

Coherence in semiconductor nanostructures

Part I: Generalities

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UNIwersYTET
WARSAWSKI



ZINTEGROWANY
PROGRAM ROZWOJU

Equipe mixte CEA-CNRS "Nanophysique et semiconducteurs"
Institut Néel - CNRS
Grenoble France

Warsaw University, October-December 2020

Plan of the Lectures, 30 hours

- 1 Semiconductors, nanostructures & excitons
- 2 Enhancing light-matter interaction with photonic devices
- 3 Spontaneous coherence in ensembles of excitons and polaritons
- 4 Notions of nonlinear spectroscopy
- 5 Retrieving single exciton coherence: experimental challenges
- 6 Single exciton coherence exploited with four-wave mixing
- 7 Exciton-cavity system in the quantum strong coupling regime
- 8 Coherent coupling in small ensembles of excitons
- 9 Coherent spectroscopy of excitons in TMDs and their heterostructures

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Outline

- 1 Semiconductors
- 2 Nano-scale
- 3 Exciton Generalities
- 4 Nano-Structures

What is a semiconductor ?

A solid in which opto-electronic properties can be tuned via size, composition & **controlled doping**

- ◉ Electronic transport



- ◉ Absorption and emission of light



Photo-Voltaics



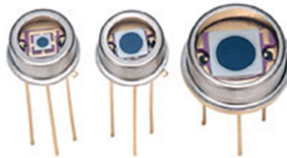
Light Emitting Diodes

What is a semiconductor ?

A solid in which opto-electronic properties can be tuned via size, composition & **controlled doping**



Tunable Laser diodes



Avalanche Photo Diodes



From UV and blue to Mid-Infra Red and THz range

What is a semiconductor?

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incandescence



LED

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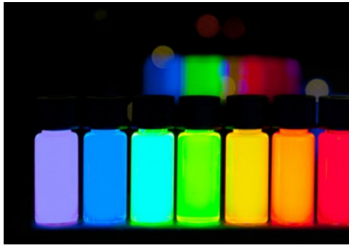


LED



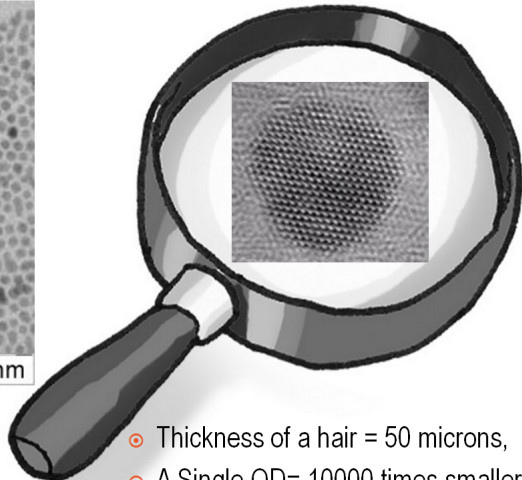
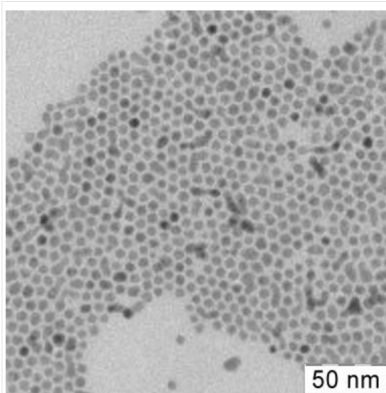
Towards the **nanophysics**

NanoMeter=0.000000001 Meter

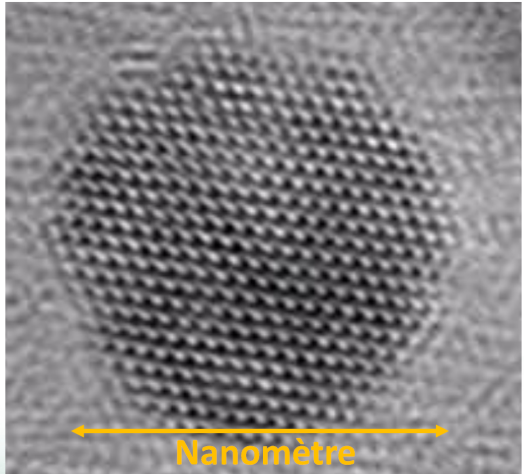


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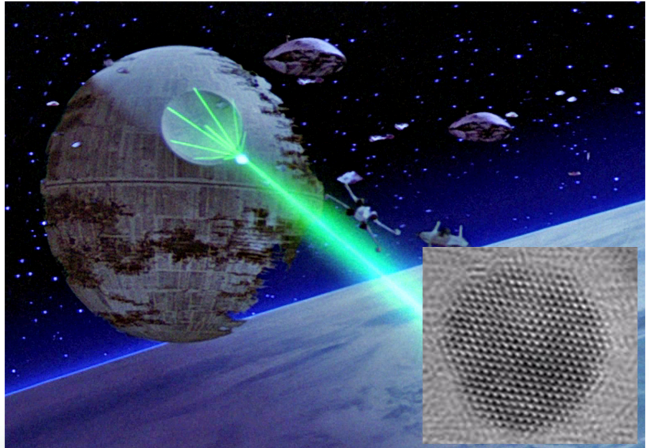
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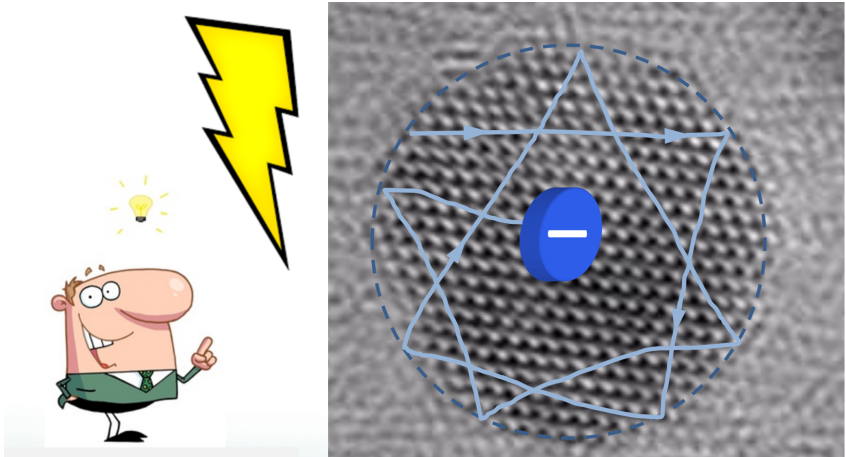
Nano-emitter \Rightarrow **A single semiconductor Quantum Dot**
nanometric trap for charge carriers, 1 photon out each nano-second



