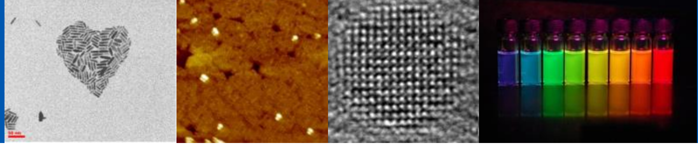

Tunable band structure in core-shell quantum dots through alloying of the core

A. Guille | D. Mourad | T. Aubert | A. Houtepen | R. Van Deun | E. Brainis | Z. Hens

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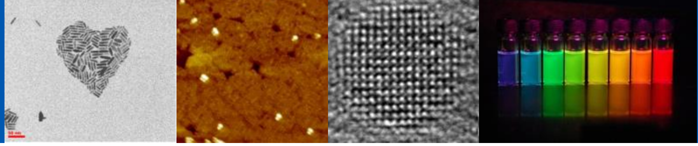
nb-photonics Center for Nano- and Biophotonics Ghent University
Belgian Science Policy Office belspo

1. Introduction



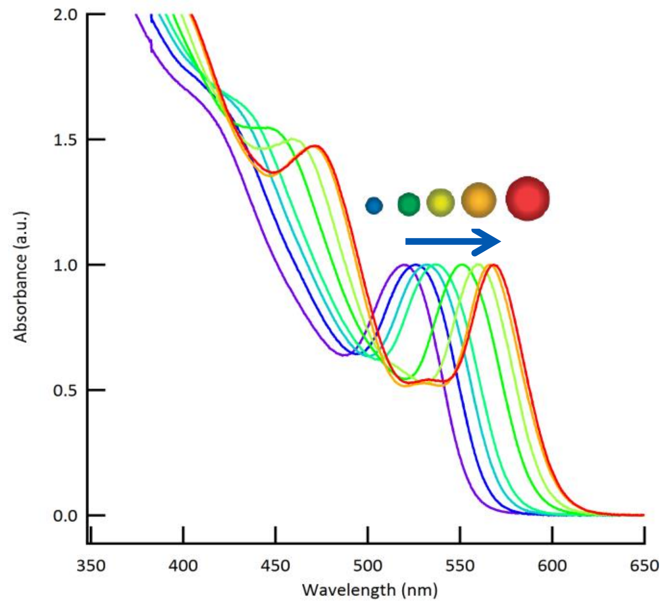
Semiconductor nanocrystals

→ High tunability

→ Different materials available (CdSe, PbSe, CdS, ZnS,…)

→ Influence of size (quantum confinement)

→ Possibility to synthesize core/shell structures



Additional degree of freedom : Composition

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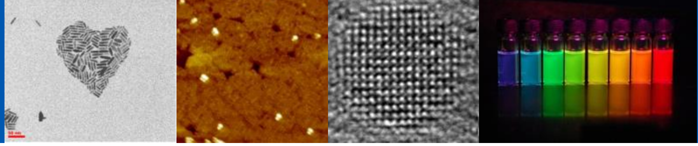
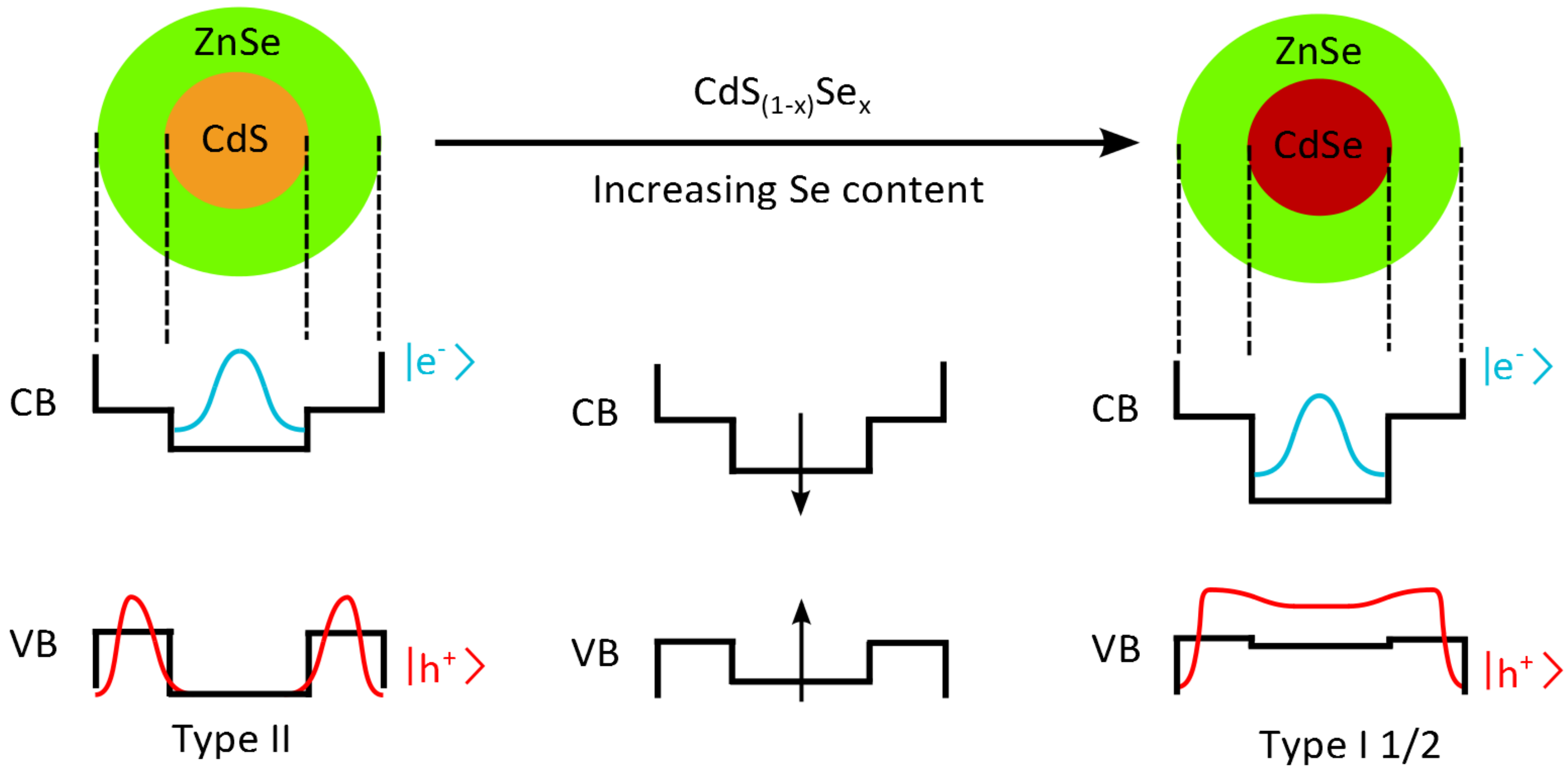


