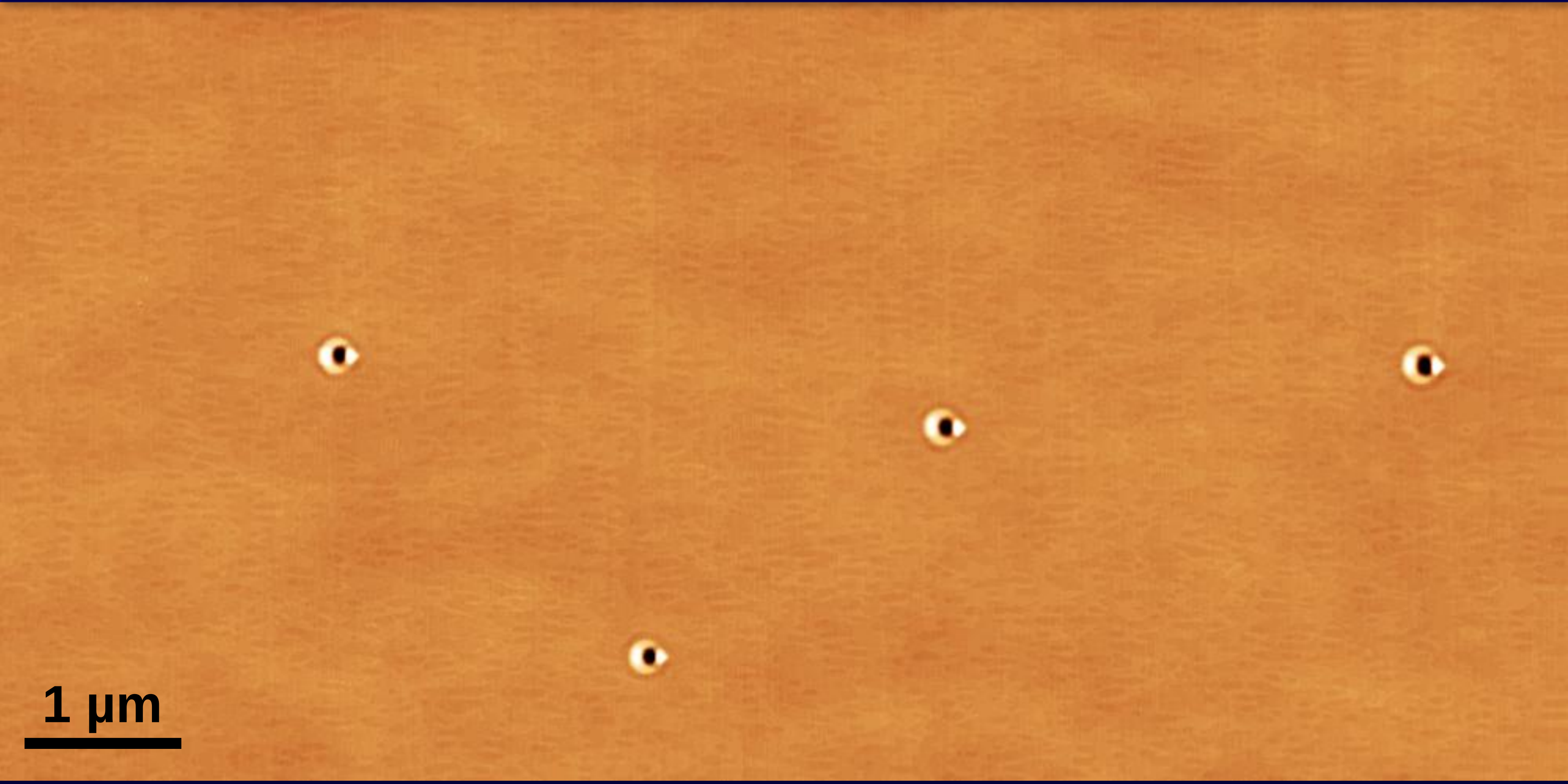
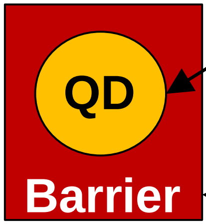
Self-assembled Droplet Etching for versatile Quantum Structures

