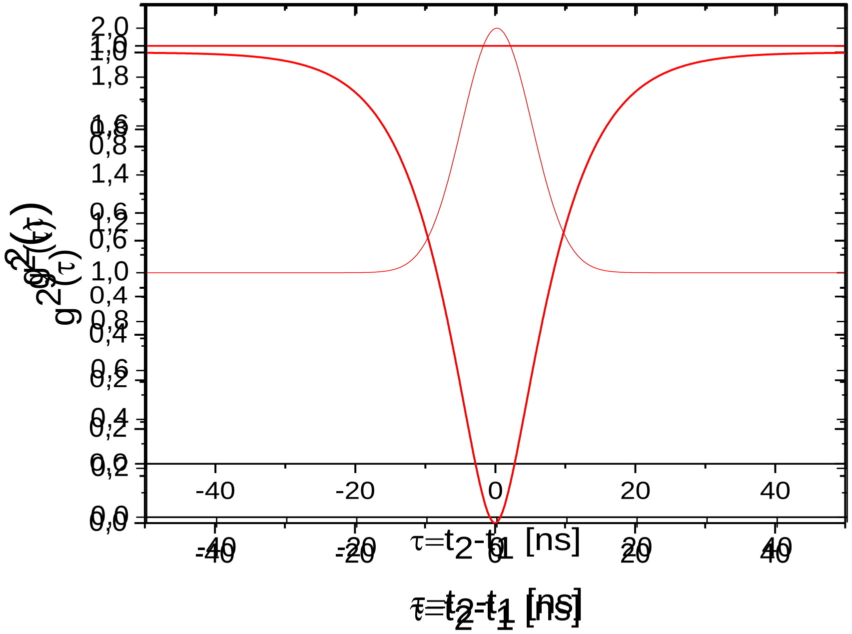
$$g^2(\tau) = \frac{\left\langle : a^\dagger(t) a^\dagger(t+\tau) a(t+\tau) a(t) : \right\rangle}{\left\langle a^\dagger(t) a(t) \right\rangle^2}$$

Laser: $g^{(2)}(\tau)=1$
Photons obey Poissonian statistics

Thermal source: $g^{(2)}(0)=2$
Photon bunching

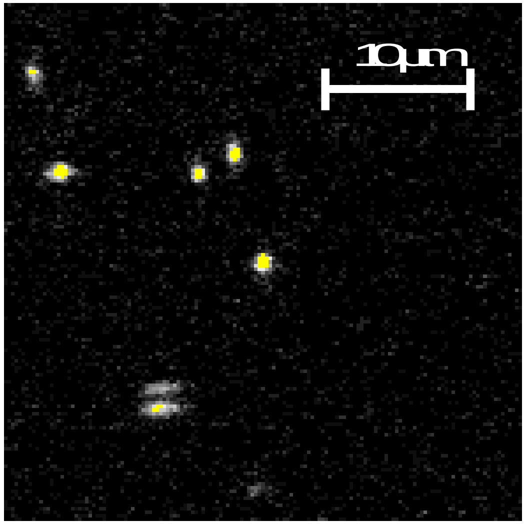
Single emitter: $g^{(2)}(0)=0$

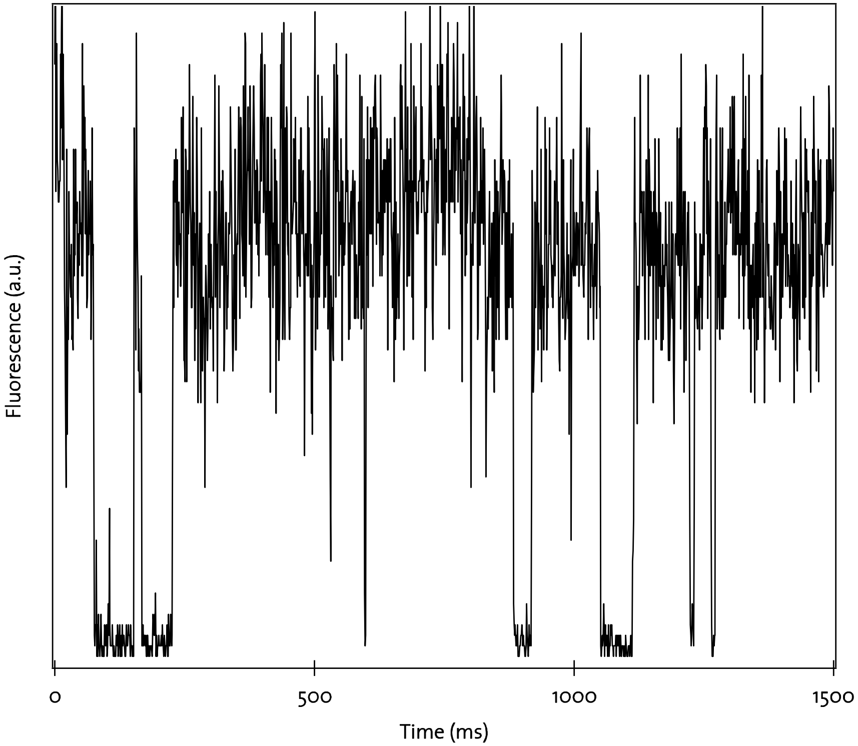




Blinking of a single emitter

Widefield image
of quantum dots

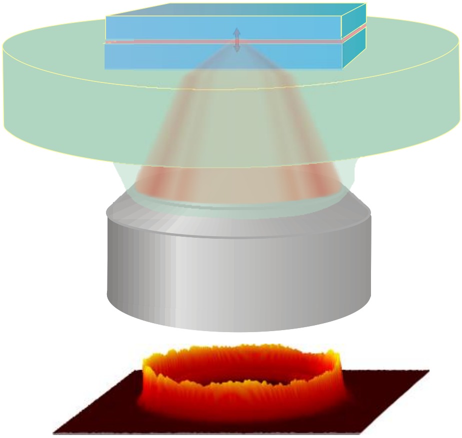




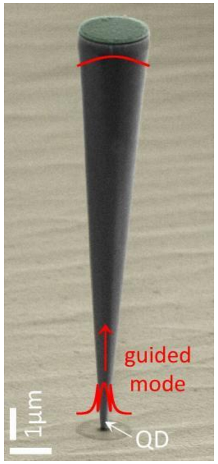
- Single spots clearly visible
- Diffraction limited spot originates from single quantum dots
- Quantum jumps (Blinking)

Part 2

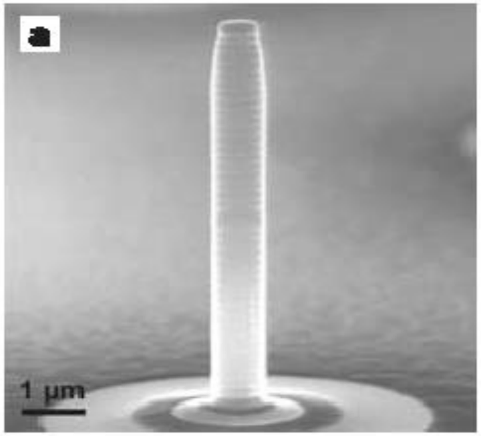
Highly efficient single-photon sources: photon collection strategies



planar dielectric antenna



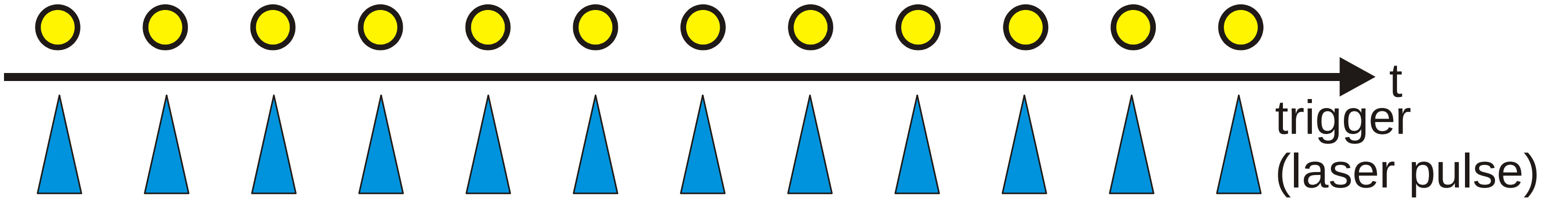
photonic trumpet



microcavity

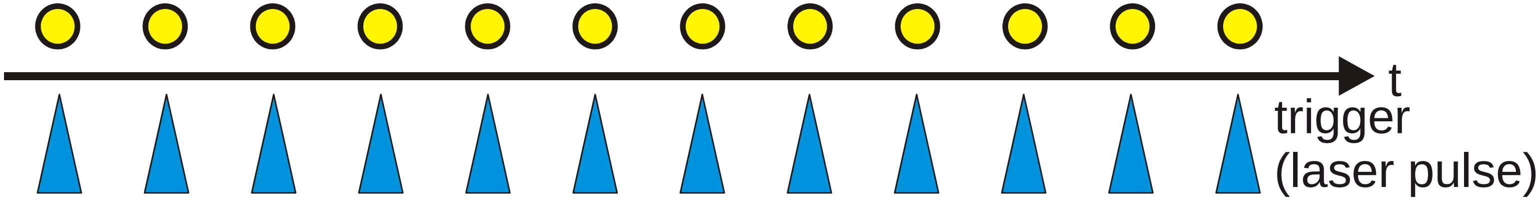
Introduction

Deterministic single-photon source:

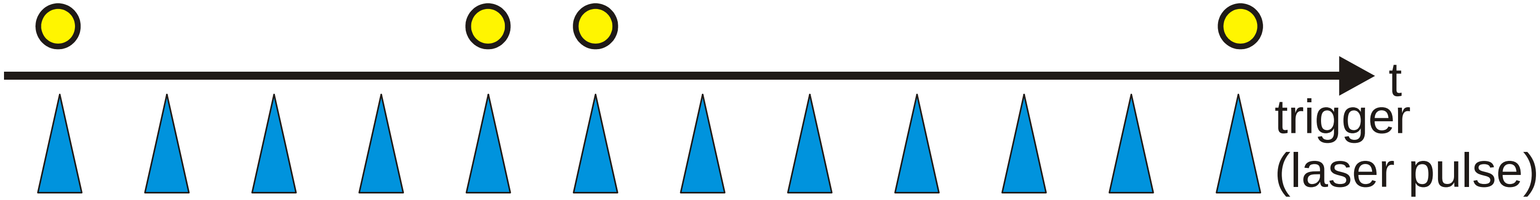


Introduction

Deterministic single-photon source:



Current single-photon sources:



Efficiency in the percent range!