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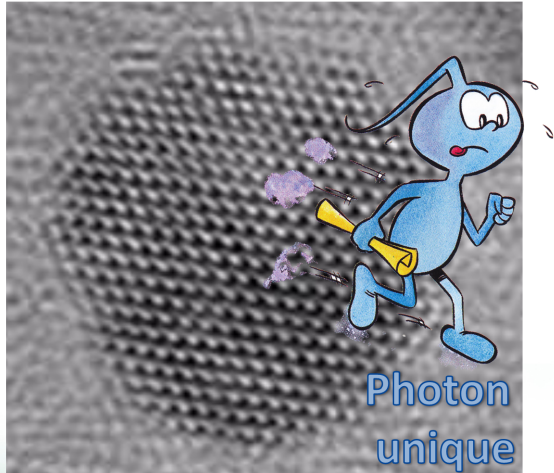
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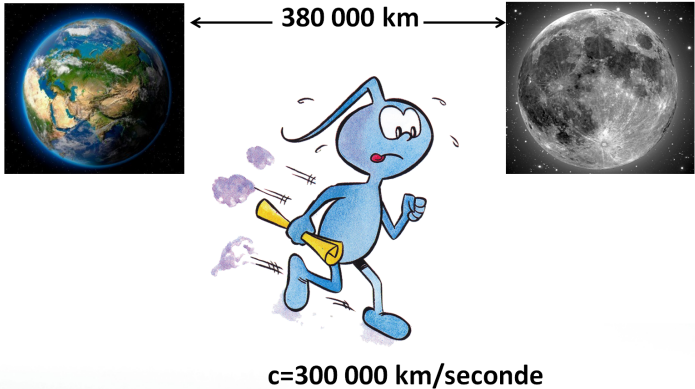
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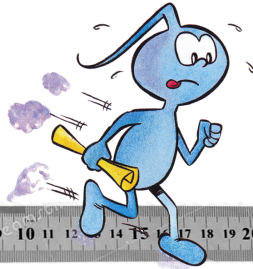


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seconde



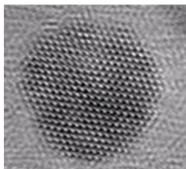
nano
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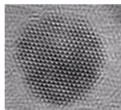
30cm

Optical spectroscopy

color = size + composition (alloys)



red



green

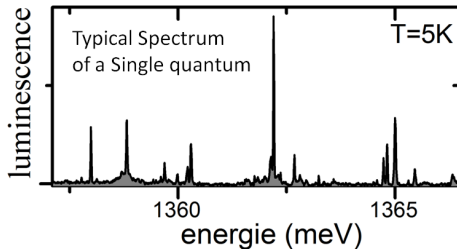


blue

Optical spectroscopy

color = size + composition (alloys)

- Intensiy vs. Color (Energy, wavelength, frequency)

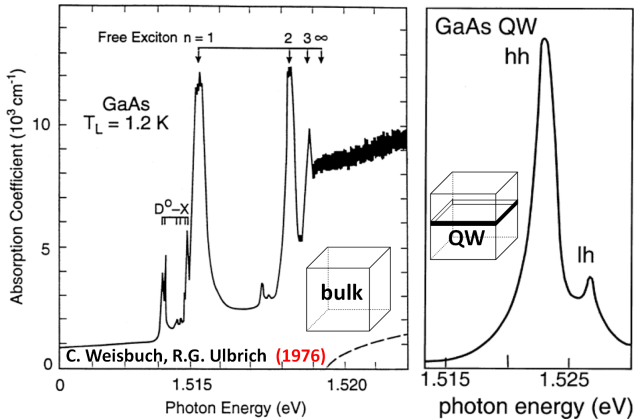


Semiconductor Optics \Rightarrow Excitons

Ryd ~ 0.01 eV, 1000 times weaker than in atoms



G. H. Wannier Phys. Rev. 52, 191 (1937): "the electron cannot escape its hole completely"

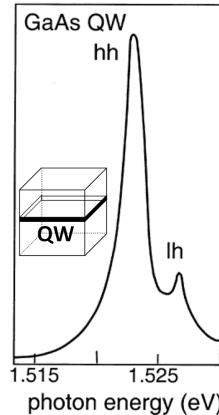
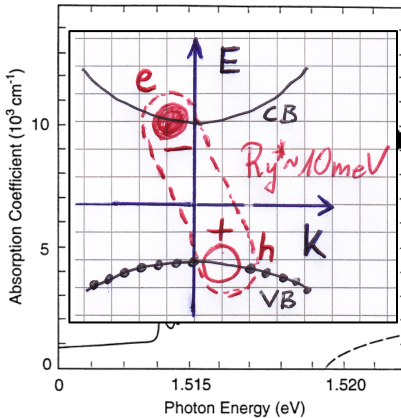


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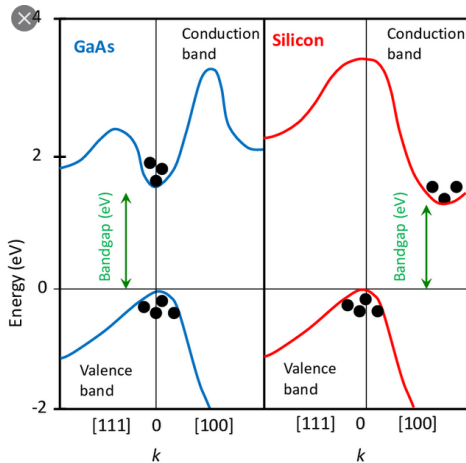
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Optical Activity \Rightarrow Bandgap: Direct vs. Indirect

Direct: GaAs, InAs, CdTe, TMD monolayers, Cu₂O

Indirect: Si, Ge, TMDs bulk and multilayers



How to enhance the light-matter interaction ?