Ch. Heyn, L. Ranasinghe, K. Denecke, S. Federsen, and W. Hansen

CHyN, University of Hamburg



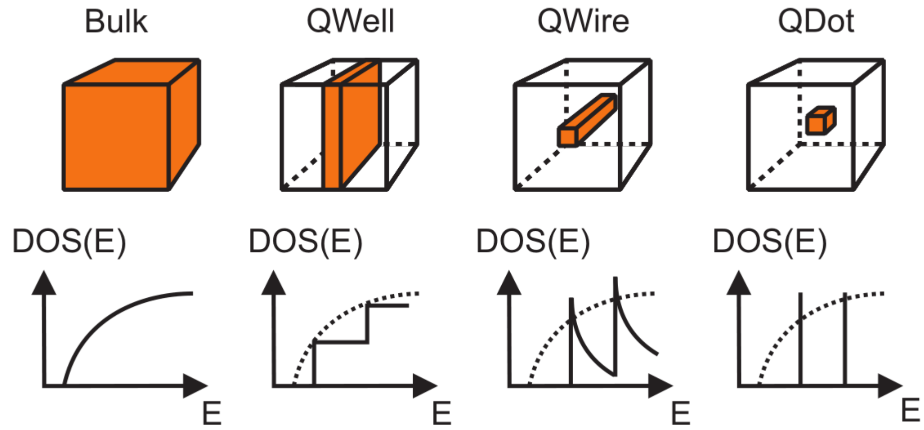
Semiconductor Quantum Dots

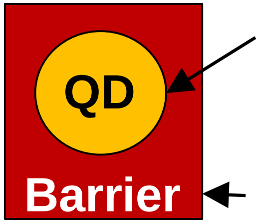


Small bandgap

Large bandgap

Size ≲ 50 nm:
atom-like
0D density
of states





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Size $\lesssim$ 50 nm:
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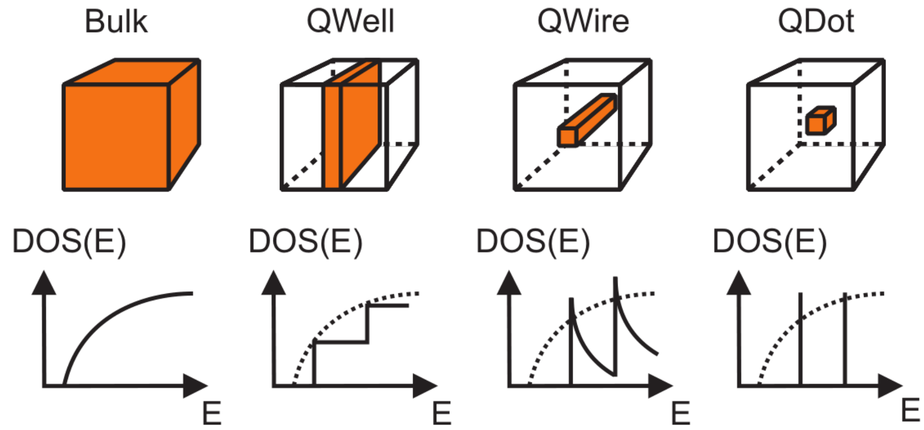
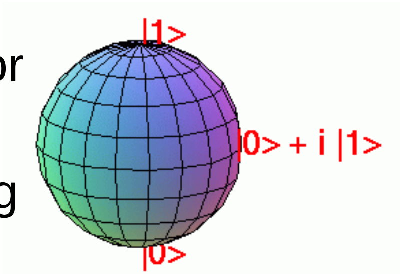


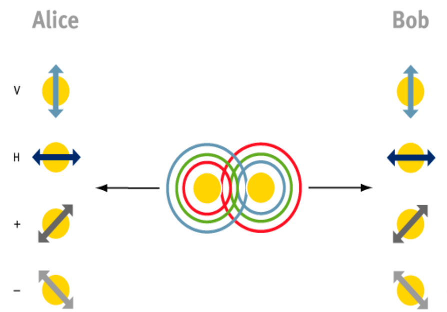
Applications

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

QuBits for quantum computing



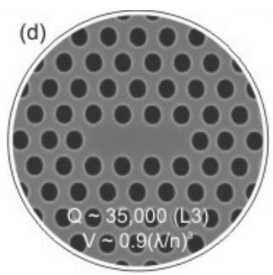
Entangled photons for quantum cryptography



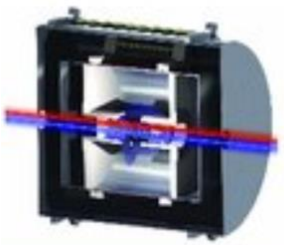
Quantum sensors, ….

- **Integration:**

Photonic environment
Quantum photonic integrated circuits, quantum network



Rb vapour cell as a quantum memory



Fabrication of Semiconductor Quantum Dots

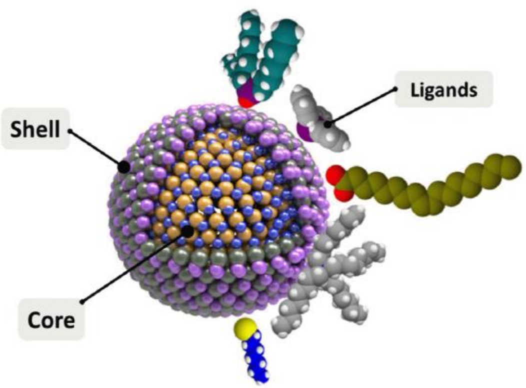
Synthesis of colloidal QDs

Powder / in a liquid

+ Mass production

- Contacts (ligands)

- Optical instability (blinking)



