Quantum optics in nanosystems

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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)
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 Zhuang²,³,⁵*
Localization and Colocalization

**Localization:**

*“Resolution“:* \( \frac{\text{FWHM}_{\text{PSF}}}{\sqrt{N}} \)

\( N \): 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)


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!

- **spectral selection**
- **spatial selection**

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

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

\[ E_0 \rightarrow E_1 \rightarrow E_2 \]

redshifted fluorescence

Typical microscope

Laser

sample with molecules

microscope objective

beam splitter

longpass filter

CCD Kamera
Single molecule microscopy

Typical microscope

- Laser
- Longpass filter
- CCD Kamera
- Sample with molecules
- Microscope objective
- Beam splitter
Single molecule microscopy

Alexa 532

5µm

Terrylene in p-Terphenyl

5µm

Size of the molecule: 1nm
Camera image reveals orientation of the molecule!
Single molecule microscopy

Alexa 532

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

Terrylene in p-Terphenyl

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**
- **Single well-separated photons**
Photon statistics of a laser beam

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

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

$|n\rangle$: 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

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

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

Impossible to generate exactly one photon!

M. Fox: Quantum optics: an introduction, Oxford
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

\[ g^2(\tau) = \frac{\langle :a^\dagger(t)a^\dagger(t+\tau)a(t+\tau)a(t) : \rangle}{\langle a^\dagger(t)a(t) \rangle^2} \]
Photon statistics of a laser beam

Second-order coherence function of a coherent state at $\tau=0$

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

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

with:

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

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

$$g^2(0) = \frac{|\alpha|^2 |\alpha|^2}{|\alpha|^2 |\alpha|^2} = 1$$
Photon statistics:
How to identify a single-photon source

Hanbury-Brown and Twiss Correlator

**Intensity correlation**

\[ g^{(2)}(\tau) = \frac{\langle a^\dagger(t)a^\dagger(t+\tau)a(t+\tau)a(t) \rangle}{\langle a^\dagger(t)a(t) \rangle^2} \]

A single emitter is needed

**Photon statistics:**

- **Laser:** \( g^{(2)}(\tau) = 1 \)
- **Thermal source:** \( g^{(2)}(0) = 2 \)
- **Single emitter:** \( g^{(2)}(0) = 0 \)

**Diagram:**

- Light source
- Beam splitter
- Detector 1
- Detector 2

**Graph:**

- Intensity vs. time delay \( \tau \)
- Data points for different time delays
Blinking of a single emitter

- 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
Deterministic single-photon source:

\( t \) (laser pulse)
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:

\[ E_{\text{photon}} = h \nu \]

\[ P = n h \nu / t \]

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)

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

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 \times 10^5 attempts neccessary
Efficiency of a single-photon source

Emitter efficiency (quantum efficiency, dark states)

Collection efficiency

Losses in the set-up

Detection efficiency

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

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

• need to be well characterized
• develop strategies to suppressing them
The problem: efficient collection of single photons

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

Free space

Emitter in solid state matrix

Collection lens

Collection efficiency by definition <50%

Emission into $4\pi$

interface

$n>1$

Total internal reflection reduces collection angle further
e.g. GaAs: 3 %

Note: The problem of efficient collection is the inverse of an efficient interaction!
Photon collection strategies: microcavities (cavity quantum electrodynamics)

**Micropillars:**

Enhancement of spontaneous emission rate:

- **Purcell-factor:** \( F \sim \frac{Q}{V} \)
  
  \( Q \): quality factor
  
  \( V \): mode volume

- Fraction of spontaneous emission coupled to cavity mode:
  
  \[ \beta = \frac{F}{F+1} \]


- Purcell-factor 61
- Funnel photons into cavity mode
- Coupling to mode 97%
- Outcoupling efficiency: few %
Photon collection strategies: microcavities (cavity quantum electrodynamics)

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


Performance:
• 31 MHz collection rate
• collection efficiency 38%
• 4 MHz detection rate

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

Photon collection strategies: nanowires

NV center in Diamond:


- 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)

QDs in GaAs:

Claudon et al., Nature Phot. 4, 174 (2010)

Collection efficiency: 72%
$g^{(2)}(\tau = 0): 0.008$
Photon collection strategies: photonic trumpet

Inverted "trumpet" taper

\[ D > 1.5 \, \mu m \]

\[ d \approx 220 \, \text{nm} \]

Compatible with electrical contacting!

Photon collection strategies: photonic trumpet

Experimental performance:

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

Photon collection strategies: photonic crystal waveguides

- \( \beta = 98.4\% \)
- Detection efficiency unknown
- \( g^2(0) = 0.2 \)

Origin of large spontaneous emission coupling factor \( \beta \):

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

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 vertical dipole at an interface

Emission is directed into high index material, if emitter is only a few tens of nm from interface!
A vertical dipole at an interface

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

Emission into angles not accessible by any objective.
Tayloring the emission pattern with thin films: A dielectric planar antenna

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

Design principles:
• keep emitter an evanescent length from interface \( \theta < \sin^{-1}(n_2/n_1) \)
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Tayloring the emission pattern with thin films: A dielectric planar antenna

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

- structure is broadband (wavelength insensitive)
- tolerant to fabrication imperfections
An ultra bright single-photon source with near-unity efficiency

**Performance:**
- Collected photons: $94 \times 10^6$ per s
- Detected photons: $48.1 \times 10^6$ per s (16 pW)
- Antibunching measurements in less than 1s!

**Theory and experiment:**
- Calculated collection efficiency $\eta = 96\%$
- Measured collection efficiency $\eta = (96\pm3)\%$

Improving the collection efficiency to 99%

4% losses into upper half space

Scheme works for molecules, diamond, quantum dots!

>99% collection efficiency for arbitrary dipole orientation

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 $\approx 1$

- Detection efficiency
  - Optimize setup
  - Det. efficiency: $\approx 70$

A sub-shot-noise single emitter quantum light source

Pulsed excitation

Strong excitation ($\rho = 99\%$)

Weak excitation $\rho = 31\%$)

1 ms

Timeinterval (ms)

A sub-shot-noise single emitter quantum light source

Low loss setup: total detection efficiency $(68 \pm 4)\%$

Excitation rate: 15 kHz
Photon count rate: 11.4 kHz

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

Intensity squeezing!

Summary: platforms for highly efficient SPSs

- Photonic nanowire (0.75)
- Micropillar (0.79)
- Photonic crystal membrane (0.98)
- Planar dielectric antenna (0.99)

Courtesy: Nils Gregersen