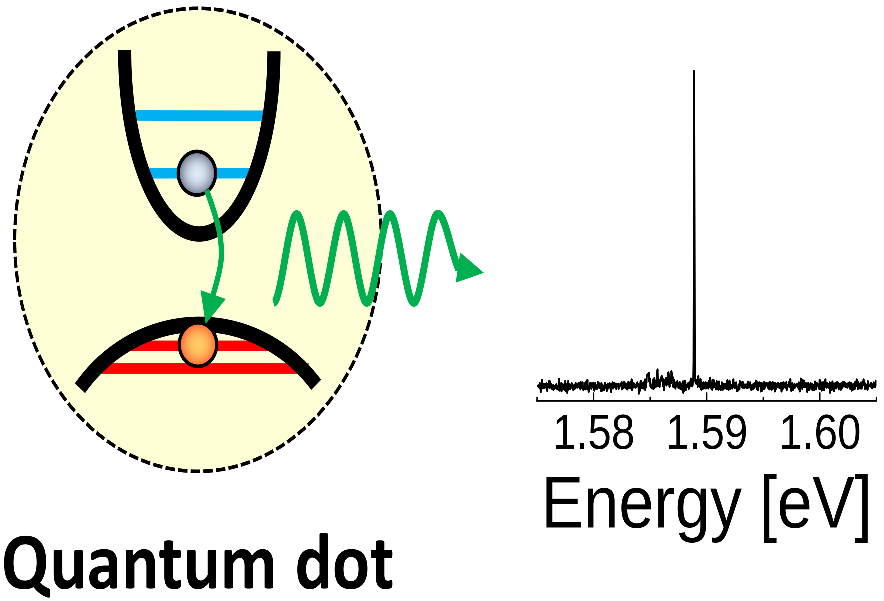
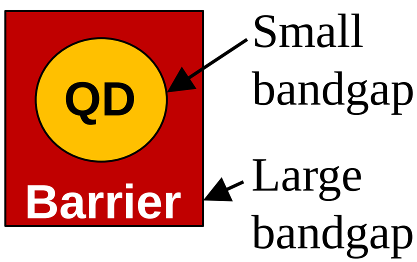
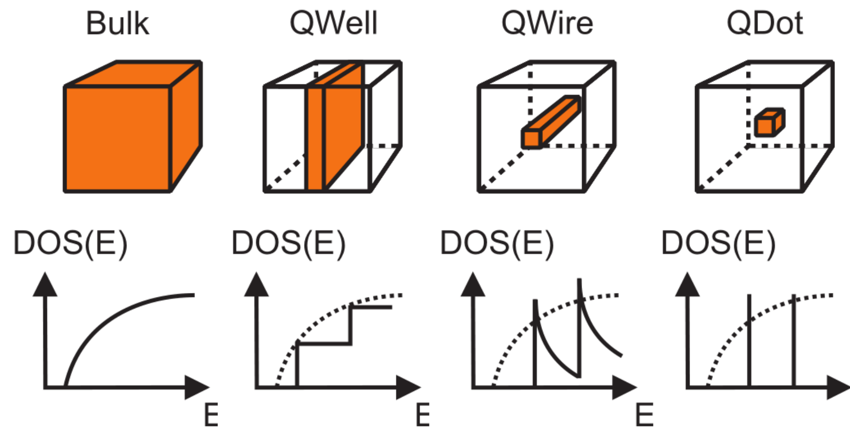
Praktikumsversuch:
Photolumineszenz von Halbleiter-Quantenpunkten



Semiconductor Quantum Dots (QD)



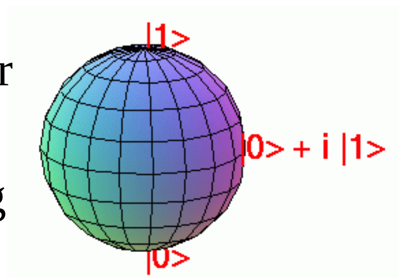
Artificial atoms
(size ≲ 50 nm)
discrete density of states (DOS) with quantized energy levels



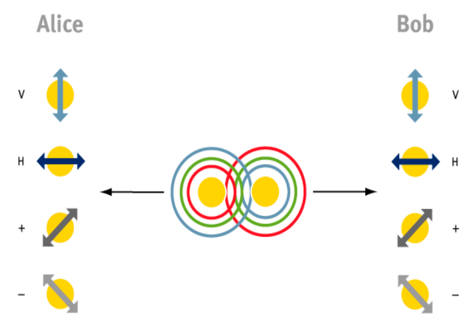
__Applications__

- **Laser, Solar cells:** dense QD ensembles
- **Quantum information:** single QDs

QuBits for quantum computing



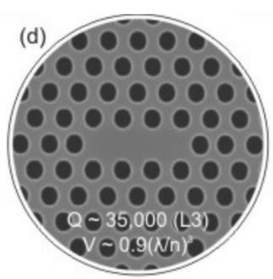
Entangled photons for quantum cryptography



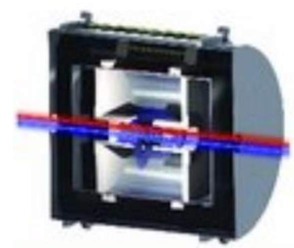
Quantum sensors, ….

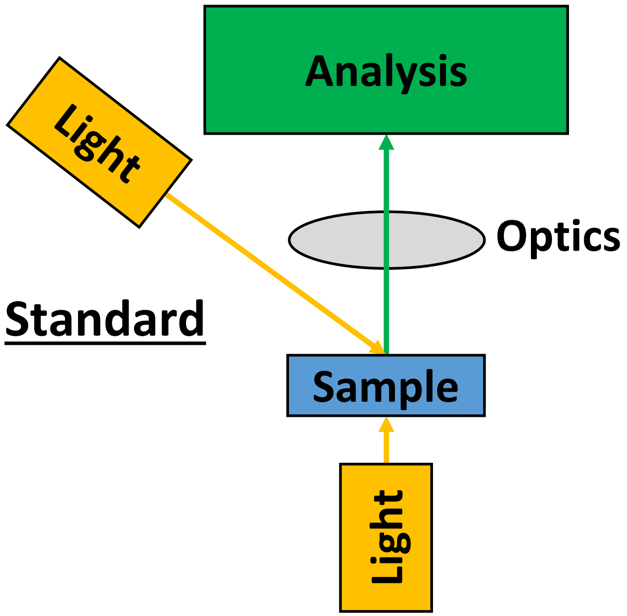
- **Integration:**

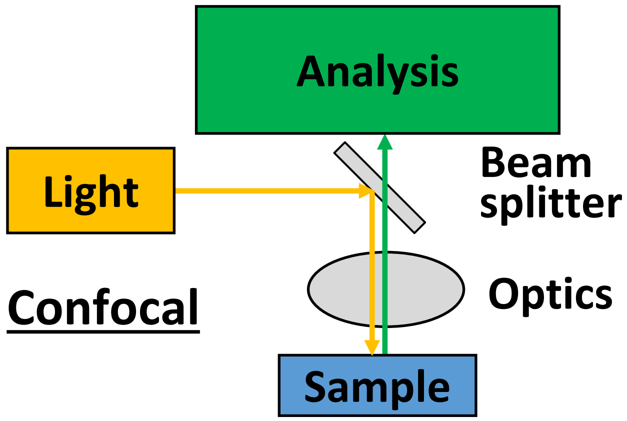
Photonic environment
Quantum photonic integrated circuits,
quantum network



Rb vapour cell as a **quantum memory**







Luminescence: „cold light"
Photoluminescence: inelastic light scattering

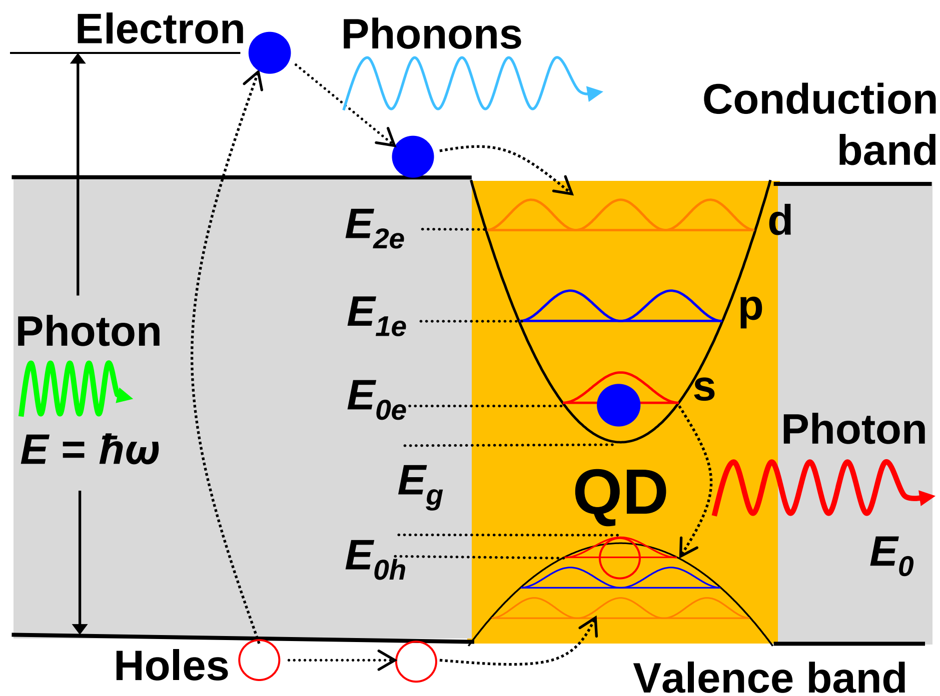
Photoluminescence (PL) spectroscopy:
- Excitation by laser in reflection
- Often excitation / emission through one objektive
- Spectral analysis of the emission
- Information: energy of the states inside the sample (absolute energy)