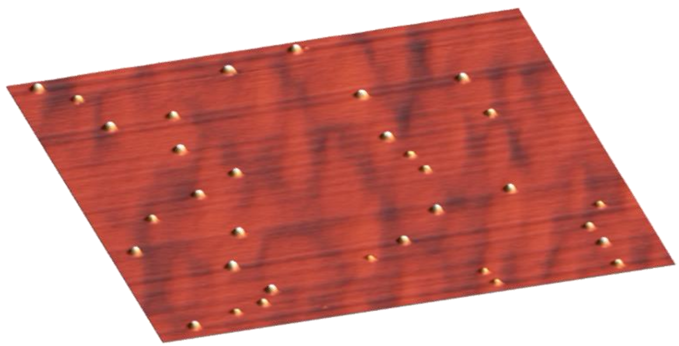
Self-assembly of epitaxial QDs

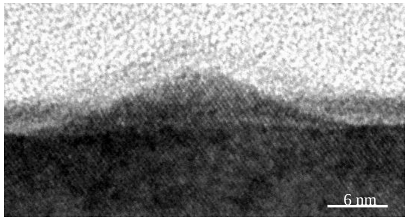
Wafer based

+ Tunable density

+ Contacts

+ Optically stable





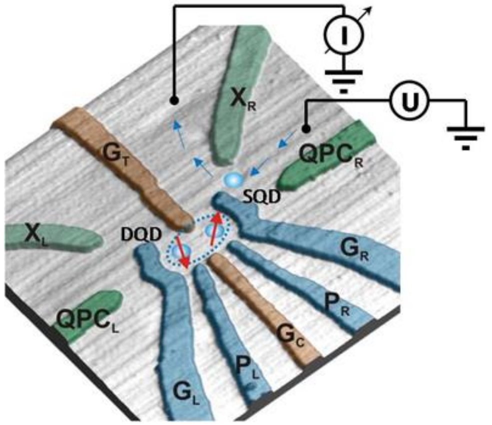
Lithography by gates, etching

Wafer based

+ Single QDs

+ Contacts

- Optical emission



Fabrication of Semiconductor Quantum Dots

Synthesis of colloidal QDs

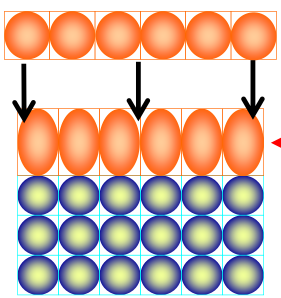
Self-assembly of epitaxial QDs

Lithography by gates, etching

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

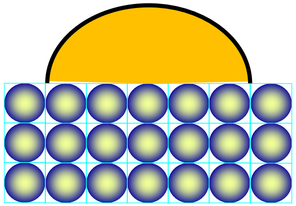
Droplet Epitaxy
(Volmer-Weber)
e.g. GaAs/AlGaAs

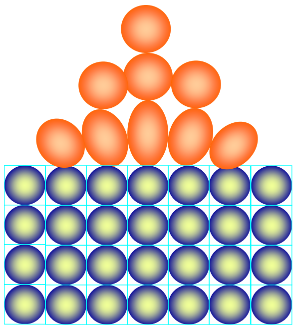
Droplet Etching
(Volmer-Weber)
e.g. GaAs/AlGaAs



strained

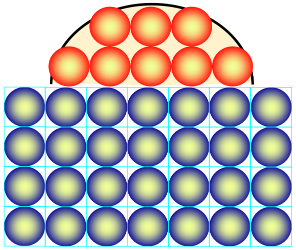
Droplet deposition (Ga, Al, In, ..)





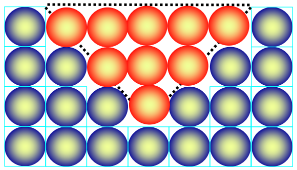
strained

Crystallization



unstrained

Nanohole drilling
Filling with QD



Fabrication of Semiconductor Quantum Dots

Synthesis of colloidal QDs

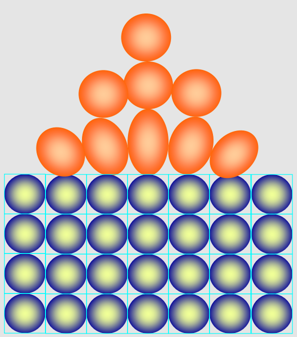
Self-assembly of epitaxial QDs

Lithography by gates, etching

Strain induced
(Stranski-Krastanov)

InAs/GaAs

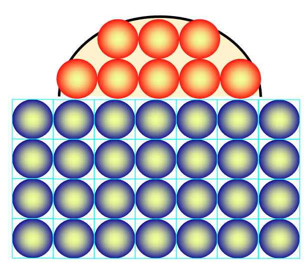
Emission: 1-1.3 µm



strained

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

Crystallization



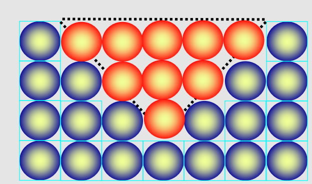
unstrained

Droplet Etching
(Volmer-Weber)