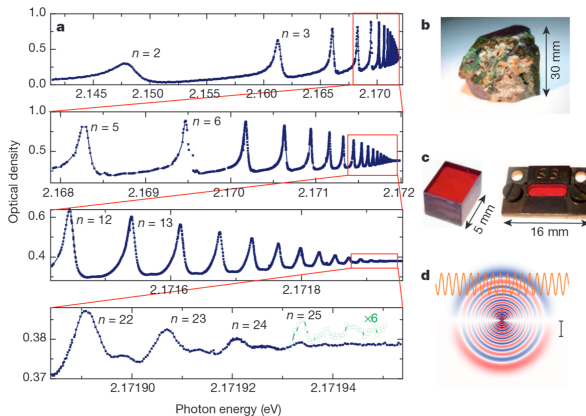
Increase exciton oscillator strength, most prominent example is **Cu₂O**



Gross (1952), ..., Kazimierzczuk et al., Nature 514, 343 (2014)

Exciton extension up to 2 μm for high Rydberg states

Giant Rydberg excitons in the copper oxide Cu₂O



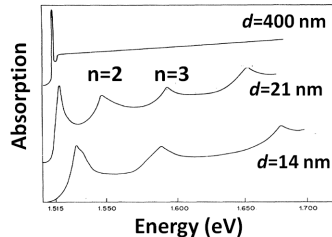
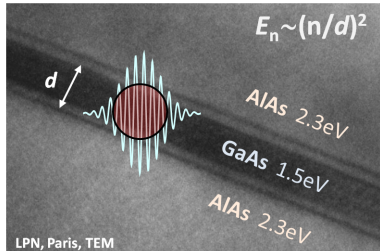
Quantum confinement in nanostructures

Material growth with atomic precision \Rightarrow wavefunction engineering

Quantum Wells: $d \sim \lambda_{dB} \sim (mT)^{-1/2}$

Quantum States of Confined Carriers in Very Thin $\text{Al}_x\text{Ga}_{1-x}\text{As-GaAs-Al}_x\text{Ga}_{1-x}\text{As}$ Heterostructures

R. Dingle, W. Wiegmann, and C. H. Henry
Bell Laboratories, Murray Hill, New Jersey 07974
(Received 24 June 1974)



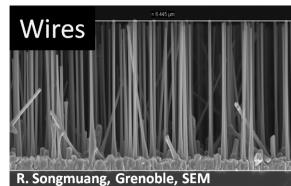
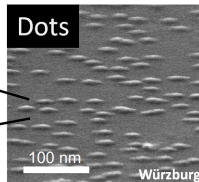
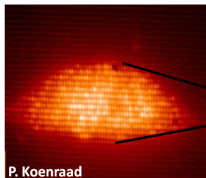
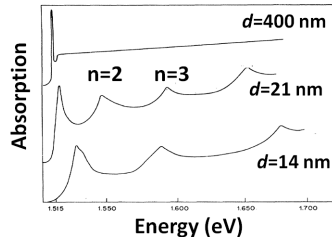
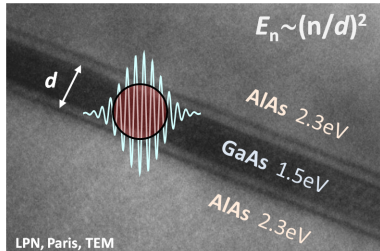
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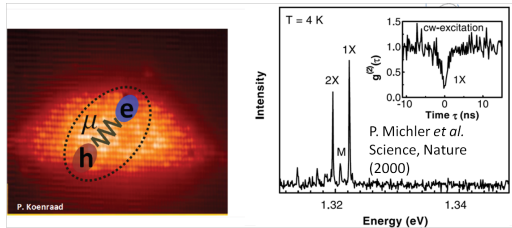
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A quantum dot exciton

Bright, nano-scopic source of quantum light from the solid

Optically driven, fast qubit ?



two-level system

embedded in the
crystal \rightarrow **phonons**

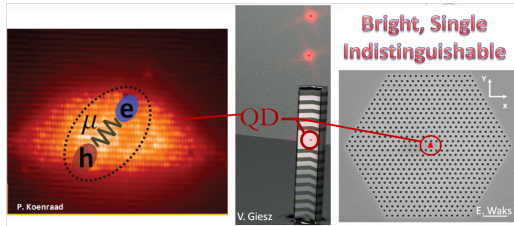
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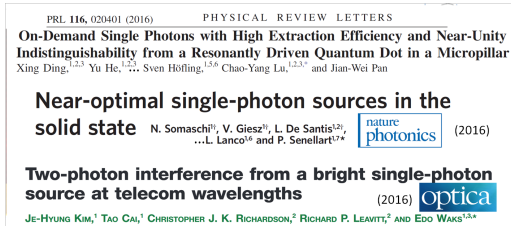
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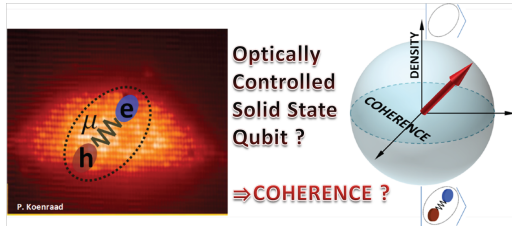
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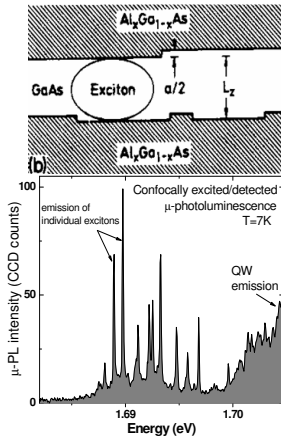
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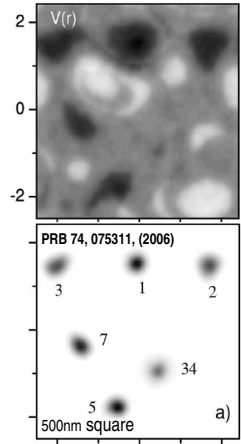
Single excitons localized on interface fluctuations of a Quantum Well