Raman spectroscopy:
- Setup like PL, in addition precise laser filter
- Information: energy loss in the sample (energy relative to the laser energy)

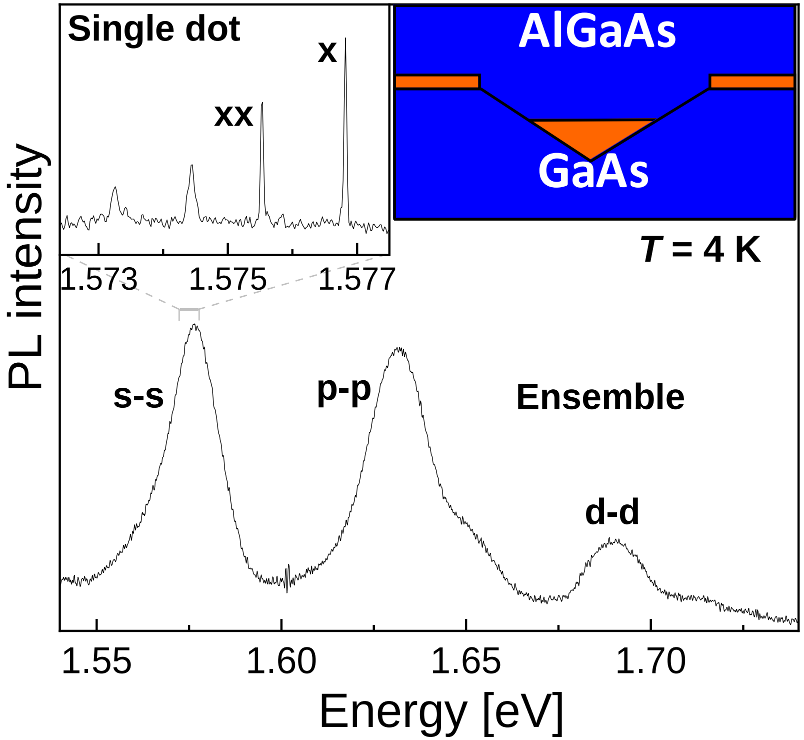
PL spectroscopy:

- Excitation with monochromatic light either resonantly or non-resonantly into the barrier material generates electron-hole-pairs (excitons)
- Excitons diffuse into positions with lowest energy (e.g. QD states)
- The excess energy is relaxed by phonon emission (Raman effect)
- Radiative recombination of excitons by emission of photons with energy:

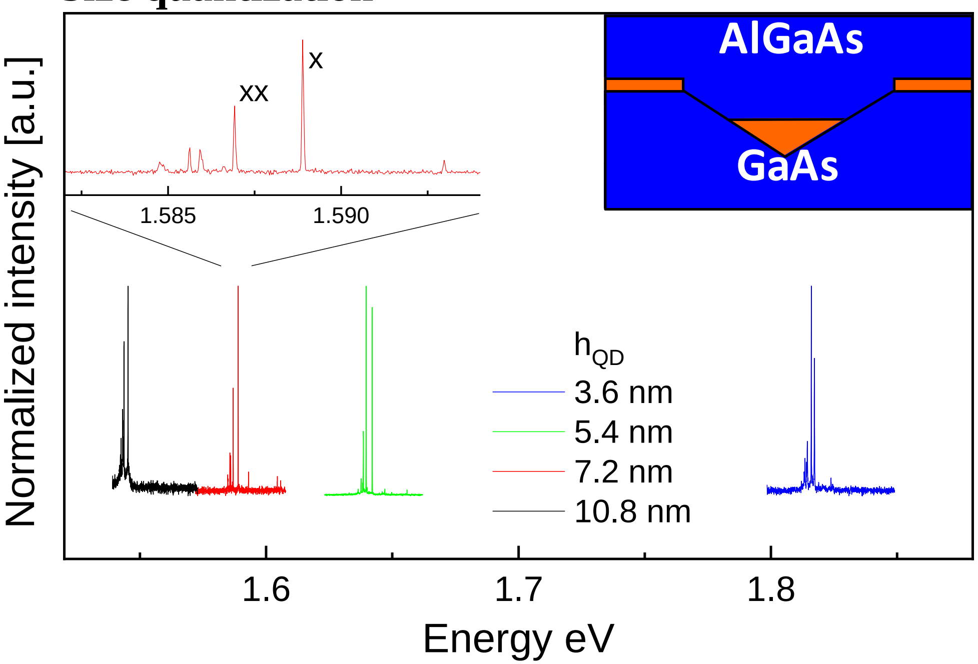
  $E_n = E_g + E_{ne} + E_{nh} - E_C$, with bandgap energy E_g of the QD material, electron and hole quantization energies E_{ne}, E_{nh}, exciton binding energy E_C (Coulomb interaction)
- Optical selection rules: transitions between e-h states with equal quantum numbers have the highest probability
- In GaAs, the light-hole (lh) density of states (DOS) is 6% of the total DOS (hh+lh): heavy holes (hh) dominate the PL emission



PL Spectra: Example GaAs QDs



[Appl. Phys. Lett. ***94****, 183113 (2009),*
New Journal of Physics ***14*** *(2012)]*



Ensemble (macro-PL):

- <u>QD shell structure:</u> splitting by <u>quantization energy</u> ΔE = 5-100 meV
- Linewidth: 10-50 meV, broadened by QD size fluctuation (Gaussian distribution)

Single-dot (mikro-PL, confocal microscope):

- <u>Excitonic complexes</u>, splitting be Coulom-interaction ΔE = 0.5-3 meV
- Linewidth: natural linewidth a few μeV (Lorentzian distribution)
  often broadened by fluctuating charges

Harmonic Oscillator: single-Particle frame

For a QD size < de Broglie wavelength: size quantization becomes relevant

Quantization energy in Cartesian coordinates: $\boldsymbol{E_n = E_x + E_y + E_z}$

QDs have often approximately equidistant energy levels ⇒ parabolic potential

1D parabolic potential: $V = \frac{1}{2} m^* \omega_x^2 x^2$, with effective mass m^*, oscillator frequency ω_x

Eigenenergies of 1D harmonic oscillator: $E_x = \hbar\omega_x \left(n_x + \frac{1}{2}\right)$,

with quantum number $n_x = 0, 1, 2, 3 \ldots.$

Approximation: QD size ≅ ground-state oscillator length L:

For $x = \frac{L}{2}$: $\quad V(x) = E_0$ and $\boldsymbol{L^2 = \frac{4\hbar^2}{m^* \hbar\omega_x}}$

Spherical QD shape ($\omega_x = \omega_y = \omega_z = \omega_0$)

$\boldsymbol{E_n = E_x + E_y + E_z = \hbar\omega_0 \left(n + \frac{3}{2}\right)}$,

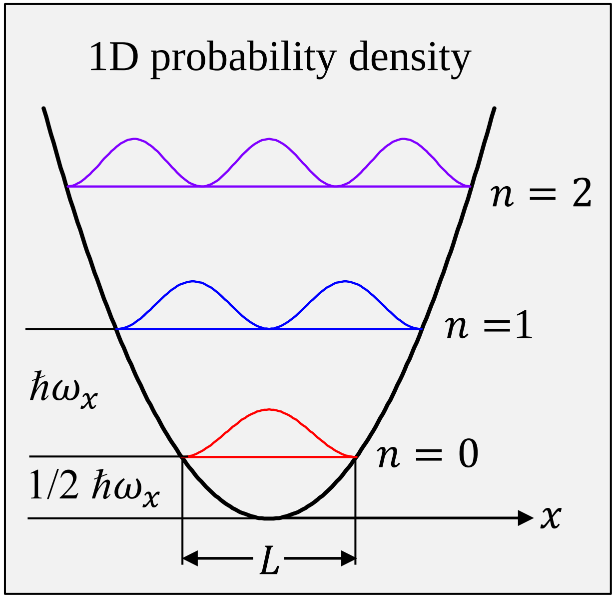
with $n = n_{xyz} = n_x + n_y + n_z$

Degeneracy (without spin):