Tunability of $CdS_{(1-x)}Se_x$\ZnSe QDs

→ Homogeneously alloyed $CdS_{(1-x)}S_x$ core

→ Tuning of overlap of charge carriers wavefunctions

→ Single exciton gain demonstrated in Type II CdS / ZnSe



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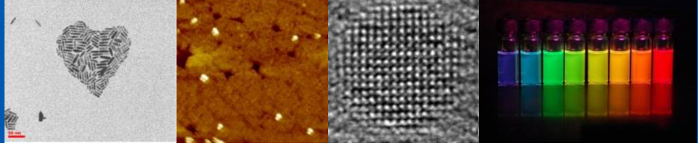
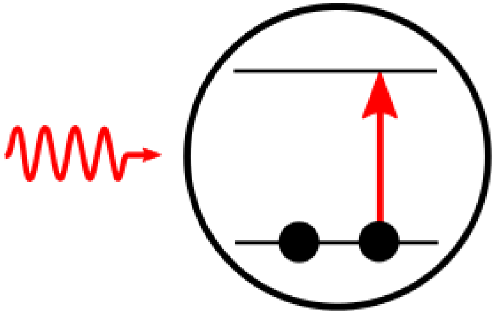
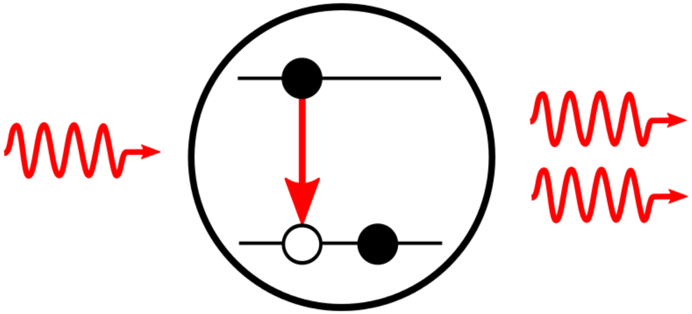


Quantum dots as gain medium : Advantages of type II QDs



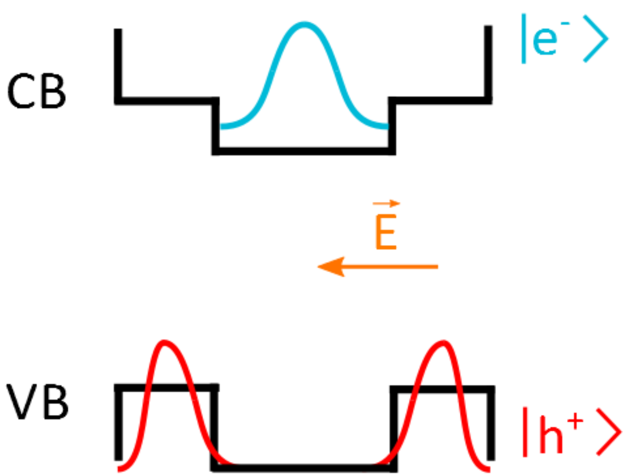
Absorption



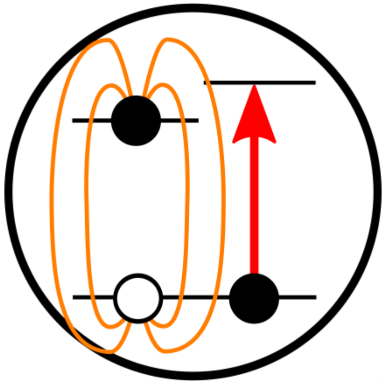
Stimulated emission

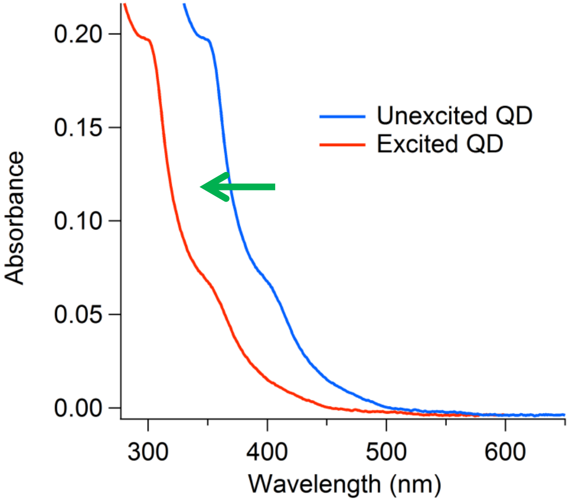
→ Gain requires more than one exciton per dot : **High excitation fluence**

*Klimov et al., Nature 2007, **447**, 441-446*



Type II





→ Reduced reabsorption : **Reduced excitation fluence**

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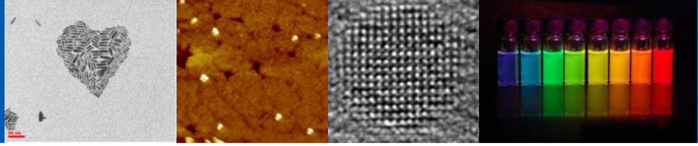
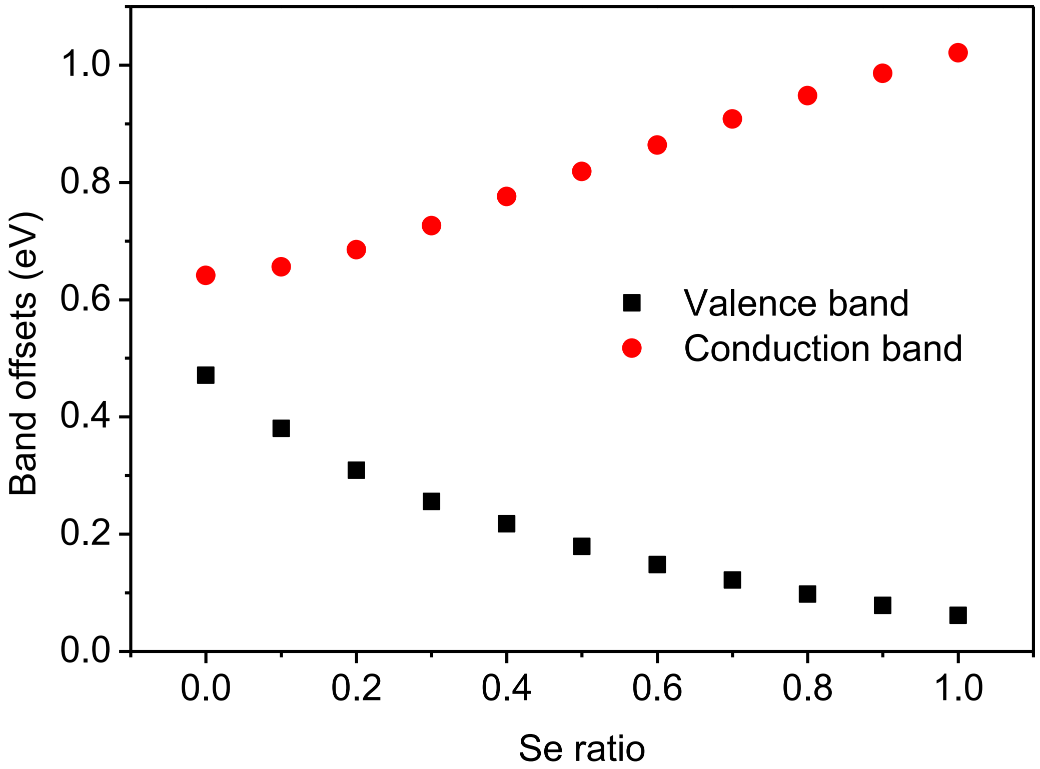