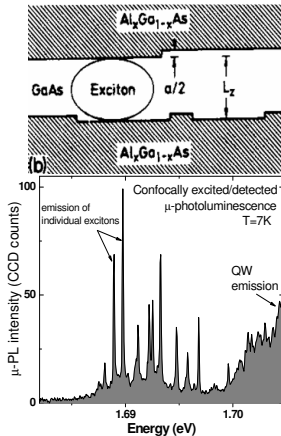
AlAs/GaAs/AlAs 5 nm single QW

- Growth Interruption \Rightarrow Formation of Monolayer Islands
- Wedge \Rightarrow Variation of Average Thickness
- Disorder Potential $\Rightarrow |\Psi|^2$ of localized X
- Large Extension \Rightarrow Large Osc. Str. \Rightarrow

Suitable for Non-Linear Spectroscopy



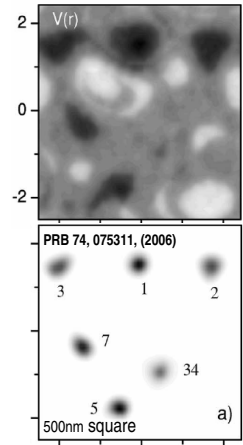
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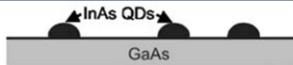


Multi-step self-assembly of Quantum Dots



Phys. Rev. Lett. 92, 166104, (2004)

1. Single QD layer



Characteristics

- emission ~ 750 nm
- strong-confinement: 200 meV
- ultra-low density
- strain-free
- controlled geometry

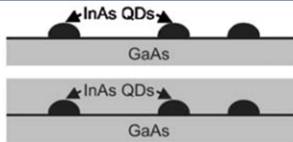
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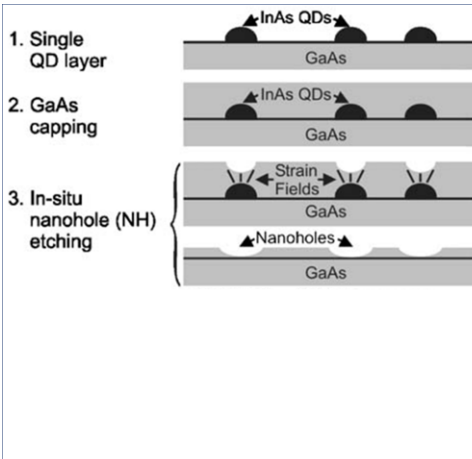
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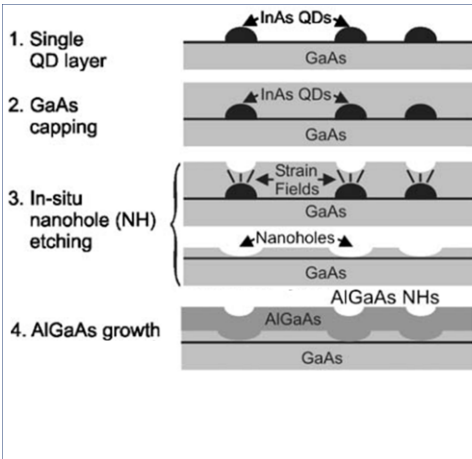
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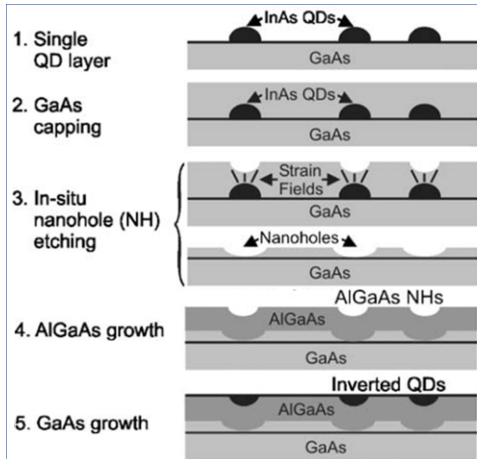
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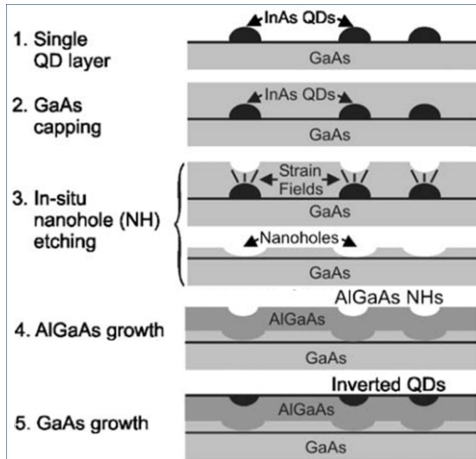
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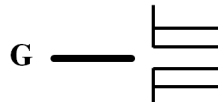
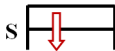
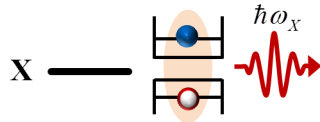
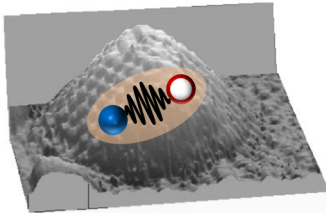
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Typical **exciton complexes** in a semiconductor QD

Neutral excitons, biexcitons, trions, radiative cascades



Y. Benny et al. Phys. Rev. B 86, 085306 (2012), PhD de Santis

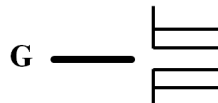
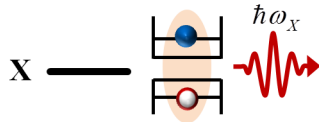
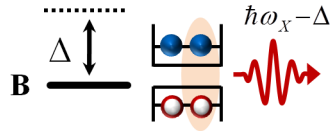
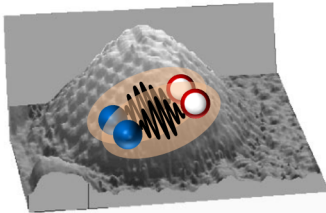