$n = 0$: no degeneracy

$n = 1$: deg. = 3 ($n_{100}, n_{010}, n_{001}$)

$n = 2$: deg. = 6 ($n_{110}, n_{011}, n_{101}, n_{200}, n_{020}, n_{002}$)



Harmonic Oscillator: Size Quantization

Optical emission from QDs: radiative recombinations of excitons

QD emission energy: $\boldsymbol{E_n = E_g + E_{ne} + E_{nh} - E_C}$,

with bandgap energy E_g of the QD material, electron and hole quantization energies E_{ne}, E_{nh}, and exciton binding energy E_C (Coulomb interaction)

Approximations: 3D harmonic oscillator, spherical QD shape, no Coulomb interaction, $n = 0$:

$$E_{ne} = \left(n + \frac{3}{2}\right)\hbar\omega_e = \frac{3}{2}\hbar\omega_e,\; E_{nh} = \frac{3}{2}\hbar\omega_h$$

With $\hbar\omega_e = \frac{4\hbar^2}{m_e^* L^2}$, $\hbar\omega_h = \frac{4\hbar^2}{m_h^* L^2}$:

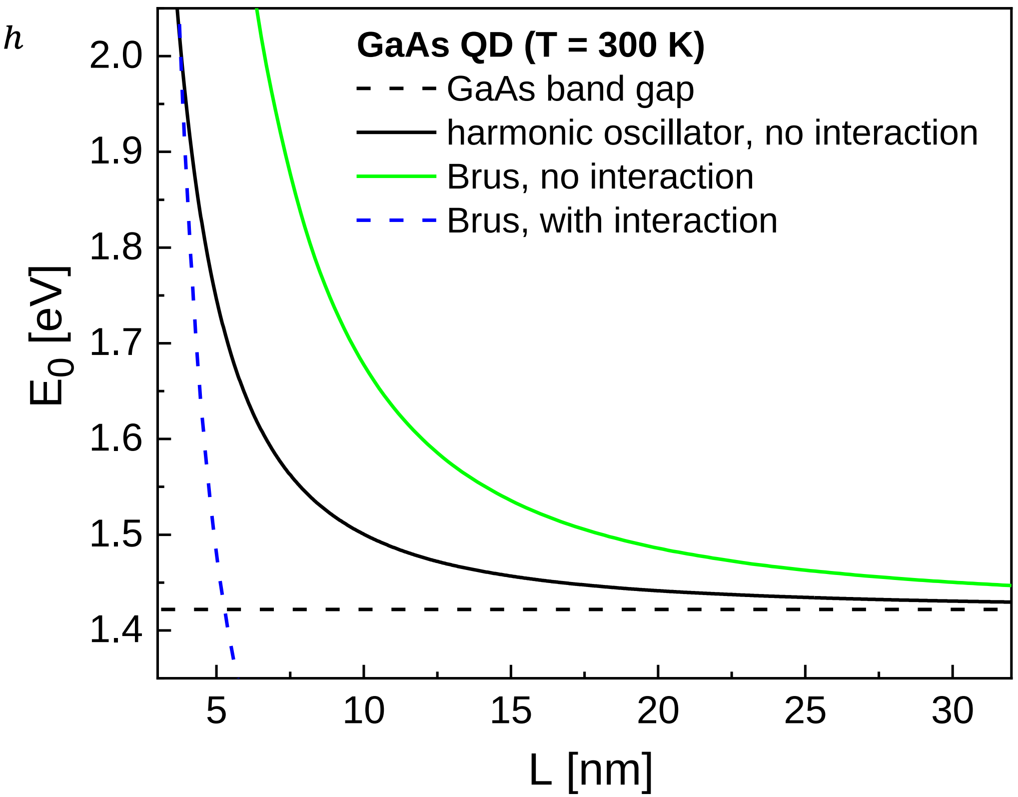
$$\boldsymbol{E_0 = E_g + \frac{6\hbar^2}{L^2}\left(\frac{1}{m_e^*} + \frac{1}{m_h^*}\right)}$$

The well known Brus equation considers also an approximation for Coulomb interaction:

$$E_0 = E_g + \frac{2\pi^2\hbar^2}{L^2}\left(\frac{1}{m_e^*} + \frac{1}{m_h^*}\right) - \frac{3.6e^2}{\varepsilon_s L}$$

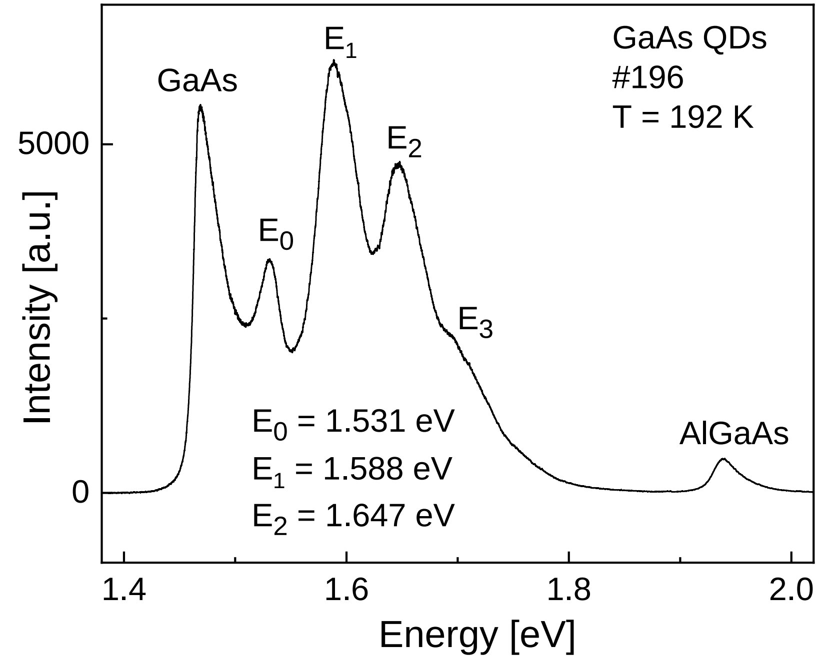
with the semiconductor permittivity ε_s

[L. Brus, J. Phys. Chem. ***90****, 2555 (1986)]*



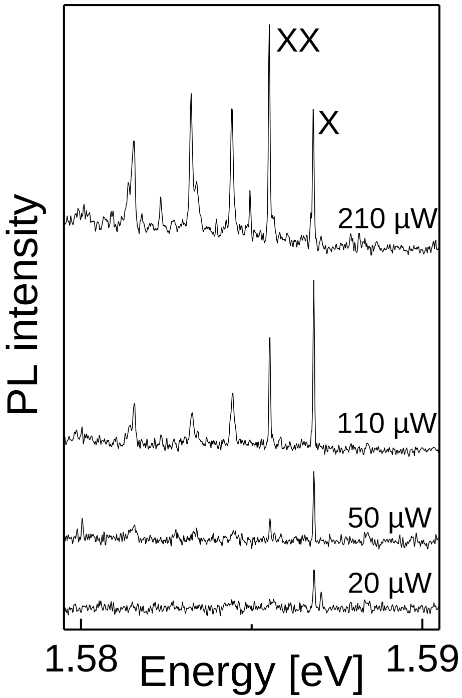
Example: PL spectrum of GaAs QDs in AlGaAs

- Bulk GaAs Peak at 1.469 eV (E_g at $T = 192$ K)
- QDs: nearly equidistant peaks
  $E_1 - E_0 \cong E_2 - E_1$, $(E_n = E_g + E_{ne} + E_{nh} - E_C)$
  Harmonic oscillator approximation can be used:
  $E_{ne} = \left(n + \frac{3}{2}\right)\hbar\omega_e,\ E_{nh} = \left(n + \frac{3}{2}\right)\hbar\omega_h$
- Electron, hole quantization (harmonic oscillator):
  $E_1 - E_0 = \hbar\omega_e + \hbar\omega_h = 57$ meV