Overlap of charge carriers

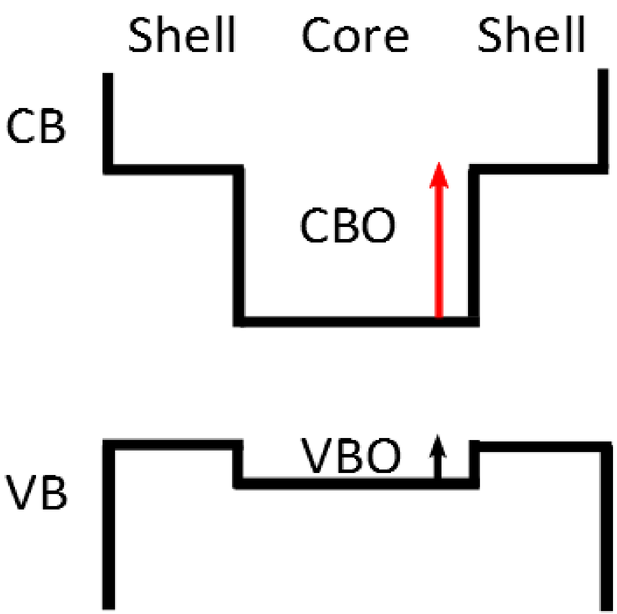
→ Valence band-offset $CdS_{(1-x)}S_x$ \ ZnSe

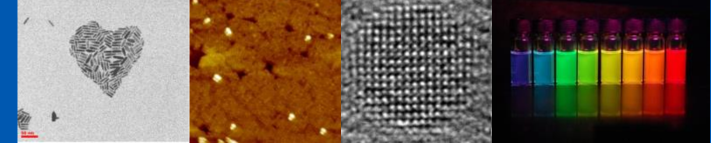
→ Calculated numerically with tight binding approach (D. Mourad)

→ From type II to type I 1/2



Se ratio: 0

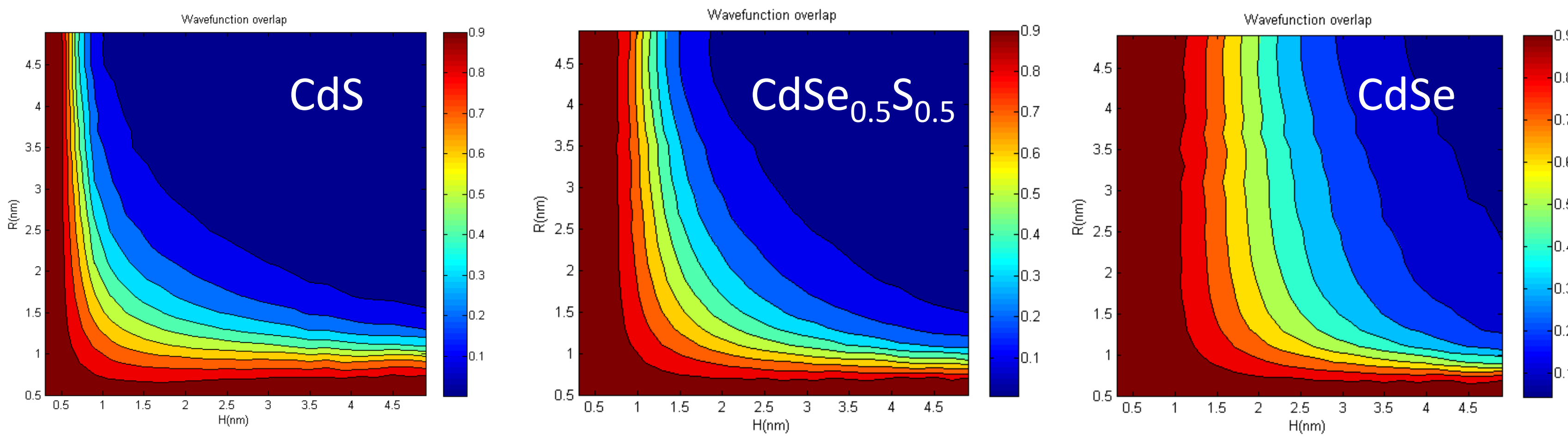




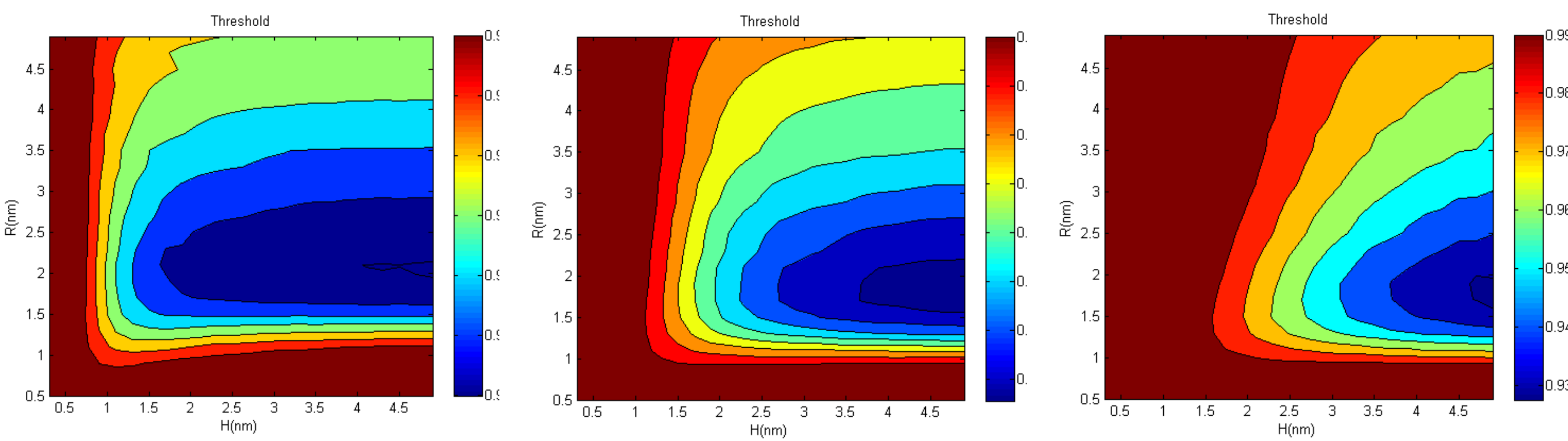
Overlap of charge carriers and gain threshold

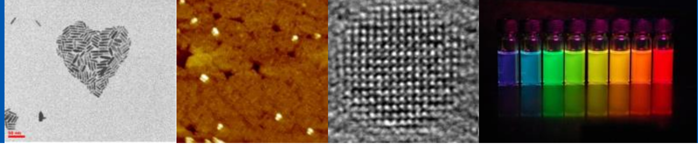
→ Calculation in effective mass approximation with calculated band offsets

Electron-hole overlap



Gain threshold





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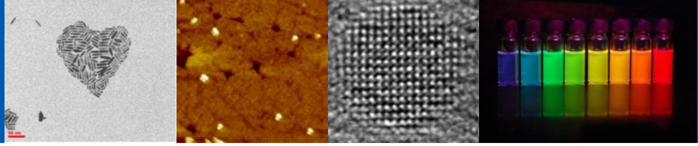


