Experimental observation of single-emitter fluorescence and photon anti-bunching

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Aim of our research

• Single-Photon Sources (SPS)
  - efficient and stable
  - non-classical properties (anti-bunching)
  - on demand
  - at room temperature

• Applications
  - Quantum computing
  - Quantum cryptography
  - Medical application
• Classical first-order coherence function

\[
\gamma^{(1)}(x_1, x_2) = \frac{\langle E^*(x_1)E(x_2) \rangle}{\sqrt{\langle |E(x_1)|^2 \rangle \langle |E(x_2)|^2 \rangle}}
\]

• Quantum first-order coherence function

\[
g^{(1)}(x_1, x_2) = \frac{G^{(1)}(x_1, x_2)}{\sqrt{G^{(1)}(x_1, x_1)G^{(1)}(x_2, x_2)}}
\]

where \( \tilde{\rho} = \sum_i P_i |i\rangle\langle i| \)

\[
G^{(1)}(x_i, x_j) = \text{Tr}\left\{ \rho \ E^{-}(x_i)\tilde{E}^{(+)}(x_j) \right\}
\]
• Second-order coherence function

\[
g^{(2)}(\tau) = \frac{\left\langle \hat{E}^-(t)\hat{E}^-(t+\tau)\hat{E}^+(t+\tau)\hat{E}^+(t) \right\rangle}{\left\langle \hat{E}^-(t)\hat{E}^+(t) \right\rangle \left\langle \hat{E}^-(t+\tau)\hat{E}^+(t+\tau) \right\rangle} = \frac{\left\langle \hat{a}^+\hat{a}^+\hat{a}\hat{a} \right\rangle}{\left\langle \hat{a}^+\hat{a} \right\rangle^2} = \frac{\langle \hat{n}(\hat{n} - 1) \rangle}{\langle \hat{n} \rangle^2} = 1 + \frac{\left\langle (\Delta\hat{n})^2 \right\rangle - \langle \hat{n} \rangle}{\langle \hat{n} \rangle^2}
\]

where \( \hat{E}(x) = \hat{E}^+(x) - \hat{E}^-(x) \propto [\hat{a}(t) - \hat{a}^+(t)] \)
Theory – Hanbury Brown and Twiss experiment
Theory – Photon bunching

- Second-order coherence function

\[ g^{(2)}(\tau) = 1 + \frac{\langle (\Delta n)^2 \rangle - \langle \hat{n} \rangle}{\langle \hat{n} \rangle^2} \]

- Classical light
  - thermal light
    \[ \langle (\Delta \hat{n})^2 \rangle = \langle \hat{n} \rangle + \langle \hat{n} \rangle^2 \]
  - coherent light,
    \[ \langle (\Delta \hat{n})^2 \rangle = \langle \hat{n} \rangle \]

\[ g^{(2)}(\tau) \leq g^{(2)}(0) \]
• Second-order coherence function

\[ g^{(2)}(\tau) = 1 + \frac{\langle (\Delta \hat{n})^2 \rangle - \langle \hat{n} \rangle}{\langle \hat{n} \rangle^2} \]

• Non-classical light
  - sub-Poissonian statistics

\[ \langle (\Delta \hat{n})^2 \rangle < \langle \hat{n} \rangle \]

\[ g^{(2)}(0) \leq g^{(2)}(\tau) \]

Photon anti-bunching
Confocal microscopy

Better resolution and collection emitted light from a single emitter

Hanbury Brown and Twiss experimental setup

TimeHarp 200

Variable delay

APD

Photon antibunching
Oil-immersion objective  Cooled EMCCD camera

microscope cover slips
objective lens
dichroic mirror
filter

single emitters in host

Exciting an emitter and collecting the emitted light from it

Observation of the fluorescence light from emitters
Avalanche Photodiodes on adjustable micrometer stage providing x, y, and z degrees of freedom.

Sample mounted on glass microscope slide and held on piezo-translation stage with magnets.

Oil immersion objective.

Non-polarizing 50/50 beamsplitter.

Neutral density filters.

Mirrors.

Camera used for beam alignment.

Removable dichroic mirror which reflects but does not transmit 532 nm.

Removable orange glass filter.

532 nm wavelength diode laser.