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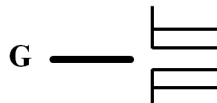
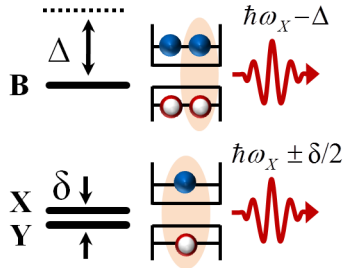
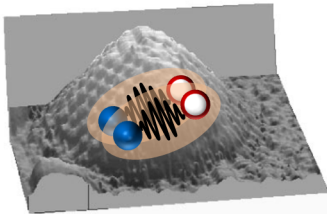


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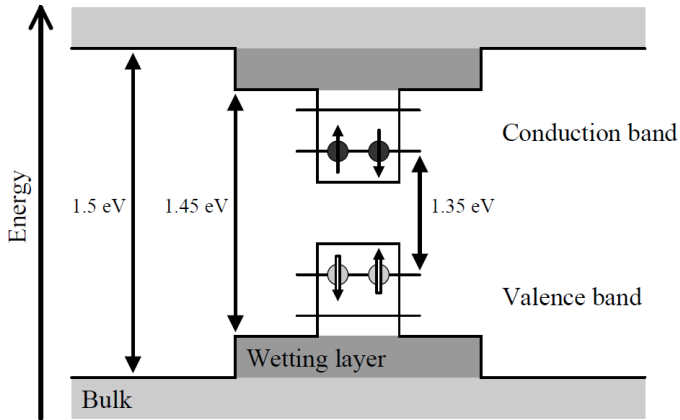


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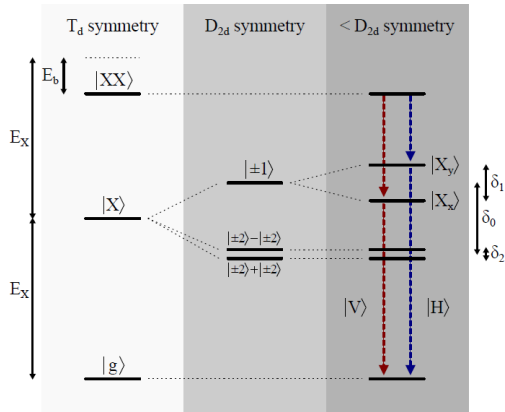


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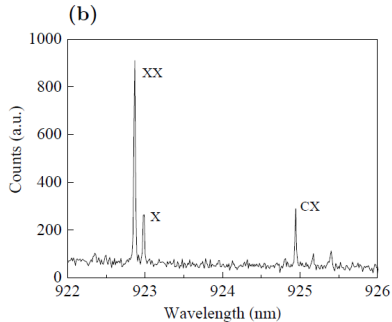
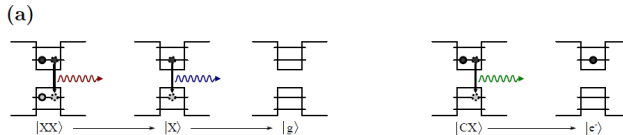


Typical **exciton complexes** in a semiconductor QD

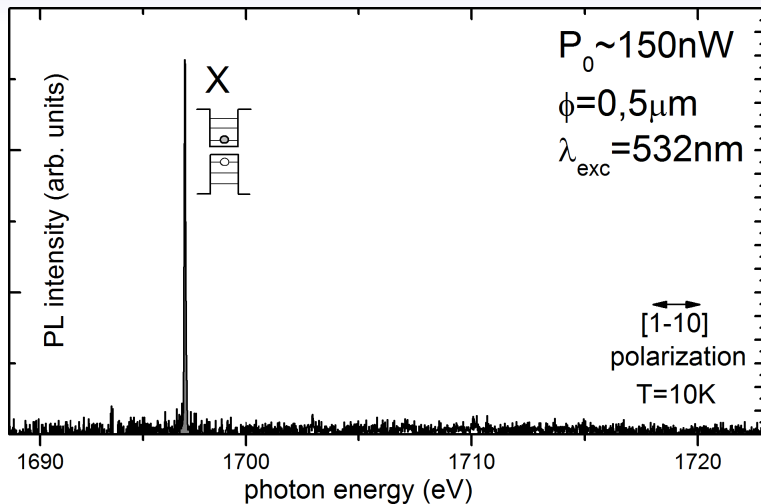
Neutral excitons, biexcitons, trions, radiative cascades



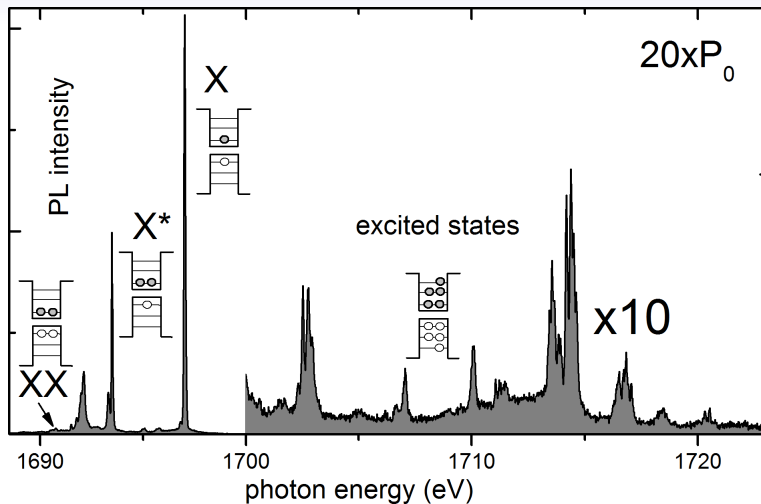
Y. Benny et al. Phys. Rev. B 86, 085306 (2012), PhD de Santis