Hot injection synthesis of $CdS_{(1-x)}Se_x$ QDs

*T. Aubert et al., Chem. Mater. 2013, **25**, 2388–2390*

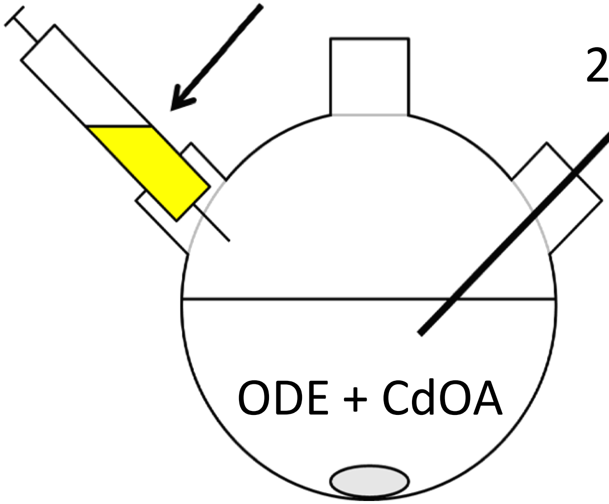
→ Precursors:

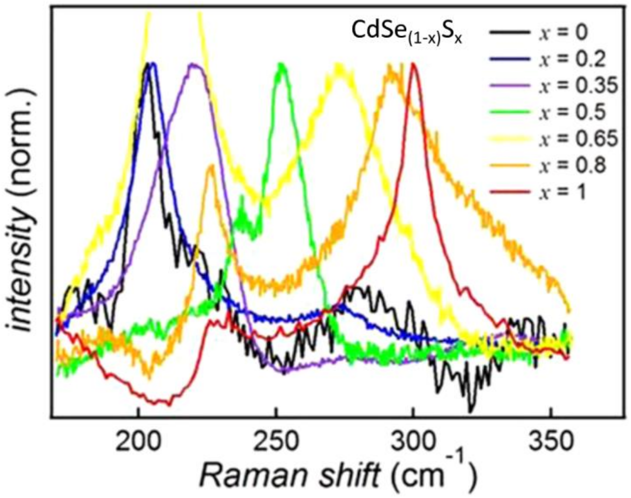
- Se powder dispersed in ODE
- S dissolved in ODE
- Cd oleate

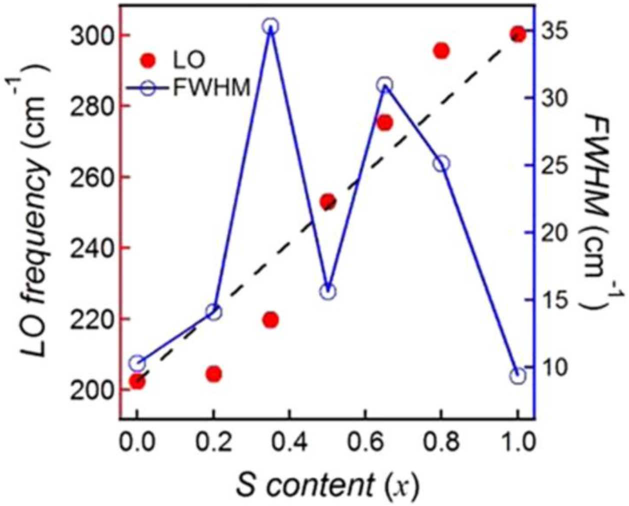
→ Balanced reactivity of S and Se precursors