Possible applications of single-photon sources with near-unity collection efficiency

Quantum information processing

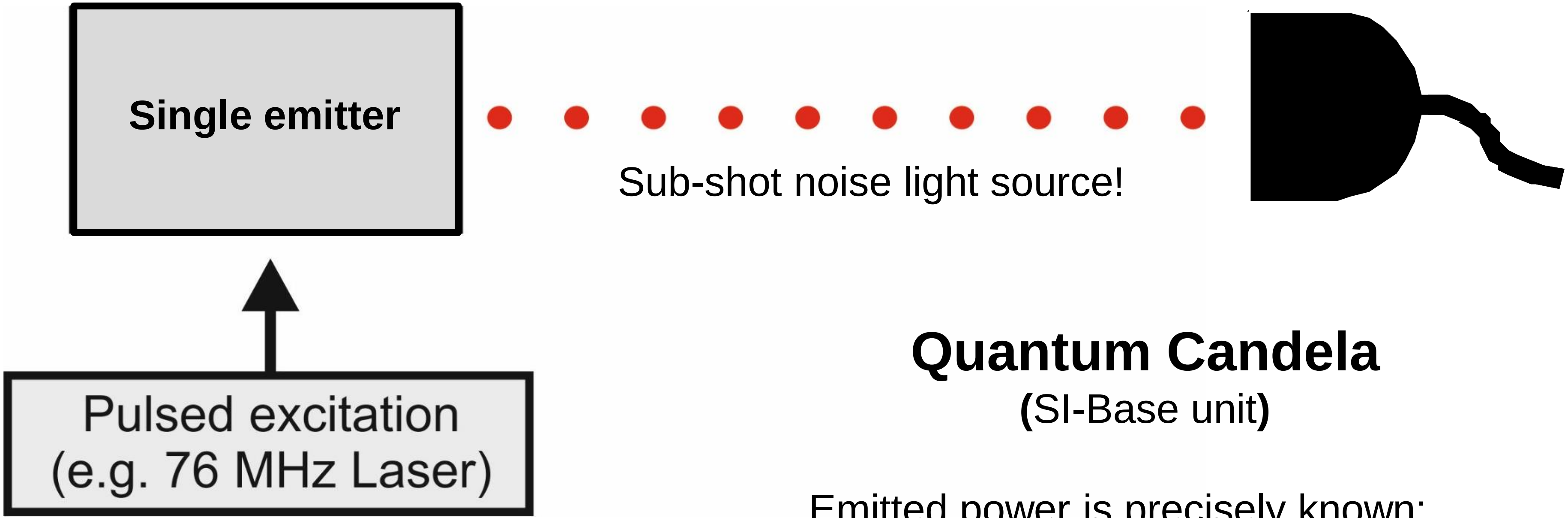
Quantum computer:

Can compute certain problems exponentially faster than a classical computer

Quantum cryptography: fundamentally secure data transmission

Possible applications of single-photon sources with near-unity collection efficiency

Intensity Squeezed light from a single emitter



Single emitter

Sub-shot noise light source!

Pulsed excitation
(e.g. 76 MHz Laser)

Quantum Candela

(SI-Base unit)

Emitted power is precisely known:

$$E_{photon}=h\,\nu$$

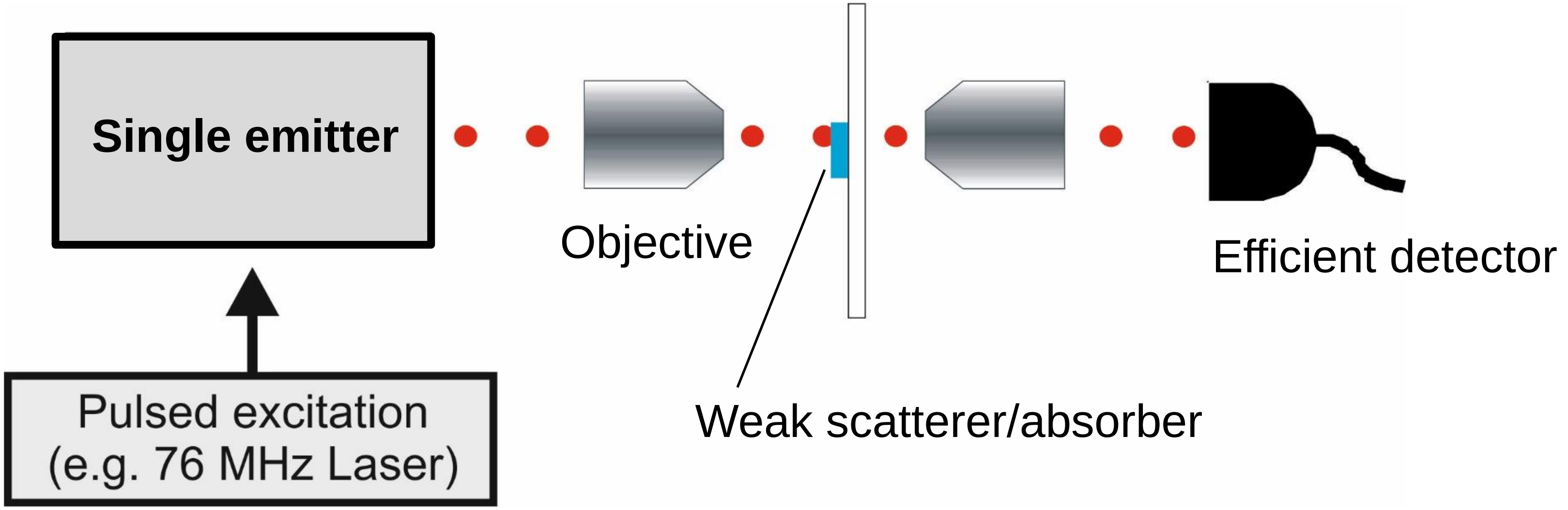
$$P= n\,h\,\nu/t$$

➡ **New primary intensity standard**

Single photon sources in metrology:
B. Rodiek *et al.*, Optica **4**, 71 (2017),
A. Vaigu *et al.*, Metrologia **54**, 218 (2017)

Possible applications of single-photon sources with near-unity collection efficiency

Quantum imaging
(Microscopy without shot noise)



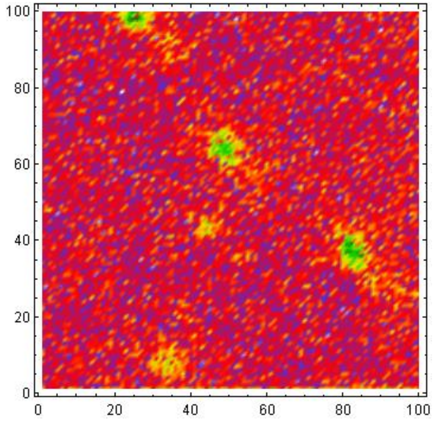
Single emitter

Objective

Efficient detector

Weak scatterer/absorber

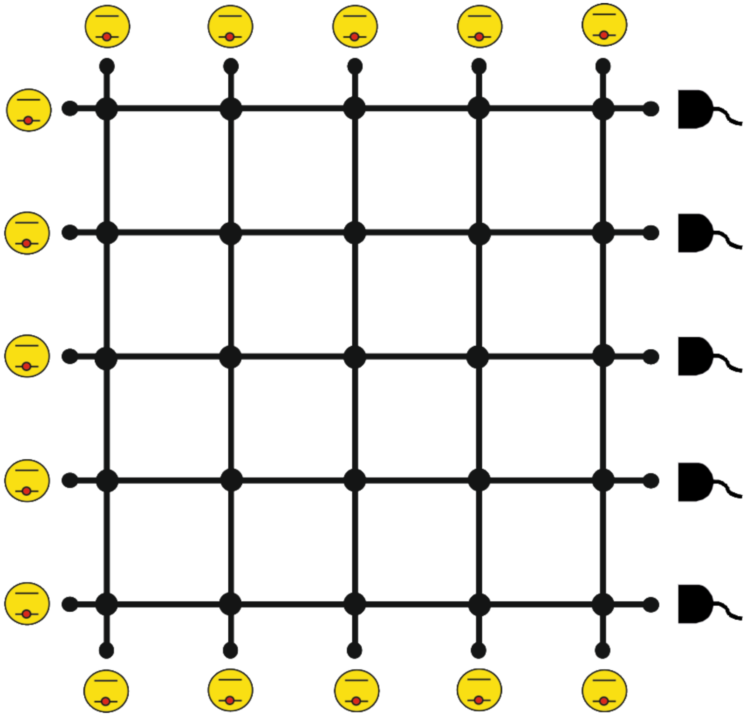
Pulsed excitation
(e.g. 76 MHz Laser)



First step taken:
Microscopy of single metallic nanoparticles
with a single molecule as light source

M. Celebrano *et al.*, Opt. Express **18**, 13829 (2010).

Effects of the efficiency on the realisation of quantum networks



Example:

Network with 1000 single-photon sources
(optical computer, quantum computer)