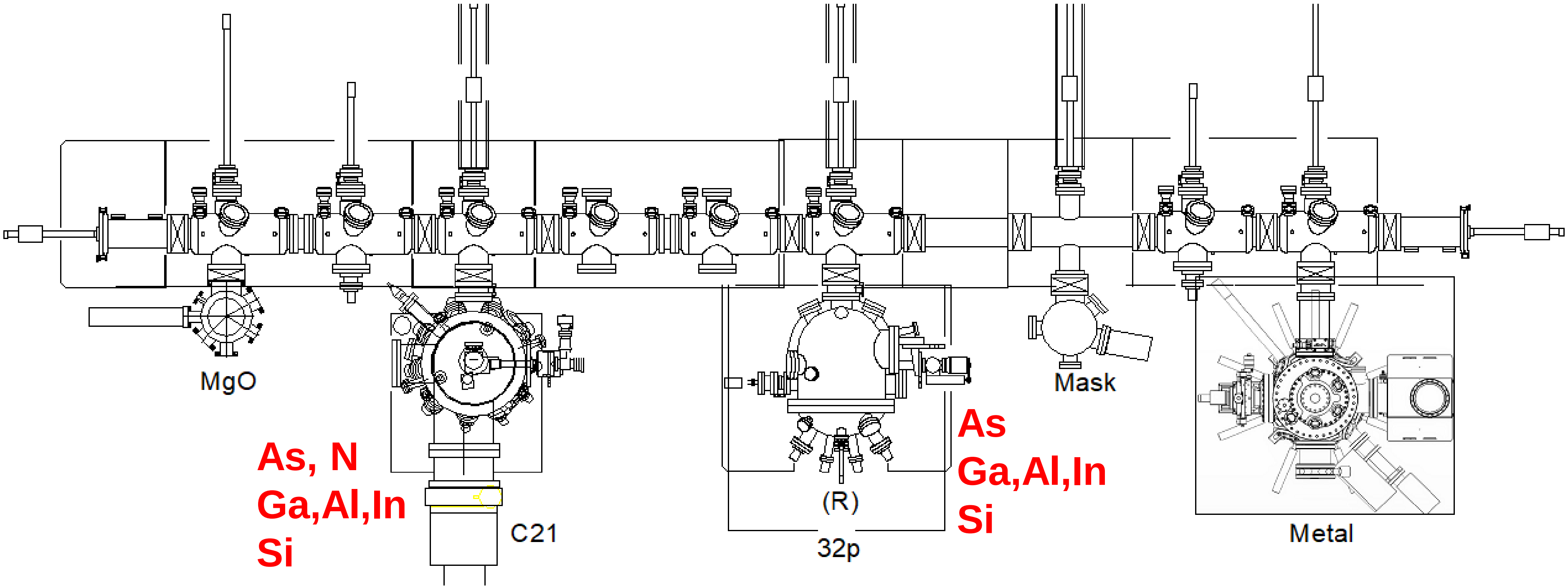
GaAs/AlGaAs

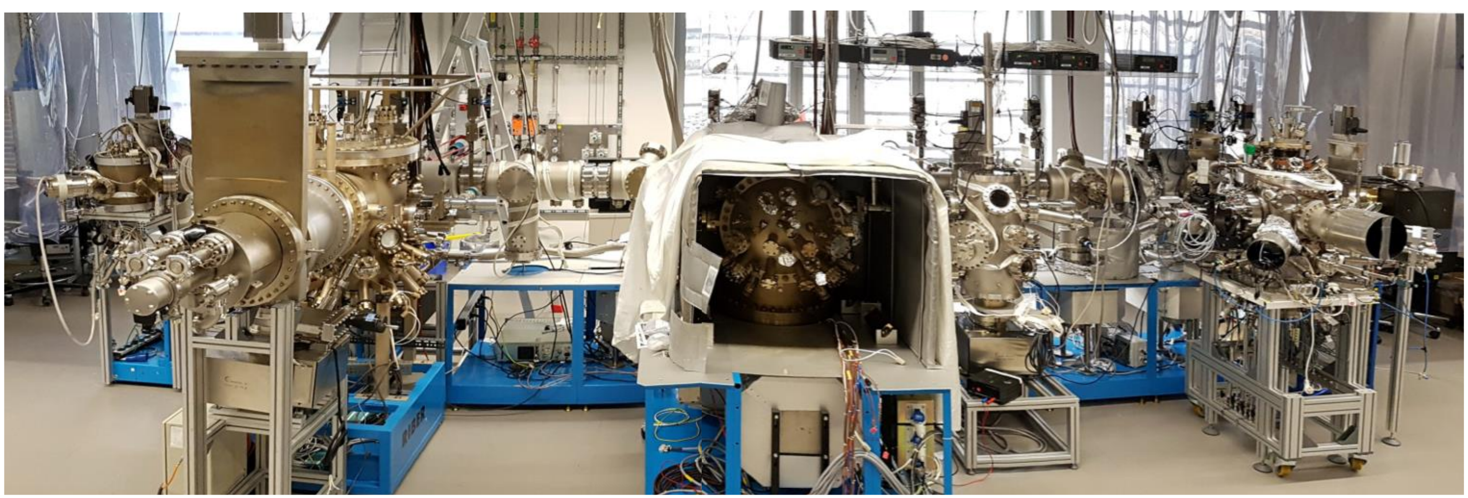
Emission: 680-810 nm

Nanohole drilling
Filling with QD



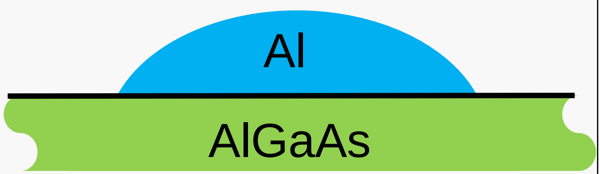
Molecular Beam Epitaxy: CHyN Cluster



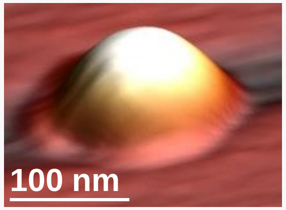


Droplet Etched QDs

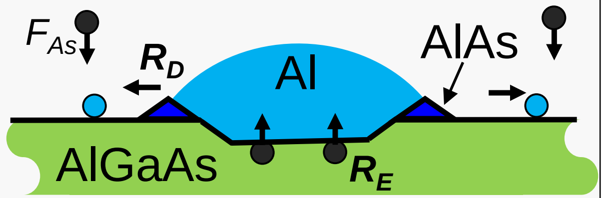
<u>Droplet deposition</u>



Minimization of surface/ interface-energies

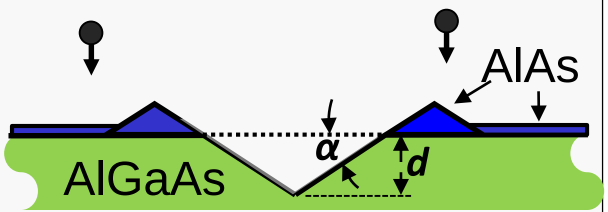


<u>Annealing</u> (early stage)

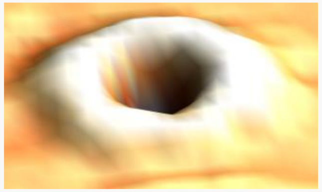


Etching, formation of crystalline wall

<u>Annealing</u> (≦ 100 s)



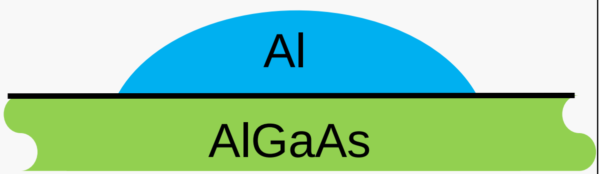
Droplet material removal



- Hole density: $2\text{x}10^6$ – $2\text{x}10^9$ cm^{-2}
- Hole depth: 2 - 120 nm

Droplet Etched QDs

Droplet nucleation



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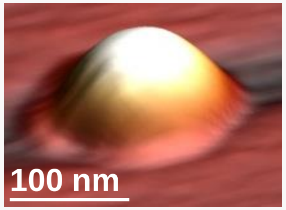

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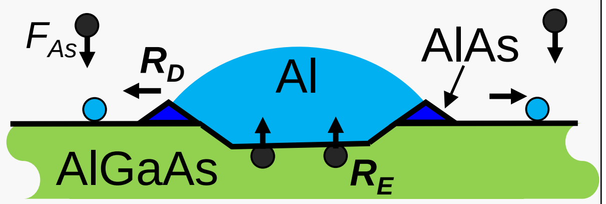