Typical photoluminescence



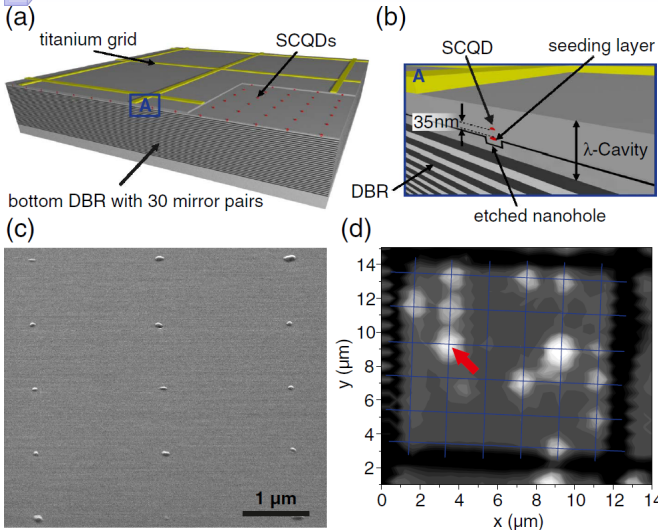
Typical photoluminescence



Site-controlled fabrication of Quantum Dots



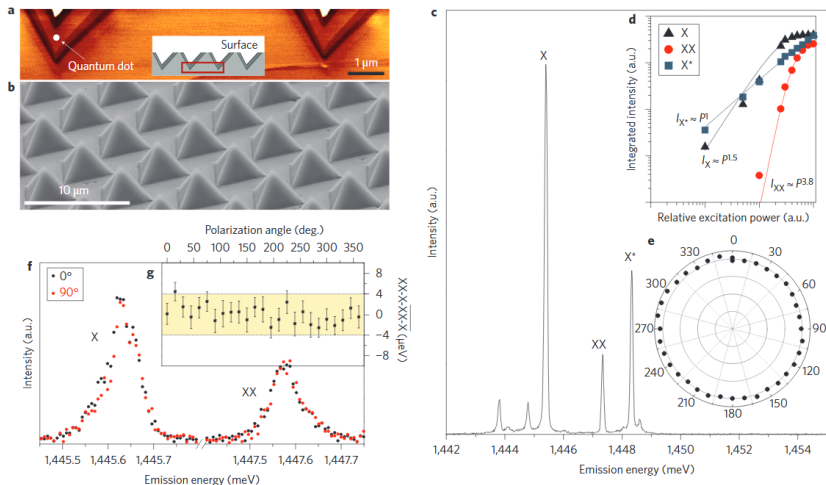
Optica 15, 2334, (2015)



Highly uniform deterministic arrays of **pyramidal** QDs

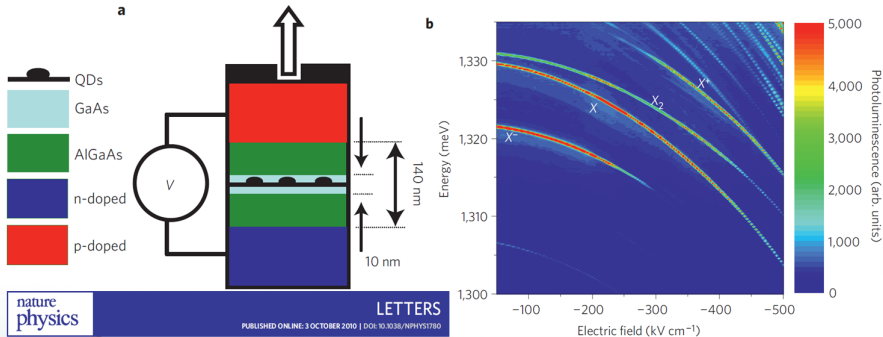
LETTERS

NATURE PHOTONICS DOI: 10.1038/NPHOTON.2013.128



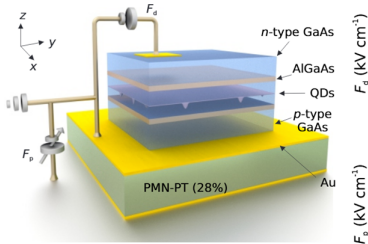
Tuning of exciton transitions in Nanostructures

Temperature, strain, magnetic field, **electric field** via Stark effect

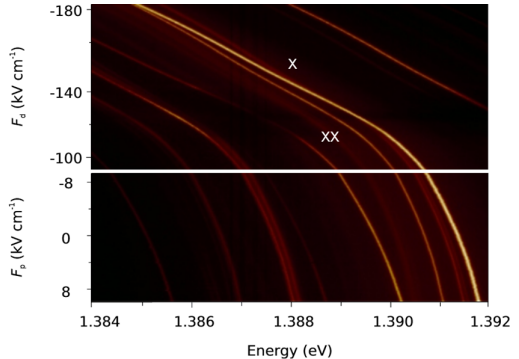


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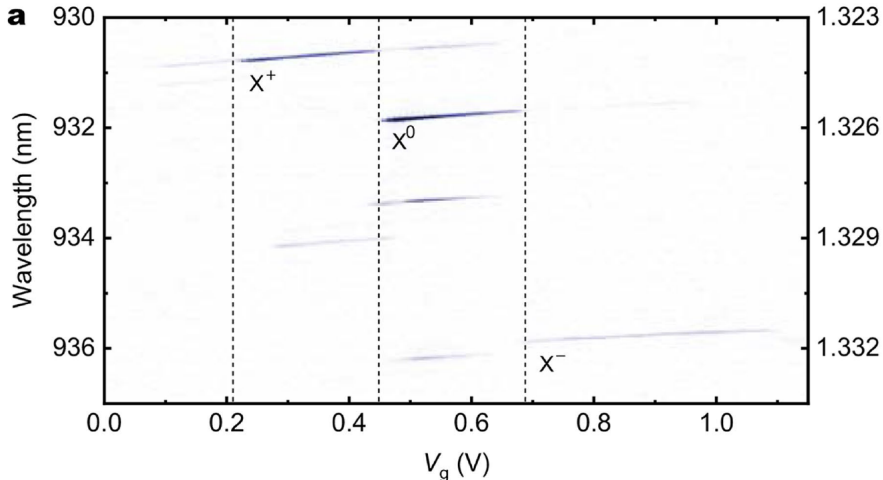


Nano Lett. 2017, **17**, 501–507



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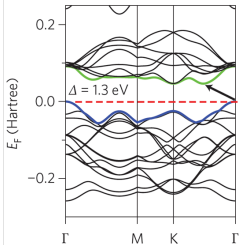
Excitons in Semiconducting TMDs