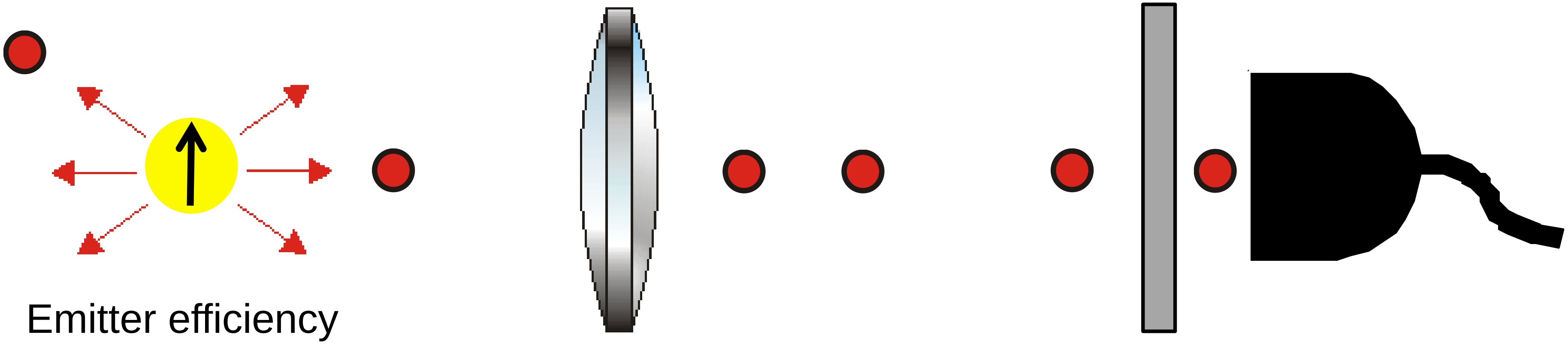
Efficiency 90%:

10^{46} attempts until success

about 10^9 tries per second possible!

Efficiency 99%:
$4*10^5$ attempts neccessary

Efficiency of a single-photon source



Losses in the set-up

Emitter efficiency
(quantum efficiency,
dark states)

Collection efficiency

Detection efficiency

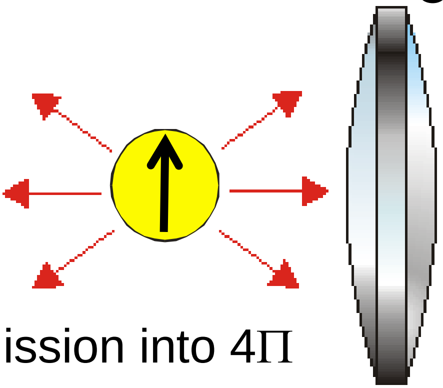
Remark on dark states:
Basically all solid state single-photon sources suffer from them

molecules:	NV centers:	quantum dots:
triplet state	singlet state	dark excitons

- need to be well characterized
- develop strategies to supress them

The problem: efficient collection of single photons

Free space



Collection lens

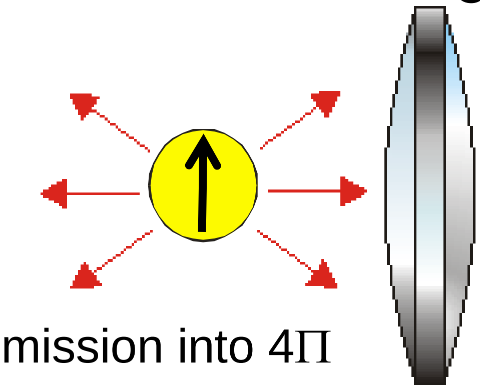
Emission into 4Π

Collection efficiency by definition <50%

Note: The problem of efficent collection is the inverse of an efficient interaction!

The problem: efficient collection of single photons

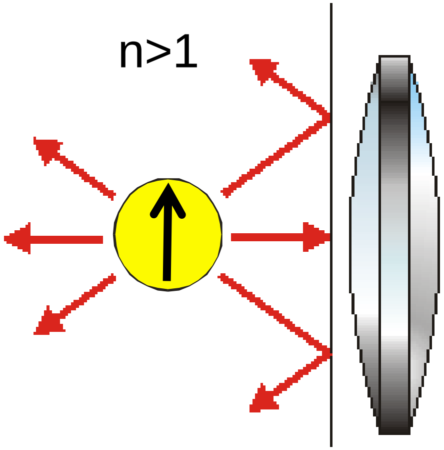
Free space



Collection lens

Collection efficiency by
definition <50%

Emission into 4Π



interface

n>1

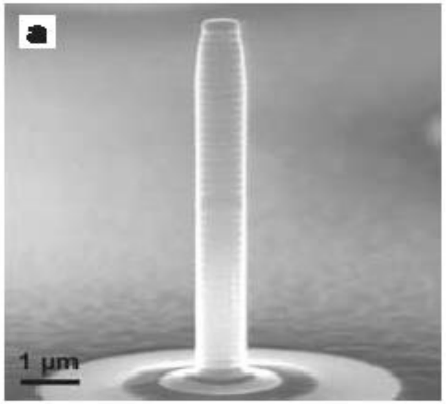
Emitter in solid state matrix

Total internal reflection
reduces collection angle further

e.g. GaAs: 3 %

Note: The problem of efficent collection is the inverse of an efficient interaction!

Photon collection strategies: microcavities (cavity quantum electrodynamics)



Micropillars:

Enhancement of spontaneous emission rate:

Purcell-factor: $F \sim Q/V$

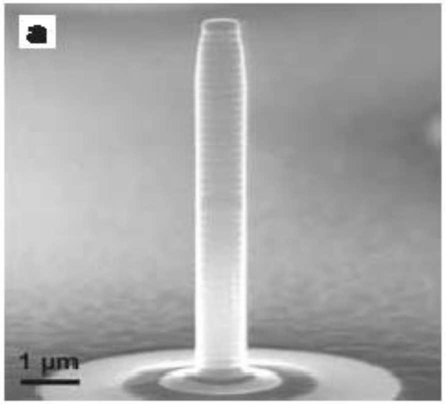
Q: quality factor
V: mode volume

Fraction of spontaneous emission coupled to cavity mode: $\beta = F/(F+1)$

D. Press, S. Götzinger, S. Reitzenstein, C. Hofmann, A. Löffler, M. Kamp, A. Forchel, Y. Yamamoto, Phys. Rev. Lett. **98**,117402 (2007).

- Purcell-factor 61
- Funnel photons into cavity mode
- Coupling to mode 97%
- Outcoupling efficiency: few %

Photon collection strategies: microcavities (cavity quantum electrodynamics)



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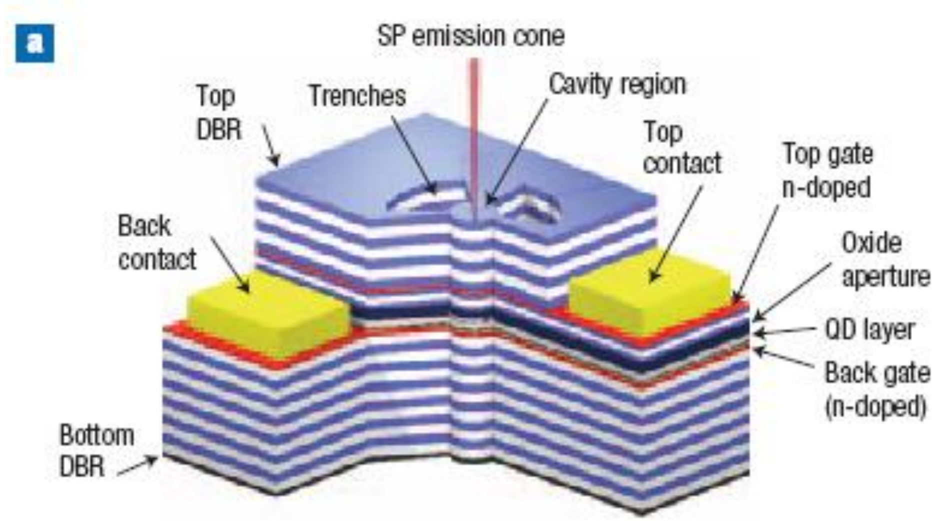
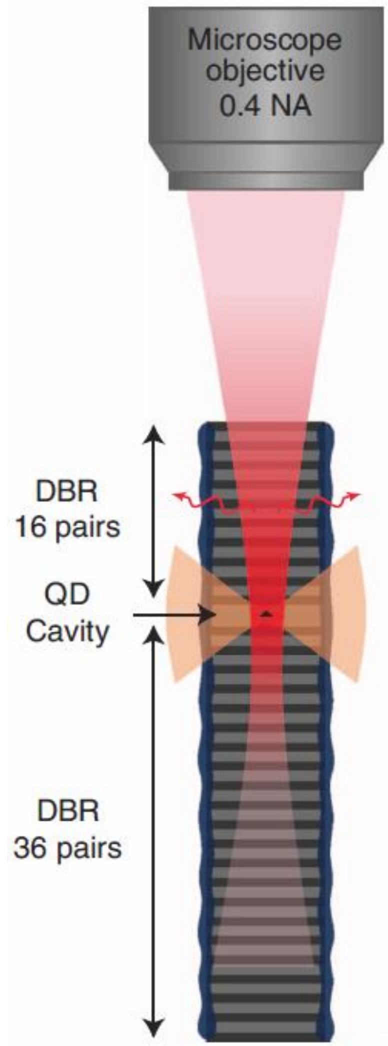


Performance:

- 31 MHz collection rate
- collection efficiency 38%
- 4 MHz detection rate

S. Strauf *et al.*, Nature Photonics **1**, 704 (2007).

Photon collection strategies: microcavities (cavity quantum electrodynamics)



- *In-situ* fabrication technique for spatial and spectral cavity-emitter alignment

- Efficiency: **0.79**
- Indistinguishability: **0.82**

☺

- Highly challenging fabrication process

☹

- Photon detection rate <1Mhz

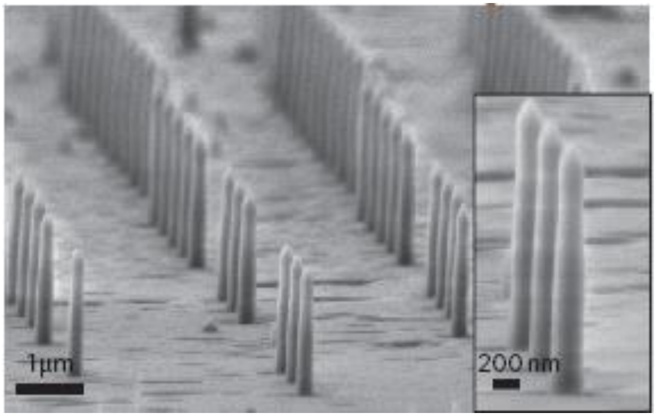
☹

Gazzano et al, Nat. Comm. **4**, 1425 (2013).