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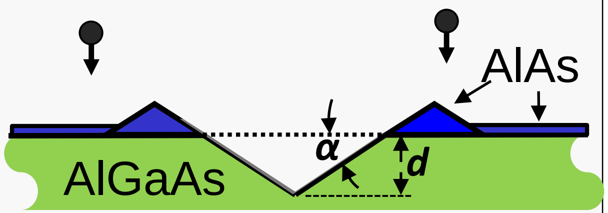
Annealing (≦ 100 s)



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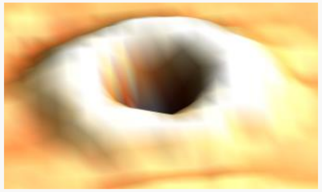
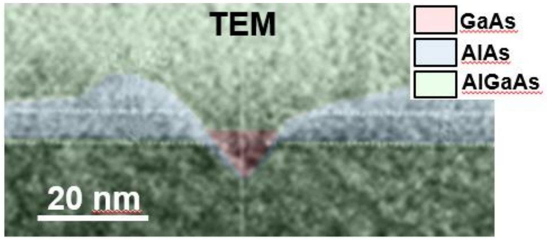
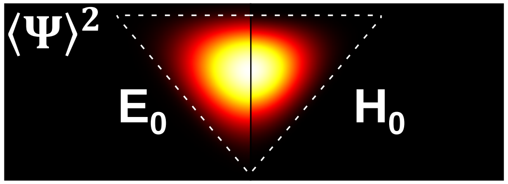


- Hole density: $2x10^6 – 2x10^9$ cm^{-2}
- Hole depth: 2 - 120 nm
- **Hole filling**: highly uniform QDs, size controlled by filling
- QD shape controlled by hole template

Cone QDs

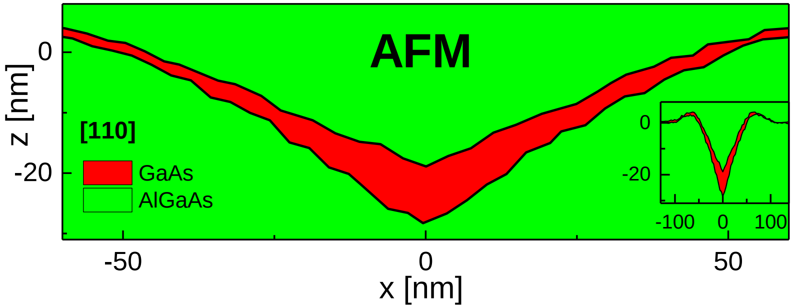
Sphere like $\langle\Psi\rangle^2$

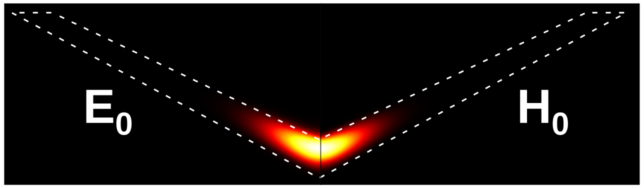




Cone-shell QDs

Disk like $\langle\Psi\rangle^2$





Optical Properties

Low excitation: exciton lines
(linewidth down to 25 µeV)

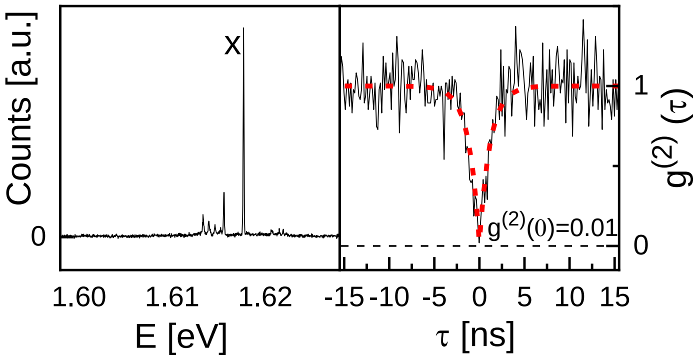
High excitation: shell structure
*[Ch. Heyn et al., APL **94** (2009)]*