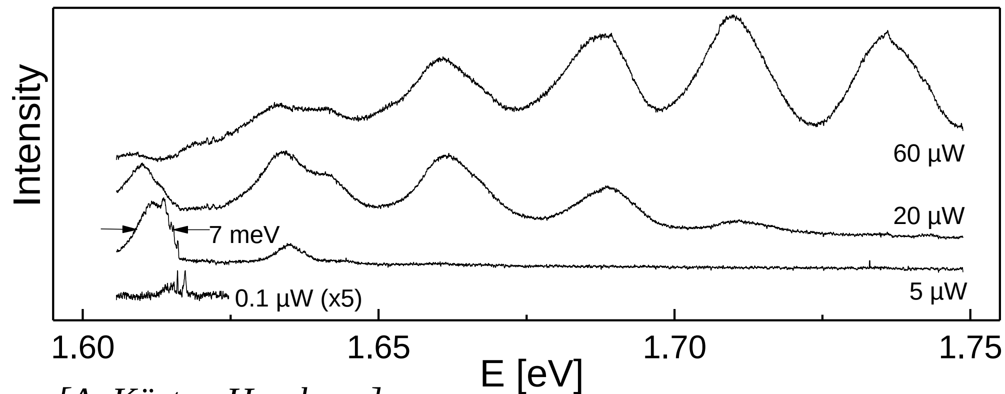


- Oscillator length: $L^2 = \frac{4\hbar^2}{m_e^*\hbar\omega_e} = \frac{4\hbar^2}{m_h^*\hbar\omega_h}$
  yields: $\frac{\omega_e}{\omega_h} = \frac{m_h^*}{m_e^*}$ and: $E_1 - E_0 = \hbar\omega_e\left(1 + \frac{m_e^*}{m_h^*}\right)$ or: $\hbar\omega_e = \frac{E_1 - E_0}{1 + \frac{m_e^*}{m_h^*}}$ and: $\hbar\omega_h = \frac{E_1 - E_0}{1 + \frac{m_h^*}{m_e^*}}$
  For GaAs ($m_e^* = 0.066$, $m_{hh}^* = 0.5$): $\hbar\omega_e = 50.4$ meV, $\hbar\omega_h = 6.6$ meV
  Estimation of QD diameter: $L = \left(\frac{4\hbar^2}{m_e^*\hbar\omega_e}\right)^{1/2} = 9.6$ nm
- Coulomb interaction (approx.: spherical QD):
  $E_0 - E_g = \frac{3}{2}(\hbar\omega_e + \hbar\omega_h) - E_C$ and $E_1 - E_0 = \hbar\omega_e + \hbar\omega_h$
  yields: $E_C = \frac{3}{2}(E_1 - E_0) - (E_0 - E_g) = 20.7$ meV

Excitonic Complexes



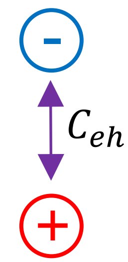
[Nanoscale Res. Lett. **5**, 1633 *(2010)]*



[A. Küster, Hamburg]

Binding energy

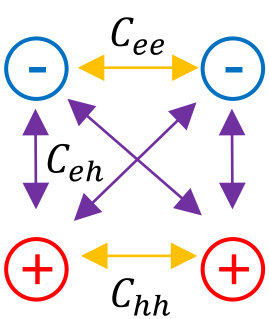
Exciton



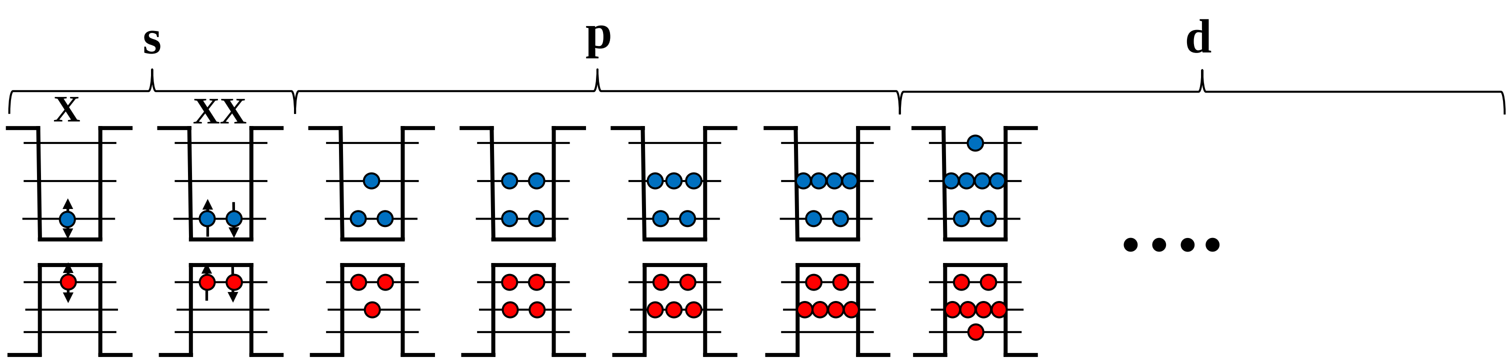
Excitonic states:

- s-level: up to 2 electrons and holes (spin-splitted): exciton X, biexciton XX
- Optical emission by radiative recombination of one exciton
- The presence of additional charge carriers shifts the emission energy due to Coulomb interactions (C_{eh}, C_{ee}, C_{hh})