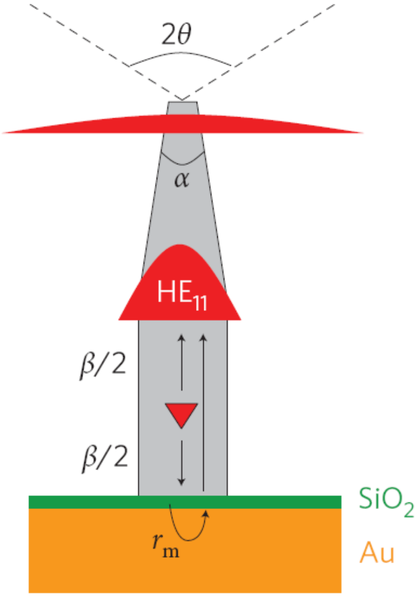
Photon collection strategies: nanowires

NV center in Diamond:

Babinec *et al.*, Nature Nanotech. **5**,195 (2010).



QDs in GaAs:



- Far-field emission pattern tailored with integrated bottom mirror and top conical taper
- adiabatic conversion of HE$_{11}$ into a strongly deconfined mode (Gaussian mode)

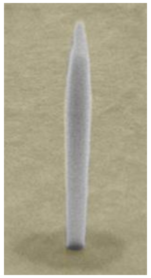
Claudon *et al.*, Nature Phot. **4**,174 (2010)
Friedler et al, Opt. Express **17**, 2095 (2009)
Gregersen et al, Opt. Lett. **33**, 1693 (2008)

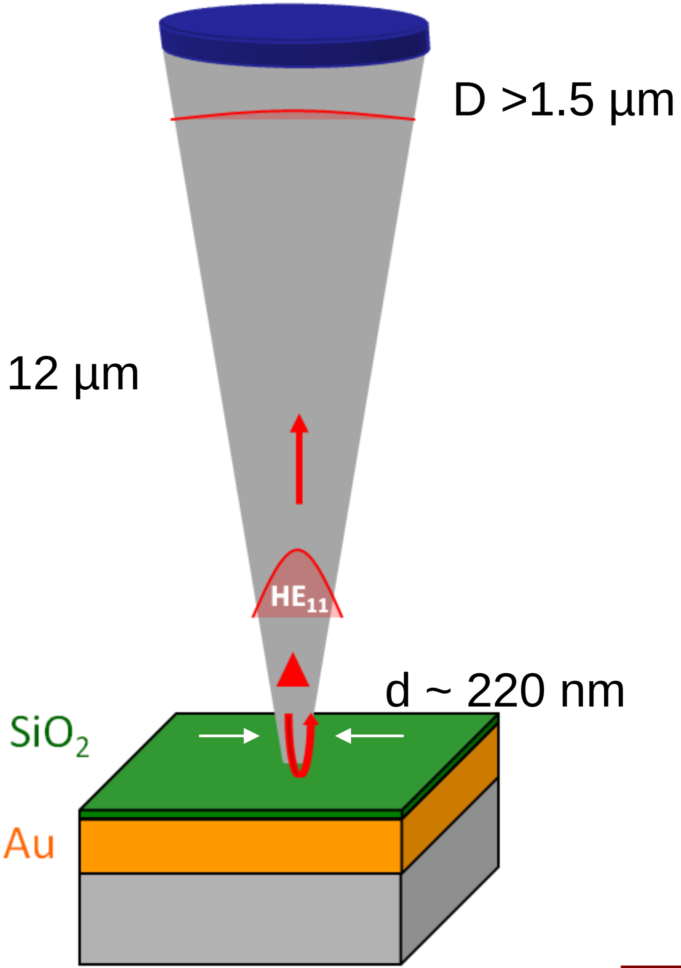
Collection efficiency: 72%

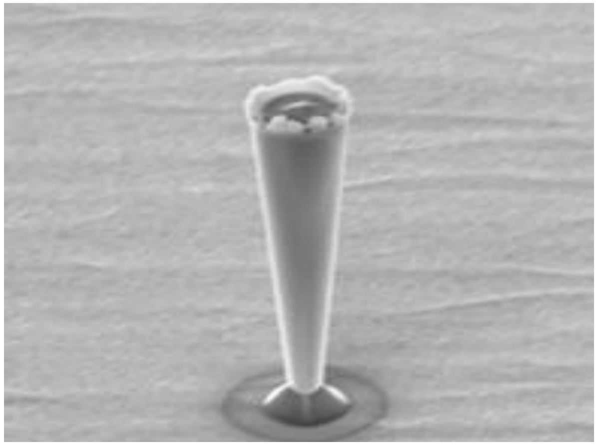
$g^{(2)}(\tau = 0)$: $\underline{0.008}$

Photon collection strategies: photonic trumpet

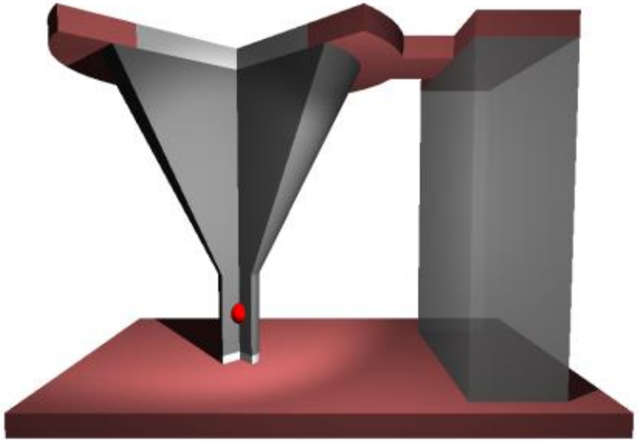
Inverted ”trumpet” taper







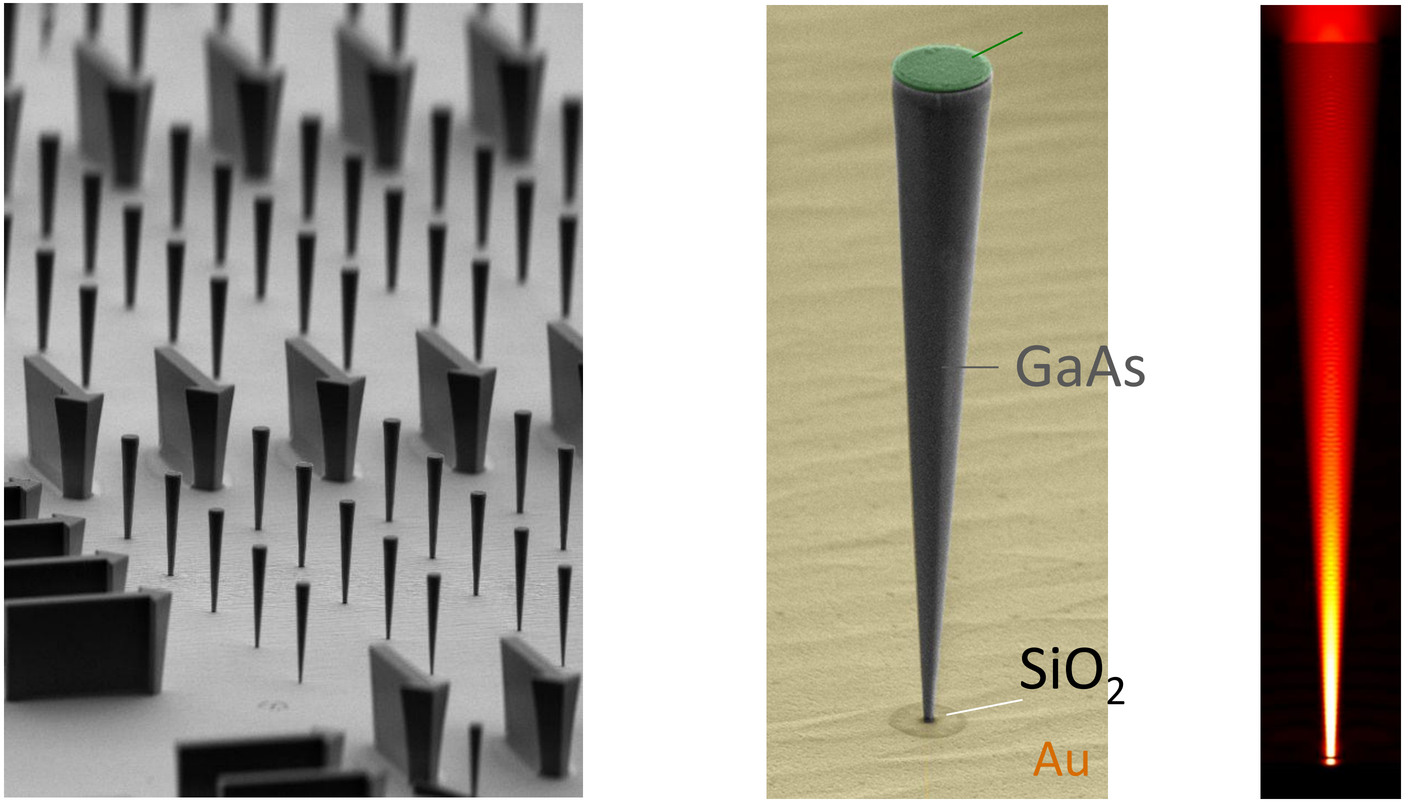
Compatible with electrical contacting!