K. F. Mak *Phys. Rev. Lett.* **105**, 136805 (2010): "a crossover to a direct-gap material in the limit of the single monolayer (...) increase in luminescence by 10^4 compared with the bulk"



WS₂ bulk

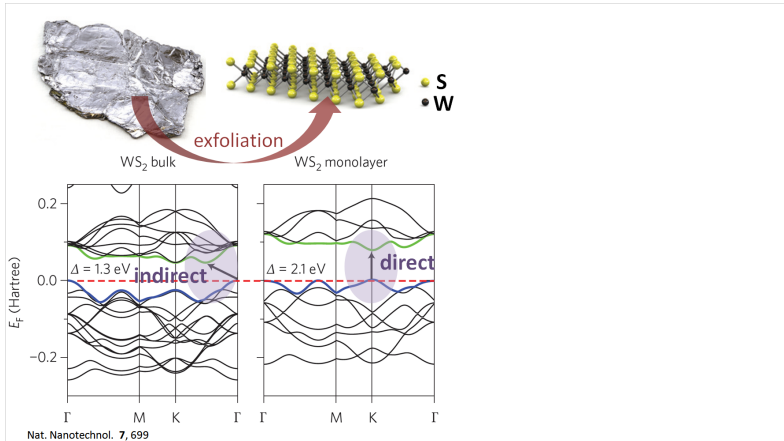


Nat. Nanotechnol. **7**, 699

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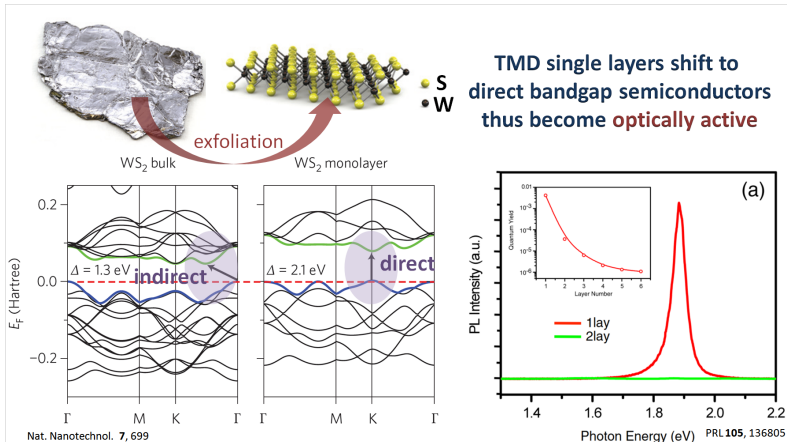
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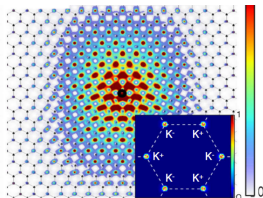
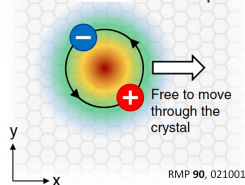
Basic characteristics of free 2D excitons



L. C. Andreani, lectures

- **Wannier** picture applicable to large extent
- Energy: $E_n(K_{||}) = E_g + E_e + E_h + \frac{\hbar^2 K_{||}^2}{2M} - \frac{R^*}{(n-1/2)^2}$,
 $R^* = \frac{\mu^* e^4}{(2\epsilon^2 \hbar^2)} > 100 \text{ meV}$
- Bohr radius: $a_{2D} = \frac{\epsilon \hbar^2}{(\mu^* e^2)} \simeq 1 \text{ nm (Compact!)}$
- 10% (**Strong!**) Linear absorption \propto oscillator strength $\propto 1/a_{2D}^2$
- **Chiral** selection rules $\sigma^\pm \leftrightarrow K^\pm$

TMD exciton: robust & compact



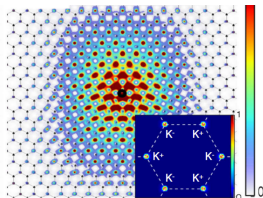
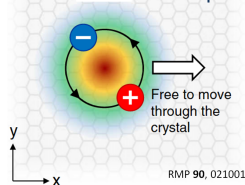
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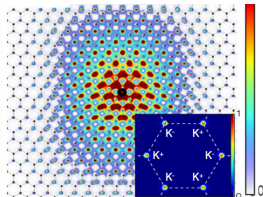
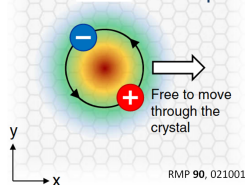
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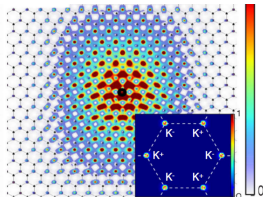
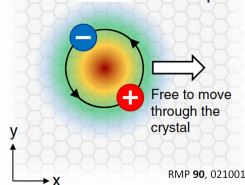
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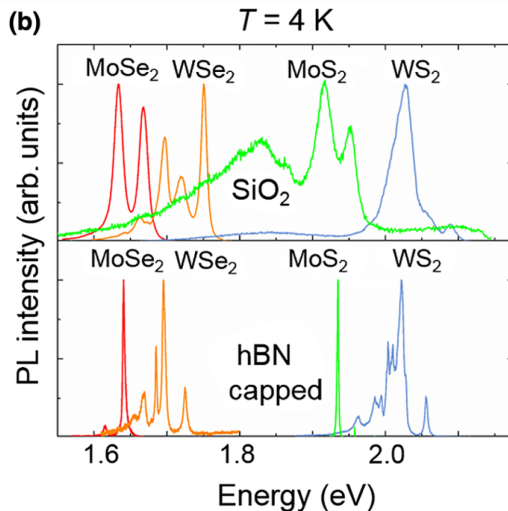
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Improving optical quality with heterostructures