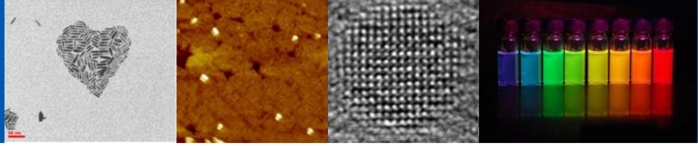
→ Composition measured by EDX

→ Alloying checked with Raman spectroscopy









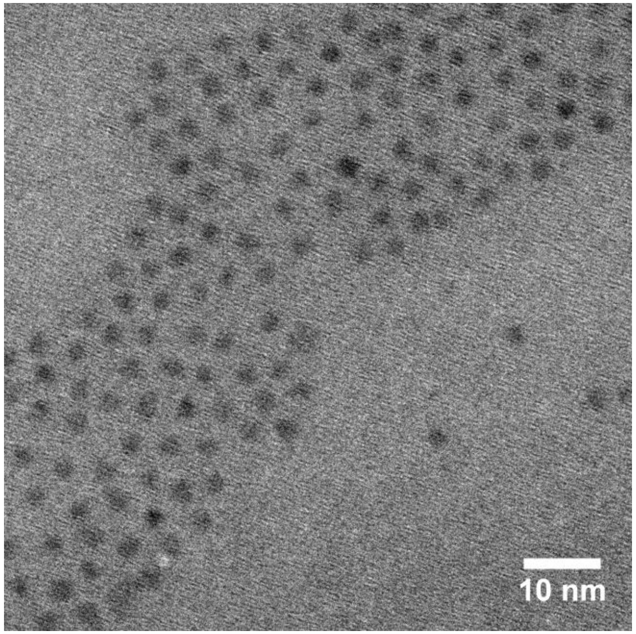
Growth of ZnSe shell

→ Cores in ODE and octadecylamine

→ Continuous injection of Zn oleate and TOP-Se

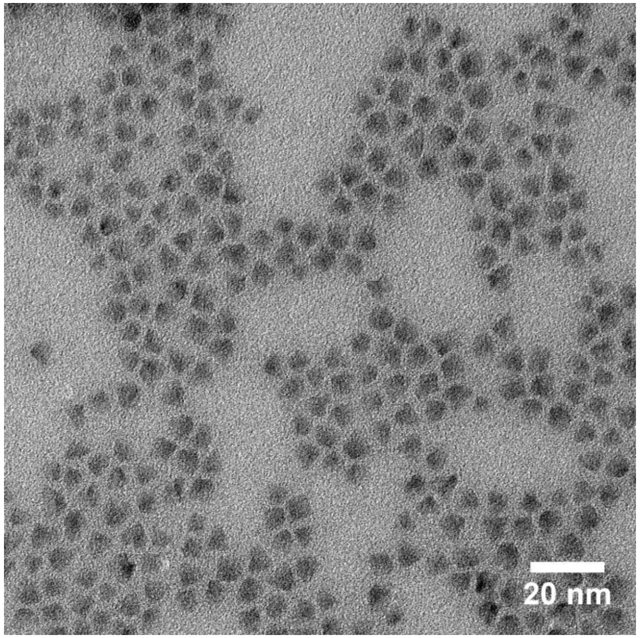
→ Stored in hexane

Core

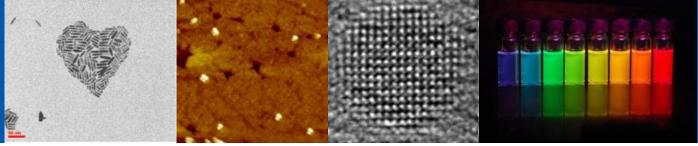


$CdS_{0.6}Se_{0.4}$

Core-shell

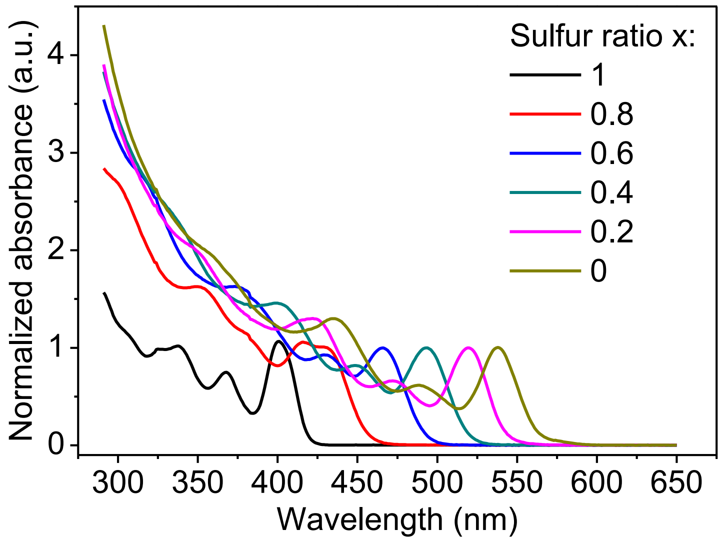


$CdS_{0.6}Se_{0.4}$ / ZnSe

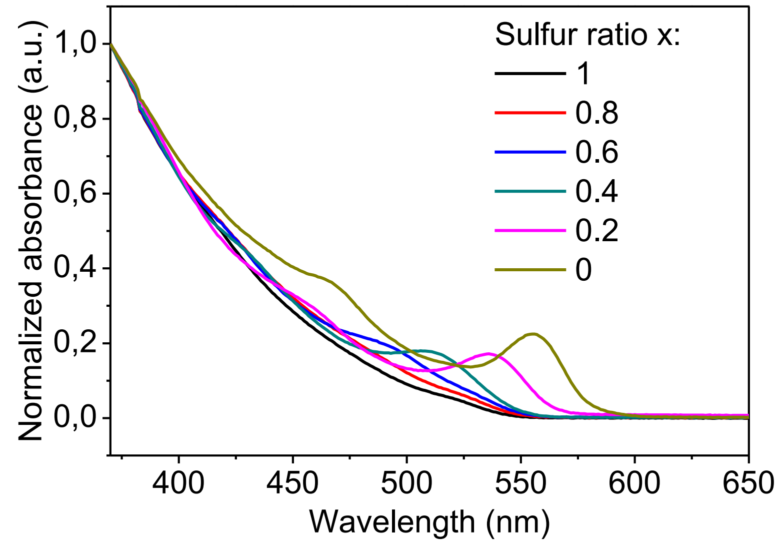


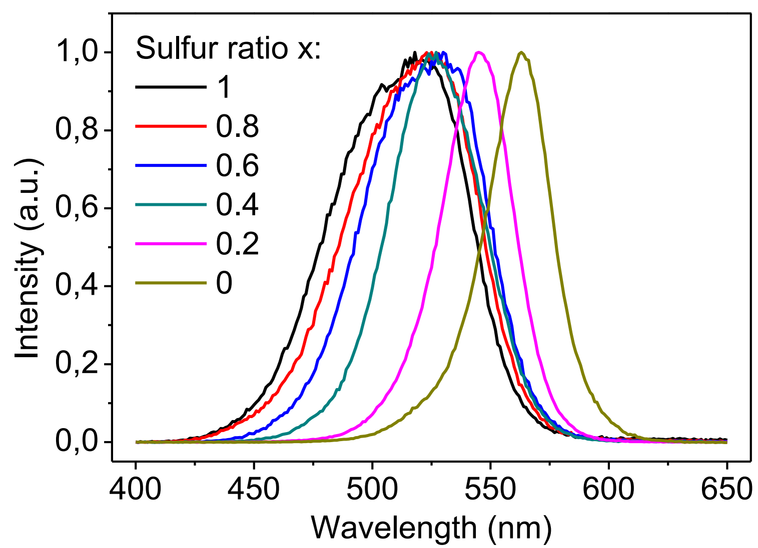
Absorption and emission spectra

Core



Core-shell





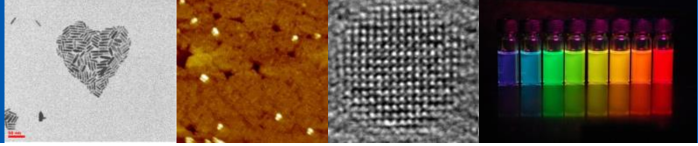
→ Core radius: 1.5 nm

→ Shell thickness: 1.5 nm