Gregersen et al, Opt. Express **18**, 21204 (2010)

Photon collection strategies: photonic trumpet

Fabricated photonic trumpet SPS

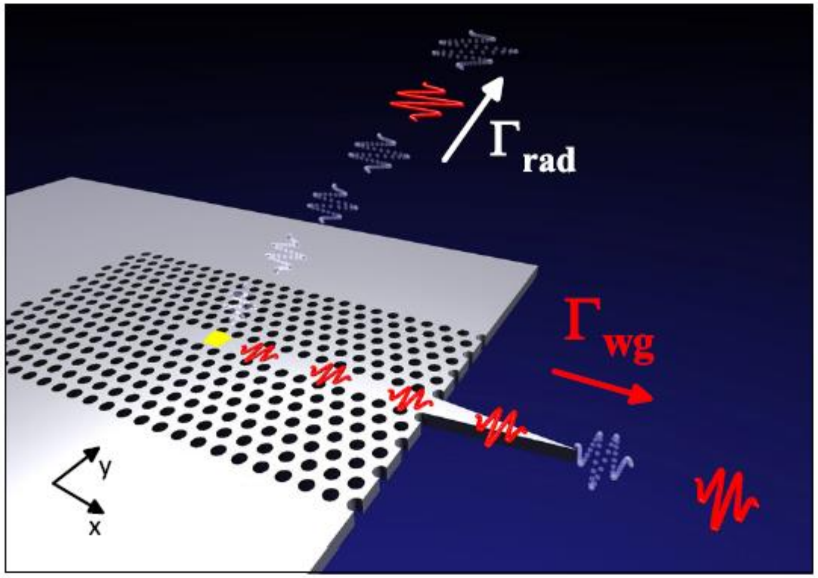


Experimental performance:

- Efficiency: **0.75** ± 0.1
- $g^{(2)}(\tau = 0)$: 0.25

Munsch et al, Phys. Rev. Lett. **110**, 177402 (2013)

Photon collection strategies: photonic crystal waveguides



- ß=98,4%
- Detection efficiency unknown
- $g^2(0)$=0.2

Origin of large spontaneous emission coupling factor ß:

- broadband Purcell effect due do slow-down factor (reduced group velocity)
- strong suppression of loss rate Γ_{rad} due to photonic crystal membrane structure

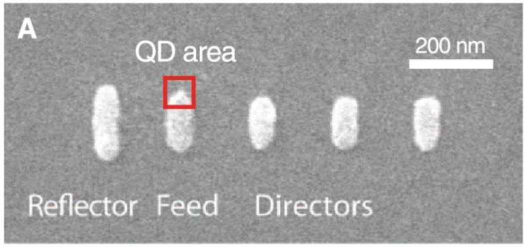
M. Arcari et al., Phys. Rev. Lett. **113**, 093603 (2014).

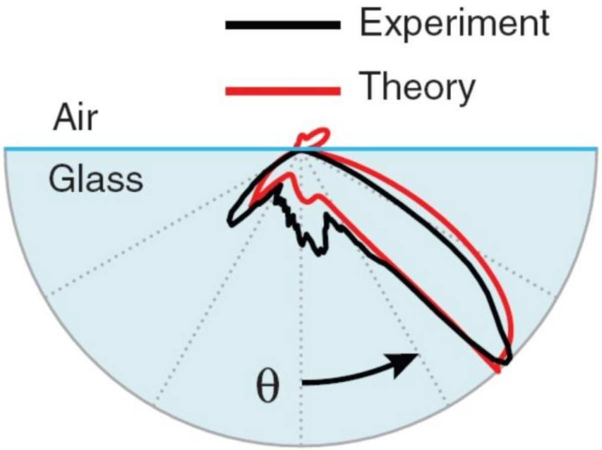
Photon collection strategies: optical antennas

Antennas are well known to direct and receive signals at microwave and radio frequencies.

Yagi-Uda antenna

Optical Yagi-Uda antenna



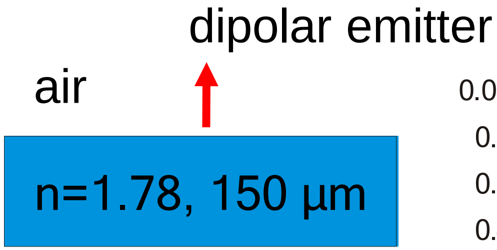


metals are lossy at optical frequencies!

Directional emission!

A. G. Curto, G. Volpe, T. H. Taminiau, M. P. Kreuzer, R. Quidant, N. F. van Hulst, Science. **329**, 930 (2010).

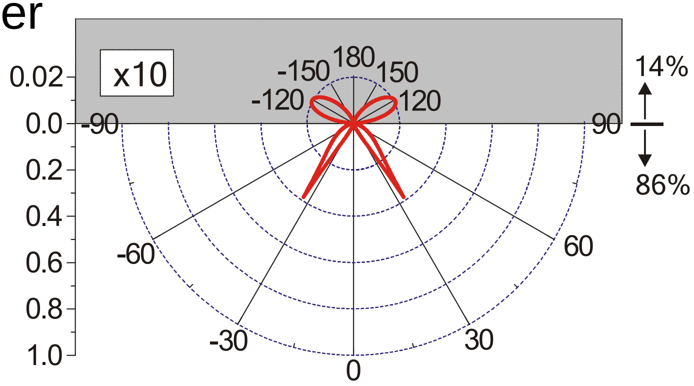
A vertical dipole at an interface

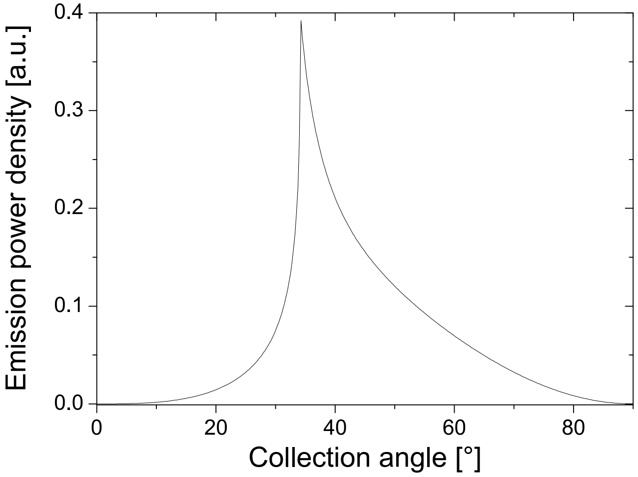


dipolar emitter

air

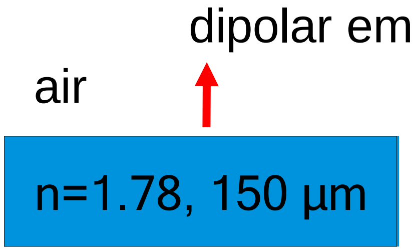
n=1.78, 150 µm

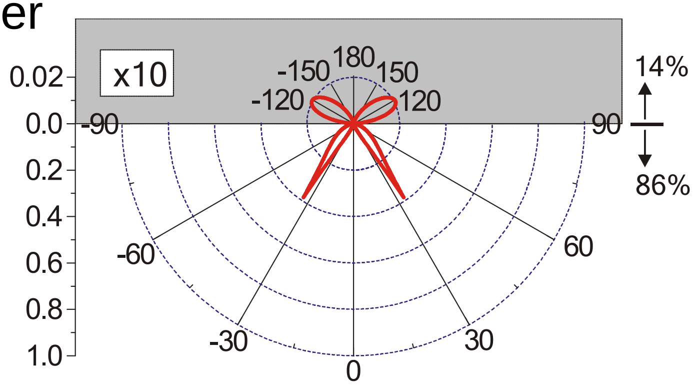


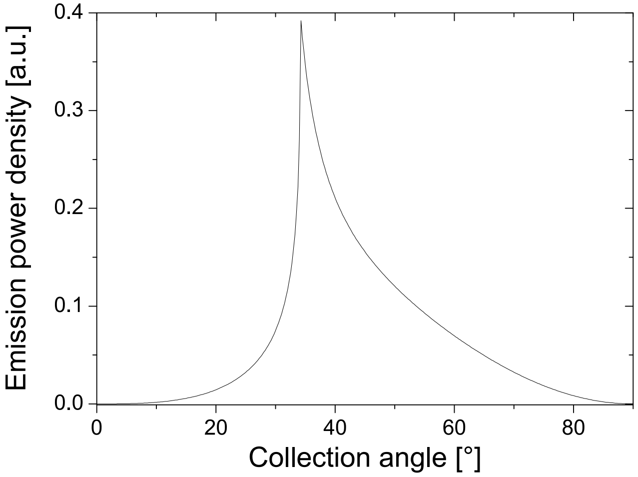


Emission is directed into high index material, if emitter is only a few tens of nm from interface!