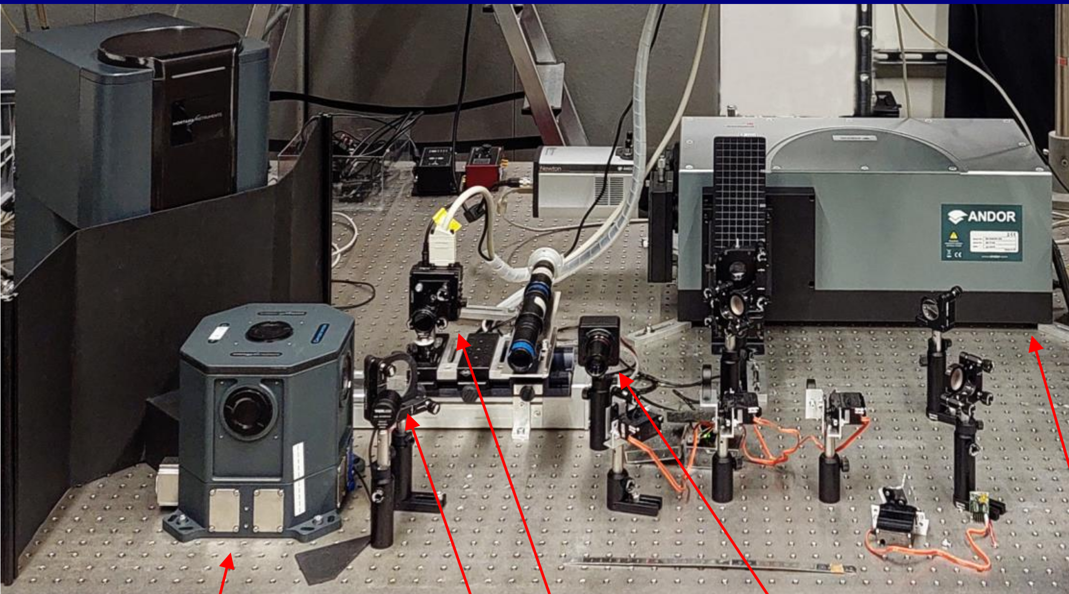
Biexciton



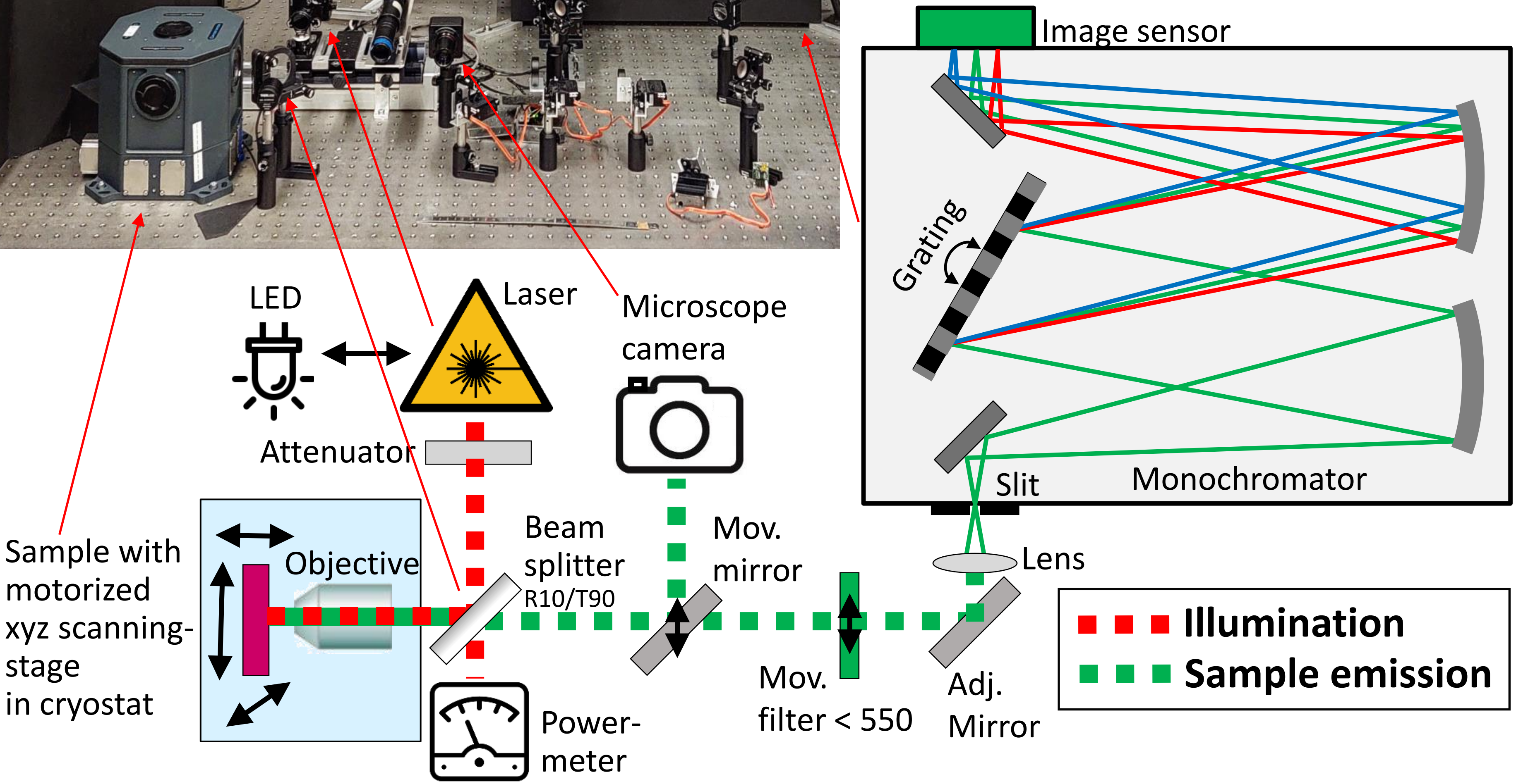
Neutral states of a disk-shaped QD

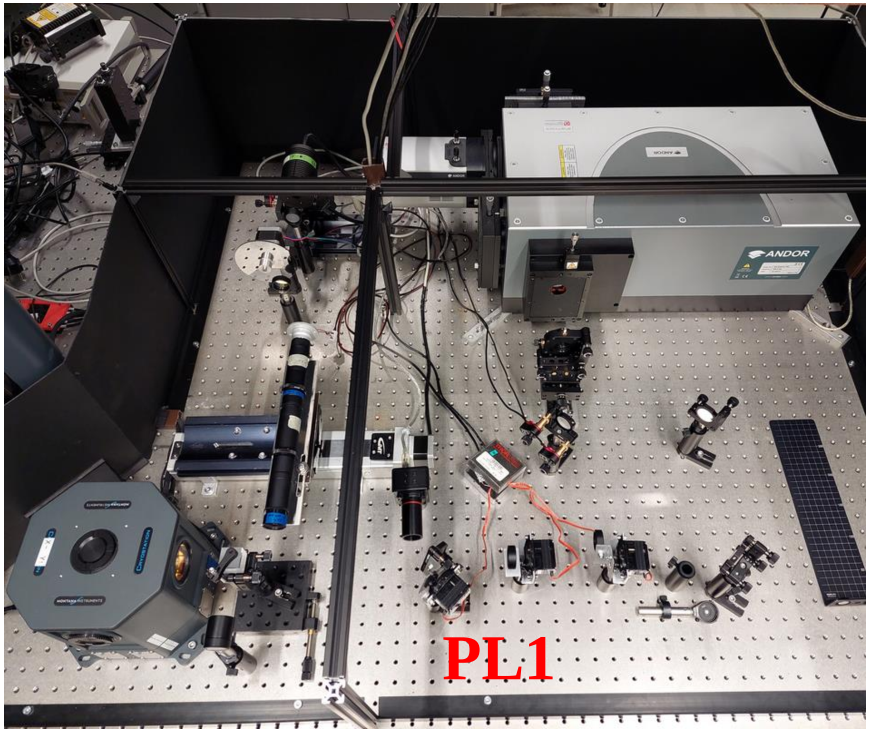


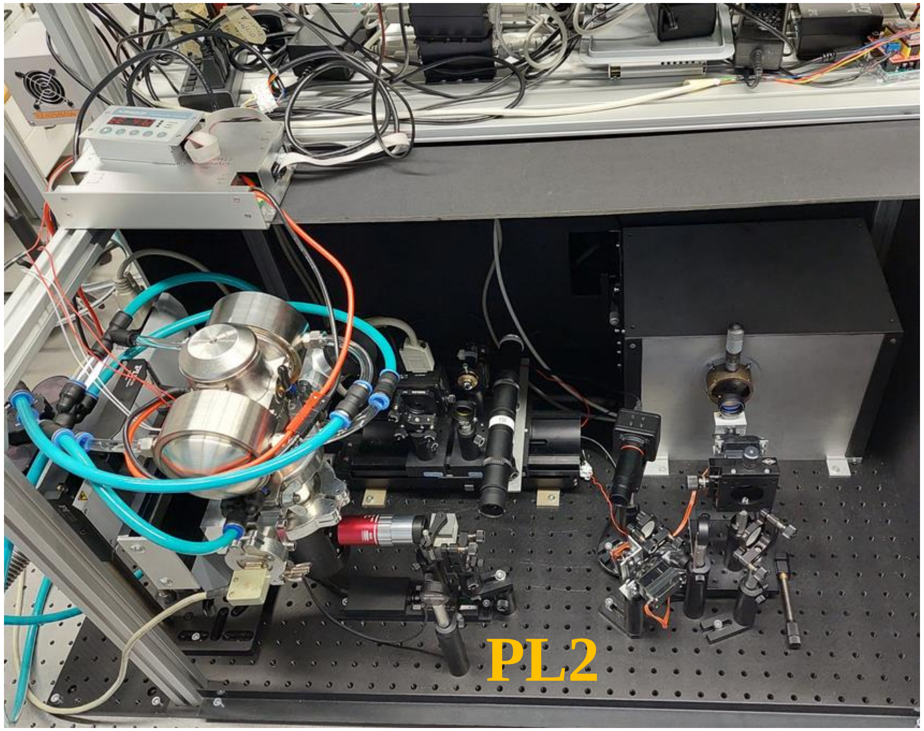
Basic Micro-PL Spectrometer



- **Sample cooling (4 K)**
- **Micro-PL (confocal microscope)**
- **Extensions: ultra-high resolution, lifetime, polarization, correlation, …**