APD data goes to photon counting card in computer.

Computer with data acquisition cards and software.
Samples—DiDye molecule and PMMA

Molecular structure

DilC18(3) dye.

Polymethyl methacrylate (PMMA)

Dye Fluorescence Spectrum

Fluorescence intensity, arbitrary units

Wavelength, nm
Samples–CdSe Quantum dots and Cholesteric LC

QD Fluorescence Spectrum

Fluorescence Intensity (arbitrary units)

Wavelength, nm

Selective transmission of the CLC hosts

Fluorescence spectrum of CdSe QD (a. u.)

Wavelength (nm)
Samples Preparation

• Emitters on slip or in PMMA host
  - diluting dye molecules or quantum dots in solvent
  - concentration of the solutions (1nM)
  - 50 μL is applied to slides on a spin coater (or mixed with PMMA solution)
  - rotation speed: 3000 rpm for 30 seconds

• Emitters in CLS hosts
  - diluting dye molecules or quantum dots in solvent
  - concentration of the solutions (1nM)
  - select proper concentration of chiral additive CB15 in nematic E7
  - One drop of solution is mixed with equal amount of CLC
  - Planar alignment using shifting two substrates relative each other in one direction
• Photon-Counting CCD observation

• Fluorescence image by confocal microscopy

• Measurement of fluorescence lifetime

• Observation of photon anti-bunching
Single emitter fluorescence

(Accumulation time: 500ms; Gain: 255)

Background

DiDye molecule single emitters
Single emitter blinking

(Accumulation time: 500ms; Gain: 255)
Dildye in PMMA

(Accumulation time: 500ms; Gain: 255)

Background

3 DilDye + 1 PMMA
CdSe quantum dots

(Accumulation time: 500ms; Gain: 255)

1:10 solution
CdSe QD blinking & bleaching

1:10 solution, 500 ms, 255 Gain
Confocal micrograph of fluorescence

- DilDye molecules

40×40 μm²
Fluorescence from a single DilDye molecule

10 × 10 μm²

2 × 2 μm²

Bleaching !!
DiDye in PMMA

40 × 40 μm²

4 × 4 μm²
CdSe quantum dots

40×40 μm²

4×4 μm²

Blinking !!
CdSe QDs in 15CB liquid crystal host

40×40 μm²

4×4 μm²
CdSe QDs in CLC4 host

40×40 μm²

2×2 μm²
Fluorescence lifetime of CdSe QDs

- Single exponential decay

\[ N = N_{BG} + N_1 \exp(-t / \tau) \]

Fluorescence Life Time; Single QD #1, 11/08/2006

Measured fluorescence lifetime of CdSe QDs without hosts (unit: ns)

<table>
<thead>
<tr>
<th></th>
<th>Data 1</th>
<th>Data 2</th>
<th>Average</th>
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<tbody>
<tr>
<td>Single QD #1</td>
<td>0.8960</td>
<td>0.9383</td>
<td>0.9172</td>
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<tr>
<td>Single QD #2</td>
<td>1.8526</td>
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<tr>
<td>Multiple QDs #1</td>
<td>1.1080</td>
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<tr>
<td>Multiple QDs #1</td>
<td>1.8300</td>
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</tr>
</tbody>
</table>
F.L. of CdSe QDs in LC hosts

Pre-prepared samples

Fluorescence lifetime (ns)
F.L. of CdSe QDs in LC hosts

Fluorescence lifetime (ns) for freshly-made samples in CB15 and CLC4.
Photon anti-bunching
CdSe QDs in CB5 host
Photon anti-bunching
CdSe QDs in CB15 host

CdSeQD 15CB Scan #1 111706

Coincidence counts vs. interphoton time (ns)
Photon anti-bunching
CdSe QDs in CLC4 host

CdSeQD CLC4 Scan #13, 112206

Coincidence counts vs. interphoton time (ns)

Central dip
1. Single-Photon Emitters are studied using Confocal Microscopy and Hanbury Brown – Twiss Setup
2. Blinking and Bleaching of single emitters are observed
3. Fluorescence lifetime are measured for CdSe QDs in different hosts
   • Hosts do not change F.L. appreciably
4. Photon anti-bunching is observed for the 1st time for QDs in CLC host