A vertical dipole at an interface







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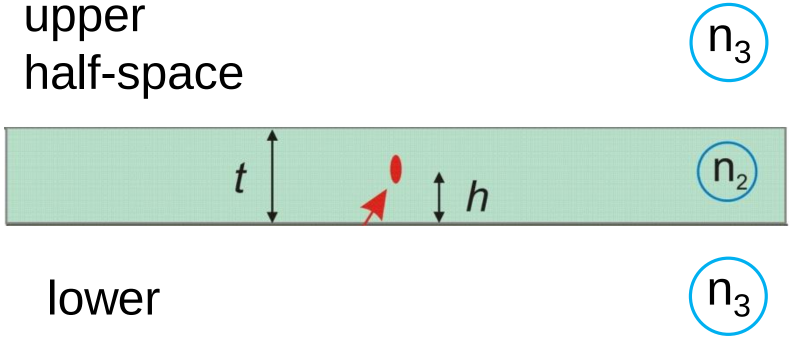
Emission into angles not accessable by any objective

Tayloring the emission pattern with thin films: A dielectric planar antenna



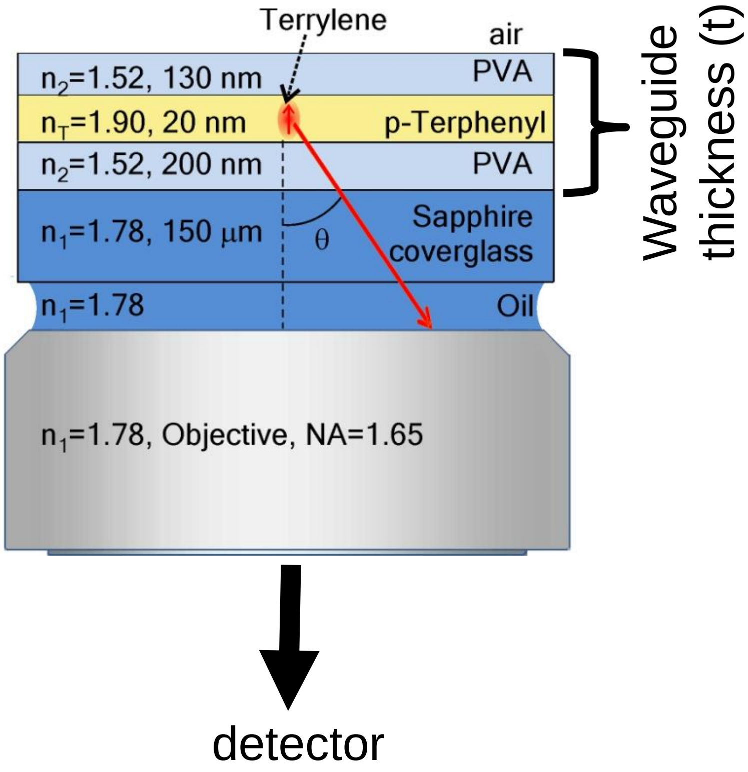
upper
half-space

lower
half-space

Design principles:

- keep emitter an evanescent length from interface ($\theta < \sin^{-1}(n_2/n_1)$)
- limit thickness of layer to form quasi-waveguide ($n_3 < n_2$)

Tayloring the emission pattern with thin films: A dielectric planar antenna



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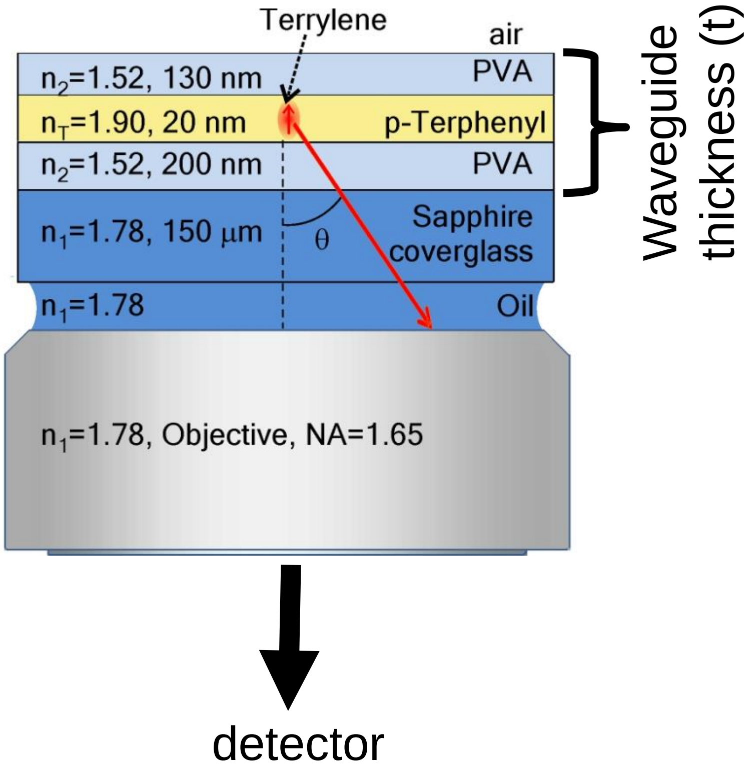
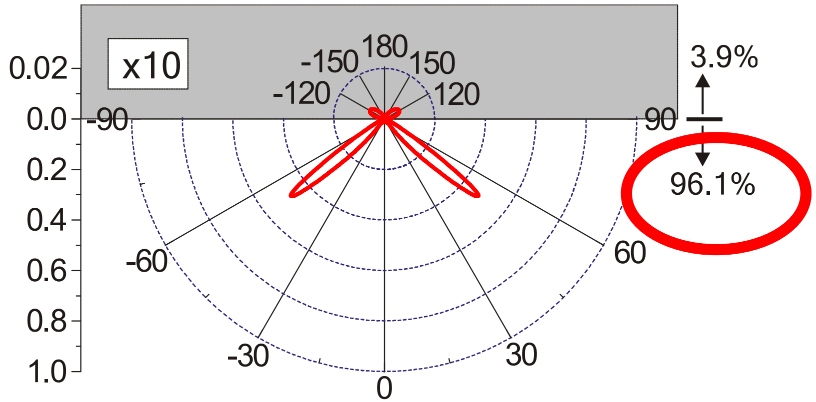
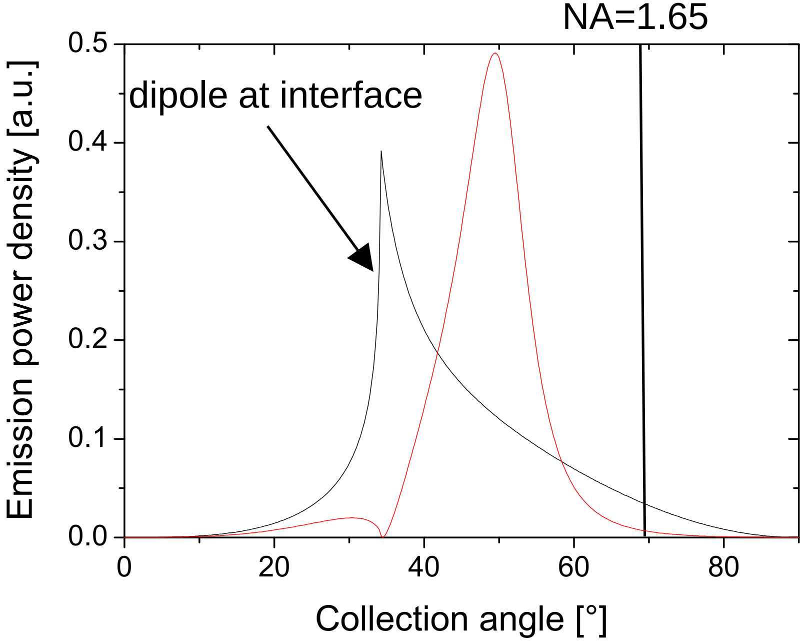
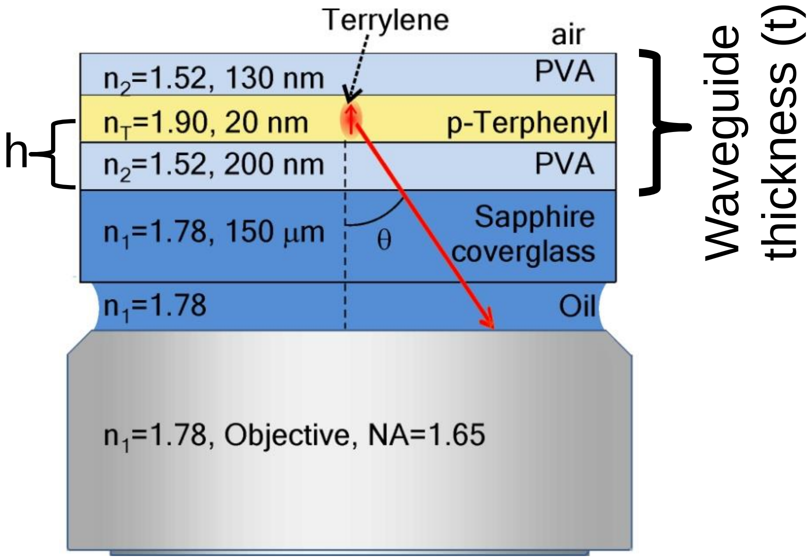


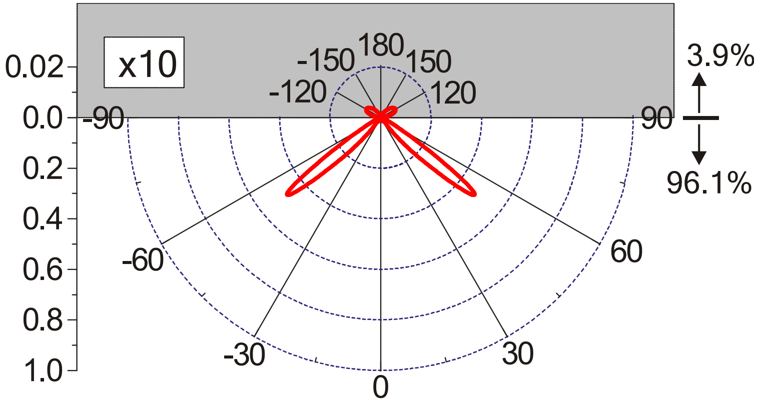




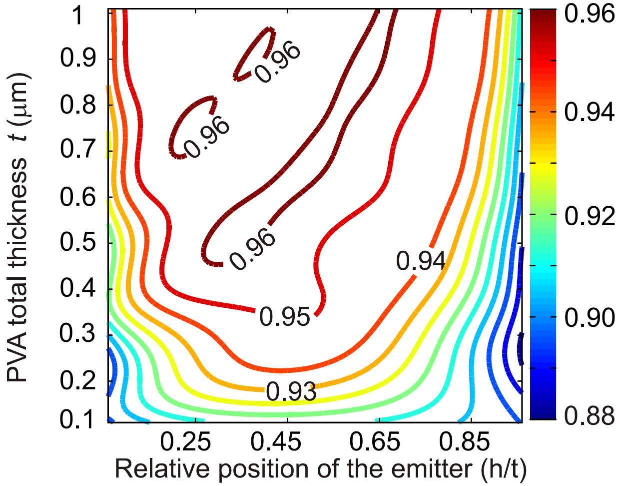
K. G. Lee, X. Chen, H. Eghlidi, P. Kukura, R. Lettow, A. Renn, V. Sandoghdar, S. Götzinger, Nature Photonics **5**,166 (2011).

