Colloidal quantum dots as single photon emitters

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1. Introduction

Semiconductor nanocrystals

→ High tunability
→ Different materials available (CdSe, PbSe, CdS, ZnS,...)
→ Influence of size (quantum confinement)
→ Possibility to synthesize core/shell structures

Can we use them as single photon sources?
1. Introduction

Single photon emission for quantum optics

→ Single photon emission demonstrated in CdSe / ZnS QDs

*Brokmann et al., APL, 85, 5, 2004*

→ Blinking behaviour, i.e. succession of ON and OFF states

*Van Sart et al., Chem Phys Chem, 3, 10, 871-879, 2002*

**Objective:** Deterministic photon source

No photon on demand behaviour
Introduction

Single QD as single photon emitter

Results and discussion

Conclusion
2. Single QD as single photon emitter

Impact of Auger recombination process

→ What is responsible for good single photon emission properties in QDs?

![Energy level diagrams showing single exciton emission, bi-exciton recombination, and Auger non-radiative recombination.]

- **1S<sub>e</sub>** and **1S<sub>h</sub>**
  - Single exciton emission
  - Bi-exciton recombination: emission

- **Strong Auger effect:** Efficient single photon emitter
  - Bi-exciton recombination: Auger, non-radiative
2. Single QD as single photon emitter

Link between Auger process and Blinking behaviour

→ Efficient Auger recombination rate: Generation of hot charge carriers
→ Trapped charge: Charged QD

\[ G_{X+e} = G_e + G_{rad} \]

In CdSe/ZnS: \( G_e \gg G_{rad} \)
No emission in charge QD
Blinking
2. Single QD as single photon emitter

Necessary control of Auger recombination rate

→ Pulsed excitation
→ Number of exciton is Poissonian

→ Low fluence:
  → No bi-exciton
  → No photon on demand
→ High fluence:
  → Almost photon on demand
  → Creation of multi-excitons

High excitation power needed
Auger process gives single photon emission and blinking behaviour
2. Single QD as single photon emitter

Non-blinking CdSe / CdS

*Cirillo et al., Chem. Mater., 26, 2, 2014*

→ High quantum yield
→ Non blinking

Usable for single photon emission?
Introduction

Single QD as single photon emitter

Results and discussion

Conclusion
3. Results and discussion

Experimental setup and emission correlation measurement

→ Measurement on single QD with fluorescence microscope and HBT detection

→ Good single photon emission in giant CdSe / CdS QDs at low excitation power
→ Better understanding of their properties is required
3. Results and discussion

Evolution of blinking trace with excitation power

→ One giant CdSe / CdS QD under different excitation power

→ Non blinking even at high power

→ Distribution changes at high power

QD in different states with different quantum yield?
3. Results and discussion

→ QD can be in different states: Confirmed by the different lifetimes
3. Results and discussion

**Lifetime in different charged state?**

- QD can take different charged state

\[ 1S_e \quad 1S_h \]

\[ G_X = 6.7 \mu s^{-1} \quad G_{X+e} = 22.2 \mu s^{-1} \quad G_{X+2e} = 55.5 \mu s^{-1} \]

- Recombination of biexciton
- Two pathways
  - Electron channel: \( G_e \)
  - Hole channel: \( G_h \)

\( G_e, G_h, G_{Xrad} \) known

Calculate correlation function in various charged states

\[ G_{Auger} = 2G_e + 2G_h \]
3. Results and discussion

Calculation of correlation function

→ QD in a succession of charged state
→ Each state shows a different correlation function
→ Evolution reproduced with calculation

![Graphs showing experimental and calculated correlation functions for QD in different charged states.](image)
3. Results and discussion

Influence of different Auger rates

→ Quantum yield of biexciton less affected by additional charge in QD
→ Responsible for degradation of single photon emission performances
→ Very low $G_e$ and very high $G_h$ recommended

\[ G_{\text{Auger}} = 2G_e + 2G_h \]
Introduction

Single QD as single photon emitter

Results and discussion

Conclusion
4. Conclusion

Colloidal QD as single photon emitter

→ Achieve deterministic photon source with single colloidal QD
→ Must be non-blinking
→ Single photon emission must be preserved at high excitation fluence

Non blinking giant CdSe\CdS QD

→ High quantum yield, non-blinking CdSe\CdS synthesized with flash method

Control of Auger recombination

→ Reduce $G_e$ : increase size of shell, alloying at interface
→ Increase $G_h$ : decrease size of the core, abrupt interface
→ Control of surface properties and crystallinity
Thank you for your attention!