→ Trap emission in S rich core QDs

Excitonic peak appears in Se rich cores

Transition from type II to type I 1/2

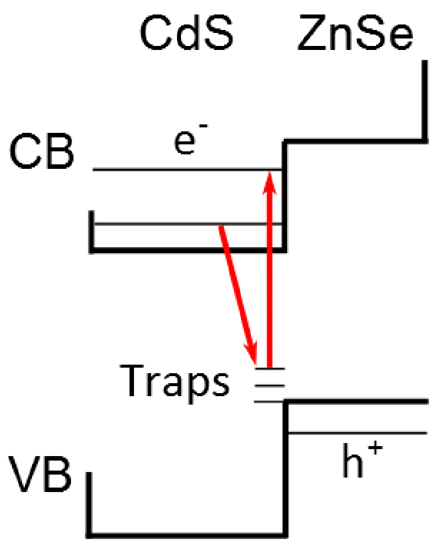


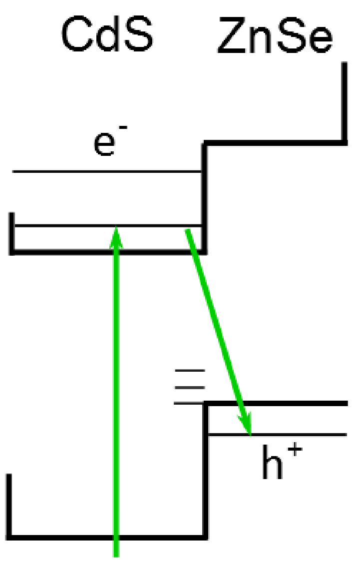
Origin of trap emission in CdS/ZnSe

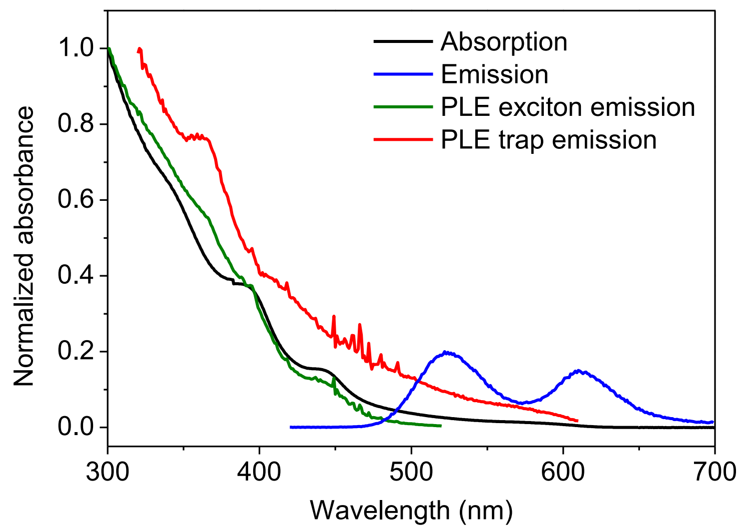
→ Larger cores : trap emission visible

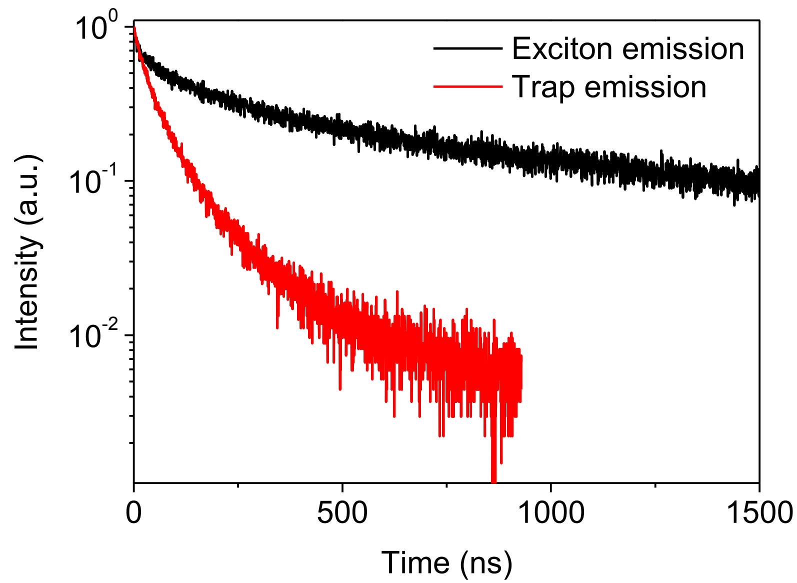
→ Different PLE spectra and decays

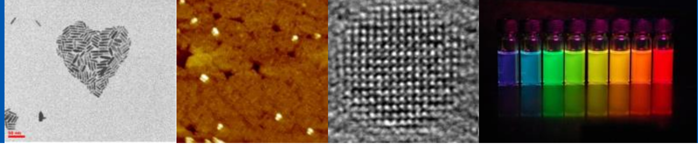
Increased absorption : Effect on gain ?











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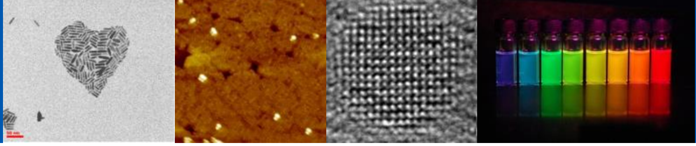
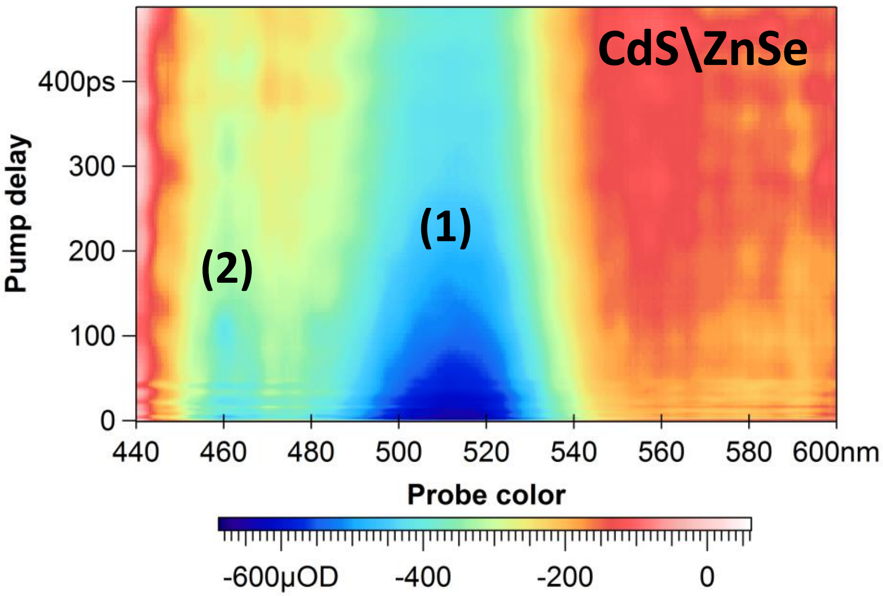
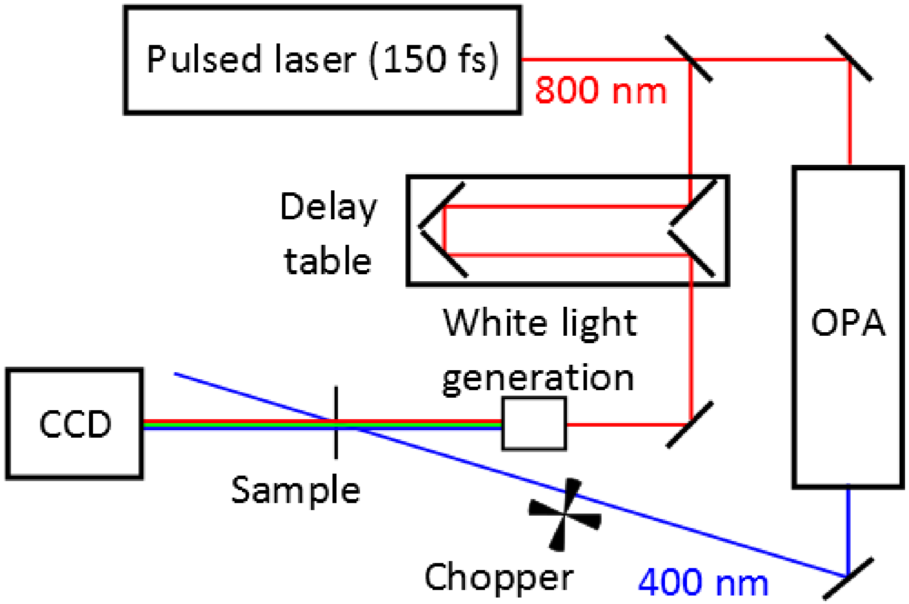
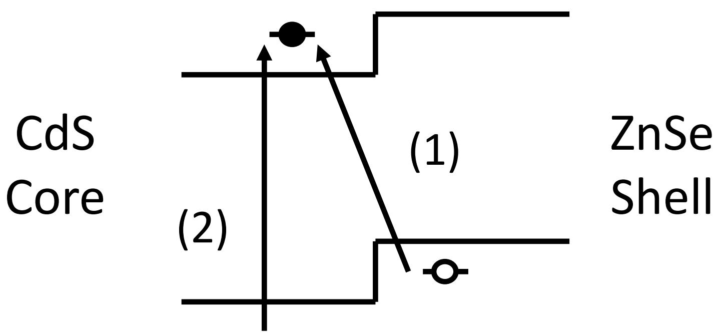