Tayloring the emission pattern with thin films: A dielectric planar antenna



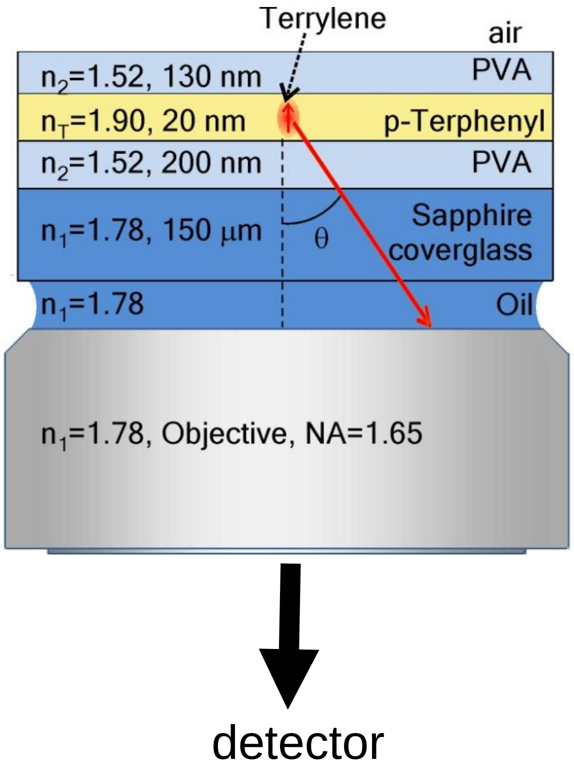


Fabrication tolerance



- structure is broadband (wavelength insensitive)
- tolerant to fabrication imperfections

An ultra bright single-photon source with near-unity efficiency



Performance:

- Collected photons: 94×10^6 per s
- Detected photons: 48.1×10^6 per s (16 pW)
- Antibunching measurements in less than 1s!

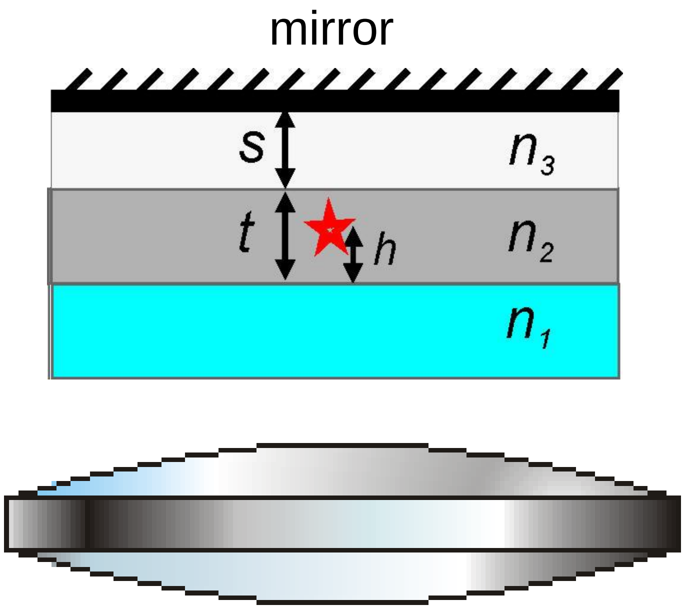
Theory and experiment:

- Calculated collection efficiency η=96%
- Measured collection efficiency η =(96±3)%

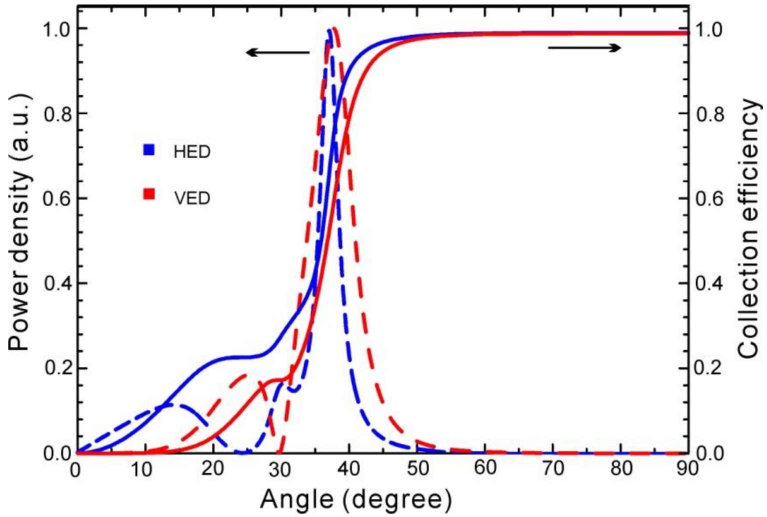
K. G. Lee *et al*., Nature Phot. **5**,166 (2011).

Improving the collection efficiency to 99%

4% losses into upper half space



radiation pattern



HED: horizontal electric dipole
VED: vertical electric dipole

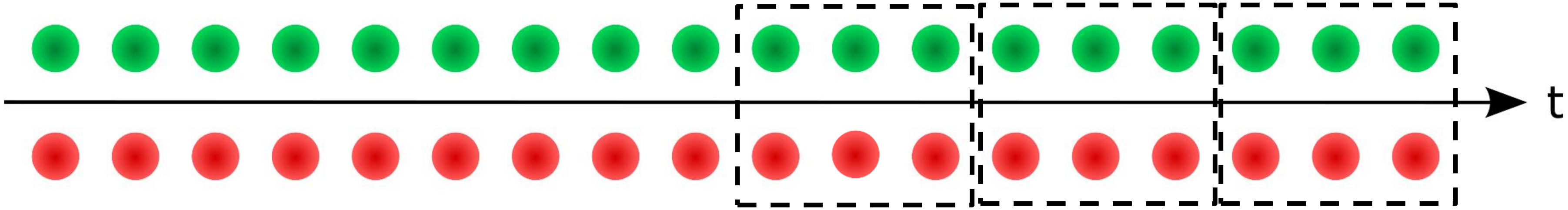
>99% collection efficiency for arbitrary dipole orientation

Scheme works for molecules, diamond, quantum dots!

X. Chen *et al.*, Opt. Lett. **36**, 3545 (2011).

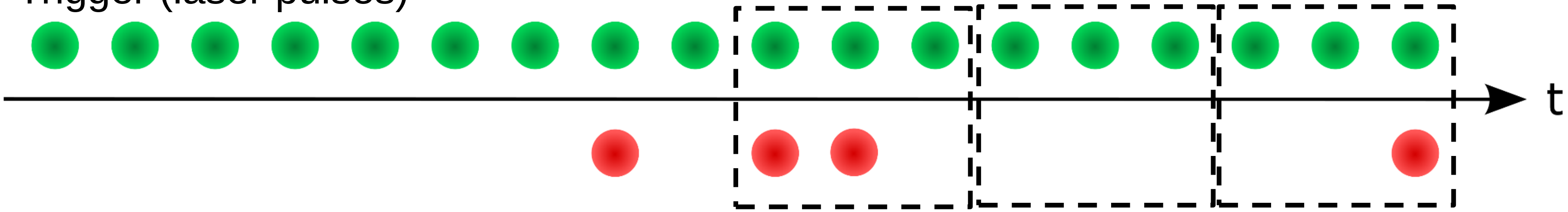
Comparing single-photon sources

Deterministic single-photon source:


Trigger (laser pulses)

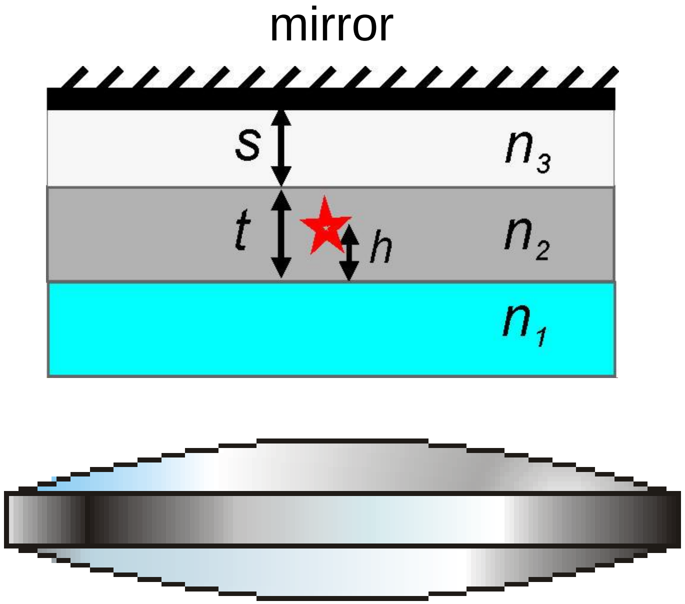
→ **Intensity squeezing!**

“Current“ single-photon sources:


Trigger (laser pulses)

Detection efficiency in the percent range!