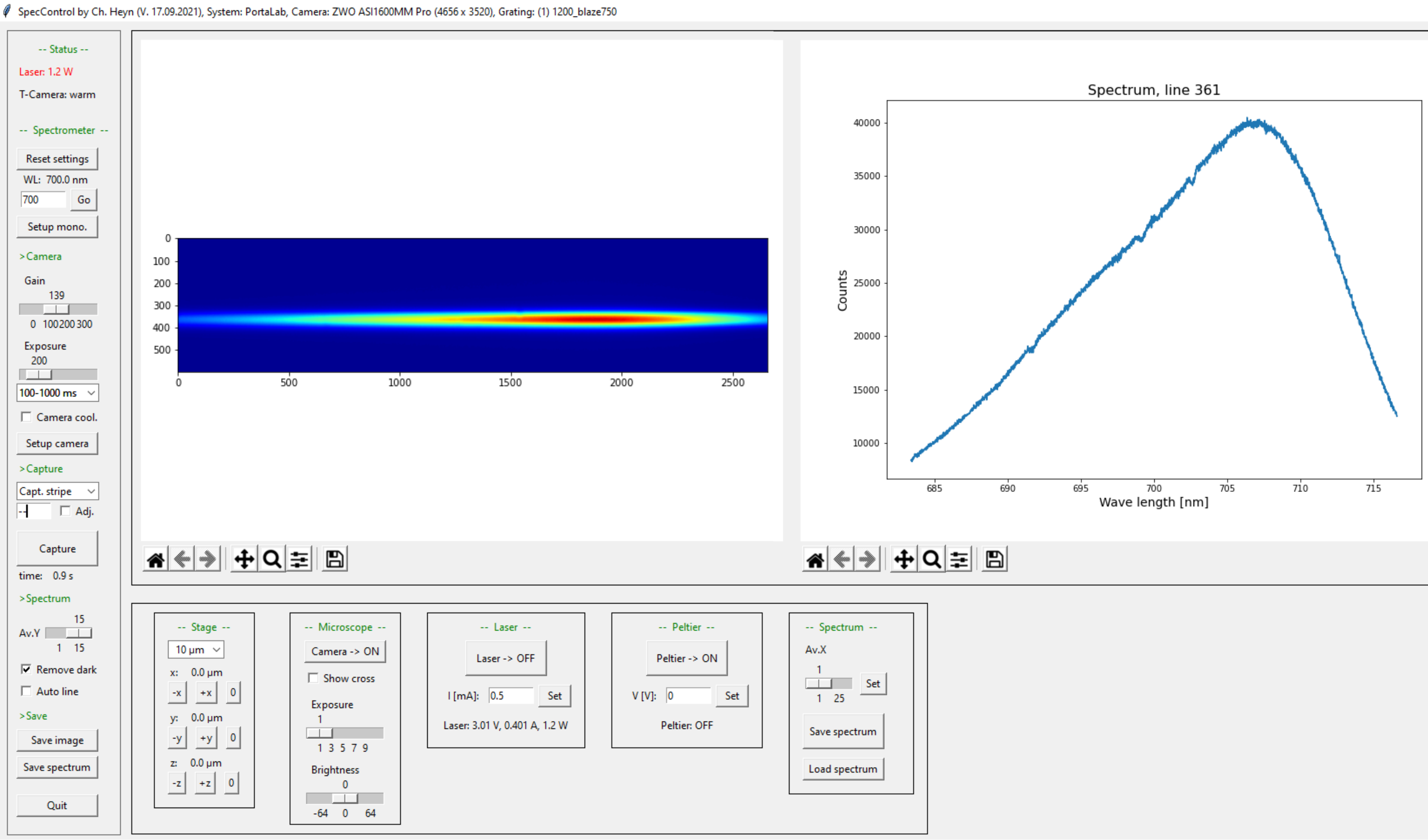
<u>PL1</u>

- **Sample cooling down to 3 K**
- **Ultra-low vibrations**
- **Extensions: high resolution, lifetime**

<u>PL2</u>

- **Sample cooling down to 30 K**

PL-Setups: Software

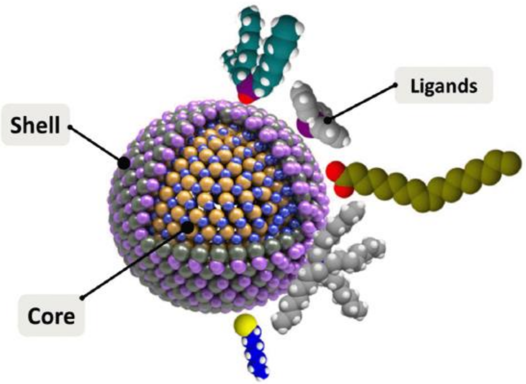


Fabrication of Semiconductor Quantum Dots

Synthesis of colloidal QDs

Powder / in a liquid

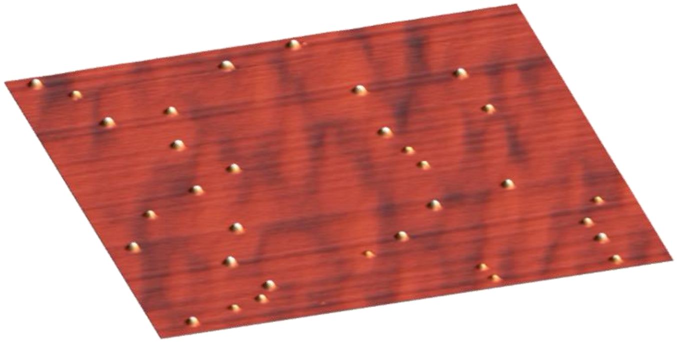
+ Mass production
- Contacts (ligands)
- Optical instability (blinking)

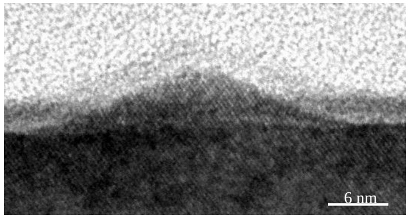


Self-assembly of epitaxial QDs

Wafer based

+ Tunable density
+ Contacts
+ Optically stable

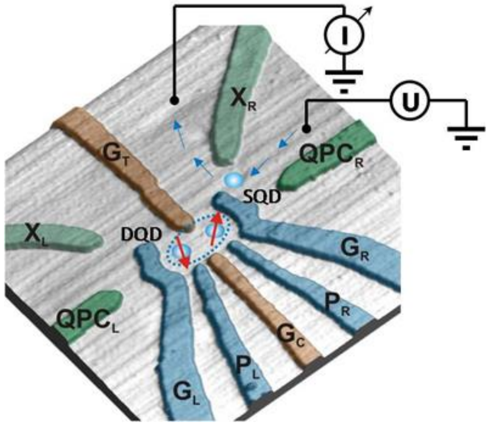




Lithography by gates, etching

Wafer based

+ Single QDs
+ Contacts
- Optical emission



Fabrication of Semiconductor Quantum Dots

Synthesis
of colloidal QDs

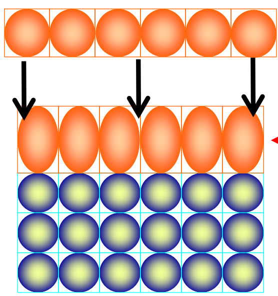
**Self-assembly
of epitaxial QDs**

Lithography
by gates, etching

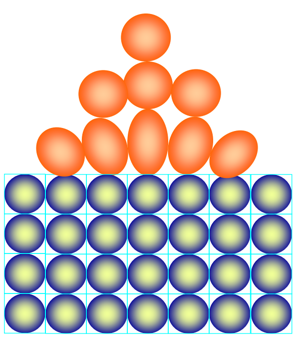
Strain induced
(Stranski-Krastanov)
e.g. InAs/GaAs, Ge/Si

Droplet Epitaxy
(Volmer-Weber)
e.g. GaAs/AlGaAs

Droplet Etching
(Volmer-Weber)
e.g. GaAs/AlGaAs

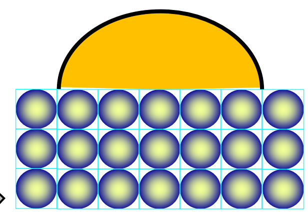


strained

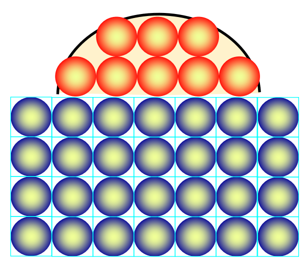


strained

Droplet deposition (Ga, Al, In, ..)