Transient absorption spectroscopy

→ Absorption spectrum at different pump-probe delays

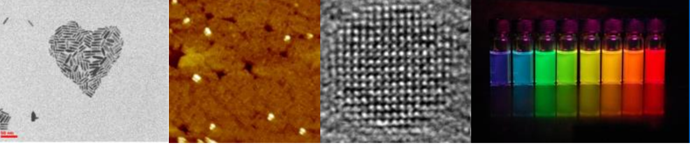




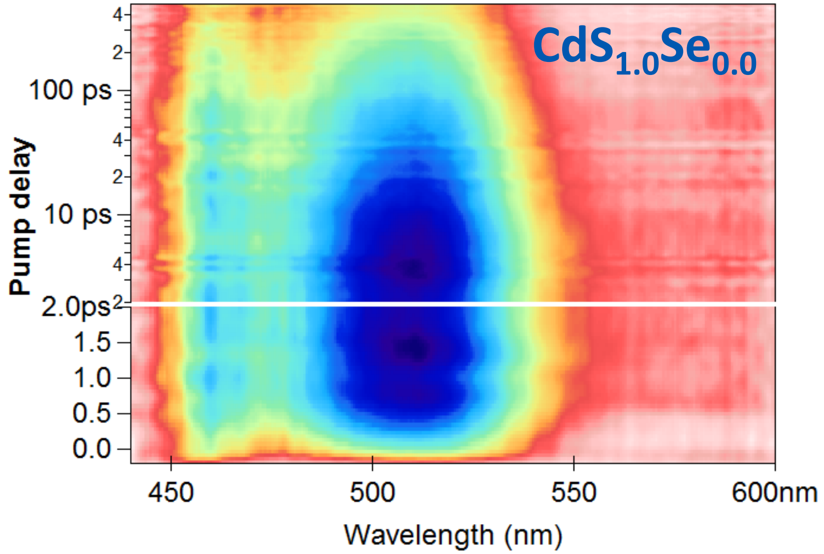


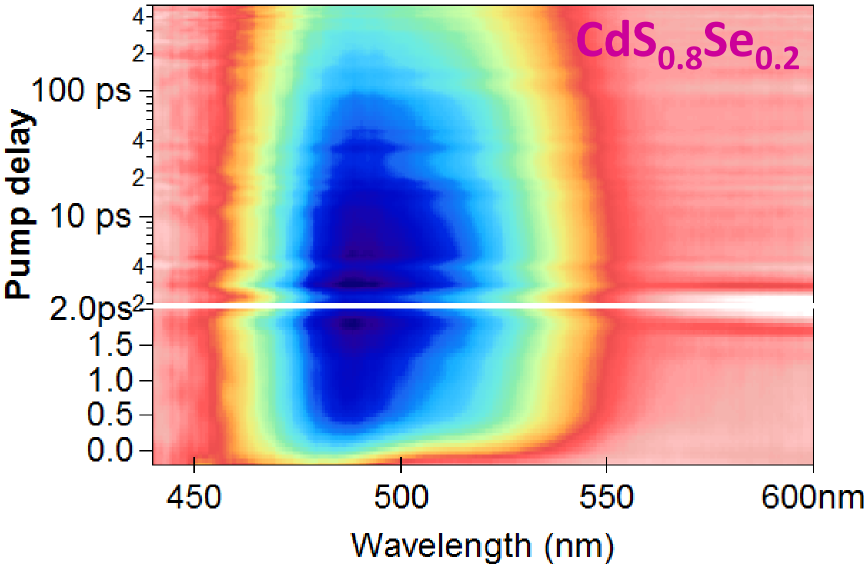
Evidence for the type II structure

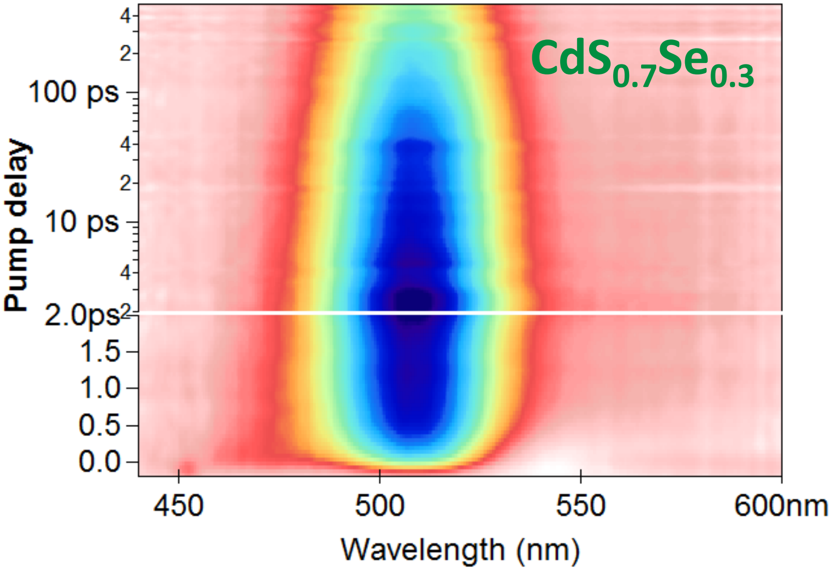
3. Results and discussion

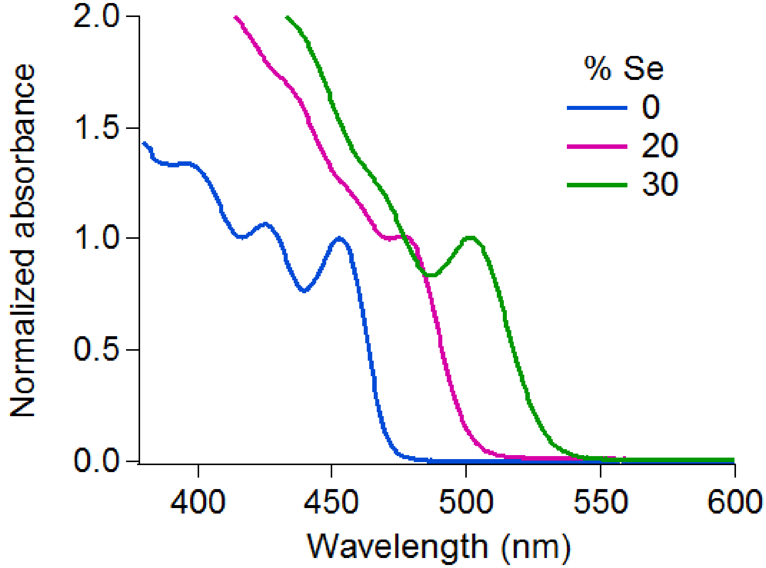


Transition Type II to type I 1/2









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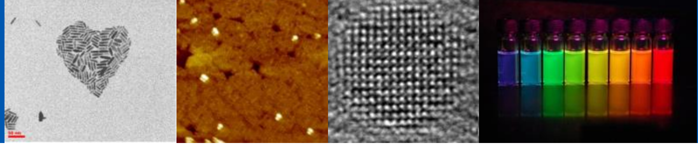
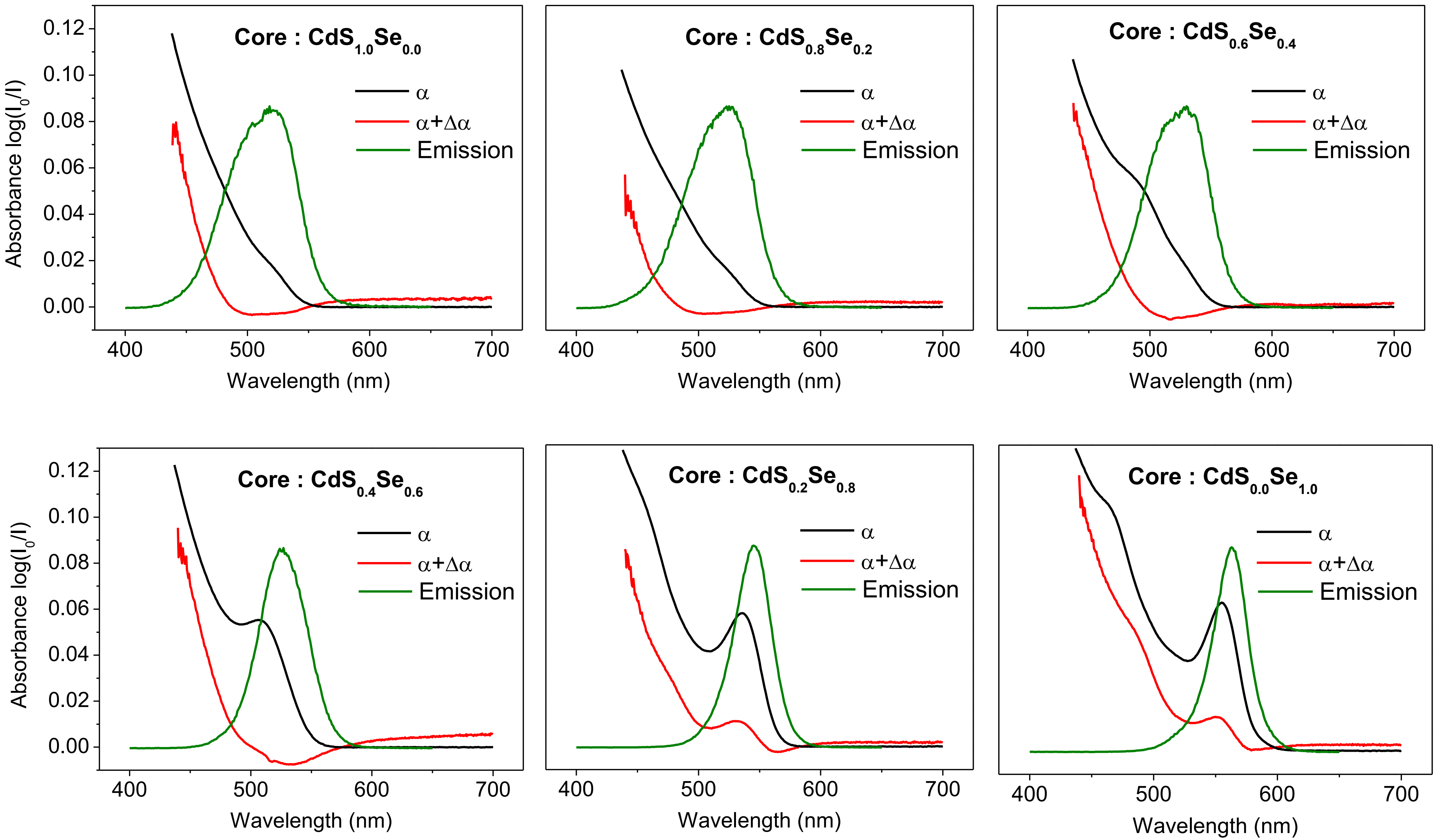
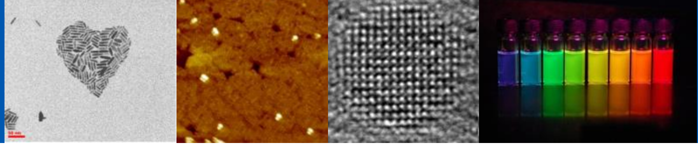


Absorption spectrum of excited QDs : Optical gain





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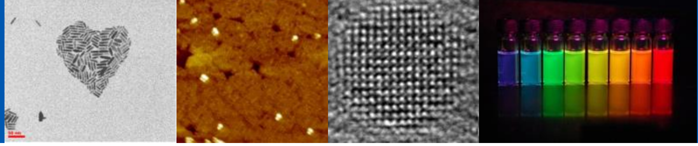