Emission wavelength controlled by hole filling d_F : 680 - 810 nm

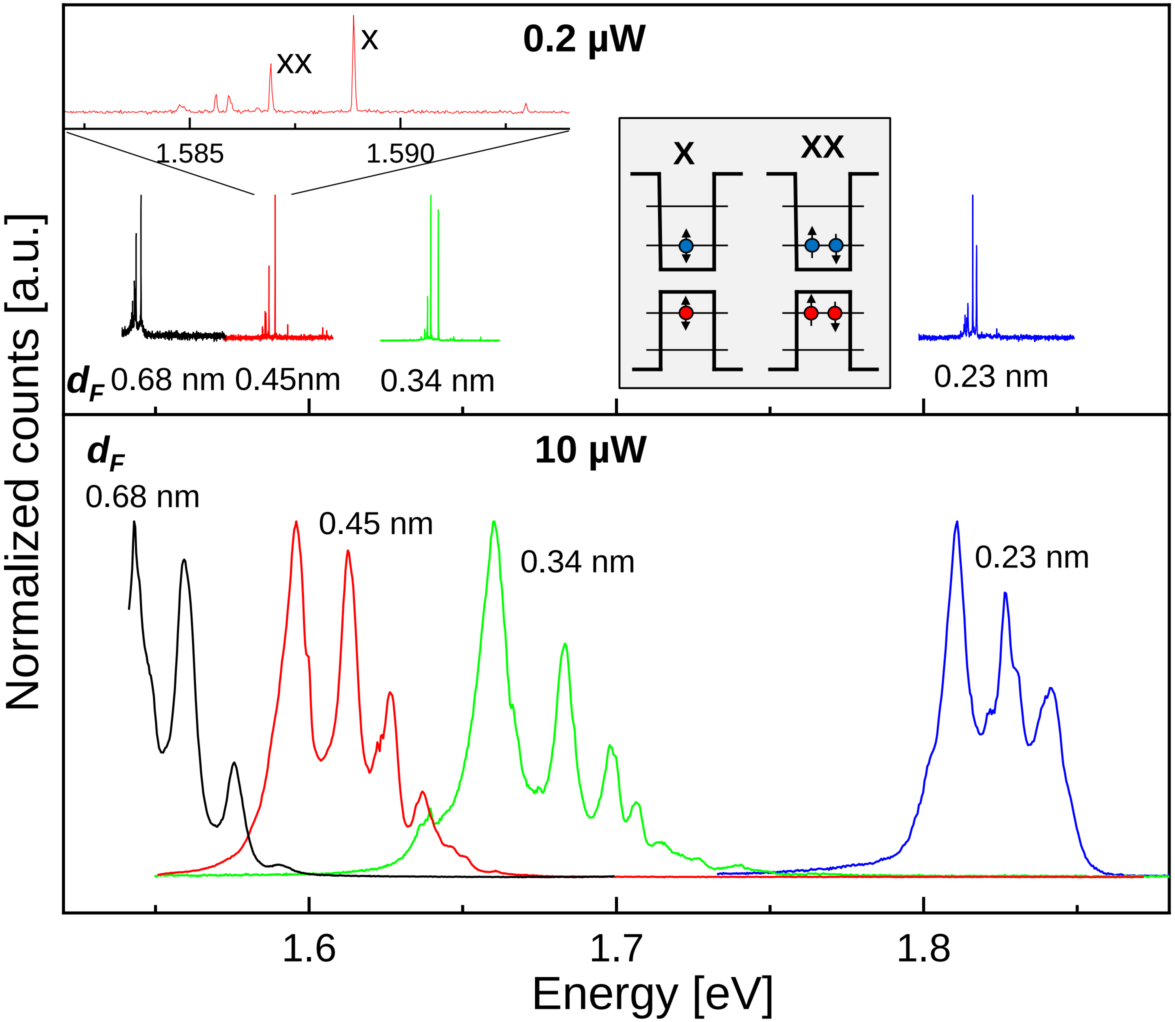
Exciton lifetime: 400 – 800 ps
*[Ch. Heyn et al., N. J. Physics **14** (2012)]*

Fine-structure splitting: 4.5 µeV

Single-photon emission $g^{(2)}(0)$: 0.01
*[A. Küster et. al., Nanoscale Res. Lett. **11** (2016)]*



Single-dot pholumineszenz, T = 8K, cone-shell QDs
Varied excitation power and filling layer thickness d_F



Cone-shell QDs: Wave-Function Tuning

Vertical electric field

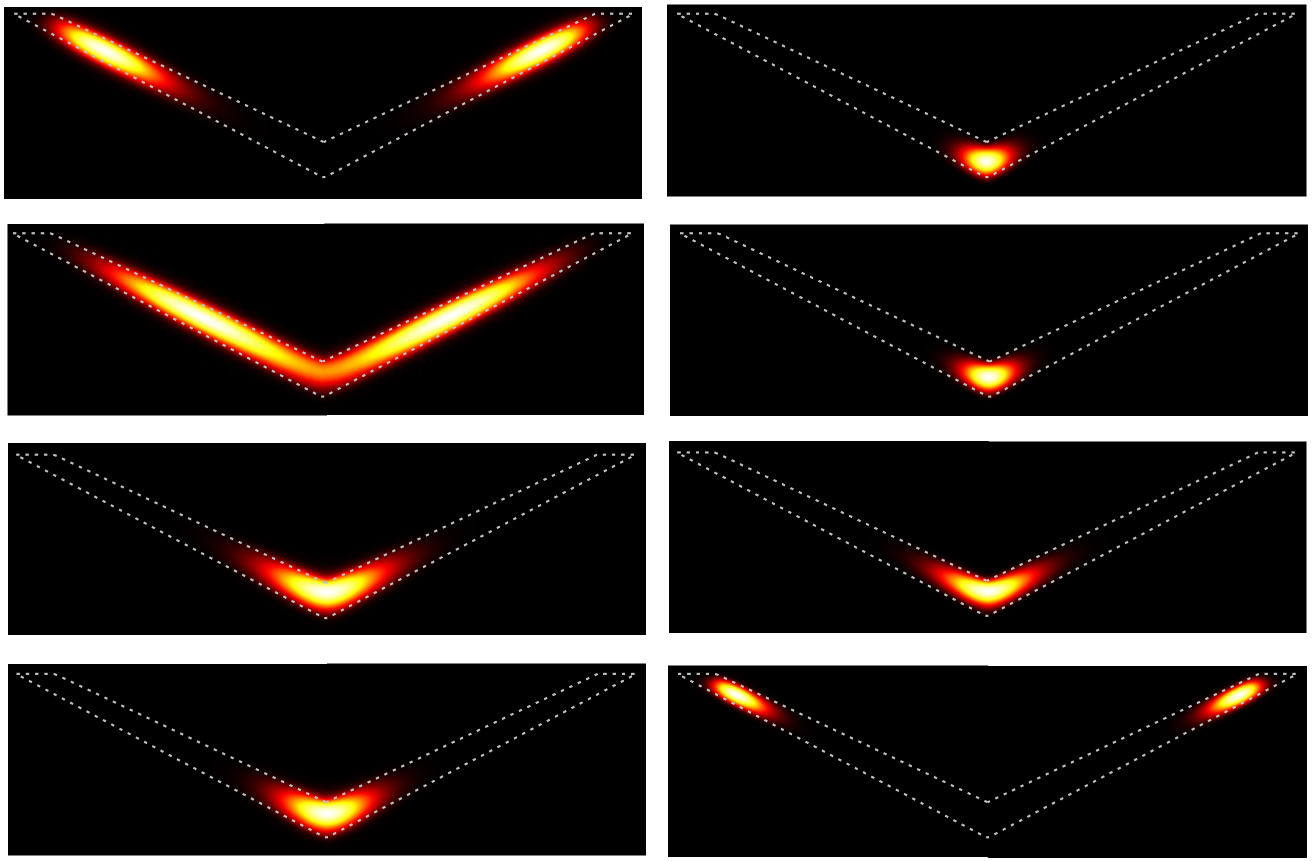
Electron, $\Psi^2_{e,0}$ | Hole, $\Psi^2_{h,0}$