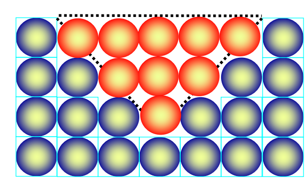


Crystallization



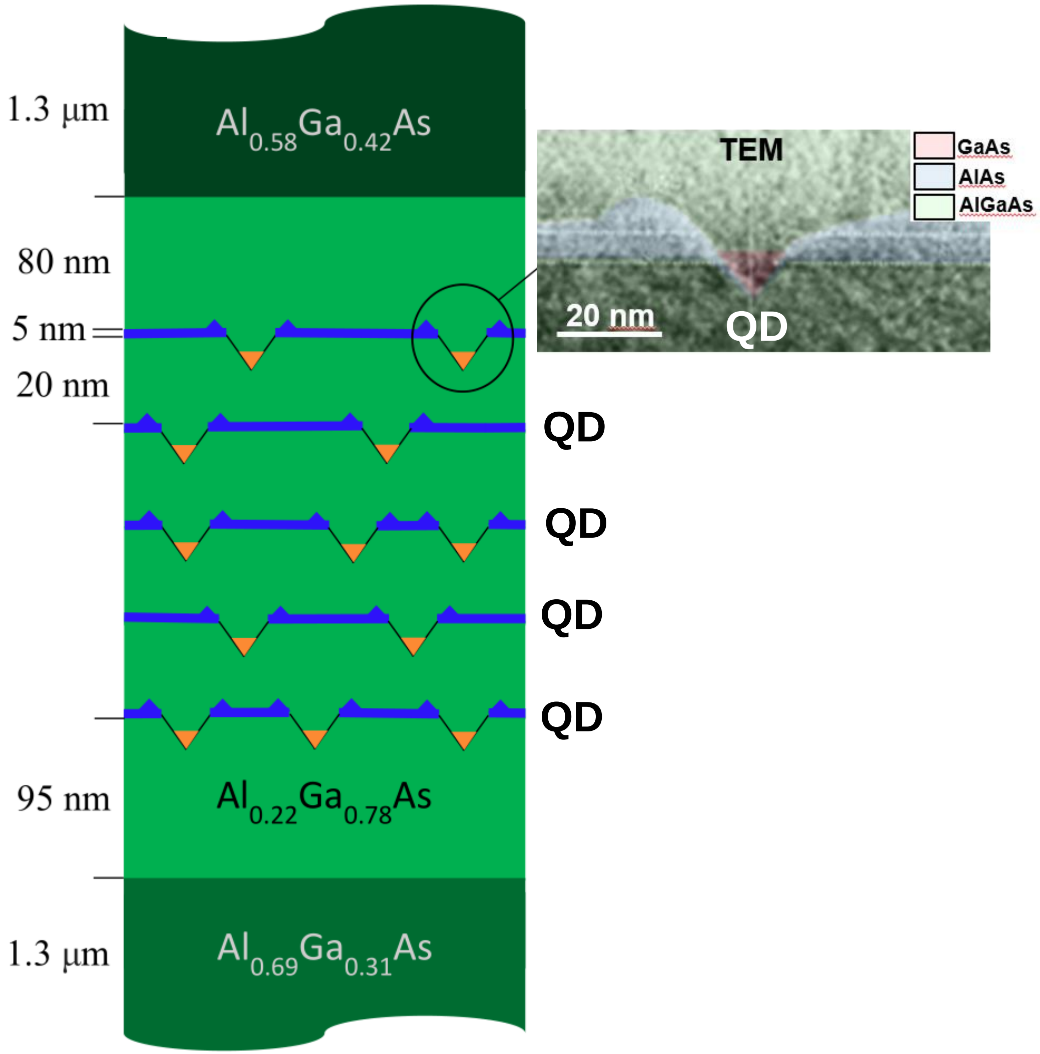
unstrained

Nanohole drilling
Filling with QD

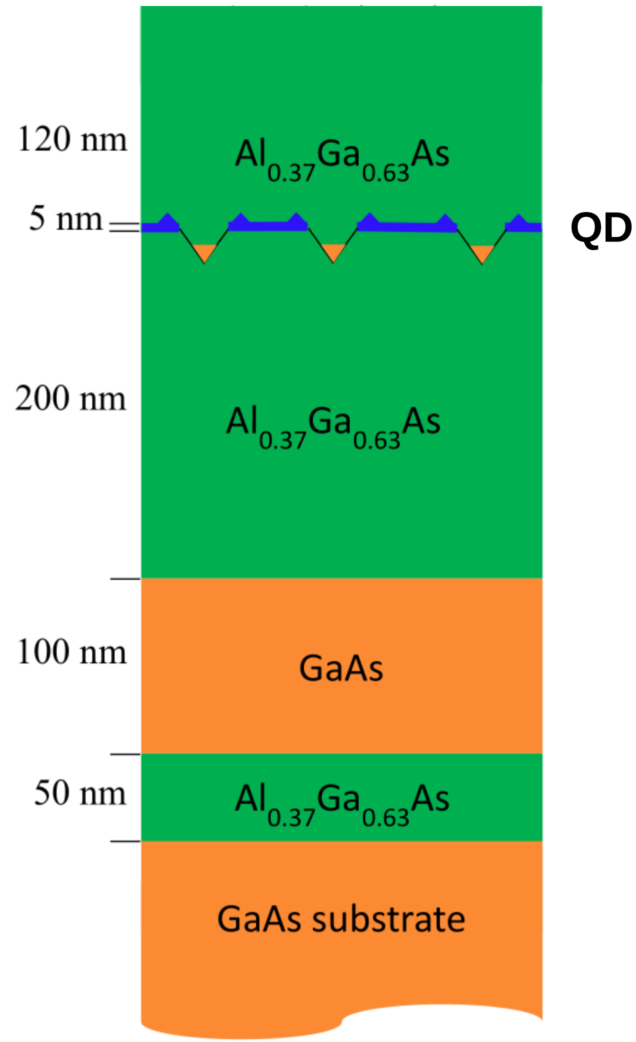


QD Samples

QD–Probe A („Spezial“)
5 Lagen GaAs QD in dickem AlGaAs

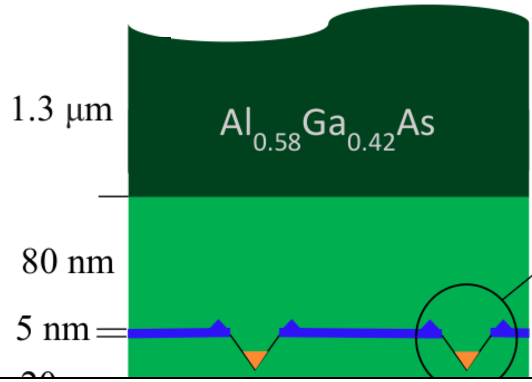


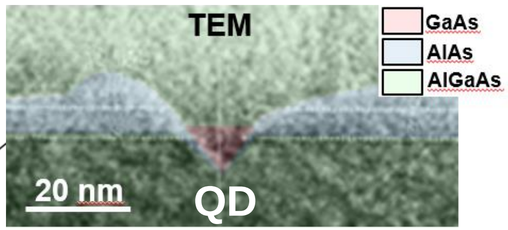
QD–Probe B („Standard“)
1 Lage GaAs QD in dünnem AlGaAs