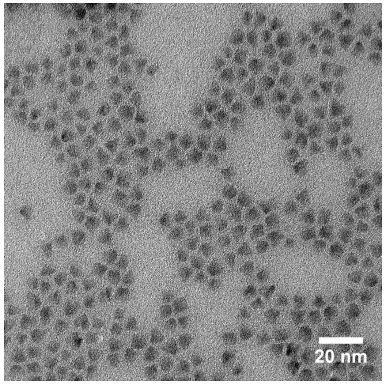


Synthesis of $CdS_{(1-x)}Se_x$ / ZnSe QDs



→ Absorption and emission from traps at the interface

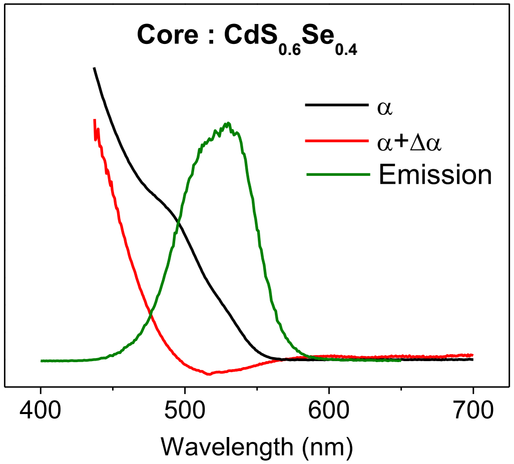
→ Excess of absorbance at the emission wavelength : synthesis to improve

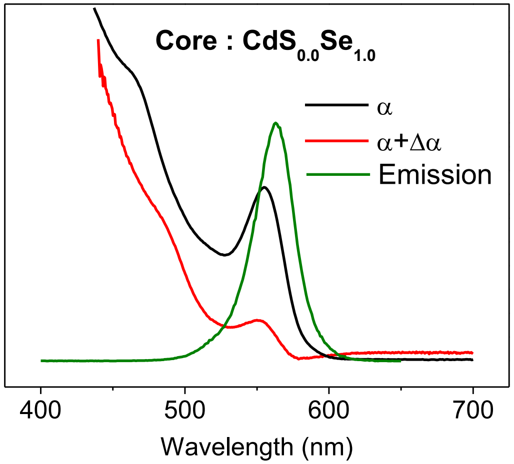
Tunability of QDs band structure

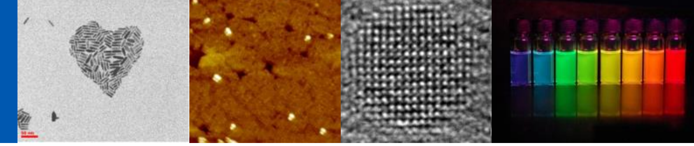
→ Continuous transition from type II to type I ½ demonstrated

→ Very large tunability of emission wavelength and overlap

→ Gain observed with transient absorption spectroscopy







Thank you for your attention !

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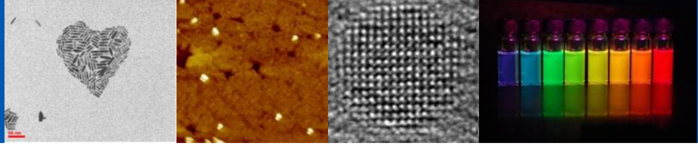

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