Flattening, Shielding & Isolating from excess charges \Rightarrow suppressing σ



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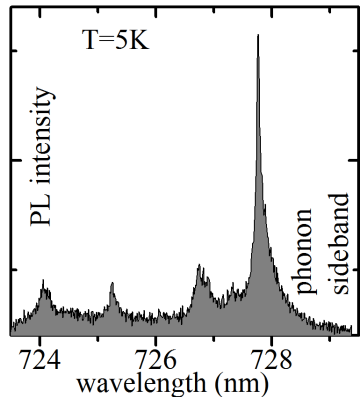
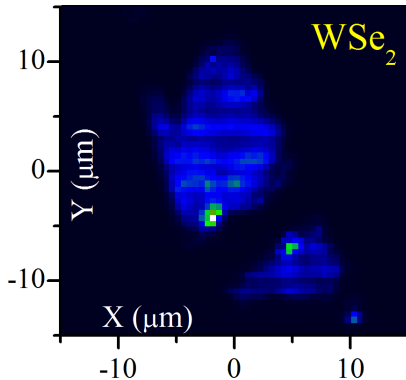


Localized quantum emitters in monolayers

Driven by local strain/disorder, ...but on-demand arrays now at hand



Koperski et al., *Nature Nanotech.* 10, 503 (2015), [arXiv:1609.04244](https://arxiv.org/abs/1609.04244) (!)

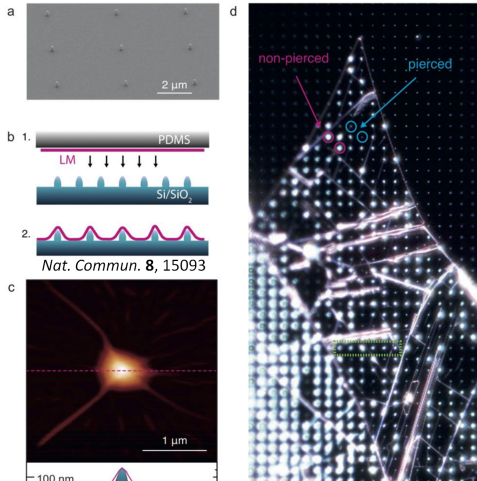


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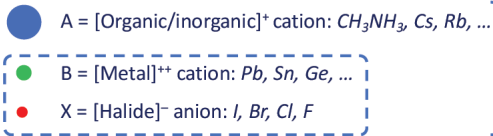
Koperski et al., *Nature Nanotech.* 10, 503 (2015), [arXiv:1609.04244](https://arxiv.org/abs/1609.04244) (!)



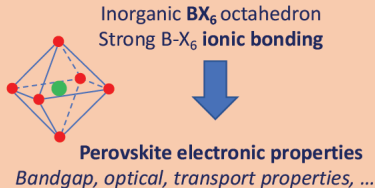
Excitons in Perovskites

Basic Physical Properties Structure

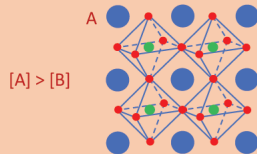
Halide perovskites ABX_3



- Hybrid (organic-inorganic) perovskites
 $CH_3NH_3 BX_3$
- All-inorganic perovskites
 $Cs BX_3$



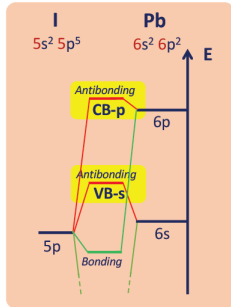
3D Corner-sharing BX_6 network Weak A- BX_6 bonding



Active "core" BX_6 - "shell" A

Excitons in Perovskites

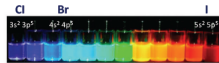
Basic Physical Properties Structure


 BX_6
 $B = Pb, X = I$


s-p dipolar interband	VB	CB
Perovskites	s	p
CdTe, GaAs, GaN, ...	p	s

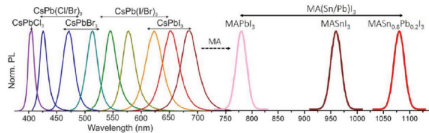
Band gap tunability 400 – 1100 nm

Saliba et al., Angew. Chem. Int. Ed. (2017)



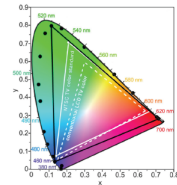
CsPbX₃ nanocrystals

Protesescu et al., Nano Lett. (2015)



Broad color gamut

CsPbX₃ NCs = black dots

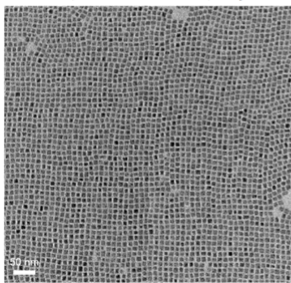


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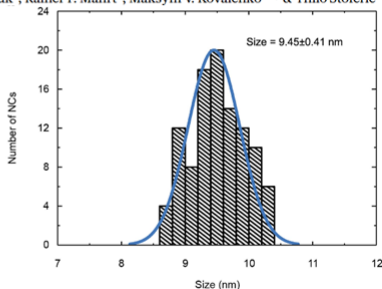
Basic Physical Properties Structure

Superfluorescence from lead halide perovskite quantum dot superlattices

Gabriele Rainò^{1,2,3,5*}, Michael A. Becker^{3,4,5}, Maryna I. Bodnarchuk², Rainer F. Mahrt³, Maksym V. Kovalenko^{1,2*} & Thilo Stöferle^{3*}



Extended Data Fig. 1 | Quantitative analysis of CsPbBr₃ NC size distribution. a, Low-resolution TEM image of the NC material used to prepare the superlattices. b, Histogram of NC sizes (of >100 NCs) as



obtained from TEM image analysis. The solid line is a fit with a normal distribution, and the given mean size (9.45 nm) and standard deviation (0.41 nm) are obtained from this fit.

Perovskites: "miracle" materials for photovoltaics?

Perovskites



Solar cell desirable features	Benefits
Broad bandgap tunability 400 – 1100 nm	Solar radiation, multi-junction
Direct band gap + absorption $\geq 10^4 \text{ cm}^{-1}$	Thin film technology
High photo-generation of free carriers	High Power Conversion Efficiency (J_{sc} , V_{oc})
Low radiative recombination losses	
Carrier diffusion length > absorption depth	
High carrier collection	Consumer market
Cost-effective growth	
Scalability, environmental impact, toxicity	
Lifespan ≥ 25 years	