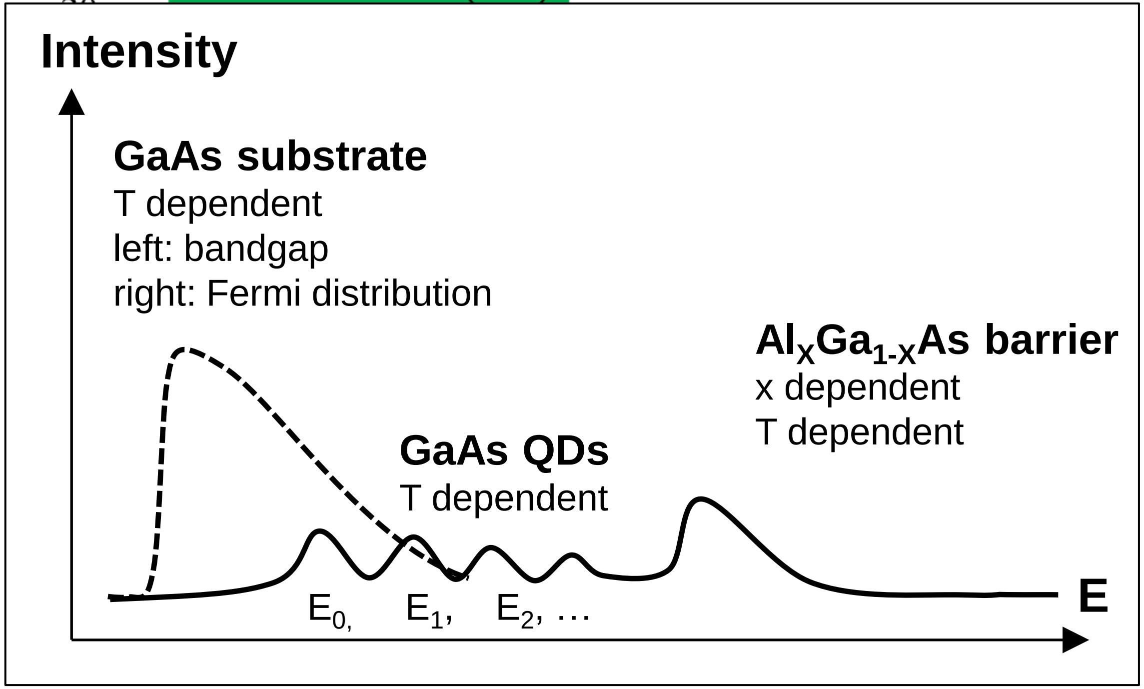


QD Samples

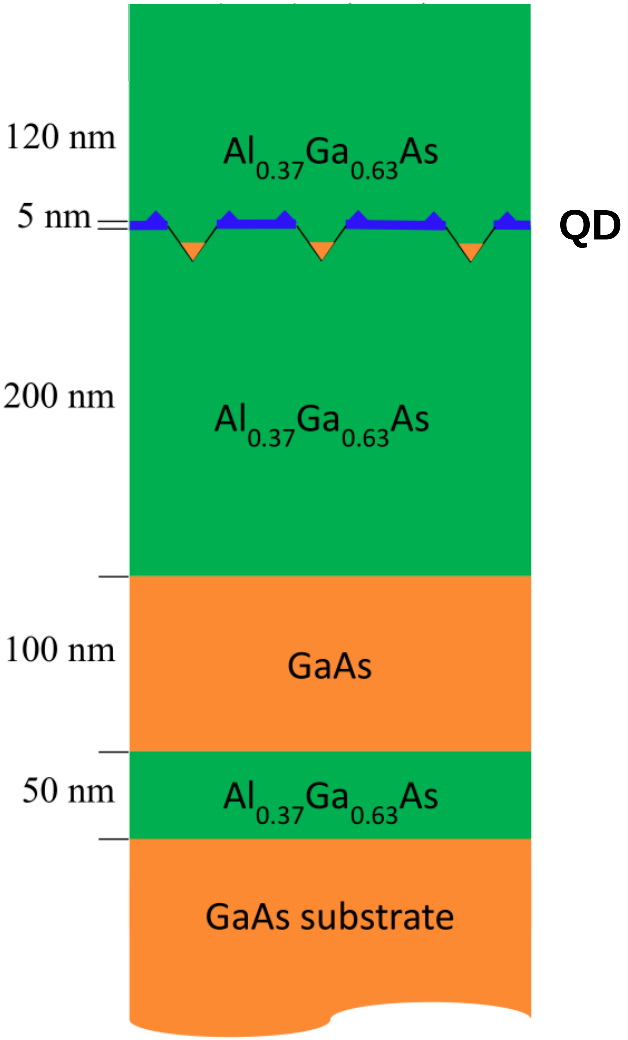
QD–Probe A („Spezial“)
5 Lagen GaAs QD in dickem AlGaAs



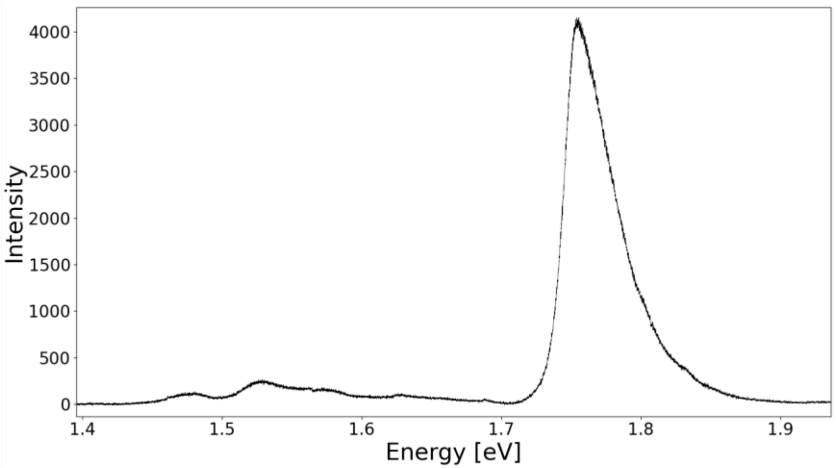




QD–Probe B („Standard“)
1 Lage GaAs QD in dünnem AlGaAs

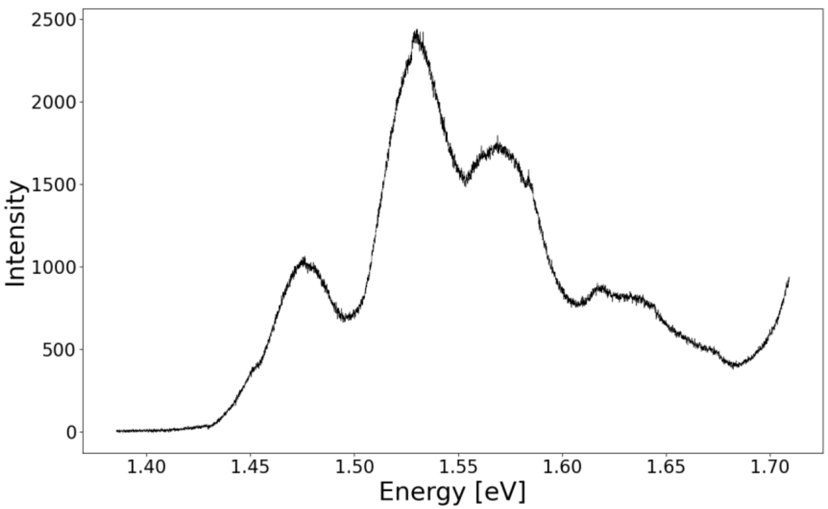


QD Sample A



Messung an QD-Probe A bei T = 300 K:
(Bereich 600-900 nm, zusammengesetzt aus Einzelspektren)

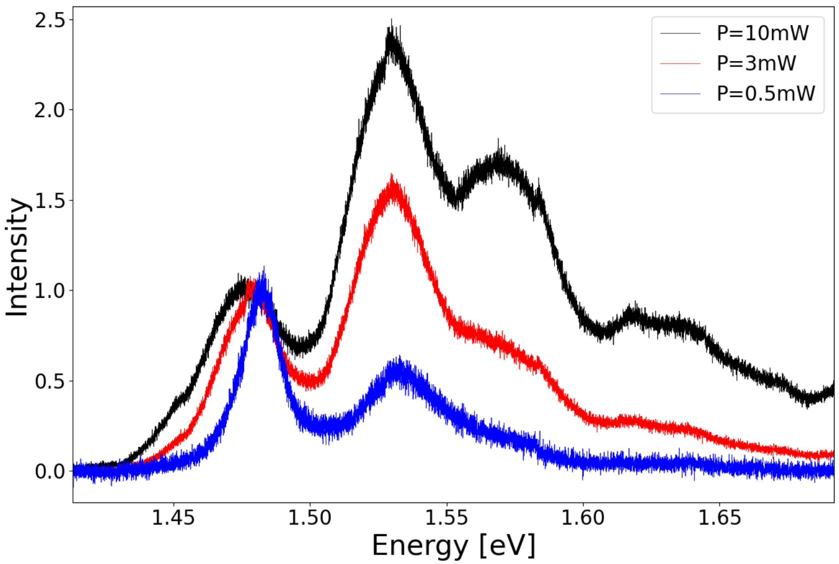
- Starker AlGaAs-Peak + schwache QD Peaks
- Zoom: QD-Peaks mit 4 Energieniveaus



Auswertungen:

- QD-Peaks sind (halbwegs) äquidistant:
  → parabolisches Potenzial
- Bestimmung der Quantisierungsenergien
- Abschätzung der QD-Größe (Ausdehnung der Wellenfunktion)
- Abschätzung der Coulomb-Wechselwirkung zwischen Elektron und Loch

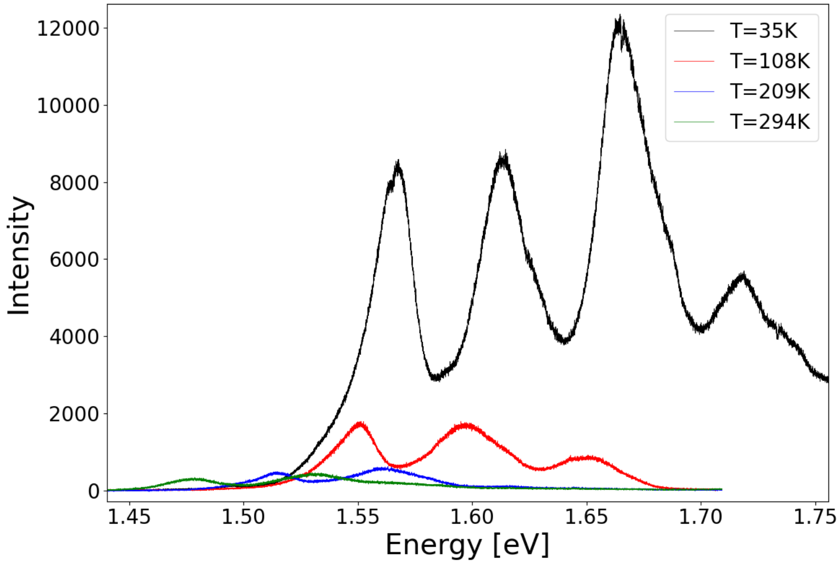
QD Sample A



QD-Probe A, Einfluss Laserleistung P

Bei reduzierter Laserleistung:

- Abnehmende Intensität
- Relative Abnahme höherer Schalen (Besetzungsstatistik)
- Blauverschiebung des Grundzustandes (Rotverschiebung durch höhere Schalen)



QD-Probe A, Einfluss Probentemperatur T

Bei Kühlung:

- Intensitätserhöhung (Reduktion thermischer Emission aus den QD)
- Blauverschiebung der Energieniveaus (Analog GaAs-Bandlücke)
- Besetzung höherer Schalen

Location

Meeting point:

Office
Christian Heyn

Center for Hybrid
Nanostructures
(CHyN)
Luruper Chaussee 149
Building 600

Room: **2.46**