F = -30kV/cm

F = -20kV/cm

F = 0

F = 20kV/cm



Ring e

Dot

Ring h

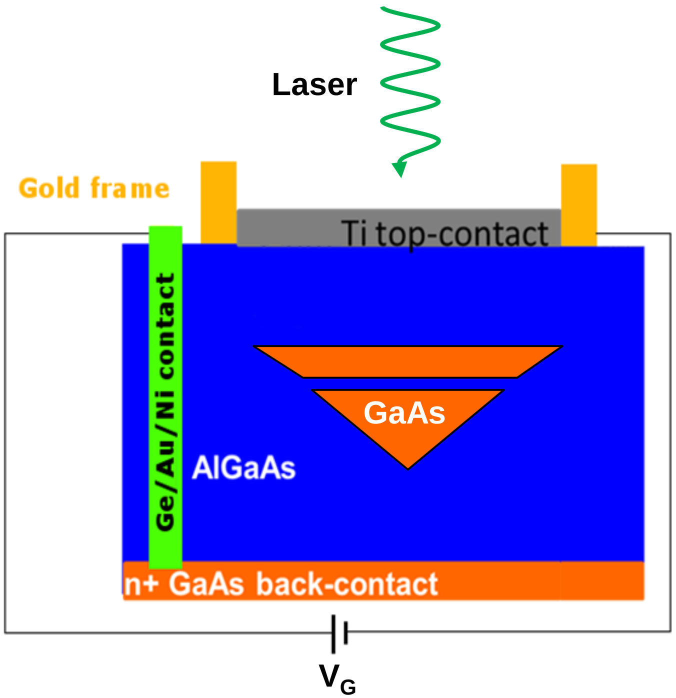
Simulations:

[Ch. Heyn et al., phys. stat. sol. RRL (2018)]

- **Field-controlled dot to ring transition of either electron or hole**
- **Elongation of radiative lifetime up to miliseconds → quantum memory?**

- Vertically self-aligned
  (2 QDs and barrier in a nanohole)
- Adjustable QD sizes and barrier thickness
- Energy-state tuning via a vertical electric field by gate voltage V_G

*[D. Sonnenberg et al., Nanoscale **6** (2014)]*

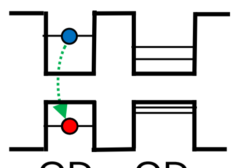


GaAs QD Molecules

- Direct X and indirect excitons iX
- Anticrossing: **delocalized molecule states** (either symmetric or antisymmetric) → strong coupling
- Simulations of X, iX, anticrossing

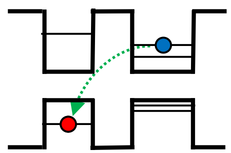
Direct exciton, X

Small Stark-shift
(small QD_T height)



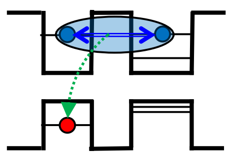
Indirect exciton, iX

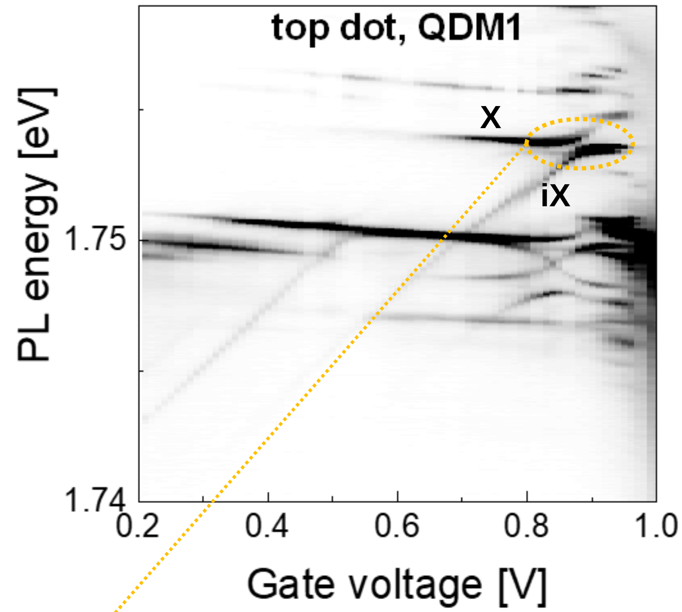
Strong field dependence
(large QD_T - QD_B distance)
blue-shift: h - QD_T, e - QD_B

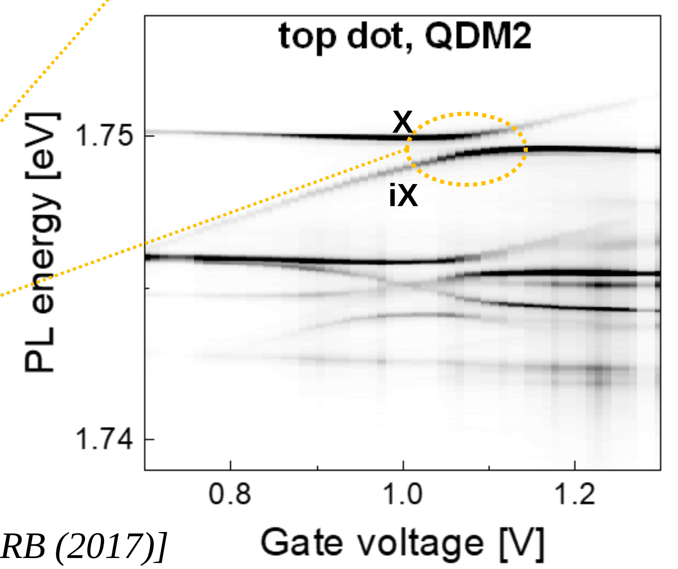


Anti-crossing

Resonance
(delocalization due to tunneling)