“Experimental demonstration“ of 99% collection efficiency



>99% collection efficiency from single quantum dot

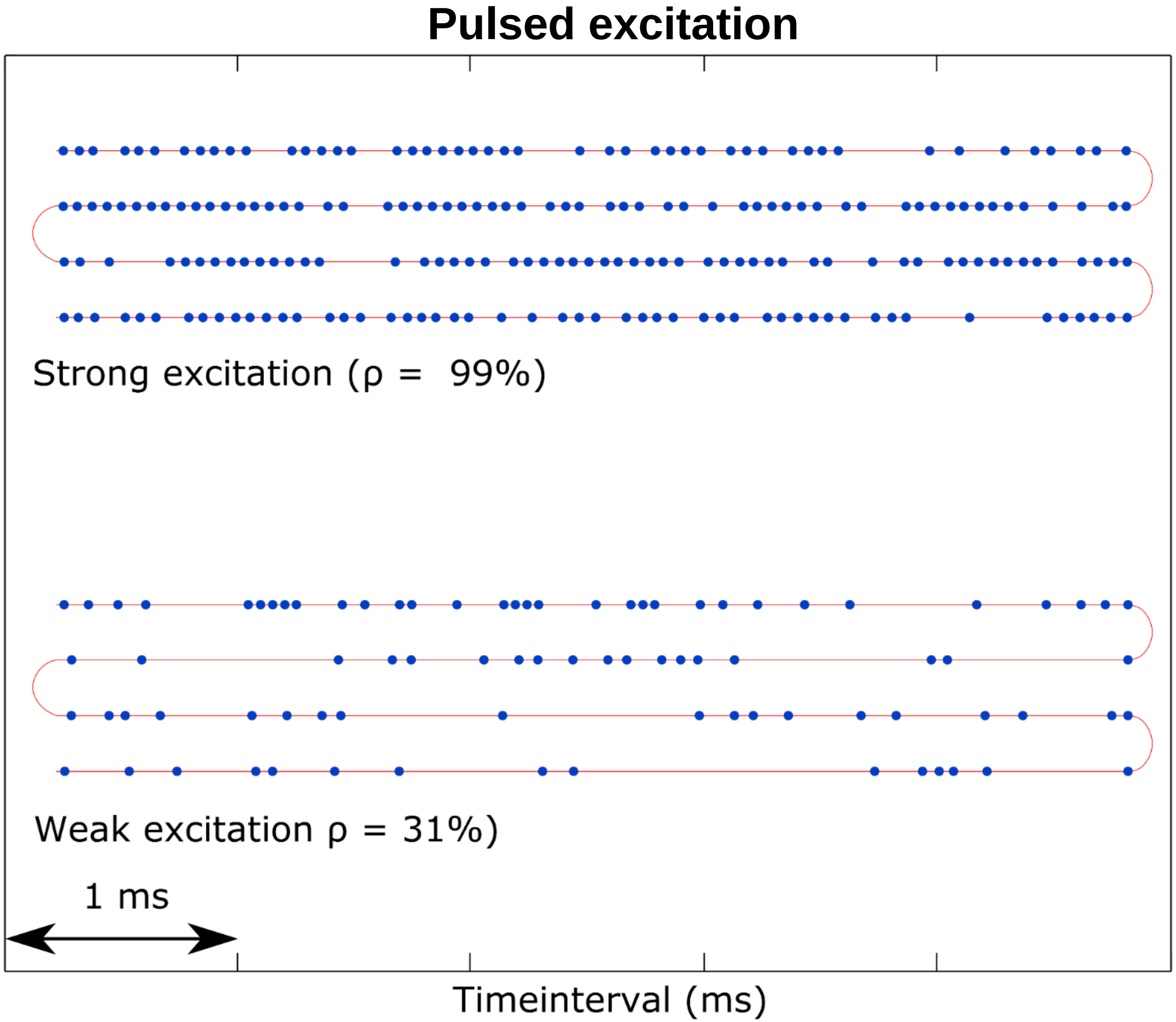
Collection problem solved!

Remaining challenges:

- **Emitter efficiency** → QE of molecules ≈1
- **Detection efficiency** optimize setup → Det. efficiency: ≈70%

X.-L. Chu *et al.* Optica **1**, 203 (2014)

A sub-shot-noise single emitter quantum light source



X.-L. Chu *et al.* Nature Photonics **11**, 58–62, (2017).

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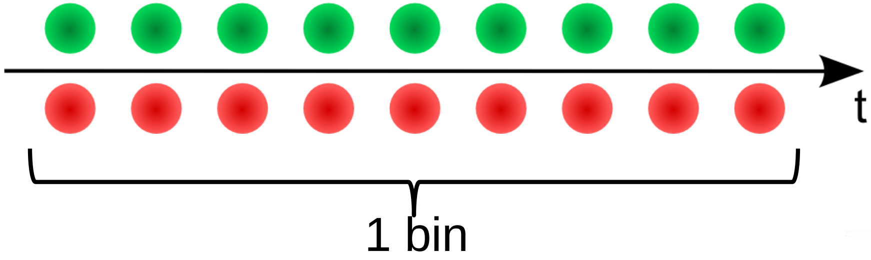


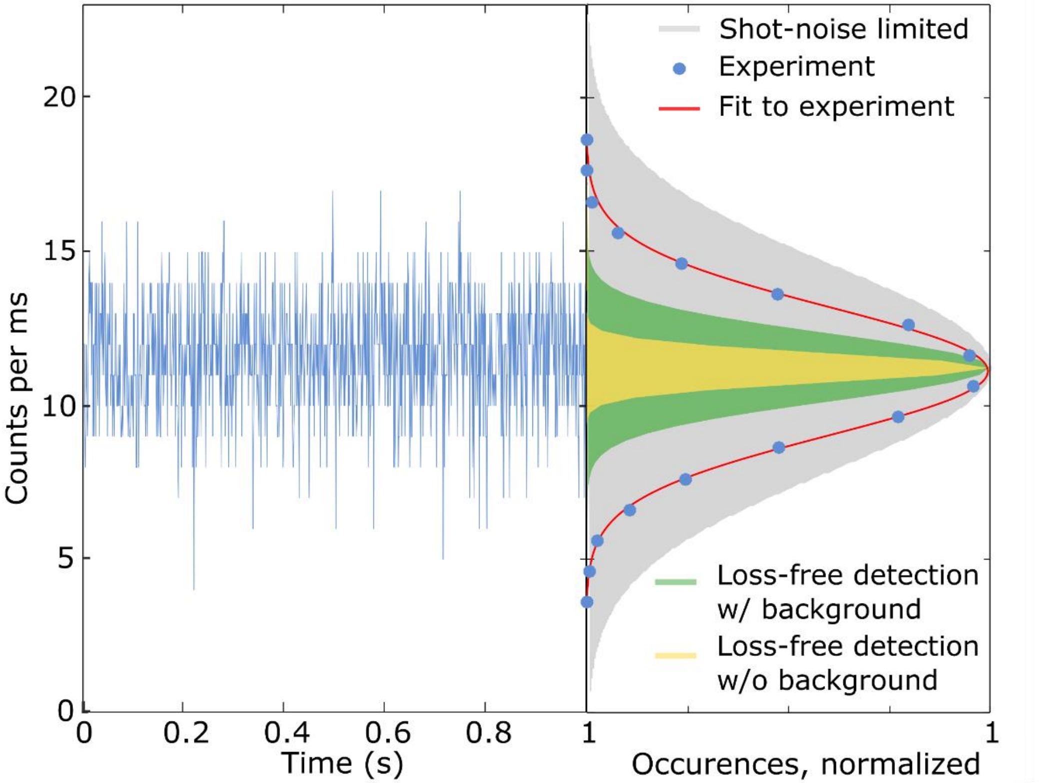
Low loss setup:
total detection efficiency **(68 ± 4)%**

Excitation rate: 15 kHz

Photon count rate: 11.4 kHz

Theory: 43% below shot-noise
Measured: 41% below shot-noise

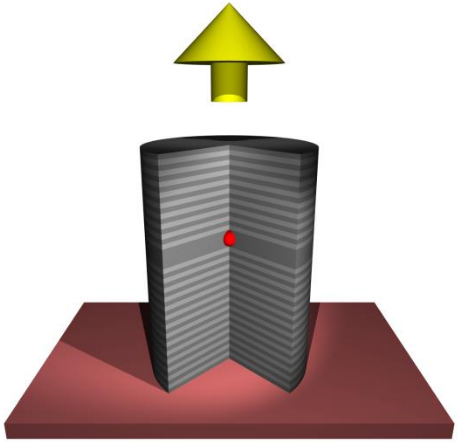
Intensity squeezing!



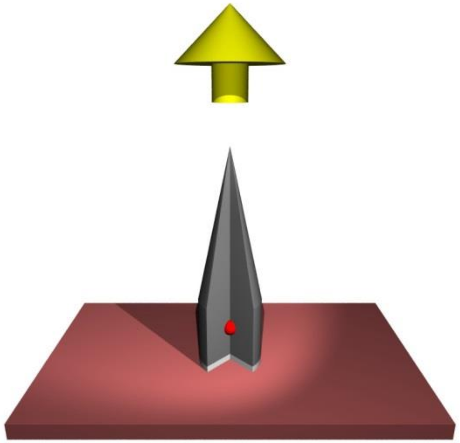
X.-L. Chu *et al.* Nature Photonics **11**, 58–62, (2017).

Summary:
platforms for highly efficient SPSs

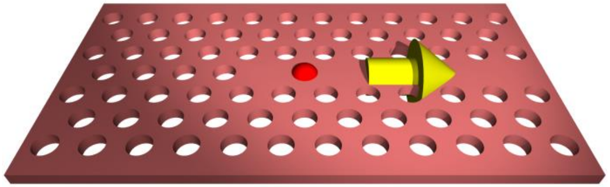
Micropillar (0.79)



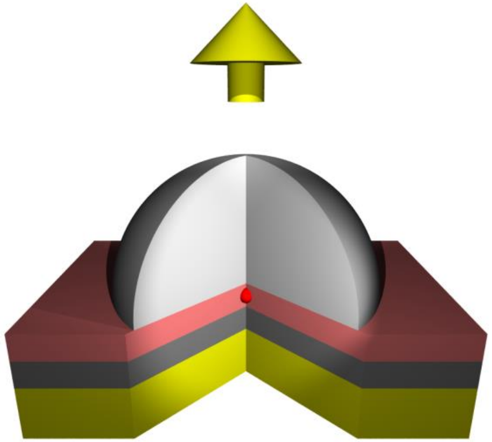
Photonic nanowire (0.75)



Photonic crystal membrane (0.98)



Planar dielectric antenna (0.99)



Courtesy: Nils Gregersen