[Ch. Heyn et al., PRB (2017)]

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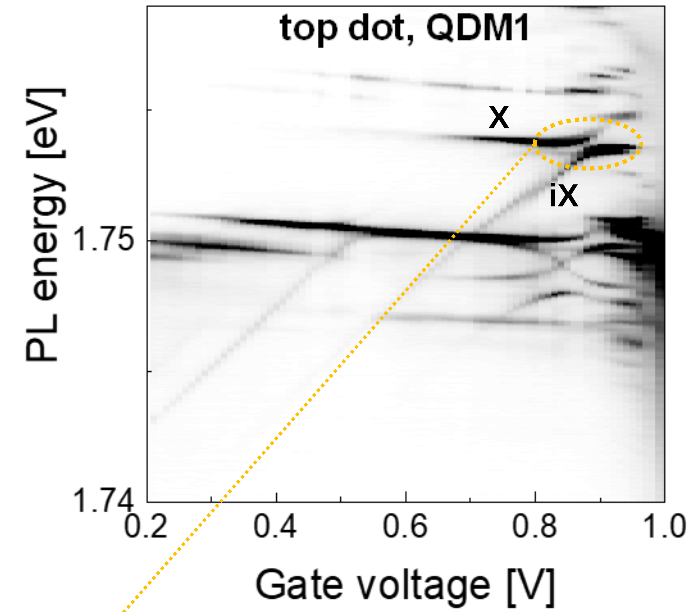
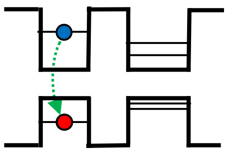


Direct exciton, X

<u>Small Stark-shift</u>
(small QD_T height)



Indirect exciton, iX

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(large QD_T - QD_B distance)
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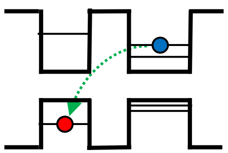

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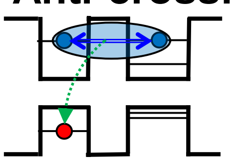
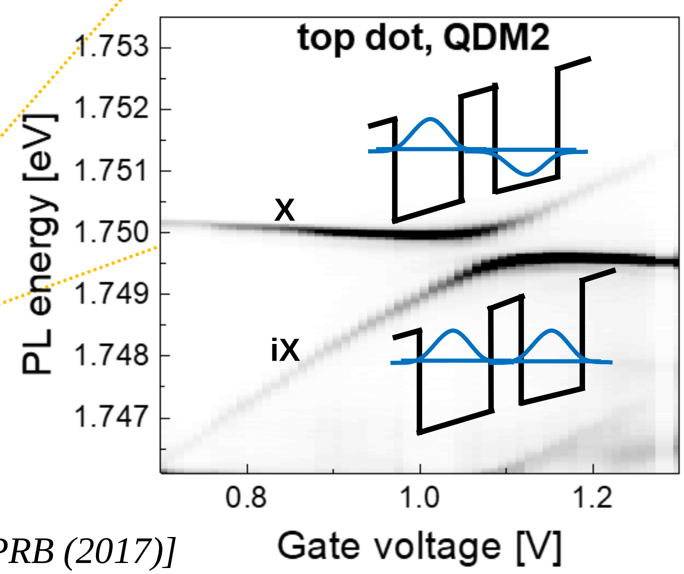
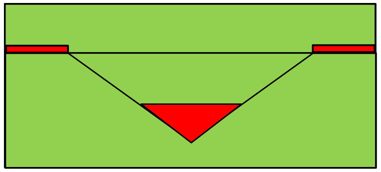




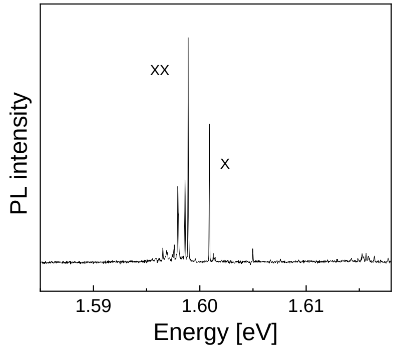
[Ch. Heyn et al., PRB (2017)]

Summary: Quantum Structures by Droplet Etching

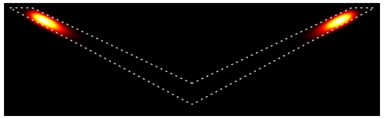
- **Epitaxial QDs:**
  Deterministic source of single and entangled photons
  Wide range of applications in photonic quantum technology



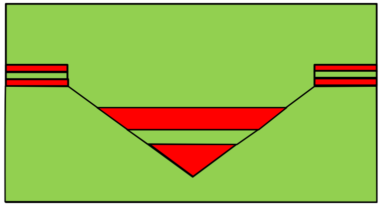
- **QDs by droplet etching**
  Morphology: strain-free, low density, size+shape tunable
  Single-dot PL: sharp exciton lines, tunable wave length, single-photon emission
  Tunable wave function → quantum rings, ms lifetime



- **QD molecules**
  Molecule-resonant electron states → strong coupling



- **Outlook**
  Hybrid with Rb quantum memory, site-control, integration into photonic environment, …



HA 2042/6+8, HE 2644/2-1
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